

NEUTRON CAPTURE BY THE ISOTOPES OF

SOME RAPE EARTH ELEMENTS

.

THE RELATIVE ABUNDANCES

AND

PILE NEUTRON CAPTURE CROSS SECTIONS

OF THE ISOTOPES OF

SAMARIUM, GADOLINIUM, DYSPROSIUM AND YTTERBIUM

by

WILLIAM HERBERT WALKER, M. Sc.

A Thesis

Submitted to the Faculty of Arts and Science in partial Fulfillment of the Requirements for the Degree Doctor of Philosophy

McMaster University

May, 1956

DOCTOR OF PHILOSOPHY (1956)

McMaster University Hamilton, Ont.

TITLE: The Relative Abundances and File Neutron Capture Cross Sections of the Isotopes of Samarium, Gadolinium, Dysprosium and Ytterbium.

AUTHOR: William Herbert Walker, B.A. Sc. (University of Toronto) M. Sc. (McMaster University)

SUPERVISOR: Professor H.G. Thode

NUMBER OF PAGES: 114

SCOPE AND CONTENTS:

A double-focussing mass spectrometer has been used to determine the relative abundances of the isotopes of naturallyoccurring platinum, samarium, gadolinium, dysprosium and ytterbium. The relative abundances of a portion of the four rare earth elements were also measured after bombardment in a high neutron flux for a period of six months. From a comparison of the results for the irradiated and unirradiated samples the neutron absorption cross sections of the individual isotopes have been calculated.

ii

ACKNOWLEDGMENTS

I should like to thank Professor H.G. Thode, under whose direction this work has been done, for his encouragement and for many discussions which were helpful in overcoming the difficulties arising during the course of the experiments and the analysis of the results.

I am indebted also to Dr. R.H. Tomlinson for his guidance in developing suitable chemical techniques for handling the rare earth materials, and to other members of the science faculty for helpful discussions on specific problems.

Dr. S.A. Kushneriuk, of Atomic Energy of Canada Ltd., was most helpful in discussions concerning the reduction of neutron flux within the samples irradiated.

The generous financial assistance of Atomic Energy of Canada Limited has made this work possible.

(iii)

TABLE OF CONTENTS

Section	Subject	Page
	Scope and Contents	11
	Acknowledgments	111
	List of Tables	vii
	List of Illustrations	ix
I	Introduction	1
II	Isotope Redistribution due to Neutron Irradiation	4
III	Apparatus	
	1. Deflection Chambers	7
	2. Collector	7
	3. Source	8
	4. Pumping System	13
	5. Electronics	14
IV	Experimental Procedure	15
	1. Sample Preparation	1 5
	2. V-Source	16
	3. Slits	17
	4. Voltages and Currents	17
	5. Treatment of Data	18
	(i) Determination of Relative Abundances (ii) Analysis of Abundance Data (iii) Contaminants	18 19 21

	Table of Contents (Cont'd)	
Section	Subject	Page
v	Results	
A -	Percent Relative Abundances	24
	1. Naturally-occurring Platinum	24
	2. Naturally-occurring Samarium	28
	3. Neutron Irradiated Samarium	37
	4. Naturally-occurring Gadolinium	43
	5. Irradiated Gadolinium	47
	6. Naturally-occurring Dysprosium	47
	7. Irradiated Dysprosium	50
	8. Naturally-occurring Ytterbium	52
	9. Irradiated Ytterbium	52
v		
B -	Determination of Isotopic Capture Cross S	Sections
	1. Introduction	56
	2. Samarium	58
	3. Gadolinium	62
	4. Dysprosium	63
	5. Ytterbium	67
VI	Discussion	
A-	Absolute Abundances of the Isotopes of Naturally-occurring Elements	71
B-	Isotopic Neutron Capture Cross Sections	80
	1. Samarium	80
	2. Gadolinium	83
	3. Dysprosium	84
	4. Ytterbium	85

		Table of Contents (Cont'd)	
Section		Subject	Page
APPENDICES	- A	Effect of Neutron Irradiation on Isotopic Abundances. Derivation of the Equations	92
	- B	Flux Depression in the Irradiated Samples	99.
REFERENCES			111

.

.

LIST OF TABLES

Number	<u>Title</u>	Page
I	Operating Currents and Temperatures for Thermionic Emission	18
II	Abundance of the Isotopes of Platinum	26
III	Mass Spectrometric Results for Unirradiated Samarium	32
IV	Time Dependence of the Relative Abundances of the Isotopes of Naturally-occurring Samarium	34
V	Mass Spectrometric Results for Irradiated Samarium	41
VI	Time Dependence of the Relative Abundances of the Isotopes of Irradiated Samarium	42
VII	Maas Spectrometric Results for the Oxide of Naturally-occurring Gedolinium	45
VIII	Mass Spectrometric Results for the Oxide of Irradiated Gadolinium	46
IX	Mass Spectrometric Results for Naturally- occurring Dysprosium	49
x	Mass Spectrometric Results for Irradiated Dysprosium	51
XI	Mass Spectrometric Results for Naturally- occurring Ytterbium	54
XII	Mass Spectrometric Results for Irradiated Ytterbium	55
XIII	Isotopic Cross Section Calculations for Samaria	um 61
XIA	Isotopic Cross Section Calculations for Gadolin ium	n- 62,63
XV	Isotopic Cross Section Calculations for Dyspro- ium	65,6 6
XVI	Isotopic Cross Section Calculateons for Ytterb:	ium 69,70

List of Tables (Cont'd)

Number	<u>Title</u>	Page
XVII	Measurements of the Isotopic Abundances of Naturally-occurring Platinum	75
XVIII	Measurements of the Isotopic Abundances of Naturally-occurring Samarium	76
XIX	Measurements of the Isotopic Abundances of Naturally-occurring Gadolinium	77
XX	Measurements of the Isotopic Abundances of Naturally-occurring Dysprosium	78
XXI	Measurements of the Isotopic Abundances of Naturally-occurring Ytterbium	79
XXII	Measurements of the Neutron Capture Cross Sections of Samarium Isotopes	88
XXIII	Measurements of the Neutron Capture Cross Sections of Gadolinium Isotopes	89
VIXX	Measurements of the Neutron Capture Cross Sections of Dysprosium Isotopes	90
XXV	Measurements of the Neutron Capture Cross Sections of Ytterbium Isotopes	91
BI	Isotopic Capture Cross Sections for Neodymium	105

LIST OF ILLUSTRATIONS

Figure	Title	Page
1.	Adjustable Slit and Collector	7a
2.	Mass Spectrometer Source	9
3.	Pumping System	12
4.	Mass Spectrum of Platinum Isotopes	25
5.	Platinum 190-192 Ratio	27
6.	Spectrum of Unirradiated Samarium with Rare Earth Contaminants	30
7.	Spectrum of Unirradiated Samarium with Hydrocarbon Peaks	31
8.	Least Squares Fit for Unirradiated Samarium	33
9.	Metal Ions Spectrum of Irradiated Samarium	36
10.	Survey of Ions from Irradiated Samarium on Platinum V	39
11.	Gadolinium Spectra	44
12.	Dysprosium Spectra	48
13.	Ytterbium Spectra	53

I. INTRODUCTION

The radiative absorption cross sections of individual isotopes for neutrons having an extended energy spectrum have been measured in several ways. The most sensitive method has been the determination of those cross sections leading to radioactive nuclei, usually referred to as "activation cross sections" 1,2,3. In this method the intensity of the radioactivity of the product of neutron capture is measured. The best accuracy obtainable is of the order of several percent, while the minimum cross section measured has been a fraction of a millibarn."

The most accurate measurements have been made with "pile oscillators" 4,5,6, in which a measurement is made of the magnitude of the oscillation produced in a neutron flux by the periodic movement of the unknown absorber. To determine the cross sections of individual isotopes, it is necessary to compare several samples each enriched in a different isotope. Although the accuracy of determination of the macroscopic sample cross sections in each case can be a few tenths of one percent or better, the microscopic (nuclear) cross section for each isotope can only occasionally be determined to better than one percent. The minimum cross section measured by this method is of the order of a tenth of a barn.

1 barn = 10^{-24} cm², 1 millibarn = 10^{-3} barn

The most flexible method of measuring isotopic absorption cross sections requires a mass spectrometer. By observing the changes produced by neutron irradiation in the relative abundances of each isotope, their cross sections can be calculated. The sample may be either the naturally-occurring element 7-13, a special selection of isotopes obtained by isotopic enrichment 14, or separated fission products 10,15,16,17. The accuracy and minimum cross section which can be measured are determined by the total irradiation and the sensitivity of the mass spectrometer.

The work reported in this thesis is an extension of the determination of isotopic cross sections initiated in the author's M. Sc. Thesis 11 on the neodymium isotopes. Where the contents of this thesis have already been covered in the M. Sc. thesis, particularly the sections on theory and instrumentation, the latter is quoted generously.

The elements discussed here - samarium, gadolinium, dysprosium and ytterbium - were irradiated along with the neodymium and some other samples inside the calandria of the NRX reactor at Chalk River for an estimated total irradiation ^{*} of 0.4 neutrons per kilobarn. For an irradiation of this magnitude, the range of cross sections which

This value was estimated from the integrated power output of the reactor over the irradiation period and the estimated flux density at the irradiation position. Accuracy is not expected to be better than 20%.

- 11

can be determined with the double-focussing mass spectrometer used is from 10 barns to about 20 kilobarns. Within this range the cross sections of Sm¹⁴⁷, Sm¹⁵⁰, Sm¹⁵², Gd¹⁵⁴, Gd¹⁶⁰, Dy¹⁶⁰, Dy¹⁶¹, Dy¹⁶², Dy¹⁶⁸, Dy¹⁶⁴, Yb¹⁷¹, Yb¹⁷², Yb¹⁷⁸ and Yb¹⁷⁴ have been determined. In addition, upper limits have been set for Sm¹⁴⁸, Sm¹⁵⁹, Gd¹⁶⁶, Gd¹⁵⁸ and Yb¹⁷⁰.

II. ISOTOPE REDISTRIBUTION DUE TO NUTRON IRRADIATION

The destruction of a nuclide by the capture of a neutron is analogous to destruction by the radioactive emission of a charged particle. In the latter case $dn/dt = -\lambda n$, where n is the number of nuclides present and λ is the "decay constant" of the nuclide. In the case of neutron capture, the absorption cross section of is defined by the equation $dn/dt = -6\pi$, where π is the neutron flux density in neutrons/cm²/sec. Thus 6 has the dimensions of area. To illustrate the mechanism of multiple neutron capture and show how the equations used to calculate the capture cross sections arise, consider the neutron capture chain A1 (n1, 61) neutron. A2 (n2, σ_2) neutron A3 (n3, σ_3) ----- neutron A_S (n_S, σ_S) where A; is the mass number of ith isotope and n; and of are its initial abundance and cross section respectively. The final abundance of the ith isotope, n1, will be the sum of the contributions, $\sum_{i=1}^{2} i(\delta n!)_{i}$, of the individual isotopes treated separately. The first isotope will contribute an amount given by 0-0700 (1) $(on!) = n_0 \delta_0 \delta_0 = 0$

$$+ \frac{e^{-\sigma_2 \varnothing t}}{(\sigma_1 - \sigma_2)(\sigma_3 - \sigma_2) - (\sigma_1 - \sigma_2)} + \frac{e^{-\sigma_1 \vartheta t}}{(\sigma_1 - \sigma_2)(\sigma_3 - \sigma_2) - (\sigma_1 - \sigma_2)} + \frac{e^{-\sigma_1 \vartheta t}}{(\sigma_1 - \sigma_1)(\sigma_2 - \sigma_1) - (\sigma_{1-1} - \sigma_{1})}$$

which is just the general formula for the number of nuclei of the i^{th} product of a radioactive chain, with \mathcal{O} replacing λ . If i = 3, we have

4.

(2)
$$n_{3}^{2} = \sum_{j=1}^{3} (\delta n_{3}^{*})_{j}^{j}$$

= $n_{1}\sigma_{1}\sigma_{2} \left[\frac{e^{-\sigma_{1}\phi t}}{(\sigma_{2}-\sigma_{1})(\sigma_{3}-\sigma_{1})} + \frac{e^{-\sigma_{2}\phi t}}{(\sigma_{1}-\sigma_{2})(\sigma_{3}-\sigma_{2})} + \frac{e^{-\sigma_{3}\phi t}}{(\sigma_{1}-\sigma_{3})(\sigma_{2}-\sigma_{3})} + n_{2}\sigma_{2}\phi t + n_{2}\sigma_{2}\phi t + \frac{e^{-\sigma_{3}\phi t}}{(\sigma_{3}-\sigma_{2})} + \frac{e^{-\sigma_{3}\phi t}}{(\sigma_{2}-\sigma_{3})} + n_{3}e^{-\sigma_{3}\phi t} + n_{3}e^{$

5

In appendix A the detailed development of these equations is treated. It is shown that equation (2) can be expressed in terms of σ_3 in the form (3) $\sigma_3 \not = \ln \left[\frac{n_3 +}{n_2 \sigma_2} (\sigma_2 - \sigma_3)^{-1} + n_1 \sigma_1 \sigma_2 (\sigma_1 - \sigma_3)^{-1} (\sigma_2 - \sigma_3)^{-1} \right] - \ln \left[n_3 + n_2^2 \sigma_2^2 \sigma_3^{-1} + n_1 \sigma_1 \sigma_2 (\sigma_1 - \sigma_3)^{-1} (\sigma_2 - \sigma_3)^{-1} \right]$

A considerable simplification can be effected if the net change in the abundance of the isotopes is small. If it is assumed that the rate of change is constant with time, rather than exponential, the cross section equations become

(4)
$$\sigma_1 p t = \frac{\Delta n_1}{(1/2)(n_1+n_1)}$$
, $\sigma_2 p t = \frac{\Delta n_1 + \Delta n_2}{(1/2)(n_2 + n_2)}$, etc.

where $\triangle n = n-n^{\dagger}$. It is shown in appendix A that these simplified equations are a more exact modification of the simple first order approximation.

These equations have been developed in terms of numbers of nuclei. As long as there is no gain or loss of nuclei for the element, no correction is required when percent relative abundances are used instead. However, where nuclei are lost to the element through radioactive decay of a capture product or gained by the capture in, and subsequent decay of, a contaminant, the uncorrected use of relative abundances can lead to erroneous values for the absorption cross sections.

III. - APPARATUS

The machine used for the experiments described here is a double-focussing mass spectrometer using an electronbombarded V-source suitable for solid samples. With the exception of the source and collector which were modified, the machine has been described in detail by R. Horsley ¹⁸.

1. DEFLECTION CHAMBER

The machine was originally built as a mass spectrograph for the measurement of mass doublets. The deflection chamber, which is dimensionally identical with Dempster's first double-focussing mass spectrograph 19,20 , was retained on conversion to a mass spectrometer for measuring isotopic abundances.

The double-focussing property of combinations of a radial electric field followed by a magnetic field has been discussed by Mattauch ²¹. He showed how certain combinations, of which the Dempster spectrometer is a particular case, will focus a beam of ions which is not monoenergetic, in addition to the "direction-focussing" of a divergent beam of which a simple magnetic spectrometer is capable. The optical analogue is the achromatic lens.

2. COLLECTOR

To provide a wide range of resolution combined with optimum peak shape a collector unit was built (Fig. 1) with a variable width slit. The slit width can be adjusted continually over its range by turning the differential screw.



designed to give an increment of 0.05 mm. per revolution. The small plate which covers the edge of the collector insulator was found necessary in order to eliminate both positive and negative secondary peaks caused by ions and electrons scattered by the beam when it reached the inner edge of the polystyrene beyond the brass plates. 3. SOURCE

The new source is based on the same principle as the Shaw crucible source²², heating by bombardment with a controlled current of energetic electrons, but designed to have a higher efficiency, and to make more easily reproducible the mechanical alignment which is used for focussing in this machine.

The source and its supporting assembly are shown in Figure 2, which is a horizontal section in the plane of the source slits. A metal rectangle about 4 mm. x 6 mm. x 0.2 mm. is shaped with a cold chisel to form a simple rightangled V. This is mounted by one inch supports of $.020^{\text{m}}$ tantalum wire onto two $.060^{\text{m}}$ tungsten posts. Two shorter posts of the same diameter ($.060^{\text{m}}$) support a $.030^{\text{m}} \times .001^{\text{m}}$ tungsten ribbon which is heated to provide the current of electrons for bombarding the V. Each tungsten post is insulated from the supporting stainless steel collar by a short glass tube to which it is sealed.



Around this unit is a radiation shield or oven which has a slit in the centre of its nichrome cover about $0.20" \ge 0.05"$. The shield is supported by a spider of three glass rods to the inner section of the supporting brass telescope. The shield and the fine support wires on the V are necessary to reduce the loss of heat from the V so that its temperature may be raised high enough to provide a satisfactory current of thermally produced ions from the samples studied.

The two heater supports and one V support are connected electrically to the three Kovar-glass terminals at the end of the brass tube which supports the source. The radiation shield is connected electrically to one side of the heater supply. Radiation shield and source are maintained at about 5000 volts with respect to the source slits mounted at the end of the electrostatic deflector.

The source and its supports are held in position and insulated from ground by a glass cap made from the bottom of a heavy-walled pyrex distillation flask. The vacuum seal at both inner and outer glass-metal contacts is made with Armstrong Adhesive ^{*} which also provides ample mechanical support.

* Armstrong Products Co., Warsaw, Indiana, U.S.A.

The inner sleeve of the brass telescope carries both the source and the radiation shield. The outer sleeve is vacuum-sealed by a flat neoprene gasket to the end of the electrostatic deflector, and carries a side tube for separate pumping of the source.

Small vertical and horizontal movement of the outer sleeve permits initial alignment of the slits. The horisontal position of the inner relative to the outer sleeve sets the distance between the fixed source slits and the radiation shield. Adjustment of the alignment and spacing of the V with respect to the radiation shield slit is accomplished by the rotation and horizontal movement, respectively, of the tube which carries the V source.

In operation, a 500 volt potential is maintained between the V and its neighbours. This serves to accelerate the regulated current of electrons from the tungsten ribbon onto the V. The ions produced by thermal ionization of the sample, which is deposited in the trough of the V, are accelerated by the same field towards the cover of the radiation shield. Those ions which pass through the slit in the shield are then accelerated towards the source slits by the accelerating potential of about 5000 volts. In addition to facilitating alignment of the source and slits, the various movements available permit a certain amount of focussing of the ion beam by proper choice of gaps for the accelerating fields.



4. PUMPING SYSTEM

The pumping system is shown schematically in Figure 3. The electrostatic deflector is a separate unit vacuumsealed to the magnetic deflector by an O-ring. The collector is sealed to the magnetic deflector in the same way.

The magnetic deflector itself consists of four parts: two shaped iron pole faces and two brass spacers, one curved and one flat. The latter is the section to which the electrostatic deflector and collector are sealed. These four pieces are joined together by screws and sealed with Apiezon "W" black wax, which is applied to the surfaces of the joints before assembly after the four pieces have been heated to about 200°F in an oven.

The source assembly pumping system is isolated as much as possible from that of the deflection chamber, the source slits providing the only path for diffusing gas between the two.

The main pumping line is from the electrostatic deflector to the trap and two-stage oil-diffusion pump (Edwards 03B) via a large Kovar-glass seal. The auxiliary line to the source is sealed to the brass pumping tube shown in figure 2 by black wax. A ball and socket joint between the small Edwards 10B two-stage oil-diffusion pump and the source provides the flexibility necessary to accommodate movements of the source assembly. Both diffusion pumps are backed by a single Welsh Duoseal two-stage rotary pump.

5. ELECTRONICS

The accelerating voltage and deflecting voltage stabilizers, and the magnet current stabilizer have been described by Horsley ¹⁸. Sporadic discontinuities in the magnet current of undetermined origin made it preferable to scan the mass spectrum by changing the current manually, in order to ensure that the maximum of each peak was recorded correctly.

The current from the Faraday collector produces a voltage across a Victoreen resistor of 5×10^{10} ohms, which is amplified by a vibrating reed electrometer ^{*} and fed through a constant impedance attenuator into a strip chart recorder ^{**}.

* Applied Physics Corporation Model 30E Vibrating Reed Electrometer ** Brown "Elektronic" Potentiometer Type with two-second response

IV EXPERIMENTAL PROCEDURE

1. SAMPLE PREPARATION

All of the rare earths investigated were obtained from A.D. McKay and Co., New York, prior to 1945. Purities were not high except in the case of ytterbium, and were not specified in some cases. Half of each sample, up to a maximum of about 30 milligrams, was placed in a small aluminium tube made from $1/8^{m}$ rod closed with a screw cap. These were sealed inside a standard NRX aluminium irradiation capsule, along with the neodymium reported previously, 11,12 and a number of other samples. Irradiation to a nominal 0.4 neutrons per kilobarn took six months, and was completed in July, 1951.

The procedure used in preparing samples for analysis followed a similar pattern for each element. The unirradiated portion was used to establish satisfactory techniques which were then applied to the irradiated portion.

In the case of samarium, which came in the form of metal filings of unspecified purity, a few milligrams were oxidized by heating to redness in a crucible. After cooling the sample could be dissolved in a few drops of concentrated hydrochloric acid. A few drops of diluted solution, estimated to contain about 100 micrograms, were then placed in the trough of the V and evaporated slowly to dryness.

Attempts to reduce unidentified contaminations in the unirradiated samarium by precipitation as the oxalate and by treatment with perchloric acid did not produce any improvement.

The other three elements came in the form of the trioxide. The gadolinium was rated as "pure", and purities of 98% and 99.99% were given for the dysprosium and ytterbium oxides respectively. The dysprosium oxide dissolved in hot dilute hydrochloric, the others in concentrated hydrochloric. Dilution and application to the V were treated in the same manner as the samarium. The charges were estimated to lie in the range 50 to 100 micrograms.

2. V-SOURCE

In all cases successful operation was obtained with a tantalum V which had strips of platinum welded to the emitting surface. The V was formed from a rectangle approximately 6 mm. x 4 mm. and $.007^{"}$ thick. The platinum strips, about $.02^{"}$ x $.002^{"}$ x 6 mm., usually three in number, were spotwelded to the surface of the rectangle before shaping it into a V.

Two other materials were used to make V's. For the first runs on dysprosium when the oxide ions were being measured, the tantalum was replaced by molybdenum because of interference in the mass spectrum from the Ta^{lel} peak. In addition, a

platinum V of about the same dimensions was used for the work on irradiated samarium, in which the oxide peaks were measured.

3. SLITS

For samarium, the source slit was 0.010 cm. wide and the collector slit was 0.006 cm. For the other three elements the source slit was reduced to 0.005 cm. The theoretical resolutions for these two cases are $\frac{1}{510}$ and $\frac{1}{780}$ respectively. 4. VOLTAGES AND CURRENTS

The accelerating voltage between the source and source slit was kept at approximately 5200 volts, small adjustments being required occasionally for optimum focussing. The deflecting voltage between the plates of the radial electrostatic deflector was held at 700 volts. For this combination, the metal ions of samarium appear at a magnet current near 60 milliamps. For the other metal ions and oxide ions the current is correspondingly higher.

No direct measurement can be made of the source temperature. However, the current of the bombarding electrons can be measured, and for V's of similar dimensions the temperatures corresponding to a particular current should be nearly equal. Since most of the ion emission occurs at temperatures in the neighbourhood of the platinum melting point. it is possible to make a rough estimate of the operating temperatures assuming radiative heat loss only. The estimated temperature ranges are shown in Table I.

TABLE I

OPERATING CU		
Ions	Current Range - milliamps	Temperature Range - °C
Pt melting point	~30	1770
Sm oxide on Pt V	≲30	~1700
Sm metal (unirrad- iated)	40-60	1900-2200
Sm metal (irrad- iated)	45-75	2000-2300
Gd oxide	30-45	1700-2000
Dy metal and Dy or	ide 30-55	1700-2100
Yb metal	20-40	1600-1900

5. TREATMENT OF DATA

(1) Determination of Relative Abundances

Relative abundances were determined by the standard techniques employed in this laboratory. The mass spectrum is scanned in one direction by varying the magnet current, then back again by reversing the current. These two scans are combined to make a single "set" from which relative abundances are computed. The purpose of the double scan is to eliminate variations due to decreasing sample size or temperature drift. Because base line drift is small with a vibrating reed electrometer, and takes place over periods which are long compared to the time required for a single scan, it is not necessary to completely resolve the peaks or make corrections for lack of resolution as was the case when a D.C. electronic amplifier was used¹¹. If care is taken that the tail of one peak does not extend to the centre of an adjacent peak it is only necessary to determine the position at each end of the mass spectrum in order to draw in the base line to which peak heights are measured.

If a, b, c, ---- i, i' ---- c', b', a' are the measured heights from peak to base line of one set of the isotopes, A,B,C --- I, and if S = (a + a') + (b + b') + ---(i + i'), then the relative abundance of isotope A for this $set is <math>\frac{a + a'}{S} \times 100\%$. Corrections for identifiable contaminants are made at this point where necessary by deducting the appropriate amount from each sum (i + i') before computing S. A succession of a dozen or more sets constitutes a "run".

(ii) Analysis of Abundance Data

The analysis of the results to obtain reliable values of the abundances, and of the associated standard deviations is based on the assumption that the errors are random and that

the "set" values have a normal or Gaussian distribution about the mean. The main features of such a distribution are discussed briefly by H.J.J. Braddick 3^4 . Adopting the symbols of that reference, let N be the number of sets in a run, let v_i be the difference between a set abundance and the calculated mean for the run, and let u be the best estimate of the root mean square (r.m.s.) deviation of the set values about the true value of the abundance. Then

$$\mu = \sqrt{\frac{\sum v_i^2}{(N-1)^2}}$$

Further, the r.m.s. error of the arithmetic mean for the run of N sets is

$$\mu_{\rm N} = \sqrt{\frac{\Sigma v^2}{N(N-1)}} = \mu/\sqrt{N}$$

A knowledge of μ and $\mu_{\rm N}$ provides a basis on which either set or run abundances may be rejected. Using Chauvenet's oriterion for rejection ³⁵ as a guide, it is found that for the number of sets used (about 12) a particular value should be rejected if it is further than 2 μ (or 2 $\mu_{\rm N}$) from the mean computed using the remaining data. Once a value has been rejected, it is then necessary to adjust the abundance of the other isotopes to equal 100% less the accepted mean of the reject. This procedure is considered to be valid as long as the remaining isotopes have a total abundance exceeding 50%; if they do not the entire set or run is rejected. Values of μ calculated for platinum (Table II), unirradiated samarium (Table III), and unirradiated dysprosium (Table IX), and spot-checked for other samples, fall predominantly in the range 0.12 ± 0.04 percent, for which the value of $\mu_{\rm N}$ for ten sets is $0.04 \pm 0.01\%$. A check on $\mu_{\rm N}$ is provided by calculating $\mu_{\rm R}$, the r.m.s. value of the deviation of the run abundances from their mean. In general, $\mu_{\rm R}$ is nearly equal to $\mu_{\rm N}$, but there is a finite probability, increasing as the number of runs decreases, of a marked deviation. The practice followed here has been to use $\mu_{\rm N}$ as a basis of rejection where it can be calculated; in other cases the value of $\mu_{\rm R}$ is used, providing it is not less than 75% of the value of 0.04% quoted above for $\mu_{\rm N}$.

(111) <u>Contaminants</u>

Interference from contaminants can distort the mass spectrum and cause serious errors in the calculated percent abundances. For this reason, careful surveys were made preceding and during the analysis of each material in order to detect, identify and correct for contaminants.

The methods used can best be illustrated by a detailed description of the treatment of contaminants in the cases of unirradiated and irradiated samarium. These fall into two classes each having its own characteristics and requiring a different method of treatment. The first class, identified as rare earth contamination in the original

sample, occurs in both the irradiated and unirradiated samples and is most prominent in the oxide region (compare figures 6 and 9). They have in common a marked sensitivity to changes in temperature. Once the rare earth peaks have been identified they are studied to ascertain whether metal, oxide or dioxide ions are present which might interfere with a spectrum of the element being studied. The results for samarium metal ions were that interference did not occur, or was negligible. In particular neodymium and gadolinium, which were known to appear as the oxide, were so small in comparison to the other peaks present that their characteristic spectrum could not be detected. Of the peaks identified in figure 6, only that due to CeO was certain. This peak appeared regularly before reaching the samarium working range along with the peaks due to Ce and CeO2 ions. For the oxide ion spectra (figure 9) the neodymium and gadolinium oxide ions were easily identified. No other interfering ions were detected.

For the other contaminant peaks, of uncertain identity and origin, no correction is possible. They were tentatively identified as hydrocarbons because of their undulating pattern, high for odd masses, low for even masses, and their sensitivity to changes in temperature. In figure 7, the peaks at mass numbers 141, 142, 143, 145, 146, 151 and 153 fall in this group. Because they maintained about the same proportion to one another at all times, only one peak, that at mass 145, was regularly measured as a gauge of their importance.

An extensive series of tests were performed in an attempt to identify and reduce or eliminate these peaks. It was found that the peaks were due in part to the sample and did not occur in a run using a samarium sample supplied by Dr. H.E. Duckworth of this university. They were also missing or very small in runs with the irradiated samarium. However, as noted previously, chemical treatment was not successful in reducing this contamination. A similar spectrum from a clean V, i.e., one without a sample deposited on it, could be reduced or eliminated by focussing or by the use of specially shaped V's, suggesting that some of these ions were formed from the bombardment of evaporated atoms by stray electrons near the edges of the V.

V RESULTS

A - PERCENT RELATIVE ABUNDANCES

1. Naturally-occurring Platinum (Table II)

In an effort to improve the emitting properties of the source platinum black was electrodeposited on the concave surface of the V and heated to a white heat. Platinum ions were found to be emitted at temperatures somewhat above the platinum melting point from molybdenum V's treated in this manner. Ten satisfactory runs on the platinum spectrum were obtained including four measurements of the abundance of Pt^{192} . The abundance of Pt^{190} , which is present to only 0.01%, was not measured in these runs.

In these runs mercury was always present, making an appreciable contribution as mass 193. The necessary correction varied from 3% to 12% of the Pt¹⁹³ peaks, except on one occasion when it was 22%. No other contaminants were detected.

Figure h shows an "average" platinum spectrum (to scale) which is not completely resolved. The mercury correction, which was determined by a measurement of the Hg¹⁹⁹ peak, was 5% in this case. The fluctuations in the base line are an indication of the sensitivity of the amplifier. Since the smaller peaks were taken using the most sensitive shunt on the vibrating reed electrometer, which gives a recorder deflection of 2.8 cm. per millivolt appearing across the 5 x 10¹⁰ ohm resistor, it can be seen that the smallest peak (Pt¹⁹²) is produced by an ion current of 7 x 10⁻¹⁵ amperes, while the limit of detection is just over 10⁻¹⁵ amperes.


TABLE II

ABUNDANCES OF THE ISOTOPES OF PLATINUM

-	7		Mass No	0. 02	Platin	im Isoti	ope			
Run	192		194		195		198		198	
1	0.89	(0.4)	32.83	(.17)	34.01	(.18) ^R	25.20	(.09)	7.12	(.05)
2	-		32.80	(.15)	33.90	(.10)	25.19	(.09)	7.20	(.09)
3	-		32.79	(.17)	33.8	5 (.14)	25.30	(.08)	7.16	(.10)
<u>4</u> .	-		32.78	(.20)	33.79	(.20)	25.24	(.15)	7.29	(.07)
5	-		32.79	(.12)	33.76	(.17)	25.28	(.16)	7.27	(.09)
6	-		32.82	(.21)	33.89	(.18)	25.18	(.16)	7.21	(.12)
7	-		32.86	(.19)	33.74	(.23)	25.25	(.12)	7.39	(.10)
8	0.68	(.05)	32.84	(.13)	33.84	(.15)	25.20	(.15)	7.24	(.12)
9	0.90	(.05)	32.70	(.14)	33.74	(.11)	25.26	(.12)	7.27	(.08)
10	0.88	(.07)	32.85	(.12)	33.82	(.18)	25.15	(.10)	7.29	(.11)
Mean	0.89	9	32.82	1	33.81		25.22		7.24	
Std. Devi- ation	0.02		0.04		0.06		0.05		0.05	

Note:

Bracketed values are the calculated r.m.s. deviations of the set abundances.

R = rejected value as described on pp. 19-21. The remaining abundances for the run have been corrected.



When platinum V's were used with irradiated samarium large singly-charged platinum peaks were observed, as well as multiply-charged peaks up to the fifth power. It was found that Pt^{190} could be detected at temperatures just below the melting point. A run of ten sets was obtained from which the ratio of Pt^{192} to Pt^{190} was calculated to be $59 \stackrel{+}{=} 2.5$. Figure 5 shows 2 sets from this run. The most striking characteristic is the very large tail from Pt^{104} due to a marked increase in the broadening of the lines as the melting point is approached. The vibrating reed electrometer is again at its most sensitive setting for the smaller peaks.

2. <u>Naturally-occurring Samarium</u> (Tables III, IV)

Twenty-nine runs were made with two samples of unirradiated samarium using a platinum-on-tantalum (Pt-on-Ta) V source. Use of the first sample was discontinued after nine runs because of a partial disintegration of the V, although the sample showed no signs of running out. The life of the second sample was also indeterminate since it was still yielding peaks of satisfactory size and stability after twenty runs, even though the last three were made at a higher temperature than the earlier ones, with ion currents about five times as large.

A plot of all results on a time scale indicated that the hydrocarbon "background" had a detectable effect on the relative abundances for a peak of mass 145 exceeding 1% of all the samarium isotopes. No correction was required in these runs for rare earth contaminants. Figures 6 and 7 demonstrate the

different behaviour of the two groups of contaminants, as discussed previously. Table III lists the percent abundances for all accepted runs. The original run number is retained since it provides a convenient time index for the analysis of the change of abundances with time which follows.

Because of the variation of the abundances with time, attributable, as in the case of neodymium ¹¹, to mass fractionation during ion formation, it was necessary to fit the results by a least squares analysis. In this case the standard deviation used as a criterion for the rejection of a particular result (see pp. 19-21) was that given by the least squares analysis. After a rejection, the least squares fit to the other abundances had to be corrected to the adjusted values. All abundances were given equal weight except those of the last three runs. Because of their superior accuracy each of these abundances were given double weight - approximately in inverse proportion to their standard deviations, relative to the other results.

Figure 8 displays this data, as well as the least squares fit and its standard deviation for each isotope. Sm¹⁵⁰ is omitted because its near zero slope reduces the least squares fit to a simple determination of the mean. The results of the least squares analysis are shown in Table IV.





TABLE III

MASS SPECTROMETRIC RESULTS FOR UNIRRADIATED SAMARIUM

Sample	Run		Iso	topic Percent	t Abundances			The second s
NO.	NO.	Sm- 44	Sm	Small	Smiles	Smr So	Smise	Smise
1	N 374 500 7-00 9	$3.42(.07)^{R}$ 3.27(.08) 3.21(.08) 3.19(.10) 3.13(.05) 3.05(.07) 3.11(.05)	15.53(.22) ^R 15.20(.24) _R 15.52(.27) ^R 15.21(.21) 15.12(.10) 15.20(.13) 15.14(.05) _R 15.26(.08) ^R	11.60(.16) ^R 11.42(.14) 11.45(.17) 11.45(.07) 11.45(.11) 11.28(.08) 11.33(.06) 11.46(.08) ^R	13.85(.19) 13.90(.10) 13.91(.11) 13.88(.25) 13.94(.10) 13.76(.08) 13.83(.10) 13.88(.07)	7.37(.14) 7.38(.08) 7.32(.08) 7.36(.06) 7.43(.07) 7.41(.05) 7.37(.06) 7.34(.06)	26.62(.21) 26.52(.24) 26.60(.25) 26.57(.27) 26.62(.24) 26.66(.16) 26.86(.10) 26.71(.13)	22225(.28) 22.32(.14) 22.29(.23) 22.18(.30) 22.30(.29) 22.36(.15) 22.51(.09) 22.46(.28)
2	2 34 158 199 20	3.35 ^R 3.25 3.17 3.16 3.13(.03) 3.16(.04) 3.10(.04)	15.25 15.23 15.24 15.10 15.05(.05) 14.97(.03) 15.02(.05)	11.45 11.28 11.44 11.28 11.23(.05) 11.25(.03) 11.22(.04)	13.92 13.76 ^R 13.86 13.75 13.77(.04) 13.72(.05) 13.78(.04)	7.39 7.38 7.35 7.39 7.34(.05) 7.39(.04) 7.38(.04)	26.51 26.50 26.62 26.60 26.80(.07) 26.75(.05) 26.78(.04)	22.26 22.29 22.32 22.58 22.67(.07) 22.75(.05) 22.73(.05)

Note: Bracketed values are the calculated r.m.s. deviations of the set abundances.

R = rejected value as described on pp. 19-21, based on the least squares

fit of Table IV. The remaining abundances in the set have been corrected.



TABLE IV

TIME DEPENDENCE OF THE RELATIVE ABUNDANCES OF THE ISOTOPES

Isotope	Sm ¹⁴⁴	Sm ¹⁴⁷	Sm ¹⁴⁸	Sm ¹⁴⁹	Sm ^{1 50}	Sm ^{1 52}	Sm ¹⁵⁴
Percent Abundance - Time Mean	3.17 ^a	15.14	11.36	13.84	7.37	26.67	22.47
Standard Devi- ation	0.040	0.028	0.023	0.035	0.024	0.041	0.037
Time Rate of ^b Change	0047	0136	0135	0096	+.0003	+.0148	+.0266
Corrected C relative time Rate of Change	0019	00103	00073	00043	00013	+.00050	+.00110

OF NATURALLY-OCCURRING SAMARIUM

- A _____ Not including the correction for hydrocarbon contamination (p. 35)
- b the change in abundance from one run to the next higher run
- c the relative time rate of change is equal to the time rate of change divided by the mean abundance. The values given here have been corrected to give a total abundance of 100% for any run (p.35).

The result of the least squares analysis gives the slope of the line of best fit (time rate of change in Table IV) as well as the abundance and the standard deviation of points from the line. However, these slopes are not correlated so that the total abundance of all the isotopes will remain 100% for any run. To correct the slopes to give a properly normalized set of abundances for a run preceding or following the mean, the quantity (yi/n1) ; d, was calculated for each isotope and the mean value determined. Here y₁ is the time rate of change, or slope for the ith isotope, n, is its time mean abundance and d, is equal to the mass number of the isotope (A_i) less the atomic weight of samarium (150.4). The values of (yi/ni) based on the mean value of $y_1 + n_1 d_1$ are shown in the last line of Table IV (corrected relative time rate of change).

In the case of Sm¹⁴⁴ it is possible to estimate and correct for the effects of the hydrocarbon contamination. The results rejected because of the magnitude of the mass 145 peak were separated into two groups corresponding to

average abundances of 1.2% and 1.5% of mass 145. The average deviations of these two groups from the least squares value for the $Sm^{1.44}$ abundance, which had a mean contamination of 0.3% at mass 145, were +0.05% and + 0.15% respectively. For zero hydrocarbon contamination it is concluded that the abundance of $Sm^{1.44}$ should be reduced by 0.03[±]0.01%.



3. Neutron Irradiated Samarium (Tables V and VI)

Where the unirradiated samarium required only two samples each of which gave many runs, the analysis of the irradiated portion required many samples which gave only a few runs each. With the Pt-on-Ta V, the maximum number of runs obtained was five, while the commonest number was two. For the runs made with a platinum V (oxide ions analyzed) the V melted after only one run was obtained, with a single exception when the number was two. The results of these runs are shown in Table V, arranged in three groups for the purpose of analysis as described below. The abundance of the samarium oxide ions which were obtained from the platinum V have been corrected for the presence of 0.2% 0¹⁸. In addition, nineteen sets were taken of Sm150 and Sm151 using a higher temperature, to determine the abundance of Sm¹⁵¹. The mean value for the ratio Sm¹⁵⁰/Sm¹⁵¹ was 135-8.

Figure 9 shows a set of the metal ions obtained from the Pt-on-Ta V. The contaminant at mass 153 was identified as Eu¹⁵⁵ by its rare-earth type behaviour. Like the cerium mentioned in the preceding section, it appeared as a peak several centimeters high on the most sensitive shunt at a temperature somewhat below the working temperature for samarium. There was no accompanying peak

at 151 which would interfere with the Sm^{151} peak. Hydrocarbon contamination was usually absent. It is worth noting that when peaks did appear at mass 145 and 146, no accompanying peak could be detected at mass 149. The inset shows a part of one of the sets used to determine the abundance of Sm^{151} . The sensitivity for Sm^{144} and Sm^{151} in the inset are both equal to that of Figures 6 and 7.

The rare earth impurities which appear in the metal ion and oxide ion regions when a Pt V is used, are shown in Figure 10. Corrections are required where the neodymium and gadolinium oxide spectra overlap the samarium oxide. Each element displays its irradiated pattern, and corrections are applied on that basis to the results of each set. For neodymium the peaks at mass 161 and 162 corresponding to Hd^{1+60} and Hd^{140} were measured and the mean of the correction from the two ¹¹ were applied to Sm^{146} O, Sm^{146} O, and Sm^{150} O at masses 160, 164 and 166 respectively. The correction for Gd^{154} was based on the results for irradiated gadolinium reported below and the measured size of the Gd^{156} peak.

Since the number of runs per sample was so small a simpler method of analysis was employed than in the preceding section. The results for each sample ware divided on a time basis among three groups corresponding to the initial, intermediate and final relative abundances obtained over the duration of each sample's useful life. Samples from which



only one run was obtained were assigned to the intermediate group; if there were two runs, to the initial and final group; if three or more, they were divided symmetrically among the three groups.

The three groups are averaged separately as shown in Table V. The least squares analysis is based on the assumption that the three sets of abundances obtained for the initial, intermediate and final groups correspond to three well-defined equally-spaced times, each having a weight equal to the number of set abundances on which it was based. In the case of Sm¹⁴⁷ for example, the analysis is carried out for 7 values of 15.10% at time -1, 7 values of 14.97 at time 0, and 5 values of 14.98 at time +1. The result for time 0 (time mean abundance) is (15.02-0.044)-0.0644 t, where t is in the units used above and the negative coefficient indicates that the abundance is decreasing with time. The results of this analysis for all the isotopes are shown in Table VI. The standard deviations are the results of the least squares analysis except for Sm¹⁵² where this was zero. Here the standard deviation was taken arbitrarily as 0.045%, in line with the other values. The corrected relative time rate of change shown in the last line of the table is calculated in the same way as the corresponding values of table IV, except that a small shift has occurred in the atomic weight of the sample due to neutron capture, raising it to 150.6 from 150.4.

TABLE V

MASS SPECTROMETRIC RESULTS FOR IRRADIATED SAMARIUM

dama da la participación de la	least statistics grant		Parcent	Isotopic	abundan	ces
Sample	Sm	Sm	Sm ⁺	Smillo	Smille	Sm+++
	ISO SHUS	POR BAC	H SOMPT			
<u>a - 1 - 1 1</u>	W4 110110	TON DAO	AL DINGA LA			A CONTRACT OF A
I II VI VI VI IX	8.23 8.13 8.10 8.14 8.14 8.11	15.19 15.06 15.05 15.05 14.99	12.03 11.92 11.87 12.01 11.95 11.95	21.06 ^R 20.91 20.94 20.86 20.85	25.66 25.53 25.69 25.69 25.60	23.07 23.26 23.44 23.18 23.28 23.28 23.09
R	2441	15.15	11.93	20.95	25.70	23.17
Mean	2.13	15.10	11.95	20.92	25.72	23.17
R.M.S. Dev.	0.03	0.08	0.06	0.05	0.07	0.085
	5	7	7	6	6	7
b - IN	ERMEDIA	re or si	NGLE RUN	is for ea	CH SAMPL	
IV VII VII VIII	3.09 3.08 3.13 3.08	14.91 14.86 14.94 15.07 _R	11.85 11.84 11.83	20.99 _R 20.83 20.91 20.98	25.77 25.83 25.77 25.68	23.39 23.43 23.41 23.36 23.40
A B C D	3.11 3.11 3.10 3.10	15.14 15.06 14.92 15.05	11.83 11.89 11.86	20.90 20.90 21.01 20.87	25.71 25.72 25.73	23.38 23.36 _R 23.29
Mean	3.10	14.97	11.85	20.95	25.74	23.39
R.M.S. Dev.	0.016	0.085	0.02	0.05	0.05	0.03
Veryes	а	7	8	7	8	7
C - LAS	T RUNS	FOR EACH	SAMPLE	P		
I II III V VI VI IX	3.12 3.11 3.08 3.04 3.04 3.15 3.11	15.04 15.97 14.95 14.72 14.95 14.95 14.98 15.18	11.88 11.79 11.81 11.79 11.87 11.75 11.75 11.95	21.00 20.88 20.90 20.89 20.83 20.76 20.76	25.71 25.78 25.74 25.76 25.78 25.83 25.83 25.75	23.40 23.29 23.52 23.55 23.42 23.58 23.43
E		14.81	11.89	20.94	25.75	23.35
Joan	3.10	14.98	11.84	20.85	25.76	23.47
R.M.E. Dev	0.035	0.04	0.05	0.07	0.04	0.09
liged	7	5	8	7	8	6
Conception of the local division of the loca	A subscription of the local division of the	the second s				

Note: Samples designated I, II, III, etc. are for metal ions from Pt-on-Te V. Those designated A,B, etc. are for oxide ions from Pt V's (corrected for 0°). R = rejected values as described on pp. 19 and 21. The remaining abundances in the set have been corrected.

TABLE VI

TIME DEPENDENCE OF THE RELATIVE ABUNDANCES OF THE ISOTOPES OF IRRADIATED SAMARIUM

Isotope	Sm144	Sm ¹⁴⁷	Sm ¹⁴⁸	Sm ^{1 50}	Sm ^{1 52}	Sm ¹⁵⁶
Percent Abundance - Time Mean -	3.11	15.02	11.88	20.91	25.74	23.35 -
Standard Deviation	0.013	0.044	0.030	0.035	0.045	0.045
Time Rate ⁸ of Change	0144	0644	0587	0368	+.0200	+.150
Corrected b Relative Time Rate of Change	0113	00614	00492	00098	+.00246	+.00590

a - The change in abundance in going from the initial to intermediate, and intermediate to final groups

b - The relative time rate of change is equal to the time rate of change divided by the mean abundance. The values given here have been corrected to give a total abundance of 100% for any run (pp. 40).

4. Naturally-Occurring Gadolinium (Table VII)

Oxide ions were the only ones found with the gadolinium sample. These peaks were found to last for only one or two runs if an attempt was made to maintain large peaks, but more runs could be obtained when peaks were used of a size similar to those shown in Figure 11. The sensitivity here is the same as in the preceding samarium spectra.

As nearly as could be ascertained no contaminabts contributed to the gadolinium oxide peaks at the temperatures used. Certainly no peaks were detected at non-gadolinium positions. The results of the mass spectrometric analyses for gadolinium oxide ions are shown in table VII. Since no time dependence was evident the mean abundances were determined by the method described previously (pp. 19, 21). Because of the low ion yield obtained with these samples, no attempt was made to measure $\mathrm{Gd}^{1.82}$, which has an abundance of about 0.2%. The last line in Table VII shows the percent abundance after correcting for 0.2% 0¹⁰, and for the absence of $\mathrm{Gd}^{1.52}$ (i.e. total calculated abundances equals 99.8%, not 100%).



TABLE VII

			Per	cent Is	ctopic .	Abundan	ces
Sample	Run	Ed a pe	CCT PP	Gda	GGT BY	Gdabo	Gdabo
I	12	2.14	14.77R	20.32 ^R	15.78	24.79	21.87
II	34	2.17	14.73	20.59	15.75R 15.88	24.84	21.90 21.96 _R
III	5678910 111	2.10 2.09 2.07 2.22 2.09 2.20 2.20 2.12	14.70 14.70 14.70 14.75 14.76 14.93 14.80	20.60 20.61 20.52 20.64 20.65 20.64 20.65	15.78 15.82 15.71 15.69 15.60 15.75 15.77	24.87 25.15 24.99	21.71 21.93 21.93 21.73 21.90 21.87 21.55
lean Ah	12 ppd.	2.00	14.75	20.77	15.74	24.97	21.93
HOULI NU		C.20	44412	LV.UL	-J+1+		
R.M.S.	Dev.	0.043	0.031	0.024	0.044	0.062	0.040
Corroat	89 d. ^a	2.10	14.74	20.60	15.70	24.81	21.85

MASS SPECTROMETRIC RESULTS FOR THE OXIDE OF NATURALLY-OCCUPRING GADOLINIUM

a - Corrected for 0.2% 0¹⁸ and absence of 0.20% 0d¹⁵²

Note: R = rejected value as described on pp. 19, 21. The remaining abundances for the run have been corrected.

MASS SPECTROMETRIC RESULTS FOR THE OXIDE OF IRRADIATED GADOLINIUM

		Perc	ent Abu	ndances	
Sample	Run	Garae	Gd too	60- 00	Qdree
I	1	1.98	35-53	40.54 ^R	21.79
II	2	2.09 ^R	35.61	40.61	21.83
	3	2.16 ^R	35.25 ^R	40.62	21.93
III	4		35.68 ^R	40.65	21.90
	5		35.59	40.68	21.78
	6		35.57	40.72	21.76
	7	1.91	35.44	40.77	21.88
	8	2.00	35.45	40.70	21.84
	9	1.99	35.52	40.64	21.85
IV	10		35.51	40.71	21.78
v	11	1.88	35.50	40.74	21.89
	12	1.92	35.46	40.77	21.84
	13	1.92	35.47	40.77	21,97 ^R
Mean Abund. (oxide)		1.94	35.51	40.70	21,84
R.M.S.	Deva	0.047	0.055	0.059	0.054
Correct	ed e Abun	d. 1.94	35.50	40.61	21.75

a - Corrected for 0.2% 0¹⁸ and absence of Gd¹⁵² (0.2\%)

Note: R = rejected value as described on pp. 19, 21. The remaining abundances for the run have been corrected.

•

5. Irradiated Gadolinium (Table VIII)

The limits imposed by the low ion yield in the unirradiated gadolinium applied to this sample as well. A spectrum from one of the irradiated runs is shown on the right of Figure 11. Table VIII shows the percent abundance for each accepted run, calculated to give a total abundance of 100% for the four isotopes measured. The last line gives the values for the percent abundances for the metal ions (corrected for $0.2\% 0^{18}$) with the assumption that there is no contribution due to neutron capture by Gd^{152} (0.2% in unirradiated 0d) or any contaminant.

6. <u>Naturally-Occurring Dysprosium</u> (Table IX)

Nine satisfactory runs were obtained using Pt-on-Ta and Pt-on-Mo V's. Of these, one was of exide ions and the remainder were of metal ions. The contaminant peaks encountered in the metal ion runs appear in the unirradiated dysprosium spectrum of Figure 12. The peak at mass 159 can only be due to Nd¹⁴³O, Tb¹⁸⁹or. a hydrocarbon ion. Since no peak appears at mass 158 or lower, the first possibility is ruled out, and the last is extremely unlikely. The peak is therefore attributed



TABLE IN

MASS SPECTROMETRIC RESULTS FOR NATURALLY-OCCURRING DYSPROSIUM

			Percent	Isotopic	Abundance	3
Sample	Run	160	161	162	163	164
I	1	-	18.85	25.55	24.98	28.34
	5	2.23	18.87	25.44	24.98	28.47
II	3	2.24	18.91	25.55	24.87	28.35
III	4	2.23	18.79	25.60	24.95 0.10-11	28.34
	5	2.31	18.88	25.55	24.85	28.31 0.11-9
	6	2.28 0.09-6	18.87	25.56 0.10-12	24.97	28.32
IV	7	2.29 0.09-12	18.88 0.09#5	25.53 0.17-7	24.97 0.16-7	28.33 0.10-8
V	8	2.32 0.13-10	18.99 ⁿ 0.12-9	25.53 0.13-13	24.91 0.11-12	28.36 0.16-12
VI	9	2.33 006-9	18.86 0.12-10	25.54 0.09-13	24.96 0.14-10	28.29 0.11-8
Kean Al	hund.	2.28	18.87	25.55	24.96	28.33
Av. Run R.M.S.	Dev.	0.10	0.11	0.12	0.11	0.12
Av. No. Sets/Ru	an	7.2	8.7	10.7	9.6	8.9
R.N.S.	Dev.	0.037	0.036	0.036	0.037	0.040

NOTE+

Entries for each run include the isotopic abundance in the first line and the corresponding r.m.s. deviation and no. of set values: from which these are calculated in the second.

The R.M.S. deviation of the last line is equal to the average r.m.s. deviation par run divided by the average number of sets per run, i.e., $\mu_{\rm N} = \mu/N$ (see p. 20)

R = rejected value as described on pp 19, 21. The remaining abundances for the run have been corrected.

to Tb. A similar argument leads to the identification of the source of the small peak at the other end of the spectrum as Ho^{les}.

The results for 5 of the dysprosium isotopes are shown in Table IX. The other, two (Dy^{156}, Dy^{150}) were too small with respect to the background to be measured accurately. Their abundances are 0.05% and 0.09% respectively. Their presence was taken into account in determining the set relative abundances, i.e., the total abundance for the 5 isotopes was 99.85% rather than 100%. The deviations assigned to the final values of the percent abundances were calculated from the r.m.s. deviation of the individual "set" values, μ , and the average number of sets used per run, N, using the equation $\mu_{\rm N}$ = $\mu \sqrt{N}$ from p. 20. This was the value used in rejecting the run abundances, rather than the r.m.s. deviation of the run values from the final mean.

7. Irradiated Dysprosium (Table X)

The spectrum of metal ions from irradiated dysprosium is also shown in Figure 12. When the operating range for dysprosium is first reached, a peak appears at mass 165 which is initially larger than any in the dysprosium spectrum, but rapidly dwindles, and for the balance of the life of the sample is very small or undetectable. This peak is undoubtedly due to the metal ions of Ho¹⁰⁵, the product of neutron capture in Dy^{164} followed by beta decay of Dy^{105} .

TABLE X

MASS SPECTROMETRIC RESULTS FOR IRRADIATED DYSPROSIUM

			Percen	t Abunda	nces	
Sample	Run	160	161	162	163	164
I	1 2	2.26	14.79 14.82	27.28	25.11	10.56
II	34	2.28	14.71	27.38 ^m	25.09	10.65 R
III	56	2.25	14.75	27.28 27.42	24.99 ^R	10.64
IV V	7 8 9 10	2.39 2.26 2.27 2.25	14.67 14.80 14.73 14.70	27.29 27.30 27.33 27.31	25.20 25.08 25.05 25.21	10.68 10.63 10.49 10.63
VI	11 12 13 14	2.29 2.27 2.22 2.32	14.77 14.67 14.72 14.61	27.28 _R 27.22 27.31 27.31	25.03 25.12 25.11 25.08	10.65 10.65 10.64 10.56
Mean Isotopic Abundance 2.26			14.72	27.30	25.09	10.62
R.M.S. Deviation 0.0		0.025	0.037	0.020	0.036	0.036

Note: R = rejected value as described in pp. 19, 21. The remaining abundances for the run have been corrected. In Table X, listing the mass spectrometer results for irradiated dysprosium, the total abundance has been taken as 80% rather than the usual 100% since Dy¹⁶⁴ was known to have a cross section sufficiently large to give a loss of this magnitude.

8. Naturally-Occurring Ytterbium (Table XI)

Vat lower temperatures than the preceding elements. The spectrum was free of contamination with the occasional exception of a small peak at mass 169. Any element which might interfere with the ytterbium spectrum would have detectable peaks at other masses besides 169.

Figure 13 shows an ytterbium spectrum of a size typical of the sixteen runs accepted. The detector sensitivity was the same here as in the spectra shown for the preceding elements. Yb¹⁰⁰ with an abundance of 0.14%, was not large enough with respect to the background fluctuations to permit accurate measurements. No allowance for the presence of this isotope was made in calculating the run abundances of table XI. This correction is made in the last line.

8. Irradiated Ytterbium (Table XII)

The results for irradiated Yb are shown in Table XII. No contaminants were detected in these runs. A portion of a set taken with irradiated ytterbium is shown on the right side of Figure 13.



TABLE XI

MASS	SPECTROMETRIC	RESULTS	FOR	NATURALLY	-OCCUPRING
and the set		TTERBIUN	1		

	1	ndances					
Sample	Run	Ybivo	Ybaya	APTA	YDAYS	Xbere	YOTIS
I II IV V	12345678901123456	2.97 3.00 3.94 3.02 3.94 3.02 9.95 3.02 9.95 3.02 9.95 3.02 9.95 3.02 9.95 3.02 9.95 3.02 9.95 3.00 9.00 9.00 9.00 9.00 9.00 9.00 9.00	14.21 14.21 14.27 14.22 14.22 14.24 14.26 14.24 14.24 14.24 14.25 14.28 14.27 14.28 14.27 14.29 14.27 14.29 14.27	21.84 21.73 21.80 21.33 21.78 21.79 21.81 21.82 21.84 21.82 21.82 21.82 21.82 21.82 21.95 21.95 21.95 21.95	R 16.10 16.20 16.31 16.21 16.18 16.05 16.16 16.22 16.12 16.21 16.21 16.21 16.21 16.25 16.18 16.25 16.18	7 31.89 32.04 32.05	12.75 12.60 12.55 12.55 12.55 12.55 12.55 12.65 12.75 12.65 12.65 12.75 12.65 12.75 12.70 12.70 12.70
Nean Fer	rcent	3.00	44.25	21.81	16.10	32.03	12.71
R.N.S.		0.027	0.036	0.027	0.035	0.051	0.027
Correcto	DC DC	3.00	14.23	21.78	16.17	31.99	12.69

Note: a - corrected for presence of 0.14% Yb100

R - rejected value as described on pp. 19, 21. The remaining abundances have been corrected.

TABLE XII

MASS SPECTROMETRIC RESULES FOR INRADIATED YTTERBIUM

			Percen	t Isotos	re nomi-	ENGER	
Sample	Run	Yb170	76272	Yb172	Vb178	Yb ²⁷³	Yb176
I II	123	3.07R 3.17 3.11	13.94 13.94 13.94	22.43 22.34 22.34	16.20 ^R 16.16 16.17	31.82 ^R 31.59 31.60	12.85 13.14 12.86
III	4502	3.12 3.01 _R 3.17	13.90 _R 13.81 13.97 13.89	22.43 22.39 22.44 22.27	16.06 16.16 16.11 15.06	31.63 31.58 31.56 31.55	12.87 13.01 12.87 12.94
IV .	8910	3.07 3.08 3.08	14.04 ^R 13.87 _R 14.08	22.35 22.44 22.39	16 07 16 15 16 15	31.63 31.60 31.54	12.95 12.31 12.97
V VI VII VIII	112745567	2,99 3.03 3.00 3.11 3.03 3.04 8.04	13.96 13.94 13.83 13.95 14.13 14.05 13.96	72.40 22.27 22.20 22.42 22.42 22.42 22.42 22.42	10.25 16.12 16.13 16.07 16.07 16.14 16.15	31.59 31.59 31.59 31.59 31.58 31.68 31.68	12.92 13.04 12.95 12.93 12.73 12.87 12.87 12.87
Mean Isotop: Abundar		3.05	13.94	22.39	16.12	31.58	12.90
R.M.S. Deviation		0.043	0.035	0.035	0.039	0.035	0.040

Note: R = rejected value as discussed on pp 19, 21. The remaining abundances of the run have been corrected.

V B - DETERMINATION OF ISOTOPIC CAPTURE CROSS SECTIONS

1. INTRODUCTION

The use of the measured abundances in the equations derived in appendix A does not permit an unequivocal determination of the capture cross sections. Two additional data are required before such a determination is possible: a normalization of the abundances of the irradiated sample to allow for losses through the beta decay of capture products, and information on the integrated flux, since the equations are expressed in terms of the product of this quantity and the isotopic cross sections.

In the rare earth region, which contains many large isotopic cross sections, it is generally not possible to determine both the correct normalization and integrated flux. However, if the cross sections leading to loss of nuclides have been measured and found to give only small losses (of less than 1%), normalization is possible, If, in addition, the absorption cross section of the element, σ_{0} , has been determined quite accurately, the contributions of the individual isotopes given by the equation

 $.01 \ge n_i \sigma_i \phi t = \sigma_a \phi t$ can be used to determine the integrated flux, ϕt . The neutron absorption cross sections of the neodymium isotopes were determined in this way¹¹; a redetermination using a new value of $\sigma_a (Nd)^{36}$, 37, 38 is included in Appendix B. This method is not applicable to the elements under consideration here. In the case of both samarium and gadolinium the elemental absorption cross section is dominated by isotopic cross sections for which this experiment can set only a lower limit. For dysprosium, the cross section of Dy¹⁶⁴ reduces the amount of the sample by about 20%, with an error which would invalidate the experimental results if it provided the only criterion. For each of these elements, a special method is adopted which will be described in their separate sections.

Ytterbium appears to have available all the information necessary to determine both cross sections and integrated flux without additional information from other samples. However, such an abalysis leads to a value of ϕ t more than twice the value obtained in the neodymium analysis. For reasons which are advanced in the discussion, the integrated flux calculated from the neodymium results has been used in every case, including the ytterbium analysis.

The flux inside each sample will be less than the "unperturbed flux" which would have existed had there been no neutron absorber. It is necessary, therefore, to estimate the time variation of the ratio of the mean flux inside the sample to the "unperturbed" flux in order to obtain an "effective integrated flux" for use in the cross section equations. The details of these calculations are given in Appendix B. The analysis of the neodymium data gives an unperturbed integrated flux of 0.50[±] 0.037 neutrons per kilobarn.

2. SAMARIUM (Table XIII)

Because the unirradiated samples of samarium were not completely used up in the mass spectrometer analysis, it is doubtful that the time mean abundances of Tables IV and VI can be compared. In fact, the only abundances known to be comparable are those corresponding to zero time. In terms of the time scales of Tables IV and VI, these abundances correspond to -10 time units for unirradiated samarium, and -(1+1/2) units for the irradiated portion. The "zero time" abundances for unirradiated and irradiated samarium given in the first two lines of Table XIII were obtained from the time means of Tables IV and VI using the corrected relative time rates of change given in the last lines of those tables.

The effective integrated flux calculated for samarium in appendix B is $0.33^{\pm}0.04$ neutrons per kilobarn. This value corresponds to irradiation of a sample in which all the Sm¹⁴⁰ has been converted to Sm¹⁵⁰, so that for these calculations the abundance of Sm¹⁵⁰ is taken equal to 13.90% + 7.38%(N140 + N150).

The possibility of a large cross section for Sm^{158} , competing successfully with the natural beta decay to Eu^{158} , is suggested by the very large increment at Sm^{154} . Although this increment can be fitted to the known cross section for Sm^{154} without assuming any transfer from lower isotopes, the corresponding cross sections for other isotopes are much larger than found by other experimenters 1,2,16,39. This possibility (large Gise) means that the mass spectrometric data alone can only be used to set limits to the isotopic cross sections.

A normalization in which it is assumed that σ_{183} g << λ_{183} λ_{155} is the beta decay constant of 47 hr. Sm¹⁵⁰, will where give a set of isotopic cross sections which are the upper limits obtainable. Sm¹⁵⁶ is effectively isolated from the lighter isotopes, so that the only change possible is a loss of atoms by neutron capture. (The possibility of contaminants affecting the abundance of irradiated Sm¹⁸⁴ was treated in the section dealing with the mass spectrometric results). The "best value" for Oise is quoted as 5.571.1 barns³⁸. For such a small cross section the percentage loss due to neutron capture is given by the equation $\triangle n_{156} = n_{156} \sigma_{156} \sigma_{156}$. Inserting the numerical values for n_{154} , σ_{154} and gt gives $\Delta n_{154} = 0.04\%$, whence $n'_{154} = n_{154} - \Delta n_{156} \pm 22.19\%$. The normalization factor, F, giving the upper limits of the isotopic cross sections is $\mathbf{F} = \frac{22.19 + 0.037}{23.14 + 0.045} = 0.9589 + 0.0025$. The calculations based on this normalization are given in Table XIII (case II).

The lower limit will be reached when the smallest cross section approaches zero. As the normalization factor is increased towards unity, σ_{140} rapidly approaches zero. Since the standard deviations associated with the cross sections are relatively large, a normalization which gives a value for σ_{140} equal to the negative of the standard deviation will be a reasonable assumption in setting lower limits to the isotopic oross sections. The standard deviation associated with sums and differences involving n_{147} , n_{147}/F , n_{148} and n_{148}/F

is 0.07. The normalization factor is then given by the equation $n_{1,7} - F(\frac{n^{*}1!.7}{F}) + n_{1,8} - F(\frac{n^{*}1!.6}{F}) + 0.07 = 0.$ Solving gives $F = 0.9835 \pm 0.0026$. The cross section determinations for this normalization factor are given in Table XIII (case I).

The cross sections were determined using the approximate equations (h) with the exception of Sm^{151} . Checks on the values of 6_{152} using the mathematically-exact logarithmic formula gave substantially the same values. In the case of 6_{151} , $6_{151} & 1^n \left[n_{150} & 6_{150} - 6_{151} \right] \right]$ $- 1^n \left[n^*_{151} + n^*_{150} & 6_{150} - 6_{151} \right] \right]$. Taking σ_{151} to be 12 kb¹⁶ and $\delta t = 0.33$ n/kb., $n_{150} & 6_{150} - 6_{151} \right] = e^{4} \approx 50$ $n^*_{151} + n^*_{150} & 6_{150} - 6_{151} \right]$ Rearranging and taking $n_{150} \approx 1.03$ n'150 $\frac{\sigma_{150}}{\sigma_{151} - \sigma_{150}} = \frac{50n^*_{151} - n_{150}}{50n^*_{151} - n_{150}} = 1.02 \frac{n^*_{151}}{n^*_{150}} = 1.02/R$ where R is the measured value of the ratio n^*_{150}/n^*_{151} , equal to 135 ± 3. In this case $\sigma_{150} = \sigma_{150}(n - 1)$ within 1%.

An estimate can be made of the capture cross section of Sm^{153} corresponding to case I of Table XIII, since we know the amount of samarium lost through neutron capture by Sm^{152} , the amount lost by the same process in Sm^{154} and the net gain in Sm^{154} . Sm^{153} has a half-life of 17 hours, corresponding to a beta decay constant, λ_{153} , of 4.1 x 10⁻⁶ sec.⁻¹. Since the

Quantity	Sm ¹⁴⁴	Sm ^{1 47}	Sm ¹⁴⁸	Sm ^{1 50}	Sm ¹⁵¹	Sm ¹⁸²	Sm ^{1 54}
n (%) ^a n'/F(%) ^a	3.20 (.040) 3.16	15.30 (.030) 15.16	11.44 (.035) 11.96	21.28 (.043) 20.94		26.55 (.041) 25.65	22.23 (.037) 23.14
I - LOWER L	(.015) MITS -	(.044) F = 0.98	(.030) 985 b	(.035)	(.009)	(.046)	(.045)
n' (%) ^b	3.12	14.99	11.82	20.70	0.153	25.36	22.87
∆n=n-n/	0.08 (0.43)	0.31 (.056)	-0.38	0.58	-0.15	(0.73)	-0.64
$\sum (\Delta n)^{c}$	0.08 (.043)	0.31 (.056)	-0.07	0.58 (.07)	0.43	1.62 (.102)	-
σ (Barns) e	.125 76-45	.0205 62-12	006 -18-18	.0276 84-10	-11,300	.0624 190-12	5.5 ^g
IL - UPPER	LIMITS -	F = 0.9	589 b		-1400		
n' (%) B	3.03	14.54	11.46	20.08	0.149	24.60	22.19
∆n=n-n'	0.17	0.76	-0.02	1.20	-0.15	1.95	0.0 4 ¹
$\Sigma(\Delta n)^{c}$	0.17	0.76	0.74	1.94	1.79	3.74	0.04
oøt d	.055	.051	.065	.094	-	.146	-
o (Barns)	167-50	155-12	197-20	205-15	- 2400 ¹	440±15	5.5

TABLE XIII - ISOTOPIC CROSS SECTION CALCULATIONS FOR SAMARIUM

- a Relative abundances corresponding to "time zero". $\sum n = 100\%$, $\sum (n'/F) = 100.165\%$. Bracketed values are standard deviations
- b The determination of the normalization factors is discussed in the text (pp 59,60).
- c The summation $\sum_{n=1}^{\infty} (\Delta n)$ is broken after Sm¹⁴⁴, ¹⁴⁸ in case I, and after Sm¹⁴⁴, ¹³² in case II.
- d Values of oft obtained using the approximate equations
- e Values of o are calculated from the corresponding value of of taking ft = 0.33-0.04 n./kb. (appendix B), except where noted.
- f 0151 = (134 =8) 0150.
- g $\sigma_{154} = 5.5 \pm 1.1$ barns (reference 38). This cross section will produce a change of 0.05% in the amount of Sm¹⁵⁴. Values of σ_{155} based on this figure are: 60-10 kb. for Case I, 0 b. for Case II.
amount of Sm¹⁵⁵ quickly comes into equilibrium due to this short half-life

$$\frac{\sigma_{153}}{\lambda_{153}} \approx \frac{\text{amount of samarium transfer Sm}^{154}}{\text{amount of samarium decaying to Eu155}}$$
$$\approx \frac{-\Delta n_{154} + n_{154}\sigma_{154}gt}{\sum_{150}^{150} \Delta n_{1}} = (-\Delta n_{154} + n_{154}\sigma_{154}gt)$$

Taking $\emptyset = 5 \times 10^{15}$ neutrons/cm²/sec, and sub stituting for the other quantities gives a value for σ_{153} of 60,000 barns.

3. GADOLINIUM (Table XIV)

The effective integrated flux in the gadolinium sample is estimated in appendix B to be 0.3170.04 neutrons per kilobarn, on the basis of an unpacked oxide density of 1.470.1 gm/ cc. This is equivalent to bombardment by the unperturbed flux for a shortened time interval in which the initial concentration of Gd¹⁵⁵ and Gd¹⁵⁷ have already been destroyed and converted to Gd¹⁵⁶ and Gd¹⁵⁸ respectively. In the determination of cross sections, therefore, the initial abundances assigned to Gd¹⁵⁶ and Gd¹⁵⁸ are (n₁₅₅ + n₁₅₆) and (n₁₅₇ + n₁₅₆) respectively.

TABLE XIV	- ISOTOPIC C	ROSS SECTION	CALCULATIONS	FOR GADOLINIUM
Quantity	Gd	0d 4 50	Gdabe	Gdree
$n(5) \stackrel{a}{\rightarrow} \\ n'(5) \stackrel{b}{\rightarrow} \\ \Delta n = n - n' \\ \Sigma (\Delta n) \stackrel{c}{\rightarrow} \\ \sigma \not > t \stackrel{c}{\rightarrow} $	2.10(.043) 1.94(.047) 0.16(.064) 0.16(.064) .079	35.34(.039) 35.48(.058) -0.14(.070) 0.02(.095) .00056	40.51(.076) 40.59(.054) -0.08(.096) -0.06(.136) 0015	21.85(.040) 21.74(.054) 0.11 (.067) 0.11 (.067) .0050
o (Barns) ^d	2567120	3711	-3714	17∓11

(References on next page.)

- a For purposes of these calculations, nise is equal to the sum of the relative abundances of Gd¹⁵⁵ and Gd¹⁵⁶ from Table VII; nise is obtained in a similar manner.
- b Obtained from Table VIII using an normalization factor of 0.9994.
- c The summation is broken following Gd¹⁵⁸.
- d The cross sections were obtained using the approximate equations for a value of gt = 0.31+0.04 heutrons/kilobarn.

To normalize the percent relative abundances of the irradiated gadolinium for comparison with those of the unirradiated element, it is necessary to estimate the losses due to beta decay of capture products. In the mass range measured here such losses occur only for the isotopes of mass 158 and 160. Taking these cross sections to be 4.5 barns and 1.0 barn respectively on the basis of available information the total loss from these two isotopes is 0.06%, giving a normalization factor of 0.9994.

The details of the cross section determination and the results are given in Table XIV. Since Δn 's are all less than 10% of the corresponding n's, the approximate equations are valid throughout. The bracketed values are standard deviations, either measured or calculated by the standard statistical method for estimating errors of sums and differences.

4. DYSPROSIUM

In the case of dysprosium the sample cross section changes by a factor of about two over the irradiation period. The effective integrated flux is0.4770.04 neutrons per a kilobarn (Appendix B) using an oxide density of 1.470.1 gms. per c.c. and assuming that the whole sample is irradiated during the entire period with a depressed flux equal to the mean of the initial and final values.

Since the neutron capture cross section of dysprosium has been measured, the method used to determine the integrated flux in the case of neodymium (Appendix B) may be reversed an order to determine the capture cross section of the isotopes. The method is as follows: a normalization factor is chosen, and the corresponding values of $\Delta n = n-n^{\circ}$ and $\sigma_0 \not{\sigma} \ddagger \frac{1}{n_0 \ast} \sum_{i=1}^{8} (\Delta n_i)^{\circ}$ are calculated. To be acceptable the Aproducts $n_0 \sigma_0 \not{\sigma}$ must fall within the limits of the product $\sigma_0 (Dy) \cdot \not{\sigma} t$ (0.517∓0.083) where $\sigma_c (Dy) = 1100$ ∓150 barns and $\not{\sigma} t = 0.47$ ∓ 0.04 n./kb. as noted above. As a final check the "correct" values of $\sigma_{1e1} \not{\sigma} t$ and $\sigma_{1ee} \not{\sigma} t$ were determined using the logarithmic equations of Appendix A.

Table XV shows two sets of calculations based on this method, one corresponding to the upper limit of $\sigma_{\overline{m}}(Dy)$ \mathscr{I} t, the other to the lower limit. To determine the cross sections corresponding to the isotopic values of \mathscr{O} , it is necessary to estimate a range of \mathscr{I} t appropriate to the limits chosen for $\sigma_{\overline{c}}(Dy)$ \mathscr{I} t. If the error in this product were due entirely to either $\sigma_{\overline{c}}(Dy)$ or \mathscr{I} t, then \mathscr{I} t would have its proper error (zero in the first case). On the other hand, if the percentage errors of both $\sigma_{\overline{c}}(Dy)$ and \mathscr{I} t were the same, then the limits of $\sigma_{\overline{c}}(Dy)$ \mathscr{I} t would correspond to most probable values for the integrated flux of approximately $(\text{@t})_n \neq 2$ (error in @t) where $(\text{@t})_n$ refers to the nominal value for @t, 0.47 neutrons/kilobarn. For our purposes, this last estimate will suffice. For case I, corresponding to the lower limit for $\sigma_c(Dy)\text{@t}$, a value of $\text{@t} = 0.47 - \frac{1}{2}(0.04)$ n/kb is used; for case II, the value used is $\text{@t} = 0.47 + \frac{1}{2}(0.04) = 0.498$.

TABLE XV - IS	SOTOPIC CROSS	SECTION	CALCULATI	LONS FOR I	YSPROSIUM
Quantity	Dyleo	Dy101	<u>Dy</u> 102	Dyles	Dy106
I - o _c (Dy)Øt	= 0.435 F =	• 0.792			
n a	2.28	18.84	25,52	24.93	28.29
n' b	2.24	14.57	27.03	24.84	10.51
∆n=n-n*	0.04	4.27	-1.51	0.09	17.78
$\sum (\Delta n)$	0.04	4.31	2.80	2.89	20.67
oøt ^c	.018	.258	.107	.116	1.065
σ (barns) ^C	41	584	242	262	2410
olog (barns)	1	588			2625
II - $\sigma_{c}(Dy) \emptyset t$	= 0.600 F	= 0.748			
n a	2.28	18.84	25.52	24.93	28.29
nt b	2.11	13.76	25.53	23.46	9.93
∆n=n-n*	0.17	5.08	-0.01	1.47	18.36
∑ (∆n)	0.17	5.25	5.24	6.71	25.07
opt ^c	.077	.322	.205	.277	1.312
σ (barns) ^C	155	647	412	556	2635
o _{log} (barns) ^d		653			2965
$\sigma (Average)^{e}$ $\sigma (barns)^{f}$ (for $\sigma_{c}(Dy) \emptyset t =$	100760 100740 0.517)	620735 62079	325∓90 330∓7	410∓150 420∓8	2800∓170 2810∓15

(References on next page.)

- a = 0.9986 of abundances of Table IX to allow for 0.11% Dy156, by153. b = Abundances of Table X are equivalent to F = 0.30.
- c These values are obtained using the approximate equations and $\mathscr{S}t$ equal to 0.1.7 \neq 1 x (the error in 0t) i.e., for case I $\mathscr{S}t = 0.1.1.2$, for case II $\mathscr{S}t = 0.1.93$.
- d Values of σ_{\log} are calculated using the logarithmic equation of Appendix A.
- e The "rounded-off" average of the values for cases I and II
- f The cross sections obtained corresponding to \$\mathcal{G}_c(Dy)\$\textsf{t}\$ = 1.10 x 0.1? = 0.51? (F = 0.7695). Errors include only mass spectrometric deviations.

The second to last line of the table gives the values of the isotopic cross sections which are the means of the values obtained in cases I and II. The errors assigned are equal to the difference between the mean and the corresponding case I or case II value. The last line shows the corresponding cross sections obtained when $G_{\rm c}(\rm Dy)$ %t takes its median value of 0.517. The normalization factor in this case is 0.7695, and the value used for the integrated flux in determining the cross sections is the median value 0.17 neutrons per kilobarn. The errors quoted in this case include only the cumulative mass spectrometric errors.

5. YTTERBIUM

Two alternative bases for normalization are available for ytterbium, one corresponding to upper limits on the cross sections of the stable isotopes under study, the other to lower limits. Table XVI gives the results for both alternatives. In case I (upper limits) it is assumed that no transfer of atoms from Yb¹⁷⁴ to Yb¹⁷⁶ occurs (i.e. σ_{175} % << λ_{175} , the beta-decay constant of Yb¹⁷⁶). It is then possible to use the equation $\Delta_{n176} = n_{176}\sigma_{176}$ % to give the normalization, since σ_{176} is known to be small (5.571.0 barns³⁸). Using the value of %t from appendix B (0.5070.04 n/kb), a value is obtained for Δn of 0.03570.007%. The normalization factor is then given by the equation

$$F = \frac{n_{176} - \Delta n_{176}}{(n'_{176}/F)} = 0.981 + .004$$

Here n'176/F refers to the relative abundance of the irradiated isotope from Table XII, n170 to the corresponding abundance in Table XI. The error assigned to F is predominantly due to the mass spectrometric errors in these quantities $(n_{176}, n'_{170}/F)$. The contributions of the ytterbium isotopes of mass 170 to 176 to the ytterbium capture cross section $(\sigma_{c}(Yb))$ in a pile neutron flux are also included. Their total contribution is 7278 barns.

To determine the lower limits consistent with the mass spectrometric data a normalization factor is chosen which in the case of one particular isotope, will give a value of off equal to the negative of the cumulative error assigned to that quantity. The first isotope to reach this limit is Yb^{172} . The corresponding normalization factor is 0.993. Such a lower limit implies the transfer to Yb^{176} of some of the Yb^{174} atoms which were transmitted by neutron capture to Yb^{175} . The neutron capture cross section of Yb^{175} which will give the measured transfer is given by the equation

 $\frac{\sigma_{1760}}{\lambda_{175}} = \frac{n_{176}\sigma_{1760}t - \Delta n_{176}}{\sum_{170}(\Delta n_{1}) - (n_{176}\sigma_{1760}t - \Delta n_{176})}$

The beta decay constant, λ_{175} , corresponding to a half-life of 101 hours is 1.9 x 10⁻⁶ sec.⁻¹. The value of σ_{175} for case II is therefore 973 kilobarns. The isotopic contributions shown in the last line give a total of 2973 barns towards the capture cross section of ytterbium.

	PADLE XVI - 1	ISOTOPIC CROS	SS SECTION C	LCULATIONS 1	OR HTTPHERIDICUL	
Quantity	$\frac{117S - F = 0}{Yb^{17S}}$	98170.04 Yb	Tb172	Yb178	YD174	Yb178
n	3.00 (.027)	14.23(.036)	21.78(.027)	16.17(.035)	32.03(.051)	12.69(.027)
n* ^b	2.99 (.043)	13.68(.065)	21.96(.094)	15.81(.074)	30.98(.125)	12.655 ^c
∆n=n-n *	0.01 (.05)	0.55 (.074)	-0.18(.098)	0.36 (.082)	1.05 (.135)	0.035 ^c
Σ(Δ n)	0.01 (.05)	0.56 (.090)	0.38(.133)	0.74 (.16)	1.79 (.20)	0.035 ^c
opt e	.0033	.0401	.0174	.0463	.0568	
σ (barns) ^e	7∓37	80715	35+15	93725	114∓20	5.5 - 1.1 °
.01 no 1	1.2	11	873	15-4	37∓6	0.7 - 0.1 ^c
II - LOWER L	INITS - F = 1	0.993				6
na	3.00 (.027)	14.23(.036)	21.78(.027)	16.17(.035)	32.03(.051)	12.69(.027)
n ^{t b}	3.03 (.043)	13.84(.035)	22.23(.035)	16.01(.039)	31.36(.035)	12.81(.040)
∆n=n-n *	-0.03(.05)	0.39(.050)	-0.45(.044)	0.16(.052)	0.67(.062)	-0.12(.048)
Σ(△n) ^d	-0.03 (.05)	0.39(.054)	-0.06(.07)	0.16(.053)	0.83(.082)	
opt e	01	.0278	0027	.0099	.0262	
σ (barns) ^e	-20737	5679	-12714	2077	5276	5.571.0 °
.01 no f	1	871	0	3+1	17∓2	1

(References on next page.)

.

- a The abundances, with their errors, taken from Table XI.
- b The abundances of Table XII are equivalent to n'/F.
- c Normalization is based on a cross section of $5.5\overline{+}1.0$ barns for Yb¹⁷⁶, giving a contribution to Δn_{176} of 0.035%.
- d The summation does not carry over negative values. The error however, is carried over; it is equal to the upper limit of the preceding sum.
- e Both oft and o are calculated using the approximate equations with ft = 0.5040.04 n/kb. Errors include mass spectrometric deviations and the uncertainty in ft
- f The isotopic contributions to $\sigma_{\alpha}(Yb)$.

VII - DISCUSSION

A - ABSOLUTE ABUNDANCES OF THE ISOTOPES OF NATURALLY-OCCURING ELEMENTS

The use of a mass spectrometer to measure the neutron capture cross sections of stable isotopes requires reliable determinations of relative isotopic abundances. To this end considerable care was taken in section V A in determining the standard deviations. It should be remembered, however, that these standard deviations are a measure of the reproducibility of results under closely-controlled experimental conditions and are not an indication of the differences between the measured abundances and the true, or absolute, abundances of the isotopic constituents of the sample.

The results obtained in this investigation for the unirradiated naturally-occurring elements are compared in Tables XVII to XXI with measurements which have been reported previously in the literature. This comparison permits an estimate of the absolute abundances of the isotopes, and of the deviations to be expected in a single mass spectrometric analysis defined by a unique set of experimental conditions. In addition, it provides a reference which is useful in detecting any errors in the relative abundance determined in this investigation which might occur due to the presence of an unsuspected contaminant.

Differences between reported relative abundances fall into two categories. The first type, which is systematic, is a function of the mass displacement of the isotope from the atomic weight of the element. This type of variation has already been noted, both in the work on neodymium ¹¹ and in the case of samarium (section V A, 2, 3). It has been attributed to mass-dependent fractionation in the production of ions at the source, and appears to depend on the nature of the emitting surface. The second type of difference is a random variation due not only to the limiting accuracy for the mass spectrometers used, but also possibly to the presence of contaminants.

In order to sort out the two categories of differences, the following method of analysis was used: for a particular element, the mean abundance for each isotope was calculated by simple averaging of all values reported in the table; the ratio of an individual measurement over the corresponding mean was then calculated and the results plotted against isotopic mass. In general, the results reported by one laboratory will fall about a straight line of slope either greater or less than unity. Difference in slope, then, will give an indication of the mass-dependent differences due to fractionation while scatter about this line will provide an indication of the random mass spectrometric scatter. In addition, certain large deviations from these lines were noted. The individual abundances so noted were given reduced weights in determining the absolute abundances given in the last line of each table. The results of this analysis are briefly discussed below, by elements.

<u>Platinum (Table XVII)</u> - In the case of platinum, there are no measurements of the isotopic abundances having accuracy comparable

with the present work. It is not possible, therefore, to estimate the absolute abundances for the isotopes of platinum. The only other analysis reported ⁴¹ is in satisfactory agreement with our results. In the case of Pt^{190} , first detected by Dr. H.E. Duckworth and his co-workers ⁴², the ratio between the abundance of this isotope and that of Pt^{102} reported by W.T. Leland ⁴³ is in satisfactory agreement with the present measurement. In vidw of the greater resolution obtained in the former work (as judged by a sample spectrum included in the report) it is probable that the ratio determined there is more nearly correct. A value for this ratio of 6372 is recommended.

<u>Samarium (Table XVIII)</u> - Mass-dependent differences between the sets of isotopic abundances reported for samarium are very pronounced. The Mattauch-Scheld values ⁴⁴ correspond to an extrapolation from the abundances of Table IV corresponding to several tens of runs. The abundances reported by Hibbs ⁴⁵ occupy an intermediate position, roughly corresponding to the last runs of Table III, while the work of Inghram, et al. ⁴⁶, closely parallels the abundances determined here.

One marked deviation was noted. The value reported by Hibbs for Sm^{144} (2.87%) was much below the other values, although for the remaining isotopes Hibbs' values lay close to the mean. In the determination of the absolute abundances this value was replaced by 3.05%. The absolute abundances were determined by assigning. weights of 0, 1/2, 1 and 1 to the sets of abundances reported in lines 1 - 4 respectively.

<u>Gadolinium (Table XIX)</u> - The three sets of abundances for the gadolinium isotopes show mass dependent deviations much less marked than those for samarium. In calculating absolute abundances, two values were given weights of 1/2 because of a marked deviation from the line corresponding to its set. These were the 2.10 value for Gd^{154} and the 20.36 value for Gd^{159} . <u>Dysprosium (Table XX)</u> - Mass dependent deviations between the three sets reported were similar to those for gadolinium. In calculating absolute abundances, the value 2.36% for Dy^{160} was given a weight of 1/3. (Its percentage deviation was greater by a factor of two than the gadolinium values given reduced weights of 1/2.)

<u>Ytterbium (Table XXI)</u> - Mass dependent deviations were less marked than in the other elements discussed above. The 31.77% value for Yb¹⁷⁴ was given a weight of only 1/3 because of a marked percentage deviation.

One result of the preceding analysis will be of particular interest in the next section, namely, that there is no evidence that an unidentified contaminant is contributing to the relative isotopic abundance values determined in this investigation.

TABLE XVII - MEASUREMENTS OF THE ISOTOPIC ABUNDANCES OF NATURALLY-OCCURRING PLATINUM

	Isotopic Mass No.							
Reference	190	192	194	195	196	198		
Inghram, Hayden ^a		0.78	32.8	33.7	25.4	7.23		
Duckworth, Black, b Woodcock	.006							
Leland C	.012							
This work d	.015	0.89	32.82	33.81	25.22	7.24		

a - See reference 41

- c See reference 43. This value is based on 0.78 for the abundance of Pt^{192} . The corresponding value of the ratio of Pt^{192}/Pt^{190} is 65.
- d These abundances are taken from Table II. The value for the abundance of Pt¹⁹⁰ corresponds to a value for the ratio Pt¹⁹²/Pt¹⁹⁰ of 59, using 0.89 for the abundance of Pt¹⁹².

b - See reference 42

Reference		Isotopże	Mass No.				
	144	147	148	149	150	152	154
Mattauch, a							
Scheld	2.95	14.62	10.97	13.56	7.27	27.34	23.29
Hibbs b	2.87	14.94	11.24	13.85	7.36	26.90	22.84
Inghram, Hayden C							
Hess	3.16	15.07	11.27	13.84	7.37	26.68	22.48
This work d	3.14	15.14	11.36	13.84	7.37	26.68	22.48
Recommended Absolute Abundances	3.13 + .05	15.07 + .09	11.30 + .06	13.84 + .05	7•39 + •03	26.70 + .11	22.57 + .15

TABLE XVIII - MEASUREMENTS OF THE ISOTOPIC ABUNDANCES OF NATURALLY-OCCURRING SAMARIUM

a - See reference 44

b - See reference 45

c - See reference 46

d - The time mean abundances from Table IV

Isotopic Mass No.											
Reference	152	154	155	156	157	158	160				
Leland a	0.20	2.16	14.68	20.36	15.64	24.96	22.01				
Hesa b	0.20	2.15	14.78	20.59	15.71	24.78	21.79				
This work ^C		2.10(.04)	14.74	20.60	15.70	24.81	21.85				
Recommended Absolute Abund.	0.20	2.14 7.03	14.73 + .05	20.54 + .11	15.68 + .04	24.84 ∓.09	21.87 + .11				

TABLE XIX - MEASUREMENTS OF THE ISOTOPIC ABUNDANCES OF NATURALLY-OCCURRING GADOLINIUM

a - See reference 47

b - See reference 48

c - The values are taken from the last line of Table VII

Isotopic Mass No.										
References	156	158	160	161	162	163	164			
Leland a	.064	.105	2.36	18.73	25.36	24.91	28.47			
Inghram, Hayden b and Hess	.052	.090	2.29	18.88	25.52	24.97	28.18			
This work ^c			2.28(.04)	18.84	25.52	24.93	28.29			
Recommended Absolute Abund.	.055	.095	2.30 +.03	18.82 + .08	25.48 + .09	24.94 + .04	28.31 + .14			

TABLE XX - MEASUREMENTS OF THE ISOTOPIC ABUNDANCES OF NATURALLY-OCCURRING DYSPROSIUM

a - See reference 47

b - See reference 49

c - These abundances are obtained from the values of Table IX, making an allowance of 0.14 for Dy^{150,150}

Isotopic Mass No.										
Reference	168	<u>170</u>	171	172	173	174	176			
Leland ^a	0.13	3.03	14.27	21.77	16.08	31.91	12.80			
Haydan, Hess b Inghram	0.14	3.03	14.34	21.88	16,18	31.77	12.65			
This work ^C	-46- 200	3.00	14.23	21.78	16.17	31.99	12.69			
Recommended Absolute Abund.	0.13	3.02	14.28 + .06	21.81 +.06	16.14 + .06	31.92 + .08	12.70 + .07			

TABLE XXI - MEASUREMENTS OF THE ISOTOPIC ABUNDANCES OF NATURALLY-OCCURRING YTTERBIUM

- a See reference 47
- b See reference 50
- c Abundances from Table XI

VII B ISOTOPIC NEUTRON CAPTURE CROSS SECTIONS

In Tables XXII to XXV previous measurements on isotopic neutron capture cross sections are listed and compared with the values calculated in section VI B (Tables XIII to XVI). An attempt is made to narrow the limits for samarium and ytterbium, and consideration is given to divergences between these results and those reported previously.

1. Samarium (Table XXII)

A comparison with results obtained by other investigators for the cross sections of Sm^{151} and Sm^{152} suggests that the lower limits of Table XIII are much nearer the true cross sections than are the upper limits. The cross sections presented in the last line of Table XXII correspond to a normalization factor which is somewhat smaller than the one giving the lower limits, since the latter cross sections represent an absolute minimum consistent with the mass spectrometer data. The upper limit is taken into account by a partial error equal to one quarter of the difference between the value appearing in Table XXII and the upper limit for the same isotope from Table XIII, i.e., it is assumed that there is a probability of only h x 10⁻⁵ that the true cross section will exceed the upper limit. For each isotope the assigned error is equal to the square root of the sum of the squares of this partial error and the larger of the experimental errors for that isotope found in Table XIII.

Several discrepancies are noted in Table XXII. In the case of Sm^{152} it is possible that the spread between the three results listed could be accounted for by a large contribution due to the resonance capture of epithermal neutrons.* Sm^{152} has one prominent resonance 38

"See Page 84 for a more detailed discussion of this effect.

which will give a contribution of the correct magnitude for a change in the spithermal flux equal to about 3% of the total neutron flux.

The value obtained by Inghram, Hayden and Hess for 6151, viz. 7200 barns, also lies well below our lower limits. Since their value for 6149, which is closely related to 6151, lies well below the integral for a pile neutron spectrum over the large resonance at 0.09 ev 38 (about 60,000 barns) it seems likely that both their cross sections should be multiplied by a factor of h/3.

The last discrepancy occurs at Sm¹¹⁴. The 1.0 \mp 0.3 barn value for G_{1144} is based on a report ¹¹⁴ which gives the mass spectrometric analyses before and after irradiation in NRX of a sample of samarium enriched in Sm¹¹⁴ (71.02%). Sm¹¹⁵ was detected in the irradiated sample, and its abundance and half-life were measured. This information, combined with the estimated integrated flux, is sufficient to determine G_{1144} . Although the value determined mass spectrometrically (35 \mp 50 barns) lies within twice the standard deviation, it is worthwhile considering the implications of the assumption that the difference is significant.

The "effective" cross section could be changed if strong resonance absorption of epithermal neutrons were possible. However, since both irradiations took place in nearby tray-rod positions where the epithermal neutron density is expected to remain proportional to the thermal neutron density, no such marked effect will occur.

Neutron filtering by the Sm^{140} resonance could increase the apparent cross section for our sample (13.8% of Sm^{140}) compared to the U.S. sample (4.5% of Sm^{149}). If all the Sm^{144} cross section were due to a resonance at a neutron energy higher than the Sm^{140} "cut-off" at 0.2 ev., neutron capture by Sm^{144} would proceed inside the "black" core taken as a model in appendix B, due to the presence of epi- Sm^{149} neutrons. Thus the "effective" integrated flux for Sm^{144} would exceed the value calculated in the appendix. The maximum correction possible on this basis would be a multiplication factor equal to the "effective integrated flux" for samarium divided by the unperturbed integrated flux, numerically equal to two-thirds.

A second explanation, namely, that $\mathrm{Sm}^{1.45}$ has a large cross section and "burns up" to form $\mathrm{Sm}^{1.46}$ appears untenable in view of the identification of $\mathrm{Sm}^{1.46}$ as a long-lived alpha emitter 52 . Such a cross section would yield about 3% of $\mathrm{Sm}^{1.46}$ in the U.S. sample, but this isotope was not detected. Short-lived isomoric states may also be formed. A short-lived $\mathrm{Sm}^{1.45}$ insomer could account for the small amount of that isotope in the U.S. sample, despite a large $\mathrm{Sm}^{1.44}$ cross section; a short-lived $\mathrm{Sm}^{1.46}$ isomer could account for the lack of that isotope if $\mathrm{Sm}^{1.46}$ and $\mathrm{Sm}^{1.45}$ had large cross sections.

It is possible that the sample used in our investigation contained an unidentified contaminant at mass 144 which was destroyed by irradiation. This contaminant could not be a rare earth, nor could it be the oxide or dioxide of an element of lower Z, since all

elements concerned would show a characteristic pattern about mass 144. In addition, the presence of such a contaminant would require a "real" abundance of Sm^{144} of 3.05% or less. An abundance this low is discounted, but not ruled out, by the results of the samarium analysis of the preceding section (VII A). The effect on the other abundances of this reduction in Sm^{144} would bring about cross section changes well within the errors quoted.

Summarizing briefly, we find that the set of cross sections given in the last line of Table XXII are in agreement with the best values determined elsewhere except in the case of Sm^{144} . In this case a more accurate measurement gives a cross section lower than our value by an amount equal to 1.7 times our quoted error. This discrepancy is most probably due to the uncertainty associated with measuring the small Sm^{144} peaks and in extrapolating abundances back to zero time. The formation of a short-lived isomeric state is also feasible.

2. Gadolinium (Table XXIII)

The irradiation period used in the present investigation was too long to permit a determination of the very large cross sections of Gd^{155} and Gd^{157} . In the case of the remaining isotopes the cross sections are such that the changes produced in isotopic abundances are not decisive. This applies even in the case of σ_{154} where the cross section just exceeds twice the assigned error. The discrepancy between this work and others for Gd^{160} could signify a cross section of about 10 barns yielding a very short-lived metastable state of Gd^{101} .

3. Dysprosium (Table XXIV)

The results obtained in Table XV are compared in Table XXIV with other measurements of σ_{160} . The cross section determined for Dy¹⁶⁰ from the mass spectrometer measurements is in good agreement with activation values determined previously.

The possibility of an increase in $\sigma_{\rm C}({\rm Dy})$ due to resonant capture of epithermal neutrons has not been taken into account. One resonance peak is particularly prominent in the spectrum which show the total cross section of dysprosium as a function of neutron energy 38. This peak has an energy, Mr, of 5.9 electron volts at the resonance maximum. The product of the maximum cross section and the peak half-width, $\sigma_{\rm T} \Gamma_{\rm T}$, is about 1850. Using a simplification of the formula 55 giving the contribution from resonance capture of epithermal reactor neutrons to the total effective cross section for reactor neutrons of all energies, we have

$$\sigma_{re} = 2 \sqrt{\pi} \sigma_{r} \left(\frac{\Gamma_{r}}{2Er} \right) \frac{\beta}{1 - \beta}$$

where σ_{re} is the cross section contribution due to capture of epithermal neutrons in a single resonance defined by σ_r , E_r , and Γ_r , and β is the ratio of the density of epithermal neutrons to thermal neutrons. For the position occupied by these samples in the reactor, β is about 0.03. The contribution from this large peak is therefore about 16 barns. It is doubtful whether the remaining peaks, which are not clearly resolved, make a total contribution greater than this. As a conservative estimate, we may take the epithermal neutron contribution to be less than 50 barns. This represents an increase in the value of $\mathcal{O}_{\mathbf{c}}(\mathbf{D}\mathbf{y})$ of about two-thirds the error assigned. The corresponding increase in the isotopic cross sections would be about 10% of the corresponding errors.

The effect of the Tb^{159} contamination noted in section V A has not been included. If all the impurity in the dysprosium sample (2%) were Tb^{159} , capture to Tb^{100} followed by /3-decay to Dy^{160} would add 0.05% to the irradiated abundance of that isotope, raising the cross section from 100 to 140 barns. It is probable, however, that Tb^{159} constituted only a few percent of all the impurities, in which case this source of error is negligible.

4. Ytterbium

The occurrence of a large cross section for Yb¹⁰⁰ complicates the problem of interpreting the measurements on ytterbium. The information available in addition to that givenin Table XXV is as follows: in the curve of total cross section vs. neutron energy one resonance peak has been identified asdue to Yb²⁰⁰ $(E_r = 0.6 \text{ ev.}, \sigma_r = 135 \text{ barns}, \Gamma_r = 0.085 \text{ ev})$, this peak contributes 1.0 barns due to resonance capture of epithermal neutrons in the reactor; other peaks give a total contribution of 3 barns due to capture of epithermal neutrons where β is taken to be 0.03 in both cases; the measured cross section for the element, σ_c (Yb), in a thermal flux is 3674 barns ³⁸, using the pile oscillator technique.

The total isotopic contribution to the capture cross section of ytterbium in a reactor flux should therefore be equal to σ_{e} (Yb) in a thermal flux plus the epithermal contributions from all known resonances minus the contribution from Dy¹⁰⁰ in a reactor flux. Using the lowest value available for this latter cross section, viz. 11,000 barns ³⁸, this latter contribution is 15 barns. Thus $\sum_{170}^{170} n_1 \sigma_1 = 36 + 3 + 1 - 15 =$ 25 barns, a value which is in reasonable agreement with that obtained using the lower limits of the ytterbium isotopic cross sections given in Table XVI. Because of this agreement, these results are repeated in the last line of Table XXV. Confirmation of these values will depend on further measurements of the cross section of Dy¹⁷⁶ and an experiment to determine whether neutron capture in Dy¹⁷⁵ does compete appreciably with beta-decay for high flux irradiations.

5. Summary

At the present time full use of the precision available in the mass spectrometric determinations is precluded by uncertainties in current cross section data. These include the experimental errors in the absorption cross sections of the naturallyoccurring elements for a reactor spectrum of neutrons, disagreements between measurements of the activation cross sections of stable isotopes and a dearth of information concerning neutron capture in short-lived isotopes. As more exact information becomes available these mass spectrometric measurements will permit estimates having higher accuracy to be made of the reactor neutron capture cross sections of the stable isotopes of these four elements.

			Isc	Mass 1	ass No.					
Reference		144	147	148	149	150	151	152	153	154
Seren, Friedlander and Turkel	a							135		5.5
Bothe	b							280		
Inghram, Hayden Hess	C				47000		7200			
Melaika, Parker Petruska, Tomlinso	đ				66200		12000			
Harwell	e				76700					
U.S.A.	ſ	1.0∓0	•3							
This work	g	85755	85725	<70	>250	00 11 5 +45	15000 +6500	250 750	30000 +12000	o
a - See reference	2									
b - See reference	1.	The value 64 barns	here : for ho:	is nor: Imium	malized	d to a	captu	re cro	ss sect	tion of
 c - See reference d - See reference e - See reference f - This value was g - These values c very near the on Page 80. 	39 16 51 cal orre 10we	culated f sponding er limits	rom ind to a no of Tab	format ormali le XII	ion apj zation I. The	pearin facto method	g in ro r of O of assi	eferen .983, : gning e	ce 14. 1.e., rror is	described

.

TABLE XXII - MEASUREMENTS OF THE NEUTRON CAPTURE CROSS SECTIONS OF SAMARIUM ISOTOPES

				Isot	opic Ma	ss No.		
Reference		152	154	155	156	157	158	160
Inghram, Hayder Hess	n ^a			41x10 ⁵		59.5x	10 ³	
Lapp, Van Horn, Demps	b ster			69x10 ^s	1	240x1	08	
Seren, Friedlander, Tu	c 1rkel	125					4	
Butement	đ						4.5	0.17
der Mateosian Goldhaber, Smit	e th							0.1
This work	ſ		250712	0 >25x10) ^s <14	>25x1	0 ³ <11	17-11
a - See referen b - See referen c - See referen	nce 39 nce 7 nce 2							

TABLE XXIII - MEASUREMENTS OF THE NEUTRON CAPTURE CROSS SECTIONS OF GADOLINIUM ISOTOPES

d - See reference 40 e - See reference 53 f - Value from Table XIV

			Isotopic I	lass No.		
Reference		160	161	162	163	164
Goldhaber Muchlhause	a					2860
Bothe	Ъ					2870
Seren Friedlander,	c Turkel					2620
This work	đ	100 7 70	620 i 35	330 i 90	420 - 150	2800 - 180

TABLE XXIV - MEASUREMENTS OF THE NEUTRON CAPTURE CROSS SECTIONS OF DYSPROSIUM ISOTOPES

a - See reference 54

b - See reference 1. The value listed here is normalized to a capture cross section of 64 barns for holmium ³⁸.

c - See reference 2

d - From Table XV. Errors include both mass spectrometric standard deviations and uncertainty in the product $o_c(Dy)gt$.

TABLE	XXV	- MEASUREN	ENTS OF	THE M	EUTRON C	APTURE	CROSS SECT	IONS OF	YTTERBIUM	ISOTOPES
						Iso	topic Mass	No.		
Refer	ence		168	170	171	172	173	174	175	176
Bothe		a,c	24000					96		8.8
Atter Bohr,	ling, Sigu	b,c rgeirson	40000					20		5
This	work	d		<18	55 ± 15	<5	20 ± 10	55 ± 2	25 9000 [±] 4000	
a - S	00 ro	ference 1.	. The v secti	values lon of	listed 1 64 barn	here ar s for h	e normaliz olmium38.	ed to a	capture cr	038
b - S	ee re	ference 50	5. The t section	values lon of	listed 1 2800 ba	here ar rns f or	e normaliz Dy164.	ed to a	capture cr	2022
o - B p	NL 32 reced	25 ³⁸ quoto ling refere	es 11000 ences.	± 300	0 barns :	for Yb ¹	.68 based o	n the ve	lues of th	ne two
d - Th		ross section	s correspo	nd to t	the lower	limita o	f Trable VVT	The owned		

.

.

d - These cross sections correspond to the lower limits of Table XVI. The errors assigned were obtained in a manner similar to that described on Page 80.

APPENDIX A

EFFECT OF NEUTRON IRRADIATION ON ISOTOPIC ABUNDANCE.

DERIVATION OF THE EQUATIONS.

An element of charge Z has isotopes with mass numbers $A + 1, A + 2, \dots A + n$, of which at least the first is stable and occurs naturally. These isotopes are present in the amounts n_1 , n_2 , \dots , n_n nuclei per c.c. before irradiation, and n'_1 , n'_2 , \dots , n'_n nuclei per c.c. after irradiation, having undergone a change of Δn_1 , Δn_2 , \dots , Δn_n nuclei per c.c., where $\Delta n = n-n'$. This change is due to the capture of neutrons in a flux of \emptyset per cm² per sec. for a time t seconds, where the capture cross-sections are σ_1 , σ_2 , \dots , σ_n respectively.

The basic equations are those for the number of atoms present in a radioactive decay chain after a time t. These are

$$n_1 = n_1 e^{-\lambda_1 t}$$

$$n_{2}^{\prime} = n_{1}\lambda_{1} \left[\frac{\frac{\partial}{\partial 1}t}{\partial 2-\lambda_{1}} + \frac{\partial}{\partial 1-\lambda_{2}} \right]$$

(A1)

$$n_{3}^{\prime} = n_{1}\lambda_{1}\lambda_{2} \left[\frac{-\lambda_{1}t}{(\lambda_{2}-\lambda_{1})(\lambda_{3}-\lambda_{1})} + \frac{-\lambda_{2}t}{(\lambda_{1}-\lambda_{2})(\lambda_{3}-\lambda_{2})} + \frac{-\lambda_{3}t}{(\lambda_{1}-\lambda_{3})(\lambda_{2}-\lambda_{3})} \right]$$

$$n_{s}' = n_{1}\lambda_{1}\lambda_{2} - \lambda_{s-1} \left[\frac{-\lambda_{1}t}{(\lambda_{2}-\lambda_{1})(\lambda_{3}-\lambda_{1})-(\lambda_{s}-\lambda_{1})} + \frac{-\lambda_{s}t}{(\lambda_{1}-\lambda_{s})(\lambda_{2}-\lambda_{s})-(\lambda_{s-1}-\lambda_{s})} \right]$$

The only change required in dealing with neutron crosssections is to replace λ by $\sigma \emptyset$, a permissible step since both represent the probability of the loss of a nucleus per second.

For the first (lightest) isotope, the equation is very simple, since nuclei are lost, but none are gained from preceding isotopes.

(A2)
$$n_1' = n_1 e^{-\sigma_1 g t}$$

For the remaining isotopes, it is necessary to treat each preceding isotope as the parent of a "capture" chain, in addition to the nuclear loss equivalent to equation (A2)

(A3)
$$n_2' = n_2^{-\sigma_2 \otimes t} + n_1^{\sigma_1} \begin{bmatrix} -\sigma_1^{\otimes t} & -\sigma_2^{\otimes t} \\ \frac{\Theta}{\sigma_2 - \sigma_1} & + \frac{\Theta}{\sigma_1 - \sigma_2} \end{bmatrix}$$

where the second part is just the second equation of (A1). Rearranging

$$n_{2}^{i} = e^{-\sigma_{2} \not \sigma t} \left[n_{2} + \frac{n_{1} \sigma_{1}}{\sigma_{1} - \sigma_{2}} \right] + \frac{n_{1} \sigma_{1}}{\sigma_{2} - \sigma_{1}} \cdot e^{-\sigma_{1} \not \sigma t}$$

$$i.e.$$

$$(A_{3}') \quad n_{2}^{i} = e^{-\sigma_{2} \not \sigma t} \left[n_{2} + \frac{n_{1} \sigma_{1}}{\sigma_{1} - \sigma_{2}} \right] + \frac{n_{1}^{i} \sigma_{1}}{\sigma_{2} - \sigma_{1}}$$

Similarly

(A4)
$$n_{3}^{\dagger} = n_{3}e^{-\sigma_{3}gt} + n_{2}\sigma_{2}\left[\frac{e^{-\sigma_{2}gt}}{\sigma_{3}-\sigma_{2}} + \frac{e^{-\sigma_{3}gt}}{\sigma_{2}-\sigma_{3}}\right] + n_{1}\sigma_{1}\sigma_{2}\left[\frac{e^{-\sigma_{1}gt}}{(\sigma_{2}-\sigma_{1})(\sigma_{3}-\sigma_{1})} + \frac{e^{-\sigma_{2}gt}}{(\sigma_{1}-\sigma_{2})(\sigma_{3}-\sigma_{2})} + \frac{e^{-\sigma_{3}gt}}{(\sigma_{1}-\sigma_{3})(\sigma_{2}-\sigma_{2})}\right]$$

Rearranging

$$n_{3}^{i} = e^{-\sigma_{3} \# t} \left[n_{3} + \frac{n_{2} \sigma_{2}}{(\sigma_{2} - \sigma_{3})} + \frac{n_{1} \sigma_{1} \sigma_{2}}{(\sigma_{1} - \sigma_{3})(\sigma_{2} - \sigma_{3})} \right]$$

+ $e^{-\sigma_{2} \# t} \left[\frac{n_{2} \sigma_{2}}{(\sigma_{3} - \sigma_{2})} + \frac{n_{1} \sigma_{1} \sigma_{2}}{(\sigma_{1} - \sigma_{2})(\sigma_{3} - \sigma_{2})} \right]$
+ $e^{-\sigma_{1} \# t} \left[\frac{n_{1} \sigma_{1} \sigma_{2}}{(\sigma_{2} - \sigma_{1})(\sigma_{3} - \sigma_{1})} \right]$

The second term is equal to

$$\left[n_{2}^{\prime}-\frac{n_{1}^{\prime}\sigma_{1}}{\sigma_{2}-\sigma_{1}}\right]\frac{\sigma_{2}}{\sigma_{3}-\sigma_{2}} \qquad \text{from (A3:)}$$

and the third term is equal to

$$\frac{\mathbf{n_1}^{\mathbf{a_1}}\mathbf{\sigma_2}}{(\mathbf{\sigma_2} - \mathbf{\sigma_1})(\mathbf{\sigma_3} - \mathbf{\sigma_1})}$$

Combining the two terms in n gives

$$n_{1}^{\prime} \left[\frac{-\sigma_{1}\sigma_{2}}{(\sigma_{2}-\sigma_{1})(\sigma_{3}-\sigma_{2})} + \frac{\sigma_{1}\sigma_{2}}{(\sigma_{2}-\sigma_{1})(\sigma_{3}-\sigma_{1})} \right] = n_{1}^{\prime} \frac{\sigma_{1}\sigma_{2}}{\sigma_{2}-\sigma_{1}} \left[\frac{-\sigma_{3}+\sigma_{1}+\sigma_{3}-\sigma_{2}}{(\sigma_{3}-\sigma_{2})(\sigma_{3}-\sigma_{1})} \right]$$
$$= n_{1}^{\prime} \frac{\sigma_{1}\sigma_{2}}{(\sigma_{1}-\sigma_{3})(\sigma_{3}-\sigma_{2})}$$

Therefore

$$\begin{array}{c} (All^{+}) \\ n_{3}^{*} = e^{-\sigma_{3} \sigma_{1}^{*} \sigma_{2}} \left[n_{3}^{*} + \frac{n_{2} \sigma_{2}^{*}}{\sigma_{2}^{*} - \sigma_{3}^{*}} + \frac{n_{1} \sigma_{1} \sigma_{2}}{(\sigma_{1}^{*} - \sigma_{3}^{*})(\sigma_{2}^{*} - \sigma_{3}^{*})} \right] + \frac{n_{2}^{*} \sigma_{2}^{*}}{\sigma_{3}^{*} - \sigma_{2}^{*}} + \frac{n_{1}^{*} \sigma_{1} \sigma_{2}}{(\sigma_{1}^{*} - \sigma_{3}^{*})(\sigma_{3}^{*} - \sigma_{2}^{*})} \right]$$

Equations (A2), (A3') and (A4') can be rearranged to give the required expressions for the capture cross-sections. (A5a) $\sigma_1 \sigma_1 = \ln n_1 - \ln n_1'$

(A5b)
$$\sigma_2 \sigma t = \ln \left[n_2 + \frac{n_1 \sigma_1}{\sigma_1 - \sigma_2} \right] - \ln \left[n_2' + \frac{n_1' \sigma_1}{\sigma_1 - \sigma_2} \right]$$

(A5c)
$$\sigma_{3}\sigma_{3} = \ln \left[n_{3} + \frac{n_{2}\sigma_{2}}{\sigma_{2}-\sigma_{3}} + \frac{n_{1}\sigma_{1}\sigma_{2}}{(\sigma_{1}-\sigma_{3})(\sigma_{2}-\sigma_{3})} \right]$$

- $\ln \left[n_{3}^{i} + \frac{n_{2}^{i}\sigma_{2}}{\sigma_{2}-\sigma_{3}} + \frac{n_{1}^{i}\sigma_{1}\sigma_{2}}{(\sigma_{1}-\sigma_{3})(\sigma_{2}-\sigma_{3})} \right]$

The extension to the nth isotope is

(A5n)
$$\sigma_{n} \sigma_{n} = \ln \left[n_{n} + \frac{n_{n-1}\sigma_{n-1}}{\sigma_{n-1}\sigma_{n}} + \dots + \frac{n_{1}\sigma_{1}\sigma_{2}-\sigma_{n-1}}{(\sigma_{1}-\sigma_{n})(\sigma_{2}-\sigma_{n})-\dots(\sigma_{n-1}-\sigma_{n})} - \ln \left[n_{n}' + \frac{n_{n-1}'\sigma_{n-1}}{\sigma_{n-1}-\sigma_{n}} + \dots + \frac{n_{1}'\sigma_{1}\sigma_{2}-\dots\sigma_{n-1}}{(\sigma_{1}-\sigma_{n})-\dots(\sigma_{n-1}-\sigma_{n})} \right]$$

A somewhat different approach to the problem leads to a set of much simpler equations, which can be shown to be equivalent to modified first order approximations to equations (A5).

If the rate of change of abundance of an isotope is small, the abundance at any time T after the start of irradiation ($T \leq t$) will differ only slightly from $n - \frac{T}{t}$ (n-n^t).

Assuming that this difference is negligible, this is equivalent to the relation,

rate of capture =
$$\frac{1}{2}$$
 (initial rate + final rate)
= $\sigma_{\emptyset} \left(\frac{n+n^{\dagger}}{2}\right)$

The total change due to capture, ôn, is, then $\delta n = \frac{1}{2}(n + n')\sigma\sigma t = n^{\frac{1}{2}}\sigma\sigma t$

Applying this formula to each isotope in turn gives

$$\Delta n_{1} = \delta n_{1} = n_{1}^{*} \sigma_{1} \sigma_{1}$$

$$\Delta n_{2} = \delta n_{2} - \delta n_{1} = n_{2}^{*} \sigma_{2} \sigma_{2} \sigma_{1} - \Delta n_{1}$$

$$\Delta n_{3} = \delta n_{3} - \delta n_{2} = n_{3}^{*} \sigma_{3} \sigma_{1} - (\Delta n_{1} + \Delta n_{2})$$

$$\Delta n_{n} = \delta n_{n} - \delta n_{n-1} = n_{n}^{*} \sigma_{n} \sigma_{1} - \sum_{s=1}^{n-1} \Delta n_{s}$$

The approximate equations for the cross-sections become

$$\sigma_{1} = \Delta n_{1} / n_{1}^{\ddagger}$$
(A6')
$$\sigma_{2} = \frac{\Delta n_{1} + \Delta n_{2}}{n_{2}^{\ddagger}}$$

(A6)

$$\sigma_{m} ct = \frac{\sum_{i=1}^{m} (\Delta n_{i})}{n_{m}^{\pm}}$$

In order to show that these equations correspond to a modification of the first order approximation, consider the specific case of the second isotope (equation (A3'))

$$n_{2}' = o^{-\sigma_{2} \otimes t} \left[n_{2} + \frac{n_{1} \sigma_{1}}{\sigma_{1} - \sigma_{2}} \right] + \frac{n_{1}' \sigma_{1}}{\sigma_{2} - \sigma_{1}}$$
where $e^{-\sigma_{2} \otimes t} = 1 - \sigma_{2} \otimes t + \frac{(\sigma_{2} \otimes t)^{2}}{2!} - \frac{(\sigma_{2} \otimes t)^{3}}{3!} + \cdots + \cdots$

To a first approximation, then

(A7)
$$n_{2}' = (1 - \sigma_{2} \sigma_{1}) \left[n_{2} + \frac{n_{1} \sigma_{1}}{\sigma_{1} - \sigma_{2}} \right] + \frac{n_{1}' \sigma_{1}}{\sigma_{2} - \sigma_{1}}$$

= $n_{2} - n_{2} \sigma_{2} \sigma_{1} + \Delta n_{1} \frac{\sigma_{1}}{\sigma_{1} - \sigma_{2}} - n_{1} \sigma_{1} \sigma_{1} \frac{\sigma_{2}}{\sigma_{1} - \sigma_{2}}$

The last term is seen to be equal to the first order approximation for Δn_1 times $\frac{\sigma_2}{\sigma_1 - \sigma_2}$. Therefore $\Delta n_2 = n_2 - n_2 = n_2 \sigma_2 \varphi t + \Delta n_1 \left[\frac{\sigma_2}{\sigma_1 - \sigma_2} - \frac{\sigma_1}{\sigma_1 - \sigma_2} \right]$ $= n_2 \sigma_2 \varphi t - \Delta n_1$

(A8) whence

$$\sigma_2 = \frac{\Delta n_1 + \Delta n_2}{n_2}$$

This equation differs from its equivalent in the set (A6) in that n_2 replaces n_2^{\pm} .
Now the equation n' = n(1-ost) is seen to represent a rate of capture which is independent of time, just as was assumed in deriving equations (A6). In the first order approximation however, this rate is taken proportional to the initial abundance, which is certainly too large, and will therefore give an expression for the capture cross-section which is too small.

Therefore, the set of equations (A6) are a modification of the first order approximation giving a more nearly correct value. As indicated in the calculation of the actual crosssections, they are a very good approximation, even when $\Delta n/n$ exceeds ten percent.

APPENDIX B

FLUX DEPRESSION IN THE IRRADIATED SAMPLES

1. Absorber in Neutron Bath

The flux within each of the rare earths samples irradiated (Nd, Sm, Gd, Dy and Yb) will be reduced by neutron capture in the material. The flux depression in an absorber located in a neutron bath has been treated for a variety of shapes and volumes of absorbing material by Case, de Hoffmann and Placzek ⁵⁷. Their treatment is used as a basis for developing a set of equations applicable to the irradiations of interest here.

It has been shown (loc. cit., p. 20) that, for a neutron absorber located in such a bath, the ratio of the average neutron density within the absorber to the unperturbed density, i.e., the density in the neutron bath in the absence of the absorber, is equal to the escape probability for a constant isotropic neutron source distribution within the absorber. The algebraic equivalent of this statement is

$$\varphi/\varphi_{\rm un} = \mathbf{P}_6 \tag{B1}$$

where β is the mean flux within the sample, β_{un} is the unperturbed flux, and P_0 is the probability of neutron escape for a constant isotropic neutron source distribution within the absorber.

As a typical example the escape probability for a homogeneous purely-absorbing (scattering cross section << capture cross section) sphere is

$$P_{0} = \frac{3}{8} (a\Sigma)^{-2} \left[2(a\Sigma)^{2} - 1 + (1 + 2a\Sigma) e^{-2a\Sigma} \right]$$
(B2)
= $\frac{3}{4} (a\Sigma)^{-2} \left[1 - \frac{1}{2} (a\Sigma)^{-2} \right]$ for $a\Sigma \gg 1$ (B2')

Here a is the radius of the sphere and \geq is the macroscopic cross section, i.e., the inverse of the absorption mean free path, 1, given by the equation

$$\Sigma = \frac{no}{V}$$
 (B3)

where n is the number of atoms in the sample,

o is the absorption cross section per atom,

and V is the sample volume.

In the case of a homogeneous purely-absorbing infinite cylinder of radius "a"

$$P_{0} = 1 - \frac{1}{3} a \Sigma + \frac{1}{2} (a \Sigma)^{2} \log (2/a \Sigma) + \frac{1}{2} (a \Sigma)^{2} (1.25 - Y) \text{ for}$$

$$a \Sigma < 0.1 \qquad (B4)$$

$$\approx \frac{1}{2} (a \Sigma)^{-1} \left[1 - \frac{3}{16} (a \Sigma)^{-2} \right] \text{ for } a \Sigma > 6 \qquad (B4')$$

$$(Y = \text{Huler's constant} = 0.577216--)$$

Tabulations of $P_c = 1-P_o$ as a function of $a/1 = a \sum$ are given for these (loc. cit., Tables 3 and 4) as well as other forms.

2. Flux Depression in a Weak Absorber

In a neutron irradiation the average destruction rate of a particular isotope will depend on the mean neutron flux to which it is subjected. As a consequence of changes occurring in the isotopic distribution during the irradiation, and hence also in the macroscopic cross section, Σ , and the escape probability, P_o , the destruction rate will be a complicated function of the irradiation history of the sample. Further, because of the flux distribution is not uniform inside the sample the changes in Σ depend on position, so that the expressions for the escape probabilities are more complex than indicated by equations (B2) and (B3).

In certain limiting cases, however, the treatment may be simplified. Thus, for a weakly absorbing sample (a $\geq \ll 1$) for which the irradiation period exceeds the mean destruction time, the flux within the sample remains nearly constant during the irradiation and does not depend appreciably on position. In this case the neutron distribution within the sample may be considered to be homogeneous, with \geq constant throughout the irradiation.

3. Flux Depression in a Strong Absorber

a) <u>Neutron Absorption in a Shell</u>

For strongly-absorbing matter $(a \Sigma \gg 1)$ the destruction rate is independent of Σ as a first approximation, and depends only on the sample surface area. This may be demonstrated for a spherical sample as follows: the number of neutrons absorbed in the sphere per second, $\delta \beta$, is equal to the mean flux within the sphere, $P_{o} \phi_{un}$, multiplied by the total absorption cross section of the sphere, $\Sigma V = n\sigma$. Now for $a \ge \gg 1$, $P_0 = \frac{3}{4} (\ge a)$ so that $\delta \varphi = P_0 \varphi_{un} \ge V = (1/4) \varphi_{un} 4 \pi a^2 = (1/4) \varphi_{un}$ (B5) where S is the capturing surface. Equation (B5) can also be shown to apply to the infinite cylinder.

Thus for $a \geq >1$ an approximation which is useful in determining how quickly the absorbing isotope is being destroyed is to assume that all the neutron absorption takes place in a shell which is thin compared to the dimensions of the absorber. This shell initially has the dimensions and shape of the surface of the absorbing material, but shrinks down as the isotope(s) with large $\sigma_{\rm d}$ are burned out. This approximation is valid only as long as the radius of the unirradiated core is large enough so that the first term of the expansion of $P_{\rm o}$ ($a \geq$), for large $a \ge$, is nearly equal to the escape probability. Thereafter, the burnup rate increases more slowly than for the "surface burnup" approximation because some neutrons which enter the core will escape.

Assuming either a spherical or infinite cylindrical form (both of radius "a") the number of atoms in the shell when its radius has shrunken to r_0 ($r_0 < a$) is given by

$$\delta n = (nR/V).S(r).\delta r_0 \qquad (B6)$$

where R = the relative isotopic abundance of the absorbing isotopes. If it is assumed that the isotopes cutside the shell do not depress the flux appreciably, neutrons absorbed per second may be equated to atoms destroyed to obtain the rate at which the radius changes with time

$$\delta r_0 / \delta t = g_{un} V / 4nR$$
 (B7)

The time required to reduce the shell to a radius, r_v , which may be taken as the limit of validity for this approximation, is given by

$$T(r_v) = \frac{4nR(a-r_v)}{\varphi_{un}} \text{ secs. (B7')}$$

b) Flux Outside the Absorbing Shell

If the material in the region outside the absorbing shell is only weakly-absorbing then the flux at any point a distance r from the centre of the cylinder or sphere is given approximately 58 by

$$g(\mathbf{r}) = \varphi_{un} \left[1 - \frac{r_o}{2r} B \left(\Sigma r_o \right) \right] \qquad (B8)$$

where r_0 is the radius of the undestroyed core and $B(\Sigma r_0) =$ $\frac{4V(n_{o})}{S(n_{o})} \stackrel{P_{o}(\sum r_{o})}{is}$ the blackness or impenetrability of the core (of surface $S(r_0)$ and volume $V(r_0)$). The ratio of mean flux inside the sample to the unperturbed flux is then given by

for a core radius of r_0 , where $V_1 = \frac{4}{3}\pi a^3$ for the sphere and Tta² for a unit length of cylander. For cylindrical geometry, the only case of interest in these irradiations,

$$V/S = r_0/2$$

so that

In the region $r_{y} \leq r_{\bar{y}} \leq a$, $P_{0} \approx \frac{1}{2r_{o} \geq 1}$

and
$$\frac{\varphi}{\varphi_{un}} = 1 - \frac{r}{a}o$$
 (B9⁺⁺)

4. Neodymium. The Unperturbed Flux

The previous treatment of the neodymium results ¹¹ did not include a small adjustment for loss of neodymium atoms through capture in Nd¹⁴⁰ followed by beta-decay. Table BI is a revision of the fourth table from that report. On the basis of $\sigma_c(Nd) = 46 \mp 2$ barns, the "best value" as quoted in the Brookhaven compilation on neutron cross sections ³⁸, combined with the value for $\sigma_c(Nd)$ Øt of 2.22 obtained by summing the contributions from Nd¹⁴²⁻⁶(row 6), the value for the effective integrated flux is 0.483 \mp 0.035 neutrons per kilobarn.

The neodymium sample consisted of 11 mg. of metal filings, contained in an $1/8^{n}$ O.D. aluminum tube having a drilled cylindrical hole of radius 0.07 cm. The effective density is taken to be 2.0 \pm 0.5 gms/cc., in line with measurements on samarium metal and the oxides of the other rare earths. The height of the sample within the tube was therefore about 0.35 cm., sufficiently greater than the radius that treatment as an infinite cylinder will not introduce a serious error.

The macroscopic cross section Z is calculated to be 0.33 \neq 0.10 cm.⁻¹ so that .025 < P_c < .042 (Table 4, reference 57,) for the infinite cylinder of radius 0.07 cm. On the other hand, if the material is assumed to be massed as a sphere $\Xi = \frac{0.62n\sigma}{v^{2/3}}$ $= \frac{0.62 n\sigma}{(mass/density)^{2/3}}$ giving .026 < P_c < .037 (loc. cit., table 3). The flux depression, \emptyset/\emptyset_{un} (equal to 1-P_c) may therefore be taken

TABLE BI - ISOTOPIC	CAPTURE	CROSS	SECTIONS	FOR	NEODY LUI
---------------------	---------	-------	----------	-----	-----------

lieodymium Isotope	142	143	144	145	146	148	150
n ^a .	27.10 (.025)	12.14 (.017)	23.83 (.024)	8.29 (.011)	17.26 (.017)	5.74 (.016)	5.63 (.023)
n* b	26.92 (.042)	10.57	25.60 (.033)	8.15 (.015)	17.36 (.027)	5.73 (.017)	5.64 (.028)
$\Delta n = n - n^*$	0.18 (.05)	1.57 (.033)	-1.77 (,04)	0.14 (.019)	-0.11 (.032)	.01 (.023)	01 (.036)
∑ ^s (∆n _i) ^c	0.18 (.05)	1.75 (.06)	02 (.07)	0.14 (.03)	0.03 (.04)	.01 (.023)	01 (.036)
ogt d	.0067	.154 f	0008	.017	.0017	.0017	0017
nogit e	0.18 (.05)	1.87 ^f (.06)	-0.019	0.14 (.03)	.029 (.04)	0.01 (.023)	01 (.036)
_ o d	14 7 4	320 - 14	f -2 + 7	35 + 7	475	3 7 9	-3 + 14

a $\Sigma n = 99.99\%$. The numbers in brackets are standard deviations.

b $\Sigma n^{1} = 99.96\%$. The extra .03% is the allowance for loss after capture in Nd¹⁴⁰.

c The summation terminates after Nd¹⁴⁴, as well as after Nd¹⁴⁸. The error assigned to the value for Nd¹⁴⁵ has been increased to take into account a portion of the preceding cumulative error.

d The cross sections are based on the value of \$t obtained above. Obtained using the approximate equations, except where noted.

e Isotopic contributions to σ_n (Nd)gt. $gt = .483 \pm .035$ n/kb. (See text)

f These values are obtained by using the exponential equation, not the approximation.

as 0.97 \div 0.01, which, combined with the effective integrated flux calculated above, gives a value for the unperturbed integrated flux, β_{un} t, of 0.50 \div .037 neutrons per kilobarn. 5. Samarium

Thirty milligrams of powdered samarium metal were irradiated in the standard aluminum capsule having an inner diameter of 0.14 cm. A measurement of the density of the uncompressed powder gave a value of 2.25 ± 0.20 g./cc. The material therefore occupied a volume in the irradiation capsule of 0.0133 cc, equivalent to a cylinder length of 0.86 cm. Since Sm¹⁴⁹ has a cross section of about 66000 barns for pile neutrons ¹⁶, it is valid to use a model in which this isotope is destroyed in a thin cylindrical shell of initial radius 0.07 cm.

An approximate value for the effective integrated flux can be obtained from equations (B7') and (B9'') on the assumption that $r_y = 0$. In this case the irradiation "lifetime" of Sm¹⁴⁹ is found to be

 $\phi_{\rm im} T_{149} = 0.35 \, \rm n/kb$.

During this interval, the Sm^{149} in the material outside the unburned core will have been completely converted to Sm^{150} . Also, according to equation (B9^{**}) the mean flux varies linearly with the radius r_0 , and hence with the irradiation period. Therefore, the effective integrated flux is equivalent to an irradiation of the "burned-out" sample, in which the amount of Sm^{150} is given

by $n_{140} + n_{150}$, by the unperturbed flux for one half the period T_{140} plus the balance of the irradiation period, i.e., $(gt)_{eff} = 0.325 n/kb.$

For a more exact calculation let $r_v = 0.04$ cm. Inside this region the possibility of a neutron passing through the core has values appreciably greater than zero. The ratio \emptyset/\emptyset_{un} is then given by equation (B9') and the rate of change of radius by the equation

$$\frac{\delta \mathbf{r}}{\delta \mathbf{t}} \mathbf{o} = - \frac{\mathbf{g}}{4\mathbf{H}\mathbf{R}} \mathbf{B}(\mathbf{\Sigma} \mathbf{r}_{\mathbf{o}}) \qquad (\mathbf{B7''})$$

Note that as $B(\sum r_o)$ decreases, \emptyset/\emptyset_n increases more rapidly than in the outer region, while $\delta r_o/\delta t$ increases more slowly. These two effects tend to cancel one another. A determination of $(\emptyset t)_{eff}$ using these equations, and taking

 $B(\Sigma r_0) = \propto r_0 \ln(\beta r_0) *$

to facilitate integration of equation 10, gives a value of 0.323 n/kb in good agreement with the approximation above.

According to Dr. S.A. Kushneriuk the approximation used (B8) to determine \mathscr{P}_{un} gives values which are somewhat low just outside the core. The calculated value of $(\mathscr{P}_{eff})_{eff}$ will be correspondingly too low. The best value for the effective integrated flux in the samarium sample is therefore taken to be $(\mathscr{P}_{eff})_{eff} = 0.33 \pm 0.04$ neutrons per kilobarn

* This equation gives a very good fit to $B(\Sigma r)$ over the region below r = 0.04 cm. \propto and β were determined by fitting at radii of 0.03 cm. and 0.004 cm.

6. Gadolinium

Seven milligrams of gadolinia powder (Gd_2O_3) were irradiated. Measurements on uncompressed oxides of samarium, neodymium and gadolinium gave densities in the range 1.4 ± 0.1 gms./cc. On the basis of this density the volume occupied by the gadolinia will be 0.005 cc., corresponding to a height of 0.32 cm. in the 0.07 cm. radius cylinder. The dimensions are such that the model of an infinite cylindrical shell in treating the "burn-out" of Gd¹⁵⁵ and Gd¹⁵⁷ will give a value for the lifetime of these isotopes somewhat on the high side, although the difference should not exceed the errors involved.

In the present case the isotopic abundance is the sum of the abundances of the isotopes of mass 155 and 157, viz. 0.305 (Table VII); also, ρ_{el} will be less than the oxide density by a factor — equal to the ratio of atomic weights for gadolinium to one half the molecular weight of the oxide. Applying the equation (B7') the irradiation lifetime is given by

 $g_{un}T_{155+157} = 0.40 \div 0.03 n/kb.$

The results with samarium indicate that the effective integrated flux should be 0.31 \pm 0.04 n/kb. For this irradiation period the appropriate abundance to be used for Gd¹⁹⁶ is $n_{156} + n_{155}$, and, for Gd¹⁹⁶, $n_{158} + n_{157}$.

7. Dysprosium

The density of loosely-packed dysprosium oxide will be taken as 1.4 ∓ 0.1 gms/cc in line with measurements referred to in the previous section. Since only 2 mg. of the oxide were irradiated, the sample volume is .0014 gms/cc which would occupy only 0.1 cm. of the container.

This material would occupy the end of the hole in the container, which would have the conical shape typical of a drill tip, since the capsules were too long to lie on their side. Treating the sample as a hemisphere (Sec. 10.4 of reference 57) will therefore give a more accurate picture than other models available, such as a sphere or oblate spheroid.

A value of 0.165 \neq 0.015 is obtained for Za, using a radius of 0.075 cm. and an elemental capture cross section of 1100 \neq 150 barns ³⁸. The collision probability for this range of Σ a values lies within the limits of 0.072 and 0.088, giving an initial flux depression of 0.92 \neq 0.01. Most of the 1100 barn cross section is due to Dy^{104} , which is considerably depleted by the end of the irradiation, so that it is necessary to estimate from the mass spectrometric data the final value of σ_{c} per residual atom. Using preliminary values for the isotopic cross sections, and assuming that all the dysprosium lost through β -docay is retained as Ho¹⁰⁵ with a cross section of 64 barns ³⁸ the value for σ_{c} at the end of the irradiation is about 500 barns per rare earth atom. Since P_{c} is changing linearly with Σ a for the magnitude of concern here, and hence with σ_{c} , the final flux depression

may be taken as $0.96 \neq 0.01$. Using the mean of the initial and final values, the effective integrated flux for the dysprosium irradiation is $0.47 \neq 0.04$ neutrons per kilobarn.

8. <u>Ytterbium</u>

Irradiation conditions - chemical form, mass, density and capsule - were identical with those for dysprosium. Since the elemental cross section is less than 100 barns 3^8 it may be assumed that flux depression was negligible. The effective integrated flux is therefore taken as 0.50 ± 0.04 neutrons per kilobarn.

REFERENCES

¥ 1

- 1. W. Bothe Z. Naturforsch 1, 179 (1946) (Cross sections reported in this paper are based on a value of 49 barns for the capture cross section of Holes.)
- 2. L. Seren, H.N. Friedlander and S.H. Turkel Phys. Rev. <u>72</u>, 888 (1947).
- 3. L. Yaffe, R.C. Hawkings, W.F. Merritt and J.H. Craven -Phys. Rev. 82, 553 (1951) L.
- 4. H. Pomerance and J.J. Hoover Phys. Rev. 73A, 1265 (1948).
- 5. H. Pomerance Phys. Rev. <u>76A</u>, 195 (1949).
- 6. H. Pomerance Phys. Rev. 88, 412 (1952).
- 7. R.E. Lapp, J.R. Van Horn, and A.J. Dempster Phys. Rev. <u>71</u>, 745 (1947).
- R.J. Hayden, J.H. Reynolds and M.G. Inghram Phys. Rev. <u>75</u>, 1500 (1949).
- 9. D.C. Hess and M.G. Inghram Phys. Rev. 76, 300 (1949) L
- 10. J. MacNamara and H.G. Thode Phys. Rev. 80, 296 (1950).
- 11. W.H. Walker M. Sc. Thesis (physics) McMaster University, Hamilton, Ontario (1952).
- 12. W.H. Walker and H.G. Thode Phys. Rev. 90, 447 (1953).
- 13. A.W. Boyd, F. Brown and M. Lounsbury Can. J. Phys. 33, 35 (1955).
- 14. Unclassified Am. Report M15C64 (AECL Library, Chalk River).
- 15. M.G. Inghram, J.H. Reynolds Phys. Rev. 78, 822 (1950).
- 16. E.A. Melaika, M.J. Parker, J.A. Petruska and R.H. Tomlinson Can. J. Chem 33, 830 (1955).
- 17. J.A. Petruska, E.A. Melaika and R.H. Tomlinson & Can. J. Phys. 33, 640 (1955).

REFERENCES (Cont'd)

- 18. R.J. Horsley M. Sc. Thesis (physics) McMaster Univ., Hamilton, Ontario (1950).
- 19. A.J. Dempster Proc. Am. Phil. Soc. 75, 755 (1935).
- 20. A.J. Dempster Phys. Rev. <u>51</u>, 67 (1937)
- 21. J. Mattauch Phys. Rev. 50, 617 (1936), 1089 (1936) Errata
- 22. A.E. Shaw MDDC 308, (Atomic Energy Commission (U.S.A.) (1946)
- 23. J.L. Hundley Phys. Rev. <u>30</u>, 864 (1927)
- 24. E.B. Jordan and N.D. Coggeshall J. Appl. Physics 13, 539 (1942)
- 25. N.D. Coggeshall J. Chem. Phys. <u>12</u>, 19 (1944), Phys. Rev. <u>71</u> 482 (1947)
- 26. M.G. Inghram Phys. Rev. 70, 653 (1946)
- 27. H.G. Thode, J. MacNamara, F.P. Lossing and C.B. Collins -J. Amer. Chem. Soc. <u>70</u>, 3008 (1948)
- 28. R.M. Reese and J.A. Hipple Phys. Rev. 75, 1332 (1949)
- 29. G. Careri and G. Nencini Nuovo Cim 7, 64 (1951), J. Chem. Phys. <u>18</u>, 897 (1950).
- 30. C.E. Berry, Phys. Rev. <u>78</u>, 597 (1950)
- 31. O.A. Schaeffer J. Chem. Phys. <u>18</u>, 1681, (1950(L
- 32. J. Mattauch Z. Naturforsch <u>6a</u>, 391 (1951)
- 33. A.O. Nier Phys. Rev. 77, 789 (1950), 79, 450 (1950)
- 34. H.J.J. Braddick "The Physics of Experimental Method", John Wiley & Sons, New York, 1954, - Chapter I.
- 35. W.N. Bond "Probability and Random Erross", Edward Arnold & Co., London, 1935 p. 64
- 36. H. Pomerance Phys. Rev. <u>83</u>, 641 (1951)
- 37. S.P. Harris, C.O. Muchlhause, S. Rasmussen, H.P. Schroeder and G.E. Thomas - Phys. Rev. 80, 342 (1950)

- 38. D.J. Hughes and J.A. Harvey "Neutron Cross Sections" BNL325 (Brookhaven National Laboratories) (July 1, 1955)
- 39. M.G. Inghram, R.J. Hayden and D.C. Hass, Jr. AECD 2759 (1950) (Atomic Energy Commission, U.S.A.)
- 40. F.D.S. Butement Phys. Rev. 75, 1276 (1949)
- 41. M.G. Inghram and R.J. Hayden Phys. Rev. 71, 130 (1947)
- 42. W.E. Duckworth, R.F. Black and R.F. Woodstock Phys. Rev. <u>75</u> 1616 (1949)
- 43. W.T. Leland Phys. Rev. 76, 992 (1949)
- 44. J. Mattauch and H. Scheld Z. Naturforsch 3a, 105 (1948)
- 45. R.F. Hibbs Y 648 (Carbide and Carbon Chemicals Corp. Y-2 Plant (U.S.A.) (1950)
- 46. M.G. Inghram, R.J. Hayden and D.C. Hess, Jr. Phys. Rev. <u>73</u> 180, (1948)
- 47. W.T. Leland Phys. Rev. <u>77</u>, 634 (1950)
- 48. D.C. Hess, Jr. Phys. Rev. <u>74</u>, 773 (1949)
- 49. M.G. Inghram, R.J. Hayden and D.C. Hess, Jr. Phys. Rev. <u>75</u> 693, (1949)
- 50. R.J. Hayden, D.C. Hess, Jr. and M.G. Inghram Phys. Rev. <u>75</u> 322 (1949)
- 51. D.J. Littler (report on U.K. work) A/Conf. 8/P/432 (Geneva, 1955)
- 52. D.C. Dunlavey and G.T. Scaborg Phys. Rev. 92, 206 (1953)
- 53. E. der Mateosian, M. Goldhaber and A. Smith ANL 4237 (Argonne National Laboratories, Chicago) 64, (1949)
- 54. M. Goldhaber and C.O. Muchlhause ANL4010, 55, (Argonne National Laboratories, Chicago) (1947)
- 55. C.H. Westcott J. Nuclear Energy 2, 59 (1955) (Last term of equation 11a)

- 56. H. Atterling, E. Bohr and T. Sigurgeirsson Arkiv. Mat. Astron. Fysik, <u>32A</u> No. 2 (1946). Cross sections reported in this paper are based on a value of 2400 barns for the cross section of Dy¹⁰⁴.
- 57. K.M. Case, F. de Hoffmann and G. Placzek "Introduction to the Theory of Neutron Diffusion - Vol. I" - U.S. Gov't. Printigg Office, Washington, D.C.
- 58. S.A. Kushneriuk and D. McPherson CRT 619 (Atomic Energy of Canada, Chalk River) (to be published)