

PROTON IRRADIATED URANIUM

by

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Ce n'est jamais par les routes les plus simples et les plus directes que l'esprit humain parvient aux verites.

LAGRANGE

Abstract

The imminent commissioning of the Spallation Neutron Source at the Rutherford Appleton Laboratory produced a requirement for the measurement of radionuclides produced in 800 MeV proton irradiated uranium. Estimates for the formation of irradiation products were obtained from the general calculation involved in the target design but it was felt appropriate to check these by direct measurement.

Both alpha and beta gamma emitters have been measured following the dissolution of the target. Gamma emitting nuclides were identified and measured directly by gamma spectrometry using an intrinsic germanium detector. Alpha emitting nuclides were measured using alpha spectrometry after chemical separation by ionexchange and solvent extraction. No measurements of Beta emitting nuclides has been made in this work.

Estimates of the production cross-sections of some of the nuclides identified have been made. These are compared with measurements made some 25-30 years ago using less sensitive equipment and with those calculated using Monte Carlo codes.

Measured cross-sections in the spallation region (200-238 amu) showed good agreement with both the Monte Carlo calculation and the previously reported work : Pa-233, Th-228, Th-227, 13.3mb, 3.3mb, 2.8mb respectively. Also included are the nuclides Po-209 and Po-208 which have not previously been reported.

Agreement with the calculated yields in the fission region, from 156 down to 88 amu are less good. The cross-sections measured were within a factor of two greater : (measured: calculated) Y-88 7.2mb : 4.48mb, Ag-110m 6.43mb : 6.17mb, Sb-124 12.87mb : 6.54mb. Other nuclides show a much greater difference from the calculated data: Zr-95 94.7mb : 34.7mb, Ru-103 114.8mb : 19.14mb, Cs-136 20.1mb : 3.56mb, Cs-137 71.4mb : 3.62mb. In the fission region comparison with the previous work is difficult as the majority of nuclides identified and measured here have not previously been reported and there is also a great difference in the irradiation energy. The cross-sections for the nuclides Y-88, Zr-95, Sb-124, Sb-125, Te-121, Te-121m, Te-123m, Ce-141, Cs-134 and Cs-136 are reported here for the first time.

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CHAPTER 11.1a SPALLATION NEUTRON SOURCE

The need for a more intense source of neutrons in the study of materials structure, and in the study of condensed matter has prompted the development, at the Rutherford Appleton Laboratories in Oxfordshire, of a Spallation Neutron Source in which the interaction of protons with a target material produces a source of neutrons by spallation and fission. At present the major source of neutrons for structural and condensed matter research is from high flux research reactors with fluxes in the order of 10^{15} n cm⁻² s⁻¹ (such a reactor is at the Institut Max von Laue-Paul Langevin Grenoble).

The interaction of high energy protons with heavy nuclei produces numbers of neutrons per incident proton which are significantly greater than those from thermal fission. The yield of neutrons from spallation in a target of depleted uranium is approximately double that of other metal targets such as lead. The increased amount is due to the fission that occurs from the interaction of primary protons and that of the cascade nucleons (see section 1.5), as well as fast fission from evaporation neutrons.

Yields have been estimated for ^{238}U with 800 MeV protons which vary from : 35 n/p^(1.1) down to 26 n/p^(1.2). The proton synchrotron at the Rutherford-Appleton laboratory will produce approximately 3×10^{13} protons per pulse cycling at 53 Hz giving 1.59×10^{15} protons per second which will in turn produce neutrons in the order of 4.8×10^{16} per second from a uranium target of 1 range-length for protons [approximately 25cms at 800 MeV.]. In addition to the production of neutrons π and μ mesons and neutrinos are produced.

Neutrons can be used in the same manner as X-rays for the study of solids, liquids and biological materials, but they give better resolution and hence more information, X-rays give only an average description of a structure whereas neutrons make it possible to study atom motions^(1.2).

When on line the SNS will provide neutron beams to research stations for neutron diffraction experiments. The facilities and their primary uses are described in brief below.

: Liquids and Amorphous Solids Diffractometer
used to study the structure of non-crystalline
materials (high intensity medium resolution
diffractometer).

- : Time Focused Crystalline Diffractometer for vibrational spectrometry (high intensity moderate resolution apparatus).
- : High Resolution Powder Diffractometer with a $\delta d/d = 4 \times 10^{-4}$, to be used for structural measurements.
- : High Energy Transfer Spectrometer for the measurement of high-energy inelastic magnetic and vibrational excitations.
- : Low-Q Diffractometer which will allow small angle neutron scattering measurements to probe structures at the 10-50 Angstrom level.
- : High Resolution Spectrometer for the investigation investigation of near-elastic processes such as diffusion and rotation of molecules, this will have a resolution in the range of 1 μ eV to 50 μ eV.

1.1b OBJECTIVE OF PRESENT WORK

It is important to note that as a consequence of the target irradiation many radioactive residual nuclei from spallation and fission reactions will be produced and they could constitute a radiological hazard.

It is therefore necessary for health physics purposes, that information on the build up of activity in the target is estimated, and calculations made for the yields of isotopes of most significance in the event of an accidental cladding failure in the target assembly. This can be done by two methods: computed Monte-Carlo calculations or analysis of a scaled down irradiated target. The latter produces direct information, that is, σ the production cross-section of spallation nuclides and isobaric mass yields from quantitative measurements of fission products.

The work reported in this thesis deals mainly with the analysis of an irradiated uranium target, but some cross-section information was obtained by Monte-Carlo calculations run at the Swiss Institute for Nuclear Research by Dr. F Aitchison. Comparison of the data from both sources is given in Chapter 4.

1.2 NUCLEAR INTERACTIONS

The interaction of high energy nucleons with target atoms are varied and numerous, and it is therefore convenient to categorise them by their general features. They are often divided into four groups
a) spallation, b) fission, c) fragmentation and
d) secondary reactions.

There are two principal initial interactions: elastic and inelastic scattering. In elastic scattering no change occurs in the nucleus following the interaction of the incoming particle. In this type of interaction the sum of the particles kinetic energy is conserved and changes in velocity will be observed. In the case of charged particles, e.g. a proton, the forces responsible for this are electrostatic Coulomb repulsion and nuclear forces. In the case of neutral particles (neutrons) only nuclear forces effect the velocity change. Some elastic scattering always occurs in nucleon-nucleus reactions independently of energy or the incident particle in the collision event.

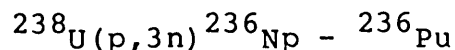
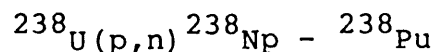
The category of inelastic scattering can be divided into several types of nuclear reactions covering all other reactions where changes occur in the struck nucleus and the incoming nucleon: spallation where nucleons or clusters of nucleons are emitted from the struck nucleus, fission in which the nucleus divides into two or more fragments of comparable mass, fragmentation giving rise to chunks of nuclear matter with masses in the range of 18 to 32 amu^(1.3) and secondary reactions in which an emitted particle from spallation interacts with a target nucleus. A final mode of interaction is the capture of the bombarding nucleon giving rise to products with a mass number greater than the target. These categories of reaction are not

at all mutually exclusive, and a target nucleus interacting with an incident nucleon may, for example, emit several particles in a spallation reaction and then undergo fission, or a fragment in an excited state may emit several nucleons.

The reactions of interest here are: capture, fission and spallation induced by protons.

1.3 CAPTURE REACTIONS

This is seen as a medium energy reaction as the Coulomb barrier stops charged nucleon induced reactions at low energies, the incoming particle needing sufficient energy to overcome this electrostatic barrier. Protons having sufficient energy are capable of kicking out one or more nucleons or larger fragments of the nucleus giving reactions of the type (p,n) , $(p,2n)$, (p,pn) . The reaction cross sections



have been measured in this work. These reactions are assumed to follow the absorption of the incident proton by the target nucleus to form a 'compound nucleus' where the incoming proton's energy is distributed throughout the nucleus leaving the target atom in an excited state.

If the energy of the incoming particle is insufficient none of the nucleons escape initially but statistical fluctuations may redistribute the energy giving sufficient energy to one or more nucleons allowing them to escape with relatively low kinetic energy.

1.4 FISSION REACTIONS

"Nuclear fission is a violent collective phenomenon in which a nucleus undergoes a series of large vibrations until it becomes so strongly deformed that it breaks into two primary fragments of comparable mass"(1.4). Fission can occur by either spontaneous processes (in actinides) or induced by the interaction of neutrons or protons with a complex nucleus. Fission induced by nucleons is dependent upon the incident energy of the nucleon. At low energy the fissioning nucleus can be thought of as a compound nucleus which then deforms to the point of scission, where two fragments are still in contact and are elongated due to nuclear forces attracting one another. At this point there is a concentration of protons away from the point of contact of the scissioning nucleus due to repulsion between the protons on either side of the saddlepoint. As the two fragments separate the influence of the nuclear forces no longer affects them and they are repelled due to the Coulomb forces.

The fragments are in an excited state as excitation energy from scission and the deformation energy of elongated fragment nucleus are liberated as the nuclei regain spherical shapes. If this energy is greater than the energy for neutron separation within the fragment, then the system de-excites by neutron emission until the energy level is below the threshold for this mode of de-excitation. Prompt neutron emission occurs 10^{-17} s after scission (thermal neutron fission). The fragments can then de-excite further by gamma-ray emission these being prompt gammas, leaving the fission fragments in either their ground state or in a long lived isomeric state.

The prompt neutron emission brings the fission fragments closer to the beta line of stability from their neutron rich formation. The fission products are not usually stable nuclei and decay by β emission to reach the bottom of the beta stability valley which can be followed through isobaric mass chains. The initial de-excitation by prompt neutron evaporation does not occur because of the neutron rich effects, but is preferential to de-excitation by proton emission which is hampered by the Coulomb barrier.

The analysis of a large number of fission events shows a wide variety of products giving a mass distribution which is asymmetric for thermal neutron fission.

Figure 1.4 shows the mass distribution for thermal fission of several nuclides. It can be seen that symmetric fission is much less frequent, this being illustrated by the peak to valley ratio of approximately 600 in ^{235}U thermal fission. The nuclear shell effect can also be seen in fig. 1.4, the high mass peak remaining relatively unchanged as the fissioning nucleus' mass is increased. The left hand side of the peak being stable as 'A' changes, this corresponds to the 'magic numbers' of protons and neutrons ($N=82$, $Z=50$) in closed shells.

The mass distribution also changes with the excitation energy of the nucleus. This is illustrated in fig. 1.4, as the energy of the incident nucleon increases the amount of symmetric fission also increases. The single peak distribution grows wider and less symmetrical, starting to bulge towards the low mass number side.

As well as fission of the heavy elements with thermal neutrons nuclei can be induced to fission if given sufficient excitation energy. For example the neutron induced fission of ^{238}U occurs when the kinetic energy of the neutron is 1.1MeV or above^(1.5) (fast fission). Fission induced by protons starts at an energy sufficient to overcome the Coulomb barrier which is approximately 10MeV^(1.6) in high atomic number elements.

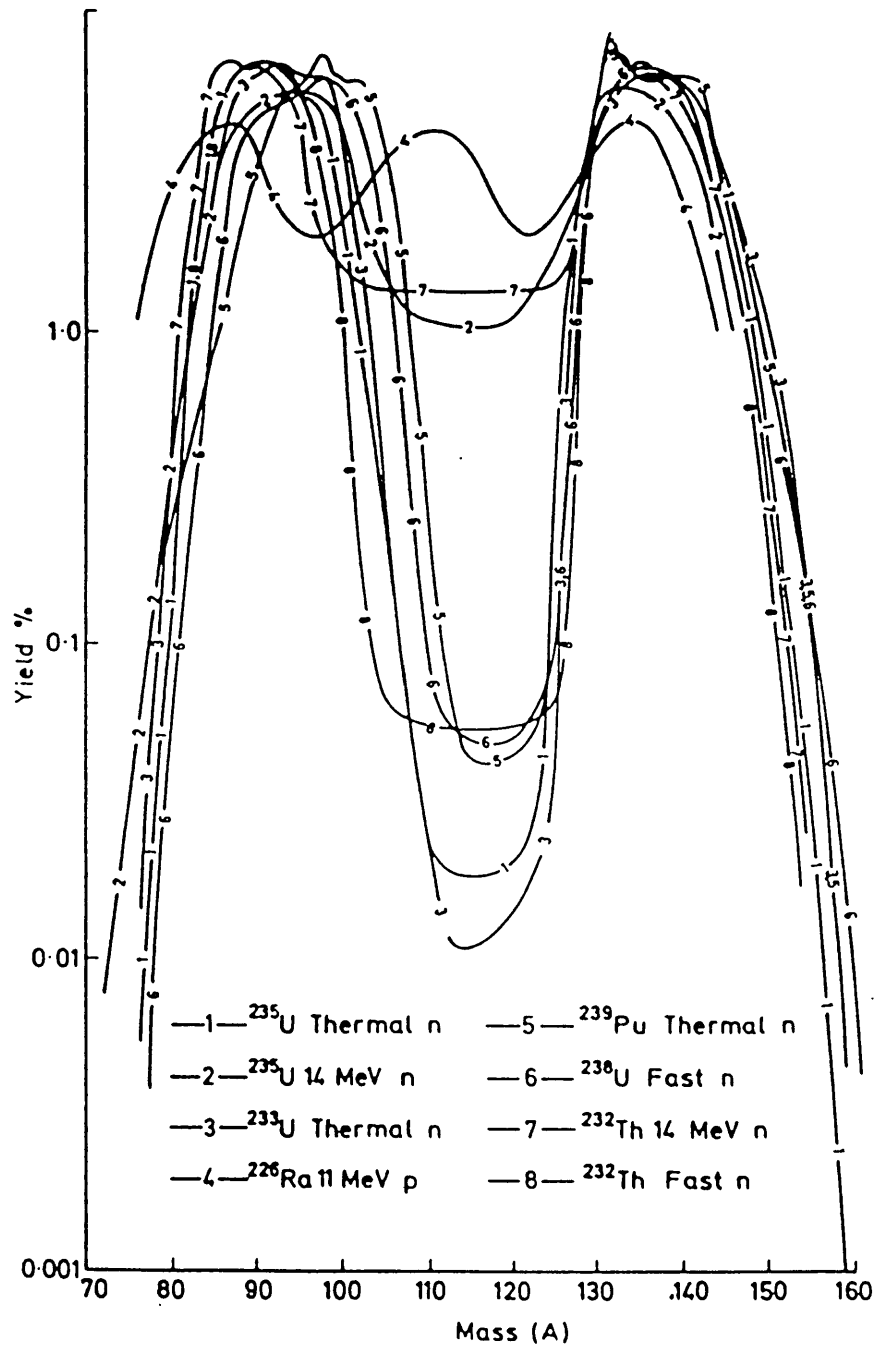


Fig. 1.4 Mass Distribution of Fission Products
(Variation with neutron energy)

In high energy proton induced fission, the distribution of fission products is not essentially determined by the fission process itself as particle evaporation from the fission fragment in its excited state alters the distribution. Proton induced fission is discussed along with spallation in section 1.5 as these reactions occur together.

1.5 SPALLATION REACTIONS (General theory)

Spallation is a term used to describe nuclear reactions in which many nucleons or fragments are emitted from the target nucleus^(1.7). As previously mentioned the energy necessary to cause the ejection of nucleons must be greater than that for nucleon separation. Consequentially it is implied that the nucleus is struck by high energy particles. The limiting energy for the interaction of protons to produce the emission of nucleons is the sum of the average Fermi energy for the nucleon plus the binding energy of the most loosely bound nucleon and the Coulomb energy for a proton at the nuclear surface. This is approximately 50.9 MeV for ^{238}U (1.8). (Where the Fermi energy is the kinetic energy of the highest filled state (Pauli principle) of the nucleon in the Fermi gas model of the nucleus^(1.9)).

Reactions at high energy are significantly different to those at low energy. The cross-section for the production of a nuclide from low energy interactions

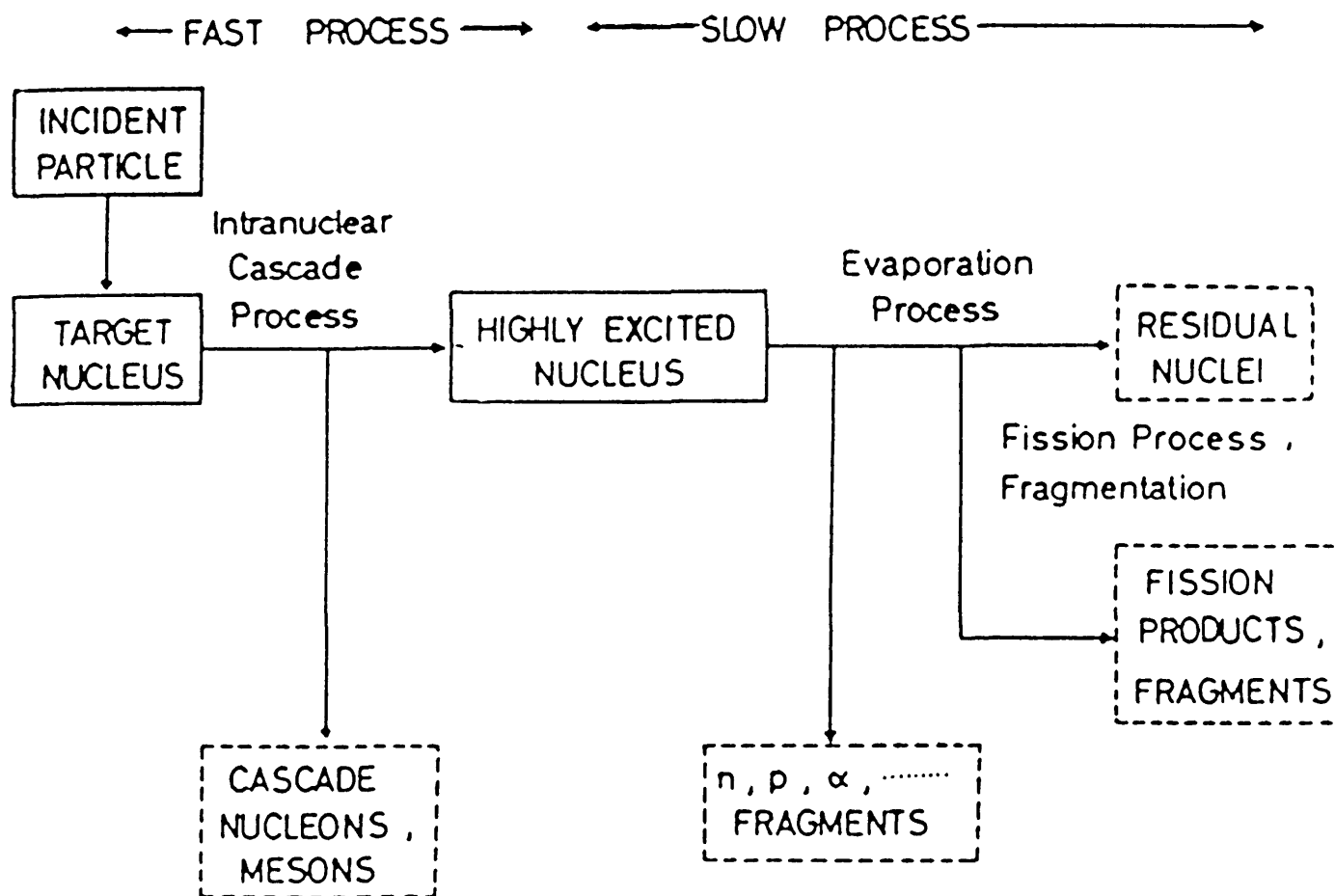
increases with energy to a well defined sharp maximum (resonance absorption) at a particular energy, this being interpreted as the formation of a compound nucleus. At high energy the variation of cross-section with energy becomes more slowly varying, which is interpreted as evidence of a two step reaction (1.10).

In spallation studies of heavy target materials it should be noted that high energy fission is an important reaction. It has been shown that fission occurs to an appreciable extent at GeV energies (1.11) even in light nuclei such as silver^(1.12), and in uranium and the trans-uranics fission accounts for about 90% of the total cross-section.

Figure 1.5 shows a schematic representation of the process of nuclear interactions at high energy. There is seen to be a two step mechanism : a fast cascade of nucleons and fragments followed by a slower step of evaporation of nucleons in competition with fission. This mechanism was proposed by Serber in 1947.

In the initial interaction phase the incident high energy proton strikes the target nucleus and ejects a number of nucleons by direct interaction leaving a residual excited nucleus. This is followed by the de-excitation through particle and photon emission or through the break up into two or more fragments (fission) leaving the final product nuclei.

Fig. 1.5



The initial phase leaves a variety of residual nuclei with a spectrum of excitation leading to a wide number of products.

1.6 CASCADE STAGE-KNOCK ON PHASE

Serber^(1.13) proposes that the collision time of incident proton with a nucleus is assumed to be small compared with nucleon-nucleon interactions within the target, thus the effect can be seen as a succession of two body collisions between the incident proton and the individual nucleons. The associated wavelength^h of the particle is less than the average distance between nucleons and the momentum of a nucleon in the nuclear potential well is much less than that of a high energy incident nucleon. The nucleus can be thought of as being a semi-transparent gas through which the particle traces a trajectory with a defined mean free path (as molecules in a gas). The struck nucleons can be treated likewise, and the interactions can be analysed in terms of free protons and neutrons within the target nucleus.

A proton striking the nuclear surface will be able to penetrate to some depth and collide with either a proton or neutron. The incident proton generates an intra-nuclear cascade which is stochastic. Each step can be described by the properties of the nucleon-nucleon collision, the two collision partners may

escape from the nucleus or undergo further collisions. As a consequence of these events a number of protons and neutrons may escape from the nucleus, but others will not have gained sufficient energy to escape the nuclear potential well. The energy they have gained will remain as nuclear excitation which effect the evaporation stage.

The progression of the cascade step is governed by the mean free path of nucleons in nuclear matter and by the Pauli exclusion principle. The effective size of the fast incident proton (wavelength $\lambda = h/p$ the de Broglie wavelength) is assumed to be small compared with the inter-nucleon distance within the nucleus, and its kinetic energy large compared to the nuclear potential of the target nucleons. [These criteria are met by protons with energies in excess of 100MeV].

Fernbach, Serber, Taylor^(1.14) treating the interactions classically have shown that the inelastic nucleon-nucleon scattering cross-section for a nucleus with a well defined boundary can be expressed as follows:

$$\sigma = \pi R^2 \frac{1 - (1 + 2R/\lambda) \exp(-2R/\lambda)}{2(R^2/\lambda^2)}$$

Where R = the nuclear radius

λ = the mean free path of the incident particle

Serber however pointed out that in some respects collisions cannot be treated as free particle interactions. The effective cross-section differing from the free nucleon approach because of the Pauli exclusion principle which makes occupied states inaccessible after scattering, that is a collision is not allowed unless both partners involved are able to end up in an unoccupied level in the nuclear potential well. Therefore the presence of other target nucleons influence the cascade, such that the collision partners have to leave the collision with an energy greater than E_f , the highest filled level in the well [where E_f is the Fermi energy of the nucleon].

The nature of the cascade interaction lends itself to Monte-Carlo calculations using the previously mentioned assumptions and the nuclear model of a Fermi momentum distribution of nucleons in a uniform density nucleus with a square potential well. [Monte-Carlo methods are discussed in section 1.8].

1.7 EVAPORATION STEP

The residual nucleus ,after the emission of particles in the cascade step, is left with a certain amount of excitation energy. The maximum amount of energy deposited in the nucleus is the sum of the kinetic energy of the incoming particle and its binding energy. The cascade of nucleons generally leave less than this, and a wide number of nuclei with a range of

excitation energies will be left (the different numbers of particles emitted in the cascades remove different quantities of the initial excitation as kinetic energy). When the excitation energy is greater than the binding energy of the least tightly bound nucleon de-excitation occurs by the emission of that nucleon or the fissioning of the nucleus.

The principle of de-excitation by particle emission is treated in the same manner as that consequent on the formation of a compound nucleus by amalgamation of an incident particle and nucleus.

In the formation of a compound nucleus as described by Neils Bohr^(1.15) the sum of kinetic and binding energies of the absorbed particle is represented as excitation energy. It is assumed that this energy is rapidly distributed in a random manner among all possible configurations of the system (because of the incoming nucleon-target nucleon collisions), this leaves none of them with sufficient energy to escape immediately. It is at this stage that the post cascade residual nuclei can be treated as a compound nucleus. The compound nucleus remains intact until sufficient energy has been concentrated onto one nucleon or a cluster for escape to occur. [the decay of the excited system shows a similarity to the alpha decay of the heavy elements].

Analysis of the evaporation stage of nuclear reactions has been from a statistical assumption and from following the formula developed by Weisskopf (1.16) using the Bohr compound nucleus model. The probability of a particle being emitted increases with the increasing energy of the particle but as the particle's energy increases the excitation energy of the residual nucleus decreases.

Weisskopf's formula

$$P(E_j) dE_j = \sigma(E_j) [g_j m_j E_j / \{\pi^2 (h/2\pi)^3\}] [\omega(E_f) / \omega(E_i)] dE_j$$

Where $P(E)$ is the probability of a nucleus with excitation energy E_i will emit a particle of energy E_j .

$\omega(E_i)$ and $\omega(E_f)$ are the initial and final level densities of the nucleus which are functions of mass, charge, and excitation energy.

$\sigma(E_j)$ is the cross-section for the formation of a compound nucleus in the inverse process (absorption of the evaporated particle).

$\sigma(E_j)$ is an unknown quantity but is thought not to be too different from the capture cross-section for the particle by a nucleus in its ground state. For neutrons with energies above the thermal region πr^2 , the geometric cross-section is used. This has to be corrected for the effect of the Coulomb barrier in the case of protons.

The calculation of the level densities $\omega(E)$ as a function of excitation energy is made using the Fermi gas model of the nucleus and this gives the particle evaporation probability:

$$P(E_j)dE_j \propto \sigma(E)E_j \exp(-E_j/T_f) dE_j$$

Where T_f is the 'nuclear temperature' which is a measure of the population of higher states in the Fermi gas. The maximum value of $P(E_j)$ for neutrons is when E is equal to the nuclear temperature. In the case of protons a higher value of E is necessary due to the effect of the Coulomb barrier on σ_c . The energy of nucleons from the evaporation stage is much lower than cascade nucleons.

As the excitation energy of the nucleus increases the mean lifetime of the nucleus between emissions decreases but the time for the equilibration of the energy decreases in this manner the emitted particle will be affecting the following particle^(1.15). Analysis of the evaporation step of highly excited nuclei by statistical methods does not explain the way that the characteristics of the "nth" emitted particle is not independent of the "(n-1)th" particle. This would indicate no real sharp separation of the two processes of knock-on and evaporation.

1.8 MONTE-CARLO METHODS

This mathematical method of analysis can be applied to the calculations of both stages of the spallation reaction. This method follows the history of the interaction of an incident particle with a nucleus where the affect of the interaction is governed by a choice of numerical values which determine the outcome. The outcome of such a calculation has many possibilities for one interaction it is therefore necessary to follow a large number of interactions to acquire reliability. Also the manner in which the initial values are chosen will influence the outcome.

Collision details between an incident proton and a nucleon are decided by the choice of random numbers which express the relative probabilities for the possible interactions. Each parameter of the interaction is determined by a random number: one number decides the point of impact of the proton another the depth within the nucleus at which the first collision occurs. The choice of collision partner and momentum are also chosen in the same manner. From these parameters the nature of the collision can be determined and a decision made on whether it is allowed by the Pauli exclusion principle. If it is allowed the results calculation continues for both of the collision partners using random numbers to describe their progress. If the collision is not allowed then the progress of the proton is followed through undisturbed until it escapes the nucleus or collides.

CHAPTER 22.1 EXPERIMENTAL PROCEDURES

The chemistry of the actinides in solution is complicated by the multiplicity of oxidation states they are capable of exhibiting. The chemical properties of these elements are different for each different valency state, whilst in the same valency they closely resemble each other.

The existence of several stable oxidation states is of great importance in the analytical chemistry of these elements. The selective maintenance of a specific state of the element in question can be used advantageously in separation techniques. A particular actinide can be obtained in a desired oxidation state by the appropriate use of a mixture of oxidising and reducing agents, enabling a mixture of the elements to be separated by choosing a combination of reagents that will hold the desired nuclides in the oxidation state for the particular separation process. The others are maintained in an unextractable state. Figure 2a shows the main oxidation states and redox potentials of the actinides.

Fig. 2.1a

OXIDATION REDUCTION POTENTIALS FOR THE ACTINIDESVALENCY STATES IN DILUTE HClO₄

Th⁴⁺ ----- -1.9V----- Th

|----- +0.32V-----|
 UO₂²⁺ ----- UO₂⁺ ----- U⁴⁺ ----- U³⁺ ----- U
 |--- +0.063V---|--- +0.058V---|--- -0.631V---|--- -1.80V---|

|----- +0.938V-----|--- +0.155V---|--- 1.83V---|
 NpO₂²⁺ ----- NpO₂⁺ ----- Np⁴⁺ ----- Np³⁺ ----- Np
 |--- +1.137V ---|----- +0.739V -----|

|----- +0.677V-----|
 |----- +0.677V-----|

|----- +1.0433V-----|--- +0.982V---|--- -2.03V---|
 PuO₂²⁺ ----- PuO₂⁺ ----- Pu⁴⁺ ----- Pu³⁺ ----- Pu
 |--- +0.913V---|--- +1.172V---|
 |----- +0.677V-----|

For the separation of plutonium, neptunium, uranium and thorium; methods based on coprecipitation, ion-exchange and solvent extraction are the most important. The coprecipitation of the large 3+ and 4+ ions in milligramme quantities using 35mg of barium sulphate has been used by C.W. Sills et al. (2.1-2.4) to give recoveries in excess of 99% in the presence of potassium ions. With a decrease in charge or ionic size there is decreased recovery, and all mono- and divalent ions show little tendency to coprecipitate.

The mechanism of the coprecipitation from this medium is unclear but the dependence of the recovery on charge and size points to the isomorphous replacement of the barium in the crystal lattice. The increase in the recovery in the presence of potassium and the conservation of charge are more easily explained by the formation of a compound, for example: $K_2Ba[Th(SO_4)_4]$. The precipitation of uranium through to americium can be prevented by prior oxidation to their highest valency states. This gives a convenient method of eliminating the bulk uranium of the target material in this study, enabling the trace quantities of the higher actinides to be identified.

The $BaSO_4$ precipitate is dissolved and the actinides then reprecipitated as the hydroxides. This new precipitate is filtered and washed on a micro-porous membrane. The hydroxide substrate is sufficiently

thin to allow high resolution alpha spectra to be obtained (FWHM 27.7 KeV).

The ion-exchange behaviour of the actinides in acid media is shown in figures 2.1b and 2.1c. These show the distribution coefficients for some oxidation states of the elements from nitric and hydrochloric acids on the strong base anion exchange resin DOWEX-1. The varying strengths of absorption allows the separation of the actinides from each other and from fission products. Thus it can be seen that uranium and fission products can be separated from plutonium (IV), neptunium, and thorium(IV) in 8 molar nitric acid. From hydrochloric acid concentrations above 8 molar thorium(IV) can be separated from plutonium(IV).

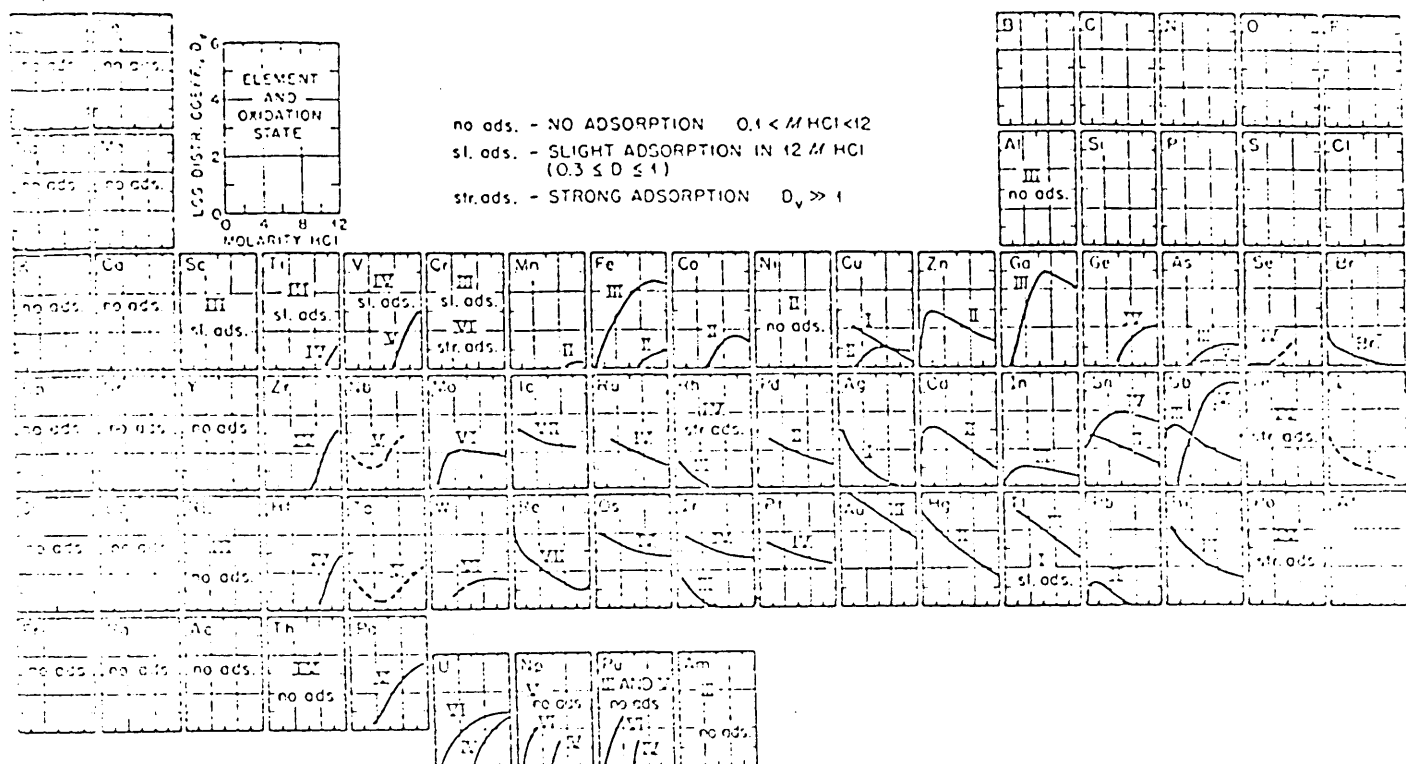
In high concentrations of nitric acid thorium forms a stable anionic nitrate $[\text{Th}(\text{NO}_3)_6]^{2-}$ which is strongly absorbed onto strong base anion resins (5 < 10 molar acid solutions). Few other metal ions are absorbed under these conditions but these include Np(IV), Pu(IV), Pa(IV), Ce(IV) and Bi(III). Plutonium (IV) forms the anionic complexes $[\text{Pu}(\text{NO}_3)_6]^{2-}$ and $[\text{Pu}(\text{NO}_3)_5]^-$.

The stability constant of the hexanitrate complex of plutonium (IV) is considerably higher than that of thorium and it is appreciably absorbed from 1 molar

acid; thorium showing a similar absorption only in strong acid media. The hexavalent states of plutonium and uranium are only slightly absorbed. The trivalent state shows no absorption at all. Therefore by selectively maintaining the tetravalent state of plutonium the maximum absorption can be obtained from nitric acid.

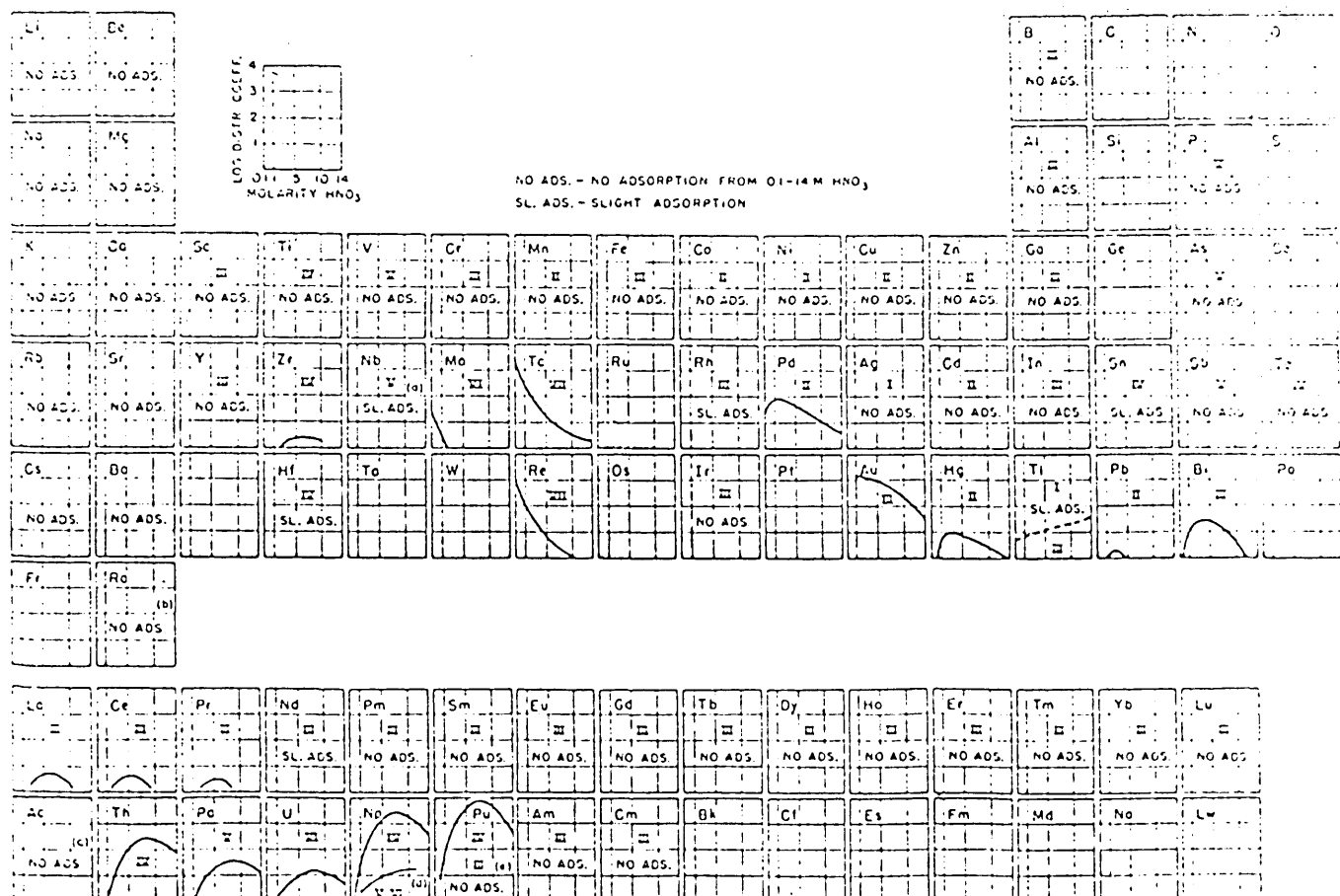
In hydrochloric acid at high concentrations uranyl ions form complexes of the composition $(\text{UO}_2\text{Cl}_3)^-$ and $(\text{UO}_2\text{Cl}_4)^{2-}$ which are strongly absorbed on strong base anion resins in the chloride form. Their absorption coefficients increase to $<10^3$ in acid above 8 molar. From figure 2.1b it can be seen that the trivalent state of trans-uranium elements and thorium(IV) are not absorbed whilst the chloride complexes of plutonium (IV) above 8 molar are.

After loading a preconditioned resin (DOWEX-1) with the sample (with Pu adjusted to Pu(IV)) in 8M nitric acid, uranium and fission products can be eluted with several resin bed volumes of 8M nitric acid. This leaves Pu(IV) and Th(IV) with a small quantity of uranium on the column. Eluting the column with several bed volumes of concentrated hydrochloric acid changes the acid medium and releases the thorium which shows no absorption from this medium.



Adsorption of the elements from HCl solutions by a strong-base anion-exchange resin, Dowex-I, as a function of HCl molarity. [From Kraus and Nelson (124); reproduced with permission of United Nations Publications.]

Fig. 2.1a



Adsorption of the elements from HNO_3 solutions by a strong-base anion-exchange resin, Dowex-I, as a function of HNO_3 molarity. [From Farris and Buchanan (138); reproduced with permission of *Analytical Chemistry*.]

Fig. 2.1b

Plutonium (IV) remains strongly absorbed, together with the remaining uranium (IV) and (VI), which have distribution coefficients $>10^3$. Changing the oxidation state of plutonium from (IV) to (III) in situ then enables the plutonium to be recovered (no absorption of tervalent ion) while decontaminating it from the remaining uranium. This is achieved using ammonium iodide in concentrated HCl as the eluent (the iodide being oxidised to iodine while reducing the plutonium).

Like plutonium, neptunium exhibits several oxidation states in aqueous solution but it has a stable (V) state which disproportionates to (IV) and (VI) only at high acidities. The partial reduction of Np(IV) to Np(III) can be achieved in the presence of hydrazine and formaldesulphoxalate in dilute HNO_3 , but Np(III) is generally of little practical importance for analysis under ordinary chemical conditions. Neptunium (IV) can be maintained quantitatively in dilute acid solutions with relatively strong reducing agents (Fe(II) ; H_2O_2). If the acid concentration is greater than 3 M, Np(V) and Np(VI) are rapidly reduced to Np(IV) by peroxide. Sulphates, chlorides, nitrates and thiocyanates form stable complexes which are useful in ion-exchange. The anionic chloride complex of Np(IV) is absorbed from strong HCl solutions and it has been shown by J.L.Ryan^(2.5) that $\text{Np(NO}_3)_6^{2-}$ is absorbed from strong nitrate solution. Neptunium

follows the path of the plutonium in the separation methods discussed in this thesis, except in the ion exchange where the neptunium does not elute with the plutonium. Table 2.1 lists some of the reagents that can be used to alter the oxidation state of plutonium and neptunium for analytical purposes.

The solvent extraction of the actinides into thenoyltrifluoroacetone (TTA) in benzene has been used in this study. Thenoyltrifluoroacetone is a β -diketone insoluble in neutral aqueous solutions, but readily soluble in organic solvents.

TABLE 2.1

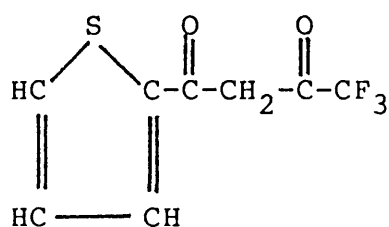
REDOX REAGENTS FOR PLUTONIUM SEPARATION

REAGENT	SOLUTION	REDUCTION
I^-	11M HCl	Pu(IV)-Pu(III)
	0.1M NH_4I	
NO_2^-	HNO_3	Pu(VI)-Pu(IV)
		Np(VI)-Np(V)-Np(IV)
NH_2OH^+	$NH_2OH.HCl$	Pu(IV)-Pu(III)
H_2O_2	HNO_3	Pu(VI)-Pu(IV)

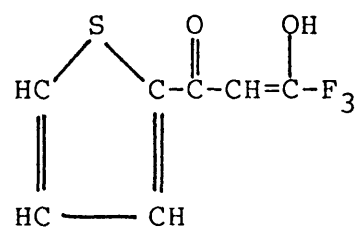
In the enol form it is highly acidic due to the trifluoromethyl group; this enables it to form stable non-ionic metal chelates at low pH which are extractable in organic solvents immiscible with water.

The reagent possesses the advantage over other complexing agents in that most of the extracts are acidic and there is a larger concentration of its anion present at the pH of extraction; and TTA complexes are stable in acid media.

The extraction of the actinides as the TTA complex involves the more acidic enol tautomer, the enol hydrogen being replaced by one of the metal ions to form the chelate.

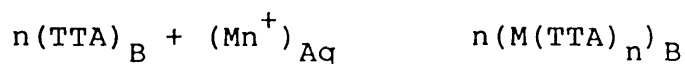


KETO



ENOL

The overall reaction of a solution of TTA in benzene in contact with an aqueous solution of an n -valent cation M^{n+} is given by :



TTA forms strong complexes with many metal ions, particularly those of high valency. The distribution coefficients show an n^{th} power dependence on the concentration of TTA, and inverse proportionality to

the n^{th} power of the aqueous acidity^(2.6). The extraction of metals is thus dependent on the pH of the aqueous medium and table 2.2 gives the $\text{pH}_{1/2}$ (this being the pH where 50% of the ion is extracted) for the extraction of the actinides.

TABLE 2.2

(Extraction)

ELEMENT	$\text{pH}_{1/2}$
Th	>1
U	3.48-8
Np	0.5
Pu	1

Due to the complexity of the solution analysed in this study, extraction of the actinides as TTA complexes was used initially as a final stage for the preparation of alpha sources. Having separated the alpha emitting nuclides into separate aliquots the final stage was the preparation of the TTA chelate giving the final decontamination from any residual interferences. The complex was then evaporated to near dryness as a thin layer on a stainless steel planchette and thermally decomposed to give a layer of oxide. By following the method of Moore and Huchens (2.7) the plutonium for example was extracted directly and was found to give greater recoveries, and resolution of the same order of magnitude as that of the more complicated ion-exchange technique.

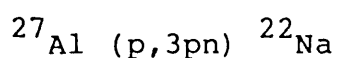
2.2 TARGET IRRADIATION & TREATMENT

Uranium targets of natural isotopic composition in aluminium holders were irradiated with protons. 2.2a shows the detail of the target construction.

The first irradiation took place on the 6th of June 1981, the target consisted of three discs of uranium having a total mass of 55.30 grammes, and dimensions : 25.4mm diameter 6.08mm total thickness. The target was irradiated on the CERN 'booster' synchro-cyclotron transfer line with 800 MeV protons. The irradiation took 364 minutes with 1.9×10^{13} protons per pulse and a flux density of 1.8×10^{16} protons per cm^2 giving a total of 9.12×10^{16} protons onto the target. [Figure 2.2b shows the irradiation configuration schematically].

Subsequently a second target was irradiated on the 24th of February 1983. The irradiation took place in the same configuration as the first target on the CERN 'booster'. The target mounting was identical to that in the first irradiation but consisted of only two discs (2.45mm x 0.43mm) with a total mass of 38.9 grammes. It recieved an integrated beam of 4.3×10^{16} protons onto the target from a beam of 10^{13} protons per pulse every 2.4 seconds for 365 minutes, equivalent to 8.486×10^{15} protons per cm^2 .

The beam was aligned for the irradiations on a scintillation screen viewed by television camera. Beam profile measurements were made with multi-wire chambers. The intensity of the beam was controlled by the beam monitoring transformer, and the integrated beam was calculated from sodium-22 production in aluminium foils centred on the target. A 14 millibarn cross-section for 800 MeV protons was assumed, for the nuclear reaction:



The target discs were dissolved in concentrated nitric acid to give solutions of the concentrations in table 2.3 . For alpha spectrometric analysis small samples were taken to ensure that sources of small mass were produced, to achieve the resolution necessary for quantitative measurements. Due to the short range of alpha particles it is extremely important to make clean sources. [See chapter 3]. The small samples taken minimised the contamination of the detector from recoil daughter nuclides, which can be a problem when analysing nuclides with a reasonably high specific activity e.g. $^{236}\text{Pu} - 5.3 \times 10^8 \text{ pCi}_\mu\text{g}^{-1}$.

TABLE 2.3

Target 1	Disc 1	19.28g
	Disc 2	16.18g
	Disc 3	19.93g
Target 2	Disc 1	19.22g
	Disc 2	17.25g

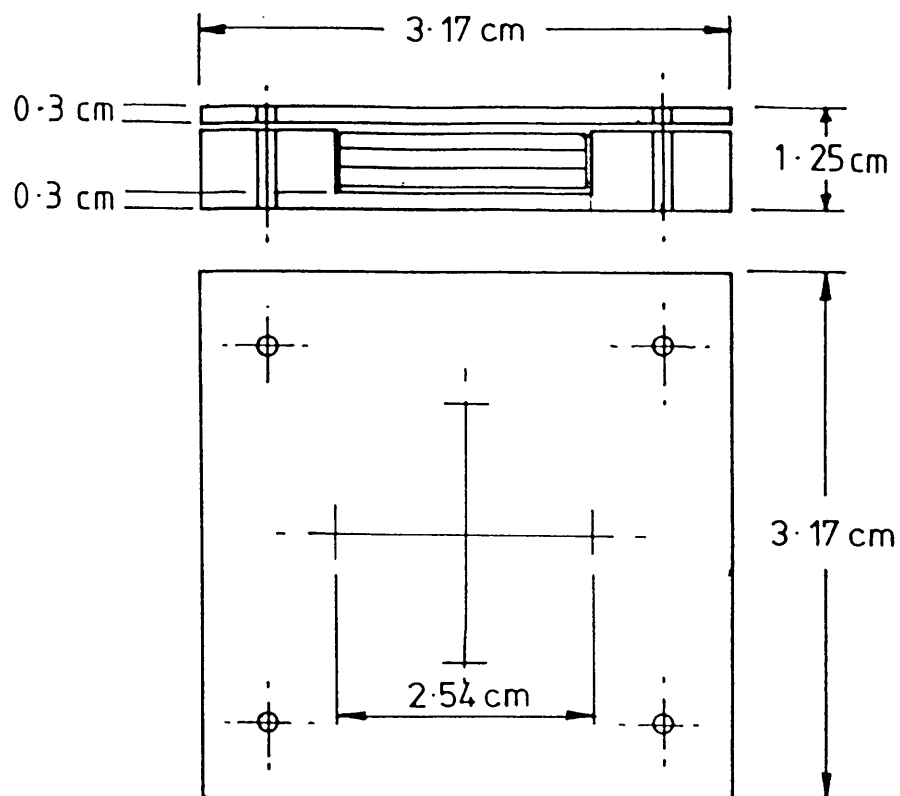


Fig. 2.2a Target construction

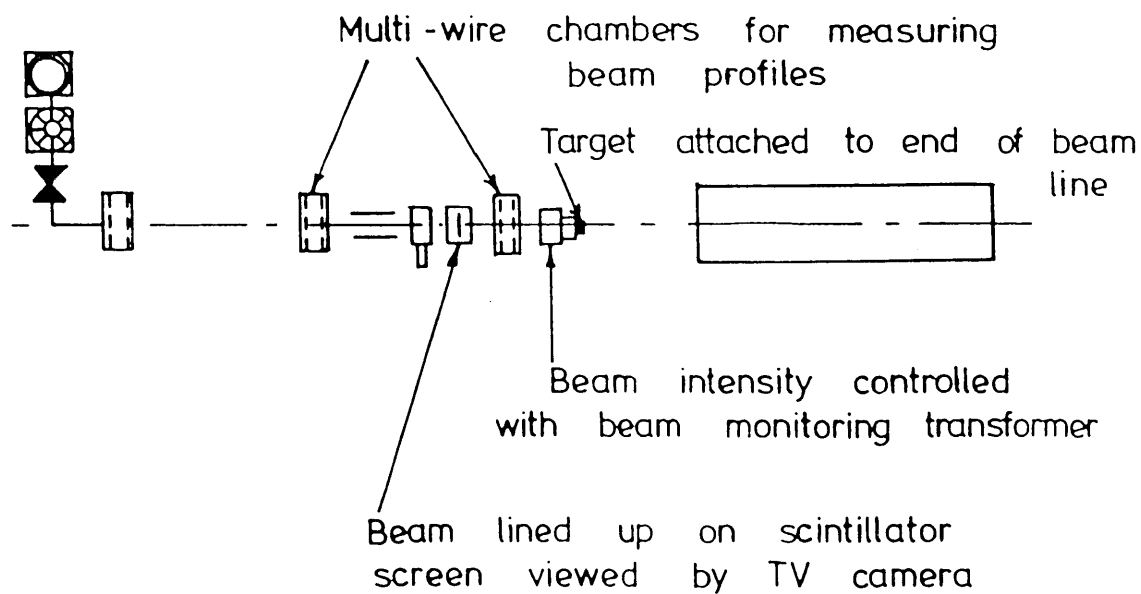


Fig. 2.2b Schematic irradiation configuration

2.3 ANALYTICAL METHODS FOR DETERMINATION OF ALPHA EMITTERS

The irradiated samples were analysed for the alpha emitting isotopes of: plutonium, neptunium, uranium, thorium and polonium.

Various combinations of the analytical techniques previously mentioned have been used and compared, so as to obtain spectra with the maximum resolution and the best yields. The combinations examined were:

- i Co-precipitation with barium sulphate and as hydroxides(gross α analysis).
- ii Direct extraction into thenoyltrifluoroacetone (benzene) for separation and source preparation.
- iii Ion-exchange, and solvent extraction as TTA complex for source preparation.
- iv Ion-exchange and electroplating from sulphate medium for source preparation.

To evaluate the percentage recovery of the nuclides being chemically separated from the bulk material, each sample analysed had a small known quantity ($<12\text{pCi}$) of a tracer isotope added. For plutonium three different tracers were used ^{238}Pu , ^{239}Pu , and ^{242}Pu , for thorium ^{232}Th and for neptunium ^{237}Np were

used. These standard tracers were added to each sample analysis with the exception of those done as gross alpha analysis. All the reagents used were of analytical grade and blank analyses were undertaken to ensure that there was no reagent contamination.

2.3a GROSS ALPHA ANALYSIS

The initial analysis of the target material was achieved by coprecipitation of the actinides with barium sulphate and as hydroxides as outlined by C.W.Sills and R.L.Williams^(2.8). These methods were developed for the analysis of environmental samples and in this study the method has not been changed.

To an aliquot of dissolved target material in a conical flask; 3g of potassium sulphate, 3ml of concentrated sulphuric acid and 6 drops of concentrated nitric and perchloric acids were added and heated and evaporated to fumes. The flask was heated over a high temperature blast burner whilst swirling continuously, until a glassy melt was obtained (pyrosulphate fusion). The melt was then heated at maximum temperature to volatilise any excess acid, and was allowed to cool to room temperature.

0.5ml of concentrated sulphuric acid, 35mls of distilled water were added and the solution boiled on an electric hotplate. To the boiling solution 1ml of 30% hydrogen peroxide was added to oxidize the uranium and to ensure complete reduction of the plutonium.

1ml of a 0.45% solution of barium sulphate was added drop-wise to the boiling solution whilst being stirred (PTFE covered magnetic stirrer). The solution was boiled for one minute before another 1ml of barium solution was added. The solution was cooled down in a bath of running cold water, transferred to a centrifuge tube using several aliquots of water giving a total volume of approximately 30ml and centrifuged at about 1500 rpm for 5 minutes. The supernate was discarded and the precipitate washed with distilled water and centrifuged.

The precipitate still in the centrifuge tube was suspended in 0.5ml of water 6mls of 0.05 M alkaline ethylenediaminetetraacetic acid added and mixed and the the solution was heated on a hot water bath for 5 minutes until clear. 0.05mg of cerium carrier was added as acidified cerous nitrate (0.1 M HNO_3) and 20 microlitres of a 25% solution of hydrazine dihydrochloride was added along with 2mls of 10 M sodium hydroxide. The solution was mixed and digested at boiling for 10 minutes. The resulting hydroxide precipitate formed was barely visible but gave the solution an opalescent effect.

When cool, the finely divided precipitate was mounted, for counting, on a 25mm Tuffryn HT100 0.1 micron pore membrane (Gelman Sciences). The membrane was placed on a stainless steel support in a demountable

polysulphone filter funnel, wetted with ethanol (A.R.), and the suspension applied whilst under suction, in such a manner that an even distribution was obtained. This was achieved by applying the first 2mls of suspension and sucking the membrane dry to plug the filter as tightly as possible. The centrifuge tube was washed with a few millilitres of 1 M NaOH and the funnel and precipitate were washed three times with 5ml portions of ethanol. The chimney of the funnel was removed without interrupting the suction (no disturbance of precipitate) and the membrane filter was dried under an infrared lamp. The dried precipitate was positioned on a stainless steel holder and counted on a 100 mm² high resolution surface barrier detector.

The results from this analysis are shown in figures 2.3. These are qualitative alpha spectra of those actinides precipitated. No plutonium, neptunium, or thorium standards were available at the time. From these spectra it can be seen that a high degree of resolution is obtainable, as exemplified by the two ²²⁸Th peaks at 5.43 MeV and 5.34 MeV. The spectra shows an average FWHM of 27.7 KeV, but there is still a little masking of the plutonium peak at 5.5 MeV by the thorium daughters. This peak would have been more easily distinguished if a smaller sample had been taken reducing the quantity of ²²⁸Th and its daughters which interfere by producing a background on which the peaks of interest now sit.

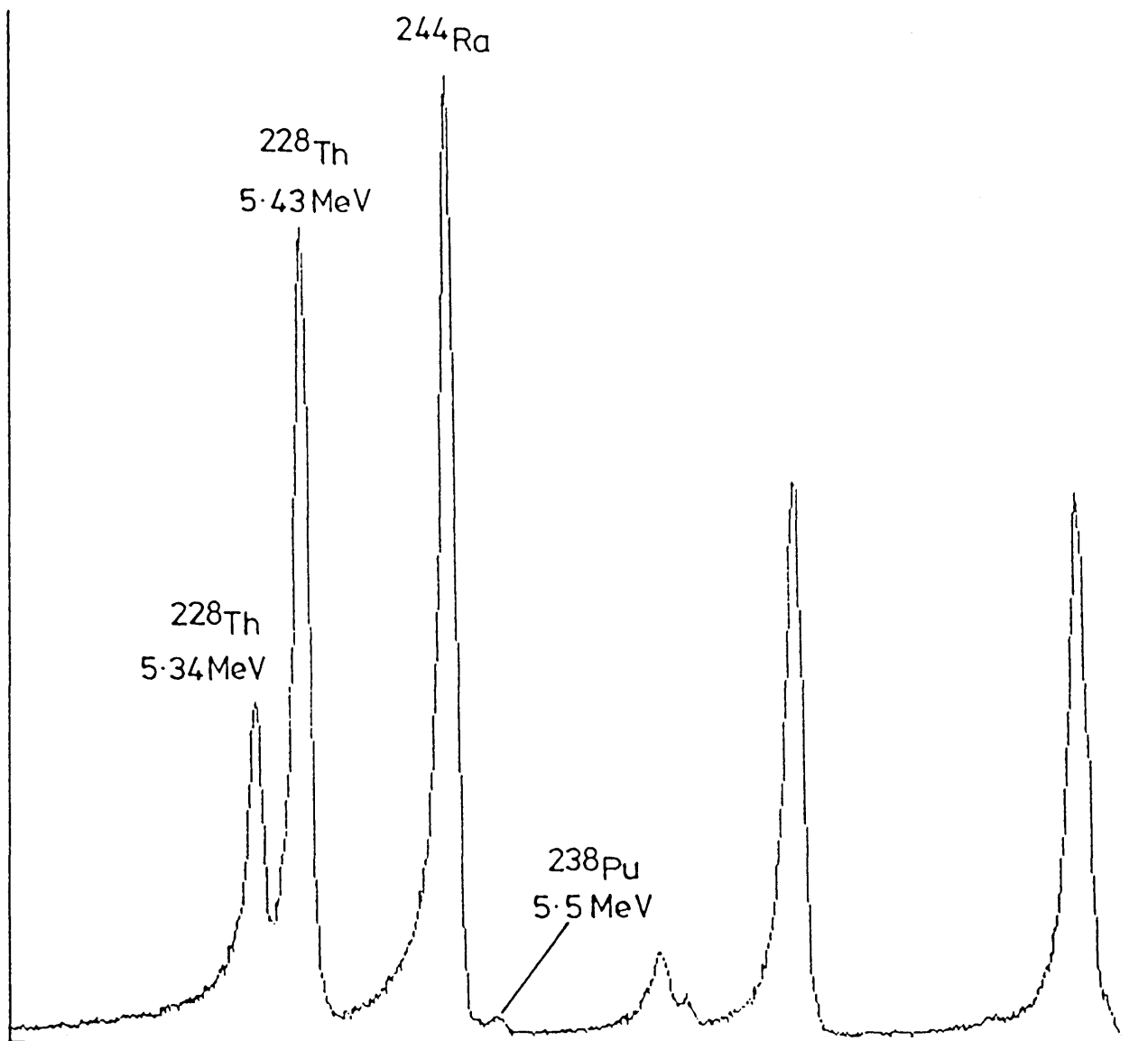


Fig. 2. 3a Alpha spectra co - precipitated on barium sulphate

2.3b EXTRACTION OF PLUTONIUM AND NEPTUNIUM AS TTA COMPLEXES

i) PLUTONIUM

The method followed was one outlined by F.L.Moore and J.E.Hudgens (2.9). An aliquot of the dissolved target in a 10ml beaker had 50 microlitres of plutonium stock solution (supplied by A.E.R.E. Harwel) of precisely known activity (see chapter 4 -results and analysis) was taken and heated carefully to just below boiling, on a hotplate, until dry. The residue was dissolved in about 3mls of 2 M HNO_3 , and 1ml of 1 M hydroxylamine hydrochloride. The solution was heated for about 5 minutes at 80° , and the volume was adjusted to 4mls to make up any losses from evaporation during heating.

The solution was transfer⁶ed to a 50ml separating funnel, and the beaker rinsed with two 1ml portions of 1 M sodium nitrite. The resulting solution was mixed thoroughly and allowed to stand until all effervescence had subsided. [The mixture was approximately 1 M with respect to HNO_3].

3mls of 0.5 M thenoyltrifluoroacetone in benzene (A.R) was added to the funnel, and the solution mixed using a mechanical shaker for at least 10 minutes for the extraction of the complex. The two phases were left to

disengage and the aqueous layer was discarded. The organic phase containing the plutonium was washed several times with an equal volume of 1 M HNO_3 , these were discarded.

The organic solution of TTA(benzene) was run into a 5ml 'narrow ended' centrifuge tube, the separating funnel was rinsed with 1ml of benzene and this added to the organic complex. The solution was then centrifuged at around 1500 rpm to separate any entrained aqueous phase (which would cause sputtering in the next stage). The organic complex was then pipetted into a 5ml beaker and the solvent (benzene) was evaporated down to <0.5ml on an electric hotplate at low temperature.

The plutonium-TTA complex was then transferred to a clean (ethanol washed) stainless steel disc, 12.7mm diameter, on an electric hotplate using a clean disposable Pasteur pipette. The complex solution was slowly evaporated to dryness. The disc was then flamed over a bunsen burner to fix the plutonium, as the oxide, to the surface and to decompose the thenoyltrifluoroacetone giving a source of minimum mass 10 μ g for counting. [Several individual aliquots were taken through this separation method for evaluation of the reproducibility]

ii) NEPTUNIUM

Analysis of the long lived alpha emitting isotope ^{237}Np was undertaken using the method described by F.L. Moore^(2.10), whereby the plutonium is reduced and held in the tervalent oxidation state by potassium iodide, KI, and the pentavalent neptunium is reduced to the extractable tetravalent state using hydroxylamine hydrochloride, HONH_3Cl . Several aliquots of the irradiated uranium were taken, and to half of these a small amount of a standard solution of ^{237}Np was added as a monitor of the analytical technique, as no other standard was available.

An aliquot of target (in nitric acid) was metathesised to the chloride form, in a 10ml beaker, by evaporating to dryness below boiling in the presence of concentrated acid. The residue was then taken up in 1 M HCl and made 1M with respect to HONH_3Cl and 2 M with respect to KI, covered with a watch glass (to reduce evaporation) and heated to approximately 80°C for 20 minutes. The solution was allowed to cool to room temperature and transferred to a 100ml separating funnel. The neptunium was extracted as the TTA complex into a 0.5 M solution of TTA in benzene. The phases were separated and the organic complex washed with 1 M HCl then treated as per the plutonium (sect. 3.3ii(a)) for the preparation of the alpha source. [All reagents used were of analytical grade and supplied by B.D.H.].

2.3c ION-EXCHANGE

i) PLUTONIUM

The plutonium was separated from other elements using a strong anion resin; solvent extraction and electro-deposition were used as methods of source preparation after decontamination from other elements. Again each analysis was repeated several times for the purpose of establishing reproducibility. Although in this method three different fractions containing uranium, thorium and plutonium are possible from each aliquot it was decided to analyse for each element individually.

Aliquots of the irradiated sample solution were taken roughly equivalent to 40 milligrammes. A precisely known amount of tracer isotope was added, in the case of plutonium, this was initially ^{238}Pu . Using this tracer the isotope ^{236}Pu was identified, and it was noted that the sample contained an undetectable amount of ^{239}Pu . Subsequent samples were spiked with ^{239}Pu tracer this enabled the identification of the presence of the isotope ^{238}Pu in the sample, a spallation product.

The samples for analysis in 50ml beakers, were made 8M with respect to nitric acid by dilution; or evaporated to dryness below boiling and taken up in 8 M HNO_3 . 10mls of 6% hydrogen peroxide was added and heated

gently until all the peroxide had decomposed, and the volume was adjusted so that the acid concentration was again 8M. This reduced the plutonium to the tervalent oxidation state. The solution was allowed to cool then a few milligrammes of sodium nitrite were added to oxidise the Pu(III) to the four valent state. The solution was left until all gas evolution had stopped.

This solution was passed through an ion-exchange column containing around 5mls of anion-exchange resin (DOWEX-1X8 100-200 mesh) which had been pre-conditioned with 8 M HNO_3 . The flow rate was approximately 1ml per minute. The beaker used for the redox preparation was rinsed out with three 1ml portions of 8 M HNO_3 , and these were added to the top of the resin bed. The solution was drained to the top of the resin column, and then eluted with 50mls of 8 M HNO_3 and drained to the resin top. [This eluent contained the uranium and fission products.]

The column was then eluted with several column volumes of 12 M hydrochloric acid (50mls). This changes the resin bed from the nitrate form to the chloride form simultaneously eluting the thorium(IV) present (this has a log distribution coefficient of in the order of 2.5 in 8 M HNO_3 but shows no absorption in hydrochloric acid (fig 2.b)). Plutonium shows strong absorption in both acid media.

The plutonium was then eluted into a clean 50ml beaker using a solution of 11 M HCl, 0.05 M with respect to ammonium iodide. This solution was prepared freshly prior to use. [30mls 12 M HCl and 1.5ml 1 M NH_4I].

The plutonium eluate was slowly evaporated (below boiling), to dryness. The ammonium salts were removed by evaporating with several portions of concentrated HCl and HNO_3 (2-3mls each).

The residue in the beaker was dissolved in 2 M HNO_3 and 1 M HONH_3Cl , it was then transferred to a separating funnel and the plutonium extracted as the TTA complex. The method used was the same as that outlined in section 2.3b i) and the sources were prepared in the same manner.

An alternative preparation of the sources was tried. After the decomposition of the iodine salts the plutonium was electroplated from sulphate solution, following the method of Talvitie^(2.11), modified by J.J. Higgo.

The residue was taken up in 0.5ml of concentrated sulphuric acid, covered, and gently heated allowing the acid to reflux down the sides of the beaker for 5 minutes. The solution was left to cool, then 5mls of distilled water were added. A few drops of thymol blue indicator (0.04%) and 1ml of concentrated ammonia solution were used to make the solution basic. It was then carefully neutralised with sulphuric acid.

The solution was transferred to an electroplating cell (designed by J.J. Higgo) having a stainless steel planchette as the cathode and a 2mm diameter platinum anode. The beaker was rinsed with 0.02 M H_2SO_4 and these washings added to the cell. The pH was adjusted to between 2-3 using ammonia. The solution was plated for two hours at 12 volts D.C. with a current of 1 Ampere.

At the end of plating 1ml of concentrated ammonia was added to the cell before switching off the current. The source was washed with distilled water then acetone, dried, flamed and counted.

ii) THORIUM

Thorium was separated from uranium, plutonium and fission products by anion exchange from nitric and hydrochloric acids. The final stage was the preparation of the TTA complex as an evaporated source, as described for plutonium and neptunium.

A solution of natural thorium nitrate, 100% ^{232}Th , as the hexahydrate, $\text{Th}(\text{NO}_3)_4(\text{H}_2\text{O})_6$, was made, to give a concentration of approximately 352mg l^{-1} (0.0001 M). The activity of the solution was then determined by making an evaporated source of 100 μl pipetted directly onto a stainless steel disc. [This was repeated several times to ensure pipetting consistency]. 1ml of ^{232}Th solution was added to each sample being

analysed, equivalent to $232\mu\text{g}$ of ^{232}Th ($1\mu\text{mole}$) about 250pCi. Samples of between 20-40mg were taken and made 8 M with respect to HNO_3 and loaded onto DOWEX-1X8. The separation followed was the same as that outlined in section 2.3a. The nitrate eluate was discarded and the thorium eluate (chloride) was evaporated to dryness, below boiling, for preparation as a source.

The beaker containing this thorium residue had several aliquots of HNO_3 and HCl which were taken to dryness. The residue was then dissolved in 1ml of 1 M HNO_3 (warmed to aid dissolution). The solution was transferred to a 50ml separating funnel and the beaker rinsed with 9 mls of distilled water giving a total volume of 10mls of solution (0.1 M in nitric acid).

The thorium was extracted into 0.5 M TTA and washed with several portions of 0.1 M HNO_3 and as previously outlined the complex was transferred to a beaker and the solvent reduced to a minimum before pipetting onto a stainless steel disc as a source.

2.4 URANIUM ANALYSIS

A sample of the uranium irradiated on the CERN booster in February 1983 was analysed for any detectable changes in the ratios of ^{238}U : ^{235}U : ^{234}U , and to identify any other alpha emitting isotopes produced by spallation, such as ^{232}U and ^{230}U . No standard isotope

was added for this analysis as the concentration of the uranium solution was precisely known. From peak areas, accounting for decay branching, the ratios of the isotopes were measured and from this their concentration calculated.

A small sample of the irradiated uranium in 16 M HNO_3 was pipetted into a 100ml separating funnel and diluted with distilled water to make it 1 M. Sufficient aluminium nitrate was added to make the solution 2.7 M with respect to the salt. This gave a total nitrate concentration of 9M.

The solution was contacted with an equal volume of 4-methylpentan-2-one, thus extracting the uranium and leaving the fission products, thorium and plutonium in the aqueous phase. The uranium was then back extracted into 0.1 M HNO_3 and the pH adjusted to between 3 and 3.5 for the extraction of uranium as the TTA complex in 0.5 M TTA(benzene). The complex was then used to give the evaporated source for alpha counting on a stainless steel disc as previously described.

2.5 POLONIUM and LEAD-210 ANALYSIS

Over the period of this project samples from all three discs of the first irradiation, and samples from the first disc of the second irradiation have been analysed. The isotopes ^{208}Po , ^{209}Po , and ^{210}Po have been

identified as spallation products in the first irradiation. There was a much shorter 'cooling' time between irradiation and analysis in the case of the second sample. In this case it was also possible to identify the short lived isotope ^{206}Po . The yields of all four isotopes have been quantified and the results are compared in chapter 4.

Trace amounts of polonium can be separated by conventional chemical means; precipitation from solution on a carrier, solvent extraction, ion-exchange, and electrodeposition.

Polonium deposits very readily onto silver metal from dilute HNO_3 and HCl solutions normally giving a very complete separation^(2.12) but during deposition a film of silver oxide or peroxide may appear on the silver surface inhibiting further deposition of the polonium^(2.13). The deposition is improved, and the silver foils are cleaner, if the solution is treated with a reducing agent such as hydrazine^(2.14). The polonium deposit becomes involatile after a few hours exposure to air or if left in the solution from which it has been deposited. The deposition has also been shown to be more efficient, and smoother from HCl than from HNO_3 , though less surface oxidation occurs in the nitrate solution. The deposition of polonium is independent of the concentration of acid provided it

is greater than $0.1 \text{ M}^{(2.15)}$ and proceeds more efficiently at elevated temperatures. At $70^{\circ}\text{--}80^{\circ}\text{C}$ approximately 99% of the polonium is deposited from 0.5 M hydrochloric acid in approximately sixty minutes.

Coprecipitation of polonium with tellurium is quantitative. The polonium can be precipitated from hydrochloric acid containing tellurous acid - H_2TeO_4 . The solution containing the polonium and tellurium are treated with tin(II) which reduces the tellurium(IV) to the element bringing down the polonium quantitatively with it.

The method of self deposition was chosen for polonium analysis, in the first instance, as there are few steps in which losses may occur. The second method of coprecipitation was used to scavenge the polonium from several aliquots of the irradiated uranium for the analysis of ^{210}Pb . ^{210}Pb has a half-life of 22.6 years and decays to ^{210}Po by β decay. A measure of the amount of this isotope produced as a spallation product, and consequently its contribution to the overall production cross-section of ^{210}Po , was made by stripping out accumulated polonium from the irradiated uranium solution and the measuring the production of ^{210}Po after several months of ^{210}Pb decay.

This component can then be subtracted to give the 'direct' production cross-section for ^{210}Po . The

detailed experimental procedures for polonium and ^{210}Pb are described below. Appendix C gives the possible routes for the production of ^{210}Po and ^{210}Pb .

i) POLONIUM

Several aliquots of irradiated uranium, in HNO_3 , between 50-100mg, were taken in 100ml beakers. The nitric acid was destroyed by heating with concentrated HCl on a hotplate at a temperature not exceeding 120°C [polonium is volatile at 150°C], until dry without baking the residue. The samples were taken up in 10mls of 2 M HCl and warmed to aid dissolution. 5mls of 20% hydroxylamine hydrochloride solution, 2mls of 25% sodium citrate solution and 1ml of Bi^{3+} holdback carrier (approximately 10mg) were added. The volume adjusted to 50 mls, the solution was then 0.5 M with respect to HCl , 0.3 M with respect to HONH_3Cl and 0.03 M with respect to sodium citrate. The pH was adjusted to 2 with 0.880 ammonia solution, and the solutions heated to between $80-90^\circ\text{C}$ on a stirrer hotplate, and gently mixed with a teflon coated magnetic stirrer. A square of clean silver foil approximately 100mm^2 was added to each solution, the beaker covered, and left to plate for 90 minutes.

After plating had been completed the silver planchette was removed, washed with distilled water then ethanol and dried under an infra-red lamp. In the method used

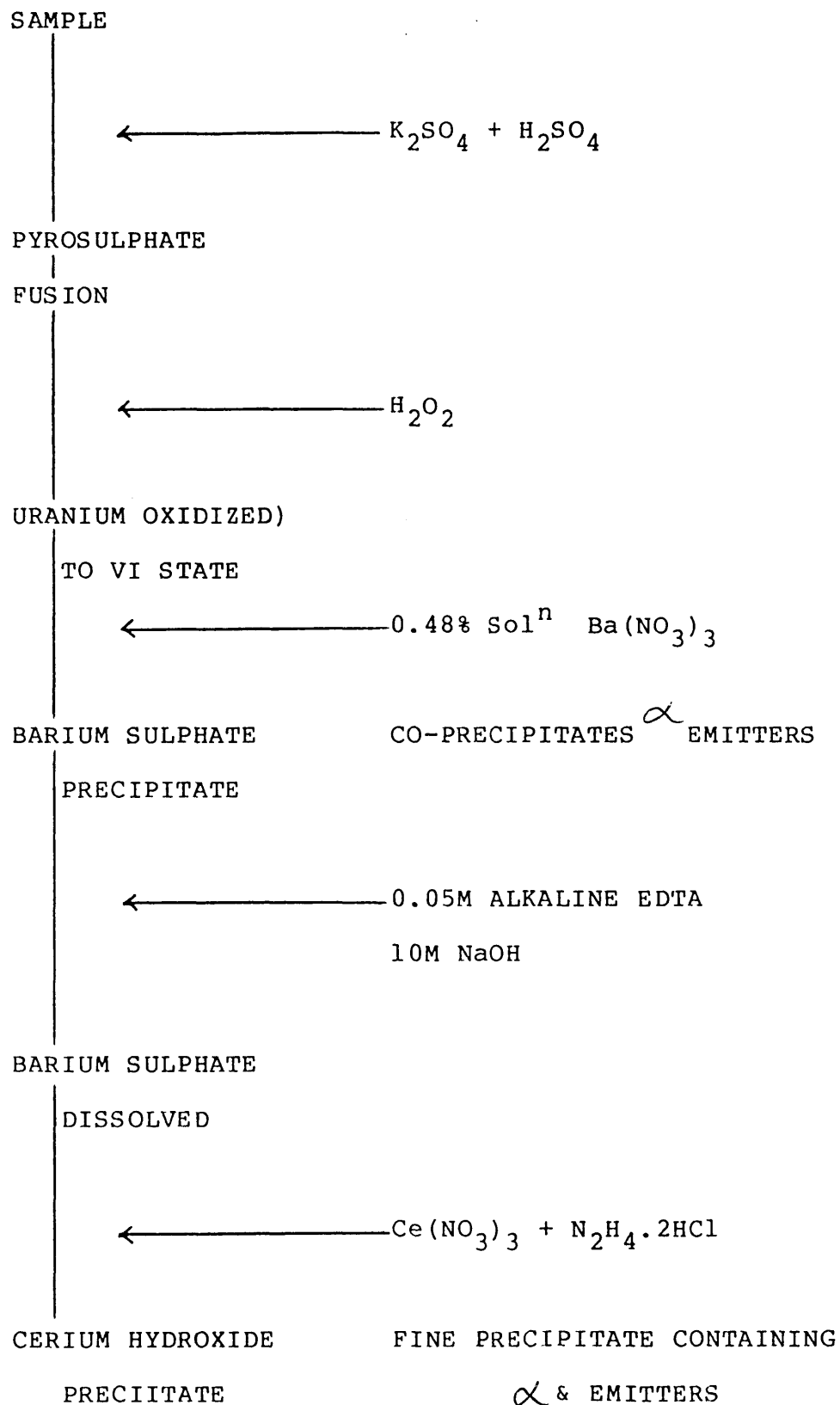
the silver was allowed to lie freely in the plating solution and consequently both sides of the source had to be counted and the count rates summed. A fresh piece of silver was then put into the plating solution and left at 80°C for a further hour to make sure that all traces of polonium had been removed. The resulting solution was used for the estimation of lead-210.

The solution was covered and stored for several months to allow ^{210}Po to grow in from the decay of ^{210}Pb . The interval between the analyses was chosen to allow the maximum yield of ^{210}Po to be produced thus enabling the detection of the minimum amount of ^{210}Pb .

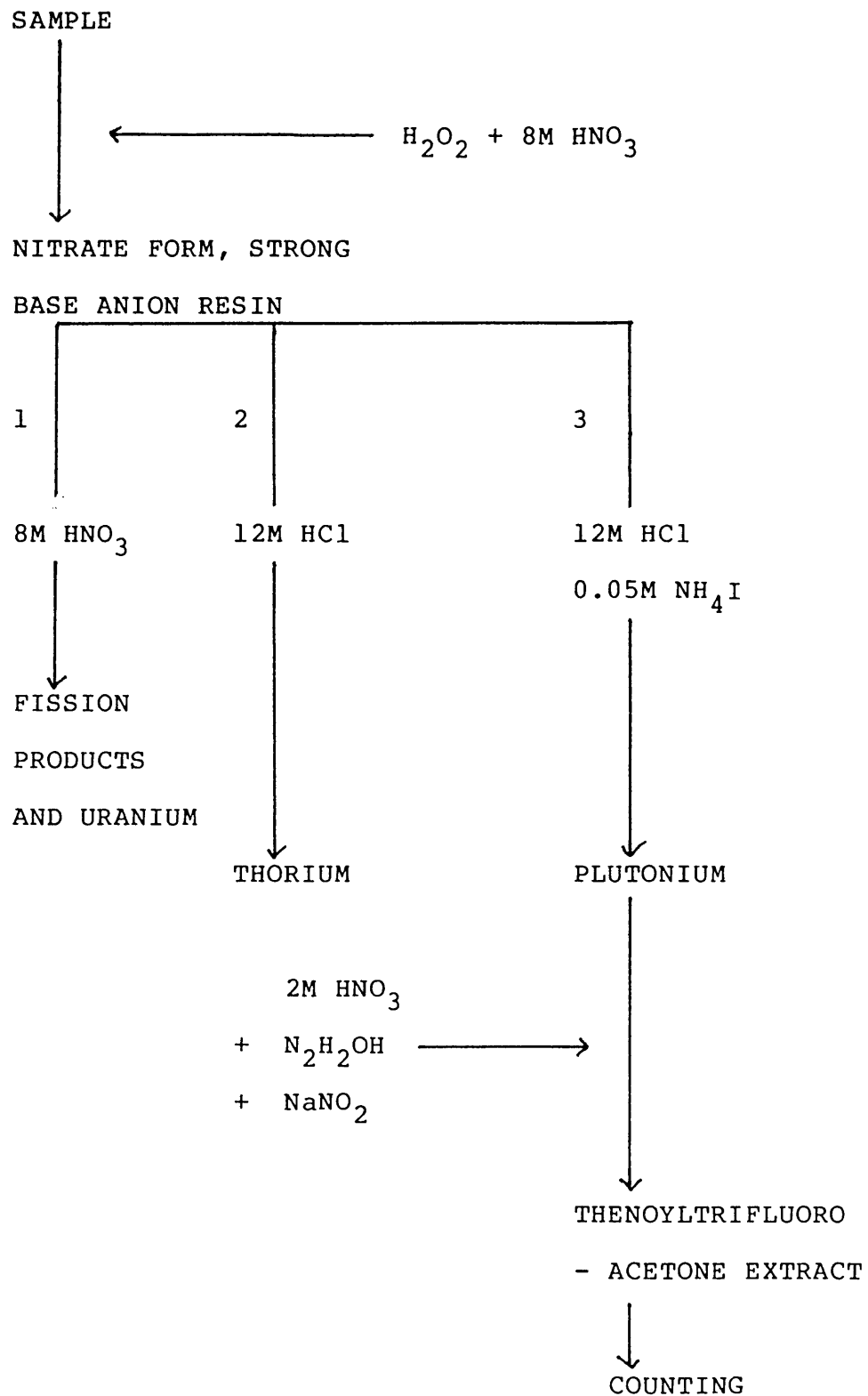
ii) LEAD

The analysis was carried out by coprecipitation with tellurium as a concentration step before the deposition of the daughter, polonium, onto silver foil as previously described above. This method was chosen as the expected quantity was less than that observed for the total polonium production. The solutions were made 5 molar with respect to HCl and between 1 to 2mg of bismuth holdback carrier was added and 1mg of tellurium added as the acid H_2TeO_4 . The polonium was precipitated along with the tellurium after the addition of a few drops of concentrated tin chloride (SnCl_2) solution. The precipitate was then filtered through a sintered glass Buchner funnel (fine porosity) and washed with deionised water.

[kaolin was used as a flocculating agent]. The tellurium / polonium was then dissolved off the filter support in a small amount of concentrated HCl. The kaolin and the filter were washed with sufficient distilled water to dilute the HCl to 6 M and the solution was adjusted as described above in section 2.5i and the polonium plated onto silver.



SCHEME FOR CO-PRECIPITATION OF ALPHA EMITTERS
FOLLOWING C.SILLS METHOD.



SCHEME FOR ION-EXCHANGE AND LIQUID EXTRACTION SEPARATIONS.

CHAPTER 3SOLID STATE SEMICONDUCTOR DIODE DETECTORS

3.1 In the detection of alpha and gamma radiation solid semiconductors have a great advantage over other detectors for both in that their stopping power is much greater than that of gas filled detectors. This is a consequence of the solid densities being in the region of 1000 times greater than those of the gas fillings. The efficiency is therefore improved, and the detector dimensions can be reduced. The important quantity for the application of a semiconductor as a radiation detector is the energy required by the primary radiation to produce an electron-hole pair. (These being the information carriers, analogous to the ion-pairs formed in a gas detector.) This is largely independent of the radiation type and its energy, as long as all the energy is deposited within the sensitive volume of the detector.

The minimum energy required to produce a single ion-pair in a solid is equal to the energy gap between the valence and conduction bands; for silicon and germanium these are 1.1 eV and 0.7 eV respectively. Effectively ϵ , the energy required to produce an ion-pair in these semiconductor materials is larger, 3.6 eV and 2.8 eV respectively, as some of the

radiation energy is deposited as excitation. This is much smaller than the ionisation energy of a gas, which requires approximately 30 eV for the formation of an ion-pair.

For a given radiation exposure the use of semiconductor detectors can therefore result in the formation of a much greater number of charge carriers, approximately 10 times that produced in gas filled detectors.

The resolution of these detectors is greatly enhanced compared to solid scintillation detectors, which require a greater number of events to convert the primary radiation into an electrical signal. The energy required to produce an information carrier in a solid scintillator is of the order of 1 keV, and the number of carriers produced by radiation interaction will be no more than a few thousand per MeV. The statistical fluctuations place an intrinsic limitation on the achievable resolution of photons.

As well as silicon and germanium there are several other semiconductor materials of possible use for radiation detection. Examples are the binary compounds: gallium arsenide (GaAs) and cadmium telluride (CdTe). Table 3.1 gives a comparison of detector types and energy required to produce one information carrier.

TABLE 3.1

<u>Material</u>	<u>Energy per information carrier</u>	
Gas	~30 eV	
Solid Scintillation		
Liquid Scintillation	~1 keV	
Germanium	(77K)	2.98 eV
Silicon	(300K)	3.61 eV

At present germanium and silicon are the only semi-conductors easily produced with sufficient purity and crystal perfection for use as solid state radiation detectors.

The significant effect of the interaction of radiation with a semiconductor is the production of many electron-hole pairs along its track. The increase in the number of charge carriers has two effects which enhance the detectors energy resolution:

- i The statistical fluctuations in the number of carriers per pulse becomes a smaller part of the total, as the number of carriers increases.
- ii The greater amount of charge per pulse gives a better signal to noise ratio.

As in insulators the majority of semiconductor electrons are fixed, but a small quantity of impurity renders it conductive. Germanium and silicon are each four valent, and each of their valence electrons are used in bonds with adjacent atoms, leaving no free electrons for conduction. The introduction of an impurity atom from group V such as phosphorus (five-valent) into the lattice gives an extra electron at that site, leaving one electron free for conduction. The five valent atom is a 'donor' impurity, and produces an n-type semiconductor material having a negative site for conduction (figure 3.1a).

A p-type semiconductor material having a positive site for conduction, is produced by the introduction of a group III 'acceptor' impurity such as gallium or boron into a lattice site (figure 3.1b). This leaves one electron missing from the bonding; giving a positive hole. Conduction in this lattice type is from the transfer of an electron from an adjacent atom, which leaves a vacancy where it came from. It is the movement of these electrons that gives the appearance of the positive hole moving in the opposite direction.

Some classes of solid state detectors are based on the favourable properties which are produced near the junction between n- and p-type semiconductor materials. Charge carriers are able to move across the junction between p- and n- material if the regions are in good thermodynamic contact.

The junction is formed in a single crystal by producing a change in the levels of impurity from one side of the junction to the other.

On the n-type side of the junction of p-n material, the concentration of conduction electrons is much higher than in the p-type, any charge carrier free to migrate diffuses from the region of high concentration to that of low concentration. These will be able to combine with the holes of the p-type material. This leaves behind a net fixed positive charge on the n-type side. Diffusion from high concentration to low of positive holes leaves a net fixed negative charge in the p-type material. The field produced stops further diffusion of charge carriers across the junction leaving an equilibrium charge distribution. The region over which the charge imbalance has occurred is called the depletion region, or intrinsic area. Figure 3.1c illustrates this process.

The depletion region of the semiconductor diode exhibits the properties desirable for the detection of radiation. The electric field between the p- and n- regions causes electrons created in the depletion region to be swept back towards the n-type region and holes towards the p-type region. Electron hole pairs created in this region by the passage of radiation will be carried out of the area by the electric field, their motion being the basic electrical signal.

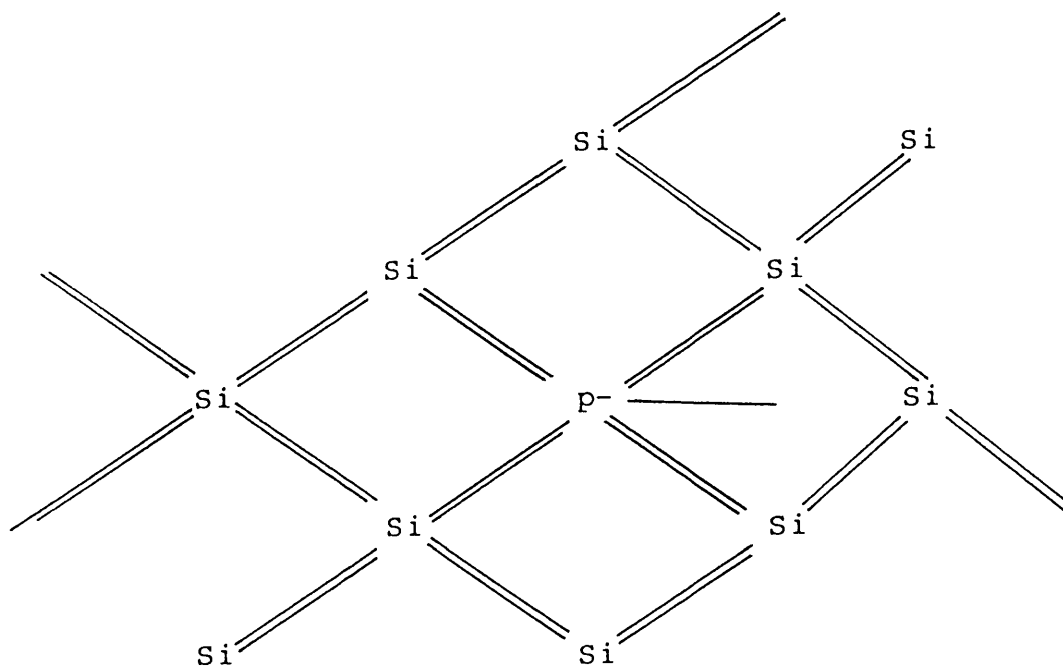


Fig 3.1a

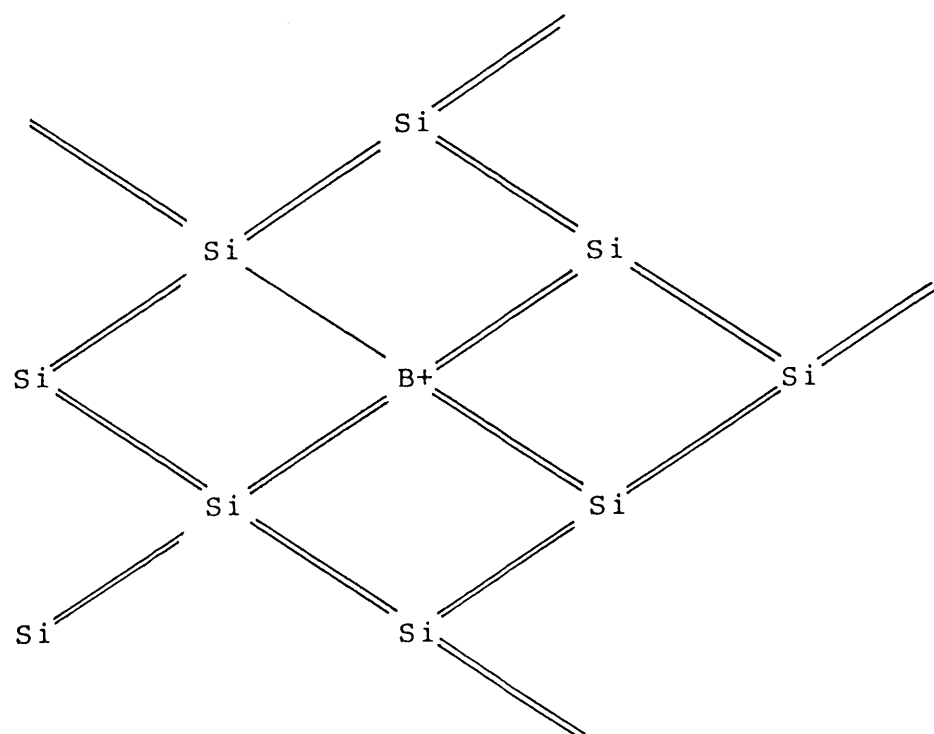


Fig 3.1b

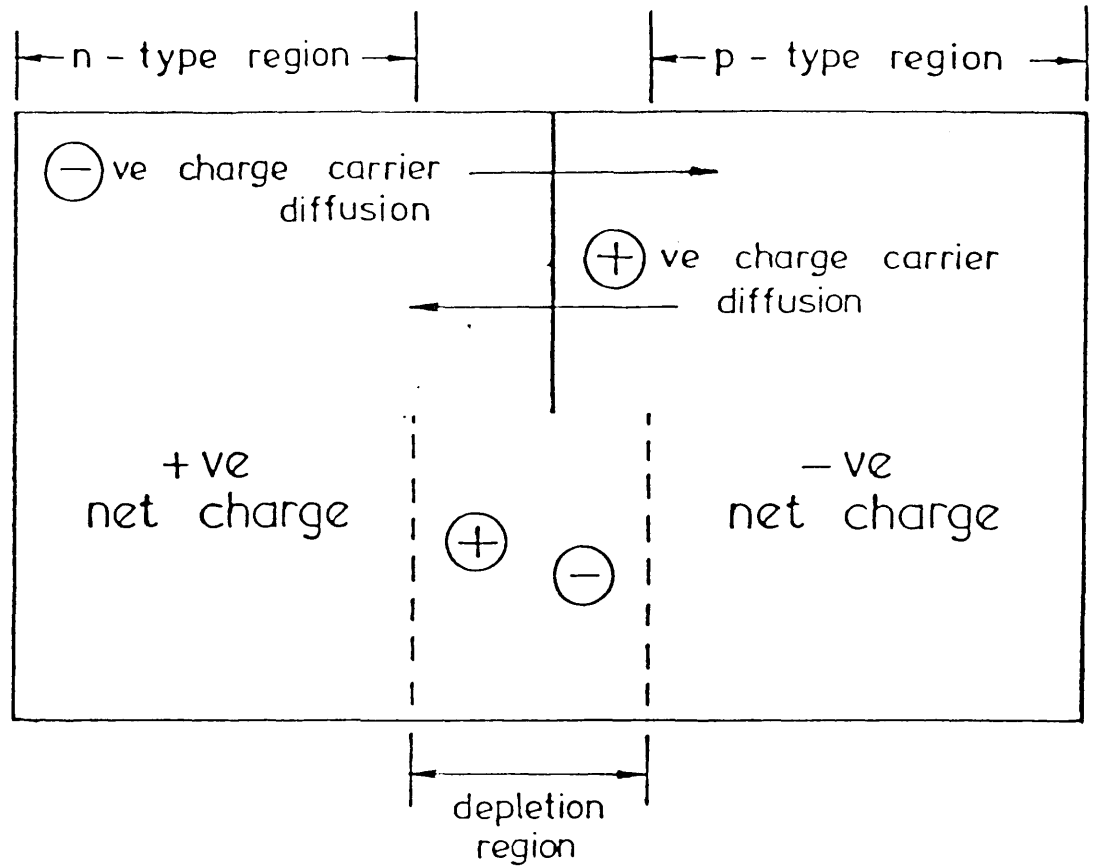


Fig. 3.1c Recombination of conduction electrons from n-type material with a vacancy in the covalent bond of the p-type, producing the depletion region

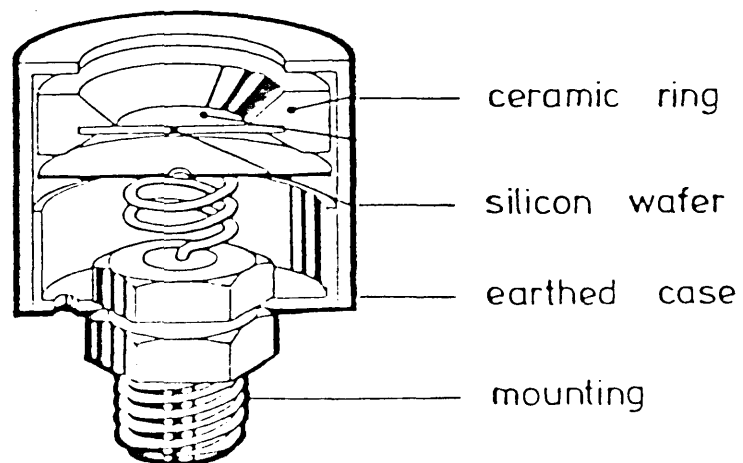


Fig 3.2 Silicon surface barrier detector (EG + G Ortec)

The potential across the junction is in itself insufficient to collect the charge rapidly giving incomplete charge collection as carriers will be lost by recombination and trapping.

The junction conducts readily if a forward voltage is applied, but conducts very little current when reverse biased. When the junction is reverse biased the natural potential difference is enhanced, the device acts as a diode rectifier since it passes current in one direction only. In this situation virtually all the applied voltage appears across the depletion region due to its higher resistivity (lack of free charge carriers) accentuating the natural potential difference of the junction. This simultaneously increases the depletion region in p- n- junctions, expanding the active volume over which radiation produced charge carriers can be collected. [This increase does not occur in compensated detectors Ge(Li)].

In the reverse bias situation only the minority carriers (holes on the n- side and electrons on the p- side) are attracted across the junction. Their low concentration produces a much lower reverse current, the leakage current. Another source of leakage current is the thermal excitation of electrons from the valence to conduction band, acting against the reverse bias.

For silicon semi-conductors this is tolerable at room temperature. In germanium at room temperature the thermal excitation is quite considerable and it needs to be cooled to at least 90K to be of use. In practice it is convenient to cool the detector to liquid nitrogen temperature (77K).

3.2 SILICON SURFACE BARRIER DETECTORS

~~The~~ p-type material for the formation of the junction in this type of detector can be formed by producing a high density of electron traps at the surface of an n-type crystal. This produces a depletion region between the two types. The production of this p-type area is usually effected by etching the surface of the n-type silicon crystal followed by the evaporation of a thin gold layer onto its surface for electrical contact (this is a positive polarity reverse biased n-type detector). Junctions can also be produced starting from a p-type crystal with the evaporation of aluminium onto the surface to give an n-type contact (negative polarity reverse biased p-type detector).

The evaporated film is outside the depletion region and is effectively a dead layer or window through which the incident radiation must travel to reach the active volume of the depletion region. It is disadvantageous to have a large dead layer for the spectrometry of charged particles as some of the particles' energy will be lost in this region before

reaching the active volume. A characteristic of surface barrier detectors is their very thin dead layer. For this reason silicon surface barrier detectors have become most popular in applications where heavy charged particles are involved, commonly in the spectrometry of alpha particles. [Figure 3.2a shows a schematic section of a silicon surface barrier detector.]

Compared with other devices such as NaI(Tl) and liquid scintillation; semiconductor detectors provide advantages in several areas:

- i They show exceptional energy resolution.
- ii Good stability - freedom from drift.
- iii Thin entrance windows.
- iv Excellent timing characteristics with observed pulse rise times in the order of 10 nanoseconds or less.

The latter corresponds to the migration of electron-hole pairs formed by the incident radiation across the high electron field of a depletion region. The time is minimised in detectors of high electric fields and small depletion widths.

The resolution of the detector is measured at its full width half maximum (FWHM) of an alpha peak. Commercially detectors are available with a FWHM of as low as 12 keV (e.g. for the 5.49 MeV peak of ^{241}Am), but usually it is in the region of 24 keV. It is not usually experimentally possible to find any significant variation in detector efficiency in the energy interval 2.5 to 8.8 MeV, this makes the quantitative measurement of alpha emitters fairly simple.

3.3 LITHIUM DRIFTED GERMANIUM DETECTORS

Surface barrier detectors find widespread use in alpha spectrometry, but because of their small depletion widths (up to 3 mm) they are not adaptable to more highly penetrating radiations such as gamma-rays.

The ion-drifting process however produces semi-conductors with large active volumes. The process consists of producing a thick region of 'compensated' material in which acceptor and donor impurities are present in equal concentrations. A region is thus produced with similar properties to the intrinsic material; which with blocking contacts on either side can act as a medium for the collection of electronhole pairs produced by the incident radiation (see Figure 3.3a).

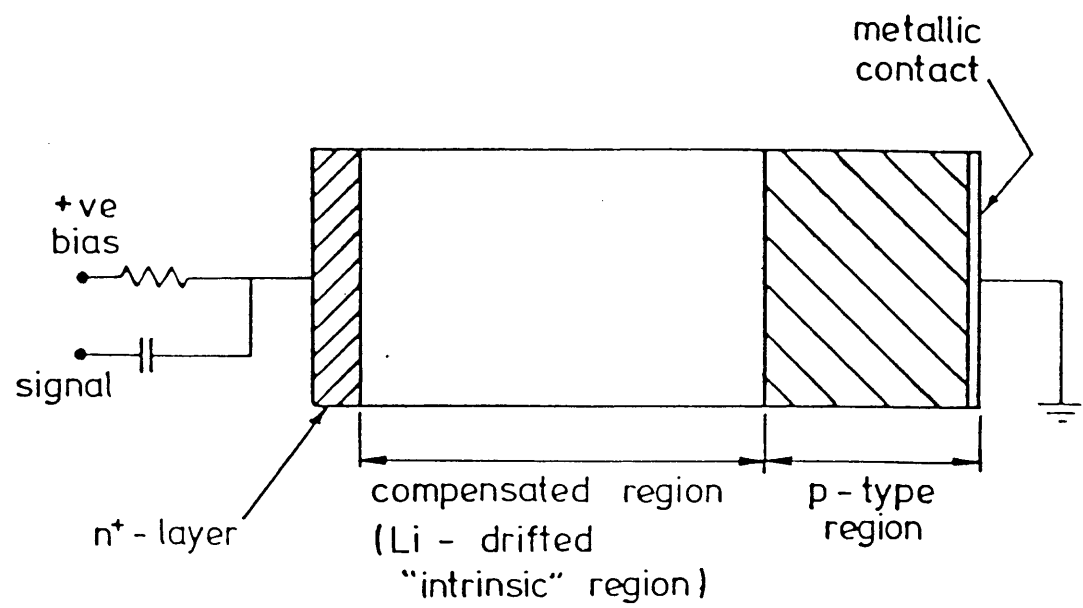


Fig. 3.3a Basic configuration of a lithium - drifted p - i - n junction detector

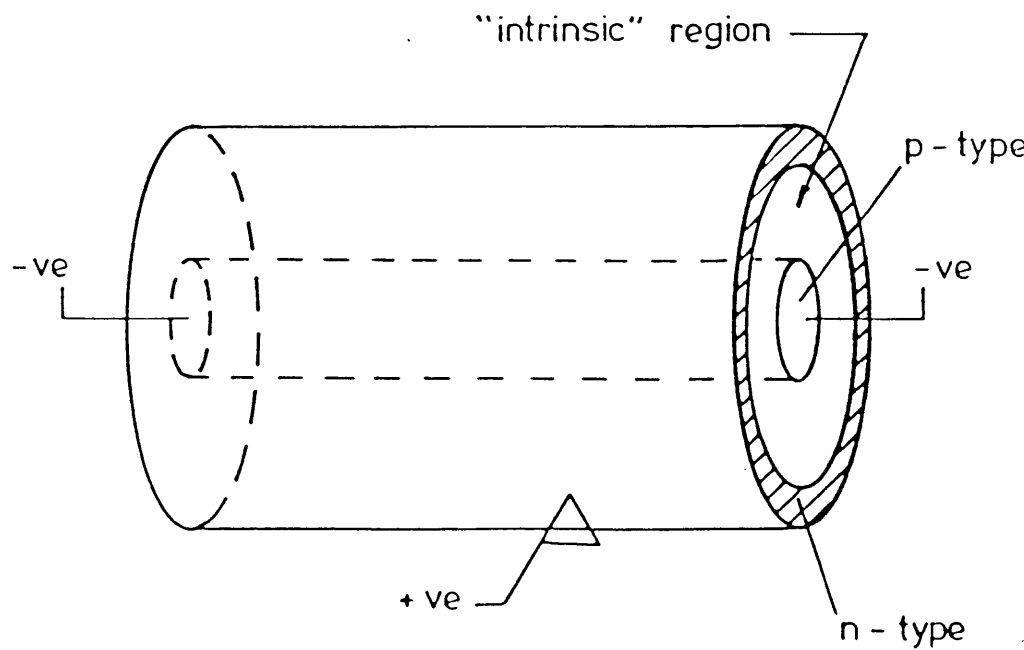


Fig. 3.3b Open - ended cylindrical, "true coaxial" configuration

In germanium the highest purity available tends to be the p-type containing acceptor impurities. The alkaline metal lithium can be used to form interstitial donors in the crystal. The diffusion of an excess of lithium through one face of a p-type material creates an n-type region, where the number of lithium donors greatly outnumber the original acceptor impurities. Reverse biasing the crystal enhances the diffusion of lithium ions into the crystal. Under the influence of the applied electric field the lithium donors reach a concentration equal to the original acceptors. A near exact compensation automatically is produced, as the drifting of lithium tends towards a state where there is zero net space charge. Through this procedure a compensated region of 8 to 10 mm is possible.

The mobility of the lithium ion in germanium remains high enough at room temperature to allow redistribution of the lithium, therefore the detector has to be permanently kept at liquid nitrogen temperatures.

The excess lithium in the region where the drifting was originated is used as the electrical contact as this region is heavily doped making a conductive n^+ -type region, where n^+ means heavily doped. The non-compensated region at the opposite end of the crystal is p-type material and electrical contact is made here by metallic contact.

The life time of charge carriers in the compensated region is much greater than the time required to collect them. Therefore good charge collection properties are attainable. For rapid charge collection high voltages in the region of 3000-4000 volts are generally applied across the detector to produce a sufficiently high electric field to attain the maximum drift velocity of charge carriers. This maximises the inherently slow pulse rise time which varies with depth of active volume. At saturated drift velocity the collection time depends on the distance to be travelled, and is in the order of 100 nanoseconds for a 1 cm thick detector.

Virtually all this applied voltage appears across the compensated region due to its high resistivity compared with the p- and n- regions. Consequently it is the size of this region which determines the active volume of the detector.

To increase the active volume in drifted detectors non-planar geometries are used where the drifting occurs over larger surface areas, examples of these are : open ended and closed ended coaxial crystals. These can produce active volumes up to 150 cm^3 . Figure 3.3b shows an open ended coaxial system.

The width of the active volume of a Ge(Li) is determined by the compensation region only, and is fixed after drifting unlike surface barrier detectors which are effected by the applied voltage; increasing with voltage to the point where electrical breakdown occurs. The active volume in Ge(Li)s being many times greater than in surface barriers results in a lower electrical field being produced by the applied voltage, the latter being limited to the surface breakdown potential.

The dominant characteristic of germanium detectors is their energy resolution, which is much greater than NaI(Tl) scintillators. This enables complex spectra to be analysed as the resolution is such that closely spaced gamma energies are separable. The resolution of Ge(Li) detectors is determined by:

- i The inherent statistical fluctuations in the number of charge carriers produced.
- ii Charge collection efficiency.
- iii Electronic noise.

The second of these factors is most significant in detectors of large active volume, where there is a low average electric field. The electronic noise encompasses the broadening effects of all the electronic components following the detector.

3.4 HIGH PURITY GERMANIUM

Germanium with purities needed for semiconductors cannot produce p-n junctions with sufficient depletion depth for gamma ray spectrometry. The lithium drifting process can create these regions with a depth in the order of 10-15 mm, but they have the disadvantage, in operation, of needing a constant supply of liquid nitrogen. If allowed to warm up they lose their intrinsic region by the re-distribution of the lithium ions.

Germanium is now available in a state of very high purity, having only 10^{10} atoms of impurity per cm^3 . Sufficient resistivity is produced, such that a depletion depth of 10 mm is obtainable with a reverse bias of 1000 volts. [The depletion depth of a p-n junction at a given voltage increases as the square root of the resistivity of the materials.]

The semiconductor properties of high purity germanium are usually p- type due to residual acceptors (Al) or lattice defects. The general configuration of this detector type is: $n^+ - p - p^+$ (see Figure 3.4). The n^+ contact is formed by the evaporation of lithium onto a surface of the p- type germanium. Reverse biasing between n^+ and p- gives the depletion region. The other contact on the opposite face is produced by

metal-semiconductor surface barrier junction, acting as a p^+ contact. This p^+ face is used as the entrance window, as the dead layer associated with the n^+ lithium contact is much greater.

The depletion region stems from the n^+ region as the reverse bias is applied extending through the p -region as the voltage is increased eventually becoming fully depleted, the active volume extending from one contact to the other. An over-voltage is applied to raise the electric field so that a minimum field is produced to reach saturation velocity of charge carriers. This minimises the collection time reducing the probability of recombination and trapping.

The voltage to produce the maximum velocity for electrons is the value used, due to the difficulties related to breakdown (electrical) and surface leakage at higher field strengths (3 to 5 times that of electrons) required to obtain saturated hole velocities.

The properties of semiconductor detectors mainly depend on the electric field in the active volume. In high purity germanium detectors approximately 75% of this field is higher than in Ge(Li) detectors, for similar volume and applied voltage, giving better timing characteristics than Ge(Li)s and improving the

collection time of holes. The gamma ray detection efficiency and response function of high purity Ge is identical to Ge(Li) detectors and the resolution is comparable for example: 0.4 - 0.5 keV for 122 keV gamma and approximately 1 keV for the 622 keV gamma ray.

3.5 AMPLIFICATION AND SIGNAL SHAPING

In spectrometry the signal from the detector is fed into a pre-amplifier for the first stage of amplification, then to the main amplifier where the signal is shaped and processed for data collection in a multichannel analyser. The pre-amplifier is mounted as close as practical to the detector to reduce input capacitance from cabling, and thus giving a greater signal to noise ratio. In most gamma spectrometry systems the detector and pre-amplifier are built as an integral unit to reduce noise (the pre-amplifier is cooled to near liquid nitrogen temperature along with the semiconductor).

In most spectrometry applications charge sensitive pre-amplifiers are used. The detector signal from semiconductors being a quantity of charge giving a current pulse lasting 10^{-9} to 10^{-5} seconds. The pre-amplifier integrates the charge on a feedback capacitor with an ideal rise time of the output pulse

equal to the width of the detector current pulse. The charge sensitive pre-amplifier converts the input charge into an output voltage pulse V_o , given by:

$$V_o = Q_d / C_f$$

where Q_d is the charge from the detector and C_f is the feedback capacitance. The charge produced by the semiconductor is a function of the incident radiation energy and is given by:

$$Q_d = E e 10^6 / \epsilon$$

E = incident radiation energy in MeV

e = electron charge

ϵ = energy for electron-hole pair production in detector material

The output of the pre-amplifier consists of a step voltage pulse which is proportional to the input amplitude and has a rise time in the order of tens of nano-seconds decaying exponentially. This is fed into the main amplifier. The amplifier expands the analog outputs from the pre-amplifier to a range that is measurable with greater accuracy.

Pulse shaping and filtering are performed in the amplifier to improve the signal to noise ratio, and to shorten the response time for each pulse, thus preventing overlap of pulses. The voltage pulse

amplitude carries the basic information, if a following pulse arrives before the preceding one has decayed to zero it is superimposed on the tail of the decaying pulse and it is no longer an accurate measure of the charge deposited.

To solve this problem the pulses are shaped eliminating the tails, but extracting the information of the amplitude maxima. As the shaped pulses (semigaussian) tend to undershoot zero subsequent events could have their amplitude reduced. This is eliminated by pole-zero cancellation and base line restoration improving resolution of closely spaced pulses at high count rates (see figures 3.5a and 3.5b).

3.6 ANALOG TO DIGITAL CONVERTER & MULTICHANNEL ANALYSER

The incoming semi-gaussian pulse from the amplifier to the multichannel analyser is converted to a digital address by a Wilkinson type ADC. The pulse charges a capacitor, which is discharged linearly. This is made to produce a strong rectangular pulse, with the original amplitude and with a duration equal to that of the discharge. The pulse is then addressed by scaling clock pulses against the discharge period, the address being directly proportional to the amplitude of the initial analog input. The multichannel analyser (MCA) provides a record of the addresses in a memory core. Successive equivalent pulses are incremented to the appropriate channel, building up a pulse height spectrum.

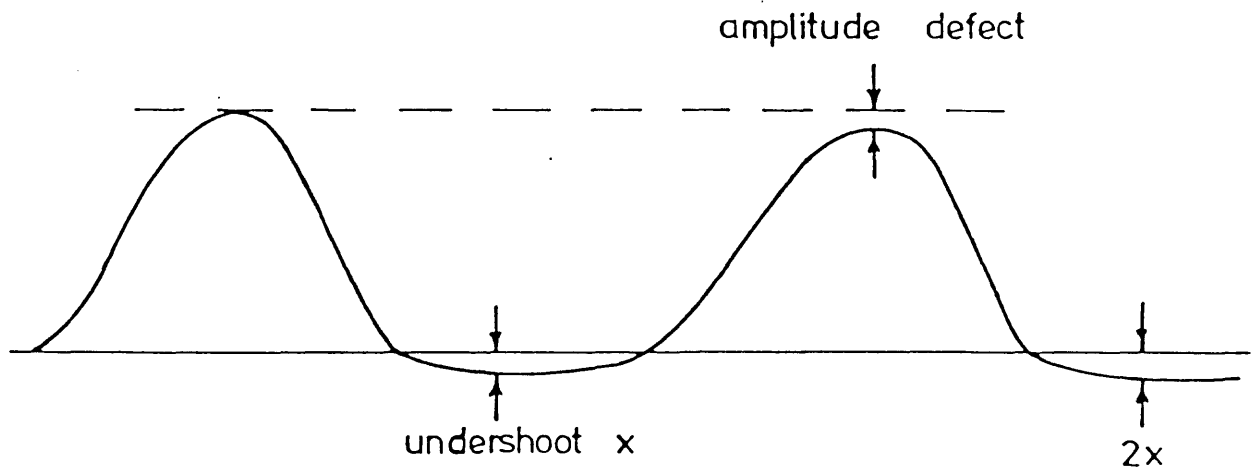


Fig. 3.5a Effect of baseline shift on amplitude

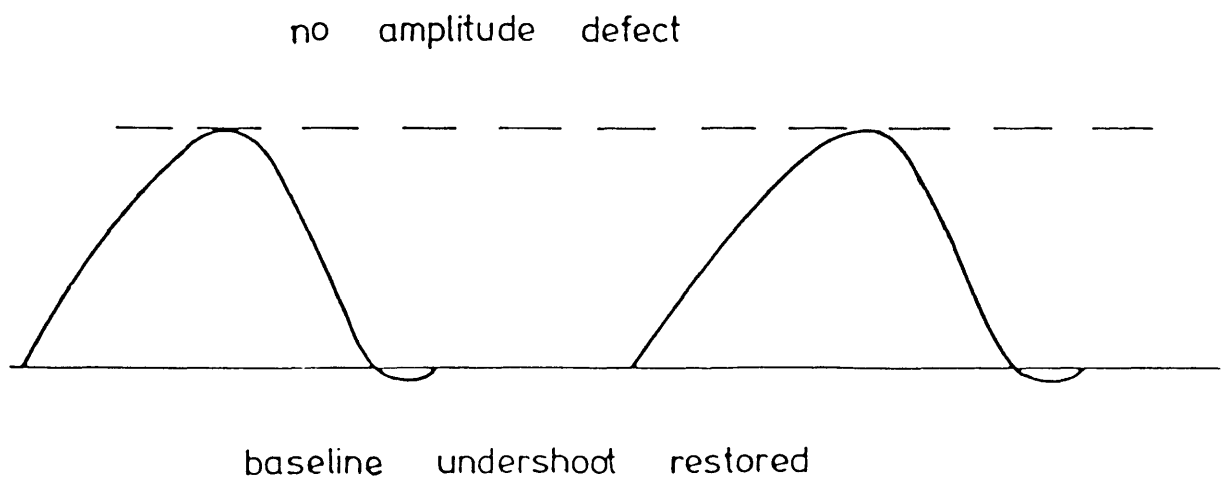


Fig. 3.5b Effect of baseline restoration

After accepting a signal, at the beginning of the address procedure an electronic gate is closed, and is not opened until the data has been stored in the memory of the MCA, preventing interferences from other pulses. This introduces a degree of dead time into the system which must be accounted for, as the pulses from the random decay of a sample which occurs during the time that the MCA is procesing a pulse will be lost. This is often automatically accounted for by the MCA. (e.g. preset live-time of 10000 secs. with 10% dead time shows a real time of 11000 secs.).

3.7 EXPERIMENTAL EQUIPMENT

3.7a ALPHA SPECTROMETRY

Small aliquots of sample were taken for analysis to ensure that 'thin' alpha sources were produced, as the range of an alpha particle is in the order of a few mg cm⁻². Hence sources with a thickness of a few tenths of mg cm⁻² of the sample were aimed for. As alpha particles are very easily absorbed, having these small masses of material reduces self absorbtion.

Thick samples degrade the quality of the spectrum, the resolution is reduced by broadening of the peaks from scattering, and deposition of energy within the sample. [The stopping power : dE/dX is approximately 50 KeV/ μ m in solids]. This has one of two effects: on the lower energy side of a major peak an adjacent one

of lower specific activity may be masked by overlapping making identification difficult and quantitative analysis impossible. Secondly with adjacent peaks of similar activity, the peak with lower energy will appear to have an increased activity due to contributions from the tailing of the other. Similarly the higher energy peak will seem to be reduced by the quantity that it is contributing to the adjacent peak making quantitative analysis unnecessarily difficult. Poorly produced alpha sources can not be improved by better electronics.

Alpha spectrometric measurements were made using a 300 mm² ruggedised EG&G Ortec p-type silicon charged particle radiation detector (Al contact/window), having a sensitive depth of 100 microns. The resolution of the 5.486 MeV ²⁴¹Am alpha decay was measured to be 20.7 KeV FWHM by the manufacturers. The surface barrier was connected to an EG&G preamplifier, model 412b, the connection being direct to the detector by a BNC fitting. An EG&G 570 spectroscopy amplifier fed signals into an ELSCINT Promeda MCA via the Wilkinson type ADC. The pulse height spectra were counted in 1024 channels, with the gain and offset set to give an energy span of approximately 3 MeV. Some measurements were made on a 100 mm² Ortec nonruggedised n-type (gold contact/window) high resolution silicon surface barrier detector with 100 microns sensitive depth and a measured resolution of 14.6 KeV FWHM using ²⁴¹Am.

The detectors were mounted in a stainless steel vacuum chamber connected to an Edwards rotary pump. The line between the chamber and pump contained a foreline trap of activated alumina to ensure that the sensitive detector surface was not contaminated with oil vapour.

For counting purposes the samples were mounted directly below the detector on a stainless steel holder. This also acted as a seal for the vacuum chamber. The chamber was evacuated before commencing counting (and applying bias voltage); the counting was done under vacuum to maximise the range of the alpha particles which lose energy by interactions with gas molecules. Under these conditions the resolution is at its optimum.

3.7b GAMMA SPECTROMETERIC MEASUREMENTS

Gamma spectrometric measurements have been made using a closed ended coaxial intrinsic germanium detector- (PRINCETON GAMMA-TECH), with a volume of 48cm^3 and a window thickness of 1mm. The detector fully depletes with a bias voltage of 2500V. The FWHM for the ^{60}Co 1.333 MeV photon is 2.25 KeV.

Supplementary measurements were made using a Ge(Li) detector (PRINCETON GAMMA-TECH) having an active volume of 42cm^3 , and on a planar intrinsic detector (7mm thick) having a detector window $<10^{-3}$ mm thick (beryllium).

The detectors were operated at liquid nitrogen temperature. The intrinsic coaxial Ge detector's pre-amplifier, built into the unit, uses room temperature field effect transistors. The detector was connected to an EG&G ORTEC spectroscopy amplifier, model 570, which was pole-zero cancelled and contained automatic baseline restoration circuitry. The quantities of activity used (<500cps) were sufficiently low so that the effect of pulse pile-up could be ignored ; pulse pile-up correction being necessary when the count rate approaches 10Kcps.

The amplified signals were analysed on a ELSCINT Promeda pulse height analyser, the signals being fed into a Wilkinson type analog to digital converter built into the multichannel analyser for data aquisition and processing. There is a maximum of 4096 usable channels for the ADC to address to with a fixed 6 μ sec conversion time.

Each gamma spectrum was allocated 1024 channels in the analyser. The gain and offset of the ADC was adjusted to give the energy range required for the particular sample, and to maintain a high degree of resolution. In the aquisition of gamma spectra the energy range 80-1350 keV was examined by looking at the spectra in three different energy bands: 80-395 keV; 356-727 keV; and 612-1350 keV.

These ranges gave: 0.36, 0.36, and 0.72 keV per channel respectively. The resolutions obtained with these ADC parameters were 2.16 keV, 2.16 keV and 4.32 keV FWHM respectively, measured for the ^{96}Zr 724 keV gamma transition.

The detector was calibrated for energy per channel, and the variation of its efficiency with energy using a calibrated standard gamma source supplied by Amersham International. The samples were prepared for counting so that they were as near as possible geometrically identical to the standards. Measurements were taken from counting small samples so that any effect of self absorption was negligible. The sample size was in the order of 2.4 milligrammes, taken as 25 microlitres of solution 96.1g l^{-1} , this was pipetted into a standard irradiation capsule of 12mm internal diameter and 6mm depth (manufactured by Imperial College Reactor Centre) and counted while centred on the detector window.

CHAPTER 4SPECTRAL ANALYSIS AND MEASUREMENTS4.1 GAMMA ANALYSIS

As previously mentioned in Chapter 3 the main quantitative analysis was performed on a large intrinsic germanium detector, this was complemented by some work on a Ge(Li) detector. The Ge(Li) was a multi-user facility with a variable source to detector distance which could not be accurately reset. Those peaks which were measured on the Ge(Li) have been calculated from their peak area ratio to major peak areas obtained from counting on the intrinsic detector. This being the case a larger sample was counted so that the lesser intensity peaks could be identified more easily.

In order to make accurate determinations of gamma photon intensities using semi-conductor detectors two important factors must be taken into account. Firstly an accurate method of determination of the peak area of the gamma transitions of interest, and secondly the determination of the efficiency of the detector for the photons in the energy region desired.

The two main methods of measuring a peak area are a) the summation of the data between two boundaries with the subtraction of the background, the latter being calculated from limits outside the peak area; and b) using an analytical function that describes the peak area and background.

P.A. Baedeker^(4.1) has compared the precision of several methods of peak area measurement. This was done by analysis of the standard deviations in twelve spectra containing nine photopeaks, over a broad energy range. A modified version of the TOTAL PEAK AREA METHOD (TPA), by J.T. Wasson, generally exhibited the highest precision. When the shape of the spectra was altered by increasing the amounts of activity the TPA method was found to be comparable. Equations I and II summarise the TPA and Wasson methods respectively.

$$\text{I} \quad A = \sum_{i=1}^{i=r} a_i - (a_1 + a_r) \times (r - 1 + 1) / 2$$

$$\text{II} \quad A = \sum_{i=-n}^{i=+n} a_i - (n + 1/2) \times (b_n + b_{-n})$$

a_i = number of counts in channel i .
 l = channel number left of peak limit.
 r = channel number right of peak limit.
 n = number of channels to the left and right
 of the peak centroid.
 b_n = background in channel n .

The background under the photopeak area consists of natural radiations and pulses from higher energy gamma photons. If these are small compared with the photopeak area then the errors in the baseline will not affect the accuracy or precision of the intensity measurements^(4.2). However the manner in which the background is evaluated affects the measured area of a peak. Therefore to remove any ambiguity in comparative measurements it is essential to analyse all peaks in the same manner.

The method of Wasson used a fixed number of channels on either side of the peak centroid. This can not be applied to the analysis of the spectra here, since as previously mentioned, spectra were obtained with different ADC conversion gains, and this effects the width of the peaks. Consequently taking a fixed number of channels would give different proportions of the peak and makes comparison meaningless. To overcome this and enable all the peaks to be related to efficiency data the TPA method was used.

The value obtained for the area depends on the boundaries set for the summation, and the interpolation of the baseline from the background. To enable all the peaks to be comparable the boundaries of each peak were calculated on a statistical basis. The number of channels chosen was calculated from the full width half maxima (FWHM) of the peak centroid, which is equivalent to 2.355σ , and taking the boundaries as: the centroid, $C \pm 3\sigma$ (4.3). Assuming a Gaussian normal distribution 99.87% of the data points (peak area) will be expected to fall within the channels selected.

The measure of the background as a straight line fit from the TPA method was calculated by taking the average value from a suitable region adjacent to the region of the peak limits, taking as many channels as possible. This modifies equation I to give:

$$A = \sum_{i=1}^{i=-1} a_i - a_i/2l + \sum_{i=r}^{i=+r} a_i/2r \times (r - 1)$$

To enable precise measurements of the gamma emission rates from the sample the detector was accurately calibrated for the full energy peak efficiency of the system as a function of photon energy. This was done experimentally using a multi-nuclide gamma standard having a range of photons from 0.088 MeV to 1.836 MeV

all with well defined emission probabilities. The standard source was centred on the detector with exactly the same source to detector distance as that of the samples being measured. This ensured that the geometric conditions were the same: significant differences in efficiency are obtained with a small change in geometry. The activity of this reference source was determined by an accurate absolute measurement by the manufacturers - $4\pi\beta\text{-}\gamma$ coincidence. Table 4.1 gives the reference source data at time of standardisation [1-2-1984]

TABLE 4.1

NUCLIDE	PHOTON	INTENSITY	ERROR
^{109}Cd	0.088 MeV	$708\gamma\text{s}^{-1}$	$\pm 2.8\%$
^{57}Co	0.122 MeV	$653\gamma\text{s}^{-1}$	$\pm 2.2\%$
^{139}Ce	0.166 MeV	$844\gamma\text{s}^{-1}$	$\pm 3.7\%$
^{203}Hg	0.279 MeV	$2155\gamma\text{s}^{-1}$	$\pm 2.3\%$
^{113}Sn	0.392 MeV	$2353\gamma\text{s}^{-1}$	$\pm 3.1\%$
^{85}Sr	0.414 MeV	$4363\gamma\text{s}^{-1}$	$\pm 2.0\%$
^{137}Cs	0.662 MeV	$2819\gamma\text{s}^{-1}$	$\pm 2.8\%$
^{88}Y	0.898 MeV	$6865\gamma\text{s}^{-1}$	$\pm 4.0\%$
^{60}Co	1.173 MeV	$3674\gamma\text{s}^{-1}$	$\pm 0.8\%$
	1.333 MeV	$3679\gamma\text{s}^{-1}$	
^{88}Y	1.836 MeV	$7318\gamma\text{s}^{-1}$	$\pm 2.9\%$

[The standards were corrected for decay since their initial standardisation using the exponential law:

$A_t = A_0 \exp(-\lambda t_d)$, A is the activity and t_d the time elapsed since preparation].

The full energy peak efficiency of each photopeak is determined according to the relationship:

$$\epsilon_p = N / A_0 D$$

the error is given by:

$$(\sigma)^2 = (\epsilon_p)^2 \times \{ (\sigma_N/N)^2 + (\sigma_{A_0}/A_0)^2 + (\sigma_D/D)^2 \}$$

ϵ_p = peak efficiency

N = peak area

A_0 = absolute activity in γ 's s^{-1} at reference time

D = correction factor for decay since standardisation

[$D = \exp(-\lambda t_d)$; λ = decay constant, t_d = decay time]

The errors in t_d and λ are taken as negligible being an order of magnitude less than the overall uncertainties of the reference source.

The measurement of gamma photon emission rates from a sample do not generally coincide with those used for the detector calibration, hence an accurate method of

interpolation between the measured values is necessary. To achieve this a semi-empirical analytical function can be fitted to the data (generally a least squares technique) to give a smooth continuous curve. Ahmad^(4.4) has proposed the following class of fitting function:

$$\epsilon_E = \{P_1 + P_2(\ln E) + P_3(\ln E)^2 + P_4(\ln E)^3 + P_5(\ln E) + P_6(\ln E)^7\} / E$$

where E is the photon energy, and P is a calculated fitting parameter

This has been shown to give satisfactory fits to several data sets from different laboratories^(4.5), and with carefully measured efficiency data gives interpolated efficiencies with a precision of about 1%. In this study a programme at Imperial College Computer Centre (ICCC) used the equation above to fit the measured efficiency data. The fit is shown in figure 4.1a and table 4.2 details the efficiency data calculated from the function above.

TABLE 4.2

ENERGY (keV)	EFFICIENCY (%)	ERROR (%)
88.034	11.102	±0.366
122.06	15.435	±0.380
165.86	14.182	±0.27
279.19	8.792	±0.171

TABLE 4.2 (contd.)

ENERGY (keV)	EFFICIENCY (%)	ERROR (%)
391.7	5.773	± 0.092
514.	4.014	± 0.071
661.66	2.851	± 0.051
898.02	1.905	± 0.027
1173.	1.366	± 0.014
1333.	1.176	± 0.012
1836.	0.835	± 0.037

4.2 GAMMA MEASUREMENTS

Measurements commenced 42 days after irradiation because of the requirement of 'cooling' before transportation. The sample was counted on the coaxial germanium detector at intervals over a six month period. Figure 4.2a shows the spectrum obtained from 80 keV to 760 keV. Figures 4.2b to 4.2e inclusive are expanded regions of 4.2a. Major peak energies and associated nuclides are indicated. Table 4.3 lists the gamma energies identified and the nuclides with which they are associated.

The assignment of γ -energies to particular nuclides has been made on the basis of the energy of the major transition and confirmed by the presence of lesser intense transitions. Energy and intensity data were taken from 'Table of the Isotopes' (seventh edition) (4.6) , and 'The Gamma Rays of the Radionuclides' (4.7).

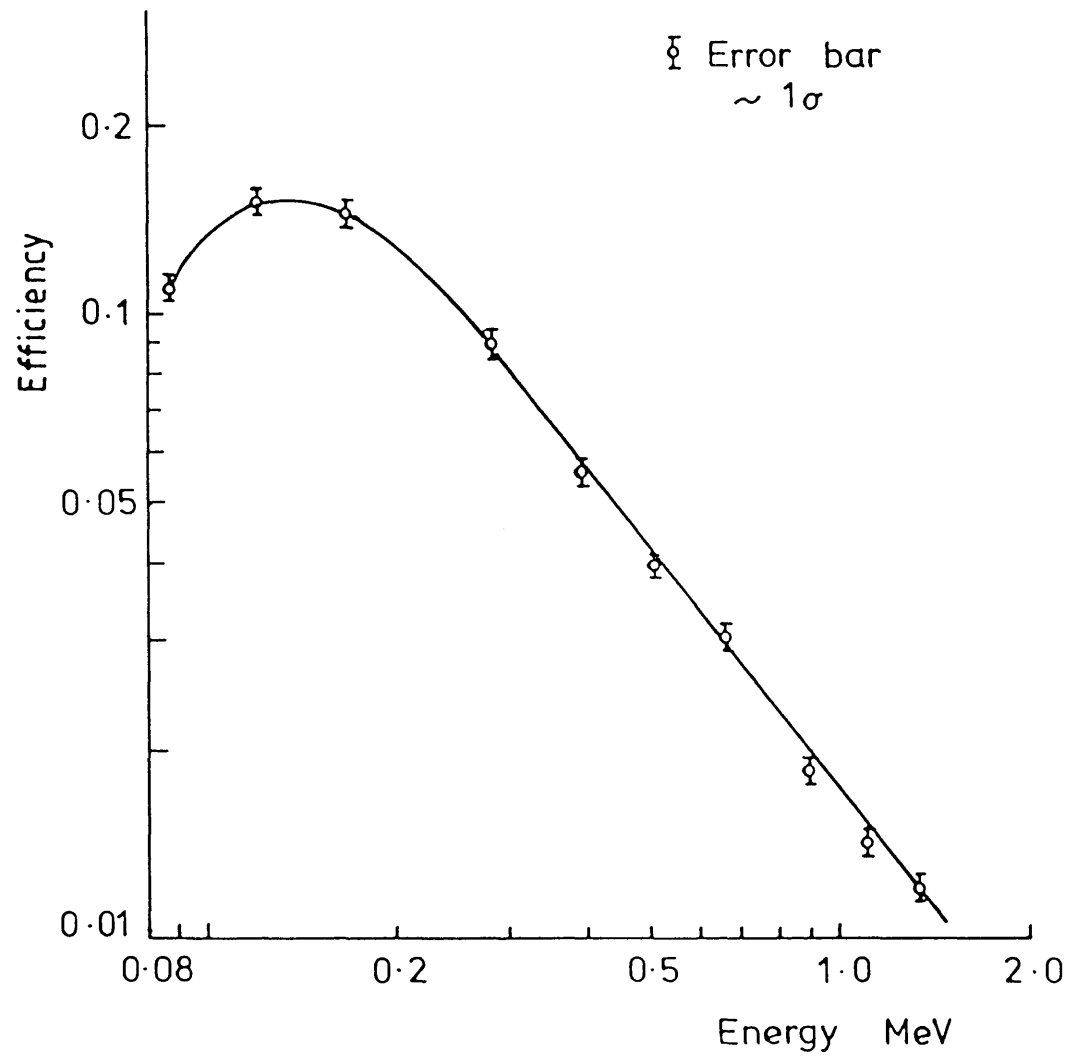


Fig. 4. 1a Efficiency curve for intrinsic Ge detector

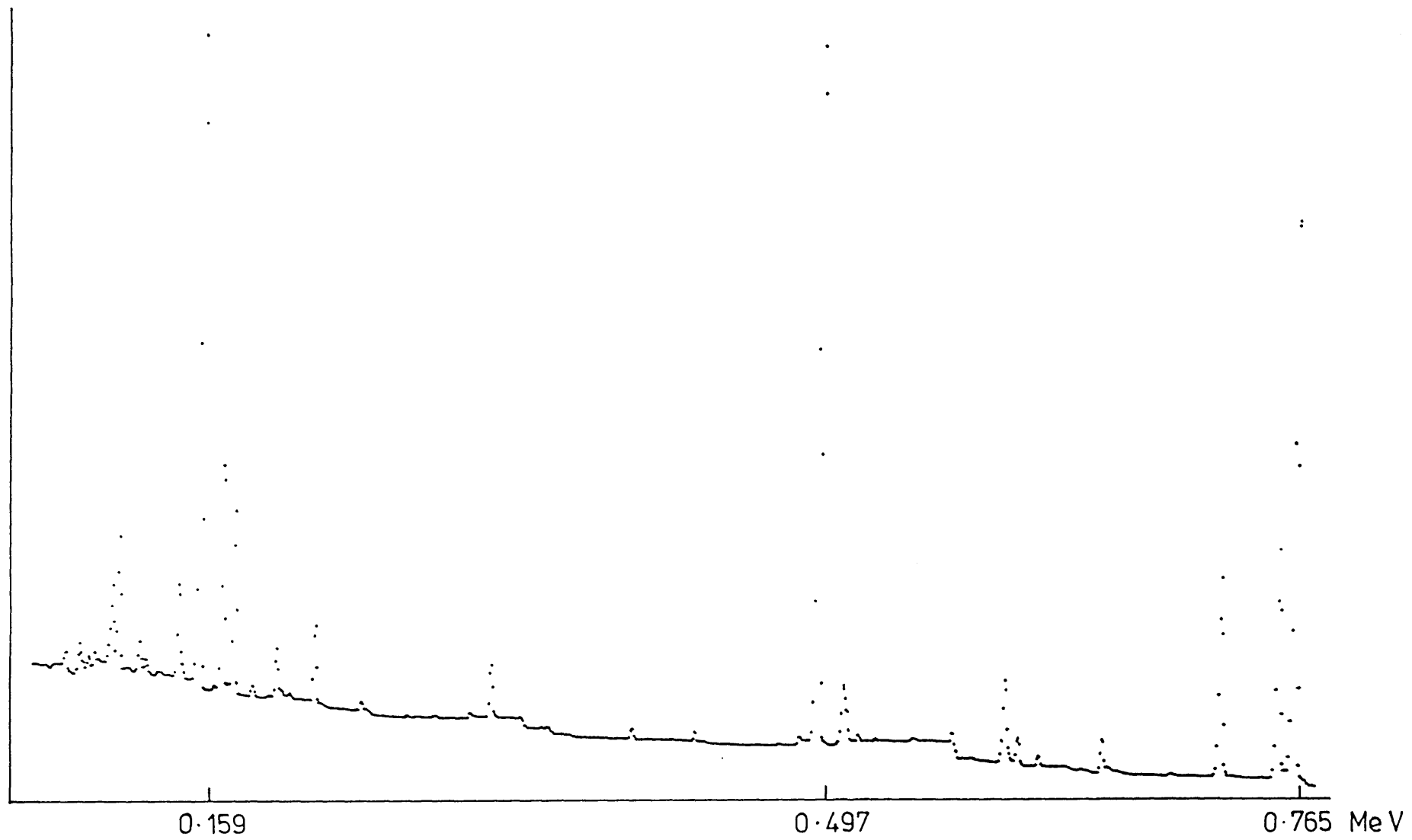


Fig. 4.2a

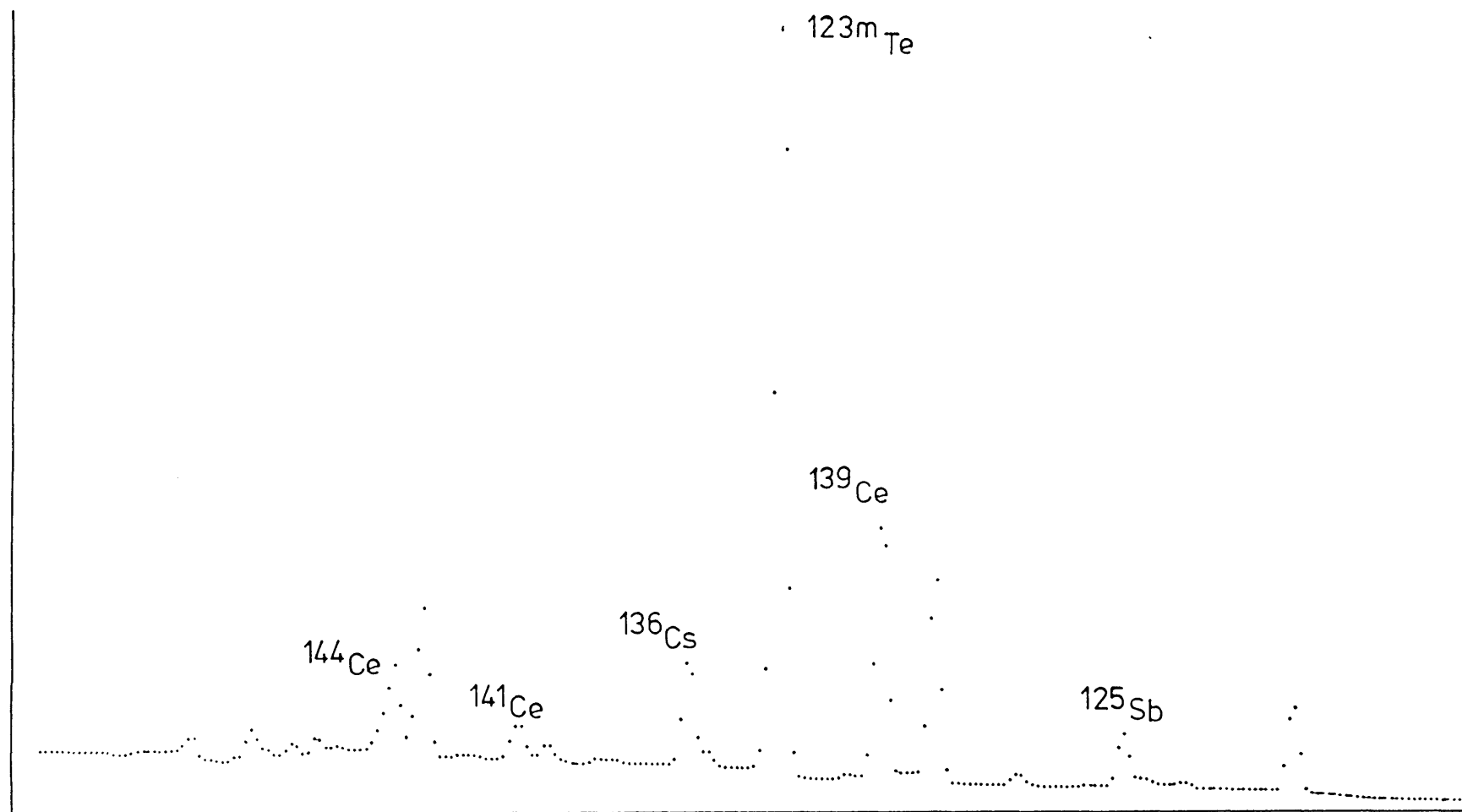


Fig. 4. 2b

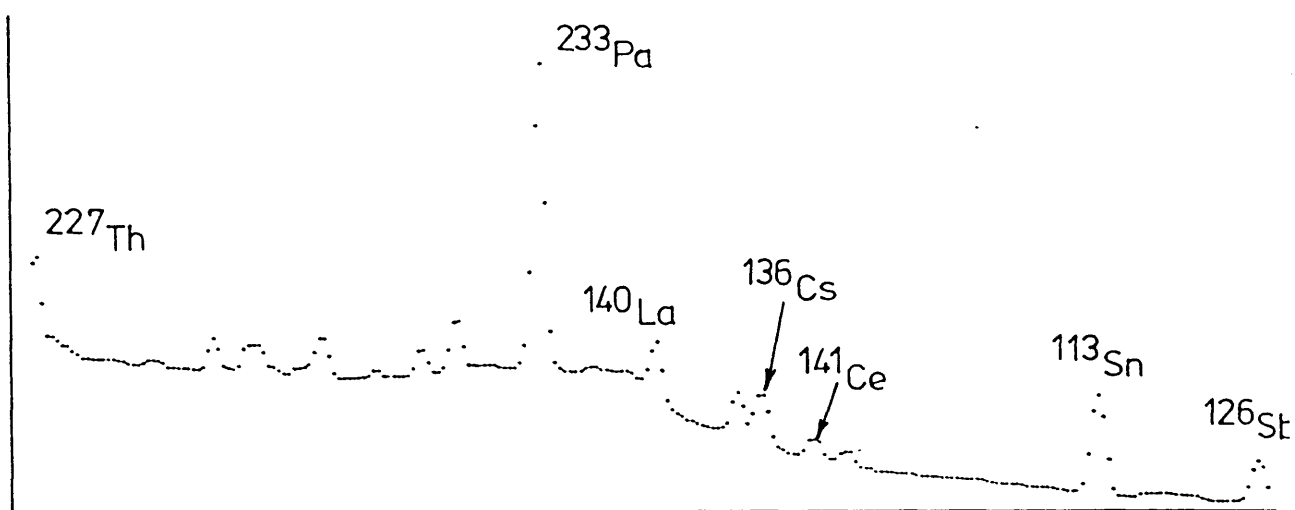


Fig. 4. 2c

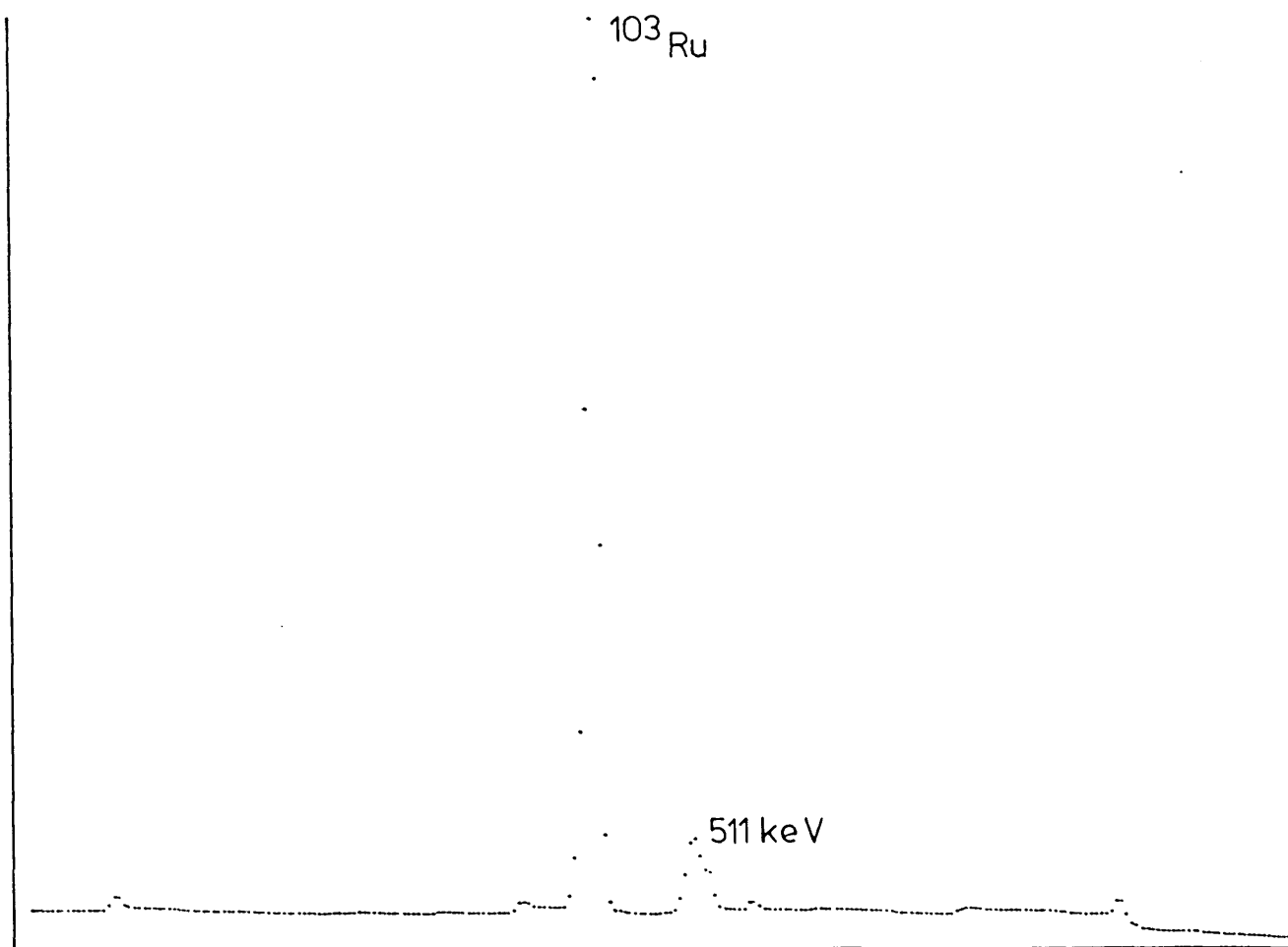


Fig. 4. 2d

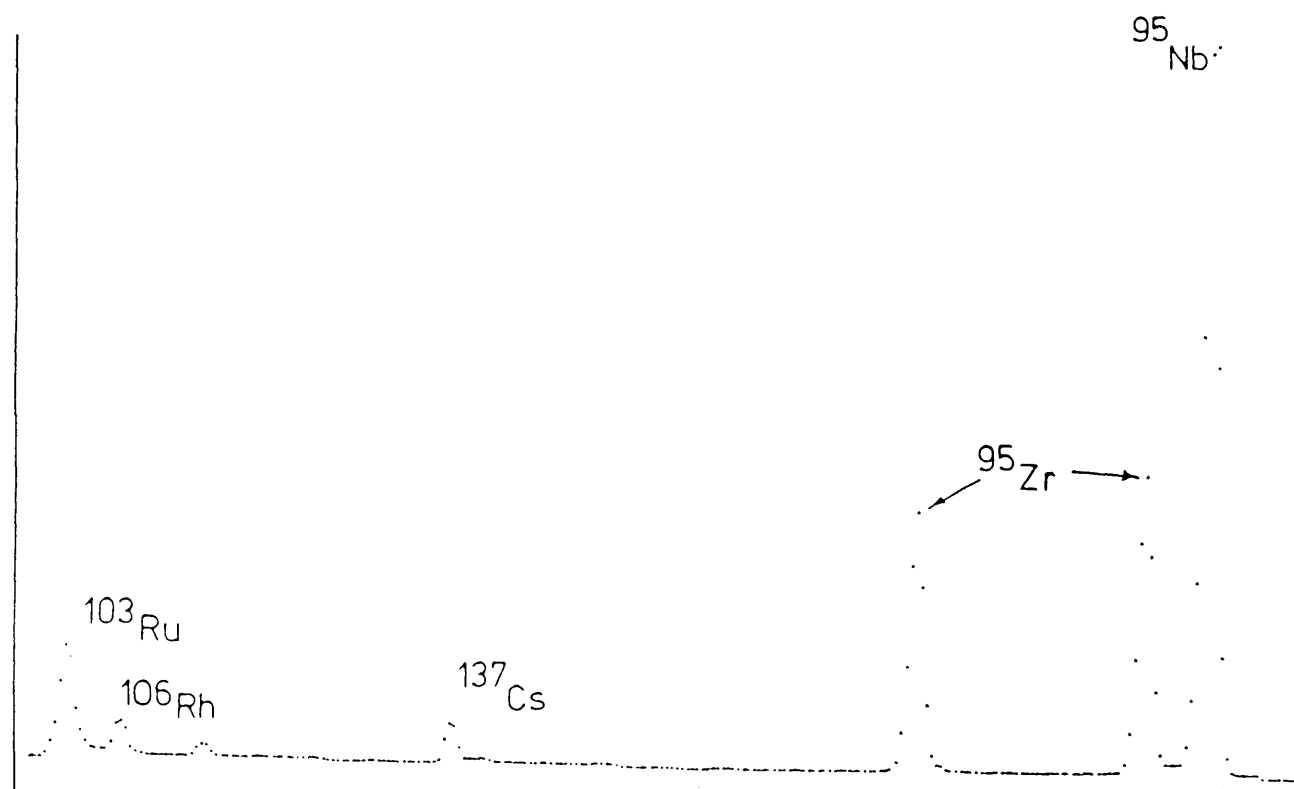


Fig. 4. 2e

The calculation of the half-life, from the data obtained after recounting the sample, assured nuclide identification. The data was fitted using a least squares algorithm to plot $\ln$ count rate versus elapsed time, the slope of the best fitted line is equivalent to $-\lambda$. Nuclides having only one gamma transition were mostly treated in this manner. Calculated half-lives agree to within a few per cent of reference data. Figure 4.2f is a plot of the measured decay rate of ^{103}Ru .

TABLE 4.3

GAMMA PHOTONS IDENTIFIED AND THE ASSOCIATED NUCLIDES

γ -ENERGY (KeV)	NUCLIDE	% BRANCHING
98.44	^{153}Gd	
101.2	^{237}U	
103.97	^{153}Gd	21.8%
105.49	^{230}Pa	
111.	^{233}Pa	
117.56	^{237}U	
121.21	^{75}Se	15.8%
124.44	^{131}Ba	28 %
133.58	^{144}Ce	11.1%
136.07	^{75}Se	53.9%
145.42	^{141}Ce	48.4%
159.09	$^{123\text{m}}\text{Te}$	
162.6	^{140}Ba	6.1%
165.96	^{139}Ce	79.9%
176.36	^{125}Sb	6.8%

TABLE 4.3 (cont.)

γ -ENERGY (KeV)	NUCLIDE	% BRANCHING
185.84	^{226}Ra	4.0%
208.04	$^{95\text{m}}\text{Te}$	70.0%
212.3	$^{121\text{m}}\text{Te}$	82.0%
216.09	^{131}Ba	21.75%
238.76	^{227}Th	15.0%
264.91	^{75}Se	58 %
280.01	^{75}Se	24.88%
312.02	^{233}Pa	37 %
344.13	^{136}Cs	53 %
356.32	^{141}Ce	
365	^{131}I	82 %
392.37	^{139}Ce	
400.91	^{75}Se	11.57%
428.2	^{125}Sb	30 %
447.33	^{230}Pa	5.3%
463.86	^{125}Sb	10.5%
475.92	^{103}Ru	
486.65	^{140}La	43 %
496.75	^{103}Ru	
507.93	^{121}Te	17.66%
520.05	^{83}Rb	46 %
529.36	^{83}Rb	30.17%
550.37	^{140}Ba	23.6%
572.43	^{121}Te	80.3%
582.9 8	$^{95\text{m}}\text{Tc}$	
602.45	^{124}Sb	98.3%
610.02	^{103}Ru	
621.65	^{106}Rh	

4.3 (cont.)

-ENERGY(KeV)	NUCLIDE	% BRANCHING
635.52	^{125}Sb	11.2%
645.55	^{124}Sb	7.23%
657.43	$^{110\text{m}}\text{Ag}$	
661.35	^{137}Cs	
666.	^{126}Sb	99.7%
677.21	$^{110\text{m}}\text{Ag}$	10.7%
686.62	$^{110\text{m}}\text{Ag}$	6.4%
696.14	^{126}Sb	31.9%
706.38	$^{110\text{m}}\text{Ag}$	16.7%
723.73	^{95}Zr	49 %
756.35	^{95}Zr	49 %
765.42	^{95}Nb	
801.77	^{134}Cs	88 %
814.15	^{140}La	22.4%
818.54	$^{110\text{m}}\text{Ag}$	72.78%
834.20	$^{95\text{m}}\text{Tc}$	
885.24	$^{110\text{m}}\text{Ag}$	
898.54	^{88}Y	
1049.8*	^{106}Rh	
1098.84	^{59}Fe	
1290.88	^{59}Fe	43.2%
1383.63	$^{110\text{m}}\text{Ag}$	
1594.74	^{140}La	95.5%
1690.47	^{124}Sb	
1836.63	^{88}Y	99.34%

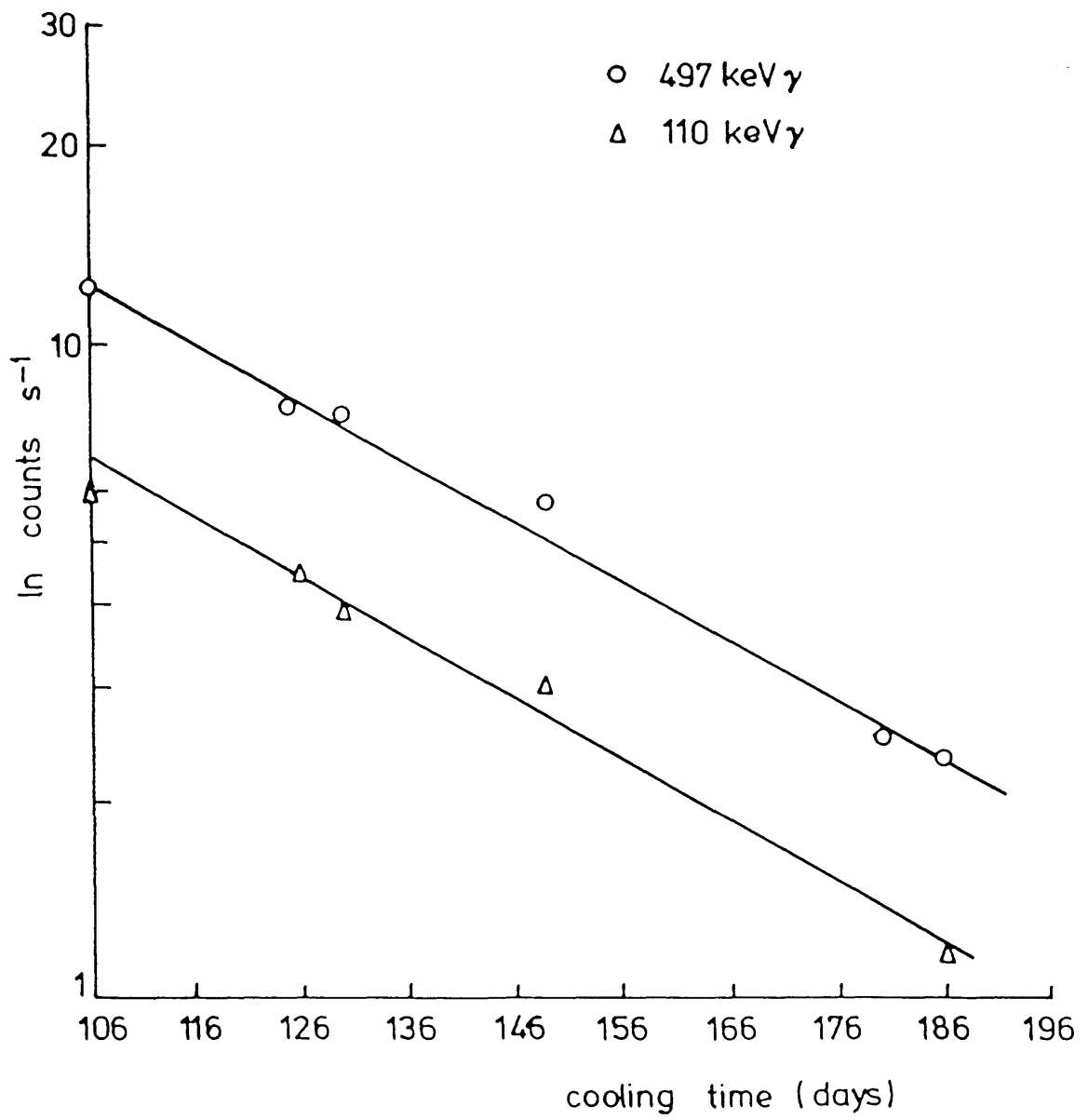


Fig. 4.2f Measured decay of ^{103}Ru

4.3 CROSS-SECTION CALCULATIONS of γ -EMITTERS

The quantities of the radio-nuclides produced were calculated from the accurately determined photopeak areas. Corrections for the calculated detector efficiency and the branching in the nuclides decay scheme were taken into account. [Appendix B lists the calculated peak areas and background areas for each gamma energy at time t_e , the time since irradiation, when counted]. Decay scheme data was taken from 'Table of Isotopes' (Seventh Edition) ^(4.8). The decay of the nuclide since target irradiation was taken into account, using the exponential decay law:

$$N_e = N_i \{ \exp -\lambda t_e \}$$

where

$$N_i = N_e / \{ \exp -\lambda t_e \}$$

N_e = amount of nuclide at time of counting.

N_i = amount of nuclide at end of irradiation.

λ = decay constant.

t_e = time since irradiation.

The quantity produced at the end of irradiation allowing for any loss of the nuclide, by decay, during the irradiation is given by the following expression:

$$R = N_i \lambda / 1 - \{ \exp -\lambda t_i \}$$

$$N_o = R t_i$$

R = rate of production of the nuclide.

t_i = irradiation time.

N_o = amount of nuclide produced by the irradiation.

The probability of a nuclear reaction occurring is expressed in terms of its 'cross-section' which can be defined as the ratio of the number events giving a product β per nucleus per unit time over the number of incident particles per unit area per unit time.

$$\sigma_{\alpha} = \frac{N_o \text{ events type } \alpha \text{ per nucleus per unit time}}{N^o \text{ incident particles per unit area per unit time}}$$

In the hypothetical case of a spherical nucleus the geometric cross-section corresponds to the area of a circle presented by the nucleus to the incoming particles. This is a simple geometric collision and $\sigma_{\text{geom}} = \pi R^2$ where R is the radius of the circle. [6×10^{-13} cm is an average value for a nuclear radius and this gives a value in the order of 10^{-24} cm² for σ_{geom}].

The calculation of the production cross-section of the nuclides listed below in table 4.3 is determined according to the relationship :

$$\sigma = N_o / N_t \times \phi_p$$

σ = cross-section

N_t = number of target atoms analysed.

ϕ_p = number of protons impinging upon the target per unit area per unit time (cm⁻²s⁻¹).

Cross-sections which are less than 10^{-24}cm^2 represent much rarer than a geometric collision. Those greater than 10^{-24}cm^2 indicate an incident is possible even if the incoming particle passes some distance from the nuclear surface.

Table 4.4 contains the cross-sections of the identified nuclides calculated from the formulae given above, at different cooling times. The variation of the mean cross-section with nuclide mass is depicted graphically in figure 4.3. The measurements of the cross-section have been made from either the most abundant gamma transition or the normalised transition given in the 'Table of Isotopes' (4.9).

TABLE 4.4 MEASURED CROSS-SECTIONS OF GAMMA EMITTERS

<u>NUCLIDE</u>	<u>COOLING TIME</u>	<u>CROSS-SECTION</u> (σ)	<u>MONTE CARLO</u> σ
^{75}Se	18.748 E+6	1.245mb	$\pm 0.055\text{mb}$
		1.570mb	$\pm 0.061\text{mb}$
	σ -average	1.408mb	$\pm 0.061\text{mb}$
^{83}Rb	7.252 E+6	3.369mb	$\pm 0.395\text{mb}$
	7.689 E+6	3.658mb	$\pm 0.349\text{mb}$
	9.417 E+6	4.327mb	$\pm 0.35\text{mb}$
	σ -average	3.785mb	$\pm 0.364\text{mb}$
^{88}Y	7.257 E+6	6.79mb	$\pm 0.4\text{mb}$ 4.481mb
	7.776 E+6	7.7 mb	$\pm 0.4\text{mb}$
	9.158 E+6	7.1 mb	$\pm 0.4\text{mb}$
	σ -average	7.2 mb	$\pm 0.4\text{mb}$

<u>NUCLIDE</u>	<u>COOLING TIME</u>	<u>CROSS-SECTION</u> (σ)	<u>MONTE CARLO</u> σ
^{95}Zr	5.788 E+6	94.7 mb ± 3.5 mb	34.72mb
	9.158 E+6	94.2 mb ± 3.5 mb	
	σ -average	94.7 mb ± 3.5 mb	
^{103}Ru	5.788 E+6	118.56mb ± 3.85 mb	19.141mb
	7.257 E+6	112.89mb ± 3.54 mb	
	9.417 E+6	112.89mb ± 3.54 mb	
	σ -average	114.82mb ± 3.65 mb	
$^{110\text{m}}\text{Ag}$	5.788 E+6	6.48mb ± 0.66 mb	6.171mb
	14.452 E+6	6.38mb ± 0.4 mb	
	17.453 E+6	6.38mb ± 0.4 mb	
	σ -average	6.43mb ± 0.5 mb	
^{121}Te	5.788 E+6	12.46mb ± 1 mb	1.984mb
	7.257 E+6	16.97mb ± 1.2 mb	
	9.504 E+6	21.9 mb ± 1.26 mb	
	σ -average	17.13mb ± 1.1 mb	
$^{121\text{m}}\text{Te}$	9.504 E+6	4.56mb ± 0.22 mb	1.984mb
	12.44 E+6	4.51mb ± 0.2 mb	
	σ -average	4.51mb ± 0.2 mb	
$^{123\text{m}}\text{Te}$	9.504 E+6	7.9 mb ± 0.2 mb	3.783mb
	12.44 E+6	7.85mb ± 0.2 mb	
	σ -average	7.85mb ± 0.2 mb	

<u>NUCLIDE</u>	<u>COOLING TIME</u>	<u>CROSS-SECTION</u> (σ)	<u>MONTE CARLO</u> σ
^{124}Sb	5.788 E+6	13.17mb $\pm 0.66\text{mb}$	6.539mb
	7.257 E+6	12.26mb $\pm 0.55\text{mb}$	
	7.689 E+6	12.97mb $\pm 0.55\text{mb}$	
	9.417 E+6	13.1 mb $\pm 0.55\text{mb}$	
	σ -average	12.87mb $\pm 0.55\text{mb}$	
^{125}Sb	7.257 E+6	24.47mb ± 2.5 mb	9.995mb
	7.689 E+6	24.12mb ± 2 mb	
	σ -average	24.3 mb ± 2 mb	
^{126}Sb	5.788 E+6	8.05mb ± 1 mb	8.745
	7.257 E+6	7.7 mb ± 1.1 mb	
	7.689 E+6	8.01mb ± 1.1 mb	
	σ -average	7.9 mb $\pm 1.05\text{mb}$	
^{134}Cs	7.257 E+6	9.27mb $\pm 0.96\text{mb}$	4.115mb
	12.139 E+6	6.08mb ± 0.4 mb	
	σ -average	7.6 mb ± 0.7 mb	
^{136}Cs	5.788 E+6	17.33mb ± 1.1 mb	3.562mb
	7.257 E+6	19.76mb ± 1.7 mb	
	7.776 E+6	24.8 mb ± 2 mb	
	σ -average	20.1 mb ± 1.6 mb	
^{137}Cs	17.453 E+6	75.65mb ± 7.6 mb	3.62mb
		67.13mb ± 8.1 mb	
	σ -average	71.4mb ± 8.1 mb	

<u>NUCLIDE</u>	<u>COOLING TIME</u>	<u>CROSS-SECTION</u> (σ)	<u>MONTE CARLO</u> σ
^{139}Ce	9.504 E+6	5.87mb $\pm 0.16\text{mb}$	2.167mb
	12.44 E+6	6.03mb $\pm 0.18\text{mb}$	
	σ -average	5.93mb $\pm 0.17\text{mb}$	
^{140}Ba	5.788 E+6	56.8 mb ± 4.5 mb	21.032mb
	7.689 E+6	53.1 mb $\pm 0.55\text{mb}$	
	σ -average	54.9 mb ± 2.6 mb	
^{141}Ce	9.504 E+6	38.18mb ± 1.2 mb	13.206mb
	12.44 E+6	38.13mb $\pm 1.26\text{mb}$	
	σ -average	38.15mb ± 1.1 mb	
^{144}Ce	9.504 E+6	32.93mb ± 3.3 mb	14.2838mb
	12.44 E+6	29.99mb ± 3.2 mb	
	σ -average	31.4 mb ± 3.3 mb	
^{153}Gd	9.504 E+6	7.7mb $\pm 0.66\text{mb}$	0.624mb
^{233}Pa	9.504 E+6	12.6 mb ± 8 mb	3.673mb
	12.44 E+6	14. mb ± 13 mb	
	σ -average	13.3 mb ± 10 mb	

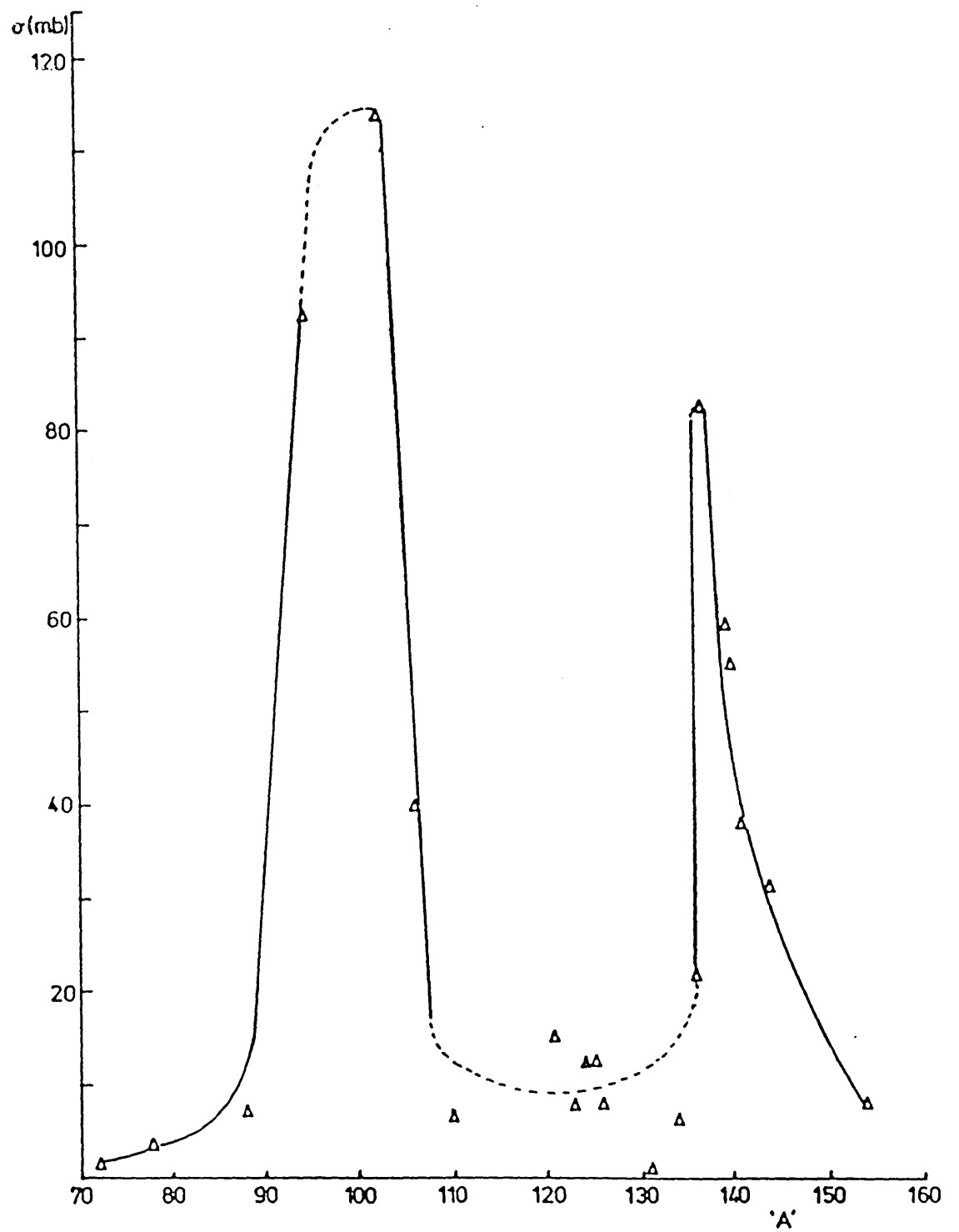


Fig. 4.3 Yields of reaction products in the mass range ~ 70 to ~ 150

4.4 ALPHA ANALYSIS

The quantitative analysis of alpha emitting actinides and polonium were carried out as previously described (Chapter 3, 3.7). Measurements of the alpha emitting nuclides have been made using silicon surface barrier detectors [EG&G ORTEC]. Initially a non-ruggedised device [BA 16-100-100] was used. This was subsequently replaced by a ruggedised detector [BR 24-300-100] due to the eventual increased background of the former detector.

For the quantitative analysis the detectors were efficiency calibrated using standardised solutions of plutonium and americium (^{239}Pu and ^{241}Am) supplied by UKAEA Harwell [See table 4.4]. Single nuclide sources were prepared by evaporation of an aliquot of solution onto the surface of a clean 12.5mm diameter stainless steel planchette. This produced sources in the same geometric form as those prepared from the analyses. Small quantities of solution were pipetted out to avoid sputtering and to produce a thin uniform film of material. These were then flamed to red heat in a bunsen flame to give an adherent oxide deposit. All samples were mounted centrally on a support directly below the detector at a fixed distance to ensure that the counting geometry was reproducible.

TABLE 4.5

ALPHA DETECTORS EFFICIENCIES

DETECTOR	ACTIVITY	PEAK AREA		EFFICIENCY	
Non-Ruggedised					
	²³⁹ Pu				
	5.63nCi	8025	89.6	12.91 ± 0.3	%
	5.63nCi	7944	89.1	12.78 ± 0.3	%
			Avg.	12.84 ± 0.3	%
Ruggedised					
	²⁴¹ Am				
	50pCi	26482	162.42	23.38 ± 0.43	%
	50pCi	22039	148.45	23.04 ± 0.44	%
	²³⁹ Pu				
	28pCi	13850	117.68	23.39 ± 0.28	%
	28pCi	4972	70.5	23.55 ± 0.63	%
	28pCi	3871	62.22	22.88 ± 0.65	%
			Avg.	23.25 ± 0.48	%

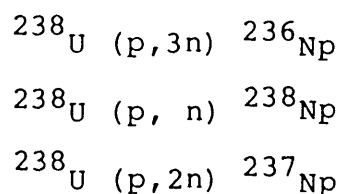
Compared with gamma spectrometry the measurement of a peak area is less complicated as the detector shows a virtual zero background initially, and there is no compton effect. The effect of overlap occurs for peaks with energies close enough to be within the FWHM of the device, i.e peaks less than 17keV or 24keV apart for the non-ruggedised and ruggedised devices respectively. If the source thickness is great enough to produce excessive self absorption broadening, and hence overlap of peaks, which otherwise would have been resolvable occurs.

4.5 DETECTION AND ESTIMATION OF α EMITTING NUCLIDES

Using the separation techniques outlined in Chapter 2 the following alpha emitting nuclides were identified and were subsequently quantified:

Plutonium: ^{238}Pu , ^{236}Pu
 Uranium: ^{230}U
 Thorium: ^{228}Th , ^{227}Th
 Polonium: ^{210}Po , ^{209}Po , ^{208}Po

Plutonium separated from the irradiated material by a combination of ion-exchange and liquid extraction gave recoveries in the order of 20-40%. This was substantially lower than those measured by direct extraction as the TTA complex, which gave recoveries in the order of 50-80%. However the results from both methods are in good agreement. The extraction of neptunium as a TTA complex was found to be up to 65% (^{235}Np being the tracer used), but no other isotopes of this element were observed. The levels of ^{237}Np which may be expected would be of the same order of magnitude as those of ^{236}Pu and ^{238}Np which are the daughters of ^{236}Np and ^{238}Np respectively formed from the reactions:



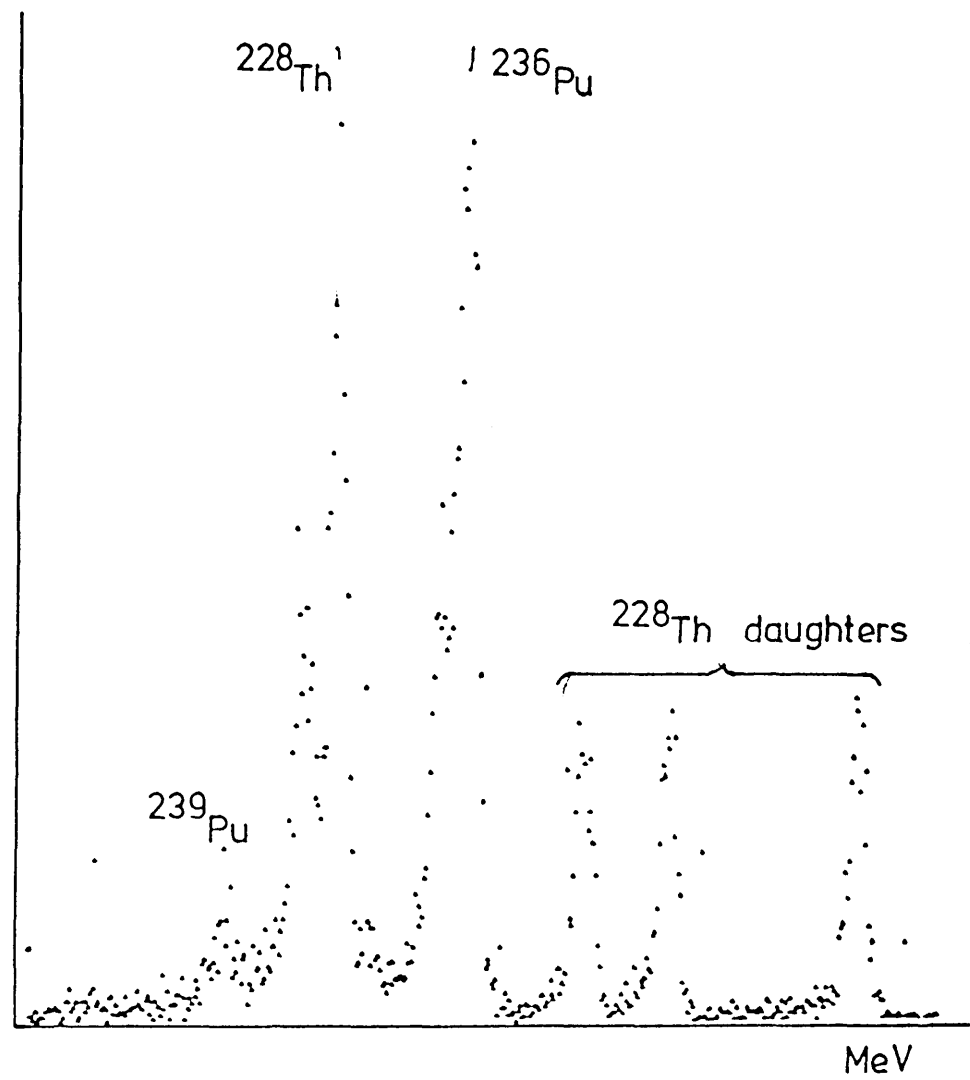


Fig. 4. 5a Electroplated plutonium and thorium

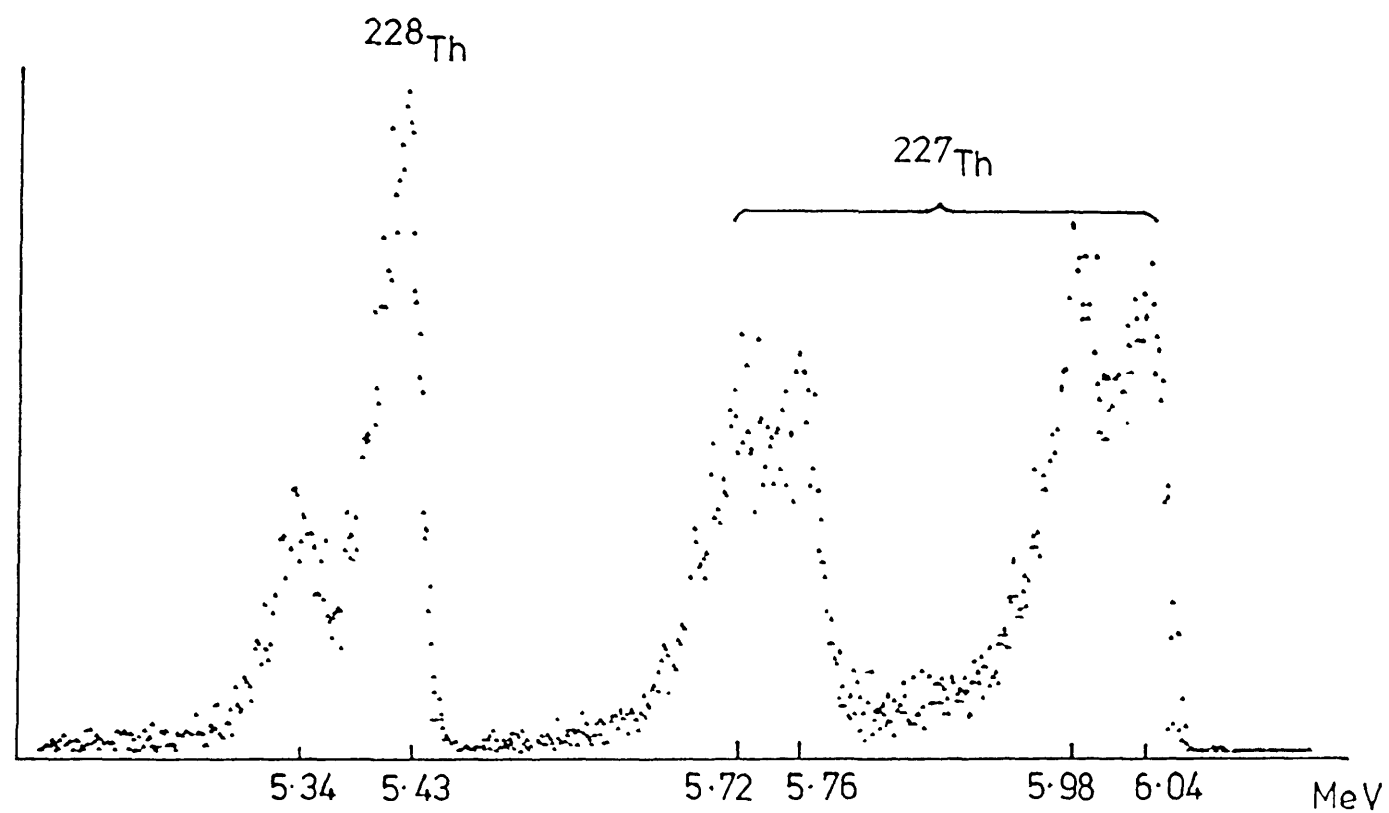


Fig. 4. 5b Alpha spectrum of thorium isotopes (TTA complex)

Analysis of the small quantities of ^{237}Np produced in these irradiations are made more difficult due to its long half-life of 2.14×10^6 years. ^{236}Np and ^{238}Np have much shorter half-lives, 22 hours and 2.1 days respectively, and can be measured from the activity of their daughters ^{236}Pu and ^{238}Pu . A comparison of the decay constants of these nuclides shows the activity of ^{236}Pu and ^{238}Pu to be in the order of 1×10^6 and 2×10^4 times greater than that of ^{237}Np respectively [λ ^{236}Pu $7.7 \times 10^{-9} \text{ s}^{-1}$; λ ^{238}Pu $2.5 \times 10^{-10} \text{ s}^{-1}$; λ ^{237}Np $1 \times 10^{-14} \text{ s}^{-1}$] Even using large quantities of the irradiated solution the presence of any ^{237}Np produced was below the limit of detection.

The alpha emitting nuclide of uranium, ^{230}U , produced by spallation was quantified without the use of a standard spike. A uranium source was prepared and the peak areas of ^{234}U , ^{235}U and ^{238}U were compared with that of ^{230}U . It was assumed that no measurable changes had occurred in the ratios of these isotopes. From this the percentage abundance of each isotope was calculated and hence the amount of ^{230}U in the whole target. The data and the calculations for this are set out below.

Thorium separations were by ion-exchange and TTA extraction consecutively as outlined in section 2.3c. The thorium being isolated from the target uranium and other nuclides to produce an evaporated source for counting.

This gave recoveries ranging between 11 and 24%. Direct extraction of thorium was not attempted as the conditions for its extraction are such that trivalent ions would also be recovered for example some lanthanides. This would produce an overly thick source giving a degraded spectrum.

The recovery of polonium was taken as 100% by plating onto several pieces of silver from dilute hydrochloric acid containing a reducing agent. The majority of the polonium was plated onto the first planchette, with a second piece plating the residual polonium. [A final piece of silver showed no polonium]. Therefore by adding the activities of the first and second planchettes quantitative measurements were made. The cross-sections for the polonium isotopes have been measured in all three target discs of the first irradiation.

The production cross-sections for all the alpha emitting nuclides were calculated from accurately determined peak areas, which had been corrected for the calculated detector efficiency and the branching in the nuclide decay schemes.[Decay scheme data was taken from Table of Isotopes seventh edition]. The decay of the nuclide since target irradiation and loss by decay during irradiation were also taken into account.

The calculations were identical to those used in section 4.3 above. The results are given below in table 4.6 .

URANIUM ISOTOPES CALAULATION

Isotope	Area (counts)	% total
^{238}U	10564	99.31%
^{235}U	460	0.681%
^{234}U	10570	$5.41 \times 10^{-3}\%$
^{230}U	695	$8.8 \times 10^{-11}\%$

The total sample from which the uranium was analysed was 19.2235 grammes. Therefore the total number of ^{230}U atoms in the sample is given by:

$$\begin{aligned} (19.2235/238.029) \times 6.023 \times 10^{23} \times (8.8 \times 10^{-11}/100) \\ = 4.28 \times 10^{10} \text{ atoms} \end{aligned}$$

When corrected for decay during and since irradiation this gives:

$$2.0567 \times 10^{11} \text{ atoms } ^{230}\text{U}$$

The cross-section σ is given by:

$$2.0567 \times 10^{11} / (4.863 \times 10^{22} \times 8.486 \times 10^{15})$$

$$\sigma = 0.498 \text{ mb}$$

TABLE 4.6

<u>ISOTOPE</u>	<u>SAMPLE</u>	<u>CROSS-SECTION</u>	<u>ERROR</u>
^{238}Pu		1.11mb	0.06mb
		1.08mb	0.05mb
		1.11mb	0.04mb
		1.01mb	0.08mb
^{236}Pu		0.97mb	0.06mb
		0.96mb	0.06mb
		0.93mb	0.05mb
		0.99mb	0.08mb
		0.91mb	0.04mb
^{237}U		251.85mb	8.21mb
^{230}U		0.498mb	0.05mb
^{228}Th		3.3 mb	0.18mb
		3.43mb	0.2 mb
		3.28mb	0.14mb
		3.32mb	0.18mb
^{227}Th		2.8 mb	0.3 mb
		2.83mb	0.25mb
		3.08mb	0.14mb

TABLE 4.6 (contd)

<u>ISOTOPE</u>	<u>SAMPLE</u>	<u>CROSS-SECTION</u>	<u>ERROR</u>
^{208}Po	19.3 gl^{-1}	9.82mb	0.93mb
		9.8 mb	0.91mb
	16.18 gl^{-1}	9.8 mb	0.93mb
		9.5 mb	0.95mb
	19.93 gl^{-1}	8.03mb	0.9 mb
		8.04mb	0.9 mb
		8.17mb	0.9 mb
^{209}Po	19.3 gl^{-1}	17.76mb	1.54mb
		17.96mb	1.9 mb
	16.18 gl^{-1}	18.16mb	0.92mb
		18.24mb	0.98mb
	19.93 gl^{-1}	15.55mb	0.6 mb
		15.15mb	0.58mb
		15.45mb	0.6 mb
^{210}Po	19.3 gl^{-1}	10.71mb	0.48mb
		10.7 mb	0.48mb
	16.18 gl^{-1}	10.63mb	0.48mb
		10.62mb	0.48mb
	19.93 gl^{-1}	11.33mb	0.52mb
		11.28mb	0.53mb
		11.22mb	0.53mb
^{210}Pb	19.93 gl^{-1}	0.72mb	

4.6 DISCUSSION

The imminent commissioning of the Spallation Neutron Source (SNS) at the Rutherford-Appleton Laboratory produced a requirement for the measurement of radionuclide production in proton irradiated uranium. The build up of radioactivity during the lifetime of the target was of concern from a radiological safety point of view in particular as regards possible accident scenarios and in connection with ultimate disposal. Although estimates of the formation of irradiation products were obtained from the general calculations involved in the target design it was felt appropriate to check these by direct measurement. Such measurements in turn would provide some corroboration of the general calculational procedure and would also provide comparison with measurements of yields made some 25-30 years ago using equipment much less sensitive than that available today.

The aim of this work was to identify and quantify the yields of spallation and fission products produced when natural uranium was irradiated with 800 MeV protons. From the quantitative analysis the direct production cross-section of some nuclides and the cumulative cross-section for nuclides in beta decay chains have been calculated and comparisons have been made with cross-sections computed using Monte-Carlo codes.

Of significance is the production of the alpha emitting nuclides of plutonium and polonium, the latter element is volatile at the predicted target operating temperature of 348°C (along the centre line in the first plates). These nuclides could constitute a particular radiological hazard to workers in the target area. The build up of nuclides can also be used as an estimate of the 'burn up' of uranium and consequently the targets useful life.

In the late 1940's, 50's, and early 1960's experimental work on the irradiation of uranium with protons and helium ions was undertaken along with irradiations of other elements. From this work models for high energy processes were developed as outlined in Chapter 1 and led to the development of Monte-Carlo (4.10) calculations. Subsequently much work was done on Monte-Carlo methods by Goldberger and Metropolis et al (1958). The model used in these codes for the transportation of particles is based on the intranuclear cascade formulae of Serber for high energy particle-nucleus interactions linked to the Weisskopf formulae for evaporations.

In the design of the target assembly of the SNS, Monte-Carlo codes were used to calculate the cascade and evaporation of nucleons from the target and their transport through the materials assembly. The codes calculate the number of neutrons produced that reach

thermal energy taking account of the losses from the original number produced by absorption and scattering. Neutron induced reactions are also calculated along with the high energy proton induced fission. These together allow the calculation of the yields of nuclides formed.

The codes used were modified versions of those supplied by the Radiation Shielding Information Centre of Oak Ridge National Laboratory. They are essentially made up of two codes: HET for high energy transportation of nucleons and O5R for neutron transport. A third code ORIGEN has been used to calculate the build up and decay of radionuclides using the Bateman equations. The transport of evaporation particles and source protons between collisions were calculated using cross-sections close to the geometric: $\pi r^2 A$ (where A is the atomic Mass).

The SNS target is composed of uranium, zircaloy cladding and D₂O coolant/moderator. Because of this inhomogeneity target calculations are carried out in five stages. The SNS is made up of 28 plates of uranium of varying thickness with coolant channels between the plates. The first section consists of 12 plates 0.65 cm thick. The samples irradiated for this study were 0.623 cm and 0.43 cm thick respectively. This is roughly equivalent to the first disc of the

SNS target and in making this assumption the comparison can give an indication of the degree of agreement between the experimentally measured and Monte-Carlo calculations for the proton interactions at 800 MeV. To enable a reasonable test of the computational technique to be carried out a tailored model was used to predict the yield of products in the irradiations undertaken as described in Chapter 2.

Monte-Carlo modelling of the target irradiated for this research was carried out by Dr. F. Aitchison. The target was 0.623 cm thick and 2.54 cm in diameter and was uniformly irradiated with 1.86×10^{16} 800 MeV protons per cm^{-2} in a 364 minutes period. For modelling purposes the samples are considered 'thin' with approximately a 5% interaction probability. Only contributions from direct proton-uranium interactions and an estimate of the neutron contribution have been considered. The codes used the intranuclear cascade and evaporation model without any internucleus cascade and gave a calculated interaction cross-section of 1890 mb. The total inelastic cross-section of protons on nuclei at high energy was found by Ashmore et al (4.11) to be given by:

$$\sigma_{\text{inel}} = 15.9 \pi A^{2/3}$$

When calculated for uranium this gives an interaction cross-section of 1918 mb. [The geometric cross-section for uranium is approximately 2040 mb, calculated from $\pi r^2 A$].

The calculation examined 10326 high energy interactions and five evaporations were performed for each residual nucleus from the cascade stage giving 51550 evaporations. 27265 of these were fissions which is approximately 55% of the interactions and gives a fission cross-section of 1 barn.

Neutron evaporation contributions are included due to the high number generated in the intranuclear cascade. They have been calculated on the assumption that the 800 MeV protons interact uniformly throughout the target and that the emission of evaporation neutrons is isotropic in the 'laboratory system'. The neutron reactions calculated are: (n,γ) , $(n,2n)$, $(n,3n)$ and $(n,\text{fission})$ in ^{238}U . The cross-sections of the neutron induced reactions total 198 mb and neutron induced fission is 317 mb giving a total of 515 mb. 80 mb is deducted for non-treated nuclides. This gives a Monte-Carlo calculated cross-section of 2330 mb which is in agreement at the 10% level with the empirical formula for inelastic interactions.

The production cross-sections of certain nuclides calculated by this Monte-Carlo method are compared to those identified and measured by gamma and alpha spectrometry in tables 4.4 and 4.6. Figures 4.6a and 4.6b show the Monte-Carlo calculated results plotted as probability against mass number.

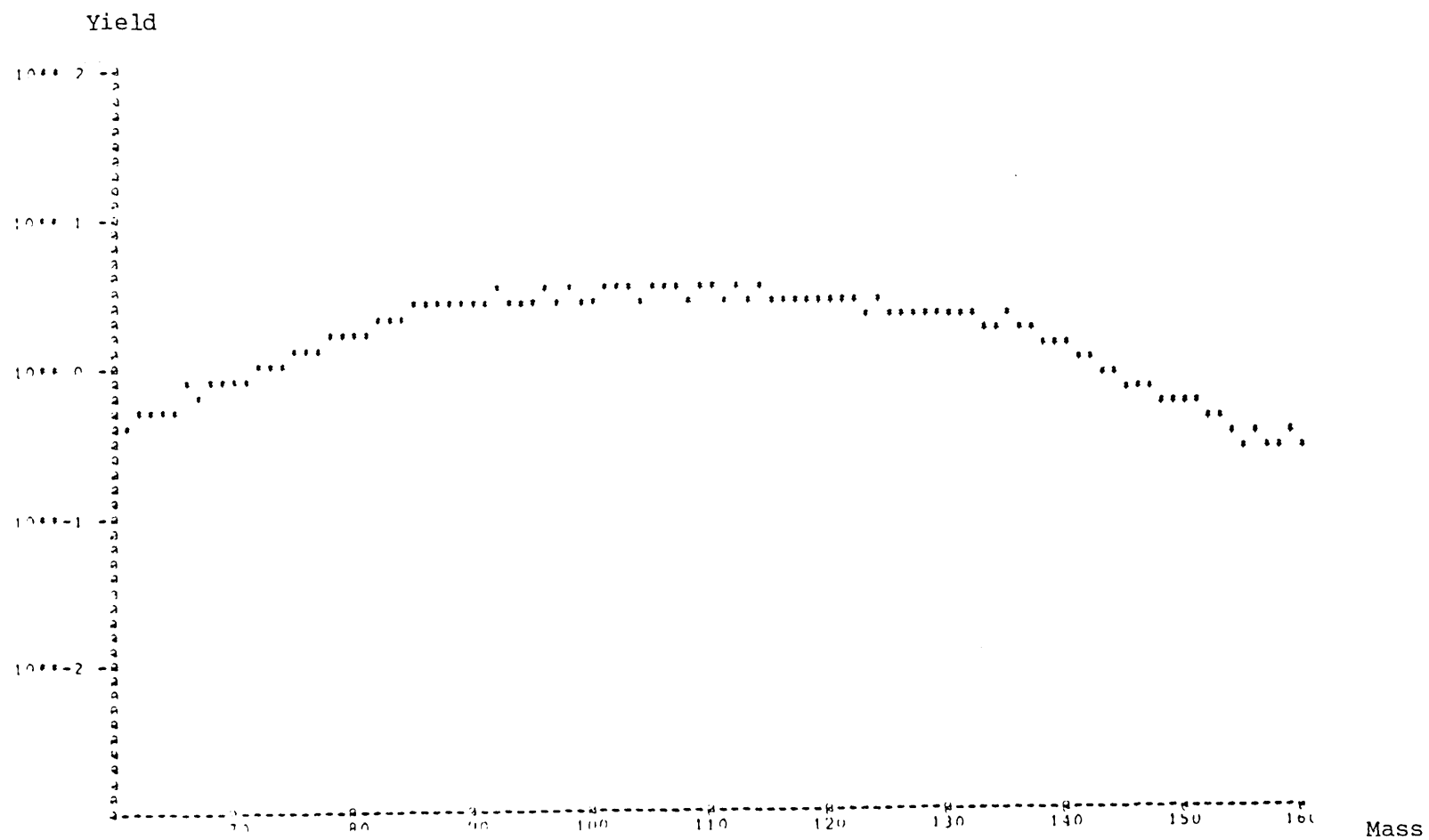


Fig. 4.6a

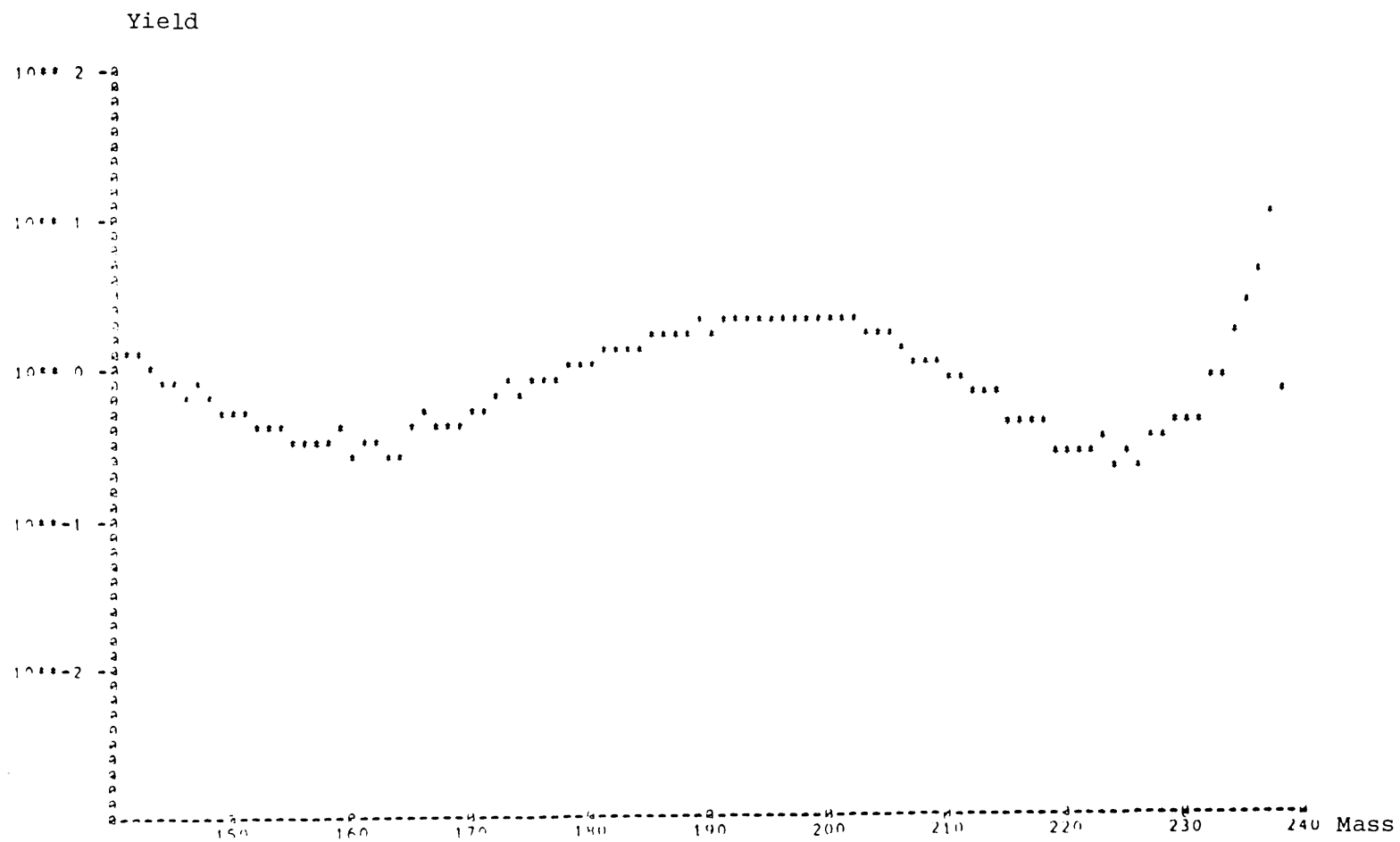


Fig. 4.6b

In figure 4.6a the region shown is that of fission; it can be compared with figure 4.3 on page 112 which is a plot of the measured probabilities for the formation of the fission nuclides identified. [It is not a continuous isobaric yield curve]. Figure 4.6c is a plot of the Monte-Carlo calculation's cross-sections for nuclides identified in this work. [This exhibits a similar form to figure 4.3 due to the omission of nuclides which were not identified experimentally]. Individual yields of some of the nuclides are substantially different though at the limits of the plotted data and in the 'trough' region both experimental and computed results have similar values.

The results obtained for the spallation products close to the mass number of the target are found to be within a factor of two of the predicted values. For example: σ ^{237}U 202 mb calculated and 251.85 mb measured, ^{233}Pa 3.673 mb calculated and 13 mb measured ^{228}Th 1.84 mb calculated and 3.1 mb measured; ^{227}Th 1.46 mb calculated and 2.9 mb measured; ^{210}Po 13 mb calculated and 11 mb measured; ^{208}Po 11.2 mb calculated 8.5 mb measured. Tables 4.7, 4.8 and 4.9 list measured cross-sections from proton irradiated uranium -238 samples of previous workers for comparison. The cross-section for ^{237}U is markedly different, in table 4.7 σ is 22mb at 660 MeV, 63mb at 680 MeV and 50mb at 1.8GeV. For ^{233}Pa σ is 13mb at 660 MeV, 19mb at 680 MeV and 10mb at 1.8 GeV. These two nuclides exhibit a marked variation in σ with

proton energy and the results measured here would fit into the general trend of the previous work. [Giving an excitation function increasing with energy to a maximum probability before decreasing. In the case of polonium the isotope ^{210}Po has been measured at 200, 340 and 660 MeV with 0.17 mb, 1.7 mb and 2.6 mb respectively. The isotopes ^{209}Po and ^{208}Po are reported here for the first time. This data in tables 4.7-4.9 is taken from a compilation of spallation data by E. Brunix of CERN (1969).

In the spallation region fair agreement might reasonably be expected between the model predictions, the earlier work and those of this work. In the first instance the calculation is dealing with comparatively few steps in the cascade process and given that the mechanisms proposed are essentially correct comparatively minor errors are likely to ensue. Although the targets used in these experiments are much thicker than those used previously the expected agreement between the sets of experimental results is not suprising. The spallation products are produced by proton reactions and secondary reactions induced by neutrons are unimportant in this mass range.

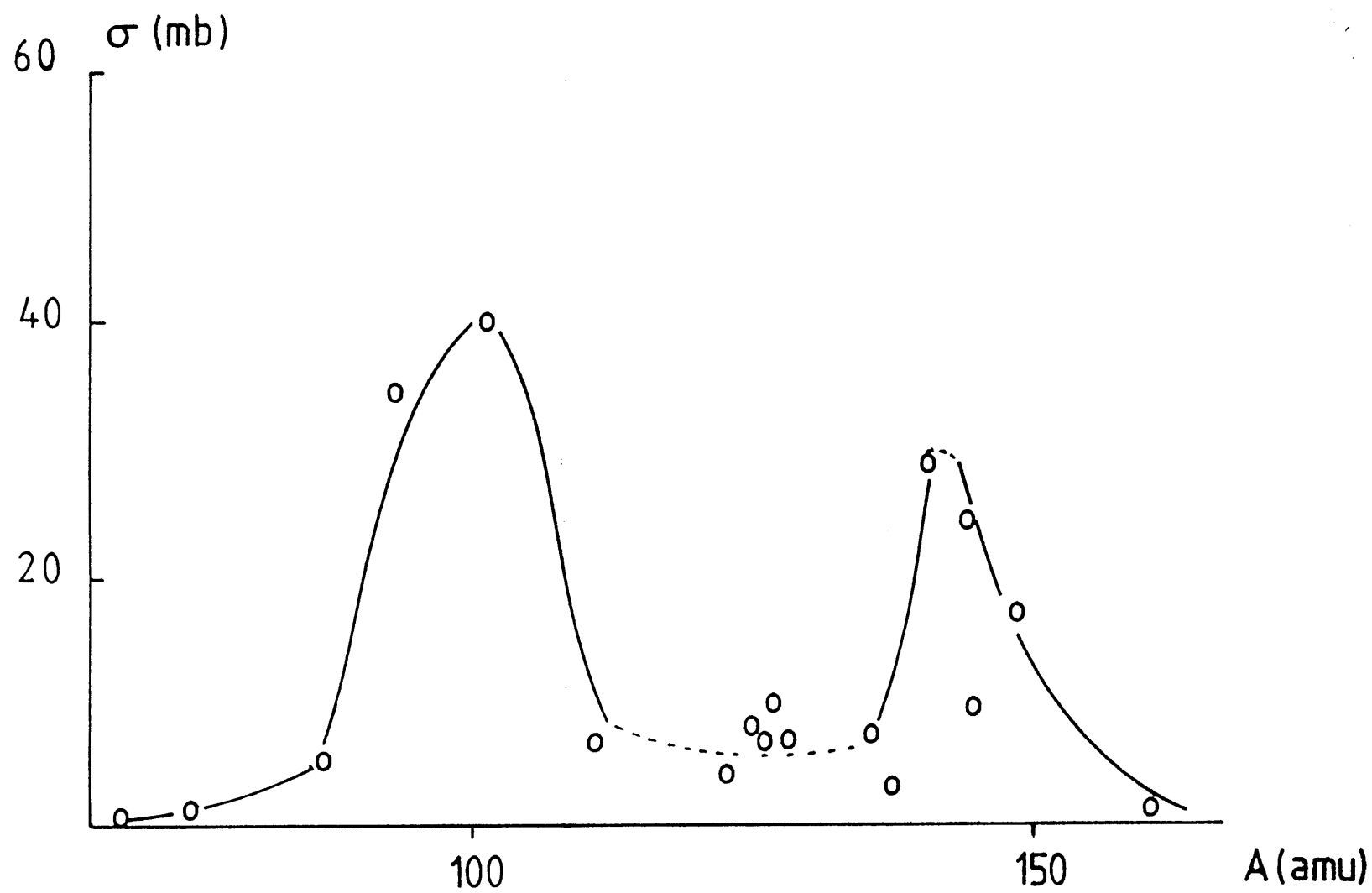


Fig. 4.6c

TABLE 4.7

<u>Nuclide</u>	<u>$\sigma(\text{mb})$</u>
^{236}Np	3.4
^{234}Np	2.2
^{237}U	22
^{230}U	0.26
^{234}Pa	13
^{233}Pa	8.7
^{232}Pa	1.7
^{228}Pa	0.4
^{234}Th	4.7
^{231}Th	12
^{226}Ac	4.8
^{225}Ac	1.5
^{225}Ra	7.8
^{224}Ra	14
^{223}Ra	8
^{212}Fr	0.2
^{211}At	1.2
^{207}At	0.2
^{210}Po	2.6
^{207}Po	1.7
^{206}Po	0.3
^{204}Po	3.7
^{212}Bi	0.3
^{210}Bi	1.1
^{206}Bi	0.9
^{204}Bi	0.52
^{203}Bi	0.15
^{209}Pb	0.2
^{203}Pb	2.8

TABLE 4.7(cont)

URANIUM

660 MeV (cont.)

<u>Nuclide</u>	<u>σ(mb)</u>
^{200}Pb	0.24
^{202}Tl	6.4
^{201}Tl	2.4
^{199}Tl	0.24
^{203}Hg	0.18
^{197}Hg	0.2
$^{193+195}\text{Hg}$	0.04
^{196}Au	0.4
^{195}Au	0.5
$^{193+196m}\text{Au}$	0.15
^{192}Au	0.16
^{197}Pt	0.5
$^{195m+191+193m}\text{Pt}$	0.5
^{198}Pt	0.45

Monitor : $^{27}\text{Al}(p, 3pn)^{24}\text{Na}$
 $\sigma(\text{mb}) = \text{not stated.}$

Error : Not stated.

- Radiochemical investigation of the products of the fission of uranium by protons with an energy of 660 MeV.
- A.K. Lavrukhina and S. Rodin.
- Radiokhimiya, 2, 83 (1960).

E_p	0.68	1.0	1.8	3.0	6.2
<u>Nuclide</u>	<u>$\sigma(\text{mb})$</u>				
^{238}Np	0.25 $\pm$ 0.04				
^{236}Np	0.26 $\pm$ 0.03				
^{237}U	63 $\pm$ 10		50 $\pm$ 9		
^{232}U	4.7 $\pm$ 0.7		2.9 $\pm$ 0.6		
^{230}U	0.23 $\pm$ 0.04		0.18 $\pm$ 0.04		
^{229}U	0.05 $\pm$ 0.01		0.036 $\pm$ 0.008		
^{228}U	0.024 $\pm$ 0.005		0.015 $\pm$ 0.004		
^{237}Pa	13 $\pm$ 2		6.3 $\pm$ 0.5	9 $\pm$ 1	7 $\pm$ 0.8
^{235a}Pa	17 $\pm$ 2		6.8 $\pm$ 0.5	7.4 $\pm$ 0.4	6.6 $\pm$ 0.4
^{234}Pa	22 $\pm$ 4		11 $\pm$ 2	10 $\pm$ 2	10 $\pm$ 2
^{233}Pa	19 $\pm$ 5		10 $\pm$ 4	10 $\pm$ 4	11 $\pm$ 3
^{232}Pa	6.5 $\pm$ 0.8		4 $\pm$ 0.5	2.9 $\pm$ 0.4	3.8 $\pm$ 0.4
^{229}Pa	4.9 $\pm$ 0.7		2.2 $\pm$ 0.5	2.1 $\pm$ 0.5	2.5 $\pm$ 0.5
^{228}Pa	1.0 $\pm$ 0.1		0.47 $\pm$ 0.07	0.55 $\pm$ 0.05	0.49 $\pm$ 0.05
^{227}Pa	0.39 $\pm$ 0.07		0.18 $\pm$ 0.04	0.14 $\pm$ 0.03	0.12 $\pm$ 0.03
^{234}Th	5 $\pm$ 3		18 $\pm$ 9	5 $\pm$ 2	
^{233}Th	7.5 $\pm$ 1		9 $\pm$ 2		7 $\pm$ 2
^{231a}Th	3 $\pm$ 1			6.4 $\pm$ 1.3	
^{228}Th	2.7 $\pm$ 0.2		2.4 $\pm$ 0.2		2 $\pm$ 0.2
^{227}Th	2.3 $\pm$ 0.2	2.2 $\pm$ 0.2	1.6 $\pm$ 0.2	1.6 $\pm$ 0.2	1.7 $\pm$ 0.2
^{226}Th	2.3 $\pm$ 0.2	2.4 $\pm$ 0.2	1.8 $\pm$ 0.2		1.7 $\pm$ 0.2
^{225}Th	1.5 $\pm$ 0.2		1.5 $\pm$ 0.2		
^{228}Ac	1.1 $\pm$ 0.2		0.8 $\pm$ 0.2		
^{226}Ac	1.5 $\pm$ 0.2		1.3 $\pm$ 0.2		
^{225}Ac	1.6 $\pm$ 0.2		1.3 $\pm$ 0.1		
^{224}Ac	2.1 $\pm$ 0.2		1.4 $\pm$ 0.1		

TABLE 4.8 (cont)

URANIUM (^{238}U)

0.68 - 6.2 GeV (cont)

Monitor : $^{27}\text{Al}(\text{p}, 3\text{pn})^{24}\text{Na}$
 $\sigma(\text{mb}) = 11.0$ at 0.68 GeV
 9.5 " 1.8 "

Note : Values used by author = 11.0 at 0.68 GeV
 10.5 " 1.8 "
Error : Mean deviations of duplicate determinations.

- Spallation of Uranium and Thorium nuclei with BeV energy protons.
- B.D. Pate and A.M. Poskanzer.
- Phys.Rev. 123, 647 (1961).

URANIUM

TABLE 4.9

50-340 MeV

	E_p	50	75	100	125	150	175	200	250	300	340
<u>Nuclide</u>		<u>$\sigma(\text{mb})$</u>									
^{140}Ba	-	29	27	23	20	-	22	18	-	17	
^{136}Cs	-	-	12	11	12	-	11	10	10	8.1	
$^{115\text{m}}\text{Cd}$	4.5	4.2	8.3	7.5	9.1	-	11	12	11	13	
^{115}Cd	73	28	30	25	23	-	24	22	21	19	
^{115}Ag	61	28	29	21	21	-	18	19	16	19	
^{113}Ag	73	35	32	-	30	-	24	-	31	29	
^{112}Ag	0.41	-	0.50	-	-	-	4.6	-	19	7.5	
^{112}Pd	128	38	32	25	28	27	27	31	-	25	
^{109}Pd	163	47	49	44	49	48	49	54	-	55	
^{97}Zr	-	55	-	-	45	-	-	42	-	38	
^{96}Nb	-	1.1	2.0	2.6	2.9	3.8	4.0	5.2	6.3	7.2	
^{89}Sr	-	32	32	29	25	25	19	30	29	30	
^{83}Br	-	16	16	15	-	13	14	14	15	14	
^{82}Br	-	0.32	0.75	0.85	1.0	1.2	1.5	1.6	2.3	2.3	
^{80}Br	-	0.21	0.40	0.46	-	0.60	0.85	-	1.4	1.8	
^{67}Cu	-	0.18	0.22	-	0.40	0.50	0.60	0.76	-	1.4	

Monitor : $^{27}\text{Al}(p,3\text{pn})^{24}\text{Na}$

$\sigma(\text{mb}) = 6.2$ at 50 MeV

9.7 " 75 "
 10.7 " 100 "
 10.2 " 125 "
 9.2 " 150 "
 8.9 " 175 "
 9.2 " 200 "
 9.9 " 250 "
 10.1 " 300 "
 10.3 " 340 "

Error : $\pm 5\%$

- Radiochemical studies of the high-energy fission process.
- A.G. Hicks and R.S. Gilbert.
- Phys.Rev. 100, 1284 (1955).

Further away from the target mass number the agreement between the experimental results and those of the model is less good. This is immediately evident from the comparison of figures 4.3 and 4.6a and 4.6c. The measured mass yield curve exhibits a degree of asymmetric fission unlike the computed yield curve. In the experiments reported here in addition to fission brought about by the protons some will be induced by the neutrons produced via the spallation and other reactions. In the earlier work using thin targets the initial proton interactions were presumably the only source of fission.

The extent of fission calculated in the Monte-Carlo model produced by proton interactions and by nuclear de-excitation by fission in the evaporation step (where there is competition between evaporation and fission) may be underestimated. [Both the evaporation of nucleons and fission are dependent on the energy deposited by the initial proton interaction, as outlined in section 1.4].

The computer codes represent the proton and neutron contributions separately and thus it is possible to compare the calculated cross-sections for fission products with those measured in the 50's and 60's for thin films. The targets used here were essentially "thin" to protons as the following discussion shows. [the term "thin" is used to imply that the flux and energy of the proton beam changes very little during passage through the target].

Protons lose energy by two types of interaction: Rutherford scattering and collision with extranuclear electrons. Rutherford scattering is an elastic nuclear collision giving rise to large changes in direction of the impinging proton but the average energy loss is not significant. Most of the energy loss is due to the orbital electron interactions following the Bethe-Bloch form:

$$-dE/dx = [4\pi^2 e^4 n / MV^2] [\ln MV^2 / I - \ln(1 - \beta^2) - \beta]$$

The range of protons in uranium can be calculated more easily from the following equation:

$$R_z / R_{air} = 0.90 + 0.0275Z + (0.06 - 0.0086Z) \log E/M$$

Where R_z and R_{air} are the ranges in air and the medium of atomic number z , E is the energy in MeV and M is the atomic mass^(4.12). This has a useful energy range from 0.1 to 1000 MeV for an elementary absorber.

For 800 MeV protons this gives a range of $346.4 \times 10^3 \text{ mg cm}^{-2}$. The calculated path length for protons is 18.28 cms which is a factor of 45 times greater than the irradiated target thickness. Like other charged particles the major energy loss occurs in the latter part of their interactions^(4.13) i.e. the energy of the protons is not significantly decreased in the target which is essentially "thin". The targets used

in the early studies were in the region of 20-50 mg cm^{-2} that is 1×10^{-3} cm to 2.7×10^{-3} cm thick (4.14-4.17). Compared with these the target used in this research was 3.75×10^3 mg cm^{-2} , (0.623 cm) which is a factor of 75-200 times greater.

An increase in fission from direct proton interaction is seen as unlikely as a corresponding increase in the spallation area would also be expected. The small amount of proton interaction is supported by the cross-sections of the spallation products which are small: in the order of millibarns, this is also in agreement with the Monte-Carlo calculations and the results from the afore mentioned research of the 1960's. The production cross-sections for spallation reactions in tables 4.7 to 4.9 show a great similarity in their magnitude to those identified in this work as would be expected from purely proton interactions. [Variations in cross-section of spallation products shown in these tables are due to proton energy].

Table 4.10 gives the cross-sections of some fission products measured at 5.7 GeV. These are the only fission products reported at sufficiently high energy for comparison. 5.7 GeV is a factor of seven times greater than that in this work; also the target was 0.005 inch thick. Comparison of these results with those measured in this work and with those of the MonteCarlo calculation is difficult. On the one hand the two experimental investigations report few

nuclides in common and on the other because of the substantial energy difference. Those that can be compared with 800 MeV and 5.7 GeV are all greater than those predicted by the Monte-Carlo calculation.

The nuclides ^{140}Ba , $^{110\text{m}}\text{Ag}$ and ^{103}Ru are reported and have also been reported in this work. The cross-section for ^{140}Ba at 5.7 GeV is 32 mb compared with 54.9 mb at 800 MeV. For $^{110\text{m}}\text{Ag}$ σ at 5.7 GeV is 3 mb, with a measured σ of 6.43 mb at 800 MeV and in the case of ^{103}Ru at 5.7 GeV σ is 28 mb and at 800 MeV 114.82 mb.

The mass distribution of fission products from proton induced reactions is symmetric. Early work established that as the energy of the incident particle increased the degree of symmetrical fission also increased with the peak to valley ratio in the fission yield curve decreasing. The enhanced yields of ^{95}Zr , ^{103}Ru measured in these experiments and their neighbours must in consequence be due to neutron induced reactions.

Many neutrons are being emitted as cascade/evaporation nucleons during irradiation. The energy of these nucleons may vary from 800 MeV plus binding energy (where all of the incoming proton energy is transferred) down to the binding energy of the least tightly bound nucleon in the nucleus (4.18).

TABLE 4.10

URANIUM

5.7 GeV

<u>Nuclido</u>	<u>σ(mb)</u>
^{210}Bi	3.0
^{212}Pb	0.47
^{209}Pb	3.9
^{204}mpb	15
^{183}Ta	8.6
^{178}Ta	5.9
$^{132}\text{La} + ^{133}\text{La} + ^{141}\text{La}$	29
^{140}La	0.9
^{140}Ba	32
^{128}Ba	20
^{114}mIn	4.8
^{111}In	5.9
^{117}mCd	13
^{115}Cd	28
^{115}mCd	9.5
^{115}Ag	7.3
^{113}Ag	15
^{112}Ag	9.5
^{111}Ag	21.5
^{110}mAg	3.0
^{112}Pd	1.15
^{111}Pd	7.2
^{111}mpd	2.7
^{109}Pd	23
^{106}Ru	25
^{105}Ru	28
^{103}Ru	28

TABLE 4.10 (Cont.)

URANIUM

5.7 GeV (cont.)

<u>Nuclide</u>	<u>σ(mb)</u>
^{99}Mo	42
^{73}Ga	6.1
^{72}Ga	5.4
^{68}Ga	7.1
^{72}Zn	0.68
$^{69\text{m}}\text{Zn}$	2.0
^{67}Cu	4.0
^{64}Cu	5.4
^{61}Cu	0.53
^{66}Ni	1.25
^{65}Ni	2.8
^{61}Co	6.5
^{59}Fe	2.3
^{56}Mn	6.3
^{52}Mn	0.68

Monitor : $^{27}\text{Al} (p, 3pn) ^{24}\text{Na}$
 $\sigma(\text{mb}) = 10.5$

Note : Original value used = 8.3 mb

Error : $\pm 20\%$

- Fission of uranium with 5.7 BeV protons
- R.E. Shudde
- UCRL - 3449.

Thus a broad spectrum of particle energies may be observed. [It is assumed that cascade nucleons are emitted isotropically with the incoming beam of protons (4.19), ie in a forward direction]. The production of a great number of neutrons from spallation reactions may give rise to the fission of further nuclei, this in turn will add to the number of fast neutrons available (the number of neutrons released in fission increases with the increase of energy of excitation of the nucleus (4.20)). Thus adding to the degree of fission observed.

In the modelling of the Rutherford Appleton laboratory's target which is thick (28.4 cms). The Monte-Carlo calculations takes the neutrons from the code O5R which is the neutron spectrum and calculated their contribution to fission by weighting it with the fission cross-section for uranium. The distribution function obtained for mass is of two components: a symmetric and asymmetric mass split. Their combination is very closely a bivariate gaussian distribution. Figure 4.6d shows the distribution calculated for spontaneous fission, 15 MeV fission and the target spectral function. The peak to valley ratios of the fission yields for these systems: irradiated target Monte-Carlo model; 15 MeV neutrons and the SNS Monte-Carlo spectral function give the following ratios (from figure 4.6d) : 0; 10; 60.

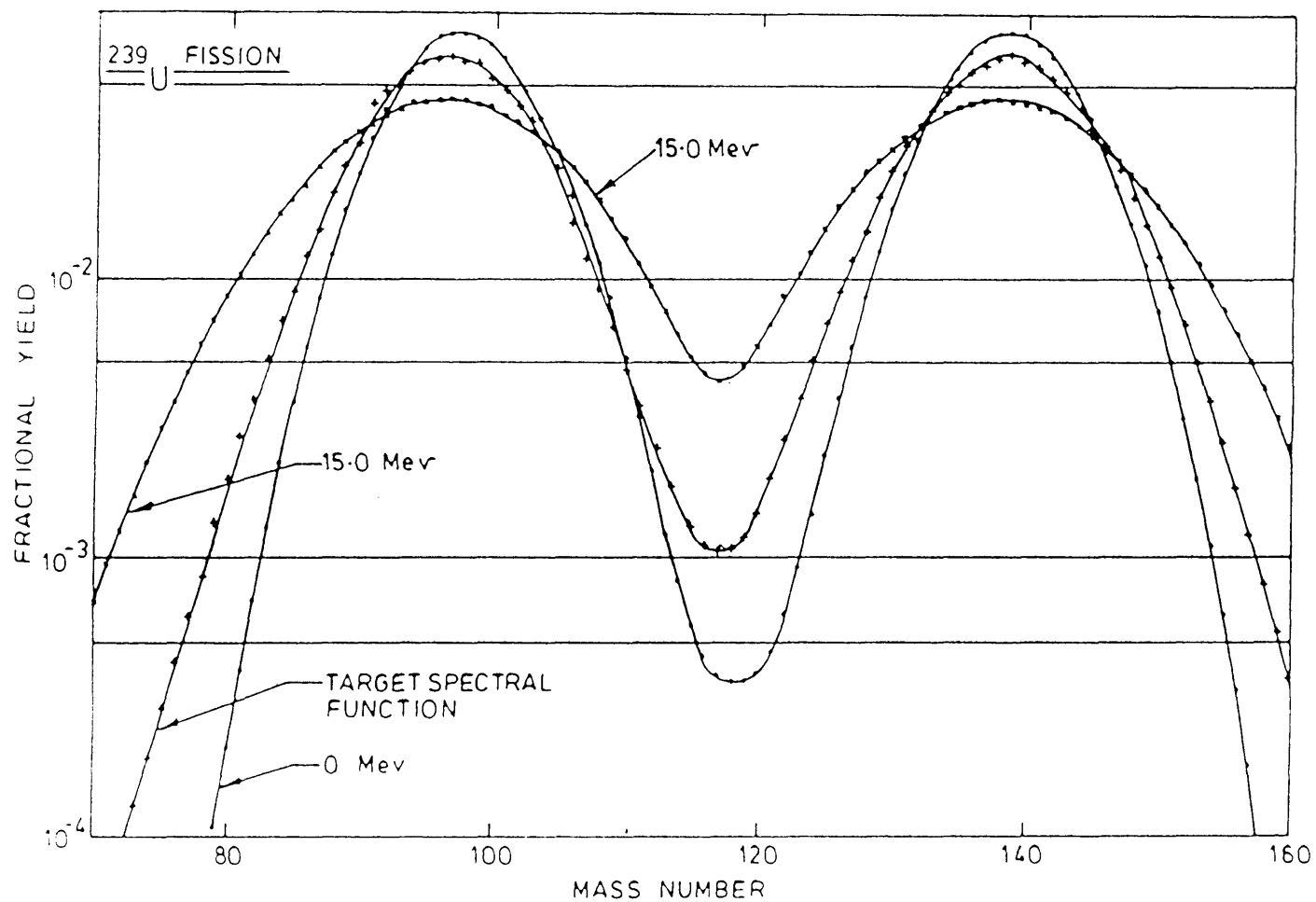


Fig. 4.6d

Mass yield in neutron induced fission of ^{238}U . Results are for spontaneous, 15 Mev and folding of spectral function

The model of the irradiated target treating the target as 'thin' has not accounted for the measured effect in the fission region whose increase in asymmetric fission can only be accounted for by an increase in neutron interactions.

A consideration is that the target is thicker than those previously analysed and the number of neutrons lost at the surface may be proportionally lower. The computer calculations treat the particle interactions quantitatively but a simple calculation can give some feel for the extent of neutron interaction. The calculation of the mean free path of neutrons from the mean logarithmic decrement ξ , gives an indication of the interaction of neutrons, which lose energy by elastic scattering with the target uranium nuclei. The fractional change in energy of the emitted neutron which is transmitted to a nucleus is independent of its initial energy and from the calculation of the mean logarithmic decrement (ξ) the number of collisions for the neutron to reach a certain energy may be calculated as exemplified below showing the mean free path for thermal neutrons in uranium:

$$\xi = 1 + \left\{ \frac{(A+1)^2}{2A} \right\} \ln \left\{ \frac{(A-1)}{(A+1)} \right\}$$

$$n = \ln(E_o/E_n) / \xi$$

$$= 2887 \text{ collisions in uranium to reach } 0.025\text{eV}$$

The calculated mean free path is given by:

$$\lambda_{tr} = 1 / N\sigma_s (1 - \overline{\cos \psi_a})$$

where $\overline{\cos \psi_a}$ the average value of the scattering angle in the 'laboratory system' ($\overline{\cos} = 2 / 238x3$ for ^{238}U)

For neutrons with an energy in the thermal region the mean free path is 2.52 cms. Although we are more interested in fast neutrons data for thermal neutrons is more readily available. The transport mean free path increases with energy and λ_{tr} calculated above therefore would be an underestimate for the target. This implies that most of the neutrons produced would escape from the target without interaction and would support the 'thin' target approximation used in the model.

There is thus good evidence that the neutron energy is not substantially degraded in the samples irradiated here and that the neutron fission product spectrum should approximate that of the 15 MeV neutrons shown in figure 4.6d, that is with a significant degree of asymmetry. The overall product spectrum then should be a suitable mix of those due to proton and neutron fission. The Monte-Carlo calculation indicates that the cross-section for neutron fission should be one third of that for the protons. Figure 4.6a shows that the resultant expected has the general characteristics of symmetric fission.

From figure 4.6d (calculated fission product yields for the SNS) the cross-section for the mass 103 is calculated to be 61.44mb for 15 MeV neutrons, 82mb for the targets spectral function and the measured ^{103}Ru gives 114.8mb. At mass 140 the calculated cross section for the SNS is 92.16mb for 15 MeV neutrons, 122mb for the spectral function compared with the measured cross-section for ^{140}Ba of 54.9mb. At mass 121, the trough area σ is 10.24, 4.1mb and 17.1 for the 15 MeV neutrons, spectral function and measured cross-section of ^{121}Te respectively.

Subsequent analysis of the second disc of the irradiated target by gamma spectrometry of the long lived fission products: ^{153}Gd ; ^{134}Cs ; $^{110\text{m}}\text{Ag}$; and $^{95\text{m}}\text{Tc}$ show a marginal increase in the degree of fission from the measurement of ratios of peak areas 2nd/1st disc: ^{153}Gd : 1.065, ^{134}Cs : 1.025, $^{110\text{m}}\text{Ag}$: 1.106 and $^{95\text{m}}\text{Tc}$: 1.044. In both the first and second discs the increase in asymmetric fission indicates that away from the area of spallation the Monte-Carlo model fails to describe the observations accurately.

It is conceivable that the enhancement of the degree of asymmetric fission could be due to a contribution of thermal neutrons produced by the copper beam dump. A calculated estimate of this neutron flux for a target station in a concrete vault with dimensions: 8x3x3 metres ($0.1 \times 10^9 \text{ cm}^2$) is given by:

$$Q/A = F^{\text{th}} \quad (4.21)$$

where: F_{th} = fast neutron yield
 A = surface area
 Q = number of neutrons produced/ proton

Two neutrons are produced per incident proton on copper (taken from Barbier: Induced Radioactivity). For a proton beam of 4.5×10^{11} protons per second this gives an evaporation yield of 9×10^{11} neutrons.

$$\begin{aligned} F_{th} &= 9 \times 10^{11} / 0.1 \times 10^9 \\ &= 9 \times 10^3 \text{ n cm}^{-2} \text{ s}^{-1} \end{aligned}$$

This is approximately $2 \times 10^{-6}\%$ of the particles onto the target. However the fission cross-section of ^{235}U is very high: $580 \times 10^{-24} \text{ cm}^2$. Below is the calculated contribution of these thermal neutrons to the production of ^{95}Zr , which has a measured cross-section of 94 mb. The number of fissions NF is given by:

$$NF = {}^{235}\text{N} \sigma_f \phi_p$$

The number of ^{95}Zr atoms N^* is given by:

$$\begin{aligned} N^* &= \%_{\text{yield}} NF \\ &= 3.5 \times 10^{21} \times 580 \times 10^{-24} \times 0.06 \times 9 \times 10^3 \\ &= 10.96 \times 10^3 \\ &= 570 \text{ atoms of } ^{95}\text{Zr g}^{-1} \end{aligned}$$

The measured number of atoms per gramme of ^{95}Zr in the target is 2×10^{12} . Consequently the contribution from the thermal flux of the target station would not make a detectable contribution to the measured cross-sections of the fission products. [$2.8 \times 10^{-8}\%$ of that measured].

To summarise: the Monte-Carlo calculation performed for the target shows good agreement with measurements made in this study and in earlier work in the spallation region. The general trend seen for the masses between that of the target and fission product area ($A = 160 < 238$) is for the production cross-sections to increase which agrees with previously reported work. The numerical values are also close. However in the fission region the model fails to agree with the measurements so well with particular nuclides. An underestimate of the proportion of evaporation stage de-excitation by fission and / or an underestimate of the neutron interactions which induced fission would account for the discrepancy.

REFERENCES

- 1.1 J. M. Carpenter
"Draft Proposal for an Intense Pulsed Neutron Source"
ANL Oct.1975 (RUTHERFORD LABORATORY REPORT
RL-77-014/B)
- 1.2 J.S. Fraser, G.A. Bartholemew, P.R. Tunncliffe
AECL - 2600, July 1966 (RUTHERFORD LABORATORY REPORT
RL-77-014/B)
- 1.3 J.M. Miller, J. Hudis
Ann. Rev. Nuc. Sci., Vol.9, 1959
- 1.4 A. Michaudon (Editor)
Nuclear Fission and Neutron Induced Fission cross-
sections Pergamon press
- 1.5 G. Friedlander, J.W. Kennedy, J.M. Miller
Nuclear & Radiochemistry 3rd Ed.
- 1.6 M. Barbier
Induced Radioactivity North Holland Press 1969
- 1.7 B.G. Harvey
Prog. Nuc. Phys. Vol.7
- 1.8 Ibid reference 1.7
- 1.9 E. Segre
Nuclei and Particles 2nd Ed. Benjemins/Cummings Press
- 1.10 Ibid reference 1.3
- 1.11 Perfilov et al.
Soviet Phys Usp (English Transl.) Vol.3, 1.
- 1.12 K. Kikuchi, M. Kawai
Nuclear Matter and Nuclear Reactions

- 1.13 R. Serber
R. Phys. Rev. vol 72, 1947
- 1.14 S. Fernbach, R. Serber, T.B. Taylor
Phys Rev. Vol.75 1946
- 2.1 C.W. Sills , R.L. Williams
Anal. Chem. vol 41, 12, Oct 1969
- 2.2 C.W. Sills
Health Phys. vol 17, 1969
- 2.3 C.W. Sills, K.W. Puphal, F.D. Hinderman
Anal. Chem. vol 46, 12, Oct 1974
- 2.4 C.W. Sills , R.L. williams
Anal. Chem. vol 53, 3, March 1981
- 2.5 J.L. Ryan
J. Phys. Chem. vol 64, 1960
- 2.6 Anil. K. de, S.M. Khopkar, R.A. Charmers
Solvent Extraction of Metals (Van Nostran Reinhold
1970)
- 2.7 F.L. Moore, J.E. Hudgens Jr.
Anal. Chem. vol 29, 12, Dec 1957
- 2.8 Ibid 2.1-2.4
- 2.9 Ibid 2.7
- 2.10 F.L. Moore Jr.
Anal. Chem. vol 29, 6, June 1957
- 2.11 N.A. Talvite
Anal. Chem. vol 44, 1972
- 2.12 O. Eracher, K. Philip
Z. Phys. vol51, 309, 1918
- 2.13 K.W. Bagnall
Quaterly Revue Chem. Soc. vol 11, 1, 1957

- 2.14 K.W. Bagnall, R.W. D'Eye
J. Chem. Soc. 1954
- 2.15 Ibid 2.12
- 4.1 P.A. Baedeker
Anal.Chem.Vol. 43, 3, 1971
- 4.2 K. Debertin, U. Schotzig
Nuc. Inst. & Methods Vol 140, 1977
- 4.3 Ibid reference 4.2
- 4.4 A. Ahmad
Imperial College Ph.D. Thesis 1982
- 4.5 A. Ahmad, P. Gray
Imperial College Reactor Centre (Unpublished).
- 4.6 C.W. Lehderer et al.
Table of Isotopes 7th Edition
- 4.7 G. Ertmann, W. Soyka
The Gamma Rays of the Radionuclides Verlag Chemie
- 4.8 Ibid reference 4.6
- 4.9 Ibid reference 4.6
- 4.10 B.G. Harvey
Proc. Nuc. Phys. Vol 7
- 4.11 A. Ashmore et al
Nuclear Matter and Nuclear Reactions K. Kikuchi,
M. Kawai North Holland press
- 4.12 Ibid 1.5
- 4.13 E. Segre
Nuclei and Particles 2nd Ed. Benjamin/Cummings press
- 4.14 H.G. Hicks, R.S. Gilbert
Phys. Rev. 100, 5, Dec. 1955
- 4.15 M. Lindner, R.N. Osbourne
Phys. Rev., 94,5 June 1954

- 4.16 T.D. Thomas, B.G. Harvey, G.T. Seaborg
Proc. 2nd Int. Conf. Peaceful uses of Atomic Energy
Geneva 1958 Vol.15
- 4.17 B.D. Pate, A.M. Poskanzer
Phys. Rev. 123, 2 July 1961
- 4.18 B.G. Harvey
Prog. Nuc. Phys, Vol.7
- 4.19 Ibid reference 4.15
- 4.20 Ibid reference 4.15
- 4.21 M. Barbier
Induced Radioactivity North Holland Press 1969

BIBLIOGRAPHY

Nuclear Radiation Detection

W.J. Price. McGraw-Hill Press 1958

Radiation Detection and Measurement

G.F. Knoll. J. Wiley & Sons 1979

Radiation Detection

W.H. Tait Butterworths 1980

Radioisotope Laboratory Techniques

R.F. Faores & G.G.S. Boswell Butterworth 1981

APPENDIX A

Calculation of relativistic mass and velocity of 800 MeV protons.

$$(1) \quad KE = (M - M_0)C^2$$

$$(2) \quad M = M_0 / [1 - (v^2/c^2)]^{1/2}$$

$$800 \text{ MeV} = (M - 1.007277) \times 1.660438 \times 10^{-24} \\ \times (2.997925 \times 10^{10})^2 \times 0.6242 \times 10^6$$

$$(M - 1.007277) = \frac{800}{1.66438 \times 10^{-24} (2.997925 \times 10^{10})^2 \times 0.6242 \times 10^6} \\ = 0.858285$$

$$M = 1.007277 + 0.85828$$

$$= 1.865562 \text{ a.m.u.}$$

$$1.865562 = 1.007277 / [1 - (v^2/c^2)]^{1/2}$$

$$[1 - (v^2/c^2)] = 1.007277 / 1.865562$$

$$v/c = 0.842 \text{ (84.2\% speed of light)}$$

$$v = 2.52338 \text{ ms}^{-1}$$

APPENDIX B

<u>GAMMA ENERGY keV</u>	<u>PEAK AREA + ERROR</u>
1-5-83	
724.83	30508.25 ± 174.66
757.34	33524.1 ± 183.1
766.	72918 ± 270
796.3	616.4 ± 24.8
803.5	724.75 ± 27
816.5	2555.5 ± 50.5
882	3990.9 ± 63
898.2	2873.6 ± 53.6
911.2	539 ± 23
658.2	2866 ± 53.5
667	1068.25 ± 32.7
695	2272.6 ± 47.7
365	1178.8 ± 74
415	1237.1 ± 35
427.4	607.7 ± 24.6
486.8	3305.7 ± 57.5
497	60684 ± 246
520.6	701.5 ± 26.5
537	1420 ± 37.7
572.8	2178 ± 47
610	2969 ± 54.5
621	683.4 ± 26
657.6	1187.1 ± 34.45
666	656.6 ± 25.6
695	940.5 ± 30.7

APPENDIX B(cont.)

	<u>GAMMA ENERGY keV</u>	<u>PEAK AREA + ERROR</u>	
19-5-83			
	390	2191	± 47
	415	1027.75	± 32
	427.3	1478.5	± 38.4
	486.8	209.46	± 45.7
	497	81265.6	± 285
	520	1583.83	± 40
	537	1055.75	± 33
	572.8	3171.9	± 56.3
	602	8174	± 90
	610	4160.3	± 64.5
	621.7	1159	± 34
	657.6	2432	± 49
	666	314.9	± 18
	695.3	941.5	± 31
	724.8	26020	± 161.3
	757	28992.3	± 170
	766	70036.9	± 265
	796.3	873.3	± 29.5
	803.5	357	± 19
	816.5	1269.5	± 35.6
	882	2836.9	± 53
	898	2597.7	± 51
	911.1	704.6	± 26.5
24-5-83			
	392	3382	± 58
	415	1201.5	± 34
	427	2247	± 47

APPENDIX B(cont.)

<u>GAMMA ENERGY keV</u>	<u>PEAK AREA</u>	<u>+ ERROR</u>
487	3168.4	± 56
497	128018	± 357.8
520	2579	± 51
537	1684	± 41
573	5205	± 72
602.5	12667.5	± 112.5
610	6386	± 80
622	2002.6	± 45
657	3460	± 69
695	1521	± 39

25-5-83

724	35331	± 188
757	39528.6	± 199
766	96473.2	± 310
796	774.15	± 28
816	140.26	± 37.4
882	3499.3	± 59
898	3714.1	± 61
911	843.8	± 29
938	1039	± 32
969	763.1	± 28
1100	722.7	± 27

13-6-83

392	4626	± 68
427	3129.8	± 56
459	878	± 29

APPENDIX B(cont.)

<u>GAMMA ENERGY keV</u>	<u>PEAK AREA + ERROR</u>		
427	2247	±	47
427	2247	±	47
	462	1214.2	± 35
486	1265	±	35
497	125990	±	355
520	2892	±	54
529	2106.5	±	46
573	6297	±	79
583	2227.6	±	47
602	14901	±	122
610	6704	±	82
622	2735	±	53
629	953.2	±	31
636	709	±	26
646	1090.6	±	33
657	5200	±	72
695	1397	±	37

14-6-83

97.6	7265.7	±	85
132	948.68	±	97
145	67600	±	260
157	3672	±	191
164	23386	±	153
184	2022	±	45
188	5969	±	77
210	14467	±	120
236	6631	±	81

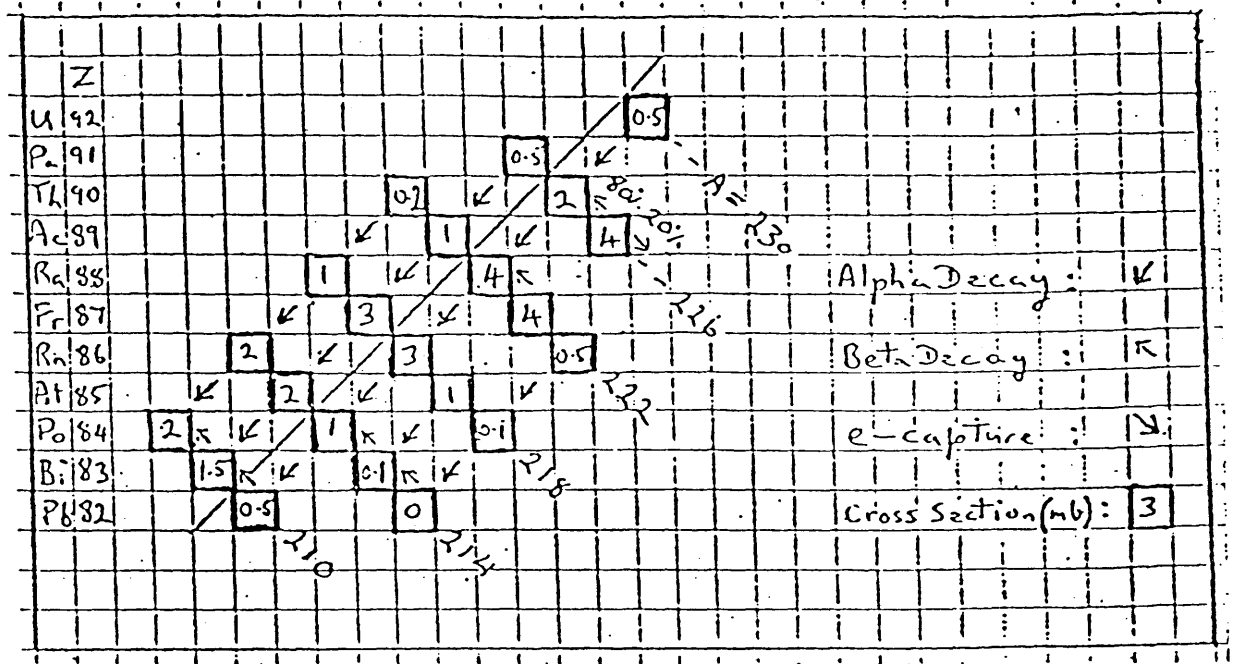
APPENDIX B(cont.)

<u>GAMMA ENERGY keV</u>		<u>PEAK AREA + ERROR</u>	
311		6282	± 79
351		2656	± 51
393		2952	± 54
14-7-83			
392		1177	±
497		23577	±
508		3505	±
520		749	±
572		1580	±
582		966	±
602		3476	±
610		1841	±
621		653	±
657		1410	±
662		545	±
15-7-83			
658		17634	± 133
697		3324	± 57
724		83515	± 57
756		93511	± 305
794		3752	± 61
881.8		11818	± 109
895		10138	± 101
908		4685	± 68
934		3808	± 61

APPENDIX B(cont.)

	<u>GAMMA ENERGY keV</u>	<u>PEAK AREA + ERROR</u>	
	965	2524	± 50
	1094	1265	± 35
	1114	2128	± 46
	1231	829	± 28
18-7-83			
	98.3	2026	± 45
	103	3137	± 56
	136	4163	± 63
	148	24053	± 155
	162	21690	± 147
	168	14914	± 122
	214	9372	± 96
	239	5442	± 73
	312	2067	± 45
	352	1893	± 43
	392	1763	± 42
	428	1685	± 41
	497	37740	± 194
	573	2588	± 50
	582	1419	± 37
	602	5565	± 74
	609	2297	± 47
	621	1354	± 36
	657	2559	± 50
	662	1324	± 36
	724	14485	± 120
	756	15109	± 123
	765	48054	± 219

APPENDIX C



Decay paths leading to Po 210 from uranium spallation products. Cross sections, in millibarns, are estimated