



**Evaluation of radiation damage on  
lutetium-aluminium and gold for practical  
applications using proton irradiation as a  
surrogate for neutrons**

By

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*A thesis submitted to the Faculty of Science,  
University of the Witwatersrand, Johannesburg  
in fulfillment of the requirements for the award of the degree of*

**Doctor of Philosophy**

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October 12, 2024

# DECLARATION

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I declare that this thesis is my own, unaided work. It is being submitted for the Degree of Doctor of Philosophy at the University of the Witwatersrand, Johannesburg. It has not been submitted before, either in its entirety or in part, for any degree or examination at any other University.



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(Samuel Terungwa TEMAUGEE)

October 12, 2024

## DEDICATION

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*To God Almighty, the all-knowing who  
guided me through every step, I dedicate  
this achievement.*

# ACKNOWLEDGMENTS

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I attribute all glory to God Almighty, the all-sufficient, for bestowing upon me grace, good health, and solace throughout my challenging academic journey, leading to its successful culmination. May His name be praised.

It is with great pleasure that I extend my heartfelt gratitude to my supervisor, Prof. I.T. Usman, whose sagacity, academic contributions, enthusiasm, meticulous planning, provision of funding, and adept coordination have been instrumental in elevating this programme to its current level.

My sincere appreciation extends to my co-supervisor, Dr. R.D. Mavunda, for your invaluable academic guidance and unwavering encouragement. I deeply cherish the early morning calls urging me to strive harder, and your meticulous review of the physics metric systems is highly valued.

I express profound gratitude to all collaborators who closely worked with me, offering corrections and invaluable insights, notably Prof. S.H. Connell and the entire RC group, Dr. G.C. Daniels, Mrs. L. Bedhesi, Mr. E. Chinaka, and the entire Necsa RRT group, whose support significantly contributed to my study.

Special thanks are also due to Prof. M.P. Short, whose mentorship guided me through this journey and afforded me the rare opportunity to conduct experimental work at MIT and Harvard University. I am deeply indebted to the members of the ShortLab group, especially Maxwell Rae, Wande Cairang, Dr. Yong Zhang, Avery Nguyen, Adria Peterkin, Dr. Kevin Woller, and Stanley Chang, whose assistance ensured the success of my experiments during my visit to MIT, USA.

Gratitude is also extended to the NMU CHARTEM group, particularly Dr. A. Janse Van Vuuren, Dr. S. Ngongo, and Dr. V. Vilane, for their contributions to the fruition of this study.

I am grateful to Prof. Deana Naidoo who promptly assented to my applications for conference funds and research visits during my programme. I am also grateful to Dr. Abishek Pande, Dr. Hilary Masenda, and Prof. D. Wamwangi for granting me access to their laboratories and for their valuable input in this work.

I extend my gratitude to the Tertiary Education Trust Fund (TETFUND) Nigeria for the funding provided through my home institution, enabling me to undertake this programme.

I am grateful to the Federal College of Education Kontagora and the Provost, Prof. F.R. Haruna for nominating me for the TETFUND Scholarship and granting me study leave for this endeavour.

My heartfelt appreciation is also directed towards the University of the Witwatersrand for the Postgraduate Merit Award and other funding support for publications. I will forever cherish my affiliation always a Witsie.

I pay homage to my late wife, Barr. Blessing Iveren Terungwa, of blessed memory, for her unwavering support, love, encouragement, and prayers during the application process and the inception of my Ph.D. programme, which she did not live to witness its completion; may she continue to rest in eternal glory.

I am indebted to my parents, my late Dad, Mr. Joseph Chiangi Temaugee of blessed memory, whose aspirations for my academic achievements remain a guiding light. To my mom, Mrs. Alice Anenge Temaugee, a mother with a golden heart and beacon of unwavering support throughout my academic journey, especially during moments of emotional stress, your presence was a source of immense strength.

Special thanks are extended to my siblings, Mr. Timothy Temaugee & Family, Mrs. Doofan Aunde & Family, Mr. Aondongusha Temaugee & Family, Mrs. Doolumun Abimiku & Family, and all extended family members, notably Dr. & Mrs. Emmanuel

Wasem & Family, Barr. Terna Tsumba & family for their unwavering support during this journey.

I express my heartfelt appreciation to my fiancée, Ms. Love I. Omokhoa, for her love, invaluable contributions, unwavering support, prayers, and encouragement throughout the development of this thesis. Your presence is a priceless gift.

I extend deep gratitude to the family of Late DCP Francis Orchia of blessed memory and my mother, Mrs. Esther Orchia, for her constant encouragement, as well as Aondoyavga Orchia, ASP. Aondongu Orchia, Erdoo Aondona, and Late Aondona Orchia of blessed memory, whose calls to check on me were a source of strength before he left for the great beyond. You are all appreciated.

I am grateful to Dr. Samson and Dr. Grace Oreofe Adeyemi and their family for their moral and financial support throughout my Ph.D. journey. May the Lord reward your kindness abundantly.

I express my profound appreciation to Rev. Prof. Credo Mangayi & family, Pst. Felix Kantonda & family, Rev. Raymond Bonglak & family, the entire Berea Baptist Mission Church Johannesburg, Bro. Gugu Zilwa & family, Bro. Sizwe Shumani & family and the entire Living Seed Brethren in Johannesburg. Prof. B.N. Atsumbe & family, Dr. Ngutor Nyor & family, and the entire Living Seed Brethren in Niger State, Nigeria.

Special thanks are also due to various families, groups, and friends whose contributions have been pivotal to the success of this program, namely: Dr. Terna Faajir & family, Mr. Ogbole Onche & family, Mr. Moses Ogianyo & family, Mr. Zecharia Msugh, Pst. James-Mark Akile & family, Barr. Msugh Palegh, Mrs. Susan Utim & family, Barr. Charlse Ogwuche, Barr. Matthew Torsaa, Dr. James Gana, Dr. Adedapo Adeyinka & family, Mr. M. Ayankaa & family, Dr. Kuma Gbanan, Mrs. Laadi Kevin, Dr. Emmanuel Ademola, Dr. N.C. Gatsi, Ms. Offor Ngozika, Mrs. M.M. Adebisi, Bro. D. Oyinloye & family, Dr. Desmond Areghan

& family, Mr. Kalu Ukagbu, Mr. Levi Ogbuaku, Mr. Emmanuel Ojuide & family, Bro. Ope Owolabi, Rev. Sr. Juliana Yarkwan, Prof. Daniel Goon, Prof. N.N. Jibiri, Dr. Raymond Tyohemba & family, Dr. Michael Bodurin, Dr. Funsho Ogun-sola & family, Bro. Patrick McGrath, Bro. Zechariah Habte, Bro. Paul and Mrs. Galateia Mykos and the entire Koinonia family in Boston, USA, Bro. Shadrach Ishegh & family, Bro. Jeremiah Iornumbe & family, Dr. O.E. Olofinniyi & family, Mr. Segun Adewara & family, Mr. Samuel Meseko & family, Dr. Bitrus Gado & family, Mr. I.O. Regha & family, Engr. J. Iorpev & family, Dr. Taiwo Afinowi & family, Dr. Lawrence Christopher, Dr. Joseph Omojola, Dr. Paul Terna & family, Dr. Tivkaa Amande & family, Dr. Abraham Avaa & family, Dr. Don Atsa'am & family, Maj. Ephraim Num & family, CSP. Peter Ilim, Dr. Henry Attah & family, Mr. Steven Olotu & family, Mr. Kamaluddeen Isa El-Kalash & family, Mr. Emmanuel Igba & family, Mr. Chia Terkuma & family, Mr. Iorwuese Mtomga & family, Mr. T. Yoosu & family, Dr. Othmane Mouane, Ms. Ongeziwe Mpantani, Dr. Nyiku Mahonisi, Ms. Saga Abdalla, Mr. Shaibu Dabo, Dr. Ahmed Gomna Kuta, Mr. Awwal Inuwa, Mr. John Aboje, Dr. Musa Bunu Ismail, Mr. A. Ayoosu, Mr. M. Saakorun & family, Ms. M. Anyanwu, Ms. P. Unaji, Dr. Mosebetsi leotlela, Mr. A. Setlhare, Ms. O. Mathlhakola, Mr. Kego Mokokwe, Mr. Kwazi Butelezi, Mr. S. Ogbanako & family, Mr. Geoffrey Mwendwa, Dr. Onesimo Mtintsilana, Dr. Khombo Dumela, Bro. Naaza Badu & family, ASP. M. Wergba & family, Mr. Samuel Adzuu & family, Mr. Femi Oyefeso & family, Dr. Olorunsola Niyi, Mrs. Augusta Ojoko, Dr. Benedict Akano, Mrs T.A. Daniel, Dr. A.R. Lasisi, Dr. C.M Dania, Mr. A.I Dangana, Mr. Abdullahi, Mr. Abubakar Umar, and Mr. K. Arzika.

Finally, to everyone who has contributed to the success of this thesis, I extend my heartfelt gratitude. Your support has been invaluable and deeply appreciated.

# ABSTRACT

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An understanding of the interaction of energetic radiations from particles such as protons, neutrons, and photons, with the microstructure of materials is crucial for predicting their bulk morphological response in extreme radiation environments. Exposure to these radiation species could lead to changes in the microstructural properties that, in turn, affect the mechanical and physical properties of the macroscopic matter. This thesis investigated the resilience of materials, specifically Au and Lu-Al, to radiation damage, employing computational simulation methods and experimental techniques. The study aims to provide critical insights into the radiation damage sturdiness of Au and Lu-Al, considering their application in reactor technology and other extreme radiation environments. Monte Carlo-based methods were employed to calculate radiation damage in the samples resulting from neutron and proton irradiation, utilizing MCNP6.2 and SRIM-2013, respectively. The objective was to compare ion beam irradiation with neutrons with a view to utilizing proton irradiation as a surrogate for neutron irradiation. Three different state-of-the-art characterization techniques—X-ray diffraction (XRD), High-Resolution Transmission Electron Microscopy (HRTEM), and Flash Differential Calorimetry (F-DSC)—were employed to evaluate damage in the materials before and after proton irradiation using the CLASS Accelerator at MIT, USA. The results of the study indicated that protons within the energy range 0.1 to 1.0 MeV produced similar types of damage in the materials as would neutrons (spectrum  $0 < E \leq 20$  MeV at SAFARI reactor), suggesting protons as an alternative to neutron irradiation. Defect characterization in the materials using XRD and HRTEM techniques revealed dislocation loops and lines in both Lu-Al and Au, as well as Stacking Faults Tetrahedra (SFT) in the Au material. These defects with proton irradiation were similar to those observed with neutron irradiation in Au and other aluminum alloys. The estimated defect number density ranged from 0.7 to  $4.8 \times 10^{14} \text{ m}^{-2}$ , showing an increase with rising displacements per atom (dpa) or proton fluence post-irradiation. Lu-Al exhibited higher defect density values than Au, consistent with Monte Carlo simulations. Furthermore, results from the Flash DSC technique revealed significant

changes in the characteristics of the power-temperature profiles (melting curves) of Lu-Al as dpa increased, offering insights into radiation-induced processes such as phase transition and precipitate stability at specific defect annealing temperatures. These findings are crucial for radiation damage studies for the binary alloy and warrant further investigation. The observed microstructural defect densities were relatively high, and prolonged exposure of the materials to higher doses could lead to further changes in microstructural properties, consequently influencing the physical and mechanical properties of the macroscopic material.

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# List of Acronyms/Abbreviations

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**CLASS** Cambridge Laboratory for Accelerator Study of Surfaces

**dpa** Displacement-Per-Atom

**DSC** Differential Scanning Calorimetry

**FOM** Figure of Merit

**FC** Full Cascade

**FIB** Focused Ion Beam

**F-DSC** Flash Differential Scanning Calorimetry

**Lu-Al** Lutetium Aluminium

**KERMA** Kinetic Energy Released in Materials

**IAEA** International Atomic Energy Agency

**MCNP** Monte Carlo N-Particle Code

**NRT** Norgett, Robinson and Torrens

**PKA** Primary Knock-On Atom

**QC** Quick Calculation

**SEM** Scanning Electron Microscopy

**HRSEM** High Resolution Scanning Electron Microscopy

**SRIM** Stopping and Range of Ions in Matter

**TEM** Transmission Electron Microscopy

**STEM** Scanning Transmission Electron Microscopy

**HRTEM** High Resolution Transmission Electron Microscopy

**TRIM** Transport and Range of Ions in Matter

**XRD** X-Ray Diffraction

**VOV** Variance of Variance

# List of Symbols

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|             |                                        |
|-------------|----------------------------------------|
| $\eta$      | Efficiency correction factor           |
| $\phi$      | Particle Flux (neutron or Ion)         |
| $\Phi$      | Particle Fluence (neutron or Ion)      |
| $K_{eff}$   | Neutron multiplicity factor            |
| $\bar{\nu}$ | Average number of neutrons per fission |
| $\rho$      | Density of material                    |
| $\rho_m$    | Atomic number density of material      |

# Chapter 1

## Introduction

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The general understanding of how damage in materials affects their efficiency during operation was largely skewed to focus on the mechanical effect on the materials. Knowledge of defects in materials was limited to overload, creep rapture, fatigue, brittle fracture, wastage, and environment-enhanced factors like corrosion, rusting, erosion, pitting, fretting, and dezincification [1, 2]. However, studies on material damage later revealed that radiation plays a significant role in inducing defects in materials exposed to harsh radiation environments, such as nuclear reactors, accelerators, and space [3–7]. In addition, smaller devices used in physics, chemistry, and radiotherapeutic processes involve the utilization of energetic particles and photons [8]. In these radiation environments, materials undergo exposure that leads to effects such as dislocations, swelling, bubbles, void nucleation, helium embrittlement, radiation-induced segregation, activation, and dissolution [1, 2].

Defects in materials originate at the atomistic level (microstructure) and accumulate over time, resulting in physical damage to the bulk morphology of materials [9, 10]. Therefore, studying the effects of radiation damage in a diverse range of materials is crucial for predicting their behaviour when exposed to energetic particles and photons. To quantify the level of radiation damage in materials, various computational and experimental methods are available to predict and characterize the damage in a material after exposure to radiation of different wavelengths [10]. Computational methods typically function as a means of introspection into real experiments and can also serve as validation tools for the results obtained in a study.

Gold (Au) and Lutetium-Aluminium (Lu-Al) alloys possess distinct properties that make them useful benchmarks for understanding radiation damage behaviour in

different materials under irradiation. Gold is a face-centered cubic (FCC) crystal structure, which has been well-studied, making it a good reference material for studies of radiation damage mechanisms in materials. Its high atomic number ( $Z$ ) and chemical inertness [11] give it resistance to oxidation and corrosion. This ensures that observed changes under irradiation are primarily due to radiation effects rather than chemical reactions. Extensive research on gold provides a baseline for radiation damage studies. One of its specific applications is in reactor monitoring and safety. Gold's high capture cross-section for thermal neutron makes it a good candidate as a neutron flux monitor [12–18].

On the other hand, Lutetium-Aluminium alloys have complex intermetallic phases that can provide insights into radiation effects in more intricate materials. Specifically, aluminium 6000 and 7000 series alloys are considered excellent materials for reactor and aerospace-based designs due to their strength and corrosion resistance ability. [19–22]. Like the Al-6000 and 7000 series alloys, incorporating rare earth elements including lutetium into aluminium could lead to unique properties that could improve high-temperature stability, corrosion resistance, radiation resistance and enhance mechanical properties such as increased strength and hardness. Lu-Al alloys could be suitable for specific applications such as cladding materials, fuel assemblies, and structural components in reactors, highlighting the need to assess the radiation damage tolerance of the material. Furthermore, Lu-Al can serve as a flux monitor. Including lutetium in the alloy with aluminum can help stabilize the material while providing additional mechanical and thermal properties beneficial for certain monitoring applications.

Studying radiation damage in Lutetium-Aluminium alloys and Gold has far-reaching impacts, aiming to enhance material performance and safety within nuclear reactors. By comprehensively understanding these materials' radiation tolerance levels and damage mechanisms, this study will contribute to the development of advanced reactor components with superior durability and stability under extreme conditions. This will lead to advanced reactor design for increased efficiency and extended lifespan of critical components at reduced cost due to maintenance. Moreover, these

insights from this study can drive innovations in material science, promoting the creation of new alloys and composites tailored specifically for high-radiation environments. The findings of this study not only support the advancement of nuclear technology but also have broader implications for fields such as space exploration, where materials are subjected to intense radiation. The research also underscores the importance of material innovation in achieving sustainable, safe and clean energy (carbon-free) solutions with nuclear energy as captured in the Sustainable Development Goals (SDG) 7 [23, 24].

This thesis is centered on employing computational algorithms and experimental characterization of materials to assess the extent to which Lutetium-Aluminium (Lu-Al) and gold (Au) demonstrate resilience to damage after radiation exposure. The subsequent sections provide background information on the concept of radiation damage, as well as the objectives and hypotheses of this study.

## 1.1 Background on Radiation Damage

When energetic particles such as neutrons, photons, protons, and other charged particles interact with crystalline matter, one potential outcome is the creation of lattice structural defects. This occurs due to the transfer of energy to atoms within the crystal [25]. Consequences of these interactions may include the generation of phonons, excitons, plasmons, and secondary particles such as electrons and photons and material heating with non-damage-producing effects that could be observed. Atoms knocked out of their lattice site hit other atoms and distort the original structure. The accumulated effect of this energy transfer results in radiation damage in materials [8].

Radiation damage can be a permanent displacement within the atomic structure in solids, brought about by these energetic particles. As radiation interacts with solids, a progression of atomic displacements may emerge, producing damage in materials that affects the properties of the material. For instance, vacancies and interstitials (Frenkel pair) shown in Figure 1.1 [26] are created in the materials by

energetic particles, that may prompt swelling and embrittlement [4, 5].

Exposure of metallic materials to radiation, especially neutrons or protons, can cause critical changes in their mechanical and physical properties over a long period (from nanoseconds to years). These changes may include an increase in the yield stress, a partial loss in ductility, and alterations in the thermal and electrical conductivity of the materials. Materials can also exhibit transitions from ductile to brittle fracture behavior when temperature reduces below the Transition Temperature (TT) of materials. [9, 27].

To fully comprehend the dynamics of radiation damage in materials, it is necessary to understand the impact of the damage that occurs from a single atom of a material (femto scale) to a macroscopic scale (Engineering Scale). Particles (neutrons/protons) with sufficient kinetic energy above the atomic displacement energy of a material, can knock out atoms from their initial lattice site and transfer kinetic energy to other atoms, creating vacancies and interstitials. This results in a build-up of more atomic displacements which are transported, ballistically by more radiation. Forming clusters and leaving an empty site (vacancy or hole) behind, while creating an interstitial site in front[28] as shown in Figure 1.1.

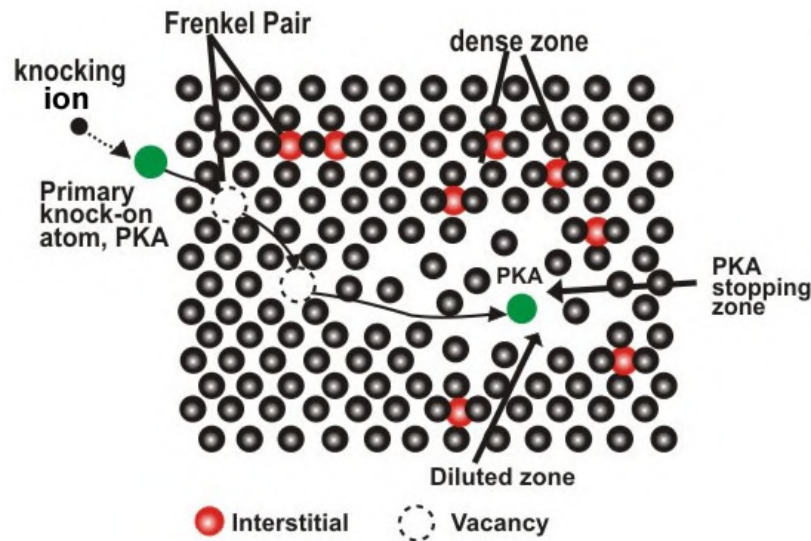


**Figure 1.1:** Lattice displacement due to collision a Primary Knock-on Atom(PKA) with atoms of the material impacted by a high energy, resulting in the formation of Frenkel pair, dense and diluted zones.( [26])

The accumulation of these displacements in materials over a protracted time gives rise to defects in materials such as dislocations, void swelling, void nucleation, embrittlement, and loss of ductility[2]. However, it is crucial to understand that the defects produced in crystals can also be much more complex: this could be in the form of defect clusters, amorphization, dislocation loops, or can even take the form of three-dimensional defects[2, 29, 30].

The radiation damage caused by neutrons/protons over time determines a material's macroscopic response to radiation and is hence vital to comprehend. Radiation damage in materials in most cases is usually categorized into two phases: first, the short-term or *Primary damage* immediately after (within the femto-pico second's range) the particle's interaction, using collision processes, that occur at an atomic level. This is far from thermodynamic equilibrium. Whereas, the *long-term* evolution of damage caused by thermally activated processes in nanoseconds to years is the second phase [8].

Understanding the impact of radiation on matter is important not just for material design, which has improved radiation damage resilience, but also in the production of novel devices by defect engineering[31]. There are two basic reasons to investigate damage occurring in nuclear materials. The first is the technological demand for the integrity of the structural materials in the reactors after exposure to radiation over several years. Secondly, there is a challenge to scientifically develop novel approaches in multiscale modeling of radiation damage effects on materials which includes going through microstructural properties to the mechanical behaviour of materials after irradiations with particles or photons[2, 8]. The basic mechanisms of radiation damage and the ensuing consequences on the properties of materials and their performance, attracted many studies of applications in the fission/fusion reactor, other harsh radiation environments, and extra-terrestrial applications[32].

A detailed study of radiation damage on a material may include a theoretical prediction via simulation methods such as Monte Carlo (MC) or atomistic modeling,

especially where there is insufficient quantitative data on the material of interest. Studies can also involve experiments, to characterize radiation defects in the material's microstructure. This usually involves:

- A pre-irradiation characterization of the sample's microstructure,
- Sample irradiation with a radiation field source and
- A post-irradiation characterization of the irradiated sample's microstructure.

Radiation sources used in this kind of study may be produced from nuclear fission/fusion reactors or particle accelerators where different types of radiation exposure are done, using particles like electrons, protons, other heavy ions, and neutrons. This irradiation is performed to induce damage in the sample under investigation to study the defects on the microstructures.

## 1.2 Neutron and Proton irradiation

Since scientists discovered that an atom can be probed, many attempts were made to measure, qualitatively and quantitatively, the properties of the atom. Analytical science has developed methodologies that enable the probing of the materials to observe the properties and structure at the single atom level. These procedures are critical in fundamental research and development and also in industrial applications since they enable the understanding of matter.

The discovery of neutrons by J. Chadwick in 1932 [33] and experimental results of F. Joliot and I. Curie in 1934 [34, 35], followed by G. Hevesy and H. Levi in 1936 [36, 37] has led to the exploration of the unique properties of the neutron in several applications including the irradiation of materials. Neutron irradiation can be done in a nuclear reactor core at defined fluences to induce radiation damage in materials and after that study the microstructural properties of the materials relating to damage. The properties of neutrons from reactor beam lines can be used to calculate reactor power levels from the velocity of neutrons emanating from the beamline, investigate how materials absorb neutrons, and determine the shape of molecules upon collision of neutrons with the materials as well as shielding effects

[38].

Irradiation of materials with neutrons in fission reactors triggers elastic and inelastic reactions, displacing the Primary Knock-on Atoms (PKA) from their lattice site, which on further collision with neighbouring atoms, creates interstitial-vacancy pairs called Frenkel Pairs (FP) [39, 40]. Neutron irradiation also induces the production of photons and heat deposition by neutrons and gammas, all of which induce radiation defects on the microstructure of materials. The evolution and diffusion of Frenkel pairs, forming higher defects in the microstructure over time translate into significant observable defects in the bulk morphology of structural materials [9].

In as much as neutrons are good radiation species for radiation damage studies, there are several challenges in usage for material damage studies. Sample irradiation with neutrons entails the use of special irradiation facilities like fission reactors and cyclotrons which are not usually easy to access in this part of the world. Doses of radiation that can be achieved within a few hours with proton irradiation could take days to achieve the same with neutron irradiation in the reactor. One important factor that may limit radiation damage studies with neutron irradiation is the level of activation of some samples post-irradiation which requires long cooling times before samples can be handled [41]. Whereas, with proton irradiation, activation of materials and radiation cooling times post-irradiation is of less concern since irradiated samples are less hazardous to handling immediately after irradiation. In addition, proton irradiation, or generally ion-beam irradiation is relatively cost-effective. [41–43]. Work with ion beams allows for the systematic control of specific irradiation conditions. This has made proton irradiation damage studies more dynamic than the use of neutrons [44].

Neutrons and protons have similar irradiation profile characteristics which is an important feature for comparing both radiations; neutrons have a neutral charge state and are highly penetrative when irradiated to solid materials, resulting in a spatially flat damage profile in the material to depths of about several millimeters [41, 45]. Protons, which are charged particles, produce a flat damage profile that

leads to a dose variation of several tens of micrometers within the energy range of 1-10 MeV. They then deposit all their energy continuously along their path through ionization, leading to a Bragg peak near the end of their range.

Many studies have shown that protons and neutrons produce similar microstructural features related to damage [41, 42, 44, 46–48]. These features include the formation of dislocation loops, precipitates, voids/bubbles, radiation-induced segregation, stacking faults, and amorphization. However, there are challenges even with ion irradiation which makes it difficult to achieve all the microstructural features that are observed with reactor (neutron) irradiation. One of these type of transmutation reactions that occur with neutrons leads to the production of gases such as hydrogen and helium, which contribute to the effects that can only be observed in neutrons. These gases exacerbate damage by promoting void swelling and blistering, especially in aluminum and its alloys [49]. This effect is sometimes simulated in proton irradiation with co-implantation or pre-implantation of hydrogen or helium and can provide valuable insights into the synergistic effects of displacement damage and gas accumulation, to emulate the effects observed during neutron transmutation of gases [50–53].

There is also a disparity in the damage rates observed between neutron and proton irradiation. Neutron irradiated dose rates for most materials are typically of the range  $10^{-8}$  to  $10^{-6}$  dpa.s<sup>-1</sup> even for fast energy regimes. Whereas, ion irradiation ranges are about  $10^{-4}$  to  $10^{-3}$  dpa.s<sup>-1</sup> leading to a compressed irradiation time [24, 54–56]. This implies that with proton irradiation, dpa rates (dose rate) up to  $10^4$  times that of neutron irradiation can be achieved, this means about 200 dpa can be achieved in days instead of years [42].

Taller et al. [24] emphasized that ion irradiation’s ability to significantly reduce irradiation time by up to three orders of magnitude makes it a viable option for studying radiation damage in materials. Their experiment demonstrated this, as shown in Figure 1.2 where similar microstructural features were obtained within 6.7 hours of dual ion irradiation as compared to 5900 hours of neutron irradiation at

approximately the same doses.

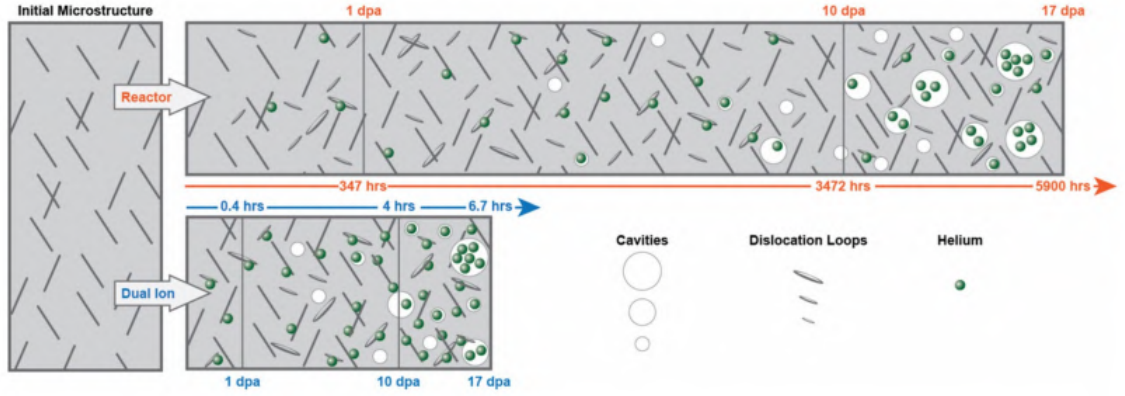


**Figure 1.2:** Microstructural features showing damage rate dependence of helium trapping and release due to neutron and ion irradiation conditions (dual ion produces higher damage rates compared to neutron irradiation)( [24])

A summary of key features between ion irradiation and neutron (reactor) irradiation shown in Table 1.1 indicates the viability of proton irradiation as a surrogate for neutron (reactor) irradiation.

**Table 1.1:** Comparison of key irradiation features with ion beam and test reactors [42]

| Variable                                      | Ion beams                                     | Test reactors                                  |
|-----------------------------------------------|-----------------------------------------------|------------------------------------------------|
| Dose                                          | > 500 dpa                                     | 10-20 dpa max                                  |
| Dose rate                                     | 100 - 10000 $\times$ reactor rates            | Few $\times$ reactor rates                     |
| Energy                                        | Depends on ion type<br>(a few keV to 100 MeV) | Neutron spectrum<br>(up to 14 MeV)             |
| Transmutants/fission products                 | Separable                                     | Controlled by nuclear physics                  |
| Temperature                                   | Better than $\pm 10^\circ\text{C}$            | Variable 10 s of $^\circ\text{C}$              |
| Residual activity                             | Low to none                                   | High                                           |
| In situ observation                           | TEM, RS, GC,etc.                              | Some T and displacement,<br>generally PIE only |
| Unit mechanisms                               | Demonstrated                                  | Challenging                                    |
| Cost                                          | Relatively low                                | Relatively high                                |
| Simultaneity<br>(e.g corrosion and straining) | Corrosion, SCC,<br>creep, diffusion, etc.     | Bulk materials,<br>doable but difficult        |
| Sample thickness                              | A few nm to 100 $\mu\text{m}$                 | Bulk                                           |

To correlate radiation damage, it is essential to have a quantity that can be used as a standard. The main parametric quantity that quantifies the radiation dose on a material is the Displacement Per Atom (dpa) [6]. The dpa offers the basis for comparing

radiation effects on crystalline materials. It is also used to quantitatively compare damage for different materials after irradiation. The dpa is also one of the important parameters because the irradiation-induced changes in the microstructure are a function of the dpa [6, 57]. Other parametric quantities that influence radiation-induced microstructural changes include dislocation density, swelling, radiation-induced segregation (RIS), helium and hydrogen content, creep rate, mechanical properties (hardness, tensile strength, residual stress), electrical conductivity, thermal conductivity, phase stability, and precipitation. [1, 6, 9, 10]. These parametric quantities provide an in-depth understanding of radiation-induced microstructural changes in materials, enabling the assessment and improvement of material performance in radiation environments

In this study, the radiation damage on Lu-Al and Au microstructures is investigated. Prediction of the damage with relevant exposure quantities, especially the dpa and other quantities such as heat deposition and fluence (dose) were examined and simulated, using Monte Carlo N-particle transport code (MCNP6.2) [58] for neutrons and the Stopping and Range of Ions in Materials (SRIM-2013) [59] for protons ion beams. The results of the predicted damage with MCNP6.2 and SRIM-2013 were compared for two purposes. Firstly, to establish the possibility for radiation damage studies in the materials under investigation with proton irradiation as a surrogate for neutron irradiation as reported in the literature, and secondly to guide the study on the level of doses or dpa required for proton ion beam irradiations. The calculated dpa serves as a dose benchmark for different damage features observed on the microstructures of Lu-Al and Au. The damage characterization methods used in the current study included Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), X-Ray Diffraction (XRD), and Flash Differential Scanning Calorimetry (F-DSC).

### 1.3 Research Questions and Hypothesis

Structural deformation of materials starts from a single point defect at the microstructure level, which can occur when a material is exposed to radiation. While

there have been studies on the formation of point defects, their nucleation, and the growth of cluster defects for Frenkel pairs in Au after irradiation with self-ions or fast neutron energies [13, 15, 16, 18, 46], no detailed work has been reported on the displacement per atom rate in Lu-Al to compare the actual damage effects characterized. The materials Lu-Al and Au are used for reactor flux monitoring and have other applications. Lu-Al is a potential alloy for reactor cladding, fuel assemblies, and structural materials [19–21]. It could also qualify for structural materials in space missions and microgravity experiments to aid the understanding of Aluminium alloy material properties such as solidification, phase stability, and microstructure evolution without the influence of gravitational forces [60, 61]. Au’s unique properties such as high reflectivity, excellent electrical conductivity, and corrosion resistance make it a choice material in microelectronic devices like detector systems. Its application spans from thermal control, design of optical instruments, electrical connectors, and coating in space technology [12, 32, 62]. Therefore, evaluating the radiation damage on these materials will enable the determination of the level of radiation resilience and the performance efficiency of these materials in the relevant radiation environment.

Research on the effect of damage in metals, alloys, and semiconductor materials has been conducted for many decades [6, 8, 10, 14, 42, 45, 46, 63–65], driven by the need to control the radiation-induced degradation of materials in reactors and the material’s response to harsh radiation environments. Such research has been beneficial for nuclear material development and other applications, particularly in assessing the capabilities of materials that can be considered for different purposes in reactor engineering or other high radiation environments, based on radiation damage resilience. This study of radiation damage on Lu-Al and Au is expected to contribute to nuclear material development capabilities and other applications. To achieve this, the section below considered the following research questions.

### 1.3.1 Research Questions

This study addresses the questions below.

- What is the level of damage predicted with MCNP6.2 for neutrons and SRIM-

2013 for protons on the materials under study?

- What are the observed microstructural defects, their sizes, and densities with the irradiation doses on the samples of Lu-Al and Au post-irradiation?

### **1.3.2 Hypothesis**

The study was guided by the following hypothesis:

- I hypothesize that through the careful application of proton irradiation, the quantities of multiple types of defects, including voids, stacking faults, dislocations, and other defect clusters can match those of neutron irradiation in Lu-Al and Au.

## **1.4 Aim and Objectives of the Study**

The study aims to evaluate the radiation damage on the Lu-Al and Au with the following objectives:

- Simulate the neutron flux, Displacement-Per-Atom (dpa), fluence, and heat deposition in Lu-Al and Au, using MCNP6.2.
- Simulate the Displacement-Per-Atom (dpa) for light and heavy ions in SRIM-2013 to enable comparison with neutron irradiation.
- Irradiate the samples of Lu-Al and Au with protons.
- perform SEM, XRD, TEM and F-DSC studies to characterize the microstructure of the samples pre and post-irradiation,
- Compare the observed radiation damage defects at different doses on the micro-structure of the materials post-irradiation.
- Compare the observed damage at different doses resulting from proton irradiation to that of neutron irradiation where applicable.

## 1.5 Thesis Outline

This thesis is organized in the following structure:

Chapter 2 discusses interactions of neutrons, photons, and charged particles with materials, emphasizing the implications of each interaction on radiation damage to crystalline materials. This chapter also provides an overview of radiation damage in materials and briefly reviews the effects of radiation damage on crystalline solids.

Chapter 3 provides an in-depth description of the Monte Carlo Simulation methods of radiation damage used to predict radiation damage to Lu-Al and Au, the experimental evaluation of proton radiation damage using CLASS accelerator irradiation facility at Massachusetts Institute of Technology (MIT). The chapter also outlined the methods for pre- and post-irradiation characterization used to observe the microstructure of the materials.

Chapter 4 reports the results of MC calculations with MCNP6.2 and SRIM-2013. This chapter also reports the results of damage characterization before and after proton irradiation of Lu-Al and Au.

Chapter 5 discusses the results of radiation damage predictions with MCNP6.2 and SRIM-2013, and the observed microstructural defects on the materials studied with a clear summary of the findings.

Chapter 6 draws valid conclusions based on the results obtained. It also recommended areas for further study of the materials under investigation. .

# Chapter 2

## Literature review and Theoretical considerations

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### 2.1 Introduction

Studying radiation damage on the microstructure of crystalline materials necessitates a comprehensive understanding of the damage mechanisms induced by various radiation species, namely neutrons, photons, and charged ions (both light and heavy). The interactions of these radiation species with materials can vary, and they result in displacements of atoms with remarkably different energies for the initial displaced atom, commonly referred to as the Primary Knock-On Atom (PKA) as well as the distinct primary recoil spectra. Therefore, a knowledge of how these energetic particles induce defects that lead to the degradation in the microstructure of materials becomes crucial. This chapter provides a review of the mechanism involved in the creation of radiation damage by neutrons, photons, protons, and other charged particles. Additionally, it delves into the concept of radiation damage, the effects of radiation on crystalline solids, and the methods used to evaluate the level of damage.

### 2.2 Properties and production of neutrons

Understanding the dynamics of radiation interaction with matter involves a cascade of events, from the particle's interaction with materials to the effects produced on the atom and the subsequent radiation damage on materials. Neutrons exhibit the most complex and elaborate radiation damage mechanism. In this section, we discuss the interaction of neutrons with matter and the resulting radiation damage

consequences on materials.

Neutrons are baryons, electrically neutral particles with a mass of  $1.674927 \times 10^{-27}$  kg or 1.008665 U, They possess a spin of 1/2 and parity +1 [66]. Neutrons are generated in various ways, ranging from small-size radioisotopes undergoing spontaneous fission and sealed tube neutron generators, to medium-size devices such as plasma focus and plasma pinch devices, light ion accelerators, and energetic bremsstrahlung photo-fission processes, but are primarily produced by large devices like nuclear fission reactors, nuclear fusion reactors, and cyclotrons [67].

Neutrons, being neutral particles, have an atomic mass higher than that of a proton. Free neutrons are typically unstable and undergo decay with an average half-life of about 636 seconds, transforming into protons through the emission of  $\beta^-$ .

$$n \rightarrow p + \beta^- + \nu_e + 0.782 \text{ MeV} \quad (2.1)$$

### 2.2.1 Classification of Neutrons

Neutron classification is arbitrarily based on their kinetic energies or speed as shown in Table 2.1.

**Table 2.1:** Classification of Neutrons by Energies [68]

| Category                            | Energy           |
|-------------------------------------|------------------|
| Cold neutrons                       | <0.0025 eV       |
| Thermal neutrons                    | 0.0025 - 0.1 eV  |
| Epithermal neutrons                 | 0.3 - 100 eV     |
| Resonance neutrons                  | 1.0 eV - 300 eV  |
| Intermediate neutrons               | 100 ev - 200 keV |
| Fast neutrons                       | 200 keV - 20 MeV |
| High Energy (Relativistic) neutrons | > 20 MeV         |

However, in neutron irradiation applications, the neutron energies are classified into thermal, epithermal, and fast neutron fluxes for convenience of operation. This is shown in Figure 2.1 below.

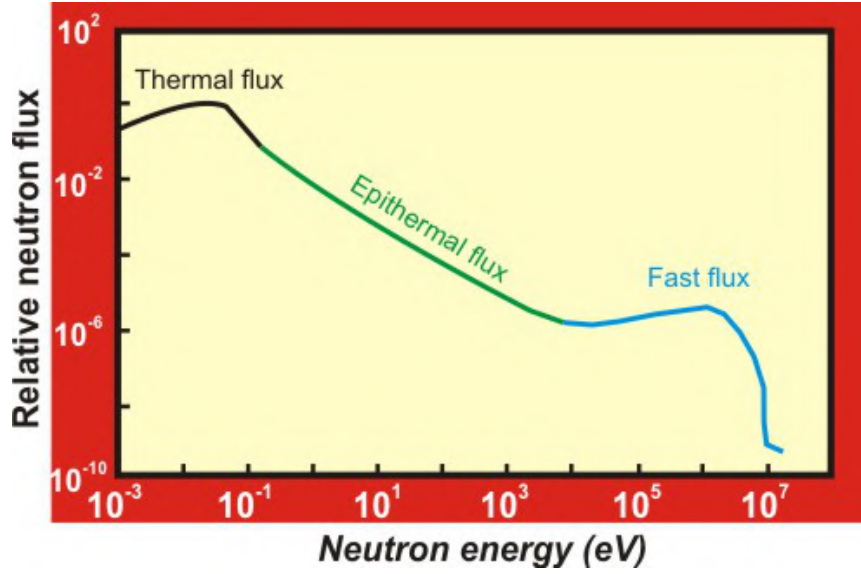


**Figure 2.1:** Relative neutron fluxes and neutron energy (eV)(taken from Ref. [69])

Neutron energies can also be expressed in form of velocity of movement ( $\text{m.s}^{-1}$ ). Additionally, neutron energy ( $E$ ) and velocity ( $v$ ) can also be expressed in terms of the wavelength( $\lambda$ ) since particles in motion also possess wave properties[70]. The  $\lambda$  of the neutron becomes an important parameter in neutron diffraction applications. The  $\lambda$ ,  $v$ , and  $E$  of the neutron are related. Knowledge of any pair, either  $E$  or  $v$ , can be used to estimate the  $\lambda$ . The velocity of neutrons is given by

$$v = \sqrt{\frac{2E}{m}}, \quad (2.2)$$

where  $m$  is the mass, and  $E$  is the kinetic energy of the neutron. Consequently, the  $\lambda$  can be obtained from the particle's mass and velocity according to the de-Broglie equation thus:

$$\lambda = \frac{h}{p} = \frac{h}{mv}, \quad (2.3)$$

where,  $h$  is the Planck's constant ( $6.63 \times 10^{-34} \text{ Js}$ ) and  $p$ , the particle's momentum.

## 2.2.2 Neutron flux and fluence

### (a) Neutron flux

The strength of a radiation field is commonly denoted as the flux. In the case of isotropic radiation fields, as seen in nuclear fission reactors, neutron radiation

interacts with materials uniformly. Consequently, the assessment of field strength is described in terms of flux density. This is in contrast to radiation fields where parallel beams of radiation interact with materials, making it easier to utilize flux intensity [71, 72]. Assuming the density of the neutron radiation at position  $\vec{r}$  to be  $n(\vec{r})$ . If the average speed of the particle incident on a material is  $v$ , then the neutron flux density or simply the flux is defined as follows:

$$\phi = vn(\vec{r}) \quad (2.4)$$

The flux density becomes the total neutron track length per unit volume per unit time,  $\phi = vn$  in  $cm^{-2}s^{-1}$ . In radiation fields such as the fission reactor, where neutrons are born with a distribution from lower to higher energies (white spectrum), the neutron density defined in space ( $\vec{r}$ ) is a function of energy  $E$  and time  $t$ ,  $n(\vec{r}, E, t)$ . Hence the neutron flux can be re-written as:

$$\phi(\vec{r}, E, t) = v(E)n(\vec{r}, E, t) \quad (2.5)$$

The units of neutron flux ( $n.cm^{-2}.s^{-1}$ ) are similar to flux intensity. However, in the case of neutron density, we look at the collision density (isotropically) in the case of neutrons where the material is exposed uniformly.

### (b) Neutron fluence (radiation dose)

The flux density provides a way of estimating the reaction rate density, which is a product of the total path length traveled by a neutron,  $\phi(\vec{r}, E, t)dE$ , and the average distance traveled by a neutron before an interaction  $\Sigma_i(\vec{r}, E, t)$ . The interaction rate density is given by:

$$\hat{R}(\vec{r}, E, t)dE = \Sigma_i(\vec{r}, E, t)\phi(\vec{r}, E, t)dE, \quad (2.6)$$

where  $\hat{R}$  is the interaction rate density, which gives good information on the calculation of the radiation effects [72]. The number of total fission occurring in volume

V, between times  $t_a$  and  $t_b$  for all neutron energies is given by:

$$N_f(t_a, t_b) = \int_{t_a}^{t_b} dt \iiint_V dV \int_0^{E_{max}} \Sigma_f(\vec{r}, E, t) \phi(\vec{r}, E, t) dE \quad (2.7)$$

Therefore, the radiation fluence can then be estimated, which is one of the implications of the radiation rate density. The dose of neutron radiation field deposits causes displacement damage. The neutron fluence between time  $t_a$  and  $t_b$  for all neutron energies is the time-integrated flux density, reflecting the reaction interaction rate of a number of fission between  $t_a$  and  $t_b$  gives.

$$N_f(t_a, t_b) = \int_{t_a}^{t_b} dt \iiint_V dV \int_0^{E_{max}} \Sigma_f(\vec{r}, E, t) \Phi(\vec{r}, E, t) dE \quad (2.8)$$

and the fluence is given by

$$\Phi(\vec{r}, E) = \int_{t_a}^{t_b} dt \Phi(\vec{r}, E, t) \quad (2.9)$$

One can easily connect the flux density or flux and fluence as the time rate of change of fluence. Calculation of neutron radiation damage effects is expressed in the unit of fluence (dose).

## 2.3 Interaction of Neutrons with Matter

Neutrons are active particles produced in nuclear fission reactors, moving in straight lines with an average mean free path and interacting with various reactor components and other materials. This section discusses neutron interaction, emphasizing the reactor setting and highlighting neutron transport. The distribution of neutrons to different reactor parts and their subsequent interactions is referred to as neutron transport. In neutron transport, the analysis of neutron interactions is conducted based on the applications of reactor technology. Some essential applications include:

- Reactor design: This requires an understanding of the distribution of neutrons and photons in the reactor and an idea of the power distribution which is deduced from the neutron population in a fission process.
- Radiation transport analysis: This entails an in-depth analysis of the various

shielding for reactor plants and components. An estimation of the heat deposition due to neutrons and photons locally and globally in the reactor and damage incurred by fuels and other components. Also, the radiation damage analysis on the Reactor Pressure Vessels (RPV) and other components, determines the reactor's life.

Neutron interactions include elastic scattering ( $n,n$ ), inelastic scattering ( $n,n'$ ), radiative capture ( $n,\gamma$ ), charged particle interactions ( $(n,p)$ ,  $(n,\alpha)$ ), neutron-producing reactions ( $n,2n$ ), ( $n,3n$ ) and fission interactions ( $n,f$ ) [71].

The reactor components and other materials placed within the reactor undergo these interactions, determined by the probability of occurrence of reactions. This section is focused on the interaction modes, specifically examining the impact of radiation damage on crystalline solids. The irradiation of crystalline materials by fast neutrons leads to the reconfiguration of the crystal lattice. This process is accompanied by the formation of lattice defects that gradually influence both the micro and macrostructure of the irradiated material over time[73].

The possible neutron interaction mechanisms with matter depend on the neutron energy, absorber material, and the interaction cross-section. The neutron interaction cross-section denotes the probability of a reaction occurring. When examining radiation damage to materials, the mechanisms recognized for displacing atoms from their lattice positions under neutron irradiation involve interactions that result in scattering and neutron capture [10].

Elastic and inelastic scattering or collision represent the initial interactions between neutrons and target nuclei. These interactions lead to subsequent displacements before the neutrons come to rest within the lattice, leaving the target nucleus unchanged in the process. Other forms of interaction include neutron capture, activation, and fission interactions.

### 2.3.1 Elastic neutron scattering (n,n)

Elastic scattering is a neutron interaction with matter where kinetic energy lost during a nuclear collision by a neutron is passed to the nucleus at ground state [71, 74]. This kind of collision can be thought of as that of billiard balls colliding with one another, where part of the kinetic energy of the particle is transferred to another without loss of energy. Hence, there is a conservation of kinetic energy in the process. When such interaction occurs with neutrons of light mass approximately the mass of the neutron, hydrogen, and other lighter atomic nuclei, the neutron energy is reduced more effectively. This mechanism is most probable for neutrons of energies 10 MeV or lower [75].

Elastic scattering sometimes takes place through the formation of a compound nucleus by the emission of a neutron that de-excites the compound nucleus to the ground state of the original nucleus[76]. When this type of resonance elastic scattering occurs, it leads to the conservation of the nuclear system of the original neutron. There could be an interaction of the neutron and the nucleus without the neutron being absorbed whereas there is the formation of a compound nucleus, which is termed potential scattering. Schematically the elastic scattering kinematics can be represented by Figure 2.2,

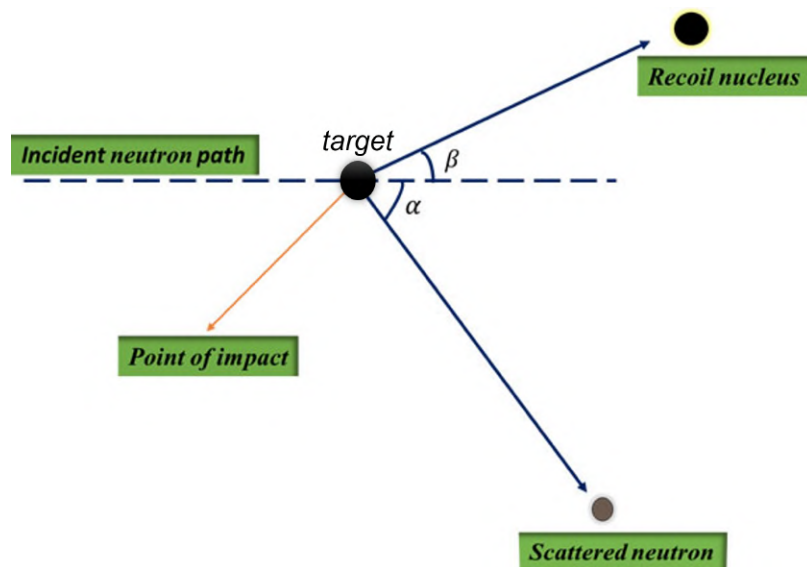


**Figure 2.2:** Neutron Elastic scattering by the impact of the neutron with the target nucleus of an atom. The neutron at an angel  $\alpha$ , while the target nucleus scatters at an angel  $\beta$

where the dashed line indicates the path of the incident neutron with a specified kinetic energy impacting a target nucleus. The nucleus undergoes a recoil at an angle  $\beta$ , while the neutron is scattered at an angle  $\alpha$  to the path of the incident neutron[70]. The kinetic energy lost ( $E_k$ ) by the neutron during collision is given by

$$E_k = \frac{4Mm_n}{(M + m_n)^2} \cos^2 \beta \quad (2.10)$$

where  $M$  is the mass of the target nucleus,  $m_n$  represents the neutron mass, and  $\beta$  the recoil angle of the nucleus. The elastic scattering mechanism is common with fast neutrons as they interact with atoms of lower elements such as hydrogen ( $^1H$ ) in light water, deuterium ( $^2H$ ), and graphite ( $^{12}C$ ) [10]. This is key in slowing down fast neutrons, and this explains the choice of light water or heavy water as moderators in the fission nuclear reactor.

The elastic interaction of neutrons ( $m_n = 1.008665$  u) with target atoms of a material can lead to the displacement of atoms from their lattice sites, the creation of defects such as vacancies and interstitials, and ultimately structural changes with the material. These structural changes contribute to radiation damage. The fractional energy lost by atoms due to neutron radiation is related to the amount of kinetic energy transferred from the neutrons to the atoms during interactions [10, 71]. The fractional energy lost ( $E_k$ ) for some nuclides is shown in Table 2.2

**Table 2.2:** Maximum fraction of kinetic energy ( $E_k$ ) a neutron can lose on impact with the nucleus of different atoms in increasing atomic mass units (u) [10]

| Nuclide    | Nuclide mass ( $M$ ) | $E_k$ |
|------------|----------------------|-------|
| $^1H$      | 1.007825             | 0.999 |
| $^2H$      | 2.014102             | 0.889 |
| $^9Be$     | 9.012182             | 0.362 |
| $^{12}C$   | 12.000000            | 0.286 |
| $^{16}O$   | 15.994915            | 0.223 |
| $^{27}Al$  | 26.98153             | 0.139 |
| $^{28}Si$  | 27.976927            | 0.134 |
| $^{55}Mn$  | 54.938047            | 0.071 |
| $^{176}Lu$ | 175.942686           | 0.023 |
| $^{197}Au$ | 196.96654            | 0.020 |

The  $E_k$  of nuclides reduces as the atomic mass of the element increases, this can be observed in Table 2.2 with the variation from  $^1H$  to  $^{179}Au$ . Higher energy transfer typically leads to more significant radiation damage. Also, the distribution of energy deposition within the material can influence the extent and nature of damage.

### 2.3.2 Neutron inelastic scattering (n,n')

Neutron Inelastic Scattering is an interaction that allows for a loss in part of the kinetic energy of the neutron in a nuclear collision. A fast neutron collision with a nucleus may occur whereby the neutron is scattered in another direction with part of its energy lost to the recoil nucleus. This lost energy enables the excitation of the nucleus, the resulting excited state can be termed a meta-stable state. The excited nucleus seeks stability by emitting characteristic gamma rays during de-excitation or maintaining the meta-stable state for some time.

In this interaction mechanism, momentum conservation is impossible between the scattered neutron and the recoil nucleus[70, 77]. Energy transfer by means of inelastic scattering can occur in the  $(n, 2n)$  and  $(n, \gamma)$  reactions. This type of scattering is possible only for neutron energies exceeding the particular threshold for an element or material. Inelastic scattering is prevalent with fast neutron impact with medium to heavy nuclei. The important role of this mechanism is that it allows the slowing down of fast and intermediate neutrons to thermal neutron energies by heavy nuclei like  $^{238}U$ . This mechanism is significant for heterogeneous reactors with highly enriched fuel.

Generally, neutron scattering, both elastic and inelastic, significantly contributes to multiple atomic displacements in materials interacting with energetic neutrons above the atomic displacement energies. Given that these mechanisms entail direct interaction with the nucleus of the atom, neutrons induce more damage to crystalline materials. Moreover, these mechanisms play a role in ionization and heat deposition caused by neutrons [78].

### 2.3.3 Neutron capture and activation

Neutron capture interaction is another mechanism of atomic displacement based on energy conservation. It is a nuclear reaction in which the target nucleus absorbs the neutron, leading to the production of an isotope in an excited state. In this reaction, the atomic nucleus collides with one or more neutrons and combines to form a heavier nucleus. There is usually the de-excitation of the activated isotope with the release of  $\gamma$ -rays as illustrated in Figure 2.3. Neutron capture is more probable at low thermal energies  $< 0.4$  eV. This type of reaction is called radiative capture ( $n, \gamma$ ), which is an exothermic reaction that contributes to atomic displacement, especially for materials that have a large thermal cross-section for this reaction.

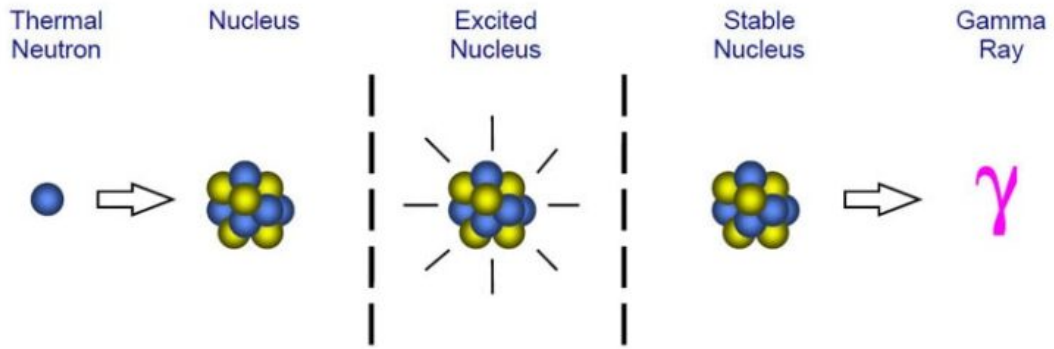


**Figure 2.3:** Neutron absorption by the nucleus producing an excited nucleus which emits a  $\gamma$  to become stable ( taken from Ref.[79])

Neutrons lack electric charge, making it easier for them to penetrate the atomic nucleus compared to protons which are electrostatically repelled due to their positive charge. The ability of a nucleus to capture a neutron depends on the type of nuclei and neutron energy.

Nuclear reactions resulting from neutron capture can emit particles such as  $^1H$  ( $n, p$ ),  $^2H$  ( $n, d$ ),  $\alpha$ -particles ( $n, \alpha$  and also neutrons ( $n, 2n$ )). Neutron-producing reactions ( $n, 2n$  and  $n, 3n$ ) are also important in radiation effects on materials because they add to the number of neutrons that are responsible for damage or nuclear transmutation in materials of interest [10]. An example of a radiative capture ( $n, \gamma$ )

reaction is the capture of neutron by  $^{235}_{92}\text{U}$ .



Subsequent nuclear reactions, such as fission and further gamma emission, may be possible. In cases where the production of fast neutrons does not accompany fission products, there may also be the production of hydrogen or helium, which could be responsible for displacements in the atom [80].

The effect of neutron capture on material damage is quite evident. From nuclear theory, neutrons at the thermal energy ranges may undergo capture during activation and may change the nucleus of the material due to an increase in the mass number of the atom thereby producing a new isotope of the material. The neutron capture process is also known to produce gases like Krypton (Kr), Xenon (Xe), and Iodine (I) [81].

These gas atoms, usually positioned at the grain boundaries of the materials sometimes migrate and coalesce, forming bubbles that accumulate especially for reactor fuel operated at higher temperatures than the rated ones. The damage production with neutron capture in metallic crystals is lower compared to the elastic and inelastic scattering since the displacement of the neutron-captured nucleus is the most significant damage event. Neutron capture is also largely responsible for impurity production in solid materials.

### **2.3.4 Neutron-induced fission**

Neutron-induced fission is observed when the interaction between a neutron with a fissile or fissionable nucleus decays and produces two daughter nuclides with the release of an enormous amount of energy[70]. A common example of fission is that of U-235 shown in Figure 2.4.

The interaction cross-section of a particular fission reaction gives a quantitative measure of the probability that the reaction would occur. The neutron-induced

fission cross-section of 0.1 barn is possible for U-235 at neutron energies of approximately 0.5 MeV. Lower neutron energy means the fission cross-section is higher, making it more likely to capture the neutron and undergo fission [71]. This interaction cross-section depends on the incident energy of the neutron and the type of nuclide with which the interactions occur. Higher fission cross-section of U-235 for lower energy (thermal) neutrons compared to higher energy (fast) neutrons [70, 71].

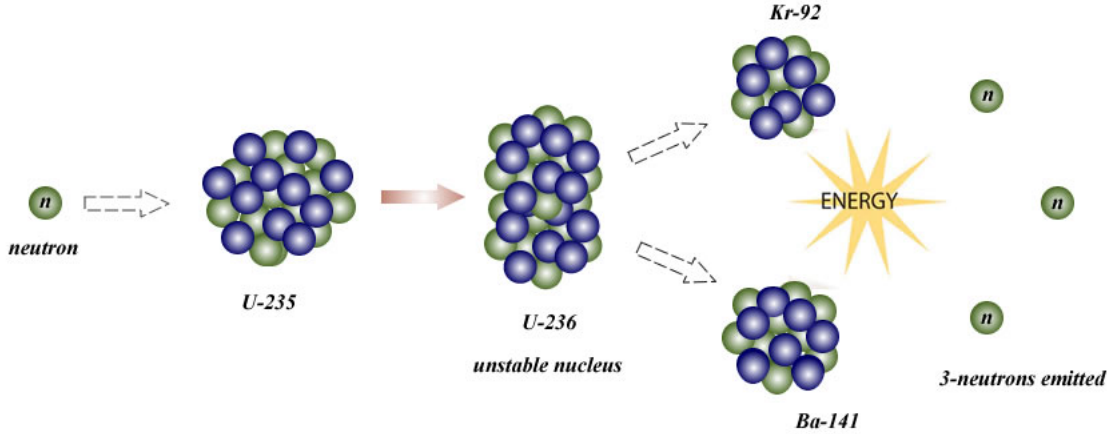


**Figure 2.4:** Neutron induced Fission in Uranium-235 nucleus

Fission products also cause ionization due to the presence of charged ions and they readily contribute to a considerable amount the heat deposition experienced in a material over a short range.[78].

In quantifying radiation damage, the mode of interaction of the neutrons with the materials becomes paramount since the neutron penetration in a material is a function of the energy of the neutrons and the interaction cross-section or the absorbing power (capturing power) of the material transverse by the neutrons. Interaction cross-section shall be considered in the following subsection.

## 2.4 Neutron interaction cross-sections

Neutron interaction cross-sections are key to understanding radiation damage in materials because they detect the energy range and flux of neutrons that interact

with the surface of a particular material. Neutron interaction cross-sections depend on the irradiated atoms, the type of radiation, and the neutron energy [73]. Neutron irradiation in fission and fusion reactors can lead to the deformation of the structural components of the reactor. Therefore, data on neutron cross-sections are essential to predict radiation damage effects in nuclear materials [82].

To define the interaction cross-section, the number of neutrons  $N_o$  incident on an area of the material strip  $dx$  as depicted in Figure 2.5 is considered. This initial number of neutrons traversed through a distance  $x$  is  $N(x)$  and after passing through the elemental strip  $dx$  is reduced to  $N(x) - dN$ .

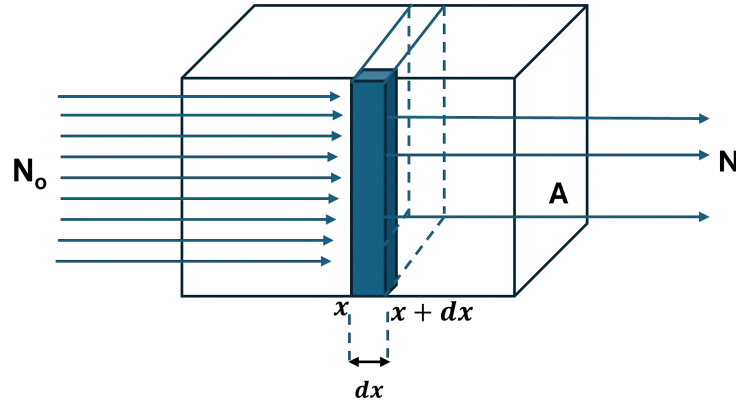


**Figure 2.5:** Description of neutron cross-section for neutrons incidents on a material strip  $dx$

If the number of atoms per unit volume (number density)  $n$  in the slab of thickness  $dx$  and Area  $A$  is considered. The number of nuclei ( $N$ ) in the target volume will be:

$$N = nAdx = nV, \quad (2.12)$$

The Area of the cross-section corresponding to one nucleus is  $n\sigma A dx$ . Where  $\sigma$  is the nuclear cross-section.

The ratio of the number of interactions to the total number of incident particles is equal to the ratio of the area of nuclear cross-sections to the total area  $A$  of the slab.

$$\frac{dN}{N} = \frac{n\sigma A dx}{A}, \quad (2.13)$$

Hence, Eq. 2.13 have that

$$\frac{dN}{N} = -n\sigma dx, \quad (2.14)$$

Using integral calculus, a negative sign is introduced, indicating a decrement in the number of neutrons after an interaction. This gives,

$$\int_{N_o}^N \frac{dN}{N} = -n\sigma \int_0^x dx, \quad (2.15)$$

$$N = N_o e^{-n\sigma x}, \quad (2.16)$$

Where the number density  $n$  is given by

$$n = \rho N_o. \quad (2.17)$$

$\sigma$  is the nuclear cross-section which is measured in barns ( $1\text{barn} = 10^{-24}\text{cm}^2$ ). The total nuclear cross-section is the summation of all the other dependent cross-sections which are designated by suffixes depending on the mechanism of interactions [71]. hence

$$\sigma_T = \sigma_c + \sigma_s + \sigma_f + \dots, \quad (2.18)$$

where:  $\sigma_c$  is the capture cross-section,  $\sigma_s$  scattering cross-section, and  $\sigma_f$  the fission cross-section. This total neutron cross-section ( $\sigma_T$ ) is the combined probability of neutron interactions with a target nucleus across all possible processes.

Neutron cross-section is broadly divided into scattering cross-section which consists of elastic and inelastic scattering and capture cross-section which accounts for the other processes like capture, fission, etc.

### 2.4.1 Elastic scattering cross-section

When neutrons transverse a solid material, there exists an infinitesimal probability that they will impact an atom at the lattice site, producing a recoil energy to the struck atom. This probability is represented by a doubly differential scattering cross-section having the energy of interaction and the angle  $\sigma_s(E_i, E_f, \Omega)$  as parameters, where  $E_i$  and  $E_f$  are the incident and final energies and  $\Omega$  is the solid angle

of the scattered neutron [10].

We can therefore express the scattering probability which is dependent on  $E_i$  and the scattering angle. in single differential form the scattering cross-section is given by:

$$\sigma_s(E_i, \Omega) = \int \sigma_s(E_i, E_f, \Omega) dE_f. \quad (2.19)$$

And the neutrons will have a total scattering probability within the energy  $E_i$  to be

$$\sigma_s(E_i) = \int \sigma_s(E_i, \Omega) d\Omega. \quad (2.20)$$

Considering a deterministic approach, the probability of a neutron with energy  $E_i$ , being scattered elastically against an atom of mass  $M$ , with the energy of recoil  $T$  imparted to the atom, can be obtained using the theory of binary collision. The transferred energy during the collision will be: [80]:

$$T = \frac{\gamma}{2} E_i (1 - \cos \phi), \quad (2.21)$$

and the mass ratio  $\gamma$  is given by:

$$\gamma = \frac{4mM}{(m + M)^2}. \quad (2.22)$$

The neutron mass is  $m$ ,  $M$  is the mass of the PKA;  $T$  the PKA's kinetic energy, and  $\phi$  is the center of mass scattering angle of the neutron in the system. The angular dependence can be shown in Figure 2.6,

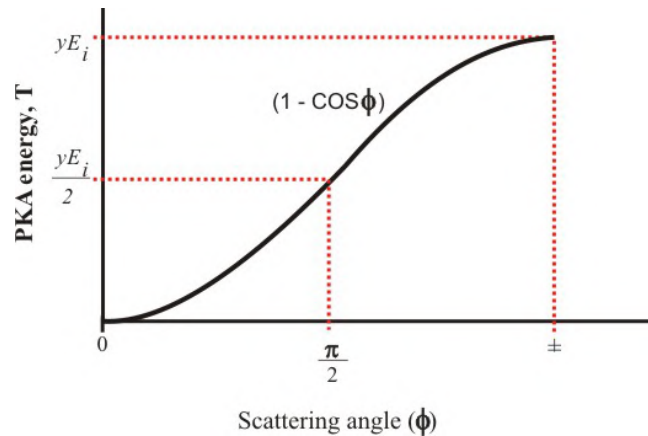


**Figure 2.6:** Energy transfer as a function of scattering angle. ( [10])

where the PKA transfer energy increases from 0 at  $\phi = 0$  to a maximum of  $\gamma E_i$  at  $\phi = \pi$  or  $T_{max} = \gamma E_i$ . If the mass  $m = 1$  and  $M = A$ , T will now depend only on one unknown,  $\phi$ . Hence,  $\gamma$  becomes:

$$\gamma = \frac{4A}{(1+A)^2}. \quad (2.23)$$

The elastic cross-section for the Au-197, Al-27, and Lu-195 nuclides is illustrated in Figure 2.7, 2.8 and 2.9.

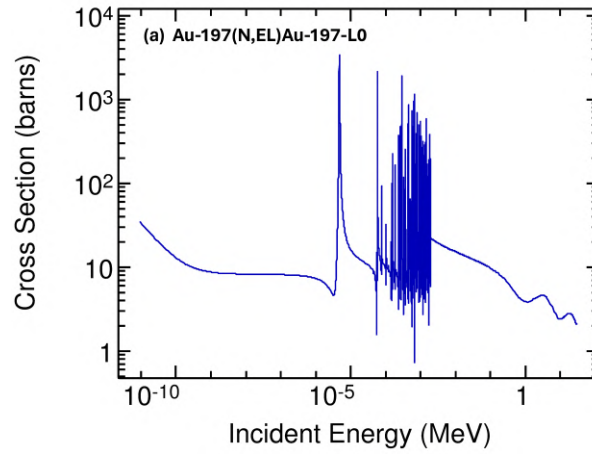


**Figure 2.7:** Elastic Cross-Section for Au-197 showing resonance dominating at the thermal neutron energy range (Plot made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org> [83])

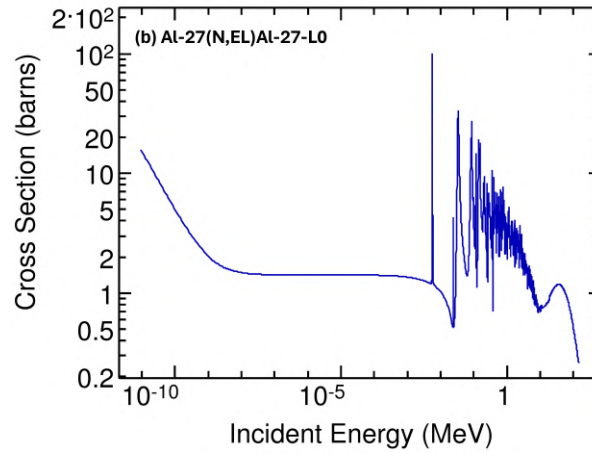


**Figure 2.8:** Elastic Cross-Section for Al-27 with resonance dominating at the fast neutron energy range (Plot made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org> [83])

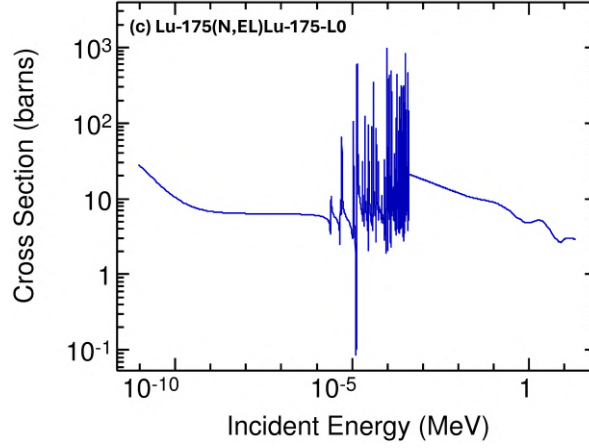


**Figure 2.9:** Elastic Cross-Section for Lu-175 with resonances around the thermal energy range (Plot made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org> [83])

The plots depict the likelihood of neutron interaction occurring elastically with these materials. The interaction probability for Au and Lu is greater at thermal neutron energies, whereas that for Al is higher in the fast neutron energy range. These cross-sections influence neutron transport, moderation, and absorption within materials containing these elements, which in turn impact the extent of radiation damage observed in the materials containing these elements [9, 76]. The extent of damage is also dependent on other factors like the composition of materials, energy spectrum, and irradiation conditions.

### 2.4.2 Inelastic scattering cross-section

An increase in the neutron energy a few MeV higher than the energy utilized in elastic scattering triggers inelastic scattering, where the recoil nucleus remains in an excited state and subsequently emits a photon. This mechanism of interaction contributes to radiation damage in materials by inducing atomic excitation and subsequent energy transfer to the material lattice. For materials such as Au, Al, and Lu whose interaction cross-section is shown in Figure 2.10, 2.11 and 2.12, neutron inelastic scattering in these materials or their alloys can result in the excitation of atomic nuclei to higher energy states. This excitation process may contribute to radiation damage through mechanisms such as phonon generation, lattice vibrations, and the creation of defects such as vacancies and interstitials [32].

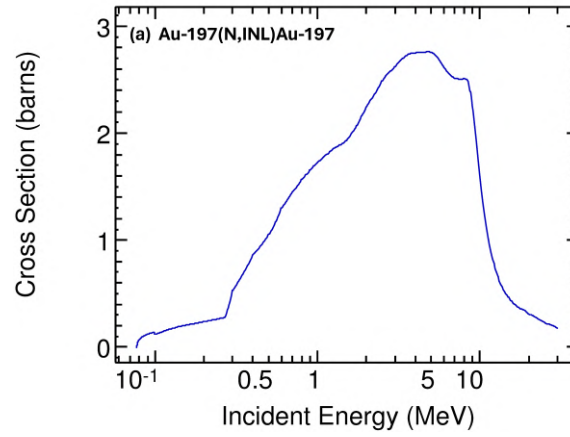


**Figure 2.10:** Inelastic Cross-Section for Au-197 ( plot made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org> [83])

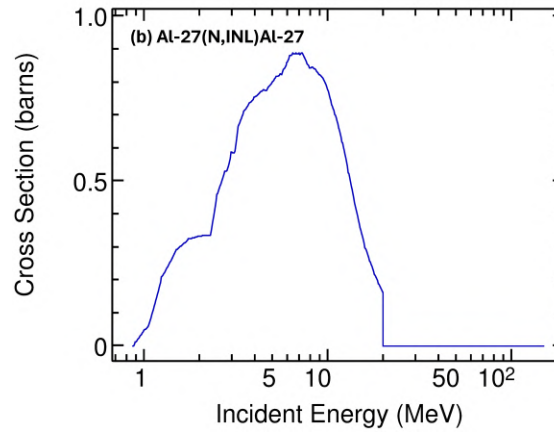


**Figure 2.11:** Inelastic Cross-Section for Al-27 ( plot made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org> [83])

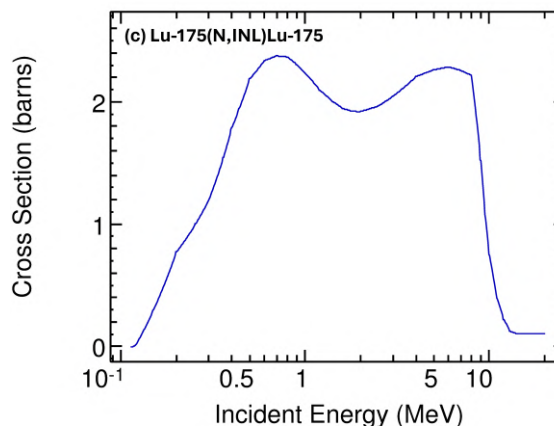


**Figure 2.12:** Inelastic Cross-Section for Lu-175 ( plot made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org> [83])

The inelastic neutron cross-sections for Au, Al, and Lu are lower than the probabilities of elastic cross-sections, and most probable at the fast neutron energies less dominated by resonances, as illustrated in Figure 2.12.

### 2.4.3 Fission cross-sections

The probability of a nuclear fission reaction occurring is referred to as the fission cross-section, denoted by  $\sigma_f$  [71]. A common example of a fission cross-section in fission reactors is Uranium-235, as depicted in Figure 2.13(b). The cross-sections of fissile materials or nuclei for neutron-induced fission heavily depend on the incident neutron energy.

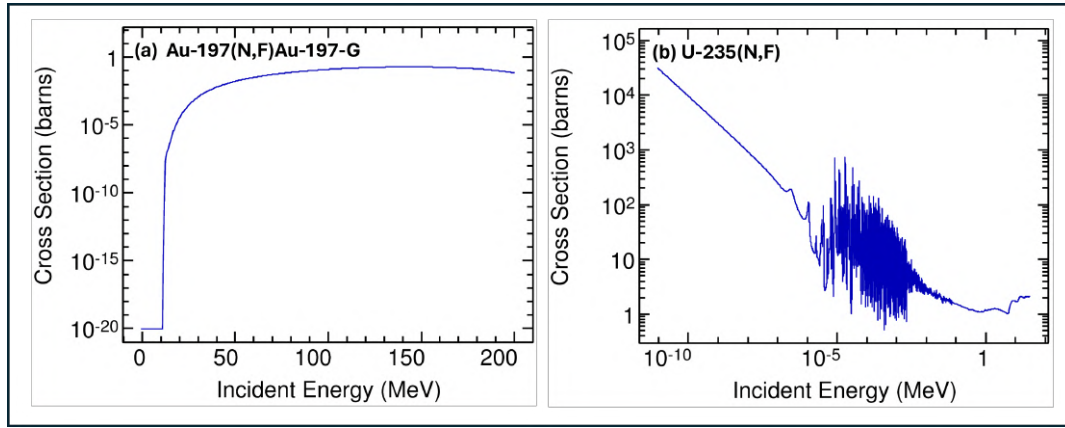


**Figure 2.13:** Fission Cross-Section for (a) Au (b) U-235 (Plot made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org/> [83] )

The neutron energy dependence of the fission cross-section reveals three basic regions. In the first region, characterized by low energies,  $\sigma_f$  is closely proportional to  $\frac{1}{v}$  [71, 76], and the fission cross-section is also large. The second region is marked by resonances, and beyond the resonance region, there is a smooth and gradually decreasing  $\sigma_f$  curve at higher energies. In contrast, Figure 2.13(a), illustrates the probability of fission in Au which is very low and is most probable at high neutron energies.

### 2.4.4 Radiative capture cross-section ( $n, \gamma$ )

The probability of neutron capture is expressed through the cross-section for radiative capture, conventionally denoted as  $\sigma_\gamma$ . Comparatively, the cross-section for

radiative capture is significantly greater at lower energies when contrasted with higher energies. The maximum cross-section is observed within the resonance region, associated with long-lived states of the compound nucleus [71, 76, 84].

The cross-section plots for Au-197 and Lu-175, as illustrated in Figure 2.14 and 2.16, exhibit elevated probabilities around thermal neutron energies, sharply diminishing at higher energies beyond the resonance region. In Figure 2.15, the probability of radiative capture in Al-27 is low, falling off rapidly, especially within the fast neutron region characterized by resonances.

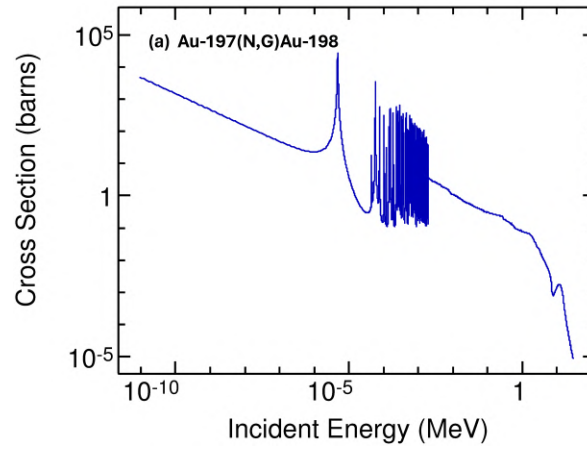


**Figure 2.14:** Radiative Capture Cross-Section for Au-197 (Plot made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org/> [83])

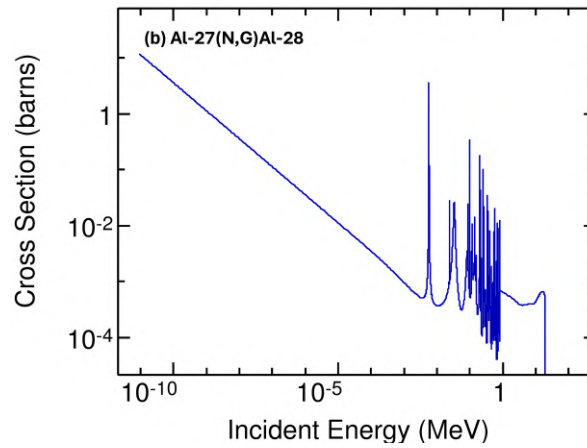


**Figure 2.15:** Radiative Capture Cross-Section for Al-27 (Plot made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org/> [83])

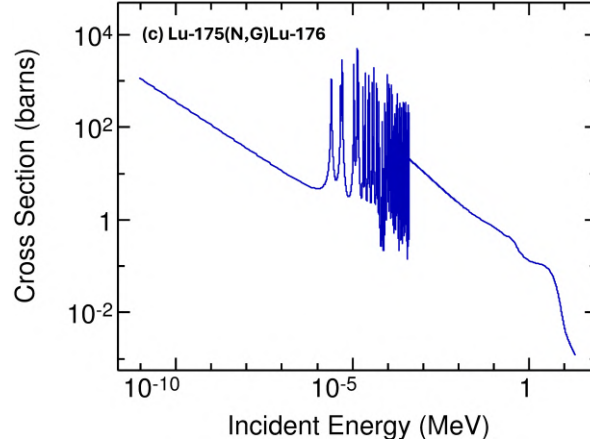


**Figure 2.16:** Radiative Capture Cross-Section for Lu-175 (Plot made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org/> [83])

Radiation damage is enhanced by the  $(n, \gamma)$  reaction due to photon emission. However, the contribution of this reaction to dpa relative to neutron is very small for most metals [10]. For instance, neutron flux ( $\phi_n$ ) of  $1 \times 10^{14} \text{ n.cm}^{-2}.\text{s}^{-1}$  and displacement cross-section  $\sigma_d$  (Al)  $4.0 \times 10^{-22} \text{ cm}^2$  for fast neutron regime in a year will contribute a neutron dpa of about 1.26 dpa on Aluminium sample, whereas gamma flux ( $\phi_\gamma = 1 \times 10^{13} \text{ } \gamma.\text{cm}^{-2}.\text{s}^{-1}$ ) with the efficiency of  $1 \times 10^{-3}$  contributes about 3.15 mdpa, three orders of magnitude less dpa in the same material. The  $(n, \gamma)$ , reactions are relevant in radiation damage studies because they contribute significantly to the material's heating. The  $\gamma$  rays emitted can be absorbed by the material, depositing their energy and thereby increasing the thermal energy of the materials [10, 58].

A particular reactor component of interest where the effect of this reaction is relevant to damage is in the Reactor Pressure Vessels (RPV), especially in circumstances where there is a large water gap between the vessel and the fuel assembly. In these cases, the flux due to  $\gamma$  is comparable to the neutron flux in the fast flux ranges [10, 85]. This has been observed in the study by Chen et al. [86] on  $\gamma$  - ray displacement damage of advanced boiling water reactor (ABWR).

The mean recoil energy of the reaction is given by the half-value of the highest

energy of recoil, denoted as  $\bar{T}$  and is calculated as follows:

$$\bar{T} \cong \frac{E_\gamma^2}{4(M + m)c^2} \quad (2.24)$$

The model of the capture cross-section is based on the Breit-Wigner single-level resonance when the intrinsic angular momentum of the target nucleus is zero. The cross-section is dependent on the neutron and gamma radiation widths ( $\Gamma_n$  and  $\Gamma_\gamma$ ), total width ( $\Gamma$ ), resonant energy  $E_0$  and the wavelength  $\lambda$  [71, 87]. Thus, can be expressed as follows:

$$\sigma_{n,\gamma}(E_i) = \pi\lambda^2 \frac{\Gamma_n \Gamma_\gamma}{(E_i - E_0)^2 + \left(\frac{\Gamma}{2}\right)^2}, \quad (2.25)$$

Eq. 2.25 can be written in terms of the maximum value of the capture cross-section  $\sigma_0$ , where  $E = E_0$  and  $\Gamma_n$  being proportional to  $\lambda^{-1}$ . The cross-section is now dependent on the total radiative capture width  $\Gamma$  thus: [10]:

$$\sigma_{n,\gamma}(E_i) = \sigma_0 \sqrt{\frac{E_0}{E_i}} \frac{\left(\frac{\Gamma_\gamma}{2}\right)^2}{(E_i - E_0)^2 + \left(\frac{\Gamma_\gamma}{2}\right)^2}, \quad (2.26)$$

where  $E_0$  is the resonant energy and  $E_i$  is the energy of the incoming neutron.

#### 2.4.5 $(n, 2n)$ Neutron cross-section

The  $(n, 2n)$  reactions are significant in radiation damage effects owing to their ability to produce extra neutrons that can cause damage or transmutation in the nuclear structure of some components of interest [10]. The  $(n, 2n)$  reaction cross-section is regarded as a special case of the inelastic scattering cross-section. Studies on  $(n, 2n)$  excitation plots of fast neutron-induced reactions have formed a significant part of the development of varieties of nuclear fission reactor technologies, prediction of depth-dose distribution, and control of flux in the reactors [82]. These key areas have the effect of radiation damage on materials.

The  $n$ ,  $2n$  reaction cross-sections for Au-197, Al-27, and Lu-175 are depicted in Figure 2.17, 2.18, and 2.19. These reactions are most probable at elevated neutron energies, forming a single broad peak spanning approximately 1 MeV to 20 MeV. Notably, Al-27, as seen in Figure 2.18, experiences a sharp drop-off at 20 MeV. The  $n$ ,  $2n$  cross-section for each material remains low, reaching a maximum of about 2.2 barns for Au-197, 2.0 barns for Lu-175, and 0.11 barns for Al-27, respectively.

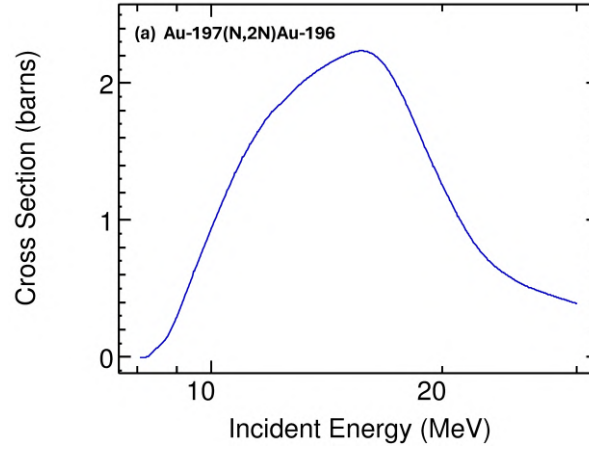


**Figure 2.17:** N, 2N Cross-Section for Au-197 (Plots made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org/> Ref: [83])

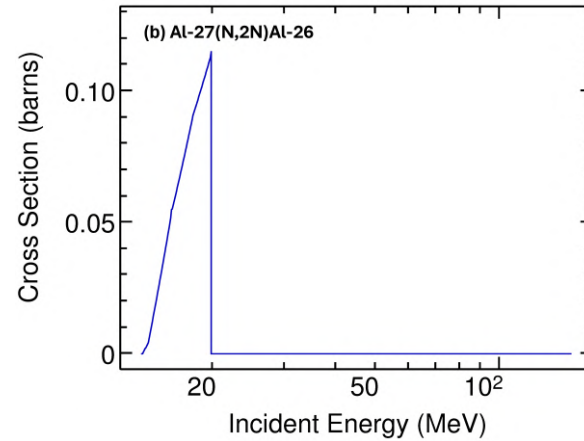


**Figure 2.18:** N, 2N Cross-Section for Al-27 (Plots made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org/> Ref: [83])

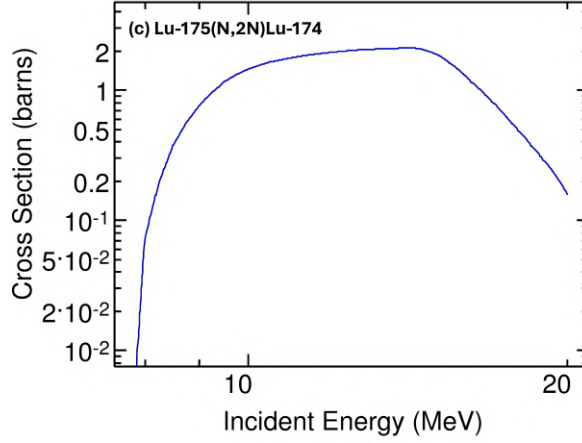


**Figure 2.19:** N, 2N Cross-Section for Lu-175 (Plots made from IAEA Evaluated Nuclear Data Files (ENDF) database, <https://www-nds.iaea.org/> Ref: [83])

#### 2.4.6 $(n,p)$ reaction cross-section

The  $(n,p)$  is a neutron-induced transmutation reaction, which is an interaction where a neutron ( $n$ ) interacts with a nucleus, resulting in the emission of a proton ( $p$ ) [88]. This type of reaction is common in nuclear physics and can occur with various target nuclei. The cross-section of this reaction, denoted as  $\sigma_{n,p}$ , is a measure of the probability that a neutron will induce a proton emission when it collides with a nucleus.

The  $(n,p)$  reaction cross-section is an important reaction for understanding radiation damage in various environments [51, 89]. This is essential in fields such as nuclear medicine, space exploration, and nuclear power generation, where exposure to neutron radiation is significant.

The  $(n,p)$  reaction cross-sections for Au-197, Al-27, and Lu-175 are illustrated in Figures 2.20, 2.21, and 2.22. Al-27 has the highest cross-section of about 0.09 barns around 14 MeV neutron energy, followed by Lu-175 whose probability of interaction illustrated in Figure 2.22 is linearly dependent on the neutron energy.

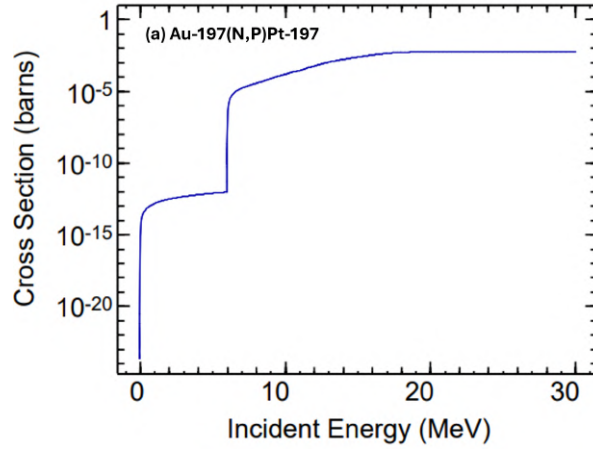


**Figure 2.20:** n,p Cross-Section for Au-197 (Plots made from IAEA ENDF database, <https://www-nds.iaea.org/> Ref: [83])

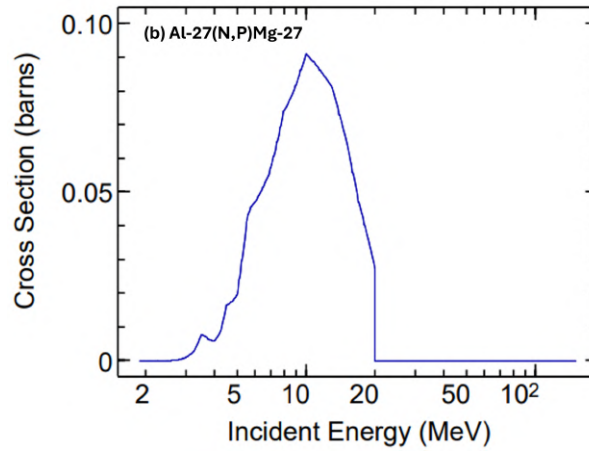


**Figure 2.21:** n,p Cross-Section for Al-27 (Plots made from IAEA ENDF database, <https://www-nds.iaea.org/> Ref: [83])

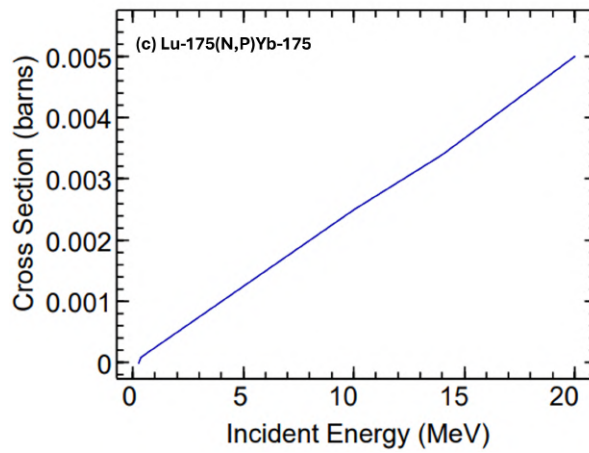


**Figure 2.22:** n,p Cross-Section for Lu-175 (Plots made from IAEA ENDF database, <https://www-nds.iaea.org/> Ref: [83])

Au-197 however has the lowest cross-section of the  $(n,p)$  reaction. This reaction influences radiation damage in structural materials like Al and its alloys. This triggers material embrittlement and the formation of gas bubbles and corrosion within the reactor materials.[89]

#### **2.4.7 $(n,\alpha)$ reaction cross-section**

The  $(n,\alpha)$  reaction involves a neutron interacting with a nucleus, resulting in the emission of an alpha particle (two protons and two neutrons). This type of reaction is significant in nuclear physics and engineering due to its implications for reactor materials and neutron economy [10].

This reaction is particularly common in materials like boron and lithium, which are often used in control rods and shielding. The alpha particles generated through  $(n,\alpha)$  reactions are highly ionizing and can cause significant damage to the crystal lattice of reactor materials, leading to embrittlement, swelling, and the formation of voids and dislocations within the material structure. This degradation affects the mechanical properties of the materials, reducing their ductility and fracture toughness, and ultimately compromising the integrity and safety of reactor components [10, 24, 50, 53]. Such damage demands a careful material selection and design considerations to mitigate these effects, ensuring the longevity and reliability of nuclear reactors.

The  $(n, \alpha)$  reaction cross-sections for Au-197, Al-27, and Lu-175 are shown in Figures 2.23, 2.24, and 2.25.

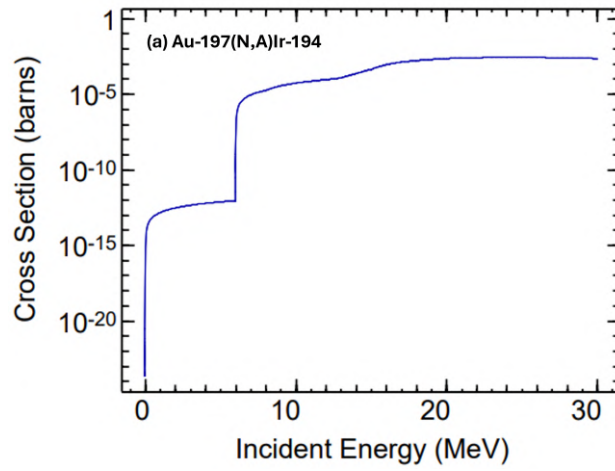


**Figure 2.23:**  $n,\alpha$  Cross-Section for Au-197 (Plots made from IAEA ENDF database, <https://www-nds.iaea.org/> Ref: [83])

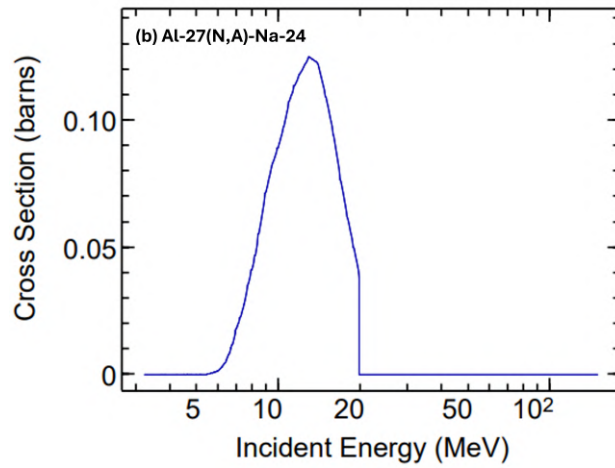


**Figure 2.24:**  $n,\alpha$  Cross-Section for Al-27 (Plots made from IAEA ENDF database, <https://www-nds.iaea.org/> Ref: [83])

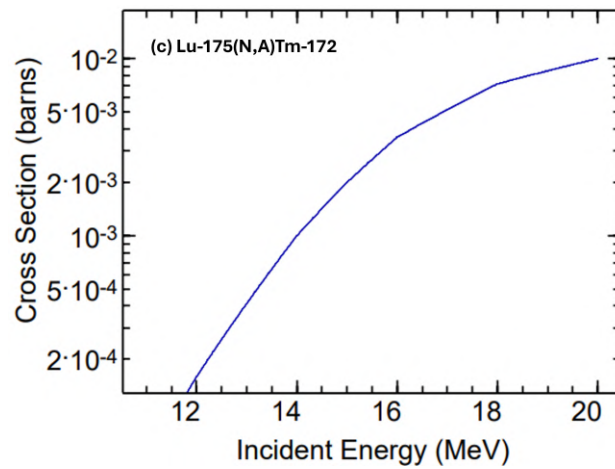


**Figure 2.25:**  $n,\alpha$  Cross-Section for Lu-175 (Plots made from IAEA ENDF database, <https://www-nds.iaea.org/> Ref: [83])

The  $(n, \alpha)$  reaction cross-section has a similar trend however, there are differences in terms of the transmutations and probabilities of occurrence. Al-27 has the highest cross-section of about 0.12 barns around 14 MeV neutron energy followed by Lu-175 with a lower reaction cross-section that is curve-linear and proportional to the neutron energy as seen in Figures 2.24 and 2.25. Au-197 in Figure 2.23 still maintains its cross-section value and shape probability due to its inertness.

Neutrons have been extensively discussed in the section above because their interactions and reactions with the nuclei of atoms in the crystalline lattice can lead to the creation of different particles and photons, such as protons, alpha particles, electron pairs, and other nuclear fragments. These secondary particles can cause additional damage as they traverse through the crystal lattice. Furthermore, these particles can be produced in accelerators and other natural processes in space. Understanding the specific particle or photon interactions with crystalline materials is crucial for assessing the potential radiation damage and designing materials for various applications, including nuclear reactors, radiation shielding, and other technological uses. The following sections discuss the interaction of photons and charged ions in materials.

## 2.5 Interaction of photons with matter

In nuclear fission reactors,  $\gamma$  radiation is produced alongside neutrons as the neutrons interact with structural and target materials. The probability of photon production depends on the neutron energy available for collisions and the neutron interaction cross-sections explored in the preceding section. Consequently, there is a necessity to delve into the interaction of photons with materials concerning damage.

Gammas, like neutrons, are neutral radiation that interacts with matter indirectly, releasing its energy to the materials. The major difference between neutrons and gamma is that neutrons interact directly with the nucleus of the atom producing significant atomic displacement and transmutation reaction leading to higher dpa, while photons interact extra-nuclearly with the electrons and in some cases,

photofission but contributes to less atomic displacement. The interaction of photon radiation with materials is important, especially when considering the reactor field for irradiation. There are many sources of gammas in the fission reactor that could interact with the material under irradiation. The most important sources are fission, decay of fission products, capture processes in fuel, inelastic scattering in the fuel, and decay of capture products [10, 71, 90].

This section discusses the interaction mechanisms of photon radiation with crystalline materials and the possible radiation damage effects they create.

### 2.5.1 Interaction mechanisms by photons

The main interaction mechanisms of photons with materials include photoelectric absorption, Compton scattering, and pair production. Other interaction mechanisms for radiation damage are photo-disintegration, Cherenkov radiation, and coherent scattering.

- *Photoelectric absorption* occurs at low energies ( $E < 200$  keV) where a photon transfers its energy completely to the orbital electron. In this way, the energy of the photon is absorbed by the material with which it interacts [78].
- *Compton scattering* is a type of elastic scattering that occurs within the energies of 200 keV to 1.5 MeV. In this interaction, the energy of the incident photon is shared between the scattered photon and the recoil Compton electron. The photon is deflected from its original track of interaction, while the difference in the energy between the incident and the scattered photons is transferred to the free electron.
- *Pair production* occurs when photons of sufficient energy interact with atoms of the absorbing materials in the coulomb field of the nucleus. The minimum energy of gamma-ray photons for pair production is 1.02 MeV. This amount of energy is sufficient enough to produce the annihilation of an electron-positron pair with rest mass energies of about 511 keV.

These interaction mechanisms detect the kind of radiation effect that a gamma will have on a material. The radiation effects of gamma radiation on materials are discussed in the following sub-section.

### 2.5.2 Radiation damage effects by gamma on materials

The radiation damage effects of photons on crystalline materials are generally low. Gamma radiation primarily induces damage through the indirect production of secondary electrons, either via Compton scattering or pair production. These secondary electrons, upon further interaction, lead to the displacement of atoms or ionization of the material [91].

However, it is important to note that damage production for  $\gamma$  radiation occurs uniformly throughout the bulk of the material they interact with [78]. Table 2.3 presents an overview of radiation effects caused by neutrons, fission fragments, photons, protons, and other charged particles based on applications related to radiation damage. Radiation effects in materials are generally classified into:

- *Atomic displacement*, where atoms of a material are displaced from their normal lattice sites, creating Frenkel pairs.
- *Production of impurity* via transmutation of nuclei into other atomic species. This is present during capture (fission and activation). There could be the production of hydrogen (H) or helium (He), which is a major concern in reactor technology [51]. The production of H and He poses a significant contribution to radiation damage in structural materials. Helium atoms are insoluble in metals and tend to cluster, forming bubbles or voids within the metal matrix. These voids can grow and coalesce, leading to void swelling and several other detrimental effects.
- *Ionization*: This is the removal of orbital electrons from a material leading to the formation of an ion.
- *Heating*: There could be the production of a large amount of energy in the form of thermal heat deposition in the material.

**Table 2.3:** Radiation types and damage effects to materials [78]

| Radiation                                     | Radiation Effect                |                             |                                            |                                        |
|-----------------------------------------------|---------------------------------|-----------------------------|--------------------------------------------|----------------------------------------|
|                                               | Displacement                    | Impurity                    | Ionization                                 | Heating                                |
| <b>Thermal neutrons</b>                       | yes, indirectly                 | yes, directly by absorption | yes, indirectly                            | yes, indirectly                        |
| <b>Fast neutrons</b>                          | yes, significant via scattering | yes, by absorption          | yes, indirectly                            | yes, high amount, very short range     |
| <b>Fission fragment</b>                       | yes, significant via scattering | yes, self impurities        | yes, charged ions, emit $\beta$ , $\gamma$ | yes, heating over short range          |
| <b>Alpha (<math>\alpha</math>)</b>            | yes                             | yes, He build-up            | yes, significant                           | yes, over a very short range           |
| <b>Proton</b>                                 | yes                             | yes, H build-up             | yes, directly                              | yes, over a very short range           |
| <b>Beta (<math>\beta</math>)</b>              | partial displacements           | No                          | yes, directly                              | yes, localized heating                 |
| <b>Photons (<math>\gamma</math> , X-rays)</b> | Rare, via Compton effect        | No                          | yes, indirectly                            | yes, $\gamma$ -heating over long-range |

Photons interacting by Compton scattering transfer the larger part of the energy to the scattered electrons and the little energy that is given to the atomic nuclei is less than the minimum energy required to create atomic displacement (the atomic displacement energy  $E_d$ ). Similarly, during photoelectric absorption, the energy acquired by the atom after ionization is too low compared to that acquired by the photo-electron and hence, insignificant to produce atomic displacement[7, 78].

Therefore, considering the damage effects above, the production of damage by gammas in crystalline materials occurs basically with ionization and heat deposition. Table 2.3 categorizes the possible radiation effects with the radiation types. It should be noted that  $\gamma$ -radiation on its own is unable to create atomic displacements because of the insufficient energy transferred to the atomic nuclei during the mechanisms of interactions. However, secondary electrons and positrons produced by the electronic excitation process may be responsible for the breaking of bonds leading to atomic displacement [92]. The probability of atomic displacement in materials with secondary electrons and positrons is small [7]. Therefore, ionization and thermal  $\gamma$ -heat deposition in the materials remain the possible means for radiation damage effects by gamma radiation in crystalline materials.

## 2.6 Interaction of charged particles with matter

Charged particles exert electrical force (the Coulomb force) on the electrons of the atoms of the matter producing excitation and ionization. The penetration of charged particles into an electron cloud can lead to elastic scattering of the particle from the nucleus. In this type of collision, both kinetic energy and momentum are conserved leading to the recoils of the nucleus. The ejection of the recoiled nucleus into a medium to another charged particle is possible especially when the incident particle is massive and has sufficient energy.[71].

Charged particles can be classified into heavy and light-charged particles or ions. Heavy ions include alpha ( $\alpha$ ) particles, Xenon ( $\text{Xe}^+$ ), gold ( $\text{Au}^+$ ) etc. These species of ions lose energy step-wise as they interact with the electrons of the materials through which they transverse. When their energy is below the required energy for excitation they tend to lose the energy via nuclear collisions. Since they are massive, their speed into materials reduces continuously due to Coulomb's interactions with the electrons of the atom in straight paths having a short range in air [71, 78].

Whereas, light charged particles typically refer to ions with relatively low atomic masses. light charged particles transverse matter through ionization, Bremsstrahlung, and elastic collisions which may be nuclear and electronic interactions alike. these include beta ( $\beta$ ), protons ( $\text{H}^+$ ), Deuterons ( $\text{D}^+$ ). The most important mechanism of interaction of Beta ( $\beta$ ) particles, for instance, is the elastic scattering resulting in the Brownian motion path in the material they transverse.

### 2.6.1 Radiation damage mechanism by charged particles

The description of radiation damage with neutrons in materials is based on their interaction cross-sections. However, for charged particle interaction with crystalline materials, it is more useful to describe their interaction based on the number of ion pairs they produce per unit track length as they interact with the material. This is referred to as specific ionization or the total energy loss in the material per unit length of the material known as stopping power (S). Specific ionization is a term

used more often when describing damage in biological tissues. The energy loss per unit length ( $-\frac{dE}{dx}$  or  $NS(E)$ ) is given by Eq. 2.27 [10, 71, 93]:

$$\left(-\frac{dE}{dx}\right)_{total} = \left(-\frac{dE}{dx}\right)_n + \left(-\frac{dE}{dx}\right)_e + \left(-\frac{dE}{dx}\right)_r = NS_n + NS_e + NS_r, \quad (2.27)$$

Where  $S$  is referred to as stopping power and  $N$  is the atomic number density of the target atom. The subscripts  $n$ ,  $e$ , and  $r$  in the equation represent the energy loss mechanisms due to nuclear, electronic, and radiation, respectively. The energy loss via radiation is often infinitesimal and can be neglected. Thus, we have that:

$$\left(-\frac{dE}{dx}\right)_{total} = \left(-\frac{dE}{dx}\right)_n + \left(-\frac{dE}{dx}\right)_e = NS_n + NS_e, \quad (2.28)$$

The stopping power is relevant to describe radiation damage charged particles cause in materials. This energy loss of the ions to the materials (target atom) takes two forms: the energy transfer by ion to the target atom known as the electronic stopping or the inelastic energy dissipation and the energy transfer from ion to target nuclei which is the nuclear stopping or the elastic energy dissipation or loss [10].

Nuclear collisions are responsible for a significant amount of energy losses and changes in the trajectory of the ion as it transverses the target material. This accounts for the crystal lattice disorder created by the atoms that are displaced from their equilibrium positions. Much lower energy dissipation is present during electronic collisions and the change in the path of the ion is insignificant. Hence, lattice disorder is negligible with electronic collisions.

The rate of energy dissipation or the stopping cross-section is given by.

$$S = \frac{1}{N} \frac{dE}{dx}, \quad (2.29)$$

Here,  $N$  represents the atomic number density. The stopping cross-section is expressed in  $eV \cdot cm^2/atom$ . The average energy transfer is presented as a function of the differential energy transfer cross-section  $\sigma(E_i, T)$  for either nuclear or electronic

stopping.

$$\bar{T} = \frac{\int T \sigma dT}{\int \sigma dT}, \quad (2.30)$$

where  $\bar{T}$  is the average energy transfer. The mean free path ( $\lambda$ ) is given by :  $\lambda = \frac{1}{N\sigma}$ , The energy loss per unit length for nuclear stopping can therefore be written as [10]

$$\frac{dE}{dx} = \frac{\bar{T}}{\lambda} = \frac{\int_{T_i}^{T_f} T \sigma(E_i, T) dT}{\int_{T_i}^{T_f} \sigma(E_i, T) dT} \times N \int_{T_i}^{T_f} \sigma(E_i, T) dT = N \int_{T_i}^{T_f} T \sigma(E_i, T) dT. \quad (2.31)$$

where the limits of the integral,  $T_i$  and  $T_f$  represent the minimum and the maximum energies losses, respectively

### (a) Energy loss by nuclear stopping mechanism

In the energy loss theory, nuclear stopping can be described in relation to damage in a target material as the binary interaction between an ion (projectile) and the nuclei of the target atoms. This interaction involves the transfer of energy from the projectile ion to the target. The process includes an elastic collision between the ion and the stationary target atom, as illustrated in Figure 2.26.

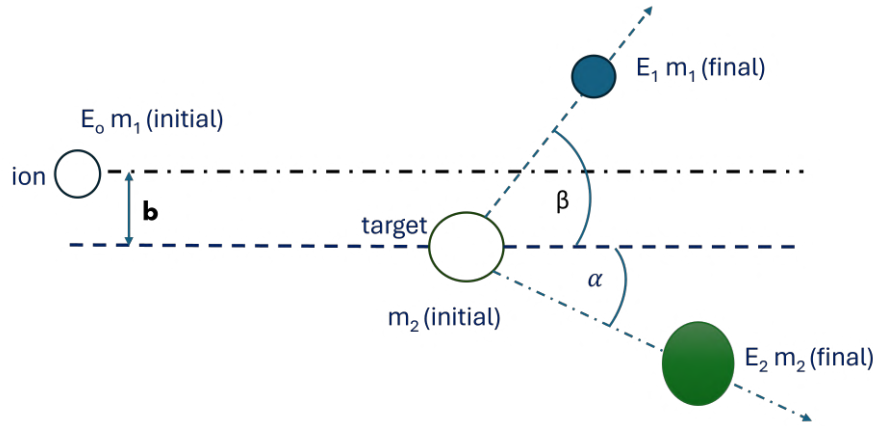


**Figure 2.26:** Elastic Collision of a projectile ion onto a stationary target atom producing a recoil atom at an angle of  $\alpha$  and the ion scattered at an angle  $\beta$

From Eq. 2.31, the nuclear stopping or energy loss by the ion during the elastic collision in low-energy interactions is described by the screened Coulomb function. This function accounts for the effects of electrons with the internuclear spacing and

is more applicable for applications in this study. The differential energy transfer cross-section is thus given in Eq. 2.32

$$\sigma(E, T) = \frac{C_m}{E^m T^{1+m}} \quad (2.32)$$

where

$$C_m = \frac{\pi}{2} \lambda_m a^2 \frac{C_m}{E^m T^{1+m}} \left( \frac{2Z_1 Z_2 \varepsilon^2}{a} \right)^{2m} \left( \frac{m_1}{m_2} \right)^m, \quad (2.33)$$

Here,  $\lambda_m$  and  $m$  are fitting variables [10, 94],  $a$  is the screening radius,  $\varepsilon$  is the unit electronic charge, and  $m_1$  and  $m_2$  are the masses of the ion and the target atom colliding. Further details on the derivation can be found in the references [10, 95]. Substituting Eq. 2.32 into Eq. 2.31, the nuclear-stopping power is obtained as:

$$S_n(E) = \frac{C_m E^{1-2m}}{1-m} \gamma^{1-m}, \quad (2.34)$$

where

$$\gamma = \frac{4m_1 m_2}{(m_2 + m_1)^2}. \quad (2.35)$$

The nuclear stopping for Au and Lu-Al generated from the SRIM-2013 database is shown in orange colour in Figure 2.27 and 2.28 respectively.

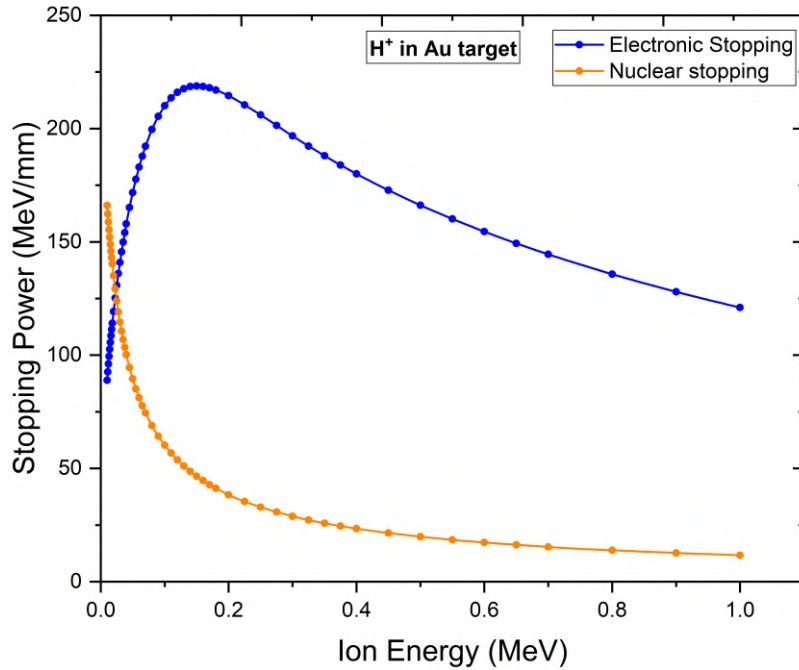


**Figure 2.27:** Nuclear and electronic stopping for Au. Generated from SRIM-2013 stopping power database. Note that nuclear-stopping power is scaled to a factor of  $10^2$  [59]

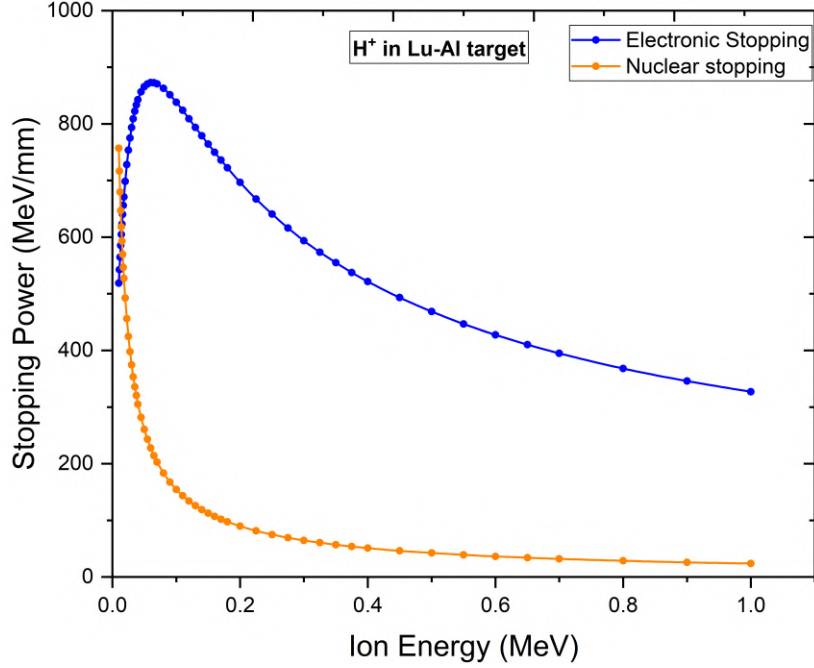


**Figure 2.28:** Nuclear and electronic stopping for Lu-Al. Generated from SRIM-2013 stopping power database. Note that nuclear-stopping power is scaled to a factor of  $10^2$  [59]

### (b) Energy loss by electronic stopping mechanism

The rate at which energy is lost due to the transfer of energy from charged particles to electrons in a material is known as electronic stopping power. The electronic stopping for Lu-Al and Au is shown in blue in Figure 2.27 and 2.28 respectively. This parameter is key in comprehending and forecasting the impact of energetic particles on the crystal lattice. These interactions result in the conversion of kinetic energy from ions to electrons, giving rise to electron-hole pairs, excitations, and various electronic defects within the material.

The electronic stopping power arising from excitation-ionization interactions is given by:

$$\left(-\frac{dE}{dx}\right)_e = \frac{n}{Z_2} \int_{\bar{I}}^{\gamma_e E_i} T \sigma_s(E_i, T) dT, \quad (2.36)$$

$$\left(-\frac{dE}{dx}\right)_e = N Z_2 \pi \frac{Z_1^2 \varepsilon^4 m_1}{E m_e} \ln \left( \frac{\gamma E}{\bar{I}} \right) \quad (2.37)$$

In Eq. 2.35, the symbol  $\gamma$  is defined with  $m_2 = m_e$ , where  $n = NZ_2$  represents the electron density, and  $\bar{I}$  denotes the average excitation-ionization energy.

The nuclear and electronic stopping behaviours exhibit distinct patterns, as illustrated in Figure 2.27 and 2.28 for Au and Lu-Al. The two mechanisms of energy loss can result in structural damage to materials, potentially occurring in different damaged regions [96, 97]. The extent of damage caused by electronic stopping is not fully understood. However, electronic energy loss predominantly influences optical and electronic properties, as well as the generation of thermal spikes in materials [8, 10, 97].

Some studies suggest that electronic energy loss can lead to the annealing of cascade damage caused by ions, thereby reducing efficiency due to electronic effects [97–99]. Nuclear stopping is primarily responsible for inducing displacement damage and is associated with most critical damage effects experienced in crystalline materials [8–10, 64].

Stopping of ions in materials is best described using inter-atomic potentials which are governed by the nature of forces acting between two colliding atoms. These potential functions are guided by the interactions. Knowledge of interatomic potential functions is critical in the description and understanding of radiation damage in materials. However, there are many potential functions to describe the interaction of radiation, the forces acting between the collision of two atoms and the ensuing energy transfer cross-sections which is essential in the description of radiation damage for a material.

The most common potential function is the Coulomb's potential upon which several other potential functions are derived and modified to suit specific conditions. A description of the variation of the interatomic potential with the separation is shown in Figure 2.38 [10].

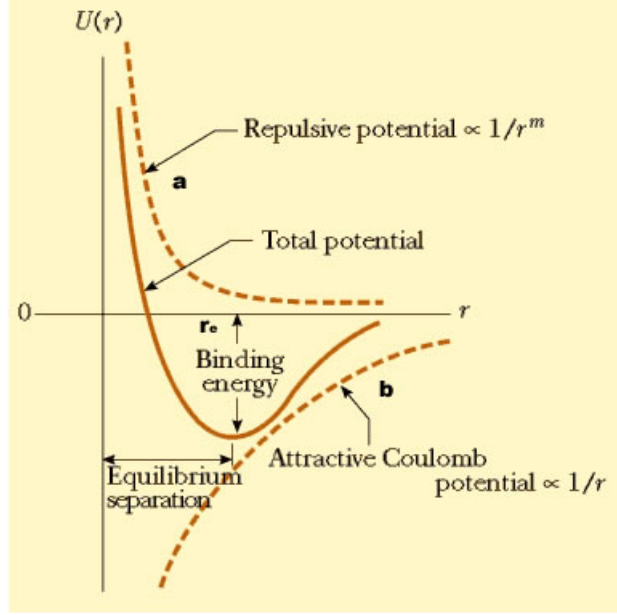


**Figure 2.29:** Changes in inter-atomic potential with separation  $R$ , with repulsive forces higher at region (a) and attractive force dominate at large separation at region (b). Ref: [100]

The Coulomb's potential function is shown in Eq. 2.38

$$U(r) = K_e \frac{q^2}{r}, \quad (2.38)$$

where the Coulomb constant  $K_e = \frac{1}{4\pi\epsilon_o}$  is evaluated to be  $8.98755 \times 10^9 \text{ Nm}^2\text{C}^{-2}$ ,  $\epsilon_o$  is the permittivity of free space, and  $q$  is the electronic charge.

Other potential functions were further developed to ensure close approximation to interactions in nature, some of which include the Morse potential, Born-Mayer (Buckingham) potential, Embedded Atomic Method (EAM), and Lennard-Jones potential mostly used for the modeling of the pairwise interaction occurring between two ions or atoms with solid states or liquids [93].

The choice of a suitable potential function to describe radiation damage in materials depends on the specificity of the collision (particle type, ions, photons, neutrons), the type/state of the target material (metals, semiconductors, liquids, and gas), and the range of separation and the nature of atoms within the interaction whether a single atom or many-body interactions [10, 93].

The calculation of stoppings due to ion beam irradiation is performed by the Stopping and Range of Ions in Materials (SRIM) [59]. An effective Binary Collision Approximation (BCA) Monte Carlo code utilized industrially for ion-beam irradiation.

### 2.6.2 Radiation damage event by charged particles in solid materials

Ion beams modify material structure by the interaction of energetic ions with the solid atoms via inter-atomic potentials. Interaction of such creates displacements of the atoms of the solid from their equilibrium positions, leading to damage in the solids.

Energetic ions moving through the target atom collide with the target atom at their stationary positions, this leads to a deflection of the ions from the original path they transverse [93]. There could also be a collision of ions with electron clouds in the solid resulting in energy dissipation during the collisions. The change in direction as a result of the binary collision of the charged ion into a target material is shown in Figure 2.30 [93].

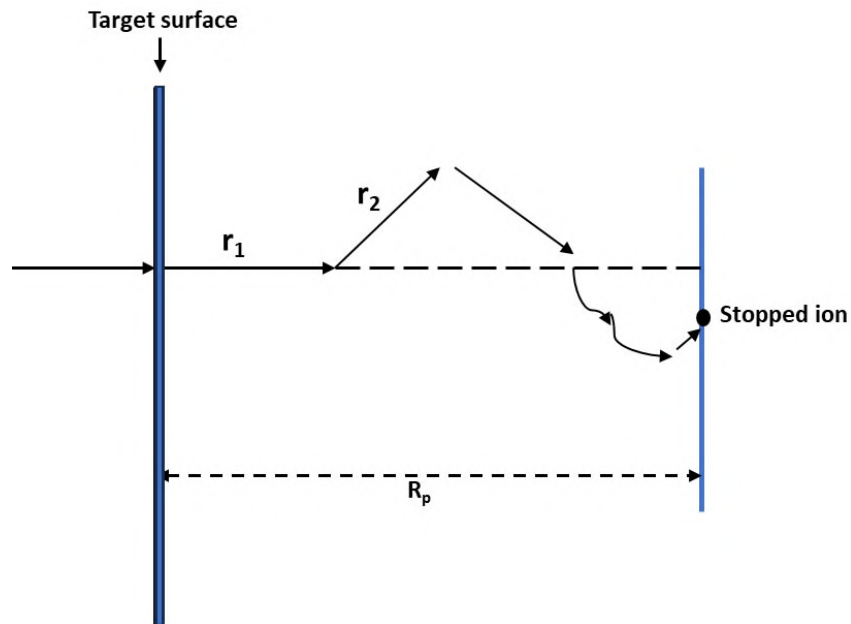


**Figure 2.30:** Incident energetic ion onto a target material with projected range  $R_p$ . The ion creates a Primary Knock-On Atom (PKA) which displaces the ion from the initial path with the mean free paths of  $r_1$  and  $r_2$  as it makes more collisions until it comes to rest

The mechanism of damage due to irradiation of ion species ( $\text{H}^+$ ,  $\text{He}^{2+}$ ,  $\text{Au}^+$ ) to solid matter begins with a transfer of energy from an energetic incident into an atom at equilibrium state in the crystal lattice, the initial recoil atom called the Primary Knock-On Atom (PKA) displaced from its equilibrium position with initial kinetic energy  $E_{PKA}$ . The PKA makes further collisions with a couple of other atoms, this creates vacancies where the atoms were displaced. The displaced atoms shove in between other atoms in the lattice structure called interstitial formation. This disrupts the original crystal lattice structure of the solid.

Some of the energy is lost during a collision with electron clouds of the target atom by electronic excitation, while the PKA energy that is left is transferred to other secondary atoms of the target called Secondary Knock-On Atoms (SKA) or recoil atoms. This energy which is capable of causing displacement damage in the material is called the damage energy ( $T_{dam}$ ) [41, 47]. Also, the recoil atoms produce a cascade of displacements in the whole target material by energy transfers. This cascade process is sustained until the energy of atoms is insufficient to create further knock-on atoms resulting in the end of the damage event.

The mechanism of damage creation in charged ions and neutrons is similar except that for neutrons there is no collision with electron clouds since they are neutral particles, they interact primarily by knocking the atoms of the materials. Ion beams like protons travel through a material and deposit all their energies at the Bragg's peak, unlike the neutrons that penetrate matter deeply depending on their kinetic energies.

### **2.6.3 Comparison of radiation damage defects between protons and neutrons**

Neutron irradiation also induces the production of photons and heat deposition by neutrons and gammas, all of which contribute to radiation defects in the microstructure of materials. The evolution and diffusion of Frenkel Pairs, forming

higher defects in the microstructure over time, result in observable defects in the bulk morphology of structural materials [9]. While neutrons are valuable for radiation damage studies, there are challenges in their usage for material damage studies.

Sample irradiation with neutrons requires special facilities like fission reactors and cyclotrons, which are not easily accessible. Doses of radiation achievable within a few hours with proton irradiation might take days with neutron irradiation in a reactor depending on the fluence [41, 42].

Another factor that sometimes limits the use of neutron irradiation studies is the activation of samples requiring extended cooling times before handling. Proton irradiation, or ion-beam irradiation in general, is relatively cost-effective [41–43, 101]. Working with ion beams allows for systematic control of specific irradiation conditions, making radiation damage studies more dynamic than using neutrons [44].

Neutrons and protons exhibit similar irradiation profile characteristics, an important feature for comparing both radiations. Neutrons have a neutral charge state and are highly penetrative when solid materials are irradiated, resulting in a spatially flat damage profile to depths of several millimeters [41]. Protons, on the other hand, produce a flat damage profile leading to a dose variation with energy and they deposit all their energy at the end of their path, known as the Bragg’s peak.

There is a variation in the radiation damage mechanism of protons and neutrons, however, several studies have shown growing evidence that protons irradiation produces effectively similar radiation damage effects just as neutrons on the microstructure of [41, 42, 44, 46–48], including the formation of dislocation loops, precipitates, voids/bubbles, radiation-induced segregation, stacking faults, and amorphization.

Was et al. [45] demonstrated that proton ions can serve as viable alternatives to neutron irradiation in certain stainless steel alloys, as depicted in Figure 2.31. In their study, microstructural characteristics such as dislocation loop size and density were quantified as a function of dpa,

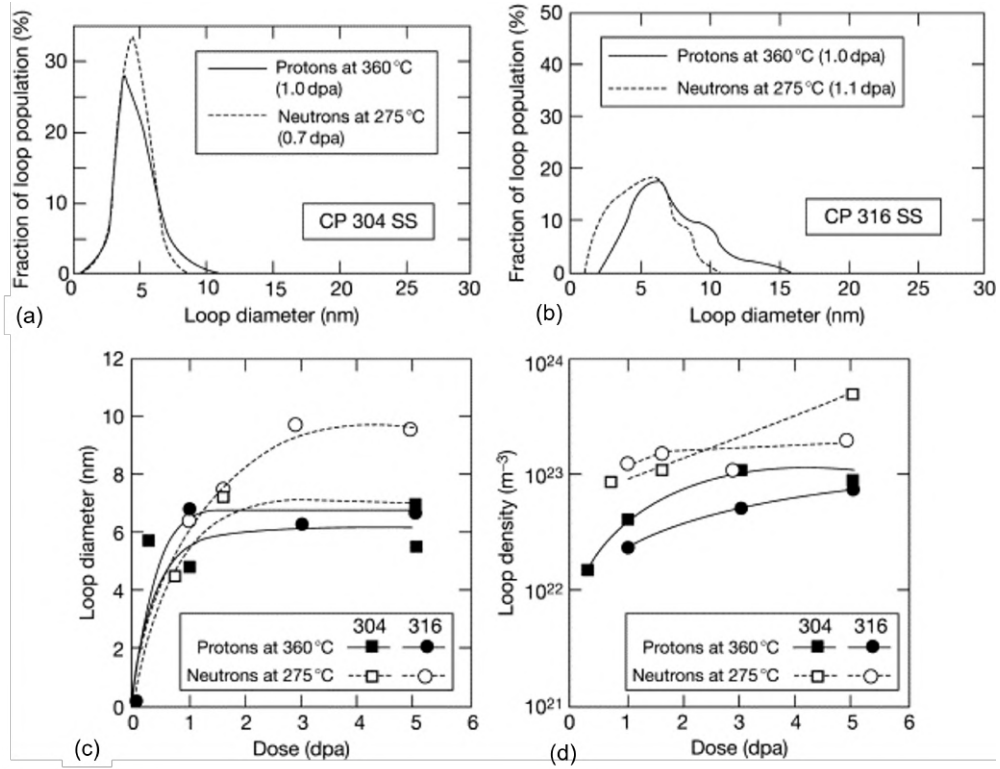


**Figure 2.31:** Comparison of proton and neutron irradiation and microstructural features in commercial purity 304 and 316 stainless steel alloys with (a) and (b) showing fractional loop population-diameter profile of protons closely approximating that of neutrons in CP 304 and 316 SS respectively. (c) and (d) represents the loop diameter and density vs Dose (dpa) profile of protons aligning closely with those of neutrons [45]

showcasing the effectiveness of proton irradiation. However, other studies have also highlighted the differences between proton and neutron irradiation. Zinkle & Busby [102] found that neutron irradiation in fusion reactors causes more severe swelling and embrittlement of structural materials compared to proton irradiation due to the higher production of helium and hydrogen. Bohanon et al. [103] observed that at higher fluences ( $\geq 1 \times 10^{20} \text{ n.cm}^{-2}.\text{s}^{-1}$ ), neutron irradiation leads to more pronounced changes in the mechanical properties of structural materials. This includes greater dislocation densities ( $\gg 10^{22} \text{ m}^{-3}$ ) observed with neutrons of lower fluences compared to the dislocation densities obtained with proton irradiation ( $\sim 10^{22} \text{ m}^{-3}$ ) even at higher fluences [104–107].

Haley et al. [108] demonstrated that 2 MeV fission neutron irradiation of Fe9Cr produced similar dislocation densities and sizes ( $\sim 5 \text{ nm}$ ) up to  $10^{22} \text{ m}^{-3}$  with 1.2

MeV protons at similar temperatures and doses ( $\sim 300^\circ$  and 2.0 dpa) respectively. The neutron and proton irradiated alloy Fe9Cr had a large number of helical dislocations close to defect clusters. There was however variation in the number density and diameter of voids formed with neutron and proton irradiation. Significant differences were also observed in the Chromium cluster features formed with the two irradiation species.

Furthermore, Was et al. [42] discussed the challenges of ion irradiation and found that transmutation reaction in neutron irradiation caused microstructural evolution, especially for voids largely due to the higher levels of helium and hydrogen production [24]. These findings highlight that careful proton irradiation can serve as a feasible alternative for reactor (neutron) irradiation in scenarios where neutron irradiation is impractical due to the aforementioned factors.

## 2.7 Solid materials and their properties

Solid Materials exist in nature either as crystalline or amorphous. In the crystalline form, there is a regular and repetitive structure of particles throughout the entire material referred to as a lattice, there are also equal inter-molecular forces acting on the particles and these have a definite melting point. Whereas, amorphous solids have an undefined geometry of the particles and unequal distribution of inter-molecular forces as illustrated in Figure 2.32.

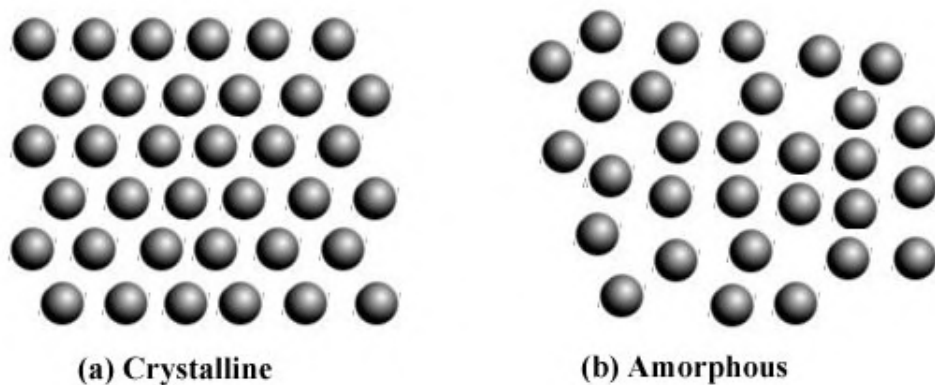


**Figure 2.32:** Illustrating arrangement of atoms in Crystalline materials with a regular repeated pattern (lattice) and Amorphous Solids with an irregular arrangement of atoms

A crystalline material may sometimes lose its regular arrangement to an amorphous nature when impacted by strong stress or radiation [9]. Crystalline materials of metals, semiconductors, alloys, and ceramics have been widely utilized in the nuclear reactor environment and other harsh-radiation environments. Therefore, an understanding of the crystal structure and the behaviour of materials in a radiation environment is key to the study of nuclear materials and their applications.

A single crystal consists of an array of many atoms (whose smallest unit is the unit cell); while several of these single crystals form a poly-crystalline material at the micro-scale level and the materials visible to us are referred to as the macro-structure. Description of material damage requires also an understanding of the interrelationships of the *processing-structure-property* which serves as a guide to predict reasonably the degree of performance for a material under practical conditions [9].

To ascertain whether or not a material has a level of damage, microstructural features such as grain size, grain boundary distribution, and inter-atomic spacing, phase orientations must be studied to be able to predict effects that will be evident at the macrostructural level over time. The arrangement of crystal structure shown in Table 2.4, provides information about the mechanical properties of materials and consequently has an effect on the material's resilience to damage.

**Table 2.4:** Basic Crystal System and Bravais lattices [9]

| System       | Structure     | Axial Lengths     | Axial angles                                    |
|--------------|---------------|-------------------|-------------------------------------------------|
| Cubic        | 4             | $a = b = c$       | $\alpha = \beta = \gamma = 90^\circ$            |
|              | Simple        |                   |                                                 |
|              | Body-Centered |                   |                                                 |
|              | Face-Centered |                   |                                                 |
| Rhombohedral | Diamond cubic |                   |                                                 |
|              |               | $a = b = c$       | $\alpha = \beta = \gamma \neq 90^\circ$         |
|              |               |                   |                                                 |
|              |               |                   |                                                 |
| Tetragonal   | 2             | $a = b \neq c$    | $\alpha = \beta = \gamma = 90^\circ$            |
|              | Simple        |                   |                                                 |
| Hexagonal    | Body-Centered |                   |                                                 |
|              |               | $a = b \neq c$    | $\alpha = \beta = 90^\circ, \gamma = 120^\circ$ |
| Orthorhombic | 4             | $a \neq b \neq c$ | $\alpha = \beta = \gamma = 90^\circ$            |
|              | Simple        |                   |                                                 |
|              | Base-Centered |                   |                                                 |
|              | Body-Centered |                   |                                                 |
| Monoclinic   | Face-Centered |                   |                                                 |
|              | 2             | $a \neq b \neq c$ | $\alpha = \beta = \gamma = 90^\circ$            |
|              | Simple        |                   |                                                 |
|              | Base-Centered |                   |                                                 |
| Triclinic    |               | $a \neq b \neq c$ | $\alpha \neq \beta \neq \gamma \neq 90^\circ$   |

There are seven basic crystal systems and the fourteen space lattice structures (Bravais) are defined using the arrangement of their unit cells [9]. The above lattice structures are important because there is a relationship between the crystal structure and atomic number density, which is a function of the Kinetic Energy Released in Materials (KERMA) used to estimate the dpa of the cell [10]. The lattice parameter, which is a function of the length of the unit cell ( $a, b, c$ ) and the mutual angles ( $\alpha, \beta, \gamma$ ), as shown in Table 2.4 provides good information about the kind of point defects a crystal may have. An alteration in the crystal's lattice parameter is closely related to the mean strain a lattice would experience from defects [109].

The Displacement per atom (dpa) also depends on the displacement energy for atomic lattice  $E_d$ . Whereas, looking at the elementary displacement theory  $E_d$  is a function of direction  $\langle 100 \rangle$ ,  $\langle 110 \rangle$  and  $\langle 111 \rangle$  as illustrated in Figure 2.33 for face-centered cubic crystals of Copper(Cu) and Gold (Au).

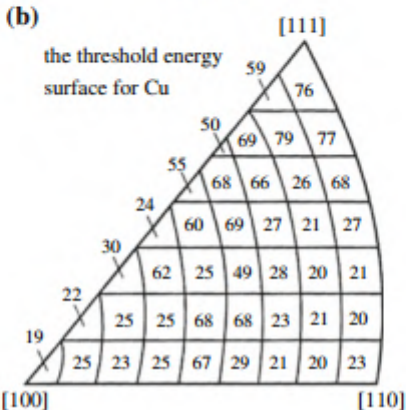


**Figure 2.33:** Dependence of displacement energy lattice direction in (a) fcc Cu and Au crystal and (b) in copper ( $[10, 110, 111]$ )

The force constant which forms the equation for displacement energy in any direction is given by:

$$k = \frac{3\nu}{a^2\beta}, \quad (2.39)$$

where  $k$  is the force constant,  $a$  is the lattice constant  $\nu$  ( $a^3/4$ ) is the specific volume of an atom and  $\beta$  is the compressibility. The threshold displacement in the  $\langle 100 \rangle$  direction is given by [10]:

$$E_d = 8U + 2(ka^2) \left( \frac{1}{\sqrt{2}} - \frac{1}{2} \right)^2, \quad (2.40)$$

where  $U$  is the energy per atom for the crystalline solid. The equation shows how the lattice parameter is dependent on the displacement energy (displacement threshold) of the atom. The formation of Frenkel pairs (vacancies and Interstitial) due to irradiation often results in a fractional change in the lattice parameter, and

how significant the change will be, depends on the type of crystal structure. Hence, the effect of radiation damage on atoms of a cubic structure will be different from that on a trigonal-structured solid.

### 2.7.1 Crystal diffraction

The phenomenon of diffraction is very useful in crystallographic studies. Most non-invasive techniques such as Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), and X-Ray diffraction, developed to study the microstructure and nanostructure utilize the principle of diffraction to qualitatively and quantitatively extract microstructural properties. The term microstructure covers all properties within the scale of structural phenomena including the grain and particle size, dislocation density, particle volume fraction, microcracking, and microporosity while nanostructure is confined to the sub-micrometer systems scales with characteristic features including crystal interfaces, point defects, and defect clusters, their nucleation, and precipitations [112]. Generally, such structural features approaching the scale of  $10^{-7}$  m and below are within the range of nanostructures.

A perfect (pristine) crystal lattice interacts elastically with radiation of wavelengths comparable to the lattice spacing of the material to high-intensity points of constructive and low-intensity points of destructive interference as it passes through the crystal lattice. These high and low points when plotted as a function of the scattering angle yield the diffraction pattern of the crystalline lattice called the diffraction spectrum. The diffraction angles and the intensities of the peak carry sensitive information about the crystal structure relevant for crystal studies [112].

Pure crystalline materials are identified when the diffraction patterns they produce match existing standard diffraction patterns of the material in some databases of material studies. When a crystal lattice has imperfections (defects), its diffraction pattern produces slight deviations from the normal peak intensities and angular peak positions. This data when processed contains the microstructural features that characterize the materials. In material damage studies we use these crystallo-

graphic diffraction patterns also to detect estimated features that are a pointer to crystal defects in the material.

Crystal diffraction studies require radiations whose wavelengths are comparable with or less than the interatomic spacing in the material. radiations frequently used for diffraction studies include X-Rays, energetic electrons, and neutrons [10, 112, 113].

To study the diffraction patterns in materials using any of the microstructural characterization techniques, Bragg's law which is a special interpretation of the Laue diffraction equation provides useful information about the orientation of single and polycrystalline materials. Bragg's law introduces a convenient way of relating geometrically, the angular distribution of the intensities of the peaks in a diffraction pattern of a regular crystalline lattice as illustrated in Figure 2.34.

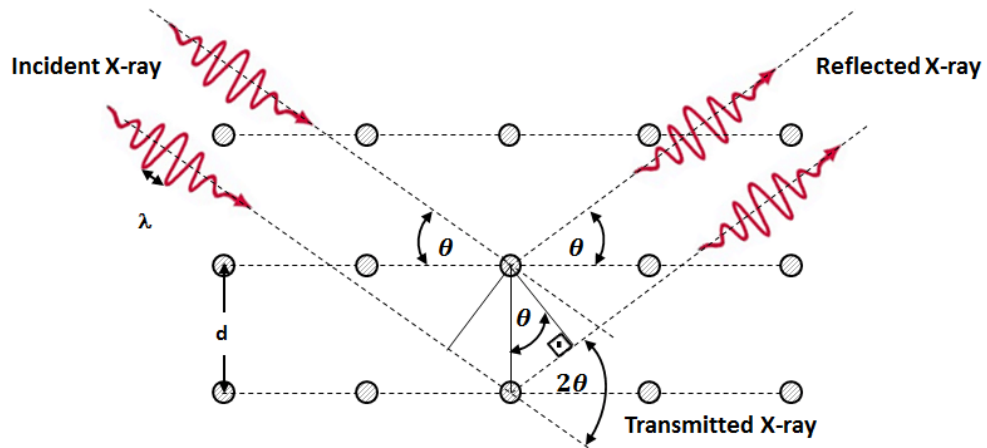


**Figure 2.34:** Diffraction in a crystalline material of lattice spacing  $d$  ([114])

From Figure 2.34 we can relate the wavelength of the radiation to the spacing and diffraction angle in Bragg's equation given Eq. 2.41:

$$m\lambda = 2d\sin\theta, \quad (2.41)$$

where  $m$  is the order of the diffraction peak,  $\lambda$  is the wavelength of the radiation,  $d$  is the spacing between lattice planes, sometimes written as  $d_{hkl}$  because it is a

function of the Miller indices of the plane, and  $\theta$  is the diffraction angle.

$$d_{hkl} = \frac{\lambda}{2\sin\theta}, \quad (2.42)$$

The  $d$  relates to the volume  $V = abc$  of the unit cell and the Miller indices ( $hkl$ ) as:

$$\frac{1}{d^2} = \frac{h^2}{a^2} + \frac{k^2}{b^2} + \frac{l^2}{c^2}, \quad (2.43)$$

For a cubic crystal lattice where  $\alpha = \beta = \gamma = 90^\circ$ . Eq. 2.43 becomes:

$$d = \frac{a}{\sqrt{h^2 + k^2 + l^2}} = \frac{a}{\sqrt{s}}, \quad (2.44)$$

where  $s = h^2 + k^2 + l^2$  must be an integer. Now equating Eq. 2.42 to 2.44, Eq. 2.45 is obtained thus:

$$d = \frac{\sin^2\theta}{s} = \frac{\lambda^2}{4a^2}, \quad (2.45)$$

where  $a$  is the lattice constant. For a given pattern, there exists a set of integers for which  $\frac{\lambda^2}{4a^2}$  yields a constant quotient for all diffraction angles in the pattern. With this, a pattern can be linked to a particular crystal structure. Each crystal structure possesses a fixed number of possible reflections. The set of allowable integers for the simple, body-centered, face-centered, and complex cubic system like diamond cubic is summarized in Table2.5

**Table 2.5:** Integral factors (s) for cubic crystal systems [112]

| Cubic system              | Integers (s)               |
|---------------------------|----------------------------|
| Simple cubic (SC)         | 1, 2, 3, 4, 5, 6, 7, 8,... |
| Body-centered cubic (BCC) | 2, 4, 6, 8, 10, 12,...     |
| Face-centered cubic (FCC) | 3, 4, 8, 11, 12, 16,...    |
| Diamond cubic (DC)        | 3, 8, 11, 16, 19, 24,...   |

The above factors are useful for the identification of crystal lattice systems when a diffraction pattern is obtained from X-Ray diffraction (XRD) data. Diffraction patterns obtained from XRD can also be used to determine microstructural features relating to damage. Such parameters include the interatomic spacing ( $d$ ), lattice

parameter ( $a$ ), and dislocation density ( $\delta$ ) [63, 113, 115].

## 2.7.2 Crystal defects

In nature, crystals are scarcely perfect even without impacting particulate or photon radiation on the material. Departure from the ideal nature of crystal structure may, however, give room for more sensitive property changes. A collection of abnormal lattice orientations and patterns, a deviation from the ideal structure is called crystal or lattice defect [9].

An increase in lattice vibrations (phonons) increases the local temperature of an atom in the lattice site, influencing changes in important physical properties like thermal conductivity, electrical conductivity, or resistivity. When the crystals absorb more radiation, there is a change in the nature of the bulk material as the phonon production (lattice vibrations) increases.

The various crystal defects that can be observed as a result of these processes include *point defects* (i.e., creation of vacancies, self-interstitials, substitutional or interstitial impurity atoms as shown in Figure 2.35), these are also referred to as 0-dimensional defects [116].

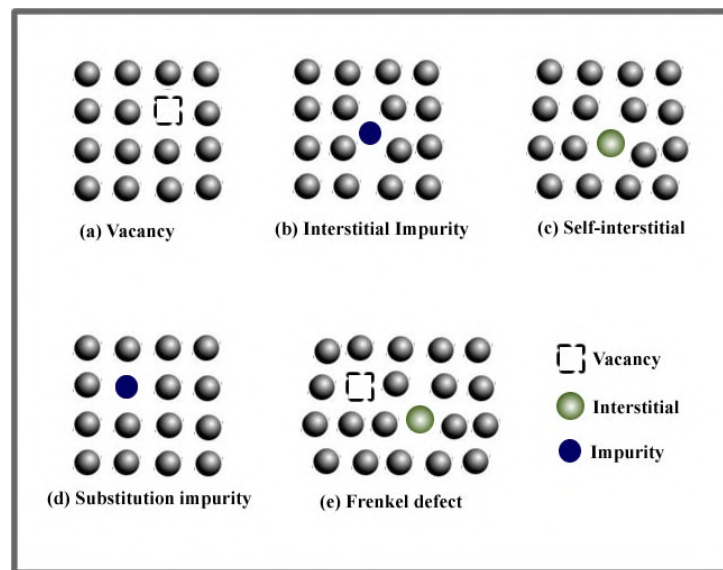


**Figure 2.35:** Types of point defects in a crystalline material showing (a) vacancy, (b) interstitial impurity (c) self-interstitial (d) Substitution impurity, and (e) Frenkel defect which consists of a vacancy and interstitial

There could also be the creation of *surface defects* that occur at grain boundaries, and line defects (dislocations, see Figure 2.36). Surface defects are mostly 1-dimensional and 2-dimensional defects, for instance, dislocation lines, and dislocation loops respectively. Another type of crystal defect that relates to the whole volume of the crystal is called the *bulk defect*. These are defects like voids, cavities, stacking-fault tetrahedra (SFT), and precipitates (see Figure 2.37). These types of defects are also referred to as 3-dimensional

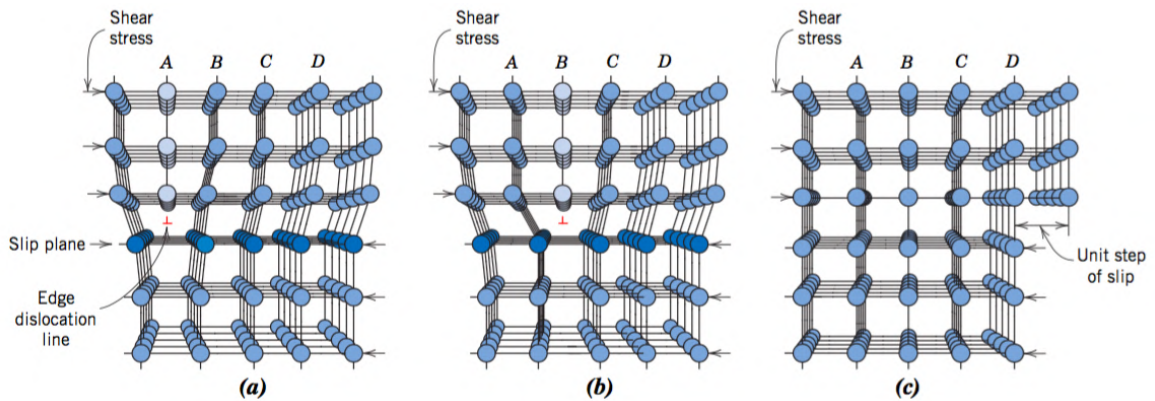


**Figure 2.36:** Formation of line defects (a) An edge dislocation motion line at the slip plane (b) Dislocation motion at extra half-plane (c) Dislocation motion due to shear stress applied in unit step of slip ([117])

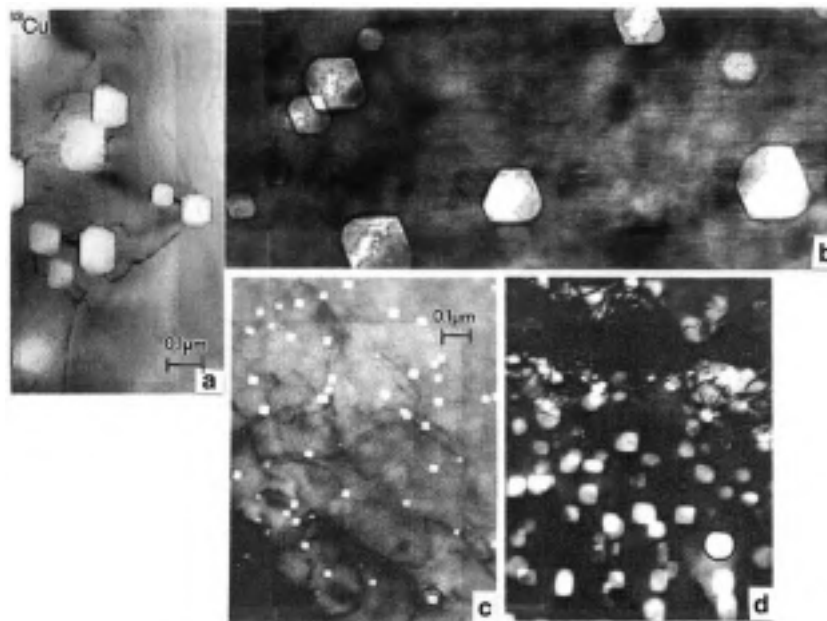


**Figure 2.37:** Transmission Electron Microscope (TEM) images showing Formation of volume defects-voids (white patches) in a material ([118])

## 2.8 Radiation damage and effects

Advancement of nuclear systems for future generations, both fission and fusion provides increased safety, better fuel efficiency, and reduced waste production. However, it poses new challenges such as higher operating temperatures and radiation doses [8, 64]. An understanding of radiation damage due to these high doses and thermal agitations, their evolution over time, and a reliable prediction of material's degradation or resilience to radiation damage will therefore help improve reactor lifetimes, economy, and reduced environmental burdens. Apart from the reactor systems, there has been a concern about radiation-tolerant materials in energetic particles-impacted radiation environments [32]. There is a need to evaluate materials based on their resilience to withstand harsh radiation environments. Hence, in this section, efforts have been made to explain radiation damage events, radiation effects on crystalline materials, and baseline principles governing the evaluation of radiation damage on materials. These concepts form the foundation upon which most of the evaluations in the work are based.

### 2.8.1 Radiation damage event

The radiation damage event is envisaged as a process in which energetic radiation collides with an initial atom in a target material. The impacted atom is referred to as the Primary Knock-On Atom (PKA). This atom is then displaced by the kinetic energy acquired by it from the incident radiation [8, 10]. If the kinetic energy is sufficient, it can lead to further collisions of the PKA with neighbouring atoms. The secondary and subsequent collision events are referred to as a collision cascade, which may result in several processes that lead to radiation damage [9, 10]. The end damage due to material irradiation can be in the form of the following:

- (a) The creation and development of faulted dislocation loops and defect clusters.
- (b) Production of Impurities: is the transition of nuclei into other nuclei which may be radioactive; neutrons produce this process via nuclear fission and capture.
- (c) Atomic Displacement: atoms in the structure of a material can be displaced

from their original positions; displaced atoms may produce lattice vacancies and interstitials called Frenkel pairs or cause a swap of dissimilar atoms in the lattice space. Long-term accumulation may lead to structural mechanical defects or damage in a material [8, 119].

The key sequence of radiation damage constitutes different processes, which include:

- (i) The interaction of a neutron(or photon) with sufficient incident energy with an atom in the material at the lattice site.
- (ii) kinetic energy is transferred from the incident particle on the atom of the material which may give rise to a PKA. For instance, the average PKA energy for neutrons displacing ions in Au-197 and Al-27 is  $\sim 20$  and  $\sim 113$  keV (calculated) respectively.
- (iii) The affected atom is displaced from its site given sufficient energy.
- (iv) There could be movement of the displaced atom around the lattice space, with the production of more atomic displacements (usually this is possible with an energetic neutron).
- (v) This cascade of displacement events sets in point defects created by the original primary knock-on atoms.
- (vi) Stoppage of PKAs motion leaves behind a Frenkel pair (vacancy and interstitial)

The end of the damage event occurs when the PKA and all other atoms are brought to rest. The surviving damage after the whole event is referred to as the *primary radiation damage* [119]. The average number of atoms displaced in the whole process depends largely on the energy of the incident particle, the neutron in the reactor setting where this study is focused and the neutron interaction cross-section possible for this energy transfer to occur between the neutron and the material's lattice atoms and the displacements created by the PKAs.

The consequences of neutron interaction with materials have been studied widely [4, 10, 46, 120] on materials such as metals, semiconductors, and other alloys. The

result of the radiation damage event produces a collection of point defects (vacancies and interstitials) and an accumulation of these defects in crystal lattices which is approximately  $10^{-11}s$ .

The timescale for the evolution can be classified into an initial atomic displacement which occurs in *femtoseconds*, termination of collision sequence when all atoms come to rest in *pico-seconds*, and a long period of thermal relaxation and possible annealing of damage which span from *nano-seconds to years*. The detailed time scale of events is shown in Table 2.6 subsequent events may involve the movement of these defects (point and cluster) or dissolution of clusters is categorized as radiation damage effects [10].

**Table 2.6:** Timescale for the formation of defects in irradiated crystalline solids[10]

| Time(s)    | Event                                                        | Result                                                                                   |
|------------|--------------------------------------------------------------|------------------------------------------------------------------------------------------|
| $10^{-18}$ | Energy transfer from the incident particle                   | Creation of a primary knock-on atom (PKA)                                                |
| $10^{-13}$ | Displacement of lattice atoms by the PKA                     | Displacement cascade                                                                     |
| $10^{-11}$ | Energy dissipation, spontaneous recombination and clustering | Formation of Frenkel pairs, single interstitial and defect clusters                      |
| $10^{-8}$  | Defect reaction by thermal migration                         | Single interstitials and vacancy recombination, clustering, trapping and defect emission |

To determine the number of displaced atoms, PKA production can be accounted for in a solid using a differential energy approximation model which is based on the energy-flux dependence of the incident particles and the energy transfer cross-section available for the collision of atoms.

### 2.8.2 Primary radiation damage and formation of PKA

The mechanism of damage creation in materials is categorized into two by time scale: Primary damage is formed just after the incident radiation or particle collides with the material causing an ejection of a PKA as depicted in Figure 2.38. This occurs in a few femto to picoseconds, and the Long-term evolution of damage

that produces bulk morphological changes in the microstructure of the material ranging from nanoseconds to years [4, 5, 8].

Irradiation of crystalline materials with energetic particles or photons may lead to a perturbed state and even re-ordering of the atomic crystal lattice of the material as shown in Figure 2.39.

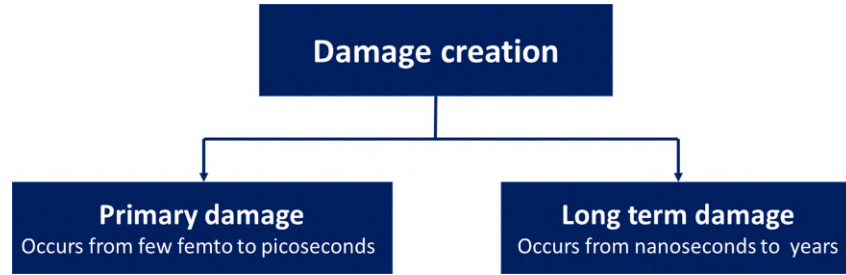


**Figure 2.38:** Description of damage creation in time Scale. Primary radiation (Short-term) damage and Long-term evolution of damage.

Processes of this nature are followed by the production of lattice defects, which affect both the micro and the macrostructural properties of such materials leading to single atom displacement events to bulk morphological changes of the irradiated material over time [32, 73].

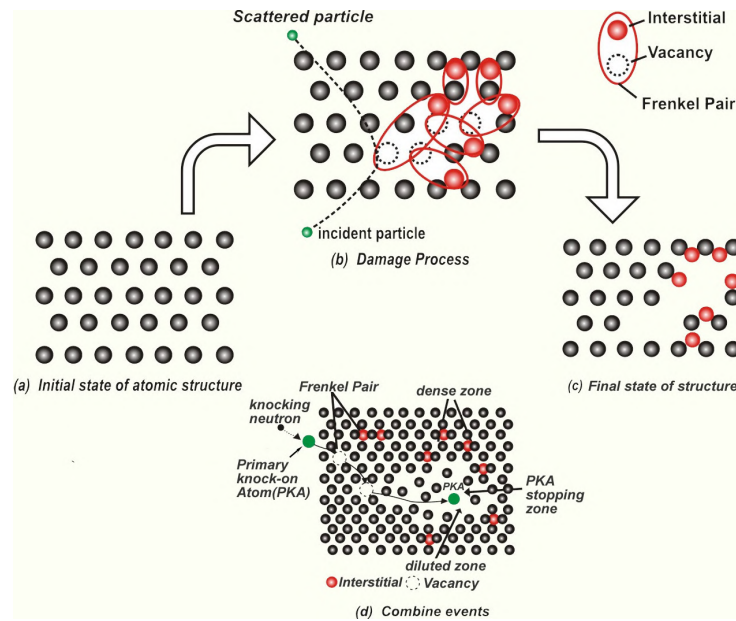


**Figure 2.39:** Radiation damage event showing (a) a pristine crystal structure (b) An incident particle hits the crystal structure creating vacancies and interstitials (c) Deformed state of the crystal (d) combined events (Modified image from Ref.[7, 26])

These changes can be damaging to some materials depending on the intended application for the material. For example, bulk morphological changes due to irradiation in a material can not be appreciated in control rods and fission fuel assembly of a nuclear reactor. However, property changes in irradiated materials could have beneficial consequences where material property modification is required for instance, in the making of special coatings for new materials [73].

An atom displaced out of its lattice position by incident radiation is referred to as a PKA, the energy of which is dependent on the mass and charge (type of PKA) and on the energy of the incident radiation. The formation of the PKA is synonymous with that of the vacancy-interstitial (Frenkel Pair) defect pair production [5, 10]. For there to be a displacement from the lattice site, a minimum energy called the displacement threshold energy ( $E_d$ ) is required. This energy is an average value that is independent of the crystallographic orientation. The mean values of the  $E_d$  are between 20 to 60 eV [10, 121]. This energy attained in lattice atoms is a function of the radiation type. It takes slightly above 1 MeV of electron energy to cause displacement in metallic lattice atoms, whereas for neutrons and ions, slightly above 1 keV and 100 eV, respectively, can initiate displacement in metallic lattice atoms. Transfer of the displacement energy from one atom to another increases lattice vibration [10, 73].

It is worth noting that in the nuclear material damage perspective, microstructural defects produced in materials due to irradiation are termed *radiation damage*, whereas, the accumulation of these defects over time (long-term evolution of damage) resulting in morphological (physical and mechanical) and chemical property changes (i.e. macrostructural changes) are termed as *radiation effects* [5, 9].

## 2.9 Radiation effects

Having examined the primary creation of radiation damage in materials, its consequences will be briefly explored. Radiation effects refer to the alterations in a material or its properties following exposure to a specific type of radiation [9], often

representing the outcome of radiation damage in a material. In general, each type of irradiation—neutrons, protons, electrons, heavy ions, and photons—exhibits common effects on crystalline solids. However, the severity of these effects varies among different types of radiation, depending on their specific characteristics, as detailed in Table 2.3.

Radiation effects in crystalline materials exposed to neutrons or photons are influenced by noteworthy defects. The concentration of defects increases with higher fluence (neutrons or photons per  $cm^2$ ). Additionally, nuclear transmutation, changes in chemical reactivity, increased diffusion, and the potential for equilibrium and non-equilibrium formation of new phases in the crystal contribute to the overall radiation effects [9]. The classification of radiation effects in materials includes:

- (i) Microstructural changes,
- (ii) Mechanical property changes,
- (iii) Physical property changes, and
- (iv) Effects on corrosion properties

The focus of this study is on the microstructural changes and some changes in physical properties since the irradiation time of the material is too small to achieve visible mechanical property changes and corrosion effects.

### **2.9.1 Microstructural changes**

Displacements of atoms from their lattice sites by energetic radiation have far-reaching effects on the microstructural properties of the material irradiated. These changes occur within femto to a few pico-seconds of a damage event. The basic effects that can be observed under high-resolution microscopy include cluster formation, extended defects such as dislocation loops and void swelling, radiation, radiation-induced segregation, and precipitation (dissolution). These effects have a chain consequence on the physical and mechanical properties of materials such as electrical and thermal conductivity degradation, irradiation creeps and radiation hardening [9, 10, 65].

The flow-chart of the damage evolution in microstructure is shown in Figure 2.40

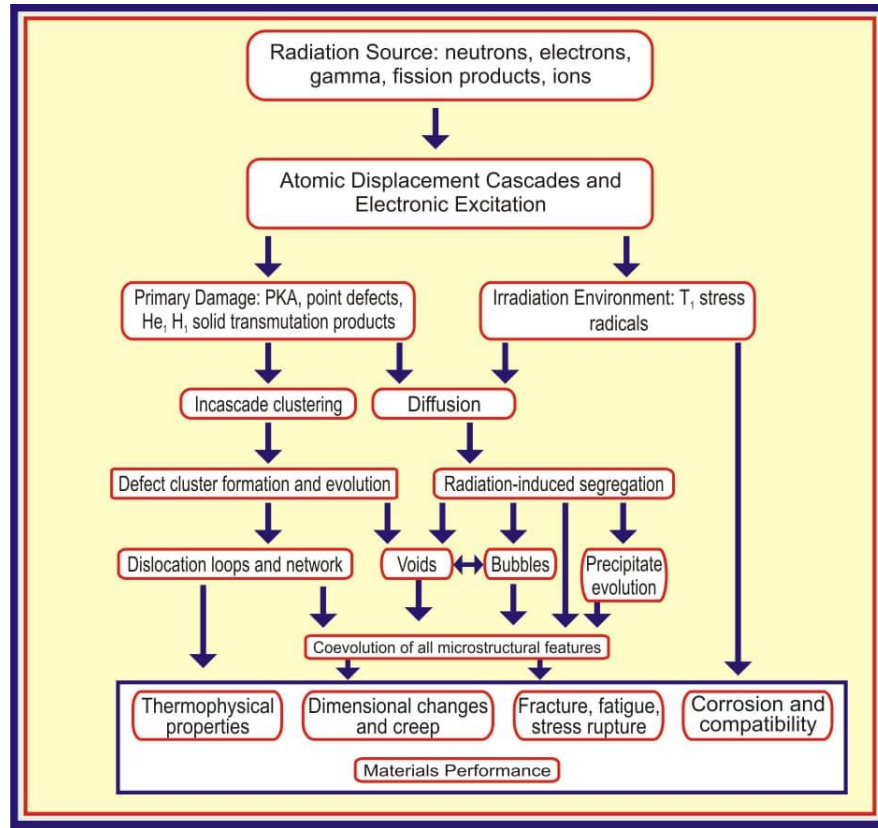


**Figure 2.40:** Radiation-induced microstructural evolution of damage to macrostructural properties in materials ( [9, 122] )

#### (a) Defect cluster formation

Cluster formation is part of the defects produced via intra-cascade recombination [9]. The greater fraction of the interstitials and vacancies produced evolve into clusters. Sometimes, vacancy clusters grow into voids, resulting in changes in microstructural dimensions and also the degradation of the bulk morphological mechanical properties.

Migration of stable states interstitial clusters away from the cascade region and subsequent absorption into dislocations and grain boundaries (which act as sinks) is a phenomenon usually observed in solid materials.[10] Vacancy clusters are unstable and less mobile than interstitial clusters which exhibit a higher level of mobility, hence, migration of clusters is common with interstitial clusters as illustrated in Figure 2.41)

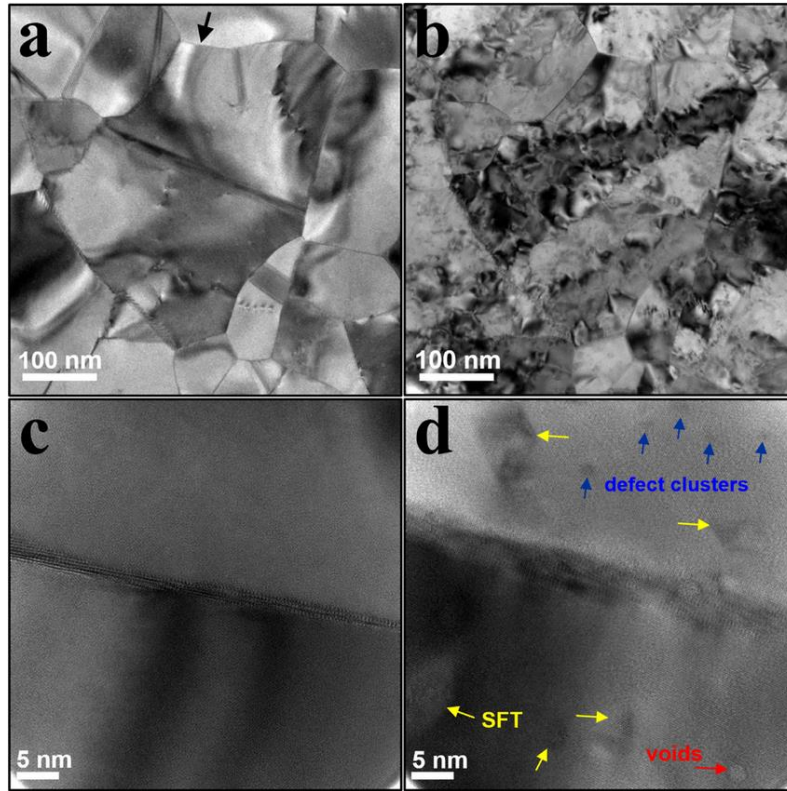


**Figure 2.41:** Evolution of Cluster defects in gold thin-film (a) CTEM Bright-field grain boundary image pre-irradiation (b) CTEM Bright-field grain boundary image of Post-irradiation with ion beam dose of 10dpa (c) pre-irradiation (d) Cluster and Void defects formation after irradiation (taken from Ref. [123] )

### (b) Extended defect formation

Defect formations that exceed 0-dimensional defects (point defects) to higher order defects like 1-D and 2-D (Surface defects), and 3-D (Volume defects) constitute extended defects. Such defects affect the occurrence and dynamics of phase transitions [8, 119]. Defects that do not migrate away from the cascade zones influence the formation of extended defects. These types of defects include nucleation and development of dislocation loops (see Figure 2.4), which results in the concentration of vacancies and interstitial, 3-D defects such as voids/bubble formation shown in Figure 2.42, are also observed.

Dislocation loops are one of the most intense effects of irradiation on the microstructural characteristics of materials since this effect is closely associated with the presence of interstitials. The deformation behaviour of materials is also greatly influenced by dislocation loops and consequently responsible for mechanical property

degradation like ductility, tensile strength, and hardening in irradiated materials [9, 10, 124].

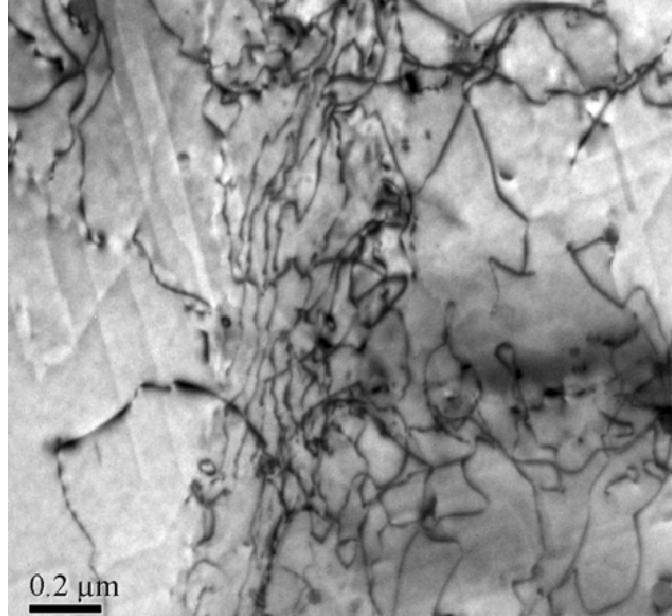


**Figure 2.42:** TEM Bright-field image showing dislocation loops in Aluminium (taken from Ref. [125])

Nucleation of loops is a process in which different types of defect clusters interfere with others resulting in what can be likened to a defect embryo that survives and grows to produce larger defects. Extended defects influence a lot of material properties as mentioned above in the case of void swelling [1, 126].

### (c) Radiation-induced segregation, and precipitation

Alloys of different materials respond to temperature changes by evolving from a homogeneous to a heterogeneous state during irradiation with fast particles thus allowing segregation due to the preferential affinity of certain alloying elements to defect fluxes. This is profound at intermediate homologous temperatures ( $T_m = 0.3 - 0.5$ ), at these temperatures, the defects migrate easily along lower energy sites like grain boundaries and dislocations than defect sinks [9, 126]. The process described above is a non-equilibrium process occurring at point defects during irradiation and is therefore referred to as Radiation-induced Segregation (RIS). This microstructural defect changes greatly the local composition close to the sinks and grain boundary. This substantially impacts the macrostructural properties of the materials in

radiation-prone environments.

Radiation-induced precipitation (RIP) is similar to RIS occurring at intermediate homologous temperatures but unlike RIS that can occur at lower doses, RIP occurs usually at higher doses of order 10 dpa. This creates instability in the phase arrangement leading to the evolution of new phases. Fast neutron irradiation has been shown to produce such defects in materials since they constitute higher doses over time [127]. Also, the formation of RIS influences the subsequent formation of radiation-induced precipitates.

### 2.9.2 Mechanical property changes

The result of radiation damage is usually observed on the bulk material microscopically in different ways. Effects like hardening, embrittlement, toughness reduction, loss of ductility, and irradiation creep are some of the visibly observed mechanical properties after exposure to radiation over time.

Irradiation of metals and their alloys increases yield strength and elongation coupled with a significant decrease in ductility (usually evaluated by uniform elongation or by measuring the total elongation). Hardening is experienced in irradiated metals when many defects are created. Radiation hardening becomes noticeable at radiation doses greater than 0.1 dpa and temperatures less than  $0.4T_m$  ( $< 0.4$  the homologous temperature) [9].

Radiation hardening in face-centered and body-centered cubic structures is enhanced by the formation of Defect clusters, Dislocations (which include dislocation loops and dislocation lines), the formation of voids and bubbles, and also precipitates. The effect of irradiation on the hardening of materials is shown in the Engineering Stress-Strain and Elongation curves depicted in Figure 2.43. Hence, it is glaring that radiation hardening depends so much on microstructural changes in materials. An increase in irradiation dose leads to gradual loss of ductility or an increase in the yield strength of the material for both fcc and bcc crystals.

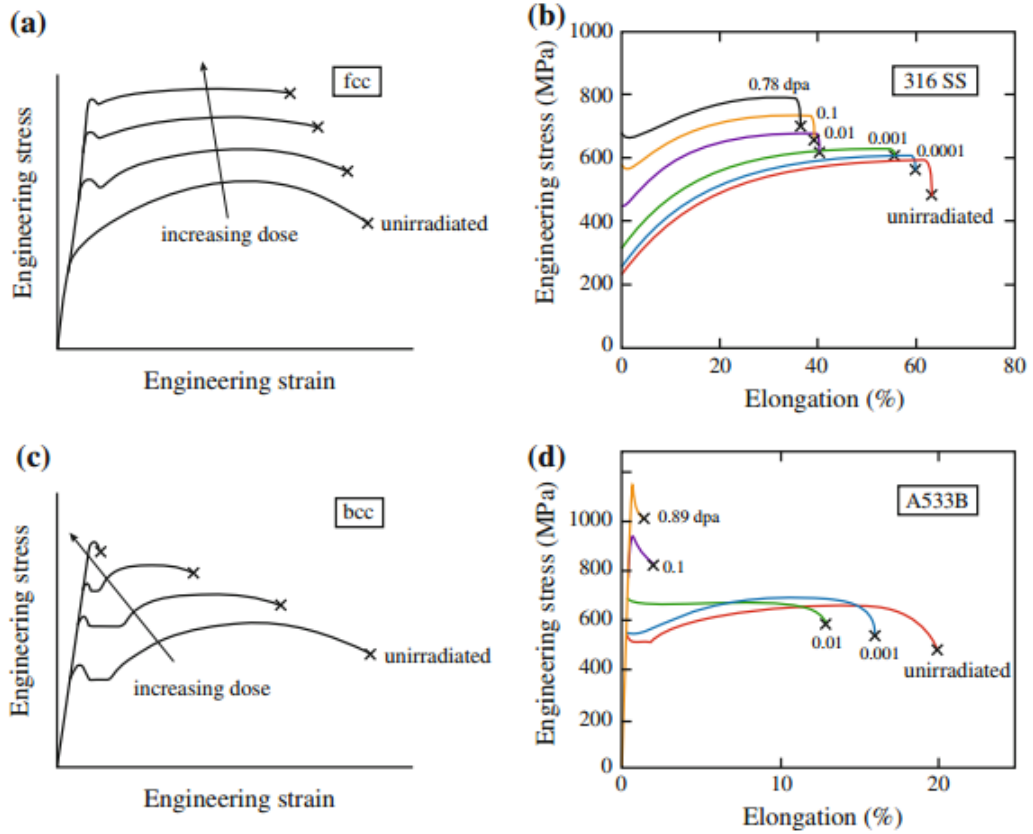


**Figure 2.43:** Stress-Strain behaviour of irradiated face-centered and Body-Centered cubics (a)Schematic diagram (b)Austenitic (fcc) stainless steel, (c) Schematic diagram (d) Ferritic (bcc) steel (taken from Ref. [10])

The level of plastic or creep deformation in a metal or alloy before it finally gets fractured is an indication of embrittlement. Embrittlement of metals is also possible as materials are irradiation, especially by fast neutrons. Radiation embrittlement is a direct consequence of radiation hardening as it reduces the ductility of the metal compared to its unirradiated state [10]. This is common in most materials and even components of nuclear reactors and it is one of the mechanical properties that are of concern in nuclear engineering.

### 2.9.3 Physical property changes

Physical property changes are inevitable as materials are exposed to high-level radiation. Properties such as electrical conductivity/resistivity, thermal conductivity, coefficient of thermal expansion, density, and stiffness of materials are bound to change when such materials are irradiated. In metals, the electrical conductivity

for instance is given by:

$$\sigma = \frac{1}{\rho} = qn\mu, \quad (2.46)$$

where  $q$  is the charge,  $n$  is the number of charge carriers and  $\mu$  is the mobility of the charge carriers. The presence of point defects in a material can result in electron deflection because of the disordered lattice structure that has shortened the mean free path for electron mobility. The drift velocity  $v$  is reduced and consequently the electron carrier mobility  $\mu$  (since carrier mobility is a ratio of drift velocity to electric field  $\mu = v/E$ ). This implies that a decrease in  $\mu$  causes a decrease in the conductivity of the metal. therefore, lattice deformation in metals leads to an increase in electrical resistivity [78]. Hence, in nuclear engineering and space technology, such changes are carefully taken into account since this can affect the overall efficiency of the material for the purpose it was designed to serve.

Irradiation of pure metals with fast neutrons causes a change in their electrical and thermal conductivity. The changes in most cases are more noticeable as the irradiation dose is increased. For copper alloys, the electrical conductivity of the DS copper alloys dropped to about 20 percent at a dose of 16 dpa and then to 40 percent when the dose was increased to about 60 dpa [128]. This also shows a dependence of dose on the physical properties of materials, especially electrical conductivity.

#### **2.9.4 Radiation damage in Gold (Au) and Lutetium-Aluminium (Lu-Al)**

Gold and Lutetium-Aluminium are used for monitoring reactor startups and activation experiments as a benchmark for thermal and fast flux respectively. Au is also being explored for several applications in radiation environments, including uses in cancer radiotherapy treatments and advanced satellite or detector applications. In applications of this nature, there could be interactions with energetic neutrons, photons, protons, and other charged particles which can cause microstructural damage ranging from single atom displacement events and could over time lead to bulk morphological changes [32].

There are existing studies of radiation damage in gold that have reported the formation of Frenkel pairs (vacancies and interstitials). These primary displacement damage defects can accumulate into clusters, dislocations, and three-dimensional (3D) effects like voids and Stacking Faults Tetrahedra (SFT)[15, 16, 129, 130]. Similarly, collision cascades resulting in defect formation such as dislocation loops, SFT, and voids have been reported with proton and ion-beam irradiation [13, 14, 16, 46, 131].

For the Lu-Al binary alloy system, there is no previous experimental work relating to damage to the microstructure. However, many studies have reported radiation damage defects on the microstructure of solid solution-strengthened Al-alloy such as Al-6000 and 7000 series. These studies reported defects such as dislocation loops, line dislocations, precipitates, Radiation-Induced Segregation (RIS), and void swelling [19, 21, 132–136]. These neutron-induced radiation defects could lead to hardening and embrittlement, impacting the alloy’s structural performance. Proton irradiation has also been shown to produce defects on both Al-6000 and 7000 series such as dislocations (loops or linear), voids, formation of Guinier-Preston (GP) zones, and  $\eta$ -phase precipitates [137–139].

These studies have also reported defect number density calculations for both Au and Al and their alloys typically within the same ranges with slight variations between ion beam and neutron irradiations. A recent simulation study with neutrons and protons on the microstructure of Lu-Al yielded dpa values that could suggest microstructural damage of Lu-Al [130].

### **2.9.5 Effect of radiation on the microstructure of materials**

Materials exposed to the irradiation of neutrons or protons from fission, and fusion reactors or accelerators can produce defects to the micro-structure of materials [9, 10, 32, 46]. Irradiation of materials with neutrons in the fission reactors triggers elastic and inelastic reactions, displacing the PKA from their lattice site, which on further collision with neighbouring atoms, creates interstitial-vacancy pairs called Frenkel Pairs (FP) [39, 40].

Neutron irradiation also induces the production of photons and heat deposition by neutrons and gammas all of which constitute radiation defects on the micro-structure ranging from 0-dimensional (points defects) to 3-dimensional defects (voids, cavities, and precipitates). Evolution and diffusion of the Frenkel pairs form higher defects on the micro-structure which may over time translate to significant observable defects in the macro-structure of materials [9]. Similar radiation defects have also been observed for proton irradiation from Ion beam accelerators [14, 41]

## **2.10 Evaluation of radiation damage in materials**

Evaluation of radiation damage in materials can be carried out using theoretical predictions (simulations) and experimental observations via different characterization techniques. Sometimes an evaluation of physical and mechanical property measurements is required to ascertain the effect of the damage on the macro-structure of the material. The simulations are done to predict the level of damage in the materials. These damage predictions may not be as accurate as desired but they give a good approximation of the level of damage in the material to an acceptable level and can be used to correlate the observed damage effects when characterization is done. On the other hand, the characterization method gives an idea of the actual physical damage to the material. In this section, the basic principles surrounding various methods of damage evaluation due to simulations and experimental characterization will be discussed.

### **2.10.1 Damage calculations using simulations**

The simulation of displacement damage is computed using different simulation codes. Prominent among them include Molecular dynamics, Kinetic Monte Carlo (KMC) methods, MCNP, SRIM, FLUKA, PHITS, and GEANT4. The damage calculation is based on the collisions of atoms with the radiation sources and the subsequent production of Frenkel pairs (vacancies and interstitial). The main parametric quantity generally accepted for quantifying radiation damage dose on a material is the displacements per atom (dpa). The dpa is the ratio of the number of atoms dis-

placed in a given volume to the number of material atoms in the same volume for a given fluence [8, 140]. In this study, the focus was on utilizing the Monte Carlo N-Particle code (MCNP) [58] and the Monte Carlo Binary Collision Approximation code SRIM [59]. These codes are specifically designed for simulating neutron and ion-beam interactions with solid materials.

### Displacement-Per-Atom (dpa) models

Radiation damage estimation models in materials have been widely developed by some researchers and also research institutes [8, 141, 142]. Radiation damage calculation requires parameters like:

- (i) the flux spectrum of a neutron as a function of both the Energy (E) of the neutron and the time of irradiation (t),  $\phi(E, t)$ .
- (ii) the displacement cross-section of the atoms for a material  $\sigma_d$  as a function of the neutron energy E,  $\sigma_d(E)$  and
- (iii) Irradiation time of the neutrons  $t_i$  [8, 143].

The displacement per atom (dpa) is defined as the average number of displacements of each lattice atom. Research has revealed that the number of displacements per atom (dpa) is one most useful quantities in estimating damage dose. Kinchin and Pease (K-P) [3] first developed the model for the calculation of dpa in which they assumed a linear dependence between the number of interstitial and vacancies (Frenkel Pairs) formed given an initial PKA this is known as the K-P Model. The number of displaced atoms per neutron under specific conditions can be obtained by expanding the displacement [144] cross-section with regard to energy transfer to PKA and using the Kinchin-Pease (KP) theory [144]. The rate of displacement is given by

$$R = \frac{\text{displacement}}{cm^3 s} = N \int_{E_{min}}^{E_{max}} \Phi(E_i) \sigma_d(E_i) dE_i, \quad (2.47)$$

which yields

$$R = \frac{N\gamma}{4E_d} \int_{E_{\frac{d}{\gamma}}}^{\infty} E_i \Phi(E_i) \sigma_s(E_i) dE_i, \quad (2.48)$$

This gives the model for the rate of displacement  $R$  of atoms as a function of flux  $\Phi$ ,

the threshold displacement energy,  $E_d$ , incident energy  $E_i$ , mass ratio  $\gamma$ , scattering cross-section  $\sigma_s$ , and  $N$  which is the atom density, from KP model.

$$R = N\sigma_s \left( \frac{\gamma E_i}{4E_d} \right) \Phi \quad (2.49)$$

The K-P model was later modified by Norgett, Robinson and Torrens(NRT) [145] to account for some factors and assumptions based on binary collision approximations (BCA). According to the NRT model,  $N_d$ , the number of offsite atoms created by primary knock-on atoms is given by:

$$N_d(T_d) = \begin{cases} 0, & T_d < E_d \\ 1, & E_d < T_d < 2.5E_d \\ \frac{0.8T_d}{2E_d}, & 2.5E_d < T_d < \infty \end{cases} \quad (2.50)$$

In Eq. 2.50,  $T_d$  is the damage energy of the Primary Knock-On Atom given by:

$$T_d(E) = \frac{E}{1 + k_d g(T/E_d)} \quad (2.51)$$

$E_d$  is the threshold atomic displacement energy also referred to as the available energy required to create atomic displacements. Heat spikes at higher energies may lead to major damage re-combinations and atomic mixing effects which are not considered by the NRT model [6]. Even though these effects may be at a nanometric scale, over some time may lead to macroscopic consequences, and therefore there is the search for improved dpa models.

Experiments have shown that the number of defects that survive after recombination during sample irradiation is usually lower than the number of defects predicted by the NRT model. Hence, the NRT model is seen to overestimate the dpa, especially at high energies. the number of defects formed in energetic cascades for metals is over-predicted by 2/3 in the NRT-dpa model. similarly, the atoms involved in atomic mixing (replacement per atom, rpa) are more than about 30 the dpa value [6]. To circumvent this, the relative defect efficiency function  $\xi_{arcdpa}$  was proposed by Nordlund [6]. The defect efficiency factor is a ratio of the actual number of defects present to the number of defects predicted by the NRT model. The athermal

recombination corrected displacement per atom (arc-dpa) model is given by:

$$N_{d,arc dpa}(T_d) = \begin{cases} 0, & T_d < E_d \\ 1, & E_d < T_d < 2.5E_d \\ \frac{0.8T_d}{2E_d} \xi_{arc dpa}(T_d), & 2.5E_d < T_d < \infty \end{cases} \quad (2.52)$$

where  $\xi_{arc dpa}(T_d)$ , is the defect efficiency function given by:

$$\xi_{arc dpa}(T_d) = \frac{1 - c_{arc dpa}}{(2.5E_d)^{b_{arc dpa}}} T_d^{b_{arc dpa}} + c_{arc dpa} \quad (2.53)$$

where  $b_{arc dpa}$  and  $c_{arc dpa}$  are material constants that are determined from molecular dynamics simulations. All other parameters remain the same as in the NRT-dpa model.

Using the above models, the dpa can be calculated. The role of dpa calculations in radiation damage estimations can not be overemphasized. To correlate radiation damage, it is essential to provide a means of comparing the data obtained from different effects observed from different irradiation sources, hence dpa is the first parametric quantity of choice for comparing the radiation damage effects on the material. The dpa offers the basis for calculating other radiation effects on crystalline materials. This dose is also used as a standard value that helps to quantitatively compare the amount of microstructural damage observed by any of the microstructural characterization methods (TEM, XRD, EBSD) on different materials for a given or different radiation source. The dpa is also an important parameter because the irradiation-induced changes in the micro-structure are a function of the dpa [6, 55].

### 2.10.2 Microstructural characterization of damage in materials

Damage at the microstructure in most cases takes a long time to manifest as physical damage on the bulk morphology of a solid metal or alloy. studying radiation-induced defects and the mechanisms of damage accumulation on the microstructure has long been shown to be a good way of understanding damage on the atomistic

level. However, this requires critical observation.

There has been a demand on the part of engineering design, for safety, reliability, and resilience of materials to radiation damage in future reactor design and other applications in harsh radiation environments [8]. Hence, there is a need for the assessment of the crystallographic structures of the materials and this requires characterization. Since the physical eyes can not be used to detect damage to the micro-structure, highly sensitive characterization techniques are required to achieve this [122]. Several characterization techniques have been developed and utilized to measure materials' properties relating to damage both at the microscopic and macroscopic levels. Some of these techniques include:

- Atom Probe Tomography (APT)
- Positron Annihilation Spectroscopy (PAS)
- Rutherford backscattering spectrometry (RBS)
- Resistivity measurements
- Neutron Diffraction (ND)
- Scanning Electron Microscopy (SEM)
- Transmission Electron Microscopy (TEM)
- X-Ray Diffraction (XRD)
- Flash Differential Scanning Calorimetry (F-DSC)

#### **(a) Scanning Electron Microscopy (SEM)**

The high-resolution SEM is used in the Secondary Electron (SE) and Back-Scatter-Electron (BSE) modes to observe microstructural changes induced by radiation using surface morphology, features like irradiation cracks, surface roughness, and delamination which are indicative of radiation damage effects observed. Elemental analysis can also be achieved with SEM using the energy-dispersive mode [122].

Changes in elemental composition or distribution can provide clues about the radiation-induced chemical reactions and phase transformation. Furthermore, crystallographic information can be obtained when the Electron backscatter diffraction (EBSD) detector is used to understand changes in the crystal structure, and orientation focusing on the level of misorientation of the crystals along grain boundaries. With EBSD, quantities like dislocation density can be calculated [63, 112]

### **(b) Transmission Electron Microscopy (TEM)**

Irradiation effects on the microstructure of materials can be observed on the high-resolution from scattered electrons off the defective crystal lattices portions [146]. TEM is a powerful technique for the characterization of radiation damage at nanoscale level which provides detailed information about the microstructure, crystallography, and defects induced by radiation.

In TEM on-zone analysis of radiation damage, microstructural changes such as dislocations, stacking faults, voids, and precipitates are visualized with high resolution [112, 146, 147]. Crystallographic information obtained through electron diffraction allows for the determination of crystal structure, orientation, and lattice parameters, aiding in the identification of radiation-induced changes. Also, energy-dispersive X-ray spectroscopy (EDS) coupled with TEM enables elemental analysis, providing insights into changes in elemental composition or distribution. TEM's high-resolution imaging capabilities allow for the visualization of atomic-scale features and defects, facilitating detailed examination of radiation-induced effects.

Observation of defects with the conventional TEM is sometimes limited by the resolution (what size of defect it can measure). Typically between 1-2 nm [148, 149]. This implies that defect clusters below this size can not be observed with TEM [63].

### **(c) X-Ray diffraction (XRD)**

Diffraction analysis of single and polycrystalline (powdered) crystal structures in XRD is a technique for characterizing radiation damage in materials, particularly in terms of changes to crystal structure and lattice parameters [146]. By analyzing

the diffraction patterns produced when X-rays interact with a material, XRD can provide detailed information about the arrangement of atoms in the crystal lattice. Radiation-induced defects such as vacancies, interstitials, and dislocations can alter the crystal structure and lattice parameters, leading to observable changes in the XRD patterns [63]. Quantitative analysis of peak positions, intensities, and widths in XRD spectra allows researchers to quantify intrinsic microstructural features like microstrain and dislocation density which determines the extent of radiation damage and assess its impact on the material's structural integrity [63, 146, 150]. Furthermore, XRD can be used to study phase transformations and amorphization induced by radiation

#### **(d) Flash Differential Calorimetry (F-DSC)**

Flash Differential Scanning Calorimetry (DSC) is a nanocalorimetry technique for characterizing radiation damage in materials by measuring the changes in thermal properties induced by irradiation [151]. The technique has been used more on ceramic and polymers [152, 153], however, recent research on metallic solids by Hirst et al. [148, 151] has shown that DSC can measure defect densities many times higher than TEM as it explores the smallest amount of defect energy stored (Wigner energy) [154–156] in the irradiated material. Flash DSC provides rapid heating and cooling cycles, allowing for the investigation of dynamic processes such as phase transitions, crystallization, and melting behavior in irradiated materials. Radiation-induced defects and structural changes can affect the material's thermal properties, leading to alterations in the heat flow signals obtained during DSC analysis. By comparing the DSC curves of irradiated and unirradiated samples, the extent of radiation damage can be identified and quantified, including changes in melting temperature, enthalpy, and heat capacity [148, 151]. Flash DSC characterization of radiation damage provides valuable insights into the thermal stability and structural integrity of materials under irradiation, aiding in the assessment of radiation effects and the development of radiation-resistant materials.

Each characterization technique mentioned above is designed to measure specific properties or defects in the microstructure of crystalline solids. Some of these

techniques have undergone upgrades to enhance measurement quality. Notable examples include High-Resolution Transmission Electron Microscopy (HRTEM), Electron Backscatter Diffraction (EBSD), and Small Angle Neutron Scattering (SANS), which serve as improved versions of TEM, SEM, and ND, respectively.

No single characterization method suffices. Even when two techniques are employed to measure a particular defect, there is variation in imaging quality and technique sensitivity [63]. Therefore, it is crucial to choose which properties are intended to be measured to provide a fair representation of the damage to the microstructure. A combination of techniques can also be employed to achieve the desired objective. Table 2.7 summarizes the different properties/defects that can be measured with these characterization techniques.

**Table 2.7:** Characterization techniques for different properties/defects [122, 157]

| S/N | Property/defect                                                                                                                         | Technique(s)                         |
|-----|-----------------------------------------------------------------------------------------------------------------------------------------|--------------------------------------|
| 1   | Bulk Structure- volume changes,<br>cracks 3D-Mapping                                                                                    | XRD, TEM, HRTEM<br>SANS, APT         |
| 2   | Surface topology                                                                                                                        | SEM, TEM                             |
| 3   | impurity states, interstitials, vacancies,<br>dislocations/dislocation loops, voids, clusters<br>bubbles, grain boundaries, segregation | HRTEM, PAS, SEM,<br>EBSD, APT, F-DSC |
| 4   | lattice structure, strain, phase shift,<br>phase orientation,spacing, phase transition                                                  | XRD, SANS, APT, F-DSC                |
| 5   | Crystallite size, grain size                                                                                                            | SEM-EBSD, HRTEM, XRD                 |

The sensitivities of the characterization techniques are important to observe a defect or property on the microstructure. It is always good to adopt a technique with better sensitivity. However, in practice, this is not always the case due to factors of resource availability in the location of the experiment, in such a case the best available technique is utilized. This study explores a combination of the techniques XRD, TEM, SEM, and F-DSC highlighted in blue for the damage characterization on the micro-structure of Au and Lu-Al.

# Chapter 3

## Materials and Methods of Radiation Damage: Simulation, Experiments and Characterizations

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### 3.1 Introduction

This chapter provides an overview of the computational methods employed in Monte Carlo simulations for calculating displacement damage arising from neutron irradiation and simulation methods for ion-induced damage from proton irradiation. The chapter further outlines the experimental techniques for sample irradiation using proton beams and discusses various methods utilized for characterizing microstructural features in the materials.

### 3.2 Computational Methods for radiation damage simulation

Over the years, computational methods have proven to be powerful and indispensable tools for modeling various physical phenomena in the nuclear reactor environment [158, 159]. These methods play a vital role in both reactor design and reactor transport analysis, allowing predictions to be made before conducting experiments. This is crucial as it saves time, money, and, most importantly, ensures the safety of lives through simulating reactor safety analysis using computational methods. These methods can be broadly categorized into deterministic and stochastic (probabilistic) methods, depending on the approach utilized to solve Boltzmann's transport equation.

### 3.2.1 Boltzmann's neutron transport equation

Describing the behaviour of individual neutrons within a nuclear system is a complex task. Nevertheless, with knowledge of neutron fluxes, reaction rates, and neutron cross-sections, it becomes possible to reasonably predict the average behaviour of a population of neutrons in certain nuclear systems, such as a nuclear reactor. The nuclear transport theory serves as the most useful model for describing this average neutron behaviour, relying on the solution to Boltzmann's transport equation [160]. This equation offers the best representation of the transport and distribution of neutral particles, such as neutrons, as they undergo collisions with atoms.

To derive this equation, the following assumptions are made:

- (i) Particles are treated as point-like entities capable of traversing multiple inter-atomic distances between collisions.
- (ii) Particles travel in straight paths between point collisions.
- (iii) The solution to Boltzmann's equation disregards particle-particle interactions. This assumption is valid in cases such as reactors and other applications where the neutron distribution is possible, as the particle density is considered to be smaller than atomic densities and thus negligible.
- (iv) Collisions are considered instantaneous.
- (v) In reactor media, the material properties are assumed to be isotropic in all directions.
- (vi) The properties of nuclei and their composition are known and remain constant over time unless otherwise stated.
- (vii) Only the expected and mean values of the particle density distribution are taken into account.

In view of the assumptions above, relevant terms contributing to the transport equation are discussed.

The *angular neutron density*, denoted as  $n(\vec{r}, E, \hat{\Omega}, t)d\vec{r}dEd\hat{\Omega}$  represents the expected population of neutrons within the volume element  $d\vec{r}$  at the position  $\vec{r}$ ,

within the energy interval  $[E, dE]$ , and in the direction  $[\hat{\Omega}, d\hat{\Omega}]$  at time  $t$ .

The *Angular Neutron Flux* denoted as  $\psi(\vec{r}, E, \hat{\Omega}, t)d\vec{r}dEd\hat{\Omega}$  is the expected neutron flux within the volume element position,  $d\vec{r}$  at position  $\vec{r}$ , the energy interval  $[E, dE]$  in the direction  $[\hat{\Omega}, d\hat{\Omega}]$  at time  $t$ . The neutron density can be related to the neutron flux by the product of the neutron density  $n$  and the velocity of neutrons  $v$ , as shown in Eq. 3.1 [160]:

$$\psi(\vec{r}, E, \hat{\Omega}, t)d\vec{r}dEd\hat{\Omega} = v(E).n(\vec{r}, E, \hat{\Omega}, t)d\vec{r}dEd\hat{\Omega}. \quad (3.1)$$

The *Angular current density*  $j$  becomes important as it connects the specific direction  $\hat{\Omega}$  with the angular flux  $\psi$  as shown in Eq. 3.2 [160]

$$j(\vec{r}, E, \hat{\Omega}, t)d\vec{r}dEd\hat{\Omega} = \hat{\Omega}.\psi(\vec{r}, E, \hat{\Omega}, t)d\vec{r}dEd\hat{\Omega} \quad (3.2)$$

The scalar product,  $j.dA$  is the net rate of neutron passage through the surface  $dA$ . The projection of the coordinates of  $u$  and the components the solid angle  $\hat{\Omega}$  for a neutron in space along  $\vec{r}$  is shown in Figure 3.1

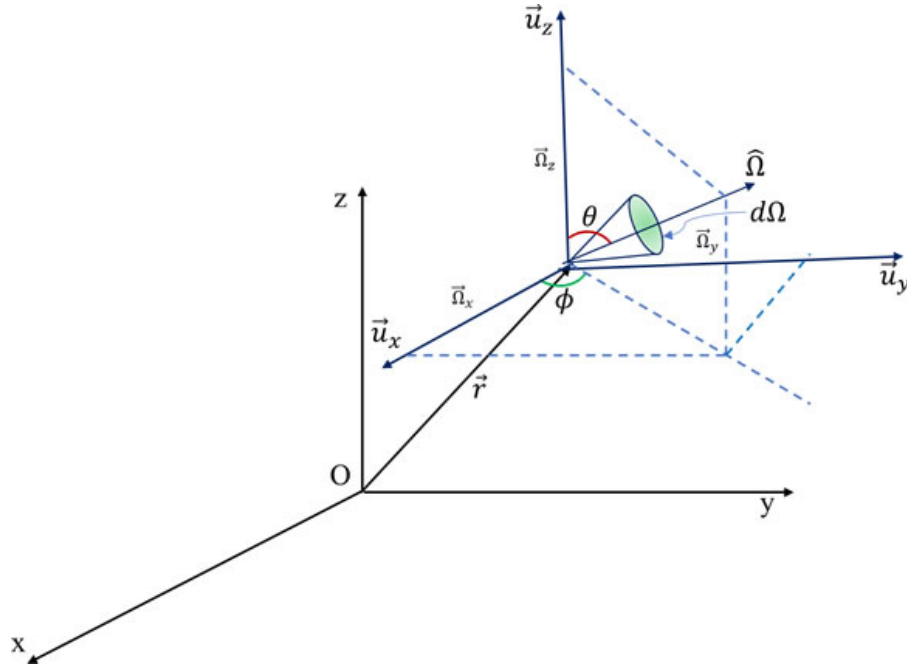


**Figure 3.1:** Neutrons phase space showing the coordinates of  $u$

where the solid angle  $\hat{\Omega}$  in the different directions is given by Eq. 3.3:

$$\hat{\Omega} = \cos \theta \vec{u}_x + \sin \theta \cos \phi \vec{u}_y + \sin \theta \sin \phi \vec{u}_z \quad (3.3)$$

Integration of the quantities, angular neutron density ( $n$ ), angular flux density ( $\psi$ ), and angular current density ( $j$ ) over different ranges of solid angles will yield the total neutron density ( $N$ ), scalar neutron flux ( $\phi$ ) and neutron current density ( $J$ ) shown in Eqs. 3.4, 4.1 and 3.6 respectively.

$$N(\vec{r}, E, t) = \int_{4\pi} \hat{\Omega} \cdot n(\vec{r}, E, \hat{\Omega}, t) \quad (3.4)$$

$$\phi(\vec{r}, E, t) = \int_{4\pi} \hat{\Omega} \cdot \psi(\vec{r}, E, \hat{\Omega}, t) \quad (3.5)$$

$$J(\vec{r}, E, t) = \int_{4\pi} \hat{\Omega} \cdot j(\vec{r}, E, \hat{\Omega}, t) \quad (3.6)$$

The Boltzmann's neutron transport equation describes the balance in some nuclear systems, where the time rate of change of the neutron population in space is equal to the addition of all sources of neutron gains and losses. Thus:

$$\frac{\partial}{\partial t} [n(\vec{r}, E, \hat{\Omega}, t) d\vec{r} dE d\hat{\Omega}] = \text{Neutron gains} - \text{Neutron Losses}. \quad (3.7)$$

By dividing Eq. 3.7 by the neutron velocity  $v(E)$  and denoting the time rate of change as  $(t_{rn})$ , we obtain:

$$\frac{\partial}{\partial t} [\psi(\vec{r}, E, \hat{\Omega}, t) d\vec{r} dE d\hat{\Omega}] = \text{Neutron gains} - \text{Neutron Losses}. \quad (3.8)$$

By moving the neutron losses term to the left-hand side of the equation and expressing the six (6) individual contributing terms, which are the time rate ( $t_{rn}$ ), neutron leakage ( $n_l$ ), neutron absorption ( $n_a$ ), neutron scatter source ( $n_{ss}$ ), fission neutron source ( $f_{ns}$ ) and the independent neutron source ( $I_{ns}$ ), we have the balance in the terms represented by Eq. 3.9

$$t_{rn} + n_l + n_a = n_{ss} + f_{ns} + I_{ns}. \quad (3.9)$$

The neutron leakage rate ( $n_l$ ) depends on the net current flowing out of some area  $dA$  enclosing the volume. The absorption or collisional term ( $n_a$ ) suggests that each collision has the potential to remove a neutron from a phase space. In cases where absorption is not possible, there is a change in direction that displaces the neutron into a different phase space. The term  $n_{ss}$  describes the rate at which neutrons from all other energies  $E'$  and directions  $\Omega'$  scatter in free space. The  $f_{ns}$  term represents the source of neutrons emitted from fission neutrons in the systems.

Multiplying all the integrals over the number of fission reactions, fission neutrons emitted in the region  $d\vec{r}$ , and the fission neutron production rate yields the expected number of neutrons born with energy  $E$  and direction  $\Omega'$ . Additionally, there may exist a source of neutrons independent of the system's neutron distribution, such as a beam of neutrons instantaneously incident on the aggregate fissile material. These independent neutron sources are accounted for by the  $q_{ex}$  function. Combining all these terms gives us Eq. 3.10.

$$\begin{aligned} & \left[ \frac{1}{v} \frac{\partial}{\partial t} + \hat{\Omega} \cdot \vec{\nabla} + \Sigma_t(\vec{r}, E) \right] \psi(\vec{r}, E, \hat{\Omega}, t) = \\ & \int_0^\infty dE' \int_{4\pi} d\hat{\Omega}' \left[ \Sigma_s(\vec{r}, \hat{\Omega}' \rightarrow \hat{\Omega}, E', t) \psi(\vec{r}, E, \hat{\Omega}, t) \right] + \\ & v\chi(E) \int_0^\infty dE' \Sigma_f(\vec{r}, E') \int_{4\pi} d\hat{\Omega}' \psi(\vec{r}, \hat{\Omega}', E', t) + q_{ex}(\vec{r}, E, \hat{\Omega}, t). \end{aligned} \quad (3.10)$$

Eq. 3.10 is the time-dependent Boltzmann's neutron transport equation. In most numerical codes, time-dependence is treated implicitly by multiplying the equation with an eigenvalue  $\frac{1}{k}$  [160]. In such a case calculates the equation is said to be time-independent Boltzmann's neutron transport equation given by:

$$\begin{aligned} & \left[ \hat{\Omega} \cdot \vec{\nabla} + \Sigma_t(\vec{r}, E) \right] \psi(\vec{r}, E, \hat{\Omega}) = \\ & \int_0^\infty dE' \int_{4\pi} d\hat{\Omega}' \left[ \Sigma_s(\vec{r}, \hat{\Omega}' \rightarrow \hat{\Omega}, E') \psi(\vec{r}, E, \hat{\Omega}) \right] + \\ & \frac{v\chi(E)}{k} \int_0^\infty dE' \Sigma_f(\vec{r}, E') \int_{4\pi} d\hat{\Omega}' \psi(\vec{r}, \hat{\Omega}', E') + q_{ex}(\vec{r}, E, \hat{\Omega}). \end{aligned} \quad (3.11)$$

The value of  $k$  is referred to as  $k_{eff}$ , determining the criticality state of a reactor. A value of  $k_{eff} = 1$  signifies a critical state, while  $k_{eff} > 1$  implies a supercritical

state. Conversely, if  $k_{eff} < 1$ , the reactor is in a subcritical condition.

Although it is possible to derive Boltzmann’s neutron transport equation analytically, obtaining an analytical solution requires significant simplifications and the imposition of boundary conditions on the energy and spatial dependence of the neutron cross-sections in the system’s materials. This limited approach can restrict the accuracy of predicting the average behavior of the neutron population, making the solution less applicable to practical systems. As a result, numerical solutions have proven to be more reliable in solving Boltzmann’s neutron transport equation, with two types of numerical solutions: deterministic and stochastic methods.

### 3.2.2 Deterministic and Stochastic Methods

Boltzmann’s Neutron Transport Equation (BNTE), as discussed in section 3.2.1, is widely employed in radiation transport simulations to address various problems. Computational codes utilize statistical methods based on two different approaches to solve the BNTE. The deterministic approach employs discrete variables, such as the  $P_N$  (P-Nodal) and  $S_N$  (Spherical Harmonics) methods. The  $S_N$  method involves a discretized representation of the BNTE, assuming a set of distinct numerical values for the direction of motion variables. In contrast, the stochastic or probabilistic approach relies on Monte Carlo statistics, obtaining answers by tracking and recording some aspects of individual particles’ mean behaviors. The mean particle behavior in a physical system is then inferred from the mean behavior of the simulated particles by applying the central limit theorem to solve the approximate problems [161].

Deterministic methods typically require shorter computational times to complete a calculation, based on various approximations that overlook fine details. However, concerns have been raised regarding the accuracy and efficiency of these methods, particularly when neutron transport involves complex interfaces. Deterministic methods have been successfully applied for quick solutions in cases involving simple geometries and boundary interfaces [161]. Conversely, for complex geometries and high-precision calculations, Monte Carlo codes are preferred. The drawback of

stochastic methods is their time-consuming nature. Nevertheless, stochastic methods are considered highly successful in approximating experimental results, especially in fission reactor transport calculations involving complex geometries. From the above discussion, it becomes evident that for reactor-type problems, numerical solutions of the BNTE using Monte Carlo methods closely approximate the behavior of the nuclear reactor system compared to deterministic solutions.

### 3.2.3 Monte Carlo Methods

Unlike deterministic methods, which explicitly approximate the mean behavior of particles to solve the transport equation, Monte Carlo methods generate multiple particle life histories through random sampling techniques [162]. Monte Carlo facilitates the prediction of real behavior by considering outcomes with the highest probabilities and enables the step-by-step tracking of particle distributions throughout the system. It provides specific information about user-requested parameters, allowing the extraction of necessary details. Consequently, solving a transport problem involves integrating the histories of particles [162]. Hence, it is not just solving an equation but giving a representation of the particles' behaviour in the system. Examples of computational codes that are based on Monte Carlo methods for particle transport include MCNP, Geant4, PHITS, SRIM, and FLUKA among others. MCNP is a powerful Monte Carlo simulation code specifically designed to model the transport of neutrons, photons, and other particles in complex geometries such as the SAFARI reactor where neutron fluence, energy deposition, and dpa can be calculated [58]. Similarly, SRIM is a Monte Carlo simulation code that can predict the creation of defects, displacement of atoms, and other forms of damage in materials subjected to ion irradiation [59, 101]. Damage calculations with these two codes were utilized in this study to aid in the understanding of the material behavior of Au and Lu-Al under neutron and proton irradiation conditions respectively.

In nuclear transport systems, the algorithms for the Monte Carlo method comprise five parts [163]:

- (i) Extraction of cross-sections: In MCNP, cross-section data are obtained from databases such as the Evaluated Nuclear Data Files (ENDF) libraries among

others [162].

- (ii) Geometry modeling: This part takes into account all the boundaries and interfaces where neutron interactions occur, and ray tracing through the geometry is performed.
- (iii) Probability distribution sampling: This step involves implementing the sampling of probability distributions.
- (iv) Tallying: This part performs the scoring of particle histories and accumulation of results.
- (v) Output interface: An interface is provided for user interactions with the program.

Monte Carlo N-Particle Transport code (MCNP) is developed and maintained at Los Alamos and distributed by the Radiation Safety Information Computational Center (RSICC). The code is a general-purpose computational code that has been successful in solving Boltzmann's neutron transport equation for a variety of reactor-type problems, especially radiation damage by electrons, neutrons, and photons to solid materials. Hence, in this study, MCNP6.2 [58] was used to simulate damage parameters.

### **3.3 Simulation of damage with MCNP6.2**

The basic architecture of MCNP is a mix of Fortran and C/C++ programming paradigm. The input file is written in an ASCII format and can be built on any Fortran compiler or any C++ compiler supporting C++ 14 standard. [58, 164].

MCNP is designed to model radiation transport using natural stochastic methods. It can simulate the interactions of neutrons, photons, and electrons in natural radiation processes [58]. Track lengths of radiation sources and their point-wise interactions cross-sections with interfaces are taken into account from the birth of a particle to its death within a defined geometry.

To simulate radiation damage using MCNP6.2, an input file is required. The input file (INP) contains geometry specification, material description, choice of cross-section libraries, source characterization, the type of solutions desired, and the preferred variance reduction techniques. The INP follows a basic block structure, with three blocks separated by a blank space. These blocks include:

1. *Cell Card*: This defines the three-dimensional geometry, materials, and number densities of objects in the modeled system.
2. *Surface Card*: This describes all the surface definitions in the cell card.
3. *Data Card*: This block describes the particle generation source, transport and tracking details, physics parameters, and the number of histories required for tally estimation [58].

To perform radiation damage simulation, the geometries of all reactor components, surfaces, and the materials' density properties for Au and Lu-Al materials were coded in the cell and surface cards. The physics quantities of interest, such as flux, displacement per atom (dpa), and heat deposition, were tallied using MCNP's tally functions.

In most cases, standard libraries are required as additional inputs to solve Physics problems, especially those involving neutrons. The Evaluated Nuclear Data File (ENDF B-VII) library, provides cross-section data for nuclides and nuclear reactions. This database contains information on various interactions, including scattering (elastic and inelastic), fission, neutron capture, and activation, as discussed in section 2.4. In this work, all possible neutron cross-sections were taken into account. Also, calculations involving the physics of photons were based on photo-electric absorption, Compton scattering, and pair production, as discussed in section 2.5.

### **3.3.1 MCNP Model of SAFARI-1**

The SAFARI-1 research reactor is classified as a tank-in-pool, beryllium-reflected, light-water-cooled research reactor with a total power output of 20 MW. The reactor's total power and flux align with its designation as a Material Testing Reactor

(MTR).[165]

To simulate reaction types that emulate the actual reactor system, detailed three-dimensional modeling of the reactor assemblies is essential for implementing the MCNP6.2 code. The geometrical surfaces consist of thousands of volumetric cells that constitute the cell card of the MCNP input file [166]. The MCNP model for SAFARI-1 was developed by the Radiation and Reactor Theory (RRT) group of the South African Nuclear Energy Corporation (Necsa) and has undergone thorough verification and validation (VV). Figure 3.2 illustrates a cross-section of the MCNP model of SAFARI-1.

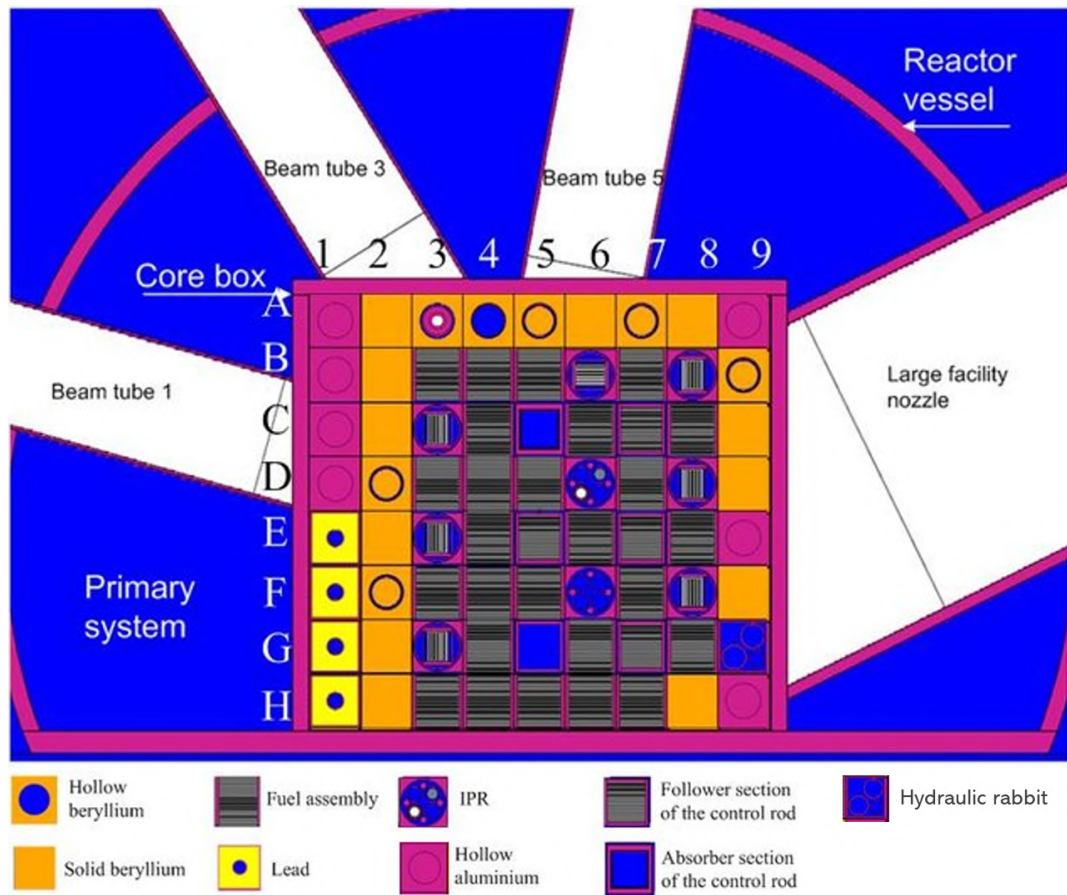


**Figure 3.2:** MCNP Model of the SAFARI-1 Research Reactor. Reactor core configuration showing the fuel assemblies and IPR positions. A grid of rows labeled A-H and columns 1-9 is shown to enhance the identification of the core structural components. ( [165, 166])

The model comprehensively describes various components of the reactor, encompassing the reactor tank, core box, reactor core, and ex-core irradiation points

(beam tubes) with varying neutron fluxes [165]. Within the reactor core, there is a grid of 8x9 different reflector elements on three sides, and fuel elements surround the irradiation rigs. Each fuel element consists of a planar array of 19-plate Uranium-Silicide-Aluminium ( $U_3Si_2-Al$ ) powder-dispersed core embedded in an aluminium alloy cladding.

The Isotope Production Rig (IPR) positions, D6 and F6, are flanked by the follower section and absorber section control rods. These positions play a crucial role in both in-core sample irradiation and the commercial production of radio-pharmaceutical isotopes. Notably, position D6 (NW), as illustrated in Figures 3.3, was specifically used as the irradiation position for the samples of Lu-Al and Au in the MCNP6.2 code.

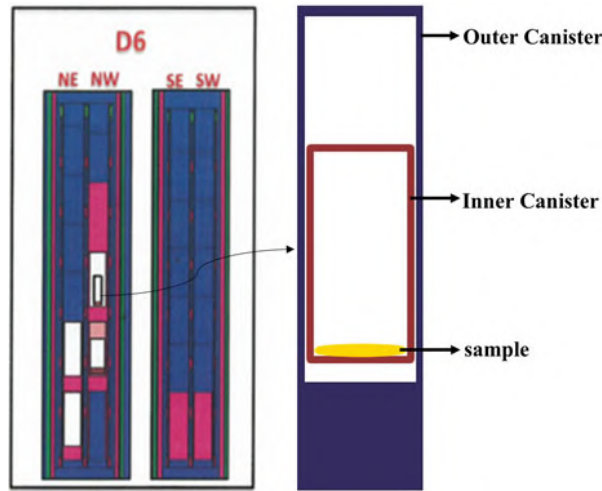


**Figure 3.3:** D6 IPR position. An extract of the D6 position illustrating the sample irradiation position

### 3.3.2 MCNP Tallies

In MCNP, the process of quantifying the parameters of interest is known as tallying. Each tally, denoted by  $F_n:a$ , is characterized by a number 'n,' defining a unique set of problems that can be addressed, and 'a,' which represents the type of particle (neutron, photon, or electron) being tracked. These tallies provide a comprehensive understanding of the physics involved in the problem, including fractional or relative errors. Additionally, tallies offer insights into track production and loss, as

well as particle energy. Information such as track population, number of collisions, mean free path, and energy of tracks in a cell can also be obtained from tallies [164].

The MCNP code incorporates seven tallies (F1, F2, F4, F5, F6, F7, and F8) [164], each capable of specifying a particular physical quantity of interest. In this study, the F4 and F6 tallies were utilized to define the physics associated with cell (volumetric) flux impinging on the cells at the irradiation position of the materials, Displacement Per Atom (dpa), and heat energy deposited in the chosen materials.

### 3.3.3 Neutron flux tally

MCNP provides two primary tallies: F2 for estimating surface flux and F4 for cell flux. The selection of F4 to tally the neutron flux is grounded in the assumption that neutron collisions with the materials will exhibit isotropic emanation from the fuel assemblies surrounding the IPR D6 position, as depicted in Figure 3.2.

The track length estimator (F4) tally, utilized to define cell flux, is scored according to the fundamental definition of fluence as the number of particle track lengths per unit volume, denoted by  $W \frac{T_l}{V}$  with units in particles/cm<sup>2</sup>. The cell flux is expressed as:

$$\bar{\phi}_V = \frac{1}{V} \int dE \int dt \int dV \int d\Omega \psi(\vec{r}, \vec{\Omega}, E, t), \quad (3.12)$$

Here,  $W$  represents the particle's weight, and  $\vec{r}$ ,  $\vec{\Omega}$ ,  $E$ ,  $t$ , and  $V$  denote the particle's position vector (cm), directional unit vector (this quantity is a bit different from the solid angle earlier defined. It specifies the orientation of interest for tallying cell particle flux relative to the incident particle direction. In contrast, solid angle quantifies the angular extent or coverage of radiation from a source as perceived from a specific point), energy (MeV), time (shakes, sh; 1 sh = 10<sup>-8</sup> s), and volume (cm<sup>3</sup>), respectively. The formula allows for scoring all particles within a cell in the designated time and energy range, thereby facilitating flux calculation.

### 3.3.4 Displacement Per Atom (dpa) tally

The calculation of the Displacement Per Atom(dpa) is relevant for crystalline materials irradiation with neutron sources. In this work, the model for dpa was based on the NRT-dpa model [145]. The number of atoms displaced is related to the total available neutron energy  $E_a$  and is given by Eq. 3.13 [167]

$$DPA = \epsilon \sum_n \frac{E_a}{2E_d}, \quad (3.13)$$

where the sum index  $n$  indicates the summation over different nuclides that make up a material [168, 169].  $E_a$  is the effective available energy for the creation of atomic displacement in the lattice of a material,  $\epsilon$  is the empirical displacement efficiency factor (0.8) obtained from Binary Collision Approximation(BCA) simulations and  $E_d$  is the displacement energy of the atom from its lattice position.

To evaluate the dpa rate for the material, a tally multiplier is applied to the standard flux in the MCNP code. The NJOY code was used to process the defect production cross-sections (DPCS) and reaction cross-sections in the Evaluated Nuclear Data Files (ENDF/B-VII) libraries into a suitable format accessible by the MCNP input file. NJOY has several functions (ACER, BROADR, GAMINR, HEATR, MODER, RECONR, etc.) for processing the ENDF data [167]. While other functions are responsible for generating other nuclear cross-section data and data conversion into suitable formats, the HEATR function is responsible for generating damage cross-sections and KERMA factors [167, 168]. The damage cross-section is usually implemented into the computation when the MT=444 is called in the MCNP INP.

The DPCS are produced for all energy ranges but there is threshold energy below which no atomic displacements can occur in a material. This is expressed in the Robinson partition function  $P_r(E)$  shown in Eq. 3.14 [167].

$$P_r(E) = \frac{E_R}{1 + F_L(3.4008\epsilon^{1/6} + 0.40244\epsilon^{3/4} + \epsilon)}, \quad (3.14)$$

where  $\epsilon$  is the ratio of the primary recoil energy ( $E_R$ ) to the energy of the lattice nuclei ( $E_L$ ).  $F_L$  is a factor relating the charge ( $Z$ ) and atomic mass ( $A$ ) of the lattice nuclei ( $L$ ) expressed by Macfarlane et al. in the NJOY16 manual [167]. The  $P_r(E)$  suggests that  $E_R$  be greater or equal to 25 eV, the threshold displacement energy before atoms are displaced from their lattice sites; otherwise, DPA is equal to zero [167].  $E_R$  values within the thermal energy region (0.003 eV - 4.0 eV) defined in the 172 energy groups in the XMAS structure [170], are below the displacement energy threshold and hence, lack sufficient energy to produce recoils leading to atomic displacements. Therefore, DPAs for such energies below 25 eV were constrained to zero in the outputs of Eq. 3.15 to be consistent with the NRT-dpa model. The F4 neutron tally was used with FM tally multipliers to estimate the dpa rate as shown in Eq. 3.15

$$F4 : Nc \quad (3.15)$$

$$FM4Km(MT)$$

where FM4 is the F4 tally multiplier which performs the integration of Eq. 3.13 implicitly during simulation when the F4 tally is called.  $MT=444$  is the material reaction,  $m$  is the material number,  $c$  is the cell number, and  $K$  is the Kinetic energy released in materials(KERMA)[169] given by:

$$K = \eta \frac{\rho_m}{2E_d}, \quad (3.16)$$

where  $\rho_m$  is the number density of the material  $m$  in atoms/barn-cm,  $\eta$  is 0.8 and  $E_d$  is the atomic displacement energy.

### 3.3.5 Heat deposition tally

The track length cell energy deposition tallies in MCNP are the F6 and F7 tallies. Heat energy deposited in the materials of interest was estimated using the F6 tally because actual heating is calculated by the combination of neutrons and photon heat deposition in the F6 tallies, which includes all neutron and photon reactions, unlike the F7 tally that estimates the heating for only the fissionable materials [164, 171]. The F6 card tallies the energy deposition for individual particles hence, it was used

to calculate the energy deposition for neutrons and photons. To ensure consistency the F6 track-length estimates for neutron heating include all heating numbers and kinetic energy for secondary particles except photons. The same applies to the estimation for photon heating. The F6 tally is given by:

$$F6 = \frac{\rho_a}{V\rho_m} \int_V \int_t \int_E \sigma_T(E) H(E) \phi(r, E, t) dE dt dV, \quad (3.17)$$

where  $\rho_a, \rho_m$  are the number and mass densities for the materials in the volume  $V$ ,  $\sigma_T(E)$  is the energy-dependent neutron cross-section in the unit barn,  $H(E)$  is the heating response in MeV/g for collision and  $\phi(r, E, t)$  is the scalar flux in  $\text{cm}^{-2}\text{s}^{-1}\text{MeV}^{-1}$ .

The individual particle's heat deposition tallies are given in Eq. 3.18 and 3.19 for neutrons and photons, respectively [166, 171].

The heat deposited by neutrons in the sample was estimated using heating tally F6:n:

$$H(E) = E - \sum_i P_i(E) [\bar{E}_{out_i}(E) - Q_i + \bar{E}_{i,\gamma}(E)], \quad (3.18)$$

where  $E$  is the energy of the incident neutron,  $P(E)_i$  is the probability for the reaction  $i$  at the incident energy  $E$  of a neutron,  $\bar{E}_{out_i}$  is the mean exiting energy of the neutron for the reaction  $i$  at incident energy  $E$  with Q-value  $Q_i$  and  $\bar{E}_{i,\gamma}(E)$  is the mean exiting  $\gamma$  energy for the reaction  $i$  at neutron incident energy  $E$ .

Similarly, for the photon heating tally, F6:p:

$$H(E) = E - \sum_{i=1}^n P_i(E) [\bar{E}_{i,out}(E)], \quad (3.19)$$

where  $P_i(E)$  is the reaction probability of gamma incident energy  $E$ ,  $\bar{E}_{out}(E)$  is the mean exiting energy of gamma for the reaction  $i$  at neutron incident energy  $E$ . Important photon reactions ( $i$ ) under consideration in the reactor environment include  $i = 1$ , representing incoherent Compton scattering,  $i = 2$  for pair production, and  $i = 3$ , for photoelectric absorption. The  $\bar{E}_{i,out}(E)$  is the mean exiting  $\gamma$  energy for the reaction at neutron incident energy  $E$ , and  $P_i(E)$  is the probability

of the reaction at  $\gamma$  incident energy  $E$ . Thus the heat deposition specifically due to neutrons and photons was evaluated.

### 3.3.6 Evaluation of uncertainties in MCNP tallies

MCNP, being a Monte Carlo code, inherently incorporates statistical variations in numerical experiments to ensure reliable results from tallies. The uncertainties in MCNP are presented as fractional standard deviations (fsd), usually accompanied by the quantity tallied expressed as *Tally Result*  $\pm$  *SD*.

In addition to fsd, several checks, often referred to as *10 statistical checks*, are performed by the code and presented in the Tally Fluctuation Chart (TFC), as detailed in Appendix A. These checks are essential for assessing the precision, accuracy, and confidence limits of the tally estimates. The checks include:

- **Relative error:** A fractional  $1\sigma$  uncertainty estimate in the mean of the tally. calculated as

$$R \equiv \frac{S_{\bar{x}}}{\bar{x}}, \quad (3.20)$$

where  $S_{\bar{x}}$  is the standard deviation of the tally's mean and  $\bar{x}$  is the mean of the tally. A value of  $R < 0.1$  is generally reliable, with  $R < 0.05$  considered meaningful for point and ring detectors [164, 172]. The relative error is an important statistic that gives an idea about the statistical precision for the estimated tally mean. However,  $R$  alone does not suffice to decide the acceptability of a tally result.

- **Figure of merit (FOM):** This is another important statistic produced in the MCNP TFC. It is the inverse of the product of the square of the relative error ( $R$ ) and the computation time ( $T$ ) in minutes.

$$FOM = \frac{1}{R^2 T} \quad (3.21)$$

FOM is a measure of the reliability of the tally results that indicate whether or not the tally is well-behaved [172]. It is necessary to check at the end

of each simulation to ascertain whether the FOMs yields are approximately constant as the number of each tally's histories.

- **Variance of Variance (VOV):** A measure of the accuracy of R, calculated as the relative VOV is a useful statistical indicator for the reliability in the confidence intervals. It is calculated as

$$VOV = \frac{S^2(S_{\bar{x}}^2)}{S_{\bar{x}}^4}, \quad (3.22)$$

where the estimated variance in  $\bar{x}$  is  $S_{\bar{x}}^2$  and  $S^2(S_{\bar{x}}^2)$  is the variance calculated in  $S_{\bar{x}}^2$ . It provides insights into the precision of a tally result and estimates the relative statistical uncertainties in the value of R.

- **Pdf SLOPE:** This check assesses the quality of the confidence interval estimate for the mean of the tally. MCNP uses the highest number of scoring histories to implicitly fit the Pareto function  $f_p(x)$  with the parameters  $a$  and  $k$  [164, 172] given by :

$$f_p(x) = \frac{1}{a(1 + kx/a)^{1+(1/k)}}, \quad (3.23)$$

the slope (SLOPE) shown in Eq. 3.24 is extrapolated to ensure it is  $\geq 3.0$  for the tally results to be deemed acceptable.

$$SLOPE \equiv \frac{1}{k} + 1, \quad (3.24)$$

### 3.4 Numerical simulations in MCNP6.2

The simulation of damage to Au foil and Lu-Al alloy materials was carried out using the model based on the SAFARI-1 research reactor, as detailed in section 3.2.1. The geometries of all reactor components and the material density properties for Au and Lu-Al were coded in the cell and surface input cards. MCNP calculations rely on tallies to track and record particle interactions with the target material. In this study, the F4 and F6 tallies, along with their tally multipliers, were utilized to calculate flux, dpa rate, and heat deposition resulting from neutrons and photons.

The calculations were conducted with the 172 energy groups in the XMAS structures [158]. This structure was employed to generate the energy-dependent flux spectrum at the D6 NW3 IPR position. Comprising 80 thermal, 47 epithermal, and 45 fast neutron energies, the spectrum ranges from  $1.10 \times 10^{-10}$  to  $1.96 \times 10^1$  MeV. This energy range has proven effective in various calculations with the SAFARI-1 MCNP model, offering results that better represent the SAFARI-1 reactor compared to other energy groups.

The simulations in this study were performed using the Beginning Of Cycle (BOC) state C1601-1, representing the normal reactor cycle for the SAFARI-1 reactor. A total of 2500 kcode active cycles and 250 inactive cycles were executed, with each cycle employing approximately 100,000 histories to ensure sufficient statistics and convergence of the results. The acceptable success criteria for MCNP uncertainties include having a tally fluctuation chart (TFC) with a constant value Figure Of Merit (FOM), a relative error, and a Variance Of Variance (VOV) all less than 0.1.

### 3.4.1 Calculation of atomic number densities and KERMA

To estimate the DPA the atomic number density of the material and subsequently the KERMA has to be calculated and the KERMA is included in the MCNP INP file. The atomic number density of a nuclide is given by:

$$\rho_m = \frac{\rho N_A}{A}, \quad (3.25)$$

where  $\rho$  is the mass density,  $A$  and  $N_A$  are the atomic mass and Avogadro's number respectively for a single nuclide. For a composite material, the number density is obtained as a summation of the individual percentage by weight composition ( $\omega_i$ ).

The atomic density for a composite material is given by:

$$\rho_{m_i} = \sum \frac{\omega_i \rho_i N_A}{A_i}, \quad (3.26)$$

where  $i$  represents the number of nuclides in the composite material. The results of the atomic densities obtained from the equations above are in  $\text{atoms.cm}^{-3}$ . How-

ever, it should be noted that  $1 \text{ cm}^2 = 10^{24} \text{ barn}$ . Therefore, the results for Au and the oxidized Lutetium-Aluminium were converted into atoms/barn-cm and presented in Table 3.1 below.

**Table 3.1:** Atomic density for Au and Lu-Al [173]

| <b>Material</b>                                             | <b>Mass Density</b><br>(g.cm <sup>-3</sup> ) | <b>Atomic density</b><br>(atoms.b <sup>-1</sup> .cm <sup>-1</sup> ) |
|-------------------------------------------------------------|----------------------------------------------|---------------------------------------------------------------------|
| Gold (Au)                                                   | 19.30                                        | $5.9070 \times 10^{-2}$                                             |
| Lutetium-Aluminium( $\text{Al}_5\text{Lu}_3\text{O}_{12}$ ) | 6.73                                         | $9.5160 \times 10^{-2}$                                             |

Eq. 3.16 was used to calculate the KERMA factors for a single nuclide. For the composite or polyatomic material, the equivalent displacement energies of the nuclides is computed using Eq. 3.27 [174]

$$E_{d,eq} = \left[ \sum \left( \frac{c_i}{E_{d,i}} \right) \right]^{-1}, \quad (3.27)$$

where  $c_i$  is the fractional composition of the individual nuclides in the polyatomic material and  $E_{d,i}$  is the displacement energy of the individual nuclides present in the material. Thus the KERMA is given by Eq. 3.28 for a polyatomic material:

$$K = \frac{\eta \rho_m}{2E_{d,eq}}, \quad (3.28)$$

The KERMA factors were calculated using known atomic displacement energies  $E_d$  [121] and the results are presented in Table 3.2

**Table 3.2:** KERMA factors for Au and Lu-Al [121, 173]

| <b>Material</b>                                             | <b><math>E_d</math></b><br>(eV) | <b>KERMA factor</b><br>(atoms.b <sup>-1</sup> .cm <sup>-1</sup> .MeV <sup>-1</sup> ) |
|-------------------------------------------------------------|---------------------------------|--------------------------------------------------------------------------------------|
| Gold (Au)                                                   | 43                              | $5.488 \times 10^2$                                                                  |
| Lutetium-Aluminium( $\text{Al}_5\text{Lu}_3\text{O}_{12}$ ) | 44 (Lu)<br>27 (Al)<br>45 (O)    | $9.897 \times 10^2$                                                                  |

The above KERMA factors were supplied as inputs in MCNP F4 modified dpa tally in Eq. 3.15.

### 3.4.2 Flux normalization

The energy-dependent neutron flux data obtained from the MCNP F4 tally requires normalization to the SAFARI-1 reactor power. This was achieved using Eq. 3.29

$$Flux(n/cm^2s) = \frac{P\bar{\nu}}{QeK_{eff}} \times F4_{Tally} = PNF \times F4_{Tally}, \quad (3.29)$$

The parameters for the flux normalization can be summarized in Table 3.3 below

**Table 3.3:** Flux normalization parameters [130]

| Parameter                                           | Value                                                             | units                |
|-----------------------------------------------------|-------------------------------------------------------------------|----------------------|
| Total reactor Power of SAFARI-1 (P)                 | 20                                                                | Mega Watts (MW)      |
| Average No. of neutrons per fission ( $\bar{\nu}$ ) | 2.456 (Au)<br>2.457 (Lu-Al)                                       | neutrons/fission     |
| Conversion unit factor (e)                          | $1.6021 \times 10^{-19}$                                          | MJ.MeV <sup>-1</sup> |
| Neutron multiplication factor ( $K_{eff}$ )         | 0.98719 (Au)<br>0.98725 (Lu-Al)                                   |                      |
| Fission Q-Value (Q)                                 | 200                                                               | MeV                  |
| Power Normalization factor(PNF)                     | $1.55288 \times 10^{18}$ (Au)<br>$1.55342 \times 10^{18}$ (Lu-Al) | n.s <sup>-1</sup>    |

The parameters average number of neutrons born per fission ( $\bar{\nu}$ ), neutron multiplication factors ( $K_{eff}$ ), and  $F4_{Tally}$  are numerical outputs from the simulations. With Eq. 3.29 and the parameters in Table 3.3, the power normalization factor (PNF) for each material was calculated and the flux was estimated by finding the product of the PNF with the flux tally results for all the energies in the XMAS group. The average absolute flux for each of the materials was also estimated.

The average neutron and photon heat deposition for each material was also converted in Watts per gram (W.g<sup>-1</sup>) using the Eq. 3.30.

$$H(E) = PNF \times \kappa \times F6(n/p), \quad (3.30)$$

where  $\kappa = 1.602 \times 10^{-13} J.MeV^{-1}$  is the energy (MeV.g<sup>-1</sup>) to power (W.g<sup>-1</sup>) con-

version factor,  $F6:(n/p)$  is the neutron or photon heat deposition tallies respectively.

## 3.5 Radiation damage Simulations with SRIM

The Stopping and Range of Ions in Matter (SRIM) [59] is a Monte Carlo simulation code that evaluates damage in solid materials using a Binary Collision Approximation. In SRIM, the directional properties of crystalline target materials are typically ignored, treating the material as amorphous. The program is divided into Stopping/Range tables and the Transport and Range of Ions in Matter (TRIM). TRIM is utilized for evaluating the three-dimensional ion distribution with associated energy losses, recoils, vacancies, target damage, sputtering yield, ionization, and phonon formation in the target [59, 101]. The physics accounts for realistic potentials, providing accurate scattering integral results over a wide range of histories, tracking in straight free-flight paths between collisions for a given individual ion energy in space.

### 3.5.1 Displacement-Per-Atom calculations using SRIM-2013

The concept of Displacement-Per-Atom (dpa) has been discussed in sections 2.10.1 and 3.3.4. In this section, the Norgett, Robinson, Torrens (NRT) dpa model is discussed based on ion-induced radiation damage in the SRIM code. Typically, a direct comparison of radiation damage for a specific neutron fluence with other radiation sources faces challenges due to variations in neutron spectrum and Primary Knock-on Atom (PKA) energy spectrum in different environments. The advantage of using NRT-dpa is that it provides an environment-independent exposure quantity, enabling comparisons with other sources.

To compute dpa in SRIM based on the NRT-dpa model, the incident ion is considered to have transferred its kinetic energy to the target material through atomic collisions, resulting in the recoil of the target atom known as the Primary Knock-on Atom (PKA). The PKA, upon further collisions with neighboring atoms, produces a collision cascade, initiating primary radiation damage as discussed in section 2.8.2. This process results in the loss of part of the PKA energy via electronic excitation,

and the remaining PKA energy, referred to as the damage energy ( $T_{dam}$ ) [47, 101], is transferred through elastic collisions of the PKA with the neighboring atoms in the lattice. The  $T_{dam}$  is the energy available for atomic displacement, sometimes denoted as  $E_a$ . According to the Robinson Partition function ( $Pr$ ) in Eq. 3.14, atomic displacement and subsequent vacancy production are only possible at a threshold where  $T_{dam} \geq E_d$ , where  $E_d$  is the atomic displacement energy. The number of displacements produced in the target material, according to the NRT model, is given by Eq. 3.31 [47]:

$$\nu_{NRT} = \frac{\eta T_{dam}}{2E_d}, \quad (3.31)$$

where  $E_d$  is the threshold displacement energy,  $\nu_{NRT}$ , the number of displacements,  $\eta = 0.8$  is the efficiency factor, and  $T_{dam}$  is the damage energy. It is important to note that SRIM calculations assume an amorphous target, while  $E_d$  is dependent on the lattice space of the material. Therefore, it is advisable to use average values of  $E_d$  in Eq. 3.31 to compensate for the different crystallographic directions [101]. For a polyatomic material, the equivalent average displacement energy ( $E_{d,eq}$ ), as shown in Eq. 3.27, should be substituted in Eq. 3.31 for the calculation of  $\nu_{NRT}$ .

The calculation of the damage energy ( $T_{dam}$ ) can be done via three approaches. However, in this work, the straightforward approach involving the arithmetic difference between the ionization energy loss of the recoil ions ( $E_{ioniz}$ ) and the energy absorbed by recoils ( $E_{recoil}$ ) [47, 101], as shown in Eq. 3.32, was used.

$$T_{dam} = E_{recoil} - E_{ioniz}, \quad (3.32)$$

where the parameters  $E_{recoil}$  and  $E_{ioniz}$  are obtained from TRIM output files *E2RECOIL.txt* and *IONIZ.txt* respectively.

The other two methods of calculating  $T_{dam}$  are presented in Eq. 3.33 and 3.34 respectively.

$$T_{dam} = E_{phr} - E_{phi}, \quad (3.33)$$

$$T_{dam} = E_{tot} - E_{non-damage} = E_{tot} - E_{ionization} - E_{phonon}, \quad (3.34)$$

where  $E_{phi}$  is the energy of the phonon generated by the incident ion,  $E_{phr}$  is the phonon energy produced by the recoil ions,  $E_{tot}$  is the total damage creating energy loss due to the incident ion beam and  $E_{non-damage}$  constitute the energy generated through ionization and phonon creation [47, 101].

The displacement per atom rate is now calculated using Eq. 3.35 [25, 47, 101].

$$dpa[s^{-1}] = \frac{\nu_{NRT}\phi}{\rho_m}, \quad (3.35)$$

Consequently, from Eq. 3.31 the dpa rate can be written as:

$$dpa[s^{-1}] = \frac{\eta T_{dam}\phi}{2\rho_m E_d}, \quad (3.36)$$

where  $\phi$  is the ion flux in  $\text{ions.cm}^{-2}.\text{s}^{-1}$  and  $\rho_m$  is the atomic number density of the material in  $\text{atoms.cm}^{-3}$ . With Eq. 3.36 dpa rate can be calculated using TRIM simulation outputs. The absolute value of dpa is obtained when ion fluence ( $\Phi$ ) with units  $\text{ion.cm}^{-2}$  instead of flux is substituted in Eq. 3.36 as shown in Eq. 3.37.

$$dpa = \frac{\eta T_{dam}\Phi}{2\rho_m E_d}, \quad (3.37)$$

where all parameters maintain their meanings as described above.

Calculating dpa using Eq. 3.37 with the three  $T_{dam}$  estimation methods shows slight differences. For instance, a 0.5 MeV proton beam with fluence  $7.7 \times 10^{16} \text{ p.cm}^{-2}$  incident in Au target will produce the dpa shown in Table 3.4.

**Table 3.4:** Estimates of dpa for Au using different  $T_{dam}$  methods

| S/N | $T_{dam}$ estimation method          | dpa    |
|-----|--------------------------------------|--------|
| 1   | $T_{dam} = E_{recoil} - E_{ioniz}$   | 0.0129 |
| 2   | $T_{dam} = E_{phr} - E_{phi}$        | 0.0119 |
| 3   | $T_{dam} = E_{tot} - E_{non-damage}$ | 0.0104 |

The first  $T_{dam}$  method in Eq 3.32 was however preferred as it is more consistent with the NRT-damage energy in Eq. 3.31 when lattice binding energy is set to zero [101].

### 3.5.2 Method of damage simulation in SRIM-2013

The radiation damage simulation was carried out on 99.99% pure gold ( $^{197}\text{Au}$ ) foil with a thickness of 0.0064 mm and Lu-Al (5.2%wt Lu) binary alloy with a thickness of 0.1016 mm. The simulation of damage in the SRIM-2013 version was performed with  $\text{H}^+$  and  $\text{Au}^+$  to compare the effect of lighter ions and heavier ions on the dpa rates. The ion projections at energies were gradually increased within the range of 0.02 to 1.0 MeV into Au and Lu-Al targets for both the Quick calculation (QC) and the Full Cascade (FC) modes for 100,000 ion histories. Agarwal et al. established that simulation histories ranging from 10,000 to 20,000 ions, representing less than 1% statistical uncertainties assuming a Gaussian probability [47], provided a favorable balance between computational cost and accuracy across a broad spectrum of ion-target conditions.

Hence, the utilization of a high ion history in this study aimed to ensure consistency in calculations with minimal uncertainties. The ion beams of  $\text{H}^+$  and  $\text{Au}^+$  were implanted into the target materials to compare their displacement per atom (dpa) rates with those of neutrons predicted in an earlier study [130]. This comparison aims to guide in selecting ion beams that are best suited for the irradiation of Lu-Al and Au, ensuring comparability with neutrons. Table 3.5 shows the in-

**Table 3.5:** Input parameters for SRIM Calculations

| Ion Data                  |               |                     |                       |       | Target Data |               |             |             |
|---------------------------|---------------|---------------------|-----------------------|-------|-------------|---------------|-------------|-------------|
| Ion Species               | Atomic number | Ion Energy<br>(MeV) | Angle<br>( $^\circ$ ) | Layer | Symbol      | $E_d$<br>(eV) | LBE<br>(eV) | SBE<br>(eV) |
| $\text{H}^+, \text{Au}^+$ | 1, 79         | 0.2 - 1.0           | 0                     | 1     | Au          | 43            | 0           | 3.36        |
| $\text{H}^+, \text{Au}^+$ | 1, 79         | 0.2 - 1.0           | 0                     | 2     | Al          | 27            | 0           | 3.80        |
|                           |               |                     |                       |       | Lu          | 44            | 2           | 4.29        |

\* *LBE* and *SBE* refer to Lattice Binding Energy and Surface Binding Energy respectively

put parameters, Ion data, and Target Data, that were utilized for the estimation of radiation damage in Lu-Al and Au materials. Applying the recommendation of Stoller et al., calculations were conducted using the NRT-dpa method. The SRIM default surface binding energies of the materials were constrained to zero for consistency with NRT-dpa models, aligning with the suggestions made in previous studies [47, 101, 175]. The threshold displacement energy ( $E_d$ ) values for Au, Lu, and Al

input into the program were determined using updated values from Konobeyev et al [121].

All SRIM input parameters were consistent for QC and FC modes of calculation. The target depth was set to ensure that all collisions of the projectile ion into the target occurred within the target region, preventing ion transmission [47, 101]. Transmission of ions could result in a loss of PKA energies at the incident ion energy and subsequently reduce the number of recoils. The number of displacements and, consequently, dpa rates were calculated for different ion energies using Eqs. 3.31 and 3.36, respectively.

### 3.6 Sample materials

The sample materials utilized in this study included a 99.99% Gold foil with a thickness of 0.0064 mm and a 99.7838% Lutetium-Aluminium alloy with a thickness of 0.1016 mm, as specified in the datasheet (see Appendix D) provided by Reactor Experiments Inc., San Carlos, California, to the South African Nuclear Energy Corporation Soc. Ltd (Necsa). The chemical composition by weight of the samples is detailed in Table 3.6.

**Table 3.6:** Elemental chemical composition of sample materials

| Material | Elemental composition (%wt) |      |      |      |      |      |      |
|----------|-----------------------------|------|------|------|------|------|------|
|          | Al                          | Lu   | Au   | Si   | Fe   | Cr   | Mg   |
| Lu-Al    | 94.72                       | 4.92 | -    | 0.18 | 0.08 | 0.06 | 0.03 |
| Au       | -                           | -    | 99.7 | 0.03 | -    | -    | -    |

Gold is a dense metal with an atomic number of 79, belonging to group 11 and period 6 of the periodic table. At room temperature, Gold has a homologous temperature ( $T_H$ ) of approximately 0.2228 times its melting temperature ( $T_m$ ), a mass density of 19.3 g/cm<sup>3</sup>, and an electrical resistivity of 2.214  $\mu\Omega \cdot \text{cm}^{-1}$  at 20 °C. It exhibits a face-centered cubic (FCC) crystal structure with a lattice parameter of  $a = 0.407$  nm in the fm-3m space group, as depicted in Figure 3.4a.

Lutetium-Aluminium alloy is a composite material consisting of aluminum, which

has an FCC crystal structure, and lutetium, a closely packed hexagonal structure, forming a binary alloy. The exact phase of this alloy is still uncertain. However, based on the XRD pattern obtained before irradiation, the sample was identified. The general Lu-Al binary alloy system exhibits a complex cubic structure, an advanced FCC system that matches the diamond cubic crystal system. This belongs to the  $Fd-3m$  (227) space group, as indicated by detailed calculations presented in Table 3.8, and is illustrated in Figure 3.4b [176].

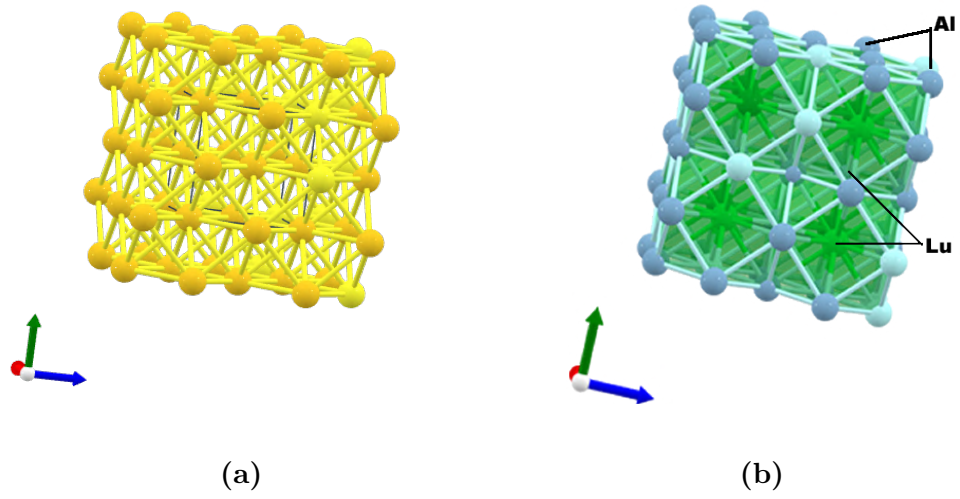


**Figure 3.4:** Crystal structures (a) Gold (Au) FCC crystal structure [177] (b) Lutetium-Aluminium complex cubic structure [178]

The XRD diffraction pattern was utilized to identify the crystal systems by calculating the lattice parameters from peak positions. As discussed in section 2.9.1, for a given pattern, there exists a set of integers for which the ratio  $\frac{\lambda^2}{4a^2}$  yields a constant quotient for all diffraction angles in the pattern. This analysis confirmed the Au system to be FCC and identified the Lu-Al complex cubic system as a Diamond cubic structure, as illustrated in Tables 3.7 and 3.8 for Cu  $K_\alpha$  X-Rays ( $\lambda=1.5406\text{\AA}$ ).

**Table 3.7:** Crystal system parameters for Au

| $2\theta(^{\circ})$ | $\sin\theta$ | hkl | $s$ (FCC) | $\frac{\lambda^2}{4a^2}$ | $a$ ( $\text{\AA}$ ) |
|---------------------|--------------|-----|-----------|--------------------------|----------------------|
| 38.2445             | 0.3276       | 111 | 3         | 0.04                     | 4.0728               |
| 44.6366             | 0.3798       | 002 | 4         | 0.04                     | 4.0569               |
| 64.7987             | 0.5358       | 022 | 8         | 0.04                     | 4.0662               |
| 71.7166             | 0.5858       | 113 | 11        | 0.03                     | 4.3613               |
| 81.6192             | 0.6536       | 222 | 12        | 0.04                     | 4.0829               |

**Table 3.8:** Crystal system parameters for Lu-Al

| $2\theta(^{\circ})$ | $\sin\theta$ | hkl | $s$ (DC) | $\frac{\lambda^2}{4a^2}$ | $a$ (Å) |
|---------------------|--------------|-----|----------|--------------------------|---------|
| 21.2826             | 0.1847       | 111 | 3        | 0.01                     | 7.2252  |
| 30.2398             | 0.2608       | 022 | 8        | 0.01                     | 8.3528  |
| 37.2335             | 0.3192       | 113 | 11       | 0.01                     | 8.0028  |
| 38.5453             | 0.3301       | 004 | 16       | 0.01                     | 9.3352  |
| 43.2495             | 0.3685       | 133 | 19       | 0.01                     | 9.1110  |
| 44.8037             | 0.3811       | 224 | 24       | 0.01                     | 9.9021  |
| 48.6473             | 0.4119       | 333 | 27       | 0.01                     | 9.7176  |
| 53.6356             | 0.4512       | 044 | 32       | 0.01                     | 9.6585  |
| 62.7767             | 0.5208       | 135 | 35       | 0.01                     | 8.7497  |
| 65.1580             | 0.5385       | 062 | 40       | 0.01                     | 9.0476  |
| 75.2683             | 0.6106       | 335 | 43       | 0.01                     | 8.2723  |
| 78.2847             | 0.6313       | 444 | 48       | 0.01                     | 8.4543  |
| 87.0581             | 0.68872      | 117 | 51       | 0.01                     | 7.9873  |

Tables 3.7 and 3.8 demonstrate a consistent quotient of  $\frac{\lambda^2}{4a^2}$ . For Au, a value of 0.04 was obtained using the FCC s-factors, while for Lu-Al with the diamond cubic s-factors, a value of 0.01 was observed. None of the other crystal systems' s-factors could yield this consistent quotient.

### 3.7 Irradiation technique with proton ion beams

In this study, proton-ion irradiation was employed as a key experimental technique to investigate the effects of radiation on the target materials. The irradiation process involved exposing the samples to a beam of accelerated protons and the impact of protons on the material's microstructure was examined. The methodology encompasses the following steps: sample irradiation setup, and subsequent analysis to assess the induced radiation damage, explore material response, and gain insights into the behavior of the crystalline structure under proton-ion irradiation.

The decision to utilize proton-ion beam irradiation instead of neutrons is primarily driven by the considerably high dose rates of  $1.02 \times 10^5$  Sv/h for Lu-Al samples and  $5.94 \times 10^5$  Sv/h for Au samples predicted for neutron irradiation (See FISPACTII inventory data in Appendix B). These dose rates are significantly high and would necessitate more than a year of cooling for Lu-Al and about 4 months of cooling for Au

before they could be safely handled for post-irradiation analyses (see Table B.1 and Table B.2 in Appendix B). Such a prolonged cooling period would significantly limit the feasibility of conducting observations with High-Resolution Transmission Electron Microscopy (HRTEM) and other characterization techniques. Consequently, proton-ion beams were utilized as an alternative for neutron irradiation, and the irradiation was performed using the TANDEM accelerator at the CLASS facility.

### 3.7.1 Proton beam irradiation at the CLASS facility

The proton ion beams used for radiation damage studies in Lu-Al and Au were generated using the 1.7 MV TANDEM accelerator at the Cambridge Laboratory for Accelerator Study of Surfaces (CLASS) at the Nuclear Science and Engineering Laboratory, Massachusetts Institute of Technology (MIT), United States of America shown in Figure 3.5. The components of the accelerator are detailed in the schematic diagram depicted in Figure 3.6.

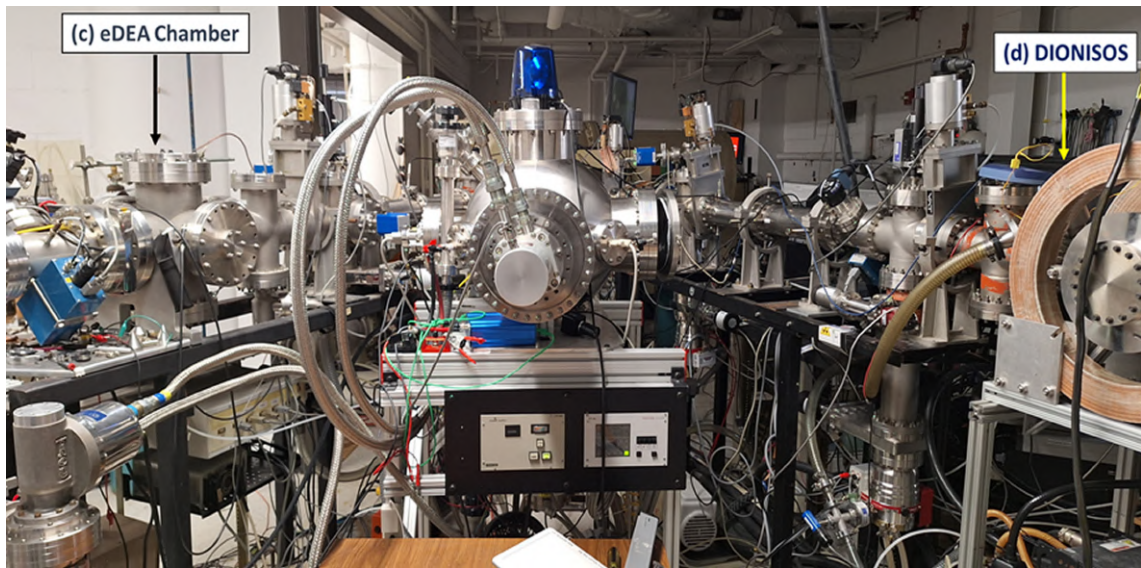


**Figure 3.5:** TANDEM Accelerator, Nuclear Science and Engineering Laboratory, MIT, USA. (Picture taken by the researcher).

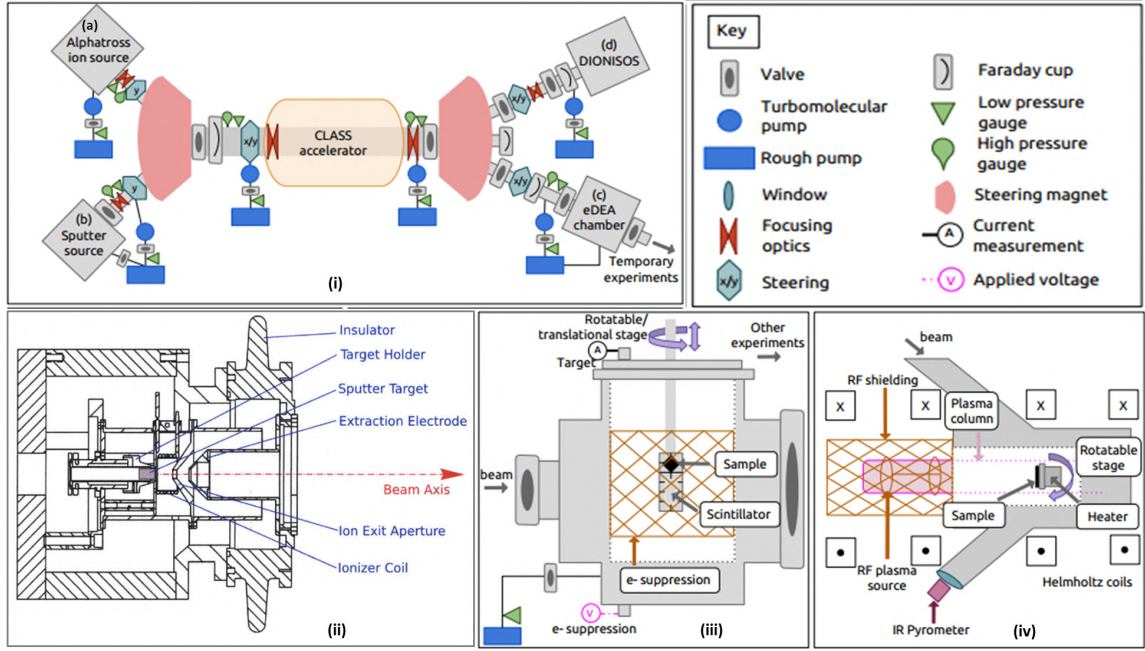


**Figure 3.6:** CLASS schematic diagram showing ion sources and beamlines (i) (a) is the Alphasource ion source. (b) is the sputter source (c) and (d) represents the beamlines. The multi-purpose beamline (c) RBS/NRA where eDEA is labeled was used for the irradiation (ii) schematics of the internal components of the Caesium sputtering source. (iii) showing the stage and the target chamber (iv) showing the top view of the target chamber. [179]

### 3.7.2 Ion sources for the TANDEM

The 1.7 MV tandem accelerator is a solid-state ion accelerator with immovable parts, designed based on a parallel-fed cascade generator utilizing a Cockcroft-Watson voltage multiplier system. The accelerator consists of two ion sources: a sputter source employing a Caesium evaporator, a solid cathode, and a hollow center with a copper cylinder packed with powder; and an Alphasource generating the desired species of Radiofrequency (RF) plasma, coupled with a Rubidium oven inducing negative charges (Figure 3.5 (i)). The CLASS accelerator features two beamlines: the RBS/NRA beamline for multipurpose material studies, and the Dynamics of Ionic Implantation and Sputtering on Surfaces (DIONISOS) beamline, dedicated to plasma-surface interaction studies [180].

The tandem accelerator primarily produces ion beam species such as H, Li, O,

Si, Cu, and Au, with energies reaching up to 10.5 MeV, using N<sub>2</sub> gas electron stripping at the terminal, at relatively high beam currents ( $< 100 \mu\text{A}$ ) [181].

### 3.7.3 Ion Energy selection and acceleration

For the irradiation, a 0.5 MeV Proton (H<sup>+</sup>) ion was used. The process of ion acceleration involves the use of a Low-Energy (LE) electromagnet for the selection of the preferred ion species that will be injected for acceleration. After the ion species has been accelerated the High Energy (HE) is then used to control the high energy ion beam to choose the preferred ion beam charge state and engage it to the beamline. These steps are summarized below,

- **Production of Negative Ions:** Negative ions are produced from an ion source,
- **Ion Species Selection and Injection:** The preferred ion species are selected and injected towards the terminal of the LE electromagnet.
- **Acceleration of Negative Ions:** The negative ions are accelerated to the high-voltage terminal (positive terminal).
- **Electron Stripping:** N<sub>2</sub> gas is injected at the terminal to strip electrons from the ions, resulting in positive ions.
- **Acceleration of Positive Ions:** Positive ions are accelerated away from the positive terminal to reach the desired final energy.
- **Focusing of Ion beams:** Magnetic quadrupole lenses are employed to obtain focused ion beams,
- **Targeting the Beamline:** The preferred ion beam species are now directed into the chosen beamline for the experiment.

The ion selection process is controlled by the LE electromagnet, functions as a mass/charge spectrometer. The Lorentz force is employed in this process.

$$F = q\mathbf{v} \times \mathbf{B}, \quad (3.38)$$

where  $\mathbf{v}$  and  $\mathbf{B}$  are the velocity and the magnetic field vector. For a uniform magnetic field with magnitude  $B_o$ , the bending radius is given by Eq. 3.39

$$R_B = \frac{\sqrt{2mE}}{qB_o}, \quad (3.39)$$

where  $q, m$ , and  $E$  are the charge, mass, and kinetic energy of the particle respectively. The magnetic current  $I_m$  is directly proportional to the magnetic field. If we assume a fixed extraction potential  $E_o$  (for the energy of the ion is  $qE_o$ , where  $q=-e$ ), the ratio of the initial and final current will be:

$$I_m \propto \sqrt{\frac{m}{q}}, \quad (3.40)$$

which follows that:

$$\frac{I_m}{I_{mo}} = \sqrt{\frac{m}{m_o}}, \quad (3.41)$$

The relationship expressed in Eq. 3.41 plays a crucial role during ion beam acceleration, providing a method for identifying and selecting the desired ion species by adjusting the currents of the electromagnet. Following  $N_2$  stripping, a spectrum of charge states is produced, resulting in a range of different ion energies. Therefore, using a HE magnet is necessary to bend the ion beam towards the appropriate beamlines for the experiment. Considering the geometry of the magnet and the accelerating axis, we can derive from Eq. 3.42 and 3.43 that [182]:

$$\sin\theta_B = \frac{d_o}{R_B}, \quad (3.42)$$

$$\sin\theta_B = \frac{qB_o d_o}{\sqrt{2mE}}, \quad (3.43)$$

where  $B_o$  is the magnitude of the magnetic field,  $d_o$  is the length of the magnet occurring in the direction of the incident beam, and  $q, m$ , and  $E$  are the charge, mass, and average kinetic energy of the particle. When ions are accelerated in the CLASS accelerator, or TANDEM accelerators generally, the final energy produced depends on both the initial charge state ( $Z(-)$ ) and the final charge state ( $Z(+)$ )

and the terminal potential ( $V_o$ ) of the high voltage, given by Eq. 3.44 [182]:

$$\sin\theta_B = \frac{\sqrt{e}B_o d_o}{\sqrt{2mE}} \left( \frac{Z_{(+)}}{\sqrt{Z_{(-)} + Z_{(+)}}} \right), \quad (3.44)$$

From the Eq. 3.44 we can deduce that:

$$I_m \propto \frac{\sqrt{mV_o(Z_{(+)} + 1)}}{Z_{(+)}}, \quad (3.45)$$

With Eq 3.45 it is easier to determine the charge state to the energy of the ion selected in the HE electromagnet.

### 3.7.4 Sample irradiation and fluence

The samples of Au and Lu-Al were irradiated in the RBS/NRA beamline chamber with 0.5 MeV protons incident at  $45^\circ$  to an ellipsoidal beam aperture of surface area  $0.04443 \text{ cm}^2$ . The beam parameters and fluence ( $\Phi$ ) are summarized in the Table 3.9. The proton fluence (Dose) was estimated based on the total collected charge on the area of the samples, thus: The number of protons ( $n_p$ ) impinging on the surface was calculated for a collected charge  $Q$  is given by:

$$n_p = \frac{\text{CollectedCharge}(Q)[C] \times 1\text{Proton}}{1.602 \times 10^{-19}[C]}, \quad (3.46)$$

where  $1.602 \times 10^{-19}C$  is the charge of 1 proton. The proton fluence ( $\Phi$ ) was now determined by the ratio of the number of protons impinging the irradiation material surface area.

$$\Phi = \frac{n_p}{A}, \quad (3.47)$$

where  $A$  is the Area of the irradiated surface. The beam aperture was an ellipsoidal patch with a radius of the major ( $a$ ) and minor surfaces ( $b$ ), 2 mm and 1 mm respectively at  $45^\circ$ . Hence the  $A$  is given by:

$$A = a \times b \times \pi = 0.2 \times \pi \times 0.1 \sin 45^\circ = 0.04443 \text{ cm}^2, \quad (3.48)$$

Using Eq. 3.46 and 3.47 the proton fluence was calculated and presented in Table 3.9.

**Table 3.9:** Irradiation parameters and fluence (Dose)

| Parameter [Units]                                   | Irradiation Batches   |                       |                       |
|-----------------------------------------------------|-----------------------|-----------------------|-----------------------|
|                                                     | (a)                   | (b)                   | (c)                   |
| Ion Energy(E) [MeV]                                 | 0.5                   | 0.5                   | 0.5                   |
| Temperature(T) [°C]                                 | 45                    | 40                    | 33                    |
| Current(I) [ $\mu A$ ]                              | 1.1                   | 1.1                   | 1.1                   |
| time(t) [s]                                         | 500                   | 1500                  | 3000                  |
| Collected Charge(Q) [mC]                            | 0.55                  | 1.65                  | 3.30                  |
| Area(A) [cm <sup>2</sup> ]                          | 0.04443               | 0.04443               | 0.04443               |
| Fluence( $\Phi$ ) [ <i>protons/cm<sup>2</sup></i> ] | $0.77 \times 10^{17}$ | $2.32 \times 10^{17}$ | $4.64 \times 10^{17}$ |

### 3.7.5 Displacements Per Atom (dpa) of irradiated sample materials

From the beam fluence ( $\Phi$ ) values in Table 3.9 and the damage energy ( $T_{dam}$ ) values  $5.5 \times 10^{-2}$  and  $3.27 \times 10^{-2}$  eV/ $\text{\AA}$ -ion for Au and Lu-Al respectively obtained in SRIM-2013 output at ion energy 0.5 MeV, the dpa values are calculated based on Eq. 3.37 as shown in Table 3.10

**Table 3.10:** Displacements Per Atom (dpa) of irradiated sample materials

| Material | Irradiation Batches |        |        |
|----------|---------------------|--------|--------|
|          | (a)                 | (b)    | (c)    |
| Au       | 0.013               | 0.039  | 0.078  |
| Lu-Al    | 0.0011              | 0.0034 | 0.0068 |

## 3.8 Scanning Electron Microscopy (SEM) characterization technique

This section focuses on the characterization study of damage in Lu-Al alloy and Au samples. Scanning Electron Microscopy (SEM) was employed to examine both the composition and surface topology.

### 3.8.1 Description of Zeiss GeminiSEM 460

The Zeiss GeminiSEM 460 (Gemini2) at the Massachusetts Institute of Technology Nano Laboratories (MIT-Nano) is shown in Figure 3.7. This instrument is a High-resolution SEM (HR-SEM) that has a resolution of 0.6 nm and 0.7 nm at 30 and 20 kV voltage respectively. The analytical resolution of 2.0 nm at 15 kV, 5 nA current in a working distance (WD) of 8.5 mm enables it to perform analysis rapidly by switching seamlessly from low to high power. The FE-gun accelerates electrons through 0.02 - 30 kV with a probe current of 3 pA - 40 nA. The schematics of the Zeiss GeminiSEM 460 is shown illustrated in Figure 3.8.



**Figure 3.7:** GeminiSEM 460, MIT-nano (Picture taken by the researcher)

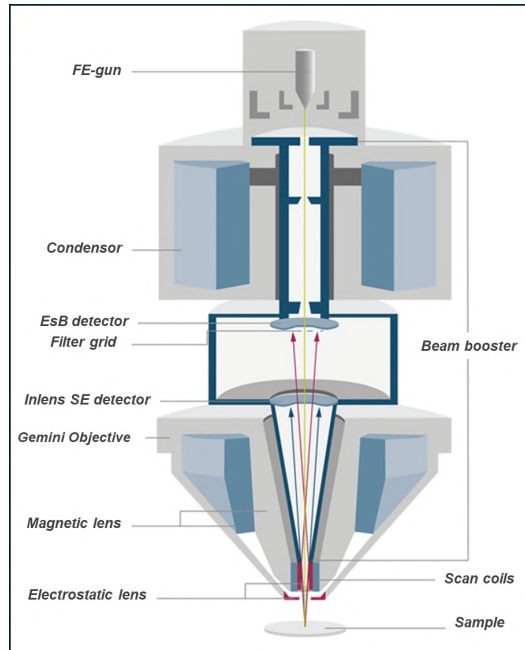


**Figure 3.8:** GeminiSEM 460 Schematics, MIT-nano (credit: RAVE SCIENTIFIC <https://ravescientific.com/products/high-resolution-fesem-zeiss-geminisem>)

The Gemini2 is integrated with a Condensor, Filter grid, Beam booster, and electrostatic and magnetic lens fields to enhance the optical performance.

### **3.8.2 SEM sample handling and imaging**

Bulk foil samples of Lu-Al and Au were cleaned with acetone and securely mounted on a stub using copper tape. The prepared sample holder was then carefully inserted into the GeminiSEM 460 sample stage. Following this, the sample chamber was vented, and the designated sample was placed on the stage. The system was then pumped to achieve the desired pressure. By selecting the appropriate sample position (1-7) button on the aluminum stub, the sample of interest was chosen.

Subsequently, the Working Distance (WD) was finely adjusted to approximately 5.8 mm, and the beam was activated for imaging. Beam parameters were configured with a high voltage value (EHT) set at 15.0 kV and a beam current of 1.0 nA for optimal high-resolution imaging. The morphology of the samples was meticulously scanned using the Secondary Electron (SE) mode of the microscope. To acquire the chemical composition of the samples, an energy dispersive spectroscopy (EDS) detector was inserted and the beam current was adjusted to suit the mode for the acquisition of the chemical composition of the samples.

### **3.8.3 Data Analyses of SEM images**

The data obtained from the imaging was processed using the SEM inbuilt software and Origin for the spectral plots. Further analysis of the images was deemed unnecessary, as no defects were observed after sample irradiation. This was expected, given the low displacement per atom (dpa) the materials were exposed to. Damage at that level will not be visible at the resolution of the SEM. However, the SEM was very useful for the pre-irradiation characterization of the samples.

## **3.9 X-Ray Diffraction (XRD) technique**

X-Ray Diffraction (XRD) was employed to analyze the sample diffraction spectra pattern, micro-strain ( $\epsilon$ ), average crystallite size (D), and average dislocation

density  $\delta$  in the materials.

### 3.9.1 Description of PANalytical X'Pert Pro XRD

The PANalytical X'Pert Pro MPD, depicted in Figure 3.9, is an XRD machine featuring a sealed 1.8 KW power X-Ray tube source with a Cu target of  $K_\alpha$  wavelength ( $=1.5406 \text{ \AA}$ ) at the MIT Material Research Science and Engineering Centre (MRSEC) [183].

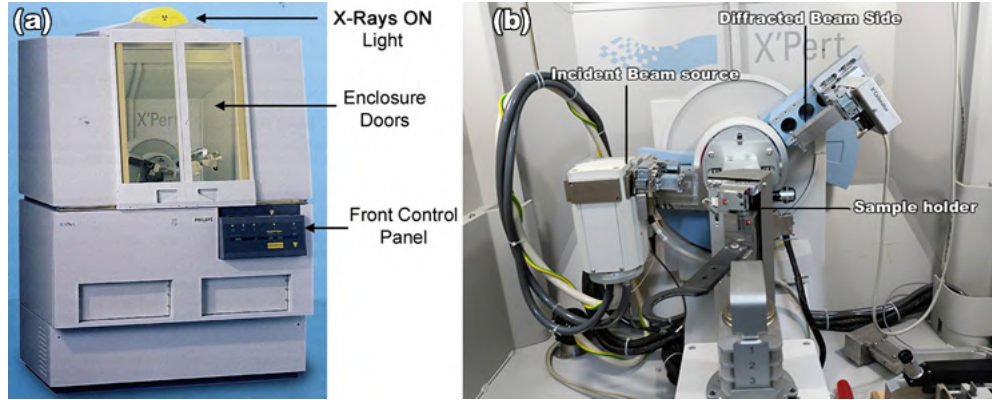


**Figure 3.9:** (a) PANalytical X'Pert Pro MPD MIT MRSEC [183] (b) Internal features of the X'Pert Pro MPD showing the Cu X-ray beam source, sample holder and the Diffracted beam side

The instrument is configured in the Bragg-Brentano para-focusing optical geometry with a high-speed, high-resolution X'Celerated detector, which covers a  $2\theta$  range of  $2.122^\circ$ , allowing for the simultaneous acquisition of a broad section of the diffraction pattern. The precision of the goniometer for the XRD machine ranges between  $0.0001^\circ$  to  $0.0005^\circ$  in  $2\theta$ .

### 3.9.2 XRD sample handling and measurements

Sample preparation for XRD analysis was straightforward for the smooth, flat-surface bulk solid samples under investigation. The bulk foil samples of Lu-Al and Au were meticulously cleaned with acetone to prevent surface contamination. Subsequently, they were mounted on a zero-background sample holder. Activating the digital height gauge involved pressing the green button to align the sample slot base. Samples were then inserted into the sample holder, ensuring that the sample's flat surface faced the top plane of the holder, making them ready for measurements.

### **3.9.3 Method of measurement and estimation of damage parameters**

#### **(a) Data acquisition method**

To acquire XRD data from the X'Pert Pro PMD, the X'Pert Data Collector program of the instrument was initiated, and the tube power was set to a tension of 40 kV and 10 mA. The Programmable Divergence Slit (PDS) was employed to generate the incident beam-side optic, while the X'Celerator detector was utilized on the diffracted beam side.

Subsequently, unirradiated samples of Lu-Al and Au were loaded onto the instrument's sample stage, and the power was adjusted to a tension of 45 kV and a current of 40 mA for X-ray production. Data from the sample were acquired with a step size of 0.01 deg in the  $2\theta$  range from 10-90°. The same procedure was replicated for the irradiated samples, and the data collected using the computer's automated data acquisition software were saved in .XRDML format. Later, these files were converted to the .xy format for ease of processing.

#### **(b) Estimation of damage parameters**

In XRD studies, microstrain and crystallite size serve as indicators of defects in the microstructure of crystalline materials, both contributing to the broadening of the peaks observed during XRD analyses. The effects of Peak line-broadening in polycrystalline samples can be analyzed to derive the microstrain in their microstructure. However, it's essential to note that other factors, such as solid solution inhomogeneity and temperature, also contribute to peak broadening. Furthermore, the average dislocation density ( $\delta$ ) in the material's microstructure can be determined from the crystallite size obtained. The microstrain can be assessed using the Williamson-Hall plot of  $\cos \theta$  versus  $4\sin \theta$ , where  $\theta$  represents the angle of diffraction [150]. Some of the limitations of this method that affect the uncertainties include the assumed uniform strain distribution and isotropic broadening, which may often not be characteristic of some material conditions, leading to potential inaccuracies [184]. Also, there could be high strain levels which may cause non-linearity between

strain and peak broadening, further complicating the analysis [185]. Furthermore, this method is sensitive to noise in the diffraction data, which can introduce considerable uncertainties in the microstrain estimates. Noise smoothening was applied before the data was subjected to analysis. The crystallite size ( $D$ ) on the other hand is determined using Scherrer's method in Eq. 3.49 [186].

$$D = \frac{K\lambda}{\beta \cos \theta}, \quad (3.49)$$

The broadening due to crystallite size is given by Eq. 3.50

$$\beta_{Crystallitesize} = \frac{K\lambda}{D \cos \theta}, \quad (3.50)$$

Eq. 3.50 is valid for crystallite sizes  $D \leq 200$  nm. Also, the broadening due to microstrain is given by Eq. 3.51

$$\beta_{microstrain} = 4\varepsilon \tan \theta, \quad (3.51)$$

The Williamson-Hall plot peak broadening can be estimated thus:

$$\beta_{Total} = \beta_{Sample} = \beta_{crystallitesize} + \beta_{microstrain}, \quad (3.52)$$

$$\beta_{Total} = \frac{K\lambda}{D \cos \theta} + 4\varepsilon \tan \theta, \quad (3.53)$$

Eq. 3.53 can be written in a simplified form as Eq. 3.54

$$\beta_{Total} \cos \theta = \varepsilon(4 \sin \theta) + \frac{K\lambda}{D}, \quad (3.54)$$

Eq. 3.54 yields a linear form when  $\beta \cos \theta$  is plotted against  $4 \sin \theta$  (Williamson-Hall plot). The slope of the graph gives the microstrain ( $\varepsilon$ ) and the crystallite size ( $D$ ) is obtained from the intercept of the graph. The dislocation density ( $\delta$ ) is the inverse square of the crystallite size with units in per square meters ( $\text{m}^{-2}$ ) shown in Eq. 3.55.

$$\delta = \frac{1}{D^2}, \quad (3.55)$$

The average crystallite size, microstrain, and dislocation density were calculated for the materials pre and post-irradiation.

### **3.9.4 Method of analysis**

The data from unirradiated and post-irradiated samples of Lu-Al and Au obtained through the X'Pert Pro MPD X-ray diffractometry were imported into Origin 2018 software. Subsequently, XRD spectra plots for the samples were generated and fitted using the Lorentzian function. The positions of Bragg's peaks and the Full Width at Half Maximum (FWHM) were then determined. Based on these values,  $4\sin\theta$  and  $\beta \cos\theta$  were estimated. These derived values were utilized in constructing the Williamson-Hall plot, allowing for the calculation of average microstrain ( $\varepsilon$ ), crystallite size ( $D$ ), and dislocation density ( $\delta$ ) for both materials.

## **3.10 Transmission Electron Microscopy (TEM) characterization technique**

This section discusses the TEM characterization technique and all the methods that were used to prepare samples, sample analyses (imaging), and methods that were utilized in analyzing the data obtained.

### **3.10.1 TEM sample preparation using Focused Ion Beam (FIB)**

TEM lamellae were prepared through the extraction of a transverse section from the specimens utilizing Focused Ion Beam (FIB) technology within the FEI Helios NANOLAB Dual-Beam 650 Scanning Electron Microscope (FIB-SEM), as illustrated in Figure 3.10(a).

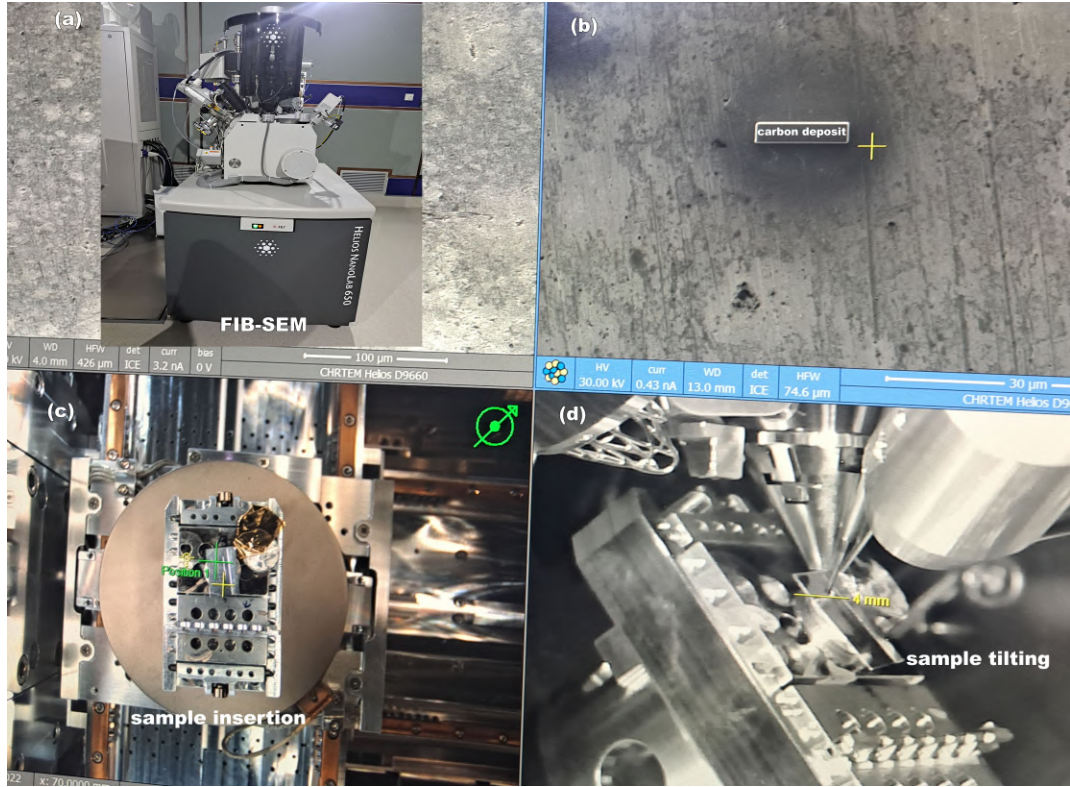


**Figure 3.10:** (a) showing the Helios NanoLab 650 FIB-SEM at CHRTEM Nelson Mandela University, South Africa, (b) carbon deposition on a site of interest (SOI) (c) sample insertion and positioning in the FIB-SEM, (d) sample tilted to dig trenches around to excavate the transverse section of the sample ( Picture taken by the researcher)

The specimens were carefully positioned in the FIB-SEM, aligning them concentrically with the electrons and ion beams and set at a eucentric height to maintain a consistent field of view and focus. This precaution aimed to prevent any lateral movement during the tilting of the sample. A carbon layer with dimensions  $10 \times 2 \times 2 \mu\text{m}$  in length, breadth, and depth was deposited at the Site Of Interest (SOI) with the sample positioned at  $0^\circ$ .

The deposition was facilitated by an Omniprobe Autoprobe<sup>TM</sup> (Omni probe) affixed to the sample during the carbon deposition process. Subsequently, the sample was tilted by  $52^\circ$  to excavate the trenches around the SOI as depicted in Figure 3.10(b)-(d).

The trenches were excavated on the bulk sample surrounding the Site of Interest (SOI), with approximate dimensions of  $22 \times 6 \times 6 \mu\text{m}$  in length, breadth, and

depth (refer to Figure 3.11(a)). This process involved the use of Ga ions at a current of 46 nA, along with sample undercutting, as illustrated in Figure 3.11(b). The current was subsequently reduced to 9 nA to eliminate the Ga ion deposit on the sample. This reduction was achieved by tilting the angle between  $49^\circ$  and  $55^\circ$ , as depicted in Figure 3.11(c).

Sample extraction occurred in four stages: an L-shaped undercut was created to partially detach the sample, followed by attaching the Omni Probe and detaching the sample from the bulk. This involved extending the L-undercut to a U-shaped undercut, as shown in Figure 3.11(b), followed by lifting the sample.

After the undercut, the Omni probe was retracted. By employing a stage rotation of  $60^\circ$  at  $0^\circ$  tilt, the Cu-grid was positioned at a concentric and eucentric height. The Omni probe, with the previously retracted sample, was then re-inserted to the eucentric height, where the sample was mounted to the Cu-grid using carbon deposition, and the Omni probe was detached.

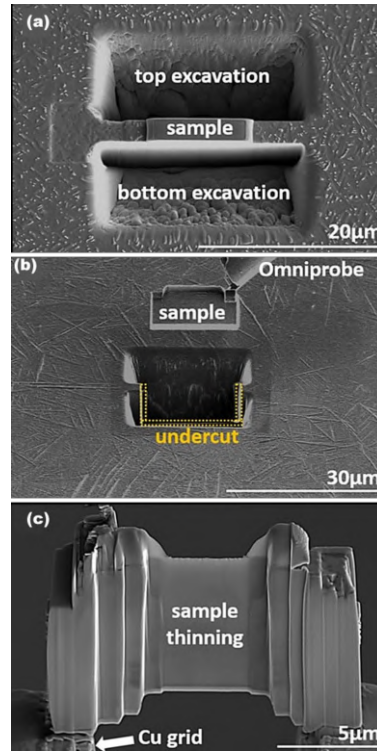


**Figure 3.11:** (a) Excavation of lamellae by digging trenches top and bottom at the SOI, (b) Sample undercut and extraction using the Omniprobe Autoprobe<sup>TM</sup> (c) Sample attached to the Cu grid for thinning

Utilizing a voltage of 30 kV and pressure of 781 Pa, with an angular tilt between  $50^\circ$  and  $53^\circ$ , Ga ions were employed to roughly thin the opposite planes of the sample, as depicted in Figure 3.11(c). The beam power was gradually reduced during fine milling to the 5 kV, 3.2 nA settings, resulting in electron-transparent lamellae with thicknesses less than 100 nm.

### **3.10.2 Basic description of TEM**

Two Transmission Electron Microscopes were utilized in this study: the JOEL JEM-F200 TEM and the FEI TECNAI (G2 Spirit TWIN) TEM.

#### **(a) JOEL JEM-F200 (F2) TEM**

The JOEL JEM-F200, as illustrated in Figure 3.12(a), represents a next-generation multipurpose operation field emission TEM boasting superior spatial resolution and enhanced analytical performance. Equipped with a Quad-Lens Condenser system, a cutting-edge technology comprising a four-stage probe-forming optical system, this microscope enables independent control of the intensity and convergence angle of the electron beam, facilitating ease of operation.

The JEM-F200 offers scanning capabilities with both high and ultra-high point resolutions of  $2.3 \text{ \AA}$  and  $1.9 \text{ \AA}$ , respectively. Additionally, the instrument incorporates the Scanning Transmission Electron Microscopy-High-angle annular dark-field imaging (STEM-HAADF) mode, featuring resolutions ranging from  $1.6 \text{ \AA}$  to  $1.4 \text{ \AA}$  for both the Cold and Schottky Field Emission Gun (FEG) in higher and ultra-high resolutions, with a beam accelerating voltage between 20 to 200 kV. These advanced features contribute to the microscope's ability to provide high-quality imaging of the micro/nanostructure of materials.

#### **(b) FEI TECNAI (G2 Spirit TWIN) TEM**

The FEI Tecnai (G2 Spirit TWIN), presented in Figure 3.12(b), is also a versatile Transmission Electron Microscope (TEM) boasting high-resolution capabilities and excellent contrast. This instrument has an accelerating beam voltage that reaches up to 120 kV. With its TWIN lens, it enables the observation of nanostructures

with up to  $2.0 \text{ \AA}$  lattice resolution and a magnification of up to  $3.6 \text{ \AA}$  point-to-point resolution. One noteworthy feature of this microscope is its maximum eucentric inclination, ranging from  $\pm 80^\circ$ . This capability makes it particularly well-suited for observing microstructural defects, such as dislocation loops, that necessitate tilting between the crystal lattice plane ( $g$ ) and the Burgers vector ( $b$ ). Both the FEI Tecnai (G2 Spirit TWIN) and the JOEL JEM-F200 were employed in this study for the observation of microstructural defects in the materials.

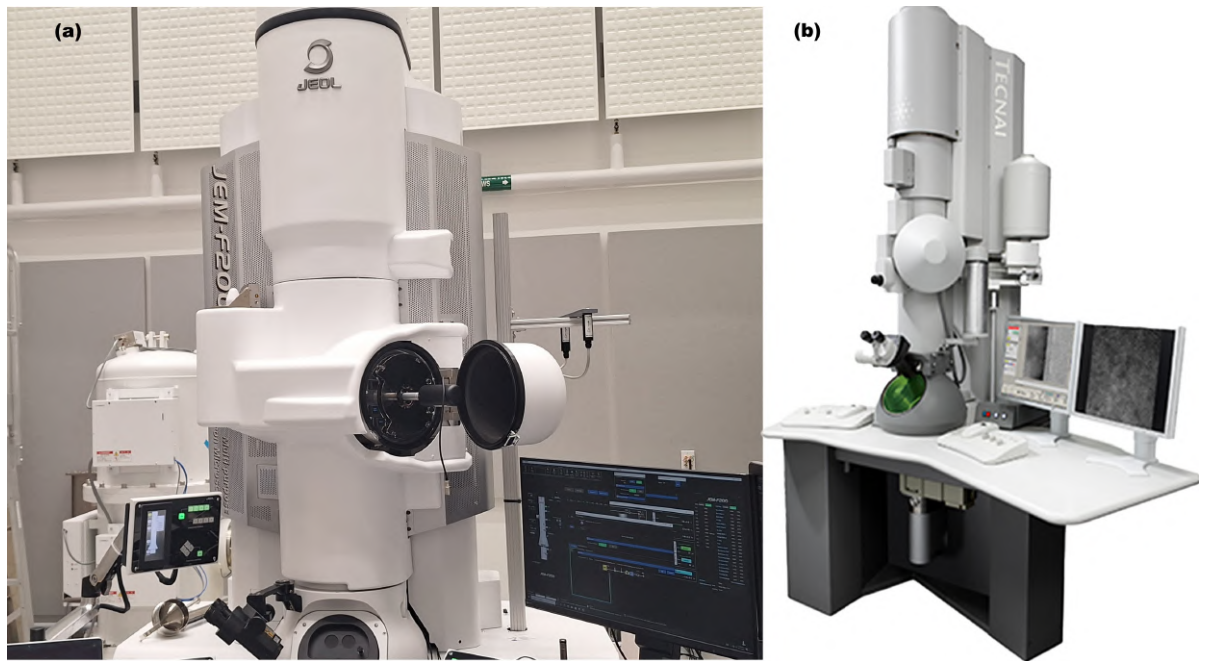


**Figure 3.12:** (a) JOEL JEM-F200 TEM at Centre for Nanoscale systems, Harvard University (b) FEI Tecnai (G2 Spirit TWIN) TEM at MIT Material Research Laboratory. ((a) Picture taken by the researcher (b) Credit MIT Nano <https://nanousers.mit.edu/characterizenano/focus-facilities/electron-microscopy/fei-tecnai-multipurpose-digital-tem> )

### 3.10.3 Loading of sample to TEM

To prepare the sample for observation, it must be inserted into the Transmission Electron Microscope (TEM). This process involves unlocking the screws on the TEM sample holder with 3x half turns, followed by gently lifting the copper clamp. The Cu-grid, carrying the samples of Au and Lu-Al on fingers A and D, is then placed on the sample holder, and the screws are locked, though not excessively tight.

Before inserting the holder, the vacuum is checked to ensure readiness. The sample holder is gently pushed with a finger to set it on the goniometer stage. The specimen chamber is pumped until the vacuum reaches a current of 49-51  $\mu\text{A}$  and a high vacuum pressure of  $< 1.0 \times 10^{-3}$  Pa. Subsequently, the holder data is selected, and the sample is completed and automatically inserted into the stage. The case is then closed, shut, and clamped in place, making it ready for TEM alignment.

#### **3.10.4 TEM alignment for image acquisition**

To achieve precise imaging, adjustments were made to the Transmission Electron Microscope (TEM) by centering the objective lens rotation in normal operating mode. Following sample insertion, the valve was opened, and the sample was maneuvered to the center of the TEM screen. The magnification was then increased to 50,000X, and brightness was enhanced to create a sharp cross on the screen. The sample of interest was located on the Cu-grid. Subsequently, the Condenser Lens Aperture was inserted and centered using the controls. Further magnification, up to approximately 200,000X, was applied to bring the sample into focus, eliminating Fresnel fringes.

Stigmatization and wobble functions were adjusted until no lateral movement was visible, ensuring the optimal configuration of the microscope to prevent artifacts in the acquired images. Finally, the camera was set on the viewing screen with the Fourier Transform window to enable the live acquisition of micrographs. With the microscope configured in this manner, various modes can be employed for data acquisition.

#### **3.10.5 Two beam condition for TEM imaging of defects**

The TEM, configured as described above, can be utilized for examining diffraction contrast using any form of electron scattering.

However, the observation of defects such as dislocation loops in the microstructure of materials necessitates strong diffraction contrast in either bright-field (BF) or dark-field (DF) mode. Therefore, a specific TEM condition known as the "Two-

Beam” condition, illustrated in Figure 3.13, was employed. This condition involves aligning  $\alpha$  and  $\beta$  by tilting the sample to an angle where the Kikuchi line leads to a zone axis. In this alignment, the diffraction mode features the direct beam and one dominant Bragg’s beam coincidence occurring prominently in the crystal diffraction pattern in reciprocal space mapping. This setup facilitates the easier observation of crystal defects like dislocation loops, dislocations, and voids/bubbles.

Typically, dislocations in TEM are not imaged directly. Instead, the strain fields around the dislocations are imaged, producing a lattice distortion from the dislocation core. For a dislocation to be visible in the TEM image, the dot product of the specific reciprocal lattice vector  $\mathbf{g}$  and the Burgers vector  $\mathbf{b}$  must not be zero ( $\mathbf{g} \cdot \mathbf{b} \neq 0$ ). Consequently, the local lattice bonding scatters electrons in a different direction, creating a diffraction contrast that renders the diffraction vector  $\mathbf{g}$  not parallel to the Burgers vector  $\mathbf{b}$ , making the dislocation visible.

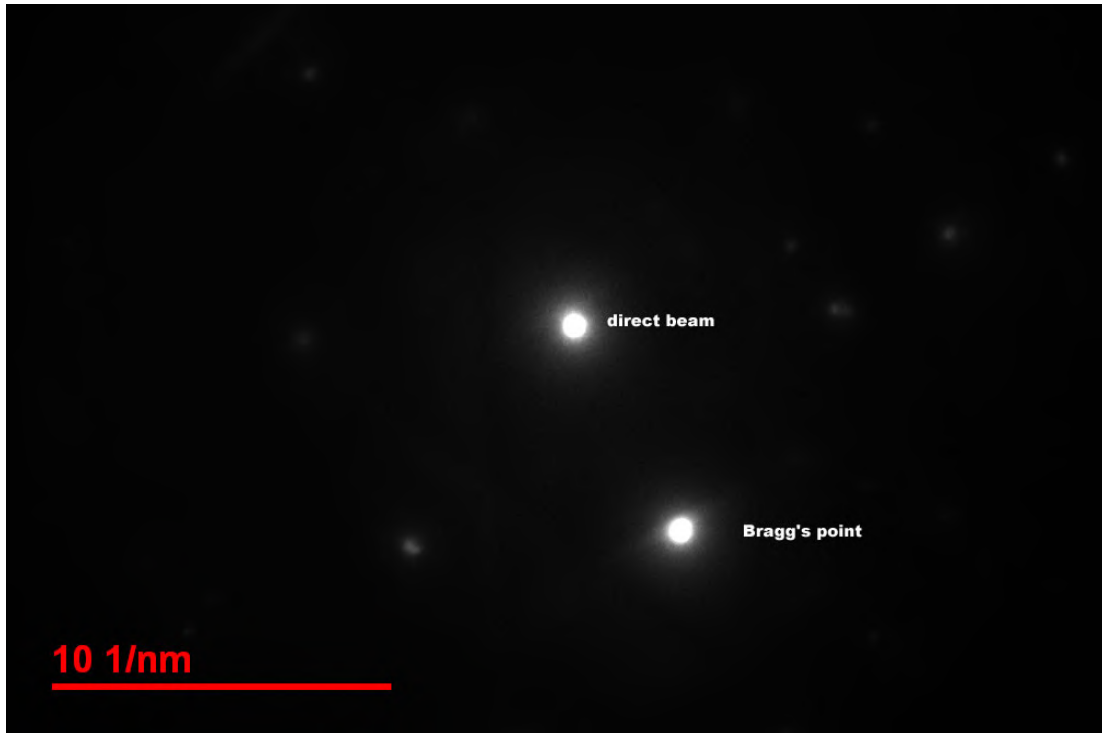


**Figure 3.13:** Two-beam condition diffraction pattern for Au in the [011] zone axis

This condition was achieved by tilting the sample to an angle that aligns the Kikuchi lines (refer to Figure 3.14(a)) to a specific zone axis. In this configuration, the electron beams undergo diffraction twice—initially through inelastic or incoherent

scattering from the lattice planes, followed by Bragg's diffraction. Within the same set of lattice planes, two Kikuchi lines are produced for each set, one with bright intensity known as the excess line, and the other with low intensity representing the defect line, as illustrated in Figure 3.14(b).

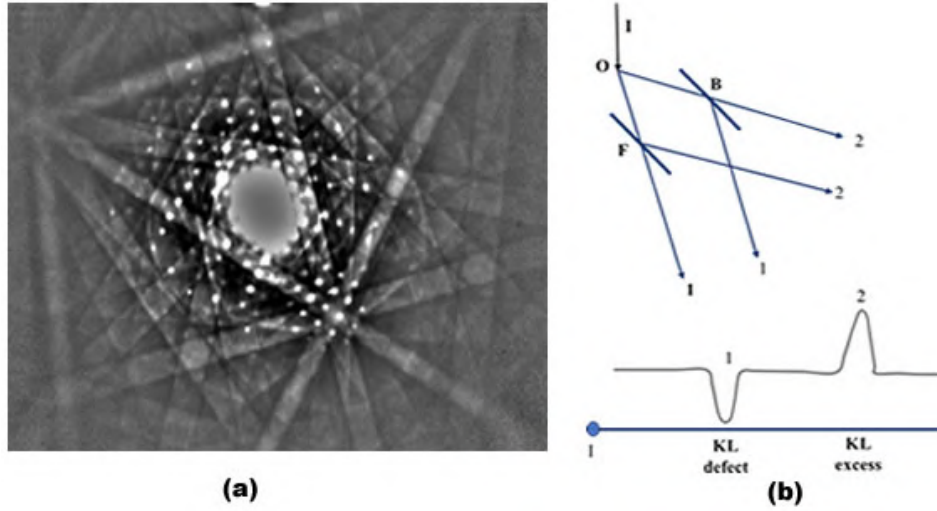


**Figure 3.14:** (a) Kikuchi patterns in transmission mode [187] (b) Illustration of two-beam condition in reciprocal mapping

These Kikuchi lines served as road maps while the samples were tilted to achieve the Two-Beam condition in the reciprocal space mapping. By utilizing the Two-Beam condition, microstructural defects were observed in the  $[011]$  and  $[110]$  zone axes.

### 3.10.6 TEM image data analyses

The data from Energy Dispersion Spectroscopy and micrographs were analyzed using Fiji ImageJ. Fiji ImageJ, an open-source Java source code designed for processing various image data types and file formats, proved suitable for TEM image analyses. This software facilitated the estimation of defect size and defect densities in the microstructure of materials post-irradiation.

In the quantitative analysis of the images, the measurement of defect size and subsequent calculation of the average dislocation density were performed. For dislocation lines, the average dislocation density in the intersection area of the surface ( $A_s$ ) and thickness ( $t$ ) of the sample was computed as the ratio of the total length

(L) of dislocation lines to the volume ( $V_s$ ) of the sample, as given by Eq. 3.56 [63].

$$\delta = \frac{L}{V_s}, \quad (3.56)$$

$$\delta = \frac{L}{A_s \times t}, \quad (3.57)$$

where,  $\delta$  is expressed in units of  $\text{m}/\text{m}^3$  or simply  $\text{m}^{-2}$ , where  $A$  represents the area in which the dislocations intersect the surface (in  $\text{nm}^2$ ), and  $t$  is the thickness of the analyzed TEM lamellae in nanometers (nm). The thicknesses for the Au and Lu-Al lamellae were approximately 97.4 nm and 114 nm, respectively. Furthermore, the dislocation line density can be calculated by determining the ratio of the number ( $N$ ) of dislocation lines intersecting the sample's surface to the area ( $A$ ) of the surface.

$$\delta = \frac{N}{A}, \quad (3.58)$$

For dislocation loops, Eq. 3.57 was modified where the length  $L$  became the mean perimeter ( $P$ ) of the dislocation loops multiplied by the number of loops ( $n$ ) in the intersecting surface area. For circular loops the average dislocation density was

$$\delta = \frac{P \times n}{A_s \times t} = \frac{2 \times \pi \times r \times n}{A_s \times t}, \quad (3.59)$$

Using the Eqs. 3.58 and 3.59 the dislocation density was calculated for the dislocation lines and the loops respectively.

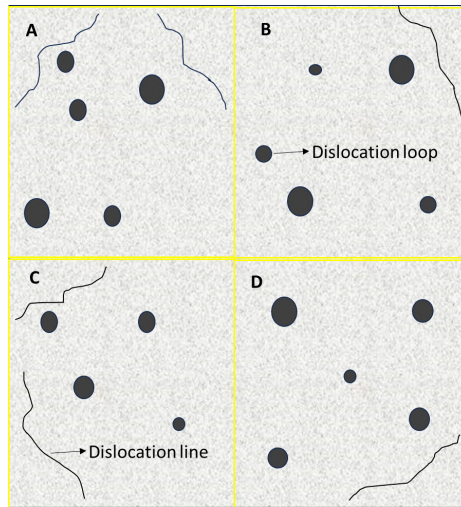


**Figure 3.15:** Estimation of dislocation density in TEM micrographs

where the dislocation loops or the lines were counted and their sizes were estimated using ImageJ software. Dislocation density was calculated with the accompanied uncertainties.

### **3.11 Flash Differential Scanning Calorimetry (F-DSC) characterization technique**

Differential Scanning Calorimetry (DSC) serves as a thermal analysis technique, gauging the heat flow in or out of a sample during its heating or cooling. Although DSC is not conventionally employed for direct measurement of material damage, it yields valuable insights into alterations in a material's thermal properties as discussed in section 2.10.2 (d). These include enthalpy changes associated with phase transitions or reactions, which can be correlated with damage or structural modifications identified through other characterization methods such as Transmission Electron Microscopy (TEM), X-ray Diffraction (XRD), and Positron Annihilation Spectroscopy (PAS).

DSC assesses defects stored in the material in the form of radiation-induced defects. The quantification of energy, known as Wigner energy (W) [154], is proposed by Wigner. This energy is released during the radiation damage recovery process known as annealing, where the excess energy from the defect is emitted. The strength of this technique lies in its capability to measure small-scale radiation damage defects in metals that remain unobservable through alternative imaging techniques such as TEM, PAS, resistivity measurement, and XRD.

Each of these characterization methods has limitations that hinder their ability to comprehensively identify defects in irradiated materials [151, 188]. TEM observes defects up to the limits of its resolution, rendering cluster defects below its resolving power invisible. The excess energies released by irradiated materials due to defects provide a more accurate account of these small defect clusters [151].

The F-DSC is an advanced nanocalorimetry technique with enhanced features that

enables the measurement of the energy release during damage annealing for small sample size in nanograms (ng). The F-DSC method has been evaluated chiefly for organic compounds. At present, only a few works have reported damage to annealing processes on metals this is discussed earlier in section 2.10.2(d). This section presents the methods for damage characterization using F-DSC and outlines some preliminary results obtained from the characterization of Lu-Al, a metallic binary alloy.

### 3.11.1 Methods of Defects Characterization with Flash DSC

The Flash DSC (F-DSC) instrument shown in Figure 3.16(a) was utilized in this investigation is a chip-based nanocalorimeter located at the MIT Nuclear Science and Engineering Laboratory, USA.

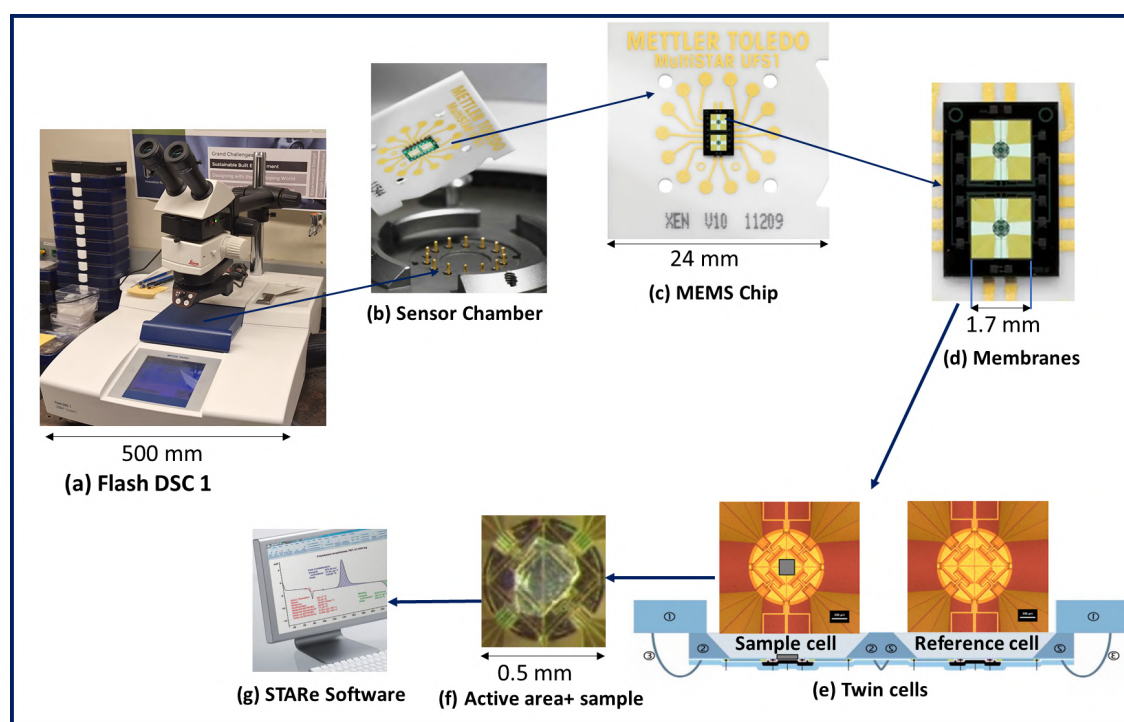


**Figure 3.16:** Description of the Mettler Toledo Flash DSC 1 (a) Flash DSC 1 instrument (b) The MultiSTAR UFS 1 Chip-Sensor Chamber where the chip with the sample is positioned directly. Sensors constitute 16 thermocouples with high sensitivity and great temperature resolution (c) The MEMS-Based technology chip (d) Illustrates the membrane of the chip hosting a twin cell (e) Schematic of the Twin cells low-temperature UFS-1 chip with sample cell and reference cell for calibration. (f) showing the active area on the sample cell where the sample is placed. (g) STARe Software for data acquisition. Images (b)-(g) taken from the Mettler Toledo Flash DSC 1 brochure.

The F-DSC features two sensors situated on a  $\text{Si}_3\text{N}_4$  membrane with a thickness of approximately  $2\mu\text{m}$ , enclosed within an  $\text{Al}_2\text{O}_3$  base frame, as depicted in Figure 3.16 (b)-(f).

The Mettler Toledo Flash DSC 1 has ultra-high heating and cooling rates, regulated by the low-temperature UFS-1 twin microchip illustrated in Figure 3.16 (e). This chip can achieve a heating rate of 2,400,000 K/min up to  $520^\circ\text{C}$  and a cooling rate of 240,000 K/min down to approximately  $-95^\circ\text{C}$ . Moreover, the Flash DSC 1 can employ the UFH-1, a high-temperature chip ( $> 1000^\circ\text{C}$ ) with even higher heating rates of around 3,000,000 K/min. It allows an intra-cooling temperature of  $-95^\circ\text{C}$  with an ultra-high cooling rate of 2,400,000 K/min when such temperature ranges are required. In this work, the UFS-1 chip was used for measurements. The active area where the sample is positioned has a 0.5 mm aluminum plate diameter. The heated region of the chip is composed of a gold (Au) disk with a diameter of 0.1 mm.

### 3.11.2 Sample Preparation

The sample preparation procedure for metallic solids is straightforward. Both irradiated and unirradiated samples of Lu-Al foil were reduced to about 100 ng in size by sectioning them with a scalpel. Subsequently, they were delicately transferred onto Chip's sample cell using an eyelash and with the aid of the microscope integrated on the F-DSC to ensure the sample was placed on the active area Figure 3.16 (e) of the sample cell. In the case of the irradiated foil, the sample was exclusively extracted from the region that was subjected to proton irradiation.

### 3.11.3 Flash DSC Measurements

The measurement conducted on Lu-Al using Flash DSC followed the methodologies outlined by Hirst et al. and Mathot et al. [148, 151, 189]. The microchip batch, provided by Mettler Toledo, was pre-calibrated using an electrical method. For damage annealing measurement, the sample was positioned on the active area of the UFS 1 chip. The procedure involved rapidly heating and cooling the samples across a wide temperature range of  $20^\circ\text{C}$  to  $450^\circ\text{C}$ . This cycle was repeated five

times, and temperature data (Temperature of sample ( $T_s$ ) and Temperature of reference ( $T_r$ )) along with power ( $P$ ) were recorded using the STARe software, as depicted in Figure 3.16 (g). The acquired data were analyzed using a MATLAB code to obtain curves for the six heating and cooling cycles.

# Chapter 4

## Results of radiation damage: Simulation, Experiments and characterization

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### 4.1 Numerical results from MCNP6.2 calculations

The radiation damage calculations were carried out using MCNP6.2, and this section presents the numerical results detailing the interactions of neutrons and photons with the sample materials. The calculated neutron flux tally, dpa tally, and heat energy deposition tally are reported along with their corresponding uncertainties, represented as fractional standard deviations (fsd). MCNP6.2 inherently performs these uncertainty calculations on the tally results, and the outcomes are illustrated in the Tally Fluctuation Chart (TFC), provided in Appendix A.

The TFC aligns with the success criteria of MCNP, displaying a random behavior with a relative error of less than 0.1. The Figure Of Merit (FOM) maintains a constant value, indicating stable and reliable tally results. The Variance Of Variance (VOV) remained below 0.1, further reinforcing the precision and accuracy of the calculated results.

#### 4.1.1 Neutron flux

The normalized neutron flux-energy spectra results for the Au sample are illustrated in Figure 4.1. Table 4.1 provides details on the average flux incident on the Au sample across different neutron energy regimes, including thermal, resonance, and fast neutron regimes.

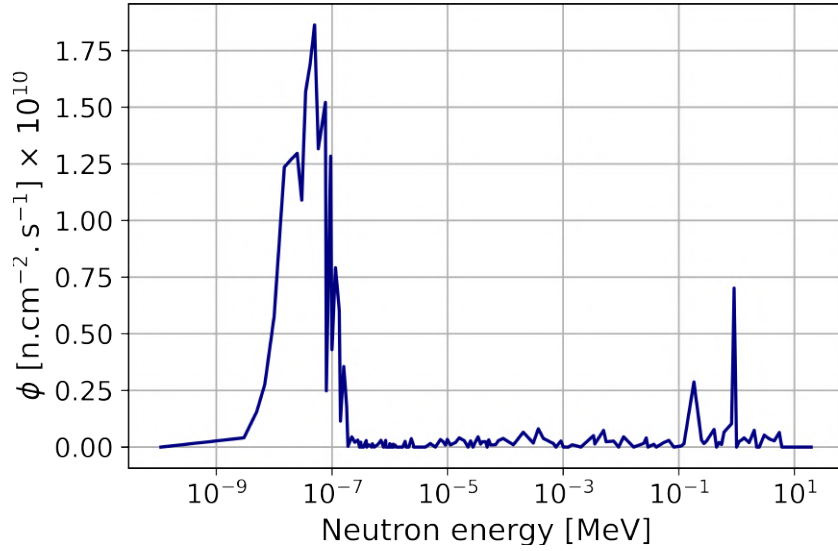


**Figure 4.1:** Neutron Flux-Energy profile for Au. [54]

Neutron fluxes were classified into three energy regimes, aligning with the XMAS structure for convenience in nuclear reactor practice. The calculated average flux for each of these energy ranges is detailed in Table 4.1.

**Table 4.1:** Neutron flux-energy dependence for Au in the energy regimes [130]

| Energy Regime | $\bar{\phi} (n.cm^{-2}.s^{-1}) \times 10^{11}$ |
|---------------|------------------------------------------------|
| Thermal       | 1.97                                           |
| Resonance     | 0.100                                          |
| Fast          | 0.191                                          |
| <b>Total</b>  | <b><math>2.263 \pm 0.048</math></b>            |

In Table 4.1, the effective neutron flux incident on Au was determined to be  $(2.263 \pm 0.048) \times 10^{11} \text{ n.cm}^{-2}.s^{-1}$ . Examining the flux-energy dependence in Figure 4.1, it is observed that higher peaks occur within the thermal energy region. This can be attributed to the absorption of thermal neutrons by the Au sample, contributing to the build-up of peaks in this region. Such behavior is possible due to the high thermal neutron capture reactions in the thermal neutron range and the relatively few inelastic scattering reactions in the fast neutron regime for gold [10]. Similarly, the average neutron flux impinging on Lu-Al was estimated and presented in Figure 4.2 and Table 4.2 below.

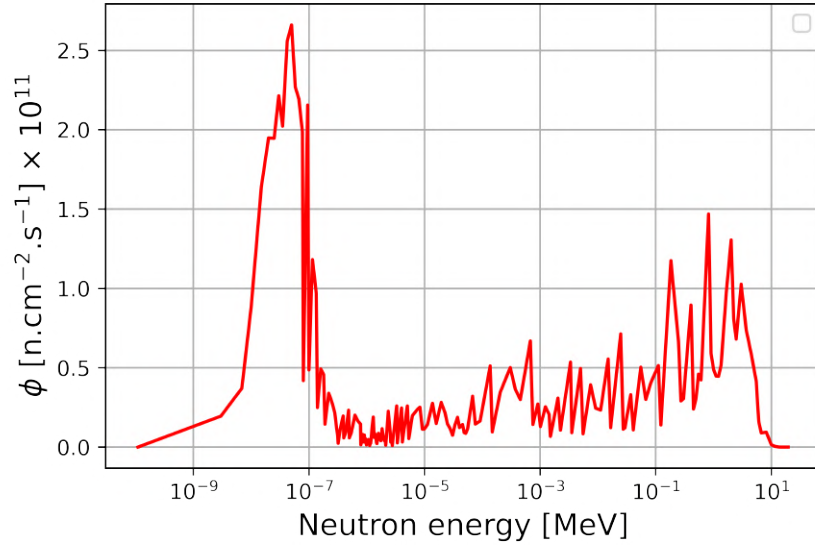


**Figure 4.2:** Neutron Flux-Energy profile for Lu-Al [130]

**Table 4.2:** Average neutron flux-energy dependence on Lu-Al in the energy regimes. In published work [130]

| Energy Regime | $\phi (n.cm^{-2}.s^{-1}) \times 10^{12}$ |
|---------------|------------------------------------------|
| Thermal       | 3.426                                    |
| Resonance     | 1.019                                    |
| Fast          | 2.048                                    |
| <b>Total</b>  | <b><math>6.493 \pm 0.007</math></b>      |

The total neutron flux incident on Lu-Al was determined to be  $(6.493 \pm 0.007) \times 10^{12} \text{ n.cm}^{-2}.\text{s}^{-1}$ . Analyzing the neutron flux distribution over energy ranges, as depicted in Figure 4.2, it is observed that the flux is dominant in the thermal and fast energy regimes, as detailed in Table 4.2. Comparing the average total flux for Lu-Al to that of Au, it is evident that the number of fission events increases with Lu-Al than Au hence flux is approximately one order of magnitude higher than that of Au. Also, Lu-Al exhibits a higher flux in the fast energy regime compared to Au, indicating the presence of more scattering reactions in Lu-Al than in Au.

### 4.1.2 Displacement Per Atom(dpa) rates in Au

The dpa is a fundamental quantitative measure of radiation damage in materials. Consequently, this sub-section presents the calculated dpa rates for Au based on the NRT-dpa model. The neutron energy profile plots for dpa rates in Au are illustrated in Figure 4.3, and the contribution of different energy regimes to the dpa rate is detailed in Table 4.3.

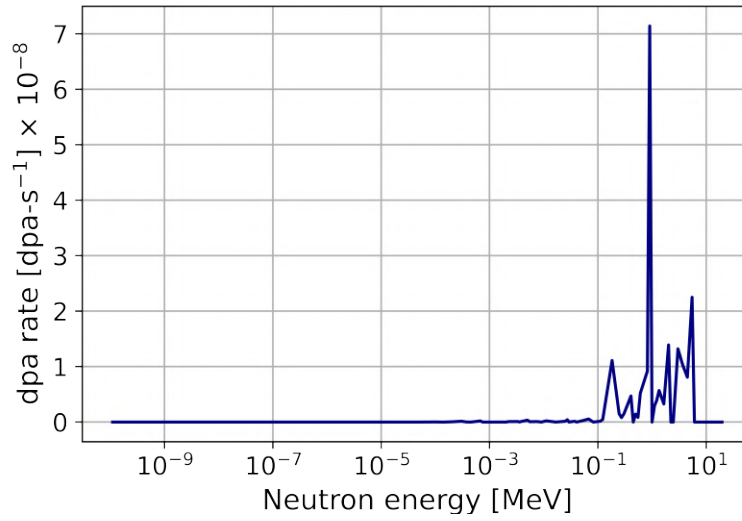


**Figure 4.3:** dpa rate-neutron energy profile. Spectra shows the displacement per atom rates across the neutron energy (thermal, resonance, and fast) regimes for Au. [130]

**Table 4.3:** Average dpa rate for Au in the energy regimes [130]

| Energy Regime | dpa rate ( $dpa.s^{-1}$ ) $\times 10^{-7}$ |
|---------------|--------------------------------------------|
| Thermal       | 0.000                                      |
| Resonance     | 0.020                                      |
| Fast          | 1.939                                      |
| <b>Total</b>  | <b>1.959 <math>\pm</math> 0.060</b>        |

Lower dpa rates occurred in the resonance region. The thermal neutron region, which encompasses neutron energies in the cold, thermal, and epithermal range, showed no contribution to the dpa of the sample since energies within this range

are below the threshold displacement energy ( $E_d$ ) values of the material. The fast energy region contributed over 99% of the neutron radiation damage for both materials. The total dpa rate for Au in this study was estimated to be  $(1.959 \pm 0.060) \times 10^{-7} \text{ dpa.s}^{-1}$ , as shown in Table 4.3.

### 4.1.3 Displacement Per Atom (dpa) rates for Lu-Al

Estimated dpa rates for Lu-Al were also done using the NRT model and presented in Figure 4.4 and Table 4.4

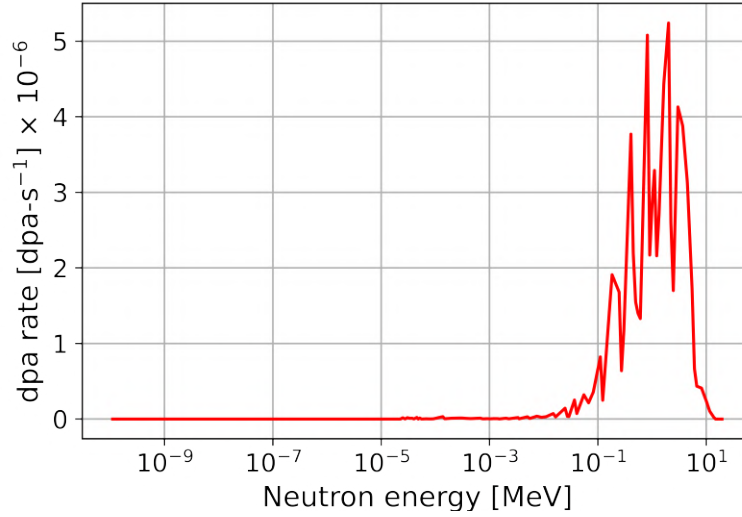


**Figure 4.4:** dpa rate-Energy profile. Spectra show the displacement per atom rates across the neutron energy (thermal, epithermal, and fast) regimes for Lu-Al [130]

**Table 4.4:** Average dpa rates for Lu-Al in the energy regimes [130]

| Energy Regime | dpa rate ( $\text{dpa.s}^{-1}$ ) $\times 10^{-7}$ |
|---------------|---------------------------------------------------|
| Thermal       | 0.000                                             |
| Resonance     | 3.550                                             |
| Fast          | 652.500                                           |
| <b>Total</b>  | <b><math>656.050 \pm 1.513</math></b>             |

From Figure 4.4, it can be observed that the fast energy region contributed over 99% of the neutron damage, dominating other energy ranges. The total dpa rate

in Lu-Al is  $(656.050 \pm 1.513) \times 10^{-7} \text{ dpa.s}^{-1}$ , as shown in Table 4.4. Examining the damage-energy profiles for both materials in Figure 4.5, it is evident that the damage in Lu-Al is largely attributed to fast neutrons.

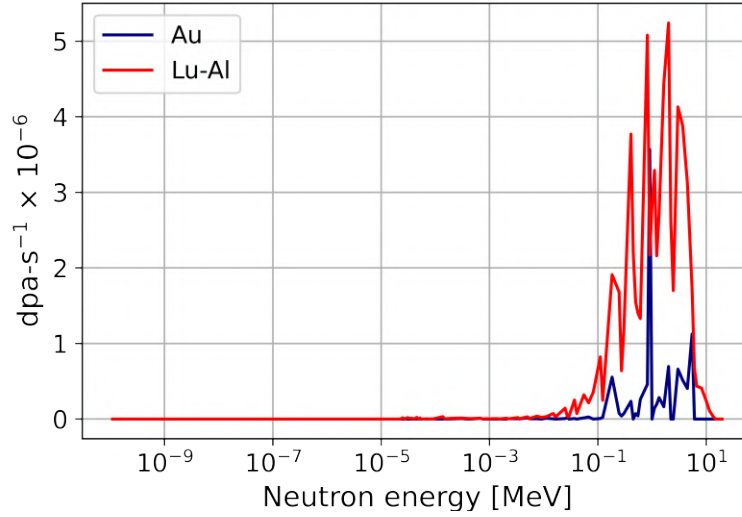


**Figure 4.5:** dpa rate vs neutron energy. The profile illustrates the dpa rate across neutron energy regions, including thermal, resonance, and fast ranges. Note that the dpa rate for Au has been scaled up to  $5 \times 10^1$  to facilitate comparison with the Lu-Al spectrum [130]

Comparing the damage for both materials, it can be observed that the dpa rate for Lu-Al is two orders of magnitude higher than that in the Au material.

#### 4.1.4 Heat deposition for Au and Lu-Al

The heat deposition from radiation types in crystalline materials is related to the thermal heating of the materials. Understanding the amount of heat energy deposited due to collisions of neutrons and photons with the nuclei of materials is essential for reactor studies. The calculated heat deposition for the materials is presented below.

#### 4.1.5 Average heat deposition in Au

Figure 4.6 and Table 4.5 present the normalized results of heat energy deposited by neutrons and photons in the Au material.

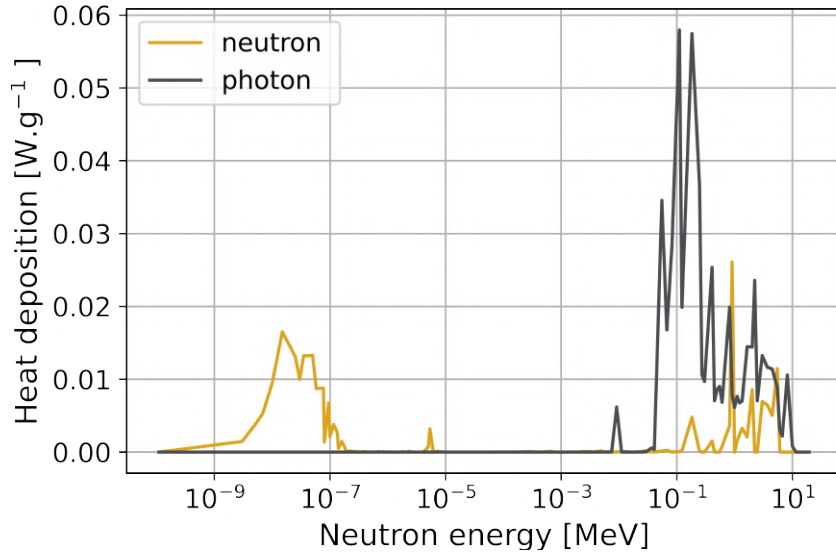


**Figure 4.6:** Neutron and photon heat deposition. The neutron heat deposition is scaled up by  $10^5$  to enable comparison with photon heat deposition which is 5 orders of magnitude higher. [130]

**Table 4.5:** Average heat deposition in Au the energy regimes [130]

| Energy Regime | Neutron-Heating<br>( $\text{W.g}^{-1}$ ) $\times 10^{-6}$ | Photon-Heating<br>( $\text{W.g}^{-1}$ ) |
|---------------|-----------------------------------------------------------|-----------------------------------------|
| Thermal       | 1.607                                                     | 0.000                                   |
| Resonance     | 0.047                                                     | 0.000                                   |
| Fast          | 0.858                                                     | 0.513                                   |
| <b>Total</b>  | $2.512 \pm 0.121$                                         | $0.513 \pm 0.011$                       |

In Table 4.5, the heat deposition by neutrons in the Au sample is negligibly small compared to the  $0.513 \text{ W.g}^{-1}$  deposited by photons in the material. The fast neutron energy region solely contributed to the heat deposited by photons in the Au sample. The average neutron heat deposition of  $2.512 \times 10^{-6} \text{ W.g}^{-1}$  is negligible compared to the photon heating which contributed over 99% of the total heat deposition in the material.

#### 4.1.6 Average heat deposition in Lu-Al

The average heat deposition by neutrons and photons was also calculated. The heating-neutron energy profile is presented in Figure 4.7.

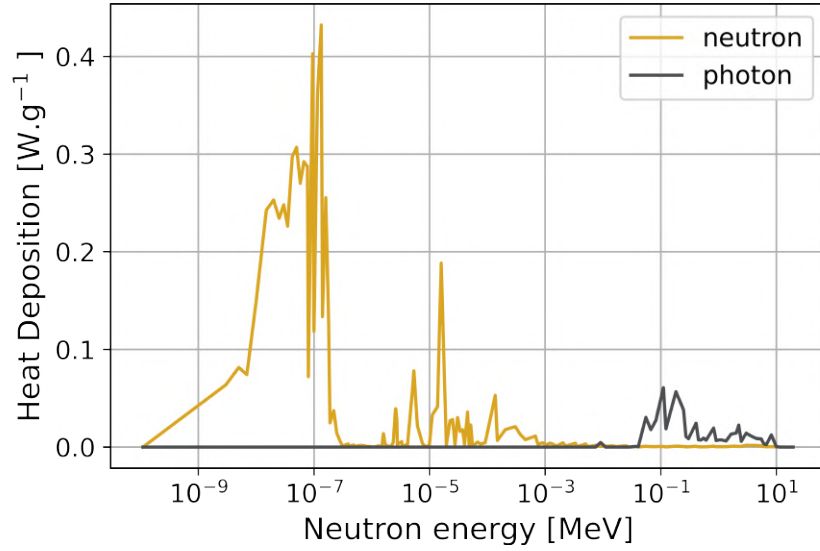


**Figure 4.7:** Neutron and photon heat deposition in Lu-Al. low deposition of heat energy by photons at fast-energy region. [130]

**Table 4.6:** Average dpa rates for Lu-Al in the energy regimes [130]

| Energy Regime | Neutron-Heating<br>( $W.g^{-1}$ ) | Photon-Heating<br>( $W.g^{-1}$ ) |
|---------------|-----------------------------------|----------------------------------|
| Thermal       | 5.1865                            | 0.0000                           |
| Resonance     | 0.7929                            | 0.0001                           |
| Fast          | 0.0279                            | 0.5026                           |
| <b>Total</b>  | $6.0073 \pm 0.0113$               | $0.5027 \pm 0.0105$              |

Results of neutron heating show that about 85% of the heating is from thermal neutrons, whereas photon-heating is purely due to fast neutron regime, as illustrated in Figure 4.7 and Table 4.6. The average neutron heat deposition of  $6.01 W.g^{-1}$  contributed over 90% of the total heat deposition in the material. It can also be observed that photon heating occurred at the fast energy range in Lu-Al.

Looking at the two materials, the Au material shows heating was mainly due to photons at the fast energy range, whereas the major contribution to heating in Lu-Al was from thermal neutrons. In both materials, however, the heating of photons at the fast energy regime is approximately the same.

#### 4.1.7 Evaluation of neutron fluence

Using the calculated neutron flux data, neutron fluence or dose was extrapolated for 1 year. The results for both materials are presented in Table 4.7.

**Table 4.7:** Total neutron fluence ( $\Phi$ ) for Au and Lu-Al [130]

| <b>Irradiation Time<br/>(Days)</b> | <b>Au<br/>(<math>n.cm^{-2}</math>) <math>\times 10^{18}</math></b> | <b>Lu-Al<br/>(<math>n.cm^{-2}</math>) <math>\times 10^{18}</math></b> |
|------------------------------------|--------------------------------------------------------------------|-----------------------------------------------------------------------|
| 5                                  | 0.098                                                              | 2.805                                                                 |
| 10                                 | 0.195                                                              | 5.610                                                                 |
| 15                                 | 0.293                                                              | 8.415                                                                 |
| 20                                 | 0.391                                                              | 11.213                                                                |

The neutron fluence (dose) for the two materials is of order  $10^{18}$   $n.cm^{-2}$  with values for Lu-Al being higher than those of the Au material in the same irradiation time. Neutron fluence is proportional to the dpa in a material. Irradiation of samples in the reactor is usually based on fluence values. Usually, higher values of fluence connote higher damage to the material.

## 4.2 Numerical results from SRIM calculations

This section presents the results of displacement profiles and the dpa for the ions calculated from SRIM-2013. The numerical results for both  $H^+$  and  $Au^+$  ion interactions with the Au and Lu-Al studied with SRIM-2013 are presented in this section.

### 4.2.1 Displacement damage profiles

The number of displacements ( $\nu_{NRT}$ ) resulting from the interaction of  $H^+$  and  $Au^+$  ions with Lutetium-Aluminium and Gold samples was calculated using Eq. 3.31. The displacement-depth profiles for the H-ion projectile are illustrated in Figure 4.8.

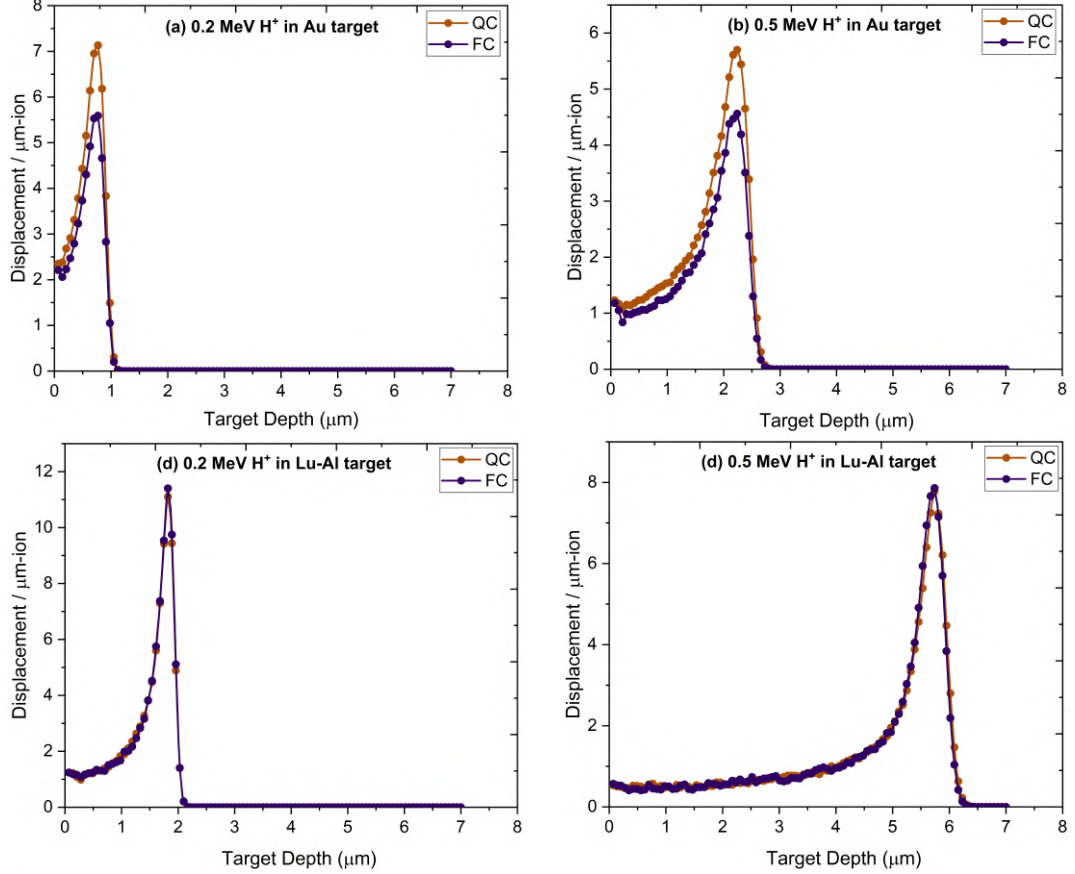


**Figure 4.8:** Displacement-target depth profile for  $H^+$  into Lu-Al and Au targets

Displacement-depth profiles in the materials indicate an overestimation in the Quick Calculation (QC) mode compared to the Full Cascade (FC) mode for H-ions into Au targets. However, the number of displacements observed in both calculation modes aligns well with the Lu-Al sample. Additionally, the number of displacements in the Lu-Al sample in both calculation modes is higher than that in the Au sample, with the maximum displacement occurring at a specific depth (Bragg peak).

Similarly, Figure 4.9 presents the displacement-depth profiles for  $Au^+$  in the targets under study. A contrasting trend emerged with  $Au^+$ . While  $H^+$  exhibited a concentration of energy deposition at a specific depth within the target, the displacement

caused by  $\text{Au}^+$  initiated from the beginning of the target and rapidly decreased. In the FC mode, the number of displaced atoms did not follow the conventional pattern of peaking before diminishing to zero.

The displacement of atoms induced by incident Au-ions in Au demonstrated better agreement in both QC and FC calculations. However, in the FC mode, higher atomic displacements were observed at shallower depths in the Lu-Al target.

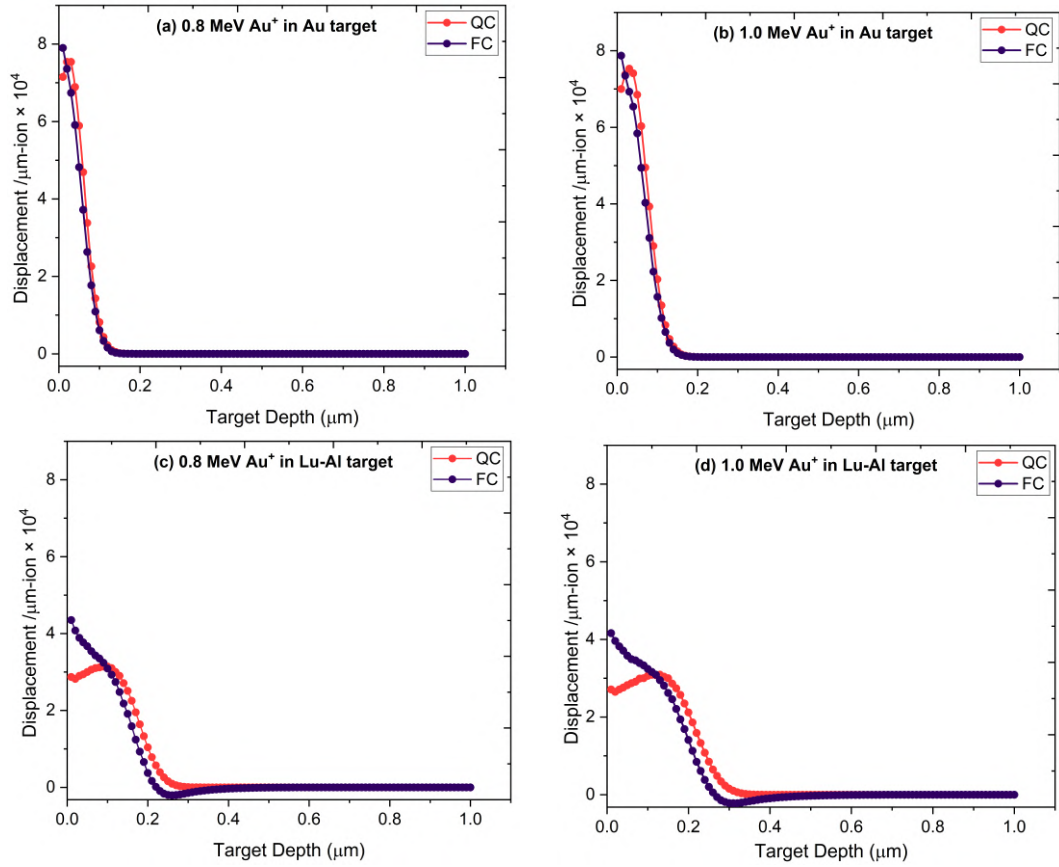


**Figure 4.9:** Displacement-target depth profile for  $\text{Au}^+$  into Lu-Al and Au targets

Overall, more atoms experienced displacement in the Au target compared to the Lu-Al target, albeit at slightly shallower depths in the Au sample, potentially due to self-ion irradiation.

### 4.2.2 Displacement per atom (dpa) rates for Au and Lu-Al samples

Displacement per atom rates resulting from  $H^+$  and  $Au^+$  irradiation with a flux of  $1.55 \times 10^{14}$  ions.cm<sup>-2</sup>.s<sup>-1</sup> on Lu-Al and Au were determined using SRIM-2013 outputs and Eq. 3.36. The obtained results are shown in Table 4.8 this section. Also, the ratios of dpa rate calculated for the QC and FC modes are shown in Table 4.9.

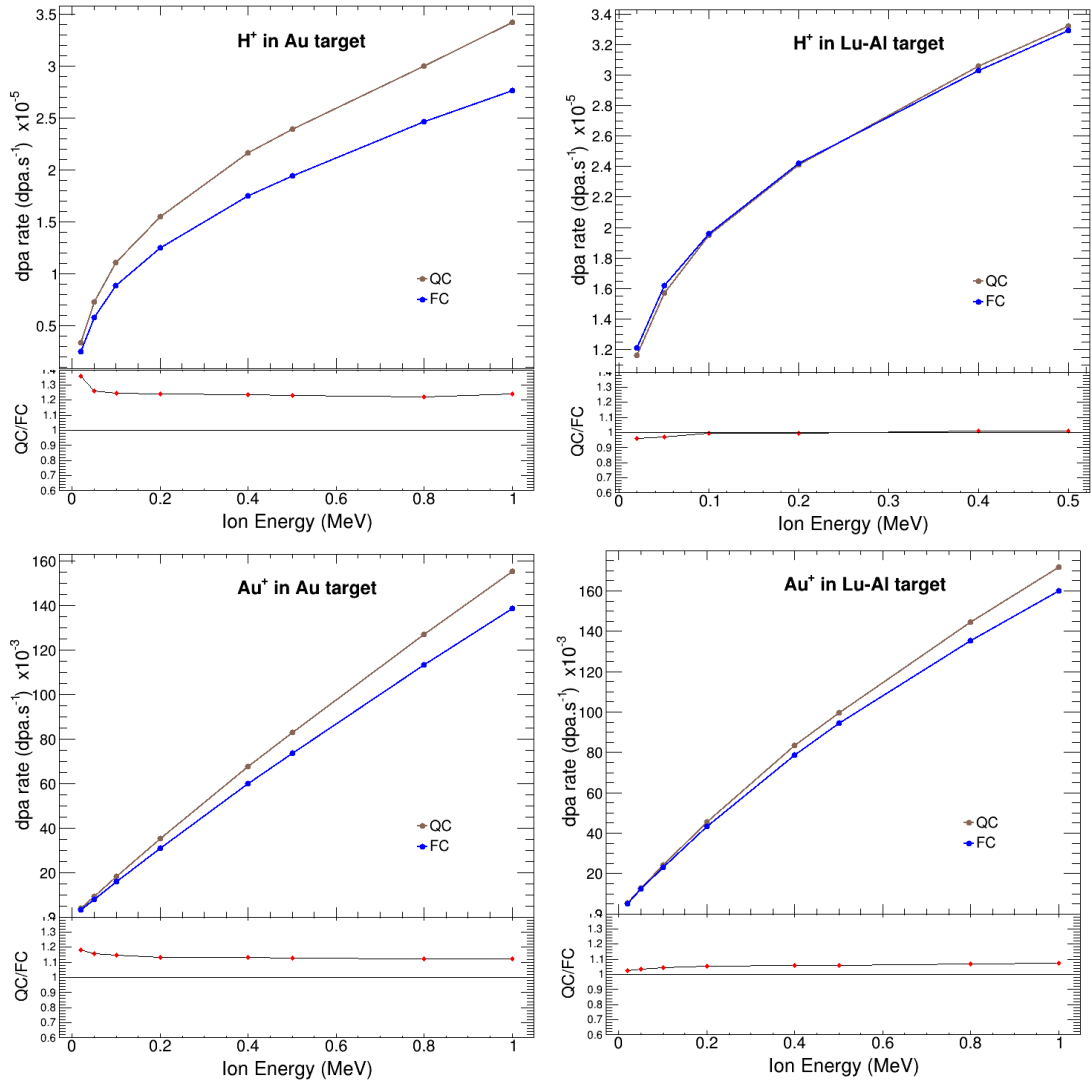
**Table 4.8:** Displacement-Per-Atom rates in QC and FC modes for Au and Lu-Al

| Ion Energy<br>(MeV) | Displacement-Per-Atom rate (dpa.s <sup>-1</sup> ) $\times 10^{-5}$ |      |                |      |              |          |                 |          |
|---------------------|--------------------------------------------------------------------|------|----------------|------|--------------|----------|-----------------|----------|
|                     | $H^+$ in Au                                                        |      | $H^+$ in Lu-Al |      | $Au^+$ in Au |          | $Au^+$ in Lu-Al |          |
|                     | QC                                                                 | FC   | QC             | FC   | QC           | FC       | QC              | FC       |
| 0.02                | 0.34                                                               | 0.25 | 1.16           | 1.21 | 380.59       | 322.04   | 523.72          | 512.25   |
| 0.05                | 0.73                                                               | 0.58 | 1.57           | 1.62 | 927.09       | 800.22   | 1265.30         | 1223.28  |
| 0.10                | 1.11                                                               | 0.89 | 1.95           | 1.96 | 1810.25      | 1580.90  | 2419.80         | 2320.4   |
| 0.20                | 1.55                                                               | 1.25 | 2.41           | 2.42 | 3513.16      | 3098.40  | 4549.10         | 4319.71  |
| 0.40                | 2.16                                                               | 1.75 | 3.06           | 3.03 | 6757.96      | 5977.30  | 8333.60         | 7874.87  |
| 0.50                | 2.39                                                               | 1.94 | 3.32           | 3.29 | 8294.97      | 7367.90  | 9977.40         | 9442.20  |
| 0.80                | 3.00                                                               | 2.46 |                |      | 12686.40     | 11320.00 | 14450.00        | 13532.50 |
| 1.00                | 3.42                                                               | 2.76 |                |      | 15540.9      | 13857.00 | 17164.00        | 16017.30 |

The calculated displacement per atom (dpa) rates for the projection of  $H^+$  into the target ranged from  $(0.25 - 3.42) \times 10^{-5}$  dpa.s<sup>-1</sup> in the gold target and  $(1.21 - 3.32) \times 10^{-5}$  dpa.s<sup>-1</sup> in Lu-Al. The self-ion irradiation of the Au target yielded dpa rates ranging from  $(3.22 - 155.4) \times 10^{-3}$  dpa.s<sup>-1</sup>. Additionally, the  $Au^+$  irradiation into the Lu-Al target resulted in dpa rates of  $(5.12 - 171.64) \times 10^{-3}$  dpa.s<sup>-1</sup> within the same energy ranges. Notably, there are no results for  $H^+$  irradiation in the Lu-Al target due to constraints on target depths to ensure proximity to the real thickness of the targets. Proton energies beyond 0.5 MeV would lead to a significant number of transmitted ions through the target, potentially resulting in an underestimation of the damage.

**Table 4.9:** dpa rate ratios of Quick calculation to Full cascade modes

| Ion Energy<br>(MeV) | $H^+$ in Au<br>QC/FC | $H^+$ in Lu-Al<br>QC/FC | $Au^+$ in Au<br>QC/FC | $Au^+$ in Lu-Al<br>QC/FC |
|---------------------|----------------------|-------------------------|-----------------------|--------------------------|
| 0.02                | 1.36                 | 0.96                    | 1.18                  | 1.02                     |
| 0.05                | 1.26                 | 0.97                    | 1.16                  | 1.03                     |
| 0.10                | 1.25                 | 0.99                    | 1.14                  | 1.04                     |
| 0.20                | 1.24                 | 1.00                    | 1.15                  | 1.05                     |
| 0.40                | 1.23                 | 1.01                    | 1.13                  | 1.06                     |
| 0.50                | 1.23                 | 1.01                    | 1.13                  | 1.06                     |
| 0.80                | 1.22                 |                         | 1.12                  | 1.07                     |
| 1.00                | 1.24                 |                         | 1.12                  | 1.07                     |

**Figure 4.10:** dpa rate vs Ion energy for  $H^+$  and  $Au^+$  into Lu-Al and Au targets for QC and FC modes. The ratio QC:FC is shown at the base of the graph.

The ion displacement-depth profiles and displacement-per-atom (dpa) rates for Lu-Al and Au were computed based on SRIM-2013 outputs at various ion energies.

In the case of  $H^+$  in the Au sample, the Full Cascade (FC) calculations yielded a lower number of displacements compared to the Quick Calculation (QC). Estimates indicated higher dpa rates in the Quick Calculation (QC) mode than in the Full Cascade (FC) mode, as shown in Table 4.9.

Conversely, a nearly equivalent number of displacements per ion was observed in the Lu-Al sample at different depths. In contrast, for  $Au^+$  in both targets, the FC calculations resulted in a higher displacement per ion count in each instance. This disparity may be attributed to both the nature of the ion beam projectile and the characteristics of the target material as earlier observed by Agarwal et al. [47]. Calculated dpa rates had a linear response with the ion energies in both  $H^+$  and  $Au^+$  ion beam irradiation as shown in Figure 4.10.

The  $Au^+$  at similar energies yielded dpa rates 2-3 orders of magnitude higher than the  $H^+$  which correlates to more damage on the microstructure of the materials when irradiated with gold-ions. However,  $Au^+$  is heavier than  $H^+$  ion hence more damage is expected but a lower penetration depth, hence damage caused in the materials for both ions at the same energy will be localized at shorter depths when  $Au^+$  ion is used.

The total displacement per atom (dpa) rates in Lu-Al were higher than those predicted for Au in both ion species used. Dpa rates are utilized to estimate how radiation-induced damage influences the physical and mechanical properties of materials. Higher dpa rates generally correlate with increased damage [9, 10, 55, 190]. The relatively higher values predicted in the Lu-Al sample indicate an elevated level of damage to the material's microstructure. In general, the dpa rates were on the order of  $10^{-5}$  dpa.s<sup>-1</sup> in both materials. These dpa rates are significant enough to cause microstructure defects in the materials after irradiation.

Most studies estimated the ratios of vacancies or displacements produced in the FC and QC mode SRIM calculations. Whereas, in this study, the ratios for the total dpa rates were reported. While some studies have reported higher estimates of

total displacement ( $\nu(T)$ ) for the Full Cascade (FC) mode [47, 101, 191], this study observed higher or nearly equal estimates of dpa rates in the Quick Calculation (QC) mode compared to the FC mode in SRIM calculations. Agarwal et al. noted that displacement damage could vary depending on the target involved, resulting in either higher or lower displacement damage in the materials [47].

The graphs in Figure 4.10 demonstrate good agreement between QC and FC modes in dpa rate estimates for the Lu-Al binary alloy. Also, a better linear dependence is observed in the  $Au^+$  irradiation with the targets. This behavior could be attributed to the ability of  $Au^+$  to cause damage without much charge interaction with the target due to its metallic inertness.

### 4.2.3 Comparison of damage in ion-beams and neutrons

Studies on the effects of radiation on microstructure have revealed that damage caused by ion beam irradiation can be likened to that induced by neutrons [45, 48, 192]. While neutrons and ions have different damage mechanisms due to the absence of charge in neutrons, ion beam irradiation has demonstrated similar radiation damage effects as those produced by neutrons on materials' microstructure [41, 42, 46, 48]. In this study, proton and gold ion beam irradiation were examined to establish a quantitative link to neutron displacement per atom (dpa) rates, as presented in Table 4.8 and 4.9.

Table 4.8 indicates that dpa rates for  $H^+$  with energies in the range of 0.02 - 1.0 MeV in the gold sample are approximately one or two orders of magnitude higher than those obtained for neutron irradiation, as shown in Table 4.10. Similarly, the dpa rates for Lu-Al fall within the same order of magnitude. However, it's essential to note that a direct comparison of dpa rates between the two radiation species is not feasible due to their distinct energy and flux spectra. An increase and or reduction in the ion flux or proton energy could potentially yield dpa rates comparable to those from neutron irradiation.

**Table 4.10:** Comparison of  $H^+$  beam with neutron dpa rates in Au and Lu-Al

| <b>Ion Energy</b> | <b>dpa rate (dpa.s<sup>-1</sup>) <math>\times 10^{-7}</math></b> |              | <b>Neutron Energy</b>        | <b>dpa rate (dpa.s<sup>-1</sup>) <math>\times 10^{-7}</math></b> |               |
|-------------------|------------------------------------------------------------------|--------------|------------------------------|------------------------------------------------------------------|---------------|
| <b>(MeV)</b>      | <b>Au</b>                                                        | <b>Lu-Al</b> | <b>(MeV)</b>                 | <b>Au</b>                                                        | <b>Lu-Al</b>  |
| 0.02              | 33.91                                                            | 115.83       |                              |                                                                  |               |
| 0.20              | 154.92                                                           | 240.83       | $1.1 \times 10^{-10} - 19.6$ | 1.959                                                            | 656.050 [130] |
| 0.40              | 215.91                                                           | 306.20       |                              |                                                                  |               |
| 0.50              | 239.10                                                           | 332.20       |                              |                                                                  |               |

From Table 4.10, at an energy of 0.5 MeV, achieving 0.26 dpa would take approximately 3 hours, while achieving the same dpa with neutron irradiation would require around 37 hours. Furthermore, manipulating beam parameters offers flexibility in reducing the time even further, a scenario challenging to achieve with neutron irradiation.

Existing literature has established a nexus between neutron and ion beam irradiation, particularly in terms of the observed post-irradiation effects using  $Au^+$  and  $H^+$  [16, 41, 46, 130]. Neutron irradiation, while effective in studying radiation damage effects in materials, presents challenges such as high costs, handling issues arising from sample activation, and extended exposure times this is discussed in section 2.6.3. From Table 4.10, it is evident that ion-beam radiation, particularly when utilizing  $H^+$  ions with lower ion energies, yields displacement per atom (dpa) rates comparable to neutrons in the Au samples. At higher  $H^+$  ion energies, dpa rates in Lu-Al targets become comparable to those obtained for neutrons. Therefore, ion-beam irradiation, especially with  $H^+$  ions, can serve as a viable surrogate for neutron irradiation in materials tested for radiation damage resilience considering the challenges of neutron irradiation outlined above.

Quantifying damage levels in crystalline materials requires a quantitative measure that analytical calculation methods struggle to achieve due to the intricate nature of the radiation damage evolution process. Utilizing Monte Carlo methods, radiation damage by neutrons to Au and Lu-Al was quantified in the Monte Carlo N-Particle code (MCNP6.2) in our previous studies [54, 130]. Similarly, damage caused by

ion beams on the studied materials was calculated using the SRIM-2013 code. The objective was to compare ion beam irradiation with neutrons as an alternative irradiation field, given the challenges associated with obtaining neutron irradiation. In addition, the predictions aimed to guide us towards a specific ion energy that would produce displacement per atom rates comparable to neutrons. The dpa (displacements per atom) serves as a key parameter for comparing radiation doses or fluences between different irradiation species, quantifying the damage incurred by crystalline or macro-molecular materials [8, 25, 140].

In ion beam irradiation, heavier ions cause more atomic displacement with shorter ranges into the materials compared to lighter ions [193], leading to higher damage rates. This, of course, depends on the energy of the ion beam. Therefore,  $\text{Au}^+$  ions resulted in higher damage rates than  $\text{H}^+$  ions. The simulation using  $\text{H}^+$  beams at lower proton energies yielded approximate dpa rates and produced similar irradiation profiles as neutrons, prompting the use of protons as a surrogate for neutron irradiation in the experiments.

### **4.3 EDS results of Lu-Al and Au samples using SEM**

To determine the chemical composition of the foils, Energy Dispersive Spectroscopy (EDS) was conducted on the samples before irradiation. The results of the EDS detector, indicating the compositions, are presented below.

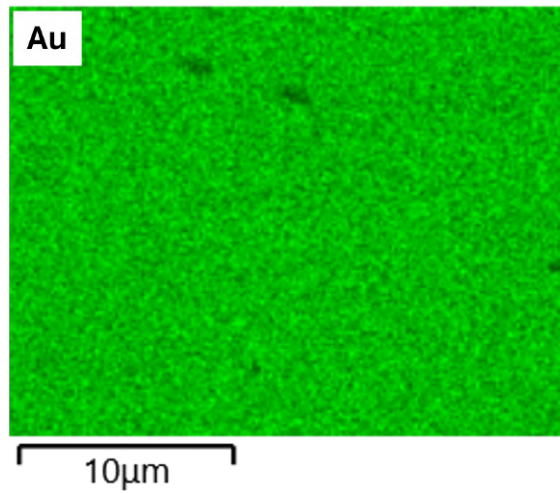


**Figure 4.11:** EDS for Au sample

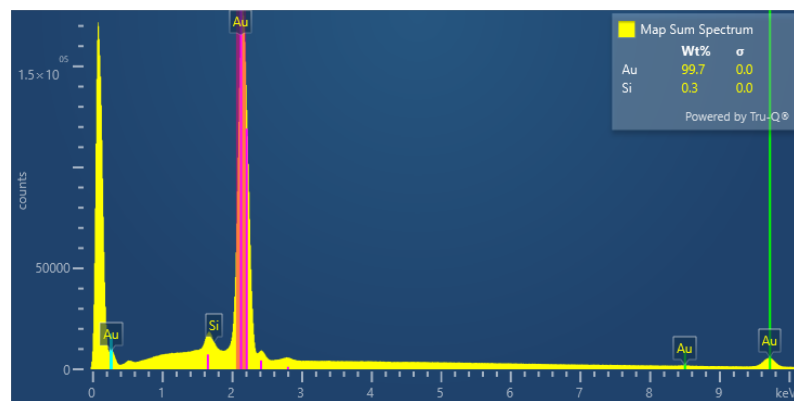


**Figure 4.12:** EDS mapping spectra for Au

From the EDS, the chemical composition of the foil is shown to be 99.7 % Au foil with a fractional impurity of silicon (Si). The chemical composition of Lu-Al alloys was also determined with the same instrument and the results are presented below.

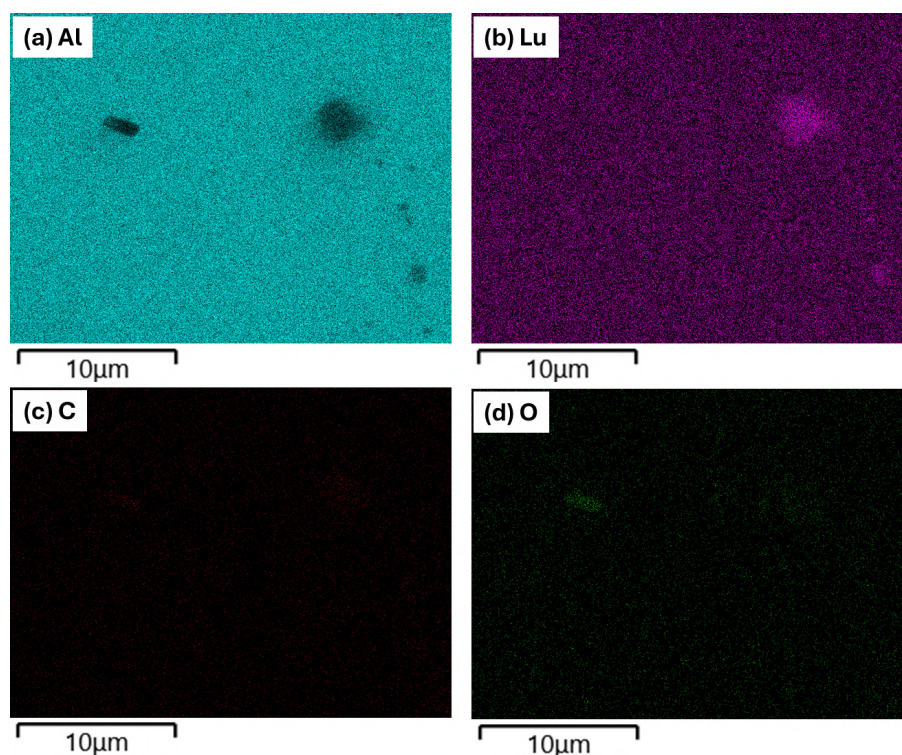


**Figure 4.13:** (a) mapping for Al (b) mapping for Lu (c) mapping for C (d) mapping for O

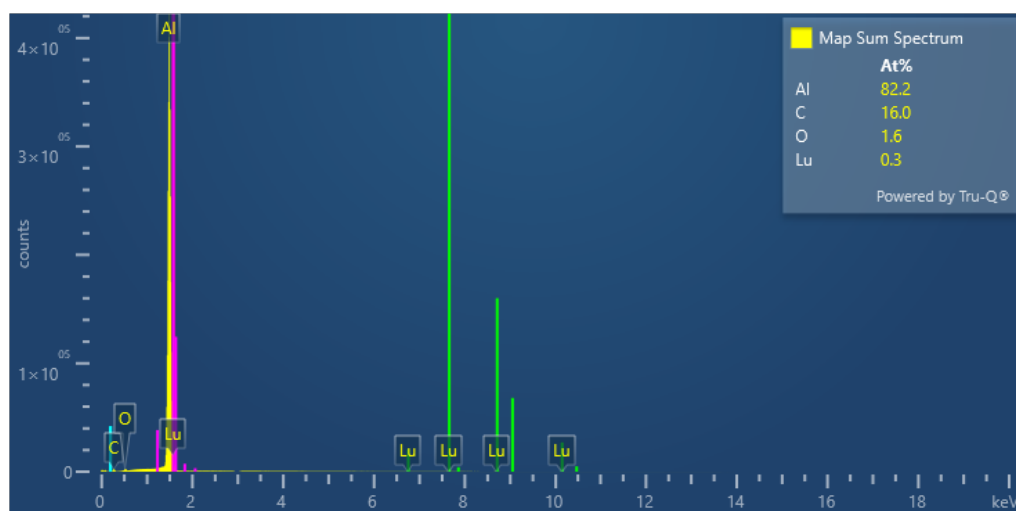


**Figure 4.14:** EDS mapping spectra for Lu-Al

The results show the chemical composition of Lu-Al by atomic percentage. The chemical composition of the foils was also complemented with HRTEM-EDS shown in section 4.4.3. Post-irradiation studies were performed on the samples however, there was no defect detected using the SE and BSE scanning modes.

## 4.4 Results of damage characterization using XRD

In this section, the response of Lu-Al and Au to local stress induced by radiation doses from proton irradiation is reported. The average microstrain, crystallite size, and dislocation density results have been calculated for Lu-Al and Au using the Williamson-Hall method described in section 3.9.3 (b).

### 4.4.1 X-Ray Diffraction patterns for Au and Lu-Al

The XRD patterns of pre and post-irradiation characterization for Au and Lu-Al using the Lorentz fit function are illustrated in Figure 4.15.

#### (a) Diffraction pattern for Au

Figure 4.15 displays the  $2\theta$  diffraction pattern spanning  $20-90^\circ$  before and after the irradiation of Au samples with 0.5 MeV protons. The pattern exhibits four diffraction peaks in Table 4.11, corresponding to the (111), (002), (022), (113), and (222) crystal planes, indicating the face-centered cubic structure of Au.

After irradiation, an increase in the intensity of peaks in the planes (111), (002), and (220) was observed. Notably, the peak in the plane (113) decreased significantly. Additionally, the (222) peak emerged after irradiation to a proton fluence of  $7.70 \times 10^{16} \text{ p.cm}^{-2}$  (b).

Further increasing the fluence to  $2.32 \times 10^{17} \text{ p.cm}^{-2}$  resulted in a significant growth in the intensity of the (002) crystal plane, accompanied by a decrease in the (111), (022), and (113) peaks. It was also observed that all the peak positions were slightly shifted to the left in spectra of samples (b) and (c) after irradiation, as shown in Table 4.11.

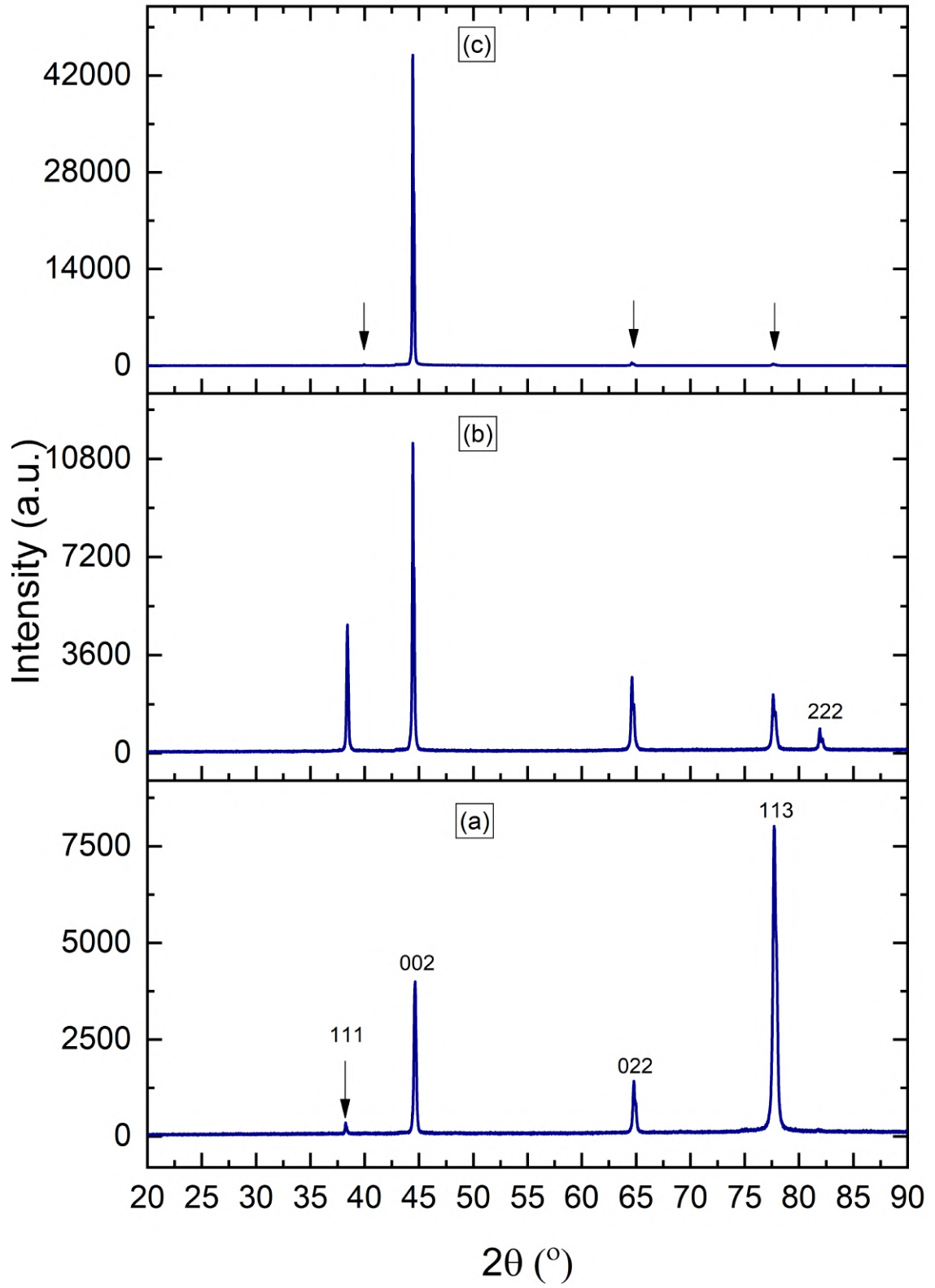


**Figure 4.15:** (a) XRD pattern unirradiated Au sample (b) XRD pattern for Au sample irradiated with protons to a dose of  $7.70 \times 10^{16} \text{ p.cm}^{-2}$ . (c) XRD pattern for Au sample irradiated with protons to a dose of  $2.32 \times 10^{17} \text{ p.cm}^{-2}$

**Table 4.11:** Peak positions, intensities, and lattice spacing for Au sample

| Sample | Fluence<br>(p.cm <sup>-2</sup> ) | Peak Position (2 $\theta$ )<br>( $^{\circ}$ ) | hkl   | Intensity<br>(a.u) | d<br>( $\text{\AA}$ ) | $\Delta d$<br>( $\text{\AA}$ ) |
|--------|----------------------------------|-----------------------------------------------|-------|--------------------|-----------------------|--------------------------------|
| (a)    | 0.00                             | 38.2445                                       | (111) | 351                | 2.3515                | -                              |
|        |                                  | 44.6366                                       | (002) | 3994               | 2.0284                | -                              |
|        |                                  | 64.7987                                       | (022) | 1426               | 1.4376                | -                              |
|        |                                  | 77.7166                                       | (113) | 8016               | 1.3150                | -                              |
| (b)    | $7.70 \times 10^{16}$            | 38.9618                                       | (111) | 4715               | 2.3098                | 0.0417                         |
|        |                                  | 44.4320                                       | (002) | 11372              | 2.0373                | 0.0089                         |
|        |                                  | 64.6192                                       | (022) | 2794               | 1.4412                | 0.0036                         |
|        |                                  | 77.6123                                       | (113) | 2159               | 1.2292                | 0.0858                         |
|        |                                  | 81.9154                                       | (222) | 905                | 1.1751                | -                              |
| (c)    | $2.32 \times 10^{17}$            | 39.9618                                       | (111) | 158                | 2.2543                | 0.0972                         |
|        |                                  | 44.4237                                       | (002) | 4497               | 2.0377                | 0.0093                         |
|        |                                  | 64.6025                                       | (022) | 439                | 1.4415                | 0.0039                         |
|        |                                  | 77.5955                                       | (113) | 275                | 1.2294                | 0.0856                         |

However, the (111) reflection peak shifted to the right after proton irradiation in both cases. Considering the (111) plane, there is a gradual contraction in the lattice spacing  $d_{hkl}$  from 2.3515 to 2.3098 and 2.2543  $\text{\AA}$ . The increments in intensity result from shorter interatomic distances within the Au crystal planes post-irradiation, as shown in Table 4.11 in the  $d$  column. This phenomenon may stem from the formation of point defects or the creation and mobility of dislocations within the crystal lattice due to the introduction of local strain introduced by the radiation stress field. Similar effects have been reported in other studies [115, 194]. There is a notable preferential growth in the intensity of the (002) crystal planes upon irradiation with high doses of proton irradiation compared to the other crystal planes. In this plane, there is a gradual increase in the  $d_{hkl}$  values, as shown in Table 4.11.

Usually, peak shifts in the XRD pattern may result from changes in the stoichiometry of dopants, Doppler broadening of the instrument, and changes in the intrinsic properties of the sample material. When there is a strong peak displacement, the calibration of the instrument or instrumental broadening is responsible. However,

slight displacements in the peak positions, as observed in the results tabulated, are indicative of changes in the intrinsic properties of the sample's microstructure.

This may include changes in lattice spacing, average crystallite size, microstrain, and binding energy. These changes could be due to the formation of point defects, dislocation, and other lattice defects related to constraints of strain fields on the crystalline structure of the Au sample post-irradiation. Analysis of the microstrain and dislocation density in the samples is performed to further explain the implications of the deviations in the diffraction pattern on the microstructure of the Au sample post-irradiation.

### **(b) Diffraction pattern for Lu-Al**

Figure 4.16 presents the X-ray diffraction pattern for the Lu-Al binary alloy system from 20-90° pre and post-proton irradiation. From the diffraction patterns, slight peak broadening occurs post-irradiation after the first dose of irradiation, leading to a decrease in all peak intensities corresponding to the crystal planes along (111), (022), (113), (004), (133), (224), (333), (044), (135), (062), (335), (444), and (117) reflections. The respective peak intensity values are shown in Table 4.12. Further peak broadening was observed for all the peaks in the second proton irradiation dose, except for the crystal planes along (224) and (335), which experienced a slight growth after irradiation with a proton fluence of  $2.32 \times 10^{17}$  p.cm<sup>2</sup> in (c).

In (b), a slight shift in the peak position was observed to the right of (a) the unirradiated sample, implying contraction due to a strain field, except for the crystal plane along (335). In (c), the peak again shifted slightly to the left of the peak positions of the unirradiated sample XRD pattern. A similar vacillating trend was also observed in the lattice spacing values presented in Table 4.12.

Slight displacements in the XRD peak positions towards the left are indicative of expansion or lattice relaxation. Hence, the Lu-Al lattice was significantly strained in the first irradiation but seemed to relax a little after the second irradiation dose. The average microstrain in the lattice is presented in section 4.4.3.

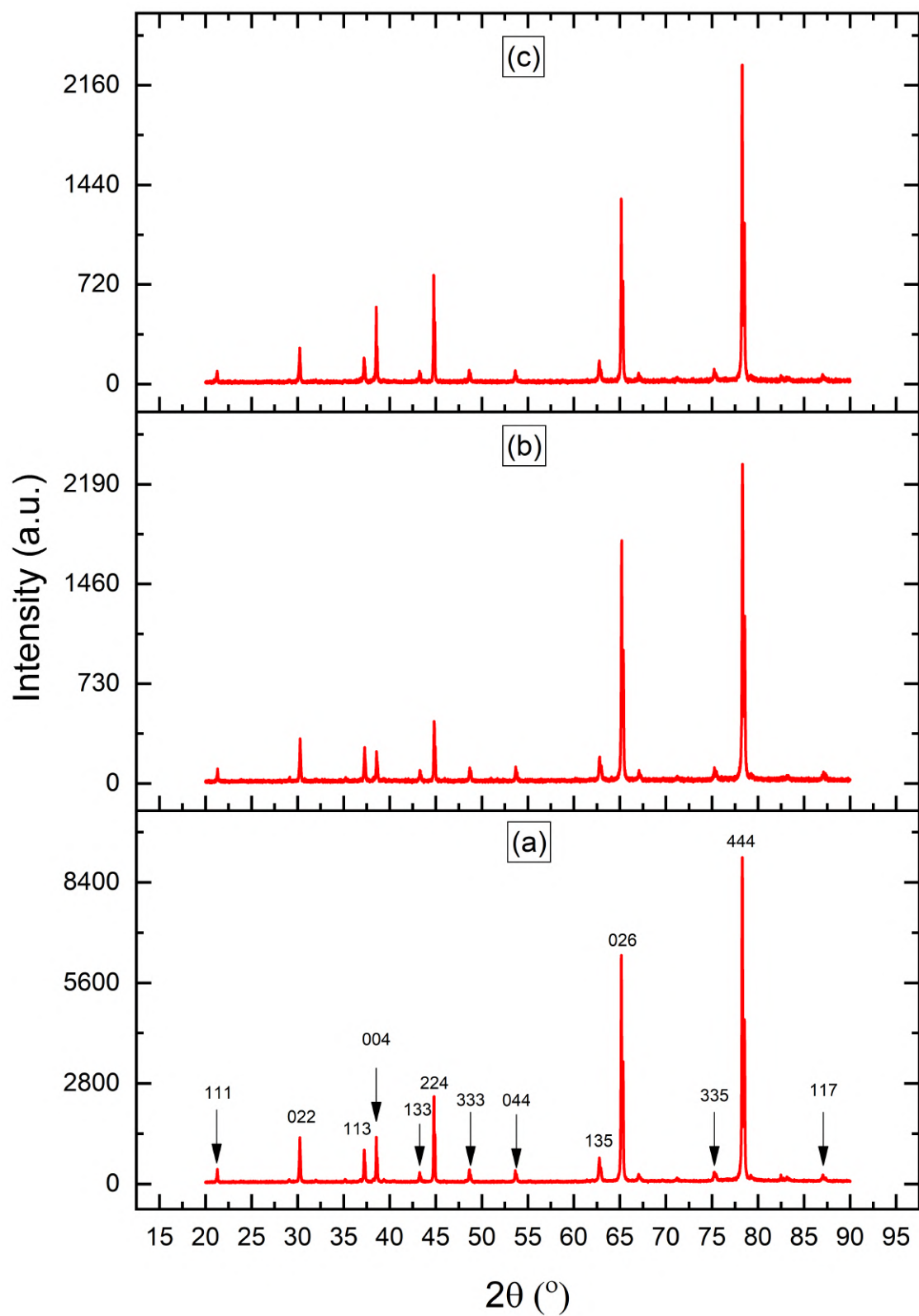


**Figure 4.16:** (a) XRD pattern for unirradiated Lu-Al sample (b) XRD pattern for Lu-Al sample irradiated with protons to a dose of  $7.70 \times 10^{16} \text{ p.cm}^{-2}$ . (c) XRD pattern for Lu-Al sample irradiated with protons to a dose of  $2.32 \times 10^{17} \text{ p.cm}^{-2}$

**Table 4.12:** Peak positions, intensities, and lattice spacing for Lu-Al sample

| Sample | Fluence<br>(p.cm <sup>-2</sup> ) | Peak Position (2 $\theta$ )<br>( $^{\circ}$ ) | hkl   | Intensity<br>(a.u) | d<br>( $\text{\AA}$ ) | $\Delta d$<br>( $\text{\AA}$ ) |
|--------|----------------------------------|-----------------------------------------------|-------|--------------------|-----------------------|--------------------------------|
| (a)    | 0.00                             | 21.2826                                       | (111) | 412                | 4.1715                | -                              |
|        |                                  | 30.2398                                       | (022) | 1291               | 2.9532                | -                              |
|        |                                  | 37.2335                                       | (113) | 945                | 2.4129                | -                              |
|        |                                  | 38.5453                                       | (004) | 1306               | 2.3338                | -                              |
|        |                                  | 43.2495                                       | (133) | 320                | 2.0902                | -                              |
|        |                                  | 44.8037                                       | (224) | 2438               | 2.0213                | -                              |
|        |                                  | 48.6473                                       | (333) | 4006               | 1.8702                | -                              |
|        |                                  | 53.6356                                       | (044) | 376                | 1.7074                | -                              |
|        |                                  | 62.7767                                       | (135) | 730                | 1.4790                | -                              |
|        |                                  | 65.1580                                       | (026) | 6372               | 1.4306                | -                              |
|        |                                  | 75.2683                                       | (335) | 326                | 1.2615                | -                              |
|        |                                  | 78.2847                                       | (444) | 9095               | 1.2203                | -                              |
|        |                                  | 87.0581                                       | (117) | 252                | 1.1184                | -                              |
| (b)    | $7.70 \times 10^{16}$            | 21.3120                                       | (111) | 107                | 4.1658                | 0.0057                         |
|        |                                  | 30.2776                                       | (022) | 328                | 2.9496                | 0.0036                         |
|        |                                  | 37.2796                                       | (113) | 263                | 2.4101                | 0.0029                         |
|        |                                  | 38.5664                                       | (004) | 233                | 2.3326                | 0.0012                         |
|        |                                  | 43.2623                                       | (133) | 95                 | 2.0896                | 0.0006                         |
|        |                                  | 44.8248                                       | (224) | 453                | 2.0204                | 0.0009                         |
|        |                                  | 48.6851                                       | (333) | 115                | 1.8688                | 0.0014                         |
|        |                                  | 53.6817                                       | (044) | 122                | 1.7060                | 0.0014                         |
|        |                                  | 62.8144                                       | (135) | 194                | 1.4782                | 0.0008                         |
|        |                                  | 65.2041                                       | (026) | 1777               | 1.4297                | 0.0009                         |
|        |                                  | 75.2643                                       | (335) | 117                | 1.2616                | 0.0001                         |
|        |                                  | 78.3225                                       | (444) | 2262               | 1.2198                | 0.0005                         |
|        |                                  | 87.0792                                       | (117) | 75                 | 1.1182                | 0.0002                         |
| (c)    | $2.32 \times 10^{17}$            | 21.2535                                       | (111) | 94                 | 4.1771                | 0.0056                         |
|        |                                  | 30.2191                                       | (022) | 262                | 2.9551                | 0.0020                         |
|        |                                  | 37.2044                                       | (113) | 189                | 2.4148                | 0.0018                         |
|        |                                  | 38.5413                                       | (004) | 556                | 2.3340                | 0.0002                         |
|        |                                  | 43.2288                                       | (133) | 94                 | 2.0912                | 0.0010                         |
|        |                                  | 44.7746                                       | (224) | 787                | 2.0225                | 0.0012                         |
|        |                                  | 48.6349                                       | (333) | 101                | 1.8706                | 0.0004                         |
|        |                                  | 53.6232                                       | (044) | 98                 | 1.7078                | 0.0004                         |
|        |                                  | 62.7643                                       | (135) | 169                | 1.4792                | 0.0003                         |
|        |                                  | 65.1457                                       | (026) | 1338               | 1.4308                | 0.0002                         |
|        |                                  | 75.2393                                       | (335) | 109                | 1.2619                | 0.0004                         |
|        |                                  | 78.2807                                       | (444) | 2307               | 1.2203                | 0.0001                         |
|        |                                  | 87.0792                                       | (117) | 64                 | 1.1182                | 0.0002                         |

#### 4.4.2 Crystallite size ( $D$ ), microstrain ( $\varepsilon$ ) and dislocation density ( $\delta$ ) for Au sample

The approach developed by Williamson and Hall [150] was used to estimate microstructural parameters relating to damage in the crystal pre and post-irradiation. Figures 4.17, 4.18, and 4.19 present the Williamson-Hall plots for the Au samples pre and post-irradiation. The microstrain, and crystallite size were extracted from the slopes and intercepts of the graph, and the dislocation density was calculated and presented in Table 4.13.

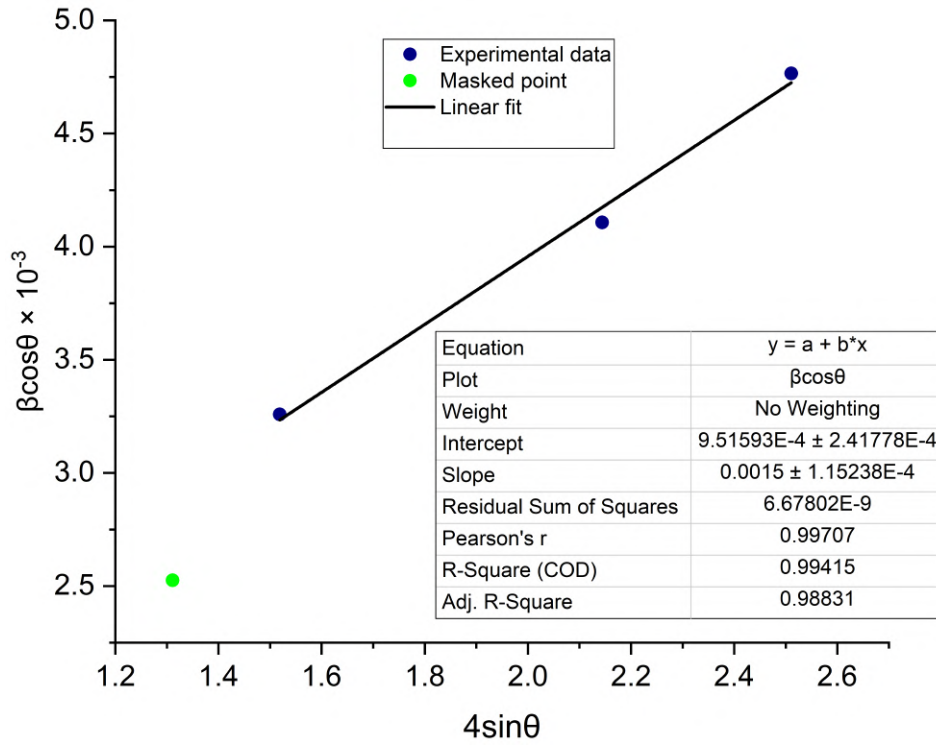


**Figure 4.17:** Williamson-Hall plot for the unirradiated Au sample

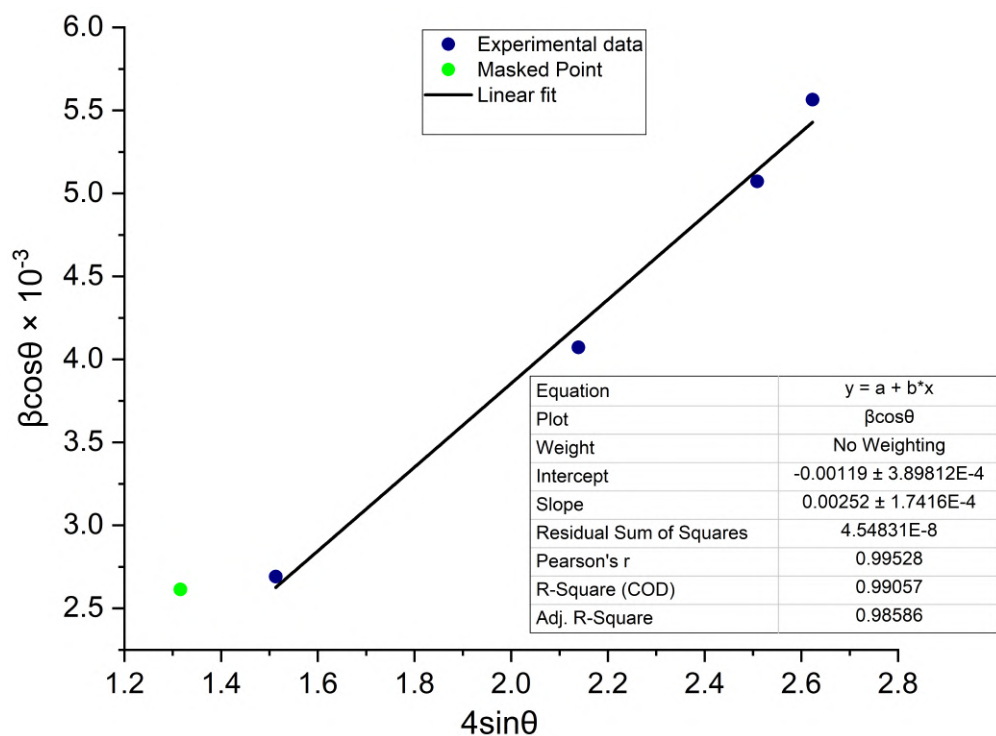


**Figure 4.18:** Williamson-Hall plot for Au sample irradiated at a fluence of  $7.70 \times 10^{16} \text{ p.cm}^{-2}$

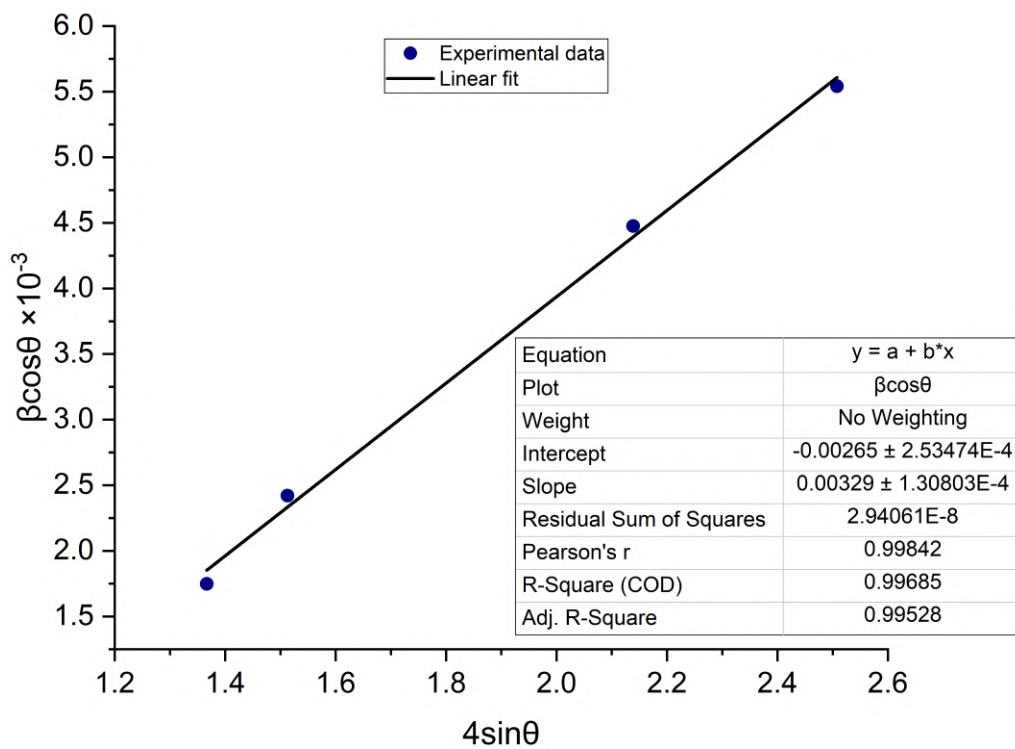


**Figure 4.19:** Williamson-Hall plot for Au sample irradiated to a fluence of  $2.32 \times 10^{17} \text{ p.cm}^{-2}$

**Table 4.13:** Intrinsic damage parameters for Au FCC lattice

| Sample | dpa   | D (nm)            | $\epsilon$<br>$\times 10^{-3}$ | $\delta$ (m <sup>-2</sup> )<br>$\times 10^{14}$ |
|--------|-------|-------------------|--------------------------------|-------------------------------------------------|
| (a)    | 0.013 | 152.18 $\pm$ 0.04 | 1.50 $\pm$ 0.11                | 0.4321 $\pm$ 0.0002                             |
| (b)    | 0.039 | 121.69 $\pm$ 0.05 | 2.52 $\pm$ 0.17                | 0.6753 $\pm$ 0.0005                             |
| (c)    | 0.078 | 54.65 $\pm$ 0.01  | 3.29 $\pm$ 0.13                | 3.3482 $\pm$ 0.0017                             |

Results of the microstrains calculated for the Au sample materials yielded a microstrain ranging from 0.15-0.33% with dislocation density ranging  $0.43 - 3.35 \times 10^{14} \text{ m}^{-2}$ . These values imply relatively high levels of dislocations in the microstructure post-irradiation.

The unirradiated Au sample had a microstrain value of 0.15%. This shows that there were lower levels of local strain in the microstructure of the materials indicating that the materials have not been subjected to stress fields that have created unrecoverable defects in the materials. After sample irradiation, the microstrain increased with an increase in the dislocation density indicating a relatively high induced radiation damage to the microstructure of Au due to proton irradiation. Usually, significant microstrains are experienced by microstructures with crystallite sizes between 100-300 nm when stress fields are applied to the material [195, 196]. This relates to the strain fields which arise from dislocations in the microstructure

Comparing these intrinsic lattice damage parameters with the irradiation fluence as shown in Figure 4.20 reveals that the average crystallite size decreased with increasing irradiation dose from (unirradiated sample) to the highest dose. In contrast, the microstrain and dislocation density increased. Studies have reported that the average crystallite size contribution is inversely proportional to microstrain and dislocation density [115, 197]. Material properties are largely affected by crystallite size, smaller crystallite sizes (small regions within a material that have a regular atomic structure) generally correspond to higher dislocation densities. Smaller grains sizes (the size of individual crystalline grains within a polycrystalline material) tend to hinder dislocation movements. This affects significantly the mechanical

and physical properties of the materials.

The trend observed in Figure 4.20 shows clearly that the change in the intrinsic microstructural features of the crystalline Au sample was chiefly influenced by proton irradiation. Microstructural degradation can occur via two possible radiation

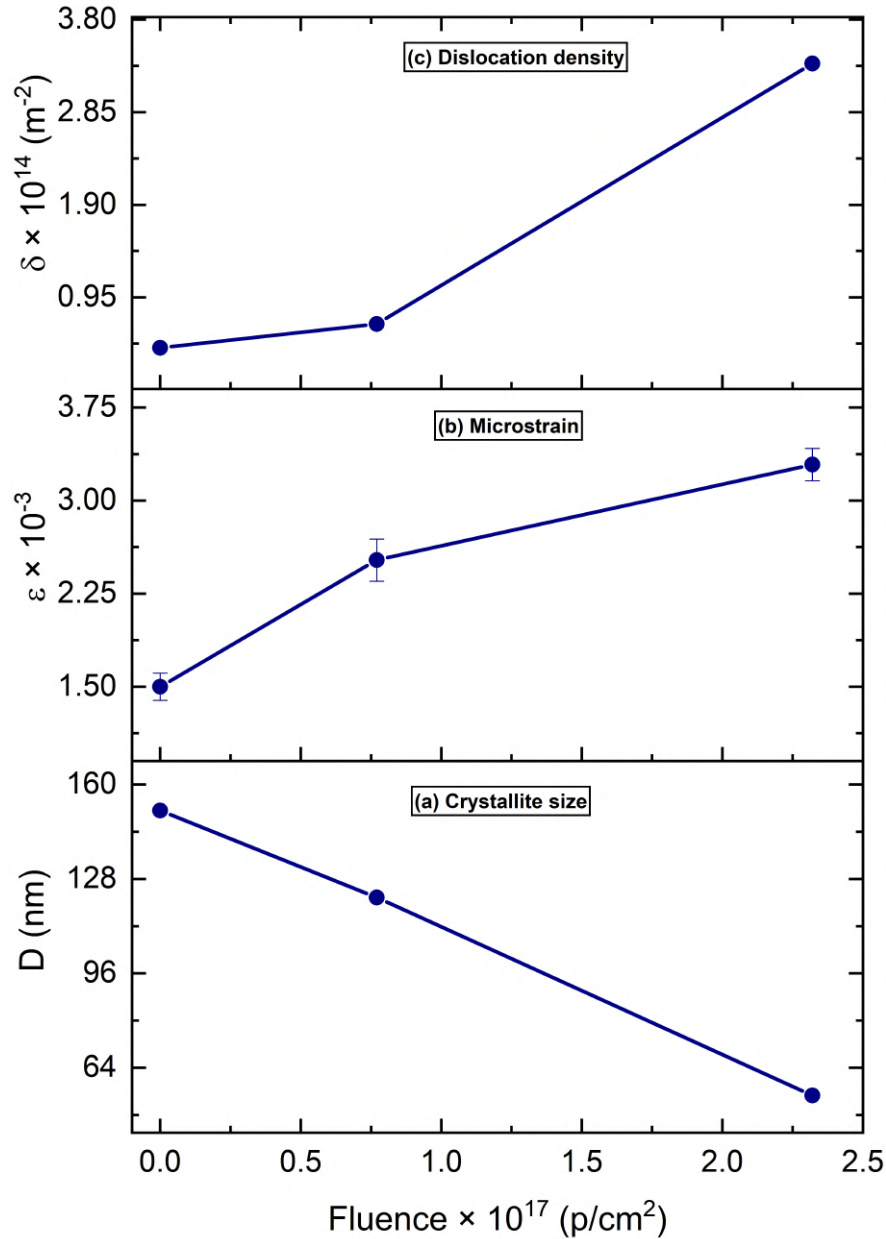


**Figure 4.20:** Plot of average (a) crystallite size, (b) microstrain, and (c) dislocation density for Au. The three data points represent proton fluence for the unirradiated (0.0), irradiated samples at fluences of  $7.70 \times 10^{16}$  p.cm<sup>-2</sup> and  $2.32 \times 10^{17}$  p.cm<sup>-2</sup>. Note that the error bars for (a) and (c) plots are close to the data points and hence not visible on the plots

damage processes that occur concurrently during sample irradiation. The vacancy creation, their accumulation, and subsequently the collapsing of the vacancies into

dislocation loops and secondary, the possible annihilation of the dislocation looks at their grain boundary sinks sites give rise to the strain fields on the microstructure of crystalline materials leading to an increase in the density of dislocations.

#### 4.4.3 Crystallite size ( $D$ ), microstrain ( $\varepsilon$ ) and dislocation density ( $\delta$ ) for Lu-Al sample

Again Lu-Al sample XRD data pre and post-irradiation was analyzed to obtain the average crystallite size, microstrain, and the dislocation density using the Williamson-Hall (W-H) approach. Results obtained from the plots are presented in Figure 4.21, 4.22, and 4.23. Similarly, the microstrain, and crystallite size were obtained from the slopes and intercepts of the graph and the dislocation density were calculated and presented in Table 4.14

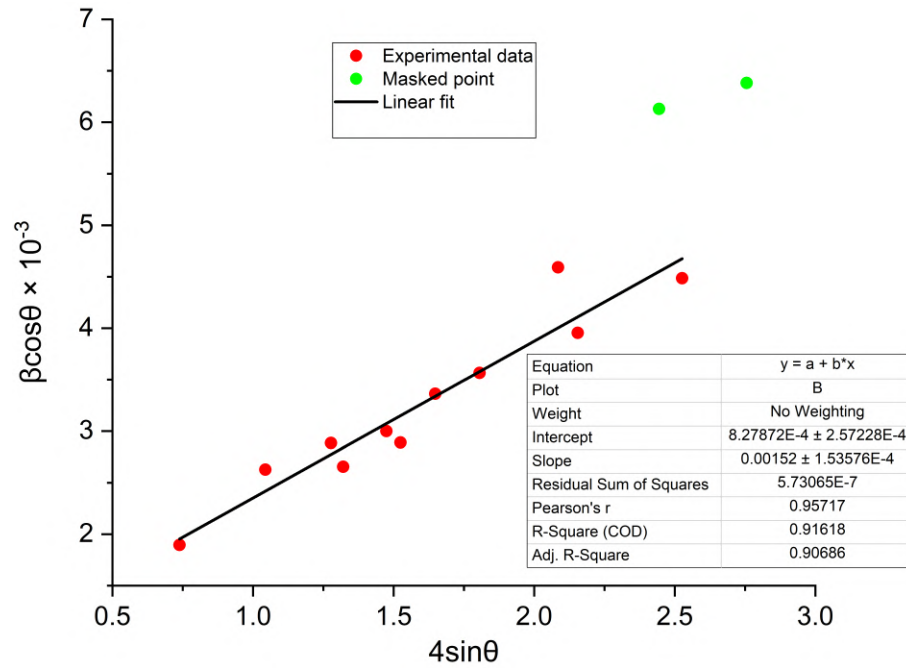


**Figure 4.21:** Williamson-Hall plot for the unirradiated Lu-Al sample

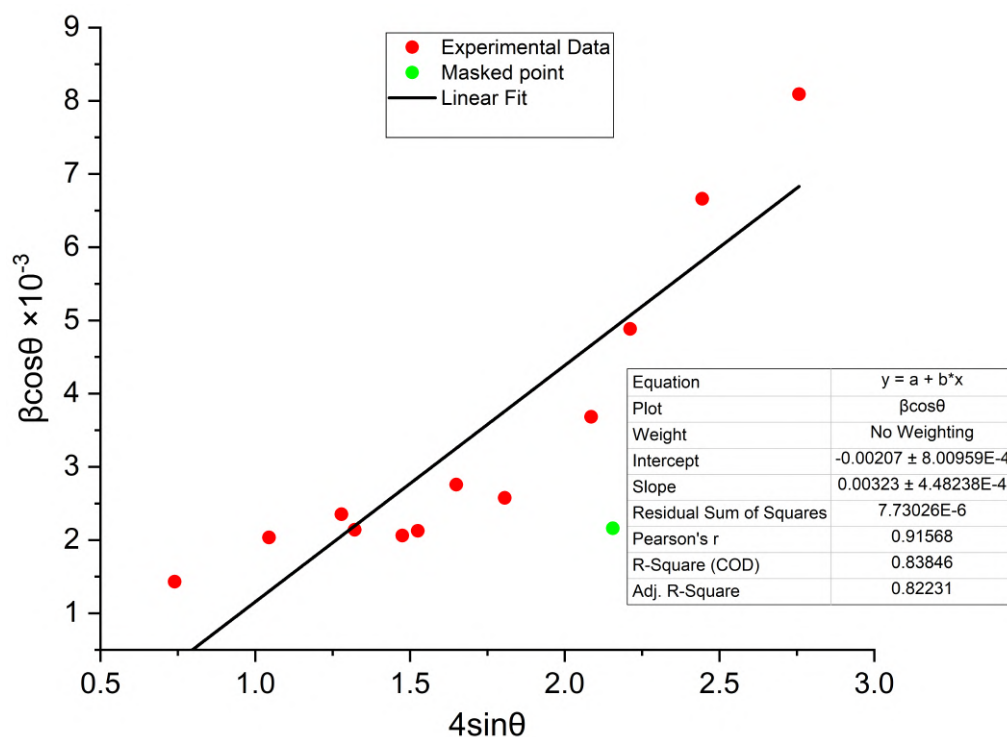


**Figure 4.22:** Williamson-Hall plot for Lu-Al sample irradiated at a fluence of  $7.70 \times 10^{16} \text{ p.cm}^{-2}$

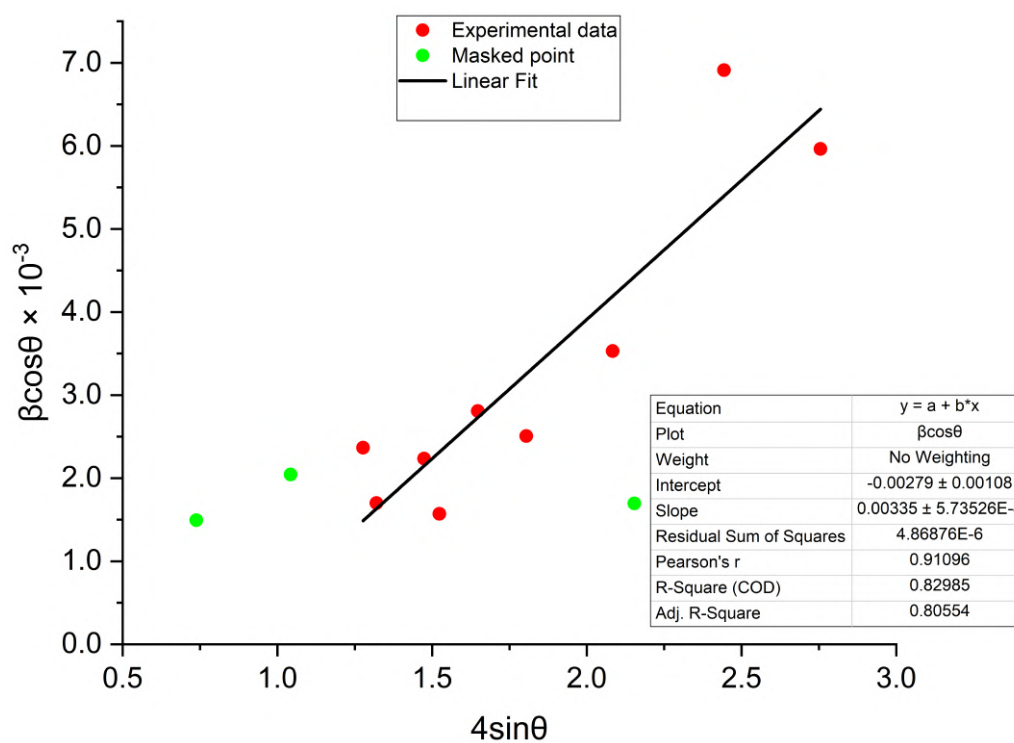


**Figure 4.23:** Williamson-Hall plot for Lu-Al sample irradiated to a fluence of  $2.32 \times 10^{17} \text{ p.cm}^{-2}$

**Table 4.14:** Intrinsic damage parameters for Lu-Al complex cubic lattice

| Sample | dpa    | D (nm)            | $\epsilon$<br>$\times 10^{-3}$ | $\delta$ (m <sup>-2</sup> )<br>$\times 10^{14}$ |
|--------|--------|-------------------|--------------------------------|-------------------------------------------------|
| (a)    | 0.0011 | 174.92 $\pm$ 0.05 | 1.50 $\pm$ 0.15                | 0.3268 $\pm$ 0.0017                             |
| (b)    | 0.0034 | 69.96 $\pm$ 0.06  | 3.23 $\pm$ 0.45                | 2.0432 $\pm$ 0.0033                             |
| (c)    | 0.0068 | 51.91 $\pm$ 0.06  | 3.35 $\pm$ 0.57                | 3.7117 $\pm$ 0.0080                             |

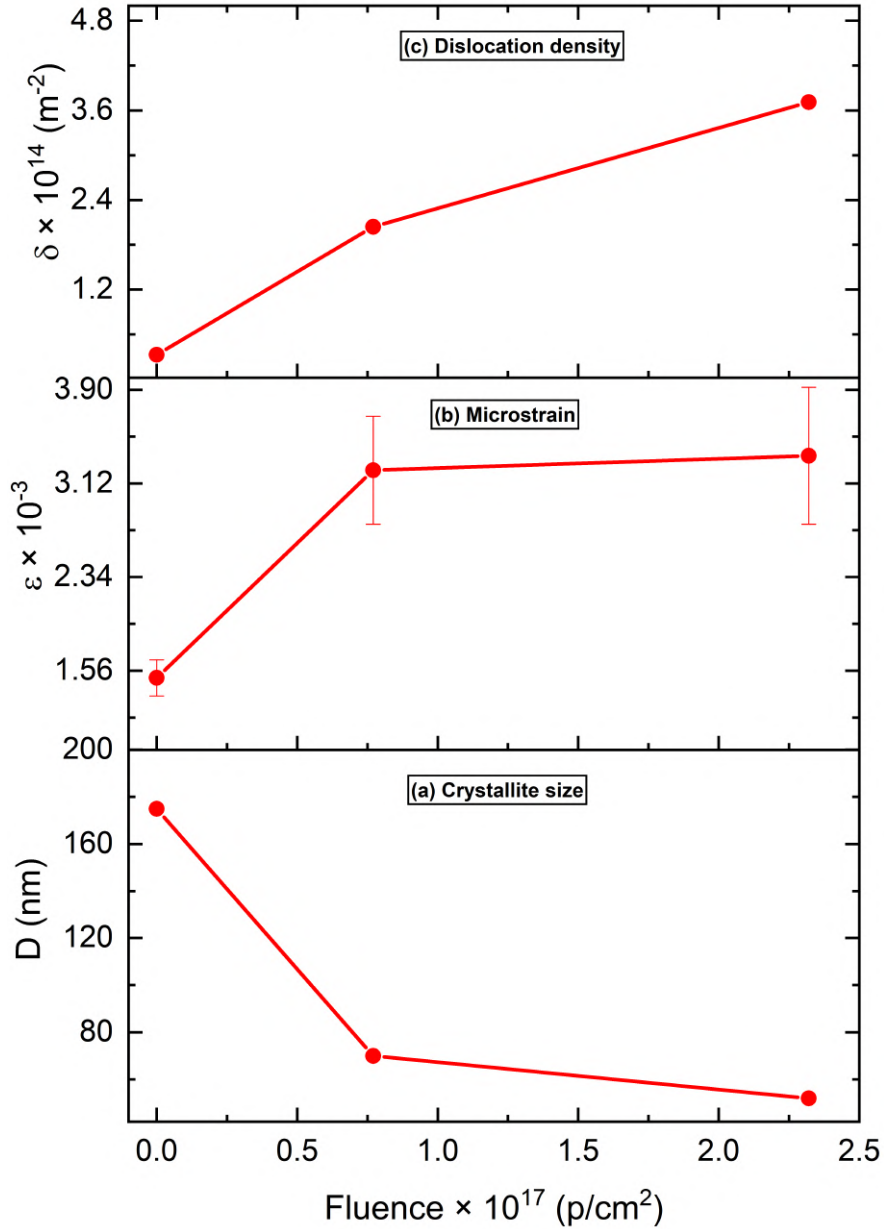


**Figure 4.24:** Plot of average (a) crystallite size, (b) microstrain, and (c) dislocation density for Lu-Al. The three data points represent proton fluence for the unirradiated (0.0), irradiated samples at fluences of  $7.70 \times 10^{16}$  p.cm<sup>-2</sup> and  $2.32 \times 10^{17}$  p.cm<sup>-2</sup>. Error bars for (a) and (c) are close to the data points and hence not visible on the plots

From Figure 4.21, The average crystallite size, microstrain, and average dislocation density were again calculated from the W-H plots and presented in Table 4.14. It can be seen that the microstrain ranged from  $1.50 - 3.35 \times 10^{-3}$  while the average crystallite size decreased. The dislocation density calculated from the average crystallite size increased significantly from  $0.3268 - 3.7117 \times 10^{14} \text{ m}^{-2}$ . The increase in the dislocation density followed an increase in the microstrain as shown in Figure 4.24.

The increase of the microstrain and dislocation density was small between (b) and (c). As earlier noted in the lattice spacing there was relaxation at (c) even though the irradiation dose was more than double the value at (b), this could mean a saturation point in the contraction of the crystallite size.

Dislocations in the microstructure of crystalline materials are a major contributing factor to the microstrain. This agreement observed in the trend of dislocation density and microstrain shows that the local strain induced in the material is due to the influence of the proton radiation field stress that the samples were exposed to during irradiation.

#### 4.4.4 Summary of XRD results

XRD analyses of the samples pre and post-irradiation showed changes in the crystalline peak intensities after irradiation for both Au and Lu-Al samples. There were also slight angular displacements of the peaks at their original positions after irradiation. These slight changes show that the contributions were due to sample broadening and not from instrumental alignment issues. As such the slight changes can be attributed to changes in the lattice spacing shown in Table 4.11 and 4.12 for both materials. Figure 4.20 and 4.24 showed a positive growth relationship between the proton fluences and the microstrain or dislocation density, indicating these microstructural properties were affected by proton irradiation. More dislocation density was observed with the Lu-Al sample than with the gold sample.

It is worthy of note however that, apart from the XRD technique, many microstruc-

tural characterization techniques measure dislocation density such as TEM and SEM-Electron Backscatter Diffraction (SEM-EBSD). In the XRD approach, dislocation density is calculated within defined crystallites. This implies that dislocations occurring close to the grain boundary sites are not accounted for. Therefore, XRD usually measures less dislocation density [63]. Also, the method of determination of crystallite sizes suffers from relatively high levels of uncertainties as such can not be used alone to conclude the level of dislocation in a crystalline material. However, measurement of dislocation density with XRD can be very informative on the relative comparison of dislocations in a crystalline material before and after irradiation with particles especially when the density of dislocation is relatively high approximating the order of  $10^{14} \text{ m}^{-2}$ .

Generally, all characterization techniques have their shortcomings and uncertainties in their measurement of average dislocation densities since different approaches are used in the computation of  $(\delta)$ . The high dislocation densities values obtained for Au and Lu-Al crystal solids in this study, ranging from  $0.33 - 3.71 \times 10^{14} \text{ m}^{-2}$  can be adequately measured using the XRD technique employed.

Utilizing microscopic imaging techniques such as TEM and SEM-EBSD may become more unreliable since at high dislocation densities there could be many overlaps of the dislocations [63]. Also, there could be difficulty in resolving the contrast during the imaging of such dislocations which may further complicate the process of estimating their densities leading to the incursion of more uncertainties in the measurements.

## **4.5 Results of damage characterization using HRTEM**

In this section, results of the pre and post-irradiation characterization of defects in the samples done with HRTEM are reported.

### 4.5.1 Chemical composition of materials

The composition of the materials was also verified using the Energy dispersive spectroscopy measurements with the HRTEM results shown in the maps and spectrum in Figures 4.27 and 4.28

#### (a) Chemical composition of Au

The chemical composition of the Au samples yielded 98.7% by weight Au and the balance Fe and Cr as shown in Figure 4.25 and 4.26. This value is close to the one in the datasheet. Usually, Fe and Cr are impurities that are present in gold samples. The EDS with HRTEM is a sensitive equipment so it was able to detect these two elements.

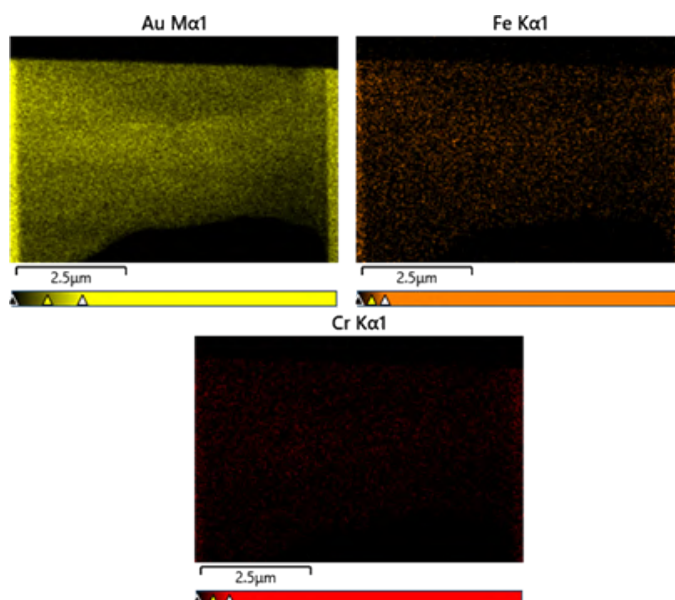


**Figure 4.25:** EDS Mapping for Au with the impurities Fe and Cr.

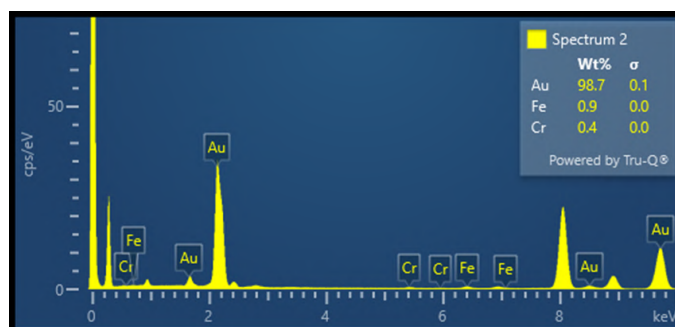


**Figure 4.26:** EDS mapping spectra for Au showing the percentage by weight (%wt) composition of the Au and the impurities Fe and Cr.

Comparing the results with those obtained through HRSEM, it is observed that the chemical composition of Au was slightly higher, and Si was detected instead of Fe and Cr. EDS results obtained with transmission electron microscopy (TEM) are generally considered more reliable than scanning electron microscopy (SEM). The chemical composition determined for Au using both methods is very close.

### (b) Chemical composition of Lu-Al

The mapping and spectrum for the chemical composition of Lu-Al are presented in Figures 4.27 and 4.28.

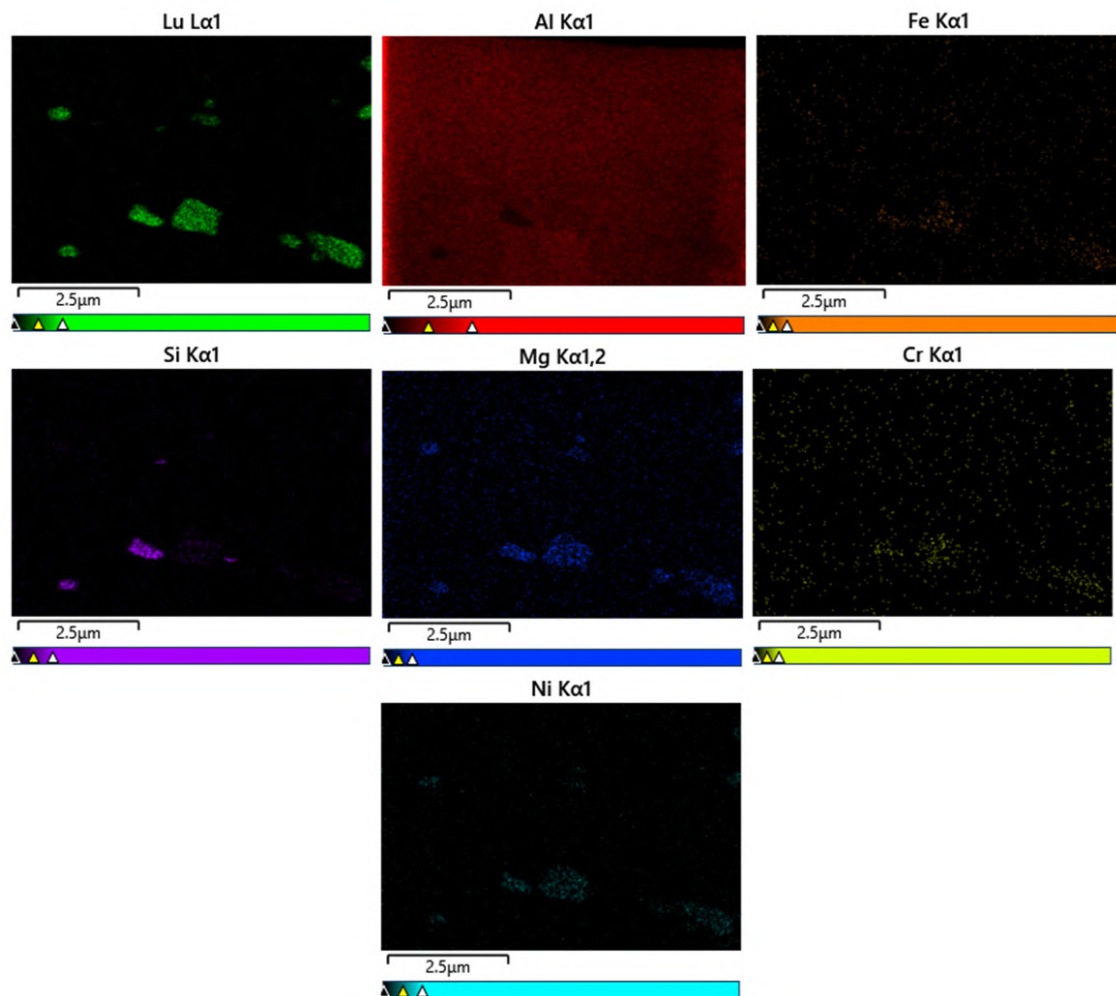


**Figure 4.27:** EDS Mapping for Lu-Al showing the constituents elements

Al and Lu had 94.72% and 4.92% respectively and the balance, impurities. These results are close to the datasheet (see Appendix D) values for this foil. Also, there is a slight variation in the composition of Lu-Al in both techniques. Considering

closely Figure 4.27 and 4.28 the EDS mapping and spectra, a peak of Ni is seen at  $\sim 1.16$  keV which corresponds to an  $M_\alpha$  line for Lu. Also, the composition of Ni is 0 which could mean that the peak is an M line for Lu. Other impurities such as Si, Mg, and Cr however have their peaks approximately at those positions as also seen on the quantified spectrum

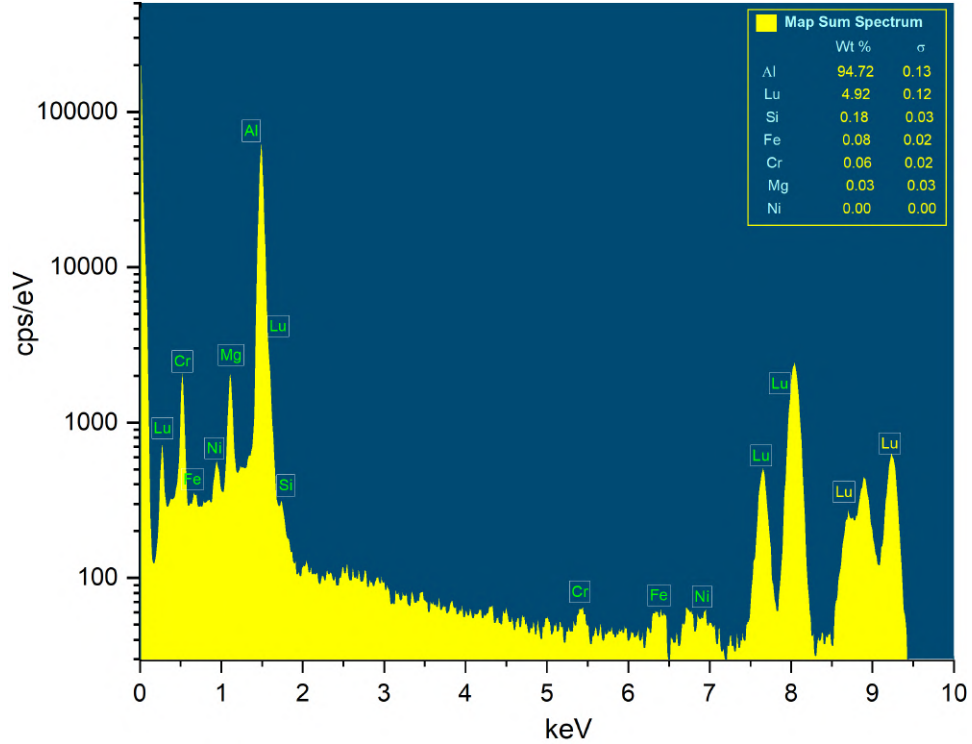


**Figure 4.28:** EDS mapping spectra for Lu-Al showing the %wt composition of Lu and Al with the impurities

Such variations are expected, as different techniques exhibit different sensitivities. This underscores the importance of verifying the composition using different techniques. In general results of the chemical composition of the materials agreed to a reasonable extent with both techniques and the datasheets provided for the materials.

## 4.6 Defects in irradiated sample materials

The defects observed in the Au and Lu-Al samples post-proton-irradiation are presented in this section.

#### 4.6.1 Defects in Au

Figure 4.29 (a)-(c) displays the STEM-BF micrographs for the unirradiated (a) and the irradiated Au at 0.013 and 0.039 dpa (b) and (c) respectively. These defects were observed at the  $[011]$  zone axis. The observed defects in these micrographs include small-sized faulted loops and dislocation loops formed alongside Stacking Fault Tetrahedra (SFT) due to the radiation-induced migration and agglomeration of many interstitial and vacancy clusters within the lattice planes.

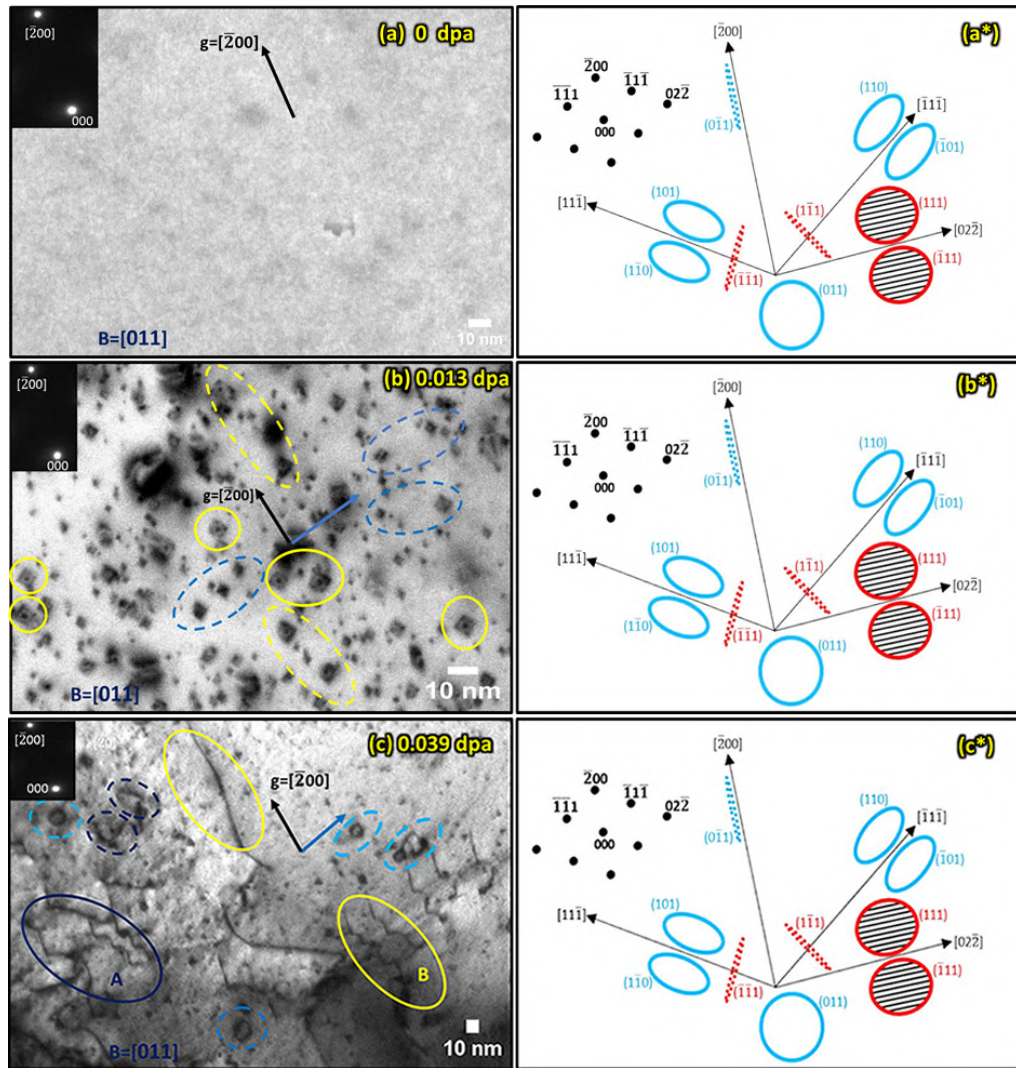


**Figure 4.29:** STEM Bright field (BF) micrographs showing the evolution of defects in the  $[011]$  zone axis (a) Unirradiated Au sample (b) Irradiated Au at 0.013 dpa showing irradiation-induced Stacking Fault Tetrahedra along with dislocation loops. (c) Irradiated Au sample at 0.039 dpa showing dislocation lines in dark blue and yellow ellipses 'A' and 'B' and some dislocation loops marked with blue and dark blue dotted ellipses.  $(a^*) = (b^*) = (c^*)$  is a simulated diffraction pattern of loop morphology for FCC-based materials under the  $[011]$  zone axis [198]

In (a), there are a few faint dots of low-gray area intensity values, suggesting the presence of some pre-existing faulted loops. In (b), numerous visible dislocation loops and faulted loops are observed on the microstructure. Some of the faulted loops may have been pre-existing but were too small to be observed. The microstructure also shows the formation of Stacking Faults Tetrahedral (SFT) along with dislocation loops. SFTs are formed in the gold microstructure through the interaction of partial dislocation loops. Previous studies [199] have suggested that SFTs are a type of complex defect with intricate arrays of vacant lattice sites formed at high dislocation points in the material. The formation of SFTs has been reported in gold materials after irradiation [46, 199, 200].

The SFTs formed in different planes had different morphology, In some the complete shape came out while others had a partial appearance. This is seen in a better perspective using the shape index map of the micrographs shown in Figure 4.30. The dotted yellow marked ellipses show SFT in the  $[\bar{2}00]$  direction having SFT that shows half the fault. The blue dotted ellipses show SFT in the  $[\bar{1}1\bar{1}]$  without the small loop inside. While the SFT along the  $[02\bar{2}]$  plane in a yellow solid circle appeared with all sides of the tetrahedra and the loop visible.

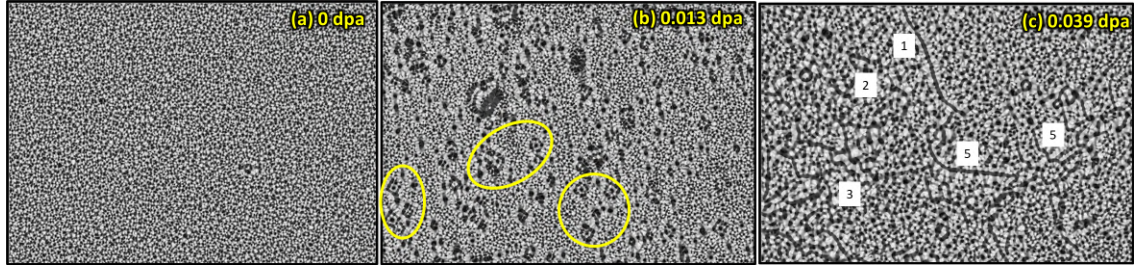


**Figure 4.30:** Shaped map index of Au foil STEM-BF images in  $[011]$  zone axis (a) Unirradiated (b) Irradiated to 0.013 dpa (c) Irradiated to 0.039 dpa

In (c), there is a formation of dislocation lines at point 'A' marked with a solid dark-blue ellipse, and with some lining up in a particular direction at point 'B' marked with a solid yellow ellipse as illustrated in Figure 4.30. The disappearance of dislocation loops and SFTs, along with the subsequent formation of dislocation lines, could result from the local strain induced by the radiation field stress during proton irradiation [200]. The interaction between Stacking Fault Tetrahedra (SFTs) and dislocation loops and the formation of dislocation lines is a complex and dynamic

**Table 4.15:** Crystallographic properties of FCC structure imaged at the  $[011]$  zone axis

| Habit plane                          | (111)         | ( $\bar{1}11$ )  | ( $1\bar{1}1$ )  | ( $11\bar{1}$ )  | (110)         | ( $\bar{1}10$ )  | (011)      | ( $01\bar{1}$ )  | (101)         | ( $\bar{1}01$ )  |
|--------------------------------------|---------------|------------------|------------------|------------------|---------------|------------------|------------|------------------|---------------|------------------|
| Family                               | faulted       | faulted          | faulted          | faulted          | perfect       | perfect          | perfect    | perfect          | perfect       | perfect          |
| <b>b</b>                             | $a/3[111]$    | $a/3[\bar{1}11]$ | $a/3[1\bar{1}1]$ | $a/3[11\bar{1}]$ | $a/2[110]$    | $a/2[\bar{1}10]$ | $a/2[011]$ | $a/2[01\bar{1}]$ | $a/2[101]$    | $a/2[\bar{1}01]$ |
| $\theta$ ( $^\circ$ )                | 35.26         | 35.26            | 90               | 90               | 60            | 60               | 0          | 90               | 60            | 60               |
| $\cos \theta$                        | 1:0.816       | 1:0.816          | edge-on          | edge-on          | 1:0.5         | 1:0.5            | 1:1        | edge-on          | 1:0.5         | 1:0.5            |
| DOIL                                 | $[01\bar{1}]$ | $[01\bar{1}]$    | $[21\bar{1}]$    | $[21\bar{1}]$    | $[1\bar{1}1]$ | $[11\bar{1}]$    | NA         | $[100]$          | $[11\bar{1}]$ | $[1\bar{1}1]$    |
| $\mathbf{g}_{-200} \cdot \mathbf{b}$ | non-zero      | non-zero         | non-zero         | non-zero         | non-zero      | non-zero         | 0          | 0                | non-zero      | non-zero         |

process that depends on various factors and radiation field stress is one of them.

The dislocation lines were aligned in the  $[\bar{1}01]$  and  $[101]$  habit planes. From Table 4.15, the angle ( $\theta$ ) of the intersection of the dislocation lines with the surface is acute ( $< 90^\circ$ ), hence these dislocation lines can be described as mixed dislocation lines. Also observed are large perfect loops along  $(\bar{1}01)$  and  $(1\bar{1}0)$  (see Table 4.15), marked with light and deep blue dotted ellipses, respectively.

The detailed crystallographic properties of the image zone axis  $[011]$ , such as the Family, Burgers vector ( $\mathbf{b}$ ), projection angle ( $\theta$ ), Aspect ratio ( $\cos \theta$ ), and the Direction of Intersection Line (DOIL) between the dislocation loop and the sample plane are shown in Table 4.15 for FCC materials [198]. Table 4.15 describes the possibility of the occurrence of a particular type of loop in a specific habit plane. In this  $[011]$  zone axis, it may be challenging to observe the dislocations along the  $(011)$  and  $(01\bar{1})$  habit planes when the two-beam condition is set at the  $\mathbf{g}_{200}$  vector direction. As the displacements per atom (dpa) increased from 0.013 to 0.039, there was an evolution of microstructural defects from dislocation loops, which formed due to the clustering of point defects (0-dimensional), to dislocation lines, which are 1 or 2-dimensional (2D) defects.

#### 4.6.2 Defects in Lu-Al

The observation of defects in the microstructure of Lu-Al, a diamond cubic material, was conducted in the  $[110]$  zone axis with the kinematic two-beam condition established between  $(000)$  and  $(220)$ . Radiation-induced defects in Lu-Al post-irradiation are depicted in Figure 4.31 (b)-(c). In (b), dislocation loops are evident, resulting from the coalescence of vacancies and interstitials. These loops, marked with yellow dotted ellipses and circles, may belong to the family of perfect loops. They exhibit clear patterns in the Lu-Al microstructure, with some aligning diagonally on a straight downward path and others following a semi-circular path.

Most of these dislocation loops are aligned along the  $(113)$  habit plane. Small black dots on the micrograph surface in (b) could represent faulted dislocation loops. In

(c), an increase in the irradiation dose leads to the evolution of these loops, with a few existing loops, the formation of dislocation lines, dislocation loops, and a high density of small-sized dislocation loops.

Dislocation lines are observed along  $(220)$ ,  $(113)$ , and  $(11\bar{3})$  habit planes, marked with solid yellow, orange, and dark blue ellipses, respectively. Faulted loops along  $(113)$  are also present. A closer examination of Figure 4.32c reveals coffee bean-type loops, marked with dotted dark-blue ellipses, which may represent perfect loops along the  $(\bar{1}\bar{1}3)$  plane when compared to Figure 4.31c.

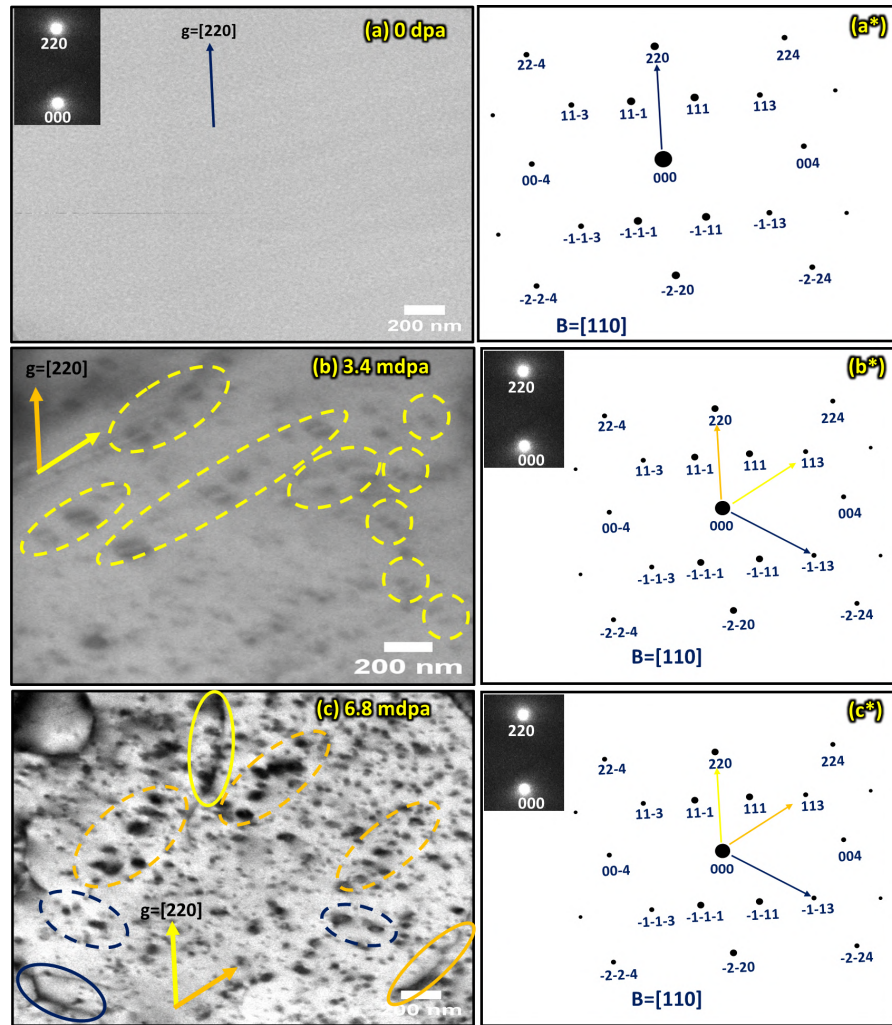


**Figure 4.31:** STEM Bright field micrographs showing the evolution of defects in  $[110]$  zone axis (a) Unirradiated Lu-Al sample (b) Irradiated Lu-Al to 0.0034 dpa (3.4 mdpa) showing irradiation-induced dislocation loops in yellow marks (c) Irradiated Lu-Al sample to 0.0068 dpa (6.8 mdpa) showing line dislocations numbered 1-6, the blue marked circles (A) are dislocation loops, the yellow circle marks (B) could be cavities and the red circle marks are high-density dislocation loops.  $(a^*)$ ,  $(b^*)$ , and  $(c^*)$  are the DC indexed pattern in the  $[110]$  zone axes

Analyzing three-dimensional images in a two-dimensional space may make it challenging to discern detailed microstructural features, particularly for materials like Lu-Al. These dislocation loops and lines are better observed when the image is transformed into a shaped map index using ImageJ software, providing clearer visibility of depth and solidity of features, as shown in Figure 4.32.

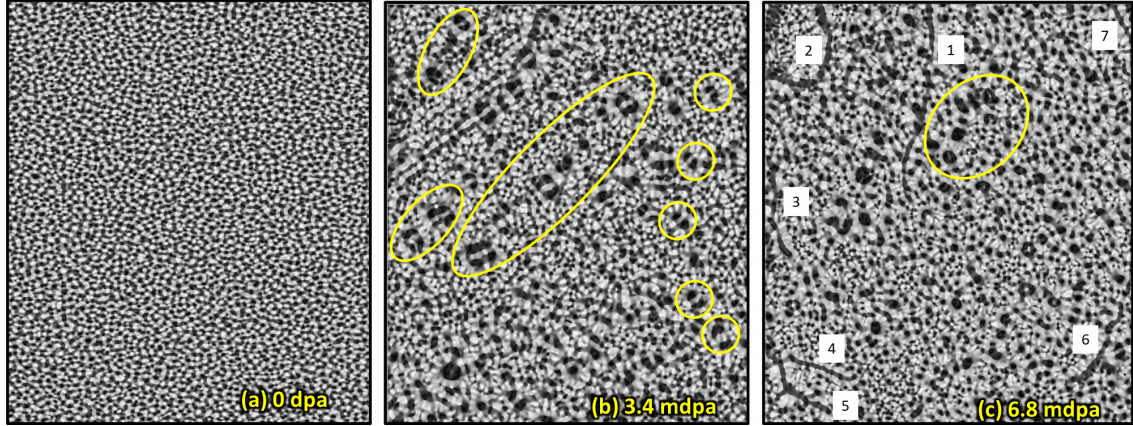


**Figure 4.32:** Shaped map index of Lu-Al foil STEM-BF images in  $[110]$  zone axis (a) Unirradiated (b) Irradiated to 3.4 mdpa (c) Irradiated to 6.8 mdpa

The microstructural features in (a) exhibit a well-shaped micrograph, similar to the one shown in Figure 4.31 above. In (b) and (c), the irradiation-induced features are revealed in a more detailed perspective. Different dislocation loop families can be clearly distinguished. In (b), the coffee bean-shaped dislocation loops and their alignment are highlighted. Additionally, in (c), faulted dislocation loops and dislocation lines 1-7 are now observed with more clarity.

## 4.7 Radiation damage levels in Au and Lu-Al

In Transmission Electron Microscopy especially in a two-beam condition, the BF signal generated at a specific location in the specimens is amplified and the corresponding signal to the point imaged on the specimen is recorded on the computer in the form of pixels [146]. The signals acquired for the whole surface are classified based on the intensity of the received beams to form images on the micrographs.

Usually, the grayscale value distribution on the micrograph has low and high values. Low gray values could be an indication of defective areas and high gray values

are areas of excess intensity where the crystal is maintaining its regular structure. Surface plots in the z-axis projection were used to compare the grayscale values on the micrographs of the unirradiated and post-irradiated samples.

Figures 4.33 and 4.34 show surface profile plots for Au and Lu-Al samples. The surface profiles show the variation in the gray value on the micrographs. The unirradiated samples of Au and Lu-Al had only the purple colour on the surface profile indicating a non-defective surface. The area of lower gray values is represented in green.

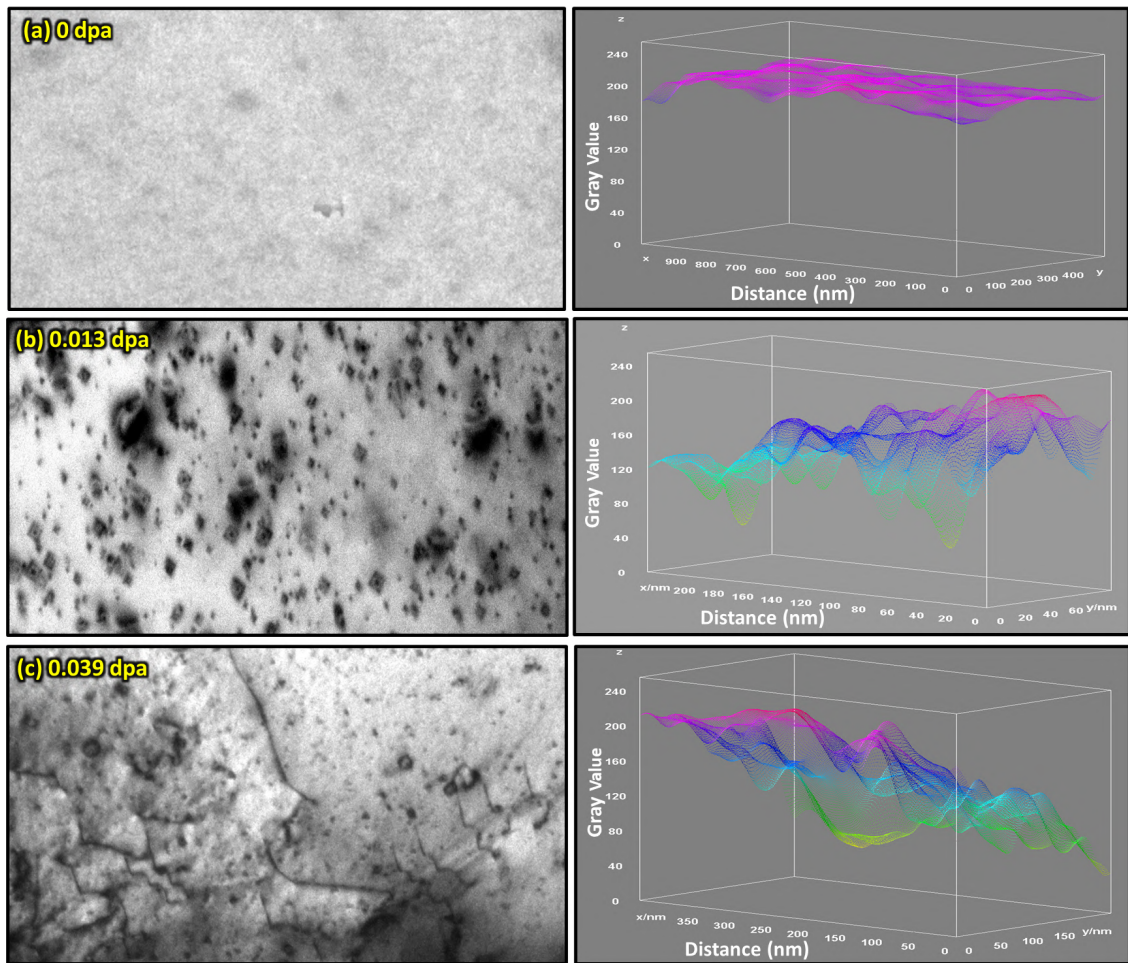


**Figure 4.33:** Surface profile of Au foil STEM-BF images (a) Unirradiated (b) Irradiated to 0.013 dpa (c) Irradiated to 0.039 dpa

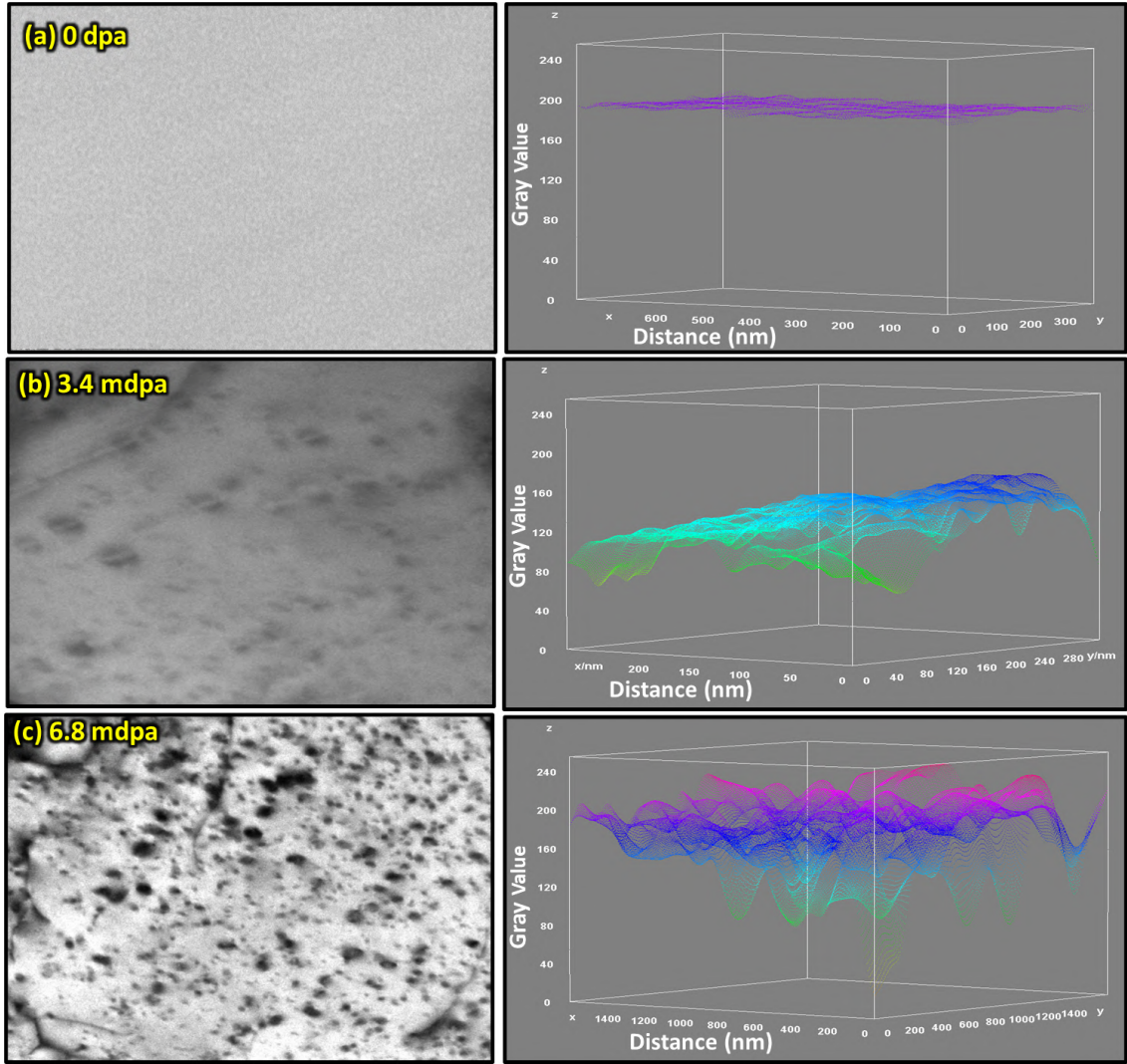


**Figure 4.34:** Surface profile of Lu-Al foil STEM-BF images (a) Unirradiated (b) Irradiated to 3.4 mdpa (c) Irradiated to 6.8 mdpa

These areas have lower gray values represented along the z-axis showing defective portions of the micrographs with weak electron transmission signals. Following the trend in both samples, it is observed that Figure 4.33c and 4.34c had the lowest gray area values indicating the highest level of damage on both samples.

#### 4.7.1 Dislocation density ( $\delta$ ) in Au

To effectively calculate the dislocation density, the surface of the analysed sample was partitioned into four quadrants 'A-D' as shown in Figure 4.35, and the number of loops and lines were counted in each quadrant. The same procedure was used for all other images. The average dislocation loop and line densities on the analysed sample were calculated using Eq. 3.58 and 3.59.

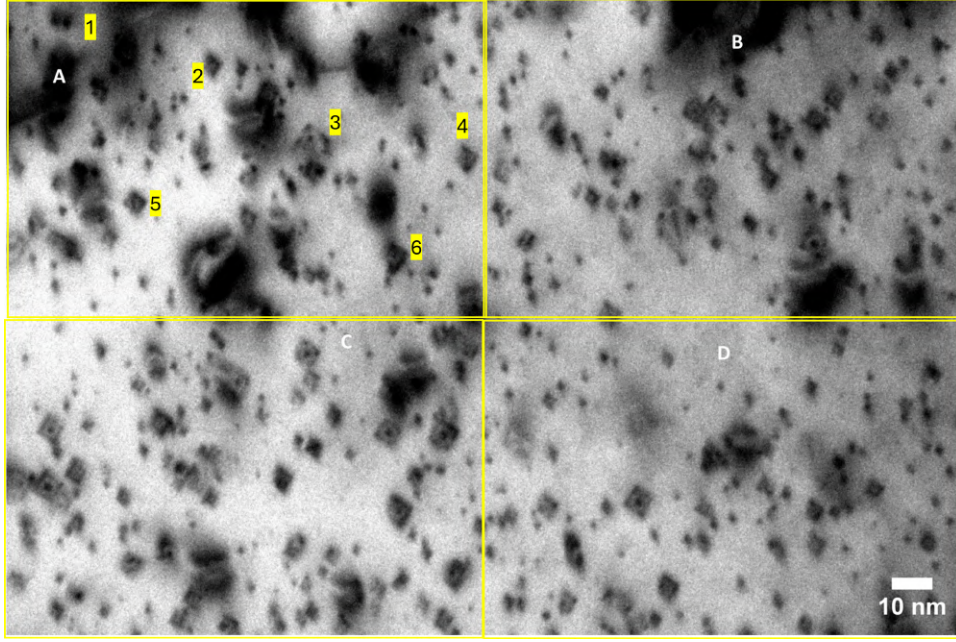


**Figure 4.35:** Estimation of dislocation density of STEM BF micrograph irradiated Au

Results of the dislocation density for the irradiated Au samples is presented in Table 4.16. In Table 4.16, the measurement of the average dislocation density for the unirradiated sample (a) was not possible because there were no intersecting dislocations on the TEM micrograph. The average dislocation density in (c) was higher than that in (b). This indicates that the damage in the Au material increased with an increase in the dpa, aligning with the qualitative analysis discussed in the earlier section. The fractional error in the measurement of the average dislocation density using this direct counting method is approximately 22% for (c).

**Table 4.16:** Dislocation density for irradiated Au

| Sample        | $\delta \text{ (m}^{-2}\text{)} \times 10^{14}$ |
|---------------|-------------------------------------------------|
| (a) 0 dpa     | -                                               |
| (b) 0.013 dpa | $1.99 \pm 0.16$                                 |
| (c) 0.039 dpa | $3.61 \pm 0.22$                                 |

#### 4.7.2 Dislocation density ( $\delta$ ) in Lu-Al

Using the same procedure of direct counting, the average dislocation density was calculated for the irradiated samples of Lu-Al shown in Figure 4.36.

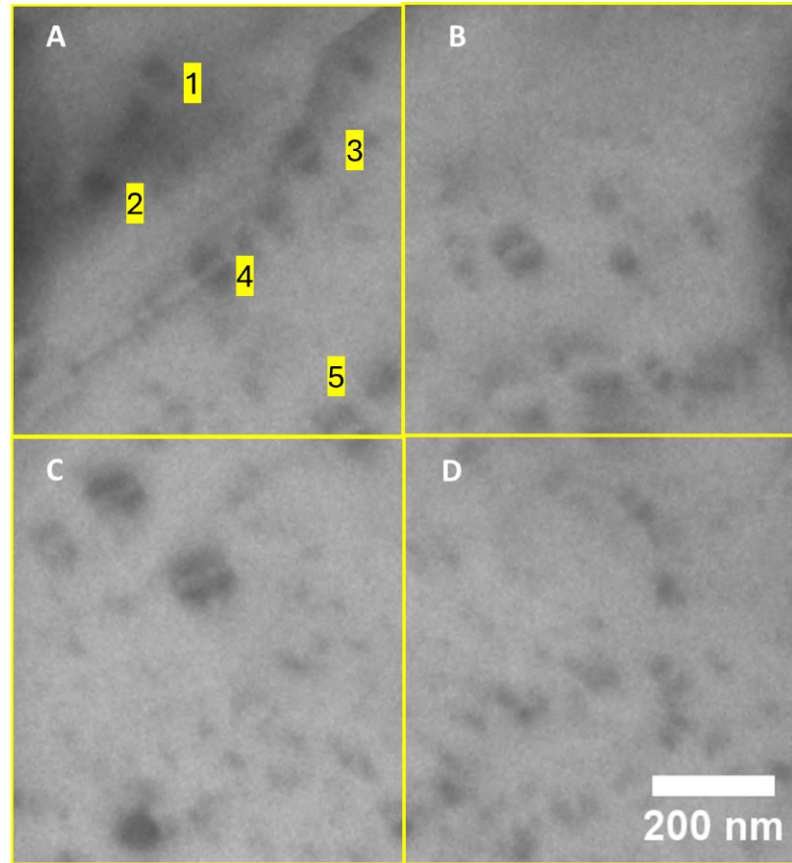


**Figure 4.36:** Estimation of dislocation density of STEM BF micrograph irradiated Lu-Al

The results of the average dislocation for the two irradiated doses and their accompanied uncertainties are presented in Table 4.17.

**Table 4.17:** Dislocation density for irradiated Lu-Al

| Sample       | $\delta \text{ (m}^{-2}\text{)} \times 10^{14}$ |
|--------------|-------------------------------------------------|
| (a) 0 dpa    | -                                               |
| (b) 3.4 mdpa | $1.33 \pm 0.15$                                 |
| (c) 6.8 mdpa | $4.76 \pm 0.21$                                 |

The average dislocation densities obtained are still in the order  $10^{14}$  which is a relatively high value of dislocation densities for a metallic alloy. Again, the fractional error for sample (c) is 21% which is relatively a high error.

## 4.8 Summary of HRTEM results

The post-irradiation characterization of damage in Au and Lu-Al samples has been accomplished. The chemical composition analysis of the sample materials was performed using electron diffraction spectroscopy. HRTEM provided more accurate chemical composition data, detecting even trace impurities inherent in the Lu-Al samples, in contrast to HR SEM. It is important to note that while EDS is not the most precise tool for chemical analysis, it provides a fair estimate of elemental distribution in the sample, offering insights into the composition of the materials as indicated in the datasheet provided in Appendix D.

Imaging defects with HRTEM is a challenging and time-consuming task. In defect analysis with HRTEM, specific methods, and zone axes are selected to enhance comparison. Setting up these specific conditions takes time and results may not be entirely free from uncertainties due to factors such as contrast and tilting of specimens.

The on-zone two-beam condition enabled the observation of defects in the TEM samples analyzed. Evident defects in the Au sample in the  $[011]$  zone axis included Stacking Faults Tetrahedra (SFT), faulted loops, perfect loops, and dislocation lines. In Lu-Al, dislocation loop families, which could be perfect and faulted loops, and dislocation lines were observed. The specific family of loops in Lu-Al remains uncertain due to limited data on the material, and no previous work on radiation damage has indexed the diffraction pattern of Lu-Al for comparison. However, since the primary focus is on assessing the level of damage at different doses of proton radiation, the obtained results provide valuable insights. Average dislocation densities were estimated for the materials, and there was a dose-response increase in dislocation densities for both Au and Lu-Al.

## 4.9 Comparison of dislocation densities measured with XRD and HRTEM

Damage characterization was done using XRD and TEM techniques. Qualitative and quantitative approaches were utilized. The most common quantitative microstructural parameter for quantification of damage on the materials' is the average dislocation density ( $\delta$ ). This was estimated in both characterization techniques and a comparison of the dislocation density for the sample materials in both techniques at different irradiation doses is illustrated in Figure 4.37.

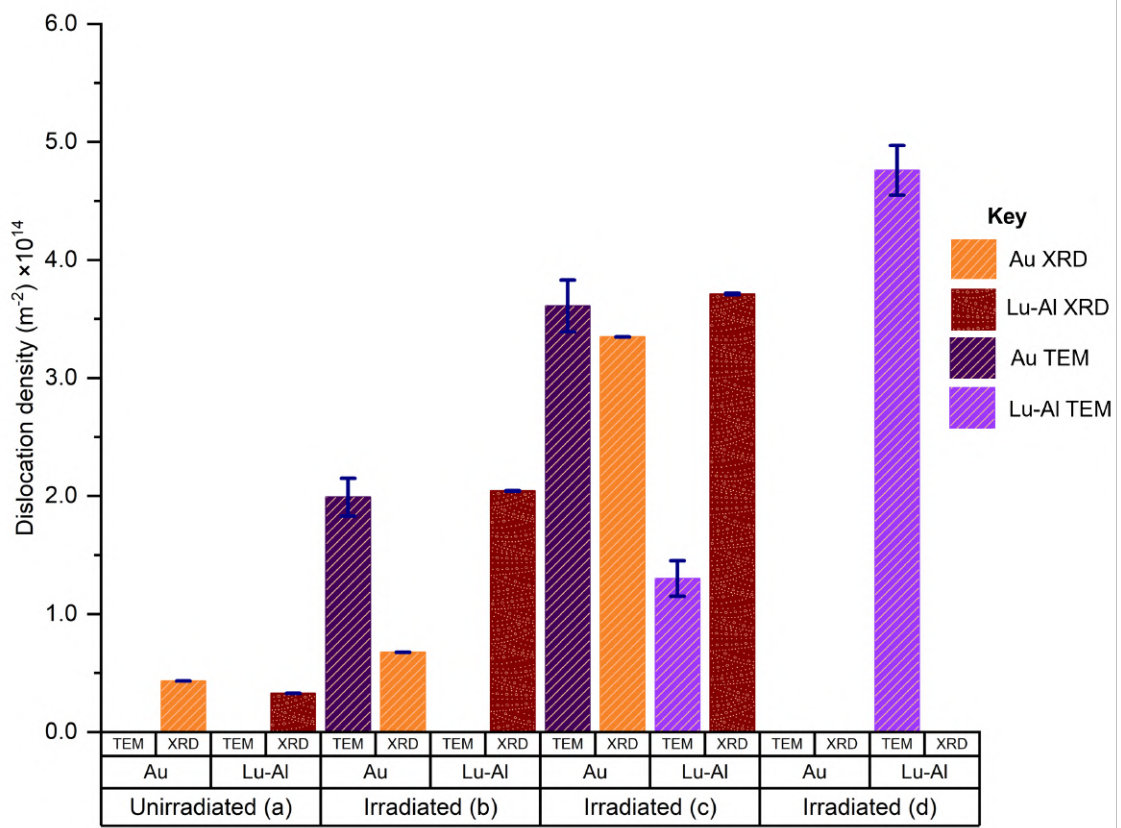


**Figure 4.37:** Average dislocation densities of Au and Lu-Al samples in XRD and TEM Techniques (a) Unirradiated sample (b) samples irradiated to  $7.70 \times 10^{16}$  p.cm $^{-2}$  (c) samples irradiated to  $2.32 \times 10^{17}$  p.cm $^{-2}$  (d) irradiated to  $4.64 \times 10^{17}$  p.cm $^{-2}$

The average dislocation densities obtained in this study ranged from  $0.67 - 3.71 \times 10^{14}$  m $^{-2}$  and  $1.3 - 4.76 \times 10^{14}$  m $^{-2}$  with XRD and TEM techniques respectively. These results of dislocation density obtained in this study are consistent with an earlier observation that XRD may miss some dislocations near grain boundaries,

leading to lower values of defect density [63]. In the second irradiation dose for both materials were of order  $10^{14}$  these values are relatively high.

Also from the XRD pattern, the crystal structure of the materials was identified. This was very important in identifying the various habit planes of the imaged zone. Estimation of crystallite size was found to decrease with increasing strain and dislocation density for both materials which is consistent with other studies [63]. From Figure 4.37, the average dislocation density calculated using the XRD and TEM techniques for Au and Lu-Al are respectively represented in the key at the top right corner.

It can be seen that the dislocation density in the unirradiated samples of Lu-Al was lower than that in the Au sample than in the Lu-Al sample. The dislocation density in Lu-Al is higher than in gold in irradiated (b) and (c) with the XRD technique. The values  $\delta$  obtained for Lu-Al are within the range reported by Sidor et al [201].

Using the TEM technique no measurement of dislocation density on the unirradiated sample. Also, the measurement for damage in TEM lamellae for the Lu-Al sample was done at fluences in the irradiated batch (c) and (d) to achieve relatively higher dpa values. Generally, for both materials, and both techniques the  $\delta$  increased with increasing radiation dose. It can be observed that the error in the TEM measurement was relatively high. This is largely influenced by how defect sizes are estimated by any particular technique used as illustrated in Figure 4.38.

This is not unexpected as the method of direct counting is prone to errors in the way the two approaches measure average dislocation density [147]. In this study, we focused on the [011] and [110] zone axes for Au and Lu-Al samples this on-zone observation limits the number of dislocations loops or lines that could intersect in the plane that is imaged. Also, much analytical processing of images is done to obtain the dislocation density with the method used in the study.

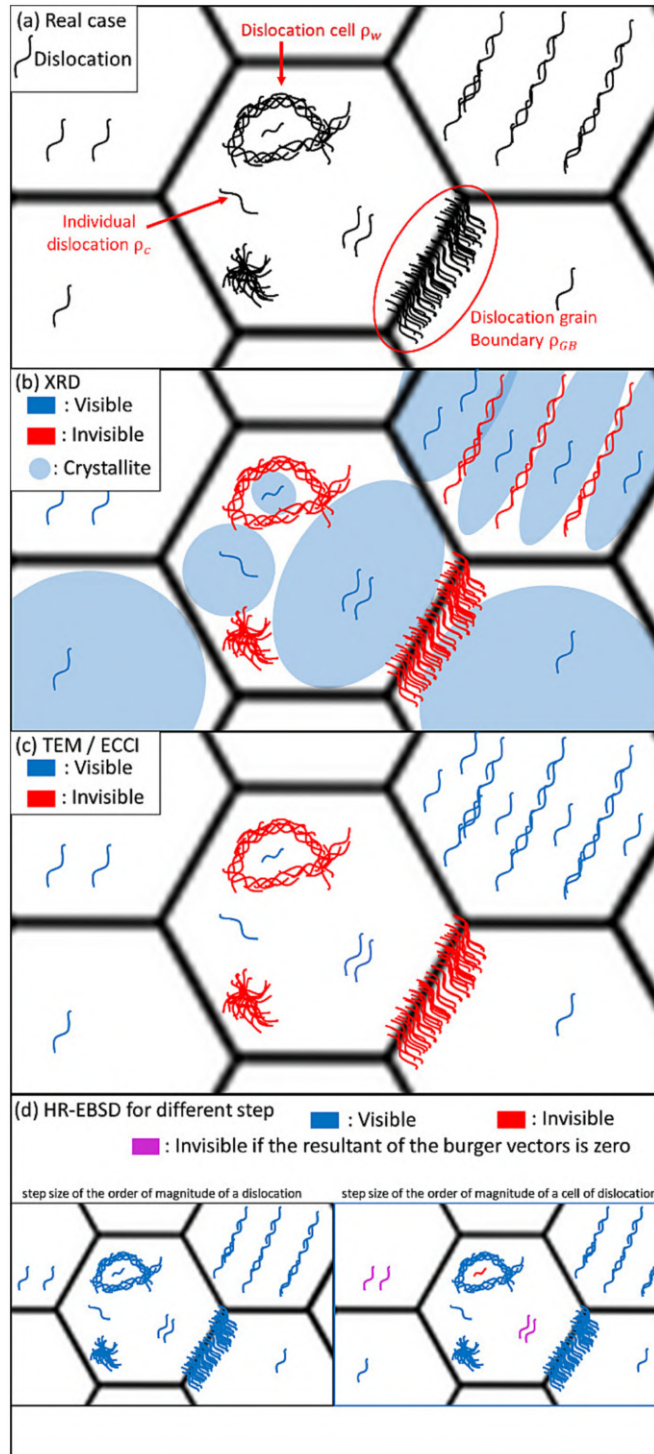


**Figure 4.38:** Illustration of approaches in the estimation of dislocation density ( $\rho = \delta$  as used in this study) based on grain pattern (a) The real case (b) using XRD where crystallite size ( $D$ ) is utilized in the calculation (c) Analysis of dislocation by TEM/ scanning electron microscopy (ECCI) imaging approach (d) HR-Electron Backscatter diffraction (EBSD) approach that depends on the distinction between pixel size. Ref.[63]

The level of uncertainty in the estimation of  $\delta$  in the microstructure with imaging methods such as TEM and SEM-EBSD becomes higher as the deformation increases

above  $1-3 \times 10^{14}$  [63]. Whereas XRD technique estimates become more relevant at higher  $\delta$  values such as those obtained in this work. It should also be noted that a direct comparison can not be made between  $\delta$  measured with XRD and other imaging methods with certainty since each technique approaches the measurement of  $\delta$  differently as shown in Figure 4.38. While XRD analysis  $\delta$  by crystallite size dependence, imaging techniques like TEM focus on the number of dislocation loops or lines intersecting the surface of the analysed sample.

## 4.10 Results of damage characterization with Flash DSC

The Figures 4.39, 4.40, and 4.41 show curves of the melting curves of the unirradiated, and irradiated samples at 1.1 mdpa and 3.4 mdpa. The observed Six (6) curves in the negative power region are the heating curves while the remaining six (6) in the positive power region are the cooling curves. The Power-Temperature profiles, representing melting curves, illustrate the first post-melt-in cycle in yellow and the last cycle in purple, where all expected damage in the samples is anticipated to have annealed.

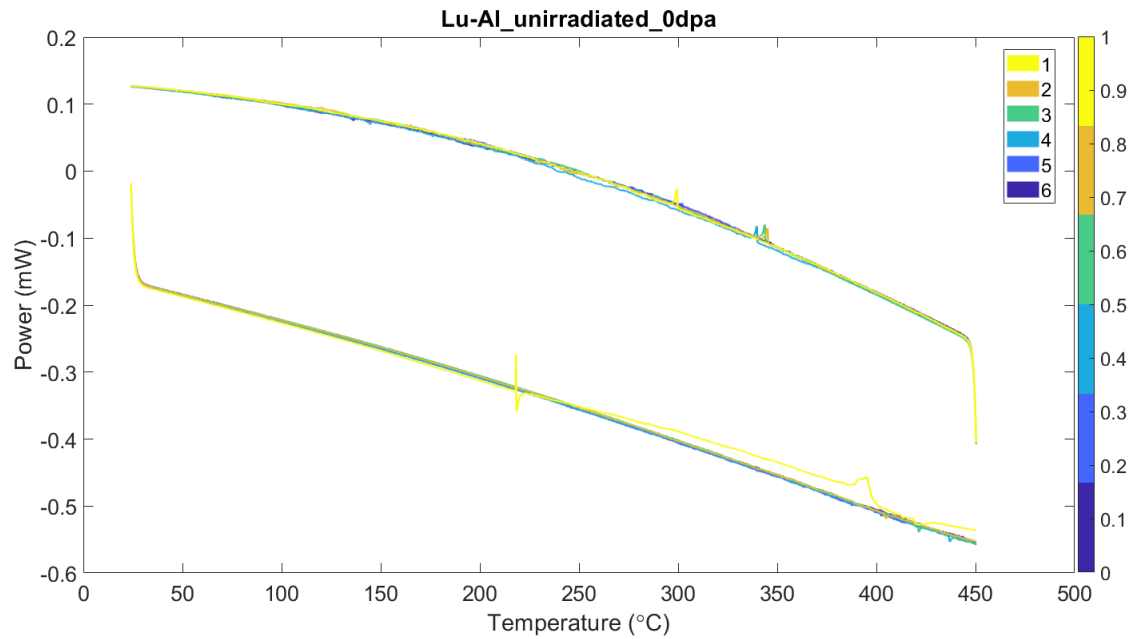


**Figure 4.39:** Power-Temperature profiles of Unirradiated (0 dpa) sample melting curves showing damage annealing in Lu-Al sample

In Figure 4.39, the unirradiated sample reveals no distinctive features of interest, indicating annealed damage. However, in Figure 4.40, the curves of the first irradiated sample are not consistently flat, showing a noticeable spike at approximately 160°C, suggesting a potential transition. Importantly, this spike is observed only during the first heating cycle (depicted in yellow).

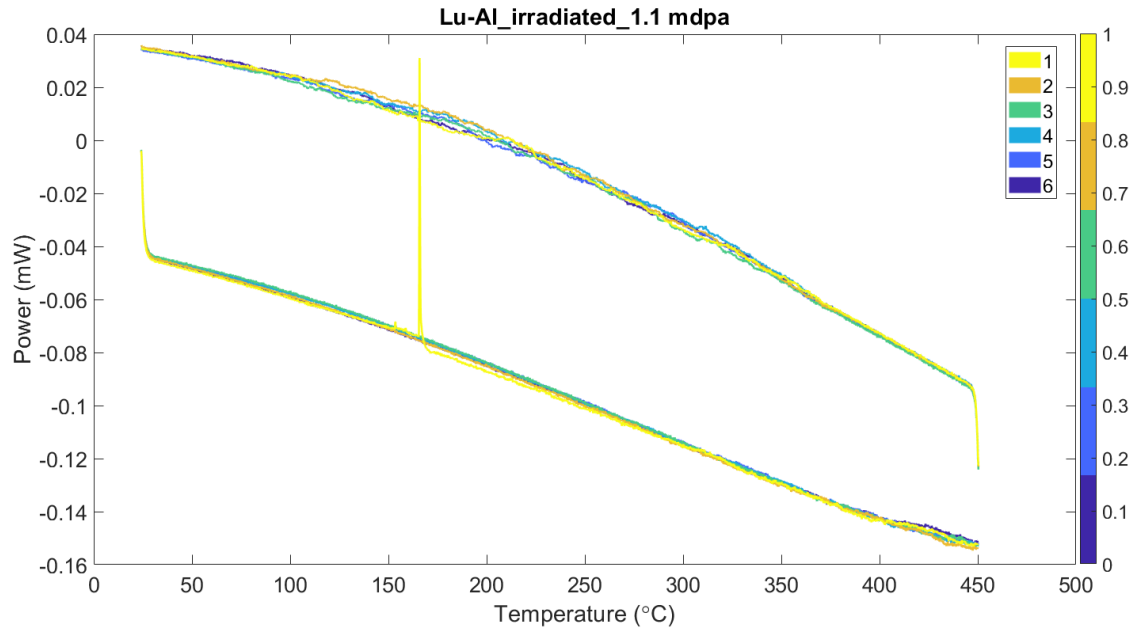


**Figure 4.40:** Power-Temperature profile of irradiated sample to 1.1 mdpa. Melting curves showing damage annealing in Lu-Al sample

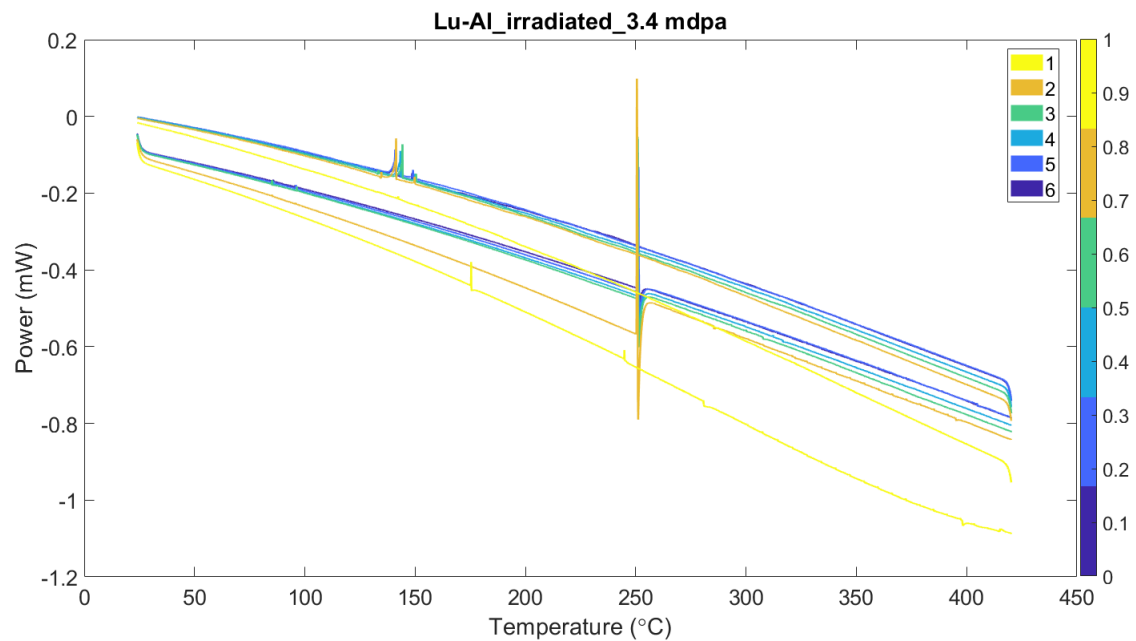


**Figure 4.41:** Power-Temperature profile of irradiated sample to 3.4 mdpa. Melting curves showing damage annealing in Lu-Al sample

Figure 4.41 presents results for irradiation at 3.4 mdpa, revealing intriguing features. Notably, there is a recurring spike around 250°C that persists beyond the first heating cycle, suggesting a possible phase transition at this temperature. Moreover, there appears to be a region of opposite curvature between 240°C and 420°C, potentially indicating a radiation-induced process not observed in the unirradiated and 1.1 mdpa irradiated samples.

In summary, these preliminary results depict alterations in the profiles of the melting curves. These variations may be attributed to the influence of proton irradiation that was performed on the materials.

## 4.11 Chapter Summary

In this chapter, results of theoretical simulations of damage and experimental quantification of actual damage on materials. The simulation of radiation damage by neutrons on Au and Lu-Al involved the use of MCNP6.2, while ion beam radiation damage on the samples was carried out using the Binary Collision Approximation (BCA) code SRIM-2013. The objective was to assess the feasibility of using protons or other ions as a surrogate for neutrons, as suggested by prior studies. The calculations of displacements per atom (dpa) rates indicated that ion beams could induce radiation damage on metallic samples similar to those employed in this study.

The simulations demonstrated that proton irradiation is cost-effective, yielding higher dpa values in a shorter irradiation time, along with other advantages outlined in preceding chapters. Importantly, the simulated data provided insights into the energy and dose requirements for proton irradiation. The CLASS accelerator beam parameters were adjusted to achieve dose rates based on SRIM-2013 calculations.

Damage characterization employed XRD and TEM techniques, using both qualitative and quantitative approaches. The average dislocation density ( $\delta$ ), which is a common quantitative microstructural parameter for assessing damage, was estimated in both characterization techniques. Also, the results obtained from Flash

DSC measurement showed increasing damage effects as the dpa increased which is consistent with the other techniques.

# Chapter 5

## Discussion of results

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This chapter delves into the results derived from two stochastic simulation methods and the experimental observations, utilizing the damage characterization techniques employed in this study. It explores the implications of these results on radiation damage for the materials investigated.

### 5.1 Radiation damage prediction and implication

This section discusses the simulation results of the interaction of neutrons and protons with the materials investigated. This covers objectives 1 and 2 which is performed in Chapter 3 and the results reported in Chapter 4. The results of neutron irradiation with MCNP6.2 and proton irradiation with SRIM-2013 provide valuable insights into the effect of radiation on the microstructure of materials. By leveraging the capabilities of MCNP and SRIM, light is shed on the intricacies of radiation-induced phenomena with neutrons and protons.

#### 5.1.1 Displacement Per Atom (dpa) rates

Quantifying damage levels in crystalline materials requires a quantitative measure that analytical calculation methods struggle to achieve due to the intricate nature of the radiation damage evolution process. Utilizing Monte Carlo methods, radiation damage by neutrons to Au and Lu-Al was quantified in the Monte Carlo N-Particle code (MCNP6.2). Similarly, damage caused by ion beams on the studied materials was calculated using the SRIM-2013 code.

The goal of using the quantity dpa was to predict the radiation dose to materials resulting from neutron and proton irradiation, as outlined in the research question

in Section 1.3.1. Additionally, we aimed to compare neutron irradiation with proton irradiation as an alternative irradiation field source, considering the challenges associated with obtaining neutron irradiation (see Section 1.4, second objective). Furthermore, these predictions were intended to guide us towards selecting a specific ion energy capable of producing displacement per atom rates comparable to those induced by neutrons

Predictions of displacement per atom rates produced by neutrons in Au and Lu-Al samples were calculated using MCNP6.2. The results revealed that neutron dpa rates for the materials were dominant in the fast neutron energy region, with Lu-Al exhibiting the highest dpa rates in that range. This suggests that the cross-section for elastic and inelastic neutron interaction in Au, and the cross-section for fast neutron capture in Aluminium and Lutetium, could be responsible for the damage accumulation in the fast energy regions [16, 32, 202, 203]. This influence of neutron cross-section on radiation damage at different energy regimes was highlighted in section 2.4

Using SRIM-2013, the dpa rates obtained for both materials with two ion species were within the same order of magnitude and comparable with results from other studies. A summary of previous studies on radiation damage with fission reactors and accelerator fields is provided by Meimei [55] in Table 5.1.

**Table 5.1:** Displacement Per Atom rates for neutron sources

| <b>Irradiation field</b>                                | <b>dpa rate<br/>(dpa.s<sup>-1</sup>)</b> |
|---------------------------------------------------------|------------------------------------------|
| Thermal fission ( Mixed spectrum reactor)[55, 134, 136] | $10^{-7}$                                |
| Fast fission reactor [55]                               | $10^{-6}$                                |
| Accelerator [55]                                        | $10^{-3}$                                |
| Current work (Fission reactor) [130]                    | $10^{-7}$                                |
| Current work (Accelerator)                              | $10^{-5} - 10^{-3}$                      |

The dpa rates for Au and Lu-Al fell within the specified ranges in the table. However, the Lu-Al dpa rates for neutron irradiation were two orders of magnitude

higher than those of Au. Also, for ion beam irradiation, the dpa rates for Au<sup>+</sup> ions for both materials were two orders of magnitude higher than those of H<sup>+</sup> ions.

These results highlight the impact of differences in the mechanism of interaction for the radiation species. In neutron irradiation, more damage was observed for Lu-Al in the fast neutron range, while the Au material exhibited less damage in that energy region.

In general, fast neutrons induce more damage than thermal neutrons, primarily due to the higher absorption cross-section of fast neutrons [10]. Thus, materials with elevated neutron absorption cross-sections in the fast region experience greater neutron damage. Consequently, neutron absorption cross-section plays a crucial role in radiation damage caused by neutrons. Recoil spectra generated by neutrons vary across different neutron spectra and materials [8]. Conversely, in ion beam irradiation, heavier ions cause more atomic displacement with shorter ranges into the materials compared to lighter ions [193], leading to higher damage rates. This, of course, depends on the energy of the ion beam. Therefore, Au<sup>+</sup> ions resulted in higher damage rates than H<sup>+</sup> ions. The simulation using H<sup>+</sup> beams at lower proton energies yielded approximate dpa rates and produced similar irradiation profiles as neutrons, prompting the use of protons as a surrogate for neutron irradiation in the experiments.

One of the objectives was to predict the resilience of Au and Lu-Al to damage based on the obtained dpas. Au material demonstrated relative resistance to radiation damage compared to Al and its alloys, attributed to its robust and densely packed metallic bonding in the lattice structure, facilitating the self-annihilation of defects. Experimental observations indicated the formation of vacancy clusters and Single Frankel Type (SFT) defects in Au when irradiated with 0.4 MeV Au-ions at a flux of  $1.0 \times 10^{13}$  ions.m<sup>-2</sup>.s<sup>-1</sup> [46].

A quick cascade mode calculation of Au dpa rates using Sekimura's self-ion irradiation parameters in SRIM-2013 resulted in a dpa rate of  $4.4 \times 10^{-7}$  dpa.s<sup>-1</sup>

when employing the NRT-dpa model. Notably, this study observed defects in the microstructure of Au with Au<sup>+</sup> that were akin to those observed in a 14 MeV neutron radiation damage study on Au thin foils to a fluence of about  $1.2 \times 10^{20} \text{ n.m}^{-2}$  [16]. These dpa rates and neutron fluence fall within the range of damage parameters recorded in this work.

For Lu-Al, there are no prior experimental or simulation works reporting dpa rates for comparison. However, previous studies on Al typically report dpa rates within the range of  $10^{-7}$  to  $10^{-4} \text{ dpa.s}^{-1}$ , observing defects such as point defects, dislocation loops, voids, and swelling in Al material [21, 134–136]. Considering the reduced homologous temperature with the presence of Lutetium in the alloy, the Lu-Al microstructure is expected to be more susceptible to damage. The calculated dpa rates for Lu-Al in this present work are approximately  $10^{-5} \text{ dpa.s}^{-1}$ , indicating an expectation of similar defects. Further discussion on the actual observed defects is presented in the subsequent section.

### 5.1.2 Heat deposition

In the reactor setting, unlike accelerators where heat deposition can be readily controlled, the heat generated through collisions of neutrons and photons with atomic nuclei of materials is significant for reactor studies. We investigated the heat deposited in materials resulting from neutrons and photons. More than 80% (see Figure 4.7) of the effective heat deposition in Lu-Al was attributed to neutron interactions. Conversely, in the Au material, heating due to neutrons was negligible.

Photon-induced heating played a predominant role in the total heating of the Au material in the fast neutron energy region. However, in the Lu-Al sample, heating was more pronounced within the thermal neutron energy region. Photon-induced heating of approximately  $0.5 \text{ W.g}^{-1}$  was observed for both materials in the fast neutron region, likely influenced by neutron capture reactions and the high energies of fast neutrons. In general, the heat deposition resulting from neutrons and photons is not significant enough to contribute to excessive thermal heating of the materials or an additional impact on the reactor’s effective power.

The results of the simulation of radiation damage with neutrons in the material matrices provided an opportunity to explore the behaviour of the intrinsic properties of these materials with respect to the neutron interaction cross-section. It is worth noting that higher dpa were estimated in the fast neutron regime in both materials. Also, neutron heating was produced more with the thermal neutron, while photon heating occurred mostly at the fast neutron energies for both materials. A careful selection of the ion energy and fluence or beam current resulted in dpa values in proton irradiation that were comparable to those of the neutron-irradiated material, with proton irradiation times roughly three orders of magnitude lower than neutron irradiation, just as reported by previous studies [24, 41, 42].

## 5.2 Radiation damage observed and implication

Three techniques were employed to characterize damage in FCC Au and DC Lu-Al materials. This part of the study explored the third to the sixth objectives. Figure 4.29 (b)-(c) illustrates the observed defects in Au samples: Stacking Faults Tetrahedra (SFT), dislocation loops, and dislocation lines, identified through HRTEM based on the zone axis. Previous studies that observed different types of defects on Au matrix are summarized in Table 5.2.

**Table 5.2:** Comparison of proton irradiation Au defects and dislocation densities with previous studies

| Material      | Source                            | dpa/fluence                                          | Defects                   | $\delta$                                             | Study                    |
|---------------|-----------------------------------|------------------------------------------------------|---------------------------|------------------------------------------------------|--------------------------|
| Au            | Ar <sup>+</sup> , Xe <sup>+</sup> | 0.063 - 1.3<br>$\times 10^{16}$ ions.m <sup>-2</sup> | SFT,<br>dislocation loops | -                                                    | Seki. et al. 1988 [13]   |
| Au            | Au <sup>+</sup>                   | 1<br>$\times 10^{14}$ ions.m <sup>-2</sup>           | SFT                       | -                                                    | Seki. et al. 1994[46]    |
| Au            | Au <sup>+</sup>                   | 0.05-1<br>$\times 10^{15}$ ions.m <sup>-2</sup>      | Vacancy clusters          | -                                                    | Seki. et al.1999 [14]    |
| Nanoporous Au | Au <sup>+</sup>                   | 0.74 dpa                                             | SFT,<br>ligament loops    | -                                                    | Briot. et al. 2019 [131] |
| Au            | neutron                           | 3<br>$\times 10^{18}$ n.m <sup>-2</sup>              | SFT,<br>dislocation loops | $\sim 2-5$<br>$\times 10^{19}$                       | Cotter. et al.1966 [129] |
| Au            | neutron                           | 1<br>$\times 10^{22}$ n.m <sup>-2</sup>              | SFT,<br>dislocation loops | $\sim 1.3$<br>$\times 10^{23}$ m <sup>-3</sup>       | Kirit. et al.1985 [15]   |
| Au            | neutron                           | 4.7<br>$\times 10^{22}$ n.m <sup>-2</sup>            | SFT,<br>dislocation loop  | $\sim 8.2$<br>$\times 10^{22}$ m <sup>-3</sup>       | Horse. et al. 1991 [16]  |
| Au            | proton                            | 10<br>$\times 10^{22}$ p.m <sup>-2</sup>             | SFT,<br>dislocation loop  | $\sim 5.4$<br>$\times 10^{23}$ m <sup>-3</sup>       | Horse. et al. 1991 [16]  |
| Au            | proton                            | 0.77-4.64<br>$\times 10^{21}$ p.m <sup>-2</sup>      | SFT<br>dislocations       | $\sim 0.43-3.61$<br>$\times 10^{14}$ m <sup>-2</sup> | Current work             |

From Table 5.2, it can be observed that almost all the previous studies on Au matrix reported SFTs and dislocation loops for heavy ion, neutron and proton irradiation fluences within  $\sim 10^{14}$  - $10^{22}$  particle/m<sup>2</sup> [13, 15, 16, 46, 129, 131]. Findings from the current study align with previous studies that reported the presence of SFT and dislocations loops/lines on the irradiated microstructure within the dpa range investigated.

The dislocations observed in this study were primarily mixed dislocation lines. Mixed dislocations are usually intermediate between screw and edge dislocations. Dislocations generally offer insights into crystallographic defects or irregular crystal structures. Studies on dislocations have reported the formation of neither screw nor edge dislocations from the interaction of SFT with either of the dislocations in FCC materials [204–206].

In Lu-Al samples (see Figure 4.31 (b)-(c)), dislocation loops and dislocation lines were observed. While dislocations may be inherent in materials due to the preparation process, they are often challenging to observe in most unirradiated materials since their size is usually small. XRD data established a pre-irradiation baseline for dislocations, crucial for assessing radiation impact. Unlike previous studies on aluminum alloys, this study did not reveal three-dimensional (3D) defects; only dislocation loops and lines were observed in Lu-Al. The formation of 3D defects such as voids/bubbles, cavities, and swelling is usually associated with long-term exposure (higher doses) of materials to radiation [5, 8–10].

The proton irradiation-induced defects in the Lu-Al matrix were compared to previous studies summarized in Table 5.3. From Table 5.3, defects observed in both neutron and proton irradiations were in agreement, predominantly, with dislocations present in almost all studies just as with the current study. Precipitates and Radiation-Induced Segregation (RIS) were also observed with both proton and neutron irradiation studies previously conducted [19, 132, 137, 138]. but these were not observed in the present study. However, the dpa/fluence in those studies that produced precipitates and RIS were at least 2-3 orders of magnitude higher than the

doses utilized in this present study. A phase transition was equally observed where heavy ion irradiation [139] with a much higher dose, with 4 orders of magnitude higher than the dose in this present study.

**Table 5.3:** Comparison of proton irradiation Lu-Al defects and dislocation densities with other studies

| Material      | Source     | dpa/fluence                           | Defects                                     | $\delta$ ( $\text{m}^{-2}$ )     | Study                   |
|---------------|------------|---------------------------------------|---------------------------------------------|----------------------------------|-------------------------|
| 6061 Al-Alloy | neutron    | 1.6 - 4.2 dpa                         | voids, dislocation loops, precipitates, RIS | $\sim 0.49 - 1.6 \times 10^{15}$ | Yan et al. 2024 [19]    |
| Al-Mg         | neutron    | -                                     | dislocations                                | $\sim 1.5-4.5 \times 10^{15}$    | Thira. et al. 2022[133] |
| Fe-Cr-Al      | neutron    | 1.8 dpa                               | dislocations, precipitates                  | $\sim 0.6-1.5 \times 10^{14}$    | Field et al.2015 [132]  |
| Fe-Cr-Al      | proton     | 0.01-0.1 dpa                          | dislocations, precipitates                  | $\sim 4.93-9.45 \times 10^{14}$  | Wei et al.2022 [138]    |
| Al and Al5083 | proton     | 0.1 -5.0 $\times 10^{21}$ p.m $^{-2}$ | point defects, RIS, dislocations            | $\sim 1.77 \times 10^{22}$       | Bhatt. et al.2017 [137] |
| Al 6061       | Ar, Cu, Au | 0.2-23 dpa                            | phase formation, vacancy clusters           | -                                | Izer. et al.2023 [139]  |
| Lu-Al         | proton     | 1.1-6.8 mdpa                          | dislocation                                 | $\sim 0.33-4.76 \times 10^{14}$  | Current work            |

Radiation induced voids were formed with neutron irradiation [19], this is sometimes typical 3D defects observed with neutron and proton irradiated Al-alloys. With proton irradiation relatively high energies and long-term exposures of the alloy to radiation are required to cause aggregation of vacancies during atomic displacements [5, 10].

It is worthy of note that some of the multiple types of microstructural defects such as Stacking Faults Tetrahedra (SFTs) and dislocations formed with proton-irradiation in Au and Lu-Al in this study matched neutron-irradiation microstructure of similar materials or series observed in previous studies (see Table 5.2 and 5.3) as hypothesized in section 1.3.2). However, this study did not observe multiple defects of 3D type (voids, precipitates and other defect clusters) with the proton irradiation. The inability to observe these defects could have been due to the low irradiation doses used in the study or some other inherent factors like material composition, especially for Lu-Al.

Quantitative measurements from XRD results (see Figure 4.20 and 4.24) included crystallite size ( $D$ ), microstrain ( $\varepsilon$ ), and dislocation densities ( $\delta$ ) for unirradiated and post-irradiation conditions. The crystallite size decreased with increasing

strain, resulting in local strain and increased dislocation density. Similar trends have been reported in other studies on FCC crystals [63, 115, 147, 197].

TEM micrographs (see Figure 4.33 and 4.34) highlighted changes in defects, indicating the effect of radiation on the microstructure of the materials. Defects evolved from point to 1 or 2D defects, such as dislocation lines and 3D defects SFTs in Au microstructure. Surface plots based on grayscale values illustrated damage levels to the microstructure of the materials but specific defects could not be isolated due to potential low-intensity values from any defective portion. These surface profiles facilitated the comparison of surface damage before and after irradiation at different dpa values. The shape index maps aided the counting of defects on the surface of the micrographs by giving an impression of depth and solidity to the 2D micrograph.

This study quantitatively measured the influence of proton irradiation dpa on microstructural changes in irradiated materials, aligning with findings in studies on proton and neutron irradiation on metals and alloys [5, 6, 14, 32, 41, 126, 207, 208].

The average dislocation density was the only quantitative damage parameter calculated using XRD and TEM to address the second research question in section 1.3.1. Results (see Figure 4.37) exhibited a correlation with radiation dose, but significant disparities were noted in TEM and XRD values. In some cases, TEM results were up to three times higher than XRD values (see Figure 4.37). As mentioned in the results summary, the two techniques employ different approaches (see Figure 4.38) in calculating  $\delta$  and have associated uncertainties, making direct comparison uncertain [147]. Higher uncertainties were recorded in  $\delta$  measurements using TEM direct counting compared to XRD, particularly at values  $\geq 10^{14} \text{ m}^{-2}$ . XRD measurements were favored for higher  $\delta$  values in this study [63, 147].

From Table 5.2, dislocation densities obtained in previously studied Au samples were between  $\sim 10^{12} - 10^{14} \text{ m}^{-2}$  [15, 16, 129] considering sample thickness in nanometers. This is within the range of dislocation densities (  $0.43 - 3.61 \times 10^{14} \text{ m}^{-2}$  ) obtained for Au matrix in this current work. Studies have suggested that neutron irradiation-

induced dislocation densities are generally higher than proton-induced dislocation densities in approximate fluences[103]. However, no clear relationship between the irradiation species can be established based on the studies at reach, reported in Table 5.2. Some of the factors responsible could be the lack of high sensitivity of the equipment and methods of data analyses as of the time these experiments were performed, more recent research with state-of-the-art facilities is needed to establish this.

Again from Table 5.3, the dislocation densities obtained for neutron-irradiated Al alloys were an order of magnitude higher than the densities ( $0.33 - 4.76 \times 10^{14} \text{ m}^{-2}$ ) obtained in this present study. However, these dislocation density values are highly dependent on the alloy composition. Even when the composition is similar, the material's preparation method can significantly affect the dislocation number densities due to different types of impurities found in the samples. Bohanon et al. [103] noted that neutron-irradiated materials generally tend to have higher dislocation densities than proton-induced radiation with comparable fluences. This could also be a factor to be considered. The dislocation density obtained when Al and Al5083 matrices [137] were considerably high probably due to the high proton fluence used. Generally, the dislocation densities obtained in most of the previous aluminium alloys irradiation with protons were of the same order of magnitude as the present study.

In this study, several challenges were encountered. One notable challenge was the inability to directly correlate the observed damage with the simulation results. While the SRIM simulation provided valuable information about the extent of damage and the required energy and doses for sample irradiation, comparing defects produced necessitates atomistic modeling. This would involve developing interatomic potentials, especially for Lu-Al, where such potentials are currently non-existent. Subsequently, atomistic simulations using Molecular Dynamics would be conducted to quantify the level of damage in the materials, enabling a direct comparison of defect densities with the characterization methods.

Another challenge lies in the limitations of the available characterization methods, particularly TEM, which has been found to underestimate defect densities in irradiated materials [148]. The application of flash Differential Scanning Calorimetry (DSC), a nanocalorimetry technique capable of quantifying defects by measuring the excess energy released during defect annealing, is reported to offer five times greater sensitivity in defect density quantification than the techniques employed in this work [148]. Additionally, certain properties of the Lu-Al alloy relevant to its radiation damage response, such as phase transitions and precipitate stability, remain unclear. A comprehensive understanding of these properties would contribute to a better assessment of the damage resilience of this binary alloy.

Based on the above we employed the Flash DSC1 to study damage on Lu-Al alloy. Preliminary observations were conducted using Flash DSC 1 on the Lu-Al alloy, revealing noteworthy features in the irradiated samples. In Figure 4.40, a spike at approximately 160°C was observed, possibly indicative of the Guinier-Preston phase transition zone, previously reported in other Al-alloys [209, 210]. Furthermore, other phenomena were noted in the sample irradiated to 3.4 mdpa in Figure 4.41, where a repeated spike occurred in all curves around 250°C, accompanied by an opposing curvature spanning temperatures from about 240°C to 420°C. This suggests potential radiation-induced processes, such as phase transitions and precipitate stability, in these temperature zones.

The significant changes in the characteristics of the Power-Temperature curves (Figures 4.39, 4.40, and 4.41) illustrate the influence of proton irradiation on the material a similar trend that was observed in XRD and TEM techniques. Comprehensive insight into the material's properties requires repeated experiments and detailed analysis.

## 5.3 Summary

This thesis explored numerical approaches to the experimental observation of radiation damage caused by protons and neutrons. The displacement per atom rates

calculated for neutrons using MCNP were achieved by proton ions in SRIM, particularly at lower proton energies (0.5 MeV). The observed proton irradiation effects on the microstructures of both Au and Lu-Al are consistent with the defects reported in other studies involving neutron irradiation. The level of damage observed ranged from 0 to 2-dimensional defects, and 3-dimensional defects in the case of Au microstructure.

This study demonstrated changes in lattice spacing in both FCC and DC materials under XRD. Intensity growth in certain preferred orientations was observed, such as the favoured (002) reflection in the Au microstructure (see Figure 4.15) and the slight increase in the (224) and (335) crystal reflections in the Lu-Al microstructure (see Figure 4.16) after each irradiation dose. The heightened intensity led to shorter inter-atomic distances in the Au FCC and Lu-Al DC crystals, consistent with findings in cubic structures reported by [115, 194], among others.

A progressive increase in dislocation density was noted as the radiation dose increased in both materials. However, in the Lu-Al DC crystal structure, the dislocation density seemed to approach saturation levels, as depicted in Figure 4.24. This argument, though, cannot be conclusively established without conducting higher proton doses to further examine this material's response to radiation damage.

The radiation damage process involved the production of vacancies and interstitials (Frenkel pairs), their clustering, evolution into dislocation loops, and subsequent accumulation and migration into line and surface defects, as observed in Figures 4.29 and 4.31. This quantitatively translated to an increase in local strain fields on the crystallites, consequently leading to an increase in the dislocation density of the samples due to the escalating radiation stress field dose.

# Chapter 6

## Conclusion and Recommendations

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### 6.1 Conclusion

This thesis demonstrates the vigor of proton irradiation in radiation damage studies as a viable alternative to neutron irradiation when carefully applied. Proton irradiation has been conducted on Au and Lu-Al binary alloy at fluences in the range  $0.77 - 4.64 \times 10^{21} \text{ p.m}^{-2}$  and stable temperature  $\sim 40^\circ\text{C}$ . Post-irradiation examination (PIE) including XRD, HRTEM, and F-DSC characterization was performed on the samples to investigate microstructural damage in the materials. Protons of energy 0.5 MeV at different irradiation fluences, induced similar microstructural defects in the materials such as those observed from previous studies that utilized neutron irradiation. Calculations conducted with MCNP6.2 and SRIM-2013 demonstrated that comparable displacement per atom rates could be achieved in significantly shorter times in the materials at lower proton energies (0.2 to 1 MeV).

Damage characterization techniques used were able to observe defect types and quantify the level of damage using the microstructural intrinsic property, dislocation density ( $\delta$ ). With HRTEM, multiple proton irradiation-induced defects such as dislocation loops, Stacking Faults Tetrahedra (SFT), and dislocation lines were observed in the Au microstructure consistent with other studies. Also, dislocation loops and dislocation lines were observed in the Lu-Al microstructure. The observed microstructural defects on the Au matrix conform with results obtained in several previous studies. Similarly, the microstructural defects on the Lu-Al matrix were comparable to those observed in other Al-6000 and 7000 series for both proton/ other ions and neutron-irradiated microstructure. Observation of defects was enhanced using the shape map index in this current study (Figures 4.30 and

4.32). It is worthy of note that three-dimensional defects such as voids/bubbles and precipitates were not observed as reported by other studies, possibly due to the low-level radiation doses and short-term radiation exposure times in this study.

The average dislocation densities obtained in this study ranged from  $0.43 - 3.61 \times 10^{14} \text{ m}^{-2}$  for Au microstructure, and from  $0.33 - 4.76 \times 10^{14} \text{ m}^{-2}$  for Lu-Al microstructure. These values were found to increase with rising dpa values or proton fluence and they suggest an elevated level of damage to the microstructure. Sustained exposure to radiation over time could lead to significant changes in the mechanical properties of the materials. The dislocation densities obtained in this present study for both materials were in the range of values obtained in other studies with approximate fluences. However, no special trend was established especially for the Au matrix and most of the studies within reach were conducted more than 2 decades ago before this nanotechnology age where there has been considerable improvement in the sensitivity of instruments and robust data analysis techniques.

Lu-Al exhibited lower ( $\delta$ ) with the unirradiated sample, but higher densities were observed post-irradiation. This suggests that Lu-Al is less resilient to radiation damage than Au as observed in the damage predictions. Similarly, radiation-induced defects observed on the microstructure of the two materials evolved from 0-dimensional to 1 or 2-dimensional defects.

Notable alterations in the characteristics of the power-temperature melting curves were evident, particularly in the sample subjected to the highest dose, suggesting potential phase transition and precipitate processes. These properties are pertinent to understanding the material's resilience to radiation damage. However, a thorough investigation is imperative, and the researcher anticipates delving into this aspect in the subsequent phase of research.

The defect density values are relatively high, and sustained exposure to higher radiation doses could induce more changes in the microstructural properties of the Au and Lu-Al materials when exposed to harsh radiation environments. Further-

more, radiation damage predicted and observed in this study may be used as a reference for inferring possible damage over long-term exposures to components built with these materials to radiations of comparable exposure levels. Dislocation densities and dpa rates calculated in this study will be an addition to the database of radiation damage studies for the Au and Lu-Al materials.

## 6.2 Recommendations

Based on the findings of this study, there is wisdom in using atomistic modeling of radiation damage to observed defects and their density to correlate with the observed damage. However, this will entail developing interatomic potentials, especially for Lu-Al using DFT-Machine learning algorithms before simulating the radiation damage; Since there are no available inter-atomic potentials for this binary alloy. This in itself is an enormous task that the research should embark on subsequently.

Observation of defect densities using the two characterization techniques used in this study is underlined by some limitations. research has shown that TEM for instance measures less the defect density present in irradiation materials. However, there is a need to leverage the technique of measuring defect densities through the excess energy released during defect annealing in the nanocalorimetry technique called the flash DSC which was partly explored in this study. This technique promises quantification of higher defect densities below the detectable limits of the HRTEM technique. Moreover, intrinsic properties such as phase transition and precipitate stability in Lu-Al binary alloy are yet unresolved. The researcher also desires in the next phase of research to use the Flash DSC technique in conjunction with the atomistic modeling of radiation damage to estimate radiation damage and these unexplored intrinsic properties that relate to damage in Lu-Al alloy.

# Bibliography

- [1] M. P. Short and S. Yip. Materials aging at the mesoscale: Kinetics of thermal, stress, radiation activations. *Current Opinion in Solid State and Materials Science*, 19(4):245–252, 2015.
- [2] M.P. Short. 27. nuclear materials — radiation damage and effects in matter, 2019. <https://www.youtube.com/watch?v=CjZjVUWMEz0>.
- [3] G. H. Kinchin and R. S. Pease. The displacement of atoms in solids by radiation. *Reports on Progress in Physics*, 18(1):1–51. Publisher: IOP Publishing.
- [4] J. Li, Y. Yang, and M. P. Short. More efficient and accurate simulations of primary radiation damage in materials with nanosized microstructural features or ion beams. In W. Andreoni and S. Yip, editors, *Handbook of Materials Modeling*, pages 1–33. Springer International Publishing, 2019. [http://link.springer.com/10.1007/978-3-319-50257-1\\_115-2](http://link.springer.com/10.1007/978-3-319-50257-1_115-2).
- [5] M. P. Short, D. R. Gaston, M. Jin, L. Shao, and F. A. Garner. Modeling injected interstitial effects on void swelling in self-ion irradiation experiments. *Journal of Nuclear Materials*, 471:200–207, 2016.
- [6] K. Nordlund, S. J. Zinkle, A. E. Sand, F. Granberg, R. S. Averback, R. Stoller, T. Suzudo, L. Malerba, F. Banhart, W. J. Weber, F. Willaime, S. L. Dudarev, and D. Simeone. Improving atomic displacement and replacement calculations with physically realistic damage models. *Nature Communications*, 9(1):1084, 2018.
- [7] I. P. Hernandez. *Simulation of atom displacements induced by photons and electrons in solids*. PhD thesis, Universiteit Antwerpen, Belgium, 2014. <https://hdl.handle.net/10067/1166870151162165141>.
- [8] Kai Nordlund, Andrea E. Sand, Fredric Granberg, and et. al. Primary radiation damage in materials, 2015. [https://www.oecd-nea.org/jcms/pl\\_19620](https://www.oecd-nea.org/jcms/pl_19620).

- [9] K. L. Murty and I. Charit. *An Introduction to Nuclear Materials Fundamentals and Applications*. Wiley-VCH, Verlag GmbH & Co. KGaA, 2013.
- [10] G. S. Was. *Fundamentals of Radiation Material Science. Metals and Alloys*. Springer, second edition edition, 2017.
- [11] G. Wittstock, M. Baumer, W. Dononelli, T. Kluner, L. Luhrs, C. Mahr, M. Oezaslan L.V. Moskaleva, T. Risse, A. Rosenauer, A. Staubitz, J. Weissjeemuller, and A. Wittstock. Nanoporous gold: From structure evolution to functional properties in catalysis and electrochemistry. *Chem. Rev.*, 123:6716–6792, 2023.
- [12] G. Steinhauser, S. Merz, F. Stadlbauer, P. Kregsamer, C. Streli, and M. Villa. Performance and comparison of gold-based neutron flux monitors. *Gold Bull*, 45(1):17–22, 2012.
- [13] N. Sekimura and S. Ishino. Stability of cascade clusters in gold under energetic heavy ion irradiation. *Journal of Nuclear Materials*, 155–157:1217–1221, 1988.
- [14] N. Sekimura, Y. Kanzaki, N. Ohtake, J. Saeki, Y. Shirao, S. Ishino, T. Iwata, A. Iwase, and R. Tanaka. High energy cascades in gold as studied by high energy self-ion irradiation. *Journal of Nuclear Materials*, 271–272:68–72, 1999.
- [15] M. Kiritani, Y. Shimomura, N. Yoshida, K. Kitagawa, and T. Yoshiie. Development of defect structures from displacement cascade damage in d-t neutron irradiated gold. *Journal of Nuclear Materials*, 133–134:410–414, 1985.
- [16] A. Horsewell, B. N. Singh, S. Proennecke, W. F. Sommer, and H. L. Heinisch. Defect structures in copper and gold irradiated with fast neutrons, 14 MeV neutrons and 600–800 MeV protons. *Journal of Nuclear Materials*, 179–181:924–927, 1991.
- [17] Y. Chimi, A. Iwase, N. Ishikawa, M. Kobiyama, T. Inami, and S. Okuda. Accumulation and recovery of defects in ion-irradiated nanocrystalline gold. *Journal of Nuclear Materials*, 297(3):355–357, 2001.

- [18] C. G. Zhang, Y. G. Li, W. H. Zhou, L. Hu, and Z. Zeng. Anti-radiation mechanisms in nanoporous gold studied via molecular dynamics simulations. *Journal of Nuclear Materials*, 466:328–333, 2015.
- [19] Zhanfeng Yan, Wei Zhou, Jian Zheng, Guanbo Wang, Xiankun Liu, Xiao Liu, Shuyu Wang, Chan Gao, Jiting Tian, Hao Wang, Wankui Yang, and Qijie Feng. Investigations of microstructure and tensile properties of neutron irradiated 6061 aluminum alloys. *Materials Science and Engineering: A*, 890:145913, 2024.
- [20] K. Farrell. 5.07 - performance of aluminum in research reactors. In Rudy J.M. Konings, editor, *Comprehensive Nuclear Materials*, pages 143–175. Elsevier, Oxford, 2012.
- [21] M. A. Amirkhani and F. Khoshahval. Evaluation of the radiation damage effect on mechanical properties in tehran research reactor (trr) clad. *Nucl. Eng. Technol.*, 52:2975–2981, 2020.
- [22] D. Yuan, L. Tan, K. Chen, S. Chen, Peng Xie, and H. Jiao. Comparison of strength, stress corrosion cracking and microstructure of new generation 7000 series aluminium alloys. *Materials Science and Technology*, 37(6), 2021.
- [23] IAEA. Nuclear power for sustainable development, 2017.
- [24] S. Taller, G. VanCoevinger, B.D. Wirth, and Gary S. Was. Predicting structural material degradation in advanced nuclear reactors with ion irradiation. *Sci Rep*, 11:2949, 2021.
- [25] K. Nordlund, S. J. Zinkle, A. E. Sand, F. Granberg, R. S. Averback, R. E. Stoller, T. Suzudo, L. Malerba, F. Banhart, W. J. Weber, F. Willaime, S. L. Dudarev, and D. Simeone. Primary radiation damage: A review of current understanding and models. *Journal of Nuclear Materials*, 512:450–479, 2018.
- [26] H. Dawson. *Friction Stir Welding of ODS Steels for Future Generation Nuclear Reactors*. PhD thesis, School of Materials, Faculty of Science and Engineering, University of Manchester, UK, 2017. [https://pure.manchester.ac.uk/ws/portalfiles/portal/66045501/FULL\\_TEXT.PDF](https://pure.manchester.ac.uk/ws/portalfiles/portal/66045501/FULL_TEXT.PDF).

- [27] S. Roberts and M. Coates. Fracture of Ferritic Steels; the effects of the Ductile Brittle Transition on Carbides within the Plastic Zone. Hawaii, 2001. ResearchGate. [https://www.researchgate.net/publication/262875587\\_Fracture\\_of\\_Ferritic\\_Steels\\_the\\_effects\\_of\\_the\\_Ductile\\_Brittle\\_Transition\\_on\\_Carbides\\_within\\_the\\_Plastic\\_Zone](https://www.researchgate.net/publication/262875587_Fracture_of_Ferritic_Steels_the_effects_of_the_Ductile_Brittle_Transition_on_Carbides_within_the_Plastic_Zone), eventtitle = 10th International Conference on Fracture.
- [28] C. Kittel. *Introduction to Solid State Physics*. John Wiley and Sons, third edition edition, 1968.
- [29] P. Partyka, Y. Zhong, K. Nordlund, R. S. Averback, I. M. Robinson, and P. Ehrhart. Grazing incidence diffuse x-ray scattering investigation of the properties of irradiation-induced point defects in silicon. *Phys. Rev. B*, 64(23):235207, 2001. Publisher: American Physical Society.
- [30] S.J. Zinkle and J. Steven. Radiation-induced effects on microstructure. In *Comprehensive Nuclear Materials*, pages 65–98. Elsevier Oxford, 2012.
- [31] Y. Yang, Y. G. Li, M. P. Short, C. Kim, K. K. Berggren, and J. Li. Nano-beam and nano-target effects in ion radiation. *Nanoscale*, 10(4):1598–1606, 2018. Publisher: The Royal Society of Chemistry.
- [32] S. A. Briggs and K. Hattar. Evolution of gold nanoparticles in radiation environments. *Gold Nanoparticles - Reaching New Heights*, pages 1–19, 2018. Publisher: IntechOpen.
- [33] J. Chadwick. Possible existence of a neutron. *Nature*, 129:312, 1932.
- [34] V. Nesvizhevsky and J. Villain. The discovery of the neutron and its consequences (1930–1940). *Comptes Rendus Physique*, 18(9):592–600, 2017.
- [35] M. F. L’Annunziata. Radioactivity hall of fame-part II. In *Radioactivity*, pages 85–117. Elsevier Science B.V., Amstadam, 2007. {<https://www.sciencedirect.com/science/article/pii/B9780124366039500065>},.
- [36] D. Lockner. George de hevesy—80 years. *The International Journal of Applied Radiation and Isotopes*, 16(9):513–516, 1965.

- [37] L. Hamidatou, H. Slamene, T. Akhal, and B. Zouranen. Concepts, instrumentation and techniques of neutron activation analysis. *Imaging and Radioanalytical Techniques in Interdisciplinary Research - Fundamentals and Cutting Edge Applications*, 2013. Publisher: IntechOpen.
- [38] M.P. Short. Inside MIT’s nuclear reactor, 2018-04-18. <https://www.youtube.com/watch?v=5QcN3KDexcU>.
- [39] F. Mota, C. J. Ortiz, R. Vila, N. Casal, A. García, and A. Ibarra. Calculation of damage function of Al<sub>2</sub>O<sub>3</sub> in irradiation facilities for fusion reactor applications. *Journal of Nuclear Materials*, 442:S699–S704, 2013.
- [40] J. A. Mascitti and M. Madariaga. Method for the calculation of DPA in the reactor pressure vessel of atucha II. *Science and Technology of Nuclear Installations*, 2011:e534689, 2011. Publisher: Hindawi.
- [41] G. Lapinska. Effect of proton irradiation on the mechanical properties of fe-10cr-4al in liquid lead, 2023. [https://www.skf.kth.se/polopoly\\_fs/1.1288236.1697719737!/GabrielaLapinska.pdf](https://www.skf.kth.se/polopoly_fs/1.1288236.1697719737!/GabrielaLapinska.pdf).
- [42] G.S. Was. Challenges to the use of ion irradiation for emulating reactor irradiation. *Journal of Materials Research*, 30:1158–1182, 2015.
- [43] S.J. Zinkle and L.L. Snead. Opportunities and limitations for ion beams in radiation effects studies: Bridging critical gaps between charged particle and neutron irradiations. *Scripta Materialia*, 143:154–160, 2018.
- [44] L. Haocheng, G. Wei, and W. Yugang. Re-evaluating ion and neutron irradiation data with calculated applicable arc-dpa. *Radiation Effects and Defects in Solids*, 176(5-6):441–452, 2021.
- [45] G.S. Was, J.T. Busby, T. Allen, E.A. Kenik, A. Jensson, S.M. Bruemmer, J. Gan, A.D. Edwards, P.M. Scott, and P.L. Andreson. Emulation of neutron irradiation effects with protons: validation of principle. *Journal of Nuclear Materials*, 300(2):198–216, 2002.

- [46] N. Sekimura, Y. Kanzaki, S. R. Okada, T. Masuda, and S. Ishino. Cascade damage formation in gold under self-ion irradiation. *Journal of Nuclear Materials*, 212–215:160–163, 1994.
- [47] S. Agarwal, Y. Lin, C. Li, R. E. Stoller, and S.J Zinkle. On the use of SRIM for calculating vacancy production: Quick calculation and full-cascade options. *Nucl. Instrum. Methods Phys. Res. B*, 503:11–29, 2021.
- [48] P. Wang, J. Bowman, M. Bachhav, B. Kammenzind, R. Smith, J. Carter, A. Motta, E. Lacroix, and G.S. Was. Emulation of neutron damage with proton irradiation and its effects on microstructure and microchemistry of zircaloy-4. *Journal of Nuclear Materials*, 557:153281, 2021.
- [49] E.F. Sturcken. Irradiation effects in magnesium and aluminum alloys. *Journal of Nuclear Materials*, 82(1):39–53, 1979.
- [50] Y Katoh, H Tanigawa, T Muroga, T Iwai, and A Kohyama. The influence of helium co-implantation on ion-induced hardening of low activation ferritic steel evaluated by micro-indentation technique. *Journal of Nuclear Materials*, 271-272:115–119, 1999.
- [51] Stephen Taller and Gary S. Was. Understanding bubble and void nucleation in dual ion irradiated t91 steel using single parameter experiments. *Acta Materialia*, 198:47–60, 2020.
- [52] N. Daghbouj, N. Cherkashin, F.X. Darras, V. Paillard, M. Fnaiech, and A. Claverie. Effect of the order of he+ and h+ ion co-implantation on damage generation and thermal evolution of complexes, platelets, and blisters in silicon. *J. Appl. Phys.*, 119:135308, 2016.
- [53] Liu H. Gao Z. Su Y. Liu Q. Ge W. Luo F. Xia S. Cao L. Xue J. Wang Y. Wang C. Huang, J. Helium-hydrogen synergistic effects in structural materials under fusion neutron irradiation. *Frontiers in Materials*, 9:849115, 2022.
- [54] S. T. Temaugee, L. Bedhesi, R. D. Mavunda, S. H. Connell, G. C. Daniels, and I. T.Usman. Evaluation of neutron radiation damage on gold microstructure using mcnp. *Journal of Physics: Conference Series*, 2586(1):012122, sep 2023.

- [55] L. I. Meimei. Experience with moving from dpa to changes in material properties, 2010. <https://accelconf.web.cern.ch/HB2010/papers/weo1b05.pdf>.
- [56] S. Taller, V. Pauly, Z. Jiao, R. Hanbury, and G. S. Was. Solute segregation and precipitation across damage rates in dual-ion-irradiated t91 steel. *Journal of Nuclear Materials*, 563:153626, 2022.
- [57] L. I Meimei. Experience with moving from DPA to Changes in Material Properties. In *Proceedings of the 46th ICFA Advanced Beam Dynamics Workshop on High-Intensity and High-Brightness Hadron Beams*, pages 468–471. CERN, 2010. <https://accelconf.web.cern.ch/HB2010/papers/weo1b05.pdf>.
- [58] C. J. Werner, J. S. Bull, C. J. Solomon, F. B. Brown, G. W. McKinney, M. E. Rising, D. A. Dixon, R. L. Martz, H.G. Hughes, L. J. Cox, A. J. Zukaitis, J. C. Armstrong, Robert A. Forster, and L. Casswell. MCNP Version 6.2 Release Notes, 2018. <https://www.osti.gov/biblio/1419730>.
- [59] J. F. Ziegler, J. P. Biersack, and M. D. Ziegler. *SRIM The Stopping and Range of Ions in Matter*. SRIM Co., 2008.
- [60] H. Jiang, S. Li, L. Zhang, J. He, and J. Zhao. Effect of microgravity on the solidification of aluminum–bismuth–tin immiscible alloys. *npj Microgravity*, 5, 2019.
- [61] S. Akamatsu, S.Bottin-Rousseau, V.T. Witusiewicz, U. Hecht, J. Mogeritsch A. Ludwig, M. Serefoglu, N. Bergeon, F.L. Mota, L. Sturz, G. Zimmermann, S. McFadden, and W. Sillekens. Microgravity studies of solidification patterns in model transparent alloys onboard the international space station. *npj Microgravity*, 9, 2023.
- [62] S. Tachikawa, H. Nagano, A. Ohnishi, and Y. Nagasaka. Advanced passive thermal control materials and devices for spacecraft: A review. *Int J Thermophys*, 43, 2022.
- [63] J. Gallet, M. Perez, R. Guillou, C. Ernould, C. Le Bourlot, C. Langlois, B. Beausir, E. Bouzy, T. Chaise, and S. Cazottes. Experimental measure-

- ment of dislocation density in metallic materials: A quantitative comparison between measurements techniques (xrd, r-ecci, hr-ebsd, tem). *Materials Characterization*, 199:112842, 2023.
- [64] S.J. Zinkle and G.S. Was. Materials challenges in nuclear energy. *Acta Materialia*, 61(3):735–758, 2013.
- [65] Steven J. Zinkle. 1.04 - radiation-induced effects on microstructure. In Rudy J. M. Konings and Roger E. Stoller, editors, *Comprehensive Nuclear Materials (Second Edition)*, pages 91–129. Elsevier, 2020. <https://www.sciencedirect.com/science/article/pii/B9780128035818120752>.
- [66] J.W. Poston. Neutrons, 2003. <https://www.sciencedirect.com/referencework/9780122274107/encyclopedia-of-physical-science-and-technology>.
- [67] A. Taylor, M. Dunne, S. Bennington, S. Ansell, I. Gardner, P. Norreys, T. Broome, D. Findlay, and R. Nelves. A route to the brightest possible neutron source? *Science*, 315(5815):1092–1095, 2007. Publisher: American Association for the Advancement of Science.
- [68] N. J. Carron. *An Introduction to the Passage of Energetic Particles through Matter*. Taylor & Francis, 2007.
- [69] N. Eby. Neutron energy relative to neutron flux, 2007. Publisher: SERC.
- [70] M. F. L’annunziata. 1 - nuclear radiation, its interaction with matter and radioisotope decay. In M. F. L’Annunziata, editor, *Handbook of Radioactivity Analysis (Second Edition)*, pages 1–121. Academic Press, San Diego, 2003. <https://www.sciencedirect.com/science/article/pii/B9780124366039500065>,.
- [71] J. R. Lamarsh and A. J. Baratta. *Introduction to Nuclear Engineering*. Prentice Hall, third edition edition, 2001.
- [72] J.K. Shultis and R.E. Faw. *Fundamentals of Nuclear Science and Engineering*. Taylor & Francis, third edition, 2017.

- [73] J. Koutsy and J. Kocik. *Radiation Damage of Structural Materials*. 79. Elsevier Science Publishers, 1994.
- [74] Schlumberger. Elastic\_neutron\_scattering, 2022. = [https://glossary.oilfield.slb.com/en/terms/e/elastic\\_neutron\\_scattering](https://glossary.oilfield.slb.com/en/terms/e/elastic_neutron_scattering).
- [75] C. Qi, Y. Wang, X. Bai, X. Jin, R. Li, W. Chen, X. Guo, J. Li, and C. Wang. Role of elastic scattering in low-energy neutron-induced seus in a 40-nm bulk sram. *IEEE Transactions on Nuclear Science*, 69(5):1057–1065, 2022.
- [76] W. M. Stacey. *Nuclear Reactor Physics*. Wiley-VCH, Verlag GmbH & Co. KGaA, second edition edition, 2007.
- [77] Schlumberger. Inelastic\_neutron\_scattering, 2022. [https://glossary.oilfield.slb.com/en/terms/i/inelastic\\_neutron\\_scattering](https://glossary.oilfield.slb.com/en/terms/i/inelastic_neutron_scattering).
- [78] K. E. Holbert. Radiation effects and damage 1. ionizing radiations. 2007. <https://api.semanticscholar.org/CorpusID:50449250>.
- [79] FTC. All about neutron activation analysis (naa): What it is and how it works, 2021. <https://frontier-cf252.com/blog/neutron-activation-analysis/>.
- [80] A. Mohammadi, S. Hamidi, and M. A. Asadabad. The use of the SRIM code for calculation of radiation damage induced by neutrons. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 412:19–27, 2017.
- [81] D. Dawson, R. Fleck, A. Wadham, and P. Bird. Radiation damage to materials, 1993. <https://canteach.candu.org/Content%20Library/20040904.pdf>.
- [82] M. Yigit. Investigation of (n,2n) reaction cross sections on pd and cd isotopes using equilibrium and pre-equilibrium models. *Applied Radiation and Isotopes*, 166:109399, 2020.
- [83] V.V. Zerkín and B. Pritychenko. The experimental nuclear reaction data (EXFOR): Extended computer database and Web retrieval system. *Nuclear*

*Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 888:31–43, 2018.

- [84] Neutron capture cross-section | nuclear-power.com.  
<https://www.nuclear-power.com/nuclear-power/reactor-physics/nuclear-engineering-fundamentals/neutron-nuclear-reactions/neutron-capture-radiative-capture/neutron-capture-cross-section/>.
- [85] Junhyun Kwon and Yong Hee Choi. Calculation of gamma displacement cross sections: Generation of recoil spectra from ENDF/b-VII, 2012.
- [86] S. Chen, D. Bernard, and C. De-Saint Jean. Calculation and analysis of gamma-induced irradiation damage cross section. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 447:8–21, 2019.
- [87] E. M. A. Hussein. Chapter three-cross-sections. In Esam M. A. Hussein, editor, *Radiation Mechanics*, pages 153–245. Elsevier Science Ltd, 2007. <https://www.sciencedirect.com/science/article/pii/B9780080450537500045>.
- [88] K. S. Krane. *Introductory Nuclear Physics*. John Wiley & Sons, 3rd edition edition, 1987.
- [89] IAEA. Assessment and management of ageing of major nuclear power plant components important to safety: Pwr pressure vessels, iaea-tecdoc-1556. Technical report, International Atomic Energy Agency, Vienna, Austria, 2007.
- [90] Matts Roos. Sources of gamma radiation in a reactor core, 1959. <https://www.ipen.br/biblioteca/rel/R17855.pdf>.
- [91] Z. Xu, L. Chen, B. Zhou, Y. Li, B. Li, J. Niu, M. Shan, Q. Guo, Z. Wang, and X. Qian. Nano-structure and property transformations of carbon systems under -ray irradiation: A review. *RSC Adv.*, 3(27):10579–10597, 2013.
- [92] F. Spaepen and T. David. *Crystallization processes*. Academic Press, New York, U.S.A, 1982.

- [93] M. Nastasi and J.W. Mayer. *Ion implantation and Synthesis of Materials*. Springer-Verlag Berlin Heidelberg, Berlin, Germany, 2006.
- [94] J. Lindhard, M. Scharff, and H.E. Schioett. Range concepts and heavy ion ranges (notes on atomic collisions ii). *Kgl. Danske Videnskab. Selskab. Mat. Fys. Medd.*, 33(14), 1 1963.
- [95] W. Moller. *Fundamentals of Ion-Solid Interactions: A Compact Introduction*. Helmholtz-Zentrum Dresden - Rossendorf, Dresden, Germany, 2017. [https://inis.iaea.org/collection/NCLCollectionStore/\\_Public/48/080/48080444.pdf](https://inis.iaea.org/collection/NCLCollectionStore/_Public/48/080/48080444.pdf).
- [96] H. M. Presby; W. L. Brown. Refractive index variations in proton-bombarded fused silica. *Appl. Phys. Lett.*, 24(10), 1974.
- [97] S. Nagata, H. Katsui, B. Tsuchiya, A. Inouye, S. Yamamoto, K. Toh, and T. Shikma. Damage process and luminescent characteristics in silica glasses under ion irradiation. *Journal of Nuclear Materials*, 386-388:1045–1048, 2009. Fusion Reactor Materials.
- [98] A. Iwase, S. Sasaki, T. Iwata, and T. Nihira. Anomalous reduction of stage-i recovery in nickel irradiated with heavy ions in the energy range 100–120 mev. *Phys. Rev. Lett.*, 58:2450–2453, Jun 1987.
- [99] A. Dunlop, D. Lesueur, J. Morillo, J. Dural, R. Spohr, and J. Vetter. High electronic excitations and damage production in gev-ion-irradiated metals. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 48(1):419–424, 1990.
- [100] R. A. Serway. *Physics for scientist and Engineers with Modern Physics, 5th edition*. Harcourt College Pub, 2000.
- [101] R. E. Stoller, M. B. Toloczko, G. S. Was, A. G. Certain, S. Dwaraknath, and F. A. Garner. On the use of SRIM for computing radiation damage exposure. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 310:75–80, 2013.

- [102] S. J. Zinkle and J. T. Busby. Structural materials for fission fusion energy. *Materials Today*, 12(11):12–19, 2009.
- [103] Brandon Bohanon, Peng Wei, Ashley Foster, Layali Bazar, Yongfeng Zhang, Douglas Spearot, Mukesh Bachhav, Laurent Capolungo, and Assel Aitkaliyeva. A critical review of irradiation-induced changes in reactor pressure vessel steels. *Progress in Nuclear Energy*, 174:105276, 2024.
- [104] H. Watanabe, S. Masaki, S. Masubuchi, N. Yoshida, and K. Dohi. Effects of mn addition on dislocation loop formation in a533b and model alloys. *Journal of Nuclear Materials*, 439(1):268–275, 2013.
- [105] Ying Luo, Yuanyuan Dong, Xiaotong Wang, Hang Peng, Dapeng Yan, Tian Hu, Shanglin Zhang, Qingyu Li, Dian Wang, and Cong Xiao. In-situ tem study of the effect of pre-existing dislocation on loop evolution in 508-iii steel during fe+ irradiation. *Journal of Nuclear Materials*, 559:153420, 2022.
- [106] E.A. Kuleshova, B.A. Gurovich, Z.V. Bukina, A.S. Frolov, D.A. Maltsev, E.V. Krikun, D.A. Zhurko, and G.M. Zhuchkov. Mechanisms of radiation embrittlement of vver-1000 rpv steel at irradiation temperatures of (50–400)°c. *Journal of Nuclear Materials*, 490:247–259, 2017.
- [107] T. Fujita, J. Hirabayashi, Y. Katayama, F. Kano, and H. Watanabe. Contribution of dislocation loop to radiation-hardening of rpv steels studied by stem/eds with surveillance test pieces. *Journal of Nuclear Materials*, 572:154055, 2022.
- [108] J.C. Haley, S. de Moraes Shubeita, P. Wady, A.J. London, G.R. Odette, S. Lozano-Perez, and S.G. Roberts. Microstructural examination of neutron, proton and self-ion irradiation damage in a model fe9cr alloy. *Journal of Nuclear Materials*, 533:152130, 2020.
- [109] D. G. Martin. Radiation damage effects in crystals. *Science Progress (1933-)*, 54(214):209–225, 1966. Publisher: Temporary Publisher.
- [110] D. J. Bacon, H. F. Deng, and F. Gao. Computer simulation of threshold displacement events in alloys. *Journal of Nuclear Materials*, 205:84–91, 1993.

- [111] W. E. King, K. L. Merkle, and M. Meshii. Threshold energy surface and frenkel pair resistivity for cu. *Journal of Nuclear Materials*, 117:12–25, 1983.
- [112] D. Brandon and D. K. Wayne. *Microstructural Characterization of Materials- 2nd Edition*. John Wiley Sons, Ltd, England, 2008.
- [113] M. Ladd and R. Palmer. *Structure determination by X-ray crystallography: Analysis by X-rays and Neutrons- 5th edition*. Springer, New York Heidelberg, Dordrecht London, Dordrecht, London, 2013.
- [114] M. Sowinska. In-operando hard x-ray photoelectron spectroscopy study on the resistive switching physics of hfo2-based rram, 2014. [https://www.researchgate.net/publication/283068066\\_In-operando\\_hard\\_X-ray\\_photoelectron\\_spectroscopy\\_study\\_on\\_the\\_resistive\\_switching\\_physics\\_of\\_HfO2-based\\_RRAM](https://www.researchgate.net/publication/283068066_In-operando_hard_X-ray_photoelectron_spectroscopy_study_on_the_resistive_switching_physics_of_HfO2-based_RRAM).
- [115] B. Mallick, T. Patel, R.C. Behera, S.N. Sarangi, S.N. Sahu, and R.K. Choudhury. Microstrain analysis of proton irradiated pet microfiber. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 248(2):305–310, 2006.
- [116] D. Kopeliovich. Imperfections of crystal structure [SubsTech], 2012. [http://ww.substech.com/dokuwiki/doku.php?id=imperfections\\_of\\_crystal\\_structure,.](http://ww.substech.com/dokuwiki/doku.php?id=imperfections_of_crystal_structure,)
- [117] H. Liu. Dislocation, 2017. <https://medium.com/@x399liu/dislocation-b1f6f36b23ac>.
- [118] L. E. Murr. Volume defects: 3d imperfections in crystals. In Lawrence E. Murr, editor, *Handbook of Materials Structures, Properties, Processing and Performance*, pages 1–11. Springer International Publishing, 2021. [https://doi.org/10.1007/978-3-319-01905-5\\_17-1](https://doi.org/10.1007/978-3-319-01905-5_17-1).
- [119] G.S. Was and P.L. Andresen. Radiation damage to structural alloys in nuclear power plants: Mechanisms and remediation. In *Structural Alloys for Power Plants Operational Challenges and High-temperature Materials*, pages 355–419. Woodhead Publishing, USA. DOI:10.1533/9780857097552.2.355.

- [120] W.J. Weber. Radiation-induced amorphization in complex silicates. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 65(1):88–92, 1992.
- [121] A. Y. Konobeyev, U. Fischer, Y. A. Korovin, and S. P. Simakov. Evaluation of effective threshold displacement energies and other data required for the calculation of advanced atomic displacement cross-sections. *Nuclear Energy and Technology*, 3(3):169–175, 2017.
- [122] IAEA. Characterization and testing of materials for nuclear reactors. Proceedings of a technical meeting IAEA-TECDOC-1545, International Atomic Energy Agency, 2007. [https://www-pub.iaea.org/MTCD/publications/PDF/te\\_1545\\_web.pdf](https://www-pub.iaea.org/MTCD/publications/PDF/te_1545_web.pdf).
- [123] P.K. Suri, J. E. Nathaniel, N. Li, J. K. Baldwin, Y. Wang, K. Hattar, and M. L. Taheri. Ion irradiation induced phase transformation in gold nanocrystalline films. *Scientific Reports*, 10(1):17864, 2020. Number: 1 Publisher: Nature Publishing Group.
- [124] H. M. Urbassek and L. Sandoval. 14 - Molecular dynamics modeling of martensitic transformations in steels. In Elena Pereloma and David V. Edmonds, editors, *Phase Transformations in Steels*, volume 2 of *Woodhead Publishing Series in Metals and Surface Engineering*, pages 433–463. Woodhead Publishing, 2012. <https://www.sciencedirect.com/science/article/pii/B9781845699710500144>.
- [125] J. Li, W. Fuliang, L. Han, and J. Zhong. Theoretical and experimental analyses of atom diffusion characteristics on wire bonding interfaces. *J. Phys. D: Appl. Phys.*, 41(13):135303, 2008.
- [126] G. S. Was, J.P. Wharry, Brian Frisbie, B. D. Wirth, D. Morgan, J. D. Tucker, and T. R. Allen. Assessment of radiation-induced segregation mechanisms in austenitic and ferritic-martensitic alloys. *Journal of Nuclear Materials*, 411(1):41–50, 2011.

- [127] C. Zheng, M. A. Auger, M. P. Moody, and D. Kaoumi. Radiation induced segregation and precipitation behavior in self-ion irradiated Ferritic/Martensitic HT9 steel. *Journal of Nuclear Materials*, 491:162–176, 2017.
- [128] S. A. Fabritsiev and A. S. Pokrovsky. The effect of neutron irradiation on the electrical resistivity of high-strength copper alloys. *Journal of Nuclear Materials*, 249(2):239–249, 1997.
- [129] M. Izerroukena, O. FadilaHaidc A. Sarib, and A. Ishaq. A comparison between neutron damage in annealed gold and in quenched gold. *Appl. Phy. Lett.*, 9, 1966.
- [130] S.T. Temaugee, L. Bedhesi, R.D. Mavunda, G.C. Daniels, S.H. Connell, I.T. Usman, and E. Chinaka. Calculation of neutron-induced radiation damage to gold and lutetium-aluminium microstructures using mcnp6.2. *Journal of Nuclear Materials*, 587:154707, 2023.
- [131] N. J. Briot, M. Kosmidou, R. Dingreville, K. Hattar, and T.J. Balk. In situ tem investigation of self-ion irradiation of nanoporous gold. *J Mater Sci*, 54, 2019.
- [132] K. G. Field, Xunxiang Hu, Kenneth C. Littrell, Yukinori Yamamoto, and Lance L. Snead. Radiation tolerance of neutron-irradiated model fe–cr–al alloys. *Journal of Nuclear Materials*, 465:746–755, 2015.
- [133] Pramote Thirathipviwat, Sega Nozawa, Moe Furusawa, Yusuke Onuki, Makoto Hasegawa, Katsushi Matsumoto, and Shigeo Sato. In-situ neutron diffraction study on a dislocation density in a correlation with strain hardening in al–mg alloys. *Materials Science and Engineering: A*, 855:143956, 2022.
- [134] R. Nabbi and J. Wolters. Investigation of radiation damage in the aluminum structures of the german frj-2 research reactor. In *Proceedings of the Eighth World Conference of Planners, Builders and Operators of Research Reactors, IGORR 8, Munich, Germany.*, 2001. [https://inis.iaea.org/collection/NCLCollectionStore/\\_Public/36/026/36026552.pdf](https://inis.iaea.org/collection/NCLCollectionStore/_Public/36/026/36026552.pdf).

- [135] S. L. Quaireau, K. Colas, B. Kapusta, B. Verhaeghe, M. Loyer-Prost, G. Gutierrez, D. Gosset, and S. Delpech. Impact of ion and neutron irradiation on the corrosion of the 6061-t6 aluminium alloy. *J. Nucl. Mater.*, 553:153051, 2021.
- [136] V. Garric, K. Colas, P. Donnadieu, M. Loyer-Prost, F. Lepretre, V. Cloute-Cazalaa, and B. Kapusta. Impact of the microstructure on the swelling of aluminum alloys: Characterization and modelling bases. *J. Nucl. Mater.*, 557:153273, 2021.
- [137] P. Bhattacharyya, N. Gayathri, M. Bhattacharya, A. Dutta Gupta, Apu Sarkar, S. Dhar, M.K. Mitra, and P. Mukherjee. Proton irradiation studies on al and al5083 alloy. *Radiation Physics and Chemistry*, 139:132–140, 2017.
- [138] B. Wei, D. Xie, W. Wu, L. Shao, and J. Wang. Quantifying the Glide Resistance to Dislocations in Proton-Irradiated FeCrAl Alloy. *JOM*, 74:4035–4041, 2022.
- [139] M. Izerroukena, O. FadilaHaidc A. Sarib, and A. Ishaq. Al 6061-t6 microstructure and mechanical properties modification under ion beam irradiation at room temperature: Application for nuclear research reactor. *Algerian Journal of Engineering and Technology*, 8:272–278, 2023.
- [140] O. Noori-kalkhoran and M. Gei. Evaluation of neutron radiation damage in zircaloy fuel clad of nuclear power plants: A study based on PKA and dpa calculations. *Progress in Nuclear Energy*, 118:103079, 2020.
- [141] Wootan, David. DPA calculational methodologies used in fission and fusion reactor materials applications. In *5th High Power Targetry Workshop*, 2014. <https://indico.fnal.gov/event/7870/book-of-abstracts.pdf>.
- [142] B. D. Wirth, P. Monasterio, and W. Stein. Calculation of radiation damage in SLAC targets. <https://digital.library.unt.edu/ark:/67531/metadc894786/>.

- [143] S. A. Santa and Suwoto. Neutron radiation damage estimation in the core structure base metal of RSG GAS. *J. Phys.: Conf. Ser.*, 962:012050, 2018. Publisher: IOP Publishing.
- [144] T.A.D. Rollett. *Materials for Nuclear Energy Radiation Damage: Atom Displacement*. Carnegie Mellon University, 2019.
- [145] M. J. Norgett, M. T. Robinson, and I. M. Torrens. A proposed method of calculating displacement dose rates. *Nuclear Engineering and Design*, 33(1):50–54, 1975.
- [146] D.B. Williams and Carter C. B. *Transmission Electron Microscopy Second Edition*. Springer, New York USA, 2009.
- [147] J. Gallet, M. Perez, S. Dubail, T. Chaise, T. Douillard, C. Langlois, and S. Cazottes. About the automatic measurement of the dislocation density obtained by r-ecci. *Materials Characterization*, 194:112358, 2022.
- [148] C. A. Hirst. *Quantifying Radiation Damage through Stored Energy Released during Defect Annealing in Metals*. PhD thesis, Massachusetts Institute of Technology, May 2022. <https://dspace.mit.edu/handle/1721.1/144559>.
- [149] Z. Zhou, S.L. Dudarev, M.L. Jenkins, A.P. Sutton, and M.A. Kirk. Diffraction imaging and diffuse scattering by small dislocation loops. *Journal of Nuclear Materials*, 367–370:305–310, 2007.
- [150] G. K. Williamson and W. H. Hall. X-ray line broadening from fcc aluminium and wolfram. *Acta Metallurgica*, 1(1):22–31, 1953.
- [151] C.A. Hirst, F. Granberg, B. Kombaiah, P. Cao, S. Middlemas, R.S. Kemp, J. Li, K. Nordlund, and M.P. Short. Revealing hidden defects through stored energy measurements of radiation damage. *Science Advances*, 8, 2022.
- [152] D. A. Toktogulova, M. N. Gusev, O. P. Maksimkin, and F. A. Garner. Influence of Neutron Irradiation on Energy Accumulation and Dissipation during Plastic Flow and Hardening of Metallic Polycrystals. *Journal of ASTM International*, 7(1):1–10, 01 2010.

- [153] K. Hajizadeh, S. Ghobadi Alamdari, and B. Eghbali. Stored energy and recrystallization kinetics of ultrafine grained titanium processed by severe plastic deformation. *Physica B: Condensed Matter*, 417:33–38, 2013.
- [154] E.P. Wigner. Theoretical physics in the metallurgical laboratory of chicago,. *Journal of Applied Physics*, 17:857–8633, 1946.
- [155] Tadao Iwata. Fine structure of wigner energy release spectrum in neutron irradiated graphite. *Journal of Nuclear Materials*, 133-134:361–364, 1985.
- [156] R. Poirier, S. Roorda, F. Schiettekatte, M. Lalancette, and J. Zikovsky. Divacancies in proton irradiated silicon: characterization and annealing mechanisms. *Physica B: Condensed Matter*, 308-310:462–464, 2001. International Conference on Defects in Semiconductors.
- [157] L. Gomell, M. Roscher, H. Bishara, E. A. Jäggle, C. Scheu, and B. Gault. Properties and influence of microstructure and crystal defects in Fe2VAl modified by laser surface remelting. *Scripta Materialia*, 193:153–157, 2021-03-01.
- [158] A. Hodgson, R. Grimes, M. Rushton, and O. Marsden. The Impact of Neutron Cross Section Group Structures on the Accuracy of Radiological Source Models. *Nuclear science and engineering: the journal of the American Nuclear Society*, 181:302–309, 2015.
- [159] D. Lee. An overview of computational methods for nuclear physics modelling, 2019. doi:10.13140/RG.2.2.32042.36808.
- [160] E.E Lewis and W.F Miller. *Computational Methods of Neutron Transport*. John Wiley & Sons Inc., 1984.
- [161] B. D. d. A. Rodriguez, M. T. Vilhena, and B. E. J. Bodmann. An overview of the Boltzmann transport equation solution for neutrons, photons and electrons in Cartesian geometry. *Progress in Nuclear Energy*, 53(8):1119–1125, 2011.
- [162] T. E. Booth, F. B. Brown, J.S. Bull, Lawrence J. C., Forster, R. A., Goorle, J. T. , Hughes. H.G., Mosteller, R.D., Prael, R.E., Selcow.E.C, Sood, A., Sweezy,

- J.E., Barrett, R.F., Post, S.E., and Roberts, T.L. MCNP — A General Monte Carlo N-Particle Transport Code, Version 5, 2003.
- [163] A. Kling, F. J. C. Barão, M. Nakagawa, L. Távora, and P. Vaz, editors. *Advanced Monte Carlo for Radiation Physics, Particle Transport Simulation and Applications: Proceedings of the Monte Carlo 2000 Conference, Lisbon, 23–26 October 2000*. Springer, 2001.
- [164] J. A. Kulesza, T. R. Adams, J. C. Armstrong, S. R. Bolding, F. B. Brown, J. S. Bull, T. P. Burke, A. R. Clark, R. A. (Art) Forster III, J. F. Giron, T. S. Grieve, C. J. Josey, R. L. Martz, G. W. McKinney, E. J. Pearson, M. E. Rising, C. J. Solomon Jr., S. Swaminarayan, T. J. Trahan, S. C. Wilson, and A. J. Zukaitis. MCNP® Code Version 6.3.0 Theory & User Manual, 2022. url <https://www.osti.gov/biblio/1889957>.
- [165] Z. Karriem and O Zamonsky. Evaluation of the Oscar-4/MCNP Calculation Methodology for Radioisotope Production in the SAFARI-1 Reactor. 2014. [https://www.researchgate.net/publication/291825019\\_Evaluation\\_of\\_the\\_Oscar-4MCNP\\_Calculation\\_Methodology\\_for\\_Radioisotope\\_Production\\_in\\_the\\_SAFARI-1\\_Reactor](https://www.researchgate.net/publication/291825019_Evaluation_of_the_Oscar-4MCNP_Calculation_Methodology_for_Radioisotope_Production_in_the_SAFARI-1_Reactor).
- [166] L. Jurbandam. Calculation of the fission q-value and spatial energy deposition in the safari-1 nuclear reactor. *undefined*, 2018. <https://www.semanticscholar.org/paper/Calculation-of-the-fission-q-value-and-spatial-in-Jurbandam/55d24e392d8485d4748bbacdaac2ab4849c689ea>.
- [167] R. Macfarlane, D. W. Muir, R. M. Boicourt, I. I. I. Kahler, and J. L. Conlin. The NJOY nuclear data processing system, version 2016, 2017-01-09. <https://www.osti.gov/biblio/1338791>.
- [168] E.A. Read and C.R.E de Oliveira. A functional method for estimating DPA tallies in monte carlo calculations of light water reactors. 2011. [https://www.researchgate.net/publication/265110861\\_A\\_FUNCTIONAL\\_METHOD\\_FOR\\_ESTIMATING\\_DPA\\_TALLIES\\_IN\\_MONTE\\_CARLO\\_CALCULATIONS\\_OF\\_LIGHT\\_WATER\\_REACTORS](https://www.researchgate.net/publication/265110861_A_FUNCTIONAL_METHOD_FOR_ESTIMATING_DPA_TALLIES_IN_MONTE_CARLO_CALCULATIONS_OF_LIGHT_WATER_REACTORS).

- [169] M. Lemaire, H. Lee, and D. Lee. KERMA and DPA tallies in the monte carlo code MCS. 2020. <https://journal-home.s3.ap-northeast-2.amazonaws.com/site/kns2020spring/presentation/20S-260.pdf>.
- [170] F. Leszczynski, Lopez A. D., and A. Trkov. Wims-d library update. International Atomic Energy Agency (IAEA) Vienna, 2007. [https://www-pub.iaea.org/MTCD/publications/PDF/Pub1264\\_web.pdf](https://www-pub.iaea.org/MTCD/publications/PDF/Pub1264_web.pdf).
- [171] Sweezy, J. E., Booth. T.E., Brown, F.B., Bull, J.S., Cox, L.J., Forster, A.R., Goorley,J.T., Hughes, H.G., Mosteller, R.D., and et al. MCNP — A General Monte Carlo N-Particle Transport Code, Version 5. Volume 1: Overview and Theory. Technical Report LA-UR-03-1987 (Revised 2/1/2008), Los Alamos National Lab. (LANL), Los Alamos, NM (United States, 2003. = <http://permalink.lanl.gov/object/tr?what=info:lanl-repo/lareport/LA-UR-03-1987>.
- [172] J.K. Shultis and R.E. Faw. An MCNP Primer, 2011. <https://www.mne.k-state.edu/~jks/MCNPprmr.pdf>.
- [173] R.S. Detwiler, R.J. McConn, T.F. Grimes, S.A. Upton, and E.J Engel. Compendium of material composition data for radiation transport modeling. Technical Report 200-DMAMC-128170, PNNL-15870, Rev. 2, Pacific Northwest National Laboratory, 2021. [https://www.pnnl.gov/main/publications/external/technical\\_reports/PNNL-15870Rev2.pdf](https://www.pnnl.gov/main/publications/external/technical_reports/PNNL-15870Rev2.pdf).
- [174] S. Chen and D. Bernard. On the calculation of atomic displacements using damage energy. *Results in Physics*, 16:102835, 2020.
- [175] Y. Lin, S.J. Zinkle, C.J. Ortiz, J. Crocombette, R. Webb, and R.E. Stoller. Predicting displacement damage for ion irradiation: Origin of the overestimation of vacancy production in srim full-cascade calculations. *Current Opinion in Solid State and Materials Science*, 27(6):101120, 2023.
- [176] K.A. Gschneidner and F.W. Calderwood. The allu (aluminum-lutetium) system. *Bulletin of Alloy Phase Diagrams*, 9:690–691, 1988.

- [177] The Materials Project. Materials data on au by materials project. doi:10.17188/1307925.
- [178] The Materials Project. Materials data on lual3 by materials project. doi:10.17188/1202177.
- [179] L. A. Kesler. *Development and Testing of an in Situ Method of Ion Beam Analysis for Measuring High-Z Erosion inside a Tokamak Using an AIMS Diagnostic*. PhD thesis, Massachusetts Institute of Technology, July 2019.
- [180] D. Whyte. Final technical report-bridging the psi knowledge gap: A multiscale approach. Technical Report DOE-MIT-2060-1, Massachusetts Inst. of Technology (MIT), Cambridge, MA (United States), December 2014. <https://www.osti.gov/biblio/1166950>.
- [181] Z. S. Hartwig. The class 1.7 mv tandem ion accelerator. Center for Science and Technology with Accelerators and Radiation, 2015. <http://cstar.mit.edu/tandetron.php>.
- [182] H.S. Barnard. *External Proton Beam Analysis of Plasma Facing Materials for Magnetic Confinement Fusion Applications*. PhD thesis, Massachusetts Institute of Technology, September 2009.
- [183] S. A. Speakman. Using the open eularian cradle (oec) using the high-speed bragg-brentano optics on the panalytical x’pert pro mpd, 2015. <http://prism.mit.edu/xray/education/downloads.html>.
- [184] J.I. Langford and A.J.C Wilson. Scherrer after sixty years: A survey and some new results in the determination of crystallite size. *J. Appl. Cryst.*, 11:102–113, 1978.
- [185] T. Ungar, J. Gubicza, G. Ribarik, and A. Borbely. Crystallite size distribution and dislocation structure determined by diffraction profile analysis: Principles and practical application to cubic and hexagonal crystals. *J. Appl. Cryst.*, 34:298–310, 2001.
- [186] P. Bindu and S. Thomas. Estimation of lattice strain in ZnO nanoparticles: X-ray peak profile analysis. *J Theor Appl Phys*, 8(4):123–134, 2014.

- [187] P. Karamched. Transmission kikuchi diffraction. oxford micromechanics group, department of materials, university of oxford, 2018. <https://omg.web.ox.ac.uk/article/transmission-kikuchi-diffraction>.
- [188] Y. Zhao, Y. Jin, S. Lv, J. Gao, Z. Zhou, T. Yano, and Z. Li. The Wigner energy and defects evolution of graphite in neutron-irradiation and annealing. *Radiation Physics and Chemistry*, 201:110401, 2022.
- [189] A. Mathot, M. Pyda, T. Pijpers, V. P. Geert, V. Ernst, V. Sander, V Floor, and L. Archi. The flash dsc 1, a power compensation twin-type, chip-based fast scanning calorimeter (fsc): First findings on polymers. *Thermochimica Acta*, 522(1):36–45, 2011. Special Issue: Interplay between Nucleation, Crystallization, and the Glass Transition.
- [190] S. Taller, G. VanCoevinger, B. D. Wirth, and G. S. Was. Predicting structural material degradation in advanced nuclear reactors with ion irradiation. *Sci Rep*, 11(1):2949, 2021.
- [191] U. Saha, K. Devan, and S. Ganesan. A study to compute integrated dpa for neutron and ion irradiation environments using srim-2013. *Journal of Nuclear Materials*, 503:30–41, 2018.
- [192] N. Galy, N. Toulhoat, N. Moncoffre, Y. Pipon, N. Béererd, M.R. Ammar, P. Simon, D. Deldicque, and P. Sainsot. Ion irradiation used as surrogate of neutron irradiation in graphite: Consequences on <sup>14</sup>c and <sup>36</sup>cl behavior and structural evolution. *Journal of Nuclear Materials*, 502:20–29, 2018.
- [193] P. Sathyavathi, S.T. Chavan, D. Kanjilal, and V.N. Bhoraskar. Heavy ion induced damage in crystalline silicon and diodes. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 156(1):72–77, 1999.
- [194] J.D. Carlson, J.E. Bares, A.M. Guzman, and P.P. Pronko. Surface property changes induced in poly(1-hexene) elastomer by high energy ion irradiation. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 7-8:507–512, 1985.

- [195] T. Ungár, S. Ott, P.G. Sanders, A. Borbély, and J.R. Weertman. Dislocations, grain size and planar faults in nanostructured copper determined by high resolution x-ray diffraction and a new procedure of peak profile analysis. *Acta Materialia*, 46(10):3693–3699, 1998.
- [196] A. Stukowski, J. Markmann, J. Weissmüller, and K. Albe. Atomistic origin of microstrain broadening in diffraction data of nanocrystalline solids. *Acta Materialia*, 57(5):1648–1654, 2009.
- [197] P. Mukherjee, A. Sarkar, M. Bhattacharya, N. Gayathri, and P. Barat. Post-irradiated microstructural characterization of cold-worked ss316l by x-ray diffraction technique. *J. Nucl. Mater.*, 395:37–44, 2009.
- [198] P. Xiu, H. Bei, Y. Zhang, H. Wang, and K.G. Field. Stem characterization of dislocation loops in irradiated fcc alloys. *Journal of Nuclear Materials*, 544:152658, 2021.
- [199] J. Takamura, M. Doyama, and M. Kiritani. Point defects, and defect interactions in metals. *Acta Cryst.*, A40:310, 1984.
- [200] B.N. Singh, S.I. Golubov, H. Trinkaus, D.J. Edwards, and M. Eldrup. Review: Evolution of stacking fault tetrahedra and its role in defect accumulation under cascade damage conditions. *Journal of Nuclear Materials*, 328(2):77–87, 2004.
- [201] J. J. Sidor, P. Chakravarty, J.G. Bátorfi, P. Nagy, Q. Xie, and J. Gubicza. Assessment of dislocation density by various techniques in cold rolled 1050 aluminum alloy. *Metals*, 11(10), 2021.
- [202] Y. Hanrong. Evaluation of thermal neutron capture cross section of gold. in: Böckhoff, k.h. (eds) nuclear data for science and technology. springer, dordrecht, 1983. [https://doi.org/10.1007/978-94-009-7099-1\\_151](https://doi.org/10.1007/978-94-009-7099-1_151).
- [203] IAEA. Atlas of neutron capture cross sections. indc(nds)-362, 1997. <https://www-nds.iaea.org/ngatlas2/>.
- [204] H. Wei and Y. Wei. Interaction between a screw dislocation and stacking faults in FCC metals. *Materials Science and Engineering: A*, 541:38–44, 2012.

- [205] Nathaniel Burbery, Raj Das, and George Ferguson. Mobility of dissociated mixed dislocations under an escaig stress. *Modelling and Simulation in Materials Science and Engineering*, 25(4):045001, mar 2017.
- [206] R. Su, D. Neffati, S. Xue, Q. Li, Z. Fan, Y. Liu, H. Wang, Y. Kulkarni, and X. Zhang. Deformation mechanisms in FCC Co dominated by high-density stacking faults. *Materials Science and Engineering: A*, 736:12–21, 2018.
- [207] E. H. Kim, M. Kim, H. S. Song, S. H. Yoo, S. Sai, K. Chung, J. Sung, Y. K. Jeong, Y. Jo, and M. Yoon. Gold nanoparticles as a potent radiosensitizer in neutron therapy. *Oncotarget*, 8(68):112390–112400, 2017. `_eprint: 29348833`  
`_eprinttype: pmid`.
- [208] R. Krishna, J. Wade, A. N. Jones, M. Lasithiotakis, P. M. Mummery, and B. J. Marsden. An understanding of lattice strain, defects and disorder in nuclear graphite. *Carbon*, 124:314–333, 2017.
- [209] L.K. Berg, J.G. Jonnes, X.Z Li, M. Knutson-Wedel, G. Waterloo, D. Schryvers, and L.R Wallenberg. Gp-zones in al-zn-mg alloys and their role in artificial aging. *Acta Mater.*, 49:3443–3451, 2001.
- [210] Z.Q. Li, W.R. Ren, H.W. Chen, and J.F. Nie. precipitate phase, gp zone clusters and their origin in al-cu alloys. *Journal of Alloys and Compounds*, 930:167396, 2023.

# Publications and Awards

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## Journals:

- [1] **ST Temaugee**, L Bedhesi, RD Mavunda, GC Daniels, SH Connell, IT Usman and E Chinaka. "Calculation of neutron-induced radiation damage to gold and lutetium-aluminum microstructures using MCNP6.2" *Journal of Nuclear Materials*, Volume 587, 2023, 154707. <https://doi.org/10.1016/j.jnucmat.2023.154707>
- [2] **ST Temaugee**, L Bedhesi, RD Mavunda, SH Connell, GC Daniels, and IT Usman. "Evaluation of neutron radiation damage on gold microstructure using MCNP" *Journal of Physics: Conference Series*, volume 2586 (2023) 012122 [doi:10.1088/1742-6596/2586/1/012122](https://doi.org/10.1088/1742-6596/2586/1/012122)

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## Conferences:

### (a) International

- [1] **ST Temaugee**, L Bedhesi, RD Mavunda, GC Daniels, SH Connell, IT Usman and E Chinaka. "Effects of nuclear radiation damage on microstructure in lutetium-aluminium and gold materials using neutron activation analysis," *The Nuclear Material Conference*, Ghent, Belgium, 2022.
- [2] **ST Temaugee**, IT Usman, RD Mavunda, SH Connell, GC Daniels, and L Bedhesi "Evaluation of radiation-induced effects on microstructural characteristics of gold using neutron irradiation," *International Nuclear Physics Conference (INPC)*, Cape Town, South Africa, 2022

### (a) Local

- [1] **ST Temaugee**, IT Usman, and RD Mavunda "Ion-induced radiation damage in Lutetium-Aluminium and Gold using SRIM-2013" *South African Institute*

*of Physics (SAIP) Conference*, University of Zulu Land, Richards Bay, South Africa, 2023,

- [2] **ST Temaugee**, IT Usman, RD Mavunda, S.H. Connell, GC Daniels, and L Bedhesi "Evaluation of Nuclear Radiation Damage on Lutetium-Aluminium for Practical Applications Using Neutron Irradiation Technique " *Advanced Nuclear Techniques and Technology Workshop*, iThemba Labs, Cape Town, South Africa, 2022,

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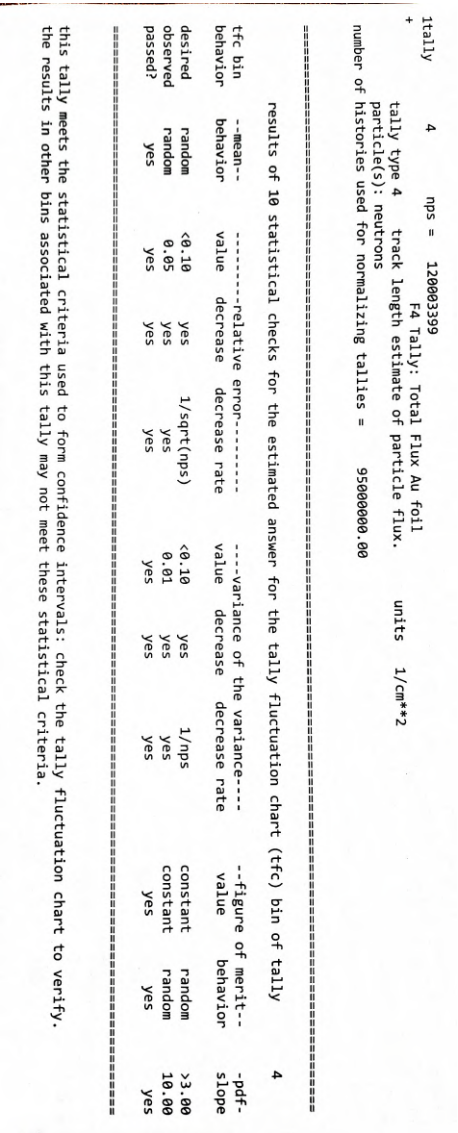
## Awards:

- **Best Oral Presentation**-Advanced Nuclear Techniques and Technology Workshop 2022. A Co-Hosted by iThemba Labs, South Africa, and the University of Brighton, Uk. (see Appendix G.)
- **Best PhD Poster presentation**- South African Institute of Physics (SAIP) Conference 2023

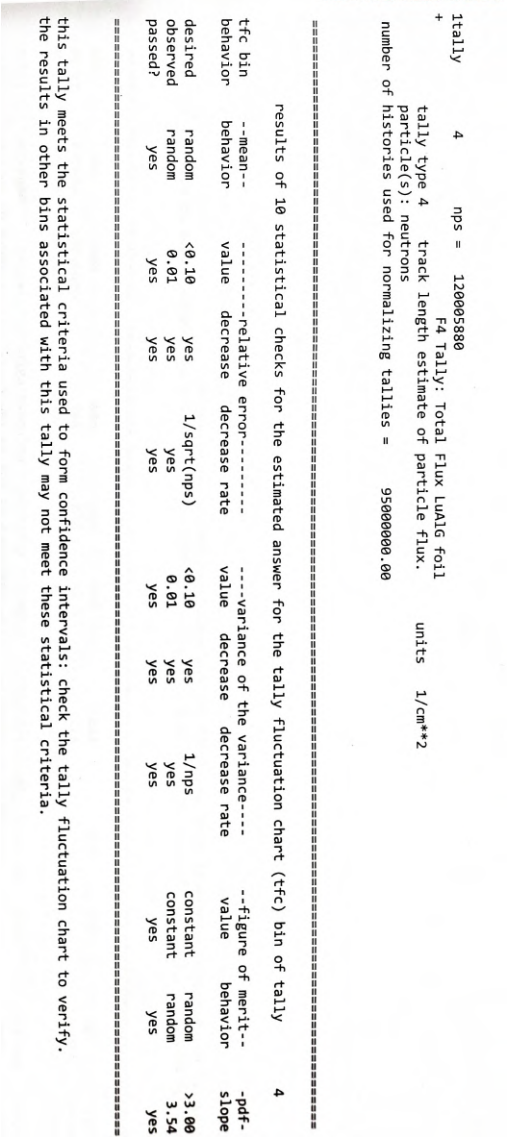
# Appendices

## Appendix A - MCNP Tally Fluctuation Chart (TFC)

### A.1 Tally fluctuation chart (TFC) for Lu-Al



## A.2 Tally fluctuation chart (TFC) for Au



## Appendix B - Post-Irradiation Cooling time from FISPACT II

The dose rate-cooling time response function obtained from FISPACT II calculations is given by

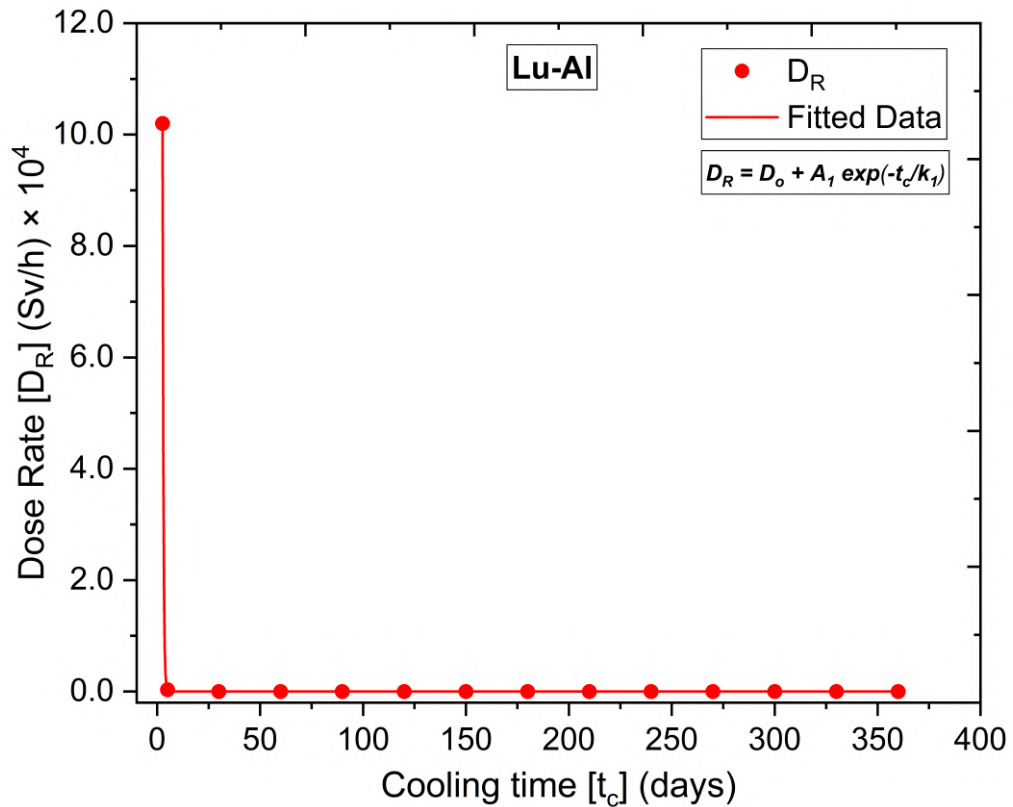
$$D_R = D_o + A_1 e^{-(t_c/k_1)}. \quad (1)$$

where  $D_R$  is the dose rate in  $Sv/h$  and  $t_c$  is the cooling time,  $D_o$ ,  $A_1$  and  $k_1$  are fitting parameters.

### B.1 Post-Irradiation Cooling time for Lu-Al

The dose rate-cooling time response decay function for Lu-Al is:

$$D_R = 1.52383 + 5.425E07 e^{-(t_c/0.4158)} \quad (2)$$

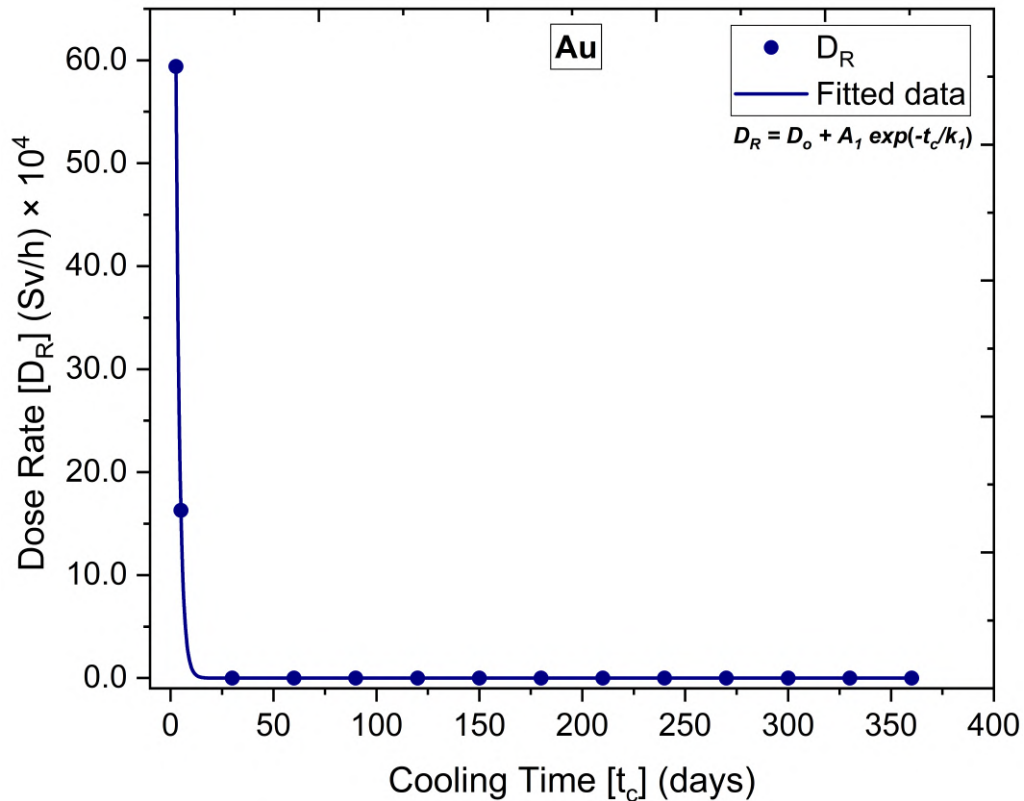


| Table B.1. Predicted Lu-Al cooling time after irradiation of 10 mg for 2.61 days. |                        |                       |                  |       |                     |       |                     |       |
|-----------------------------------------------------------------------------------|------------------------|-----------------------|------------------|-------|---------------------|-------|---------------------|-------|
|                                                                                   | Cooling Time<br>(step) | Cumulative<br>(Years) | Activity<br>(Bq) |       | Dose rate<br>(Sv/h) |       | Heat output<br>(kW) |       |
| Irradn                                                                            | 2.610 d                | 7.15E-03              | 2.33E+12 +/-     | 13.0% | 1.02E+05 +/-        | 9.2%  | 3.63E-04 +/-        | 8.8%  |
| Cooling                                                                           | 5.000 d                | 1.37E-02              | 2.61E+11 +/-     | 16.7% | 3.27E+02 +/-        | 16.6% | 7.57E-06 +/-        | 16.7% |
| Cooling                                                                           | 30.000 d               | 9.58E-02              | 1.14E+10 +/-     | 16.6% | 1.47E+01 +/-        | 16.1% | 3.32E-07 +/-        | 16.6% |
| Cooling                                                                           | 30.000 d               | 1.78E-01              | 5.30E+08 +/-     | 16.1% | 1.10E+00 +/-        | 10.9% | 1.69E-08 +/-        | 14.7% |
| Cooling                                                                           | 30.000 d               | 2.60E-01              | 4.44E+07 +/-     | 11.5% | 4.44E-01 +/-        | 11.1% | 2.61E-09 +/-        | 9.7%  |
| Cooling                                                                           | 30.000 d               | 3.42E-01              | 1.68E+07 +/-     | 9.2%  | 3.62E-01 +/-        | 11.9% | 1.65E-09 +/-        | 11.4% |
| Cooling                                                                           | 30.000 d               | 4.24E-01              | 1.37E+07 +/-     | 9.5%  | 3.17E-01 +/-        | 11.9% | 1.42E-09 +/-        | 11.6% |
| Cooling                                                                           | 30.000 d               | 5.07E-01              | 1.19E+07 +/-     | 9.6%  | 2.78E-01 +/-        | 11.9% | 1.24E-09 +/-        | 11.6% |
| Cooling                                                                           | 30.000 d               | 5.89E-01              | 1.05E+07 +/-     | 9.6%  | 2.44E-01 +/-        | 11.9% | 1.09E-09 +/-        | 11.6% |
| Cooling                                                                           | 30.000 d               | 6.71E-01              | 9.19E+06 +/-     | 9.6%  | 2.15E-01 +/-        | 11.9% | 9.56E-10 +/-        | 11.6% |
| Cooling                                                                           | 30.000 d               | 7.53E-01              | 8.07E+06 +/-     | 9.6%  | 1.88E-01 +/-        | 11.9% | 8.40E-10 +/-        | 11.6% |
| Cooling                                                                           | 30.000 d               | 8.35E-01              | 7.09E+06 +/-     | 9.6%  | 1.65E-01 +/-        | 11.9% | 7.38E-10 +/-        | 11.6% |
| Cooling                                                                           | 30.000 d               | 9.17E-01              | 6.23E+06 +/-     | 9.6%  | 1.45E-01 +/-        | 11.9% | 6.48E-10 +/-        | 11.6% |
| Cooling                                                                           | 30.000 d               | 9.99E-01              | 5.47E+06 +/-     | 9.6%  | 1.28E-01 +/-        | 11.9% | 5.69E-10 +/-        | 11.6% |

## B.2 Post-Irradiation Cooling time for Au

The dose rate-cooling time response decay function for Au is:

$$D_R = 6.86142 + 2.4383E06e^{-(t_c/1.8482)} \quad (3)$$



| Table B.2. Predicted Au cooling time after irradiation of 10 mg for 2.61 days. |                        |                       |                    |                     |                     |  |
|--------------------------------------------------------------------------------|------------------------|-----------------------|--------------------|---------------------|---------------------|--|
|                                                                                | Cooling Time<br>(step) | Cumulative<br>(Years) | Activity<br>(Bq)   | Dose rate<br>(Sv/h) | Heat output<br>(kW) |  |
| Irradn                                                                         | 2.610 d                | 7.15E-03              | 6.92E+10 +/- 32.6% | 5.94E+05 +/- 38.5%  | 7.14E-06 +/- 36.2%  |  |
| Cooling                                                                        | 5.000 d                | 1.37E-02              | 1.96E+10 +/- 31.9% | 1.63E+05 +/- 38.4%  | 1.98E-06 +/- 35.8%  |  |
| Cooling                                                                        | 30.000 d               | 9.58E-02              | 1.34E+07 +/- 27.9% | 8.25E+01 +/- 38.0%  | 1.13E-09 +/- 32.2%  |  |
| Cooling                                                                        | 30.000 d               | 1.78E-01              | 1.41E+04 +/- 28.8% | 5.54E-02 +/- 37.1%  | 9.47E-13 +/- 28.5%  |  |
| Cooling                                                                        | 30.000 d               | 2.60E-01              | 1.43E+01 +/- 33.4% | 2.86E-05 +/- 34.5%  | 7.45E-16 +/- 28.5%  |  |
| Cooling                                                                        | 30.000 d               | 3.42E-01              | 4.22E-01 +/- 32.5% | 1.16E-06 +/- 33.1%  | 2.40E-17 +/- 26.9%  |  |
| Cooling                                                                        | 30.000 d               | 4.24E-01              | 1.12E-02 +/- 37.6% | 2.18E-07 +/- 84.1%  | 1.63E-18 +/- 69.5%  |  |
| Cooling                                                                        | 30.000 d               | 5.07E-01              | 3.01E-03 +/- 84.9% | 1.92E-07 +/- 84.9%  | 1.18E-18 +/- 84.9%  |  |
| Cooling                                                                        | 30.000 d               | 5.89E-01              | 2.68E-03 +/- 84.9% | 1.71E-07 +/- 84.9%  | 1.05E-18 +/- 84.9%  |  |
| Cooling                                                                        | 30.000 d               | 6.71E-01              | 2.38E-03 +/- 84.9% | 1.52E-07 +/- 84.9%  | 9.37E-19 +/- 84.9%  |  |
| Cooling                                                                        | 30.000 d               | 7.53E-01              | 2.12E-03 +/- 84.9% | 1.35E-07 +/- 84.9%  | 8.33E-19 +/- 84.9%  |  |
| Cooling                                                                        | 30.000 d               | 8.35E-01              | 1.88E-03 +/- 84.9% | 1.20E-07 +/- 84.9%  | 7.41E-19 +/- 84.9%  |  |
| Cooling                                                                        | 30.000 d               | 9.17E-01              | 1.68E-03 +/- 84.9% | 1.07E-07 +/- 84.9%  | 6.60E-19 +/- 84.9%  |  |
| Cooling                                                                        | 30.000 d               | 9.99E-01              | 1.49E-03 +/- 84.9% | 9.51E-08 +/- 84.9%  | 5.87E-19 +/- 84.9%  |  |

## Appendix C - Defect Size measurements

### C.1 Defect sizes for Au 0.013 dpa

|    | Area | Mean  | Angle  | Length | breadth |
|----|------|-------|--------|--------|---------|
| 1  | 0.3  | 73.1  | -18.4  | 5.5    | 4.3     |
| 2  | 0.3  | 108.1 | -42    | 4.7    | 4.1     |
| 3  | 0.3  | 38.3  | -31.8  | 4.3    | 4.5     |
| 4  | 0.3  | 102.1 | -37.3  | 4.6    | 4.3     |
| 5  | 0.2  | 116.2 | -41.2  | 3.7    | 2.9     |
| 6  | 0.3  | 137.7 | -40.1  | 4.3    | 4.1     |
| 7  | 0.2  | 133.9 | -31    | 3.1    | 3.1     |
| 8  | 0.2  | 108.8 | -36.4  | 4.1    | 4.4     |
| 9  | 0.1  | 129.3 | 69     | 4.4    | 1.6     |
| 10 | 0.2  | 57.6  | -40.2  | 3      | 3       |
| 11 | 0.3  | 153.9 | -37.6  | 5.7    | 4.9     |
| 12 | 0.3  | 78.8  | -34.3  | 4.6    | 5.2     |
| 13 | 0.5  | 112.2 | -37.6  | 7.7    | 6.8     |
| 14 | 0.2  | 102.8 | -30.1  | 3.8    | 3.6     |
| 15 | 0.2  | 100.5 | -41.4  | 4      | 3.3     |
| 16 | 0.2  | 100.1 | -33    | 4.2    | 3.1     |
| 17 | 0.3  | 107.3 | -37.7  | 4.8    | 4.1     |
| 18 | 0.3  | 72.6  | -34.7  | 5.5    | 4.2     |
| 19 | 0.3  | 148.7 | -34.2  | 5.3    | 4.7     |
| 20 | 0.3  | 102.8 | 55.7   | 4.6    | 4.3     |
| 21 | 0.4  | 69.1  | 59.9   | 6.3    | 4.9     |
| 22 | 0.3  | 110.3 | -38.7  | 4.5    | 5.5     |
| 23 | 0.2  | 144.5 | -41.2  | 3.7    | 3.4     |
| 24 | 0.3  | 84.3  | -33.7  | 5.7    | 6       |
| 25 | 0.2  | 165.9 | -38.2  | 3.1    | 4.4     |
| 26 | 0.2  | 97.1  | -40.9  | 3.5    | 2.7     |
| 27 | 0.3  | 119.7 | -35.5  | 4.5    | 4.1     |
| 28 | 0.3  | 100   | -29.7  | 5.6    | 5.2     |
| 29 | 0.3  | 77.5  | -40.4  | 4.6    | 5.3     |
| 30 | 0.2  | 151.9 | -29.1  | 3.6    | 3.2     |
| 31 | 0.2  | 67.3  | -32    | 3.3    | 3.1     |
| 32 | 0.2  | 112.4 | -119.1 | 3.6    | 3.9     |
| 33 | 0.2  | 77.3  | -36.4  | 4.1    | 3.6     |
| 34 | 0.2  | 93.7  | -35.5  | 3      | 2.7     |
| 35 | 0.2  | 61.6  | -35.8  | 3.9    | 4.1     |
| 36 | 0.3  | 94.5  | -31.8  | 4.3    | 4.2     |
| 37 | 0.2  | 85.2  | -40.6  | 3.2    | 2.1     |
| 38 | 0.2  | 97.5  | -39.8  | 2.7    | 2.5     |
| 39 | 0.1  | 103.7 | -32.5  | 2.3    | 1.4     |
| 40 | 0.1  | 83.4  | -36    | 2.4    | 2       |
| 41 | 0.1  | 63.5  | -38.7  | 2.2    | 2.2     |
| 42 | 0.1  | 92    | -52.1  | 2      | 2       |
| 43 | 0.2  | 70.5  | -25.3  | 3.7    | 3.2     |
| 44 | 0.2  | 120.3 | -28.1  | 3      | 3.1     |
| 45 | 0.2  | 140   | -45    | 3.5    | 3.8     |

## C.2 Defect sizes for Lu-Al 0.0034 dpa

|    | Area     | Mean                | Angle              | Length   |
|----|----------|---------------------|--------------------|----------|
| 1  | 6.2      | 96.6                | -38.3              | 12.1     |
| 2  | 6.5      | 80.9                | -40.1              | 12.4     |
| 3  | 9.7      | 112.8               | -48.2              | 18.7     |
| 4  | 7.5      | 106.6               | -49.2              | 14.5     |
| 5  | 7.5      | 119                 | -46.4              | 14.5     |
| 6  | 7.2      | 107.2               | -39.3              | 14.2     |
| 7  | 9.2      | 141.9               | -50.6              | 18.1     |
| 8  | 9.2      | 117.4               | -48.4              | 18       |
| 9  | 6        | 144.7               | -48.6              | 11.3     |
| 10 | 9        | 122.9               | -56.8              | 17.3     |
| 11 | 10.7     | 134.3               | -46.9              | 21.2     |
| 12 | 4.7      | 126.6               | -45                | 9.2      |
| 13 | 9.2      | 124.8               | -38.2              | 17.8     |
| 14 | 4.7      | 131.7               | -49.4              | 9.2      |
| 15 | 6        | 125.2               | -59                | 11.6     |
| 16 | 5.5      | 123.7               | -70.7              | 10.6     |
| 17 | 11.7     | 133                 | -57.3              | 23.1     |
| 18 | 13.2     | 139.6               | -49.6              | 26.2     |
| 19 | 8.2      | 142.6               | -55.3              | 15.8     |
| 20 | 7        | 108.4               | -56.9              | 13.7     |
| 21 | 5.7      | 127.3               | -57.7              | 11.2     |
| 22 | 6.7      | 146                 | 15.6               | 12.9     |
| 23 | 9.5      | 147.6               | 122.8              | 18.4     |
| 24 | 8.2      | 136.4               | 136.3              | 15.9     |
| 25 | 5.5      | 124.5               | 135                | 10.6     |
| 26 | 7.5      | 126.5               | -52                | 14.6     |
| 27 | 5.5      | 125.5               | -54.8              | 10.4     |
| 28 | 6.2      | 123.1               | 90                 | 12       |
| 29 | 5        | 124.6               | 105.5              | 9.3      |
| 30 | 4.5      | 127.7               | 96.7               | 8.5      |
|    |          |                     |                    | 14.44333 |
|    |          |                     | radius             | 7.221667 |
| N  | Area     | Dislocation density | Standard deviation |          |
| 10 | 24736.15 | 0.000161            | 2.81627E-05        |          |
| 7  | 24736.15 | 0.000113            | 2.01162E-05        |          |
| 9  | 24736.15 | 0.000145            | 1.20697E-05        |          |
| 7  | 24736.15 | 0.000113            | 2.01162E-05        | Error    |
|    |          | 0.000133            | 2.01162E-05        | 0.151515 |

## Appendix D-Sample Material's Datasheet

**REACTOR EXPERIMENTS, INC.**  
963 TERMINAL WAY      SAN CARLOS, CALIFORNIA 94070

**MATERIAL DATA SHEET**

Material: 5.2% Lutetium-Aluminum      Thickness: .004 inch      Purity: 99.7838%  
Catalog No.: Special      Diameter: .1016 mm      Lot No.: 529

**CHEMICAL ANALYSIS**

| Impurity | Maximum Percent | Impurity | Maximum Percent | Impurity | Maximum Percent | Impurity | Maximum Percent |
|----------|-----------------|----------|-----------------|----------|-----------------|----------|-----------------|
| Fe       | 0.002           |          |                 |          |                 |          |                 |
| Cu       | 0.003           |          |                 |          |                 |          |                 |
| Ni       | <0.001          |          |                 |          |                 |          |                 |
| Cr       | 0.002           |          |                 |          |                 |          |                 |
| Mn       | 0.001           |          |                 |          |                 |          |                 |
| Si       | 0.20            |          |                 |          |                 |          |                 |
| Mg       | 0.002           |          |                 |          |                 |          |                 |
| Ca       | 0.0002          |          |                 |          |                 |          |                 |
| Ti       | 0.005           |          |                 |          |                 |          |                 |

Foil Code      Weight (grams)      Foil Code      Weight (grams)

# Appendix E - Published Articles

## E.1 Journal of Nuclear Materials

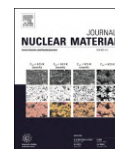
Journal of Nuclear Materials 587 (2023) 154707



Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: [www.elsevier.com/locate/jnucmat](http://www.elsevier.com/locate/jnucmat)



### Calculation of neutron-induced radiation damage to gold and lutetium-aluminium microstructures using MCNP6.2

S.T. Temagee<sup>a,d,\*</sup>, L. Bedhesi<sup>b</sup>, R.D. Mavunda<sup>a,c</sup>, G.C. Daniels<sup>a,b</sup>, S.H. Connell<sup>c</sup>, I.T. Usman<sup>a</sup>, E. Chinaka<sup>b</sup>

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#### ARTICLE INFO

##### Keywords:

Displacement-per-atom (dpa)  
MCNP6.2  
Microstructure  
Neutron  
Radiation damage

#### ABSTRACT

Interaction of particles and photons with materials in extreme radiation environments like the nuclear reactor may lead to basic alterations in the microstructural properties of crystalline solids. These changes accumulate over time into defects on the macrostructure translating to changes in the material's physical and mechanical properties. Studying the level of damage in the materials requires a good prediction of damage using statistical approaches like Monte Carlo simulations. This study aims to calculate the level of damage to Gold (Au) and Lutetium-Aluminium (Lu-Al) due to neutron irradiation, owing to the applications of the materials in reactor technology and other extreme radiation environments. Neutron fluxes, displacement per atom (dpa) rates, and heat deposition expected during irradiation were calculated with Monte Carlo N-particle transport code, MCNP6.2, using the SAFARI-1 research reactor model. The total neutron flux incident on gold and Lutetium-Aluminium was  $2.26 \times 10^{11} \text{ n cm}^{-2} \text{ s}^{-1}$  and  $6.94 \times 10^{12} \text{ n cm}^{-2} \text{ s}^{-1}$  respectively, while the dpa rate in Au and Lu-Al was estimated to be  $1.96 \times 10^{-7} \text{ s}^{-1}$  and  $6.56 \times 10^{-5} \text{ s}^{-1}$ . The calculated neutron dpa rates and fluence for the materials suggest an elevated level of damage to the microstructure of the materials.

#### 1. Introduction

Effects of radiation on crystalline solids have been studied by exposing materials to different irradiation sources, such as high-energy electron/ion beams, fission, and fusion reactors. Defects to the microstructure of materials often occur when they are exposed to high-energy particles and photons, under these harsh irradiation environments [1–3]. Irradiation of materials with neutrons in a fission reactor triggers elastic and inelastic reactions, displacing the Primary Knock-On Atoms (PKA) from their lattice site, which together on further collision with neighbouring atoms, creates interstitial-vacancy pairs called Frenkel Pairs (FP) [4,5].

Neutron irradiation also induces the production of photons which cause additional secondary radiation defects. Sustained exposure of materials to this radiation and the accompanying heat deposition causes dynamical effects (migration, aggregation, complexation, and anneal-

ing) on the microstructure ranging from zero-dimensional (point defects) to three-dimensional defects (voids, cavities, and precipitates). Evolution and diffusion of the Frenkel pairs, therefore form higher order defects on the microstructure which may, over time, translate to significant observable defects in the bulk morphology of crystalline materials [1]. To correlate radiation damage sources and effects, it is essential to provide a means of comparing the data of various effects obtained from different irradiation sources. One of the main parametric quantities for comparing the radiation damage effects on a crystalline or macro-molecular material is the displacement per atom (dpa). It is the ratio of the number of atoms displaced in a given volume to the number of material atoms in the same volume for a given fluence [6,7]. Displacements per atom offer the basis for extrapolating other radiation effects on crystalline materials such as material degradation, radiation-induced effects, radiation phase transformations, and radiation-induced swelling. Increases in dpa values indicate higher damage to materi-

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<https://doi.org/10.1016/j.jnucmat.2023.154707>

Received 28 February 2023; Received in revised form 29 August 2023; Accepted 1 September 2023

Available online 7 September 2023

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## E.2 Journal of Physics: Conference Series

28th International Nuclear Physics Conference (INPC 2022) IOP Publishing  
Journal of Physics: Conference Series 2586 (2023) 012122 doi:10.1088/1742-6596/2586/1/012122

### Evaluation of neutron radiation damage on gold microstructure using MCNP

S T Temaugee<sup>1</sup>, L Bedhesi<sup>2</sup>, R D Mavunda<sup>3</sup>, S H Connell<sup>3</sup>, G C Daniels<sup>2,1</sup> and I T Usman<sup>1</sup>

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**Abstract.** The effects of radiation interaction with materials have been studied over the years on metals, semiconductors, and other alloys. The result of these interactions constitutes microstructural effects, like point defects, dislocation loops, and void swellings. The accumulation of these defects results to damage on the macroscopic scale. This study is aimed to predict the magnitude of radiation damage in gold sample due to neutron irradiation. Neutron flux, displacement-per-atom rate, as well as heat deposition, were calculated in MCNP6.2, using the SAFARI-1 reactor model. The total neutron flux and dpa rate in the gold sample were determined to be  $2.262 \times 10^{11} \text{ n.cm}^{-2}.\text{s}^{-1}$  and  $5.209 \times 10^{-7} \text{ s}^{-1}$  respectively. Also, the total heat deposition due to neutrons and photons was  $2.515 \times 10^{-6} \text{ W.g}^{-1}$  and  $0.513 \text{ W.g}^{-1}$ , respectively. Hence, the predicted neutron dpa and flux for the gold sample in this study suggest a heavy damage regime.

**Keywords:** Displacement-Per-Atom(dpa), MCNP, microstructure, neutron, radiation damage.

#### 1. Introduction

Radiation damage in crystalline materials starts from a single point defect at the microstructure level when a material is exposed to radiation such as neutrons and photons. Radiation effects on the microstructure can over time, translate into observable defects in the form of physical property changes [1]. The formation of interstitial-vacancy (Frenkel) pairs evolves from 0-dimensional (point defects) to 3-dimensional defects (voids, cavities, and precipitates) in the material after exposure to a high dose of radiation. These constitute microstructural damage in crystalline solids [2]. To quantify the level of damage in materials, a radiation exposure quantity is essential as a standard. Universally, the quantity used to correlate displacement damage in materials is known as displacement per atom (dpa), which can be defined as the ratio of the number of atoms displaced to the number of material atoms in the same volume for a given fluence of radiation [3]. It offers the basis for calculating other radiation effects on crystalline materials since radiation-induced changes in the microstructure are a function of the dpa [3,4]. It is also a measure for comparing radiation effects for different types of radiation (neutrons, photons, electrons, ions) quantitatively. Research on the effect of damage in crystalline materials has been conducted for many decades, driven by the need to control the radiation-induced degradation of materials in reactors and the response of the material to harsh

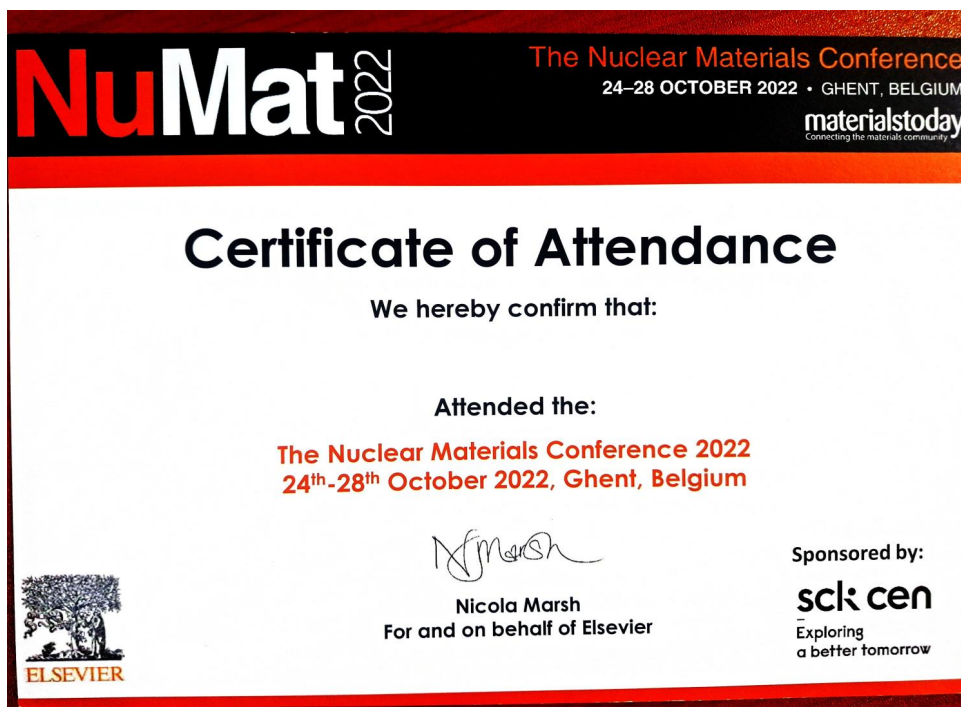
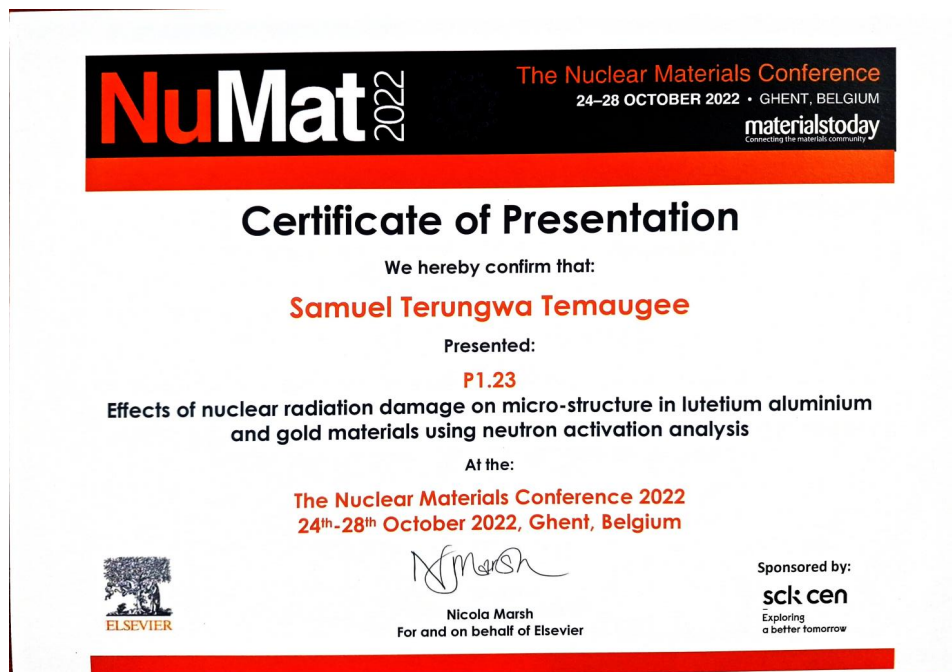


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## Appendix F - Conference Certificates





### Certificate of Attendance

This is to certify that

**Mr Samuel Terungwa Temaugee**

Has attended and participated in the 28th International Nuclear Physics Conference (INPC 2022) in Cape Town South Africa.

  
Faïçal Azalez  
INPC 2022 Chair



  
Mathis Wiedeking  
INPC 2022 Co-Chair



## **CERTIFICATE OF ATTENDANCE**

This is to certify that

*Samuel Terungwa Temaugee*  
*South African Institute of Physics*

attended the **67<sup>th</sup>** annual conference of the South African Institute of Physics

hosted by the University of Zululand

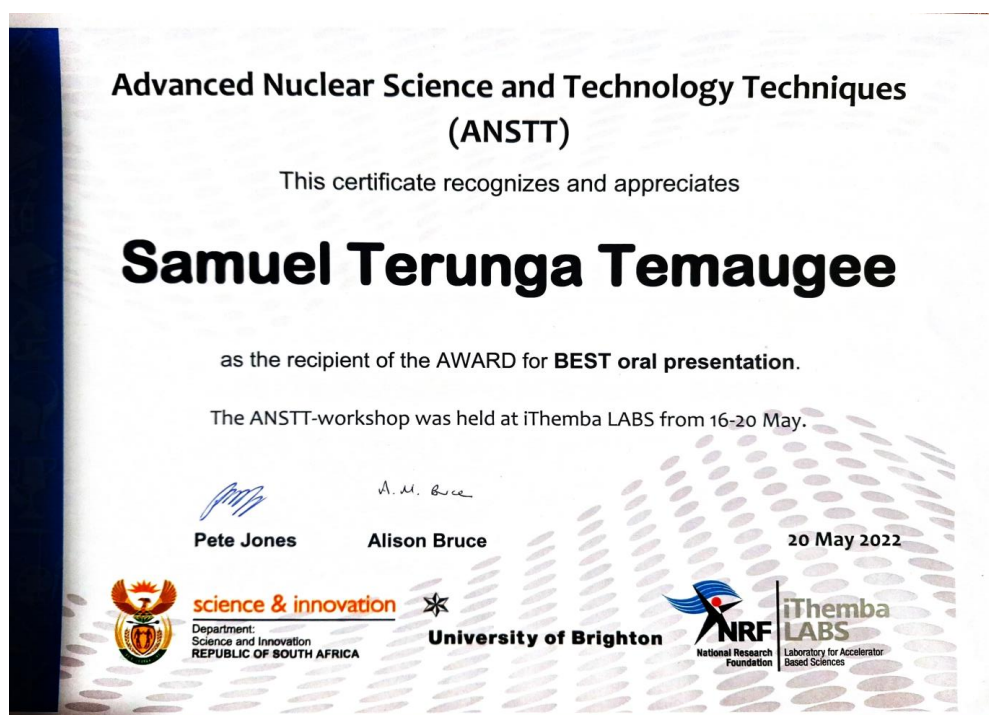
03 – 07 July 2023

A handwritten signature in black ink, appearing to read "Masara", with a stylized initial "B" to the left.

Brian Masara  
**Executive Officer - SAIP**

## Appendix G - Awards obtained from Ph.D. work

### G.1 ANSTT2022-Best Oral Presenter



Link: [https://twitter.com/Ln\\_z](https://twitter.com/Ln_z)

## G.2 SAIP2023-Best Poster Presenter (Applied Physics)

7 July 2023

Dear Samuel

Congratulations on the Applied Physics Division PhD Poster Award for your presentation entitled:  
"Ion-induced radiation damage in Lutetium-Aluminium and Gold using SRIM-2013"

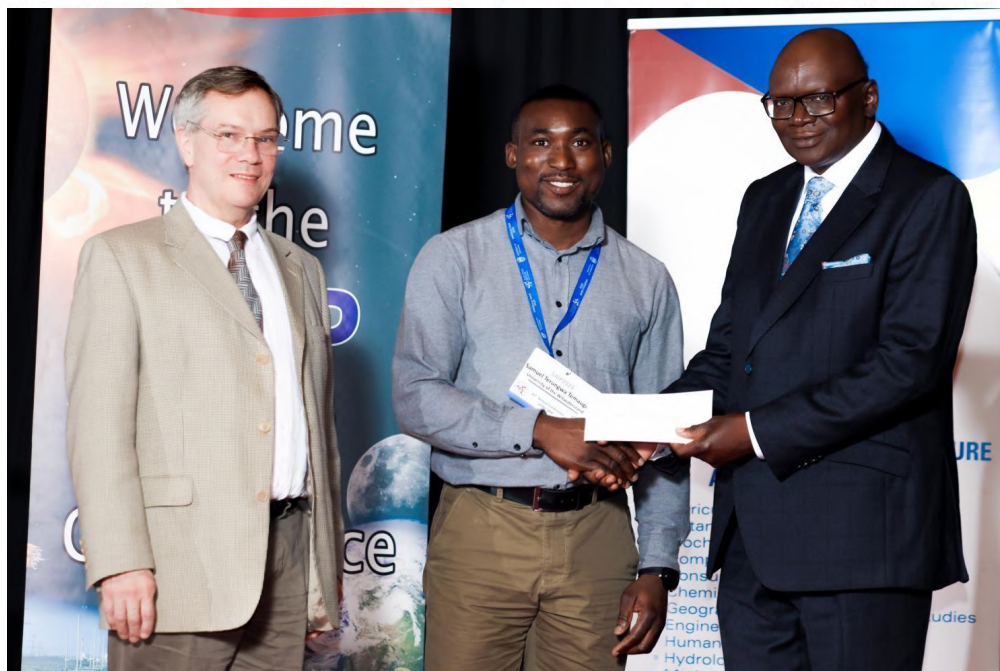
The award value is R 5 000 and will be paid by EFT once you provide the Applied Physics Division with your banking details. Please email the details to Prof van Dyk (ernest.vandyk@mandela.ac.za).

Regards

Prof Alan Matthews



Chair: Applied Physics Division



Link: <https://appliedphysics.co.za/awards/>