

CRITICALITY ANALYSIS FOR A URANIUM STORAGE FACILITY

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DECLARATION

I declare that this research report is my own, unaided work. It is being submitted for the Degree of Master of Science in the University of the Witwatersrand, Johannesburg. It has not been submitted before for any degree or examination in any other university.



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ABSTRACT

Protection against accidental criticality is of utmost importance due to the potential of extremely high doses (such as the 17 Sv in the case of the Toikamura accident) to personnel involved in a criticality accident.

In this study a criticality study is performed on a storage facility for uranium-containing residue. A criticality safety evaluation is the method used to determine the criticality safety of a system. Various internationally accepted methods exist which can be used during a criticality safety evaluation, including the use of a validated calculational technique.

The three-dimensional Monte Carlo code, MCNP, was used in this study to determine multiplication factors, k_{eff} , for several different theoretical storage configurations. The internationally accepted subcritical limit of $k_{\text{eff}} = 0.95$ was used to determine whether a specific configuration is safe in terms of criticality.

This study determines whether the theoretical storage configurations will be safe in terms of criticality, and it also evaluates the influence of various moderators on the multiplication factor, as well as the effect of distance between individual canisters on the multiplication factor.

This criticality safety evaluation aims to show that the proposed theoretical storage configurations are safe in terms of criticality. It also provides recommendations of how the storage capacity can be increased, based on the results of the study.

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TABLE OF CONTENTS

DECLARATION	ii
ABSTRACT	iii
ACKNOWLEDGEMENTS	iv
TABLE OF CONTENTS	v
LIST OF FIGURES	vii
LIST OF TABLES	ix
GLOSSARY	x
CHAPTER 1 – INTRODUCTION	1
1.1 Criticality accidents and the need for safety assessments	2
1.2 Molybdenum production	3
1.3 Hot cell	3
1.4 Conversion from HEU to LEU target plates	6
1.5 Evaluating the design for use with LEU target plates	8
1.6 Nuclear Security	8
1.7 Proposed methodology for the Criticality Safety Evaluation	9
1.8 Research Report structure and chapter outline	10
CHAPTER 2 – THEORETICAL CONSIDERATIONS	12
2.1 Nuclear interactions	12
2.1.1 Scattering	12
2.1.2 Absorption reactions	13
2.2 Nuclear fission	14
2.3 The fission chain reaction	15
2.3.1 The multiplication factor	16
2.4 Criticality Principles	17
2.5 Criticality Control	18
2.5.1 Factors that affect criticality	19
2.5.2 Control principles	25
2.5.3 Criticality data	26
2.6 Anomalies of Criticality	26

2.7	Radiation transport code MCNP	28
2.7.1	A brief description of the Monte Carlo technique	29
2.7.2	Criticality calculations with MCNP	31
CHAPTER 3 – CRITICALITY ANALYSIS FOR STORAGE CELL:		
RESULTS AND DISCUSSION		33
3.1	Neutron transport evaluation using MCNP illustrated with a worked example	33
3.1.1	MCNP input data card organisation	35
3.1.2	Discussion of the input data set for MCNP example	35
3.2	Modelling Assumptions	39
3.2.1	Cellulose composition	39
3.2.2	Water mist/steam moderation	40
3.2.3	Residue canister geometry	40
3.3	Models	41
3.3.1	Model 1	41
3.3.2	Model 2	43
3.4	Modelling Results	44
3.4.1	Model 1	44
3.4.2	Model 2	46
3.5	Discussion of results	48
CHAPTER 4 – RECOMMENDATIONS AND CONCLUSIONS		50
4.1	Conclusions from the calculations	50
4.2	Inputs to the design	50
REFERENCES		53

LIST OF FIGURES

Figure 1.1: Highly shielded hot cells for extremely high activities (source: https://inlportal.inl.gov/portal/server.pt/community/materials_characterization/698/hot_cells/8075).....	4
Figure 1.2: Rear side of a hot cell with a Padirac fitted to the cell door (source: http://www.getinge.com/nuclear/).	5
Figure 1.3: Major ^{99}Mo producers around the world (source: www.nature.com/news/2009/090715/full/460312a.html)	7
Figure 2.2: Liquid Drop Model of fission [DOE93].	14
Figure 2.3: Thermal neutron fission yield of ^{235}U as a function of mass number [www.science.uwaterloo.ca/~cchieh/cact/nuctek/fissionyield.html].....	15
Figure 2.4: The fission chain reaction [http://www.jaymaron.com/nuclear/nuclear.html].....	16
Figure 2.4: How geometry can influence neutron leakage [source: internal Necsa training presentation].	20
Figure 2.5: An increased mass favours the production of neutrons [source: internal Necsa training presentation].	20
Figure 2.7: How storage in close proximity to each other can cause criticality [source: internal Necsa training presentation].	21
Figure 2.8: A container designed to keep a safe distance from another container [source: photo obtained from a colleague who visited Springfields Fuel Ltd.].....	22
Figure 2.9: A moderation safe storage area for canisters containing fissile material [Source: photo obtained from a colleague who visited Springfields Fuel Ltd.].....	23
Figure 2.10: Triple potential for criticality induced by increasing water content between and within uranium storage units (Author's own representation).....	28
Figure 2.11: Flow diagram for an analog Monte Carlo method used to calculate the effective multiplication factor k_{eff} [KNI81].	30

Figure 3.1: Visual representation of the geometry of the MCNP example – side view.....	34
Figure 3.2: Visual representation of the geometry of the MCNP example – top view.....	34
Figure 3.3: MCNP input file structure [BRE09]	35
Figure 3.4: Side and top view of a residue canister.....	40
Figure 3.5: Top and side view of Array D as applied in Model 1.	42
Figure 3.6: Top and side view of an array of 6 x 6 x 13 residue canisters placed a few centimetres (varied from 0 cm to 16 cm) apart as applied in Model 2.	43
Figure 3.7: Graphical representation of Model 1 results.	45
Figure 3.8: Graphical representation of Model 2 results.	47

LIST OF TABLES

Table 3.1:	MCNP example input data set.....	36
Table 3.2:	Arrays in Model 1.	42
Table 3.3:	Model 1 results.....	45
Table 3.4:	Results of the 36 model runs in Model 2.	46
Table 3.5:	Maximum k_{eff} values for Model 2.....	47

GLOSSARY

Criticality Accident	<i>“The release of energy as the result of accidentally producing a self-sustaining fission chain reaction” [MCL00].</i>
Criticality Safety Evaluation	<i>“The analysis and documentation that the fissile material covered by the scope of the evaluation will remain subcritical under both normal and credible abnormal conditions” [DOE07].</i>
Fission products	<i>“Nuclides produced by fission of by subsequent radioactive decay of nuclides formed in this manner” [MCL00].</i>
High Enriched Uranium (HEU)	Uranium enriched to at least 20% ^{235}U by weight.
Low Enriched Uranium (LEU)	Uranium enriched up to 19.999% ^{235}U by weight.
Proliferation	The spread of biological, chemical and/or nuclear weapons, and their delivery systems.
Subcritical limit	<i>“The limited value assigned to controlled parameter that results in a system known to be subcritical” [ANS75].</i>
Thermal energy	The energy level at which neutrons are in equilibrium with the thermal motion of surrounding materials.
Thermal neutron	Neutrons with very low kinetic energy levels because they are in equilibrium with the thermal motion of surrounding materials.

CHAPTER 1

INTRODUCTION

The word “nuclear” often brings to mind visions of exploding bombs and the resulting mushroom clouds. However, most of the general public is totally oblivious to the difference “nuclear” makes daily in the lives of many patients undergoing life-saving diagnostic or therapeutic procedures in the field of nuclear medicine. As such NTP Radioisotopes SOC Ltd (a subsidiary of Necsa) has evolved into a very important global player in the nuclear medicine business. The most important isotope they produce is Molybdenum-99, ^{99}Mo . This is used as the “parent” in a generator that results, after decay, in Technetium-99m, $^{99\text{m}}\text{Tc}$.

The $^{99\text{m}}\text{Tc}$ is used mostly for bone scans, but is also used to assess cardiac function and for functional brain imaging. Other uses include imaging of the liver, kidney, gall bladder and various other organs.

The radioisotope $^{99\text{m}}\text{Tc}$ is the one most widely used in medicine, employed in approximately 80% of all nuclear medicine procedures - 70 000 every day in the USA alone [SNM08]. It is a metastable nuclear isomer of ^{99}Tc and it has almost ideal characteristics for a nuclear medicine scan, amongst which are:

- it has a short half-life of six hours, which allows sufficient time for scanning procedures, yet it is short enough to keep the radiation exposure to the patient low;
- $^{99\text{m}}\text{Tc}$ decays by emitting γ - rays with a characteristic energy of 140 keV and since there is no high-energy β - emission, the radiation dose to the patient is low and
- the low energy γ - rays it emits easily escape the human body and are easily detected by a γ -ray camera, minimising the radiation dose to the patient.

There are, however, certain risks involved with the production of ^{99}Mo . One of which is the risk of inadvertent criticality, the other the perceived proliferation risk.

1.1 Criticality accidents and the need for safety assessments

A criticality accident occurs when sufficient fissile material to sustain a nuclear chain reaction is unintentionally brought together. For example, when the nucleus of ^{235}U splits, two or three neutrons are released, and each can cause another nucleus to split. However, if the total mass of the ^{235}U is insufficient to sustain a chain reaction, the neutrons simply escape.

There are two different types of criticality accidents: process accidents and research reactor accidents.

In non-reactor nuclear facilities, administrative or engineered controls are usually in place to prevent criticality from occurring, but fail (process accidents). In research reactor accidents, criticality is achieved intentionally, under conditions that are strictly controlled, but it ends up getting out of control [MCL00].

[MCL00] studied 60 reported criticality accidents since 1945. Although most of these accidents did not lead to fatalities, there have been 21 deaths from criticality accidents. Nine of these fatalities resulted from accidents in non-reactor nuclear facilities, and 12 were due to research reactor accidents.

On 17 June 1997 a criticality accident occurred at the Russian Federal Nuclear Centre (formerly known as Arzamas-16) in the town of Sarov. The accident happened while an experienced technician was busy with routine work on a critical assembly. The accident was immediately recognised and the victim received prompt medical attention, but he died 66 hours later [IAE01].

On 30 September 1999 a criticality accident occurred at the Tokai nuclear fuel plant in Tokaimura, Japan. The accident happened while three workers were preparing a batch of fuel. The chain reaction continued intermittently for

approximately 20 hours. The workers were exposed to doses of up to 17 Sv, causing severe radiation sickness. The worker exposed to the highest dose died 12 weeks later. The worker exposed to the second highest dose of 6 - 10 Sv died another 7 months later. The plant's operating licence was revoked early in 2000 [HOR09].

World-wide the occurrence of nuclear criticality accidents has caused processing and manufacturing facilities to be out of operation for extended periods of time for investigation purposes. In some instances, the affected facilities have never returned to production.

Criticality accidents, such as those mentioned above, can put a country's entire nuclear enterprise under scrutiny.

1.2 Molybdenum production

The first step in the production of ^{99}Mo is irradiating the enriched uranium target plates in a high neutron flux nuclear reactor, such as the 20 MW Oak Ridge-type SAFARI-1 reactor at Necsa. The resulting fission products are a number of radionuclides of different half-lives and energies, including the radioisotope of interest (in this case ^{99}Mo).

The irradiated target plates are then transferred to a hot cell for further processing in order to separate the isotope of interest. The target plates containing fission-products are dissolved, which effects separation of the fission products into two streams, the ^{99}Mo - and (enriched) U-streams. The uranium residue from the production is then stored in a dedicated storage hot cell.

1.3 Hot cell

A hot cell is specially designed to minimise radiation exposure and to prevent the uncontrolled spread of radioactive contamination. The word "hot" refers to

radioactive. The main design objective for a hot cell is to provide and maintain confinement and shielding.

Since the viewing of operations within the hot cell is necessary, hot cell design includes the use of leaded glass shield-windows. The window should provide the same attenuation as the other parts of the hot cell shielding. Windows are sized and installed to give wide ranges of view through the cell, although optical distortion may occur at extreme viewing angles. Figure 1.1 shows a series of hot cells, each equipped with a viewing window and two remote manipulators.



Figure 1.1: Highly shielded hot cells for extremely high activities (source: https://inlportal.inl.gov/portal/server.pt/community/materials_characterization/698/hot_cells/8075).

Hot cells are equipped with remote handling master-slave manipulators for processing radioactive materials as shown in Figure 1.1. The application of hot cells is based on the principle that radiation protection is the cheapest and most effective if the shielding is closest to the radiation source [IAE04].

Hot cells are generally made of stainless steel or ordinary mild steel covered by a good-quality epoxy resin paint (containment box). Operations are carried out through one of the larger faces by using manipulators. Hot cells have to be shielded against gamma radiation. Various types of shielding material can be used, such as concrete, lead, lead-glass and depleted uranium. The thickness of the shielding is calculated based on the type, energy and activity of the radiation in order to keep doses to the operating personnel As Low As Reasonably Achievable (ALARA).

The rear of the cell is fitted with a door through which products, waste and consumables can enter/exit the cell. A transfer system, such as the Padirac by Getinge La Calhène, can be used to move items into or out of the cell without breaking the containment and with full protection against radiation (see Figure 1.2).



Figure 1.2: Rear side of a hot cell with a Padirac fitted to the cell door (source: <http://www.getinge.com/nuclear/>).

The hot cells also need to maintain negative pressure to prevent radioactive contamination escaping to the environment. The hot cell should be leak tight. Ventilation is used to provide dynamic containment in addition to the static containment provided by the containment box. A system of dynamic containment is where an area with higher contamination levels is kept at a lower pressure than

their surroundings. Air from a volume, therefore, always has a tendency to flow towards a volume with higher contamination levels, thus preventing the spread of airborne contamination to less contaminated volumes. The inlet air and the exhausted air should pass through HEPA (High Efficiency PARTICULATE) filters to prevent the introduction of dirt and exhaust of any contaminated particles.

1.4 Conversion from HEU to LEU target plates

In 2007, the International Physicians for the Prevention of Nuclear War (IPPNW) have launched the International Campaign to Abolish Nuclear Weapons (ICAN). The goal of ICAN is to “*reawaken public concern about the growing threat posed by nuclear weapons, and to mobilize civil society to demand a nuclear-weapon-free world through the negotiation and adoption of a Nuclear Weapons Convention*”. [WIL07].

IPPNW doctors have written about HEU: “*How many doctors are aware that they contribute to a nuclear proliferation and terrorist risk every time they order a nuclear bone or lung scan, or other procedure utilising an isotope produced using HEU, directly usable in a nuclear weapon? Almost certainly very few. How many of the patients involved are aware of this? Probably even fewer.*” [WIL07].

Although statements such as the above are usually made to shock the public, it is very successful in affecting the layman’s opinion about the nuclear industry negatively. The concerns over nuclear weapons proliferation have also led a drive to reduce and eliminate international commerce in HEU.

The majority of the radioisotopes used in diagnostic medical procedures are currently produced using HEU. The major competitors in the international medical isotope marketplace (as shown in Figure 1.3) provide more than 95% of the global supply, from only five different reactors: Canada’s NRU, the Netherlands’ HFR, Belgium’s BR-2, France’s Osiris and South Africa’s SAFARI-1. Australia’s Opal reactor, which started up in 2007, has the capacity to produce up to half the world’s ^{99m}Tc demand but at present has only a small molybdenum processing facility and supplies only domestic demand.

In July 2005, the South African Department of Minerals and Energy (DME) authorised the conversion to LEU of the South African Research Reactor (SAFARI-1) and the associated fuel manufacturing at Pelindaba [PIA06]. In 2008, SAFARI-1 has successfully been converted to LEU fuel, and NTP is in the process of converting to LEU target plates although they are currently using both HEU and LEU target plates.



Figure 1.3: Major ^{99}Mo producers around the world (source: www.nature.com/news/2009/090715/full/460312a.html).

The decision to convert from HEU to LEU target plates for molybdenum production was motivated based on two considerations. The first is based on the international initiatives, such as the US Global Threat Reduction Initiative and Reduced Enrichment for Research and Test Reactors (RERTR) Programme, to reduce and eventually remove international commerce in HEU. These initiatives are widely supported internationally and acceptable methods are being developed by various institutions for the production of ^{99}Mo using such target plates.

A second reason for converting is based on the fact that uranium enrichment activities were stopped in South Africa in the early 1990s and given the complexities of international procurement, Necsa and NTP will have to revert to alternative materials for the fuelling of SAFARI-1 and its target plate

requirements. SAFARI-1 has successfully been using LEU fuel since 2009, and NTP is currently in the process of converting to LEU target plates.

1.5 Evaluating the design for use with LEU target plates

In order to design a storage facility for uranium containing residue, a few important factors need to be taken into consideration. The first two aspects, shielding and confinement, are achieved by using a hot cell. The third important function is criticality prevention. With the change-over from HEU target plates to LEU target plates, the volume required for storage is increased, because of the lower enrichment grade. Since the total amount of uranium will increase with the lower enrichment grade, the storage capacity will fill up much faster.

The purpose of this Criticality Safety Evaluation is to provide input into the design of the storage facility, and to determine a first order estimation of the amount of residue canisters that can be safely stored in the storage hot cell.

1.6 Nuclear Security

[BRO10] describes the three-part strategy President Obama presented in April 2009 in order to address international nuclear threats. This strategy entails:

- *“proposing measures to reduce and eventually eliminate existing nuclear weapon arsenals”*;
- *“strengthening the Non-Proliferation Treaty (NPT)”*; and
- *“preventing terrorists from acquiring nuclear weapons or materials”*.

The International Atomic Energy Agency (IAEA) has developed four potential nuclear security risk categories [BRO10], namely:

- *“theft of a nuclear weapon”*;

- “the acquisition of nuclear materials for the construction of nuclear explosive devices”;
- “the malicious use of radioactive sources, including in so-called dirty bombs”; and
- “the radiological hazards caused by an attack on or sabotage of a facility or transport vehicle”.

The Nuclear Security Summit (NSS) has been established as global summit with the objective of preventing nuclear terrorism around the world. The first summit was held in Washington, D.C. in 2010.

The 2014 Nuclear Security Summit will be the third and will be attended by a South African delegation.

The Department of International Relations and Cooperation said “*South Africa’s participation in the NSS process is informed by the shared vision of a world free of weapons of mass destruction, and in particular nuclear weapons. The 2014 Nuclear Security Summit takes place within this broader framework of South Africa’s triple objectives of nuclear disarmament, nuclear non-proliferation, and the peaceful uses of nuclear energy*” [DIR14].

In the interest of South Africa’s commitment to nuclear security, this report will give no details of the storage location, enrichment grade, mass of uranium or residue composition. In addition, the theoretical storage configuration will be modelled.

1.7 Proposed methodology for the Criticality Safety Evaluation

In order to establish that a proposed system or process will be subcritical under normal operating conditions and under postulated abnormal conditions, it is first necessary to establish acceptable subcritical values for the operation and then show that the proposed operation will not exceed those values.

There are currently four acceptable methods for the establishment of these values [DOE07]:

- *“by reference to national standards that present Subcritical Limits;*
- *by reference to widely accepted handbooks of Subcritical Limits;*
- *by reference to experiments with appropriate adjustments to ensure subcriticality when the uncertainties of parameters reported in the experiment documentation are considered; and*
- *by validated calculational techniques”.*

Examples of calculational methods are:

- a Monte Carlo code, such as MCNP,
- transport theory codes, such as ANISN and
- hand calculations.

Due to the limited criticality data in the intermediate enrichment range (~6% to ~93% ^{235}U) from which subcritical limits for criticality control can be deduced, the chosen method for the present study is the use of a calculational technique. More specifically, the Monte Carlo code MCNPX 2.6.0 will be used for all calculations [RSI08].

1.8 Research Report structure and chapter outline

The layout of this Research Report is as follows:

- Chapter 2 describes the theoretical considerations applicable to nuclear criticality events and nuclear criticality safety,

- Chapter 3 gives more detail on the MCNP model built specifically for the criticality safety evaluation and also includes a presentation of the results, while
- Chapter 4 presents the conclusions and recommendations.

CHAPTER 2

THEORETICAL CONSIDERATIONS

Of all the potential radiation hazards with which a Radiation Protection Specialist deals, that of an accidental supercriticality is among the most serious according to Cember [CEM96].

In this chapter the theoretical considerations applicable to nuclear criticality events and nuclear criticality safety is addressed. It includes concepts such as nuclear interactions, nuclear fission, criticality principles, the nuclear fission chain reaction, criticality control, control principles, anomalies of criticality and the MCNP code.

2.1 Nuclear interactions

Neutrons are electrically neutral and are not affected by the electrons in an atom or the positive charge of a nucleus. Therefore, a neutron passes through the electron cloud of an atom and interacts directly with the nucleus. The different interactions that a neutron can have with a nucleus are described below.

2.1.1 Scattering

A neutron scattering reaction occurs when a neutron collides with a nucleus, and emits a single neutron. Usually the incident neutron and scattered neutron are not the same, but the net effect of the reaction is as if the projectile neutron had merely scattered from the nucleus. The two categories of scattering reactions, elastic and inelastic scattering, are described in the following paragraphs.

2.1.1.1 Elastic scattering

In an elastic scattering reaction between a neutron and a target nucleus, kinetic energy and momentum are conserved. This type of reaction is often described as a “billiard ball” type reaction. Elastic scattering is the most likely interaction

between fast neutrons (energy of up to 1 MeV) and low-atomic numbered absorbers (such as water and carbon). This interaction is denoted by (n, n) .

2.1.1.2 Inelastic scattering

In inelastic scattering, the incident neutron is captured by the nucleus, forming a compound nucleus. The kinetic energy of the neutron is transferred to the nucleus, leaving the nucleus in an excited state, and emitting a neutron of lower kinetic energy. The excited nucleus is de-excited by emitting one or more γ -rays. Generally the energy at which the first excited state is found decreases with increasing mass number. This reaction is abbreviated by the symbol (n, n') .

2.1.2 Absorption reactions

When the interaction of a neutron with a nucleus results in the loss of the neutron, it is known as an absorption reaction. The original neutron is absorbed by the nucleus and a charged particle or γ -ray is emitted. Radiative capture, charged-particle reactions, and fission are all absorption reactions and are described in the paragraphs below.

2.1.2.1 Radiative capture

In radiative capture the incident neutron is absorbed by the target nucleus forming a compound nucleus. The excited compound nucleus then decays to its ground state by emitting γ -rays. This interaction is denoted by (n, γ) . These reactions can take place at all neutron energies.

2.1.2.2 Charged-particle reactions

In this type of reaction, the incident neutron is absorbed by the target nucleus forming a compound nucleus. The compound nucleus is excited to a high energy level causing it to emit a charged particle. Such reactions can be abbreviated by (n, α) , (n, p) and $(n, 2n)$. The cross-sections for these reactions usually have threshold energies from 5 to 10 MeV.

2.1.2.3 Fission

When a neutron collides with certain nuclei, the nucleus absorbs the neutron and splits into two similarly sized parts. Fission will be discussed in more detail in the next paragraph.

2.2 Nuclear fission

The fission process is often described by considering the nucleus as a liquid drop. In the liquid drop model, it is assumed that nuclear forces hold the nucleus together in the same way that a drop of water is held together by the molecular forces that tend to make the drop spherical in shape.

The liquid drop model for fission is illustrated in Figure 2.1. The nucleus in the ground state is represented as a sphere or a liquid drop (A). When an incident neutron is absorbed by the target nucleus, a compound nucleus is formed.

The energy imparted on the compound nucleus causes the sphere to become distorted, and, under certain conditions, to become dumbbell-shaped (B). If given sufficient energy, the dumbbell shape splits at the neck into two similar sized fragments (C) and several neutrons.

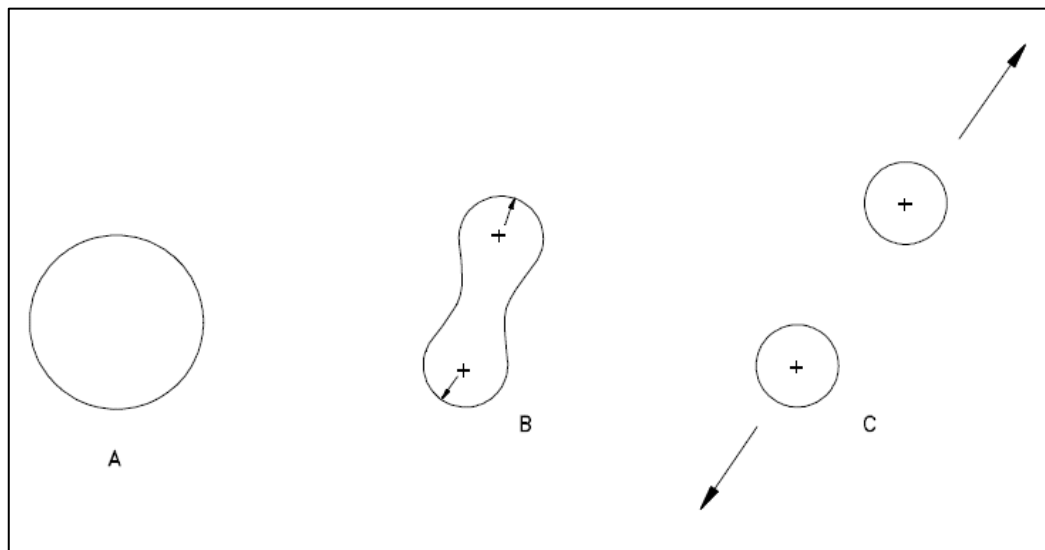


Figure 2.1: Liquid Drop Model of fission [DOE93].

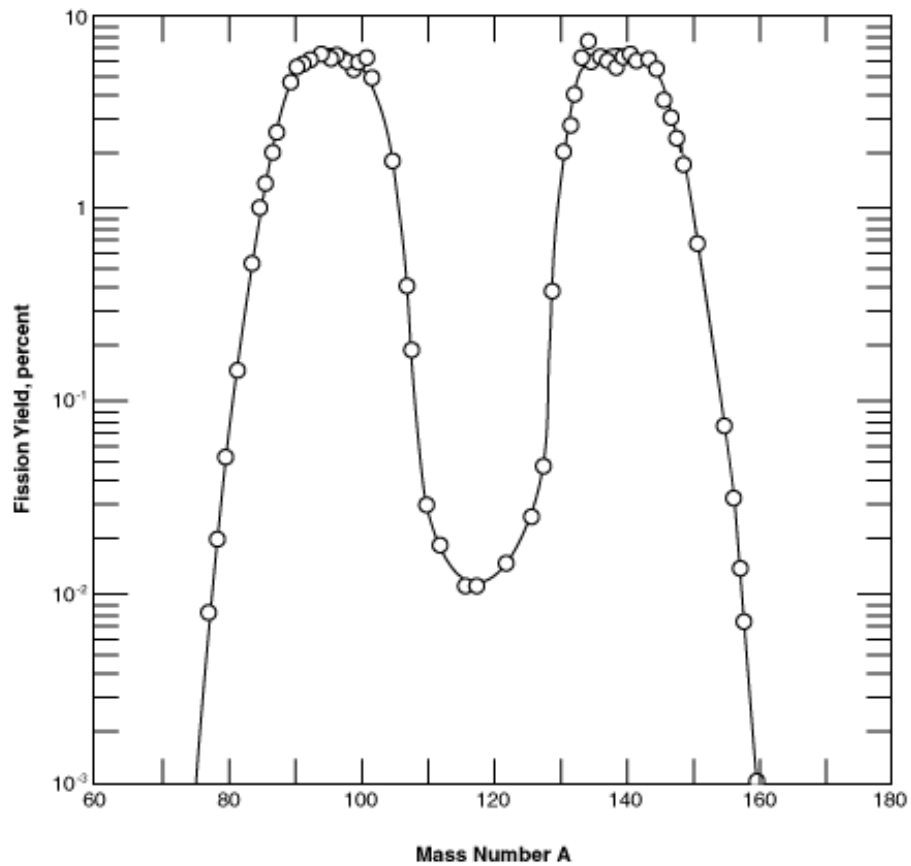


Figure 2.2: Thermal neutron fission yield of ^{235}U as a function of mass number [www.science.uwaterloo.ca/~cchieh/cact/nuctek/fissionyield.html].

One would expect that a fissioning nucleus should split more or less in half [LAM01]. However, fission is usually asymmetric with fission products falling into two definite groups, one light group with mass number of around 95u and a heavy group with mass number of around 140u. A uranium nucleus can split in many ways, yielding over 80 primary fission products. Mass numbers of the fission products vary between 72 (Zn) and 160 (Tb). This is illustrated by Figure 2.2 which shows the fission yield as a function of mass number.

2.3 The fission chain reaction

According to Cember [CEM96] “criticality may be defined as the attainment of physical conditions such that a fissile material will sustain a chain reaction”.

According to Lamarsh [LAM01], a fission chain reaction (see Figure 2.3) happens when “neutrons emitted by fissioning nuclei induce fissions in other fissile or fissionable nuclei; the neutrons from these fissions induce fissions in still other fissile or fissionable nuclei; and so on”.

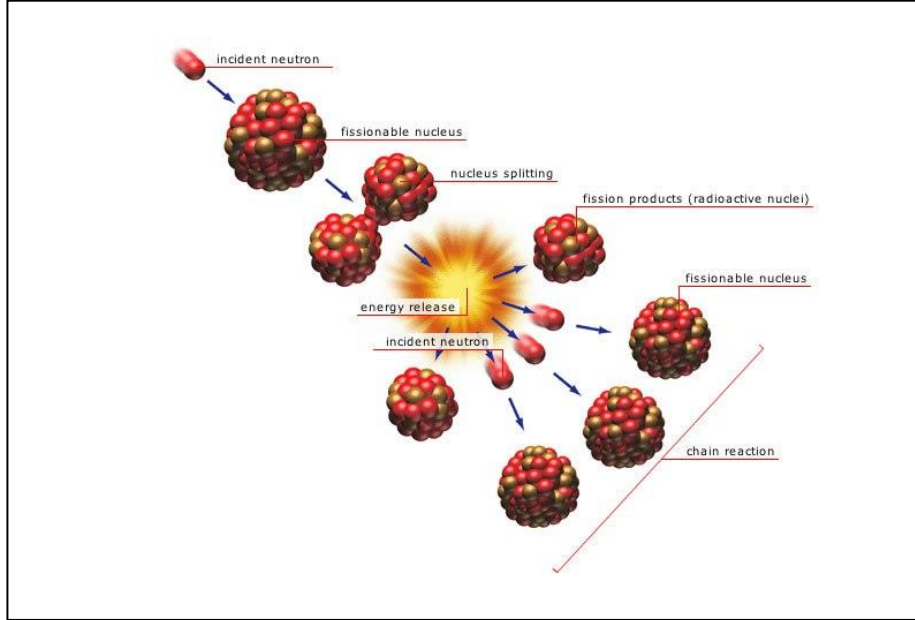


Figure 2.3: The fission chain reaction
[\[http://www.jaymaron.com/nuclear/nuclear.html\]](http://www.jaymaron.com/nuclear/nuclear.html).

2.3.1 The multiplication factor

Not all of the neutrons produced by fission will cause new fissions. Some neutrons will be absorbed without causing fission, and some will leak out of the system. In order to maintain a chain reaction, each nucleus that is split, should produce at least one neutron that causes another fission. This ratio is expressed in terms of a multiplication factor:

$$k_{\text{eff}} = k = \frac{\text{number of neutrons in one generation}}{\text{number of neutrons in the preceding generation}} \quad (2.1)$$

Alternatively,

$$k_{\text{eff}} = k = \frac{\text{neutron production rate}}{\text{neutron loss rate}} \quad (2.2)$$

When k_{eff} is greater than 1, the system is described as supercritical. The number of fissions increases with time, because the rate of neutron production is greater than the rate at which neutrons are absorbed or leak out of the system.

When k_{eff} is less than 1, the system is described as subcritical. The number of fissions decreases from generation to generation, because the rate of neutron absorption and leakage is greater than the rate of neutron production.

When k_{eff} is equal to 1, the chain reaction proceeds at a constant rate, because the rate of neutron production is equal to the rate of neutron absorption and leakage. Such a system is said to be critical.

The value of k_{eff} can be seen as an index of criticality. The internationally accepted limit to ensure that a system remains subcritical is $k_{\text{eff}} \leq 0.95$ [VEN03].

In a nuclear reactor, the system is designed to obtain a chain reaction in a controlled manner. However, outside of a nuclear reactor one would want the system to remain subcritical. Criticality outside of a nuclear reactor is an accident situation, since it would be uncontrolled. It is caused by the building up of enough fissile material to cause a nuclear chain reaction.

2.4 Criticality Principles

Meggitt [MEG06] describes the fission process in some detail. When a uranium nucleus fissions, an average of 2.4 neutrons are emitted with an average energy of 2 MeV. There is one of three things that could happen with each neutron:

- it can collide with a nucleus through inelastic scattering,
- it can be absorbed by a nucleus or
- it can cause another fission.

With natural uranium, about 80% of neutrons will cause inelastic scattering with a ^{238}U nucleus. Almost all of the other neutrons will cause fission events in ^{238}U

with only approximately 0.3% causing fission events in ^{235}U . These fission events will not produce enough neutrons to sustain a chain reaction. The emitted neutrons will lose a substantial portion of their energy and most of these neutrons will cause further inelastic collisions. Neutron-induced fission of ^{238}U will not happen because the neutron energy is now below the threshold for fission of ^{238}U . The fission cross-section for ^{238}U only becomes appreciable at energies in excess of 1.3 MeV.

About 15% of the neutrons will now cause ^{235}U fission events. This is still not enough fission events to sustain a chain reaction, and the dominant effect is that the neutron continues to lose its energy by inelastic scattering collisions. When the energy falls below 1 keV, most of the neutrons are absorbed by the ^{238}U nucleus (radiative capture). The few neutrons that do escape capture, reach an energy region where ^{238}U fission is the most likely fate. Due to the small amount of ^{235}U , a chain reaction can, however, not be sustained.

If the uranium is enriched, the probability of ^{235}U fission becomes equal to that of the inelastic scattering, and sustainability of a chain reaction now becomes a possibility.

When neutrons collide with moderators, they lose a high proportion of their energy in each collision. The energy falls until it is similar to the thermal energy of the nuclei of the moderator. In this region the probability of fission of ^{235}U is at its highest and a chain reaction becomes possible (even with natural uranium). Moderators are always a low-mass-number material with low neutron absorption characteristics and a high scattering cross-section [LAM01]. Common materials present in facilities that could cause moderation (even though not their primary function) are: water, oil, polythene, carbon and paint.

2.5 Criticality Control

Nuclear criticality safety has been defined as “*the prevention or termination of inadvertent nuclear chain reactions in non-reactor environments*” [ANS75].

It has also been defined as the “*art of avoiding a nuclear excursion*” by Paxton [PAX72].

Another definition states that criticality safety may be considered “*protection against the consequences of a nuclear excursion*” [PAX72], or “*protection against the consequences of an inadvertent nuclear chain reaction, preferably by prevention of the chain reaction*” [ANS83].

Out of the above, it is clear that prevention of criticality in a non-reactor scenario is the ultimate goal. The use of the word “art” also shows that criticality prevention is not totally scientific. Criticality is highly dependent on human factors, i.e. the actions by operators and supervisors. The definitions also show the importance of proper design to provide sufficient mitigation of the consequences of a criticality excursion.

2.5.1 Factors that affect criticality

It is possible to control the neutron economy of a system containing fissile material. The factors that allow for control of the neutron economy are listed and discussed below [WIL96].

2.5.1.1 Geometry

The shape of systems containing fissile material can be designed so that the likelihood of collisions is significantly reduced.

For example, a system may be required for operational reasons to be a flat slab. The slab can be made sufficiently thin so that it makes no difference how much fissile material is present in the system. A sufficient fraction of neutrons will escape and a criticality cannot occur. The same amount of fissile material is therefore safer in a shape with a large surface area (such as a slab) than in a small compact shape such as a cube or a sphere.

Figure 2.4 shows how fewer neutrons are available for fission in the “flat” geometry than in the cube geometry, resulting in a lower k_{eff} .

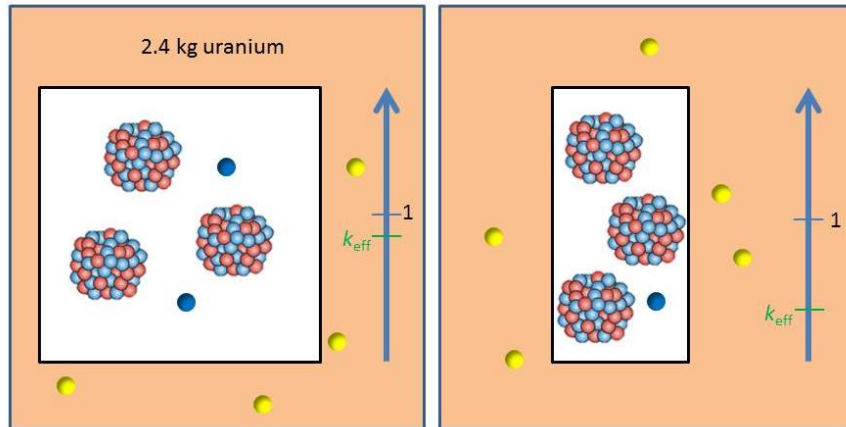


Figure 2.4: How geometry can influence neutron leakage [source: internal Necsa training presentation].

2.5.1.2 Mass

If there are only a few fissile atoms in a given location, most of the neutrons from those which undergo fission would be able to escape the system. If there is a significant mass of fissile material, then a neutron will have a high probability of being captured and causing fission (see Figure 2.5). The minimum mass at which a criticality can occur is called the “critical mass”.

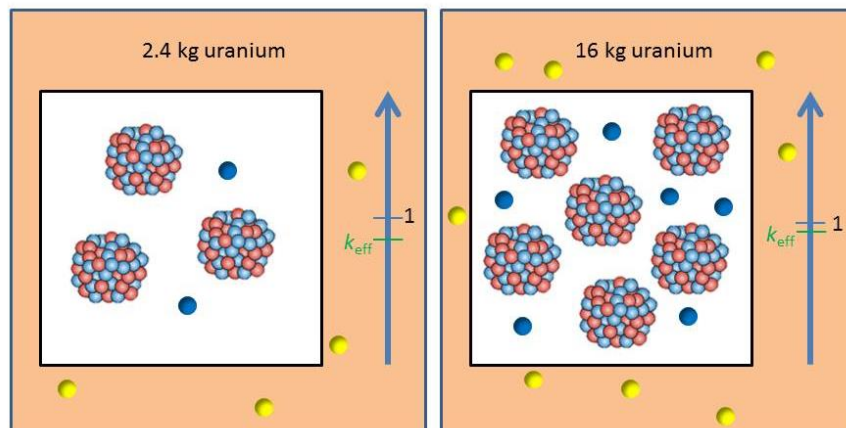


Figure 2.5: An increased mass favours the production of neutrons [source: internal Necsa training presentation].

2.5.1.3 Density

It follows from the discussion of mass that the density of nuclei is important (this refers to uranium density and not material density). The closer packed the nuclei are, the greater the likelihood of collisions with the neutrons and, therefore, the higher the potential to sustain a chain reaction.

2.5.1.4 Interaction

Fissile material in one container cannot distinguish between neutrons generated within it and neutrons generated in another container of fissile material. Storage of two or more containers of fissile material in close proximity of each other can, therefore, increase the probability of a criticality occurring, as shown in Figure 2.6.

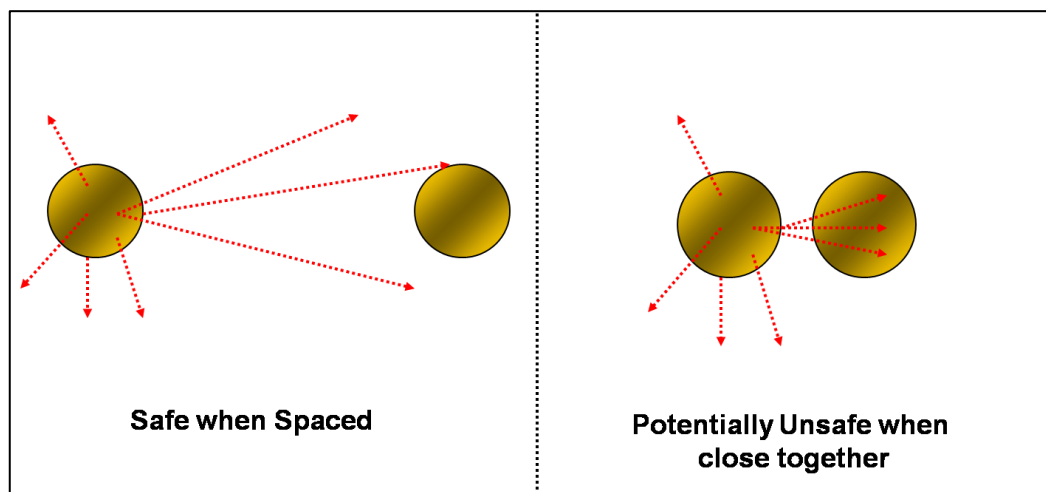


Figure 2.6: How storage in close proximity to each other can cause criticality [source: internal Necs training presentation].

By way of example, vessels can be designed with spacer rings in order to reduce interaction with other vessels as shown in Figure 2.7.



Figure 2.7: A container designed to keep a safe distance from another container [source: photo obtained from a colleague who visited Springfields Fuel Ltd.].

2.5.1.5 Moderation

Neutrons which move relatively slowly through fissile material have a greater fission cross section (such as 583 barn for ^{235}U) than neutrons that are moving more rapidly (such as 1 barn for ^{235}U). The slowing down of neutrons is known as moderation and is used to produce controlled criticality in nuclear reactors. The simplest method of slowing neutrons down is to increase the potential for them to strike non-fissile and non-absorbing nuclei with a small atomic mass. Such collisions may be regarded as elastic scattering impacts between spheres, and the nearer the masses of the colliding spheres, the more effective is the transfer of momentum. It follows that the most effective nuclei in slowing neutrons down are the lighter ones, such as hydrogen. This is why water is considered to be one of the most dangerous substances to associate with fissile material without due precautions.

Figure 2.8 shows an example of how a storage area can be designed to limit possible moderation. The following precautions are taken – there are no water or steam lines in the area, fire fighting with water is not permitted, drums are lidded and branded, drums are off the floor and the roof is watertight.



Figure 2.8: A moderation safe storage area for canisters containing fissile material [Source: photo obtained from a colleague who visited Springfields Fuel Ltd.].

Other common moderating materials are oil, polyethylene, carbon and paint.

2.5.1.6 Concentration

Even where fissile material is dispersed in a moderator (e.g. dissolved in water) it is possible for the concentration of the fissile material to be so low that criticality is not possible, because the neutrons are more likely to be captured by non-fissile than by fissile material.

2.5.1.7 Poisons

A poison, in the context of criticality control, is a material that absorbs neutrons, thus reducing its chance of colliding with fissile nuclei. Criticality can, therefore,

be controlled by mixing poisons with the fissile material. The most effective poisons are considered to be boron and cadmium.

2.5.1.8 Reflection

If a moderating material is placed around a piece of fissile material, some of the neutrons that would have escaped will be reflected back into the fissile material. This process is called reflection. The effect is similar to that of interaction. The properties that make a material a good moderator also make for a good reflector. However, a good reflector is not necessarily a good moderator (e.g., iron is a good reflector but also a poison). Good reflectors are graphite, beryllium, water and natural uranium.

2.5.1.9 Enrichment

Uranium consists of two main isotopes: ^{235}U which is fissile and can cause criticality if present in sufficient quantities and ^{238}U which is not fissile. In natural uranium there is only about 0.72% ^{235}U [LAM01] which makes criticality almost impossible, since the mass of ^{235}U is very small. The ^{238}U has a variety of properties. It is slightly fissionable and may undergo fission as a result of fast (but not slow) neutron absorption. It may also absorb neutrons without undergoing fission. Hence, the probability of fission resulting from neutron capture by ^{238}U nuclei tends to be low, especially in unmoderated systems. ^{238}U also acts as a reflector, as a separator between ^{235}U nuclei affecting the fissile density of the system, and has some effect as a poison. When the uranium is enriched, the amount of ^{235}U is increased and criticality becomes possible.

2.5.1.10 Inhomogeneity

A homogeneous system is one where all the nuclides are evenly distributed throughout the system (such as in a solution). An inhomogeneous system is one where the fissile material is physically separate from the other parts of the system (for example in a reactor where fissile material in the rods is physically separated from the moderator). At a high ^{235}U enrichment a homogeneous uranium salt solution is more reactive than any comparable inhomogeneous system. At

enrichment close to natural uranium, however, an inhomogeneous system may be more reactive than a comparable homogeneous one.

2.5.2 Control principles

It is generally accepted that nuclear safety should depend as little as possible on decisions and actions of operating personnel, especially for routine operations. The [ANS83] recommends that, where practicable, criticality control be based on limited geometrical size. Even when geometrical favourable equipment is used, care is required to assure maintenance of dimensions.

When geometrical safe design is not possible, the plant operation is governed by the so-called double-contingency principle [DOE92]:

“Process designs shall incorporate sufficient factors of safety to require at least two unlikely, independent, and concurrent changes in process conditions before a criticality accident is possible. Protection shall be provided by either”:

1. *“The control of two independent process parameters (which is the preferred approach, if practical)” or*
2. *“A system of multiple (at least two) controls on a single parameter”.*

“In all cases, no (credible) single failure shall result in the potential for a criticality accident.”

An example of application of the double contingency principle can be found in the fabrication facilities for Light Water Reactor (LWR) fuel. Since the slightly enriched uranium can be critical only if appropriately moderated or reflected, full water flooding is the “first contingency” for nearly all analyses. Then, even if flooded, an additional independent change, for example major geometrical rearrangement, would be required for criticality under the double contingency principle.

2.5.3 Criticality data

Data for criticality safety guidance are distributed through a number of publications. “Critical Dimensions of Systems Containing ^{235}U , ^{239}Pu , and ^{233}U ” [PAX87] is a convenient source of critical dimensions. Nuclear Safety Guide TID-7016 [THO78] contains and references a substantial amount of general information. ANS-8.1 [ANS83] and LA-3366 (Rev) [PAX72] are both very useful references.

When using data from these publications, it is important to understand the limitations of the data. For example, [ANS83] states: *“This standard is applicable to operations with fissionable materials outside nuclear reactors, except the assembly of these materials under controlled conditions, such as in critical experiments. Generalised basic criteria are presented and limits are specified for some single fissionable units of simple shape containing ^{233}U , ^{235}U , or ^{239}Pu , but not for multi-unit arrays. Requirements are stated for establishing the validity and areas of applicability of any calculational method used in assessing nuclear criticality safety. This standard does not include the details of administrative controls, the design of processes or equipment, the description of instrumentation for process control, or detailed criteria to be met in transporting fissionable materials.”*

Unfortunately data on the criticality of uranium in the intermediate enrichment range (~6% to ~93% ^{235}U) from which subcritical limits for criticality control can be deduced are limited, and this enrichment range is not adequately covered in [ANS83].

2.6 Anomalies of Criticality

The Oxford dictionary defines an anomaly as “*something that deviates from what is standard, normal, or expected*”. Since 1943 several anomalies of criticality have come to light and have been published in order to add awareness to the persons involved with criticality control and prevention [CLA10].

A few examples are given below:

The cube and the sphere

It is a well-known expectation that a sphere has the smallest critical volume of any shape. The reason for this being that neutron production depends on volume, and neutron leakage on surface area and the ratio of surface area to volume is a minimum in the case of a sphere. Results from experiments have, however, indicated that a reflected cube might under certain circumstances have a smaller critical volume and mass than if the fissile material were in the shape of a sphere [BIE69, BIE71].

Added scatterers and moderation

In some cases the effect of added scatterers can significantly reduce the critical dimension, whereas in others the result can be precisely the opposite. For example, when adding water to mixed oxides of Pu and U, the critical volume initially decreases. Once the point of saturation is reached, further addition of water actually reduces the density of the mixed oxides and the critical volume starts to increase [CLA10].

Fog, mist or flooding: a potential for triple criticality

This example looks at the effect on criticality of increasing the water content of the intervening airspaces within the storage array. This criticality anomaly is caused by the competing effects of increasing water content density between and within units. It looks at an array that is subcritical when totally dry (see Figure 2.9).

At first k_{eff} increases quite fast with increased water content density due to internal moderation, external reflection and enhanced interaction. A small amount of water will typically slow down some neutrons, therefore enhancing the interaction. If the water content is further increased, more neutrons will be absorbed between the units, reducing interaction, and therefore lowering k_{eff} . Then, as the water content density approaches unity, k_{eff} increases again.

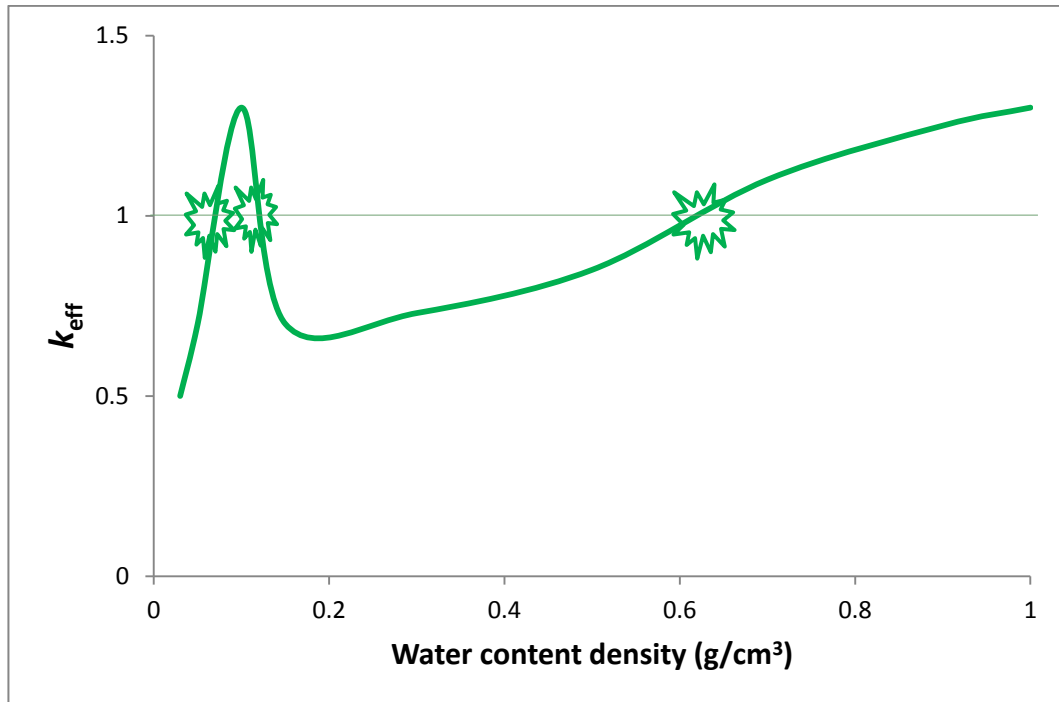


Figure 2.9: Triple potential for criticality induced by increasing water content between and within uranium storage units (Author's own representation).

As shown in Figure 2.9, criticality can occur at three different water content densities (where k_{eff} equals unity), separated by two subcritical regions of water content density, when moving from a completely dry condition to fully flooded.

This problem is particularly of interest for the purpose of this research project. The temperature in the hot cell is high due to the decay heat of the residue stored in the hot cell and any water present in the hot cell evaporates within several hours. It is, therefore, very possible that water vapour is present in the hot cell at a density much lower than that of normal water. Thus, it is important that the safety of a given storage array be examined over the full range of densities that may be encountered.

2.7 Radiation transport code MCNP

The Monte Carlo N-Particle (MCNP) code is an internationally recognized code for analysing the transport of photons and neutrons. Applications for MCNP

include, but are not limited to, accelerator applications, radiation shielding, and nuclear criticality. The code was developed and is maintained by the Los Alamos National Laboratory [SHU07].

2.7.1 A brief description of the Monte Carlo technique

In general, any Monte Carlo technique mathematically traces the paths of individual neutrons, collision by collision, obeying certain fundamental laws of probability. Although very different in form, the procedure can be shown to be equivalent to using a random-sampling technique to evaluate the double integral in the Boltzmann equation (as demonstrated by Schaeffer [SCH73] and Carter and Cashwell [CAR75]).

Knief [KNI85] describes the Monte Carlo method in the context of nuclear criticality safety as the “*tracking of individual (fictitious) neutrons through a material medium containing fissile and other species*”. A complete description of neutron travel paths accounts for: types and distances between interactions, energies and direction of travel, production from fission or other reactions and loss by capture or leakage. It is well known that the outcome of any such event cannot be identified beforehand. Instead, each may be described by a probability distribution that quite accurately predicts the composite behaviour of a very large number of events. These “laws” are usually described or otherwise characterised by such familiar parameters as reaction cross sections, mean free paths, scattering distributions, and energy spectra. Since the outcome of an event is random, the Monte Carlo method makes event selection equivalent to choosing a random number. The overall results of following a number of neutrons through their “path of events” in a particular medium provide a reasonable approximation to the behaviour of the actual system represented by the model.

A simplified flow diagram for a multiplication factor calculation is shown in Figure 2.10.

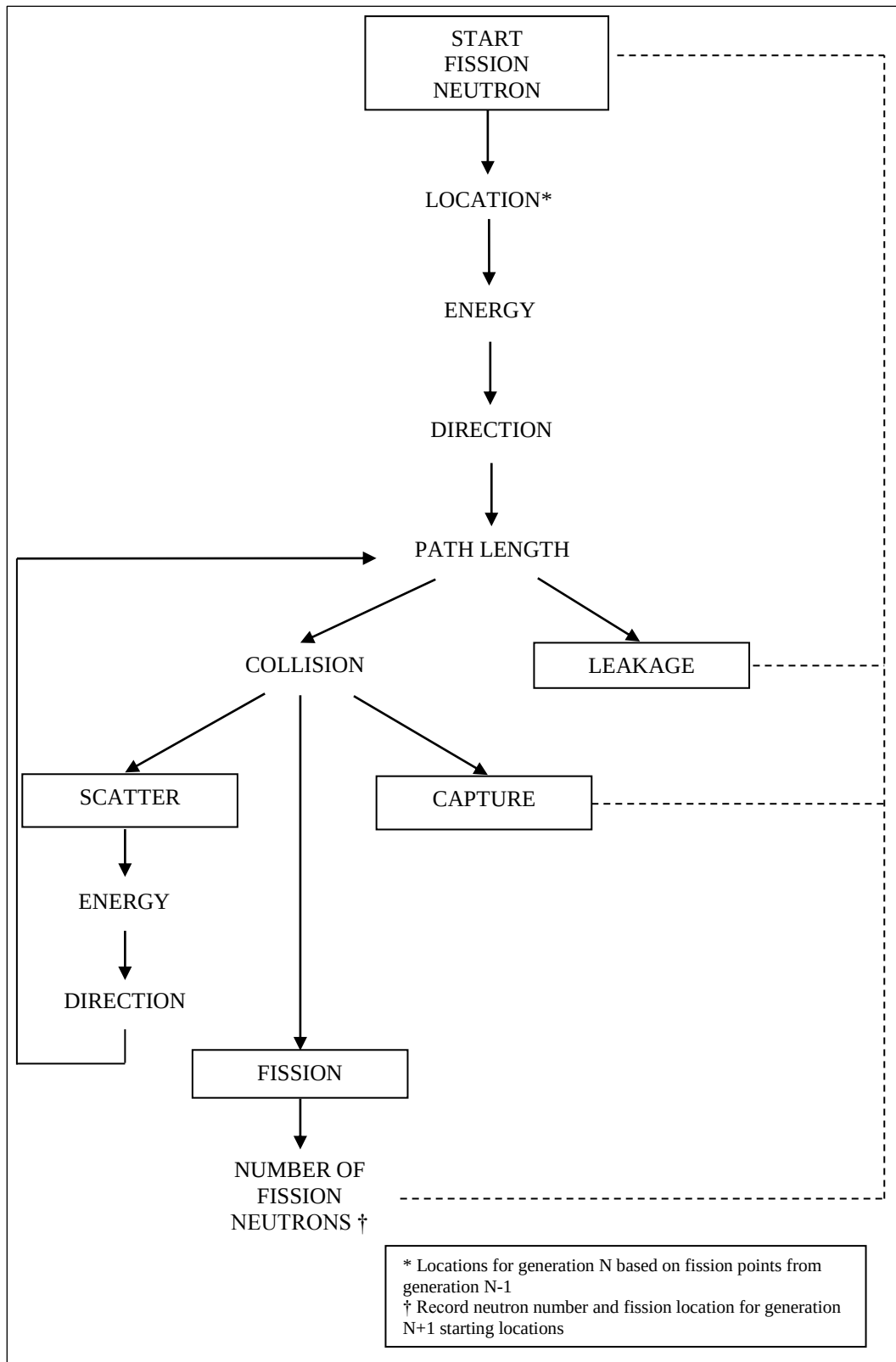


Figure 2.10: Flow diagram for an analog Monte Carlo method used to calculate the effective multiplication factor k_{eff} [KNI81].

The procedure, a form of unbiased or analog Monte Carlo, follows generations of neutrons and compares the number started to the number produced to calculate k_{eff} . In an arbitrary generation, the locations for starting individual neutron histories are selected from those of the previous generation, the first generation having started neutrons from an arbitrary distribution. The energy and direction are selected randomly from appropriate cumulative distribution functions. Neutron path lengths between collisions depend on the total macroscopic cross-section. The geometry determines whether a neutron leaks or experiences a collision at the end of its path length. Collision types are selected randomly in accordance with the appropriate reaction cross-sections. Scattering events change the energy and direction of the neutron before it continues through the system. Leakage, capture and fission terminate the history and signal the start of the next fission neutron. For fission reactions, the number of neutrons is randomly selected with the resulting number and the location if the event stored for use in starting neutrons of the next generation. Since it is typical to start with a fixed number of neutrons in each generation N , the number of fission points in generation $N - 1$ may be adjusted upward or downward by random duplication or elimination, respectively.

An advantage of Monte Carlo over transport methods is that it is inherently less limited by geometry – the computing effort required to track neutron paths is relatively insensitive to geometric complexity. As a result, Monte Carlo can handle any geometry, generally as an assemblage of cells such as finite cylinders and hemispheres, or combinations thereof, and readily accommodates finite arrays of units [KAL68].

2.7.2 Criticality calculations with MCNP

When calculating k_{eff} , MCNP estimates the mean number of fission neutrons produced in a single generation per fission neutron started. In MCNP, a fission generation is called a k_{eff} cycle. In other word, a cycle is an estimation of an actual fission generation, as calculated by MCNP. Processes such as (n,2n) and (n,3n) are considered internal to a cycle and do not act as termination. Because fission neutrons are terminated in each cycle to provide the fission source for the

next cycle, a single history can be viewed as continuing from cycle to cycle. MCNP uses three different estimators for k_{eff} , based on collision, absorption, or track length. Urbatsch *et al.* [URB95] recommend using, for the final k_{eff} result, the statistical combination of all three.

It is extremely important to emphasise that the result from a criticality calculation is a confidence interval for k_{eff} that is formed using the final estimated k_{eff} and the estimated standard deviation.

CHAPTER 3

CRITICALITY ANALYSIS FOR STORAGE CELL: RESULTS AND DISCUSSION

This chapter gives the reader a basic understanding of the MCNP code, by means of a simple example. It also discusses the assumptions made during the present study, it gives a description of the configurations modelled and the parameters changed, and it gives a summary and discussion of the results obtained.

3.1 Neutron transport evaluation using MCNP illustrated with a worked example

In order to better explain the operation of the code and in preparation for understanding the actual model, an example has been chosen that is relatively simple, but contains the necessary principles.

A cylinder representing a residue canister has been defined. Shown in **Figure 3.1** is a representation of a vertical cut through the residue canister (not to scale). The cylinder is inside a box, representing a 30.0 cm thick water reflection. The horizontal cut is shown in Figure 3.2.

The MCNP code requires dimensions in centimetres as input data. The residue canister is 10.2 cm high, with an outside diameter of 16 cm and the wall thickness is 0.1 cm. The box representing the water is 76 cm long x 76 cm wide and 70.2 cm high.

The dimensions of the surfaces are defined in the MCNP input file according to a Cartesian coordinate system. The origin of this axial system can be chosen by the user. In this example, the origin is chosen as the bottom centre of the inside of the residue canister.

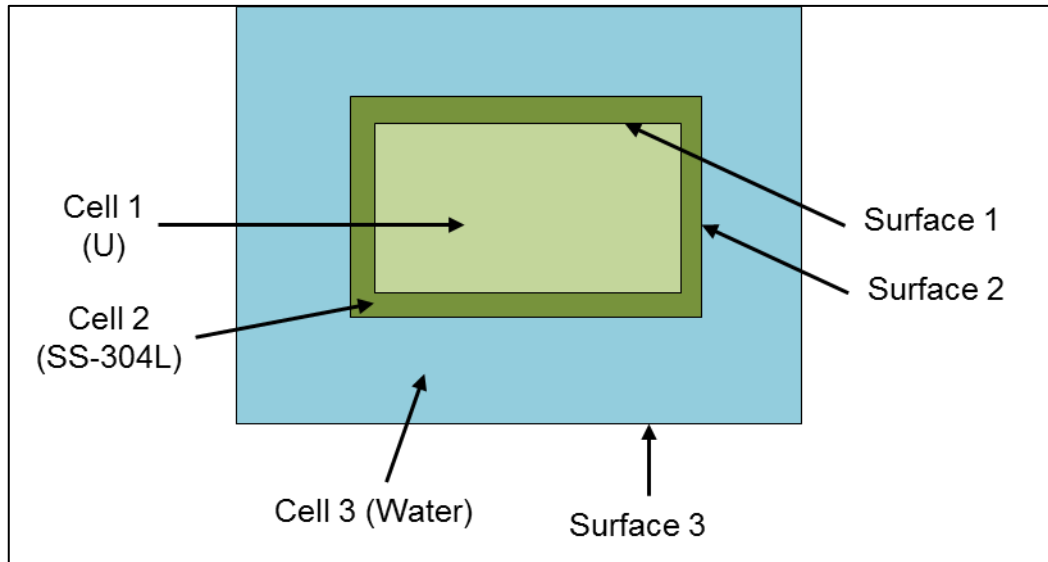


Figure 3.1: Visual representation of the geometry of the MCNP example – side view.

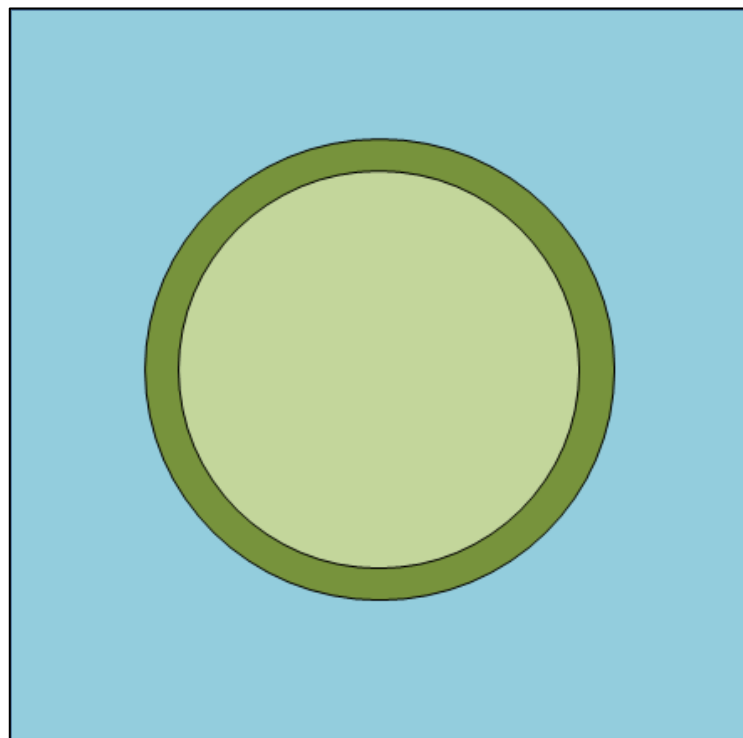


Figure 3.2: Visual representation of the geometry of the MCNP example – top view.

3.1.1 MCNP input data card organisation

An MCNP input file must contain the descriptions of the problem geometry, source and materials. Figure 3.3 shows how the input file is organised [BRE09].

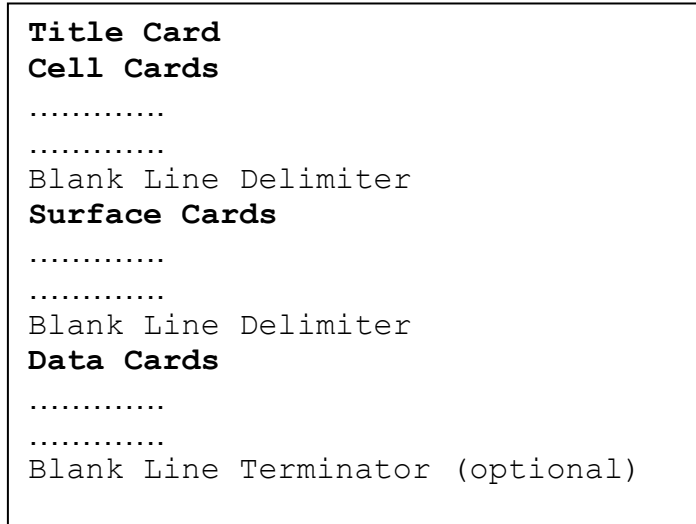


Figure 3.3: MCNP input file structure [BRE09].

The comment character, (“c”), can be used at the beginning of a line. Everything following the comment character is not used by MCNP. The card name, surface number or data card name must begin within the first five characters of a line. A line cannot contain more than 80 characters.

3.1.2 Discussion of the input data set for MCNP example

The input data set for the MCNP example is shown in **Table 3.1**.

A line by line explanation of the input data set follows:

```
Single residue canister
```

This is the title of the MCNP run, known as the Title Card. It contains information about the problem being modelled. The title should be clear enough to enable the user to distinguish among different input files and to help identify the content of output files.

Table 3.1: MCNP example input data set.

```

Single residue canister
c Cell Cards
1      1      -0.833  -1      imp:n,p=1  $residue in canister
2      2      -8.00   -2 +1  imp:n,p=1  $SS-304L canister
3      3      -1.00   -3 +2  imp:n,p=1  $300 mm water reflection
4      0              +3  imp:n,p=0  $External void (UmWelt)
c =====

c =====
c Surface Cards
1      rcc  0  0   0.0   0  0  10.0   7.9   $Canister inside surface
2      rcc  0  0  -0.1   0  0  10.1   8.0   $Canister outside surface
c
3      rpp  -38 +38  -38  +38   -30.1  +40.1  $300mm water reflection
c =====

c =====
c Material Cards
m2      6000      -0.03              $SS-304L
      14028      -0.55              $SS-304L
      14029      -0.03              $SS-304L
      14030      -0.02              $SS-304L
      15031      -0.02              $SS-304L
      16000      -0.03              $SS-304L
      24050      -0.83              $SS-304L
      24052     -15.920             $SS-304L
      24053      -1.81              $SS-304L
      24054      -0.45              $SS-304L
      25055      -1.70              $SS-304L
      26054      -3.980             $SS-304L
      26056     -62.93              $SS-304L
      26057      -1.47              $SS-304L
      26058      -0.22              $SS-304L
      28058      -6.78              $SS-304L
      28060      -2.64              $SS-304L
      28061      -0.12              $SS-304L
      28062      -0.37              $SS-304L
      28064      -0.10              $SS-304L
c
m3      1001      +1.99975           $Water
      1002      +0.00025           $Water
      8016       +1                 $Water
c =====
c
c =====
ksrc    0  0  4.9
kcode   1e3    1    20    50

```

For the purposes of this explanation the next two sections are swopped around. Cell Cards precede the Surface Cards in the input file, but it is more logical to define the surfaces first.

```
C Surface Cards
1      rcc  0  0   0.0   0  0  10.0   7.9      $Canister inside
surface
2      rcc  0  0  -0.1   0  0  10.1   8.0      $Canister outside
surface
c
3      rpp  -38 +38  -38  +38   -30.1  +40.1  $300mm water
reflection
```

Surface 1 is a right circular cylinder, hence the surface mnemonic “rcc”. In the case of an rcc surface, the x_1 , y_1 , and z_1 coordinates (coordinates from the bottom centre) are defined. Thereafter the x_2 , y_2 and z_2 coordinates are defined relative to the origin. Lastly the radius is given. The inside surface of the canister (surface 1) has a height 10 cm and a radius of 7.9 cm.

Surface 2 is the outside surface of the canister. The bottom centre has the same x_1 and y_1 coordinates as surface 1, but the z_1 coordinate is 0.1 cm below that of surface 1, because of the wall thickness of 0.1 cm. This surface has a height of 10.2 cm and a radius of 8 cm.

Surface 3 is a rectangular parallelepiped “rpp” with surfaces normal to major axes. In the case of an rpp, the x , y and z coordinates are defined in the order $x_{\min}$, $x_{\max}$, $y_{\min}$, $y_{\max}$ and $z_{\min}$, $z_{\max}$. Surface 3 represents the 30.0 cm thick water reflection.

```
C Cell Cards
1      1      -0.833  -1      imp:n,p=1  $residue in canister
2      2      -8.00   -2 +1  imp:n,p=1  $$SS-304L canister
3      3      -1.00   -3 +2  imp:n,p=1  $300 mm water reflection
4      0                      +3  imp:n,p=0  $External void (UmWelt)
```

Referring to Cell Cards, the number in the first column is the number of the cell. The number in the second column is the material number that fills the cell. The number in the third column is the density of the material. The fourth column is reserved for reference to the surface numbers that form the boundaries of the cell. The importance (imp) of tracing particles is shown in the fifth column. The \$ sign

signifies the beginning of an in-line comment; everything to the right of the \$ sign, is only a comment.

Cell 1 is composed of material number 1, which has a density of 0.833 g/cm^3 (negative densities mean the quantity is mass-density given in units of g/cm^3 ; positive densities mean that the unit is given as the number of atoms per barn.cm). Cell 1 consists of the volume inside surface 1. The importance of neutrons (n) and photons (p) inside Cell 1, is 1.

Cell 2 is composed of material number 2, which has a density of 8 g/cm^3 . Cell 2 consists of the intersection of all space outside surface 1 and inside surface 2.

Cell 3 is composed of material number 3, which has a density of 1 g/cm^3 . Cell 3 consists of the intersection of all space outside surface 2 and inside surface 3.

Cell 4 is a void region with zero importance. The moment a particle enters this region, it is not tracked any more by the Monte Carlo code. The comment term, “UmWelt” is a short, descriptive German word that means “environment.”

By looking at the surface cards, one now understands that cell 1 is the U inside cell 2, a cylinder consisting of SS 304-L. Cell 3 is water that surrounds the cylinder. Cell 4 is all space outside the water reflection.

Three materials are defined in this input data set. Material 1 is residue, and material 2 is SS304-L and material 3 is water. For the purposes of this report the composition of the residue is omitted. The ZAID (Z-A identity code) of an isotope is $(1000Z + A)$. If a negative number follows the ZAID, it is the mass fraction of the material composed of that element. If a positive number follows the ZAID, it designates a number fraction and not a mass fraction. In the Material Card above, the composition of Stainless steel is given as mass fractions and the composition of water is given as a number fraction.

C Material Cards			
m2	6000	-0.03	\$SS-304L
	14028	-0.55	\$SS-304L
	14029	-0.03	\$SS-304L
	14030	-0.02	\$SS-304L
	15031	-0.02	\$SS-304L
	16000	-0.03	\$SS-304L
	24050	-0.83	\$SS-304L
	24052	-15.920	\$SS-304L
	24053	-1.81	\$SS-304L
	24054	-0.45	\$SS-304L
	25055	-1.70	\$SS-304L
	26054	-3.980	\$SS-304L
	26056	-62.93	\$SS-304L
	26057	-1.47	\$SS-304L
	26058	-0.22	\$SS-304L
	28058	-6.78	\$SS-304L
	28060	-2.64	\$SS-304L
	28061	-0.12	\$SS-304L
	28062	-0.37	\$SS-304L
	28064	-0.10	\$SS-304L
c			
m3	1001	+1.99975	\$Water
	1002	+0.00025	\$Water
	8016	+1	\$Water

The “ksrc” card shows the location where the fission source point is placed. In this case, it is near the centre of the residue canister. The initial “ksrc” source is used only for the first k_{eff} cycle. A new spatial fission source is generated during each cycle and is used as the source for the next cycle.

```
Ksrc  0 0 4.9
kcode 1e3 1 20 60
```

A “kcode” card is required to run a criticality calculation. This example will be run with 1000 neutrons per cycle. The initial estimation for k_{eff} is 1.0. Accumulation of k_{eff} data will only start after 20 cycles have been completed, 60 cycles will be run in total.

3.2 Modelling Assumptions

3.2.1 Cellulose composition

Low density waste present in the cell consists of mainly contaminated papers and plastics which also have a moderating effect due to its low mass numbers. For

modelling purposes it is simplified as cellulose (11.7% hydrogen, 50% carbon, 38.3% oxygen). It is the general composition of cellulose with 8.5% water added. The density of the cellulose is varied to determine the effect that the density has on criticality.

3.2.2 Water mist/steam moderation

In the event of water ingress into the cell, the heat in the cell would rapidly vaporise the water, resulting in mist or steam in the cell. Whenever mist or steam is considered, the top (empty) part of the canister is also filled with mist/steam.

3.2.3 Residue canister geometry

For simplicity, the residue canister is approximated as a stainless steel cylinder (as illustrated in Figure 3.4) with an overall height of 10.2 cm, an outer diameter of 16.0 cm and 0.1 cm wall thickness. The container is filled to a height of 9.5 cm with residue, while the air gap is 5.0 cm. The inside top part of the canister is always filled with air unless stated otherwise.

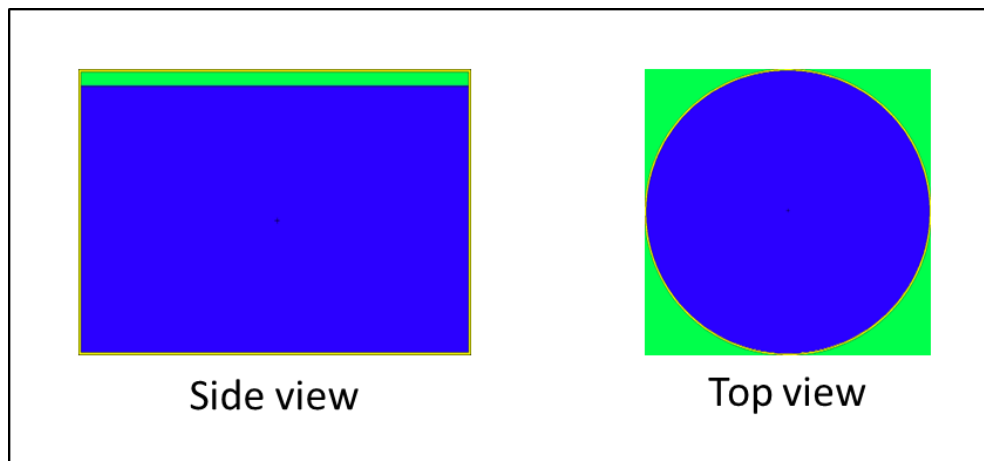


Figure 3.4: Side and top view of a residue canister.

3.3 Models

In this section the two different models are described.

3.3.1 Model 1

The aim of this model is to determine a maximum storage capacity for the storage hot cell. In order to represent a sphere as closely as possible (the most reactive configuration), residue canisters were modelled in different sized “blocks/cubes” (Array A, B, C and D). One configuration representing a slab was also modelled (Array E), which is expected to be much less reactive than the configuration representing cubes.

This model is an approximated block of tightly packed residue canisters moderated by the hydrogen therein, and reflected by 30.0 cm of water. The number of canisters was varied from 360 to 768 residue canisters in five different theoretical arrays. The exact configurations modelled are shown in Table 3.2. Figure 3.5 shows the top and side view of Array D respectively. The side view shows the canisters stacked on top of each other.

The thick red part around the canister in Figure 3.5 represents water and the blue represents the material inside the residue canisters. The green parts between the canisters represent air. The top view shows the 8 x 8 canisters (length x width). The side view shows how the 8 x 8 configuration is stacked 12 canisters high.

Table 3.2: Arrays in Model 1.

Array	Configuration (length x width x height)
A	6 x 6 x 10
B	7 x 7 x 11
C	7 x 7 x 12
D	8 x 8 x 12
E	24 x 10 x 2

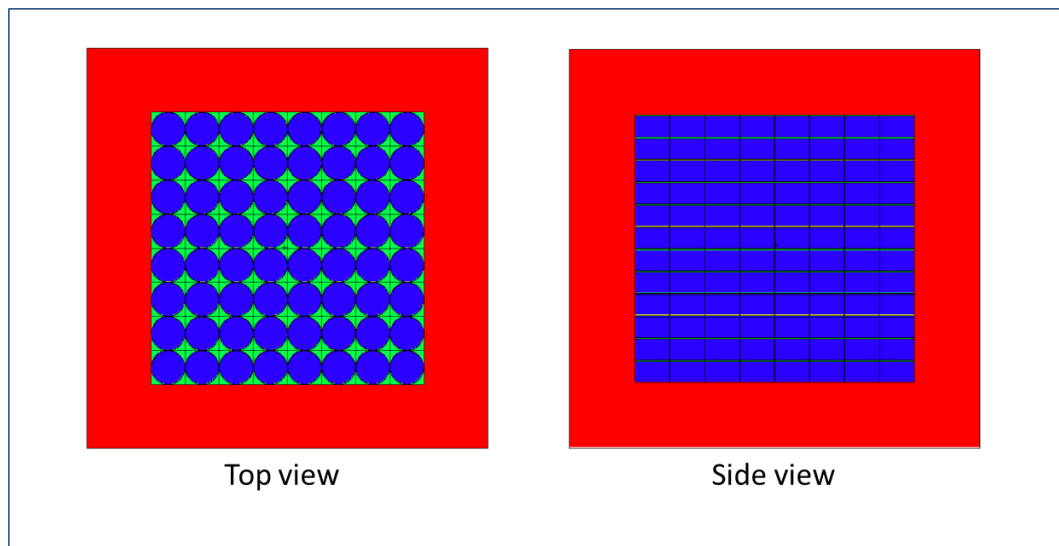


Figure 3.5: Top and side view of Array D as applied in Model 1.

3.3.2 Model 2

This model is similar to Model 1. Since there is also paper towelling and plastic bags present in the hot cell, the effect of cellulose with varying densities (0.024 g/cm^3 , 0.048 g/cm^3 and 0.100 g/cm^3 , respectively) as an interstitial moderator was determined.

This model is an approximated block of tightly packed residue canisters with full cellulose moderation and 30.0 cm water reflection. The residue canisters were modelled in a $6 \times 6 \times 13$ array. The spacing between the individual canisters was varied from 0 cm to 16 cm in the horizontal direction (x- and y-direction) only. In the z-direction, the canisters remain stacked on top of each other as in Model 1. The cellulose was also replaced with water mist/steam to calculate the effect it will have on the different geometries. Figure 3.6 shows the top and side view of the $6 \times 6 \times 13$ array, respectively.

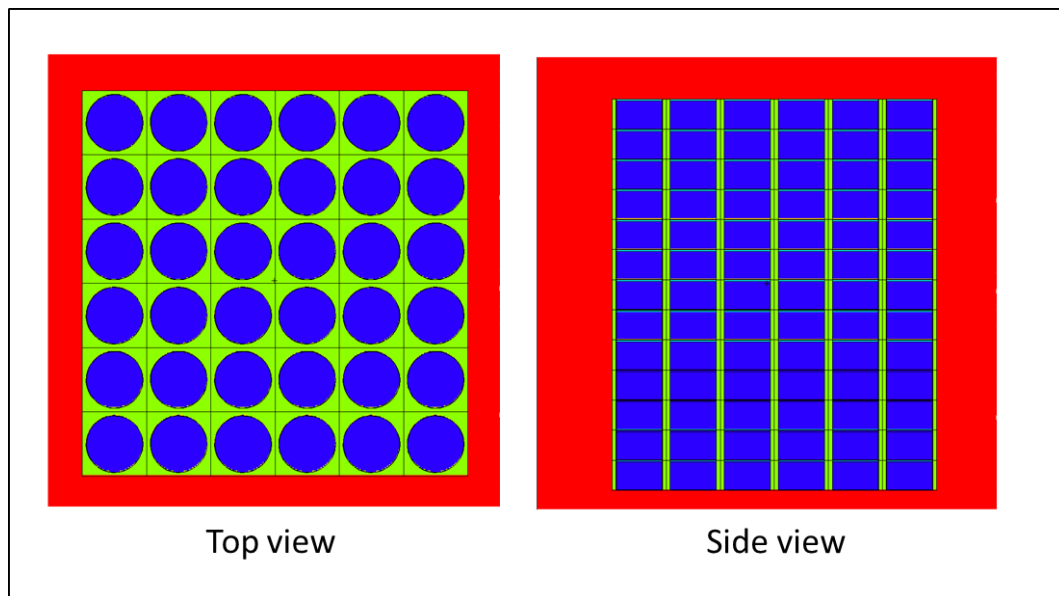


Figure 3.6: Top and side view of an array of $6 \times 6 \times 13$ residue canisters placed a few centimetres (varied from 0 cm to 16 cm) apart as applied in Model 2.

The thick red part around the canisters in Figure 3.6 represents water and the blue represents the material inside the residue canisters. The green parts between the canisters represent cellulose.

3.4 Modelling Results

The Monte Carlo code MCNPX 2.6.0 was used on an Intel® Core™ i7 CPU, 2.67 GHz with 4 GB installed memory (RAM). A total of 41 runs were performed, with an average run time of 8 minutes per run. However, in order to optimise the model, Model 1 went through 14 iterations and Model 2 through 6, adding significantly to the total run time before results were obtained.

Initial programming time for Model 1 was approximately 21 hours, with an additional 1 – 2 hours to make the changes for the different arrays. For Model 2 an additional 10 minutes were needed to adjust the input file for each data point in the model.

The following is a summary of the results that were obtained from the two models ran on MCNP. The k_{eff} , as well as the 2σ value, is obtained from the individual MCNP output files.

3.4.1 Model 1

The results of the five different storage configurations analysed (see Table 3.3) are presented in Figure 3.7.

The reactivity (i.e. k_{eff}) increases as the number of residue canisters increase (for the selected configurations). Model 1 analysed 360 residue canisters while Model 2 analysed 768 residue canisters. The configuration representing a slab containing 480 residue canisters has a much lower reactivity than all of the cube configurations. It is noted that all five configurations are subcritical. However, the result for Array D (768 residue canister) is above the internationally accepted subcritical limit of $k_{\text{eff}} = 0.95$ (indicated by the red line in Figure 3.7).

Table 3.3: Model 1 results.

Configuration	k_{eff}	2σ error (95% confidence)
Array A (6 x 6 x 10)	0.8820	± 0.0013
Array B (7 x 7 x 11)	0.9342	± 0.0015
Array C (7 x 7 x 12)	0.9453	± 0.0012
Array D (8 x 8 x 12)	0.9825	± 0.0002
Array E (24 x 10 x 2)	0.5214	± 0.0002

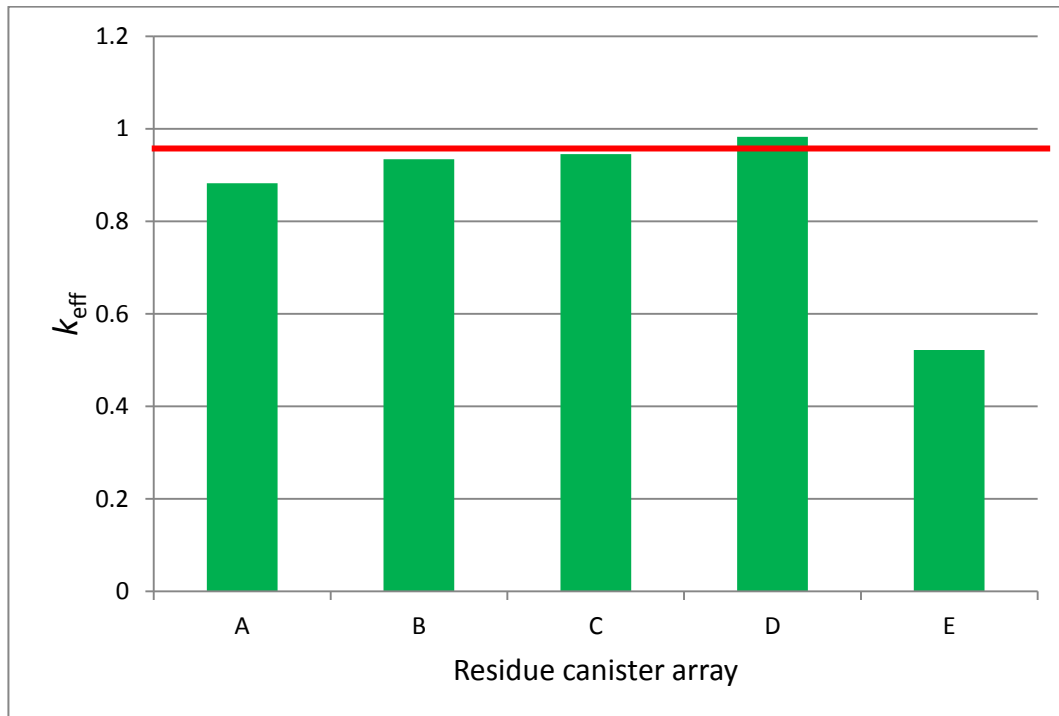


Figure 3.7: Graphical representation of Model 1 results.

3.4.2 Model 2

Table 3.4 shows the results of the 36 model runs, and Figure 3.8 illustrates the effect of interstitial moderator, of varying densities, on the reactivity of the configuration modelled. The x-axis denotes the distance that the canisters are apart from each other in vertical columns (stacked on top of each other). The y-axis denotes the k_{eff} . The legend of Figure 3.8 distinguishes the different densities from each other. The k_{eff} decreases as the distance increases for the lower density moderators while the 0.100 g/cm³ cellulose reaches a peak at 8 cm spacing.

Table 3.4: Results of the 36 model runs in Model 2.

Distance (cm)	k_{eff}			
	Cellulose (moderator)			Water Mist
	0.024 g/cm ³	0.048 g/cm ³	0.100 g/cm ³	
0	0.92569	0.93964	0.97063	0.91083
2	0.88839	0.91658	0.97752	0.85642
4	0.85931	0.90065	0.98477	0.82214
6	0.83568	0.88478	0.99021	0.78685
8	0.81412	0.87283	0.9923	0.75616
10	0.79479	0.8612	0.99076	0.72004
12	0.77896	0.85027	0.98643	0.69707
14	0.76158	0.84018	0.97883	0.68251
16	0.74813	0.82947	0.96741	0.66256

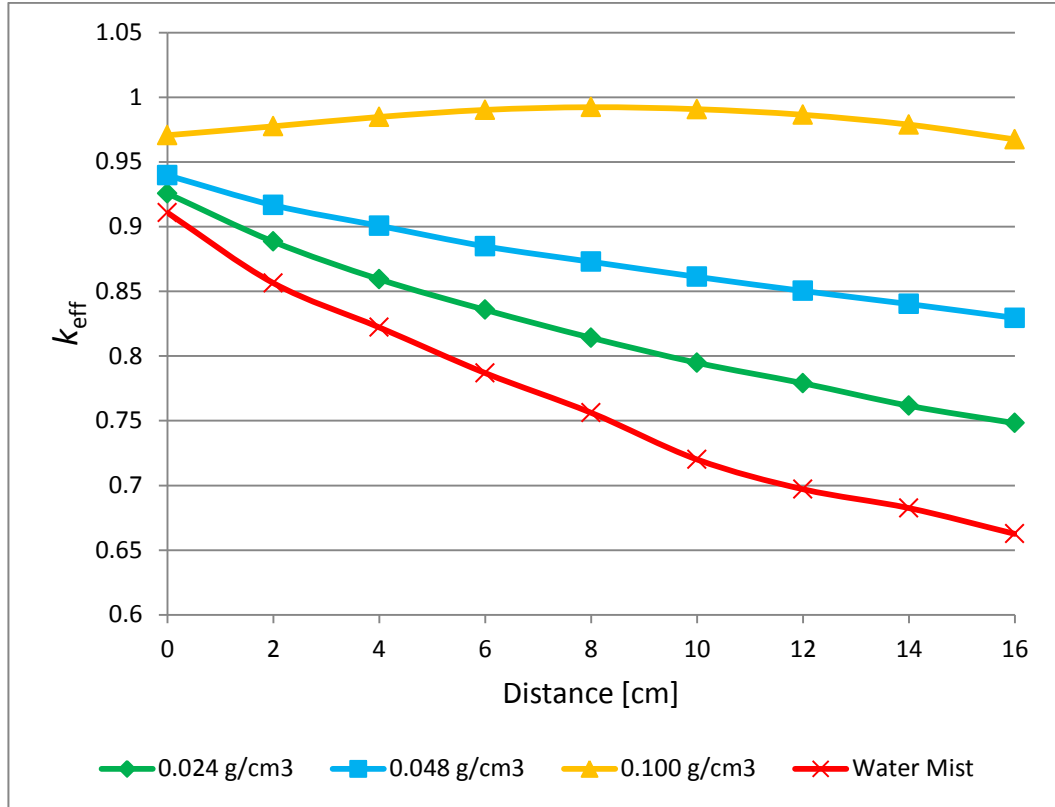


Figure 3.8: Graphical representation of Model 2 results.

Table 3.5: Maximum k_{eff} values for Model 2.

Moderator	Max k_{eff}	Distance at which the k_{eff} peaks	2 σ error (95% confidence)
Cellulose (0.024 g/cm ³)	0.9257	0 cm	± 0.0005
Cellulose (0.048 g/cm ³)	0.9396	0 cm	± 0.0010
Cellulose (0.100 g/cm ³)	0.9923	8 cm	± 0.0023
Water mist/steam	0.9120	0 cm	± 0.0010

Table 3.5 shows the maximum k_{eff} for each of the moderators modelled, and corresponds to the graph in Figure 3.8.

3.5 Discussion of results

The internationally accepted subcritical limit of $k_{\text{eff}} = 0.95$ allows for uncertainties in the calculations and experimental data used in its derivation. The results of Model 1 indicate that the theoretical storage configurations represented by Arrays A, B, C and E are acceptable in terms of criticality safety, with the k_{eff} below 0.95. This equates to a maximum of 588 residue canisters being stored in the storage hot cell in a rectangular assembly with no interstitial moderation and full water reflection.

With full homogeneous interstitial cellulose moderation of a density up to 0.048 g/cm^3 , the configuration in Model 2 remains subcritical when spaced horizontally from 0 cm to 16 cm. For a cellulose density of 0.100 g/cm^3 , k_{eff} exceeds the subcritical limit but does not reach supercriticality. Canisters are stored in the storage hot cell in a rectangular assembly with interstitial cellulose moderation (density $< 0.1 \text{ g/cm}^3$) and full water reflection.

Water mist or steam calculations show the lowest k_{eff} and are, therefore, bound by the interstitial cellulose moderator.

From this study, it is derived that the storage of up to 588 residue canisters will be safe in terms of criticality, without placing any restrictions on the exact geometry. The number of residue canisters that can be safely stored can, however, be increased by introducing a fixed geometry. There are two ways that this can be implemented.

Firstly – by increasing the surface area to volume ratio of the configuration, the reactivity of the system is reduced (as shown in Model 1), since the same amount of fissile material is safer in a shape with a large surface area (such as a slab) than in a small compact shape (such as a cube or a sphere), because the number of neutrons that will escape the system is higher, resulting in a lower k_{eff} . This is supported by the theory presented in Section 2.5.1.1.

Secondly – by introducing spacing between canisters, the reactivity (k_{eff}) of the system is reduced (as shown in Model 2), since the probability of neutrons interacting is reduced when the fissile material is spaced further apart. This is supported by the theory (see Section 2.5.1.4).

CHAPTER 4

RECOMMENDATIONS AND CONCLUSIONS

The conclusions and recommendations resulting from this study are presented in this chapter.

4.1 Conclusions from the calculations

The results of the MCNP model predictions are presented in Chapter 3. From the model runs, it is clear that all the array configurations are safe in terms of criticality safety, except for Array D.

4.2 Inputs to the design

From the present study, the following recommendations can be made as inputs into the design of the facility.

When deciding on a maximum capacity for the storage cell, any of the theoretical configurations modelled in Array A, Array B and Array C will remain subcritical without the need to implement any restrictions on the storage configuration.

Should the capacity represented by the above arrays be not sufficient, the capacity can be increased by:

- 1) increasing the surface area to volume ratio, i.e. move more towards a “flatter” configuration such as a slab (Scenario 1); or
- 2) increasing the distance between the canisters (Scenario 2).

In the case of high density moderator, it is clear that there will be a preferred spacing between canisters.

Whether the design team decides to choose 1) or 2) above to increase the capacity, additional restrictions (controls) will need to be implemented to ensure that the canisters remain in a safe configuration.

It is relatively simple to introduce restrictions that will keep the configurations safe in term of criticality. For Scenario 1, the stacking height of the canisters can be limited. For Scenario 2, a prescribed distance between canisters can be introduced. These are, however, administrative controls.

The biggest concern with administrative controls is that it is entirely dependent on the operator, and the probability of non-adherence to the restrictions is relatively high. Furthermore, it is also often difficult to detect non-adherence.

When implementing controls, the order of preference should be:

- First – Passive Engineered Controls;
- Second – Active Engineered Controls;
- Third – Administrative Controls.

The design team should therefore be advised to rather look at the possibility of engineered controls. Possible engineered controls for the 2 different scenarios are described below.

Scenario 1:

The surface area to volume ratio of the configuration can be increased by decreasing the height of the arrays. This can be done, for example, by not allowing any stacking of the canisters. By modifying the lid design, the canister can be manufactured such that it is impossible to stack canisters on top of each other (Passive Engineered Control). The disadvantage of prohibiting any stacking is that the area (floor space) required for storage can increase significantly.

Scenario 2:

The distance required between canisters can be fixed by, for example, introducing a floor grid which will prevent the operator from placing canisters closer to each other than the required distance (Passive Engineered Control).

Recommendations:

Based on the finding of this report, the recommendation to the design team is to implement a combination of Scenario 1 and Scenario 2. A storage rack should be designed that consists of fixed positions for the canisters, thereby fixing the geometry as well as the distance between individual canisters.

In addition it is recommended that the exact storage configuration be modelled upon completion of the design of the facility.

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