

of trace-elements in human blood and tissues. The accuracy of this analytical method has led to its use in the medico-legal field. Hair, even hundreds of years old, can be analysed successfully for arsenic and other residues. It has even been used to identify the geographical source of opium by identifying the trace-elements the poppy plants absorb from the different soils in which they grow.

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A QUALITATIVE AND COMPARATIVE STUDY OF ELEMENTS IN TEETH BY NEUTRON ACTIVATION AND HIGH RESOLUTION GAMMA SPECTROMETRY

I. INTRODUCTION

AS the exact mineral composition of teeth often has to be known, improved methods for the detection and quantitative analysis of the elements they contain continually are being sought. In 1937 Lowater and Murray¹ detected nineteen elements by spectrographic methods; and later Swift² found a large number quantitatively by means of spark source mass spectrometry. A further advance was the use of electron probe x-ray microanalysis by Suga *et al*³ to determine the topographical distribution of some of the elements present.

A recently developed sensitive analytical method—neutron activation followed by gamma-ray spectrometry—may be used as a qualitative and quantitative method for analysis of elements in teeth. Söremark and Samsahl^{4,5,6} employed it for the analysis of trace-elements in enamel, dentin and calculus. The samples were irradiated for 20 hours in a neutron flux of approximately $2 \times 10^{12} \text{n.cm}^{-2} \text{sec}^{-1}$. The activated samples were sub-

jected to a chemical group separation followed by gamma spectrometric analysis. A NaI(Tl) scintillation crystal was used as a detector. Eleven elements (Ca, P, Na, Cl, Zn, Sr, Br, Mn, Cu, W and Au) were determined quantitatively, and an additional eight long-lived radioisotopes qualitatively. This work was continued by Söremark and Lundberg⁷, who determined six of these long-lived radioisotopes (Pb, Cr, Ag, Fe, Co and Pt) quantitatively by employing a higher neutron flux of approximately $6.5 \times 10^{13} \text{n.cm}^{-2} \text{sec}^{-1}$.

The inherent high sensitivity of neutron activation analysis for many elements makes this method suitable for the analysis of trace-elements. Since Ge (Li) semiconductor detectors⁸ with a superior resolution have superseded the Na I (Tl) scintillation counters for the detection and measurement of gamma-rays, this method has become even more versatile. The high resolution permits studies of complex gamma spectra in much more detail and has made possible the non-destructive

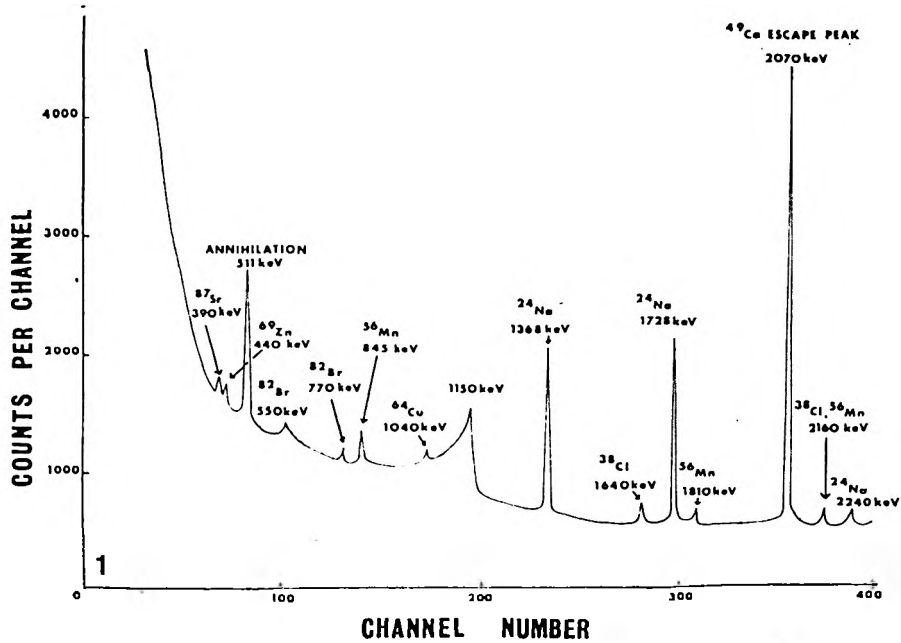


FIG. 1.—Gamma spectrum of tooth 3 after short irradiation. 4 of patient A. Total weight of tooth 1.0643 gm.

neutron activation analysis of a great variety of samples^{9,10}.

This investigation was undertaken to determine whether non-destructive neutron activation analysis and high resolution gamma spectrometry is a suitable method for the analysis of elements in human teeth. At the same time the results obtained were used to compile "nuclear fingerprints" of different teeth which were then compared.

II. EXPERIMENTAL PROCEDURE

(a) Preparation of the teeth prior to irradiation

Thirty-six sound, freshly extracted, unrestored teeth were used. Three were obtained from each of 12 individuals and consisted of a pair of contralateral teeth, and an odd one. Obvious debris or tartar was removed with plastic instruments. The teeth were then thoroughly washed in de-ionised water for a few minutes and dried in an oven at 105° C to constant weight, and the weight of each tooth was recorded. The teeth were sealed in polyethylene tubes for short irradiation, and subsequently some were sealed in quartz ampoules for long irradiation.

(b) Irradiation of the samples

For the detection of short-lived radioisotopes, each tooth in its polyethylene container was irradiated for exactly one minute. The irradiation was carried out in the pneumatic facility of the nuclear reactor Safari I* in a thermal neutron flux of approximately $1 \times 10^{13} \text{ n. cm}^{-2} \text{ scc}^{-1}$.

The long-lived radioisotopes were detected after irradiation of the teeth in the poolside facility of Safari I. They were irradiated for 50-100 hours in a thermal neutron flux of approximately $2 \times 10^{13} \text{ n. cm}^{-2} \text{ scc}^{-1}$.

(c) Measuring of the gamma activity

The measurement of the gamma activity of the samples that were irradiated for one minute was commenced exactly two minutes after each irradiation. The teeth were placed 7.0 cm below a Ge (Li) detector; that used at Pelindaba is a 5.0 cc coaxial Ge (Li) diode† mounted in a cryostat and cooled with liquid nitrogen.

*Safari I — An ORR-type reactor of the Atomic Energy Board at Pelindaba.

†Princeton Gamma Tech., Princeton, J. J., U.S.A.

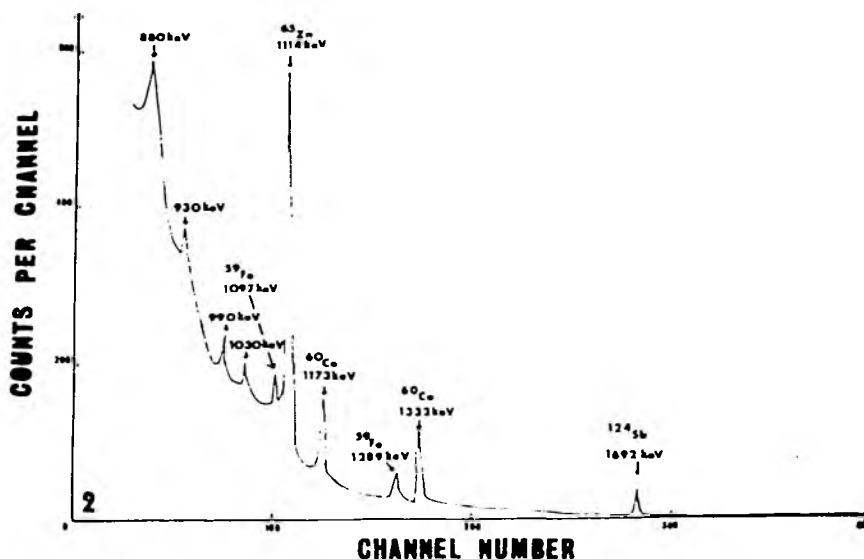


FIG. 2.—Gamma spectrum of the same tooth as in Fig. 1 after long irradiation.

An uncooled TC-130 Tennelec pre-amplifier, a TC-200 Tennelec amplifier and a R I D L bias-amplifier (only the cut amplifier section) were used to obtain low noise amplification of the diode output signals. The analysis of the gamma-spectrum was done on an Intertechnique 400 channel pulse height analyser (model SA 40B), bypassing its built-in amplifier. The resolution of the counting equipment is 4.6 KeV (fwhm) for the 0.662 MeV photopeak of ^{137m}Ba .

The teeth that were irradiated for 50-100 hours were allowed to undergo radioactive decay for approximately three weeks before counting was undertaken. The same recording equipment was used.

The results were recorded graphically with a point plotter and printed digitally.

III. RESULTS

Graphs were drawn from the digitally printed results. The energies of the peaks in the gamma spectrum obtained from each tooth were determined from the calibration curve. The elements responsible for these peaks were identified from available tables^{11,12}. Although the determination of the short-lived and long-lived radioisotopes were undertaken as a non-destructive analysis, it was found that whenever the teeth were irradiated for

long periods they became brittle and disintegrated.

(a) Qualitative analysis of elements in teeth

In 36 teeth analysed Sr, Zn, Br, Mn, Cu, Na, Cl, Ca, Fe, Co and Sb were identified. Fig. 1 represents a typical spectrum obtained from one of the teeth subjected to short irradiation, and Fig. 2 the spectrum obtained from the same tooth after long irradiation.

(b) Comparative study of the "nuclear fingerprints" obtained after short irradiation

Figs. 3, 4, 5 and 6 represent the gamma spectra obtained after short term irradiation of a matched pair and another tooth from one individual and a tooth from a different individual. The basic "nuclear fingerprint" pattern obtained from the different teeth is similar. Certain minor differences occur such as the absence of the first chlorine peak in Fig. 4. There are also differences in the heights of some of the peaks of corresponding radioisotopes in the various spectra, indicating a possible difference in the concentration of the respective isotope.

IV. DISCUSSION

In this preliminary investigation only

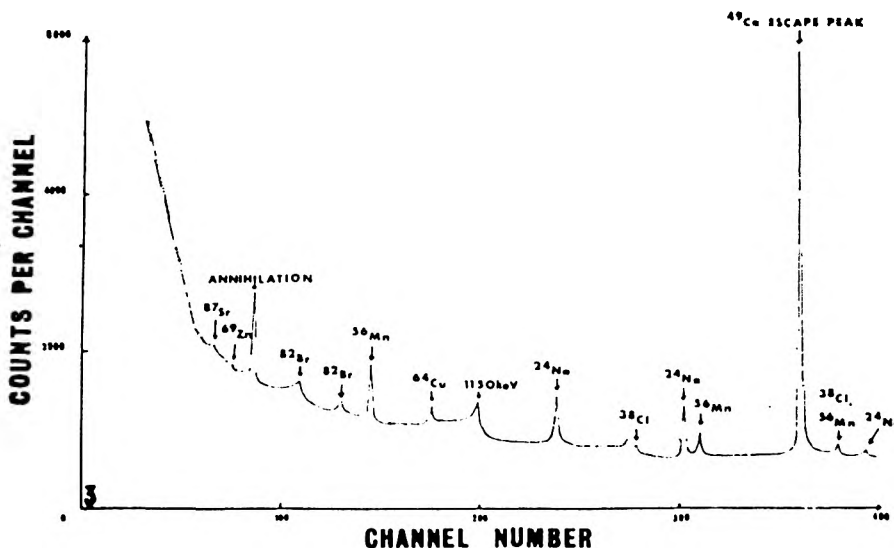


FIG. 3.—Gamma spectrum of tooth 10 after short irradiation. 11 of patient B. Total weight of tooth 0.9482 gm.

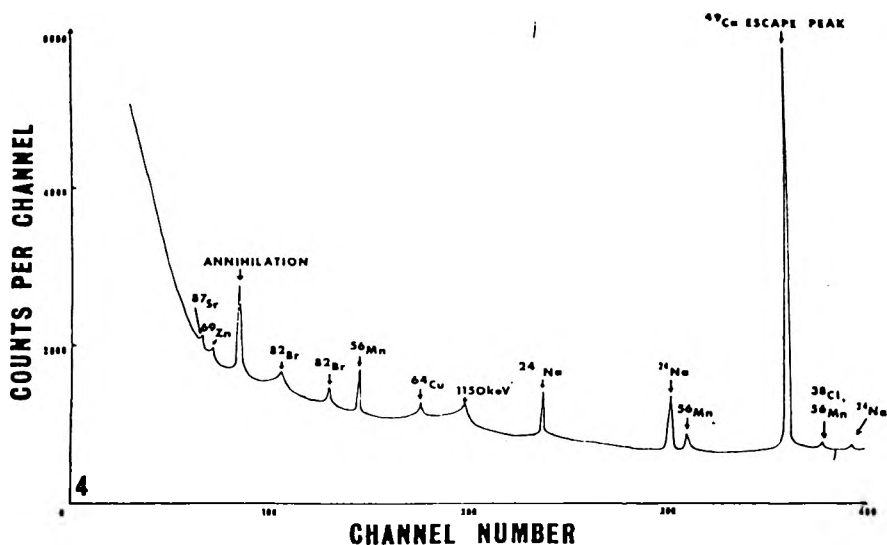


FIG. 4.—Gamma spectrum of tooth 11 after short irradiation. 11 of patient B. Total weight of tooth 0.9786 gm.

11 of the elements present in teeth could be determined qualitatively. The relatively small number detected can be attributed to two factors. First, the samples could not be irradiated for longer than one minute if counting was to commence two minutes after irradiation. Because of

the amount of sodium present in these teeth, the activity of its isotope, ^{24}Na , is such that the recording instrument would be damaged if counting instrument would be commenced two minutes after a longer irradiation. The latter limitation can be overcome by a chemical separation technique which, by

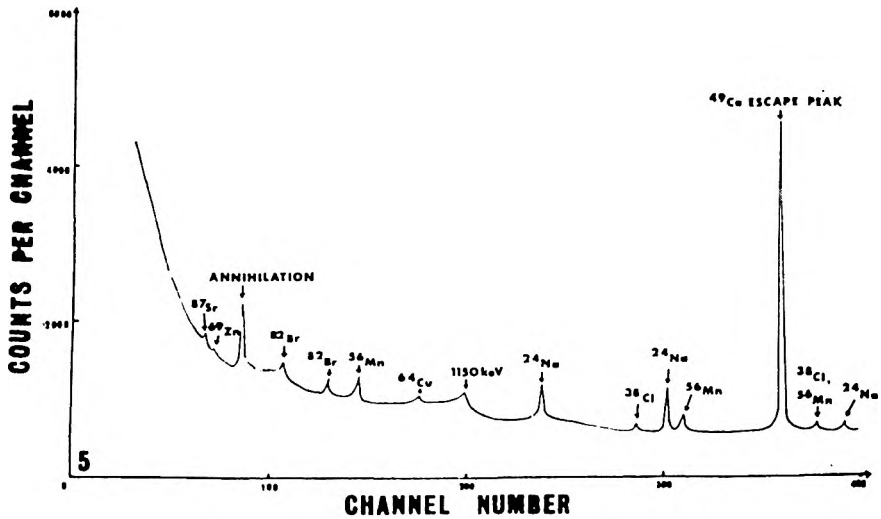


FIG. 5.—Gamma spectrum of tooth 12 after short irradiation. $\frac{3}{1}$ of patient B. Total weight of tooth 0.8870 gm.

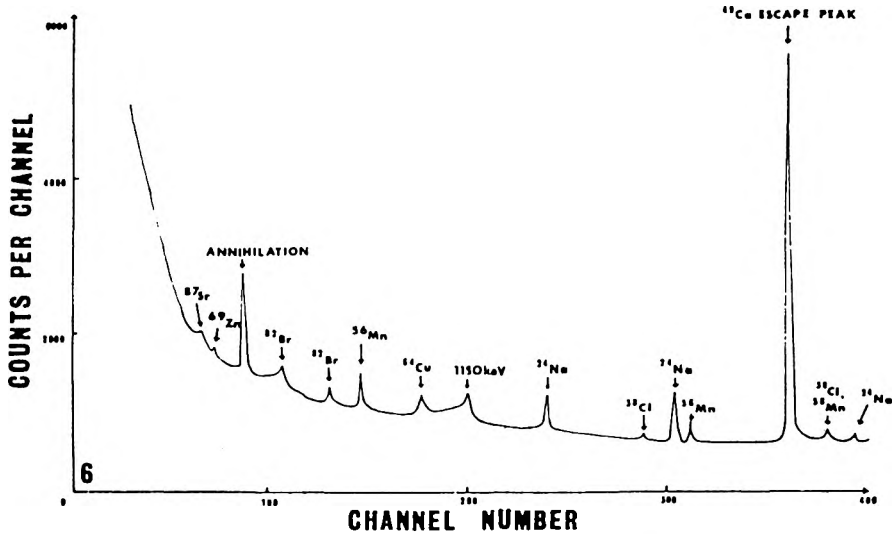


FIG. 6.—Gamma spectrum of tooth 13 after short irradiation. $\frac{3}{3}$ of patient C. Weight 0.9684 gm.

means of an ion exchange resin to remove the sodium prior to or after irradiation, will allow the sample to be irradiated for a much greater length of time. A longer irradiation will result in the detection of more short-lived radioisotopes and at the same time increase the heights of the peaks of the elements already determined.

Secondly, with the facilities available at Pelindaba it was not possible to commence counting sooner than two minutes after irradiation of the sample. The detection of very short-lived radioisotopes such as ^{20}F , with a half-life of 11 seconds, becomes very difficult.

The advantages of a non-destructive

analysis are obvious. Apart from the fact that it excludes a time-consuming chemical separation technique, the tooth itself is not destroyed, which is important in the medico-legal field. This advantage of the non-destructive neutron activation analysis is, however, limited to the determination of the short-lived radioisotopes in teeth, it was found that whenever they were irradiated for long periods they become brittle and disintegrated. The disintegration may be accompanied by the evolution of gases resulting in an increase in pressure within the sealed ampoule. This may have been the cause of the disintegration of one of our sealed quartz ampoules which led to the contamination of the laboratory with radioactive material. It therefore is recommended that teeth to be irradiated for long periods be sealed in quartz ampoules under vacuum.

The "nuclear fingerprints" of the two paired teeth (Fig. 3 and 4) are practically identical. The "nuclear fingerprint" pattern, besides being very similar to that of the other tooth of the same person (Fig. 5), can also be much the same as that of a tooth from a different individual (Fig. 6). In this investigation the "nuclear fingerprints" obtained from the teeth after irradiation showed no significant differences.

The variation in the heights of corresponding peaks which were apparent need not necessarily be attributed to the differences in concentration of the respective isotopes; it could be due to the difference in weight of the teeth compared or to the variation in the relative value of the neutron flux in the reactor at the time of irradiation. The latter problem can be overcome by determining the relative value of the neutron flux for each irradiation.

IV. CONCLUSION

This investigation confirms that neutron activation followed by high resolution gamma spectrometry is a sensitive method for the qualitative analysis of some of the elements in teeth. The anticipated advantages of the non-destructive analytical method did not materialize as the teeth disintegrated when they were irradiated for long periods. A technique possibly

could be developed employing neutron activation followed by high resolution gamma spectrometry in conjunction with chemical separation (destructive analysis) for the qualitative and quantitative analysis of a great number of elements in mineralized dental tissues.

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