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A. INTRODUCTION

(a) GENERAL

Nuclear reactions, in which a highly energetic particle (or photon) hits a nucleus and knocks out one or more nucleons or clusters of nucleons, are called 'spallation' reactions. The study of spallation reactions, in particular the study of 'excitation functions' - variation of production cross section with incident energy - is a simple and often very useful means of investigating the mechanism of nuclear reactions. As discussed later, excitation functions for spallation reactions may be divided roughly into two classes, those for which the threshold is in the region where the Bohr compound nucleus mechanism⁽¹⁾ is said to be applicable (< 50 Mev), and those with the threshold in the Serber⁽²⁾ knock-on region (> 50 Mev). Fig. 1 reported by Meadows⁽³⁾ shows a good example of the compound nucleus (1) type of In this figure it is seen that the cross section reaction. for a typical low-energy reaction (p,n) rises sharply to a maximum and declines rapidly because of increased competition from other reactions which become energetically possible as the energy is raised. Fig. 2, taken from the same publication, is a case where one can see compound nucleus formation at low energies (ζ 50 Mev). At higher bombarding energies the curve levels off, which is to be expected as the knock-on interaction⁽²⁾ replaces that of compound nucleus formation.

Figure 1

MEADOWS⁽³⁾ VALUES OF THE CROSS SECTIONS FOR THE Cu⁶³(p,n)Zn⁶³ REACTION

,





MEADOWS:(3) VALUES OF THE CROSS SECTIONS FOR THE Cu⁶⁵(p,pn)Cu⁶⁴ REACTION



3a

The work reported here is concerned with the study of excitation functions of erbium, when bombarded with protons ranging from 6 to 87 Mev. In addition, search for new nuclides in the more neutron-deficient regions, reconfirmation of nuclear properties of some of the product nuclides, comparison of theoretically predicted cross sections, and experimental cross sections wherever it is possible, and finally, providing a test for the compound nucleus formation in the low-energy reactions were also some of the important aspects of the present work.

(b) COMPOUND NUCLEUS THEORY

Nuclear reactions induced by particles of energy \angle 50 Mev are usually considered in the framework of compound nucleus theory, as originally proposed by Bohr⁽¹⁾. Bohr's concept has been extended by others, and the general features of the resulting statistical model have been discussed by Blatt and Weisskopf(4). In this model, a nuclear reaction is described as taking place in two stages. The first stage is a strong interaction between the projectile and the target nucleus in such a manner that the energy and the momentum of the incident particle are quickly distributed over the whole system, resulting in the formation of a compound nucleus. The second stage is the break-up or decay of the compound nucleus in a manner predicted by statistical theory. The two stages are considered as separate and independent processes, so that the break-up of

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the compound nucleus should not depend on the particular manner in which it was formed, but only on its overall properties such as energy and angular momentum. One would expect that this concept of compound nucleus formation should be applicable to spallation reactions as well as fission.

Compound nucleus theory predicts that if one compares excitation functions for reactions induced by two particles A and A' - the comparison being made at the same excitation energy - the ratios of the corresponding cross sections should be the same. That is, if B represents the emission of a different number of particles, then

 $\frac{\sigma(A,B)}{\sigma(A,B')} = \frac{\sigma(A',B)}{\sigma(A',B')}$

where $\sigma(A,B)$ represents the cross section for the reaction

X + A = Y + B.

This interesting test was applied for the first time by $Ghoshal^{(5)}$ who bombarded Ni^{60} with helium ions and Cu^{63} with protons to produce the compound nucleus Zn^{64} . He experimentally verified that the relation:

(p,n): (p,2n): $(p,pn) = (\alpha,n)$: $(\alpha,2n)$: (α,pn) was valid at each excitation energy. This test strikingly verified the assumption of independent formation and decay of the compound nucleus. A similar test for the compound nucleus Po²¹⁰ has been applied by John⁽⁶⁾ who compared the excitation functions for Pb²⁰⁶ which he bombarded with helium ions, with the excitation function studies of Kelley⁽⁷⁾ who bombarded Bi²⁰⁹ with protons. John found that the ratios of corresponding cross sections were in agreement but that the excitation functions were displaced in energy by an amount slightly different from the amount one would calculate to match the excitation energies. Gibson⁽⁸⁾ also tested the compound nucleus formation of Am^{241} by comparing the excitation functions of Pu^{239} which he bombarded with deuterons, with the excitation functions for Np²³⁷ which he bombarded with helium ions.

Test for compound nucleus formation of Zn⁶⁴ served as a good example in the region of medium weight Similarly Po²¹⁰ and Am²⁴¹ have served as good elements. examples in the region of non-fissile heavy elements and fissile heavy elements respectively. We have attempted to test the compound nucleus formation of Tm¹⁶⁹, that is, in the region of medium heavy elements. The details will be described later. However, there is experimental evidence indicating the importance of other processes rather than compound nucleus formation at energies less than 50 Mev. Energy and angular distribution of the reaction products which are in disagreement with predictions from the statistical theory have been measured (9,10,11,12). The probability for charged-particle emission has often been found to be much larger than expected (13,14) from statistical theory considerations. Whether these discrepancies are due to failure of the statistical model or simply to the contributions of other reaction mechanisms such as direct

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interaction is not yet understood.

In 1956 Weisskopf⁽¹⁵⁾ suggested that nuclear reactions can be described by a three-stage process. The first stage he termed the 'independent particle stage', and it describes the stage in which the incident particle, although influenced by the nucleus, can be considered as distinct from the target nucleus. The second stage is reached when the incident particle, removed from the entrance channel, forms the 'compound system'. This concept of a compound nucleus is a more general one than that used in the Bohr theory. The compound system includes the compound nucleus, but also includes states created by direct collisions, or by surface excitations, etc. The third stage of the reaction is the break-up of the compound system into the residual nucleus and the emitted particle or particles. If, in the compound system, the incident particle shares its energy with all its constituents and forms a compound nucleus, then the third stage should take place as predicted by statistical theory. However, such processes as surface direct interactions, volume direct interactions, multiple collisions and collective excitations can also take place in the compound system stage and lead directly to the third stage, without the formation of the compound nucleus. Although this three-stage mechanism of nuclear reaction provides a convenient frame work in which to discuss low energy nuclear reaction mechanisms, there is yet much to be learnt about the details of what happens in

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the various stages.

(c) KNOCK-ON CASCADE THEORY

In general the compound nucleus theory explains much about nuclear reactions that occur at energies below 50 Mev, but it does not explain the reactions that occur at Serber⁽²⁾ was the first to explain energies above 50 Mev. theoretically the observed phenomena in the high energy reactions (> 50 Mev). He pointed out that at high energies the collision time between the incident particle and a particle in the nucleus is short compared to the time between collision of the particles in the nucleus. Thus, the first step in the reaction can be considered as the collision of the incoming particles with a single nucleon in the nucleus. This collision will not be exactly analogous to the interaction of nucleus in free space, since the Pauli exclusion principle will exclude those encounters with small momentum transfers, the allowed collisions result in somewhat higher momentum transfers. At high energies there is even a finite probability that the bombarding particle traverses the nucleus without any interaction. That is, the nucleus begins to be transparent to the bombarding particles, and the compound nucleus picture is not applicable. When collisions do occur the average momentum transferred to the struck nucleon is small. Since the struck particles have much lower energy and shorter mean free paths, it is rather likely that they, in

turn, will undergo further collisions and that this energy will be distributed over many nuclear particles, thus contributing to the excitation of the nucleus. The incident particle in most cases will still retain most of its energy and may emerge from the nucleus without a second collision. However, if the first collision occurs near the centre of the target nucleus, there is a chance that the bombarding particle will make a second, a third or even more collisions before it is finally captured or escapes with much reduced energy.

The first part of the reaction is referred to as the nucleonic cascade, the 'knock-on' cascade, or simply as the cascade. It may consist of a single collision or of many collisions, including those made by the original particle and those made by the struck particles while escaping or being captured by the nucleus.

By the end of the cascade, which takes place in a very short period of time ($\sim 10^{-21}$ seconds) the interacting target nuclei will be converted to a distribution of product nuclei excited to a variety of excitation energies. There are a variety of possibilities ranging from the bombarding particle emerging with most of its energy in-tact to loss of the entire incident energy to the nucleus. From the excited state the nucleus dissipates its energy by a process called 'evaporation'. The course of a particular reaction is determined first by the amount of excitation energy deposited during the knock-on phase, and secondly, by the manner in which this excitation is given up during the evaporation phase, by the number of particles evaporated.

Serber's theory was able to give a qualitative explanation of high energy nuclear reactions. That is, it accounted for the high energy prompt particles emitted in the forward direction as a result of the cascade and explained the high yields of simple reactions like (p,2n), (p,3n) and (p,4n) etc. as being produced by collisions in which one or two of the prompt particles carry off most of the energy, leaving the residual nucleus enough excitation energy to boil off one or a few more nucleons. Serber's mechanism seems to be applicable for bombarding energies of a few hundred Mev or more. Thus one could see compound nucleus formation until about 40 Mev with a gradual transition to the knock-on cascade type as the energy is increased. However, it should be pointed out that this differentiation into two distinct types is certainly an over-simplification.

(d) SPALLATION PRODUCTS

With the advent of high energy particle accelerators, a large number of spallation yield studies have been made at various energy ranges. A variety of target elements have been bombarded with various particles. Miller and Hudis⁽¹⁶⁾ have given an excellent summary of this work. The cross sections, or at least the relative yields of different products from a given bombardment, are of interest. In general, the products in the immediate neighbourhood of the target element are found to be in highest yields.

The spallation yields cluster quite strongly in the region of stability in the case of medium-weight elements, and somewhat more to the neutron-deficient side of stability for heavier elements. This is what one would expect from evaporation theory. The experimental evidence in the case of medium-weight elements has been given by several authors, a typical reference being that of Rudstam⁽¹⁷⁾. In the case of non-fissile heavy elements, typical examples are Bell and Skarsgard⁽¹⁸⁾ and Nervik and Seaborg⁽¹⁹⁾.

Another important aspect of spallation studies is the mass yield curve. Fig. 3 provides a very good example of the difference in yields at low energies to those at high energies. At high energies fission competes with spallation, with the result that spallation products become less prominent and fission products become prominent, as is evident from Fig. 3. Erbium oxide has been bombarded by several authors in search of new activities, but not for yield studies. The pertaining references will be mentioned in the part of this thesis dealing with results. In the present work an attempt has been made to study the yields of various spallation products of erbium.

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Figure 3

CROSS SECTIONS AS A FUNCTION OF PRODUCT

MASS NUMBER FOR THE INTERACTION OF

LEAD OR BISMUTH WITH PROTONS OF

VARIOUS ENERGIES. FROM MILLER AND HUDIS⁽¹⁶⁾



(p,xn) reactions (x = 1,2,3,4) in non-fissile heavy elements have been intepreted by Jackson⁽²⁰⁾ in terms of compound nucleus formation and a simple neutron evaporation chain. The main assumptions in Jackson's model calculations are:

- (i) If it is energetically possible for a neutron
 to be evaporated, a neutron will be evaporated.
- (ii) The competition from the other modes of de-excitation (e.g. proton knock-out) can be neglected.
- (iii) That the neutron energy spectrum is given by $CE \exp^{(-E/T)}$, where C is a normalization constant and E the kinetic energy of the neutron and T the nuclear temperature.
- (iv) That the nuclear temperature is independent of excitation energy.

Jackson compared theoretically-predicted excitation functions with the experimental curves of Bell and Skarsgard⁽¹⁸⁾ and showed good agreement between theory and experimental results. Ramler et al.⁽²¹⁾ compared their experimental results for the reactions $Bi^{209}(\alpha,2n)$, $Bi^{209}(\alpha,3n)$, $Bi^{209}(\alpha,4n)$ and $Bi^{209}(d,n)$, $Bi^{209}(d,2n)$ and $Bi^{209}(d,3n)$ with the predictions of Jackson's evaporation model, and got good agreement except for a high energy 'tail' which they attributed to a stripping process. Jackson's calculations have been extended by Vandenbosch et al. (22) for cases where there is a competition between fission and neutron evaporation, and they could compare their experimental results for the reactions $U^{233}(\alpha,xn)$ and $U^{235}(\alpha.xn)$ with the calculated values on the basis of a modified Jackson's evaporation model. Wing et al. (21a) studied the reactions $U^{238}(\alpha, xn)$ and $U^{233}(d, xn)$ and got good agreement between experimental and calculated values. Although Jackson has applied his calculation to the nonfissile heavy elements, in the present work his calculation has been applied to medium heavy elements. Thus comparison has been made between experimental and calculated results based on Jackson's model for the reactions $Er^{168}(p,n)$, $Er^{168}(p,2n)$, $Er^{170}(p,3n)$, and $Er^{170}(p,n)$. The details of the theoretical calculations and comparison with the experimental values are described in the latter part of this thesis.

(f) MONTE CARLO CALCULATIONS

(i) <u>Nuclear Cascade Calculations</u>

Serber's theory of knock-on cascade mechanism was treated quantitatively by Goldberger (23) who used the Monte Carlo technique of Ulam and Von Neumann(24) to follow the cascade. Since then many other workers (17,25,26,27,28,29,30,31,32) have used the method for a variety of reaction conditions and each has added some improvements to the techniques. Metropolis et al. (31,32) followed a thousand cascades for several representative target elements and for a variety of bombarding energies. Their treatment is considered to be more complete and elaborate.

The Monte Carlo method consists of choosing a model for the nucleus and following the cascade step by step, until the particle is captured or the particle escapes from the nucleus. The calculations are of the same general type and each employs the same nuclear model (degenerate Fermi gas in a sharply defined potential).

The cascade calculations consist of the following steps:

- (i) Determination of energy of the incident particle, e.g. proton.
- (ii) Determination of the point where the proton enters the nucleus.
- (iii) Determination of the place for the first collision.
- (iv) Determination of the collision partner (proton or neutron).
- (v) Determination of the scattering angle.
- (vi) Construction of the new particle paths if the collision is not forbidden by the exclusion principle.
- (vii) If the collision is not permitted, the particle is returned to the collision site and given another chance to escape the nucleus or undergo another collision.

The above steps are applicable to the knock-on

calculation for incident particles of energies below the meson production threshold. In the case of incident particle energies above the meson threshold, the nuclear cascade becomes more complex and the attempt to follow it is more difficult.

Although it is easy to realize that the complete cascade calculation is still a relatively crude attempt to deal with a very complicated process, yet it can give a lot of information as listed below.

- (i) The nature and energy of the particle going into the nucleus.
- (ii) The number of collisions inside the nucleus.
- (iii) The nature and number of the outgoing particles.
- (iv) The atomic number and mass number of the residual nucleus.
- (v) The energy and angular distribution of the outgoing particles.
- (vi) The excitation energy of the residual nucleus.
- (ii) Evaporation Cascade Calculations

At the end of the nucleonic cascade, the target nucleus has been converted into a highly-excited nucleus with a different composition of neutrons and protons. Le Couteur (33) and Fuzimato and Yamaguchi (34) used Weisskopf's (4)evaporation formalism to calculate analytically the average behaviour in the de-excitation process.

Rudstam⁽¹⁷⁾, Dostrovsky et al.⁽³⁵⁾, and Friedlander et al.⁽³⁶⁾ used the Monte Carlo method, since various probabilities can be calculated afresh after the emission of each individual particle and the necessary nuclear constants (mass number, nuclear charge, nuclear radius, level density parameter, potential barrier, etc.) can be adjusted to the most appropriate values for each step of the evaporation cascade. The process is repeated until the initial excitation energy is almost all removed.

Dostrovsky et al. computed 1,000 cases, each for a wide variety of initial nuclei.

The general information available from each series of cascades may be summarized as follows:

- (i) The average number of each type of evaporated particle.
- (ii) The average energy spectrum for each type of particle.
- (iii) The A, Z distribution of the residual nuclei.

Although a few discrepancies have been observed, detailed comparisons of theory and experiment can be made if Monte Carlo calculations of the nuclear cascade and the evaporation stage are combined.

In the present work, although the Monte Carlo calculations were not done, an attempt has been made to see how experimental values compared with the values available in the literature.

(g) VARIOUS EXPERIMENTAL METHODS FOR SPALLATION STUDIES

Spallation reactions of various target elements

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have been studied at various bombarding energies ranging from a few Mev to a few Bev employing various techniques. Several types of bombarding particles, e.g. protons, deuterons, alphas, neutrons, and pions, etc., have been used. The techniques used in the spallation reactions include radiochemistry, mass spectroscopy, nuclear emulsions and counters.

Radiochemical methods are often used to determine the formation cross sections of radioactive nuclides in spite of the inherent experimental difficulties which lead to results usually good only to 15 - 30%. Very briefly, the technique involves irradiation of a target with a known number of particles, chemical separation and purification of the desired element, and determination of the number of radioactive atoms present in the purified sample by some sort of radiation detection apparatus. According to the suitability of the half-lives of the product nuclides, it is necessary to work out a separation in time, with Sometimes it is difficult to follow reasonable yields. the decay of the spallation products owing to the presence of several isotopes with similar half-lives in the desired The half-lives of the desired nuclides should element. not be too short or too long. Growth of daughters in the parent fraction may sometimes limit the processing of the decay data. The present work was done using radiochemical techniques.

Mass spectrometry may be used for the accurate

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determination of formation cross sections. This method may sometimes be used for radioactive products with reasonably short and very long half-lives, as well as for stable species. Although this technique has been extensively used for studying fission yields, it has not been much used for spallation studies due to the inherent Recently, however, formation cross problems involved. sections at Bev energies were determined by Gordon and Friedman⁽³⁷⁾ who estimated mass spectrometrically the yields of cesium isotopes from the 3-Bev fission products Schaeffer and Zäringer⁽³⁸⁾ determined the of gold. formation cross sections of rare gases formed in high energy fission using mass spectrometry techniques.

The counter telescope method may be used to determine the energy and angular distribution of emitted charged particles by allowing the particles to pass through a group of counters which define a direction with respect to the incident beam.

Nuclear emulsion techniques make use of a different approach. In this technique one looks at individual reactions, and may determine the relative number, nuclear charge, energy, and angular distribution of charged particles emitted during the reaction. Disadvantages of this method are that the estimation of absolute cross section is difficult and the neutrons emitted in a reaction are not detected.

Amongst the above methods, the radiochemical

technique has been chosen for the present work.

(h) ION EXCHANGE SEPARATION OF RARE EARTHS

One of the important steps in studying the spallation products by radiochemical techniques is the separation and purification of the desired element from the spallation products (that is, from the monitor and other heavy rare earths). In this connection the previous literature pertaining to individual heavy rare earth separations is described.

For a long time one of the most difficult and challenging problems in inorganic chemistry has been the separation of the rare earths* from each other. The introduction of cation exchange techniques which took place on the Manhattan project, and which is due to the working groups of Boyd, Cohn, and especially of Spedding, is largely responsible for the recent development of macro and

*The rather peculiar group of elements with atomic numbers from 57-71 is mostly designated by the rare earth group. J.K. Marsh proposed the name 'Lanthanons' /J.K. Marsh, Quarterly Reviews, 1, 126 (1947)7. Yttrium, while not belonging to that group, occurs with and behaves chemically like a rare earth and will be treated as an 'extra' member of that group. In addition to the name Lanthanons, modern literature includes the names: Lanthans, Lanthanates, Lanthanides and Lanthanoids. Based on atomic and geochemical reasons [I.D. Mainsmith, Nature, 120, 853 (1927); W. Klemm, Angew chem. 51, 77 (1938)7 rare earths are distinguished between the 'cerium group' (also known as light rare earths) La-Eu and the 'yttrium group' (also known as heavy rare earths) Gd-Lu, including yttrium in the latter.

micro quantities of rare-earth separations. An excellent history of this development of the separation of these elements has been given by Johnson, Quill and Daniels (39) and Spedding et al.⁽⁴⁰⁾. As would be expected from similarities in inner and outer shell atomic configurations, in aqueous electrode, gaseous ionization potentials $(M \Longrightarrow M^{+3} + 3e^{-})$, and ionic and atomic sizes, all of the lanthanide elements form the same general types of compounds and these compounds have very similar properties. However, the gradation of certain compounds, in which the oxidation state of each lanthanide is +3, is of sufficient magnitude to permit the separation. As have been pointed out by Nachod and Wood⁽⁴¹⁾ and emphasized by Schubert, Boyd and Adamson⁽⁴²⁾, the hydrated ionic radii govern the ion exchange separation. The series of hydrated ionic radii are just the reverse of the crystal radii, since the smaller the ion, the greater the tendency to hydration. The stabilities of complex ions containing the tripositive lanthanides increase with increasing atomic number, (that is, the hydrated ionic radii also increase with increasing number) due to well-known lanthanide 'contraction effect'. This explains why rare earths are eluted in reverse order of the atomic number (Lu -- La) in the so-called cationexchange separation process.

In brief, the ion exchange procedure depends upon adsorption of the mixed tripositive lanthanides on a small

portion of a column of cation exchanger, followed by elution with a complexing ion such as citrate, lactate, or α -hydroxy isobutyrate, etc. Although the tendency of a cation in solution to exchange with, for instance, hydrogen ion on the exchange resin increases with cation size, the opposing tendency to form complex ions with citrate (or any similar ion) increases much more rapidly with decreasing cation size. As the solution passes through the exchange column, a series of competitive equilibria between the ions on the exchanger and in complex form are established. These lead to progressive enrichment of the smallest (most highly complexed) lanthanide in the material passing down If several chemical and physical controlling the column. factors (which will be described later on) are adjusted to optimum separation condition, separation into bands ultimately occurs and, if the eluate from the column is collected in fractions, the various lanthanide ions appear separately in reverse order of atomic numbers (lutetium first and lanthanum last).

Since the present work is concerned with the quick separation of milligram quantities of heavy rare earths, only relevant references are discussed. Ketelle and Boyd (43,44) were the first to develop a quick micro scale separation of heavy rare earths as well as light rare earths, using the recently developed resin Dowex-50 (270-325 mesh). The elution temperature was kept at 100° C. The eluant was 4.75% citrate of pH 3.2 and 3.45. They

discussed the importance of temperature and careful loading in attaining good separations.

Although Spedding's group worked on macro-scale separations of rare earths at room temperature, employing citrate as eluant and Amberlite IR-120 as resin column material, still all the controlling factors discussed by him are applicable for any type of rare-earth separation. According to Spedding⁽⁴⁵⁾ the important variables which influence purity, zone shape and degree of separation are (1) pH, (2) citrate concentration (any complex ion used as eluant), (3) column load or rare-earth concentration, (4) flow rate, (5) dimensions of the column, (6) particle size of the resin, (7) temperature, and (8) mould growth in the eluant solution.

Since then, in connection with the interest of studying nuclear properties of rare-earth activities in fission products, a gratifying success has been achieved in the separation chemistry of the rare earths. Freiling and Bunney⁽⁴⁶⁾ described a method for the separation of rare earths from the fission products employing lactic acid of pH 3 as eluant and at 87° C temperature.

Stewart (47) developed a quick separation of heavy rare earths using glycollic acid as an eluant. In another paper by Stewart (48), he described the experimental arrangement for the glycollic acid method of rare-earth separation and compared the separation factors from the literature data of several other eluants, reported by several authors.

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Various eluants: lactate, EDTA, tartrate, citrate, malate glycollate; in some cases at room temperature and some at high temperatures (87° and 100°C) have been compared. Vickery⁽⁴⁹⁾ also compared the elution curves obtained, using several complexing agents: acetate, citrate and EDTA. Mayer and Freiling⁽⁵⁰⁾ also compared several complexing agents and proved EDTA > lactate = glycollate > malate > citrate which are placed in above order of preference. Fitch and Russel^(51,52) employed amino acetic acids for the rare-earth separations.

Apart from the above few references, several authors separated rare earths with improved conditions using those eluants that are described above mainly by a heated cation exchange column of Dowex 50 x 12 (fine mesh). Notable improvements are the concentration gradient elution of Bunney et al. (53), temperature gradient elution of Stewart (54), pH gradient elution of Nervik (55). These three gradient methods depend on the fact that the concentration of the complexing ion can be controlled for optimum separations by varying one of the three variables (pH, temperature, concentration) keeping the others constant.

Another recent eluant reported in the literature by Choppin and Silva⁽⁵⁶⁾ is α -hydroxy isobutyric acid. They compared the separation factors of EDTA, citrate, lactate, and isobutyrate and showed that isobutyrate is comparable to EDTA and more efficient than even citrate and lactate. They attribute the merit of isobutyrate to the

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replacement of hydrogen by a methyl group on the alpha carbon position.

For the present work, isobutyrate has been preferred because of the better separation factors reported by Choppin and Silva⁽⁵⁶⁾ when they separated Dy, Tb, Cf, Gd, Eu, Sm, Cm, and Pm. EDTA has been discarded because of its low solubility at low pH values which is not very useful for the separation of heavy rare earths. Of course, EDTA is good for light rare earths.

The present work was performed with dilute solutions of ammonium salt of α -hydroxy isobutyric acid as an eluting agent as developed by Choppin and Silva⁽⁵⁶⁾, using the method of Ketelle and Boyd⁽⁴³⁾ and the set-up of Harvey, Choppin, Thompson, Silva and Seaborg⁽⁵⁷⁾. The exact control of various factors for quick and pure separations of individual heavy rare earths required for the present work will be described later.

(i) MEASUREMENT OF RADIOACTIVITY

(i) <u>General</u>

For measurement of alpha, beta or gamma radiations, techniques involving the detection and measurement of individual particles or photons are almost universally employed. Among the gas ionization devices are the pulse ionization chamber, proportional counter and the Geiger-Müller counter. Scintillation methods are available for measuring alpha and beta particles, but are especially popular for gamma-ray

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measurement. Some of the principles involved in these measurements are described in the following paragraphs, since together they form the basis of practically all the common measurement techniques. Photographic techniques involving the use of nuclear emulsions may also be used for the detection and identification of individual particles.

Ionization chambers may be used as detectors for individual particles. Such chambers have the advantage of high stability and are frequently used for the most accurate work. They do, however, require the use of highly stable, sensitive electronic equipment.

The proportional counter takes its name from the proportionality between the output pulse and the initial ionization. The design of the counter and the voltage applied are such that a very high voltage gradient exists in the vicinity of the positive electrode. Under this voltage gradient, the liberated electrons undergo a high acceleration and are themselves capable of further Under suitable conditions gas multiplications ionization. of 1,000 or more are possible, the total ionization (and hence the pulse size) being strictly proportional to the initial ionization. The following diagram shows the effect of increasing voltage on the current produced, and the different regions which are commonly used for the measurement purposes.

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In the proportional region, since the output pulse is proportional to the number of electrons in the initial ionizing event, one can discriminate between particles of varying specific ionization.

The Geiger-Müller counter is a popular type of radiation detector, since, within its limitations, it is capable of detecting alpha, beta and gamma radiations and does not require the use of a high-gain amplifier.

Scintillation methods have, in the last few years, gained rapid popularity, especially in the measurement of gamma emitters. The scintillation detector is composed of a 'scintillator' which responds to incident radiations by emitting photons in the light region, and a photomultiplier which amplifies this. The introduction by Hofstadter of thallium-activated sodium iodide brought about an important advance in the application of scintillation techniques for gamma measurement and energy determination.
In the present work alpha and beta particle measurements, gamma and X-ray radiation measurements were needed. In this connection, the principles involved in each type, in particular mostly of gamma and X-ray measurements, are given below.

(ii) Alpha measurements

The simplest device for measuring this type of radiation consists of two charged electrodes and an external circuit for measuring the current produced.

Another possible technique is to utilize the individual pulses of the current produced by each ionizing event. These pulses can be amplified electronically and pulses per unit time are measured.

Amongst the instruments that can be used are the ionization chamber, alpha scintillation counter and gas-flow proportional counter. These instruments can maintain low background counting rates and high efficiencies. In the present work, a gas-flow 2π -proportional counter has been used for alpha measurement. The actual alpha measurement and conditions for operation will be described later, (B, section g).

(iii) Beta measurements

For this type of measurement, scintillation counters, gas-flow proportional counters, and the standard Geiger-Muller tube can be used. The scintillation and gasflow proportional counters are both proportional counters; that is, by suitable adjustment of the operating voltages, the counters can be made to respond to different beta particle energies. This may be advantageous in discriminating unwanted isotopes but causes difficulties in that operating conditions must be fixed within very small tolerances.

In the present work a typical gas-flow 2πproportional counter has been used for beta measurement. The operating conditions, and the actual measurements will be described later (B, section g).

(iv) Gamma and X-ray measurement

These are electromagnetic radiations. X-rays originate from transitions in the electronic levels, and the radiations are emitted when outer electrons fall into empty inner shells of the atom. An electron from an inner shell of an atom can be removed by ionizing interaction with a particle (alpha or beta) or by absorption of a sufficiently energetic photon to remove it from the atom, e.g. by a gamma ray.

Gamma rays interact with matter in the following ways:

Photoelectric effect:

In this interaction the gamma ray gives all of its energy to an electron, detaches it completely from its atom and gives it kinetic energy. In short, the energy of the electron, resulting from such interactions, corresponds to the full gamma-ray energy minus the binding energy of the electron.

Compton effect:

The gamma ray may collide with an electron in the same fashion as billiard balls collide. Thus, only part of the original gamma-ray energy is given to the electron and the rest is removed by a scattered gamma ray. This type of interaction gives rise to a continuous spread of secondary electron energies and hence a continuum of pulse heights corresponding to the electron energies.

Pair production:

A gamma ray, the energy of which is greater than the mass of two electrons (2 x 0.51 Mev) may be totally converted into a positive (positron) and a negative (negatron) electron. These electrons then lose energy by ionization and excitation until they are stopped. The positron then combines with an electron, both are converted into 'annihilation' quanta or gamma rays of electromagnetic energy of 0.51 Mev each at an angle of 180° to each other. This interaction, taking place in a scintillator, gives rise to several pulse heights on the spectrum, e.g. the positive and negative electrons are stopped in the scintillator, when the full incident primary gamma energy (less 1.02 Mev) will appear. If either or both annihilation gammas are stopped photo-electrically in the scintillator, 0.51 or 1.02 Mev will be added to the primary pulse height. If either annihilation guanta interacts by the Compton effect, one or two continua will be superimposed. The simplest example of such gamma-ray spectrum

is shown in Fig. 4. The effect of interaction of gamma rays of various energies with NaI may be well understood from Fig. 5.

A gamma spectrum is complicated by several ways in which the gamma rays interact with matter (NaI(Tl) activated crystal) as described above. By combination of these interactions, the gamma spectrum of a commonly used nuclide like Cs^{137} as seen by an ideal detector would look like that shown in Fig. 6(a). Since the gamma energy is less than 1.02 Mev, pair production is not seen. In practice, a scintillation detector produces a spectrum as shown in Fig. 6(b). Three characteristics of the scintillation detector cause the differences (1) 'statistical broadening' (2) 'Back-scattering' (3) photomultiplier 'noise'.

(1) This broadening of the photoelectric peak arises primarily from the statistical spread in the relatively small number of photo-electrons finally formed at the photocathode. The term 'resolution', used to denote the degree of broadening, is defined as the full width of the peak as measured by the pulse height at the peak. The term 'resolution' should not be confused with 'resolving time' (the measure of the system's ability to distinguish pulses which occur with short time intervals between them). The width and the pulse height may be measured in energy units (electron volts) or in discriminator volts.

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Figure 4

Na²⁴ GAMMA-RAY SPECTRUM



ABSORPTION COEFFICIENT OF NaI(T1)

FOR VARIOUS GAMMA-RAY ENERGIES



PHOTON ENERGY (kev)

Figure 6(a)

GAMMA SPECTRUM AS SEEN BY IDEAL DETECTOR

Figure 6(b)

GAMMA SPECTRUM AS SEEN BY

SCINTILLATION SPECTROMETER



(2) A broad peak* may be noticed in the middle of the Compton distribution. This phenomenon, known as 'backscattering', derives from the photoelectric interaction and detection of gamma rays which have been scattered backwards or through about 180° from material below and surrounding the scintillation crystal. The effect varies directly with the amount and weight of the surrounding material and inversely with the source-crystal distance.

(3) The sharp increase in the observed counting rate with decreasing discriminator settings is due to the photomultiplier 'noise', caused by burst of thermal electrons emitted spontaneously from the photocathode at normal room temperature. These small noise pulses limit the practical application of scintillation counting to a minimum of a few thousand electron volts.

*The energy of this peak is given by the simple formula

$$E_{\theta} = \frac{E_{y}}{1 + \frac{mc^{2}}{(1 - \cos \theta)E_{y}}}$$

or when $\theta = 180^{\circ}$: $E_e = \frac{E_y}{1 + \frac{mc^2}{2E_y}}$

where $E_e =$ energy of the scattered electron,

 $E_v = energy$ of the primary photon,

 θ = the angle between the primary photon direction and the secondary scattered gamma ray,

 $mc^2 = 0.511$ Mev.

In the present work a commercially available $3" \times 3" / \overline{NaI(T1)}7$ crystal of 12.9% resolution for the photoelectric peak of Cs^{137} (0.661 Mev) has been used as a detector in conjunction with a commercial 100 channel pulse-height analyser (Computing Devices of Canada Limited) for gamma-ray and X-ray measurement. The description of the instrument and processing of the data are discussed in the Experimental Approach chapter, (B Section g).

(j) <u>SEARCH FOR NEUTRON-DEFICIENT NUCLIDES AND</u> <u>MASS ASSIGNMENTS</u>

If one bombards any element with protons of energy ranging from 6 to 85 Mev, or more, one would expect predominantly (p,xn), (p,pxn) and less predominantly (p,2pxn) and (p,3pxn) reactions, whichever are energetically possible (where x = 1, 2, 3, 4, 5, 6, 7, 8). Since erbium had not previously been bombarded at high energies, one may expect to identify new activities of measurable half-lives in the spallation products of erbium when bombarded with protons of energy > 40 Mev.

In addition, it is useful to confirm the nuclear properties of known nuclides employing refined techniques. Special advantages in the present work are: quick separation of the heavy rare earths, availability of enriched stable isotopes of erbium (since December 1959), and availability of a synchrocyclotron that can accelerate protons up to 87 Mev. Mass assignments in the present type of work are generally made by the study of genetic relations, cross bombardments, and bombarding enriched isotopes, study of nuclear properties and also by subjecting the product activities for direct mass separation. Some of the types of studies that have been made will be described later. The chart of the nuclides in the region of erbium as it was at the end of 1958, when the present work began, is presented in Fig. 7. It clearly indicated the possibility of identifying new neutron-deficient isotopes.

(k) SEARCH FOR NUCLIDES THAT EXHIBIT ALPHA EMISSION

Nuclides sufficiently far to the neutron-deficient side of the stability curve may very well exhibit alpha emission. This was shown to be a valid postulate when, in 1949, Thompson and co-workers reported the discovery of alpha radioactivity in neutron-deficient isotopes of gold and mercury and in rare earth isotopes (58). Rasmussen and co-workers reported on a detailed study of such isotopes in the rare-earth region (59). Very recently, Toth (60)reported a study of alpha emission in the neutron-deficient isotopes of dysprosium and terbium. In view of the above experimental facts, a search for alpha emitters has been made in the heavy rare-earth region. The experimental details of the search are described later.

NUCLIDE CHART IN THE REGION OF ERBIUM

(as of October, 1958)

	*				,						
						Talff	Tm167	Tmi68	Te i69	Tm 170	
					25h 720, 81, 16,	7.7h #*2: ; 000 32	964	87 d	* *	129 d 4' 97, 89 7 0840.4' 5 97 + 2000	1.9y #io My tio
	Er 160 29h *	Er161 3h K, 41 12 7 42,112	Er 162	Er163 75m ; 43,110	Er 164 156	Er 165	Er 166 314	Er 167. 22.9	Er 168 27,1 17 117,004	Er169	Er 170 14.9
		Ho 160 5.0h y 19,72, 94, K	Ho 161 2.5h	Hol62		Ho 164		Hol66	Hol67 3.0h 4° 10, 20 7 35, 70		Ho169 #- 1.6h
Dy 157 8.2h * 7.32	Dy 158	Dy 159 134d *.1		Dy IGI	Dy 162 25.5 #1977	Dy 163 25.0	Dy 164 20.2	Dy 165	Dy 166		
Tb 156			TE 159	Tb 160 72 d # 36, 45, 734, 0840-145 6181 # 400	Tb 161 7d 7049.00 64	Tb162					
		• 1 ~									

(1) CROSS-SECTION CALCULATION FORMULAE

The cross-section, σ , of a particle-induced nuclear reaction can be written in the form

 $\sigma = \frac{\text{Number of product nuclei formed by the reaction}}{(\text{Number of parent nuclei/area})(\text{Number of bombarding particles})}$

The dimensions of the cross section are then an area measured in

$$\frac{\mathrm{cm}^2}{\mathrm{nucleus}} \text{ or better in millibarns} = \frac{10^{-27} \mathrm{cm}^2}{\mathrm{nucleus}} .$$

The cross-section calculation formula for a reaction induced by a charged particle (of charge Z)

$$\sigma = \frac{4.44 \times 10^{-17}. (Z) D_{p}^{\circ}. t}{q \cdot \frac{n}{A} (1 - e^{-\lambda t})}$$

D_p^o = number of disintegrations per minute of the product nuclei at the end of the bombardment.

- Z = charge of the bombarding particle, e.g. proton or alpha = 1 or 2 respectively.
- t = the length of the bombardment in minutes.

n/A = number of parent nuclei per cm², where

 $\frac{n}{A} = \frac{\text{weight of parent nuclei in mg.x 6.02 x 10}^{20}}{\text{atomic weight of the parent nucleus/area}}$ in cm²

secondary charges can leave this cup.

 $\lambda = \text{decay constant} = \left(\frac{0.693}{\frac{1}{2} \text{ of the product nuclei}}\right)$ q = number of microamp. hours. $\frac{4.44 \times 10^{-17} \cdot (Z)}{q} = \text{number of bombarding particles of charge Z.}$ This may be determined by collecting the charged particle beam in a Faraday Cup(61) which is insulated in such a way that no

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If the charge particle beam is monitored, that is, by bombarding the target element and a monitor simultaneously, the cross-section calculation formula is

$$\sigma_{\mathbf{p}} = \sigma_{\mathbf{m}} \frac{D_{\mathbf{p}}^{\mathbf{o}} W_{\mathbf{m}} M_{\mathbf{T}} (1 - e^{-\lambda_{\mathbf{m}} t})}{D_{\mathbf{m}}^{\mathbf{o}} W_{\mathbf{T}} M_{\mathbf{m}} (1 - e^{-\lambda_{\mathbf{p}} t})}$$

$$\lambda$$
 and λ are decay constants of monitor and product nuclei.

(m) CHOICE OF MONITOR

The monitor reactions often used are the following: $Al^{27}(p,3pn)Na^{24}(15 \text{ hours}) \longrightarrow Mg^{24}$ (stable) $c^{12}(p,pn)c^{11}$ (20.5 min.) $\longrightarrow B^{11}$ (stable) $B^{11}(p,n)c^{11}$ (20.5 min.) $\longrightarrow B^{11}$ (stable)

It was not possible to use the above monitoring reactions because, in some cases, it was difficult to mix uniformly the monitoring element and the target material (erbium oxide). In some instances, problems arose involving chemical separation, and subsequently problems involved in measurement due to inconvenient half-lives. In others, the cross sections do not adequately cover the required range (6.1 to 86.5). Copper was chosen as a monitor in order to minimize the above inconveniences to a reasonable degree.

Copper is one of the typical medium weight elements which has been bombarded with protons by several workers (3,5,62,63,64,65,66,67) and the formation cross sections at various energies have been reported.

The monitoring reactions used for the present work were the following:

(a) $Cu^{63}(p,n)Zn^{63}$ (38.3 min.) $\longrightarrow Cu^{63}$ (stable) (b) $Cu^{65}(p,pn)Cu^{64}(12.8 \text{ hrs.}) \longrightarrow Ni^{64}$ (stable)

The reaction (a) has been used for low-energy (6.1, 10.6 and 12.8 Mev) irradiations. The cross sections used in the present work were that of Meadows⁽³⁾ as shown in Fig. 1. Although Meadows' values are low by 40% when compared to those of Ghoshal⁽⁵⁾ and Howe⁽⁶⁷⁾, still the Meadows' values have been preferred owing to the following reasons:

- (1) Bell and Skarsgard⁽¹⁸⁾, who preferred Meadows' values rather than Ghoshal's values, gave a very convincing argument in support of the choice of Meadows' values.
- (2) In addition, Jackson's ⁽²¹⁾ calculated values of formation cross sections were in good agreement with those of Bell and Skarsgard ⁽¹⁸⁾ who used Meadows' values.
- (3) The Meadows' cross sections for reaction (b), shown in Fig. 2, were in good agreement with those of

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Coleman and Tewes⁽⁶²⁾. If the cross section for reaction (b) is considered correct, then one can thus conclude by presumptive evidence that the values for reaction (a) are also correct, since the techniques employed were the same.

For the irradiations at energies from 12.8 to 86.5 Mev, reaction (b) has been chosen as a monitoring reaction. The cross sections used in the present work were those of Meadows⁽³⁾, shown in Fig. 2. Some of these cross sections were checked by Coleman and Tewes⁽⁶²⁾, and the values were in good agreement with those of Meadows. In addition, Turkevich, in a private communication, has put more faith in Meadows' and Coleman's values rather than in his⁽⁶⁵⁾ reported values which are low, since there is the wide-energy spread involved in low-energy (\leq 100 Mev) irradiations with the cyclotron at Chicago.

B. EXPERIMENTAL

(a) TARGET MATERIALS AND PREPARATION OF TARGETS

Target materials:

It is important that very pure materials are to be used as targets, since small impurities might cause large errors in cross-section determinations. In the case of rare earths, the impurities of the adjacent rare earths are very likely to be present because of the difficulty involved in the individual rare-earth separations.

Erbium oxide and copper oxide of natural isotopic abundance were obtained from Johnson Matthey and Co., London. Enriched isotope samples of erbium were obtained from Oak Ridge National Laboratory, Union Carbide Nuclear Company, Oak Ridge, Tennessee. Holmium oxide was obtained from Research Chemicals Company, Burbank, California. All the samples were fine powders. Data, as submitted by the supplier, regarding the impurities are presented in Tables I(a) and I(b). In Table II abundances of the stable isotopes in the enriched as well as natural isotope sample of erbium are presented.

(1) Targets for Proton Irradiations

In the case of low-energy bombardments (6.1, 10.6, 12.8 Mev) an atom ratio of erbium to copper of 1:1 was picked, while at high-energy bombardments the atom ratio of erbium to copper picked was 1:2. Erbium oxide and cupric oxide were weighed in the required proportions and thoroughly

m 4 4	Impurities					
Target material	Element	Amount %				
Cupric oxide	Ni	0.000002				
(natural)	Fe	0.000001				
	Mg	0.000001				
	Ag	0.000001				
Erbium oxide	Ho	0.05				
(natural)	Tm	0.01				
	Fe	0.000002				
	Na	0.000002				
	Si	0.000001				
	Mg	0.000001				
Holmium oxide (natural)	Rare Earths	0.01				

Table I(a)

Table I(b)

Target	Impurities							
Material	Element	Amount %	Element	Amount %				
Erbium	Al	0.05	Ti,V	0.02 of each				
oxide	В	0.01	Zr	0.1				
	Ba	0.02	Y	0.015				
	Be	0.001	La	0.04				
	Ca	0.1	Ce, Pr,Nd	0.2 of each				
CI	b,Co,C r ,Cu,Fe	0.05 of each	Sm	0.1				
	Mg	0.02	Gd, Tm, Lu	0.02 of each				
	Mn,Mo,Na	0.05 of each	Eu	0.008				
	Ni	0.01	Чъ	0.004				
	Pb,Si	0.05 of each	Ho	0.1				
	Sn	0.02	ТЪ	0.05				

Ta	b 1	e	Ι	Ι
				_

		ISOTOPE MASSES							
		170	168	167	166	164	162		
			% Abundances						
Natural	l erbium	14.9	27.1	22.9	33.4	1.56	0.136		
Erbium in	enriched 167	1.93	25.3	58.8*	14.0	0.1	0.1		
Erbium in	enriched 166	0.8	8.5	17.7	72.9*	0.1	0.1		
Erbium in	enriched 164	1.9	7.8	14.3	60.4	15.6*	0.1		
Erbium in	enriched 162	4.8	15.1	18.2	44.2	11.6	6.1*		

*Indicates the abundance in the enriched sample.

Natural copper has two stable isotopes:

Cu⁶³ of 69% abundance; Cu⁶⁵ of 31% abundance. Natural holmium is monoisotopic. mixed, manually and mechanically for 40 hours. The mixture was uniform because the copper oxide and erbium oxide were fine powders and of similar densities.

The uniform mixture was placed in a piece of aluminium tubing obtained from the Precision Tube Company, Philadelphia, Pa. The dimensions of the tubing were as follows:

Outside diameter 0.0625" ± 0.005"

Wall thickness 0.0015" - 0.0005"

A length of tubing (about $1^{1}/3^{*}$) was cut from the main stock. This piece was pressed shut at one end with a dissecting forceps. About 10 to 15 mgs of the powdered mixture to be bombarded was weighed into the tubing. The target assembly is pictorially represented in Fig. 8. The tube containing the target mixture was fixed to the target holder in such a way that the target mixture was right opposite the mounting screw, where the target was presented to maximum intensity of protons.

(2) Targets for Alpha Irradiations

The holmium oxide target was prepared by the method developed by Dodson et al. (68). This method in brief consisted of the following steps:

- (i) Preparation of paint and its composition:
 - A known amount of holmium oxide was dissolved in concentrated HNO₃ and evaporated almost to dryness (leaving a trace of acid) on a water bath, and the holmium nitrate was dissolved in

PROTON IRRADIATION TARGET FASTENED

TO TARGET HOLDER



1% zapon solution (0.4 cc *zapon syrup or Awuanite-A, 7 cc ethyl acetate and 28 cc of 95% alcohol). The concentration of holmium oxide in such a mixture (paint) was maintained at 6 mg/cc.

(ii) Preparation of foil:

Very carefully flattened and ironed aluminium foil of thickness 6.6935 mg/cm² has been used for target backing.

- (iii) A record player has been used as a rotating spindle of speed 78 R.P.M.
- (iv) Painting with a fine quality camel hair brush was done on the aluminium foil. The foil was fixed on the glass plate which in turn was firmly fixed to the rotating disc (spindle). After each painting and subsequent air drying of the paint, the aluminium foil was dropped on to the surface of a hot plate (400°C). This process was repeated several times. After each painting the surface was smoothened with lens paper and by ironing.
- (v) Circular targets were punched out of the foil with an arch punch (C.S. Osborne No. 149) of diameter ³/₈ⁿ.

Obtained from Glidden Paints Co., Cleveland, Ohio, U.S.A.

The targets thus prepared were fixed in the target holder shown in Fig. 9, along with necessary aluminium absorbers reported in the literature (69) to degrade the energy to the required values.

(b) IRRADIATION PROCEDURES

Proton irradiations:

The target, fixed to the target holder, was bolted to the end of the probe. Control of the bombarding energy was achieved by inserting the target holder to different distances from the centre by means of a probe entering through a sliding vacuum seal in the outer wall of the dee Thus the targets were irradiated with protons of chamber. energies varying from 6 Mev to 86.5 Mev in the circulating The calibration beam of the McGill synchrocyclotron. curve of proton energies vs. probe length in inches was supplied by Professor Bell⁽⁷⁰⁾ and is shown in Fig. 10. In the same private communication, Professor Bell indicated that the uncertainty and the spread in beam energy was about $\stackrel{+}{-}$ 2 Mev. The duration of the bombardment varied from 15 minutes to 2 hours according to need.

Alpha irradiations:

Two alpha irradiations^{*} were done at the Brookhaven National Laboratory cyclotron. The alpha particle energy of the B.N.L. cyclotron was 39.8 Mev.

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^{*}I am greatly indebted to Dr. J. Hudis of Brookhaven National Laboratory for arranging the two alpha irradiations.

ALPHA IRRADIATION TARGET HOLDER



ENERGY CALIBRATION CURVE FOR THE

McGILL SYNCHROCYCLOTRON

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5**1a**

I

The uncertainty in beam energy has been estimated at Brookhaven to be of the order of + 0.5 Mev. A Faraday cup arrangement was used to measure total beam current of alpha particles. Seven targets were irradiated at the same time by the stacked foil techniques. Previous experience at Brookhaven has shown that there is no loss of beam by scattering by this technique. In one of the two irradiations, VYNS* (a polymer) and Al backing was used as target backing material. This irradiation proved to be unsuccessful because the VYNS burnt due to heat produced during irradiation in spite of the reduced beam current and The second irradiation, for which the water cooling. targets were prepared according to Dodson's method⁽⁶⁸⁾. was very successful.

(c) ESTABLISHMENT OF CONDITIONS FOR THE SEPARATION OF INDIVIDUAL HEAVY RARE EARTHS

In preliminary experiments to determine the characteristics of elution and optimum separation conditions for the heavy rare earths, both inactive and active materials have been used. In the case of erbium, holmium, dysprosium and terbium, hydroxide precipitation was the test. In the case of thulium, Tm^{170} , and for sodium, Na^{22} have been used, and they were traced by scintillation detectors.

In some procedures, by not adding one of the

A polyvinyl chloride-acetate copolymer available from Bakelite Co., New York, U.S.A.

elements, e.g. erbium or holmium, a gap has been observed for the element that has not been added. The gap was identified by applying the hydroxide precipitation test to the eluate. Finally, a few high energy irradiations of erbium were made, and the individual rare earths were separated.

A typical elution pattern is shown in Fig. 11. This separation was done for a natural erbium target irradiated for half-an-hour with 80 Mev protons. For a quick, reproducible and pure separation of milligram quantities of individual heavy rare earths, as shown in Fig. 11, the following physical and chemical operation conditions were used:

(i) Preparation and purification of resin:
The resin employed was that portion of a ¼ pound lot of minus 400 mesh, 12% cross-linked Dowex-50 which settled out in half-an-hour after the resin had been suspended in 1¼ feet of distilled water. The graded resin had been washed with 6 M NH4OH, hot distilled water, HCl and hot distilled water. This type of cycling was done several times and finally stored as the ammonium form in degassed distilled water.

(ii) Preparation of elutriant solution:
 Elutriant solutions were prepared by direct
 weighing of an especially purified^{*}, crystallized

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^{*}I am grateful to Dr. D.C. Stewart of Argonne National Laboratory for pointing out the possibility of the impurities in commercial α-hydroxy isobutyric acid and for suggesting the method of purification, by re-crystallization from ether.

TYPICAL ELUTION PATTERN OF

HEAVY RARE EARTHS



and dried Eastman Kodak α -hydroxyisobutyric acid, followed by dilution and simultaneous adjustment of pH with fresh ammonium hydroxide. The optimum pH and concentration determined for the present work, after a series of column separations of Tm, Er, Ho, Dy and Tb proved to be 0.2 M concentration and a pH of 4.1. The elutriant solution gave reproducible, quick (70 minutes) good separations. All the solutions were prepared in degassed distilled water. Care was taken to approach the pH 4.1 from the acid side. so that readjustment with acid was not necessary, since it is important for good separation to keep the ionic strength at a minimum.

(iii) Dimensions and preparation of resin column:

A resin bed 20 cm long and 0.8 cm in diameter has been used. The free column volume was estimated to be roughly 60% of the settled resin column volume. The column tip was plugged with glass wool. A slurry of resin was introduced and was allowed to settle. The uniformly deposited bed of resin, free from air bubbles or channels was essential for a good separation.

In Fig. 12(b) the cation exchange column is shown.

(iv) Temperature control of the column:

Elutions were performed at 87°C using a set-up similar to that of Thompson et al.⁽⁵⁷⁾. The

- 55 -
Figure 12(a)

ANION EXCHANGE COLUMN

Figure 12(b)

CATION EXCHANGE COLUMN



arrangement is shown in Fig. 13. A column at a temperature of 87°C has given better separations than one at room temperature.

(v) Channelling:

Careful precautions were taken to prevent channelling by using degassed solutions and the regenerated columns instead of fresh columns. Regeneration was partly done by passing several column volumes of eluant and partly by passing several free-column volumes of degassed distilled water at room temperature.

- (vi) Preparation of loading mixture:
 - To an aliquot containing the erbium spallation product mixture, the necessary carriers (2 to 4 mg quantities) along with 1 mg of sodium chloride <u>/</u>see later under (xi)7 have been added. The loading mixture was evaporated down to a residue. The residue was dissolved in a minimum quantity of 0.06 M HCl, and this process was repeated three times. Then the residue was finally dissolved in a few drops of hot 0.06 M HCl.

(vii) Loading of rare earths:

Since initial adsorption of the rare-earth mixture was one of the key steps to achieve good separation, sufficient care was necessary at this stage also. Ketelle and Boyds: (43) slurry method and Stewart's (48) method of forcing the

HEATING JACKET ARRANGEMENT FOR

RARE-EARTH SEPARATIONS



rare-earth solution into the top bed of the resin were not employed, since the former gives poor yields and the latter poorer separations because of disturbances caused due to force. The method employed for the present work was to add the rare-earth solution drop by drop on to the top of the resin column bed and allow it to be adsorbed (without an external pressure). As soon as all the rare-earth solution was adsorbed, the sides of the column were washed with a few drops of hot distilled water, and the top of the column was filled with hot eluant while the bottom end was closed immediately (to prevent elution). The column was inserted into the jacket maintained at 87°C, as described above, and allowed to stand. Elution was started 10 minutes after insertion into the jacket.

(viii) Capacity resin:

Overloading of the column causes the elution bands to overlap. This was prevented by keeping the amount of loading mixtures below 10 milligrams.

(ix) Flow rates:

The elutriant solution reservoir was adjusted in such a way that the flow was 0.75 ml per minute (14 drops a minute). This flow rate was found to be convenient because each element was eluted in a volume of 10 mls, collected in about six centrifuge tubes, out of which a few fractions in the middle were used for radiation measurements.

- (x) Fraction collection and further treatment of eluate:
 - The fractions were collected every two minutes with the help of a MISCO-TYPE Fraction Collector, No. 6500, provided with interval timer, No. 6510. The fractions were collected into 12 ml capacity pyrex centrifuge cones. The cones that were expected to contain a particular element were combined (before being tested for purity with the help of a pulse height analyser), the rare earths were precipitated in a centrifuge cone as hydroxides, and the precipitate was washed three times with distilled water. Finally, the precipitate was dissolved in a minimum quantity of dilute HCl, transferred into a 5 ml capacity volumetric flask and made up to volume, from which aliquots were taken for radiation measurements.
- (xi) Introduction of foreign ions:
 - From the previous researches of Ketelle and Boyd⁽⁴³⁾ and Stewart⁽⁴⁸⁾, sodium ion was known to be eluted near thulium. Trace experiments were performed to find out the elution behaviour of Tm^{170} and Na^{22} activities. It was found that Na^{22} was eluted directly after Tm^{170} . Based on this

observation, separation of Tm and Er has been tried introducing 1 mg of NaCl. It was seen that the separation factor was increased. This experimental observation was rather qualitative. However, in the present work, rare-earth separations were done introducing less than milligram quantities of inactive sodium ion.

(xii) Time of separation:

It was necessary to determine the exact time of separation in order to calculate the amount of daughter activity which grew in due to the presence of parent before chemical separation. In this connection, the time of separation was determined by following the path of Tm^{170} (t₁ 129 d.: 0.95 Mev β^{-}) after its adsorption on the top of the resin bed, (all the other experimental conditions were kept as mentioned above) and subsequent elution with 0.2 M ammonium salt of alpha-hydroxy isobutyric acid of pH 4.1. The path of Tm¹⁷⁰ after five minutes and after 15 minutes from the start of the separation has been indicated in Fig. 14. This behaviour gave an indication that the separation must have been complete after 15 minutes. It has been assumed that erbium. holmium and dysprosium followed thulium one after another in a similar pattern.

Tm¹⁷⁰ ELUTION PATTERN (BAND SPREAD)

a - initial adsorption

- b after five minutes elution
- c after fifteen minutes elution



(xiii) Mould formation:

To prevent mould formation, the stock solution of elutriant was kept in a refrigerator. This was done since mould formation may alter the elution characteristic of alpha-hydroxy isobutyrate solutions.

(d) TARGET CHEMISTRY FOR LOW-ENERGY PROTON IRRADIATION PRODUCTS (6.5 to 12.8 Mey)

In the three low-energy irradiations chemical separation was not necessary, since only (p,n) reaction products were formed from erbium and copper. That is, the following nuclear reactions occurred:

Since the half-lives are easily separable, the use of a multi-channel pulse height analyser and the proper choice of photopeaks made the analysis relatively simple.

In view of the above, the spallation product mixture from the aluminium tube was poured into a 10 ml volumetric flask and dissolved in a few drops of warm concentrated HCl. The solution was cooled and the volume made up to the mark. An aliquot was taken for radiation measurement purposes of rare-earth and monitor activities. In the above procedure, it was safe to assume erbium acted as carrier for thulium activities and copper for zinc activities.

(e) TARGET CHEMISTRY FOR THE HIGH-ENERGY PROTON IRRADIATION PRODUCTS (16.4 to 86.5 Mev)

Separation of copper (monitor activity):

The method employed for the present work was based on Kraus and Moore's (71) publication in which they determined the elution constants of Ni, Mn, Cu, Fe, Co, and Zn in O-14 M HCl with a Dowex-1 resin column. Another relevant publication for the present separation is that of Kraus and Nelson(72) who reviewed extensively the anion exchange studies of fission products.

The separation procedure consisted of the following steps:

- (i) After irradiation the aluminium tube containing copper and erbium spallation products was cut open with a fine razor blade, and the contents were transferred into a 40 ml capacity centrifuge tube into which a few drops of concentrated HCl were added until all copper and rare earths went into solution.
- (ii) The spallation products in concentrated HCl were transferred on to the top bed of a column

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- (iii) The column was eluted with 6 mls of concentrated HCl (12 M). Rare earths and Ni passed through, leaving the remaining copper spallation products (Cu, Zn, Mn, Co, Fe,) on the column.
- (iv) 10 mls of 4M HCl were run through the column which caused Mn and Co to be eluted.
- (v) Copper was eluted with 1.5 M HCl. It was easy to observe the blue-green band of copper passing through the column.

The copper, thus separated, was made up to 10 mls, and an aliquot (1 ml) was taken for radiation measurement and another for chemical yield determinations.

Individual rare-earth separations:

To the eluate (obtained from step 3 of the monitor separation) from the anion exchange column, about 2 mg of nickel ion was added as a hold-back carrier (for the nickel obtained as a spallation product from copper), and the rare earths were precipitated as hydroxides with 6 M $\rm NH_4OH$. The nickel remained in solution as the nickel-ammonium complex.

The hydroxide precipitate was washed with distilled water and dissolved in a minimum amount of HCl and transferred into a 10 ml volumetric flask, then diluted up to the mark. This was considered as a stock solution. Until

^{*}Baker analysed grade Dowex 1 x 8 resin was washed six times with distilled water and concentrated HCl alternately and finally the resin column was prepared in concentrated HCl.

now erbium acted as a non-isotopic carrier for all rareearth activities. From the stock solution a 1 ml aliquot, diluted in a vial to 2 mls, was used for measuring thulium activities. (All the thulium activities, Tm^{170} , Tm^{168} , Tm^{167} , Tm^{166} , Tm^{165} , Tm^{164} , Tm^{163} , could be followed for decay in the rare-earth mixture by proper choice of photopeaks; and it was also safe to assume that erbium acted as a carrier for thulium activities.)

From the stock solution, a 2 ml aliquot was pipetted into a standard 40 ml capacity centrifuge tube, to which milligram quantities of individual rare earths as carriers (Tm, Ho, Dy and Tb) along with one milligram of sodium chloride also were added. From this mixture the loading mixture, prepared as mentioned earlier, was loaded on the top bed of the resin column, and the column was inserted into the heating jacket maintained at 87° C. The elution was started 10 minutes after insertion into the heating jacket.

All the separation chemistry was done in the glove boxes. The total time required for complete separation, from the end of bombardment, was about 4 hours. The cation exchange separation took 70 minutes, and the anion exchange separation 60 minutes. The remainder of the time was taken up with purification of individual rare earths from the eluate and allowing short-lived parents to decay before the start of separation.

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(f) TARGET CHEMISTRY FOR ALPHA IRRADIATION PRODUCTS

It was not necessary to do any separation chemistry on the targets which were irradiated with alpha particles, as in the case of low-energy proton irradiations.

The holmium targets, obtained from B.N.L. a few days after the irradiation, were transferred into separate vials of specific dimensions and dissolved by the addition of two drops of 10% H₂O₂ and 10 drops of concentrated HCl. The total solution was made up to 2 ml volume.

The 0.2 Mev photopeak of 86-d Tm¹⁶⁸, and 9.6-d Tm¹⁶⁷ could be easily followed for decay. The decay curves did not show any impurity.

(g) <u>IDENTIFICATION AND DETERMINATION OF THE</u> SPALLATION PRODUCT NUCLEI

- (1) Gamma Radiation Measurement
- (i) Source detector arrangement:

The source detector consisted of a hermatically sealed 3" diameter x 3" cylinder of NaI(T1) crystal obtained from the Harshaw Chemical Co. The crystal was optically coupled to a Dumont 6364 photomultiplier tube. The high voltage supply for the photomultiplier tube (P.M. tube) was a Baird-Atomic Model 318. The scintillations produced in the crystal, due to gamma-ray interaction, are converted into amplified electrical pulses by the photomultiplier tube. In order to

improve the operation of the P.M. tube, it was shielded from magnetic fields by a mu-metal shield. Background radiation was reduced by placing the crystal-P.M. tube assembly inside a cylinder of lead, 12" thick. Fluorescent X-rays from the lead shield were absorbed by a graded shield made of 🛃 of iron and $\frac{1}{8}$ of lucite. The detector and the shielding are shown in Fig. 15. The electrical pulses from the P.M. tube were fed via a cathode follower (Hamner Electronics Model N-351) into a linear non-overloading amplifier with variable gain (Baird Atomic, Model 215). The positive output pulse was passed through an anode follower into the 100-channel pulse height analyser. Important units are the magnetic core storage unit, oscilloscope unit, pen recording unit, and the printing unit. A block diagram of the above equipment is presented in Fig. 16. The dead time of the pulse height analyser varied from about 35 microseconds to 135 microseconds, depending on the channel. These values were rather high. However, the unit was equipped with a microammeter which indicated the percentage loss due to dead time for a random distribution of pulses. The resolution losses, in the case of the short-lived Tm isotopes, varied from 20-30%, as indicated by the meter, and corrections were made for these.

NaI(T1) DETECTOR, PHOTOMULTIPLIER

TUBE AND SHIELDING



BLOCK DIAGRAM OF 100-CHANNEL PULSE

HEIGHT ANALYSER AND AUXILIARY EQUIPMENT



Processing of the photopeak spectra:

(ii)

Although the gamma-ray scintillation pulse height analyser is finding increasing acceptance as a standard piece of equipment in nuclear research laboratories, its more general application to the routine quantitative analysis of radioactive nuclide mixtures has been hindered by the lack of a quick and convenient method for estimating abundances from the pulse distribution data. Lazar⁽⁷³⁾, Heath⁽⁷⁴⁾ and Lee⁽⁷⁵⁾ have suggested methods for analysing composite spectra into their gamma-ray components. Lazar's and Heath's methods essentially consisted of fitting a 'gaussian' distribution to the highest energy The distribution can be subtracted gamma ray. from the rest of the spectrum, and similarly for each gamma-ray component, until the entire spectrum has been analysed. Lee described a method of estimation of gamma-ray abundances in radionuclide mixtures by making use of a unique 'complementsubtraction' feature of the Argonne-type 256-channel Lazar(73) stated that the methods. analyser. similar to the one described by him, were rather tedious especially when there were many spectra on hand, as in the present work.

In the present work, counting rates of gamma rays were obtained by integrating the approximate photopeaks in the scintillation spectra of the samples. Figs. 17 and 18 illustrate the manner in which the integration was performed in an 'easy case' and in a 'difficult case' respectively. When the situation was such that the photopeaks were distorted, due to the presence of the photopeaks of short-lived isotopes, the gamma-ray measurements were done after the decay of the short-lived components, and in some cases proper choice of the gamma peaks saved the situation, e.g. Tm^{165} (1.16 Mev) Tm^{166} (2.05 Mev).

- Apart from the high energy gamma rays, measurement of the decay of the K-X-rays of some of the nuclides (e.g. Ho¹⁶⁰, Ho¹⁶¹, Er¹⁶⁰, Er¹⁶⁵ and Dy¹⁵⁹) presented a problem partly because of the variation of shape of the peak with counting rate and partly due to multi-component systems, which made it necessary to follow the decay even at low counting rates in order to resolve the decay curves. On the left hand side of Fig. 19 typical X-ray spectra have been presented, whereas on the right hand side of the same figure the same spectra are shown after background has been subtracted.
- After a careful study of several types of X-ray spectra of the nuclides, it was concluded that it was safe to assume a 'gaussian' distribution for X-rays after background subtraction, and the peak

BACKGROUND SUBTRACTION PROCEDURE

FOR PHOTOPEAKS 'EASY CASES'



73a

BACKGROUND SUBTRACTION PROCEDURE

FOR PHOTOPEAKS 'COMPLICATED CASES'



74a

TYPICAL X-RAY PEAKS AT VARIOUS COUNTING

RATES TO ILLUSTRATE EASY BACKGROUND

SUBTRACTION PROCEDURE

- l Iodine escape peak
- 2 K-X-ray peak



75a

height of such peaks was given by the vertical distance between the dotted lines, as shown in the spectra on the left hand side of Fig. 19. These distances were the same as the vertical distances between the maximum of the peak and the base line, as are shown in the spectra on the right hand side of Fig. 19. This procedure assumes the background is a straight line drawn parallel to the baseline from the channel that accumulated the minimum number of counts, and the peak height is the vertical distance between the maximum of the peak and the straight line drawn parallel to the baseline. The peak height thus obtained was converted into total counts under the whole area by multiplying by conversion factors determined by averaging typical cases.

The total counts obtained, by measuring counts under the photopeaks, were converted into counting rates, and decay curves were drawn. The counting rates at the end of irradiation were converted into disintegration rates by applying corrections for crystal efficiency (as described later), branching ratio and for internal conversion. In addition, in the case of K-X-ray peaks, fluorescence yield corrections were also necessary. To determine the number of product nuclei, it was essential to multiply by the chemical yield factor determined. using colorimetric methods described in Sections 4 and 5, and the dilution factor as well. This procedure was used for the nuclides formed independently and also for cumulative yields. In the case of nuclides produced independently as well as by the decay of parents, the contribution due to the parent is given in Appendix II.

- (iii) Source preparation:
 - Although in the literature several authors (76,77,78) have used point, disc, or collimated sources, 2 ml volume liquid sources in glass vials of specific dimensions have been used for the present work because of the ease and quickness involved in preparing the sources. In addition, ease in handling the source for measurement and, in some cases, accuracy were a few more points in favour of this type of source. Sources were prepared by pipetting a 1 ml aliquot from the stock solution and a 1 ml wash of the pipette into a vial.
- (iv) Energy calibration of photopeaks:

Several long-lived standard sources (Na²², Co⁶⁰, Eu¹⁵²⁻¹⁵⁴) have been used to plot photopeak energy vs. channel number (or pulse height). The unknown sample and the standards were measured under identical settings, e.g. channel width, gain and resolution loss (R.L.) reading, as indicated by the microammeter. It is necessary to maintain the same R.L., since it was observed that the pulse height shifted with change in counting rate. From the calibration plot unknown energies were determined.

(v) Efficiency calibration of the crystal:

The sodium iodide crystal mounted by the manufacturer (Harshaw Chemical Co.) was different from the one described by Bell(76), and in this case the theoretically computed curves could not be used. In addition, the sources used in the present work were of different geometry from those described by Bell, so that the theoretically computed curves could not be used for low-energy photopeaks. The crystal mounting is shown in Fig. 20. The manufacturer of the crystal stated that the modification in the construction was necessary for a good optical contact between the crystal and the lucite window. In view of the above modifications, it was necessary to calibrate the crystal. The efficiency^{*} calibration is described in Appendix I.

(2) Beta Measurement

A standard 2π -proportional counter has been used for beta measurement of the nuclide $\operatorname{Er}^{169}(t_{\frac{1}{2}} = 9d, \beta^{-} = 0.34$ Mev). The operating voltage was determined to be 3200 volts from Fig. 21, and discriminator setting was determined to be

*Mr. G. Grant, Mr. M. May and myself collaborated on the efficiency calibration of the 3" x 3" NaI(T1) crystal.

DIAGRAM OF HARSHAW 3" x 3"

NaI(T1) CRYSTAL



PLATEAU CURVE FOR A 2π -PROPORTIONAL

BETA COUNTER

Discriminator bias setting - 25 volts



25 volts. Sc^{46} ($t_{\frac{1}{2}} = 85$ d, $\beta^{-} = 0.36$ Mev) has been used as a standard for standardization and calibration relative to a 4π counter. The techniques used for determining the disintegration rate by 4π counting were developed by Yaffe et al. (79,80,81,82). The conversion factor, for converting 2π data to 4π data, was determined to be $1.5 \stackrel{+}{=} 0.03$.

Source separation:

An aliquot of slightly acidic erbium chloride solution was pipetted into a clean copper planchet and evaporated gently under infra-red lamp. The source thickness was around $40\,\mu\,\mathrm{gm}\,\mathrm{cm}^2$.

(3) Alpha Measurement

In connection with search for alpha emitters, the above-mentioned 2π -proportional counter has been used at an operating voltage of 1750 volts as determined from Fig. 22. A polonium source, available in the laboratory, has been used as a standard.

Source preparation:

The rare-earth source has been prepared by evaporating and subsequent strong heating of rare-earth solution on a clean copper planchet.

(4) Spectrophotometric determination of Copper

Colorimetric determination of copper with diethyldithiocarbamate⁽⁸³⁾ was chosen because of its convenience and suitability for concentrations of copper that were encountered in the present work. Gravimetric and volumetric methods were discarded because of lack of
PLATEAU CURVE FOR A 2π ALPHA

PROPORTIONAL COUNTER

Discriminator bias setting = 25 volts



suitable (high) concentrations, whereas colorimetry using dithizone and polarographic methods were discarded because of the errors encountered in using such low concentrations (1 - 5y).

The diethyl dithiocarbamate method, which has been found to give very excellent results between the concentration range 10Y to 70Y with a minimum error of 1% to a maximum of 2%, is described below.

- (i) An aliquot containing copper of suitable concentration is transferred into a 25 ml stoppered measuring cylinder.
- (ii) Dilute NH4 OH is added until the solution is alkaline.
- (iii) 2 gms of A.C.S. grade NaCl (solid) and 1 cc of 2% solution of diethyldithiocarbamate are added and the solution is shaken.
- (iv) The yellow complex of copper-diethyldithiocarbamate is extracted with 5 ml portions of isoamyl alcohol and the extract is transferred into a 25 ml volumetric flask and made up to the volume. If the solution is cloudy, a pinch of anhydrous sodium sulphate is added.
- (v) The absorbance is measured at 430 mµ against a blank of the reagents with a Beckman D.U. model spectrophotometer. The calibration curve is shown in Fig. 23.
- (5) <u>Spectrophotometric determination of Rare Earths</u> Colorimetric estimation of rare earths with

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ABSORBANCE vs. CONCENTRATION CURVE

FOR COPPER-DIETHYLDITHIOCARBAMATE



a

Alizarin Red-S⁽⁸⁴⁾ has been chosen in preference to gravimetric and titrimetric procedures which were not suitable for the concentrations that were encountered in the present work. Alizarin Red-S method, besides being very quick and convenient when compared to all other colorimetric procedures listed in the literature, works very satisfactorily in the concentration range 40 to 130Ywith an error of 3 to 5%.

An aliquot containing up to 120 y of the individually purified lanthanide, which is slightly acidic, is transferred into a 10 ml volumetric flask, into which two drops of 1% phenol red had been added. 0.2 M NaOH and 0.03 M HCl are used to turn phenol red indicator colour to 0.5 ml 1 M ammonium acetate and acetic acid vellow. buffer and 2 ml of 0.1% solution of sodium salt of Alizarin sulphonate are now added. The solution is diluted to 10 ml and, after allowing the solution to mix, to develop maximum colour for five minutes, the absorbance is measured at 550 m μ , against a blank of the reagents. Measurements should be done within half-an-hour after the development of the colour because of the instability of the complex. This method can be employed for all rare earths. The standard calibration curve is presented in Fig. 24.

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Figure 24

ABSORBANCE vs. CONCENTRATION CURVE FOR

RARE-EARTH-ALIZARIN RED-S COMPLEX



C. RESULTS

(a) General

Natural and enriched isotope samples of erbium oxide mixed with copper oxide have been irradiated with protons of various energies in the McGill Synchrocyclotron in order to determine the formation cross-sections of various spallation products. In Table III a list of various product nuclides that were identified, as well as the nuclides whose formation cross-sections were determined, are given. Since natural erbium is multi-isotopic, the experimental cross-sections determined were often not directly attributed to only one isotope of erbium, (e.g. Tm^{167} was produced from $Er^{167}(p,n)$ and $Er^{168}(p,2n)$ as well). In order to determine the contribution of individual reactions, natural and isotopically enriched samples of erbium oxide were irradiated with protons. In all the above experiments individual elements of interest were separated and purified, then radiation measurements and chemical yield determinations were made. The measurement of Tm¹⁶⁶ presented a problem because of the non-availability of a convenient photopeak. However, a high energy photopeak of 2.05 Mev was found to occur in the transition, and this was used. The decay of Ho^{161} and Ho^{160m} has been followed using their K-X-rays. Their half-lives being quite similar, 2.5 hrsand 5 hrs respectively, it was necessary to develop a relative and convenient method with

Table III

PROTON IRRADIATION ENERGY													
NUCLIDE	6.5	10.1	12.8	16.4	22.2	28.5	34	39.8	47.4	57	67	77	86.5
_{Tm} 170	ø	x	x	x	ø								
Tm 168	ø	x	x	x	x	x	x	ø					
Tm167	ø	x	x	x	x	x	x	x	x	x	x	x	x
Tm 166	x	x	x	x	x	x	x	x	x	x	x	x	x
Tm165			ø	x	x	x	x	x	x	x	ø		
Tm164*													
Tm ¹⁶³							ø	x	x	x	x	x	x
Tm ¹⁶² Ø													
_{Tm} 161#													
Er ¹⁶⁹				x	x	x	x	x	x	x	x	x	x
Er ¹⁶⁵				x	x	x	x	x	x	x	x	x	x
Er163Ø													
Er ¹⁶¹							x	ø	x	x	x	x	x
Er ¹⁶⁰										x	x	x	x
H0 ¹⁶¹										x	x	x	x
Ho ^{160m}										x	x	x	x
_{Ho} 160*													
Dy ¹ 59											x	x	x
Dy ¹⁵⁷											x	x	x
Dy ¹⁵⁵													ø
x Forma	ation	n cro	ss-se(ction	s wer	e det	erm:	ined.					
* It wa bea	t was not possible to determine formation cross-sections because of short half-lives.												
Ø Ident	tifi	ed, bi	it the	e deca	ay was	s dif:	ficu	alt to	o foli	low	•		
# Ident	tifie	ed by	its g	geneti	ic rel	lation	1.						

which one could follow the decay even at low counting rates. This method was previously indicated in Fig. 19.

In the case of calculation of formation crosssections, particularly in the cases where nuclei were produced by the decay of the parent as well as independently, knowledge of decay schemes was necessary to correct for the decay. In most of the cases literature values helped. However, it is necessary to mention that the decay schemes are not fully known in the rare earth region.

Natural and enriched samples of erbium oxide alone were bombarded with protons in connection with the search for new nuclides as well as mass confirmation studies. A search for Tm^{164} , Tm^{163} , Tm^{162} , and Tm^{161} and for alpha emitters has been made. A few genetic relations have been studied.

In the case of irradiations of the holmium oxide targets, the products of interest were Tm^{168} and Tm^{167} . Radiation measurements did not present any problem. The decay curves showed only a 9.6d component and an 86d component when the 200 kev photopeak was followed.

(b) SPALLATION PRODUCTS AND MONITOR NUCLIDES

In Table IV a list of nuclear properties of various spallation products observed are listed. Nuclear properties of the product nuclides observed in the present work are described in the following pages.

Table IV

Nuclide	Half-life observed	Characteristic photopeaks observed
170	126 [±] 10 d	80 kev
Tm ¹⁶⁸	86 + 3 a	80,100,200,270,490,750,800,1280 & 1450(?) kev
Tm ¹⁶⁷	9.6 ⁺ 0.2 d	115,200,500 & 720 kev
_{Tm} 166	7.7 <u>+</u> 0.1 hr	80,180,700,790,1200,1850 & 2050 kev
Tm ¹⁶⁵	28 <u>+</u> 1.5 hr	200,800,1160 & 1400 kev
Tm^{164}	1.85 ± 0.15 min	
Tm ¹⁶³	1.8 ⁺ 0.15 hr	110 kev
Tm162	90 ⁺ 10 min	
_{Tm} 161	30 ⁺ 10 min	
Er ¹⁶⁹	9.0 ⁺ 0.2 d	
Er ¹⁶⁵	$10.4 \pm 0.1 hr$	
Er ¹⁶¹	$3.1 \stackrel{+}{=} 0.2 \text{ hr}$	200,300,820,1160,1400,1700 & 2000(?) kev
Er ¹⁶⁰	29.5 ± 0.5 hr	·
H0 ¹⁶¹	2.5 ± 0.2 hr	90 kev
Ho ^{160m}	5.0 ± 0.2 hr	90,190,650,730,890 & 970 kev
Dy ¹⁵⁹	138 <u>+</u> 4 a	-
Dy ¹⁵⁷	8.2 <u>+</u> 0.15 hr	320 kev

(i)
$$\underline{Tm^{170}}_{2}$$
 (t₁ = 128d)

This nuclide was found at low energies (up to 16.4 Mev), produced by the (p,n) reactions on Er^{170} (14.9% natural abundance). The decay scheme data were taken from Strominger, Hollander and Seaborg⁽⁸⁵⁾. It was not convenient to measure the β^- radiation alone even though 100% of the decays occur in this manner because of 86-d Tm¹⁶⁸ which emits conversion electrons. The decay of the 84-kev photopeak was followed, and the contribution due to 86-d Tm¹⁶⁸ subtracted. For subtraction purposes, Tm¹⁶⁸ was produced exclusively by a 30-Mev proton irradiation on natural erbium. At this energy Tm^{170} is not produced, since the $Er^{170}(p,n)$ reaction is almost negligible. In Fig. 25 the Tm^{168} spectrum alone and the combined Tm^{170} + Tm¹⁶⁸ spectrum are given. The decay curve of 84-kev photopeak is shown in Fig. 26. The half-life was determined to be $128 \stackrel{+}{=} 10$ days, measured over a period of two half-lives. The large error is due to the low counting rate observed. The half-life observed in the present work is in agreement with the values of Ketelle⁽⁸⁶⁾, Caldwell⁽⁸⁷⁾ and Bothe⁽⁸⁸⁾ whose values are 129d, 120d and 127d respectively. This nuclide was found only at 10.1, 12.8 and 16.4 Mev proton irradiation. It was not possible to observe any trace of it at 22.2 Mev.

(ii) Tm^{168} (t_{1/2} = 86d)

This nuclide was produced by the reactions: $Er^{168}(p,n)Tm^{168}$ and $Er^{170}(p,3n)Tm^{168}$. The gamma-ray

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Figure 25
Low Energy Gamma-ray Spectrum
of Tm ¹⁰⁰ .
Low Energy Gamma-ray Spectrum
$_{of Tm}^{168} + _{Tm}^{170}$



TYPICAL	DECAY	CURVE	OF	THE
84-KEV	PHOTOR	PEAK C	OF Tn	n170



93a

spectrum is shown in Fig. 27, and the decay curve of the 0.2 Mev gamma ray, measured over a period of five half-lives, is presented in Fig. 28. The half-life was determined to be 86 $\stackrel{+}{}$ 3d which was in agreement with Wilkinson's⁽⁸⁹⁾ value of 85 d and Handley's⁽⁹⁰⁾ value of 87d. The decay scheme data were obtained from the Nuclear Data sheets⁽⁹¹⁾. The observed gamma peaks are in agreement with Jacob et al.⁽⁹²⁾ and Kochevanov et al.⁽⁹³⁾. At 6.1 Mev proton irradiation, the amount produced was so small that the decay could not be followed, whereas at proton energies greater than 39.8 Mev this nuclide was not observed in the spallation products.

In the case of the alpha irradiations of holmium oxide, this nuclide was quantitatively determined at alpha energies of 15, 20 and 25 Mev only. The threshold for the reaction is 10 Mev, so the nuclide was not observed below this bombarding energy. At 30 Mev the cross-section for the (α, n) reaction had already fallen to very low values.

(iii) $\underline{Tm^{167}}(t_{\frac{1}{2}} = 9.6d)$

This was produced by the reactions: $Er^{167}(p,n)$, $Er^{168}(p,2n)$, and $Er^{170}(p,4n)$. This nuclide was found in all the proton irradiations of natural erbium. At 6.1 Mev the decay could not be followed because of very low production. However, traces of the production have been observed. The very low production was due to the fact that the irradiation energy is just at the threshold. Gamma peaks of energies 110, 200, 510 and 710 kev, as well as

TYPICAL GAMMA-RAY SPECTRUM OF Tm¹⁶⁸

- Low Energy gamma-ray spectrum of Tm¹⁶⁸.
- (2) High Energy gamma-ray spectrum of Tm¹⁶⁸.



TYPICAL DECAY CURVE OF THE 0.2 MEV

PHOTOPEAK (UP TILL 34 MEV IRRADIATIONS

• $t_{\frac{1}{2}} = 9.6 \text{ d.}$ • $t_{\frac{1}{2}} = 86 \text{ d.}$



a K-X-ray, have been observed, decaying with 9.6 + 0.2d half-life. In Fig. 29, a typical decay curve of 0.2 Mev gamma ray, measured over a period of six half-lives decaying with $9.6 \stackrel{+}{-} 0.2d$ half-life, is given. Up till bombarding energies of 39.8 Mev the decay curve was resolved, as is shown in Fig. 28. Above 39.8 Mev the 0.2 Mev photopeak decayed with a 9.6 d half-life only. The gamma peaks observed are in agreement with those published by Nervik and Seaborg (19) and Narasimhia and Pool (94)who recently published the complete decay scheme of Tm^{167} . According to them, the 200 kev photopeak is an E3 transition with 98% branching ratio. The conversion coefficient was obtained from Rose's (95) tables to be 1.669. The crosssections reported in the present work are independent and absolute.

In the case of alpha irradiations, this nuclide was observed at alpha energies greater than 20 Mev. The decay curves were similar to those observed in the proton spallation products, as mentioned above.

(iv) Tm^{166} (t₁ = 7.7 hr)

This nuclide was produced in the present work by the nuclear reactions: $Er^{166}(p,n)$, $Er^{167}(p,2n)$, $Er^{168}(p,3n)$ and $Er^{170}(p,5n)$. The photopeaks observed are in agreement with the recent work of Pool and Wilson⁽⁹⁶⁾. Poor resolving power of the detector made the identification of all the photopeaks quite difficult. However, the 1850 kev and 2050 kev gamma peaks were identified by us one year

TYPICAL DECAY CURVE OF THE 0.2 MEV

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PHOTOPEAK IN A Tm ACTIVITY

(From 40 Mev irradiation onwards)



earlier than the publication of Pool and Wilson. The two high-energy photopeaks are shown in Fig. 30. In the present work, the 2050 kev photopeak has been used to calculate the relative cross-sections. The decay curve of the 2050 photopeak, measured over a period of 8 half-lives, is presented in Fig. 31. The half-life of 7.7 $^{\pm}$ 0.1 hrs is in agreement with Wilkinson and Hicks⁽⁸⁹⁾, Michel and Templeton⁽⁹⁷⁾ and Pool and Wilson⁽⁹⁶⁾. It is necessary to mention that Pool and Wilson's⁽⁹⁶⁾ decay scheme is not complete, so that only relative cross-sections are reported. (v) Tm^{165} (t₁ = 28 hr)

This nuclide was produced by the reactions: $Er^{166}(p,2n)$, $Er^{167}(p,3n)$, $Er^{168}(p,4n)$ and $Er^{170}(p,6n)$. The photopeaks observed were 220, 300, 830, 1160, and 1380 These observations are in agreement with Handley (98) kev. and Gorodinskii⁽⁹⁹⁾. The decay of the 1160 kev photopeak is plotted in Fig. 32. The half-life determined in the present work was 28 ± 1.5 hrs, measured over a period of six half-lives, whereas Handley's value was 24.5 hrs and that of Gorodinskii 29 hrs. Since the decay scheme is not known, only relative cross sections are reported here. This nuclide was not observed at bombarding energies greater than 67 Mev. This was due to very low production of this nuclide at high energies and to the fact that the photopeak chosen might have been masked due to the presence of other nuclides.

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Figure 30

HIGH ENERGY GAMMA-RAY SPECTRUM OF

Tm ACTIVITIES



100a

TYPICAL DECAY CURVE OF THE 2050 KEV

PHOTOPEAK OF Tm¹⁶⁶ ACTIVITY

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TYPICAL DECAY OF THE 1.16 MEV

PHOTOPEAK OF Tm165



(vi)
$$Er^{169}$$
 (t_{1/2} = 9d)

This nuclide was produced chiefly by the reaction $Er^{170}(p,pn)$. The decay of this nuclide was followed by measuring the decay of the 0.34 Mev β^- with the help of a $2\pi\beta$ -proportional counter calibrated relative to a $4\pi\beta$ proportional counter. A typical decay curve is presented in Fig. 33. The half-life of $9 \stackrel{+}{-} 0.2$ days, measured over a period of six half-lives, is in agreement with Butement's (100) value of 9d, but not with Ketelle's (101) and Pavlotskaia's (102) whose values are respectively 9.4 and 9.5 days, which are The counting rate measured was converted into rather high. a disintegration rate by multiplying by 1.5 \pm 0.03 which was determined earlier. The decay scheme data were taken from the reference (85) and it was possible to calculate independent and absolute cross sections.

No attempt has been made to observe the 8.4 kev X-ray which has been reported in the reference (103) because erbium was not carrier-free.

(vii) Er^{165} (t₁ = 10.4 hr)

This nuclide was produced by the nuclear reactions $Er^{166}(p,pn)$, $Er^{167}(p,p2n)$, $Er^{168}(p,p3n)$, and $Er^{170}(p,p5n)$ and by a small decay from the parent Tm^{165} ($t_{\frac{1}{2}} = 28$ hr) until the chemical separation had taken place. The decay of the nuclide was followed through the measurement of its characteristic K-X-ray. The decay curve is presented in Fig. 34 in which it was resolved by subtracting the 29.5 hr

TYPICAL DECAY CURVE OF THE

0.34 Mev β^- of Er¹⁶⁹





	TYPICAL	DECAY	CURVI	OF THE
	K-X-RAY	OF AN	ERBIUN	FRACTION
	(For in grea	radiat ter th	ions (an 55	of energy Mev)
•	t <u>1</u> 2	10.4	hr of	Er ¹⁶⁵
0	t <u>1</u> =	29.5	hr of	Er ¹⁶⁰


Er¹⁶⁰ component. This type of decay curve was obtained when erbium was bombarded with protons of energy greater than 55 Mev, in which case the purified erbium fraction will have 3.1-hr Er^{161} . 10.4-hr Er^{165} and 29.5-hr Er^{169} . typical decay curve of the pure 10.4-hr Er¹⁶⁵, obtained at less than 50 Mev irradiations, is shown in Fig. 35. The half-life of 10.4 ± 0.1 hr, measured over a period of six half-lives, is in agreement with Grigorev et al. (104) whose value is 10.5 hr, whereas Butement's (105) value and that of Kundu et al. (106) of 10 \pm 0.1 hr and 9.9 hr respectively were not favoured. The formation cross sections reported in the present work are independent and absolute. However. as discussed later. there is a bit of uncertainty because the decay schemes of Tm^{165} and Er^{165} are not fully known.

Three irradiations were done on holmium oxide in search of gamma rays and to establish the half-life of ${\rm Er}^{165}$. After chemical separation, the decay was followed for ten half-lives using a well-type scintillation counter (Baird Atomic 810A). The half-life was found to be 10.4 $\stackrel{+}{-}$ 0.1 hrs. As has been reported by several authors, no gamma ray has been observed. Two extra irradiations were done in order to verify whether an unknown activity of half-life about 2 hours belonged to erbium. Fluoride precipitation followed by cation exchange separation of individual rare earths proved the foreign activity was due to an impurity in the holmium oxide.

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TYPICAL DECAY CURVE OF THE K-X-RAY

OF AN ERBIUM FRACTION

(For proton energies less than 50 Mev)



(viii) $Er^{161} (t_{\frac{1}{2}} = 3.1 hr)$

This nuclide was produced by the nuclear reactions: Er¹⁶²(p,pn), Er¹⁶⁴(p,p3n), Er¹⁶⁶(p,p5n), Er¹⁶⁷(p,p6n) and $Er^{168}(p,p7n)$. It was also produced through the decay of the short-lived (\sim 30 minute) Tm¹⁶¹ which was produced by (p,xn) reactions on erbium. Since Er^{162} and Er^{164} isotopes are of very low abundance in natural erbium, the amount of activity that was produced through these isotopes was assumed to be negligible. The activity was observed in measurable amounts from 34 Mev onwards. Individual separations were done $2\frac{1}{2}$ hours after the end of irradiation in order to allow Tm¹⁶¹ to decay virtually completely. The gamma-ray spectrum is presented in Fig. 36. The photopeaks observed were confirmed by the recent work of Grench and Burson⁽¹⁰⁷⁾ who published the complete decay scheme, which helped in calculating the independent formation crosssections of Er¹⁶¹ and Ho¹⁶¹. It was not possible to identify their very low intensity photopeak of energy 1800 A typical decay curve of the 820 kev photopeak is kev. presented in Fig. 37. The half-life of $3.1 \stackrel{+}{-} 0.2$ hr determined is in agreement with Nervik's (19) value of 3.01 hrs but not with Handley's (108) and Michel's (97) whose values are 3.6 hrs and 3.5 hrs respectively.

Analysis of the erbium fraction obtained from 57, 67, 77 and 86.5 Mev proton irradiations was complicated by the fact that Ho^{160m} ($t_{\frac{1}{2}} = 5$ hr) grows due to the decay chain $Er^{160} \rightarrow Ho^{160m}$, which masks the 820 kev peak. Of course

TYPICAL GAMMA-RAY SPECTRUM OF

3.1 hr Er¹⁶¹

(1) Low Energy Gamma-Ray Spectrum

(2) High Energy Gamma-Ray Spectrum



CHANNEL

109a

TYPICAL DECAY CURVE OF THE 820 KEV

PHOTOPEAK OF THE Er¹⁶¹



110a

this complication arose only after Er¹⁶¹ had been measured for a few half-lives. The growth is indicated in Fig. 38, which, after some time, finally looks like a Ho^{160m} spectrum as shown in Fig. 40.

(ix) $\operatorname{Er}^{160}(t_{\frac{1}{2}}=29.5 \text{ hr})$

This nuclide was produced by the reactions: Er¹⁶²(p,p2n), Er¹⁶⁴(p,p4n), Er¹⁶⁶(p,p6n), Er¹⁶⁷ (p,p7n) and $Er^{168}(p,p8n)$ and also through the decay of Tm^{160} (?) of The nuclide, Er¹⁶⁰, was reported for very short half-life. the first time by Nervik and Seaborg⁽¹⁹⁾ from tantalum spallation products. Since then it has been reported in several references (109,110). The decay of the nuclide was followed by measuring its K-X-ray. The decay curve is shown in Fig. 34, in which 10.4-hr Er^{165} and 29.5 \pm 0.5 hr Er¹⁶⁰ have been resolved. The half-life determined is in agreement with values previously obtained, viz. 29.4 hr⁽¹⁹⁾, 29.5 $hr^{(109)}$, and 30 $hr^{(110)}$. By the time the measurement had begun (around 20 hours after the irradiation), Ho^{160m} was already in secular equilibrium with Er^{160} and decaying with a half-life of 29.5 hrs. So, in the decay curve, it was easy to resolve 10.4-hr Er^{165} and 29.5-hr Er^{160} , as is shown in Fig. 34. The cumulative cross sections reported are absolute. The K-X-ray abundance factor reported by Nervik and Seaborg, as mentioned by Grover (109), has been used.

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VARIATION OF THE SHAPES OF THE GAMMA-RAY SPECTRA WITH TIME IN THE ERBIUM FRACTION INDICATING THE GROWTH OF Ho^{160m}

(1) $1\frac{1}{2}$ hours after separation of erbium fraction

(2) 14 hours after separation of erbium fraction



(x)
$$Ho^{161}$$
 (t₁ = 2.5 hr)

This nuclide was produced independently in the present work by the reactions $Er^{162}(p,2p)$, $Er^{164}(p,2p2n)$, Er¹⁶⁶(p,2p4n), Er¹⁶⁷(p,2p5n), Er¹⁶⁸(p,2p6n) and Er¹⁷⁰(p,2p8n). It was also produced through the decay of 3.1 hr parent Er^{161} . The decay curve was followed by measuring its K-Xray. The decay curve is shown in Fig. 39, in which the 2.5-hr decay was resolved from the 5-hr activity. The half-life of 2.5 ± 0.2 hr, measured over a period of four half-lives, was in agreement with Toth's (60) and Handley and Olson's (108) values which are consistently 2.5 hr. The reported cross sections in the present work were independent and absolute. The correction for the parent decay was possible by the decay scheme published recently by Grench and Burson⁽¹⁰⁷⁾.

The K-X-ray decay curve resolution into 2.5-hr and 5-hr components was possible with the X-ray measurement method developed earlier. Computer methods developed to resolve decay curves of close half-lives would have given more accurate values than the graphical way of resolution method used in the present work.

(xi) Ho^{160m} (t₁ = 5 hr)

This nuclide was produced in the present work as a spallation product by the reactions $Er^{162}(p,2pn)$, $Er^{164}(p,2p3n)$, $Er^{166}(p,2p5n)$, $Er^{167}(p,2p6n)$, and $Er^{168}(p,2p7n)$, as well as by the decay of Er^{160} . The spectrum of this nuclide is presented in Fig. 40. The intense photopeaks

	- 114 -
	Figure 39
	<u></u>
YPICAL	DECAY CURVE OF THE K-X-RAY
	OF A HOLMIUM FRACTION
-	
٠	$t_{\frac{1}{2}} = 2.5 - hr Ho^{161}$
0	$t_{\frac{1}{2}} = 5 - hr Ho^{160m}$



TYPICAL GAMMA-RAY SPECTRUM

of Ho^{160m}



of energies 87, 190, 650, 730, 890 and 970 kev plus X-rays of 46 kev were observed. The decay curve was followed through the measurement of the K-X-ray. The 2.5-hr component and 5-hr component are shown in Fig. 39. The halflife measured was $5 \stackrel{+}{-} 0.2$ hr, measured over a period of five half-lives, is in agreement with Nervik's (19), Mihelich's(111) and Wilkinson's (112) values which are consistently 5 hours. The independent formation crosssection calculations were possible with the help of the K-X-ray measurement. The K-X-ray abundance for Tm¹⁶⁰ and Ho^{160m} was reported to be 0.82 by Nervik and Seaborg as obtained from Grover's thesis⁽¹⁰⁹⁾. In spite of the error involved in resolving 2.5-hr and 5-hr half-lives from the composite decay curve, the K-X-ray measurement was preferred because of the availability of an abundance factor, and due to the reliable method developed for K-X-ray measurement as described earlier.

(xii) Dy^{159} (t₁ = 138d)

In the present work this nuclide was produced by the decay of the $\backsim{30}$ -minute Ho¹⁵⁹ independently by $\mathrm{Er}^{162}(\mathrm{p},\mathrm{3pn}), \mathrm{Er}^{164}(\mathrm{p},\mathrm{3p3n}), \mathrm{Er}^{166}(\mathrm{p},\mathrm{3p5n}) \mathrm{Er}^{167}(\mathrm{p},\mathrm{3p6n}),$ and $\mathrm{Er}^{168}(\mathrm{p},\mathrm{3p7n})$. In order to allow the 30-minute parent Ho¹⁵⁹ to decay almost completely, the rare-earth separations were done $2\frac{1}{2}$ hours after the end of irradiation. The decay of Dy¹⁵⁹ was followed through the K-X-ray. Extremely low counting rates led to poor statistics. A typical decay curve is shown in Fig. 41, in which the peak height was

TYPICAL DECAY CURVE OF THE K-X-RAY

of Dy¹⁵⁹



plotted against decay time (days). The half-life found, 138 ± 5 days, measured over a period of two half-lives, is in reasonable agreement with the previous work of Butement⁽¹¹³⁾ and Ketelle⁽¹¹⁴⁾ whose values are respectively 140 days and 144.4 days. The cross sections reported in the present study are cumulative.

(xiii) Dy^{157} (t₁ = 8.2 hr)

This nuclide was produced by the decay of shortlived parent Ho¹⁵⁷ (unknown) which was produced by (p,2pxn) reactions on erbium, as well as independently just like Dy^{159} . Direct formation was possible through $Er^{162}(p, 3p3n)$, Er¹⁶⁴(p,3p5n), Er¹⁶⁶(p,3p7n), and Er¹⁶⁷(p,3p8n) reactions. The decay was followed by measuring the counting rate under the 320 kev peak. A typical decay curve is shown in Fig. The spectrum is presented in Fig. 43, which is in 42. agreement with the work of Toth and Nielsen⁽¹¹⁵⁾. The half-life, $8.2 \stackrel{+}{-} 0.2$ hrs measured over a period of six halflives, is in agreement with Toth and Nielsen⁽¹¹⁵⁾, Handley and Olson⁽¹¹⁶⁾ and Dzelepove et al.⁽¹¹⁷⁾ whose values are consistently 8.2 hrs. The 8.5-hr half-life, as reported by Gorodinskii et al. (110), might be due to contribution from 10-hr Dy¹⁵⁵ which has a photopeak adjacent to 320 kev peak. The decay scheme data were obtained from Toth and Nielsen⁽¹¹⁵⁾. The abundance of the 320 kev photopeak is 100%. The cross sections reported in the present work are cumulative and absolute.

TYPICAL DECAY CURVE OF THE 320 KEV

PHOTOPEAK OF Dy157



119a

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TYPICAL GAMMA-RAY SPECTRUM

of Dy¹⁵⁷



$$(xiv) Zn^{63} (t_1 = 38.4 min)$$

This nuclide was used as a monitor in the lowenergy irradiations (up to 12.8 Mev). It was possible to follow the decay of the 511 kev photopeak directly from the erbium spallation product mixuture in the case of low-energy irradiations. A typical decay curve is shown in Fig. 44. The monitor cross-section data were taken from Fig. 1 and the decay scheme data from the literature⁽⁸⁵⁾.

 $(xv) Cu^{64} (t_1 = 12.8 hr)$

This nuclide was used as a monitor for proton irradiations greater than 12.8 Mev. A typical decay curve of the 511 kev photopeak is shown in Fig. 45 in which, at the start of the measurement, a short-lived component is present. This was due to 3.3-hr Cu⁶¹, a positron emitter, produced as a copper spallation product from 30 Mev proton energy onwards. In practice, the monitor nuclide was measured about 20 hours after the irradiation. The decay scheme data were taken from the literature⁽⁸⁵⁾. The monitor cross section data were derived from Fig. 2.

(c) NEW ISOTOPES AND MASS ASSIGNMENTS

(i) Search for alpha emitters:

Alpha emitters in the present work are highly neutron-deficient nuclides. One can predict the probability for producing alpha emitters in the mediumheavy elements by combining α -decay systematics in this region with known data deduced from mass differences

TYPICAL DECAY CURVE OF THE 511 KEV

PHOTOPEAK OF Zn⁶³



122a

		- 123	-			
	I	Figure	45			
	-					
TYPICAL	DECAY	CURVE	OF	THE	511	KEV
PHO	TOPEAK	OF Cu ⁶	54	ACTIN	VITY	



obtained from the Cameron mass formula⁽¹¹⁸⁾. Since the possible alpha emitters will be very neutron deficient, they will decay by positron emission or electron capture with half-lives of a few hours at most. On the other hand, because the available alpha energies are relatively low, the partial half-lives for alpha decay would be very long, and therefore the chances for detection of new alpha emitters are quite small.

Even though Toth⁽⁶⁰⁾ has observed alpha emitters in the very neutron-deficient region of Tb and Dy, in the present work no alpha emitters of measureable half-life (>30 mins) could be identified in the more neutron-deficient regions of Tm and Er. Irradiation of erbium enriched in Er^{162} with protons of energy 86.5 Mev, chemical purification of individual elements and subsequent measurement of activity, using a $2\pi \alpha$ -proportional counter, led to this conclusion. This finding is in agreement with that of Merz⁽¹¹⁹⁾ who, on bombarding erbium with 400 Mev protons, also could not identify any alpha emitters.

(ii) New neutron-deficient isotopes:

Tm¹⁶³, Tm¹⁶¹ and Tm¹⁶⁴, Tm¹⁶², the odd and even mass nuclides respectively have been discussed.

During the progress of the work, five other groups were also working on the study of new isotopes of thulium. These groups are Harmatz et al. at Oak Ridge, the first to obtain enriched isotopes of erbium; Pool and Wilson at Ohio; Butement and Glentworth at Liverpool, England; Abdurazakov et al. in Russia, and Nielsen in Denmark.

Enriched and natural isotope samples of erbium have been irradiated in connection with the search for new isotopes of thulium, and the details of the study are presented below.

(1)
$$\text{Tm}^{163}$$
 (t₁ = 1.8 hr)

- This nuclide was first observed tentatively in 34 Mev proton irradiation products of erbium and more definitely in the 39.8 Mev proton irradiation products of erbium. In spite of the poor resolution of the detector, a 110 kev peak was observed. The gamma spectrum and the decay curve are shown in Figs. 46 and 47 respectively. The half-life and the photopeak being reasonably convenient, relative cross sections have been reported.
- About this time Harmatz et al.⁽¹²⁰⁾ reported on Tm¹⁶³ and Tm¹⁶¹. They analysed enriched isotope sample irradiation products by means of a conversion electron spectrometer, photographic recorders and permanent magnet spectrographs, and established the decay chains:

 $Tm^{163} \xrightarrow{2 \text{ hr.}} Er^{163} \xrightarrow{75 \text{ min}} Ho^{163} \xrightarrow{.8 \text{ sec}} Dy^{163} (\text{stable})$ $Tm^{161} \xrightarrow{30^{+} 10 \text{ min}} Er^{161} \xrightarrow{3.1 \text{ hr}} Ho^{161} \xrightarrow{2.5 \text{ hr}} Dy^{161} (\text{stable})$

LOW ENERGY GAMMA-RAY SPECTRUM OF Tm ACTIVITY

1출 HOURS AFTER THE 40 MEV IRRADIATION

(110 kev photopeak decayed with 1.8 hr half-life)



TYPICAL DECAY CURVE OF THE 110 KEV

PHOTOPEAK OF Tm¹⁶³


After this publication, enriched isotope samples of erbium were obtained from Oak Ridge. Erbium enriched in Er¹⁶⁴ was irradiated with 20 Mev protons and the decay of Tm^{163} , which was produced through the Er¹⁶⁴(p,2n) reaction, was followed. The halflife observed was 1.8 - 0.15 hr measured, whereas the value given by Harmatz et al. (120) was 2 hrs. Much importance has not been given to Glentworth and Butement's work⁽¹²¹⁾ which guoted a value of In their work they irradiated natural 1.9 hr. erbium with a few hundred Mev protons and followed the gross decay of thulium fraction in which several decays and several growths can be observed. This type of study limits the validity of the value. It can only be good if the characteristic photopeaks have been followed like Harmatz et al. (120). In the present work no attempt has been made to look for the other photopeaks as reported by Nielsen⁽¹²²⁾ who obtained the data using a mass analyser and a high resolution scintillation spectrometer. Nielsen reported the intensities of the photopeaks of Tm^{163} as 115 (8), 171 (1), 240 (5), 290 (3) and 355 (3) kev. Intensities in the brackets are in percent of total X-ray intensity. (2) Tm^{161} (t₁ = >20 min and \sim 30 min)

This nuclide was identified in the present work only through the study of its daughter 3.1 hr-Er¹⁶¹.

Erbium oxide enriched in Er¹⁶² was irradiated with 20 Mev protons and the thulium fraction was measured 100 minutes after the end of the irradiation. No photopeak decaying with about 30-minute half-life was observed. However, an 820 kev photopeak decaying with about 3-hr half-life was observed, which was attributed to 3.1-hr Er¹⁶¹. The decay curve is presented in Fig. 48. If the enrichment of Er¹⁶² were higher and the separation faster, much concrete information would have been obtained. The 3.1-hr Er¹⁶¹ could be seen in the thulium fraction even after 100 minutes after the irradiation, in spite of the fact that very little activity is produced on account of the low (6%) abundance of The half-life of the Er¹⁶¹ parent must then Er^{163} . have been more than 20 minutes and round about 30 This observation is in line with that of minutes. Harmatz et al. (120) who claim 30 - 10 mins, whereas Glentworth and Butement's (121) claim, 32 mins, was not at all considered to be genuine due to the lack of reasonable experimental support. On the assumption that no reasonable information could be obtained on account of the delay involved in the separation and the low enrichment of the Er¹⁶² available (6%), the present work was therefore not extended to determine the half-life precisely.

Figure 48

DECAY CURVE OF THE 820 KEV OF PHOTOPEAK OF Er¹⁶¹ ALREADY GROWN IN THE SEPARATED Tm FRACTION; DUE TO THE DECAY OF ~ 30-MIN Tm¹⁶¹



130a

(3)
$$\text{Tm}^{164}$$
 (t₁ = 1.85 $\stackrel{+}{-}$.15 min)

- Harmatz et al.⁽¹²⁰⁾ concluded in the case of Tm^{164} and Tm^{162} , the even-mass isotopes, that either the associated half-lives were too short ($\langle \frac{1}{2} hr \rangle$) or the energies available were not sufficient to allow decay to levels which depopulate with observable internally converted transitions.
- In connection with the search for Tm^{164} , erbium oxide enriched in Er^{164} was irradiated for two minutes with protons of energy 7 Mev, and the decay of the K-X-ray was followed immediately after the irradiation, without doing any chemical separations, with the help of a multichannel pulseheight analyzer* close to the Cyclotron.
- The decay of the K-X-ray is presented in Fig. 49. The half-life observed is almost in agreement with Pool and Wilson⁽¹²³⁾ whose value was 2 mins. The existence of the isotope was also confirmed by Abdurazakov et al.⁽¹²⁴⁾ who established the decay chain:

Yb¹⁶⁴ 75 min Tm^{164} 2 min Er^{164} (stable). The 90-kev transition reported by Pool and Wilson⁽¹²³⁾ could not be observed because of Compton scattering and the other contributions from the 511 kev peak of N¹³ produced through O¹⁶(p, α)N¹³. The 90-kev

^{*}I am indebted to Professor R.E. Bell of the Radiation Laboratory, McGill University, for allowing me to use their 256 channel pulseheight analyser.

Figure 49

DECAY CURVE OF THE K-X-RAY OF

	Tm ACTIVITY				
	1 <i>6b</i>				
۲	1.85-min Tm ¹⁰⁴				
0	∽ 7.7-hr Tm ¹⁶⁶				



132a

photopeak could have been identified if the N^{13} was driven out by dissolving the target in hydrochloric acid. In the present work mass assignment would have been more complete if the (p,n) excitation function would have been studied. This study may avoid the doubt, if any, in not doing chemical separations on the target.

(4) Tm^{162} (t₁ = 90 ⁺ 10 min)

An attempt has been made to prove the existence of this nuclide.

- Erbium oxide, enriched in Er¹⁶², was irradiated with protons of energy 7 Mev for a duration of around two hours; the thulium fraction was separated and the decay of the K-X-ray was followed. The decay curve is shown in Fig. 50. In the first ten hours' duration the composite decay curve has a half-life around six hours and later on lengthens to around eight hours. The behaviour was observed consistently in two experiments.
- If there were no Tm^{162} of measurable half-life, the decay curve would have shown only a half-life of eight hours or longer due to the presence of Tm^{166} (t₁ = 7.7 hr) and Tm^{167} (t₁ = 9.6d).
- The decay curve, as is shown in Fig. 50, on resolution indicated definitely a short-lived 90 ⁺ 10 min component. No specific photopeak has been observed decaying with this half-life except the K-X-ray.

Figure 50

i

DECAY CURVE OF THE K-X-RAY OF THE

	SEPARATED Tm ACTIVITY
0	Tm^{166} (t ₁ = \sim 7.7 hr)
٠	Tm ¹⁶² (t <u>i</u> = ∽90 min)
	:



been highly converted.

The experiments in the present work were not very conclusive, with special reference to assignment of correct half-life, because of low (up to 6%) enrichment of Er^{162} in erbium target. An obvious extension of this work is to irradiate highly enriched Er^{162} . Pool and Wilson⁽¹²³⁾ reported a half-life of 77 mins when they irradiated up to 14% enriched Er^{162} with 6-Mev protons.

(iii) <u>Mass Assignments</u>

In the present work the following decay chains have been studied:

(1)
$$\operatorname{Tm}^{161} \longrightarrow \operatorname{Er}^{161} \longrightarrow \operatorname{Ho}^{161} \longrightarrow \operatorname{Dy}^{161}$$
 (stable)

and (2)
$$\operatorname{Tm}^{160}$$
? \longrightarrow Er^{160} ? $\xrightarrow{\operatorname{Ho}^{160}}$ Dy^{160} (stable).

Erbium oxide enriched partially in Er^{162} (up to 6%) was irradiated with protons of energy 20 Mev. The thulium fraction was separated and an 820-kev photopeak decay has been followed. The decay curve indicated the presence of 3.1-hr Er^{161} . The growth could not be followed due to the following reasons:

- At the time of measurement only about 12% of the activity of Tm¹⁶¹ was present.
- (2) Since the nuclide was produced from 6% abundant Er^{162} , the production was very small.

- 135 -

(3) The intense contribution of the other nuclides of thulium.

It is necessary to mention that the decay of Tm^{161} could not be measured because no photopeak of reasonable intensity was observed. No attempt has been made to resolve the K-X-ray peak which consisted of six decays and three growths (decay of Tm^{168} , Tm^{167} , Tm^{166} , Tm^{165} , Tm^{163} and Tm^{162} , and growth of Er^{165} , Er^{161} and Er^{163}).

However, it has been concluded that the Tm^{161} half-life is of the order >20 mins and 50 mins, since the 3.1-hr daughter was observed in the thulium fraction even after 100 minutes after the end of irradiation. If the half-life were $\langle 20 \text{ mins}$, the 3.1-hr Er^{161} would have decayed by the time of separation (100 minutes after the irradiation).

From the erbium fraction another separation was made to obtain holmium, in which the 2.5-hr activity was observed. The gamma spectrum and the decay curve are shown in Figs. 51 and 52 respectively. This observation clearly indicated the activity to be an isotope of holmium. Since the activity had not been observed in bombardments of unenriched samples and only in those enriched in Er^{162} , the 2.5-hr activity is confirmed as belonging to Ho^{161} .

Thus, using reasonably quick ion-exchange separation techniques and multichannel pulseheight analysis, the final result has been established:

 Tm^{161} <u>30 min</u> Er^{161} <u>3.1 hr</u> Ho¹⁶¹ <u>2.5 hr</u> $\operatorname{Dy}^{161}(\operatorname{stable})$

Figure 51

GAMMA-RAY SPECTRUM OF Ho¹⁶¹



Figure 52

DECAY CURVE OF THE K-X-RAY OF Ho¹⁶¹

anne gelille stern fin a wennen – "s.a. menn alter den al feite beis fin a dag some fin i ven beg post gen gen gen after den den det Bis bereiten an gegener



This decay chain has been studied also by Harmatz et al.⁽¹²⁰⁾ using a conversion electron spectrometer, photographic recorder and permanent magnet spectrometer.

In the case of the second decay chain, natural erbium was irradiated with 70-Mev protons for a duration of one hour, and erbium was separated $2\frac{1}{2}$ days after the irradiation. Immediately after the separation, the growth of the 650 + 730 kev photopeak was followed. The decay curve is shown in Fig. 53. The $\backsim 30$ -min component corresponded to Ho¹⁶⁰ ($t_{\frac{1}{2}} = 22$ min), the 5-hr component corresponded to Ho¹⁶⁰ ($t_{\frac{1}{2}} = 5$ hr), and the 29.5-hr component indicated that Ho¹⁶⁰ is in secular equilibrium with Er^{160} . Thus the decay chain:

$$Tm^{160}$$
 (unknown and ?) $\to Er^{160}$ Ho^{160m} Dy^{160} (stable)

was confirmed.

The chart of the nuclides, filled in according to the results obtained in the spallation product studies and new isotope studies, is presented in Fig. 54.

(d) FORMATION CROSS SECTIONS

As stated earlier in the Introduction, Section (1), the formation cross sections can be calculated from the formula:

Figure 53

GROWTH CURVE OF THE 650 + 730 KEV PHOTOPEAK OF

Hol	60m	IN	THE	SEPAR	ATED	ERBIUM	FRACTI	ON
•	-	01	igir	nal de	сау с	urve.		
0	-	De	ecay	curve	afte	er subti	raction	of
			29.5	5-hr E	r ¹⁶⁰ .			
		D			- 64 -			. 0
•	-	De	ecay	curve	arte	r subti	action	01
			5-h	. но ¹⁶	Om ac	tivity.		



Figure 54

CHART OF THE NUCLIDES IN THE REGION OF ERBIUM, BROUGHT UP TO DATE (AUGUST 1961)

AFTER INCLUDING THE RESULTS OBTAINED

IN THE PRESENT WORK



NEUTRON

NUMBER

$$\sigma_{p} = \frac{\sigma_{m} D_{p}^{o} W_{Cu} M_{Er} (1 - e^{-\lambda_{m}t})}{D_{m}^{o} W_{Er} M_{Cu} (1 - e^{-\lambda_{p}t})}$$

where:

- σ_{p} and σ_{m} = the cross sections in millibarns of product and monitor nuclei. D_{m}^{o} and D_{p} = disintegration rates of monitor and product
- nuclei at end of bombardment.
- W_{Cu} and W_{Er} = weight of monitor and target respectively corrected for isotopic abundance.
- ^MCu and ^MEr = atomic weight of monitor and target respectively.

$$\lambda_{m}$$
 and λ_{p} = decay constants of monitor and product nuclei.
. t = length of bombardment in minutes.

The most recent decay scheme data have been taken into account for the monitor nuclides, as well as for the rare earth nuclides of interest, to calculate D_m^o and D_p^o respectively. In this connection, Tables V(a),(b),(c) and (d) have been presented in which necessary decay scheme data and conversion factors for peak heights to areas are given.

In the case of Tm¹⁶⁶, Tm¹⁶⁵ and Tm¹⁶³, no complete decay schemes have been published. In these instances, counting rates only have been used instead of disintegration rates, to calculate relative cross sections instead of absolute cross sections in millibarns.

As explained earlier, Meadows' values have been preferred for monitor cross sections obtained from Figs. 1 and 2. Thus σ_m values were obtained. In Table VI, σ_m , D_m^o , W_{Cu} and t values are given. The value for $\frac{M_{Er}}{M_{Cu}}$ is

TABLE V(a)

CASE OF NUCLIDES WHOSE GAMMA RAYS HAVE BEEN MEASURED

N		t <u>1</u>	Energy of the characteristic	Branching ratio		Type of cross-	Reference	
Nuclide	Present work	Literature values	photopeak measured	factor B.R.	l + αT factor	section measured	of B.R.and aT factors	
_{Tm} 170	128 d	127, 129	0.084 Mev	0.24	5.6	A, I	(85)	
Tm^{168}	86 d	85, 87	0.200 Mev	0.81	1.207	A, I	(91)	
$_{Tm}$ 167	9.6 đ	9.6, 9.4	0.200 Mev	0.98	2.669	A, I	(94) (95)	
Tm166	7.7 hr	7.7	2.05 Mev	-	-	R, I		
$_{Tm}$ 165	28 hr	24.5, 29	1.16 Mev	-		R, I		
Tm ¹⁶³	1.8 hr	2 hr	0.105 Mev	-	-	R, I		
Er ¹⁶¹	3.1 hr	3.6, 3.5	0.820 Mev	0.56	1	A, C	(107)	
Dy ¹⁵⁷	8.2 hr	8.2,8.5,8.5	0.320 Mev	1.00	1	A, C	(115)	

- A represents absolute cross section
- C represents cumulative cross section
- I represents independent cross section
- R represents relative cross section

TABLE V(b)

CASE OF NUCLIDES WHOSE K-X-RAY HAS BEEN MEASURED

		t ₁	Fluenceence	Totol	Type of	Reference of	
Nuclide	Present work	Literature values	yield factor (F.Y.F.)	X-rays/ K-X-rays	section measured	K-X-rays and F.Y.F.	
Er ¹⁶⁵	10.4 hr	11.2,9.9,9.5	•97	2.2	A, I	(91),(135)	
Ho ¹⁶¹	2.5 hr	2.5	•96	1.33	A, I	(18),(135)	
Dy ¹⁵⁹	138 d	140,134,144.4	•95	1.35	A, C	(91),(135)	
Er ¹⁶⁰	29.5 hr	30, 29.4, 29	K-X-rays per	disin-	A, I	(109)	
Ho ¹⁶⁰	5 hr	5.6, 5, 5.3	tegration =	.82	A, I	(109)	

A represents absolute cross section

C represents cumulative cross section

I represents independent cross section

Table V(c)

CASE WHERE BETA RADIATION WAS MEASURED

Nuclide	Present work	t <u>1</u> Liter val	rature lues]	Decay	Scheme	data	Reference
Er ¹⁶⁹	9d.	9.4,	9.95	0.34	Mev	100%	abundance	(85)

Table V(d)

AVERAGE VALUES OF VARIOUS CONVERSION FACTORS FOR PEAK HEIGHTS TO AREAS FOR THE PEAK HEIGHTS OF NUCLIDES MEASURED

200 kev peak of Tm ¹⁶⁸ & Tm ¹⁶⁷	K-X-ray peak of Er ¹⁶⁵ & Er ¹⁶⁰	820 kev peak of Er ¹⁶¹	K-X-ray peak of Ho ^{160m} , Ho ¹⁶¹ & Dy ¹⁵⁹	511 kev peak of Zn ⁶³ & Cu ⁶⁴
10.83	5.9	6.77	5.98	4.77

Table VI

Bomb. No.	Proton Energy (Mev)	Bomb. Dura- tion mins.	Zn ⁶³ 0 (mb)	Cu ⁶⁴ σ (mb)	Dm	W _{Cu}
1	6.5#(7)	120	200	-	8.4478×10^7	1.4496
2	10.1#(10.6)	120	384	-	81.209 x 10 ⁷	1.4826
3	12.8 [#] (13.3)	60	408	-	10.380×10^8	1.3945
4	16.4	60	-	150	9.9166 x 10^6	1.3570
5	22.2	60	-	476	16.067×10^{7}	1.5700
6	28.5	60	-	500	12.050×10^{7}	1.1688
7	34.0	60	-	3 65	9.1634 x 10^7	1.3248
8	39.8	30	-	323	12.804×10^{6}	0.8617
9	47.4	60	-	280	3.6548×10^7	1.0267
10	57.0	60	-	255	17.7748×10^6	1.8570
11	67.0	60	-	232	43.788×10^{6}	1.6096
12	77.0	60	-	208	89.911 x 10 ⁶	1.4239
13	86.5	60	-	184	25.689×10^{5}	2.0429
14	57-2	60	-	255	17.515×10^{6}	1.5353
15	67-2	61	-	232	27.44×10^6	2.3988
16	22.2-2	60		476	15.764×10^{7}	0.6438
17	16.4-3	56	-	150	3.7947 x 10 ⁷	0.7181
18	28.5-2	60	-	500	4.8459 x 10 ⁷	0.5076
19	34.0-2	60	-	365	3.5030×10^7	0.8419
20	39.8-2	60	-	323	8.8744×10^6	0.4953
21	16.4-2	60	-	150	8.057 x 10 ⁶	0.7132
22	16.4 E	15	-	150	24.6317 x 10 ⁵	2.0058
23	22.2 E	15	-	476	62.949 x 10 ⁵	2.6075
24	28.5 E	20	-	500	89.407 x 10 ⁵	1.1143
25	34.0 E	20	-	365	53.374 x 10 ⁵	1.1415

E indicates enriched isotope irradiations, and -2, -3 indicate repetitions.

indicates these energies were the net after attenuation correction (125).

The energy degradation due to attenuation was about 0.5 Mev. The energy values in the parentheses were the actual bombarding energies.

In the case of all other energies, it was assumed the attenuation correction was negligible.

calculated to be 2.6325.

With regard to the spallation products of erbium, except in respect to Tm^{170} , Tm^{168} , Tm^{167} , and Er^{169} , in all the other cases (Tm^{166} , Tm^{165} , Tm^{163} , Er^{165} , Er^{163} , Er^{160} , Ho¹⁶¹, Ho^{160m}, Dy¹⁵⁹, and Dy¹⁵⁷) the total weight of erbium recovered has been assumed to be $W_{\rm Er}$. In the case of Tm^{170} calculations, 14.9% erbium recovered has been the target weight because it was produced only through the reaction $Er^{170}(p,n)$, and in the case of Er^{169} also the weight of erbium was assumed to be 14.9% of the erbium recovered since it was assumed to be produced through the

In the case of Tm^{168} , for the irradiation with protons of energy up to 16.4 Mev, the target weight was assumed to be 29.1% of erbium, since it was assumed to be produced by $\text{Er}^{168}(\text{p,n})$ and for the proton irradiation at energies 22.2, 29.8 and 34 Mev, the target weight has been assumed to be 14.9% of erbium recovered, since it has been assumed that Tm^{168} was produced only by the $\text{Er}^{170}(\text{p,3n})$ reaction. These assumptions have been checked qualitatively through enriched isotope irradiations.

For Tm^{167} calculations, the weight of target has been assumed to be 66.9% of erbium because Er^{170} , Er^{168} , and Er^{167} only contribute in the production of Tm^{167} whereas Er^{166} , Er^{164} , and Er^{162} cannot produce Tm^{167} .

In the case of alpha irradiations, since holmium is mono-isotopic, the weight of holmium is the weight of the target.

For alpha irradiations, only the saturation factor of the product, the disintegration rate of the product, total beam current, weight of holmium and duration of irradiation were needed. The calculation is straightforward, according to the formula mentioned previously.

In the case of enriched isotope sample irradiations of erbium, individual $Er^{168}(p,2n)$ cross sections have been calculated by solving two simultaneous equations with results obtained from the natural erbium irradiations and partially enriched isotope sample irradiations.

In the case of a particular product (e.g. Ho^{160m} or Ho^{161} or Er^{165}) directly produced and also produced through the decay of the parents during the irradiation and until the end of separation, calculations of the type are outlined in Appendix II.

The formation cross sections have been reported in the last columns of Tables VII, VIII, IX and X.

In Table XI, the formation and cross sections of Tm^{168} and Tm^{167} , through the reactions $Ho^{165}(\alpha,n)Tm^{168}$ and $Ho^{165}(\alpha,2n)Tm^{167}$ are presented. The irradiation data used for the calculation are:

Length of irradiation - $3\frac{1}{2}$ hours. Total α -beam current - 5.014 Ahours.

The cross-section values for Tables VII, VIII, IX and X are plotted in Figs. 55 to 68 inclusive. Cross-section values for the α -irradiationsfrom Table XI are plotted in Fig. 70.

<u>Table</u>	VII
--------------	-----

Nuclide	Bombard- ment No.	Proton Energy in Mev	$\sigma_{m} \cdot \frac{M_{Er}}{M_{Cu}}$	D ^o p	$\frac{\frac{D_{p}^{o}}{p}}{D_{m}^{o}} \cdot \frac{W_{Cu}}{W_{Er}}$	Ratio of Saturation Factors	σ (mb)
Tm ¹⁷⁰	2	10.1	1010.880	2.169×10^4	0.4652×10^{-4}	1999	95.52
,,	3	12.8	1074.060	3.228 x 10 ⁴	0.5416×10^{-4}	2996	174.28
,,	17	16.4	394.875	1.4124 x 10 ⁴	0.25964×10^{-3}	237.9	24.39
Tm^{168}	2	10.1	1010.88	51.492×10^3	0.60706×10^{-4}	1153.0	70.76
,,	3	12.8	1074.06	48.112 x 10^3	0.43088 x 10 ⁻⁴	2540.5	117.60
,,	17	16.4	394.875	39.203 x 10^3	3.963×10^{-4}	155.06	24.30
,,	5	22.2	1253.07	25.348 x 10 ⁴	26.772×10^{-4}	155.06	519.99
,,	6	28.5	1316.25	27.447×10^4	30.589 x 10 ⁻⁴	155.06	624.57
,,	7	34.0	960.862	12.4759 x 10 ⁴	17.680×10^{-4}	155.06	263.42
$_{Tm}$ 167	2	10.1	1010.88	76.7482×10^4	3.778×10^{-4}	148.23	56.65
,,	3	12.8	1074.06	201.786 x 10 ⁴	7.547×10^{-4}	221.733	179.72
,,	4	16.4	394.875	581.865 x 10 ⁴	16.8282×10^{-2}	17.573	1167.72
,,	5	22.2	1253.07	126.831 x 10 ⁵	30.762 x 10 ⁻³	17.573	677.20
,,	6	28.5	1316.25	57.073 x 10 ⁵	14.602×10^{-3}	17.573	377.75
,,	7	34.0	960.862	79.1016 x 10 ⁵	25.7329×10^{-3}	17.573	434.50
,,	8	39.8	850.298	168.39 x 10 ⁴	33.727×10^{-3}	17.78	509.73
,,	9	47.4	737.1	252.053 x 10 ⁴	12.007×10^{-3}	17.573	155.53

t

Nuclide	Bombard- ment No.	Proton Energy in Mev	o _m . $\frac{M_{Er}}{M_{Cu}}$	D_p^{o}	$\frac{\frac{D_{p}^{o}}{D_{m}^{o}} \cdot \frac{W_{Cu}}{W_{Er}}}{W_{Er}}$	Ratio of Saturation Factors	σ _p (mb)
	10	57.0	671.287	99.882 x 10 ⁴	0.9742×10^{-2}	17.573	114.96
,,	11	67.0	610.74	219.204 x 10 ⁴	0.8796×10^{-2}	17.573	94.40
,,	12	77.0	547.56	33.249 x 10 ⁵	0.6142×10^{-2}	17.573	59.10
,,	13	86.5	484.38	83.118 x 10 ³	0.5681 x 10 ⁻²	17.573	48.35
,,	* 23	22.2 E	1253.07	22.652 x 10 ⁴	0.7278 x 10 ⁻¹	18.203	1660.17
,,	* 24	28.5 E	1316.25	90.599 x 10 ³	0.1744×10^{-1}	17.890	406.12

Table VII (Contd.)

- *In these two enriched isotope irradiations, the formation cross sections have been calculated on the basis that contributions from ${\rm Er}^{167}(p,n)$ reactions and from ${\rm Er}^{170}(p,4n)$ reactions on 1.5% abundant ${\rm Er}^{170}$ are negligible. Weight of ${\rm Er}^{168}$ is weight of the target element.
- In the case of irradiations 22 and 25 performed at 16.4 Mev and 34 Mev, cross sections for the (p,2n) reactions themselves were calculated to be 548 [±] 109.6 and 120 [±] 20 mb respectively. This was done by solving two simultaneous equations obtained from the natural erbium irradiations (Nos. 4 and 7) and enriched erbium irradiations.

Τa	ble	VI	II

Nuclide	Bombard- ment No.	Proton Energy in Mev	σ _m • ^M Er MCu	C.R.	$rac{{}^{\mathrm{D}}{}^{\mathrm{o}}_{\mathrm{p}}}{{}^{\mathrm{O}}_{\mathrm{m}}}\cdot rac{{}^{\mathrm{W}}_{\mathrm{Cu}}}{{}^{\mathrm{W}}_{\mathrm{Er}}}$	Ratio of Saturation Factors	o (rel)
Tm ¹⁶⁶	1	6.5	52.65	3.7857×10^4	0.112865×10^{-3}	5.3900	0.32
, ,	2	10.1	1010.88	53.5714 x 10 ⁵	0.17112×10^{-2}	5.3900	9.32
,,	3	12.8	1074.06	12.50×10^6	0.3032×10^{-2}	7.7286	25.17
,,	4	16.4	394.87	57.2917 x 10 ⁶	0.3945	0.6125	95.41
,,	5	22.2	1253.07	8.20×10^7	0.1290	0.6125	99.00
,,	6	28.5	1316.25	7.30×10^{7}	0.12116	0.6125	97.68
,,	7	34.0	960 .86 2	4.35×10^7	0.09185	0.6125	54.05
,,	8	39.8	850.298	7.70×10^{6}	0.07000	0.6060	36.07
,,	9	47.4	737.10	33•5743 x 10 ⁶	0.01038	0.6125	46.85
,,	10	57.0	671.2875	81.2520 x 10 ⁵	0.05142	0.6125	21.14
,,	11	67.0	610.74	22.857×10^6	0.0595	0.6125	22.20
,,	12	77.0	547.56	31.4285 x 10 ⁶	0.03768	0.6125	12.64
,,	13	86.5	484.38	10.000 x 10 ⁵	0.04438	0.6125	13.16

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Nuclide	Bombard- ment No.	Proton Energy in Mev	σ _m • $\frac{M_{Er}}{M_{Cu}}$	C.R.	$\frac{D_p^o}{D_m^o} \cdot \frac{W_{Cu}}{W_{Er}}$	Ratio of Saturation Factors	op (rel)
$_{\rm Tm}$ 165	17	16.4	394.875	6.6497 x 10 ⁵	0.1821 x 10 ⁻²	2.1590	1.553
,,	16	22.2	1253.07	15.533 x 10 ⁵	0.04434×10^{-2}	2.1560	1.198
,,	18	28.5	1316.25	14.619 x 10 ⁵	0.29625 x 10 ⁻²	2.1560	8.407
,,	19	34.0	960.862	14.010 x 10 ⁵	0.3850×10^{-2}	2.1560	7.976
, ,	20	39.8	850.290	7.716 x 10 ⁵	0.52922×10^{-2}	2.1560	9.700
,,	9	47.4	737.1	12.1827 x 10 ⁵	0.3763×10^{-2}	2.1560	5.980
,,	14	57.0	671.287	4.1421 x 10 ⁵	0.2293×10^{-2}	2.1560	3.320
Tm^{163}	19	34.0	960.862	3.687×10^6	0.010125	0.16498	1.605
,,	20	39.8	850.29	2.375×10^7	0.16297	0.16498	22.860
,,	9	47.4	737.1	7.8125×10^7	0.24148	0.16498	29.360
,,	10	57.0	671.287	5.875 x 10^7	0.3720	0.16498	41.200
,,	11	67.0	610.74	13.00×10^7	0.3385	0.16498	34.120
, ,	12	77.0	547.56	16.25×10^7	0.19486	0.16498	17.590
,,	13	86.5	484.38	10.00 x 10 ⁶	0.05555	0.16498	0.443

Table VIII (Contd.)

Nuclide	Bombard- ment No.	Proton Energy in Mev	$\sigma_{m} \cdot \frac{M_{Er}}{M_{Cu}}$	Dp	$rac{\mathrm{D}_{\mathrm{p}}^{\mathrm{o}}}{\mathrm{D}_{\mathrm{m}}^{\mathrm{o}}}\cdot rac{\mathrm{W}_{\mathrm{Cu}}}{\mathrm{W}_{\mathrm{Er}}}$	Ratio of Saturation Factors	σ (mb)
Er ¹⁶⁹	17	16.4	394.875	2.475×10^3	0.0553×10^{-2}	16.440	3.59
,,	16	22.2	1253.070	3.675×10^3	0.02071×10^{-2}	16.423	4.26
,,	18	28.5	1316.250	5.587×10^3	0.0275×10^{-2}	16.423	5•94
,,	19	34.0	960.862	2.469 x 10 ³	0.1858 x 10 ⁻²	16.423	29.32
,,	20	39.8	850.290	5.2125 x 10 ³	0.3277×10^{-2}	16.423	45.77
,,	9/	47.4	737.100	30.75 x 10 ³	0.5706×10^{-2}	16.423	69.07
,,	14	57.0	671.287	22.500 x 10 ³	1.0812×10^{-2}	16.423	119.20
,,	15	67.0	610.740	31.50 x 10 ³	1.1286×10^{-2}	16.423	113.20
,,	12	77.0	547.560	90.750 x 10 ³	0.9888×10^{-2}	16.423	88.92
,,	13	86.5	484.380	28.50×10^3	0.7785×10^{-2}	16.423	61.93

<u>Table IX</u>

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Nuclide	Bombard- ment No.	Proton Energy in Me v	$\sigma_{m} \cdot \frac{M_{Er}}{M_{Cu}}$	D ^o p	$\frac{D_p^o}{D_m^o} \cdot \frac{W_{Cu}}{W_{Er}}$	Ratio of Saturation Factors	op (mb)
Er ^{165xxx}	16	22.2	1025.64	5.0984 x 10 ⁶	0.1457×10^{-2}	0.8181	1.49
,,	19	34.0	960.86	24.074 x 10 ⁶	6.403×10^{-2}	0.8181	50.332
,,	20	39.8	850.30	8.617 x 10 ⁶	0.5913×10^{-2}	0.8181	41.133
,,	9	47.4	737.10	65.287 x 10 ⁶	0.2018 x 10 ⁻¹	0.8181	121.689
,,	14	5 7	671.29	62.072 x 10 ⁶	0.3437	0.8181	188.75
,,	15	67	610.74	16.531×10^6	0.7717	0.8171	385.05
,,	12	77	547.56	35.6555 x 10 ⁶	0.5064	0.8181	226.854
, ,	13	86.5	484.38	16.1396 x 10 ⁵	0.5749	0.8181	227.80

Table IX (Contd.)

xxxIn the calculations of independent formation cross section of Er¹⁶⁵, it has been assumed that the abundance of 1.16 Mev photopeak of Tm¹⁶⁵ is to be 1%. As mentioned earlier, while outlining the nuclear properties of Tm¹⁶⁵ (under spallation products), it could be observed only until around 60 Mev. Therefore from 60 Mev onwards it has been assumed that Er¹⁶⁵ has been produced only independently, and the correction for decay of Tm¹⁶⁵ was not necessary. The assumption of 1% abundance of 1.16 Mev photopeak is justified because the relative formation cross sections of Tm¹⁶⁵ appear to be 100 times lower than that expected.

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Nuclide	Bombard- ment No.	Proton Energy in Mev	o _m . $\frac{M_{Er}}{M_{Cu}}$	D ^o p	$\frac{D_{p}^{o}}{D_{m}^{o}} \cdot \frac{W_{Cu}}{W_{Er}}$	Ratio of Saturation Factors	σ (mb)
Er ¹⁶¹	19	34.0	960.86	15.115×10^4	0.03357	0.2632	8.49
,,	9	47.0	737.1	31.11 x 10 ⁵	0.086056	0.2632	16.69
,,	14	57.0	671.288	97.778 x 10 ⁵	0.69998	0.2632	123.67
,,	15	67.0	610.74	47.0509 x 10 ⁶	2.5125	0.2632	404.028
,,	12	77.0	547.56	127.567 x 10 ⁶	2.071	0.2632	298.47
,,	13	86.5	484.38	50.444 x 10 ⁵	2.0539	0.2632	261.85
Er ¹⁶⁰	14	57.0	671.2875	387.459 x 10 ³	0.02774	2.266	42.10
,,	15	67.0	610.74	108.56 x 10 ⁴	0.057916	2.266	80.12
,,	12	77.0	547.56	95.409 x 10 ⁵	0.15493	2.266	192.23
, ,	13	86.5	484.38	547.264 x 10 ³	0.22283	2.266	244.75
Ho ¹⁶¹	14	57.0	671.288	11.162 x 10^6	0.3090	0.2177	45.15
,,	15	67.0	610.74	25.9239 x 10 ⁶	0.6890	0.2210	93.0
,,	13	86.5	484.38	3.419 x 10 ⁶	0.7583	0.2177	79.96

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Nuclide	Bombard- ment No.	Proton Energy in Mev	° _m ∙ $\frac{M_{Er}}{M_{Cu}}$	Dp	$\frac{D_p^o}{D_m^o} \cdot \frac{W_{Cu}}{W_{Er}}$	Ratio of Saturation Factors	σ (mb)
Ho ^{160m}	14	57.0	671.288	5.0577 x 10 ⁵	0.1401 x 10 ⁻¹	0.4073	3.83
,,	15	67.0	610.74	25.043 x 10 ⁵	0.6658 x 10 ⁻¹	0.4072	16.56
,,	12	77.0	547.56	130.492 x 10 ⁵	0.7824 x 10 ⁻¹	0.4073	17.45
,,	13	86.5	484.38	5.602 x 10 ⁵	0.1243	0.4073	24.52
Dy ¹⁵⁹	15	67.0	610.74	6.96×10^2	0.3373×10^{-4}	254.86	5.25
,,	12	77.0	547.56	115.193×10^2	2.378×10^{-4}	251.05	32.68
,,	13	86.5	484.38	10.658 x 10 ²	2.692×10^{-4}	251.05	32.73
Dy ¹⁵⁷	15	67.0	610.74	8.8154 x 10 ⁴	0.04269×10^{-1}	0.64999	1.69
,,	12	77.0	547.56	72.3077×10^4	0.14925 x 10 ⁻¹	0.6505	5.316
,,	13	86.5	484.38	8.9231 x 10 ⁴	0.2254 x 10 ⁻¹	0.6505	7.10

Table X (Contd.)
Nuclide	Target No.	Alpha Energy in Mev	D ^o p	*n/A	Saturation Factor	o p (mb)
_{Tm} 168	2	15	0.2976×10^4	1.3562×10^{18}	12.00×10^{-4}	6.81
,,	3	20	3.045 x 10 ⁴	2.40996 x 10 ¹⁸	, ,	39.16
,,	4	25	0.2076 x 10 ⁴	1.1515 x 10 ¹⁸	,,	5.60
$_{Tm}$ 167	3	20	1.6989 x 10 ⁵	2.40996 x 10 ¹⁸	10.45×10^{-3}	25.1
,,	4	25	15.6299 x 10 ⁵	1.1515 x 10 ¹⁸	,,	484.17
,,	5	30	10.6465 x 10 ⁵	0.85296 x 10 ¹⁸	,,	445.25
,,	6	35	3.092 x 10 ⁵	0.98107×10^{18}	,,	112.17
,,	7	39	1.9254 x 10 ⁵	1.1515 x 10^{18}	9 9	59.64

<u>Table XI</u>

*n/A varied from target to target because targets were chosen from three holmium painted aluminium foils.

(e) EVALUATION OF ERRORS

The probable error in the formation cross-section calculations may be estimated at best from a knowledge of the errors in the individual terms used to calculate the cross section. The formula as described earlier is

$$\sigma_{p} = \sigma_{m} \frac{D_{p}^{\circ} W_{Cu} M_{Er} (1 - e^{-\lambda_{m}t})}{D_{m}^{\circ} W_{Er} M_{Cu} (1 - e^{-\lambda_{p}t})}$$

in which the disintegration rate of the product or monitor involves the terms counting rate of the particular photopeak of interest at the end of the irradiation (C_{peak}°) and efficiency factor E_{f} , obtained from Figs. 71(a) and 71(b) in Appendix I, branching ratio (B.R.), total conversion coefficient factor α_{T} , and the dilution factor (Dil.).

In other words,

$$D_{p}^{\circ} = C_{peak}^{\circ} \cdot \frac{1}{E_{f}} \cdot \frac{1 + \alpha_{T}}{B \cdot R \cdot} \cdot \frac{1}{Dil}.$$

or
$$D_{m}^{\circ} = C_{peak}^{\circ} \cdot \frac{1}{E_{f}} \cdot \frac{1}{2 \times B \cdot R \cdot} \cdot \frac{1}{Dil}.$$

C^opeak: It is estimated that the error involved in calculating counts under the photopeak was about ± 10%.

*The factor '2' has been used because for every positron decay two annhilation photons are emitted. For example, in the case of Zn^{63} , 93% positron decay, there are 2 x 93 annhilation photons emitted for every 100 decays of Zn^{63} . In the case of Cu^{64} , 19% positron branch, there are 2 x 19 annhilation photons emitted for every 100 decays of Cu^{64} .

- E_f : The error involved in the factor obtained from the efficiency curves was estimated to be $\frac{+}{-}$ 5%. The error involved in the case of photopeaks of energy 400 kev may be a bit less. However, $\frac{+}{-}$ 5% has been chosen.
- $\alpha_{\rm T} = \alpha_{\rm K} + \alpha_{\rm L} + \alpha_{\rm M}$, B.R. = Branching ratio. These two factors were obtained from the Nuclear Data Sheets and recent literature, and in some cases $\alpha_{\rm T}$ was taken from Rose's tables ⁽⁹⁵⁾. No error was quoted for these since improved values can be inserted as they appear.
- Dil.: The error involved in pipetting and diluting was taken to be $\frac{+}{-}$ 1%.
- λ_{m} t: The nuclides, whose formation cross-sections reported in the present work have half-lives greater than $l\frac{1}{2}$ hr, and the duration of irradiation were known within a few seconds. The decay constant that depends on half-life was determined reasonably well. The error involved in this factor was estimated to be $\frac{+}{2}$ 1%.
- WEr & W_{Cu}: The errors involved in determining the weight of erbium recovered and copper recovered were estimated to be + 5% and + 2% respectively. O_m: The monitor cross sections used were those of Meadows⁽³⁾ for the bombarding energies obtained from the cyclotron calibration curve

which was shown in Fig. 10, corrected for radial oscillations by Professor Bell⁽⁷⁰⁾. The uncertainty involved in the irradiation energy was $\stackrel{+}{-}$ 2 Mev. No errors were assigned to the value of σ_m , since again improved values can be inserted as they appear.

The total error is estimated to be -14%. This error was assigned to Tm¹⁶⁸, Tm¹⁶⁷, Tm¹⁶⁶, Tm¹⁶⁵, Er¹⁶¹, Er¹⁶⁰ and Tm¹⁶³.

In the case of Tm^{170} it was estimated to be $\div 20\%$ because of the error involved in estimating Tm^{170} activity from $\text{Tm}^{170} + \text{Tm}^{168}$ activity which involved measuring two samples of low counting rates.

For Er^{169} a 2π β -proportional counter has been used, and the counting error is $\stackrel{+}{=} 2\%$. The error in chemical yields of rare earth is $\stackrel{+}{=} 5\%$, whereas for copper yields the error is $\stackrel{+}{=} 2\%$. Cross-contamination error is $\stackrel{+}{=} 3\%$. Error involved in dilution is $\stackrel{+}{=} 1\%$. In all the total error is estimated to be $\stackrel{+}{=} 7\%$.

In the case of holmium activities, Ho^{160m} and Ho^{161} , $\frac{+}{2}$ 17% error is due to counting, efficiency factor, resolving decay curves of close half-lives and cross contamination. Chemical yield errors are $\frac{+}{2}$ 2% and $\frac{+}{2}$ 6% for the monitor and holmium respectively. Error due to dilution is $\frac{+}{2}$ 1%. Error involved in noting the time of separation is $\frac{+}{2}$ 5%. The total error is determined to be $\frac{+}{2}$ 19%.

The total error for dysprosium activities, Dy^{159}

and Dy^{157} , is estimated to be \div 18%.

For alpha irradiations, the errors involved in target weight and area are $\stackrel{+}{=}$ 2%, and $\stackrel{+}{=}$ 10% for the counting and $\stackrel{+}{=}$ 5% for the efficiency factor. The total error is estimated to be $\stackrel{+}{=}$ 11%.

D. DISCUSSION

(a) EXCITATION FUNCTIONS

The study of formation cross-sections of different spallation product nuclides was one of the main aspects of the present work. From the study of the excitation functions some rather qualitative conclusions are given below.

In the case of (p,xn) reactions (production of thulium radioisotopes) their excitation function shapes followed a general trend, that is, the peaked shape expected from the compound nucleus formation followed by neutron evaporation, successive (p,xn) cross sections were shifted up the energy scale by about 10 Mev per neutron emitted. In the cases where the product nuclide was produced through adjacent target isotopes from the (p,n) and (p,2n) reactions, the peaked shapes of the excitation functions This was predicted in Jackson's were superimposed. schematic model calculations⁽²⁰⁾. In the present work Jackson's type of calculations proved reasonably well the superimposition of the peaks of (p,n) and (p,2n) reactions as was experimentally observed as well, whereas (p,3n) and (p,4n), the peaked shapes of the excitation functions, could be identified at 30 and 40 Mev proton bombarding In the case of (p,5n) excitation function onwards energy. the peak heights decrease and the peaks broaden, with the result that the peaks overlap and the excitation functions

slowly level off. This was what was observed experimentally by Grant⁽¹²⁶⁾, Bell and Skarsgard⁽¹⁸⁾as well as in the present work, and also theoretically predicted by Jackson⁽²⁰⁾. Since Er^{170} and Er^{168} are stable, and Er^{169} is unstable, no (p,xn) reactions are attributable to the latter. Thus a gap is provided, which in some cases helped to observe well-separated peaks in the excitation functions. This also helped in plotting pure excitation functions of Tm^{168} and Er^{169} and, to some extent, assisted in resolving the mixed excitation function of Tm^{167} .

In the case of (p,pxn) reactions, the excitation functions started around the threshold and from the threshold rose rapidly and levelled off. The levelling off or slowly decreasing nature is very well explained by the knock-on cascade mechanism rather than by Bohr's model, as explained earlier. It was experimentally observed that the cross-section values of (p,pxn) and (p,xn) reactions were almost comparable at high energies.

In (p,pxn), (p,2pxn) and (p,3pxn) reaction excitation functions, it was not possible to observe any sort of indication that the nuclide was produced through different targets of erbium; unlike (p,xn) reaction excitation functions where the contribution from different target isotopes could be seen from the peaked shapes.

For (p,2pxn) excitation functions, the cross sections were found to be an order of magnitude lower than the (p,pn) excitation functions. The (p,2pxn) excitation In the case of (p,3pxn) and (p,2pxn) products, the cumulative cross sections only were plotted. Their excitation functions increase monotonically with bombarding energy of protons just like (p,2pxn) reaction products.

The excitation functions are discussed in detail below.

(i) (p,xn) Reaction Excitation Functions

available in the McGill synchrocyclotron.

In the present work Tm^{170} , Tm^{168} , Tm^{167} , Tm^{166} , Tm^{165} and Tm^{163} were produced through this type of reaction. The excitation functions are shown in Figs. 55, 56, 57, 58, 59, and 60.

(1) Tm^{170} (t₁ = 128 d) Fig. 55:

The radiation measurement of this nuclide was difficult, with the result that large errors are associated with experimentally calculated formation cross-sections. This may be attributed to:

- (a) low counting rate produced due to short duration of bombardment (one to two hours' duration of irradiation whereas the half-life is 128 d).
- (b) measurement of the highly converted 84 kev gamma transition.
- (c) the interference of $86-d \text{ Tm}^{168}$ photopeaks with the 84 kev photopeak of Tm^{170} .

However, the excitation function showed a maximum around 12 Mev proton energy. Experimental cross



Er¹⁷⁰(p,n)Tm¹⁷⁰ REACTION



ENERGY (Mev.)

sections agreed reasonably well with theoretically predicted values within experimental errors. This comparison is shown later.

- (2) Tm^{168} (t₁ = 86 d) Fig. 56:
- This nuclide was first produced by a (p,n) reaction on Er^{168} and secondly through a (p,3n) reaction on Er^{170} . The peaked shapes around 12 and 30 Mev represent the maxima of the (p,n) and (p,3n) reactions respectively. The formation crosssections have been compared with theoretically predicted values. This aspect is decsribed later. (3) Tm^{167} (t₁ = 9.6 d) Fig. 57:
 - This nuclide was produced by (p,n), (p,2n) and (p,4n) reactions on erbium. In the excitation function a superimposed peak of (p,n) and (p,2n) around 20 Mev and a peak due to (p,4n) at about 40 Mev were observed.

(4)
$$\operatorname{Tm}^{166}$$
 (t₁ = 7.7 hr) Fig. 58:

This nuclide was produced by (p,n), (p,2n), (p,3n)and (p,5n) reactions on erbium. The composite excitation function has a peak at about 20 Mev. This was due to superimposition of (p,n) and (p,2n)excitation functions and a hump around 50 Mev is due to the (p,5n) reaction on Er^{170} . The clear separation of this hump is due to the gap due to radioactive Er^{169} . The peak due to (p,3n) reaction

EXCITATION FUNCTIONS FOR

Er¹⁶⁸(p,n)Tm¹⁶⁸

and

Er¹⁷⁰(p,3n)Tm¹⁶⁸ REACTIONS



ENERGY (Mev.)





ENERGY (Mev.)

- 169 -Figure 58 RELATIVE EXCITATION FUNCTION FOR $Er^{166}(p,n)Tm^{166} + Er^{167}(p,2n)Tm^{166} +$ Er¹⁶⁸(p,3n)Tm¹⁶⁶ + Er¹⁷⁰(p,5n)Tm¹⁶⁶ REACTIONS



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on Er^{168} is not seen. However, it has broadened the (p,n) and (p,2n) peak at 30 Mev, indicating the presence of the (p,3n) reaction.

- (5) Tm^{165} (t₁ = 28 hr) Fig. 59:
 - This excitation function is similar to that of Tm^{166} in that both are relative and in both cases the whole weight of erbium was assumed to be the target weight. The data are very sparse and the positions where the peaks for the (p,2n), (p,3n) and (p,4n) reactions probably occur are sketched in.

(6)
$$\text{Tm}^{163}$$
 (t₁ = 1.8 hr) Fig. 60:

- This excitation function is also relative like that of Tm^{166} and Tm^{165} , since the decay scheme of the nuclide is not fully established. The broad peak around 60 Mev is due to the sum effect of (p,4n) and broad (p,5n), (p,6n) and (p,8n) excitation functions.
- (ii) Excitation Functions for (p,pxn) Reactions

(1)
$$\operatorname{Er}^{169}(t_{\frac{1}{2}} = 9 \text{ d})$$
 Fig. 61:

This nuclide was produced by (p,pn) reaction on Er^{170} . The typical (p,pn) reaction excitation function increased rapidly and reached maximum around 50 Mev and slowly decreased. The peaking around 50 Mev was in agreement with $Grant^{(126)}$ who studied the reaction $Ir^{193}(p,pn)Ir^{192}$. However, Kavanagh⁽¹²⁷⁾ reported a maximum between 40 and 45 Mev for the (p,pn) reaction on gold, whereas Gusakow et al.⁽¹²⁸⁾

 $\frac{\text{RELATIVE EXCITATION FUNCTION FOR}}{\text{Er}^{166}(p,2n)\text{Tm}^{165} + \text{Er}^{167}(p,3n)\text{Tm}^{165} +}$ $\text{Er}^{168}(p,4n)\text{Tm}^{165} + \text{Er}^{170}(p,6n)\text{Tm}^{165} \text{ REACTIONS}$







EXCITATION FUNCTION FOR Er¹⁷⁰(p,pn)Er¹⁶⁹ REACTION



ENERGY (Mev.)

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80 Mev which is rather high. In the present work the (p,pxn) excitation functions rapidly increased and, after attaining a maximum, decreased slowly. (2) $Er^{165}(t_{\frac{1}{2}} = 10.4 \text{ hr})$, $Er^{161}(t_{\frac{1}{2}} = 3.1 \text{ hr})$ and $Er^{160}(t_{\frac{1}{2}} = 29.5 hr)$ Figs. 62, 63 & 64 respectively. The general tendency of the excitation function is similar to that of Er¹⁷⁰(p,pn)Er¹⁶⁹ reaction excitation function. The excitation functions start from the threshold, rise to a maximum and slowly decrease or level off. In the case of Er^{161} and Er^{160} the cumulative cross sections only could be plotted instead of independent cross sections because of the very short-lived Tm parents. Therefore the cross-section values are high. As mentioned earlier, in the case of Er^{165} , Er^{161} and Er^{160} they were produced from different stable isotopes of erbium.

The high (p,pxn) cross sections cannot be explained by the compound nucleus mechanism which favours neutron evaporation rather than charged particle emission, which is contrary to experimental observations. An attempt has been made to explain the high reaction cross section on the basis of non-compound nucleus reactions, and also on the basis of neutron, proton binding energies.

Reactions referred to as non-compound nucleus reactions such as 'knock-on', 'pick-up', 'stripping' or 'hot spot'

Figure 62 EXCITATION FUNCTION FOR $Er^{166}(p,pn)Er^{165} + Er^{167}(p,p2n)Er^{165} +$ Er¹⁶⁸(p,p3n)Er¹⁶⁵ + Er¹⁷⁰(p,p5n)Er¹⁶⁵ REACTIONS

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CUMULATIVE YIELDS FOR THE FORMATION OF

Er¹⁶¹ BY Er(p,pxn) REACTION

AND THROUGH THE DECAY OF Tm¹⁶¹



CUMULATIVE YIELDS FOR THE FORMATION OF

Er ¹⁶⁰ B	Y Er(p,pxn)	REACTION
	فحدد محجز تصنع مقتل عظتا محت البروا الهبوالث عرقمة تنجها	

AND THROUGH THE DECAY OF Tm¹⁶⁰



reactions have been suggested to explain the high reaction cross sections. A 'knock-on' reaction is one in which the projectile collides with individual nucleons, transferring sizable amounts of energy directly to one or to a small group of particles and allowing them to escape. The bombarding particle may or may not be captured by the nucleus. 'Pick-up' reactions and 'stripping' reactions are another closely allied pair of direct interaction processes. Assumption of a 'pick-up' reaction, such as the (p,d) process, might not explain the high cross sections at high energies because the (p,d) process is negligible for complex nuclei at high energies ⁽⁶⁵⁾. In the 'pick-up' reaction the projectile combines with a nucleon in the nucleus and the combination escapes. For example, a proton projectile may combine with the neutron which carries most of the energy of the incident proton. The stripping process is the reverse case where the nucleus removes or 'strips'a nucleon from the projectile.

Another type of reaction which can be included in this direct interaction is local excitation or 'hot spot' reactions. In this case a portion of the nucleus is excited by the incoming particle, and the emission phase of the reaction or 'evaporation' takes place before the excitation energy is distributed throughout the nucleus.

It may be possible to explain high reaction cross sections on the basis of neutron and proton binding energies reported in tables of Cameron⁽¹¹⁸⁾. The neutrons in the residual excitation energy) will be bound tighter than the protons, so that proton knock-out will be favoured compared with neutron knock-out. For example, take Tm^{162} as the resulting excited nucleus. The neutron binding energy is 7.791 Mev and the proton binding energy 3.717 Mev. Thus proton boil-off may occur giving Er^{161} . This is a possible explanation for the high formation cross section for any result of a (p,pxn) reaction.

(iii) Excitation Functions for (p, 2pxn) Reactions

Ho¹⁶¹ ($t_{\frac{1}{2}} = 2.5$ hr) and Ho¹⁶⁰ ($t_{\frac{1}{2}} = 5$ hr)

The two independent excitation functions given in Figs. 65 and 66 start from the threshold and increase monotonically with energy but did not level off because it was not energetically possible. Ho¹⁶⁰ ($t_{\frac{1}{2}} = 28$ min) could not be followed for decay because of its very short half-life.

(iv) Excitation Functions for (p,2pxn) + (p,3pxn) Reactions

 Dy^{159} and Dy^{157} have been produced by these types of reactions. The excitation functions in Figs. 67 and 68 belong to these two nuclides and have trends similar to the (p,2pxn) reaction excitation functions. They rise monotonically from the threshold with the increase in energy. Only cumulative cross sections were plotted. It was not possible to determine independent cross section because of the short-lived parents which decayed to dysprosium by the start of measurement.

EXCITATION FUNCTION FOR THE

Er(p,2pxn)Ho¹⁶¹ REACTIONS



EXCITATION FUNCTION FOR THE Er(p,2pxn)Ho^{160m} REACTIONS


Figure 67

CUMULATIVE YIELD FOR THE FORMATION OF $\frac{Dy^{159}}{AND}$ BY THE REACTION OF Er(p, 3pxn) AND THROUGH THE DECAY OF Ho¹⁵⁹



ENERGY (Mev.)

- 183 -Figure 68 CUMULATIVE YIELD FOR THE FORMATION OF Dy¹⁵⁷ BY THE REACTION OF Er(p, 3pxn) AND THROUGH THE DECAY OF Ho157



(b) <u>COMPARISON OF EXPERIMENTAL AND</u> THEORETICAL CROSS SECTIONS

(i) (p,xn) reactions

The multi-isotopic nature of natural erbium which has been used as a target element limited the possibilities of comparison of theoretical and experimental cross sections. However, Tm^{168} , Tm^{170} and Er^{169} , whose formation cross sections are independent as well as being produced by only one type of reaction, have been used for comparison studies. In addition to this, the formation cross sections of Tm^{167} , which was produced by a (p,2n) reaction on Er^{168} , have been used for comparison purposes after resolving the mixed excitation function through enriched isotope irradiations.

As mentioned earlier, Jackson⁽²⁰⁾ used a schematic model for predicting (p,xn) cross section in non-fissile heavy elements. Jackson combined the Monte Carlo results of McManus et al.⁽²⁶⁾ for the nuclear cascade with a simplified evaporation theory in order to calculate the formation cross sections.

In the evaporation stage, it was assumed that a nucleus excited to an energy E would de-excite only by neutron emission as long as it is energetically possible. Charged particle (e.g. proton) emission was assumed to be negligible because of the high coulombic barrier in the case of high Z elements.

It was assumed that the neutron spectrum was given by

 $P(x) = E e^{-E/T}$

where the nuclear temperature T was assumed to be constant. This assumption of constant nuclear temperature was contrary to the assumptions inherent in most nuclear models.

Jackson showed that the probability that a nucleus with initial excitation energy E will evaporate x neutrons, is given by

 $P(E,x) = I(\Delta x, 2x - 3) - I(\Delta x + 1, 2x - 1).$ In this expression the probability P(E,x) for several values of x was evaluated using Pearson's incomplete gamma functions⁽¹²⁹⁾. The first incomplete gamma function represents the probability that at least x neutrons will be evaporated from the excited nucleus while the second gives the probability that at least (x + 1) neutrons will be emitted. The difference of the two terms gives the probability that exactly x neutrons will be emitted.

$$I(Z,n) = \left(\frac{1}{n!}\right) \int_{0}^{Z} y^{n} e^{-y} dy$$

where I(Z,n) is Pearson's incomplete gamma function, and $\Delta x = (E - \sum_{i}^{x} B_{i})/T$ where B_{i} is the binding energy of the ith neutron. In the present calculations the average binding energy of the neutron in this region was taken from Cameron's⁽¹¹⁸⁾ tables to be 7.4 Mev. Δx then is just the energy above the threshold for the emission of x neutrons.

An approximate nuclear temperature T was calculated from the expression

$$\Delta E = \overline{B} + 2T$$

where $\triangle E$ represents the energy separation in Mev between

successive peaks of (p,xn) excitation functions. According to Hollbach⁽¹³⁰⁾ ΔE should be 13 Mev for Z = 69. The average binding energy per nucleon, \overline{B} was taken as 8.2, giving a nuclear temperature, T = 2.4 Mev.

In the range of bombarding energies where all the interactions of the bombarding particle with the target nucleus involve the initial formation of a compound nucleus followed by the neutron evaporation, the formation cross sections are given by simple functions of the type

$\sigma(p,xn) = \sigma_c(E_o)P(E,x)$

where $\circ_{c}(E_{o})$ is the cross section for compound nucleus formation at incident energy of E_{o} . The values of $\circ_{c}(E_{o})$ were taken from Shapiro⁽¹³¹⁾. The nuclear radius R was taken to be 1.3 x 10⁻¹³ A^{1/3}. Shapiro's values have been extrapolated to 50 Mev using the formula given by him. The P(E,x) and $\sigma_{c}(E_{o})$ values are given in Figs. 72 and 73 respectively in Appendix III.

The comparison between experimental and calculated values is shown in Table XII and plotted in Fig. 69. The solid lines represent the theoretical curves, whereas the experimental cross sections were fitted by the dotted (broken) curves. Reaction cross sections for $\mathrm{Er}^{168}(\mathrm{p,n})\mathrm{Tm}^{168}$ and $\mathrm{Er}^{170}(\mathrm{p,n})\mathrm{Tm}^{170}$ have been used for comparison of the theoretical and experimental (p,n) results. The formation cross sections for the former reaction agreed very well within experimental error, whereas the results for the latter reaction were high by 30%.

Table XII

Proton Bombarding Energy in Mev	Type of Nuclear Reaction	Experimental Cross Sections measured (mb)	Theoretical Cross Sections calculated (mb)
10.1	Er ¹⁷⁰ (p,n)Tm ¹⁷⁰	70.76	70
12.8	11	117.60	120
16.4	n	24.30	25
10.1	Er ¹⁶⁸ (p,n)Tm ¹⁶⁸	95.52	70
12.8	11	174.28	120
16.4	n	24.39	25
16.4	Er ¹⁶⁸ (p,2n)Tm ¹⁶⁷	537.08	250
22.2	11	1660.17	550
28.5	π	406.17	130
34.0	n	117.54	40
22.2	Er ¹⁷⁰ (p,3n)Tm ¹⁶⁸	519.99	450
28.5	n	624.57	700
34.0	11	263.42	350

Figure 69

COMPARISON OF EXPERIMENTAL EXCITATION

FUNCTIONS WITH THOSE PREDICTED

BY THE JACKSON MODEL

	Theoretical
	Experimental
⊽	Er ¹⁷⁰ (p,n)Tm ¹⁷⁰
٠	Er ¹⁶⁸ (p,n)Tm ¹⁶⁸
0	Er ¹⁶⁸ (p,2n)Tm ¹⁶⁷
	$Er^{170}(p, 3n)Tm^{168}$



In the case of $\mathrm{Er}^{168}(p,2n)\mathrm{Tm}^{167}$ reaction cross sections, the comparison was poor. The experimental cross sections are about 200% higher than the calculated values. Jackson's calculated cross sections when compared with Bell and Skarsgard's results show the experimental values to be 60% higher than the calculated values. However, it is difficult to explain the much higher values which were encountered in the present work. This may well be due to the use of incorrect decay scheme data.

The excitation function of the $Er^{170}(p,3n)Tm^{168}$ reaction agreed very well with the calculated one within experimental error, as is shown in Fig. 69 and as compared in Table XII.

This type of comparison would have been better if the target element were monoisotopic, in which case comparison could be extended to other reactions as well.

(ii) (p,pn) reactions

Unfortunately, the radiochemical method measures only the total cross section or the sum of the various contributing mechanisms. Theoretical predictions based upon Monte Carlo cascade data are very useful, since the Monte Carlo method breaks down the total cross section into contributions from various mechanisms, but the difficulty of comparing total cross sections remained. Agreement between experiment and theory can thus be looked upon only as an indication of various mechanisms considered in the calculation of the predicted cross section. Strohal and

Caretto⁽¹³²⁾ presented an excellent discussion to explain the mechanism of (p,pn) reactions occurring in the 200 to 400 Mev proton energy range. This situation in this energy region is quite different when compared to the present work in which there is a transition between compound nucleus formation and knock-on cascade mechanism. At 200 Mev the fraction of inelastic events leading to compound nucleus decreases to less than 1% for copper and about 8% for uranium. It does not appear likely that compound nucleus mechanisms can play a significant role at energies about 100 Mev. The results of the Monte Carlo cascade calculations indicate that the fraction of all events leading to compound nucleus formation decreases with increasing energy and increases with atomic weight. In view of the above discussion, in the present work the compound nucleus mechanism explains the rapid increase of (p,pn) reaction until 50 Mev and from that energy onwards the excitation function starts to decrease and level off at higher energies (60 Mev onwards). This is a situation where the compound nucleus mechanism is fading and the knock-on cascade mechanism starts to play a part. Not very much work was done in theoretical predictions between 60 to 100 Mev.

Yule and Turkevich⁽⁶⁵⁾ combined the results of the intranuclear cascade and nuclear evaporation theories and calculated (p,pn) reaction cross sections for the energies 83, 156, 235 and 366 Mev and plotted cross section, (mb) vs. mass number. They assumed the cross section to be the sum of the following reactions:

- (1) a (p,pn) cascade leaving a nucleus with too little excitation to evaporate any other particles.
- (2) a (p,p') cascade followed by neutron evaporation.
- (3) a (p,n) cascade followed by proton evaporation.

According to them the (p,pn) cross section at 83 Mev is about 30 mb for mass number 169, whereas the present experimental value was about 60 mb.

It has been suggested (16,65) that this discrepancy arises from the use of a sharp nuclear boundary in these calculations, thus giving an incorrect number of low-energy depositions. However, it is not clear that making use of a diffuse nuclear boundary will lead to varying contributions from different reaction mechanisms with energy. It is interesting to note that this type of difference between the experimental and calculated values was observed by several authors (133, 65, 64, 132).

In general, little can be deduced from the present study about the relative contribution of individual direct interaction processes. Occasional suggestions may be made as to the probably contributing factors, but the unambiguous resolution of this problem awaits other, more detailed experiments involving the measurement of energy and angular distribution of the reaction products. (c) EXPERIMENTAL TEST FOR COMPOUND NUCLEUS THEORY

In the present work the compound nucleus Tm^{169} was produced through alpha irradiation of Ho^{165} and proton irradiation of Er^{168} . The following reactions were studied:

$$\begin{array}{c} \operatorname{Ho}^{165}(\alpha, n) & & \operatorname{Ho}^{165}(\alpha, 2n) \\ \operatorname{Er}^{168}(p, n) & & \operatorname{Tm}^{168} & & \operatorname{Er}^{168}(p, 2n) & & \operatorname{Tm}^{167} \end{array}$$

Both alpha and proton excitation functions are plotted in Fig. 70 and in Tables 13(a) and 13(b). Crosssection ratios are given at the same excitation energy. It can be seen that the ratios given below agreed very well within experimental error of $\stackrel{+}{-}$ 17.5%, (proton cross-section error is $\stackrel{+}{-}$ 14% and the alpha cross-section error $\stackrel{+}{-}$ 11%) at the same excitation energy for the compound state.

$$\frac{\sigma(p,n)}{\sigma(\alpha,n)} = \frac{\sigma(p,2n)}{\sigma(\alpha,2n)} \quad \text{or} \quad \frac{\sigma(p,n)}{\sigma(p,2n)} = \frac{\sigma(\alpha,n)}{\sigma(\alpha,2n)}$$

Ratio calculations have been made at the same excitation energies for both proton and alpha irradiations. Some excitation energies can be calculated from the following relation:

alpha excitation energy = proton excitation energy to to produce Tm^{169} produce $Tm^{169} + /proton$ binding energy 5.900 Mev - alpha binding energy (-0.534 Mev)7.

In other words 6.434 Mev, as obtained from reference (118), has to be added to the proton bombarding energies in order to produce Tm^{169} at the same alpha





193a

Proton Energy	α-Energy	0 (α,n)	0(p,n)	$\frac{\sigma(p,n)}{\sigma(\alpha,n)}$
8.5	15	6.81	20	2.94
13.5	20	39.16	115	2.94
18.5	25	5.6	14	2.5

Table XIII (a)

Table XIII (b)

Proton Energy	α-Energy	σ(α,2n)	0(p,2n)	<u>σ(p,2n)</u> σ(α,2n)
18.5	25	484.17	1400	2.89
23.5	30	445 .2 5	1500	3.37
28.5	3 5	112.17	400	3.57
32.5	39	59.64	180	3.02

excitation energy. Experimentally it has been observed that the excitation functions matched when the proton energy scale was shifted by $6.5 \stackrel{+}{-}1$ Mev.

In the case of Ghoshal's (5) experiment, the relation

 $c(p,n) : o(p,2n) : o(p,pn) = o(\alpha,n) : o(\alpha,2n) : o(\alpha,pn)$ was observed to hold good when 7 Mev was added to the proton energy to match the excitation energy of the compound nucleus.

John⁽⁶⁾ tested the relation

 $o(p,2n) : o(p,3n) : o(p,4n) = o(\alpha,2n) : o(\alpha,3n) : o(\alpha,4n)$ He added 11.9 Mev to the proton energy to match the curves.

On the other hand, Gibson⁽⁸⁾ tested the compound nucleus Am^{241} by checking the ratios

 $\sigma(d,2n)$: $\sigma(d,3n) = \sigma(\alpha,2n)$: $\sigma(\alpha,3n)$

and $\circ(d,2n)$: $\circ(\alpha,2n) = \circ_{c}(d) : \circ_{c}(\alpha) = \circ(d,3n) : \circ(\alpha,3n)$. In this case the deuteron curves have been shifted by adding 13.9 ⁺ 1 Mev to the deuteron energy to match the curves.

In the case of John⁽⁶⁾ and Ghoshal⁽⁵⁾, they got almost perfect superimposition of the (p,xn) excitation functions and (α ,xn) excitation functions. In the case of the present work, and Gibson's work as well, the ratios only compared very well within experimental error, while superposition of the curves was not obtained.

Ghoshal's experiment provided a good test for compound nucleus formation in the region of medium weight elements. John and Gibson provided good tests for compound nucleus formation in the regions of non-fissile heavy elements and fissile heavy elements respectively. In the present work the test was applied in the region of medium heavy elements.

According to ancient Hindu Mythology, God Siva accepted with equanimity, not only the most wanted products like Moon, nectar and several other good things, but also the vicious poison, from gods and demons during their exploration of the sea.

Similarly the present work: spallation of multiisotopic natural erbium yielded many wanted things as listed in the summary and less informative composite excitation functions, which were accepted by the author with equanimity!

E. SUMMARY AND CONTRIBUTIONS TO KNOWLEDGE

1. Mixtures of erbium oxide and copper oxide of natural isotopic abundance have been irradiated in the McGill Synchrocyclotron with protons of energies up to about 86.5 Mev. Enriched isotope samples of erbium oxide mixed with copper oxide have been irradiated with protons to resolve the $\mathrm{Er}^{168}(p,2n)$ reaction excitation function. Holmium oxide targets have been irradiated in the Brookhaven National Laboratory Cyclotron with alpha particles of up to about 39 Mev energy.

2. Anion exchange separation has been developed to separate the monitor nuclide activity (Cu^{64}) . Cation exchange separation has been developed to separate quickly (in about 70 minutes) milligram quantities of individual heavy rare earths (Tm, Er, Ho and Dy).

3. Spallation products that were identified covered $66 \le z \le 69$ and $157 \le A \le 170$, and they are: 128-d Tm^{170*}, $86-d Tm^{168*}$, 9.6-d Tm^{167*}, 7.7-hr Tm^{166*}, 28-hr Tm^{165*}, $1.85-min Tm^{164}$, 1.8-hr Tm^{163*}, $50-min Tm^{161}$, 9-d Er^{169*}, $10.4-d Er^{165*}$, 75-min Er¹⁶³, 3.1-hr Er^{161#}, 29.5-hr Er^{160#}, $2.5-hr Ho^{161*}$, 5-hr Ho^{160m*}, 22-min Ho¹⁶⁰, 138-d Dy^{159#}, and 8.2-hr Dy^{157#}.

4. Transitions of 1850 and 2050 kev belonging to 7.7-hr Tm^{166} have been definitely identified.

6. The existence of 1.85-min Tm^{164} , 90-min Tm^{162} , 1.8-hr Tm^{163} and \sim 30-min Tm^{161} has been confirmed.

7. The following decay chains have been confirmed: $Tm^{161} (\underline{(30 \text{ min})} \text{ Er}^{161} \underline{3.1 \text{ hr}} \text{ Ho}^{161} \underline{2.5 \text{ hr}} \text{ Dy}^{161} (\text{stable})$ and $Tm^{160} \underline{\text{ short lived}} \text{? Er}^{160} \underbrace{\text{ Er}^{160}}_{\text{Ho}} Ho^{160} \underbrace{\text{ Dy}^{160} (\text{stable})}_{\text{Ho}^{160}}$

8. Independent excitation functions have been obtained for the nuclides marked with \mathbf{x} ; cumulative yields have been obtained for the nuclides marked with #, as indicated above. In addition, $\mathrm{Ho}^{165}(\alpha,n)$ and $\mathrm{Ho}^{165}(\alpha,2n)$ reaction excitation functions have been obtained.

9. $Er^{170}(p,n)$, $Er^{168}(p,n)$, $Er^{168}(p,2n)$ and $Er^{170}(p,3n)$ reaction excitation functions have been compared with the predictions of Jackson's schematic model. The (p,n) and (p,3n) reaction excitation functions were in good agreement, whereas (p,2n) reaction excitation function values are 200% higher than the predicted values.

10. The experimentally determined $Er^{170}(p,pn)$ reaction cross section at ~ 83 Mev has been compared with

the literature value calculated from Monte Carlo cascade data, combined with evaporation theory, and found to be high by a factor of 2.

11. Experimental test for the Tm¹⁶⁹ compound nucleus formation was provided by the following relation:

$$\frac{o(p,n)}{o(\alpha,n)} = \frac{o(p,2n)}{o(\alpha,2n)}$$

APPENDIX I

PHOTOPEAK EFFICIENCY CALIBRATION OF 3" x 3" NaI(T1) CRYSTAL

According to the availability, standards covering the useful energy range 30 kev to 3000 kev have been chosen for the calibration of the crystal, bearing the following requisites in mind:

- The nuclide should have a known and preferably simple decay scheme.
- The half-lives of the nuclides should be well known and preferably not too short.
- 3. The gamma-ray energies should be known precisely.
- 4. It should be possible to standardize the nuclides by
 - 4π $\beta\text{-measurement}$ or by coincidence techniques.
- 5. Nuclides should be pure, otherwise they should be

purified from any possible impurities.

Marion⁽¹³⁴⁾ listed the following nuclides according to the above requisites.

	Nuclide	Gamma-ray energy	Half-life		
	Am ²⁴¹	0.05968	458 y		
	Co ⁵⁷	0.12198	268 d		
x	Ce ¹⁴¹	0.14550	32.51 d		
	In ^{114m}	0.1903	50.0 d		
x	Hg ²⁰³	0.27912	49.9 d		

*Complete details of calibration have been described by George Grant in his thesis 'Spallation of Iridium' submitted to McGill University (1961, May).

	Nuclide	Gamm	a-ray energy	<u>Half-life</u>
	Cr ⁵¹		0.32100	27.8 d
x	Au ¹⁹⁸		0.41176	2.7 d
	Be ⁷		0.4780	53.4 d
x	Cs ¹³⁷		0.6616	28 y
	1 ¹³¹		0.7239 0.6380 0.3644 0.2843	8.066 d
	Mn ⁵⁴		0.835	313.5 d
	Zn ⁶⁵	Annihilation	1.114 0.5109	244.4 d
x	sc ⁴⁶		1.1180 0.8920	83.9 d
x	Na ²²	Annihilation	1.127634 0.5109	2.58 y
x	c° ⁶⁰		1.3325 1.1727	5.27 y
	¥ ⁸⁸		1.841 0.900	105 d
	S b ¹²⁴		2.088 1.692	60.6 d
x	Na ²⁴		2.7535 1.3679	14.87 hr
		y = year,	d = day, $hr = hour$	

*These nuclides were used for the purpose of efficiency calibration.

In the case of low-energy range (< 100 kev) use was made of the characteristic X-rays emitted following internal conversion of the gamma rays in the following sources:

Nuclide		<u>X-ray</u>	energy	in	kev
Cs ¹³⁷	(Ba ^{137m})		32		
Ce^{141}	(Pr ¹⁴¹)		3 5		
Hg ²⁰³	(T1 ²⁰³)		72		

All the activities were radiochemically purified.

The next step was the determination of the absolute disintegration rate of each nuclide, by measurement of suitable aliquots in a gas flow 4π β -proportional counter. The measured rate was corrected for resolution losses, source-mount absorption and self-absorption. All these corrections and 4π -counting techniques used, have been developed in this laboratory by Yaffe et al. (79,80,81,82).

Standard sources for Y-scintillation measurement were made by pipetting suitable aliquots and making up to exactly 2 ml volume into small screw capped glass vials. The sources were then measured on each of ten shelves of reproducible geometry situated on the top of the crystal.

Photopeak efficiency was calculated in brief as follows:

(i) Photopeak efficiency = $\frac{A}{N_{\gamma}}$

where A is photopeak area in counts per minute and N γ is photon emission rate in gammas/minute.

- (ii) Disintegration rate = $N\gamma$ + No. of converted X branching ratio = lectrons
- (iii) $N_{\gamma} = \frac{\text{Disintegration rate X branching ratio}}{1 + \text{total conversion coefficient}}$

Total conversion coefficients and branching ratios have been obtained from the literature (91,85).

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In the case of nuclides whose K-X-rays have been measured, the right-hand side of equation (iii) was multiplied by the fluorescence yield for K-shell, W_k , obtained from the reference (135) and by the internal conversion coefficient for the K-shell.

Experimental photopeak efficiency curves are shown in Figs. 71(a) and 71(b). Most of the points fitted within 5%. In Fig. 71(b) the dashed line which is the theoretical efficiency curve for the tenth shelf is high, whereas the experimental curve is low, especially in the low-energy region.

This could be attributed to the fact that the theoretical efficiency curves were drawn based on the fact that the source is an isotropic point source, whereas the experimental source is a 2 ml volume of liquid in a glass vial. Figure 71(a)

PHOTOPEAK EFFICIENCIES OF 3" x 3" NaI(T1)

CRYSTAL vs. GAMMA-RAY ENERGY FOR

SHELVES 0 - 4 INCLUSIVE



Figure 71(b)

PHOTOPEAK EFFICIENCIES OF 3" x 3" NaI(T1)

CRYSTAL	Vs.	GAM	MA-	RAY	ENE	RGY	FOR
SHE	LVES	5 -	10	INC	CLUS	IVE	
	0		Exp	erin	nent	al	
		_	The	oret	tica	1	



a

APPENDIX II

CALCULATION OF DISINTEGRATION RATE OF INDEPENDENTLY PRODUCED B IN THE CASE OF THE CHAIN $A \rightarrow B \rightarrow C(STABLE)$

The number of nuclei of B, N_B ', grown due to the decay of A during the irradiation time, t, is given by the expression:

$$N_{B}' = \frac{D_{A}^{O}}{\lambda_{B}(1 - e^{-\lambda_{A}t})} \left[1 + \frac{\lambda_{B} e^{-\lambda_{A}t} - \lambda_{A} e^{-\lambda_{B}t}}{\lambda_{A} - \lambda_{B}}\right]$$

or

$$D_{B}' = \frac{D_{A}^{O}}{1 - e^{-\lambda_{A}t}} \left[1 + \frac{\lambda_{B} e^{-\lambda_{A}t} - \lambda_{A} e^{-\lambda_{B}t}}{\lambda_{A} - \lambda_{B}}\right]$$

where $D_B^{\circ} =$ number of disintegrations of B, grown in from A, during irradiation at end of bombardment. D_A° and $D_B^{\circ} =$ disintegration rates of A and B at end of bombardment, produced independently. λ_A and $\lambda_B^{\circ} =$ disintegration constant of A and B respectively.

At a time, T, after the irradiation, the disintegration rate of those nuclei formed from the decay during the irradiation is given by

$$D_{B}' e^{-\lambda}B^{T}$$

In the time interval, T, B has continued to grow from A and the disintegration rate of B, $D_B^{"}$, from this source is given by

$$D_{B}^{"} = \frac{\lambda_{B}}{\lambda_{B} - \lambda_{A}} D_{A}^{O} (e^{-\lambda} A^{T} - e^{-\lambda} B^{T})$$

Therefore, the experimentally measured disintegration rate

of B, D_B^{exp} , at a time T, after irradiation is given by the sum

$$D_{B}^{exp} = D_{B}^{o} e^{-\lambda} B^{T} + D_{B}' e^{-\lambda} B^{T} + D_{B}''$$

from which D_B^o can be calculated, and hereby independent formation of B can be calculated. This type of calculation needs the knowledge of decay schemes of A and B to calculate D_A^o and D_B^{exp} . In addition, it is necessary to know accurately the duration of irradiation, t, and the time interval, T, between the end of irradiation and the end of separation.

APPENDIX III

Figs. 72 and 73

Figure 72

THE PROBABILITIES P(E,x) FOR NEUTRON

EMISSION vs. PROTON ENERGY IN MEV



P(E,x)


C VERSUS PROTON ENERGY

SHAPIF	10(1	31)		
	Z	-	60	
	Z	=	70	



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