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ABSORBER BURN-UP EXPERIMENT

Final Report

By

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April 1962

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VALLECITOS ATOMIC LABORATORY

GENERAL ELECTRIC

ATOMIC POWER EQUIPMENT DEPARTMENT

SAN JOSE, CALIFORNIA

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ABSTRACT

Experimental results are presented of pile oscillator measurements of cross section of irradiated samples of seventeen elements. Mass spectrometer analysis of some samples was performed. Quantitative theory-experiment correlation yields some new thermal absorption cross section information and also establishes the reliability of control rod lifetime calculations for each element investigated. Application of the results to calculation of the nuclear lifetime of reactor control rods is discussed.

I. INTRODUCTION

This document is the final report of results of the absorber burn-up experimental program. The purpose of the program was to provide information about the change of effective cross section of reactor control element materials due to transmutation by neutron capture. The method used was to measure the reactivity effect, by the pile oscillator technique in a low power reactor (the Nuclear Test Reactor), of small samples of interesting control materials which have been subjected to varying amounts of neutron irradiation in a high flux engineering test reactor (the General Electric Test Reactor).

The samples were irradiated in the reflector of the GETR and hence in a thermal spectrum. The reactivity measurements were made in the center of the NTR with a more complex spectrum. In order to provide some spectral information, and to separate resonance and subcadmium cross section effects, reactivity measurements were also made with samples oscillated inside a fixed cadmium tube in the NTR core.

The results of the experiment consist of reactivity effect in the NTR (both bare and cadmium-covered) vs. exposure in GETR for each sample. Theory-experiment correlation consists of the correlation of the nvt behavior of sample reactivity with basic isotopic cross sections. Some cross section information was obtained by mass spectrometer analysis of selected irradiated samples.

Based on the results of these experiments, a set of curves has been computed for the relationship between the cross section of an absorbing element and the number of neutrons (per atom) captured by the element. This information permits calculation of the nuclear lifetime of a control rod⁽¹⁾.

The final report duplicates the description of experimental techniques presented in the interim report⁽²⁾.

This is followed by a discussion of theoretical models for comparison with experiment; tabulation of data and discussion by element of the conclusions which can be drawn from the data obtained on this program; and a description of a method for using the results of this experiment for prediction of control element burn-up.

II. EXPERIMENT

The experiment consists of measuring the change in macroscopic absorption cross section of various chemical elements due to irradiation in the GETR. The elements to be investigated include most of those with thermal absorption cross section greater than ≈ 20 barns. Test samples are cylinders, 1/4" in diameter and 2" long. Most of the absorbers are diluted in glass* to provide samples of three different self-shielding factors for each element (corresponding to $\Sigma_a t$ of about 0.2, 1, and 5). Several samples of each concentration of each element are welded into individual 3/8" O.D. by 2-1/4" long aluminum capsules. One sample of each concentration is set aside as a standard for comparison, and one or more of the rest are irradiated for one or more cycles in the GETR.

The capsules are loaded, end to end, in an aluminum tube (basket) during irradiation. The basket position is in a GETR pool facility adjacent to the core. Peak fluxes in this location are about 10^{14} nv. Sixteen capsules fill the 36" tube. (The reactor core is 36" tall.) The flux is monitored by aluminum-cobalt wires which run the length of the basket equally spaced around the circumference of the samples. These are γ -scanned to determine axial flux distribution, and a central section of each wire is analyzed to determine absolute nvt.

After each irradiation, the hot samples are transported to the NTR. Each sample, in turn, is inserted into the core region with an 18-ft. rod 7/8" in diameter.

* Samples prepared by Research and Development Division of Corning Glass Works, Corning, New York.

The rod has an accessible sample position in which a standard cold sample can be placed. The rod is mechanically symmetrical about the point midway between the cold and hot sample positions. A measurement consists of oscillating the rod between two positions such that the cold and hot sample positions alternately occupy the point of maximum flux in the reactor; and recording the variation in power level due to this oscillation. A second experiment is performed in which the samples are oscillated in a 20-mil, fixed cadmium sleeve in the same core location. The square-wave motion is accomplished by an air cylinder attached to the end of the rod. The frequency of oscillation is $1/20$ cycle per second. The power level variation is measured by two independent detection and recording channels. Timing of the entire system is based on a single instrument to eliminate instrument drift phase shifts between the two recording channels and the sample motion.

A data channel consists of a γ -compensated ion chamber, a bucking current source, a linear DC amplifier, a filter (i.e., a one-second R-C integrator), an analogue-to-digital voltmeter, and an automatic paper tape punch. The current that is amplified and recorded is the difference between the output of the ion chamber and a constant DC bucking current. This arrangement permits detecting small fractional power changes. The system samples and records the signal once each second, i.e., 20 times per cycle. A data run consists of about 25 cycles. Header information is added to the data tapes which are then converted to IBM cards for machine analysis.

The daily calibration of data channel consists of replacing the ion chamber with a variable current source. Differential measurements permit conversion of an observed change in bucked voltmeter reading to fractional change of unbucked ion chamber current. The number provided by the calibration (for each scale of the DC amplifier) is designated by z on the following diagram.

From the diagram we obtain the following relationship:

$$\frac{\Delta I}{I} = \frac{\Delta V}{z + A} \quad (1)$$

In summary, the raw data from this experiment consist of wire traverses of the GETR flux near the irradiated samples; tapes representing flux level variation with oscillation of the samples, both with and without the cadmium sleeve in the reactor; and a z-value, or calibration, for each amplifier scale.

A. Equipment and Procedure

During the two-year course of this experiment there has been a gradual evolution of experimental equipment and technique. The most significant change in procedure was made possible by installation of a hot cell (independently of this program) at the

end of the NTR thermal column. Conversion to automated data handling (using a paper tape system, rather than a digital typewriter output) has considerably eased data reduction.

The following is a description of the hot sample handling procedures, both before and after installation of the hot cell; the pile oscillator instrumentation; and the flux monitoring system for measuring sample exposure in the GETR.

1. Hot Sample Handling

During the first 12 months of operation a special cask was used to handle the irradiated samples. The 16 hot samples, irradiated in GETR each cycle, were loaded into lead lined aluminum holders (Figure 1) and inserted into the special cask. The central portion of this 3-ton cask consists of a revolving lead drum with 16 slots in it (Figure 2). The loaded cask was positioned horizontally (supported by a steel dolly) adjacent to the thermal column end of the NTR. The offset access hole of the cask and the horizontal "glory hole" of the NTR were aligned. A push rod having a cavity at one end (Figure 3) with an unirradiated sample, were attached to the sample holder containing the irradiated sample through the access hole. This assembly was then pushed into the "glory hole" and the other end of the rod was attached to an air driven cylinder, then the oscillator actuated. The length of the oscillator stroke was the same as the separation of the center points of the samples. During the oscillation the samples alternately occupied the point of maximum flux in the reactor. The assembly (push rod and sample holder) was mechanically symmetrical about the point midway between the standard and irradiated sample positions. The variation of flux level produced by the oscillation was recorded by two independent detection and recording channels.

The above procedure was used for all 16 samples in the cask. Each

Figure 2. ABSORBER BURN-UP CASK NO.1

Figure 3. PUSH ROD

sample was measured twice, once with an aluminum insert in the "glory hole" and once with a cadmium insert. During a typical day's operation, 16 measurements were made and the instrumentation calibrated twice.

All sample holders were oscillated without samples, with and without the cadmium liner, in order to obtain the holder background. The holder background for the bare measurements was quite reproducible. However, it was erratic in the cadmium sleeve measurements. In spite of considerable effort, the trouble was never completely eliminated, and therefore all cadmium-covered data taken prior to the change in procedure were discarded. This is not a total loss since the samples are still available for measurement.

In September of 1960 a shielded experimental room was completed at the end of the NTR thermal column. The wall separating the control room from the experimental room is 8 feet high, 26 inches deep, made of high density concrete bricks and lined on both sides with 1/8" steel sheets. Two "master-slave" remote manipulator units are installed over this wall, an auto crane is positioned next to them and the experimental area is sealed. These modifications of the facility made it possible to change the experimental procedure and setup.

Under the procedure now followed, the 16 irradiated samples are brought to the NTR on a rack in a standard cask (Figure 4). The cask is positioned under the slave portion of the manipulators and the lid removed with the auto crane. The rack with hot samples is picked up by remote manipulators and lowered down a chute leading to a small enclosure at the face of the NTR thermal column. This operation is facilitated by adjustable mirrors positioned in strategic locations. The enclosure is made of stacked lead bricks. The front wall is 8 inches wide with a 8 x 8 x 8 inch lead glass

Cask Wt. = 3200 Lbs.

Figure 4. ABSORBER BURN-UP CASK #2 OUTLINE

window (sp. gr. 6.4) and the back wall is lined with mirrors. A table inside the enclosure supports the sample rack, a flood light, and a hair dryer which is used to dry the irradiated samples in case they are wet. Inside the enclosure the samples are handled by means of a flexible "pick up tool" which is mounted through a castle mount on top of the enclosure. A homogeneous (95 percent magnesium, 3 percent aluminum, 1 percent zinc) push rod passes through a $3/4$ inch hole in the front wall of the enclosure. Two slots in this push rod accommodate the two samples to be oscillated. The standard sample is loaded into one of the slots outside the enclosure and the irradiated sample is inserted inside the enclosure. The rod is then connected to the air cylinder and the oscillations are performed.

This second procedure proved to be very satisfactory. Uncertainties in the cadmium-covered data are reduced to tolerable size. Figure 5 shows experimental setup No. 1. Figure 6 shows experimental setup No. 2.

2. Instrumentation

The desired result of the measurement with the pile oscillator is the variation of power level due to sample oscillation divided by the average power level during oscillation. The signal used in this report as a measure of the reactivity effect of the sample is the coefficient of the $1/20$ cycle per second harmonic of the ion chamber output divided by the average ion chamber output. Ion chamber output is measured by two independent detection and recording channels (Figure 7). Two γ -compensated ion chambers are inserted into the 4" x 4" vertical cavity (Figure 8). The signals obtained from the CIC's are fed into two micromicroammeters. A large portion of the signal is bucked out by bucking current sources. The difference between the output of each ion chamber and the constant DC bucking current is amplified and recorded. Two analogue-to-digital voltmeters measure the signals and the information is punched once

Figure 5. EXPERIMENTAL SET-UP #1

Figure 6. EXPERIMENTAL SET-UP #2

Figure 7. INSTRUMENTATION CHANNELS

Figure 8. CORE ARRANGEMENT

per second on paper tape using an 8-hole code. A line frequency controlled clock times both channels, the tape punch, and the motion of the pile oscillator. The use of the single clock assures identical timing for both channels, reproducible oscillation periods, and reproducible phasing.

A stable voltage divider which is battery supplied provides the voltage in each bucking current source. One channel is continuously controllable and the other has a step control so that a large number of reactor powers can be chosen and so that the second channel can be set to make the micromicroammeters read zero simultaneously. The system is calibrated before and after each day's operation. The calibration is done by replacing the current from each compensated ion chamber by an independently measured battery supplied current source. Several scales on each micromicroammeter are calibrated by varying the current from the current source.

After each measurement additional information is punched on the paper tape to identify the oscillation and then it is used to punch IBM cards. The data are then Fourier analyzed by a computer.

3. Integrated Neutron Flux Exposures

The activation of cobalt is used to monitor the integrated neutron flux exposure for each set of 16 samples irradiated in the GETR. Three 38-inch long, 30-mil diameter, 0.010 w/o cobalt-aluminum wires are loaded into the sample irradiation basket (Figure 9) along with the samples to be irradiated. Following the completion of a cycle the wires are transferred from the basket into aluminum tubes and are then γ -counted using a collimated sodium iodide crystal. An activity profile of each wire is determined by measuring the count rate at 1-inch intervals along the entire length of the wire. A set of typical profile curves is illustrated in Figure 10.

All Material 6061 - T6 Aluminum Except As Noted

Figure 9. ABSORBER BURNUP EXPERIMENT IRRADIATION BASKET

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Figure 10. TYPICAL FLUX WIRE ACTIVITY

One-inch sections are then cut from the peak activity region of each wire. These sections are counted on a 256 channel gamma spectrometer and compared with a NBS cobalt standard in order to determine the absolute Co^{60} activity. From these results integrated neutron flux values are calculated.

Figure 11 shows the radial locations of GETR experimental facilities. Table 1 lists the GETR cycle numbers, the basket numbers used for control rod materials irradiation, basket position in the reflector region, dates and effective full power (30 MW) days of each cycle, and the exposure received by each sample from each cycle.

B. Reduction of Balanced-Pile-Oscillator Data

The output of the data channels depends upon the time-dependence of the position in the core of the hot sample, the cold sample, and the holder-and-rod assembly. Because the calibration is in terms of fractional power, the signal does not depend upon power level of the core (although pile noise considerations lead to restrictions on minimum useable power level). To analyze the results of the experiment, two assumptions must be made. First, it is assumed that the effect on signal of each of the 3 components, hot sample, cold sample, and holder, are linearly additive. Second, it is assumed that the effects on signal of the hot sample and the cold sample are proportional to the number of neutrons they absorb, though not necessarily with the same constant of proportionality.

The time dependence of the signal can be expanded in a Fourier series such that

$$\frac{\Delta I(\omega t)}{I} = \frac{V(t)}{z + A_{c-h}} = \frac{1}{z + A_{c-h}} (B_{c-h} \sin \omega t + C_{c-h} \cos \omega t + \dots). \quad (2)$$

The linearity assumption permits writing:

Figure 11. RADIAL LOCATIONS OF G.E.T.R. EXPERIMENTAL FACILITIES

$$V(t) = \sum_h f_h (\beta_x \sin \omega t + \gamma_x \cos \omega t + \dots) + \sum_c f_c (\beta_s \sin \omega t + \gamma_s \cos \omega t + \dots) + \sum_H f_H (\beta_H \sin \omega t + \gamma_H \cos \omega t + \dots) \quad (3)$$

where A_{c-h} , B_{c-h} , and C_{c-h} are the constant, the coefficient of the sine term and the coefficient of the cosine term, of the Fourier expansion of $V(t)$ for the case of sample C in the s-position of the holder, and sample h in the x-position of the holder. $\sum_y f_y$ is the macroscopic absorption cross section times the self-shielding factor for sample y. β and γ are constants.

From Eqs. (2) and (3):

$$\begin{aligned} B_{c-h} &= \sum_c f_c \beta_s + \sum_h f_h \beta_x + \sum_H f_H \beta_H \\ C_{c-h} &= \sum_c f_c \gamma_c + \sum_h f_h \gamma_x + \sum_H f_H \gamma_H \end{aligned} \quad (4)$$

For this analysis the Fourier expansion will be limited to the fundamental frequency since the information content of the higher harmonics is very small. Because the terms of a Fourier expansion form a complete orthogonal set, neglect of the higher order terms does not affect the analysis of the fundamental.

Because of the similarity of B and C, only B will be followed through the analysis. The expression for B can be re-arranged into an indefinite number of equivalent representations. The following appears to be the most useful for our purposes:

$$B_{c-h} = \beta_x (\sum_c f_c - \sum_h f_h) + \sum_c f_c (\beta_s + \beta_x) + \sum_H f_H \beta_H \quad (5)$$

The first term in Eq. (5) is the useful component of the signal, i.e., that component which is proportional to the difference between the number of neutrons absorbed by the hot and cold samples. The last term

is the contribution of the holder. The central term is called the asymmetry contribution. The latter terminology can be appreciated by examining Eq. (3). β_s must be approximately equal to $-\beta_x$ because the two samples are oscillating 180° out of phase across a nearly symmetrical flux distribution. If the detector locations were symmetrically located, and the flux distribution perfectly symmetrical about the peak flux location, then $(\beta_s + \beta_x)$ would be zero, and there would be no asymmetry contribution.

Because different scales, hence different z , may be used to measure signals of different size, it is necessary to define a normalized B' which is proportional to $\Delta I/I$ rather than V . This is accomplished by defining $\beta' = \beta/(z + A)$ such that $B' = B/(z + A)$. Eq. (5) then becomes

$$B_{c-h}' = -\beta_x'(\sum_c f_c - \sum_h f_h) + \sum_c f_c(\beta_s' + \beta_x') + \sum_h f_h \beta_h' \quad (6)$$

The useful component of Eq. (6) is then:

$$b_{c-h} = B_{c-h}' - \sum_h f_h \beta_h' - \sum_c f_c(\beta_s' + \beta_x') = -\beta_x'(\sum_c f_c - \sum_h f_h) \quad (7)$$

The rod contribution to the signal, $(\sum_h f_h \beta_h')$, is the observed signal with no samples. The asymmetry term is the observed signal less the holder signal with identical samples in the cold and hot positions. The asymmetry correction, from Eq. (6) is proportional to the strength of the sample in the cold position. The procedure followed is to measure the asymmetry correction for a few pairs of identical samples, and to correct the rest of the data in proportion to the strength of the sample in the cold position for each measurement. There is, of course, no asymmetry correction for the cases which have a pure glass sample in the cold position. For these experiments the asymmetry correction is between 7 and 12 percent of the cold sample worth depending upon detector location in the core.

A similar development for the coefficient of the cosine component, c_{c-h} , leads to

$$c_{c-h} = C_{c-h}' - \sum_H f_H \gamma_H' - \sum_c f_c (\gamma_s' + \gamma_x') \quad (8)$$

The signal used in analysis of data is taken to be

$$d_{c-h} = \sqrt{(b_{c-h}')^2 + (c_{c-h}')^2} = (\sum_c f_c - \sum_h f_h) [(\beta_x')^2 + (\gamma_x')^2]^{1/2}, \quad (9)$$

where b_{c-h}' and c_{c-h}' are the b_{c-h} and c_{c-h} corrected for asymmetry.

The above analysis pre-supposes a Fourier expansion of the time dependence of the ion chamber output. This expansion is performed by FOURANAL, a Transac 2000 program. The following is a description of the operations performed upon the data to obtain A, B, and C of Eq. (2).

The first operation by the computer is to average each point in the cycle by summing every Nth data point and dividing by the number of cycles. This reduces all of the data to one average cycle ($N = 20$, for this experiment).

The Fourier expansion of such a function is not rigorously defined unless the behavior of the function between the discrete data points is specified. Rather than specify this behavior as an assumption, it is assumed that the following approximations are valid.

$$A = \frac{1}{N} \int_0^N V(t) dt \approx \frac{1}{N} \sum_{i=0}^{i=(N-1)} V_i \quad (10)$$

$$B_r = \frac{2}{N} \int_0^N V(t) \sin\left(\frac{2\pi}{N} t\right) dt \approx \frac{2}{N} \sum_{i=0}^{i=(N-1)} V_i \sin \frac{2\pi i}{N} \quad (11)$$

$$C_r = \frac{2}{N} \int_0^N V(t) \cos \left(\frac{2\pi}{N} t \right) dt \approx \frac{2}{N} \sum_{i=0}^{i=(N-1)} V_i \cos \frac{2\pi i}{N} \quad (12)$$

Inherent in these equations is the assumption that a cycle is N time units long.

The sums of Eqs. (10), (11), and (12) are included in the output of FOURANAL.

B_r and C_r must be corrected for ramp (systematic deviation of the average reactor power level) before they can be utilized in Eqs. (2) through (9). Included in FOURANAL output is the average value of the data over the first cycle and the last cycle. From this can be calculated R , the average increase in average power between data samplings (the increase in one second, for this experiment). If there were no signal, but only ramp, FOURANAL would yield a value for B_r and C_r . The derivation of the correction consists of calculating what this would be and subtracting. This leads to

$$\begin{aligned} B &\approx B_r + \frac{N}{\pi} R \\ C &\approx C_r + R \end{aligned} \quad (13)$$

This correction is usually less than one percent.

In summary, reduction of a tape to a quantity, d_{c-h} , proportional to the difference between the number of neutrons absorbed by the cold standard and the number absorbed by the hot sample includes:

- (a) FOURANAL analysis for A , B_r , and C_r .
- (b) Ramp correction of B_r and C_r to B and C by Eq. (13).
- (c) Normalization of B and C to B' and C' by dividing by the appropriate $z + A$.

(d) Correction of B' and C' for holder worth and asymmetry.

(e) Combine sine and cosine contributions in Eq. (9) to obtain d_{c-h} .

d_{c-h} , from the derivation, is proportional to the difference between the number of neutrons captured by the samples in the cold and hot positions. Small changes in absorption cross section can be measured directly in the balanced pile oscillator by making the measurements with an unirradiated sample in the standard position. This is preferable to measuring each sample independently to obtain the small difference of two large numbers. Conversely, after appreciable irradiation, best accuracy is obtained by using the oscillator in the conventional fashion without the balancing feature. Also, when accuracy is limited by background considerations, as is usually the case for the cadmium-covered runs, the balancing feature does not improve accuracy.

III. DISCUSSION OF THEORY

Interpretation of results consists of comparing NTR signal with the predictions of theory. A complete calculation of all effects is beyond the present state of the art. Therefore the theory-experiment correlation is a measure of the validity of the mathematical approximations used in the theory as well as the reliability of the fundamental cross section information used in calculation.

The calculational model must account for production and destruction of isotopes in the sample by neutron capture and radioactive decay. From the calculated isotopic composition of the sample at the time of measurement, the NTR signal must be calculated.

The general equation which described the concentration of the i^{th} isotope, N_i , in the experimental sample is:

$$\frac{dN_i}{dt} = \sum_j \sum_k \left(\int_0^\infty dE \sigma_{j \rightarrow k} \phi + \lambda_{j \rightarrow k} \right) (\delta_{ik} - \delta_{ij}) N_j \quad (14)$$

where: N_i = position dependent concentration of the i^{th} isotope

$$\delta_{ij} = \begin{cases} 0 & \text{for } i \neq j \\ 1 & \text{for } i = j \end{cases}$$

$\lambda_{j \rightarrow k}$ = spontaneous decay constant for isotope j to k

$\sigma_{j \rightarrow k}$ = energy dependent neutron cross section for formation of isotope k from isotope j

ϕ = time, space and energy dependent neutron flux in sample.

Solution of Eq. (14) requires a burn-up dependent description of the neutron flux in space and energy in the sample. However, the most detailed equation that can be solved, at present, with reasonable computer and programming expense does not include such a description of neutron transport represented simultaneously with isotopic burn-up.

If the assumption is made that the entire sample experiences the same flux, then Eq. (14) reduces to the tractable form, Eq. (15), which is used to analyze the results of this experiment. In Eq. (15), the flux is multiplied by the factor f (which varies the sample cross section) to account for the difference between the flux of interest in Eq. (15), which is the average flux in the sample, and the flux measured in the experiment by the cobalt wires which were outside the sample almost one-half inch from the sample surface. Eq. (15) is solved by VARI-II, a Fortran computer code⁽³⁾.

$$\frac{dN_i}{dt} = \sum_j \sum_k \left(f \sigma_{j \rightarrow k} \phi + \lambda_{j \rightarrow k} \right) (\delta_{ik} - \delta_{ij}) N_j \quad (15)$$

where: ϕ = flux at cobalt wire monitor as determined by cobalt activation

$$\sigma_{j \rightarrow k} = \frac{\int_0^{\infty} \phi(E) \sigma_{j \rightarrow k} dE}{\int_0^{\infty} \phi(E) dE}$$

f = self-shielding factor

Eq. (15) can be an adequate model for analysis of the dilute (thin to

neutrons) samples of this experiment. The time variation of the flux is important only for isotopes which have spontaneous decay rates comparable to neutron capture transfer rates. If the time decay is very fast ($\lambda \gg \phi \sigma$), then it can be assumed instantaneous with small error. If it is very slow, it can be assumed stable. For these two situations, the time dependence of ϕ is unimportant. There are a few samples in this experiment bearing isotopes for which neither assumption is valid and these are discussed in the relevant section on analysis of data. Exposure, ϕt , is used as the independent variable in this report rather than time, t .

Important aspects of the history of the samples which must be considered are:

A. Neutron Spectrum of GETR at Sample Location

The samples are irradiated in the water reflector region of the GETR reactor near the location of the thermal flux reflector peak. The unperturbed spectrum in this region can be described as a Maxwellian at the water temperature plus a small $1/E$ tail. The water temperature in the pool depends upon weather and time of operation; however maximum variation is from about 26°C to 36°C. In the analysis, 0.0253 ev cross sections (20°C) are used both for calculating exposure and for calculating burn-up. This correctly handles isotopes with approximately $1/v$ cross sections in the thermal region. However, severely non- $1/v$ isotopic cross sections must be modified by the factor:

$$\frac{\int_0^{\infty} \phi(E) \sigma(E) dE}{\int_0^{\infty} \phi(E) \sigma(.025) \sqrt{\frac{.025}{E}} dE}$$

The minimum amount of epithermal absorption expected during irradiation would correspond to an infinite medium pure water spectrum. This would amount to 0.7 percent epicalcium absorption for a $1/v$ absorber. General arguments indicate that the actual spectrum should be much harder than

this minimum. Gold foil cadmium ratios⁽⁴⁾ indicate a value of about twice the minimum. This is further perturbed by the samples and basket. The episcadmium capture is neglected in computing sample burn-up. This approximation is seriously invalid for a few isotopes and is discussed in the relevant section on analysis of data.

B. Neutron Flux and Exposure

The definition of exposure is based on absolute counting of the cobalt wire activity, and conversion of this to nvt with a cobalt cross section of 37.0 barns. This is a commonly used definition of exposure. However, 37 barns is the 2200 mps cross section for cobalt and does not correspond to a spectral averaged cross section. The exposure as defined is:

$$E = \frac{1}{\sigma_a(2200 \text{ mps})} \int_{t_1}^{t_2} dt \int_0^{\infty} \phi(E, t) \sigma_a(E) dE \quad (16)$$

This definition of exposure can only be used consistently for calculating burn-up of absorbers which have the same energy dependence of absorption cross section as the monitor. In the analysis it is assumed that cobalt has a $1/v$ absorption cross section, since the irradiation was performed in a spectrum characterized primarily by the thermal component. It is estimated that about 2 percent of the absorption in a $1/v$ absorber in the GETR location used are above the cadmium cut-off. Since the resonance component of the episcadmium (>0.6 ev) resonance integral of cobalt is about three times that of the $1/v$ component⁽⁵⁾ the error in assuming the monitor to be a $1/v$ detector is about 6 percent.

The self-shielding model must determine the ratio of the average flux in the sample to the flux in the cobalt monitors near the sample. The model used is a P-3 cylindrical cell calculation with Maxwellian averaged cross sections. The calculations were for a large cell centered about the absorber. Results were obtained for a wide range of absorber cross sections. The calculated self-shielding factor was taken

to be the ratio of the average flux in the absorber region to the flux at the monitor radius. The range of calculated self-shielding parameters was fitted with a rational polynomial in $\Sigma_a r$ where Σ_a is the 2200 mps cross section and r the sample radius. The rational polynomial so obtained is:

$$f(\Sigma_a r) = \frac{1 + 0.3707 \Sigma_a r + 0.0049 (\Sigma_a r)^2}{1 + 1.2091 \Sigma_a r + 0.6802 (\Sigma_a r)^2} \quad (17)$$

C. Time of Irradiations and Measurements

Any amount of detail in time dependence of flux can be included in the solution of Eq. (15). However, for most samples the results can be adequately described by exposure (ϕt), and time dependence of ϕ is unimportant. For the cases where half lives and $1/\phi\sigma$ are in the range of a few days, the uncertainty in ϕ can introduce significant error. The flux at the sample location in the GETR varied by as much as a factor of 2 during a single irradiation cycle because of the gradual withdrawal of control rods to compensate for U-235 burn-up. Further, the self-shielding in many samples varied significantly during irradiation. The cobalt wire monitor is a measure of only the integrated neutron dose for the run. Therefore little information is available about the time dependence of the impinging flux. However, the flux depression in the sample was accounted for by the f factor in Eq. (15).

D. Neutron Spectrum of NTR at Location of Measurement

The spectrum in the NTR at the location of the measurement is not markedly harder than the irradiation spectrum. The spectrum is characterized by about 4 percent epicadmium absorption (>0.6 ev) by a $1/v$ absorber.

The measurements performed in the cadmium tube provide a measure of resonance capture. The effective low energy cut off for the tube is about 1 ev⁽⁶⁾. The difference between bare and cadmium tube data corresponds to a spectrum with a high energy cut off at about 1 volt.

The operating temperature of the NTR is $38 \pm 2^{\circ}\text{C}$.

E. Self-shielding of the Sample During Both Irradiation and Measurement

Within the model limitation of uniform flux in the sample, two kinds of self-shielding must be considered. The difference between the average flux in the sample and the flux at the monitor during irradiation in the GETR is accounted for by calculation. The sample self-shielding during measurement in the NTR results in a non-linear relationship between signal and cross section. This relationship is determined empirically for $1/v$ absorbers by measurement of the unirradiated boron samples. Although the measurement is not strictly applicable to non- $1/v$ absorbers, it is used with fair success on all samples.

IV. RESULTS

The experimental data and interpretation of results for each element investigated are presented together. To aid in discussion, each package has, besides a tabulation of the data, the relevant decay scheme taken from the tri-linear chart⁽⁷⁾; the cross sections and decay constants used to calculate the theoretical burn-up; and a graphical presentation of data and theory.

A quantitative analysis of the sub-cadmium data (bare minus cadmium tube signal) is performed using 2200 mps cross sections modified by non- $1/v$ factors where appropriate.

The procedure followed is:

1. Assume isotopic cross sections.
2. With VARI-II, calculate elemental cross section versus the integrated sample nvt. This calculation requires as input the isotopic cross sections, sample self-shielding equation coefficients, sample initial atom densities. The output includes the integrated

sample nvt, the isotopic composition of the sample vs. exposure, as well as the microscopic and macroscopic sample cross sections.

3. The calculated macroscopic cross section could be converted to sub-cadmium signal via the boron calibration curve, and for the most straight forward comparison of theory and experiment, the data (sub-cadmium signal versus wire nvt) could be plotted on a graph of the calculated sub-cadmium signal versus wire nvt. (This is actually done for the boron samples). However, the results would not be simply related to cross section. Also, for several elements, data were obtained for samples of different initial atom densities. In order to graphically present the data in terms of cross section vs. exposure the following procedure is followed:

- (a) The experimental sub-cadmium signal is used to obtain an experimental macroscopic cross section (and hence microscopic cross section) via the boron calibration curves. For unirradiated samples, this cross section should agree with the published 2200 mps cross section for the element except for the non- $1/v$ factor applicable to the sub-cadmium cross section as measured in the NTR. To compensate for this non- $1/v$ factor, the experiment cross sections of the irradiated samples are multiplied by the ratio of the published 2200 mps cross section of the element to the experimental cross section for the unirradiated sample. Obviously, this procedure may obscure unknown effects. Therefore the multipliers used for the data are listed on each graph. In this type of plot the error in cross section is large for heavily self-shielded samples because the slope of the boron calibration curve (signal vs. cross section) is then small.

- (b) The experimental wire nvt is used to obtain an experimental sample nvt via the calculated curve of sample nvt vs. wire nvt.

(c) For comparison, the modified data (microscopic cross section versus sample nvt) are plotted on a graph of the calculated cross section vs. sample nvt.

4. Where mass spectrometer data are available, the predicted isotopic ratios are compared with the experimental ratios. This data provides information about individual isotopic cross sections. However, only in a few cases is the information cleanly interpretable.

After examining the results of the above comparisons, it is sometimes apparent that certain of the assumed isotopic cross sections are in error. These cross sections are modified accordingly and the procedure repeated. The choice of cross sections is discussed in detail with each element data package.

TABLE 1

SAMPLE EXPOSURE AND CYCLE HISTORY

nvt x 10²⁰ of sample for each cycle of GETR

GETR cycle #/MWD:	5/633	6/437	7/668	8/528	9/467	10/773	11/742	12/613	13/547	14/662	17/824	18-20/1422	21-23/1895
Start Date:	9/5/59	10/10/59	11/8/59	12/8/59	1/6/60	2/9/60	3/14/60	4/20/60	5/26/60	6/27/60	10/6/60	11/13/60	3/21/61
End Date:	10/2/59	11/3/59	11/30/59	12/28/59	2/1/60	3/7/60	4/10/60	5/15/60	6/19/60	7/22/60	11/5/60	2/5/61	6/4/61
Pool Pos./Bskt. #:	Y-6/1	Y-6/2	Y-6/3	Y-6/4	Y-6/5	Y-6/6	Y-6/7	Y-6/8	Z-11/9	Z-11/7	Z-3/8	X-5/9	X-5/9
SAMPLE													
CD-2	0.16	0.19		0.38		1.04					0.73		
CD-3			1.23										
CD-4							0.91						
DY-2	0.67	0.47		0.79		2.12		1.24			2.71	4.31	5.14
DY-3							1.92						
DY-2A					1.31				1.28	2.31		3.47	4.47
ER-2	0.29	0.40		0.70		1.95		1.47			2.75	4.25	5.38
ER-3					1.33								
ER-4							1.72						
EU-2	0.54	1.14		0.93		1.96		1.88			2.67	3.92	
EU-3							2.17						
EU-2A					0.77				1.49	2.04			
EU-2B									1.59	1.73		4.03	4.66
EU-2C											2.13		4.09
GD-2	0.43	0.26		0.49					1.64				
GD-3							0.58						
GD-2A					0.25			0.47					
GD-2B					0.30			0.61					2.44
AU-2			0.35						1.81				
HF-2		0.81		0.40		0.92		1.93				0.46	1.15
HF-2A			0.69					1.42			2.27	3.87	3.95

TABLE 1

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GETR cycle #/MWD:	5/633	6/437	7/668	8/528	9/467	10/773	11/742	12/613	13/547	14/662	17/824	18-20/1422	21-23/1895
Start Date:	9/5/59	10/10/59	11/8/59	12/8/59	1/5/60	2/9/60	3/14/60	4/20/60	5/26/60	6/27/60	10/6/60	11/13/60	3/21/61
End Date:	10/2/59	11/3/59	11/30/59	12/28/59	2/1/60	3/7/60	4/10/60	5/15/60	6/19/60	7/22/60	11/5/60	2/5/61	6/4/61
Pool Pos./Bskt.#:	Y-6/1	Y-6/2	Y-6/3	Y-6/4	Y-5/5	Y-6/6	Y-6/7	Y-6/8	Z-11/9	Z-11/7	Z-3/8	X-5/9	X-5/9
SAMPLE													
HO-2	0.30	1.0E		0.78		1.54			1.99	1.63		0.92	1.88
HO-3							1.96						
IN-2A			0.71					1.17			1.84	2.81	
IN-2B			0.29						1.90	0.95			
IN-2C					0.98				0.38				
LI-2A			1.03										
LI-3A					1.06				1.17	0.74		1.44	0.50
LU-2	0.67					2.11							
LU-3							2.17			2.26		2.11	2.78
SM-2	0.54	1.05		0.98		2.24							
SM-3							1.16						
SM-2A					0.96								
SM-2B						1.19		1.87					
AG-2			0.95										
AG-3					0.68			1.45			1.01		
AIC-2								0.84			1.45	2.65	3.81
TB-2											1.35	2.37	3.69
TM-2A							0.94			1.73		3.34	
YB-2					0.66								
YB-3							1.74			1.41		1.92	3.12

BORON, ${}_5^B$

Nuclear Data⁽⁸⁾

<u>Isotope (Abundance, $T_{1/2}^1$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element --	755 ± 4
B-10 (.198)	3813
B-11 (.802)	~ 0

Boron is the classical $1/v$ absorber. The reaction products of the B-10 capture (He-4 and Li-7) have negligible thermal absorption cross sections, so no important absorption chain is produced.

Natural boron is used as the cross section standard for interpretation of NTR signal. The experimental curve of sub-cadmium signal versus macroscopic 2200 mps cross section of the unirradiated boron samples permits relating all sub-cadmium signals to an equivalent $1/v$ cross section.

Boron is also used to determine the reliability of the experimental technique. By irradiating and measuring several boron samples, the experimental accuracy can be estimated, since the cross section of boron is known with greater accuracy than the precision of this experiment.

The samples were glasses of several B_2O_3 to SiO_2 ratios. Table 2 lists the results on unirradiated boron with the physical description of the samples. Fig. 12 is a plot of the bare signal vs. 2200 mps macroscopic cross section of the sample. Also in Fig. 12 is a plot of the sub-cadmium signal vs. 2200 mps macroscopic cross section. This is the curve used to relate signal and cross section for all analyses other than boron in this report. To improve reproducibility of small signal conversion, the equation

$$\text{sub-cadmium signal} = 4.302 \times 10^{-2} \Sigma - 8.65 \times 10^{-3} \Sigma^2 \quad (18)$$

was used for signals below <0.03 .

Figure 12. BORON CALIBRATION

The epicadmium data for the boron samples are plotted in Fig. 13.

The pyrex glass samples (4.05 w/o boron) were irradiated in several GETR cycles. The results are tabulated in Table 3.

The calculated signal is plotted with the experimental data in Fig. 14. The calculated values are based on the bare calibration curve of Fig. 12. The unshielded boron curve is plotted for comparison.

The self-shielding of these samples is large (initial $f = 0.423$), and varies significantly during irradiation. The agreement between theory and experiment for the boron samples implies that the self-shielding model is adequate.

The scatter of the data is used to estimate the experimental uncertainty. The error for small exposure is primarily due to signal while the error for large exposure is primarily due to exposure. The rms deviation of the experimental signal from the calculated signal for all points of less than 1×10^{20} nvt is 1.2 percent. Conversely, the scatter can be related to an uncertainty in the nvt determination. However, this is not a valid check for very low exposure. The rms deviation of the theory from the data for the 5 points above 2.0×10^{20} nvt is 6.5 percent.

Reactor noise, mechanical non-reproducibility, holder subtractions, and other unidentified sources of noise place the limit of reproducibility for small signals at about ± 0.0001 . This corresponds to a sample cross section of about 0.002 cm^{-1} . The various diluent glasses may differ in absorption cross section by about $\pm 0.002 \text{ cm}^{-1}$ from the standard glass sample. The uncertainty associated with each signal is taken to be:

$$\sqrt{(0.01 d)^2 + (.0002)^2}$$

The uncertainty in the cross section is this error divided by the slope of the calibration curve.

Figure 13. BORON CADMIUM COVERED SIGNAL

TABLE 2

BORON STANDARDS

Sample	Wt.(gm)	w/o B	Signal Bare	Signal Cd. Cov.	Σ_a Calculated
B-1	3.553	4.05	.0733 $\pm$.0007	.0051 $\pm$.0002	3.78
B-1A	3.849	0.934	.0328 $\pm$.0003	.0013 $\pm$.0002	0.94
B-1B	3.886	4.67	.0816 $\pm$.0008	.0061 $\pm$.0002	4.72
B-1C	3.676	11.2	.1026 $\pm$.0010	.0117 $\pm$.0002	10.8
BS-1D	3.797	0.61	.0227 $\pm$.0003	.0008 $\pm$.0002	0.60
BS-1E	3.881	2.03	.0547 $\pm$.0005	.0028 $\pm$.0002	2.10
BS-1F	3.909	3.25	.0706 $\pm$.0007	.0044 $\pm$.0002	3.38
BS-1G	3.900	4.07	.0765 $\pm$.0008	.0056 $\pm$.0002	4.16
BS-1H	3.760	8.12	.0936 $\pm$.0009	.0097 $\pm$.0002	8.00
BS-1I	3.264	20.4	.1126 $\pm$.0011	.0151 $\pm$.0003	17.9

Figure 14. IRRADIATED BORON

TABLE 3

BORON

DATA						CALCULATION			
Sample	Wt. (gm)	w/o B	nvt x 10 ²⁰ Wire	Signal Bare	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ sample	E_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
B-1	3.553	4.05 ↓	0	.0733 ± .0007	741 ± 54	0	3.78	.0738	755
B-2	3.559		0.504	.0702 ± .0007	677 ± 39	0.221	3.48	.0711	695
B-3	3.528		1.17	.0659 ± .0007	593 ± 29	0.530	3.09	.0675	617
B-4	3.583		0.724	.0695 ± .0007	663 ± 36	0.321	3.35	.0700	669
B-5	3.542		2.65	.0583 ± .0006	475 ± 21	1.30	2.30	.0582	459
B-6	3.543		3.73	.0506 ± .0005	365 ± 13	1.95	1.79	.0505	358
B-7	3.574		0.897	.0677 ± .0007	629 ± 33	0.402	3.25	.0691	649
B-8	3.585		3.82	.0496 ± .0005	352 ± 13	2.01	1.75	.0499	350
B-9	3.571		3.35	.0510 ± .0005	372 ± 14	1.72	1.97	.0535	393
B-10	3.574		1.54	.0631 ± .0006	551 ± 27	0.720	2.88	.0654	575
B-14	3.544		1.04	.0662 ± .0007	601 ± 30	0.469	3.16	.0681	631
B-15	3.593		1.06	.0667 ± .0007	611 ± 32	0.477	3.15	.0680	629
B-16	3.562		0.862	.0680 ± .0007	635 ± 33	0.385	3.27	.0693	653
B-17	3.577		0.477	.0707 ± .0007	687 ± 40	0.210	3.49	.0714	697
B-18	3.562		0.163	.0724 ± .0007	721 ± 46	0.070	3.68	.0730	735
B-19	3.577		0.147	.0723 ± .0007	719 ± 46	0.061	3.70	.0730	739
B-20	3.535		0.433	.0703 ± .0007	679 ± 37	0.190	3.52	.0715	703
B-22	3.527		0.763	.0692 ± .0007	655 ± 36	0.340	3.33	.0699	665
B-8	3.585		7.961	.0252 ± .0003	134 ± 4	5.16	.526	.0202	105

CADMIUM, ^{48}Cd
Nuclear Data ⁽⁸⁾

<u>Isotope (Abundance, $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element	2450 ± 50 (non- $1/v$ factor = 1.3)
Cd-106 (.0122)	1.0 ± 0.5
Cd-.08 (.0087)	
Cd-110 (.1239)	0.2 ± 0.1
Cd-111 (.1275)	
Cd-112 (.2407)	$.03 \pm .015$
Cd-113 (.1226)	$20,000 \pm 300$
Cd-114 (.2886)	$1.2 \pm .3$
Cd-116 (.0758)	$1.5 \pm .3$

The high cadmium cross section is due to the single isotope Cd-113 which captures to the stable low cross section isotope Cd-114. The other published isotope cross sections are activation cross sections.

The samples were prepared by dissolving cadmium in lead. Results are tabulated in Table 4 and plotted in Figure 15.

The shape of the absorption cross section is non- $1/v$. The Maxwell-averaged cross section is 1.3 times the Maxwell average of a $1/v$ absorber of the same 2200 mps cross section. The calculated burnup is based on a Cd-113 cross section of 1.3 times the 2200 mps cross section.

The corrected cross section does not reproduce the unirradiated cadmium signal. This is qualitatively as expected, because the harder spectrum of the NTR should yield a higher ratio of spectral-averaged cadmium absorption to $1/v$ absorption than the 1.3 of a Maxwellian spectrum.

TABLE 4

CADMIUM

DATA					CALCULATION				
Sample	Wt. (gm)	w/o Cd*	nvt x 10 ²⁰ Wire	Signal Bare	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
CD-1	18.299	0.85	0	.0531 ± .0005	4255 ± 77	0	1.70	.0490	3273
CD-2	18.436	↓	1.77	.0015 ± .0002	67 ± 11	1.58	0.025	.0011	48
CD-4	18.021		0.913	.0094 ± .0002	454 ± 10	0.763	0.231	.0095	445

* Cadmium in lead diluent

Figure 15. CADMIUM

DYSPROSIUM, ^{66}Dy

Nuclear Data ⁽⁸⁾

<u>Isotope (Abundance, $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element --	950 $\pm$ 50
Dy-156 (.00052)	
Dy-158 (.00090)	96 $\pm$ 20
Dy-160 (.02298) 105*	
Dy-161 (.1888) 725*	
Dy-162 (.2553) 245*	
Dy-163 (.2497) 160*	
Dy-164 (.2818) 3090*	2800 $\pm$ 300
Dy-165(139 m)	5000 $\pm$ 2000

* cross sections from ref. ⁽⁹⁾⁽¹⁰⁾KAPL.

The data are tabulated in Table 5 and plotted in Fig. 16 as σ versus sample exposure since samples of two different blacknesses were measured. The cross sections are calculated from the subcadmium signal via the subcadmium calibration curve, Fig. 12.

Sample DY-3 was analyzed by mass spectrometer after an exposure of 1.78×10^{20} nvt (corresponding to a wire exposure of 1.92×10^{20} nvt). Results are tabulated in Table 6.

The complex capture chain for dysprosium is shown in Fig. 17 along with the simpler chain used in calculation. Dy-156 and Dy-157 are neglected because of the small quantity of these isotopes in natural dysprosium. The 5000 barn cross section of Dy-165 to Dy-166 is neglected because Dy-165 decays so rapidly to Ho-165 that only ~0.5 percent of the Dy-165 formed captures a neutron before decaying. The 2.32 hour half-life is neglected by assuming capture directly to the stable Ho-165. The chain is terminated after the 65 barn holmium because the 27.3 hour Ho-166 cross section is

TABLE 5
DYSPFOSIUM

DATA						CALCULATION				
Sample	Wt. (gm)	$\frac{w/o}{Dy_2O_3}$	nvt x 10^{20} Wire	Signal Bare	Signal Cd. Cov.	$\sigma_a \times 10^{-24}$	nvt x 10^{20} Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
DY-1	4.332	3.99	0	.0143 ± .0003	.0013 ± .0002	785 ± 26	0	0.329	.0132	947
DY-2	4.360	3.99	4.04	.0087 ± .0002		423 ± 32	3.79	0.183	.0076	526
↓	↓	↓	5.29	.0070 ± .0002		325 ± 23	4.98	0.158	.0066	455
			8.00	.0060 ± .0002	.0015 ± .0002	253 ± 29	7.59	0.123	.0051	354
			12.31	.0049 ± .0002	.0015 ± .0002	186 ± 27	11.77	.095	.0040	273
			17.45	.0042 ± .0002	.0014 ± .0002	158 ± 20	16.80	.082	.0035	236
↓	↓	↓								
DY-3	↓	3.99	1.918	.0117 ± .0003		612 ± 23	1.780	.244	.0100	702
DY-1A	4.317	7.06	0	.0230 ± .0003	.0021 ± .0002	743 ± 16	0	0.582	.0221	947
DY-2A	4.380		1.31	.0197 ± .0003		610 ± 19	1.14	0.476	.0185	774
↓	↓	↓	2.59	.0175 ± .0003		525 ± 20	2.28	0.400	.0158	650
			4.90	.0143 ± .0003	.0023 ± .0002	403 ± 19	4.39	0.300	.0121	488
			8.37	.0109 ± .0003	.0023 ± .0002	285 ± 14	7.62	0.216	.0089	351
↓	↓	↓	12.84	.0087 ± .0003	.0022 ± .0002	208 ± 20	11.88	0.167	.0069	272

Figure 16. DYSPROSIUM

TABLE 6

MASS SPECTROMETER DATA - DYSPROSIUM

1.92×10^{20} nvt

ISOTOPES	156	157	158	160	161	162	163	164
NATURAL	.052		.090	2.298	18.88	25.53	24.97	28.18
MASS SP. (NATURAL)	.054		.104	2.39	18.92	25.56	24.89	28.08
MASS SP. (IRRAD)	.10*	.010	.11	2.33	17.0	26.1	24.9	19.4
CALC.	.052		.090	2.39	16.68	26.66	25.38	18.75

* Residium Ho¹⁶⁵ ~ 10.02%

FIGURE 17

DYSPROSIUM CAPTURE CHAIN

SIMPLIFIED CHAIN USED IN CALCULATION

* Indicates isotope with isomeric state, only dominant transition indicated.

unknown, and the stable Er-166 is believed to have a relatively small cross section (less than ~ 50 barns, see erbium results).

The irradiation data require a complex absorption chain to explain the non-exponential behavior of sample cross section with exposure. The KAPL cross sections were used in the analysis with the exception of Dy-164. These cross sections are inconsistent with the total cross section for dysprosium, and they also predict a too rapid initial fall off in cross section with exposure. Both of these problems can be remedied by choosing Dy-164 to be about 2500 barns (consistent with 2800 ± 300 barns).

The mass spectrometer data in Table 6 is compared with the measured natural abundance to avoid problems of systematic error. The assumed initial abundance and calculated final abundance are also tabulated. The direct experimental results are ratios of the abundance of isotopes of different masses. However, the experimental ratios are not quantitative between different elements. The measured abundance of Ho-165 was about 7 percent, which is qualitatively consistent with the calculated 10.0 percent. However, the calculated ratio was used with the measured dysprosium ratios to obtain the irradiated experimental abundances.

The small abundance in masses 156 through 158 are not determined with sufficient accuracy to provide even qualitative cross section information. The exposure was small so that only cross sections of about 200 barns are experimentally significant, i.e., $(200 \times 10^{-24}) \times (1.78 \times 10^{20})$ gives a 3.56 percent change in abundance due to burn-up. The measured changes in isotopic abundance are consistent with the cross sections used in the calculation (Fig. 17). This verification of choice of cross section is $\sim \pm 30$ percent for Dy-161, ± 20 percent for Dy-164, qualitative for the other isotopes.

The correct prediction of residual cross section at large exposure, Fig. 15, implies correctness of the choice of the smaller cross sections in the chain, in particular, Dy-162 and Dy-163.

ERBIUM, ^{68}Er

Nuclear Data ⁽⁸⁾

<u>Isotope (Abundance, $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element --	137 ± 17
Er-162 (.00136)	$2.03 \pm .20$
Er-164 (.0156)	$1.65 \pm .17$
Er-166 (.334)	
Er-167 (.229)	
Er-168 (.271)	$2.0 \pm .4$
Er-170 (.149)	9 ± 2

The data are tabulated in Table 7 and plotted in Fig. 18 as normalized cross section vs. sample exposure.

Sample ER-4 was analyzed by mass spectrometer after an exposure of 1.60×10^{20} nvt (corresponding to a wire exposure of 1.72×10^{20} nvt). Results are tabulated in Table 8.

The only isotopes with significant cross section are Er-167 and possibly Er-166. The absorption chain consists of:

The two low energy resonances in Er-167 at 0.47 ev and 0.58 ev imply a high thermal cross section for this isotope. The total cross section of erbium is consistent with assigning 750 ± 75 barns to Er-167 if Er-166 is given no cross section. The mass spectrometer data, Table 8, show that only Er-167 has a cross section greater than about 100 barns and that Er-167 is about 750 ± 200 barns. However, using 750 barns leads to a slightly more rapid burn-up of erbium than is observed experimentally, Fig. 18.

The data are not inconsistent with a small cross section of about 50 barns or less assigned to Er-166.

TABLE 7
ERBIUM

DATA							CALCULATION			
Sample	Wt. (gm)	Er % 203	nvt x 10 ²⁰ Wire	Signal Bare	Signal C.G. Cov.	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
ER-1	4.857	20.6	0	0202 ± .0003	.0027 ± .0002	191 ± 5	0	0.344	.0138	173
ER-2	4.885		3.35	0177 ± .0002		163 ± 5	3.12	0.275	.0112	142
			4.82	0155 ± .0002		135 ± 4	4.48	0.248	.0101	131
			7.57	.0137 ± .0003	.0021 ± .0002	122 ± 5	7.07	0.208	.0086	112
			11.82	0110 ± .0003	.0018 ± .0002	95 ± 5	11.10	0.157	.0065	89
			17.20	.0081 ± .0003	.0015 ± .0002	66 ± 5	16.25	0.112	.0047	68
ER-3	4.854		1.33	.0192 ± .0002		179 ± 5	1.23	0.314	.0127	161
ER-4	4.885		1.72	.0183 ± .0002		168 ± 5	1.60	0.307	.0124	156

Figure 18. ERBIUM

TABLE 8

MASS SPECTROMETER DATA - ERBIUM

Sample ER-4 1.6×10^{20} nvt

ISOTOPES	162	164	166	167	168	170
NATURAL	0.136	1.56	33.41	22.94	27.07	14.88
MASS SP. (NATURAL)	0.145	1.62	33.49	23.18	26.89	14.71
MASS SP. (IRRAD)	0.139	1.672	33.383	20.491	29.294	15.021
GALC.	0.136	1.56	33.19	20.54	29.67	14.88

The resonance capture rate of erbium decreases significantly with exposure. This implies that most of the epicadmium capture is due to Er-167 and that Er-168 does not have a large resonance integral compared with Er-167.

EUROPIUM, ${}^{63}\text{Eu}$

Nuclear Data (8)

<u>Isotope (Abundance, $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element	4300 ± 100
Eu-151 (.4777)	7700 ± 80
Eu-152 (13y)	5500 ± 1500
Eu-153 (.5223)	440 ± 25
Eu-154 (16y)	1500 ± 400
Eu-155 (1.7y)	$14,000 \pm 4000$

The data are tabulated in Table 9 and plotted in Figure 19 as cross section versus sample exposure. Two samples were analyzed by mass spectrometer after an exposure of 1.27×10^{21} nvt (corresponding to a wire exposure of 1.30×10^{21} nvt) and an exposure of 2.17×10^{20} (wire exposure of 2.05×10^{20} nvt). Results are tabulated in Table 10.

The absorption chain of europium is shown in Figure 20, along with the simpler chain used in calculation. The 13-year and 16-year isotopes are treated as stable. The Gd-156 cross section is known to be small and the Eu-156 is unknown, so the chain is terminated at mass 156. The 9.2 hour transition is assumed instantaneous, and the 26 per cent of the Eu-152 mass which decays to Sm-152 is instead routed to Gd-152. This latter approximation re-routes only 2 per cent of the atoms in the sample and has negligible effect on the final isotopic composition.

The simplified chain in Figure 20 should account for any significant cross section change during the irradiation of these samples. The pile oscillator results are as expected except for the residual cross section. Only one sample was carried to very high burnup, hence experimental certainty is not high, but this sample does indicate that the predicted residual cross section is above the experimental points by a probably significant amount. More significantly, the mass spectrometer data for both samples analysed is inconsistent with calculation.

The sequence of the burnup chain allows a quantitative interpretation of the mass data. Samples of two different exposures were analysed for the low exposure sample EU-3, the experimental ratio of Eu-153 to Eu-151 is a measure of the accuracy

TABLE 9
EUROPIUM

DATA							CALCULATION			
Sample	Wt. (gm)	$\frac{w}{o}$ Eu ₂ O ₃	nvt x 10 ²⁰ Wire	Signal Bare	Signal Cd. Cov.	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
EU-1	4.223	0.80	0	.0122 ± .0002	.0006 ± .0002	4083 ± 102	0	0.281	.0114	3950
EU-2	4.240		4.57	.0031 ± .0002		850 ± 111	4.39	0.0804	.0034	1115
			6.45	.0025 ± .0002		683 ± 83	6.23	0.0632	.0027	878
			9.12	.0021 ± .0002	.0005 ± .0002	530 ± 69	8.86	0.0563	.0024	780
			13.04	.0021 ± .0002	.0005 ± .0002	530 ± 69	12.73	0.0533	.0023	739
EU-3	4.228		2.17	.0068 ± .0002		2174 ± 156	2.05	0.147	.0061	2048
EU-1A	4.161	2.04	0	.0259 ± .0003	.0014 ± .0002	3785 ± 55	0	0.715	.0263	3950
EU-2A	4.146		0.765	.0228 ± .0003		3237 ± 50	0.650	0.585	.0222	3232
			2.26	.0166 ± .0003		2193 ± 45	1.96	0.384	.0152	2122
			4.29	.0107 ± .0002	.0011 ± .0002	1331 ± 40	3.85	0.229	.0094	1265
EU-1C	4.877	25.6	0	.0972 ± .0010	.0111 ± .0002	3286 ± 150	0	10.3	.1008	3950
EU-2C	4.846		2.13	.0944 ± .0009	.0110 ± .0002	2740 ± 135	0.414	9.04	.0975	3450
			6.22	.0880 ± .0009	.0073 ± .0002	2309 ± 115	1.626	5.339	.0844	2038

Figure 19. EUROPIUM

TABLE 10

MASS SPECTROMETER DATA - EUROPIUMSample EU-2 12.7×10^{20} nvt

ISOTOPIC RATIOS	153/151	153/152	153/154	153/155	153/156
NATURAL	1.09				
MASS SPEC. NATURAL	1.08				
MASS SPEC. EXP.	<2,000	9* $\begin{matrix} \nearrow 13 \\ \searrow 5 \end{matrix}$	6.0 ± 0.4	12* $\begin{matrix} \nearrow 13 \\ \searrow 10 \end{matrix}$	115 $\begin{matrix} \nearrow 160 \\ \searrow 70 \end{matrix}$
CALC.	16,278	398	3.5	33	3.1

Sample EU-3 2.05×10^{20}

MASS SPEC. EXP.	4.9	2 to 5**	25	225	1,000
CALC.	5.7	3.8	14.5	205	189

* Ratios were time dependent.

** Erratic measurement.

FIGURE 20

EUROPIUM CAPTURE CHAIN

SIMPLIFIED CHAIN USED IN CALCULATION

* Indicates isotope with isomeric state, only dominant transition indicated.

of the exposure determination and the Eu-151 cross section. If the 0.95 non-1/v factor for elemental europium is applied to the Eu-151 cross section, then the exposure so determined is 11% lower than that determined by the cobalt wire measurement.

The total amount of material in all mass positions 154 and higher originate in the 153 position. The cross section for the 153 can be derived from the exposure and the amount of higher mass material.

The cross section so derived is 250 ± 60 barns which is significantly smaller than the published 440 ± 25 barns⁽⁸⁾ at 2200 mps.

A similar argument can be applied to the Eu-154 cross section. The 156 mass is not quantitative so the result can only be approximate. The (qualitative) result is that Eu-154 has a cross section of the order of 1000 barns.

The small amount of material in mass 156 implies that the Eu-155 cross section is several times smaller than 14,000 barns.

The above conclusions are consistent with the mass data for the long exposure sample, EU-2. For this sample the Eu-151 was virtually all burned to mass 152. The mass 152 branch with high cross section has also proceeded to 153 by another neutron capture, leaving about 10% of the total europium material in mass 152. The cross section for Eu-153 can be derived from the exposure and the total amount of material in mass 154 and higher. The result is again 250 ± 60 barns.

The samples were mass analysed several months after irradiation. During this period a fraction of the 1.7 year Eu-155 β decayed to Gd-155. The 15 day Eu-156 all decayed to Gd-156. It was found that the experimental mass ratios of the gadolinium to europium isotopes varied a factor of two with time during the measurement, while the europium to europium isotope ratios remained constant. The 153/155 ratio varied about 30% with time. From this it is concluded that the europium to gadolinium ratios are quantitative to within a factor of two. On this basis, the Eu-154 cross section is 800 ± 250 barns and the Eu-155 cross section is less than 600 barns. The presence of material in mass 156 can be fully accounted for by the 1.7 year β decay to Gd-155 and subsequent reaction capture to Gd-156.

Masses 157, 158 and 159 were unobservable.

In summary, this data is consistent with the published cross section for Eu-151 and with the ratio of the 153 cross section to the 154 cross section. It is inconsistent with the published values for Eu-153 and 154 by almost a factor of two and inconsistent with the Eu-155 cross section by a factor of 20.

The discrepancy with Eu-153 cross section may be explainable in terms of a non- $1/v$ factor.

The cross sections derived from the mass data alter only the residual cross section in the calculated burnup curves of Figure 19, (reduces residual by about factor of two).

GADOLINIUM, ${}_{64}\text{Gd}$

Nuclear Data ⁽⁸⁾

<u>Isotope (Abundance, $T_{1/2}^1$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element --	46,000 $\pm$ 1000 (non-1/v .85)
Gd-152 (.0020)	
Gd-153 (.0215)	
Gd-155 (.1473)	56,200 $\pm$ 1000
Gd-156 (.2047)	
Gd-157 (.1568)	242,000 $\pm$ 4000
Gd-158 (.2487)	3.9 $\pm$ 0.4
Gd-160 (.2190)	

Gadolinium has two absorbing isotopes of extremely high cross section. The elemental cross section is non-1/v by 0.85. This is included in the Gd-157 isotopic cross section.

The data are not amenable to graphical representation. The tabular comparison of theory and experiment, Table 11, shows poor agreement which is attributed to the several decade variations in cross section. The GD-2 samples were measured to be 1.5 and 2.5 standard deviations below zero cross section for exposures which theory indicates as being substantially zero. A signal of one standard deviation (in the positive direction, of course) would correspond to about a 60 barn cross section for the element. GD-2B burned to -1 standard deviation. One standard deviation corresponds to about 20 barns for this sample. It is concluded that the residual gadolinium cross section is probably less than 20 barns.

For several years it has been recognized that if the unknown cross section of Gd-156 was appreciable (~ 1000 barns), then gadolinium would be a long burning control material of relatively high cross section. The low residual cross section does not support this speculation. The upper limit on the residual implies that Gd-156 has a cross section of less than 30 barns.

TABLE 11

GADOLINIUM

Sample	DATA					CALCULATION			
	Wt. (gm)	w/o Gd ₂ O ₃	nvt x 10 ²⁰ Wire	Signal Bare	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
GD-1	4.231	0.81	0	.0503 ± .0005	27,955 ± 548	0	2.92	.0657	40,951
GD-2	4.265	↓	1.18	-.0003 ± .0002	0	1.11	0.001	.0000	14
↓	↓	↓	2.82	-.0005 ± .0002	0	2.75	0.000	.0000	0
GD-3	4.228	↓	0.578	.0041 ± .0002	1,403 ± 282	0.514	0.028	.0011	393
GD-1A	4.096	0.204	0	.0229 ± .0003	36,497 ± 521	0	0.708	.0261	40,951
GD-2A	4.115	↓	0.252	.0029 ± .0002	4,049 ± 289	0.239	0.040	.0017	2,314
↓	↓	↓	0.723	.0004 ± .0002	578 ± 174	0.709	0.002	.0001	116
GD-1B	4.164	2.41	0	.0702 ± .0007	19,096 ± 573	0	8.56	.0982	40,951
GD-2B	4.163	↓	0.304	.0429 ± .0004	7,161 ± 119	0.125	1.39	.0432	6,636
↓	↓	↓	0.910	.0076 ± .0002	907 ± 33	0.687	0.029	.0012	138
↓	↓	↓	3.353	-.0002 ± .0002	0	3.310	0.0000	.00000	0

GOLD, ^{79}Au

Nuclear Data⁽⁸⁾

<u>Isotope (Abundance, $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Au-197 (1.0)	98.8 ± 0.3
Au-198 (2.7d)	$26,000 \pm 1200$
Au-199 (3.15d)	30 ± 15

Data are tabulated in Table 12. Mass analysis was not performed on this sample nor were the measurements pursued beyond the one irradiation. The capture chain for gold is shown in Fig. 21. If gold is exposed to a flux that is sufficiently high to produce a significant amount of Au-198, then the effective cross section of the sample will increase significantly during irradiation. The self-shielding of the gold sample used, combined with the long delay (2 days) from end of exposure to time of measurement in NTR, made the increased cross section unobservable in this experiment. Rather than pursue this point for the relatively well-known gold isotopes, this portion of the experiment was discontinued.

TABLE 12

DATA			GOLD			CALCULATION				
Sample	Wt. (gm)	w/o Au	nvt x 10 ²⁰ Wire	Signal Bare	Signal Cd. Cov.	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
Au-1	30.35	100	0	.0962 ± .0009	.0147 ± .0002	123 ± 6	0	5.83	.0750	96.8
Au-2	30.40	100	2.158	.0947 ± .0009		113 ± 5	0.579	5.94	.0778	100.7

FIGURE 21
GOLD CAPTURE CHAIN

* Indicates isotope with isomeric state, only dominant transition indicated.

HAFNIUM, $_{72}^{Hf}$

Nuclear Data (BNL-325)

<u>Isotope (Abundance, $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element	105 ± 5
Hf-174 (.0018)	1500 ± 1000
Hf-176 (.0515)	15 ± 15
Hf-177 (.1839)	380 ± 30
Hf-178 (.2708)	75 ± 10
Hf-179 (.1378)	65 ± 15
Hf-180 (.3544)	14 ± 15

The data are tabulated in Table 13 and plotted in Figure 22 as cross section vs. sample exposure. This plot is different from the other burnup curves in this report, in that for this case the data are not normalized at zero exposure to the barn book elemental cross section.

The absorption chain for hafnium is shown in Figure 23 along with the simpler chain used in calculation. The burnup behavior of hafnium is dominated by the 380 barn Hf-177. The 1500 barn Hf-174 accounts for only 3% of the total cross section, because of its low concentration.

The hafnium samples were prepared by vacuum arc puddling hafnium and zirconium to form small bars of alloy which were then machined to size, the unirradiated standard and irradiated sample both coming from the same bar. The inconsistency between irradiated and unirradiated data implies that the hafnium and zirconium were not adequately mixed during the puddling process. This means that the concentrations are probably in error.

The slope of the data is not sensitive to assumed concentration and is consistent with the slope of the calculated curve.

TABLE 13

HAFNIUM

DATA							CALCULATION			
Sample	Wt. (gm)	w/o Hf	nvt x 10 ²⁰ Wire	Signal Bare	Signal Cd. Cov.	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
HF-1	11.150	10	0	.01699 ± .0003	.0050 ± .0002	126 ± 6	0	.253	.01033	105
HF-2 ↓	11.311 ↓	↓	2.131	.0181 ± .0003		137 ± 7	1.99	.241	.0099	100
			4.064	.0173 ± .0003		131 ± 6	3.80	.231	.0095	96
			4.528	.0173 ± .0003		131 ± 6	4.22	.229	.094	95
			5.676	.0169 ± .0003	.0052 ± .0002	123 ± 6	5.28	.224	.092	93
HF-1A	11.737	20	0	.0364 ± .0004	.0089 ± .0003	149 ± 4	0	.557	.0213	105
HF-2A ↓	11.631 ↓	↓	2.111	.0310 ± .0003		119 ± 4	1.820	.533	.0205	100
			4.380	.0300 ± .0003	.0079 ± .0003	114 ± 4	3.80	.512	.0198	97
			8.250	.0284 ± .0003	.0075 ± .0003	107 ± 3	7.19	.478	.0186	90
			12.197	.0279 ± .0003	.0073 ± .0003	105 ± 3	10.60	.449	.0176	85

Figure 22. HAFNIUM

FIGURE 23

HAFNIUM CAPTURE CHAIN

SIMPLIFIED CHAIN USED IN CALCULATION

* Indicates isotope with isomeric state, only dominant transition indicated.

HOLMIUM, 67^{Ho}

Nuclear Data (BNL-325)

<u>Isotope (Abundance, $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Ho-165 (1.0)	65 $\pm$ 3

The data are tabulated in Table 14 and plotted in Figure 24 as normalized cross section versus sample exposure. Sample Ho-3 was analysed by mass spectrometer after an exposure of 1.90×10^{20} nvt (corresponding to a wire exposure of 1.96×10^{20} nvt). Results are in Table 15.

The single holmium isotope captures a neutron to Ho-166 which β decays with 27 hour half life to Er-166 of unknown cross section (see erbium data).

The burnup data, Figure 24, are consistent with expectation. However, the burnup rate is so small that the data do not provide a quantitative indication of the cross section. The mass data in Table 15 is likewise not quantitative since the relevant experimental ratio is between erbium and hafnium. Mass data is in qualitative agreement with expectation.

For comparison, the amount of Er-167 is listed which would be produced if Er-166 had a 9 barn cross section, and is well below the sensitivity of the measurement.

TABLE 14

HOLMIUM

DATA

CALCULATION

Sample	Wt. (gm)	w/o H ₂ O O ₃	nvt x 1020 Wire	Signal Bare	Signal Cd. Cov.	$\sigma_a \times 10^{-24}$	nvtx1020 Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
HO-1	4.859	20.6	0	.0085 ± .0002	.0027 ± .0002	74 ± 4	0	.129	.0054	67
HO-2	4.898		3.701	.0089 ± .0002		78 ± 4	3.576	.128	.0054	67
			5.691	.0083 ± .0002	.0027 ± .0002	70 ± 4	5.501	.127	.0053	66
			7.321	.0082 ± .0002		69 ± 5	7.075	.127	.0053	66
			8.236	.0081 ± .0002	.0027 ± .0002	67 ± 6	7.961	.127	.0053	66
			10.119	.0080 ± .0002	.0026 ± .0002	67 ± 6	9.762	.127	.0053	66
HO-3	4.861		1.964	.0083 ± .0002		70 ± 4	1.898	.126	.0054	67

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Figure 24. HOLMIUM

TABLE 15

MASS SPECTROMETER DATA - HOLMIUMSample HO-3 1.90×10^{20} nvt

	<u>Ho</u>	<u>Er</u>	<u>Er</u>
ISOTOPES	165	166	167
NATURAL	100%	0	0
MASS SPEC.	99.191	.809	.03
CALCULATED	98.77	1.218	.00444*

* Assuming 9 barn cross section for Er-166

INDIUM, $_{49}\text{In}$

Nuclear Data (BNL-325)

<u>Isotope (Abundance or $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, barns</u>
Element	191 ± 3
In-113 (.0423)	58 ± 12
In-115 (.9577)	202 ± 16

The data are tabulated in Table 16 and plotted in Figure 25 as normalized cross section versus exposure.

In-113 accounts for only 1% of the elemental cross section. In-115 captures a neutron to In-116 which quickly (54 minutes) β decays to the stable low cross section Sn-116. The data are in excellent agreement with prediction.

TABLE 16
INDIUM

DATA			CALCULATION							
Sample	Wt. (gm)	w/o In	nvt x 1020 Wire	Sample Bare	Sample Cd. Cov.	$\sigma_a \times 10^{-24}$	nvt x 1020 Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
IN-1A	17.179	5	0	.0282 ± .0003	.0042 ± .0002	231 ± 6	0	.579	.0220	201
IN-2A	17.203		0.708	.0275 ± .0003		224 ± 5	0.60	.572	.0218	198
			1.879	.0266 ± .0003		215 ± 4	1.59	.560	.0214	194
			3.716	.0265 ± .0003	.0041 ± .0002	213 ± 4	3.22	.542	.0208	188
			6.526	.0256 ± .0003	.0040 ± .0002	204 ± 4	5.67	.517	.0199	179
IN-1B	15.970	20	0	0.636 ± .0006	.0077 ± .0002	231 ± 6	0	2.128	.0557	201
IN-2B	16.026		0.291	.0632 ± .0006		227 ± 6	0.171	2.121	.0556	200
			2.191	.0632 ± .0006		227 ± 6	1.300	2.073	.0550	195
			3.141	.0623 ± .0006	.0076 ± .0002	220 ± 6	1.865	2.049	.0546	193
IN-1C	12.351	80	0	.0997 ± .0010		350 ± 26	0	6.246	.0886	201
IN-2C	12.056		0.980	.0991 ± .0010		334 ± 26	.284	6.210	.0884	199
			1.361	.0981 ± .0010		320 ± 25	.394	6.196	.0883	199

Figure 25. INDIUM

LUTETIUM, ^{Lu}₇₁

Nuclear Data (BNL-325)

<u>Isotope (Abundance, T_{1/2})</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element	112 ± 5
Lu-175 (.9740)	35 ± 15
Lu-176 (.0260)	4000 ± 800

The data are tabulated in Table 17, and plotted in Figure 26 as normalized cross section versus exposure. Sample LU-2 was analysed by mass spectrometer after an exposure of 2.64×10^{20} nvt (corresponding to a wire exposure of 2.78×10^{20} nvt). The complex capture chain for lutetium is shown in Figure 27 along with the simpler chain used in calculation.

The early burnup behavior is dominated by the 4000 barn Lu-176 and the residual is determined by the 35 barn Lu-175. Pile oscillator and mass data are in substantial agreement with expectation. The hafnium isotopes were not observed, so the 175/176 mass ratio is probably quantitative lutetium with negligible hafnium interference.

TABLE 17
LUTETIUM

DATA							CALCULATION			
Sample	Wt. (gm)	w/o Lu ₂ O ₃	nvt x 10 ²⁰ Wire	Signal Bare	Signal Cd. Cov.	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ Sample	Σ_a (220 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
LU-1	4.870	20.6	0	.0145 ± .0003	.0030 ± .0002	154 ± 8	0	0.261	.0106	138
LU-2	4.909		2.78	.0099 ± .0002		80 ± 7	2.64	0.141	.0059	75
LU-3 ↓ ↓ ↓	4.358 ↓ ↓ ↓		2.17	.0103 ± .0002		85 ± 6	2.06	0.158	.0066	84
			4.43	.0086 ± .0002	.0040 ± .0002	59 ± 6	4.23	0.112	.0047	59
			6.536	.0074 ± .0002	.0037 ± .0002	49 ± 4	6.29	0.094	.0040	50
			9.318	.0068 ± .0002	.0038 ± .0002	39 ± 5	9.01	0.084	.0036	44

Figure 26. LUTETIUM

FIGURE 27

LUTETIUM CAPTURE CHAIN

SIMPLIFIED CHAIN USED IN CALCULATION

TABLE 18

MASS SPECTROMETER DATA - LUTETIUM

Sample LU-2 2.64×10^{20} nvt

MASS RATIO	175/176
NATURAL	375
IRRADIATED	113 ± 1
CALC.	108

SAMARIUM, ^{62}Sm

Nuclear Data (BNL-325)

<u>Isotope (Abundance, $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element	$5,600 \pm 200$
Sm-144 (.0317)	< 2
Sm-147 (.1507)	87 ± 60
Sm-148 (.1127)	
Sm-149 (.1384)	$40,800 \pm 900$
Sm-150 (.0747)	
Sm-151 (73y)	$12,400 \pm 400$
Sm-152 (2663)	216 ± 6
Sm-154 (.2253)	5.5 ± 1

The data are tabulated in Table 19 and plotted in Figure 28 as normalized cross section versus exposure. Sample SM-2 was analysed by mass spectrometer after a sample exposure of 4.66×10^{20} nvt (corresponding to a wire exposure of 4.81×10^{20} nvt).

The capture chain for samarium is shown in Figure 29. Initial burnup of samarium is dominated by the rapid conversion of Sm-149 to Sm-150. The residual cross section is based largely on Sm-152 which feeds the europium chain, and Sm-150 which goes to 12,400 barn Sm-151.

The cross section of Sm-150 can be obtained from the mass ratio 150/151 and the known cross section of Sm-151 since the isotopes were in "secular" equilibrium. The result is 100 ± 30 barns.

The mass spectrometer results are consistent with the other cross section assignments. However, because of the low exposure this is quantitative for only Sm-152 and Sm-150. The mass data also implies that the cross section of Sm-148 is less than 50 barns.

TABLE 19

SAMARIUM

DATA					CALCULATION				
Sample	Wt. (gm)	w/o SM ₂ O ₃	nvt x 10 ²⁰ Wire	Signal Bare	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ Samples	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
SM-1	4.292	3.04	0	.0589 ± .0006	9666 ± 214	0	2.35	.0590	8520
SM-2	4.312	↓	4.81	.0023 ± .0002	199 ± 15	4.66	.0336	.0014	119
SM-3	4.292	↓	1.16	.0022 ± .0002	185 ± 21	1.03	0.0345	.0015	123
SM-1A	4.086	1.01	0	.0318 ± .0003	10600 ± 189	0	0.766	.0279	8520
SM-2A	4.078	↓	0.961	.0014 ± .0002	389 ± 78	0.923	0.0125	.0005	139
SM-1B	4.496	13.1	0	.0877 ± .0009	7299 ± 434	0	10.80	.1026	8520
SM-2B	4.560	↓	1.19	.0593 ± .0006	2162 ± 47	0.531	0.548	.0210	432
85 ↓	↓	↓	3.06	.0078 ± .0010	155 ± 19	2.314	0.148	.0062	117

Figure 28. SAMARIUM

FIGURE 29

SAMARIUM CAPTURE CHAIN

SIMPLIFIED CHAIN USED IN CALCULATION

* Indicates isotope with isomeric state, only dominant transition indicated.

** See Figure 20.

TABLE 20

MASS SPECTROMETER DATA - SAMARIUMSample SM-2 4.66×10^{20} nvt

	ISOTOPES	Sm-144	Sm-147	Sm-148	Sm-149	Sm-150	Sm-151	Sm-152	$\frac{\text{Sm}}{\text{Eu}}$ 153	$\frac{\text{Sm}}{\text{Eu}}$ 154	Eu-155
	NATURAL	2.87	14.94	11.24	13.85	7.36		26.90		22.84	
∞	MASS SPEC. (NATURAL)	3.17	14.89	10.96	13.70	7.51		26.91		22.84	
	MASS SPEC. (IRRAD)	2.99	14.35	11.38	0	20.18	0.17	24.82	2.75	23.20	0.13
	CALC.	2.87	14.47	11.81	.002	20.42	0.165	24.84	2.33	22.79	.10

SILVER, $_{47}\text{Ag}$

Nuclear Data (BNL-325)

<u>Isotope (Abundance, $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element	63 ± 1
Ag-107 (.5135)	31 ± 2
Ag-109 (.4865)	87 ± 7

The data are tabulated in Table 21 and plotted in Figure 30 as normalized cross section versus exposure.

Both silver isotopes capture and then β decay to stable cadmium isotopes. Because of the relatively low cross section of both silver isotopes, the burnup rate is too slow to observe in this experiment. The results are consistent with expectation.

TABLE 2:

SILVER

DATA							CALCULATION			
Sample	Wt. (gm)	w/o Ag	nvt x 10 ²⁰ Wire	Signal Bare	Signal Cd. Cov.	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
AG-1		100	0	.0850 ± .0009	.0148 ± .0002	68 ± 3	0	4.67	.0805	80
AG-2			0.950	.0848 ± .0003		68 ± 3	.35	4.65	.0803	79
AG-3			0.875	.0849 ± .0008		68 ± 3	.32	4.65	.0803	79
			2.324	.0822 ± .0008		62 ± 3	.85	4.63	.0802	79
			3.339	.083E ± .0008	.0145 ± .0002	67 ± 3	1.22	4.61	.0800	78

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Figure 30. SILVER

THULIUM, ${}^{69}\text{Tm}$

Nuclear Data (BNL-325)

<u>Isotope (Abundance, $T_{1/2}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Tm-169 (1.0)	127 ± 4
Tm-170 (129d)	150 ± 20

The data are tabulated in Table 22 and plotted as normalized cross section versus exposure in Figure 31.

The thulium capture chain is shown in Figure 32. The expected result of no observable change in cross section with the attained exposure is verified by the data.

TABLE 22
THULIUM

Sample	Wt. (gm)	w/o Tm ₂ O ₃	nvt x 10 ²⁰ Wire	Signal Bare	Signal Cd. Cov.	$\sigma_B \times 10^{-24}$	nvt x 10 ²⁰ Sample	Σ_a (2200 m/sec)	Sample Bare	$\sigma_a \times 10^{-24}$
TM-1A	5.255	35	0	.0201 ± .0002	.0043 ± .0002	115 ± 3	0	.466	.0182	130
TM-2A	5.228		0.940	.0201 ± .0002		115 ± 3	0.84	.467	.0182	130
			2.670	.0202 ± .0002	.0044 ± .0002	115 ± 3	2.36	.468	.0182	130
			6.010	.0200 ± .0002	.0045 ± .0002	112 ± 3	5.26	.470	.0183	131

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Figure 31. THULIUM

FIGURE 32

THULIUM CAPTURE CHAIN

* Indicates isotope with isomeric state, only dominant transition indicated.

YTTERBIUM, ^{70}Yb

Nuclear Data (BNL-325)

<u>Isotope (Abundance, $T_{\frac{1}{2}}$)</u>	<u>2200 mps Absorption Cross Section, Barns</u>
Element	37 ± 4
Yb-168 (.0014)	$11,000 \pm 3000$
Yb-170 (.0303)	
Yb-171 (.1431)	
Yb-172 (.2182)	
Yb-173 (.1613)	
Yb-174 (.3184)	60 ± 40
Yb-176 (.1273)	5.5 ± 1.0

The data are tabulated in Table 23 and plotted as normalized cross section versus exposure in Figure 33. Mass spectrometer analysis was performed on sample YB-2 after an exposure of 0.65×10^{20} nvt (corresponding to a wire exposure of 0.66 nvt) and are listed in Table 24.

The ytterbium capture chain is shown in Figure 34. The observable burnup is due to the 11,000 barn Yb-168. The implication of both the mass analysis and the pile oscillator data is that the Yb-168 cross section is significantly less than 11,000 barns. However, the errors of both these measurements are large and hence certainty is not assured. Because of the low exposure of the mass analysed sample, no information was obtained for the other ytterbium isotopes but possibly still within the above quoted uncertainty.

TABLE 25

YTTERBIUM

DATA							CALCULATION			
Sample	Wt. (gr)	W/o Yb ₂ O ₃	nvt x 10 ²⁰ wire	Signal Bare	Signal Cd. Cov.	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
Yb-1	4.963	24.3	0	.0059 ± .0002	.0014 ± .0002	47 ± 4	0	.0810	.0034	35
Yb-2	4.965		.660	.0059 ± .0002		47 ± 4	.648	.0624	.0027	26
Yb-3	4.978		1.736	.0055 ± .0002		43 ± 4	1.687	.0502	.0021	21
			3.146	.0050 ± .0002	.0013 ± .0002	38 ± 4	3.100	.0459	.0020	20
			5.066	.0048 ± .0002	.0014 ± .0002	35 ± 4	4.996	.0450	.0019	19
			6.193	.0046 ± .0002	.0013 ± .0002	34 ± 4	6.090	.0450	.0019	19

Figure 33. YTTERBIUM

TABLE 24

MASS SPECTROMETER DATA - YTTERBIUMSample YB-2 0.65×10^{20} nvt

ISOTOPES	168	170	171	172	173	174	176
NATURAL	.14	3.03	14.34	21.88	16.18	31.77	12.65
MASS SPEC.	.109	3.189	14.886	21.792	16.493	31.045	12.491
CALC.	.07	3.03	14.34	21.88	16.18	31.72	12.70

NOTE:

Yb ¹⁶⁹	0.06	} CALC.
Tm ¹⁶⁹	0.006	
Tm ¹⁷⁰	0.00002	
Lu ¹⁷⁵	0.005	
Lu ¹⁷⁶	0.00004	
Lu ¹⁷⁷	0.000003	

FIGURE 34
YTTERBIUM CAPTURE CHAIN

SIMPLIFIED CHAIN USED IN CALCULATION

* Indicates isotope with isomeric state, only dominant transition indicated.

SILVER - INDIUM - CADMIUM

The pile oscillator data for the silver - indium - cadmium sample is in Table 25 and plotted in Figure 35. This dramatically demonstrates the rapid burnup of the high cross section Cd-113.

However, the total reactivity effect of the sample on the NTR varied only 110% while the effective thermal cross section of the sample varied a factor of two to three.

TABLE 25

DATA			SILVER - INDIUM - CADMIUM*			CALCULATION			
Sample	Wt. (gm)	nvt x 10 ²⁰ Wire	Signal Bare	Signal Cd. Cov.	$\sigma_a \times 10^{-24}$	nvt x 10 ²⁰ Sample	Σ_a (2200 m/sec)	Signal Bare	$\sigma_a \times 10^{-24}$
AIC-1	16.545	0	.1141 ± .0011	.0175 ± .0003	299 ± 18	0	12.28	.1048	217
AIC-2	16.546	0.838	.1114 ± .0011	----	248 ± 15	0.144	10.50	.1011	186
↓	16.546	2.29	.1069 ± .0011	.0178 ± .0003	178 ± 10	0.441	8.08	.0944	143
		4.937		.0177 ± .0003		1.16	5.85	.0868	103
		8.754	.0944 ± .0010	.0172 ± .0003	101 ± 5	2.40	5.17	.0833	91

* MANUFACTURER'S SPECIFICATION

Ag 80.11 w/o

In 14.82 w/o

Cd 4.92 w/o

Figure 35. SILVER - INDIUM - CADMIUM

V. CONTROL ROD BURNUP

The problem of control rod burnup is highly complex because of the very high self-shielding of the absorber. Although a rigorous attack of this problem may not be justified on the basis of control rod economics because other variables in a control system are not predictable, a reliable, but not necessarily highly precise, estimate of control system lifetime is a necessity for design of economic nuclear power plants.

The following is a description of a semi-empirical method for providing the nuclear data for this estimate.

The method (1) takes into account the fact that some isotopes form absorber chains in which each successive absorption product can absorb another neutron. In order to make the problem tractable, it is assumed that the neutrons are mono-energetic (constant isotopic cross section approximation) and that the competition between absorbing isotopes is always in proportion to the cross sections (dilute absorber approximation). For chemical elements which have only a single absorbing isotope, and no chain burners, the validity of the assumption is not dependent upon the two approximations. For the highly self-shielded mixtures of absorbing isotopes found in control rods, the neglected effect of spatial shielding of the high cross section isotopes in the interior by the lower cross section isotopes remaining on the surface (at early to intermediate stages of burnup) leads to prediction of a too rapid early burnup.

The quantity F is defined:

$$F = \frac{\sigma(n)}{\sigma(o)} \quad (19)$$

where n = the number of neutrons captured by
the absorbing element (per atom)

$\sigma(o) = \sum_i N_i(o) \sigma_i$ = element cross section, i.e., sum of
isotopic cross sections weighted by
abundances, for no irradiation

$\sigma(n) = \sum_i N_i(n) \sigma_i$ = element cross section after capturing n neutrons per atom.

N_i = abundances ($\sum_i N_i = 1$)

$\sigma(n)$ is defined by the parametric (in x) equation (neglecting spontaneous decay).

$$n = \int_0^x \sum_i \sigma_i N_i(x) dx \quad (20)$$

$$\frac{dN_i(x)}{dx} = -\sigma_i N_i(x) + \sum_{j \rightarrow i} \sigma_j N_j(x) \quad (21)$$

To illustrate the behavior of equations 19 and 20, consider the simple case of an element with a single absorbing isotope of abundance N_0 , and no absorbing chain. Then:

$$N = N_0 e^{-\sigma x} \quad (22)$$

$$\text{and: } n = \int_0^x \sigma N_0 e^{-\sigma x} = N_0 (1 - e^{-\sigma x}) \quad (23)$$

$$\text{solving: } F = 1 - \frac{n}{N_0} \quad (24)$$

Applying equation 24 to the case of natural boron, for which N_0 is 0.188 leads to the boron line in Figure 36. This merely demonstrates the obvious conclusion that boron, after absorbing 0.188 neutrons per atom, has zero cross section because only 18.8% of the atoms of natural boron are B-10, the absorbing isotope.

For the elements presented, which have more than one absorbing isotope, the competition between absorbing isotopes must be considered. The parametric equations were solved, with the VARI-II code, using cross sections which also predicted the results of the reported pile oscillator measurements. Therefore the F curves of Figure 36 are experimentally verified to the degree of burnup investigated.

The F curves neglect all resonance capture effects. They are computed on the basis of thermal burnup and the experimental verification was with a thermal flux.

Figure 36. FRACTIONAL CROSS SECTION VERSUS FRACTIONAL BURNUP

Figure 36.

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