CHARGE DISPERSION OF PRODUCTS IN THE SYMMETRIC MASS REGION

SARKAR
NUCLEAR CHARGE DISTRIBUTION IN THE REGION OF
SYMMETRIC FISSION OF $^{238}\text{U}$ BY PROTONS OF ENERGY 20-85 MeV

by

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ABSTRACT

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NUCLEAR CHARGE DISTRIBUTION IN THE REGION OF

SYMmetric Fission of $^{238}$U BY PROTONS OF ENERGY 20-85 MeV

Charge dispersion curves around the most probable charge $Z_p$ in the fission of $^{238}$U with protons of energy 20-85 MeV were constructed on the basis of radiochemical studies on the fission products in the symmetric mass region. Independent cross-section for the nuclides $^{111}$Pd, $^{111}$Ag, $^{112}$Ag, $^{115}$Cd, $^{115}$In, $^{117}$In and cumulative yields for the nuclides $^{112}$Pd, $^{112}$Ag, $^{113}$Ag and $^{115}$Ag were measured.

Absolute isotopic yield data were obtained by monitoring the incident proton beam studying the reaction cross-section of $^{65}$Cu(p,pn)$^{64}$Cu using the external beam facility of the McGill synchrocyclotron coupled to a Faraday cup charge collection assembly.

The experimentally determined $Z_p$ values were deduced directly from the charge dispersion curves. The $(Z_A - Z_p)$ variation with incident proton energy is intermediate between the values obtained from light mass fission fragments of natural uranium and those obtained from the heavy mass fission fragments. The estimated total number of neutrons emitted in the symmetric mode of fission is higher than...
in the process of asymmetric fission attributable to fission events with higher than average excitation energy.
DISTRIBUTION DE CHARGE DES PRODUITS DE FISSION
SYMETRIQUE DE $^{238}$U PAR PROTONS DE 20 À 85 MeV

Les courbes de distribution de charge des produits résultant de la fission symétrique de $^{238}$U par protons de 20 à 85 MeV ont été construites à partir des mesures radiochimiques des sections efficaces de formation indépendantes de $^{111}$Pd, $^{111}$Ag, $^{112}$Ag, $^{115}$Cd, $^{115}$In, $^{117}$In et des sections efficaces cumulatives de $^{112}$Pd, $^{112}$Ag, $^{113}$Ag et $^{115}$Ag.

Les valeurs absolues des sections efficaces ont été obtenues à l'aide de la réaction de monitorage $^{65}$Cu(p,pn)$^{64}$Cu dont l'étude a été effectuée en faisceau externe auprès du synchrocyclotron de McGill, au moyen d'une cage de Faraday.

La charge la plus probable, $Z_p$, a été directement déduite des courbes de distribution de charge à chaque énergie incidente. Quand celle-ci croît, $Z_p$ se rapproche de la ligne de stabilité $Z_A$, les valeurs de $(Z_p-Z_A)$ se situant entre celles observées dans le cas des produits lourds et des produits légers de fission de l'uranium naturel. Le nombre total de neutrons émis accompagnant la fission symétrique est supérieur à celui déduit dans le cas de fissions asymétriques, ceci
pouvant être attribué à une plus grande énergie d'excitation des fragments.
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# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>ABSTRACT</td>
<td>i</td>
</tr>
<tr>
<td>ACKNOWLEDGEMENTS</td>
<td>v</td>
</tr>
<tr>
<td>TABLE OF CONTENTS</td>
<td>vii</td>
</tr>
<tr>
<td>LIST OF FIGURES</td>
<td>xi</td>
</tr>
<tr>
<td>LIST OF TABLES</td>
<td>xvi</td>
</tr>
<tr>
<td>I. INTRODUCTION</td>
<td>1-87</td>
</tr>
<tr>
<td>A. THE PHENOMENON OF FISSION</td>
<td>1</td>
</tr>
<tr>
<td>B. NUCLEAR REACTIONS</td>
<td>5</td>
</tr>
<tr>
<td>(1) Introduction</td>
<td>5</td>
</tr>
<tr>
<td>(2) Mechanisms</td>
<td>6</td>
</tr>
<tr>
<td>C. FISSION MECHANISM</td>
<td>14</td>
</tr>
<tr>
<td>(1) Introduction</td>
<td>14</td>
</tr>
<tr>
<td>(2) Nuclear Models</td>
<td>14</td>
</tr>
<tr>
<td>D. FISSION YIELDS AND THEIR CORRELATIONS</td>
<td>34</td>
</tr>
<tr>
<td>(1) Introduction</td>
<td>34</td>
</tr>
<tr>
<td>(2) Distribution of Yields of Fission</td>
<td>35</td>
</tr>
<tr>
<td>Fragments as a Function of Mass Number</td>
<td>35</td>
</tr>
<tr>
<td>(3) Distribution of Nuclear Charge in Fission</td>
<td>51</td>
</tr>
<tr>
<td>E. PREVIOUS WORK</td>
<td>61</td>
</tr>
<tr>
<td>(1) Charge Distribution in Low-Energy Fission</td>
<td>61</td>
</tr>
<tr>
<td>(2) Charge Distribution in Medium-Energy Fission</td>
<td>66</td>
</tr>
<tr>
<td>F. CHEMICAL PROCEDURES - GENERAL CONSIDERATIONS</td>
<td>80</td>
</tr>
<tr>
<td>G. PRESENT WORK</td>
<td>86</td>
</tr>
</tbody>
</table>
II. EXPERIMENTAL PROCEDURES

A. TARGET ARRANGEMENT

B. IRRADIATION

C. PROTON BEAM MONITORING

   (1) Introduction

   (2) Experimental

      (i) Targets and Irradiation Procedure

      (ii) Measurement of Proton Energy

      (iii) Energy Spread in Proton Irradiations

      (iv) Determination of the Integrated Bombardment Current

      (v) Results and Discussion

D. CHEMICAL PROCEDURES - TECHNIQUES

   (1) Preparation and Standardization of Carrier Solutions

   (2) Filtering, Drying and Weighing

   (3) Mounting of Samples

E. CHEMICAL PROCEDURES - SPECIAL METHODS FOR THE SEPARATION OF FISSION PRODUCTS IN THIS WORK

   (1) Isolation of Indium Isotopes from a Mixed Fission Product Solution

   (2) Isolation of Silver Isotopes from a Mixed Fission Product Solution

   (3) Isolation of Palladium Isotopes from a Mixed Fission Product Solution
F. RADIOCHEMISTRY OF COPPER................................. 121
G. MEASUREMENT OF ACTIVITY.......................... 123

III. TREATMENT OF DATA......................................................... 139-185
A. ANALYSIS OF SPECTRA.................................................. 139
B. GENERAL EQUATION FOR CROSS-SECTION CALCULATION........... 143
C. RESOLUTION OF RADIOACTIVE DECAY DATA...................... 145
D. SPECIAL EQUATIONS USED IN COMPUTATION OF FISSION YIELDS.... 147
E. DECAY SCHEMES AND FISSION PRODUCT DECAY CHAIN IN THE MASS REGION, \( A=111-A=117 \)...... 154
F. RELEVANT DETAILS OF DECAY SCHEMES, AND, COMPUTATION OF ACTIVITIES AT END OF IRRADIATION OF NUCLIDES STUDIED IN THE PRESENT WORK................................. 155
(1) Chain \( A=117 \)........................................... 155
(2) Chain \( A=116 \)........................................... 159
(3) Chain \( A=115 \)........................................... 161
(4) Chain \( A=113 \)........................................... 168
(5) Chain \( A=112 \)........................................... 173
(6) Chain \( A=111 \)........................................... 176
G. BETA DECAY CURVES............................................... 182
H. ERROR ANALYSIS.................................................. 184

IV. RESULTS.......................................................... 186-189

V. GENERAL CONCLUSIONS AND DISCUSSION......................... 200-221
(1) Charge Dispersion and Most Probable Charge 200
(2) Excitation Functions of Nuclides Formed in the Symmetric Mass Region .................. 213
(3) Variation of N/Z_p with Mass .................. 214
(4) Most Probable Charge from UCD and ECD Mechanism ............................... 214
(5) Total Numbers of Neutrons Emitted .......... 217

VI. SUMMARY AND CONTRIBUTION TO KNOWLEDGE .................. 220

VII. REFERENCES ................................. 222
**LIST OF FIGURES**

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>Figure 1</td>
<td>Scission configuration in the fission process and neutron emission.</td>
<td>17</td>
</tr>
<tr>
<td>Figure 2</td>
<td>Schematic diagram of the fission process.</td>
<td>18</td>
</tr>
<tr>
<td>Figure 3</td>
<td>Liquid-drop model.</td>
<td>19</td>
</tr>
<tr>
<td>Figure 4</td>
<td>Energy of a fissioning nucleus as a function of the distance of the fragments.</td>
<td>21</td>
</tr>
<tr>
<td>Figure 5</td>
<td>Diagram on statistical model of the nucleus.</td>
<td>32</td>
</tr>
<tr>
<td>Figure 6</td>
<td>The measured mass yields from fission of $^{235}\text{U}$ and $^{238}\text{U}$ induced by neutrons of various energies.</td>
<td>42</td>
</tr>
<tr>
<td>Figure 7</td>
<td>Schematic summary of fission properties.</td>
<td>45</td>
</tr>
<tr>
<td>Figure 8</td>
<td>Fission product mass distribution, $^{238}\text{U}(P,f)$ (composite curves at 75 and 100 MeV).</td>
<td>47</td>
</tr>
<tr>
<td>Figure 9</td>
<td>Fission product mass distribution, $^{238}\text{U}(P,f)$ (composite curves at 40 and 55 MeV).</td>
<td>48</td>
</tr>
<tr>
<td>Figure 10</td>
<td>Fission product mass distribution, $^{238}\text{U}(P,f)$ (composite curves at 32 and 35 MeV).</td>
<td>49</td>
</tr>
<tr>
<td>Figure 11</td>
<td>Fission product mass distribution, $^{238}\text{U}(P,f)$ (composite curve at 20 MeV).</td>
<td>50</td>
</tr>
<tr>
<td>Figure 12</td>
<td>The average number of post-fission neutrons as a function of fragment mass.</td>
<td>76</td>
</tr>
<tr>
<td>Figure 13</td>
<td>The average kinetic energy of the post-fission neutrons in the c.m. systems of the fragment as a function of the fragment mass.</td>
<td>77</td>
</tr>
</tbody>
</table>
Figure 14  The average number of post-fission neutrons as a function of the fragment mass for events of high total kinetic energy $E_k$ of the two fission fragments and for events with low $E_k$ .......................... 78

Figure 15  Target assembly-foils........................... 89

Figure 16  Target assembly and end of cyclotron probe........................................... 91

Figure 17  Irradiation energy as a function of the radial distance of the cyclotron probe........................................... 93

Figure 18  Proton beam position of McGill synchrocyclotron as a function of energy....... 94

Figure 19  Targets and irradiation assembly for external beam proton bombardments..... 98

Figure 20  The sectional view of the external beam irradiation assembly.......................... 99

Figure 21  Fraction of transmitted beam as a function of absorber thickness............. 100

Figure 22  Energy degradation of the external beam............................................. 102

Figure 23  Absolute cross-section for the reaction $^{65}$Cu$(p, pn)^{64}$Cu as a function of bombardment energy................................. 105

Figure 24  Experimental and theoretical excitation function for the reaction $^{65}$Cu$(p, pn)^{64}$Cu in the energy range 25-102 MeV............. 106

Figure 25  Radiochemical separation of indium - flowchart........................................ 114

Figure 26  Radiochemical separation of silver isotopes - flowchart............................. 117

Figure 27  Radiochemical separation of palladium isotopes - flowchart......................... 120

Figure 28  Radiochemical separation of copper - flowchart........................................ 122

Figure 29  Block diagram of the detector system used.............................................. 127
Figure 30 Absolute photopeak efficiency of 30 c.c. Ge(Li) detector

Figure 31 Absolute photopeak efficiency of 40 c.c. Ge(Li) detector

Figure 32 Fraction of the gamma-ray absorbed as a function of energy of the γ-ray

Figure 33 Calculated absolute efficiencies ($\varepsilon_{20}$) for beta-standards as a function of mean beta-energies

Figure 34 Chain A = 117

Figure 35 Chain A = 116

Figure 36 Chain A = 115

Figure 37 Chain A = 113

Figure 38 Chain A = 112

Figure 39 Chain A = 111

Figure 40 Schematic representation of the decay path for the nuclide $^{111}$Ag

Figure 41 Sample decay curves from experiments involving beta-decay of pure radiochemical samples

Figure 42 Independent isomeric yield ratios $^{117m}_{\text{In}}/^{117}_{\text{In}}$

Figure 43 $^{117}_{\text{In}}(m+g)$, independent formation cross-section

Figure 44 $^{115m}_{\text{Cd}}$, independent formation cross-section

Figure 45 $^{115}_{\text{Cd}}$, independent formation cross-section (estimated)

Figure 46 $^{115}_{\text{In}}$, independent formation cross-section

Figure 47 $^{111}_{\text{Ag}}$, independent formation cross-section

Figure 48 $^{112}_{\text{Ag}}$, independent formation cross-section
Figure 49  $^{111}$Pd, independent formation cross-section

Figure 50  $^{112}$Pd, cumulative formation cross-section

Figure 51  $^{113}$Ag, cumulative formation cross-section

Figure 52  $^{111}$Ag, cumulative formation cross-section

Figure 53  $^{115}$Ag, cumulative formation cross-section

Figure 54 Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 85 MeV

Figure 55 Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 70 MeV

Figure 56 Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 55 MeV

Figure 57 Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 40 MeV

Figure 58 Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 30 MeV

Figure 59 Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 20 MeV

Figure 60 Variation of $Z_a - Z_p$ function with bombarding energy

Figure 61 Variation of $(Z_a - Z_p)$ with mass number at a specific incident proton energy in the fission of natural uranium
Figure 62  Variation of $N/Z_p$ with mass .................. 215

Figure 63  Variation of $Z_p$ with the average excitation energy of intermediate nuclei for the \( ^{238}\text{U} + \text{p} \) system .................. 218
LIST OF TABLES

TABLE I  Cross sections for the reaction 65Cu(p, pn)64Cu................................. 104(b)

TABLE II Random and systematic errors in the absolute cross-section for the reaction 65Cu(p, pn)64Cu 106(b)

TABLE III Relative absolute efficiencies for gamma-lines of spread solid sources used in this work .................. 131(b)

TABLE IV Summary of the decay properties of observed radioisotopes .................................................. 138(b)

TABLE V  Fission product decay chains.......................... 154(b)

TABLE VI Independent isomeric yield ratio (117mIn/117In) ......................................................... 186(b)

TABLE VII Independent formation cross-sections (m barn) of products from 20-85 MeV proton fission of 238U.................. 188(b)

TABLE VIII Cumulative formation cross-sections of products from 20-85 MeV proton fission of 238U.................. 188(c)

TABLE IX Independent cross-section values used in charge dispersion curves....... 200(b)(c)

TABLE X  Most probable charge from UCD and ECD mechanism.................. 216(b)

TABLE XI Total number of neutrons in the 238U(p, f) reaction; study of products in the symmetric and near-symmetric mass region.................. 218(b)

TABLE XII Total number of neutrons emitted in the proton-induced fission of 238U........ 219(c)
I. INTRODUCTION
A. THE PHENOMENON OF FISSION

The splitting of a heavy nucleus into roughly equal parts was first defined as "fission" by Meitner and Frisch (Me 39). Ternary and quaternary fissions are very rare events compared to binary fission [The observed ratio of ternary to binary events in thermal-neutron fission of uranium is of the order of $10^{-6}$ as reported by Muga (Mu 69) and approximately .03 for similar modes of fission of uranium using 400-MeV argon ions, Brandt (Br 71)].

Shortly after the discovery of the interaction of uranium with neutrons by Fermi and co-workers (Fe 30), Noddack (No 34) considered the possibility of explaining their results by a 'fission' process. No supporting evidence was presented with her argument, however, and the fission process was really first almost reluctantly postulated five years later by Hahn and Strassman (Ha 39) on the basis of their studies of the chemical nature of the radioactive products occurring when uranium is bombarded with neutrons.

Since the discovery of nuclear fission, a vast amount of work has been done by nuclear physicists and chemists and a variety of fission processes using different projectiles at various bombarding energies and fissile targets have been studied.

Much of the early work was that of thermal-neutron
induced fission of uranium. It was shown very early by Bohr and Wheeler (Bo 39) that it is the rare isotope $^{235}\text{U}$ (0.71 per cent abundance), that undergoes fission when uranium is bombarded with thermal neutrons. This fact was later verified experimentally by Turner (Tu 40). In the fission of $^{235}\text{U}$, the fissionable nucleus captures a thermal neutron forming an excited compound nucleus $^{236}\text{U}$, which then undergoes fission. This process has been represented by the following equation:

$$^{235}\text{U} + \frac{1}{0}\text{n} \rightarrow [^{236}\text{U}] \rightarrow (Z_1A_1) + (Z_2A_2) + \nu\frac{1}{0}\text{n} + \gamma + Q$$

(I-1)

and is commonly referred to as $^{235}\text{U}$ fission, even though the nucleus that breaks apart actually is the compound nucleus $^{236}\text{U}$.

The mass distribution is asymmetric and a primary light fragment ($Z_1A_1$) and a primary heavy fragment ($Z_2A_2$) are formed in the fission process (fission fragments are primary or independently-formed nuclides, and fission products are all products formed in fission including both those from direct formation and those from decay of primary fission fragments). The total number of neutrons released is characterized by $\nu$ and these neutrons, in thermal-neutron fission are not emitted from the compound nucleus $^{236}\text{U}$ during the fission process but from the fission fragments themselves after the occurrence of the fission act,
Wilson (Wi-47). The major part of the neutrons are emitted within $10^{-14}$ sec., Fraser (Fr-52), after the splitting of the compound nucleus and are therefore included in the above equation. They are called 'prompt' neutrons in order to distinguish them from the delayed neutrons which themselves account for only about one percent of the total neutron emission. A certain amount of energy, $\gamma$, is released as electromagnetic radiation at the instant of fission, and $Q$ is the kinetic energy of the fission fragments and fission neutrons.

In the equation above, the laws of conservation of mass ($A$) and charge ($Z$) must be fulfilled, i.e., the sum of the mass numbers of complementary fission fragments plus the actual (integral) number of neutrons emitted must for a given pair be

$$A_1 + A_2 + \nu = 236.$$  

The sum of the nuclear charges of complementary fission fragments with average $A_1$ and $A_2$ must equal 92.

Due to the coulomb effect, the nuclides in the heavy mass region have a rather high neutron-to-proton ratio and the primary fission fragments must have, therefore, a similar ratio. The total coulomb energy of the two fragments, however, is only about one half of the coulomb energy of the parent nucleus.
Consequently, these nuclides have a much higher neutron-to-proton ratio than the ratio for stable nuclides in the fission product region. The primary fission fragments \((Z_1A_1)\) and \((Z_2A_2)\) hence are far removed from stability and must reduce the high neutron-to-proton ratio by some transformation until the ratio corresponding to stability is reached. Two processes are available:

(i) neutron emission, and, (ii) emission of negative \(\beta\) particles (negatrons).

(i) As the fission fragments usually have more excitation energy than that corresponding to the binding energy of the last neutron in the fragment, neutron emission will be highly probable. The neutron emission, however, does not reduce the neutron-to-proton ratio appreciably and the most important decay mechanism which follows neutron emission is therefore (ii) the emission of \(\beta^-\) particles.

(ii) In the \(\beta^-\) emission process a neutron in the nucleus is transformed into a proton in the nucleus with simultaneous ejection of a \(\beta^-\) particle and a neutrino. This transformation does not change the mass-number \(A\), but is associated with an increase in the atomic number.
B. NUCLEAR REACTIONS

(1) **Introduction**

When a layer of some target material is bombarded with a beam of particles, many different phenomena may be observed. If the particles are charged, the vast majority of them will lose energy by interactions with the atomic electrons of the target material until they stop or emerge from the rear face of the target layer with reduced energy. Other particles will pass close enough to a nucleus in the target layer to feel the effect of the Coulomb-repulsion. These particles will suffer a change of direction (Coulomb scattering) and a small loss of kinetic energy. Other particles will pass close enough to a target nucleus to feel the effect of the short-range nuclear forces. Like Coulomb scattering, such interactions cause a change in the direction of motion of the particle, and transfer a small amount of kinetic energy to the target nucleus. The nucleus may be left in an excited state or in its ground state. In the latter case, the total kinetic energy of the system of particles and nucleus remains the same before and after the event. Such particles are said to be **elastically-scattered**. When the target nucleus is left in an excited state, the energy is derived from the kinetic energy of the particle. In such events, the particle is said to be **inelastically-scattered**.
In some cases, the incident particle may undergo a nuclear reaction with the target nucleus. The particle (or a part of it if it is a complex particle such as a deuteron) may be captured by the nucleus, and one or more new particles may be emitted.

In nuclear reactions induced by low energy (below a few hundred MeV) bombarding particles, the number of protons and neutrons in the initial and final systems separately remain constant. Thus in the reaction $^{209}_{83}\text{Bi}(\alpha,2n)^{211}_{85}\text{At}$, there are 85 protons and 128 neutrons on each side of the equation. In reactions in which mesons are absorbed or emitted, the total charge and the total number of nucleons must remain constant, but the numbers of protons and neutrons separately may change. For example, in the reaction $^{63}_{29}\text{Cu}(p,p^+)^{63}_{28}\text{Ni}$, the total charge is 30 units before and after the reaction, but the numbers of protons decreases by unity and the number of neutrons increases by unity. The total number of nucleons remains the same (since the emitted $p^+$ meson is not a nucleon).

(2) **Mechanisms**

The different nuclear reactions which take place may involve several different kinds of mechanisms.

(i) **Compound Nucleus Mechanism.** This is valid especially when the incident particles are of low energy (less
than about 50 MeV) and involves the capture of the incident particle by the struck nucleus to form what is called a compound nucleus. The theory of reactions of this type was first discussed by Niels Bohr in 1936. Reactions of nuclei with very low energy neutrons, for example, are of this type. The compound nucleus formed by the capture of the incident particle will be in a state of high excitation energy. The net resulting excitation will be equal to the binding energy of the captured particle to the nucleus plus the kinetic energy of the captured particle in the center-of-mass system. For example, a 25-MeV (laboratory system) helium ion incident upon a nucleus of $^{65}_{29}$Cu will, if it is captured, form an excited nucleus of $^{69}_{31}$Ga. The center-of-mass kinetic energy of the helium ion will be $(25 \times 65)/69$ or 23.6 MeV. From the atomic-masses, $Q$ is found to be +4.48 MeV. The excitation energy of the $^{69}$Ga nucleus is therefore $23.6 + 4.5$ or 28.1 MeV. Its kinetic energy is $(25-23.6)$ MeV.

At excitation energies of many MeV, nuclei must be in a chaotic state. Many nucleons will be excited into energy levels well above their normal ground-state levels, and the nucleus may contain a considerable amount of rotational and vibrational energy. At such high excitation, the levels may be so closely-spaced in energy that their widths are greater than the energy separation between levels. It is then no longer possible to recognize discrete energy
levels, and the nucleus no longer exists in a single, well-defined quantum state. Its energy and its angular-momentum, of course, remain constant, but there may be an enormous number of different nuclear configurations of the same energy and angular momentum, and the nucleus will oscillate very rapidly between all of these different possible states. In these circumstances, we can treat the energy levels statistically. The properties of the excited compound nucleus may therefore be described by means of the level density and the nuclear-temperature concepts.

Decay of the compound nucleus:

Immediately after their formation, the highly excited compound nuclei will begin to decay. They are energetically capable, in most cases, of emitting one or more particles such as neutrons, protons, deuterons, and helium ions, or energy may be emitted in the form of γ-radiation. The emission of γ-rays, however, is a slow process by comparison with the emission of particles, so that whenever the nuclear excitation energy exceeds the binding energy of a particle, emission of the particle will nearly always take place in preference to the emission of the energy as a γ-ray. Since there is no Coulomb-barrier, emission of neutrons is easier than emission of charged particles, particularly from nuclei of high atomic number.
Hence reactions such as \((\alpha, 2n)\) usually have larger cross sections than \((\alpha, pn)\) or \((\alpha, 2p)\), but there are exceptions.

The average time delay required for the concentration of energy on a single nucleon is of the order of \(10^{-20}\) seconds, and although this is very short when compared with the time required for the emission of a gamma-ray, it is long enough to permit the energy brought in by the incident particle to be randomly distributed through the nucleus. In these circumstances, the direction of emission of the evaporated nucleons need bear no relation to the direction of the incident particle. When many over-lapping levels are formed in the compound nuclei, the particles emitted from a large collection of compound nuclei are found experimentally with equal intensity in all directions in the center of mass system; the emission is isotropic.

In the laboratory system, there will be more particles emitted in the forward direction (that is, the direction of motion of the incident particle), because the decaying compound nuclei are themselves moving in this direction.

If the compound nuclei exist in a well-defined quantum state, and if they decay by particle emission to new nuclei which are also in a well-defined state, the emission of the particles need not be isotropic. However, an argument based on the conservation of parity requires that the probability of emission shall be symmetric about a
plane at 90° to the direction of the incident particle.

The difficulty of concentrating a large amount of energy on a single nucleon suggests that the "evaporated" nucleons will be emitted with rather low kinetic energies. This is found experimentally to be the case.

In a few cases, it is possible to form the same compound-nucleus by two different reactions. If the assumptions of the compound nucleus theory are correct, the decay of the compound nuclei formed in different ways should be independent of the method by which they were formed. It was shown by Ghoshal (Gh 50), the compound nucleus $^{64}$Zn was produced by the bombardment of $^{63}$Cu with protons and $^{60}$Ni with helium ions. The reaction $^{60}$Ni(a,n)$^{63}$Zn was compared with $^{63}$Cu(p,n)$^{63}$Zn, and so on for other pairs of corresponding reactions. From the excitation functions, the correspondence between the pairs of reactions became obvious, showing that the mode of decay of the $^{64}$Zn compound nuclei is indeed independent of the way in which they are formed.

(ii) High Energy Nuclear Reactions

The assumption that the energy of the incident particle is randomly shared by all the nucleons of the struck nucleus is an approximation which becomes worse as the energy of the incident particle is increased. For energies above about 50 MeV, it is no longer able to explain
even the main features of nuclear reactions. Particles are no longer emitted isotropically; rather, they are concentrated into the forward direction. Reaction cross sections vary little with energy; the peaks characteristic of evaporation reactions are not found.

Reactions of this kind take place by collisions of the incident particle with individual nucleons within the target nucleus. The struck nucleons will quite frequently receive sufficient kinetic energy from the collision to permit them to escape from the nucleus instantaneously, usually in the forward direction. The two most common situations could be:

(a) The incident proton strikes a neutron, and both particles escape from the nucleus, so that the reaction is \((p, pn)\).

(b) Both the incident particle and the struck nucleon may make further collisions which can result in the emission of a shower of particles. Reactions in which several particles are emitted are often called "spallation", regardless of the mechanism by which they take place.

Calculations about nuclear reactions falling under the 'high-energy' category, are often made with the aid of the technique called the Monte Carlo method. The dynamics of the collision processes being so complex, the essence of the Monte Carlo method is the use of random numbers.
The path of the single incident proton through the nucleus is calculated, records being kept of all the collisions which it made, the paths of all the struck nucleons, and their kinetic energies and directions of emission. The calculation is repeated over and over again, using a fast electronic computer, until a reliable picture has been created of what happens when a large number of protons strike a target. The incident protons may strike nuclei at any point on their surfaces. To give all points of impact equal weight, the computing machine picks a number from a collection of random numbers, and this number determines the impact point. Another number from the collection determines the point of impact for the second case, and so on.

The results of the Monte Carlo calculations agree quite well with experimental results such as those reported, for instance, by Harvey (Ha 69) which compares the mass yield distribution for bombardment of copper with 340 and 570 MeV protons.

The scattering of the high energy particles occurs through a direct interaction mechanism. The incident particle interacts through the nucleon-nucleon forces (and the Coulomb force if it is charged) with the nucleons of the target nucleus. Its direction of motion is thereby changed. This direct reaction mechanism was first observed with deuterons. The deuteron is a very loosely bound structure.
As it approaches a nucleus, the nucleon forces may break off one of the nucleons of the deuteron and cause it to be captured by the target nucleus, while the other nucleon of the deuteron continues on its path almost unaffected by the loss of its partner. The net reaction is therefore (d,p) or (d,n), depending on which part of the deuteron was captured. This reaction mechanism is called stripping. Such reactions may take place with incident deuterons of high or low energy.

The product nucleus may be left in its ground state or in an excited state. In the latter case, the energy of the emitted particle must be reduced, and a study of the energy spectrum of the emitted protons or neutrons can be used to measure the energy of the levels of the product nucleus. Since the process occurs by the capture of single nucleon, the levels most likely to be formed are those in which only that single nucleon is excited above its normal shell-model state. More complex levels, in which several nucleons are in excited states, are much less likely to be formed.

The captured nucleon may bring integral amounts of angular momentum $l_c$ into the target nucleus. The probability that the emitted nucleon will be found at any angle $\theta$ relative to the direction of the incident deuteron depends both on $\theta$ and $l_c$. 
C. FISSION MECHANISM

(1) Introduction

In recent years the wealth of new discoveries concerning the many aspects of the fission process has led to new insights into the mechanism of this very complicated process. The purpose of this section of the thesis is to examine briefly many of these aspects of the fission process by particularly considering fission induced by excitations up to tens of MeV - the range of energy for experimental work described in this thesis. Greater bombarding energies involve uncertainties in the nature of the deposition energy that are apart from the fission process itself. The results of neutron-induced fission will be considered, mainly because of simplicities of barrier penetration and absorption into nuclear matter.

(2) Nuclear Models

An important problem in understanding the massive motion of nuclear matter that leads to nuclear fission into two fragments is the determination of the factors which cause asymmetry - the inequality of the two fragment masses. Several different approaches have been used in attempts to explain asymmetry, as well as other fission parameters. Among these are:
(a) LIQUID-DROP MODEL

The mass formula of V. Weizsäcker (We 35) has been applied by Bohr and Wheeler (Bo 39) to deformed nuclei, and, use of this model has been made to explain aspects of fission as described in fundamental articles by Hill and Wheeler (Hi 53), Nix and Swiatecki (Ni 65) and Businaro and Gallone (Bu 55).

Although it does not provide a complete theory of fission, the liquid-drop model throws some light on the nature of the fission process. According to this point of view, a nucleus tends to assume a spherical shape (just like a water drop), under the influence of a surface tension. This surface tension arises because nucleons in the nuclear surface have fewer close neighbours than nucleons in the center, and are therefore less strongly bound. The surface tension causes a loss of total binding energy which is proportional to the surface area. In any liquid drop, the surface energy is just the product of the surface-tension and the surface area, so that work must be done to cause an increase in surface area.

However, in the case of an electrically-charged drop, such as a nucleus, the Coulomb repulsion is greatest for the very compact spherical shape. Thus the Coulomb repulsion tries to make the drop assume a non-spherical shape, while the surface tension tries to keep it spherical.

In light nuclei, the Coulomb repulsion is small, and hence a nearly spherical shape is preferred. For the
very heavy nuclei, however, the repulsion is much stronger and very little excitation energy is needed to cause it to overcome the surface tension and for the nucleus to divide or fission. The surface area, and hence the surface energy, is greater after the nucleus has divided, but the Coulomb energy is very much lower, so that the fission is accompanied by a net drop in the potential energy.

These ideas can be made quantitative by assuming that a nucleus which is in the process of fissioning passes through a sequence of shapes such as those shown in Figure 1. It is possible to calculate the surface and Coulomb energies as a function of the nuclear shape. The shape of an axially symmetric nucleus may be described by the equation:

\[ R = R_0 \left[ 1 + \alpha_2 Y_2^0(\theta) + \alpha_4 Y_4^0(\theta) + \ldots \right] \]  

(I-2)

If coefficients \( \alpha_2 \) and \( \alpha_4 \) vary with time in an appropriate way, the nuclear shape given by the above equation will trace out the sequence of shapes as shown in the sequence of steps leading up to the scission configuration in Figures 1 and 2. In Figure 3 we consider a "volcano-crater" of potential energy, over the lip of which a fissioning nucleus must pass to undergo fission. Here the radial co-ordinate is deformation, while the azimuthal co-ordinate is the degree of fission asymmetry - the inequality in the size of the two resulting fission fragments. The height of the pass
FIGURE 1

Scission configuration in the fission process and neutron emission. After Leachman (Le 60).
FIGURE 2

Schematic diagram of the fission process.

After Hyde (Hy 62).
Nuclear Reaction
such as particle capture or a direct reaction, e.g.
\( \alpha \) or \( \beta \)

\( ^{239} \text{U} \)
Target nucleus in equilibrium deformation

\( ^{235} \text{U} \)
Transition state nucleus with saddle deformation
and \( E = 5 \text{MeV} \) of excitation energy

\( 10^{-21} \text{sec} < t < 10^{-20} \text{sec} \)
Scission configuration, big kinetic energies, few fragments, high deformation, small kinetic energies of fragments, occasional small particle emitted although neutron yield may be relatively large

\( ^{235} \text{U} \) and \( ^{232} \text{U} \)
Accelerating primary fragments under Coulomb interaction

\( ^{235} \text{U} \) and \( ^{232} \text{U} \)
Neutron emission in time period of the order of
\( 10^{-13} \text{ to } 10^{-14} \text{ sec} \)

\( ^{235} \text{U} \) and \( ^{232} \text{U} \)
Primary fission products, secondary decay products, and their radioactive decay products

\( ^{235} \text{U} \) and \( ^{232} \text{U} \)
Stable end products
FIGURE 3

Liquid-drop model. After Leachman (Le 60).
LIQUID DROP MODEL

~6 MEV

~200 MEV
corresponds to the fission barrier. It is this barrier which prevents the heavy nuclei from instantaneous decay by spontaneous fission. The barrier height is found experimentally to be about 6 MeV for nuclei between $^{232}\text{Th}$ and $^{239}\text{Pu}$; Figure 4 shows a cross section of the potential energy surface taken along the radial axis.

The pioneering calculations by Bohr and Wheeler (Bo 39) were later improved by Frankel and Metropolis (Fr 47). In view of this work reliable estimates are now available of the critical or activation energy as a function of the mass number and the atomic number of the nucleus. The predictions based on the liquid-drop model consequently can be compared with experiments.

The liquid-drop model is able to explain some aspects of the fission phenomenon, as for example, the observation that a nucleus will be more fissionable, the larger the fission parameter $Z^2/A$. The model also predicts the limit of stability of nuclei against fission, the critical deformation or threshold energy, the fission-energy, and the spontaneous fission rate. As a consequence of the odd-even fluctuations in neutron-binding energy it was predicted, using this theory, that $^{235}\text{U}$, $^{233}\text{U}$, and $^{239}\text{Pu}$ will undergo fission with thermal neutrons, as observed. It was hoped that the liquid-drop model would explain the observed asymmetry of low-energy fission. The calculations lead, however, to the symmetrical as the most probable mode.
FIGURE 4

Energy of a fissioning nucleus as a function of the distance of the fragments.
Potential Energy

Distance Along Radial Axis

Saddle Point
FiSSion Barrier
Stable Spherical Shape

To Fission
of fission. However, experiments clearly show that fission proceeds asymmetrically into unequal sizes of fragments, except in the case of very high-energy fission. There is no evidence that the effects producing asymmetric fission are represented in this model, and no conclusive indications have been given that the minimum in the yield-mass curve for thermal neutrons and low-energy particle and neutron fission can be explained by the liquid-drop model alone.

Dynamic models such as that proposed by Hill (Hi 53) and earliest works of Swiatecki (Sw 65), which follow the motion of the compound nucleus from its ground state or its saddle point up to configurations of tangent or even infinitely separated fragments, led to reasonable results. The most precise and detailed calculations of Nix and Swiatecki (Ni 65), however showed that the asymmetric mass distribution, which is observed in the low-energy fission of heavy nuclei, cannot be explained better than as gross approximations. The calculations were further applied recently by Plasil and Schmitt (Pl 72) to their experimental data obtained from 77.3-MeV, \(^4\)He-induced fission of \(^{181}\)Ta, \(^{209}\)Bi, and \(^{233}\)U. It was concluded by them that the model describes fission of relatively light nuclei in general terms, but that it should not be used in those cases where accurate quantitative predictions are required.
(b) **THE SHELL MODEL**

When the liquid-drop model failed to explain the asymmetry of thermal and low-energy fission, it was proposed that nuclear shells are, in some way, responsible for this phenomenon. Thus the asymmetry in the fission-yield curve may be correlated with the existence of closed shells of 50 and 82 neutrons. Explanations based on this assumption have been given by Mayer (Ma 48), Meitner (Me 50, Me 52) and others.

The shells are assumed to persist in the nucleus at the moderate excitations giving rise to asymmetrical fission. The splitting of the nucleus then takes place in a mode so as to distribute the neutrons in the fissioning nuclide in fragments containing >82 neutrons and >50 neutrons. This distribution gives rise respectively to the heavy and the light group in the yield-mass curve. In a highly-excited nucleus, however, the shell structure cannot persist and high-energy fission will therefore be symmetrical, as is observed, and also predicted by the liquid-drop model.

The theory of nuclear shells gives a superficial qualitative explanation of the process, but since no detailed mechanism for the fission process is known, it has not been possible to apply this theory in a quantitative manner.

(c) **THE HYBRID MODELS**

We have seen how the liquid-drop model has
successfully been used to explain many aspects of the fission process, but not the asymmetry of fission. On the other hand the shell model seems to offer some explanation of the asymmetry of fission.

Since real nuclei show properties that are characteristic of both the previous models, it is reasonable to assume that the real picture is a hybrid of the liquid-drop and the independent-particle (shell model).

One of the first attempts to use the idea of a hybrid model was that of Wick (Wi 49). He made use of the idea of nuclear shells by assuming that there is a connection between fission yields and the oscillations of the packing fraction curve and compared the fission process with the dissociation of a diatomic molecule.

(i) **Adiabatic Model of Asymmetric Fission.** According to Dickmann et. al. (Di 69) it is assumed that, in the process of fission, a potential barrier develops at a position \( Z = Z_0 \) (the symmetry axis of the nucleus is chosen as the \( Z \)-axis). Consequently, a neck is formed in the nuclear shape which leads to the final separation of the fragments. The barrier is represented by a repulsive \( \delta \)-function which is added to the single-particle potential. The position \( Z_0 \) of the neck is determined by requiring that the energy \( E(Z) \) needed to insert the barrier should be minimized. This picture presupposes an adiabatic fission process at the time
of neck formation. Preliminary calculations using this model were performed for the nuclei $^{246}\text{Cf}$, $^{236}\text{U}$, and $^{226}\text{Ra}$ using a cylindrical box as a highly simplified single-particle potential. For $^{246}\text{Cf}$ and $^{236}\text{U}$ the absolute minima of $E(Z)$ were located at asymmetric positions.

The objections levied against this kind of approach is identical to any approach treating the nucleus thermodynamically "cold", as that originally proposed by Wilets (Wi 64). In essence the adiabatic approach restricts the characteristic time for the collective motion of the nucleus to be much greater than that for particle motion to adjust itself adiabatically to the collective motion of the nuclear surface. According to these concepts, the low-lying levels of nuclei - in particular, even-even nuclei - are rotational and vibrational bands separated from excited intrinsic states by an energy gap. A similar arrangement of levels is considered to hold at the saddle point in an adiabatically sealed "cold" nucleus. There is no viscosity in the adiabatic model. Therefore, there is no effective mixing or exchange of energy between levels separated by the energy gap as the nucleus proceeds from saddle point to scission point. Low-energy fission would then proceed exclusively by the low-lying states and the fragment distributions should be representative of the properties of these states.

Fong (Fo 64) has argued that the adiabatic model cannot be applied to the fission process from saddle to
scission. He calculates that a level spacing of $\gg 8.4$ MeV at the ground state is required for this model to be valid. Heavy odd-A nuclei have four or five intrinsic levels per MeV and, hence, a level spacing which is very small compared to 8.4 MeV. Another argument advanced by Gindler and Huizenga (Gi 68) points out that, since there is no viscosity in the model, the kinetic energy of the fragments should increase with an increase in excitation energy of the compound nucleus. Experimentally, this is generally not observed. It appears that strong non-adiabatic processes become important between the saddle point and scission.

(ii) The Dynamic Model

A liquid-drop kinetic energy obtained by the assumption of ideal hydrodynamic flow of nuclear matter is essential for the work of Nix and Swiatecki (Ni 65) described earlier. Unfortunately, the calculations predict mass distributions peaked at symmetry, and the method is, therefore, applicable only to symmetric fission cases, i.e., to the fission of nuclei lighter than radium or to the fission of heavier nuclei at high excitation energies. This limitation is due to the fact the liquid-drop intrinsically favors symmetric configurations and it is likely that asymmetric fission results from shell-structure effects, as mentioned previously.

To make this dynamic model of Swiatecki also
applicable to the low-energy fission of heavy elements, Hasse (Ha 68, Ha 69) has proposed a model modified by adding a simple shell-energy term to the liquid-drop potential and including a curvature energy as proposed by H. von Groote (Gr 69). The shell-energy terms used are those from Myers and Swiatecki (My 66) based on single-particle levels in highly-deformed nuclear potentials. It also contains a phenomenological attenuation factor to allow for non-spherical ground states.

Myers and Swiatecki shell energies, which vanish near the saddle-point, were applied to the region beyond the saddle and to the vicinity of the scission point. This was achieved by defining two "clusters" in the nucleus undergoing fission. The proton and neutron numbers of the clusters are $Z_H$, $Z_L$ and $N_H$, $N_L$ respectively, where subscripts H and L refer to heavy and light clusters. It is then assumed that in the region between the saddle and the scission point, the sum of the shell energies of the clusters applies, multiplied by an arbitrary attenuation factor.

The main discrepancies between the experimental and those of the dynamical model in the version presented above are:

(i) The heavy fragment mass distribution does not peak at the experimentally observed $A \approx 140$ but at $A \approx 128-118$, 
depending on the value of the attenuation factor. This could be due to any neglected nuclear interaction terms between the two clusters, and/or the fact that the magic numbers seem to be shape-dependent, as is known from Strutinsky's (St 69) work.

(iii) The Cluster Model

The conceptual guide-line incorporated in the dynamical model of Hasse of using the cluster model of the nucleus, is based on the pioneer work of Faissner and Wildermuth (Fa 62, Fa 64, Fa 66). The idea is that large clusters (for instance with the above-mentioned magic numbers) can also be formed within the fission-prone compound nucleus during its migration through the many energetically possible nucleon configurations. This is an improbable configuration - about as improbable as the concentration of the excitation energy on one neutron, which then can escape. But as soon as such a two-cluster state is reached, the energy gained by cluster formation enhances the probability for fission considerably.

The thermal-neutron fission data of Wahl et. al. (Wa 62) and Konecny et. al. (Ko 65), using radiochemical procedures, have been used by Faissner to compute charge distributions for fixed fragment mass number for these sets of data. The only definite discrepancy is that the computed distributions have broader wings, in particular towards
larger charges for light fragments (i.e. smaller charges for the heavy fragments).

(iv) The Fission Probability and the Statistical Model.

The probability of fission seems to be related to the relationship between the nuclear charge (Z) and mass number (A) of the fissioning nucleus. In order to distinguish between fissile and non-fissile nuclides, a parameter, x, called the "fissionability parameter" is obtained by considering the Coulomb and surface energies of the spherical nucleus, Halpern (Ha 59). The Coulomb energy is proportional to \((Ze)^2/R^3\), where \(R\) is the radius of the nucleus, and the surface energy is proportional to \(4\pi R^2\). Thus the parameter \(x\) is proportional to \(Z^2/R^3\) and hence the term \(Z^2/A\) enables one to estimate the fissionability of a nuclide. Those nuclei with a value of \(Z^2/A\) greater than about 45 are unstable towards fission.

However, measurements made for photo-fission and cross-sections for neutron fission indicated that the fission thresholds do not depend upon the values of \(Z^2/A\) alone. Similar conclusions were drawn from the studies of half-lives of spontaneous fission of even-even nuclides. Seaborg (Se 52) has developed an empirical relation to calculate the slow-neutron fission threshold, \(E_b\), as given by the following equation:
\[ E_b = (19.0 - 0.36 \frac{Z^2}{A} + \varepsilon) \text{ MeV} \]

where

\[ \varepsilon = 0 \text{ for even-even} \]
\[ \varepsilon = 0.4 \text{ for even-odd, and} \]
\[ \varepsilon = 0.7 \text{ for odd-odd nuclides.} \]

Since a measurable amount of neutron-induced fission occurs at an excitation energy less than the fission threshold, \( E_b \), it was further proposed that the activation energy, \( E_a \), is about 0.9 MeV less than \( E_b \). The values of \( E_b \) and hence \( E_a \) can be calculated and also the neutron-binding energy, \( B_n \) for a nuclide with mass number \( A + 1 \) can be obtained. If the difference between \( (B_n - E_a) \) is negative, then the fission cross-sections will be less than about one barn. However, even though the correlation between calculated and observed values for slow-neutron fission is good, this line of demarcation of fissile and non-fissile is quite arbitrary.

In fission induced by particles with high energies, where the competition between fission and neutron evaporation has to be considered and the probability of neutron emission is comparable to that of fission, the relative probability for neutron emission or fission is expressed in terms of the partial "width" ratio \( \Gamma_n/\Gamma_f \). This ratio, though subject
to a mass number dependence, decreases as the nuclear charge increases. There is no marked dependence of the above ratio on excitation-energy.

Fong (Fo 64) has tried to calculate the fission process by the use of statistical mechanics. The fact that the fission probabilities of each mode vary considerably while the energy dependence changes only little, suggested the idea of using the Fermi gas model of the nucleus. He assumes that the probability of occurrence of a given mode of fission is proportional to the number of quantum-statistical states of the corresponding nuclear configuration at the moment just before the fission fragments separate. In order to give the statistical theory of fission, the gas model is used to calculate the number of states each mode can have. Fig. 5 depicts the concepts outlined.

The premise on which the foundation for this model lies is on the assumption that equilibrium exists at the moment of scission. Swiatecki (Sw 65) visualizes an implication in the statistical model that there exists a strong coupling between the collective and internal (single particle) motions, and that the model is essentially an extension of the single-particle model at high energy. This is borne out by the discrepancies and inability to explain the low-energy fission data in spite of Fong's theoretical arguments in his papers. Fine structure in the n + 235U mass distribution was not reproduced by the
FIGURE 5

After Leachman (Le 60).

Description: Diagram on statistical model.
Statistical Model predicts that the most probable fission asymmetry would result in fragments with the greatest density of nuclear excitation.
In the accompanying figure, the excitation energy is the difference between the two surfaces. The bottom surface (with identical meaning of radial and azimuthal co-ordinates as before) is the energy liberated by the excess of mass of the fissioning nucleus over the mass of the two fragments, and the upper surface is the Coulomb repulsion energy of the two fragments.
F R A G M E N T
S E P A R A T I O N

0 MEV

M₀ ENERGY

C O U L O M S
E N E R G Y E₀

M₀ = M_L + M_H + E_K + E^*

S Y M M E T R I C
F I S S I O N
M_H / M_L = 1

F R A G M E N T
E X C I T A T I O N E^*

M_H / M_L = 1.8

M_L + M_H ENERGY

M AX I M U M Y I E L D

M_H / M_L = 1.3

S T A T I S T I C A L M O D E L
Fong calculation. Application of the model to $^{239}\text{Pu} + n$ fission yielded a four-lumped mass distribution shown in the work of Perring and Storey (Pe 55). The original Fong calculations were well defined and free of arbitrary parameters but at the present stage considerable amount of experimental data could only agree with the theoretical predictions made by Fong on the basis of properly selected input parameters in the latter calculations. By modifications of the original theory and the employment of a certain amount of empiricism, reasonable agreement is obtained with various experimental distributions, e.g., Newton (Ne 73), Erba et al. (Er 64). The important question remains: Can these empirical changes be justified theoretically?

It is apparently clear that the state-of-the-art in the development stage of nuclear models to explain the fission phenomenon is still rudimentary, still grappling with explanation of neutron-induced fission. The fission of a heavy element at moderately high excitation energy presently remains well beyond the realm of any of these models considered.
D. FISSION YIELDS AND THEIR CORRELATIONS

(1) Introduction

The concept and first measurement of fission yields is due to Anderson, Fermi, and Grosse (An 41), and Moussa and Goldstein (Mo 41). As each fragment (i.e., primary-formed fission nuclide) gives rise to a fission-product decay chain ending in a stable nuclide, it is possible for all members of a decay chain, except the first, to have two modes of formation, either through decay from a parent nuclide or independent formation. Consequently, we can define two types of fission yields:

(i) The total or cumulative fission yield, \( y \), of a given fission product is defined as the percentage of fission acts giving the nuclide in question by independent or direct formation and through decay of precursors, without reference to half-life.

(ii) The independent yield, \( y \), of a fission fragment is similarly defined as the percentage of fission acts giving the nuclide in question by direct formation in the ground state and very short-lived excited states. Isomers are treated independently. Consequently, in the study of the fission process, one will be faced with two problems:

(i) The fission yield of a given mass number (total chain-yield) as a function of mass \( A \). This will
give a mass-distribution curve.

(ii) The variation of yields of primary fragments along a fission product decay chain for a given mass, giving a charge-distribution curve.

Once a fission fragment is formed and the prompt neutrons emitted there is no change in the mass number (except for very small effects as a result of delayed neutron emission). The number of fissions leading to a given mass number as a function of mass are therefore time independent. The yield as a function of charge, however, is affected by time due to the change in nuclear charge caused by the $\beta$-decay in the fission product chains.

(2) The Distribution of Yields of Fragments as a Function of Mass Number.

The yield-mass curve is a characteristic of the nucleus undergoing fission and also depends on the energy of the particle undergoing fission. The distribution of mass in fission has been determined by two types of measurements: (a) physical-measurements, and (b) chemical measurements for which there are two methods available, (i) radiochemical (ii) mass spectrometric. In the mass spectrometric technique, the yields of stable and long-lived members at the end of the mass chain are determined, Thode et. al. (Th 60)
(a) **Physical Measurements**

Kinetic energy distribution of fission fragments is determined either by measuring the ionization produced by fragments in an ionization chamber or by measuring the velocity distribution of the fragments by the time-of-flight method. Both techniques are outlined below:

(i) **Ionization Measurements**

The experiments by Jentschke (Je 43), by Flammersfeld (Fl 43), and by Deutsch and Ramsey (De 45), are of special interest for the present discussion of the mass distribution curve.

In these experiments the energies for both fission fragments were measured simultaneously with a double "back-to-back" ionization chamber with a thin uranium film sandwiched between the chambers. The fragments from each act of fission will travel in opposite directions into the two chambers and produce ionization. For the translation of the pulse sizes into fragment energies it is assumed that the total kinetic energy loss of a pair of fragments is proportional to the ionization produced. Due to conservation of momentum the ratio of the kinetic energies must be equal to the inverse ratio of the masses (neglecting momentum of the emitted prompt neutrons) and a mass distribution curve can be estimated.
Because, \[ M_L V_L = M_H V_H \] \hspace{1cm} (I-3)

where \( M \) = mass of fission fragment
\( V \) = velocity of fission fragment, and
\( L \) and \( H \) refer to light and heavy fragments respectively.

Also, it can be easily derived from the above eqn. that

\[
\frac{E_L}{E_H} = \frac{1/2 M_L V_L^2}{1/2 M_H V_H^2} = \frac{M_H}{M_L}
\] \hspace{1cm} (I-4)

where \( E \) = Kinetic Energy of the fission fragment.

(ii) **Velocity Measurements**

The distribution of the velocity of fission fragments is measured directly by irradiating a target film deposited on a thin foil. Measurements are made by the time-of-flight method. The fission fragments are detected by scintillation-detectors. One fragment travels only about a centimeter before striking a detector, while the other fragment travels about 350 centimeters along an evacuated tube. The pulses are projected on a cathode ray tube and photographed. The distance between the two peaks gives the velocity which is then converted into the kinetic energy of the fission fragments.

The energy distributions of the fission products
formed in thermal-neutron induced fission of $^{233}\text{U}$, $^{235}\text{U}$, and $^{239}\text{Pu}$ have been determined by Brunton et. al. (Br 50a, Br 50b), and, in fast-neutron fission of $^{235}\text{U}$, $^{238}\text{U}$, $^{232}\text{Th}$, and $^{239}\text{Pu}$ by Friedland (Fr 51) and Wahl (Wa 54). Leachman (Le 52), and Milton and Fraser (Mi 62) have measured the velocities of fission fragments produced in the thermal-neutron fission of $^{233}\text{U}$, $^{235}\text{U}$, and $^{239}\text{Pu}$.

Britt and Whetstone (Br 64) studied the fission of $^{230}\text{Th}$, $^{232}\text{Th}$, and $^{233}\text{U}$ induced by alpha particles of various energies and determined the mass and total kinetic-energy distributions of the fragments. They also reported the average number of prompt neutrons emitted as a function of the fragment mass. Milton and Fraser (Mi 62) have reported the energies, angular distributions, and yields of the prompt neutrons from individual fragments in the thermal-neutron fission of $^{233}\text{U}$ and $^{235}\text{U}$ by velocity measurements.

The yield-mass curves, calculated from the physical experiments, show the same overall trend, with two maxima and a minimum in between, as the curves obtained by the chemical methods (see below). The physical methods, however, have much lower mass resolution than the chemical ones.

(b) Chemical Measurements

Since the early days of the investigation of the fission process, radio-chemical methods have been used to
determine yields of fission products. In general low-energy fission of a heavy nucleus results in two unequal mass-fragments which fall in the mass region between about 70 and 170. The task of a radio-chemical fission-yield determination consists in the isolation and measurement of a radiochemically pure sample of nuclides.

In contrast to the physical methods the radio-chemical methods have led to a much more precise and extensive determination of the mass distribution curve. The reason for this is the advantage of a complete and unambiguous mass resolution; such resolution is possible by the chemical isolation of the fission products and the identification of nuclides with known and distinct mass numbers. Furthermore the sensitivity of chemical methods is not, as with the physical methods, greatly reduced for nuclides with very low yields. Fission yields have been determined by chemical methods over a range covering nearly a factor of $10^8$.

**Absolute and Relative Fission Yields:**

In the introductory part of the section, the definition of 'fission-yields' is given, and it follows that the absolute cumulative-yield (often termed only absolute yield) of a fission nuclide is given, expressed in per cent, by:
\[ Y = \frac{\text{number of atoms formed directly and by decay} \times 100}{\text{total number of fissions}} \]

and similarly the absolute independent yield:

\[ Y_i = \frac{\text{number of atoms formed directly} \times 100}{\text{total number of fissions}} \]

Consequently, the determination of absolute fission yields requires knowledge of:

(i) The disintegration rate of the fission nuclide in the bombarded sample at the steady state condition of uranium-irradiation, referred to as "saturation activity",

and

(ii) The number of fissions occurring in the sample per unit time; this number is referred to as the absolute fission rate.

If two nuclides A and B are produced in the sample of fissionable material, the yield of A will be related to that of B by the equation:

\[ Y_A = Y_B \frac{D_A}{D_B} \]  \hspace{1cm} (I-5)

Here \( D_A \) and \( D_B \) are respectively the disintegration rates at saturation.

If \( Y_B \) is known from an absolute determination, the yield of A can be determined by the method of activity
comparisons, when the saturation disintegration rates of A and B are known from radiochemical analysis and absolute β-activity measurements. Then B is used as a fission yield standard.

In the early stages of the study of the fission phenomena involving thermal-neutron fission, the only absolute yields reported were those for $^{132}$Te and $^{140}$Ba. The largest error in the determination of fission yields lies in the converting of observed counting rates to absolute disintegration rates.

(c) **Mass Spectrometric Method**

This is a very sensitive method with good precision (±1%) in some cases, to determine relative or absolute fission product yields. It is possible to determine the yields of stable and long-lived isotopes that grow from active fission products and that may be formed directly in fission. Fission yields for several mass chains can be determined at the same time for a mixture of fission products from a single irradiation.

(c) **Experimental Results**

Figure 6, taken from compiled works of Katcoff (Ka 58), Ford and Gilmore (Po 56), and, J. G. Cuninghame (Cu 57), shows the experimental results of many researchers who studied neutron-induced fission. The data taken from
The measured mass yields from fission of $^{235}\text{U}$ and $^{238}\text{U}$ induced by neutrons of various energies.

The abbreviation Th. is for thermal neutrons and F.S. for fission spectrum neutrons.

<table>
<thead>
<tr>
<th>Legend</th>
<th>Th</th>
<th>FS</th>
<th>5</th>
<th>8</th>
<th>14</th>
</tr>
</thead>
<tbody>
<tr>
<td>Katcoff (Ra 58)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ford and Gilmore (Fo 56)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cuninghame (Cu 63c)</td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
</tbody>
</table>
Cunninghame's work have been renormalized to the $^{99}$Mo yield of Terrell et. al. (Te 53), to be consistent with other data shown in the Figure. The mass yields from neutron fission of the important uranium isotopes $^{235}$U and $^{238}$U shown in the figure 6 indicate the following salient features:

(a) The curves are approximately symmetric in nature but the mode of fission is asymmetrical, as exhibited by the occurrence of light and heavy mass peaks. The nuclides which lie at the bottom of the valley typify symmetric fission and those which lie at the peaks typify asymmetric fission.

(b) The maxima of the heavy-mass peaks seem stationary near about mass number 137, and the mass of the light fragment shifts to conform with the different masses of the fissioning nuclei. This fixed position of the heavy fragment peak is more clearly demonstrated in the review work by Weinberg and Wigner (We 59) using the data for a variety of fissioning nuclides ranging from $^{229}$Th to $^{244}$Cm. However, a tendency for the heavy peak to shift to heavier masses is found for the fission of heavier nuclides, particularly for $^{254}$Fm.

(c) The data of Ford and Gilmore represented in figure 6 confirm in some detail the rise in the
valley between the mass peaks with increasing energy. This is accompanied by a slight broadening of the wings of the distributions.

(d) An interesting detail in the yield from thermal-neutron induced fission of $^{235}\text{U}$ is the small, but well-established, fine-structure peaks at masses roughly corresponding to the 82-neutron shell. This has been postulated as resulting from the shell influence, either on the fission act or neutron preferential emission to the observed masses. However, this is difficult to prove quantitatively. For 14-MeV fission no fine structure is observed, perhaps because the variation in the greater number of emitted neutrons prevents the formation of structure resulting from preferential neutron emission.

With the compilation of more experimental data, the review article by Halpern (Ha 59) clearly indicates that fission occurring in highly excited nuclei of the heaviest elements as obtained by high-energy bombardment shows decreasing asymmetry until for very high excitation, fission becomes symmetric. Figure 7 gives a schematic summary as the relative yield of very asymmetric fission also increases. The twin-peaked distribution characteristic of low-energy fission, eventually, gives way to a broad single-humped curve with a rather flat top.
FIGURE 7

Schematic summary of fission properties [from Hyde et al. (Hy 64)].
### Fission Probability and Excitation Energy Effects

<table>
<thead>
<tr>
<th>Highly Fissile Elements</th>
<th>Near Threshold</th>
<th>Exc. energy 10-40 MeV</th>
<th>Exc. energy &gt; 40 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Thorium and heavier elements)</td>
<td>Asymmetric fission</td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
</tr>
<tr>
<td><strong>Fission probability</strong></td>
<td>threshold has lower, ( f_1/f_2 ) moderate to high, ( \epsilon ) approaches ( \epsilon_{\text{ex}} ), not strongly dependent on excitation energy.</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Intermediate Elements</th>
<th>Near Threshold</th>
<th>Exc. energy 10-40 MeV</th>
<th>Exc. energy &gt; 40 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Actinium, radium, etc.)</td>
<td>Symmetric and asymmetric fission thresholds about equal, ( f_1/f_2 ) low. Asymmetric fission does not increase with excitation. Symmetric fission increases rapidly and soon washes out asymmetric fission.</td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Slightly Fissile Elements</th>
<th>Near Threshold</th>
<th>Exc. energy 10-40 MeV</th>
<th>Exc. energy &gt; 40 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Lead-bismuth)</td>
<td>Symmetric fission threshold has lower, ( f_1/f_2 ) very low but increases markedly with energy; but ( \epsilon ) never approaches ( \epsilon_{\text{ex}} ). Symmetric fission predominates. ( f_1/f_2 ) levels off at high excitation.</td>
<td><img src="image" alt="Diagram" /></td>
<td><img src="image" alt="Diagram" /></td>
</tr>
</tbody>
</table>

**Shape of mass yield curve**

- Near threshold:
  - Highly fissile elements: Asymmetric fission
  - Intermediate elements: Symmetric and asymmetric fission
  - Slightly fissile elements: Symmetric fission

- Exc. energy 10-40 MeV:
  - Highly fissile elements: Asymmetric fission
  - Intermediate elements: Symmetric and asymmetric fission
  - Slightly fissile elements: Symmetric fission

- Exc. energy > 40 MeV:
  - Highly fissile elements: Asymmetric fission
  - Intermediate elements: Symmetric and asymmetric fission
  - Slightly fissile elements: Symmetric fission
Stevenson et. al. (St 58), from their work together with data of Lindner and Osborne (Li 54), and Hicks and Gilbert (Hi 55), have reported the fission-product distribution of $^{238}\text{U}$ at various proton energies. It can be seen that, with increasing projectile energy the distribution becomes broad and the two humps slowly disappear, as mentioned previously. Qualitatively stated the results of Lavrukhina et. al. (La 67), Hagebo (Ha 66), Stevenson et. al. (St 58), Baba et. al. (Ba 71), and, Shirato et. al. (Sh 64) indicate that the trough in the mass distribution disappears above about 100 MeV excitation and the fission cross-section remains almost constant above 30 MeV. The composite data indicated in figures 8, 9, 10, 11, are taken from the above sources, private communications with Stevenson (St 73) and Umezawa (Um 73), and total chain yields as reported by Miller (Mi 70). Primarily the data of Baba et. al. have been used covering the mass region of interest in this work between 30-55 MeV bombarding energy of protons, and this reference has been exclusively used for mass distribution at 20 MeV. The mass-yield data of Shirato et. al. (Sh 64) at 55 MeV, are not used in this work because the physical measurements using a back-to-back pair of detectors yield result somewhat different from those obtained by radiochemical means and are difficult to reconcile. This is because at this energy the probability of neutron emission per fission is large.
FIGURE 8

Fission product mass distribution, $^{238}\text{U}(P,f)$ (composite curves at 75 and 100 MeV).

Legend

L. Miller (Mi 72)
Hicks et al. (Hi 55)
Stevenson et al. (St 58)
Lindner and Osborne (Li 56)
MASS REGION
OF INTEREST

\[ \sigma_{(mb)} \]

\[ \begin{array}{c}
40 \quad 60 \quad 80 \quad 100 \quad 120 \quad 140 \quad 160 \\
0.1 \quad 1 \quad 10 \quad 100
\end{array} \]

75 MeV

-100 MeV
Fission product mass distribution, $^{238}\text{U}(P,f)$ (composite curves at 40 and 55 MeV). Ref. Baba and Umezawa (Ba 71).
MASS REGION OF INTEREST

55 MeV
BABA, UMEZAWA
NUCL. PHYS. A175, 175 (1971)

40 MeV
BABA, UMEZAWA (1971)
FIGURE 10

Fission product mass distribution, $^{238}\text{U}(p,f)$ (composite curves at 32 and 35 MeV. Ref. Baba and Umezawa (Ba 71) and Stevenson et al. (St 58).
MASS REGION
OF INTEREST

35 MeV
BABA, UMEZAWA
(1971)

32 MeV
STEVENS\(\text{E}^\text{N}\)SON ET AL.
(1958)
FIGURE 11

"Fission product mass distribution, $^{238}\text{U}(P,f)$ (composite curve at 20 MeV). Ref. Baba and Umezawa (Ba 71)."
The Distribution of Nuclear Charge In Fission

The variation of yields of primary fission fragments (independent yields) along a fission product decay chain of a given mass number gives the charge distribution in fission. These primary yields of individual fission products are usually far removed from beta stability so that most of the radioactive decay half-lives are very short. Thus primary yields for most fission products are difficult to measure and consequently relatively few have been determined.

Direct measurements of independent yields of some fission fragments have been made possible by the existence of shielded and semi-shielded nuclides. A shielded-nuclide is one which cannot be formed by beta decay because its immediate precursor in the isobaric decay chain is stable (e.g. $^{136}$Cs). A semi-shielded nuclide is one which can be chemically isolated in a time short compared with the half-life of its beta decaying precursor (e.g. $^{140}$La).

The primary fission fragments are not completely stripped of electrons, Lassen (La 45), and these charges decrease rapidly as the fragments are slowed down in passing through matter, and no physical method exists that enables us to determine the nuclear charge of fission at the instant of fission. Therefore, the physical methods which have added much to our knowledge on the distribution of kinetic energy in fission have been of very little help in the charge-distribution problem.
Successful solutions of the problem will therefore depend on X-ray mass spectrometric and radiochemical methods. Carter, Wagman and Wyman (Ca 58) observed for the first time the energy spectrum of X-rays in coincidence with fission fragments by using a thin NaI crystal for K X-rays and a proportional counter for L X-rays.

The chemical approach too is difficult since it involves the determination of the most probable primary charge, $Z_p$, of fission fragments, for a given mass $A$, and also the charge distribution curve around $Z_p$. Consequently, it is a problem of determination of independent yields and the difficulty of the experimental approach arises when the object is to obtain complete data for a given fission-product chain. This object is impossible to achieve as the first few members of the chain have very short half-lives, and determination of their independent yield is therefore not feasible. The only way to resolve the difficulty is to accumulate as many independent yields as possible on various masses, and evaluate semi-empirically that in a limited portion of a given mass region the assumption that the charge distribution curve is the same for different masses.

Determination of independent yields of accessible nuclides in chains of different mass number must be followed by correlation of these data, in order to obtain the charge distribution. Correlation is customarily effected by means of theoretical or semi-theoretical relationships that give
the most probable charge, $Z_p$, for a given mass number, $A$.

The various postulates that have been put forward to obtain a value of the most probable charge, $Z_p$, are summarized below:

Postulates regarding the distribution of Nuclear-charge in the fission process.

(i) **Minimum Potential Energy (M.P.E.)**:

This postulate was suggested by Way and Wigner (Wa 51) and theoretical foundations are based on the work of Present (Pr 47). In its original form it states that the most probable charge distribution in fission is that in which the two fragments as spheres in contact just before scission have a minimum potential-energy. The prescription of McHugh (Mc 68), based on the liquid-drop mass formula of Green (Gr 54) gives the value of $Z_p$ (the most probable value of the charge) as follows:

$$Z_p = \frac{Z_C (a_4 A_H^{-1} + a_3 A_H^{-1/3} - 1/2Q^2 D^{-1})}{a_3 (A_L^{-1/3} + A_H^{-1/3}) + a_4 (A_H^{-1} + A_D^{-1}) - Q^2 D^{-1}}$$  \hspace{1cm} (I-6)

where

- $Z_C$ = charge of the fissioning nucleus
- $A_L, A_H$ = masses of the light and heavy fragments
- $Q$ = unit of electrostatic charge
- $D$ = effective separation distance of the fragment centres.
The constants given by Green are $a_3 = 0.718$ and $a_4 = 94.07$

(ii) **Unchanged Charge Distribution (U.C.D.)**

Sugarman and Turkevich (Su 51) put forward a modified version of this hypothesis and it was first stated by Goeckermann and Perlman (Go 49) to account for the results of their study of 190-MeV deuteron-induced fission of bismuth. According to this hypothesis the fission products will have the same charge-to-mass ratio as the fissioning nucleus. It can be stated in a modified way that if there exists a correlation of excitation-function shapes with $N/Z$ ratios of fission fragments - that UCD proposes that the compound nucleus fissions in such a way that the fission products will have the same neutron-to-proton ratio ($N/Z$) as the parent fissioning nucleus, Friedlander et al. (Fr 63).

The value of $Z_p$ could be calculated using the expression:

$$Z_p = \frac{Z_f}{A_f - \nu_T}$$  \hspace{1cm} (I-7)

where $Z_f =$ charge of the fissioning nucleus

$A_f =$ mass of the fissioning nucleus

$\nu_T =$ total number of prompt neutrons emitted (both fragments)
(iii) **Equal Charge Displacement (E.C.D.)**

This hypothesis was first proposed by Glendenin et. al (Gl 51) and later modified by Pappas (Pa 66). It states that when fission occurs, the most probable charge, $Z_p$ (not necessarily integral), for the light and heavy fragments will be equally displaced from stability. This can be given by the following equation:

$$ (Z_A - Z_p)_L = (Z_A - Z_p)_H $$  \hspace{1cm} (I-8)

where $Z_A$ and $Z_p$ are the most stable charge and the most probable charge respectively for a given mass chain A. The light and heavy fragments are designated by L and H respectively.

The equal charge displacement hypothesis was found to explain fairly well the mode of the charge division in the types of fission with low excitation, as indicated in the works by Wahl et. al (Wa 62) and, Glendenin et. al. (Gl 69). In the case of medium-energy fission, some doubt was, however, cast on the general applicability of the postulate. Instead the UCD hypothesis was found to give a more reliable estimate of $Z_p$ value as that obtained from the experiments with $^{239}$Pu and $^{235}$U targets as reported by Yaffe (Ya 69). Recently in this energy region, Diksić et. al (Di 74) invoked the UCD mechanism and calculated the total number of neutrons $v_T$ emitted in the
proton-induced fission of $^{238}\text{U}$ and found good agreement with the value of $\nu_T$ calculated from the experimental $Z_p$ value. In his very careful mass-spectrometric study of fission yields from $^{235}\text{U}$ and $^{238}\text{U}$ plus 45.7-MeV helium ions, Chu (Ch 69) concluded that perhaps something intermediate between a non-shell-affected ECD and a UCD rule might best explain the experimental results. Del Marmol (De 66) studied fission of $^{235}\text{U}$ with 13.6-MeV deuterons and 23.5-MeV helium, and concluded that the ECD rule gave a "satisfactory" interpretation of the fractional-chain yields for the excitation studied. Similarly Pappas and Hagebø (Ha 66) use this formalism to explain high-energy fission of $^{238}\text{U}$, Cuninghame et. al. (Cu 63) for photofission of $^{238}\text{U}$, Alexander and Coryell (Co 61) for fission of $^{238}\text{U}$ and $^{232}\text{Th}$ induced by 13.6-MeV deuterons.

The results reported by Blann (Ma 60) show that the MPE theory gives the best prediction of the empirical $Z$'s for the system gold plus carbon ions at 112 MeV, Colby and Cobble (Co 61a) have found that while the UCD rule gives excellent correlation for nuclides away from the 82-neutron shell, in their later studies (Co 61b) they had to invoke both the MPE and UCD rules. Very recently Fried, Anderson, and Choppin (Fr 68) interpreted their results on proton-induced fission of $^{232}\text{Th}$ with the ECD rule and deuteron-induced fission with the MPE postulate. In the neutron-induced fission of $^{232}\text{Th}$ and $^{238}\text{U}$ carried out by
Kuroda et al. (Ra 69, Ra 72) they found that the ECD hypothesis agreed well for the fission products produced by the asymmetric mode of fission, and the UCD or constant-charge-ratio (CCR) postulate showed the best correlation for the fission products produced by the symmetric mode of fission.

It is obvious from the compilation above that none of the rules have succeeded in becoming a general rule. Coryell et al. (Co 61) derived an empirical prescription describing Z values for any type of fission in relation to those in thermal-neutron fission of $^{235}$U. This prescription has been found totally invalid at medium to higher excitation by Umezawa et al. (Um 71).

Regarding the shape of the charge dispersion curve, the Gaussian function applied in the thermal-neutron fission of $^{235}$U is understandable in terms of the minimum potential energy prescription. The nuclear mass surface has a parabolic section along the charge axis and two of such parabolae are synthesized into a single parabola, since the mass surface rises so steeply along the charge axis that it will be expressed by a parabola in a rather wide range of the charge Z. In this description, the probability of formation or fractional independent yield of a product with a given atomic number, Z, within a particular mass chain is P(Z). Each mass chain is characterized by a single most probable atomic number, Z_P, and P(Z) is assumed to be given...
by the Gaussian equation:

$$P(Z) = A \exp \left[-\frac{(Z - Z_p)^2}{c}\right]$$  \hspace{1cm} (I-9)

The constant, $c$, reflects the relative width of the Gaussian function and has been found to have about the same value of $\sim 0.8$ for many decay chains, in the thermal-neutron fission work. The maximum amplitude of the function is given by $A$ which is related to $c$:

$$A = (\pi c)^{-1/2}$$  \hspace{1cm} (I-10)

The variation of $Z_p$ with mass number is called the $Z_p$ function and is determined empirically.

A Gaussian charge distribution may also be represented in cumulative form, as shown by Wahl et. al. (Wa 62). This representation gives unity for the sum of fractional independent yields in a decay chain. The fractional cumulative yield of a member of a decay chain with charge $Z$ is given by the equation

$$P(n) = (2\pi \sigma^2)^{-1/2} \left(\frac{Z + 1/2}{\sigma^2}\right) \exp \left[ -\frac{(n - Z_p)^2}{2\sigma^2} \right] dn$$

$$= \frac{1}{2} + \frac{1}{2} [f(Z - Z_p + \frac{1/2}{\sigma})]$$  \hspace{1cm} (I-11)

The function $f(x)$ is a normal probability integral and is
given by the equation (26.4)

\[ f(x) = (2\pi)^{-1/2} \int_{-x}^{x} \exp\left(-\frac{\alpha^2}{2}\right) dx \]  \hspace{1cm} (I-12)

The constants \( \sigma \) and \( c \) in the two representations for charge distribution are related approximately through Sheppard's correction

\[ c = 2(\sigma^2 + 1/12). \]  \hspace{1cm} (I-13)

To determine \( Z_p \) and \( c \) for a decay chain, it is necessary to determine the independent yields of several members of a mass chain. Equation (I-9) can then be fitted to the data by variation of \( c \) and \( Z_p \).

Because the charge dispersion is centered away from beta stability, most of the fission-products undergo beta decay to nuclide of higher atomic number and greater stability. This behavior makes determination of cumulative chain yields relatively easy; one waits after the end of an irradiation until all of the shorter-lived products decay, then an isobar near the end of the chain of decays is separated and measured. Independent yields are generally divided by the chain yield to obtain fractional independent yields. This serves to normalize all independent yield data.

Measurement of the independent yields of short-lived isotopes requires rapid chemical separations shortly after termination.
of irradiation to prevent build-up of the product by precursor beta decay. Also, since many of the decay chains have members with the same proton number, chemical distinction of these members is impossible. The members of the various chains with the same atomic number are distinguished by careful selection of duration of irradiation, separation time, purification procedures, and measurement procedures. As the half-life of the nuclides of interest decrease, the requirements on the selection of conditions become more stringent. When nuclides of the same atomic number in different chains have very similar half-lives and independent yields the problem of separation can become exceedingly difficult. The investigations reported in this dissertation employed either (a) Ge(Li) gamma-ray detector for measurement, or, (b) Beta detector, after an acceptable degree of purification of samples or measurement of activity for those isotopes having very unique Beta transition energies.

The excellent energy resolution of the Ge(Li) gamma-ray detector allowed measurement of gamma rays unique to a particular nuclide of interest in the presence of several other nuclides. Use of the Ge(Li) detector along with chemical purifications allowed measurement of several nuclides that would otherwise be poorly resolved from one another.
E. PREVIOUS WORK

(1) Charge Distribution in Low-Energy Fission.

Charge distribution studies in this energy region has been most extensive. A great deal of work has been done by Wahl et al. (Wa 62, Wa 65). The work showed that the charge distribution for fission products (final) in six isobaric chains, 91, 139-143, and, recent data by Fowler and Wahl (Fo 74) on chains 128 and 130, can be represented by a Gaussian distribution. On the basis of the neutron emission problem it can of course be argued, as was done by Fong (Fo 64) while proposing his statistical model, that there is no reason to believe that the final charge dispersion should always be a Gaussian even if it appears empirically so in the region of high-mass yields. According to the statistical model, Gaussian distribution is expected for the initial charge dispersion. Fong suggests that the final charge dispersion is broader than the initial one and may well be non-Gaussian, for, neutron emission may not be equal for all members of an isobaric chain. Glendenin et al. (Gl 69) have been able, by means of high-resolution studies of K X-ray emission from 252Cf fission fragments, to support this assumption for charge distributions in the region of high mass yields.

This result agrees also with the conclusions of Gordon and Aras (Go 66) based on calculations of neutron evaporation.
in fission under the assumption that the initial mass distribution and charge dispersion are smooth curves for thermal-neutron-induced fission of \( ^{235}\text{U} \). To-day, however, this assumption is not strictly valid for the initial mass distribution which has been shown \((\text{Wa} 69)\) to be perturbed as a result of pairing effects. Wahl \((\text{Wa} 69)\) has reported approximately 10\% narrower full-width at half-maximum (FWHM) of the Gaussian curve in his recent review of neutron-induced fission work. The difference is due largely to the newly observed \((\text{Wa} 69)\) odd-even effect in charge dispersion.

While making attempts toward finding systematics in charge distribution \(Z_p\) values, one faces the choice between two kinds of parameters; namely mass and mass fraction of the primary fragment. If the charge distribution is governed by the nuclear properties of the individual fragment, the most probable charge should be expressed in terms of the fragment mass, regardless of the fissile system. On the contrary, quantities relating to the collective motion of the fissioning nucleus are expected to be functions of the mass fraction, \(\eta\), the ratio of the primary fragment mass to the fissioning mass. Umezawa et. al. \((\text{Um 71})\) have compiled \(Z_p\) values obtained in the thermal-neutron-induced fission of \( ^{235}\text{U}, \ ^{233}\text{U}, \text{and} \ ^{239}\text{Pu} \) and in the spontaneous fission of \( ^{242}\text{Cm} \) and \( ^{252}\text{Cf} \) from works of several experimentalists. The observed nuclei listed in this compilation could be characterized as nothing but the secondary fragments.
after neutron emission is completed, therefore it is necessary to transform the secondary mass to the primary mass. Immediately after the scission. The mass of the nucleus at the moment of scission gives also the necessary information to find the complementary fragment mass or the mass fraction of a given fragment. The numbers of neutrons emitted before and after the scission has taken place, therefore, need to be known. The former, \( v_{\text{pre}} \), determines the fissioning mass while the latter, \( v_{\text{post}} \), changes the mass of the primary fragment.

Terrell (Te 65) has derived a general behaviour of neutron multiplicities by analysing the difference between the radiochemical yield-mass curve and the prompt yield-mass curve based on time-of-flight data. His systematics are approximately represented by two straight lines in the light and heavy fragment mass regions, that is:

\[
\begin{align*}
\nu &= 0.08 (M_L - 82), & 84 \leq M_L \leq 120 \\
\nu &= 0.10 (M_L - 126), & 130 \leq M_M \leq 160
\end{align*}
\]

The relationship gives the number of postscission neutrons, \( v_{\text{post}} \). As far as spontaneous or thermal-neutron-induced fission is concerned, the number of pre-scission neutrons, \( v_{\text{pre}} \) Neutrons emitted at the moment of scission and not involved in the mechanism of charge division.
as reported by several authors, e.g., Fraser (Fr 52), Ramanna and Rama Rao (Ra 58), and, Bowman et. al. (Bo 63), is small.

Based on the above assumptions there has been one prominent attempt to discuss the systematics in the most probable charge $Z_p$ for the $^{235}$U fission with thermal neutrons in terms of the heavy fragment mass. Milton (Mi 62) has ascribed the charge distribution to be governed by the nuclear shell effect of the heavy fragment, rather than the collective characters. In this treatment, the difference between $Z_p$ of a given fragment and the charge $Z_{UCD}$, to be given to the fragment if the charge distribution is unchanged is designated $\Delta Z_p$. He plotted $\Delta Z_p$ versus the heavy fragment mass $M_H$ from radiochemical data of several fissile systems (such as $^{235}$U + $n_{th}$, $^{233}$U + $n_{th}$, $^{239}$Pu + $n_{th}$ along with the results of the direct measurement of the spontaneous fission of $^{252}$Cf obtained by Kapoor et. al. (Ka 65). It was found that $\Delta Z_p$ is a function of the primary heavy fragment mass $M_H$ only in the peak region of the yield-mass curve, while the mass fraction $\eta$ defined as follows:

$$\eta = \frac{M + \nu_{\text{post}}}{A_c} - \nu_{\text{pre}}$$

where $M =$ mass of the secondary fragment

$A_c =$ mass of the compound nucleus

$\nu_{\text{post}} =$ no. of post-fission neutrons

...
\[ \nu_{\text{pre}} = \text{no. of pre-fission neutrons, governs} \]

de the value of \( \Delta Z_p \) elsewhere. It was therefore concluded that the shell-effect superimposes on the collective nature of the nucleus in the region corresponding to the peak of the yield-mass curve.

The Wahl plot (Wa 69), as applied to all cases known currently for spontaneous and up to 14-MeV neutron-induced fission, shows an average deviation from the unchanged charge distribution \( Z_p^\text{UCD} = \Delta Z_p \), to be of the order of +0.5 charge units and −0.5 charge units for light and heavy fragments respectively, Wolfsberg (Wo 65), and favours the equal-charge-displacement hypothesis of Glendenin. The trend given by this hypothesis seems to find support in the trend expected from the fragment shell theory as shown by Pappas et. al. (Pa 69). In the fragment shell theory the specific scission point shapes are calculated by minimizing the potential energy.

The experimental \( Z_p \) values which are corrected for neutron emission follow the calculated trends in the peak regions while in the valley region it looks as if the experimental \( Z_p \) approached the unchanged charge distribution faster than is expected from the theory. Whether this is a result of how the data are treated or indicates a contribution from the symmetric fission mode which is known to follow the U.C.D. mechanism is not clear.
(2) **Charge Distribution in Medium-Energy Fission.**

This process has been studied in a number of fissile systems of which the following works are prominent: Pate et. al. (Pa 58), $^{232}$Th with protons by Benjamin et. al. (Be 69), Holub et. al. (Ya 73), T. McGee et. al. (Mc 72), and by Choppin et. al. (Fr 68), with deuterons by Choppin et. al. (Fr 68), with alpha particles by McHugh and Michel (Mc 68), system $^{238}$U + p - studied by the Radiochemistry group at McGill as summarized by Yaffe (Ya 69) and recent published works by Miller et. al. (Mi 73), and, Dikic et. al. (Di 74), system $^{237}$Np with protons studied by McGee et. al. (Mc 72), with alpha-particles by Wogman et. al. (Wo 66), system $^{233}$U + p - studied by Khan et. al. (Kh 70) and Marshall et. al. (Ma 73), $^{239}$Pu with protons by Saha et. al. (156), and, with alpha particles by Wogman et. al. (Wo 66).

Although, available data are limited compared with those at thermal energy, still all the data indicated thus far show a Gaussian charge-dispersion. Broadening of isotopic charge dispersion curves for cesium isotopes with increasing energy has been observed by Friedlander et. al. (Fr 63), Davies and Yaffe (Da 63) and Tracy et. al. (Tr 72). Umezawa (Um 71) prefers a constant charge dispersion width for his data on mass-148 and mass-160 chain in line with thermal-neutron fission data and that of McHugh and Michel (Mc 68), with no variation with energy and mass number. The shift of the most probable charge, $Z_p$, 
towards stability with increasing proton energy is another feature of the experimental observation as summarized by Yaffe (Ya.69).

Physical methods, such as characteristic X-rays from fission fragments have also been employed with a solid-state detector in coincidence with the kinetic energies, by Glendenin et. al. (Gl 51). Other physical techniques have been also applied to this problem, e.g., Konecny et. al. (Ko 65). These approaches have the advantage that the phenomena observed can be related to the primary fission fragment. On the other hand, as pointed out earlier, radiochemical studies provide certain and accurate data of formation cross sections of each member of a fission-product chain, even though the information is related to the secondary fragment which has already evaporated prompt neutrons. Experimental measurements indicated in this section show that the charge of nuclides for which the independent yields are measurable lie closer to the most probable charge than in the case of low excitation. This gives much higher precision to the observed values in this case. This could be seen from the comprehensive compilation of fractional independent yields observed in various fissile systems at medium excitation energies and experimental Zp values, in the review paper by Umézawa et. al. (Um 71).

On the other hand, much ambiguity begins to appear with respect to the number of emitted neutrons as
At the excitation energy is raised. This is probably responsible for the lack of any general rule governing fissile systems subjected to the range of excitation energy covered in this section. In the plots of \( Z_p \) values versus excitation-energy, Umezawa (Um 71) has shown that the \( Z_p \) values deviate considerably to the lower side in the fission with the excitation-energy below 20 MeV. This paper from quantitative information draws the conclusion that the charge distribution in fission with medium excitations (\( >20 \) MeV) is somewhat different from that in fission with the lower excitations. Hence, Terrell's systematics found for the low-energy fission do not apply in the medium energy fission. Instead, the number of post-scission neutrons, \( v_{\text{post}} \), must be either independent of or linearly dependent on the fragment mass aside from the energy dependence.

**Emission of Neutrons in Medium-Energy Fission.**

An excited nucleus (a compound nucleus or a residual nucleus formed in the direct interaction) can either undergo fission or emit a particle, essentially a neutron. On the basis of residual excitation energy, the nucleus may again undergo a similar competition between fission and neutron evaporation. The fission fragments thus originating from different fissioning nuclei will have a distribution of excitation energies which are then
dissipated by the emission of neutrons and gamma rays to yield the final product of interest. A schematic representation of this simple model is as follows:

\[
\begin{align*}
&[A_f \, (E_1^*)] \xrightarrow{n} [A_f \, (E_2^*)] \xrightarrow{n} [A_f \, (E_3^*)] \xrightarrow{n} \text{etc} \\
&\text{fission} \quad \text{fission} \quad \text{fission}
\end{align*}
\]

In this fission chain the excited nucleus \( A_f \) with excitation energy \( E_1^* \) can either fission in a variety of modes yielding different fragments or emit a neutron to give \( A_f \) with excitation energy \( E_2^* \). Depending on \( E_2^* \), \( A_f \) can again undergo a similar process. The fission branching ratio, \( G = \Gamma_f / (\Gamma_f + \Gamma_n) \), of successive residual nuclei have been denoted by \( G_1, G_2, G_3 \ldots \). At each step of the chain a distribution of fission fragments is obtained with a spectrum of excitation energies. A secondary product will be formed from only those fragments which have at least sufficient excitation energy to emit the desired number of neutrons.

In this energy range, covering a transition from a compound-nucleus mechanism to a direct interaction mechanism of nuclear reaction, the concept of an 'intermediate' nuclei was introduced by Stark et al. (St 71), which include compound-nuclei and, in terms of the two-step
Serber model (Ch 68), those nuclei formed following the first "prompt-cascade" step. The average excitation energy of these 'intermediate nuclei' have been recently calculated by Dikšić (Di 74) with the aid of the Vegas–Monte Carlo STEPNO code of Chen et al. (Ch 68) and Metropolis et al. (Me 58), for the system $^{238}\text{U} + \text{p}$.

The excitation energies of the successive fissioning nuclei could be computed using the concept of nuclear temperature $T$ evaluated from the Vandenbosch and Huizenga (Vo 64, Hu 62) formalism, given by

$$U' = aT^2 - 4T \quad \text{(I-14)}$$

where $U'$ is the effective excitation energy of the residual nucleus, and $a$ is the level-density parameter, equal to $10.5 \text{ MeV}^{-1}$.

After each evaporation step, the excitation energy $U''$ of the residual nucleus is given by the expression

$$U'' = U' - B_n - 2T, \quad \text{where } U' \text{ is the excitation-energy of the preceding nucleus, } B_n \text{ is the binding energy and } 2T \text{ is the kinetic energy of the emitted neutron. Vandenbosch and Huizenga (Hu 62) formalism uses the value of } r_0 \text{ the radius parameter of } 1.2 \text{ fermis, makes the correction for the odd-even nature of the nucleus, and, a pairing energy } \delta$$
is assigned a value of 1.1 MeV.

With all this information the fission branching ratio, \( G \), is computed according to this methodology by using the expression,

\[
G = \frac{\Gamma_n}{\Gamma_f} = \frac{4A^{2/3}a_f(E - B'_n)}{K_0a_n[2a_f^{1/2}(E - E'_f)1/2 - 2a_f^{1/2}(E - E_f')1/2]} \exp \{2a_n^{1/2}(E - B'_n)1/2 - 2a_f^{1/2}(E - E_f')1/2\}
\]

where, \( A \) and \( E \) are the mass and excitation energy of the fissioning nucleus; \( a_f \) and \( a_n \) are the level-density parameters for the fissioning nucleus at the saddle point and residual nucleus after neutron emission respectively. \( E_f' \) and \( B'_n \) are the effective fission threshold and the effective neutron binding energy corrected for the odd-even character of the nucleus. \( K_0 = \hbar^2/(\text{gmr}_o^2) \) where \( g \) is the spin factor 2 for the neutron, \( m \) is the mass of the neutron and \( r_o \) the radius parameter.

Saha and Yaffe (Sa 70) have formulated a procedure to calculate \( Z_p \) values invoking UCD and ECD mechanistic models by a random selection of fragment pairs in the mass region of interest, for each fissioning nucleus. The average kinetic energy, taken from Milton and Fraser (Mi 62), was used in computing the total average excitation energies, \( E^* \), of the two primary fragments produced in a fission event according to the relationship:-
\[ \bar{E}^* = \Delta M(A_f) - \Delta M(A_1) - \Delta M(A_2) - \langle \text{K.E.} \rangle_{f,p} \quad (I-16) \]

where, \( \Delta M(A_f) \) = energy of the fissioning nucleus (sum of the ground-state mass excess plus the excitation energy due to the bombarding energy)

\( \Delta M(A_1), \Delta M(A_2) \) = Mass excesses of the complementary-primary fragments.

\( \langle \text{K.E.} \rangle_{\text{ave}} \) = average total kinetic energy of the two fragments, mentioned above.

The procedure then calls for evaporation of neutrons from fragments according to the relationship stated before, namely,

\[ U'' = U' - (B_n + 2T) \]

and pick only those fragments which end up thus at the most probable mass value in the region of study. \( Z_p \) could then be calculated for each fragment corresponding to each fissioning nucleus, according to the relations corresponding to ECD and UCD mechanism, thus

ECD: 
\[ Z_p = Z_{A_1} - \frac{1}{2}(Z_{A_1} + Z_{A_2} - Z_f) \quad (I-17) \]

UCD: 
\[ Z_p = (Z_f/A_f)A_1 \quad (I-18) \]

The calculated \( Z_p \) values are then weighted by fission branching ratios, \( T_f/(T_f + T_n) \), calculated for different fissioning nuclei by the Huizenga-Vandenbosch formalism explained before; thus,
From the data reported by Yaffe (Ya 69), the UCD postulate is favored due to the weight of evidence in its favor. However, if the gross assumption is made that the heavy fragment receives 1.5 times the excitation energy instead of either sharing of excitation energy between the fragments in proportional to their masses or the heavy fragment acquiring even only half the energy determined in this way, it could alter the situation in favor of the ECD. Therefore, it seems the assumption implicit in the calculations that the fragment excitation energies are proportional to fragment mass is a very sensitive one. Holub (Ya 73) emphasizes the simplistic nature of this assumption and proposes that the peak energy for the formation of a fission product vs N/Z plot, for various fissile targets should be parallel lines and discusses the deviations from these "regular" pattern based on a very qualitative approach.

We shall now turn to the problem of seeing what information can be obtained about neutron emission from a knowledge of the energy variation of $Z_p$ of the fission products for different fissioning systems. McHugh and Michel (Mc 68) have proposed equations relating the rate of change of neutron emission with excitation energy to the rate of change of $Z_p$ with excitation energy. The McHugh
and Michel formalism can be symbolized as follows:

\[
\frac{\partial Z_p}{\partial E} = -\left(\frac{\partial Z_p}{\partial A}\right)_{A} \left(\frac{\partial A}{\partial E}\right)_{E} Z_p
\]  
(I-20)

Since

\[
\frac{\partial Z_p}{\partial E} + \frac{\partial Z_p}{\partial A} = 0
\]  
(I-21)

then

\[
\frac{\partial Z_p}{\partial E} = \frac{\partial Z_p}{\partial A} / \left(\frac{\partial Z_p}{\partial A}\right)_{A} \left(\frac{\partial Z_p}{\partial A}\right)_{E}
\]  
(I-22)

At a given excitation energy the variation of \(Z_p\) with mass number can be approximated by the variation of \(Z_A\) with mass number,

\[
\frac{\partial Z_p}{\partial A} = \frac{\partial Z_A}{\partial A} / 0.38
\]  
(I-23)

Coryell et al. (Co 61) have given \(\frac{dZ_A}{dA} = 0.38\) for all masses. Hence,

\[
\frac{d\nu}{dE} = \frac{\partial Z_p}{\partial E} / 0.38
\]  
(I-24)

From a knowledge of \(\frac{dZ_p}{dE}\) one can easily obtain \(d\nu/dE\) and therefore, the number of neutrons at a given excitation energy. Cheifetz and Fraenkel (Ch 68) directly measured neutron energies in coincidence with the fragment energies by the time-of-flight techniques. The fissile system chosen was \(^{238}\text{U}\) bombarded with \(17\)-MeV protons, where they found \(0.62 \pm 0.25\) neutrons with velocity above
1.0 cm/n sec were emitted before scission, and fission of 209\textsuperscript{Bi} and 238\textsuperscript{U} by 155-MeV protons. Some recent experimental work seem to have confirmed the results of Cheifetz and Fraenkel, and, important generalizations from these works can be summarized as follows:

(i) The number of post-fission neutrons emitted with the 12-MeV and 155-MeV bombardment energies are almost identical.

(ii) The average number of post-fission neutrons as a function of fragment mass was found to increase strongly with increasing fragment mass. The comparison of results at the two energies mentioned above, also indicated that almost all the additional excitation energy is concentrated in the heavy fragment. Such a conclusion could also be based on the graphical relationship, determined from radiochemical data by Yaffe et al. (Ya 69), between $Z_A - Z_p$ and proton bombardment energy and will be discussed later.

The figures (12-14) taken from Cheifetz et al. (Ch 68) show the total number of neutrons, the prefission-neutrons and post-fission neutrons as a function of mass division. Two extreme assumptions used in the calculation are:

(1) Upper limit for post fission neutrons defined by the assumption that all neutrons emitted in the direction of fragment 1 ($\theta = 0^\circ$) were emitted from fragment 1; likewise
The average number of post-fission neutrons as a function of fragment mass. After Cheifetz et al. (Ch 68).

**Explanation**

Max, result for upper limit assumption.
Min, result for lower limit assumption.

The energy spectra are obtained by an iteration procedure, the dashed lines correspond to results from the first iteration of the upper limit and lower limit assumptions. Solid line represents the best estimate of the final result. The statistical error (based only on counting statistics) and mass distribution (dotted line) are also shown.
**MASS RATIO**

**a)** $^{209}\text{Bi} + p$ 155 MeV

**b)** $^{238}\text{U} + p$ 155 MeV

**MASS (A.M.U)**

**ERROR**

**STATISTICAL ERROR**
FIGURE 13 (a), (b)

The average kinetic energy of the post-fission neutrons in the c.m. system of the fragment as a function of the fragment mass. After Cheifetz et al. (Ch 68).
The average number of post-fission neutrons as a function of the fragment mass for events of high total kinetic energy $E_k$ of the two fission fragments and for events with low $E_k$. After Cheifetz et al. (Ch 68).
for fragment 2. The spectra are obtained in the c.m. systems of the two fragments.

(2) A lower limit for post-fission neutrons is defined by the assumption that all neutrons emitted at 90° to the fission fragments are pre-fission neutrons. In order to obtain the post-fission spectrum, a subtraction of the 90° spectrum from the spectra at 0 and 180° and transformation of the results to the c.m. system of the two fragments is required.
(i) **Factors Considered:**

The fission process results in a very complex mixture containing a large number of products in varying yields, each decaying with its characteristic half-life to give a new product. Furthermore, in experiments on the laboratory scale, problems are involved in the separation and characterization of fission products because only minute quantities of fission products are formed.

The chemical methods that are adopted for separation of fission products are in many aspects similar to ordinary analytical separation procedures. To cover the needs of radiochemistry, however, these must be suitably revised by taking into account very important dissimilarities. This revision very often results in separation methods completely different from those used in standard analytical chemistry. The most significant of these differences are perhaps the time factor involved when working with radioactive nuclides especially in case of short-lived one and/or nuclides formed in the fission acts by direct independent formation in the ground state and very short-lived excited states.

A best estimate for optimum separation time of fission product nuclides in question has been derived making use of trial and error separation times and by using
a computer programme called FEDG III. This program calculates, by matrix operations, activities or number of atoms from up to 19 members of a radioactive decay chain from given initial numbers of atoms or rates of formation for each member. The activities or number of atoms may be calculated for up to 100 different evenly-spaced intervals. The original programmes FEDG or FEDG II, were written by G. P. Ford of Los Alamos for the CDG 6600 computer. The two programmes were combined into one and modified for the IBM-360 computer by Wahl et al. (Wa 72).

High yield and chemical purity are two common requirements in general analytical practice. In radiochemical work, however, high yield is of secondary importance provided the exact yield can be evaluated and corrected for when necessary. Chemical purity may or may not be necessary, but, radiochemical purity, i.e., the absence of non-isotopic radioactive impurities, is of very great importance and has to be extremely good. For good radiochemical purity high decontamination factors of $10^3$ to $10^6$ are usually necessary. That the observed activity is really due to the fission product element studied is checked by repeated separations and purifications, after which the half-life, absorption characteristics, and the specific activity (expressed as activity per unit carrier mass) must be unchanged. In all the experimental work, for each of the radioactive species under examination, the decaying
activities are measured for several half-lives, a resolvable decay curve for the nuclide in question derived from a composite decay curve and the half-life value checked with the latest compilation by Wakat (Wa 71). Standard least-squares analysis computer programmes treating the Gamma and Beta decay data, are used to obtain the best value for half-lives of the nuclides under consideration and subsequently compared with the literature values.

Techniques Involved

(a) Carrier Techniques

Radiochemical separations of minute quantities of radioactive elements are aided by the addition of stable isotopes of the elements concerned in relatively appreciable quantities (in the present investigation 10-20 mg). Isotopic carrying is most effective when complete exchange or inter-change is achieved, e.g., when the radioactive atoms participate in any given chemical reaction to the same extent and in the same manner as the stable carrier atoms. Complete exchange usually occurs readily when both radioactive and stable atoms are in the same oxidation state. If they are not, precautions must be taken to ensure complete chemical exchange. Exchange is slow with elements that form colloids readily as in the present work. In these cases, additional operations are necessary to ensure that the radioactive and carrier atoms are in states which exchange
readily, e.g., by addition of complexing agents, by heating to promote coagulation.

Furthermore, the sequence of the addition of reagents has an important influence on the carrying effect of the carriers. When the carrier atoms are present in the solution together with the radioactive atoms in a state of possible exchange, complete exchange will occur; the carrier will be precipitated congruently with the radioactive atoms when the precipitant is added. If the precipitant, however, is present in the solution before the carrier is added (pre-formed) the exchange may be incomplete. Procedures involving preformed precipitates are therefore avoided when possible.

(b) Decontamination

In a mixture of fission products, active species of more than forty elements may be present together. The added carrier is subjected to chemical separations that are specific for the element of interest. Some precipitates, however, especially the gelatinous ones, have a marked tendency to absorb partially or completely and carry down non-isotopic substances, which may even be normally soluble and will thus give rise to contaminated products. The co-precipitation of unwanted radioactive atoms can be minimized by adding soluble carriers, isotopic with the impurities, to act as hold-back carriers. Thus, if the
desired element is unavoidably contaminated with very small amounts of the impurity element, it will be with almost completely stable isotopic forms of the element. The radioactive impurities will remain in solution. In the procedures used in the present investigation, hold-back carriers are only added when interfering contamination from co-precipitation of undesired activities made it necessary.

Normal purification methods may very often result in samples not pure enough in the radiochemical sense. Further purification is often achieved by using scavengers to remove interfering activities before the final separation of the desired fission product element. The scavenger used in the present work in some stages of silver purification was a precipitate of gelatinous nature - Fe(OH)_3 which shows a large tendency to co-precipitate or incorporate significant quantities of various cations when the latter occur in minute quantities.

(c) Chemical Yield

For the quantitative determination of fission product species present in a solution, the separations performed need not be chemically quantitative as long as complete exchange has been attained between the radioactive element and the added isotopic carrier. If this is the case, and the exact amount of carrier added is known, the fraction of the added carrier actually isolated for activity
measurement (chemical yield) will represent the radiochemical yield. The final form of the separated element must therefore be a stable stoichiometric compound suitable for weighing. If the chemical yield is known, the total activity originally present is obtained by multiplying the observed activity by the ratio of the carrier added to carrier recovered and measured.
G. PRESENT WORK

Within the general area of studying nuclear charge distribution in the proton fission of natural uranium, experiments were undertaken to determine isotopic yields of nuclides in the symmetric and near-symmetric mass regions. These results could then supplement fission yield and charge distribution data obtained in the light and heavy mass regions of fission products.

There has been very little experimental data in the symmetric region of fission products, primarily due to very complicated decay schemes, isomerism, and low production cross-sections at medium energy and from thermal-neutron fission. Therefore, this endeavour is an attempt to measure as accurately as possible the independent cross-sections experimentally and estimate total independent cross sections from isomer values available for nuclides providing partial independent yield only. The charge distribution curves thus obtained could be then checked for systematics in the medium-energy proton fission data in other mass region, and compared and also correlated with very high-energy fission data leading to symmetric mass products.

An accurate determination of the method to monitor the proton beam for these experiments was made by absolute cross-section measurements for the reaction $^{65}\text{Cu}(p,pn)^{64}\text{Cu}$.
using a Faraday cup assembly and the external-beam of the McGill synchrocyclotron facility, covering the entire range of incident proton energy used for the present work.
II. EXPERIMENTAL PROCEDURES
A. TARGET ARRANGEMENT

The uranium foil target was of natural isotopic composition (99% $^{238}$U). The thickness of the foil used varied from 170 mg/cm$^2$ to 230 mg/cm$^2$ depending on the nature of the experiment. The degradation in the proton bombardment energy, introduced by the target thickness, was computed at each energy using the data of Williamson et al. (Wi 66) and was found to lie within the ±2 MeV limit of the internal beam energy spread of the McGill synchrocyclotron. The target assembly consisted of the following foils, numbered for representation in Fig. 15 (a) & (b).

No. 1 Natural uranium foil, cleaned in dilute HNO$_3$ to ensure complete dissolution of the oxide coating and subsequent washing and drying with distilled water and acetone respectively.

No. 2 and No. 3 High purity aluminum catcher foils, approximately 10 mg/cm$^2$ thickness sufficient to absorb the recoil products of the fission fragments studied here based on the estimates by Ramamoorthy et al. (Ra 70) and Sugarman (Su 66).

No. 4 The copper monitor foil (> 99.99% pure) of superficial density 44 mg/cm$^2$. Estimates of the range of recoil copper ions, based on range-energy tables of Northcliffe and Schilling (No 70) and information based on Dewanjee et al. (De 68) indicate that the maximum loss
FIGURE 15
Target assembly-foils.
Diagram (a) shows a beam passing through foils 1 and 4, with foils 2 and 3 in between.

Diagram (b) illustrates a beam passing through foils 1, 2, and 3, with an angle of 45° indicated between foils 2 and 3.
of radioactivity due to nuclear recoil from the Cu target, is less than .003%. Consequently, no catcher foils were used in the target assembly with the copper monitor foil.

The target assembly, consisting of the above-mentioned foils with the leading edge of the target inside the catcher foils, as shown in Fig. 16 prevents escape of recoils from the edge of the target. The entire assembly was wrapped with an aluminum foil of identical purity and density and the catcher foil was then fixed on a target holder which in turn was attached to the cyclotron probe for irradiation (see Fig. 16). The monitor foil in the stack was placed up stream from the target, so as to measure a beam which has not been degraded in the heavy target and to avoid possibilities of reactions in the monitor foil induced by secondary neutrons produced in the target (see Fig. 16).

The foil stack was very carefully trimmed on three sides prior to wrapping with Al foil to make sure that identical surface areas of the target and monitor foil were exposed to the cyclotron beam.
FIGURE 16

Target assembly and end of cyclotron probe
Target clamping screw

Aluminum target holder

Target

End of cyclotron probe
B. IRRADIATION

The irradiations were performed in the internal circulating proton beam of the McGill synchrocyclotron. Bombarding energies of 20, 35, 40, 55, 70, and 85 MeV were obtained from insertion of the cyclotron probe at appropriate radial distances according to the data obtained by Portner (Po 64) and shown graphically in Fig. 17. In irradiations involving very rapid chemical separations, the procedure was modified by taking into consideration the following factors:

(i) The maximum exposure of the target to the proton beam was ensured by positioning the target at definite distances in the holder according to the energy variation of the vertical oscillation of the proton beam. The proton beam position of the cyclotron relative to the median plane is graphically illustrated in Fig. 18, based on Skarsgard (Sk 58) and Whitehead (Wh 61).

(ii) The relative thickness of the target foil in these experiments was reduced to a lower limit in order to minimize the dissolution time in mineral acids after irradiation. Consequently, to increase the effective thickness of the target, the target assembly was tilted at an angle during irradiation Fig. 15 (b). The duration of irradiation was a direct function of the formation cross-section of the nuclides of interest, nature of fission yield, and, beam intensity of the cyclotron and varied within a time range of two to twenty-five minutes.
FIGURE 17

Irradiation energy as a function of the radial distance of the cyclotron probe. After Portner (Po 64).
Proton beam position of McGill synchrocyclotron as a function of energy.
C. PROTON BEAM MONITORING

(1) **Introduction**

The cross-section for the reaction $^{65}\text{Cu}(p,pn)^{64}\text{Cu}$ at various bombarding energies using the McGill synchrocyclotron proton beam has been used in the Radiochemistry laboratory to monitor the internal beam for a long time. The absolute value used had been obtained from determination relative to other nuclear reactions and no absolute determinations had been made. The reaction is useful because of the high cross-section for the reaction in this energy region, the ease of target preparation and subsequent chemical purification, and the convenient half-life of the induced activity, 12.8 hours. An excitation function for $^{65}\text{Cu}(p,pn)^{64}\text{Cu}$ based on the work of Meghri (Me 62), has been employed in the past. This work only determines the relative cross-section for the aforementioned nuclear reaction. The possible errors inherent in this and other $^{65}\text{Cu}(p,pn)^{64}\text{Cu}$ excitation functions, largely due to the employment of other excitation functions as references gave the necessary impetus towards redetermination of this excitation function. Absolute measurements were made in association with L. Yaffe, D.A. Newton, and R.B. Moore and have been published (Ne 73). The external proton beam facility was coupled to a Faraday cup assembly for measuring the proton beam intensity in this work. Degradation of the beam by beryllium degraders of appropriate thickness provided proton beams of different incident energies on the
target.

The published measurements of the excitation function for this reaction have previously been made using internal proton beams of cyclotrons, with another known proton monitor excitation function to measure the beam intensity.

The stacked-foil technique has been the most common procedure, and the earliest work reported is that due to Meadows (Me 53) covering the energy range 20 to 100 MeV. The proton beam was monitored by the $^{12}\text{C}(p,pn)^{11}\text{C}$ reaction by Lee Aamodt et al. (Aa 52), who had determined absolute cross-section for this reaction. Mertz and Caretto (Me 62) obtained formation cross-sections by integral recoil studies over the range 73 to 400 MeV by the stacked-foil technique. To monitor the beam they used the $^{27}\text{Al}(p,3pn)^{24}\text{Na}$ excitation function given by Hicks et al. (Hi 56). This was based on an absolute determination of the $^{12}\text{C}(p,pn)^{11}\text{C}$ reaction by Crandall et al. (Cr 56). Cohen et al. (Co 54) used stacked-foils over the 12 to 23 MeV region, using the $^{63}\text{Cu}(p,n)^{63}\text{Zn}$ reaction of Blaser et al. (Bl 51) as a monitor. Yule and Turkevich (Yu 60) also used the $^{27}\text{Al}(p,3pn)^{24}\text{Na}$ reaction as a monitor for work in the 90 to 450 MeV region, but quote their results against the $^{12}\text{C}(p, pn)^{11}\text{C}$ reaction by Crandall et al. (Cr 56). Coleman and Tewes (Co 55) have measured the reaction at 90 to 190 MeV, essentially relating their values to those of Crandall et al. (Cr 56).
(2) **Experimental**

(i) **Targets and Irradiation Procedure**

Two circular discs of copper (> 99.99% pure) of superficial density 44 mg/cm\(^2\) were used as the target material. The targets were aligned and irradiated as shown in an expanded view in Fig. 19.

The cross-sectional view is depicted in Fig. 20. The beam was directed by a closed-circuit T.V. camera through a 0.2 mm thin aluminum window after the necessary degradation to obtain the required incident energy on the target by use of the beryllium degraders. The final focussing magnets of the beam transport system were then finely tuned to produce a beam spot about 5 mm diameter at the center of the screen. The target was placed over the collimator hole with thin adhesive tape (2.5 mg/cm\(^2\)), the Faraday cup was moved very close to the target and irradiation commenced.

(ii) **Measurement of Proton Energy**

The large solid angle subtended by the mouth of the Faraday cup gave very reliable observation of the widely diverging beam which straggled through the aluminum absorbers when the total absorber thickness was near the proton range. The reliability of the measurements is shown by the small scatter of points on a graph of the transmitted beam as a probability function (integral of the Gaussian) of the absorber thickness. (Fig. 21). **Portner and Moore** (Po 65) who made measurements on
FIGURE 19

FIGURE 20

FIGURE 21

PERCENT OF BEAM TRANSMITTED

RANGE = 3153 Mg/cm²
ENERGY = 52.1 MeV

ΔR = 120 Mg/cm²

ABSORBER THICKNESS - Mg/cm²
the external beam facility of the McGill synchrocyclotron, report an empirical relationship between the beam absorption rate as a function of absorber thickness of the aluminum stock, and estimate the accuracy of determination of the mean energy of the proton beam to be better than 0.5% at all energies.

(iii) **Energy Spread in Proton Irradiations**

The graphical representation (Fig. 22) of the work of Portner and Moore (Po 65) indicates the total energy spread of the degraded external beam in the region of irradiations for this experiment. They explain the energy spread at 100 MeV, and, that below 50 MeV as being due to the selective properties of the beam transport system. As a result of this energy spread of the proton beam, there was also a spread in the observed range of protons which was higher than the uncertainty associated with the straggling of the mono-energetic beam. The observed spread in the range was directly correlated to determination of the energy spread (FWHM) of the proton beam.

The data of Williamson et al. (Wi 66) provided the functional relationship between the target thickness and energy degradation of the beam introduced by it. The combined results of the energy-degradation due to thickness of the target and energy spread (FWHM) assumed as a result of range analysis is depicted in Fig. 22. Portner and Moore (Po 65) argue that summation of approximately rectangular bombarding
FIGURE 22

OBSERVED BEAM ENERGY SPREAD (FWHM)

ENERGY SPREAD DUE TO TARGET

ESTIMATED TOTAL ENERGY SPREAD

ENERGY SPREAD - MeV

BOMBARDMENT ENERGY - MeV
energy spread due to the thickness of the target into the approximately Gaussian low-energy spread gives an approximately Gaussian result for the energy variation. This is shown in Fig. 22 to have a small difference with the proton beam energy spread except at low incident proton energies. The cross-sections for these experiments were computed by using the "mean energy" values obtained by subtracting half the incident proton energy degradation through the target from the mean energy of the proton beam incident on the target face.

(iv) Determination of the Integrated Bombardment Current

The specific details are given by Newton et al. (Ne 73). Essentially, the Faraday cup output during the course of the experiment was monitored by an accurately calibrated current electrometer, and the output was integrated by employing a voltage-to-frequency converter coupled to a counting scaler. The integrated beam current was thus obtained from the output of the scaler.

The Faraday cup design was based on Kavanagh et al. (Ka 64) for which an accuracy of ±0.1% was reported due to the very large solid angle subtended by the mouth of the cup at the target. The modified version of the cup used in the present work has a thin front window to allow the observation of targets in air. This front window can cause secondary electrons to be scattered into the cup but use of magnetic suppression causes the current due to these electrons to be
less than 0.5% of the proton current.

(v) Results and Discussion

The final cross-sections at various energies and summary of estimated errors are given in appropriate sections in this dissertation. However, a graph using the present values and those obtained by others is indicated on Fig. 23. The data of Meghir (Me 62), obtained in the internal beam and re-normalized by Saha et al. (Sa 71), are in good agreement with the present results, especially above 40 MeV.

At proton energies between 25 and 40 MeV, where the cross-section is decreasing rapidly, according to Kirkaldy (Ki 54), the energy spread of the internal beam would have the effect of increasing the measured cross-section at a mean irradiation energy, and this probably accounts for the observed discrepancies. The relative cross-sections obtained by other workers have been normalized up to 50 MeV with respect to the absolute values for the $^{12}C(p,p\alpha)^{11}C$ reaction obtained by Cumming (Cu 63). He used a Faraday cup to measure the proton beam in the Brookhaven Linac and corrected for the diffusion of the $^{11}C$ from the polythene targets. Above 60 MeV Cumming used the data of Hintz and Ramsay (Hi 52), normalizing them to his own value at 50.5 MeV.

Yule and Turkevich (Yu 60) and Mertz and Caretto (Me 62) show cross-section data overlapping the high-energy end of the present experiment. Their normalized values are lower than those shown here, probably due to difficulties in precise
TABLE I

Cross sections for the reaction \( ^{65}\text{Cu}(p,\text{pn})^{64}\text{Cu} \)

<table>
<thead>
<tr>
<th>Proton Range ( A^l ) (mg/cm(^2))</th>
<th>Mean Bombardment Energy (MeV)</th>
<th>Cross-section (mb)</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 10320 \pm 10 )</td>
<td>( 102.1 )</td>
<td>( 127.6 )</td>
</tr>
<tr>
<td>( 9490 \pm 10 )</td>
<td>( 97.3 )</td>
<td>( 121.1 )</td>
</tr>
<tr>
<td>( 8260 \pm 10 )</td>
<td>( 89.8 )</td>
<td>( 140.7 )</td>
</tr>
<tr>
<td>( 7280 \pm 10 )</td>
<td>( 83.0 )</td>
<td>( 141.3 )</td>
</tr>
<tr>
<td>( 6580 \pm 10 )</td>
<td>( 78.0 )</td>
<td>( 156.9 )</td>
</tr>
<tr>
<td>( 5830 \pm 10 )</td>
<td>( 73.0 )</td>
<td>( 154.2 )</td>
</tr>
<tr>
<td>( 5060 \pm 10 )</td>
<td>( 67.3 )</td>
<td>( 155.6 )</td>
</tr>
<tr>
<td>( 4400 \pm 10 )</td>
<td>( 62.2 )</td>
<td>( 157.3 )</td>
</tr>
<tr>
<td>( 3735 \pm 5 )</td>
<td>( 56.6 )</td>
<td>( 162.5 )</td>
</tr>
<tr>
<td>( 3153 \pm 5 )</td>
<td>( 51.3 )</td>
<td>( 186.6 )</td>
</tr>
<tr>
<td>( 2542 \pm 5 )</td>
<td>( 45.1 )</td>
<td>( 194.4 )</td>
</tr>
<tr>
<td>( 2140 \pm 5 )</td>
<td>( 40.1 )</td>
<td>( 244.6 )</td>
</tr>
<tr>
<td>( 1703 \pm 5 )</td>
<td>( 35.8 )</td>
<td>( 249.8 )</td>
</tr>
<tr>
<td>( 1240 \pm 10 )</td>
<td>( 29.5 )</td>
<td>( 398.4 )</td>
</tr>
<tr>
<td>( 840 \pm 10 )</td>
<td>( 23.2 )</td>
<td>( 429.1 )</td>
</tr>
</tbody>
</table>

*Measured range of protons before passing through aluminum window (50 mg/cm\(^2\)) or copper target (88 mg/cm\(^2\)). Limits refer to standard error (random) in range determination as defined in text.
FIGURE 23

Absolute cross-section for the reaction $^{65}\text{Cu}(p,\text{pn})^{64}\text{Cu}$ as a function of bombardment energy. From: D. Newton, S. Sarkar, L. Yaffe and R.B. Moore (Ne 73).

Legend

† Present work
□ Meghir normalized values (Me 62)
▲ Coleman and Tewes (Co 55)
○ Meadows (Me 53)
‡ Yule and Turkevich (Yu 60)
× Mertz and Caretto (Me 62)
FIGURE 24

Experimental and theoretical excitation function for the reaction $^{65}\text{Cu}(p, pn)^{64}\text{Cu}$, in the energy range 25-102 MeV.

Legend

- - - Theoretical values (Dostrovsky et al. Do 59)

\[ \text{Theoretical values (Chen et al. Ch 68).} \]

--- This work. (experimental values)
TABLE II

Random and systematic errors in the absolute cross-section for the reaction $^{65}\text{Cu}(p,pn)^{64}\text{Cu}$. Taken from Newton et al. (Ne 73)

<table>
<thead>
<tr>
<th></th>
<th>Random</th>
<th>± %</th>
<th>Systematic</th>
<th>± %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chemical yield</td>
<td>3.0</td>
<td></td>
<td>Number of protons</td>
<td>1.0</td>
</tr>
<tr>
<td>Activity measurements</td>
<td>2.5</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Lifetime estimation</td>
<td>1.5</td>
<td></td>
<td>Standard calibration</td>
<td>1.6</td>
</tr>
<tr>
<td>Pipetting, dilution and weighing</td>
<td>1.0</td>
<td></td>
<td>Counter efficiency</td>
<td>1.5</td>
</tr>
<tr>
<td>Decay curve analysis</td>
<td>1.0</td>
<td></td>
<td>Decay scheme</td>
<td>1.0</td>
</tr>
<tr>
<td>Photopeak areas</td>
<td>3.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Uncertainty</td>
<td>5.3</td>
<td></td>
<td></td>
<td>2.6</td>
</tr>
</tbody>
</table>

Uncertainty = ± 5.9%
energy determination in their low-energy region. The very low energy data of Cohen et al. (Co 54) (12-23 MeV) have not been included. Their data were relative, compared to the $^{63}$Cu(p,pn)$^{63}$Zn studies of Blaser et al. (Bl 51), for which no information regarding absolute values they used could be found. The values of Coleman and Tewes (Co 55) at 90 MeV agree quite well. The values of Meadows (Me 53) are systematically higher than the present data. This is probably due to their poor energy resolution, due to use of the stacked-foil technique, and difficulties in activity measurements. Bell and Skarsgard (Sk 55) who compared the cross-sections for the $^{63}$Cu(p,n)$^{63}$Zn and $^{63}$Cu(p,pn)$^{62}$Cu reactions concluded that the values for the same reactions, reported by Meadows, were 10-15% too high.

Fig. 24 shows a comparison between the present data and those obtained by calculation. The calculations of Dostrovsky et al. (Do 59) agree well at lower energies, but the curve falls off too rapidly. This is not surprising since the calculation took into account only the evaporation mode. In Fig. 22 are shown values at 80 and 102 MeV from the data of Chen et al. (Ch 68). This was a cascade calculation which used a seven-step potential for the nucleus. The data shown are those for STEPNO, involving no refraction or refraction at the surfaces of changing potential. The agreement with the experimental data is better than obtained by the use of the evaporation calculation alone.
D. CHEMICAL PROCEDURES - TECHNIQUES

(1) Preparation and Standardization of Carrier Solutions

The carrier solutions were made up in volumetric flasks to contain about 10 mg per ml. of the carrier element. Stock solutions of silver, palladium, indium, cadmium, cobalt and iron were made using C.P. grade chemicals. Of these only the first four had to be standardized as the others were only used as hold-back carriers or scavengers. All standardizations were done in duplicate and periodically checked.

(2) Filtering, Drying and Weighing

After the end of the irradiation the uranium targets were dissolved in an adequate solvent. Although it is possible to isolate many fission products from a single sample it is usually impractical to do so. In the present work only a single element was isolated from each aliquot of the solution. The techniques of chemical analysis on the semi-micro scale were employed. Operations were performed in 40-ml centrifuge tubes in which all precipitations and dissolutions were made. Precipitates were separated from their supernatant liquids by centrifugation.

The final step was to precipitate the fission product element in a convenient form of known or previously standardized composition. If this product was in a crystalline or other filterable form it was filtered with suction by use of
a Millipore filter assembly consisting of glass fiber 25 μm Millipore filter of suitable pore size placed in a filter holder of fitted glass base. The final precipitate was transferred without loss and collected in a uniform layer (3.5 cm²) on the filter paper, then washed 3 times with 5 ml. of convenient washing liquids. Precipitates that are not easy to filter were centrifuged, washed with suitable liquids, mixed with acetone and transferred to the filter paper, using a transfer pipette.

(3) Mounting of Samples

The final weighed sample, evenly distributed on the filter paper and ready for activity measurement was mounted in the center of a pre-cut standard mounting card 3 inches x 2½ inches using a double-faced scotch-tape mount. The whole assembly was later covered by 1.75 mg/cm² (= 0.5 mil) MYLAR.

The specific details of the above-mentioned experimental techniques were followed in consultation with the pertinent sections elaborated in excellent reference material on Radioisotope Methodology such as, Overman and Clarke (Ov 60), Yaffe (Ya 62), Lavrukhina et al. (La 67), G.D. O'Kelley (Ke 63), Choppin (Ch 61) and Chase et al. (Ch 70).

*obtained from Millipore, Ltd., 55 Montpelier Blvd., Montreal.
E. CHEMICAL PROCEDURES - SPECIAL METHODS FOR THE SEPARATION OF FISSION PRODUCTS IN THIS WORK

Methods for the separation of radioactive fission product elements were worked out by the use of separation methods that are specific for the element of interest. The radioactive purity of the product containing the element to be isolated was examined in the different stages of the procedure, together with the path of the contaminating activities. By comparing different methods and introducing purification methods suitable to remove contaminants, it was possible to choose methods which give the element of interest isolated in a radiochemically pure form within the time limitations.

After the proper irradiation time, a leading edge of the target was sheared off for chemical processing and radiochemical assay. This was dismantled, the monitor foil was used for subsequent copper radiochemistry and the target foil was dissolved in a minimum volume of conc. HNO₃. The aluminum catcher foils and the aluminum sheath covering the target assembly were dissolved in a minimum volume of conc. NaOH and the solution, after total dissolution of the metal foils, was mixed with the hot master acid solution containing the target.

(1) Isolation of Indium Isotopes from a Mixed Fission Product Solution

From a mixed fission product solution containing In⁺³
carrier and Cd$^{+2}$ hold-back carrier, the extraction of InBr$_3$ from 4-5N HBr into isopropyl ether as used by Hudgens et al. (Hu 52) was the primary separation step in the radiochemical isolation of indium in this investigation. Hudgens et al. report 98% indium extraction into two-fold volume of isopropyl ether. The subsequent purification steps are largely those used by Sunderman et al. (Su 59) and, taken partly from the procedure outlined recently by Wa'sson et al. (Wa 67) in a paper dealing with determination of indium in meteoritic samples.

The critical point in the procedure is the separation of indium from cadmium. Tracer experiments with $^{115m}$Cd showed that Cd extracted from 4.5N HBr into a two-fold volume of isopropyl-ether was approx. 9 percent, and, similar experiments of extraction into 2-5 times the volume of ethyl ether by Hagelberg (Ha 65) also indicated that a similar percentage of Cd is lost from the mother solution, which has to be taken into consideration in the yield determination. A step was therefore incorporated prior to the ether extraction step of the above procedure whereby an almost quantitative removal of Cd from indium very rapidly became possible based on Cowan (Co 58). The separation is accomplished by precipitation of indium hydroxide by means of conc. ammonium hydroxide, cadmium remaining in solution as an ammonia complex. Through gross-fission product experiments after an initial separation of nuclides of this mass region, and, from In samples prepared by the chemical procedure outlined above, significant interfering impurities of Sn and Sb nuclides
were observed. Sunderman et al. (Su 57) also describe the decontamination factors for Sn and Sb nuclides in a typical experiment to be quite low using the standard radiochemical procedure outlined with slight modifications above. Moreover, similar half-life values of some of these nuclides necessitated an effective and quick procedure to decontaminate the tin and antimony impurities from indium samples. Recently Erdal and Wahl (Er 71, Er 66) have reported the use of a Cds-cellulose filter bed to absorb preferentially Sn-Sb fission products based on the use by Schüssler, Hermann and Ziegler (Sh 70, Zi 61, Zi 62, Zi 63) of cellulose powder impregnated with cadmium sulfide as an effective adsorbing medium.

Simple tracer experiments have been performed using \( ^{114m}\text{In} \) and \( ^{115}\text{Cd} \) in this work to extend the application of this procedure for clean separation of indium from Sn-Sb fission products. A funnel with a coarse sintered glass filter was used as the filtration assembly with the stem of the funnel leading into a beaker placed inside a bell-jar with a suction outlet and ground glass bottom. The filter-bed was formed by suspending \ (*\text{Cds-cellulose (8\% Cds)} *\) in 5 ml. of 0.6N HCl, and the suspension was saturated with \( \text{H}_2\text{S} \) for 30 seconds. The procedure used for the tracer experiment was as follows: several milliliters of the solution under investigation (always 0.6N in HCl) were saturated with \( \text{H}_2\text{S} \) for 30 seconds. A 1 ml. aliquot of this solution was removed and added dropwise to the filter-bed. The

*obtained from: Brinkmann Instruments (Canada) Ltd., 50 Galaxy Road, Rexdale, Toronto, Ontario. manufactured by: Machery, Nagel and Co., Düren, Germany.
contact time was estimated to be less than 1 second since the drops disappeared immediately. The bed was then washed with 1 ml. of the same type of solution which did not contain the tracer.

The indium (III) in the filtrate and wash was estimated by following the decay of the 1.73-hour $^{113}$In. The amount of the equilibrium activity of $^{113}$Sn($^{113}$In) was subtracted. With $^{114m}$In in 0.6M HCl used in the tracer experiments, the results showed 70% of the indium going into the filtrate on the first step, 25% going in the wash, and 5% remaining on the bed. With $^{115m}$Cd in 0.6N HCl in the tracer experiment procedure, < 0.1% of the tracer was found in the filtrate. When H$_2$S was not used, 85% of the cadmium tracer was found in the filtrate.

From 30% to 50% of the indium and usually less than 0.2% of the tin passed through the CdS-cellulose bed in the Sn$^{113}$/In$^{113}$ procedure. The presence of indium carrier improved the reproducibility of the amount of indium coming through the bed. Almost 98% of In passed through the filter bed when it was washed with warm 6N HCl.

The above procedure yields a very good and rapid separation of Sn and In without employing the intricate pressure filtration apparatus described by Sam and Love (Sa 66). The precipitation of the 8-hydroxyquinoline derivative of indium has been utilized in radiochemical procedures to obtain the indium in a final form suitable for weighing and measure-
FIGURE 25

Radiochemical separation of indium - flowchart.
Indium chemistry
Cd and Pd separations

U target foil + 5ml conc.
HCl (+12N) + 1-2 drops 10%
H₂O₂ (target solution = 10ml)

to dilution and addition of carriers

20mg IN + 20mg Pd + 10mg Cd
Total vol. = 50ml.

Aliquot #1 to Pd chemistry

Addition of excess conc NH₄OH,
aerosol, heat to coagulate
Supernate contains Cd.

Adjustment of pH
Gradual addition of 6N HCl to
dissolve the ppt. & adjust pH (to Bromocresol yellow endpt.)
to selective adsorption

Passage through a suspension of
CdS cellulose in 5ml of 6N HCl
saturated with H₂S.
to transformation to bromide

Saturate the solution with
solid KBr.
to ether phase extraction

25ml. disopropyl ether
saturated with 4-SN HBr
Shake 30 seconds

Ether phase, to combination

Very rapid combination of ether phases

Acid phase to washing
10ml. of 4-SN HBr addition
shake vigorously for 10 secs.
ether phase, to washing

25ml. disopropyl ether
Shake 30 seconds

Ether-phase to combination

Acid phase to milking

20mg In

Wait for
6½ hours

Discard

Acid phase

Discard

Acid phase

Discard

Ether phase

10ml of 6.0N HCl (shake, 30 seconds)
Acid phase, dilute 6 fold
to precipitation

Gradually add 5ml of
HC₂H₃O₂·NaC₂H₃O₂ buffer soln,
and 2ml. of 2-hydroxyquinoline
soln.
Precipitate, to washing

Wash the precipitate with two
portions of 5ml distilled water

To timed extraction (milking of daughter)
ment after addition of sodium acetate-acetic acid buffer based on Cowan (Co 58). A flow-sheet illustrating the above steps is shown in the next page, Fig. 25. In the present work, the first separation of In from Cd typically took place six to eight minutes after end of irradiation of duration, never exceeding ten minutes. The second separation was started approximately six to seven hours later.

(2) Isolation of Silver Isotopes from a Mixed Fission Product Solution

The target foil was dissolved in conc. HNO₃ containing Ag carrier to prevent adsorption losses of active silver, Folger et al. (Fo 49). The separation actually followed the procedure of Glendenin (Gl 54) and was based on silver chloride precipitations alternated by scavenging precipitations. The time of separation of Ag from the precursor Pd nuclides was taken as the time of centrifugation of the silver chloride, first precipitated about two minutes after precipitation. This takes care of exchange between silver chloride and active silver after precipitation, Kjelberg et al. (Kj 61). A detailed study has been made by Hamester and Kahn (Ha 63) of the formation of radiocolloids of silver in solutions of low pH values and adsorptive properties depending on the concentration of HNO₃ solution and the precipitating agent, that is, HCl concentration. Based on this work the master-fission product solution was made = 1N in HNO₃ before 0.1M HCl solution was added for AgCl
precipitation, to minimize adsorption of silver by pyrex and siliceous/organic dust particles.

The contamination of a silver chloride precipitate by thirteen typical tracer activities has been studied by Sunderman (Su 57), and it was determined that 97% I\textsuperscript{131} was carried with the AgCl precipitate and 27% Sb\textsuperscript{124}. The quantitative precipitation of iodine with the silver was expected, as AgI is more insoluble than AgCl. Antimony is difficult to keep in solution during any change in acidity, particularly when a precipitate is being formed, and SbOCl often precipitates with AgCl. The other interferences reported by Sunderman, which stand out are those of the platinum group, iridium and ruthenium. This is shown by the gross-fission product spectrum obtained from the master solution after separation of Sb and iodine activities by Đikšić et al. (Di 74). Silver does not co-precipitate to any great extent with ferric hydroxide in ammonia solution. In the presence of 10 mg. of silver carrier, Sunderman has shown that 91.3% remains in solution. Repeated ferric hydroxide scavenging cycles were therefore necessary for purification of silver, coupled with specific precipitation of silver sulfide from an ammoniacal solution. Glendenin (Gl 54) reports a decontamination factor of more than 10\textsuperscript{6} for a similar procedure. It was found in this work that in spite of the successful implementation of the above-mentioned purification steps, about one to five percent of I\textsuperscript{131} activity was detected.
FIGURE 26

Radiochemical separation of silver isotopes - flowchart.
Radiochemical separation and purification of Ag isotopes from a mixed fission-product solution

Target (uranium) + conc. HNO₃ (2 ml)

Dilute solution to 1N HNO₃

Add 2 ml of 10 mg/ml Ag Carrier + 1 ml 1N HCl + few drops aerosol

Boil with constant stirring with a glass rod

Wash ppt. twice with 10 ml 1N HNO₃ containing 1 drop 2N HCl

Dissolve AgCl ppt. in conc. NH₄OH

Add FeCl₃ + two cycles of Fe(OH)₃ scavengings in the same test tube

Dissolve AgCl ppt. in conc. HNO₃

Add 1 ml saturated (NH₄)₂S solution, stir vigorously and centrifuge

Ag₂S + 2 ml conc. HNO₃
Boil till ppt. dissolves. Dilute to 1N HNO₃ conc.

Add 5 mg I⁻ + 5 mg Na₂S₂O₃ powder.
Boil off I₂ gas.

Add 1 ml 0.1N HCl + AgCl ppt. Wash twice with 1N HNO₃ containing 1 drop 2N HCl

Filter, wash the ppt. with 95% ethanol, acetone and dry under suction
in the purified AgCl sample. Consequently, incorporation of a step involving addition of $I^-$ hold-back carrier and subsequent boiling the solution with $\text{Na}_2\text{S}_2\text{O}_3$ made it feasible to provide purified Ag samples free of $^{131}\text{I}$ activity, suitable for beta counting.

A flow-sheet summarizing the above-mentioned chemical steps is shown in Fig. 26.

(3) Isolation of Palladium Isotopes from a Mixed Fission Product Solution

In the present investigation, after an initial separation of the palladium isotopes radiochemically from the mixed fission-product solution, a milking procedure was followed in order to be able to measure the activity of palladium daughters. This procedure is useful due to the ease of preparation of Ag isotopes with sufficient purity for beta-measurements.

Boswell (Bo 67) has shown that the solvent extraction of palladium acetylacetonate leads to separation very rapidly and efficiently of palladium isotopes from a fission product solution from silver.

The majority of radiochemical separation techniques reported for palladium in the review by Ove Hogdahl (Ho 61) depend on the formation of dimethylglyoxime complex. These methods involve lengthy procedures to remove silver contaminants.

In this procedure acetylacetone acts as a chelating
agent and solvent extraction into benzene is based on the pH dependence of metals extracted as the acetylacetonate. The greatest contaminant of Pd, extracted as the acetylacetonate, is reported by Boswell to be $^{99}$Mo (1.5%). The extraction of molybdenum can be reduced from 1.5 to 0.13% by addition of $\text{H}_2\text{O}_2$ (30%). They also report that the presence of Cl$^-$ ion (concentration = 0.001M) reduces the yield of extraction to only 58%.

The information outlined above along with specific scheduling of timed extractions and milking intervals for separation of the Pd isotopes under investigation appears in the flow-chart illustrated in Fig. 27.
Radiochemical separation of palladium isotopes - flowchart.
Radiochemical separation of Pd-isotopes, especially from silver, in a mixed-fission-product solution

To radiochemical purification of silver

**PD\(^{111}\)M + Ag\(^{111}\)**

1. Acidity & ppt. AgCl
2. Wait 2\(\frac{1}{2}\)-3 days
3. Make solution ammonical
4. Add Ag carrier
5. Three AgCl scavenges

**Initial separation of Pd from Ag**

1. Mixed fission product solution + 10mg Pd\(^{4+2}\) carrier
2. Evaporate to dryness with HNO\(_3\) until Cl\(^{-}\) test is negative
3. Dissolve residue in conc. H\(_2\)SO\(_4\), total vol. = 5ml
4. Add 0.5ml of 30% H\(_2\)O\(_2\)
5. Equilibrate 5ml soln. with equal vol. of 0.1N acetylacetone in benzene by mechanically stirring in 40ml tube
6. Separate phases by centrifugation, wash organic phase with 5 ml. H\(_2\)SO\(_4\) = 30 sec:
7. Acetylacetone phase
8. Back extract Pd by addition of 10ml 6N HCl solution
9. (To Pd\(^{111}\) + Ag\(^{111}\) decay) Aliquot #3
10. Aliquot #2 (To Pd\(^{112}\) + Ag\(^{111}\) decay)
11. Aliquot #1 (To Pd\(^{112}\) + Ag\(^{112}\) decay)
12. Three AgCl scavenges
13. Make solution ammonical
14. Acidify and ppt. AgCl with 0.1N HCl
15. To radiochemical purification of silver

**PD\(^{111}\)G + Ag\(^{111}\)**

1. Acidity &ppt. AgCl with 0.1N HCl
2. Wait 3\(\frac{1}{2}\) hrs.
3. Make solution ammonical
4. Three AgCl scavenges
F. RADIOCHEMISTRY OF COPPER

The radiochemical processing of Cu foils, used for beam monitoring purposes for each irradiation, is based on the procedure of Kraus and Moore (Kr 53), and, subsequent purifications modified from Meinke (Me 49). The flow-chart appears in Fig. 28. For preparation of samples for activity measurement, a glass liquid scintillation measurement vial was heated at 100°C for 2 hr, cooled and weighed to ±0.0001 g. A slurry of the cuprous thiocyanate in ether was added, and the ether was evaporated by gentle warming until there was no further weight loss. The vial was then re-weighed, and the precipitate (usually ≈70 mg) was dissolved in 10.0 ml. of ammonium hydroxide. Finally the vial was sealed.
FIGURE 28

Radiochemical separation of copper - flowchart.
Radiochemical separation of copper

Cu foils + 3M HCl (minimum amount) + few drops H₂O₂ (10% solution). Gentle heating.

Evaporate soln. to dryness. Redissolve with few drops of 4.5M HCl

to ion-exchange separation

Load solution on to 20x0.7 cms ion-exchange column of purified Dowex-X8 anion exchange resin (mesh size 100-200), which has been pre-treated with 4.5M HCl

elution of Cu²⁺ ions

Wash column continuously with 4.5M HCl till yellow Cu²⁺ band is ready for elution. Elute with 1.5 HCl

Collect Cu²⁺ ions after end of elution in a 40 ml. tube. Discard first 10% Cu fraction

to precipitation of Cu²⁺

Add 6M NaOH to get Cu(OH)₂ ppt. Redissolve ppt. in minimum amt. of 1N H₂SO₄

to reduction of Cu²⁺ → Cu⁺

Add two-fold excess 0.1M ferrous ammonium sulphate in 0.1N H₂SO₄

to precipitation of thiocyanate

Add drop-wise 10% NH₄CNS soln., very slight excess

to digestion of the precipitate

Place the tube containing the ppt. in a water bath for at least 20 minutes at 90°C.

to washing & drying

Wash ppt. with dil. soln. of ferrous-ammonium sulphate, 30% ethanol, 95% ethanol, acetone

to transfer of ppt.

Transfer the ppt. in a minimum amt. of isopropyl ether to a previously baked weighing bottle

Drive off ether at 110°C

Dissolve ppt. in 10.0ml. NH₄OH
G. MEASUREMENT OF ACTIVITY

Fission yields of indium, cadmium and some silver members in decay chains studied in this work were determined by making use of the excellent resolving power of Ge(Li) detectors to select the characteristic gamma rays associated with these nuclides. Gordon et al. (Go 66) first recognized the potential of the Ge(Li) detector in fission product studies and reported measurements of many of the larger fission yields using this type of detector. One can routinely determine gamma-ray energies to ±0.5 KeV, so that if the gamma-ray energies from the decay of the fission-product nuclides of interest are known, assignments of the lines in a spectrum to specific products can be made with a high degree of certainty. Current Ge(Li) detectors have sufficiently high resolution that gamma-ray peaks which are only 5-10 KeV apart can be completely resolved, and this allows simultaneous determination of several nuclides in the presence of each other, even when they have similar half-lives. Furthermore, the high resolution of the detector allows a much larger tolerance for impurities and daughter products in the sources than beta-activity measurement permits.

When gamma-ray spectra are measured as a function of time, the decay of the various peak areas with time is generally simple, usually having only a single component and at most 2 or 3 components. The decay properties of the peak
areas establish the half-lives, genetic relationships, and help to identify nuclides. The use of the Ge(Li) detector for quantitative measurements has some important limitations relative to beta-activity measurement. Current Ge(Li) detectors are of relatively low efficiency, having photopeak efficiencies of 0.01-0.1% when measuring 500 KeV gamma-ray activities from sources ≈10 cm from the detector. While larger detectors are continually being produced, it seems unlikely that for reasonable cost one will obtain very much greater efficiencies. This is because of fabrication cost and physical limitations such as size limitations of Ge crystals, incomplete charge collection in large crystals, and limitations on maximum Li drift depth possible. An excellent discussion on these and related topics exist in works such as Camp (Ca 67), Ortec Catalogue (Or 71) and Heath (He 67).

In the present work, statistical considerations made it mandatory to resort to the beta-activity measurements for nuclides such as $^{112}\text{Ag}$, $^{111}\text{Ag}$, $^{111}\text{Pd}$. The formation cross-sections for these nuclides being very small at the energy range of this work-resolution of gamma-decay curves for them was a very difficult task.

Another limitation closely related to the efficiency of the detector is the maximum counting rate that the detector and counting system can record. At the present time, the main limitation in count rate arises from speed limitations of the analog to digital convertor (ADC) and the
memory of the electronic system associated with the detector. However, as the gross counting rate increases, there is a loss in energy resolution due to limitations in the detector and in the electronics other than from the ADC. These limitations can be reduced by using fast amplifiers, base-line restorers, and direct-coupled systems, but even with these devices gain shifts and lower resolution are experienced at high counting rates.

The solid-state detection systems used for the present work were:

(i) 30 cm³ crystal of lithium-drifted germanium (ORTEC model 8001-0536) detector coupled to a 4096-channel pulse-height analyser (Nuclear Data-2200). The detector was encapsulated in a cryostat which was itself plunged into a Dewar filled with liquid nitrogen. The voltage bias of the detector was +2000V. The complete system comprised of a pre-amplifier (ORTEC model 120-2F), a spectroscopy amplifier (ORTEC model 451), and an Analog to Digital convertor unit, part of a Nuclear Data 4096-channel analyser. The whole system was placed in a thick shielding enclosure made out of lead bricks to minimize background from local radioactivity as well as environmental radioactivity mainly from cosmic radiation. The interior of this "lead-cave" was covered with a copper foil and a lucite sheet to reduce and degrade gamma back scattering. A block diagram of the system is shown in Fig. 29. The counting data were recorded on magnetic tape and analysed using the
McGill IBM 360-75 computer. A computer code which subtracts straight line background from "clean" gamma-ray peaks, takes into account overlapping peaks following the method of Bowman (Bo 72) and "smoothes" the gamma-ray spectrum based on Yule (Yu 72) was used.

In activity measurements, the sample in any experiment was placed at such a distance from the detector that dead time was always below 10%. Under these conditions, the "live-time" mode of the system self-corrects for dead-time losses.

The resolution of this set-up was 3.1 KeV (full-width at half maximum) at the 1333-keV $^{60}$CO line.

(ii) $40 \text{ cm}^3$ Ge (Li) Detector (model 8101-0725) and a 120-2B Pre-amplifier

The analyser was a Victoreen Scipp 1600-channel model SD-2P. The resolution of this Ge(Li) detector was about 0.4% (full-width at half-maximum) in the energy range of interest. A scintillation detection system consisting of a 3" x 3" thallium-activated sodium iodide crystal and a RIDL 400-channel analyser were used to measure the activities of the 511 KeV $\gamma$-rays produced by the annihilation of the 0.7 MeV $^+$ particles from $^{64}$Cu, produced by the monitor reaction $^{65}$Cu (p,pn)$^{64}$Cu. A block diagram is shown in Fig. 29. In the set-up used in the laboratory, for greater versatility, two identical crystals and their pre-amplifier could send pulses
FIGURE 29

Block diagram of the detector system used.

(i) Ge(Li) solid state detector.

(ii) NaI scintillation detector.
alternatively to the analyser through a switch. An external cycle-timer could trigger the switch and the analyser.

As indicated in Newton et al. (Ne 73) the live timer of the analyser was found to give results accurate to \( \pm 1.5\% \) of the true count rate when the indicated dead time was below 8\%. As the dead time rose above 8\% the live timer error became significant, reaching 10\% at 23\% indicated dead time. Therefore all calibration and testing data, as well as those portions of decay curves used for calculating \(^{64}\text{Cu}\) activities, using the NaI(Tl) detector, were gathered at or below 8\% indicated dead time.

For low activity measurements, a low-background Beckman beta-counter called "Widebeta II" was used. The detector chamber in this counting system is heavily shielded with high-purity aged lead and copper to minimize the natural background. It operates in anticoincidence with the environmental background and cosmic rays by means of a guard detector. Samples for the counter were prepared on stainless steel planchets which had low background. Background measurements gave values of 1.8-2.0 cpm and the system resolving time was 0.5 microsecond equivalent at 10^6 cpm using live-timing. The stability of the instrument was regularly checked by standard \(^{89}\text{Sr}\), \(^{90}\text{Sr}\), and \(^{90}\text{Y}\) sources. The beta-counting assembly is equipped with an automatic sample changer which has the capacity of one hundred samples. Furthermore it has provision for single cycle, recycle, or single-sample repeat modes of
operation. The print-out is on a Teletype 33 unit, it prints the pre-set date, sample number, time of day, total counts, duration and twice the standard deviation of count. The dead-time loss can be automatically compensated by using the "live-time" mode gate. The features of the Beckman Widebeta II counter can be summarized as:

- Operating voltage (V) : 1775 Volts
- Plateau length (ΔV) : 400
- Detector diameter : 2.25 inches
- Window thickness : 500 (μg/cm²)
- Resolving time : 0.5 μs
- Source mount : stainless steel
- Background rate : 2.0 ± 0.2 cpm
- Counter gas : 90% argon + 10% methane

**Efficiency Calibration**

(i) Solid-state Detection System

The Ge(Li) system must be calibrated for both energy and efficiency. Energy calibration, necessary for identification of various peaks, may be done by counting standards along with the source of interest. Ge(Li) systems generally show a linear relationship between channel number and energy, but if energies are to be measured to better than ±0.5-keV, careful calibration must be done and non-linear effects must be taken into consideration. Absolute efficiency calibration, necessary for quantitative measurement of intensities, must be
done with at least one absolute standard and one or more relative intensity standards. In order to make absolute measurements one must also know the absolute abundance of the gamma-rays from the nuclide being measured. The primary calibrations for both the 30-c.c. and 40-c.c. detectors were done at a distance large enough to reduce errors due to source geometry and summing effect to a negligible level (<1%). All calibration measurements were carried out using a 0.375 "thick plastic absorber in case of the 30-cm$^3$ Ge(Li) and $\frac{1}{2}$" plastic absorber in case of 40-cm$^3$, placed immediately in front of the detector to prevent $\beta^-$-$\gamma$ summing. The IAEA standards used were point sources and cross checking to other standards was done wherever possible. The shape of the low-energy portion of the efficiency curve has been determined using $^{182}$Ta and $^{133}$Ba point sources. The relative intensities of $^{182}$Ta gamma-rays have been reported by White et al. (Wh 70) and have been recently revised by Jardine et al. (Ja 71). M. Fowler (Fo 72) has carried out extensive experimentation pertaining to various aspects of the 30-c.c. Ge(Li) calibration in our laboratory. To eliminate to a large extent the errors due to,

(i) Source-geometry effects

(ii) True coincidence events like $\beta^-$-$\gamma$ summation, and cascading $\gamma$-rays

(iii) Random coincidences, e.g., pile-up spectrum because of finite resolving time of the electronic system allowing over-lapping of two events in time,
Heath (He 67) suggests measurement of activity to be performed at a small solid angle, less than approximately that corresponding to 10 cm distance from the detector used in this work and that true coincidence events like β-γ summation could be eliminated by using plastic absorbers. M. Fowler (Fo 72) has shown that if measurement of activity is performed at a distance of 106.5 mm (source-to-detector can) defined by him as "standard-shelf" using the plastic absorber, the efficiency of the detector can be accurately known for both extended or point sources. The recommendation is made by Fowler in his study that estimation of efficiency close to the detector be made relative only to the efficiency on the "standard shelf". By doing this, one may avoid problems with summing, source geometry, lack of absorber, and so on.

A subroutine for the least-squares programme, ORGLS, (described elsewhere in this dissertation) has been written to allow calculation of the relative efficiency coefficient from measurement data on two shelves, even if the source is decaying appreciably during the measurements. Thus, the activities for samples of nuclides with low emission rates, measured on shelves closer to the detector, could be converted to equivalent rates on the standard shelf by the use of empirically-determined shelf-ratios for the nuclide in question. A listing of these shelf-ratios for the various nuclides in this study appears in Table III for both the 30-cm³ Ge(Li) and 40-cm³ Ge(Li) systems. The graphs for photopeak efficiency as
TABLE III

Relative absolute efficiencies for gamma-lines of spread solid sources used in this work

| Gamma-line observed (KeV) | Nuclide | Detector | Experimental shelf distance (can to shelf) | Standard shelf distance (can to shelf) | Best value for the "shelf-ratio"
|--------------------------|---------|----------|---------------------------------------------|----------------------------------------|-----------------------------
| 158.6                    | 117In   | 30 cm$^3$ Ge(Li) | 32 mm                                      | 108 mm                                 | 5.4 ± .3                   |
| 336.3                    | 115In   | 30 cm$^3$ Ge(Li) | 32 mm                                      | 108 mm                                 | 5.3 ± .5                   |
| 527.9                    | 115Cd   | 30 cm$^3$ Ge(Li) | 32 mm                                      | 108 mm                                 | 5.6 ± .2                   |
| 298.2                    | 113Ag   | 30 cm$^3$ Ge(Li) | 32 mm                                      | 108 mm                                 | 5.9 ± .3                   |
| 229.7                    | 115Ag   | 30 cm$^3$ Ge(Li) | 32 mm                                      | 108 mm                                 | 5.3 ± .4                   |
| 354.0                    | 111Ag   | 40 cm$^3$ Ge(Li) | 5.4 mm                                     | 108 mm                                 | 20.8 ± .2                  |
| 617.0                    | 112Ag   | 40 cm$^3$ Ge(Li) | 5.4 mm                                     | 108 mm                                 | 19.9 ± 0.1                 |
| 511.0                    | 64Cu    | 30 cm$^3$ Ge(Li) | 12 mm                                      | 108 mm                                 | 15.0 ± 0.3                 |
| 511.0                    | 64Cu    | 30 cm$^3$ Ge(Li) | 20.7 mm                                    | 108 mm                                 | 10.4 ± .02                 |
| 511.0                    | 64Cu    | NaI(Tl)           | (10 mm)                                    | (10 mm)                                | 1.0                        |
|                          |         | NaI(Tl)           | (15 mm)                                    | 10 mm                                  | 1.7                        |
|                          |         | NaI(Tl)           | (20 mm)                                    | 10 mm                                  | 2.8                        |
|                          |         | NaI(Tl)           | (25 mm)                                    | 10 mm                                  | 4.1                        |
a function of gamma-ray energy on "standard shelves" appear in Figs. 30 and 31. The latter data are taken from Kanelo (Ka 72). Some samples, due to low activity, had to be measured on the shelf nearest to the 40-cm$^3$ Ge(Li) detector and with a $\frac{1}{4}$" plastic absorber in place. It was not possible to interpose an absorber of the standard 3/8" thickness while measuring activity on the shelf closest to the 40-cm$^3$ Ge(Li) detector. In such cases, a correction is applied as determined from the graph (Fig. 32) depicting the relationship between fraction absorbed versus energy of gamma rays. The basis for the diagram is from the work by Fowler (Fo 72) with the 30-cm$^3$ Ge(Li) detector employing a 3/8" plastic absorber. Absorption is considered small, and one assumes that the fraction absorbed varies linearly with absorber thickness leading to a correlation between the fraction absorbed and the energy of the $\gamma$-ray (Fig.32) for the $\frac{1}{4}$" plastic absorber.

Activity measurements of some of the $^{64}$Cu - $^{61}$Cu monitor samples were generally determined on the "standard-shelf" of the 30-cm$^3$ Ge(Li) detector. These samples were sandwiched between two 0.02 inch thick aluminum absorbers to ensure annihilation of the positrons in a well defined geometry. The measured activities were corrected for the absorption of the 511-KeV gamma rays by the aluminum absorbers (1.3%). The NaI(Tl) detection system was calibrated using a liquid $^{22}$Na standard obtained from the New England Nuclear Corporation. The sample, in liquid form, was measured under conditions
Absolute photopeak efficiency of 30 c.c. Ge(Li) - measured in shelf 6 or the "standard" shelf. The plotted data obtained from Fowler (Fo 72a). (Data shown are IAEA standards and $^{182}$Ta data normalized to the IAEA data).
FIGURE 31

Absolute photopeak efficiency of 40 c.c. Ge(Li).
After Kantelo (Ka 72).

Legend
- Absolute standards with $(\epsilon_{\text{obs}} - \epsilon_{\text{calc}}) < 1.0\sigma_{\text{obs}}$
- Absolute standards with $1.0\sigma_{\text{obs}} < (\epsilon_{\text{obs}} - \epsilon_{\text{calc}}) < 2.0\sigma_{\text{obs}}$
- Calculated efficiency
Fraction of the gamma-ray absorbed as a function of energy of the $\gamma$-ray. After Kantelo (Ka 72).
identical to those for the $^{64}\text{Cu}$ samples prepared for monitoring the proton beam. (The maximum energies of the positrons are not very different). The contribution of the 1.27-MeV $\gamma$-radiation to the 511-KeV peak by pair production was assumed to be negligible.

(ii) Calibration of the Widebeta Detector for $E_{\beta_{\text{max}}} = 1.1$-MeV Transition

Bayhurst and Prestwood (Ba 59) have reported a method to correlate the average energy of beta particles and counting efficiency ($\epsilon$) for a given measurement and mounting set-up, and a modified version as given by Corn et al. (Co 71) of this procedure, was applied in the present work.

A standard solution of $^{210}\text{Pb} - ^{210}\text{Bi}$ from New England Nuclear (Boston, Mass., USA) was used to make duplicate samples of PbCrO$_4$ precipitating it by addition of CrO$_4^{2-}$ ions to the standard solution containing Pb carrier. The source prepared was of specific thickness within 1 mg/cm$^2$ of the specific thickness of $^{111}\text{Ag}$ samples used for activity measurement on the Widebeta counter, and were measured under identical conditions.

The nuclide $^{111}\text{Ag}$ has $E_{\beta_{\text{max}}} = 1.06$-MeV for the "allowed" transition to its daughter, and the standard spread source of $^{210}\text{Pb}$ for its transition to $^{210}\text{Po}$ has $E_{\beta_{\text{max}}} = 1.1$ MeV for a "normal" first forbidden transition. Therefore, it was determined from the National Bureau of Standards Table of Spectra (Na 53) that it has the same beta energy "shape factor" as the $^{111}\text{Ag}$ nuclide. Consequently, it was safe to assume
that the standard $^{210}_{\text{Pb}} - ^{210}_{\text{Bi}}$ source was identical in all respects. $\varepsilon_{20}$ is the value of calculated efficiency for a 20-mg sample, samples were 0.75-inch in diameter covered with $\frac{1}{4}$-mil Mylar ($\approx 0.9$ mg/cm$^2$) and mounted on glass-fiber filter paper of appropriate size on two sided 3M transparent tape (No. 666) on a backing of 2-4" diameter stainless steel planchets of 0.025" thickness. Fig. 33 is a composite curve showing the variation of $\varepsilon_{20}$ with mean beta-energies $\bar{E}$ for systems at Washington University, and here at McGill. Mean beta energies can be estimated from the maximum beta energy ($E_{\text{max}}$) values reported in the literature and the calculated ratios ($\bar{E}/E_{\text{max}}$) for various types of beta spectra compiled by Widman et al. (Wi 68). The graph shows the values of $\varepsilon_{20}$ for $^{111}_{\text{Ag}}$ as $46.3 \pm 2.0$ and $47.2 \pm 1.8$, and for the hard-betas of $^{112}_{\text{Ag}}$ ($E_{\text{max}}^\beta = 4.0$ MeV) a value of $57.2 \pm 3.0$ was obtained by cross-calibration with the 617-keV $\gamma$-line of the nuclide measured on the 40-cm$^3$ Ge(Li) detector, with the sample covered by aluminum foil of thickness 80Q mg/cm$^2$. 
Calculated absolute efficiencies ($e_{20}$) for beta-standards as a function of mean beta-energies.

Legend

Present work ▲

M. Fowler (Fo 72a) ■
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Mode of decay</th>
<th>Half-life</th>
<th>Radiation detected</th>
<th>Emitted radiation per disintegration</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{117m}$In</td>
<td>$46%$ I.T.; $54%$ $\beta^-$</td>
<td>1.94 hrs.</td>
<td>158.6 KeV $\gamma$ from daughter</td>
<td>0.87</td>
</tr>
<tr>
<td>$^{117g}$In</td>
<td>$\beta^-$ (100%)</td>
<td>44 min.</td>
<td>158.6 KeV $\gamma$ from daughter</td>
<td>0.87</td>
</tr>
<tr>
<td>$^{117m+g}$Cd</td>
<td>$\beta^-$ (100%)</td>
<td>3.4 and 2.4 hrs. respectively</td>
<td>daughter</td>
<td>0.87</td>
</tr>
<tr>
<td>$^{115}$Ag</td>
<td>$\beta^-$ (100%)</td>
<td>21 min.</td>
<td>229.7 KeV $\gamma$</td>
<td>1.00</td>
</tr>
<tr>
<td>$^{115m}$Cd</td>
<td>$\beta^-$ (100%)</td>
<td>43 d</td>
<td>336.3 KeV $\gamma$ (daughter)</td>
<td>0.45</td>
</tr>
<tr>
<td>$^{115g}$Cd</td>
<td>$\beta^-$ (100%)</td>
<td>2.3 d</td>
<td>527.9 KeV $\gamma$</td>
<td>0.275</td>
</tr>
<tr>
<td>$^{115m}$In</td>
<td>$95%$ I.T.; $5%$ $\beta^-$</td>
<td>4.5 hrs.</td>
<td>336.3 KeV $\gamma$</td>
<td>1.00</td>
</tr>
<tr>
<td>$^{113}$Ag</td>
<td>$\beta^-$ (100%)</td>
<td>5.37 hrs.</td>
<td>298.6 KeV $\gamma$</td>
<td>1.00</td>
</tr>
<tr>
<td>$^{112}$Pd</td>
<td>$\beta^-$ (100%)</td>
<td>21.0 hrs.</td>
<td>daughter</td>
<td></td>
</tr>
<tr>
<td>$^{112}$Ag</td>
<td>$\beta^-$ (100%)</td>
<td>3.2 hrs.</td>
<td>$\beta^-$</td>
<td>1.00</td>
</tr>
<tr>
<td>$^{111m}$Pd</td>
<td>$71%$ I.T.; $29%$ $\beta^-$</td>
<td>5.3 hrs.</td>
<td>daughter</td>
<td></td>
</tr>
<tr>
<td>$^{111g}$Pd</td>
<td>100% $\beta^-$</td>
<td>22 min.</td>
<td>daughter</td>
<td></td>
</tr>
<tr>
<td>$^{111}$Ag</td>
<td>100% $\beta^-$</td>
<td>7.5 days</td>
<td>$\beta^-$</td>
<td>1.00</td>
</tr>
</tbody>
</table>
III. TREATMENT OF DATA
A. ANALYSIS OF SPECTRA

In the gamma-decay data analysis, the evaluation of the area under the photopeak, which is representative of the disintegration rate, requires an accurate determination of the contribution made by the Compton background to the photopeak area. This Compton background was treated in one of several ways contingent upon (1) the intensity of the photopeak and amount of statistical fluctuation in the neighbouring background, (2) the slope of the Compton background on each side of the photopeak, and (3) the presence of peaks adjacent to the peak of interest. The procedures adopted in this work fall under the following categories commensurate with the above mentioned cases:

Procedure No. 1 - Large number of channels covered by the photopeak, for example, NaI(Tl) peak area computation.

A constant fraction of the photopeak is used for area determination, and for that purpose the method developed by Covell (Co 59) has been used. The procedure is as follows: measure n channels on each side of the channel \( a_0 \) which has the largest number of counts in the photopeak. Add the contents of these 2n+1 channels together and subtract \( (a_{-n} + a_n) (2n+1)/2 \). The top part of the photopeak which has been cut off has an area which is proportional to the full area. Two major advantages inherent in this procedure are that the subjective measurements
of area, or stripping of partly unknown $\gamma$-spectra, are avoided, and that the dividing line through the photopeak changes its slope according to the background of other gamma-radiation present. Tests and theoretical justification have been presented by Covell.

However, Menon (Me 67) and Yule (Yu 69) report that use of the above procedure for Ge(Li) detector data results in inaccuracies at moderate or high counting rates. In the present work, use of Covell's method for peak area determination using the Ge(Li) detector data indicate that peak areas are too low at high or moderate counting rates resulting in inaccurate decay curves. Therefore, only NaI(Tl) detector activity measurement data have been treated in this manner.

Procedure No. 2 - Peak area computation of Ge(Li) detector data with considerable interference from several nuclides.

A computer programme which searches for any and all peaks in the spectrum based on Yule's (Yu 69, Yu 72) first-derivative method has been used. This employs the simplified least-squares data convolution technique to generate, from the observed spectrum, a new spectrum which has essentially all statistical scatter removed, and has been used in case of few gamma-decay data with a large amount of interference. In this programme, peak boundary channels are located by studying the behavior of a smoothed spectrum and a smoothed first-derivative spectrum, each formed from the original spectrum. Net peak
areas are computed from that portion of the spectrum enclosed by the peak boundary channels, overcoming changes in the peak shape due to resolution losses and to other causes.

Procedure No. 3 - Peak area computation from Ge(Li) detector data, without interference from various nuclides.

This procedure was used for the majority of the data in this work. The area under the photopeak was determined by a simple computer programme which finds the area under a Gaussian peak superimposed on a slowly varying background. A five-channel smoothing of the background as described by Bevington (Be 69) is used in this computer code which utilizes a simple summation of the spectrum above a straight line background.

Procedure No. 4 - Analysis of a doublet from Ge(Li) detector data, without recourse to a computer code.

This procedure was adopted for analysis of overlapping peaks of data obtained through the use of the Monroe printer coupled to the 40-c.c. Ge(Li) detector-analyser system. Since computer analysis is not possible in dealing with this form of output the doublet peaks were hand-drawn and, for boundaries which are common to neighbouring peaks, e.g. channel C, Y(c) is proportioned between the neighbouring peaks according to the relative magnitude of Y(c-1)-Y(c) and Y(C+1)-Y(c).

This procedure is based on Bowman (Bo 70).
The photopeak areas as computed above were then corrected for efficiency of the detection system, source-detector geometry, dead-time loss, chemical yield, and dilution factor according to the following relation, to give the absolute photon emission rates, \( N \):

\[
N = A_p \times \frac{100}{(100-D.L.)} \times \frac{1}{E_p} \times \frac{100}{C.Y.} \times F
\] (III.1)

where

- \( A_p \) = photopeak area,
- \( D.L. \) = dead-time loss in percent,
- \( E_p \) = efficiency for a particular source position,
- \( C.Y. \) = chemical yield of the nuclide of interest in percent,
- \( F \) = dilution factor.

The absolute photon emission rate, \( N_0 \), at the end of bombardment was obtained by computer analysis of the decay curves.
B. GENERAL EQUATION FOR CROSS-SECTION CALCULATION

Once the disintegration rate of a particular nuclide at the end of bombardment was known, the formation cross-section of that nuclide could be calculated according to the following expression:

$$D_A^o = \ln T \sigma_A (1 - e^{-\lambda_A t})$$  \hspace{1cm} (III.2)

where

- $D_A^o$ = disintegration rate/second of the nuclide $A$ at the end of bombardment,
- $\sigma_A$ = formation cross-section of the nuclide $A$,
- $I$ = proton intensity as the number of protons per second
- $n_T$ = number of target nuclei per cm$^2$,
- $\lambda_A$ = decay constant of the nuclide (sec$^{-1}$)
- $t$ = period of bombardment in seconds.

The proton beam was monitored by the $^{65}$Cu(p, pn)$^{64}$Cu reaction, the absolute cross-section which was determined in this work (Ne 73). Since for each irradiation copper monitor foil was bombarded under similar conditions as the target itself, then the following relation holds true for the monitor also.
where the subscript $M$ refers to the monitor, and the other symbols are as previously defined. From (III.2) and (III.3), by division and rearrangement,

$$D_M^o = \ln M \sigma_M (1-e^{-\lambda M t})$$

(III.3)

$$\frac{\sigma_A}{\sigma_M} = \frac{n_M}{n_T} \frac{D_A^o}{D_M^o} \frac{(1-e^{-\lambda M t})}{(1-e^{-\lambda A t})}$$

(III.4)

If the weight of the target and the monitor are known, the numbers of atoms can be calculated as

$$n = \frac{W}{A.W.} \times 6.02 \times 10^{23}$$

where $W$ is the weight and $A.W.$ is the atomic weight of the target or the monitor.

Also, taking N.A. (natural abundance) into account, we have finally the cross-section expression for a particular nuclide $A$ in terms of $M$ (monitor) and $T$ (target) variables as follows:

$$\sigma_A = \sigma_M \frac{N.A_M}{N.A_T} \frac{A.W_T}{A.W_M} \frac{W_T}{W_M} \frac{D_A^o}{D_M^o} \frac{(1-e^{-\lambda M t})}{(1-e^{-\lambda A t})}$$

(III.5)
C. RESOLUTION OF RADIOACTIVE DECAY DATA

Extensive use has been made of computer-aided analysis for interpretation of the experimental data. All computer calculations were carried out using the McGill University IBM-360 computer.

Decay data which were obtained using gas-flow beta counters were resolved into the various components through the use of the Brookhaven National Laboratory least-squares programme (CLSQ) written originally by J.B. Cumming (Cu 63), and modified for use on the IBM-360 by B.R. Erdal (Er 66).

The programme uses a least-squares analysis procedure to compute the activity of each component at some arbitrary initial time, and allows up to ten different components to be resolved. The observed counting rate, \( A_t \), at some time, \( t \), is the sum of the contributions of each of the \( m \) components (\( m \leq 10 \)):

\[
A_t = \sum_{j=1}^{m} [I_j \exp (\lambda_j t) + V(t)]
\]  

(III.6)

The symbol \( I_j \) represents the initial activity due to the component which is associated with the decay constant \( \lambda_j \). The symbol \( V(t) \) represents the residual at time \( t \) due to statistical fluctuations and experimental errors. For \( n \) observations of \( A_t \), there are \( n \) equations of the above form. The initial activity of each component enters the \( n \) equations linearly so that a least-
squares solution for the $I_j$'s can be obtained. The solution is obtained by minimizing the sum of the squares of the properly weighted residuals. The CLSQ programme also has options to allow subtraction of up to fifteen components of known $I_j$ before the least-squares analysis, and to allow the experimental data and components to be plotted using a Calcomp digital plotter. For gamma-decay analysis, two least-squares programmes were used:

(i) **Oak Ridge General Least-Squares Programme**

This programme was originally devised by Busing and Levy (Bu 62), modified for use on the IBM-360 by B. R. Erdal (Er 66), updated and modified for more rapid execution adaptable to the McGill IBM 360-75 by M. Fowler (Fo 72). This non-linear least-squares programme fits experimental data to any functional form. One may have 200 data points described by up to 20 parameters, any 10 of which can be iterated for best fit of the data.

(ii) **Exponent Programme**

This is a linear weighted least-squares computer programme computed and adapted to McGill IBM 360-75 by Newton (Ne 71) based on the mathematical method of von Holdt (Ho 59) and was used primarily for analysis of the copper monitor data obtained for activity measurements with the NaI(Tl) detector.
D. SPECIAL EQUATIONS USED IN COMPUTATION OF FISSION YIELDS

The fission yield of a nuclide relative to the monitor can be calculated when the respective absolute disintegration rates and the chemical yields are known. Furthermore, the genetic relationships of the nuclide and half-lives must be known. An exact timing of the different phases of the experiments is of course necessary.

The general equations will be developed in this section which will be modified to bring them in accord with the different experimental conditions.

The rate, \( R \), at which fission takes place will depend on the proton intensity \( I \), the fission cross-section \( \sigma_F \), and the number of atoms \( N \) of the fissioning nuclide present.

That is, \[ R = \sigma_F I N \] (III.7)

During a bombardment, the number of protons per second remains relatively constant and the fission rate will therefore be a constant for a given experiment. As the fission products have two modes of formation, independent and through decay of precursors, the cumulative yield of a nuclide in fission is given by the sum of the independent yield of the particular nuclide and the total yield of its immediate precursors or:

\[ Y = \Sigma(Y_i) \] (III.8)
The production of the nuclide U can be illustrated in the following way:

\[
R \rightarrow S \rightarrow T \rightarrow U \rightarrow \]
\[
S \rightarrow T \rightarrow U \rightarrow \quad \text{Through decay} \quad (\text{III.9})
\]
\[
T \rightarrow U \rightarrow
\]
\[
U \rightarrow \quad \text{Independent}
\]

Hence, the cumulative fission yield of U:

\[
Y_U = (Y_i)_R + (Y_i)_S + (Y_i)_T + (Y_i)_U \quad (\text{III.10})
\]

or

\[
Y_U = Y_T + (Y_i)_U \quad (\text{III.11})
\]

Equations representing each process in (III.9) as a function of irradiation time must be developed. The formation of U will then be given by the sum of these equations, weighted according to the independent yields of the first member of the different chains. The absolute disintegration rate of U at any time subsequent to the irradiation will be a complicated function of the half-lives of the species involved, their independent yield, the irradiation time, and the decay time. The classical mathematical analysis for successive radioactive transformations was developed by Bateman (Ba 20), modified by Rubinson (Ru 49), and further updated by Ford (Fo 72) and will form the basis for the present calculations. Fortunately, in most of the fission
product decay chains, the half-life of a nuclide is much longer than that of its grandparent. It can be easily proved that in such cases it will be entirely sufficient in the calculation of the disintegration rate of a nuclide as a function of irradiation time and subsequent decay time to make the following simplified picture:

\[ A \rightarrow B \] (III.12)

The yield of B will be given by:

\[ Y_B = Y_B + (Y_i)_B \] (III.13)

where \( Y_A \) is the sum of the independent yields of the very short precursors of A plus the independent yield of A, i.e. \( Y_A \) is the cumulative yield of A. When A or B shows nuclear isomerism it may be necessary to take into account three members of the chain (the isomer included). Very often, however, one of the isomeric states has a half-life short compared to the rest of the half-lives of interest. Then the problem simplifies to (III.13). From the laws of radioactivity the number of atoms of independently-formed nuclides of B during an irradiation time T is:

\[ N_B^i = R \left( \frac{(Y_i)_B}{\lambda_B} \right) \left( 1 - e^{-\lambda_B T} \right) \] (III.14)
and the corresponding disintegration rate:

\[ D_B^i = \lambda_B N_B = R(Y_i)_B (1 - e^{-\lambda_B T}) \]

Similarly, one calculates the disintegration rate of the amount of B formed during fission from decay of A:

\[ D_B^A = \lambda_B N_B = RY_A [1 + \frac{\lambda_B - \lambda_A}{\lambda_A} \lambda_B T] \]

After the end of the irradiation these amounts decay exponentially:

\[ (D_B^i)_t = (D_B^i) e^{-\lambda_B t} \]

and

\[ (D_B^A)_t = (D_B^A) e^{-\lambda_B t} \]

At the same time B continues to grow from A. The disintegration rate of the amount grown subsequent to the irradiation is given by:

\[ (D_B^A)_t = R \frac{\lambda_B \lambda_A}{\lambda_A + \lambda_B} (1 - e^{-\lambda_A T}) (e^{-\lambda_B t} - e^{-\lambda_A t}) \]

The total disintegration rate of the amount of B present after an irradiation time T and a decay time t is:

\[ D_B = (D_B^i)e^{-\lambda_B t} + (D_B^A)e^{-\lambda_B t} + (D_B^A)_t \]
or, \[ \dot{D}_B = R \left\{ (Y_i)_B (1-e^{-\lambda_B t}) e^{-\lambda_B t} + Y_A \left[ (1 + \frac{\lambda_B e^{-\lambda_A t} - \lambda_A e^{-\lambda_B t}}{\lambda_A - \lambda_B}) e^{-\lambda_B t} + \frac{\lambda_B}{\lambda_A - \lambda_B} (1-e^{-\lambda_A t}) (e^{-\lambda_B t} - e^{-\lambda_A t}) \right] \right\} \] (III.21)

Often it is more convenient to use the abridged form:

\[ D_B = R \frac{Y_A}{\lambda_A - \lambda_B} \left[ \lambda_A (1-e^{-\lambda_B T}) e^{-\lambda_B t} - \lambda_B (1-e^{-\lambda_A T}) \right] e^{-\lambda_A t} + R [(Y_i)_B (1-e^{-\lambda_B T}) e^{-\lambda_B t}] \] (III.22)

where \( D_B \) = disintegration rate of B formed from A during and after bombardment.

The resolution of parent-daughter mixtures and computation of disintegration rates is elaborately discussed for various cases by Buchanan (Bu 65) and the following important categories are covered by decay equations to be prescribed specifically for the radionuclides under investigation in this work.

Case: \( \lambda_A >> \lambda_B \)

When the decay time \( t \) is large compared to the half-life of A this nuclide will have decayed away and (III.22) reduces to:

\[ D_B = R[Y_A + (Y_i)_B] (1-e^{-\lambda_B T}) e^{-\lambda_B t} \] (III.23)

or, using (III.13)
The activity at saturation of bombardment is

$$D_B = R Y_B (1 - e^{-\lambda_B T}) e^{-\lambda_B t}$$  \hspace{1cm} (III.24)

where $Y_B$ is the saturation factor.

Case: $\lambda_A > \lambda_B$

In this case the experiment can be designed in such a way that the nuclide $A$ will have decayed away before the isolation of nuclide $B$. Under such conditions (III.22) gives:

$$D_B = R \frac{\lambda_A}{\lambda_A - \lambda_B} (1 - e^{-\lambda_B T}) e^{-\lambda_B t} \left[ Y_A + (Y_B) - \frac{\lambda_B}{\lambda_A} (Y_B) \right]$$  \hspace{1cm} (III.26)

or

$$D_B = R Y_B \frac{\lambda_A}{\lambda_A - \lambda_B} \left( 1 - \frac{\lambda_B}{\lambda_A} f \right) (1 - e^{-\lambda_B T}) e^{-\lambda_B t}$$  \hspace{1cm} (III.27)

Here

$$\frac{\lambda_A}{\lambda_A - \lambda_B} \left( 1 - \frac{\lambda_B}{\lambda_A} f \right)$$  \hspace{1cm} (III.28)

where $f = \frac{(Y_B)}{Y_B}$ is the fractional increase in the chain
yield in going from A to B, measured relative to the yield at B. This factor is always less than unity, and for the last members of the chain, i.e., two or more charge units away from the most probable primary charge, $Z_p$, $f$ is of the order of $10^{-2}$ or less.

If $\lambda_B/\lambda_A < \text{unity} \ (10^{-1} - 10^{-2})$, the expression in parenthesis in the parent correction factor is very close to unity.

In this case, the parent correction factor reduces to $\lambda_A/(\lambda_A - \lambda_B)$, and

$$D_B = RY_B = \frac{D_B}{\lambda_A/(\lambda_A - \lambda_B)} \quad (\text{III.29})$$

If, however, $(1 - \frac{\lambda_B}{\lambda_A} f)$ deviates appreciably from unity the yield computed from the saturation activity as given in (III.29) is too low. A more precise value can then be reached by successive approximation.

Case: $\lambda_A < \lambda_B$

In the growth of B from A if we have a transient equilibrium and further, the decay time $t$ is long enough so that independently-formed B has decayed away before isolation of B, we have, using (III.20)

$$D_B = RY_A \left( \frac{\lambda_B}{\lambda_B - \lambda_A} \right) \frac{1}{(1 - e^{-\lambda_A T})e^{-\lambda_A t}} \quad (\text{III.30})$$

from which $D_B^\infty$ and the yield of A can be computed.
E. DECAY SCHEMES AND FISSION PRODUCT DECAY
CHAIN IN THE MASS REGION, A=111-117

The fission product decay chains investigated are shown on the next page in Table V. The half-life values listed are those used in all subsequent calculations. Numbers over arrows indicate fraction of parent decaying via that path and detailed references pertaining to the decay schemes are given in section F.
<table>
<thead>
<tr>
<th>Element</th>
<th>Mass</th>
<th>45Rh</th>
<th>46Pd</th>
<th>47Ag</th>
<th>48Cd</th>
<th>49In</th>
<th>50Sn</th>
</tr>
</thead>
<tbody>
<tr>
<td>111</td>
<td>v.short?</td>
<td>5.3h</td>
<td>74s</td>
<td>48.6m</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>22m</td>
<td>7.5d</td>
<td>stable</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>112</td>
<td>v.short?</td>
<td>21hrs</td>
<td>3.2hrs</td>
<td>stable</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>113</td>
<td>v.short?</td>
<td>1.4m</td>
<td></td>
<td>stable</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>5.3h</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>115</td>
<td>v.short?</td>
<td>39s</td>
<td>20s</td>
<td>0.72</td>
<td>0.085</td>
<td>2.3d</td>
<td>6.104y</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.915</td>
<td>0.95</td>
<td></td>
<td></td>
</tr>
<tr>
<td>116</td>
<td>v.short?</td>
<td>?</td>
<td>2.5min</td>
<td>stable</td>
<td>22sec</td>
<td></td>
<td></td>
</tr>
<tr>
<td>117</td>
<td>v.short</td>
<td>v.short</td>
<td>3.4hrs</td>
<td>1.95hrs</td>
<td>1.0</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
F. RELEVANT DETAILS OF DECAY SCHEMES, AND, COMPUTATION OF ACTIVITIES AT END OF IRRADIATION OF NUCLIDES STUDIED IN THE PRESENT WORK

(1) Chain 117, Isobars Studied: $^{117}\text{Cd}$, $^{117}\text{In}$

The composite decay scheme, shown in Fig. 34 is based on del Marmol et al. (De 66) for the Cd-In decay and the part pertaining to the In-Sn decay incorporates information contained in Hagebø (Ha 70) and Baedecker et al. (Ba 70). The half-lives for the isomers involved were taken from the work of Cline and Heath (Cl 69).

The time scheme for separation of the indium isotope under examination follows the flow-chart illustrated in Fig. 25 in the experimental section of this work, and, can be depicted on a time-scale as follows:

<table>
<thead>
<tr>
<th>$t$</th>
<th>Duration of Milking ($\Delta t$)</th>
<th>$t_0$</th>
<th>$t_1$</th>
<th>Time of measurement of In activity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Duration of Irradiation</td>
<td>(\approx 10 \text{ min})</td>
<td></td>
<td></td>
<td>first In-Cd Separation</td>
</tr>
<tr>
<td>2.4 h $^{117}\text{Cd}$</td>
<td></td>
<td></td>
<td></td>
<td>end of Milking (2nd In+Cd separation)</td>
</tr>
<tr>
<td>1.94 h $^{117m}\text{In}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>3.4 h $^{117m}\text{Cd}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The genetic relationship of the two indium isomers can be illustrated as follows:

\[ \begin{array}{c}
2.4 \text{ h }^{117}\text{Cd} \\
\text{a} \quad 1.94 \text{ h }^{117m}\text{In} \quad \gamma \quad 44 \text{ min }^{117}\text{In} \\
\text{b} \quad T_4 \quad \lambda_2 \\
3.4 \text{ h }^{117m}\text{Cd} \\
\end{array} \]

\[ \text{117Sn} \]
FIGURE 34

Chain A = 117.
CHAIN A = 117

\[ ^{117}\text{Cd} \rightarrow \frac{1}{2}^{+} \rightarrow \frac{1}{2}^{-} \rightarrow 1.94\text{h} \rightarrow 1\text{.}94\text{h} \rightarrow 314\text{ (46\%)} \rightarrow 117\text{In} \rightarrow 44\text{m} \rightarrow 117\text{Sn} \]

\[ 117\text{Cd} \rightarrow 117\text{In} \rightarrow 117\text{Sn} \]
where $\alpha$, $\beta$, and $\gamma$ are branching ratios. If $\Delta t$ is the duration of milking, for each separation, the initial activities $A_1(t_0)$ and $A_4(t_0)$ of $^{117}\text{Cd}$ and $^{117m}\text{Cd}$ respectively can be related to the indium activities observed at the end of this milking period in the following manner:

Define:

\[
\begin{align*}
\frac{\lambda_2}{\lambda_2 - \lambda_1} &= a \\
\frac{\lambda_3}{\lambda_3 - \lambda_2} &= c \\
\frac{\lambda_3 - \lambda_4}{\lambda_3 - \lambda_4} &= f \\
\frac{\lambda_3}{\lambda_3 - \lambda_1} &= b \\
\frac{\lambda_2}{\lambda_2 - \lambda_3} &= d \\
\frac{\lambda_2}{\lambda_2 - \lambda_4} &= g
\end{align*}
\]

and

\[
\begin{align*}
C_1 &= ab \exp(-\lambda_1 \Delta t) - ac \exp(-\lambda_2 \Delta t) - bd \exp(-\lambda_3 \Delta t) \\
C_2 &= a \left[ \exp(-\lambda_1 \Delta t) - \exp(-\lambda_2 \Delta t) \right] \\
C_3 &= b \left[ \exp(-\lambda_1 \Delta t) - \exp(-\lambda_3 \Delta t) \right] \\
C_4 &= gf \exp(-\lambda_4 \Delta t) - gc \exp(-\lambda_2 \Delta t) - fd \exp(-\lambda_3 \Delta t) \\
C_5 &= g \left[ \exp(-\lambda_4 \Delta t) - \exp(-\lambda_2 \Delta t) \right] \\
C_6 &= f \left[ \exp(-\lambda_4 \Delta t) - \exp(-\lambda_3 \Delta t) \right]
\end{align*}
\]
For the path indicated by (III.31), at time $t$, the activities of $^{117}\text{In}$ and $^{117m}\text{In}$ will be related to $A_1(t_0)$ and $A_4(t_0)$ by:

$$A_3(t_1) = \gamma [\alpha C_1 A_1(t_0) + \beta C_4 A_4(t_0)] \quad (\text{III.33})$$

$$A_2(t_1) = \alpha C_2 A_1(t_0) + \beta C_5 A_4(t_0) \quad (\text{III.34})$$

and for (III.32) the activity of $^{117}\text{In}(A_3')$ formed by both $^{117}\text{Cd}$ and $^{117m}\text{Cd}$ will be:

$$A_3'(t_1) = (1-\alpha) C_3 A_1(t_0) + (1-\beta) C_6 A_4(t_0) \quad (\text{III.35})$$

After milking the observed activity of $^{117}\text{In}$ will be:

$$A_3(t) = \gamma C A_2(t_1)[\exp (-\lambda_2 t) - \exp (-\lambda_3 t)] +$$

$$[A_3(t_1) + A_3'(t_1)] \exp (-\lambda_3 t) \quad (\text{III.36})$$

By means of a least-squares analysis programme the decay data of In activity, observed at any time $t$, can be resolved into the 1.94-h component ($^{117m}\text{In}$) and 44-m component ($^{117}\text{In}$) - these activities extrapolated to time $t_1$ are given by the following mathematical expressions:

$$A_{117m}^{t_1} = \gamma C [\alpha C_2 A_1(t_0) + \beta C_5 A_4(t_0)] \quad (\text{III.37})$$

$$A_{117}^{t_1} = [\gamma C_1 + (1-\alpha) C_3 - \gamma C C_2] A_1(t_0) +$$

$$[\gamma C_4 + (1-\beta) C_6 - \gamma C C_5] A_4(t_0) \quad (\text{III.38})$$
A set of similar expressions will relate the activities of $^{117m}\text{In}$ and $^{117g}\text{In}$ for samples separated at time $t_0$ to the Cd activities at the end of bombardment. The net In-isomer activity would result from beta-decay through the chain in addition to that formed independently. Hence, one can write:

$$A_{117m\text{In}}^{t_0} = \gamma C(a_2 A_1(t_0) e^{\lambda(t_0-T)} + \beta C_5 A_4(t_0) e^{\lambda(t_0-T)}) + A_{117m\text{In}}^{\text{EOB}}(i)$$

(III.39)

$$A_{117g\text{In}}^{t_0} = [\gamma a C + (1-a) C_3 - \gamma a C_2] A_1(t_0) e^{\lambda(t_0-T)} + [\gamma B C + (1-B) C_6 - \gamma B C C_5] A_4(t_0) e^{\lambda(t_0-T)} + A_{117g\text{In}}^{\text{EOB}}(i)$$

(III.40)

where $A_{117m+g\text{In}}^{\text{EOB}}(i)$ is the activity at end of bombardment due to the independently-formed $^{117m+g}\text{In}$.

From the four equations (III.37), (III.38), (III.39) and (III.40) one can obtain values for the four unknowns, namely, $A_1(t_0), A_4(t_0), A_{117m\text{In}}^{\text{EOB}}(i)$ and $A_{117g\text{In}}^{\text{EOB}}(i)$.

(2) **Chain A=116, Nuclide Studied In**

The decay scheme indicated in Fig. 35 is based on Lederer and Hollander (Le 67) and Rabenstein et al. (Ra 72).
FIGURE 35

Chain A = 116.
The most intense gammas are 0.41 MeV (30%), 0.83 MeV (61%),
1.10 MeV (61%), 1.29 MeV (84%), and 2.12 MeV (16%), based on
Hansen et al. (Ha 62). The areas under the 0.41, 1.10, and 1.29
MeV photopeaks calculated by use of PDP-8 computer, were found to
be in the ratio 0.90:0.76:1.00. Thus, the detection limits
will be very similar for measuring the activity of either the 0.41
or 1.29 MeV peaks. It was found that it was most satisfactory to
follow the 1.29 MeV peak because of fewer observed interferences.

Computation of End of Bombardment Activity

This is a case of growth of a long-lived daughter from
an initially pure short-lived parent. One can express the net
counting rate of the parent-daughter mixture [A] as follows:

\[
[A] = [A^0_p - A^\infty_d] \left[e^{-\lambda_p T}\right] + [A^\infty_d]
\]  

(III.41)

where \(A^\infty_d\) is the saturation activity of \(^{116}\text{Sn}\) and is the y-intercept
of the straight line, \(A^0_p\) is the activity of the parent \(^{116m}\text{In}\)
at the end of bombardment and can be obtained as the sum of the
slope and the intercept after a least-squares analysis of the
decay data. The maximum daughter activity was recorded typically
about three to four hours after end of irradiation.

(3) Chain A=115, Isobars Studied: \(^{115}\text{Cd}, ^{115}\text{In}, ^{115}\text{Ag}\)

The decay scheme (Fig. 35) is based on the works by Hicks
and Gilbert (Hi 55) and, Kjelberg and Pappas (Kj 69). The
chemical separation entails timed-extraction of indium.
FIGURE 36

Chain A = 115.
CHAIN A = 115
isotopes from their cadmium parents, in the mixed fission product solution using a milking procedure. Likewise cadmium isotopes are allowed to grow in from silver parents after removal of all independently-formed cadmium. The segment of the decay chain dealing with silver-cadmium decay can be drawn in an abbreviated form as follows:

\[ T \xrightarrow{P} A \]

\[ T \xrightarrow{P} B \]

where, \( T \) = target, \( A = ^{115}\text{Ag} \) silver isotope, \( B = ^{115}\text{Cd} \) isotope.

The independent cross-section for the formation of \(^{115m}\text{Cd} \) may be computed since a chemical separation of \(^{21m}\text{Ag} \) is done very soon after irradiation (\( \approx 5-10 \) minutes) and the amount of \(^{115m}\text{Cd} \) growing in from \(^{115}\text{Ag} \) is estimated from the equation of timed-extraction. The cross-section for the formation of \(^{115}\text{Ag} \) can be expressed as:

\[
\sigma^{*}_{\text{Cd}} = \sigma^{i}_{\text{Cd}} + 0.28 \sigma^{C}_{\text{Ag}}
\]

where \( \sigma^{i}_{\text{Cd}} \) = the cross section for the independent formation of \(^{115}\text{Cd} \), and

\( \sigma^{C}_{\text{Ag}} \) = the cross-section for the cumulative formation of \(^{115m}\text{Ag} \).
Let \( t_1 \) be the duration of bombardment and \( t_2 \) the time of chemical separation of cadmium from silver, measured from the end of bombardment. The cumulative cross-section of \( ^{115}\text{Ag} \) plus 0.72 of the cumulative cross-section of \( ^{115}\text{mAg} \) is the measured cross-section \( \sigma_A \).

\[
\sigma_A = \sigma^* = 1.00 \sigma^{^{115}\text{Ag}} + 0.72 \sigma^{^{115}\text{mAg}} \quad \text{(III.42)}
\]

The disintegration rate of \( B \) at the time of separation \( (t_2) \) of \( B \) from \( A \) (i.e. of cadmium from silver) is made up of two parts:

1. (1) that grown from \( A \) during the bombardment and subsequent decay (c.f. section D):

\[
D_B^1 = \frac{D_A^0}{1 - e^{-\lambda_A t_1}} \left[ 1 - \frac{\lambda_A e^{-\lambda_B t_1} - \lambda_B e^{-\lambda_A t_1}}{\lambda_A - \lambda_B} \right] e^{-\lambda_B t} \quad \text{(III.43)}
\]

2. (2) that grown from \( A \) during the time interval covering the period from the end of bombardment to the separation time, i.e., during time \( t_2 \), plus that formed independently and subsequently decayed:

\[
D_B^2 = \frac{\lambda_B}{\lambda_B - \lambda_A} \left( D_A^0 e^{-\lambda_A t_2} - e^{-\lambda_B t_2} \right) + \frac{D_B e^{-\lambda_B t_2}}{\lambda_B - \lambda_A} \quad \text{(III.44)}
\]

Thus the experimentally-measured disintegration rate at the time of separation is:
\[ D_B^t = D_B^1 + D_B^2 \]  

(III.45)

from which \( D_B^o \) can be calculated. Since \( t_1 \) and \( t_2 \) are large compared with \( \lambda_B \) for \( 115mCd \) or \( 115gCd \), \( e^{-\lambda_B t_1} \) and \( e^{-\lambda_B t_2} \approx 1 \)

and since \( \lambda_B <\lambda_A \), \( \lambda_A - \lambda_B = \lambda_A \).

Therefore,

\[ D_B^1 = \frac{D_A^o}{1 + \frac{\lambda_B e^{-\lambda_A t_1}}{\lambda_A}} \]

\[ = \frac{D_A^o}{1 - e^{-\lambda_A t_1}} \quad (III.46) \]

and

\[ D_B^2 = \frac{\lambda_B}{\lambda_A} D_A^o (1 - e^{-\lambda_A t_2}) + D_B^o e^{-\lambda_B t_2} \]  

(III.47)

Therefore

\[ D_B^t = \frac{\lambda_B}{\lambda_A} D_A^o [1 - e^{-\lambda_A t_2}] + \frac{e^{-\lambda_A t_1}}{1 - e^{-\lambda_A t_1}} \]

(III.48)

\[ D_B^o e^{-\lambda_B t_2} \]

Therefore

\[ D_B^o = e^{\lambda_B t_2} [D_B^t - \frac{\lambda_B}{\lambda_A} D_A^o F] \]  

(III.49)

where

\[ F = 1 - e^{-\lambda_A t_2} + \frac{e^{-\lambda_A t_1}}{1 - e^{-\lambda_A t_1}} \]
For the ground-state $^{115}\text{gCd}$,

$$D^0_{B}\text{g} = e^{\lambda_{B}t_2} \left[ D^0_{B}\text{g} - \frac{\lambda_{B}}{\lambda_{A}} D^0_{A} \text{F} \right] \quad (\text{III.50})$$

Now

$$D^0_{A} = \frac{1}{91} (D^0_{B}\text{g})_{aq} \frac{\lambda_{A}}{\lambda_{B}} e^{\lambda_{A}t_2}$$

$$= \frac{1}{91} A_4 Y_a \frac{\lambda_{A}}{\lambda_{B}} e^{\lambda_{A}t_2} \quad (\text{III.52})$$

and

$$D^0_{B}\text{g} = A_2 Y_0 \quad (\text{III.52})$$

where $A_2$ and $A_4$ are the measured activities of $^{115}\text{gCd}$ before and after milking operations, and, $Y_0$ and $Y_a$ are the chemical yield factors in these two instances. (III.52) can be elaborated as follows taking into account the decay during the milking process,

$$D^0_{B}\text{g} = e^{\lambda_{B}t_2} \left[ A_2 Y_0 - A_4 Y_a \cdot e^{\lambda_{A}t_2} \text{F} \right]$$

$$= e^{\lambda_{B}t_2} \left[ A_2 Y_0 - A_4 Y_a \cdot \text{F}' \right] \quad (\text{III.53})$$

where $\text{F}' = e^{\lambda_{A}t_2} \text{F}$. 

Similarly for the metastable-state $^{115}\text{mCd}$,
where $A_1$ and $A_3$ are the measured activities of $^{115m}$Cd before and after milking operations, respectively.

By substituting the above-mentioned equations in the general expression for cross-section calculations (section B), one gets

\[
D_{Bm}^0 = e^{\lambda Bm t^2} \left[ A_1 Y_0 - A_3 Y_a \cdot F' \right] \quad (\text{III.54})
\]

for $^{115m}$Cd,

\[
\sigma_m = \sigma_m \frac{N_M}{N_T} \frac{(1-e^{-\lambda t^1})}{(1-e^{-\lambda t^1})} \frac{A_1 Y_0 - A_3 Y_a}{D_{Bm}^0} \quad (\text{III.55})
\]

for $^{115g}$Cd,

\[
\sigma^* = \sigma_m \frac{N_M}{N_T} \frac{(1-e^{-\lambda t^1})}{(1-e^{-\lambda t^1})} \frac{A_2 Y_0 - A_4 Y_o}{D_{Bm}^0} \quad (\text{III.56})
\]

The decay of $^{115}$Cd into $^{115m}$In can be schematically represented as:

\[\begin{array}{c}
\text{43d} \quad ^{115g}_{\text{Cd}} \\
\alpha \\
\lambda_1 \\
\downarrow 4.5 \text{ h} \\
\downarrow ^{115m}_{\text{In}} \quad \text{I.T.} \\
\downarrow ^{335 \text{ KeV.}} \\
\downarrow \text{335 KeV.} \\
\downarrow ^{115g}_{\text{In}} \quad \text{(stable)} \\
\text{2.3d} \\
\downarrow ^{115m}_{\text{Cd}} \\
\beta \\
\lambda_4
\end{array}\]

Activity of $^{115m}$In measured at a time $t^*$ can be related to known end-of-bombardment $^{115}$Cd activities from the same experiment as follows:

\[
A_{115m}^{t*} = a_1 C_{115} A_{\text{EOB}} + \beta_2 C_{115} A_{\text{EOB}} + A_{\text{EOB}} \\
^{115m}_{\text{In(i)}}
\]

where: $a$, $\beta$ are branching ratios.

*usually about 24 hours after end of irradiation.
\[
C_1 = \frac{\lambda_2}{\lambda_2 - \lambda_1} \left[ \exp\left(-\lambda_1 \Delta t\right) - \exp\left(-\lambda_2 \Delta t\right) \right] \quad (III.59)
\]
\[
C_2 = \frac{\lambda_2}{\lambda_2 - \lambda_4} \left[ \exp\left(-\lambda_4 \Delta t\right) - \exp\left(-\lambda_2 \Delta t\right) \right] \quad (III.60)
\]

\[ A_{EOB} = \text{activity of the isotope at end of bombardment}. \] Hence, from (III.58) one can compute end of bombardment activity of independently formed 115mIn.

(4) **Chain Au=113, Nuclide Studied: \(^{113}\text{Ag}\)**

The composite decay scheme shown in Fig. 37 is based on Raman (Ra 71), Matumoto (Ma 70), and Liptak et al. (Li 69). The schematic representation of the decay scheme (Fig. ) for the purpose of activity computation will be:

The activity of the ground \((g)\) state of the end of irradiation comes from four different sources,

(a) As a directly formed fission product: \((D^i_g)\)
(b) Decay of \(A\) into \(g\): \((D^A_g)\)
(c) Decay of the directly formed fission product \(m\) into \(g\): \((D^m_g)\)
(d) Decay of \(m\) produced by disintegration of \(A\): \((D^M_g)\)
where:

\[ T = \text{duration of bombardment} \]
\[ D = \text{disintegration rate of the isotope in question}. \]

These activities can be evaluated as follows:

\[ (D_i^g)_T = \frac{R Y_i}{g} (1 - e^{-\lambda g T}) \]  
(III.61)

where \( Y_i^g \) = independent fission-yield of the ground-state isomer

\[ R = n f \sigma, \text{the symbols are as defined in section III (part D) of this work}. \]

\[ (D^A_g)_T = \frac{R Y_i^A}{A_g} (1 - \frac{\lambda A}{\lambda g} e^{-\lambda g T} - \frac{\lambda A}{A - \lambda g} e^{-\lambda g T}) \]  
(III.62)

\[ (D_m^g)_T = \frac{R Y_i^m}{m_g} (1 - \frac{\lambda m}{\lambda m} e^{-\lambda m T} - \frac{\lambda m}{m - \lambda g} e^{-\lambda g T}) \]  
(III.63)

\[ (A_{D_m}^g)_T = \frac{R Y_i^A}{A_m g} \left[ 1 - \frac{\lambda m A^g}{(\lambda A - \lambda m) (\lambda g - \lambda m)} e^{-\lambda A T} \right. \\
- \frac{\lambda A^g}{(\lambda A - \lambda m) (\lambda g - \lambda m)} e^{-\lambda m T} - \frac{\lambda A m}{(\lambda A - \lambda g) (\lambda m - \lambda g)} e^{-\lambda g T} \]  
(III.64)

Adding these four activities, the total activity of \( g \) at the end of irradiation will be:

\[ (D_g)_T = (D_i^g)_T + (D^A_g)_T + (D_m^g)_T + (A_{D_m}^g)_T \]  
(III.65)

The activity of \( A \) at end of irradiation will be:
FIGURE 37

Chain A = 113.
CHAIN A = 113

\[ \text{Pd}^{113} \rightarrow 1.5 \text{ m} \rightarrow \text{Ag}^{113} \]

\[ 1.2 \text{ m} \rightarrow 100\% \]

\[ (\frac{7}{2}^+) \rightarrow 5.37 \text{ h} \]

\[ (\frac{1}{2}^-) \]

\[ \text{Ag}^{113} \rightarrow 7.5\% (\frac{1}{2}^+) \]

\[ <0.5\% \]

\[ 1\% \]

\[ 42\% (\frac{3}{2}^+) \]

\[ 50\% \]

\[ 298 \text{ K\text{eV}} \rightarrow \text{Cd}^{113} \]
\[ (D_A)_T = RY_A (1 - e^{-\lambda_A T}) \] (III.66)

and the activity of \( m \) will be given by:

\[ (D_m)_T = RY_A \gamma_m (1 - \frac{\lambda_m}{\lambda_A} e^{-\lambda_A T} - \frac{\lambda_A}{\lambda_m} e^{-\lambda_m T}) \]

\[ + RY_m (1 - e^{-\lambda_m T}) \] (III.67)

After an interval of time \( t \), starting from the end of irradiation (chain interruption), the total activity of \( g \) will be given by adding the following four terms:

(a) Activity of \( g \) from direct decay of \( A \)

\[ (D_A^g)_t = (D_A)_T \frac{\lambda_A}{\gamma_A} (1 - e^{-\lambda_A t}) \] (III.68)

(b) Activity of \( g \) formed by direct decay of \( m \) into \( g \)

\[ (D_m^g)_t = (D_m)_T \frac{\lambda_m}{\gamma_m} (1 - e^{-\lambda_m t}) \] (III.69)

(c) Activity of \( g \) formed from the decay of \( A \) into \( m \) and then to \( g \) during interval \( t \)

\[ (A_D^m)_g^T = (D_A)_T \frac{\lambda_m}{\gamma_m} (1 - e^{-\lambda_m t}) \left[ \frac{e^{-\lambda_A t}}{(\lambda_m - \lambda_A)(\gamma_m - \lambda_A)} + \frac{e^{-\lambda_m t}}{(\lambda_A - \lambda_m)(\lambda_m - \lambda)} \right] \] (III.70)
(d) The remaining activity of $g_i$, $(D_g)_T$

\[
(D_g)^T_t = (D_g)_T e^{-\lambda g_t} = \{(b_i^T + (D^A)_T + (D^m)_T + (D^m)_T\}
\]

\[
+ (D^m)_T e^{-\lambda g_t}
\]

(III.71)

If (III.60), (III.69), (III.70) and (III.71) are added and the values of $(D_A)_T$, $(D^m)_T$, $(D^i)_T$, $(D^A)_T$, $(D^m)_T$ and $(D^m)_T$ introduced from their corresponding expressions and the terms rearranged, the total activity of $g_i$, after an interval of time $t$ (chain interruption), will be given by:

\[
(D_g)_T = R_Y A \left\{ \frac{\gamma^m_{g}^A \lambda^g e^{-\lambda^g T}}{(\lambda^m - \lambda^g)(\lambda^m - \lambda^g)} (1 - e^{-\lambda^g T}) + \right. \\
\frac{\lambda^m_{g}^m \lambda^g e^{-\lambda^m T}}{(\lambda^m - \lambda^g)(\lambda^m - \lambda^g)} (1 - e^{-\lambda^m T}) + \frac{\lambda^m_{g}^A \lambda^m e^{-\lambda^g t}}{(\lambda^A - \lambda^g)(\lambda^m - \lambda^g)} (1 - e^{-\lambda^g T}) +
\]

\[
\frac{\lambda A^m_{g} \lambda^A e^{-\lambda^g t}}{(\lambda^A - \lambda^g)(\lambda^m - \lambda^g)} (1 - e^{-\lambda^g T}) + R_Y A \left[ \frac{\lambda^m_{g}^m \lambda^m e^{-\lambda^m T}}{(\lambda^m - \lambda^g)(\lambda^m - \lambda^g)} (1 - e^{-\lambda^m T}) \right] \\
+ \frac{\lambda^m_{g}^m \lambda^m e^{-\lambda^g t}}{(\lambda^m - \lambda^g)} (1 - e^{-\lambda^g T}) + R_Y g \left[ e^{-\lambda^g t} (1 - e^{-\lambda^g T}) \right] \right. \\
\]

(III.72)
(5) Chain A=112, Isobars Studied $^{112}\text{Pd}$, $^{112}\text{Ag}$

The composite decay-scheme for this chain as shown in Fig. 38, is derived from Pate et al. (Pa 69), Lipták et al. (Li 69), Macdonald et al. (Ma 70) and, Lingeman et al. (Li 68).

**Computation of End-of-Bombardment Activities**

The independent cross-section for semi-shielded $^{112}\text{Ag}$ could be obtained by experimentally following the procedure outlined by Bent Schröder (Sc 70). This method involves varying the timing of irradiation and extraction of the daughter activity $^{112}\text{Ag}$ at different time intervals from the same master solution. The first sample contained the activity formed independently and that produced during bombardment and in the short time interval before the separation from the precursor in the chain. The second sample contained activity formed by decay of the precursor only and gave information on the chain yield. Now by symbolically characterising the various terms in the decay equation as follows:

- $A =$ Parent isotope ($^{112}\text{Pd}$ in this case)
- $B =$ Daughter isotope ($^{112}\text{Ag}$ in this case)
- $T =$ Time of irradiation
- $t_1 =$ Time interval between end of bombardment and first separation of the isotope B from its precursor A
- $t_2 =$ Time interval between the first and the second separation
FIGURE 38

Chain \( \lambda = 112 \).
CHAIN A = 112

- - - - - - - - 0.7 s

$^{112}\text{Ru}$

- - - - - - - - 4.65 s

$^{112}\text{Rh}$

$^{2+} \rightarrow 0^+ \rightarrow 20.12 \text{ hr}$

$^{112}\text{Pd}$

$^{100\%} \rightarrow 11^+ \rightarrow 3.1 \text{ hr}$

$^{112}\text{Ag}$

$18\% \rightarrow 39 \text{ Levels}$

$76\% \rightarrow 39 \text{ Levels}$

$\rightarrow 0^+ \rightarrow <0.1 \text{ ns}$

$\rightarrow 2^+ \rightarrow 0.348 \text{ s}$

$\rightarrow 4.655 \text{ s}$

$\rightarrow 0.617 \text{ hr}$

$\rightarrow 112\text{Cd}$
\[ D_b(T, t_1) = \frac{D_0^b}{\lambda_b^T} \left[ \frac{\lambda_b^T}{\lambda_b - \lambda_a^T} \right] \left( 1 - e^{-\lambda_a^T t_1} \right) \frac{\lambda_a}{\lambda_b - \lambda_a} \]

\[ = \frac{D_0^b}{\lambda_b^T} \left[ (1 - e^{-\lambda_b^T}) e^{-\lambda_b^T t_1} \right] \quad (III.73) \]

and,

\[ D_b(t_2) = \frac{D_0^a}{\lambda_a^T} \left( 1 - e^{-\lambda_a^T t_2} \right) e^{-\lambda_a^T t_1} \frac{\lambda_b^T}{\lambda_b - \lambda_a} \left( e^{-\lambda_b^T t_2} - e^{-\lambda_b^T t_1} \right) \quad (III.74) \]

From (III.73) and (III.74), values of \( D_0^a \) and \( D_0^b \) can be evaluated. Although the values of the disintegration rates at zero time for \(^{112}\text{Ag}\) can be used for final cross-section computation, \( D_0^b \) cannot be used for that purpose without some necessary corrections as pointed out by Roche et al (Ro 71). These authors report that in the chemical isolation of palladium and subsequent milking of its daughter, tracer experiments indicate that the...
exchange between $^{112}$Pd and palladium carrier was only 96.5% complete. Also, they point out that during build-up or the milking period one has to take into account the loss of silver activity formed due to the incomplete separation of $^{112}$Pd in the initial precipitation process, and, that is approximately $0.07 \lambda_{112\text{Pd}} (t_2-t_1)$. Taking these into consideration, the ratio of the number of $^{112}$Ag to $^{112}$Pd atoms at the end of bombardment can be written as follows:

$$(N_{Ag}^0/N_{Pd}^0) =$$

\[
(0.965) \frac{\lambda_{112\text{Pd}}}{(\lambda_{112\text{Ag}} - \lambda_{112\text{Pd}})} \left\{ \frac{D_b^{(T_1t_1)}}{D_b^{(T_2\exp)} \lambda_{112\text{Pd}} (t_1 + t_2 - T)} \right\} - \lambda_{112\text{Pd}} t_1 - 0.07 \lambda_{112\text{Pd}} (t_2-t_1) \]  

\text{(III.75)}

In the above equation $N_{Ag}^0$ can be substituted from the $D_a^0$ value obtained from (III.74), and, thus $N_{Pd}^0$ becomes known.

(6) Chain A=111, Isobars Studied: $^{111}$Pd and $^{111}$Ag

The composite decay-scheme in Fig. 39 is drawn based on information obtained from Panontin (Pa 68), Shick and Talbert (Sc 69), Hnatowich and Coryell (Hn 68), and Nagarajan et al. (Na 71). As indicated by the decay scheme, the large difference in half-life between the two palladium isomers makes it possible to determine their relative yields by radio-
CHAIN A = 111

\[
\begin{align*}
\text{Rh} & \quad >99\% \\
\text{\(^{111}\text{Rh}\)} & \quad (1/2^-) \quad 0.17 \quad 5.3 \text{ M} \quad 71.3\% \quad 22\text{ M} \quad 28.7\% \quad 100\% \\
\text{Pd} & \\
\text{\(^{111}\text{Pd}\)} & \\
\text{\(^{111}\text{Ag}\)} & \quad 7/2^+ \quad 0.059 \quad 74\text{ S} \quad 1/2^- \quad 99.7\% \quad 7.47\text{ d} \quad 100\% \\
\text{\(^{111}\text{Cd}\)} & \quad 3/2^+ \quad 86.6(0.14) \quad 52.44(0.061) \quad 5/2^+ \quad 2.45(0.17) \quad 342(0.90) \quad 7/2^- \quad 96.3(2.5) \quad 1/2^+ \quad 1.80 \quad 1.50 \quad 1.20 \\
\end{align*}
\]
chemically separating $^{111}\text{Ag}$ several hours and several days after an irradiation. The chemical procedure which takes into account scavengings to remove any silver activity from part of the fission-products solution to be used for palladium yield determination, and, then timed milking to produce the requisite $^{111}\text{Ag}$ activity from 22-min $^{111}\text{gPd}$ and 5.5 hrs $^{111}\text{mPd}$ respectively, is based primarily on the method used by Aumann et al. (Au 69, Au 70), and, Panontin et al. (Pa 68). The modified chemical procedure used in this work is described in the "Experimental Procedures" part of this thesis.

Computation of End of Bombardment Activities for $^{111}\text{mPd}$ and $^{111}\text{gPd}$

An expression has been derived which relates the initial activity of these isomers soon after irradiation to the total measured activity of $^{111}\text{Ag}$ based on the above-mentioned chemical procedure. The schematic diagram indicating the decay path and the time-scale used in the separations is also shown in Fig. 40.

$$A^{(111}\text{Ag}) = 0.29 \sum A^0_m e^{-\lambda_m t_1} \left\{ \frac{\lambda^{(\text{Ag})}}{\lambda^{(\text{Ag})} - \lambda_m} \right\} \left[ e^{-\lambda_m t_2} - e^{-\lambda^{(\text{Ag})} t_2} \right] +$$

$$0.71 \sum A^0_m e^{-\lambda_m t_1} \left( \frac{\lambda^{(\text{Ag})}}{\lambda^{(\text{Ag})} - \lambda_m} \right) \left( \frac{e^{-\lambda_m t_2}}{(\lambda_m - \lambda_g)(\lambda^{(\text{Ag})} - \lambda_g)} + \frac{e^{-\lambda^{(\text{Ag})} t_2}}{(\lambda_m - \lambda^{(\text{Ag})})(\lambda_g - \lambda^{(\text{Ag})})} \right)$$
FIGURE 40

Schematic representation of the decay path for the nuclide $^{111}_{\text{Ag}}$. 
The diagram illustrates the decay processes and timelines for isotopes 111m_Pd, 111g_Pd, and 111_Ag. The decay of 111m_Pd results in 111g_Pd with a half-life of 5.3 hours, and 111_Ag with a half-life of 22 minutes. The variation of time intervals gives an independent yield of 111_Ag.

**Time Scale**

- Start of irradiation
- End of irradiation
- Silver separation time
- End of Milking

**Palladium Separation**

- 111g_Pd, 111m_Pd yields.

Define:

- \( \lambda_g \) = decay constant of 111g_Pd
- \( \lambda_m \) = decay constant of 111m_Pd
- \( \lambda(Ag) \) = decay constant of 111_Ag
- \( t_1 \) = length of time from end of bombardment to the milking period
- \( t_2 \) = length of the milking period
\[ + \frac{\lambda(Ag)}{\lambda(Ag)-\lambda g} \{ \frac{\lambda_g}{\lambda g-\lambda m} \frac{1}{\frac{\lambda g}{\lambda g-\lambda m}} \ \text{Am} e^{-\lambda t_1} - e^{-\lambda(Ag)t_2} \} \]

where the total measured activity of $^{111}$Ag consists of terms describing contributions to that activity from the following sources:

(a) from the direct decay of $^{111m}$Pd into $^{111}$Ag

(b) from the indirect decay of $^{111m}$Pd via $^{111g}$Pd into $^{111}$Ag

(c) from the direct decay of $^{111g}$Pd which in turn is the cumulative product of its formation during the period of bombardment as well as that formed by the decay of $^{111m}$Pd during the time period denoted by $t_1$

**Computation of End-of-Bombardment Activity for $^{111}$Ag**

The chemical procedure, as described in the "Experimental Procedures" section of this thesis, entails four or five separations of aliquots of mixed fission product solution for assay of radioactive silver within an hour after end of a very short irradiation ($\approx$ 2 minutes). On a time scale it essentially involves varying the time interval depicted by $t_1$ in the schematic diagram in Fig. 40. The measured silver activity, therefore, can be expressed as a sum of the activity due to the independently-formed $^{111}$Ag and that formed due to decay through the chain. An equation of the type,
Total $^{111}\text{Ag}$ Activity =

$\text{(Activity }^{111}\text{Ag)}_{\text{independent}} + \text{(Activity }^{111}\text{Ag)}$

can thus be written for each of the 4-5 separations and a value for the independently formed activity due to $^{111}\text{Ag}$ can be evaluated. The $^{111}\text{Ag}$ activity for each of these expressions (beta decay through the chain) was obtained by substituting $t_1 = 0$ and $t_2 = t$ (i.e. time from mean irradiation time to time of silver separation) in (III.76)
G. BETA DECAY CURVES

The decay curves of representative nuclides from some of the beta-decay experiments appear on the following pages Figs. 41(a) - 41(c).

The measurement of activity for the hard betas of $^{112}$Ag were obtained by using the 800 mg/cm$^2$ Al absorbers.
FIGURE 41(a) - 41(c)

Sample decay curves from experiments involving beta-decay of pure radiochemical samples.
CLSPLOT
70 MeV Ag 112 Decay, July 17, 1972. 880 mGy/SqCm. Al Absorber
The final results of cross-sections as tabulated for the absolute determination for the monitor reaction $^{65}\text{Cu}(p,pn)^{64}\text{Cu}$ in Chapter II, and subsequent cross-section tabulations in Chapter IV are given with the uncertainties associated with the reported experimental results. In Table II is given a summary of the estimated standard errors for the monitor cross-section determinations. "Systematic" errors are those which cause a constant deviation for each measurement, or which causes a systematic variation with energy of irradiation, Bevington (Be 69). "Random" errors arise because of imprecisions in actual measurements. The random errors arise mainly from determination of disintegration rates, chemical yields, weights of the target and the monitor. The main source of error lies in the determination of the photo-peak area. This error was estimated to be $\pm 5$-$15\%$ depending on the complexities. The error in the initial disintegration rate could vary from two-percent (extrapolation of $^{64}\text{Cu}$ decay curve) to ten-percent for specific nuclides in an experiment. An error of $1.5$-$2\%$ was allowed in weighing and pipetting. An error of $1$-$3\%$ were allowed depending on individual cases for timing of separations, the upper limit was taken for the $^{111}\text{Ag}$ independent yield experiments requiring many separations in a short interval of time.

In chemical yield determinations, some representative
samples for the nuclides in this work the agreement was between ±3-5%. Error in the efficiency calibration of the detector varied between 2.0% for the 30-cm$^3$ and 40-cm$^3$ Ge(Li) to about 7% for $^{112}$Ag beta measurements using the cross calibration technique. An error of ±3% was allowed for sample geometry reproducibility.

The "systematic" errors, involve monitoring the beam using the standard cross-section for the $^{65}$Cu(p,pn)$^{64}$Cu reaction, and as shown previously is now known to within ±5.9%. In the region of work undertaken for this dissertation the errors due to uncertainties in the decay schemes is of paramount importance involving 10-12% uncertainty in the intensity of weak peak to 1% for more intense peaks. The branching ratio uncertainties range from 2-3%. The error associated with separate detectors being used for monitor activity measurements and actual sample activity measurement is in the range 1-3%. The spread in the incident internal proton beam has already been described graphically in Chapter II, and is on the average known to within ±2 MeV of the incident energy.

The total error was calculated by taking the square root of the sum of squares of individual errors cited above considering both the nuclide under study and the monitor. Taking specific uncertainties involving assumptions of isomer ratio for nuclides and scavenging errors for some of the nuclides as described in Chapter IV, the total error quoted in this work varied between ±11% and ±25%. 
IV. RESULTS

The independent formation cross-sections for the nuclides $^{117}$In, $^{112}$Ag, and $^{111}$Ag at each energy were directly computed as explained in Chapter III (Treatment of Data), taking into account corrections for chemical yield, detector efficiency, aliquot, parent-daughter factors etc. as explained in that chapter. The independent cross-section of $^{115}$Cd was computed from the measured $\sigma^{115m}\text{Cd (independent)}$ and the ratio of the isomer $^{115m}\text{Cd}/^{115g}\text{Cd}$ given by Tilbury and Yaffe (Ti 63) in the energy range of this work. It has been pointed out by Schmitt and Zumwalt (Sc 63) that the cross-section of $^{111m}\text{Pd}$ could be considered to be independent since the decay of $^{111}$Rh is greater than 99 percent to the ground state, the isomer ratio $\sigma(111m\text{Pd})/\sigma(111g\text{Pd})$ ind. is reported to be $2.5 \pm 0.6$ by Panontin and Porile (Pa 68) and they point out the small variation of this value in low and high energy fission. A value of 2.0 was used and thus the total independent cross-section was computed for this nuclide. From the experimentally-determined $115m$In independent formation cross-section, a total value for the cross-section was computed by assuming at each energy of experimentation a similar high spin-to low-spin isomer ratio as that of $^{117}$In determined in this work with identical spins. These isomer ratios are plotted in Fig. 42 and they agree well with similar measurements by Hagebø (Ha 65). The activities of $^{117}$In were increased by 4% to take into account scavenge losses.

* Table VI
TABLE VI

Independent isomeric yield ratios
($^{117}$In/$^{117m}$In)

| Irradiation Energy (MeV) | $^{117}$In/$^{117m}$In | Present work | Hagebø (1964) *
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>15 ± 3</td>
<td>0.8 ± 0.5</td>
<td>0.6 ± 0.4</td>
<td></td>
</tr>
<tr>
<td>29 ± 3</td>
<td>1.6 ± 0.4</td>
<td>1.2 ± 0.2</td>
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<tr>
<td>43 ± 4</td>
<td>2.5 ± 0.5</td>
<td>2.7 ± 0.7</td>
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<tr>
<td>57 ± 4</td>
<td>4.8 ± 0.8</td>
<td>4.2 ± 0.8</td>
<td></td>
</tr>
<tr>
<td>70</td>
<td>8.5 ± 0.8</td>
<td></td>
<td></td>
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<tr>
<td>85</td>
<td>9.8 ± 0.9</td>
<td>9.0 ± 1.0</td>
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<tr>
<td>110</td>
<td></td>
<td>12.0 ± 1.0</td>
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</table>

* Ref. (Ha 65)
FIGURE 42

Independent isomeric yield ratios $^{117g}_{\text{In}}/^{117m}_{\text{In}}$. 
PRESENT WORK

HAGEBÖ (1964)

INDEPENDENT YIELD RATIO $^{117}\text{g In}/^{117}\text{m In}$

PROTON ENERGY
as determined from tracer work described previously. Similarly, in the determination of $^{111}$Pd, activities were increased by three percent to correct for scavenging losses. No self-absorption corrections were made since specific weights of all samples were within 1 mg/cm$^2$ of the specific standard thickness used in the calibration. For the particular beta energies of these nuclides, the corrections for self-absorption are one percent or less. The cumulative yield recorded for $^{113}$Ag is considered "partial" to account for the partial loss from the decay of $^{113m}$Ag to $^{113}$Cd.

In a subsequent part of this chapter, measured independent yields and cumulative yields are tabulated and drawings of the excitation functions are shown in figs. 43-54.
<table>
<thead>
<tr>
<th>$E_p$ (MeV)</th>
<th>$117\text{In}$</th>
<th>*$115\text{In}$</th>
<th>*$115\text{Cd}$</th>
<th>*$111\text{Pd}$</th>
<th>$112\text{Ag}$</th>
<th>$111\text{Ag}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>85</td>
<td>7.9 ± 1.2</td>
<td>2.6 ± 0.4</td>
<td>10.6 ± 1.6</td>
<td>12.3 ± 2.0</td>
<td>7.0 ± 1.0</td>
<td>4.0 ± 0.6</td>
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<td></td>
<td>6.0 ± 0.9</td>
<td>1.5 ± 0.5</td>
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</tr>
<tr>
<td>70</td>
<td>5.3 ± 0.8</td>
<td>0.9 ± 0.2</td>
<td>4.5 ± 0.7</td>
<td>9.0 ± 1.4</td>
<td>3.8 ± 0.5</td>
<td>1.5 ± 0.2</td>
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<tr>
<td>55</td>
<td>2.6 ± 0.5</td>
<td>0.2 ± 0.03</td>
<td>3.9 ± 0.6</td>
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<tr>
<td>40</td>
<td>1.2 ± 0.2</td>
<td>0.05 ± 0.01</td>
<td>1.9 ± 0.3</td>
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<td>0.2 ± 0.03</td>
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<td>30</td>
<td>0.7 ± 0.1</td>
<td>0.03 ± 0.005</td>
<td>1.2 ± 0.3</td>
<td>2.0 ± 0.3</td>
<td>0.4 ± 0.06</td>
<td>0.08 ± 0.01</td>
</tr>
<tr>
<td></td>
<td>0.4 ± 0.06</td>
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<td></td>
</tr>
<tr>
<td>20</td>
<td>0.16 ± 0.04</td>
<td>0.01 ± 0.003</td>
<td>0.5 ± 0.1</td>
<td>1.8 ± 0.3</td>
<td>0.18 ± 0.03</td>
<td>0.05 ± 0.008</td>
</tr>
<tr>
<td></td>
<td>0.9 ± 0.2</td>
<td>0.05 ± 0.01</td>
<td>0.3 ± 0.08</td>
<td>4.0 ± 0.8</td>
<td>0.4 ± 0.08</td>
<td>0.15 ± 0.03</td>
</tr>
</tbody>
</table>

*Estimated cross-sections based on independent isomer ratio values explained in text.*
TABLE VIII

Cumulative formation cross-sections of products from 20-85 MeV proton fission of $^{238}$U.

<table>
<thead>
<tr>
<th>$E_p$ (MeV)</th>
<th>$^{112\text{Pd}}$</th>
<th>$^{111\text{Ag}}$</th>
<th>$^{115\text{Ag}}$</th>
<th>$^{113\text{Ag}}^*$</th>
</tr>
</thead>
<tbody>
<tr>
<td>85</td>
<td>43.7 ± 7</td>
<td>46.8 ± 5.6</td>
<td>37.8 ± 5.6</td>
<td>48.2 ± 6.0</td>
</tr>
<tr>
<td>70</td>
<td>35.6 ± 4.8</td>
<td>28.2 ± 4.2</td>
<td>32.3 ± 4.8</td>
<td>38.6 ± 5.8</td>
</tr>
<tr>
<td>55</td>
<td>30.2 ± 4.5</td>
<td>26.4 ± 5.3</td>
<td>30.5 ± 4.6</td>
<td>32.1 ± 4.8</td>
</tr>
<tr>
<td>40</td>
<td>22.6 ± 3.4</td>
<td>22.0 ± 3.3</td>
<td>21.3 ± 3.2</td>
<td>22.7 ± 3.4</td>
</tr>
<tr>
<td>30</td>
<td>14.2 ± 2.2</td>
<td>16.1 ± 2.4</td>
<td>16.2 ± 2.4</td>
<td>17.8 ± 2.6</td>
</tr>
<tr>
<td>20</td>
<td>11.2 ± 1.7</td>
<td>11.2 ± 1.7</td>
<td>7.6 ± 1.1</td>
<td>10.9 ± 1.6</td>
</tr>
</tbody>
</table>

*A partial cumulative yield, since there is a partial loss from the decay of $^{113\text{Ag}}$ to $^{113\text{Cd}}$. 
FIGURE 43

$^{117}\text{In}^{(m+g)}$, independent formation cross-section.
$^{117}_{\text{In}}(^{m+g})$ INDEPENDENT CROSS-SECTION

$^{238}_u(p,f)$  N/Z = 1.387

\begin{align*}
\sigma_{(\text{mb})} & \text{ vs. } E_p \text{ (MeV)} \\
20 & \text{ to } 80
\end{align*}
FIGURE 44

$115m^m_{Cd}$, independent formation cross-section.
FIGURE 45

$^{115}\text{Cd}$, independent formation cross-section (estimated).
$^{115}\text{Cd}$ INDEPENDENT CROSS-SECTION

$^{238}\text{U}(p,t)^{\text{N/Z}} = 1.395$
FIGURE 46

$^{115}$In, independent formation cross-section.
INDEPENDENT CROSS-SECTION

$^{238}\text{U}(p, f) \quad N/Z = 1.346$
FIGURE 47

$^{111}\text{Ag}$, independent formation cross-section.
$^{311}$Ag INDEPENDENT CROSS-SECTION

$238\; u(p,f)\quad N/Z = 1.361$
$^{112}\text{Ag}$, independent formation cross-section.
\( ^{112}\text{Ag} \) INDEPENDENT CROSS-SECTION

\[ ^{238}\text{U}(p,f) \quad N/Z = 1.382 \]
$^{111}$Pd, independent formation cross-section.
$^{117}$Pd INDEPENDENT CROSS-SECTION
$^{238}$U($p_f$) N/Z = 1.413
FIGURE 50

$^{112}_{\text{pd}}$, cumulative formation cross-section.
$^{113}$Ag, cumulative formation cross-section.
FIGURE 52

111Ag, cumulative formation cross-section.
$^{115}\text{Ag}$, cumulative formation cross-section.
\[ \sigma_{115g \text{ Ag}} \text{ (mb)} \]

- PRESENT WORK
- HICKS AND GILBERT
  (Hi 65)

ENERGY (MeV)
V. GENERAL CONCLUSIONS AND DISCUSSION

(1) Charge dispersion and Most Probable Charge

(i) Graphical Representation

The curves shown in Figs. 54-59 constitute the charge distribution of nuclides measured in this work in the symmetric, and near-symmetric mass regions. The prior assumption that the dispersion along isotopic yields in mass regions studied in medium and high-energy fission of natural uranium could be given by a normal Gaussian equation, which has been so successfully used by Wahl et al. (Wa 62) for explaining the vast amount of thermal-neutron data, forms the basis of this present work.

The single-peaked charge distribution curve has been drawn with fractional mass-yield plotted against the value of $N/Z$ of the product in order to conform to the method of presentation of the data in terms of $N/Z$ dispersion in this McGill Radiochemistry Laboratory. This procedure was originally used by Friedlander et al. (Fr 63) to circumvent situations arising out of the shell edge discontinuities. The fractional yields were obtained for a particular product at a specific energy by dividing the formation cross-section by the total chain yields read off from the plots given in Figs. 8-11. Table IX gives the fractional independent yields used for fitting the Gaussian-curves with the additional usual constraint.
### TABLE IX

Independent cross-section values used in charge dispersion curves

<table>
<thead>
<tr>
<th>$E_p$ (MeV)</th>
<th>$^{117}$In, (N/Z=1.387)</th>
<th>$^{115}$In, (N/Z=1.346)</th>
<th>$^{115}$Cd, (N/Z=1.395)</th>
<th>$^{111}$Pd, (N/Z=1.413)</th>
<th>$^{112}$Ag, (N/Z=1.382)</th>
<th>$^{111}$Ag, (N/Z=1.361)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\sigma$</td>
<td>$\sigma_f$</td>
<td>$\sigma$</td>
<td>$\sigma_f$</td>
<td>$\sigma$</td>
<td>$\sigma_f$</td>
</tr>
<tr>
<td>85</td>
<td>7.9 ± 1.2</td>
<td>2.6 ± 0.4</td>
<td>10.6 ± 1.6</td>
<td>12.3 ± 2.0</td>
<td>7.0 ± 1.0</td>
<td>2.6 ± 0.4</td>
</tr>
<tr>
<td>70</td>
<td>5.3 ± 0.8</td>
<td>0.9 ± 0.13</td>
<td>4.5 ± 0.7</td>
<td>9.0 ± 1.4</td>
<td>3.8 ± 0.5</td>
<td>1.5 ± 0.2</td>
</tr>
<tr>
<td>55</td>
<td>2.6 ± 0.5</td>
<td>0.2 ± 0.03</td>
<td>3.9 ± 0.6</td>
<td>4.8 ± 0.8</td>
<td>1.8 ± 0.3</td>
<td>0.8 ± 0.2</td>
</tr>
<tr>
<td>40</td>
<td>1.2 ± 0.2</td>
<td>0.05 ± 0.003</td>
<td>1.9 ± 0.04</td>
<td>3.9 ± 0.08</td>
<td>0.8 ± 0.02</td>
<td>0.2 ± 0.004</td>
</tr>
</tbody>
</table>
### TABLE IX (cont'd)

<table>
<thead>
<tr>
<th>( E_p ) MeV</th>
<th>( \sigma_{mb} )</th>
<th>( \sigma_{fly} )</th>
<th>( \sigma^*_{fly} )</th>
<th>( \sigma_{fly} )</th>
<th>( \sigma_{fly} )</th>
<th>( \sigma_{fly} )</th>
<th>( \sigma_{fly} )</th>
<th>( \sigma_{fly} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>0.4 ± 0.01</td>
<td>0.03 ± 0.0008</td>
<td>1.2 ± 0.03</td>
<td>2.0 ± 0.05</td>
<td>0.4 ± 0.009</td>
<td>0.08 ± 0.002</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>± 0.006 ± 0.002</td>
<td>± 0.005 ± 0.0002</td>
<td>± 0.3 ± 0.005</td>
<td>± 0.3 ± 0.008</td>
<td>± 0.06 ± 0.001</td>
<td>± 0.01 ± 0.003</td>
<td></td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>0.16 ± 0.003</td>
<td>0.05 ± 0.0015</td>
<td>0.3 ± 0.009</td>
<td>4.0 ± 0.025</td>
<td>0.18 ± 0.0029</td>
<td>0.05 ± 0.0025</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>± 0.04 ± 0.006</td>
<td>± 0.01 ± 0.0003</td>
<td>± 0.8 ± 0.001</td>
<td>± 0.8 ± 0.005</td>
<td>± 0.03 ± 0.004</td>
<td>± 0.008 ± 0.0005</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

**NOTES:**

\( \sigma_{fly} \) = fractional independent yield

\( \sigma^* \) = best estimated values (see text for details)
FIGURE 54

Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 85 MeV.
85 MeV
$^{238}\text{U}(p, f)$

Fractional Yield

$N/Z_p = 1.455$

FWHM = 2.36Z

$= 0.115 N/Z$
FIGURE 55

Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 70 MeV.
70 MeV
$^{238}\text{U} (p, f)$

$N/Z_p = 1.463$

$^{112}\text{Pd}$

FWHM = 2.16 Z

$= 0.11 \, N/Z$

Fractional Yield

N/Z

$^{115}\text{In}$

$^{115}\text{Cd}$

$^{112}\text{Ag}$

$^{117}\text{In}$

$^{113}\text{Ag}$
FIGURE 56

Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 55 MeV.
55 MeV
$^{238}u(P,t)$

$N/Z_p = 1.468$

FWHM = 1.96Z

$= 0.01 \frac{N}{Z}$

FRACTIONAL YIELD

$\text{III}_Pd$

$\text{II}_5 Cd$

$\text{II}_7 In$

$\text{II}_2 Ag$

$\text{II}_5 In$

$\text{III}_Ag$

$N/Z$

0.0001

1.30 1.40 1.50 1.60

0.001

0.01

1.0
FIGURE 57

Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 40 MeV.
$N/Z_p = 1.475$

FWHM = 1.92 Z

$= 0.098 N/Z$

$40 \text{ MeV}$

$^{238}\text{U}(p,t)$
FIGURE 58

Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 30 MeV.
$N/Z_p = 1.479$

FWHM = $1.74Z$

$= 0.093 N/Z$

$^{238}U(p,f)$
FIGURE 59

Charge distribution of nuclides in the symmetric mass region resulting from proton fission of natural uranium at 20 MeV.
that the right-hand portion of the Gaussian curve be drawn so that the sum of the independent cross-sections read from the curve approximate the measured cumulative yield of a nuclide measured in that chain.

The N/Z value at the peak of the charge dispersion curves and the mass number of that silver isotope having the neutron-to-proton ratio nearest the peak value were used to obtain the most probable charge, $Z_p$, for this mass. The values of N/Z at half-maximum obtained similarly from the charge dispersion curve were converted to Z values for the same mass to obtain the full-width at half-maximum (FWHM) in Z units.

(ii) Features

Characteristics of the charge dispersion curves become apparent from the listing of the parameters for these curves in Table X. These features conform with the findings of others in this energy region but working with different mass chains, i.e., the most probable charge $Z_p$ shifts to less neutron-rich nuclides with increasing bombarding energy (although the shift is less drastic than most of the studies made in the asymmetric mass region, Fig. 60) and especially the adjoining mass region ($A = 124-127$) as indicated by Miller and Yaffe (Mi 73). $Z_A$ values were estimated for the chain $A = 115$ from Coryell (Co 53), and these latter values have stood the test of time as evident from comparisons made by Diksić et al. (Di 74) and McGee et al. (Mc 72) with more recent measurements.
### Parameters of charge dispersion curves for $^{238}$U

<table>
<thead>
<tr>
<th>Proton Energy (MeV)</th>
<th>Full-width at half-maximum</th>
<th>Peak Position</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>N/Z</td>
<td>Z</td>
</tr>
<tr>
<td>85</td>
<td>0.115</td>
<td>2.36</td>
</tr>
<tr>
<td>70</td>
<td>0.111</td>
<td>2.16</td>
</tr>
<tr>
<td>55</td>
<td>0.101</td>
<td>1.96</td>
</tr>
<tr>
<td>40</td>
<td>0.098</td>
<td>1.92</td>
</tr>
<tr>
<td>30</td>
<td>0.093</td>
<td>1.74</td>
</tr>
<tr>
<td>20</td>
<td>0.088</td>
<td>1.28</td>
</tr>
</tbody>
</table>
Variation of $Z_A - Z_P$ function with bombarding energy

$^{238}\text{U}_1$, $^{238}\text{U}(p,f)$ - light-mass products

$^{235}\text{U}_1$, $^{235}\text{U}(p,f)$ - light-mass products

$^{238}\text{U}_s$, $^{238}\text{U}(p,f)$ - symmetric mass products

$^{238}\text{U}_h$ - heavy-mass products of $^{238}\text{U}(p,f)$ reaction

$^{232}\text{Th}_h$ - heavy-mass products of $^{232}\text{Th}(p,f)$ reaction

$^{239}\text{Pu}$, $^{237}\text{Np}$ - heavy-mass products of $^{239}\text{Pu}$ and $^{237}\text{Np}$
of Wing and Fong (Wi 64) and Dewdney (De 63).

A systematic correlation of the \((Z_A - Z_P)\) function with mass number at any specific incident proton energy is apparent. This is shown in the graphical representation (Fig. 61) of a compilation of data at 30, 50 and 85-MeV proton bombardment of natural uranium. For purposes of inter-comparison, the data of some earlier workers at the McGill Radiochemistry Laboratory were computer-fitted to a regular Gaussian curve and \(Z_P\) value evaluated on the basis prescribed by Friedlander et al. (Fr 63).

The \(Z_A\) and \(Z_P\) differential \((Z_A - Z_P)\) as a function of fragment mass indicates that fragments formed in the region of symmetric fission would carry more excitation energy than fragments resulting from the asymmetric mode of fission, yielding a larger number of post-fission neutrons. The \(Z_A - Z_P\) function relationship with incident proton energy (Fig. 60) indicates that the light fragments formed in the fission process in the energy range of experimentation (20-85-MeV) carry almost identical excitation energy. Also, the nature of the target seem to be inconsequential in the studies leading to the formation of light fission fragments. Consequently, excitation energy of heavy fragments would increase as bombarding energy of protons \((E_p)\) is increased and this could be one of the reasons leading to a decrease in the magnitude of \(Z_A - Z_P\) as \(E_p\) increases.

The magnitude of the FWHM of the charge dispersion curves themselves have proved difficult to interpret for inter-comparison of the charge-dispersion curves in different mass
Variation of \( (Z_A - Z_p) \) with mass number at a specific incident proton energy in the fission of natural uranium.

Legend:
1. Khan et al. (Kh 70), normalized
2. Present work
3. Miller et al. (Mi 73)
4. Dikšić (Di 74)
5. Parikh (Pa 67), normalized
6. Galinier (to be published)
\[ \begin{align*}
\Delta -30 \text{ MeV data} \\
\times -50-55 \text{ MeV data} \\
\bullet -85 \text{ MeV data}
\end{align*} \]
regions of work, and at different incident proton energies, due mainly to differing mechanisms leading to the products under consideration. However, it could be generally systematized that increased FWHM width with increasing incident proton energy in the 20-85-MeV region of work is now well established. Pate et al. (Pa 58) have attributed this observation to the competition between neutron evaporation and fission competing at each successive step and/or the effect of post-fission neutron evaporation. Since the same fissioning nuclei form both the heavy and light fragments, the role of post-fission neutron emission seems quite important. Miller et al. (Mi 73) from their experimental points near the peak of the charge-dispersion curves have suggested a more flat-topped curve than a Gaussian, and have fitted two over-lapping Gaussians to their experimental values above 30-MeV. This result implies that there may be a change from one to two separate distributions. This kind of conjectural analysis has been performed by Panontin and Porile (Pa 70), in their 11.5 GeV $^{238}$U$(P,f)$ work. This kind of analysis could not be performed from the data in the present work due to the small number of experimental points located on the wings of the charge distribution curves.

A more analytical approach towards the decomposition of the charge dispersion curves seems to be based more appropriately on a study of the ratio of the range of a product at two different energies by a thick-catcher recoil experiment. This ratio, called the "relative range" was recently determined
by Starzyk and Sugarman (St 73) for a range of products produced in the interaction of 11.5-GeV and 450-MeV proton interaction with uranium, and they have used it to decompose the charge dispersion curve for mass III based on the experimental data at these energies of Panontin and Porile (Pa 70). This relative range behaviour could be used for decomposing charge dispersion curves into components corresponding to a fission and a non-fission (spallation) mechanism.

The results available at the moment in the energy interval 170-720 MeV show that the charge dispersion width for mass chains about symmetric fission is relatively constant (between 2.8 and 2.9 charge units) and independent of bombarding energy, based on the data of Panontin and Porile (Pa 68), Pappas and Hagebø (Ha 66), Hagebø (Ha 69) and Alexander et al. (Al 63). A slow variation of average deposition energy $E^*$, in the symmetric mass region could account for the above observations. This is borne out by Panontin and Porile (Pa 70) on the basis of their results at 11.5-GeV proton bombardment of natural uranium that the average deposition energy $E^*$ leading to products in the symmetric mass region is not very different at 450-MeV than at 12-GeV, suggesting that the selectivity of the fission process for cascade events leading to the most probable product in GeV bombardments is quite similar to that found in the hundreds of MeV range of bombarding energy.

The recoil studies of Ramomoorthy et al. (Ra 70) covering the energy range of the present work, and leading to
some of the products in the symmetric mass-region show a significant variation in the excitation energy of primary fission fragments indicating a variable FWHM of the charge dispersion as the proton bombarding energy is increased from 20-MeV to 85-MeV.

(2) Excitation Functions of Nuclides Formed in the Symmetric Mass Region

Friedlander et al. (Fr 63) have shown that the excitation functions of almost all the nuclides under observation in this work should reach a maximum in the BeV-region of energy. In this work, the energy of the proton beam at which the cross-sections of the individual fission products reach their maxima, $E_p(\text{max})$, are plotted against the neutron-to-proton ratio of these fission products. Very recently Chang and Sugarman (Ch 74) have reported cross-sections for some Ag and Pd nuclides covered in this dissertation at a bombarding energy of 300-GeV in the fission of natural uranium. They conclude that the average cross section of all products is about lower at 300-GeV than at 11.5-GeV. Reported cross-sections for $^{111}\text{Pd}^m$ (i) and $^{112}\text{Pd}$ (c) show a maximum value around 3-GeV as indicated by Porile (Po 66), while the cumulative cross-section for $^{111}\text{Ag}$ shows a maximum around 11.5-GeV according to Panontin and Porile (Pa 70). In the present work, it was observed that the formation cross-sections of the various fission products increase monotonically with bombarding energy, therefore it was not possible to estimate
$E_p$ (max) of these products from experimental data in the range of energy considered.

(3) **Variation of $N/Z_p$ with Mass**

In this work it was assumed that it was not necessary to apply any corrections with respect to the variation of $N/Z_p$ with mass number as prescribed by Hogan and Sugarman (Ho 69) for higher energies. The composite graph showing the variation of $N/Z_p$ with mass number $A$ using the experimental data covering the energy range 20-85-MeV along with similar data from Pappas and Hagebø (Pa 69) and Wahl et al. (Wa 69) appear in Fig. 62. It is apparent that the above assumption is valid covering the mass region in this work. The slight variation at the lowest energy of experimentation, i.e. 20-MeV, is well within the experimental uncertainty of obtaining the data at that energy.

(4) **Most Probable Charge from UCD and ECD Mechanism**

$Z_p$ values for some of the fission products in the mass region of the present work were determined from record studies by Ramamoorthy et al. (Ra 70) according to the equations for UCD and ECD mechanisms given in Chapter I.

Despite small deviations in charges and masses of the cascade nuclei at different bombarding energies, the average charge 93.0 and the average mass 238.0 for the cascade nucleus was used at all bombarding energies. The calculation of
FIGURE 62

Variation of $N/Z_p$ with mass

The thermal neutron fission data are taken from Wahl (1969)
The 160-MeV proton fission data are from Pappas and Hagebø (1966)
Other data points from Radiochemistry Group at McGill (20–85-MeV)

- Khan et al. (Kh 70)
- Davies and Yaffe (Da 63)
- Parikh et al. (Pa 67)
- Miller et al. (Mi 73)
- Present work (1974)
THERMAL - NEUTRON FISSION
WAHL ET AL. (1969)

McGILL

160 MeV PAPPAS AND HAGEBØ (1966)

$\frac{N}{Z_p}$
the excitation energy of primary fragments which lead to the measured fission products needed the knowledge about the average fissioning nucleus. For the 25-MeV results, the pure compound nucleus $^{239}\text{Np}$ was used. The procedure adopted has been outlined previously in Chapter I. Proton evaporation was ignored. The average fissioning nucleus can lead to a number of primary fragments which in turn lead to the formation of the observed fission products. Corresponding to each fission product five trial fragments whose mass numbers are higher by 5 or less mass units, were chosen. Since proton evaporation was considered negligible, only neutron excessive isotopes of the observed fragments were chosen. The results are tabulated in Table X for the selected products in the mass region of the present study.

From the table, it could be observed that for $^{113}\text{Ag}$ and $^{111}\text{Ag}$, $(Z_p)_{UCD}$ is about one charge unit higher than that obtained through the ECD procedure at higher energies, while at lower energies both postulates predict identical values.

The calculated values from the recoil studies for A=112 are only slightly higher (less than half-charge unit) and nearest, at the highest bombarding energy, to the $Z_p$ determined with $A_p=115$ in the present work. Therefore, it is perhaps possible to argue qualitatively that the UCD mechanism provides a much better estimate of the $Z_p$ values in the region of symmetric fission products.
### TABLE X

Most probable charge from UCD and ECD mechanism (from Ramamoorthy et al. (Ra 70)).

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>$E_p$ (MeV)</th>
<th>$E^*$ (MeV)</th>
<th>$\langle A_f \rangle$</th>
<th>$\langle Z_f \rangle$</th>
<th>$Z_{p\text{ECD}}$</th>
<th>$Z_{p\text{UCD}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{111}$Ag</td>
<td>85</td>
<td>58.4±9.3</td>
<td>235.5</td>
<td>92.7</td>
<td>44.3</td>
<td>45.4</td>
</tr>
<tr>
<td></td>
<td>70</td>
<td>41.9±9.6</td>
<td>236.2</td>
<td>92.7</td>
<td>44.3</td>
<td>45.2</td>
</tr>
<tr>
<td></td>
<td>55</td>
<td>37.9±8.6</td>
<td>236.4</td>
<td>92.8</td>
<td>44.3</td>
<td>44.9</td>
</tr>
<tr>
<td></td>
<td>40</td>
<td>26.3±10.1</td>
<td>237.0</td>
<td>92.9</td>
<td>44.3</td>
<td>44.6</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>31.2</td>
<td>236.7</td>
<td>93.0</td>
<td>44.3</td>
<td>44.2</td>
</tr>
<tr>
<td>$^{112}$Ag</td>
<td>85</td>
<td>41.8</td>
<td>234.9</td>
<td>92.7</td>
<td>47.0</td>
<td>47.0</td>
</tr>
<tr>
<td></td>
<td>70</td>
<td>31.0</td>
<td>236.0</td>
<td>92.7</td>
<td>47.0</td>
<td>47.0</td>
</tr>
<tr>
<td></td>
<td>55</td>
<td>26.6</td>
<td>236.0</td>
<td>92.8</td>
<td>47.0</td>
<td>47.0</td>
</tr>
<tr>
<td></td>
<td>40</td>
<td>18.3</td>
<td>237.0</td>
<td>92.9</td>
<td>47.0</td>
<td>47.0</td>
</tr>
<tr>
<td>$^{113}$Ag</td>
<td>85</td>
<td>76.6±9.4</td>
<td>234.9</td>
<td>92.7</td>
<td>45.1</td>
<td>46.0</td>
</tr>
<tr>
<td></td>
<td>70</td>
<td>48.0±8.6</td>
<td>236.0</td>
<td>92.7</td>
<td>45.1</td>
<td>45.8</td>
</tr>
</tbody>
</table>

Notations:

- $E_p$ = bombarding energy
- $E^*$ = cascade deposition energy
- $A_f$ = mass of average-fissioning nucleus
- $Z_f$ = charge of average fissioning nucleus
- $Z_p$ = most-probable charge
(5) **Total Numbers of Neutrons Emitted**

The utilization of the McHugh & Michel (Mc 68) equation to estimate the number of neutrons emitted at a given excitation-energy has been already covered in Chapter I.

At a given excitation energy the variation of \( Z_p \) with mass number can be approximated by the variation of \( Z_A \) with mass number i.e. \( (\partial Z_p / \partial A)_E = (\partial Z_A / \partial A)_E \). Coryell et al. (Co 53) and recently Dikšić et al. (Di 74) have experimentally determined \( (\partial Z_p / \partial A)_E = 0.38 \pm 0.02 \) for isobaric studies along various mass chains. Fig. 63 shows a plot of \( Z_p \) versus average excitation energy of intermediate nuclei for the \(^{238}\text{U} + \text{p}\) system.

Total neutron emission rates \((d\nu_T / dE)\) are calculated as a first approximation from the expression,

\[
(d\nu_T / dE) = d\nu_h / dE + d\nu_l / dE
\]

where \( h \) and \( l \) refers to heavy and light fragments respectively. The assumption that \( \nu_h / \nu_l = 2 \) (Ya 69) for asymmetric fission modes could be certainly modified more appropriately to \( \nu_h / \nu_l = 1 \) for studies leading to products from a symmetric fission.

This would yield \((d\nu / dE)_h = 0.048 - \text{MeV}^{-1}\) leading to \((d\nu_T / dE)_T = 2 \times 0.048 = 0.096 - \text{MeV}^{-1}\), and the total number of neutrons (post-fission) have been calculated on this basis. The number of pre-fission neutrons have been calculated at different excitation energies using both the Vegas STEPNO code and a Monte Carlo evaporation code by Dikšić et al. (Di 74) for the same fissioning system from 30-85-MeV proton bombardment.
FIGURE 63

Variation of $Z_p$ with the average excitation energy of intermediate nuclei for the $(^{238}\text{U}+\text{p})$ system.
\[ U(p, f) \]

\[ \frac{\partial U}{\partial E_p} = 0.018 \text{ MeV}^{-1} \]

\[ \frac{\partial \Gamma}{\partial E} = 0.048 \text{ MeV}^{-1} \]

\[ \frac{\partial \Gamma_f}{\partial E} = 0.096 \text{ MeV}^{-1} \]

\[ \frac{\partial Z_p}{\partial E} = 0.011 \text{ MeV}^{-1} \]

\[ \frac{\partial \Gamma}{\partial E} = 0.095 \text{ MeV}^{-1} \]

EXCITATION ENERGY \((E^*)\) - MeV
energies. At 20-MeV the number of cascade neutrons were calculated using a pure compound nucleus mechanism. The final results are tabulated in Table XI. The total number of neutrons indicated in the table support the general attribution that symmetric fission is a result of fission events with higher than average excitation energy; therefore, one would expect more total neutrons emitted than in the asymmetric fission, as could be confirmed by comparison of data in the mass region $A = 130-135$ reported by Dikšić et al. (Di 74), as given in Table XII.
TABLE XI

Total number of neutrons in the $^{238}\text{U}(p,f)$ reaction, study of products in the symmetric and near-symmetric mass region

<table>
<thead>
<tr>
<th>$E_p$</th>
<th>20 MeV</th>
<th>30 MeV</th>
<th>40 MeV</th>
<th>55 MeV</th>
<th>70 MeV</th>
<th>85 MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;E&gt;</td>
<td>(25.2)</td>
<td>(26.7)</td>
<td>(32.09)</td>
<td>(39.62)</td>
<td>(45.44)</td>
<td>(53.93)</td>
</tr>
<tr>
<td>$\nu$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total number of neutrons $\nu_t$</td>
<td>3.1</td>
<td>3.9</td>
<td>5.1</td>
<td>6.7</td>
<td>8.2</td>
<td>9.6</td>
</tr>
<tr>
<td>Prefission cascade neutrons $\nu_c$</td>
<td>-</td>
<td>0.29</td>
<td>0.45</td>
<td>0.60</td>
<td>0.74</td>
<td>0.81</td>
</tr>
<tr>
<td>Prefission evaporated-neutrons $\nu_E$</td>
<td>0.72</td>
<td>1.12</td>
<td>1.63</td>
<td>2.38</td>
<td>3.09</td>
<td>3.63</td>
</tr>
</tbody>
</table>
TABLE XII

<table>
<thead>
<tr>
<th>$E_p$ (MeV)</th>
<th>30</th>
<th>40</th>
<th>50</th>
<th>70</th>
<th>85</th>
</tr>
</thead>
<tbody>
<tr>
<td>$&lt;E&gt;$</td>
<td>26.7</td>
<td>32.1</td>
<td>35.8</td>
<td>45.4</td>
<td>53.9</td>
</tr>
<tr>
<td>$v_T$</td>
<td>3.9</td>
<td>4.8</td>
<td>5.7</td>
<td>7.1</td>
<td>7.6</td>
</tr>
<tr>
<td>$v_C$</td>
<td>0.29</td>
<td>0.45</td>
<td>0.59</td>
<td>0.74</td>
<td>0.81</td>
</tr>
<tr>
<td>$v_E$</td>
<td>1.12</td>
<td>1.63</td>
<td>2.16</td>
<td>3.09</td>
<td>3.63</td>
</tr>
</tbody>
</table>

where: $E_p =$ proton bombardment energy  
$<E>$ = average excitation energy of the intermediate-nuclei  
$v_T =$ estimated total number of neutrons emitted  
$v_C =$ number of cascade neutrons  
$v_E =$ number of evaporated neutrons
VI. SUMMARY AND CONTRIBUTION TO KNOWLEDGE

Charge dispersion curves around the most probable charge \( Z_p \) in the fission of \(^{238}\text{U} \) with protons of energy 20-85 MeV were constructed on the basis of radiochemical studies on the fission products in the symmetric and near-symmetric mass regions.

Absolute isotopic yield data for the relevant nuclides were obtained by monitoring the incident proton beam by means of the reaction \(^{65}\text{Cu}(p,pn)^{64}\text{Cu} \). The absolute cross-section for the latter reaction was pre-determined at each energy of experimentation using the external beam facility of the McGill synchrocyclotron coupled to a Faraday cup charge collection assembly.

Rapid chemical separation techniques were worked out in the present study to yield products of sufficient radiochemical purity and independent cross-section data of \(^{111}\text{Ag} \) have been reported for the first time.

The experimentally-determined \( Z_p \) values were deduced directly from the charge dispersion curves. The recoil data in this mass region at these energies seem to verify the results of this work, and establish a preference for the unequal charge distribution hypothesis in this mass region and energy range.

The \((Z_A - Z_p)\) variation with incident proton energy is intermediate between the values obtained from light mass
fission fragments of natural uranium and those obtained from the heavy mass fission fragments.

The estimated total number of neutrons emitted in the symmetric mode of fission (both pre-fission and post-fission) are higher than in the process of asymmetric fission, attributable to fission events with higher than average excitation energy. The variation of average deposition energy is relatively more in this energy range leading to symmetric products than that in the very high energy fission. The latter seems to result from relatively slow average deposition energy variation and possibly more than one mode of fission.
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