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Extraction of Gadolinium
from High Flux Isotope Reactor
Control Plates

M. W. Kohring

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EXTRACTION OF GADOLINIUM
FROM HIGH FLUX ISOTOPE REACTOR
CONTROL PLATES

Operations Division

M. W. Kohring

Date Published - April 1987

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CONTENTS

	<u>Page</u>
LIST OF FIGURES.....	v
LIST OF TABLES.....	vii
ABSTRACT.....	ix
INTRODUCTION.....	1
GADOLINIUM-153 PRODUCTION AT ORNL.....	5
Irradiation of Europium Targets.....	5
Alternate Production Scheme.....	11
HFIR CONTROL PLATES.....	13
Description.....	13
Control Plate Motion.....	16
COMPUTATIONAL ANALYSIS	21
Description of ORIGEN2.....	21
Assumptions and Validity of Initial Estimates.....	22
Predicted Optimum Location of Gadolinium-152.....	24
EXPERIMENTAL ANALYSIS.....	31
Overview.....	31
Axial Gamma Scans.....	32
Dissection of Control Plate and Separation of Gadolinium Fraction.....	35
Mass Spectrographic Analysis.....	36
DETERMINATION OF OPTIMUM LOCATION.....	39
Quantitative Analysis.....	39
Completion of Experimental Analysis Phase.....	42
Comparison of Experimental and Computational Results	45
CONCLUSION.....	51
ACKNOWLEDGEMENTS.....	53
REFERENCES.....	55
DISTRIBUTION.....	59

LIST OF FIGURES

	<u>Page</u>
1. Cross-sectional view of HFIR at horizontal midplane	2
2. Curies of ^{153}Gd produced at ORNL, 1980-1986.....	4
3. Nuclear reaction and decay paths in the production of gadolinium isotopes.....	6
4. Schematic view of HFIR core region.....	14
5. Cutaway view of HFIR control components.....	15
6. HFIR control plate positions during a cycle.....	17
7. Calculated thermal flux per unit power in critical core at beginning of HFIR cycle.....	18
8. Typical input program to ORIGEN2 for control plate calculations.....	25
9. Normalized concentrations vs effective neutron flux intensity.....	27
10. Radioactivity vs effective neutron flux intensity.....	27
11. Gadolinium isotopic compositions vs effective neutron flux intensity.....	28
12. Gadolinium concentration vs effective neutron flux intensity.....	29
13. Measured radioactivity vs axial location.....	33
14. Isotopic composition of major gadolinium isotopes vs axial location.....	38
15. Total gadolinium per linear centimeter of control plate.....	41
16. Control plate axial location vs effective neutron flux intensity.....	48

LIST OF TABLES

	<u>Page</u>
1. Estimated neutron flux intensities at selected control plate locations.....	23
2. Measured gamma intensity in photons/s at selected control plate locations.....	33
3. Mass spectrographic analysis results for europium isotopes.....	37
4. Mass spectrographic analysis results for gadolinium isotopes.....	37
5. Quantitative analysis at selected control plate locations.....	41
6. Effective neutron flux intensities at selected control plate locations.....	46
7. Comparison of calculated gadolinium concentration in selected control plates.....	49

ABSTRACT

Gadolinium-153 is an important radioisotope used in the diagnosis of various bone disorders. Recent medical and technological developments in the detection and cure of osteoporosis, a bone disease affecting an estimated 50 million people, have greatly increased the demand for this isotope.

The Oak Ridge National Laboratory (ORNL) has produced ^{153}Gd since 1980 primarily through the irradiation of a natural europium-oxide powder followed by the chemical separation of the gadolinium fraction from the europium material. Due to the higher demand for ^{153}Gd , an alternative production method to supplement this process has been investigated. This process involves the extraction of gadolinium from the europium-bearing region of highly radioactive, spent control plates used at the High Flux Isotope Reactor (HFIR) with a subsequent re-irradiation of the extracted material for the production of the ^{153}Gd .

The ORIGEN2 computer code was used to predict the gadolinium and europium isotopic concentrations and compositions at various axial positions along one spent HFIR control plate. The calculational results, although based on rough estimates of the perturbed neutron flux intensities present at each local site, supported the decision to proceed with the experimental determination of the viability of the proposed supplemental ^{153}Gd production scheme.

To initiate the research, one spent control plate was assayed at several axial positions. At each location, a gamma scan was conducted to determine the relative intensity of the radioactive europium and gadolinium isotopes. From this information, the region of peak ^{152}Gd concentration could be inferred from the maximum activity levels of its parent isotope, ^{152}Eu , and its activation product, ^{153}Gd . The control plate was cut into thin strips for further evaluation. Following separation of the gadolinium and europium elements from each strip, a mass spectrographic analysis was made on each sample to obtain the isotopic composition. Based on these analyses, the "lower bound" to the usable portion of the control plate for ^{152}Gd extraction could be established. A "best fit" correlation between axial control plate location and an "effective" total neutron exposure over the plate lifetime was obtained and judged as sufficient for obtaining estimates of the gadolinium concentrations of other control plates.

Based on the results of the experimental and calculational analyses, up to 25 grams of valuable gadolinium ($\geq 60\%$ enriched in ^{152}Gd) resides in the europium-bearing region of the HFIR control components of which 70% is recoverable. At a specific activity yield of 40 curies of ^{153}Gd for each gram of gadolinium re-irradiated, 700 one-curie sources can be produced from each control plate assayed.

INTRODUCTION

The High Flux Isotope Reactor (HFIR) is a 100-MW (thermal), light-water-cooled and -moderated, beryllium-reflected, flux-trap type research reactor operated by Martin Marietta Energy Systems, Inc., at the Oak Ridge National Laboratory (ORNL). The HFIR was designed and built in the 1960s primarily for the production of transuranic isotopes in a high thermal-neutron flux field on the order of $3 \text{ to } 5 \times 10^{15}$ neutrons/cm²-s, one of the highest in the world.

Thirty-eight vertical irradiation facilities are located in the 30-cm concentric ring of beryllium which surrounds the fuel region (Fig. 1). This arrangement was designed to accommodate various materials irradiation damage studies and lighter radioisotope production. Since the HFIR first achieved criticality in 1965, these facilities have been the site of over 5000 irradiations to produce isotopes for a variety of applications ranging from the preservation of food through gamma irradiation (cobalt-60) to the nondestructive testing of structural welds in submarines (iridium-192).

One of the most important radioisotopes produced in the HFIR is gadolinium-153 (¹⁵³Gd), an extremely useful radionuclide for the diagnosis and monitoring of osteoporosis, a disease which affects the mineral content and structural integrity of the bones of an estimated 50 million people. The low energy decay characteristics (gamma doublet at 100 keV and x-ray doublet at 44 keV) and the 242-day half-life combine to make this isotope an ideal source for a medical imaging technique known as dual-photon absorptiometry or

Fig. 1. Cross-sectional view of HFIR at horizontal midplane.

commonly referred to as DPA. This bone-scanning method measures the difference between the emitted intensity of the source and the detected intensity after passing through the soft tissue and bone of the scan medium. The bone mineral content can be calculated from this difference and compared to normal values to determine the severity of the disease or to monitor the progress of the prescribed treatment. By using a radioisotope such as ^{153}Gd with photon emissions at two different energy levels, the need for a constant soft tissue thickness (required for single-photon absorptiometry) across the scan path can be eliminated. This allows for the use of DPA in previously inaccessible areas such as the spine where bone loss first becomes apparent due to a greater rate of mineral turnover.¹⁻⁵

ORNL first began producing ^{153}Gd in HFIR in 1980, and until recently has been the world's sole supplier. The demand for ^{153}Gd sources in 1985 necessitated a rapid step-up in its production at ORNL (Fig. 2).⁶ Experts in the field of radiation medicine practices estimated that due to technological developments like DPA and improvements in the treatment of bone diseases, the demand for imaging services would increase to the point where the use of absorptiometry to diagnose and monitor osteoporosis could possibly exceed all other medical procedures combined.⁷

For eleven consecutive HFIR cycles from the summer of 1985 to the spring of 1986, every available vertical experiment facility (VXF) position was used for the irradiation of target capsules to produce ^{153}Gd . To supplement this

Fig. 2. Curies of ^{153}Gd produced at ORNL, 1980-1986.

production, additional targets were exposed in the Oak Ridge Research Reactor (ORR). However, with the ORR scheduled for decommissioning in October 1987, the need for an alternate means to supplement the production of ^{153}Gd became evident.

GADOLINIUM-153 PRODUCTION AT ORNL

Irradiation of Europium Targets

Gadolinium-153 is produced primarily by a neutron capture reaction in the stable isotope, ^{152}Gd . Various methods of producing ^{153}Gd have been investigated. One involves the neutron activation of natural gadolinium. However, due to the scarcity of ^{152}Gd occurring in nature (0.2%) and the large absorption cross sections and higher abundance of the heavier gadolinium isotopes (^{154}Gd through ^{158}Gd and ^{160}Gd), the resulting specific activity of ^{153}Gd (curies of radionuclide per combined unit mass of all gadolinium isotopes) is too low to be useful for medical radiography. A higher specific activity is attainable if the gadolinium is enriched in the stable isotope ^{152}Gd ; however, the cost of such enrichment raises the price per curie of ^{153}Gd considerably. A third method, also very expensive, consists of the accelerator activation of europium-153 (^{153}Eu) by a (p,n) reaction.⁸

The most successful production method for ^{153}Gd at ORNL has been through the irradiation of ^{151}Eu and the resultant reaction chain:

This process is accomplished through the irradiation of a natural-europium (0.48 ^{151}Eu and 0.52 ^{153}Eu) oxide powder (Eu_2O_3) in the HFIR VXF's followed by the chemical separation of the resultant gadolinium fraction from the

europium fraction. The key to this production scheme is to irradiate the powder in a neutron flux field for an optimum time period to maximize the specific activity yield of ^{153}Gd . Exposure past this optimum continues to increase the total concentration of ^{153}Gd , but also results in the production of the heavier gadolinium isotopes which acts to reduce the specific activity. A detailed production and decay reaction chain which describes this process is shown in Fig. 3. This figure has not been extended beyond the 156 atomic weight since the contributions from the higher mass europium and gadolinium isotopes are negligible. The nuclear data shown were obtained from the most recent values tabulated in the literature.^{9,10}

Fig. 3. Nuclear reaction and decay paths in the production of gadolinium isotopes.

Europium-oxide powder is routinely loaded into a standard target rod designed specifically for isotope production in one of the HFIR VXF's. While the reactor is operating, the target materials are exposed to a neutron flux on the order of 2×10^{14} neutrons/cm²-s. Europium-151 undergoes a neutron capture to one of three isomers of ¹⁵²Eu -- ^{152g}Eu, ^{152m1}Eu, and ^{152m2}Eu. The relatively large capture cross section for the production of the ground state and first isomeric state (5900 barns and 3300 barns respectively) result in a rapid burnup of ¹⁵¹Eu. The second isomeric state is characterized by a small production cross section (3.8 barns) and decays to the other metastable isomer, ^{152m1}Eu with a 96-min half-life. Its contribution can be neglected and both metastable isomers can be expressed together as ^{152m}Eu.

Both ^{152g}Eu and ^{152m}Eu are removed from the chain by (1) β^+ decay to ¹⁵²Sm, (2) β^- decay to ¹⁵²Gd, and (3) an (n, γ) reaction to ¹⁵³Eu. Despite its extremely large absorption cross section (70000 barns), the contribution of ^{152m}Eu to the production of ¹⁵²Gd far exceeds that of ^{152g}Eu because of its much shorter decay half-life (9.3 h compared to 13.6 years) and the branching ratio which favors ¹⁵²Gd over ¹⁵²Sm.

As the activation time is lengthened, the specific activity of ¹⁵³Gd begins to decrease as the heavy gadolinium isotopes, primarily ¹⁵⁴Gd and ¹⁵⁶Gd, are produced. These stable isotopes are formed both through the decay of the heavy europium radionuclides resulting from neutron capture reactions on europium isotopes and the activation of ¹⁵³Gd (36000 barns) and ¹⁵⁵Gd

(60900 barns). These products are often referred to as "dead" gadoliniums because of their unfavorable contribution to the specific activity of ^{153}Gd .

Two of the heavy europium radioisotopes produced, ^{154}Eu and ^{155}Eu , decay with relatively long half-lives (8.8 years and 4.96 years respectively). These two isotopes and the 13.6-year half-lived ^{152}Eu are the major contributors to the high radiation levels encountered during the separation of the gadolinium and europium fractions. Gadolinium-153 must be virtually "europium-free" (<0.01% radiochemically pure) to be a marketable product. On the average, for every gram of Eu_2O_3 powder irradiated in HFIR, 2.6 curies of ^{153}Gd are produced. The europium-to-gadolinium ratio of the irradiated target material is on the order of 17 to 1 which results in a large volume of radioactive by-products generated by the chemical separation process used to achieve this high chemical purity.¹¹

The average specific activity levels of ^{153}Gd resulting from the irradiation of Eu_2O_3 -powder targets in HFIR are in the 40-45 Ci/g range. The calculated theoretical maximum specific activity is about 75-80 Ci/g. This idealized calculation assumes that the target material is irradiated for an optimum time and that self-shielding and flux depression phenomena are not present. Since the VXF's can only be accessed during HFIR shutdowns which occur every three weeks, target irradiations are flux rather than time dependent. Unperturbed flux intensities at the HFIR irradiation sites are known; however, due to the large capture cross sections of the ^{151}Eu , an accurate determination

of the perturbed flux levels within the target capsules is hampered by the self-shielding and flux depression present within the targets. Without the knowledge of an accurate local neutron flux and the dependency on the HFIR cycle length for target exposures, the optimum irradiation length can only be determined through trial and error. Several configurations have been attempted and thus far, a one-cycle irradiation in one of the inner VXF positions has been concluded as the closest to optimum arrangement. Since ^{153}Gd is sold by the curie, an attempt to maximize the quantity without adversely affecting the quality of the product has been achieved by loading the target capsules to capacity with Eu_2O_3 powder. This tends to perturb the local flux within the capsules additionally, but does not significantly affect the product specific activity.

Two additional factors that contribute to the reduction of the specific activity are the ingrowth of the heavy gadoliniums and the decay of ^{153}Gd during processing. The first of these factors is minimized by the relatively short (21-day) irradiation periods. Mass spectrographic analyses have shown that about 84% of the product gadolinium is ^{152}Gd and only 14% is ^{154}Gd and ^{156}Gd . The second factor, however, can not always be avoided. Quite often the processing time (time from reactor shutdown or "push" time until product shipment) exceeds three to four weeks resulting in a 6 to 8% reduction in the specific activity of ^{153}Gd due to decay.

The ideal ^{153}Gd production process would be one in which the target material consists of greater than a 70% enrichment of ^{152}Gd and the irradiation time is dictated by the available flux rather than the flux being varied to suit the exposure length. One of the facilities at the HFIR consists of a hydraulic tube inserted into the area of maximum thermal neutron flux -- the central flux-trap region. It is designed to irradiate milligram quantities of material for exposure times independent of the reactor cycle time. Irradiation of a small amount of gadolinium enriched in ^{152}Gd for a short time period in the high flux region of the HFIR could result in a product with a specific activity comparable to or exceeding that of the "traditional" process. In addition, process and decay losses could be eliminated since further separation might not be required after "push" time. The only missing link to this process is the supply of gadolinium enriched in the 152 isotope which has already been stated as essentially nonexistent in nature. In the early 1970s, a study was undertaken to determine the feasibility of extracting the gadolinium isotopes from the europium-bearing region of the spent HFIR control plates whose usable lifetime had expired.¹² The primary objective of this investigation was to extend this research by showing that these control plates are a valuable resource of enriched gadolinium which can be used as an alternate target material in the production of ^{153}Gd , and to substantiate this claim through computer calculations coupled with experimental analysis.

Alternate Production Scheme

Since the HFIR control plates have as a constituent the same Eu_2O_3 powder as is irradiated for the europium target production process, an identical neutron activation and isotope decay scheme will effect the conversion of a large portion of the europium to gadolinium. The intention of the 1970's research was to locate and extract the portions of the control plates which contained a high concentration of ^{152}Gd , separate out the gadolinium fraction, and re-irradiate the product gadolinium oxide powder using the HFIR hydraulic tube as an irradiation vehicle for the production of ^{153}Gd . This process is attractive in that the chemical separation of the europium fraction occurs before the ^{153}Gd production irradiation, thus reducing process and decay losses and, most significantly, a higher specific activity of ^{153}Gd can be obtained since the concentration of heavy gadoliniums can be significantly reduced by selecting a portion of the plate with a low total exposure and thus, a high ^{152}Gd isotopic assay.

The control plate research of the 1970s was abandoned at that time due primarily to the inability to chemically separate the gadolinium and europium fractions to a radiochemical purity level sufficient to obtain ^{153}Gd specific activities above 30 Ci/g. In addition, reliable yields and specific activities could not be predetermined since the flux and cross section parameters vary greatly depending upon the region and total exposure of the plate analyzed.¹³ A review of the data obtained in the 1970's analyses raised some doubt as to the

validity of some of the tests and calculations that were performed. Current revised calculations of the "expected" gadolinium isotopic compositions in the control plates using updated flux and cross section information supported the decision to proceed with a program of experimental analyses to validate the computer calculations. Plans were formulated to dissect a representative spent control plate into small pieces and analytically determine the optimum locations within the plate for the extraction of ^{152}Gd .

HFIR CONTROL PLATES

Description

The neutron-generation chain reaction of the HFIR is controlled by altering the effectiveness of the beryllium reflector. This is accomplished by moving the control elements in such a manner as to regulate the flow of thermal and epithermal neutrons from the fuel region to the reflector and back into the core. Figure 4 is a schematic view of the HFIR core region showing the relative positions of the fuel, the control components, and the beryllium reflector.

The control components are located in the annular region between the fuel assembly and the beryllium reflector. They consist of two 6.4-mm-thick concentric cylinders separated by a 3.3-mm coolant channel. The outer cylinder is divided into four shim-safety quadrants each having its own drive rod and scram mechanism. The inner cylinder, which is a single unit, serves as both a shim and regulating element. Since the inner cylinder is the equivalent of four outer control plates welded together, references to the term "control plates" will generally include the inner cylinders as well. To control the reactor reactivity, the absorber section of the control elements is driven out of the core. The outer control plates are driven upward to increase reactivity while the inner cylinder is driven downward for the same effect.

The control cylinders are divided into three longitudinal sections, each having different neutron absorption characteristics (Fig. 5).¹⁴ The lower section

Fig.4. Schematic view of HFIR core region.

Fig. 5. Cutaway view of HFIR control components.

of the inner cylinder and the upper section of the outer plates contain 31 vol % Eu_2O_3 dispersed in aluminum. These sections are highly neutron absorbing and are called the "black" regions. The central sections contain 38 vol % tantalum dispersed in aluminum. They are less absorbing and are called the "gray" regions. The upper section of the inner cylinder and lower section of the outer plates are comprised solely of aluminum and are known as the "white" regions.

Control Plate Motion

The HFIR control plates and cylinders are designed for 100,000 MWd of operation in the reactor. This corresponds to about forty-five to fifty 21-day reactor cycles. Each cycle begins with the control cylinders (the outer plates are operated "ganged" as a cylinder) completely inserted (Fig. 6). The black regions are adjacent to the core to absorb the fission-generated neutrons and thus, maintain the reactor in a subcritical or shutdown condition. The reactor becomes critical by withdrawing these neutron-absorbing regions from the core which allows some of the neutrons to "escape" from the fuel through the open neutron "window." The beryllium reflector adjacent to the control plates reflect the neutrons back into the core. As the fuel is depleted, the cylinders are withdrawn further to maintain the HFIR critical at its design full power of 100 MW.

By the end of each cycle, the absorbing region of the cylinders have been completely withdrawn from the core after having traveled a distance of

Fig. 6. HFIR control plate positions during a cycle.

66 cm. This wide range of motion has a considerable effect on the neutron exposure of the control plate. The axial thermal neutron flux distribution in the HFIR at the beginning of a fuel cycle is demonstrated in Fig.7. Similar plots have been made for fluxes of other spectral groups for both the beginning and end of cycle. These two-dimensional representations, based on computational analysis performed during the design of the HFIR¹⁵ and validated with measurements made subsequent to initial reactor startup, convincingly display the effect of the strong absorbent, ^{151}Eu , on the neutron flux intensity at the control plate location. This variation in local neutron fluence in the control plate contributes to a large diversity in the concentration and composition of the gadolinium isotopes along the length of the europium-bearing region. It was initially conjectured in the early research that the highest concentrations could

Fig. 7. Calculated thermal flux per unit power in critical core at beginning of HFIR cycle.

be extracted from that portion of the control plate near the europium-tantalum ("black-gray") interface. However, due to the relatively higher fluxes near this interface, burnup of the lighter gadolinium isotopes and subsequent production of the heavier ones, resulting in a low ^{152}Gd isotopic composition, would appear to argue against this supposition. The actual flux intensities within the control plates had not been determined at this time and without some knowledge of the europium exposure and burnup, the residency of the optimum gadolinium fraction could not be resolved.

COMPUTATIONAL ANALYSIS

Description of ORIGEN2

The ORIGEN2 (Oak Ridge Isotope Generation and Depletion)¹⁶ computer code was used to calculate the optimum location of ¹⁵²Gd in a selected HFIR control plate whose usable lifetime had expired. This code, an updated version of the ORIGEN code written by the Chemical Technology Division at ORNL in the late 1960s and early 1970s, is used primarily to calculate the buildup and depletion of isotopes in nuclear materials. It accomplishes this through a matrix exponential method which solves a large system of coupled, linear, first-order ordinary differential equations of the form

$$\frac{dX_i}{dt} = \sum_{j=1}^N f_{ij}\lambda_j X_j + \phi \sum_{k=1}^N g_{ik}\sigma_k X_k - (\lambda_i + \phi\sigma_i) X_i, \quad i = 1, \dots, N \quad (1)$$

where

X_i = atom density of nuclide i ,

N = number of nuclides,

f_{ij} = fraction of radioactive disintegrations by other nuclides (j) which lead to the formation of nuclide i ,

λ_j = radioactive decay constant of nuclide j ,

ϕ = position- and energy-averaged neutron flux,

g_{ik} = fraction of neutron absorptions by other nuclides (k) which lead to the formation of nuclide i ,

σ_k = spectrum-averaged neutron absorption cross section of nuclide k .

The integration of this set of simultaneous differential equations for each of the existing isotopes (dependent on the input material) yields the amount of each nuclide present at the end of each desired time step.

The ORIGEN2 computer code employs three principal types of input data bases: cross section, radioactive decay, and photon production. The set of cross sections used by ORIGEN2 are effective, one-group cross sections developed for a variety of reactors and fuels. The library of cross sections for a ^{235}U -enriched UO_2 pressurized water reactor (PWR), arrived at through the processing of the 84-group AMPX master cross-section library obtained from the ENDF/B-IV file,¹⁷ is typically used for HFIR calculations. The decay and photon libraries are obtained from data listed in the ENDF/B-IV file and the Evaluated Nuclear Structure Data File.¹⁸ In addition to the data provided in ORIGEN2, an option may be used to substitute decay or cross section data for specific isotopes. The original ORIGEN2 data bases were used for all calculations with an estimated value for those isotopes for which no cross section and decay parameters existed in the library.

Assumptions and Validity of Initial Estimates

The variation of the intensity of the HFIR neutron flux field is a function of the position of the control plates. In the axial direction, this variation is even more pronounced in the vicinity of the highly absorbent europium-bearing regions of the control plates. Estimated flux intensities taken from flux maps similar to Fig. 7 are listed in Table 1. They typify the variation in the thermal and

Table 1. Estimated neutron flux intensities at selected control plate locations

Control plate location	Thermal flux ^a		Epithermal flux ^{a,b}	
	BOC	EOC	BOC	EOC
Eu-Ta interface	7.5	1.0	25.0	1.5
100 mm from Eu-Ta interface	1.0	0.5	15.0	0.9
250 mm from Eu-Ta interface	0.5	0.3	10.0	0.6

^aIn units of 10^{13} neutrons/cm²-s.

^bFlux from 0.414 to 101 eV.

BOC - beginning of cycle.

EOC - end of cycle.

epithermal fluxes at three control plate locations at the beginning and end of the operating cycle. The europium-tantalum (Eu-Ta) interface is the line of demarcation between the "black" and "gray" regions of the control plate.

The large time-dependent variations and uncertainty in determining the local neutron flux intensities from these flux maps make it extremely difficult to model the control plates to obtain a time-dependent or cycle-integrated neutron flux at each location of the control plate with flux intensity as a variable.

However, the accuracy necessary to determine the region of useful gadolinium does not require that the flux be "pinpointed" at each location. Thus, a range of values were used as input parameters to ORIGEN2. For each value chosen, ORIGEN2 results were obtained for the isotopic compositions and concentrations that would be present if the control plate had been exposed to the input "effective" neutron flux over its entire lifetime. This analysis requires

the following assumptions:

1. azimuthal variation in neutron flux intensity is negligible;
2. the effective flux is constant from cycle to cycle;
3. the control plate position vs. cycle time is approximately the same for each cycle.

Since the fabrication process for the control plates requires a homogeneous distribution of the Eu_2O_3 powder and all fuel elements and cycles are essentially identical in regard to element fabrication, fuel burnup, and cycle length parameters, these assumptions are all considered valid.

Predicted Optimum Location of Gadolinium-152

A sample ORIGEN2 program for a typical input flux intensity (1.5×10^{13} neutrons/cm²-s) is displayed in Fig. 8. The input irradiation/decay scheme is based on the actual installation and removal dates of control plate 7-1 (the first plate from the seventh set used in the HFIR), which was chosen for the computational and experimental analysis. These alternating irradiation and decay periods accounted for a total of 922 days of neutron exposure at 100 MW and almost 18 years of radioactive decay. One gram of natural europium was used as the input material for each case analyzed so that all output would be based on a per gram basis rather than for the entire control plate. The effective neutron flux was varied from 10^{11} to 10^{15} neutrons/cm²-s for the initial set of computer runs. The output results included the concentration of each isotope in grams, the total radioactivity of the radioisotopes, and the isotopic composition

```

TIT   IRRADIATION OF CONTROL PLATE 7-1 FOR 92178 MWD IN HFIR
CUT -1
RDA   SPECIFY DATA BASES TO BE PRINTED
LIP   0 0 0
RDA   SPECIFY DECAY AND CROSS SECTION DATA TO BE SUBSTITUTED
LPU   641530 631540 -1
LPU   631521 641530 -1
RDA   READ DECAY AND CROSS SECTION DATA BASES
LIB   0 1 2 -3 204 205 -206 9 3 0 1 1
RDA   INPUT CASE BASIS FOR CALCULATION
BAS   1 GRAM NATURAL EUROPIUM OXIDE POWDER
INP   -1 1 -1 -1 1 1
MOX   -1 1 0 1.0
BUP
RDA   INPUT IRRAD/DECAY SCHEME AND EFFECTIVE FLUX - TIME IN DAYS
IRF   93.04 1.500E13 1 2 4 2
DEC   105.96          2 3 4 2
IRF   92.10 1.500E13 3 4 4 2
DEC   111.90          4 5 4 2
IRF   93.15 1.500E13 5 6 4 2
DEC   121.85          6 7 4 2
IRF   94.42 1.500E13 7 8 4 2
DEC   1454.58         8 9 4 2
IRF   91.69 1.500E13 9 10 4 2
DEC   177.31          10 11 4 2
IRF   67.01 1.500E13 11 12 4 2
DEC   787.99          12 1 4 2
IRF   91.19 1.500E13 1 2 4 2
DEC   1802.81         2 3 4 2
IRF   153.59 1.500E13 3 4 4 2
DEC   447.41          4 5 4 2
IRF   145.59 1.500E13 5 6 4 2  IRRAD TO END OF LIFE 10/2/84
DEC   378.0           6 7 4 2  DECAY TO GAMMA SCAN DATE 10/15/85
DEC   549.0           7 8 4 0  DECAY TO 1ST MASS SPEC ANALYSIS DATE 4/4/86
DEC   724.0           8 9 4 0  DECAY TO LAST MASS SPEC ANALYSIS DATE 9/26/86

BUP
RDA   PRINT CALCULATED RESULTS
OPTL  24*8
OPTA  24*8
OPTF  3 3*8 3 8 3 17*8
RDA   INPUT OUTPUT COLUMN HEADINGS
HED   6 10/2/84
HED   7 10/15/85
HED   8 4/4/86
HED   9 9/26/86
OUT   9 1 -1 0
END
3 631510 0.479 631530 0.521 0.0 0.0 1 GM NATURAL EU203 POWDER
0
/*
//GO.FT03F001 DO *
/** SUBSTITUTION DATA FOR DECAY CONSTANTS AND CROSS SECTIONS
3 641530 4 241.6 0 1.0 3*0.0
3 2*0.0 0.1524 0.0 3.000E-09 2.0E-04
3 631540 5 8.8 5*0.0
3 2*0.0 1.509 0.0 1.0E-10 2.0E-05
206 631521 12000 5*0.0 -1.0
206 641530 6000 5*0.0 -1.0
/*
//
ENDINPUT

```

Fig. 8. Typical input program to ORIGEN2 for control plate calculations.

of each element by atom fraction that were present following each time step of the input irradiation/decay scheme of the control plate.

The concentrations of the parent and activation product of ^{152}Gd (^{152}Eu and ^{153}Gd respectively) calculated by ORIGEN2 for control plate 7-1 were individually normalized to their maximum concentration. As shown in Fig. 9, these peak concentrations occur at fluxes of 2.2×10^{13} (^{152}Eu) and 3.8×10^{13} (^{153}Gd) neutrons/cm²-s. As expected, the highest total radioactivity of these two radionuclides occurs at the flux intensities yielding the peak isotopic concentrations (Fig. 10). The maximum concentration of ^{152}Gd , located at a flux of 2.7×10^{13} neutrons/cm²-s, falls between these peaks. Thus, the highest concentration of ^{152}Gd in the control plate should be located at a point between the maximum radioactivity levels of ^{152}Eu and ^{153}Gd which can be measured by gamma scanning. However, due to burnup of the ^{152}Gd at this flux intensity, which may result in a low isotopic composition of ^{152}Gd relative to the heavier gadoliniums, this control plate region may not contain the optimum material for the production of a high specific activity ^{153}Gd product.

To establish the effective flux for the optimum ^{152}Gd concentration requires a determination of the assay of the gadolinium isotopes for each flux intensity. At a flux of 2.7×10^{13} neutrons/cm²-s, ^{152}Gd comprises about 65% of the total gadolinium fraction. The two primary "dead" gadoliniums, ^{154}Gd and ^{156}Gd , makeup the bulk of the remaining 35%. As shown in Fig. 11, as the flux intensity increases (or for the locations of the control plate closer to the Eu-Ta interface), the percent composition of ^{152}Gd decreases due to its

Fig. 9. Normalized concentrations vs effective neutron flux intensity.

Fig. 10. Radioactivity vs effective neutron flux intensity.

Fig. 11. Gadolinium isotopic compositions vs effective neutron flux intensity.

increased burnup rate and the accelerated production of the heavier gadoliniums. This isotopic breakdown is undesirable for ¹⁵³Gd production. Therefore, even though the maximum quantity of ¹⁵²Gd exists for effective fluxes in the vicinity of 2.7×10^{13} neutrons/cm²-s, the optimum qualitative portions of the control plate are located further away from the Eu-Ta interface.

Figure 12 graphically shows how rapidly the ¹⁵²Gd concentration relative to the total gadolinium concentration falls off as the neutron flux intensity increases. Based on the results of the ORIGEN2 calculations, it was determined that the "lowest" usable segment of the control plate (toward the Eu-Ta interface) would be that location corresponding to an effective flux of

Fig. 12. Gadolinium concentration vs effective neutron flux intensity.

2.7×10^{13} , the point of maximum ^{152}Gd concentration. The "upper" bound would depend on the actual quantity of gadolinium found in those locations during the experimental analysis phase.

EXPERIMENTAL ANALYSIS

Overview

The experimental phase of this research was conducted using the same outer control plate chosen for the computational analysis. To facilitate management and transfer between the HFIR and the High Radiation Level Experiment Laboratory (HRLEL) where the initial assays were to be performed, one of the spent outer control plates was chosen for this investigative program over the more cumbersome inner cylinders. The control plate chosen had been retired from service on October 2, 1984, after accumulating 92,178 MWd. The black region was separated from the white region by an underwater tool that cuts through the material with a "nibbling" type action. The europium-bearing portion was then transferred to the HRLEL in a shipping cask where it was moved into one of the hot cells for further study.

The overall plan for the control plate research included five separate operations and assays:

1. an axial scan at the HRLEL to locate the regions of highest gamma activity for each of the four major contributing radioisotopes;
2. the dissection of the control plate into thin strips and small pieces for localized analysis;
3. the separation of the gadolinium and europium fractions;
4. the mass spectrographic analysis at various axial locations to establish the local isotopic compositions; and

5. the irradiation of the resultant gadolinium portion in the HFIR to determine the achievable ^{153}Gd specific activity for each enrichment of ^{152}Gd .

The results obtained from the various phases of the experimental program were used to validate the ORIGEN2 calculations.

Axial Gamma Scans

Following transfer to the HRLEL in October 1985, control plate 7-1 was subjected to a series of gamma scans performed at ten different axial locations. Due to the diverse neutron exposures at each axial location, a large variation in the total gamma intensity, as well as the individual activities of the four major radioisotopes, was expected. The gamma intensities of the dominant photon energy assayed at each axial location is detailed in Table 2. Since these energy levels ranged from 86.6 keV (^{155}Eu) to 1.27 MeV (^{154}Eu), an absolute determination of the isotopic concentration of each radionuclide could not be obtained. However, since the geometry and counting duration (50 min) were the same for each scan, the individual relative concentration of each radioisotope as a function of axial control plate location was established. This allowed the normalization of the data measured (Fig. 13) and the determination of the location of peak concentration for each individual radionuclide. The Eu-Ta interface was fixed as the "zero" point for all experimental measurements. All other axial locations are relative to this reference.

Table 2. Measured gamma intensity in photons/s at selected control plate locations

Axial location (mm)	^{153}Gd (97.4 keV)	^{152}Eu (344.3 keV)	^{154}Eu (1274.5 keV)	^{155}Eu (86.6 keV)
-25	0.70 ± 0.12	*	31.4 ± 0.11	1.0 ± 0.12
-12.5	0.60 ± 0.13	1.3 ± 0.18	154.1 ± 0.26	12.3 ± 0.21
25	*	18.4 ± 0.25	173.2 ± 0.31	11.5 ± 0.34
50	2.80 ± 0.19	38.9 ± 0.30	155.0 ± 0.31	10.3 ± 0.27
75	2.75 ± 0.19	51.0 ± 0.29	131.5 ± 0.33	6.2 ± 0.40
100	2.31 ± 0.19	58.0 ± 0.28	89.3 ± 0.23	4.7 ± 0.23
125	1.00 ± 0.12	52.9 ± 0.31	54.9 ± 0.17	2.3 ± 0.18
150	0.69 ± 0.12	46.2 ± 0.18	34.2 ± 0.14	1.5 ± 0.18
250	•	19.8 ± 0.04	6.4 ± 0.02	0.3 ± 0.02
325	•	5.0 ± 0.03	1.6 ± 0.02	0.3 ± 0.03
ARE ^a	13.0	2.13	0.35	6.46

*No data obtained.

^aAverage relative error in percent.

Fig. 13. Measured radioactivity vs axial location.

The accuracy of the gamma scan measurements is dependent on the energy level and abundance of the photons detected. The error associated with the gamma detection rate for each radioisotope is a function of the difference between the detected energy and the centroid of the energy peak for each radioisotope decay photon in the gamma scan computer library.¹⁹ Thus, the error is much smaller for the dominant ^{154}Eu gammas than for the weaker and less abundant ^{153}Gd . The energies chosen for this analysis were selected based on their dominance (>25%) and separation from other detected gamma energies of interest emanating from the control plate.

The first two scans (at the -25- and -12.5-mm locations) were performed on the gray region of the control plate which does not contain any rare earth elements. However, due to the scanning geometry, some activity overlap was observed accounting for the low readings that were measured in the scans of the tantalum-bearing region. This explains the large slope that appears for ^{154}Eu and ^{155}Eu whose peak activities occur in the region of highest neutron exposure. At the 250- and 325-mm locations, the intensity of the ^{153}Gd decay gammas could not be discerned from the dominant europium radioisotopes. These scans were not taken until five months after the previous eight scans which allowed an additional 35% decay of the ^{153}Gd .

A comparison of the curves displayed in Fig. 13 with the normalized intensity curves shown on Fig. 10 from the ORIGEN2 output indicates that the peak isotopic activities occur in the same order when one recognizes that the neutron flux intensity decreases as the distance from the Eu-Ta interface

increases. The gamma scan measurements mark the peak concentration of ^{152}Gd between the 75- and 100-mm locations where the peak activities of its parent and activation product isotopes occur. However, the lower boundary for the purpose of the experimental assays was set at the 25-mm location to ensure that all possible residencies of gadolinium concentrations were investigated. The next step in the procedure was to dissect the control plate into smaller pieces so that localized analyses could be performed.

Dissection of Control Plate and Separation of Gadolinium Fraction

At the HRLEL, a tool similar to the underwater nibbling tool at the HFIR was used to divide the control plate into smaller segments. Ten "cuts" were made radially through the control plate. By the action of the "nibbler," each cut resulted in several small, approximately 6-mm cube, pieces, or "nibbles" of control plate. The first cut was made 25 mm from the inferred Eu-Ta interface based on the gamma intensities observed in the gamma scans. Additional cuts were made every 25 mm out to the 250-mm position. The nibbles from each cut were segregated into a small canister and labelled for further study. The resultant strips of control plate between each cut (approximately 19-mm wide) were marked and stored for possible future gadolinium extraction after the experimental analysis phase was completed.

Following the preparation of the localized samples, the plan called for the chemical separation of five nibbles from selected axial locations. The first set to be analyzed was taken from the 50-mm lot which corresponded to the

axial location where the highest total activity levels were measured during the gamma scans. The technique used at ORNL to chemically separate gadolinium from europium involves a series of operations which include the dissolution of aluminum to separate the rare earth fraction, the electroreduction of europium precipitated as EuSO_4 , and the final separation of gadolinium and europium in a high-pressure ion exchanger (HPIE) column. The effluent gadolinium fraction is subsequently evaporated and fired to a Gd_2O_3 powder and pressed into a pellet for final use.^{20,21} For the control plate research, an aliquot containing a representative sample from the HPIE gadolinium output was prepared for each of the sets of selected nibbles (locations 50, 100, 125, 150, 200, and 250). These prepared specimens were analyzed using a mass spectrograph to determine the isotopic composition of the gadolinium.

Mass Spectrographic Analysis

The purpose of the mass spectrographic analyses was to determine the isotopic assay of ^{152}Gd and the "dead" gadoliniums at each location in the control plate. By determining these percentages, the control plate could be qualitatively classified for its usefulness as a source of ^{152}Gd . Since the chemical separation process does not completely remove the europium fraction, it was possible to also determine the europium compositions from five of the six control plate locations. The results of the six mass spectrographic analyses are listed in Tables 3 and 4. The gadolinium data points are plotted in Fig. 14.

Table 3. Mass spectrographic analysis results for europium isotopes

Axial location (mm)	Percent of total europium				
	^{151}Eu	^{152}Eu	^{153}Eu	^{154}Eu	^{155}Eu
100	13.00 ± 0.01	8.00 ± 0.01	70.78 ± 0.11	6.58 ± 0.01	1.64 ± 0.01
125	18.10 ± 0.27	8.00 ± 0.12	67.40 ± 1.00	5.40 ± 0.08	1.20 ± 0.02
150	29.89 ± 0.06	6.40 ± 0.02	60.75 ± 0.06	2.62 ± 0.01	0.34 ± 0.003
200	37.11 ± 0.04	4.91 ± 0.02	56.70 ± 0.06	1.19 ± 0.006	0.09 ± 0.001
250	45.75 ± 0.05	1.25 ± 0.01	52.76 ± 0.05	0.25 ± 0.003	0.004 ± 0.0002

Table 4. Mass spectrographic analysis results for gadolinium isotopes

Axial location (mm)	Percent of total gadolinium				
	^{152}Gd	^{153}Gd	^{154}Gd	^{155}Gd	^{156}Gd
50	7.44 ± 0.04	0.044 ± 0.01	25.85 ± 0.08	1.34 ± 0.01	63.18 ± 0.13
100	37.20 ± 0.07	0.151 ± 0.01	33.62 ± 0.07	2.00 ± 0.01	26.68 ± 0.05
125	43.80 ± 0.44	0.147 ± 0.01	33.40 ± 0.33	2.00 ± 0.02	20.40 ± 0.20
150	60.79 ± 0.12	0.150 ± 0.01	29.76 ± 0.06	1.55 ± 0.005	7.68 ± 0.02
200	71.60 ± 0.36	0.130 ± 0.01	24.40 ± 0.12	0.98 ± 0.005	2.81 ± 0.01
250	74.50 ± 1.49	0.060 ± 0.03	24.60 ± 0.49	0.49 ± 0.02	0.26 ± 0.01

In performing the mass spectrographic analyses, extreme care was taken to discriminate between the isobars of europium and gadolinium. The errors associated with each analysis are variable depending on the quantity of each isotope present in the sample analyzed and the corrections applied to each measurement.²² At the 250-mm location, the expected concentration of europium relative to gadolinium is expected to be very high due to the low neutron exposure. Thus, the error associated with the europium analysis is on

Fig. 14. Isotopic composition of major gadolinium isotopes vs axial location.

the order of 0.1 to 5% while the error for the gadolinium nuclides varies from 2 to 50%. Overall, the results of the mass spectrographic analyses were considered to be extremely reliable and were used extensively in the final analysis of this research program.

Even though the maximum concentration of ^{152}Gd was inferred to occur between the 75- and 100-mm locations as evidenced by the axial gamma scans, the composition of the gadolinium in this region (<40% ^{152}Gd) is not enriched enough for quality ^{153}Gd production. Therefore, the lower boundary for extraction of gadolinium for the production of ^{153}Gd through re-irradiation was established at the 150-mm location where the ^{152}Gd enrichment is at least 60%.

DETERMINATION OF OPTIMUM LOCATION

Quantitative Analysis

The remaining variable to be determined at this point in the analysis was the quantity of gadolinium available for use. To obtain this information, three sets of nibbles from the 150-, 200-, and 250-mm locations were transferred from the HRLEL for chemical separation. The intent of this phase of the program was to separate the gadolinium fraction, prepare an aliquot for a mass spectrographic analysis for comparative purposes, and load a portion of the extracted gadolinium oxide (Gd_2O_3) powder into a small quartz capsule for irradiation in the HFIR hydraulic tube. The amount of Gd_2O_3 recovered from each set of nibbles would determine the number of production capsules prepared. The optimum time period in the hydraulic tube could then be bracketed for the maximum specific activity yield of ^{153}Gd .

Prior to the separation of the gadolinium fraction, a rough "material balance" was attempted on one gram of nibble from the three selected locations. A set of gamma scans was performed on an aliquot of dissolved, pre-weighed nibble. The data obtained included the measured activity per unit volume of solution which was converted to grams of nuclide per gram of control plate. The isotopic percents from the mass spectrographic analyses were applied to the quantities of radionuclide obtained from the gamma scans. This produced an estimate of the quantity of each gadolinium and europium

isotope per gram of control plate. Table 5 lists the calculated total quantity of rare earths (europium, gadolinium, and samarium) and the estimated gadolinium quantities by material balance. To measure the relative accuracy of each material balance, the ratio of the calculated rare earth quantity to the initial quantity is also listed. Since each control plate is fabricated with 1922 g of Eu_2O_3 powder (1660 g of natural europium) and blending operations ensure a homogeneous mixture of the powder throughout the black region of the control plate,²³ each 1-cm strip of control plate (33 cm in length) contains 29.7 g of rare earth (europium) initially and the expected quantity of gadolinium per strip can be calculated from the material balance.

Using the calculated gadolinium concentrations and the conservative assumption that the total gadolinium present at the 350-mm location is negligible, a "best fit" curve (Fig. 15) was drawn to represent the total gadolinium residing in each linear centimeter strip of control plate. Integration of this curve from the 150- to the 350-mm location results in a total quantity of gadolinium on the order of 33 g with an average enrichment of just over 70% ^{152}Gd . This 20-cm region of the control plate (36% of the black region) was determined to be the usable region for the extraction of highly enriched gadolinium in control plate 7-1 based on the analysis of the experimental assay data. The actual quantities would be determined following the chemical separation process performed on each set of nibbles.

Table 5. Quantitative analysis at selected control plate locations

Axial location (mm)	Quantity of rare earths (g/g _{nibble})	Quantity of gadolinium (g/g _{nibble})	Percent of initial rare earth ^a	Quantity of Gd per cm strip (g/cm)
100	.357	.077	.849	5.43
150	.439	.041	1.043	2.89
200	.333	.033	.791	2.33

^aInitial rare earth quantity - 0.421 g/g_{nibble}.

Fig. 15. Total gadolinium per linear centimeter of control plate.

Completion of Experimental Analysis Phase

The initial intent of the final phase of the experimental analysis was to separate the rare earth fractions and determine the amount of recoverable gadolinium for each strip of nibbles from the 150- to the 250-mm location. Then, depending on the quantities recovered and the evaluation of the separation operations, the decision would be made regarding how much of the remaining control plate to process into nibbles for gadolinium extraction. However, a series of unexpected events transpired which resulted in the premature termination of this program.

In the summer of 1986, it became apparent that the demand for ^{153}Gd and the use of DPA for the treatment of osteoporosis would fall short of the predicted levels. This was primarily due to the prohibitive expenses of DPA that were not being absorbed by insurance providers of the elderly, the prime victims of osteoporosis, such as Blue Cross/Blue Shield and Medicare. Other imaging techniques such as single-photon absorptiometry and computed tomography (CT) scanning are covered by most insurance plans and have been improved significantly such that DPA is no longer competitive.^{24,25} The demand on ORNL for producing ^{153}Gd decreased drastically and could quite easily be supplied by the traditional production method.

Only three control plate locations (150-, 200-, and 250-mm) were further analyzed due to the loss of a permanent high radiation storage site where the material to be investigated was readily retrievable. The HRLEL was shut down

for an extensive ventilation system overhaul in the fall of 1986 which forced the storage of the control plate nibbles and strips into an inaccessible shipping cask while the lengthy shutdown was in progress. Since no other facility existed to store and handle all of the highly radioactive nibbles, three sets which would provide the best information were selected for chemical separation and re-irradiation of the gadolinium fraction. The three data points obtained with regard to the total gadolinium extracted, the isotopic compositions, and the achievable specific activities for each enrichment would be extrapolated to supply information concerning the remainder of the control plate beyond the 250-mm location.

Due to an unanticipated overloading of the ion exchange column and some dissolution chemistry problems, the chemical separation process was only carried out to completion on one set (150-mm location) of nibbles. This was attributable to a miscalculation in the total quantity of rare earths contained in the nibbles which resulted in unknowingly loading the electroreduction cell and the HPIE column to more than their maximum allowable capacity. Normal chemical separation procedures require only a single pass through the electroreduction cell and HPIE column with a recovery efficiency >70%.²⁶ However, for this batch of material, four complete passes through the chemical separation process were required to achieve the required radiochemical purity. In addition, it was discovered that the rare earth fraction was contaminated with cesium-137 (^{137}Cs) which had originated in the HRLEL hot cells. By the time this problem was detected, four gadolinium product runs had been cross

contaminated with the ^{137}Cs . The untimely detection of this problem coupled with the erroneous reporting of the rare earth quantities left the 200-mm location samples in an irrecoverable state.²⁷

Only 85 mg of 60% ^{152}Gd -enriched powder was recovered from the separation process performed on the 150-mm nibbles, less than 5% of the amount calculated to be present. This extremely low recoverability can be explained by:

1. the failure to efficiently separate the rare earths resulting in much of the desired material being entrained in the aluminum fraction of the control plate;
2. the overloading of the HPIE column with an excess amount of rare earth beyond that allowed by procedure; and
3. the multiple passes required, each pass reducing the amount of gadolinium recovered.

By attempting to dissolve the aluminum through a batch process for the 250-mm location nibbles rather than in small quantities as the 150-mm nibbles had been handled, a gelatinous mixture of solvent and rare earth materials was obtained. This material is presently being stored until a decision is made concerning further separation of the mixture.

Despite this setback, plans were formulated to continue with the experimental program by establishing the optimum irradiation time in the HFIR hydraulic tube and determining the specific activity yield for this target material. However, the HFIR was shut down in November of 1986 for an indefinite period

of time to investigate the nature of a possible brittle fracture of the reactor pressure vessel. No further analytical data could be obtained until the HRLEL overhaul was completed and the HFIR restarted. Thus, the gadolinium extraction research experimental phase was terminated.

Comparison of Experimental and Computational Results

The computational estimates that preceded the experimental phase of the gadolinium extraction program were compared to the analytical data by a "best fit" method of associating an effective neutron flux intensity with an axial location along the control plate. The ORIGEN2 outputs used for this comparison were the normalized activities of the four major contributing radioisotopes (Fig. 10), the gadolinium isotopic compositions (Fig. 11), and the europium isotopic compositions. These data were compared to the normalized activities from the axial gamma scans and the measured compositions of the mass spectrographic analyses respectively. The method used to find a correlation between estimated effective flux intensity and axial location was to assume that each set of analytical compositions for an individual isotope was correct and then to associate the control plate location for that percentage to an effective flux from the ORIGEN2 data that would yield the same percentage composition. This procedure was used for each radionuclide in the axial gamma scans and for the major gadolinium and europium isotopes that were present in the mass spectrographic analyses. The results are tabulated in Table 6.

Table 6. Effective neutron flux intensities^a at selected control plate locations

Isotope	Axial location (mm)				
	0	50	100	150	200
<i>Axial Gamma Scans</i>					
¹⁵³ Gd	14.6	4.5	2.0	0.7	0.3
¹⁵² Eu	17.0	5.5	2.2	0.9	0.4
¹⁵⁴ Eu	18.0	12.0	5.0	2.0	0.5
¹⁵⁵ Eu	25.0	15.0	7.5	3.5	2.0
<i>Mass Spectrographic Analyses</i>					
¹⁵² Gd		14.0	6.0	3.2	2.0
¹⁵⁶ Gd		>25.0	12.0	5.0	3.0
¹⁵¹ Eu			2.0	0.8	0.4
¹⁵³ Eu			2.5	1.0	0.5
¹⁵⁴ Eu			4.0	1.5	0.8

^aIn units of 10^{13} neutrons/cm²-s.

Upon analysis of this data, the observation was made that as the mass of the nuclide (or the "distance" from the initial constituents in the production and decay chain) increased, the correlated effective flux for each control plate location also increased. The thermal cross sections and resonance integrals of some europium and gadolinium isotopes (in particular, ¹⁵³Gd and ¹⁵²Eu) have not been experimentally determined with any degree of certainty. Thus, the accuracy of the calculations resulting from input errors in the effective cross sections for these two isotopes and any others in this process will be multiplied as the production and decay chain reaction progresses.

The magnitudes of the peak isotopic compositions for ^{153}Gd and ^{154}Gd as calculated by ORIGEN2 (0.28% and 60% respectively) were nearly twice that assayed in the mass spectrographic analyses (see Table 4). Therefore, no correlation data could be obtained for these isotopes. Since these percentages as determined by ORIGEN2 were considered to be erroneous, the isotopic compositions of ^{152}Gd and ^{156}Gd were also questionable. Thus, the values listed in Table 6 for ^{152}Gd and ^{156}Gd were not used in deriving the "best fit" correlation curve of control plate location vs effective neutron flux intensity shown in Fig. 16.

This relationship was obtained as an approximation to the best average flux that exists at each axial location over the lifetime of the control plate. Total flux intensities (thermal and resonance) as listed in Table 1 for the beginning and ending of the HFIR cycle are plotted for comparison. The derived effective flux curve appears to underestimate the actual flux intensity beyond the 150-mm location; however, the flux information for this region of the control plate is highly uncertain.

Since each control plate has its own irradiation history, the total neutron exposure or fluence will vary from plate to plate. By inputting this irradiation history and the effective flux into the ORIGEN2 code, an estimate of the concentrations by flux, and thus, by axial location, can be acquired. To compare the results of the experimental data and the ORIGEN2 data for plate 7-1, computational analyses were conducted for three other spent HFIR control plates.

Fig. 16. Control plate axial location vs effective neutron flux intensity.

The calculated neutron fluence at the 150- and 350-mm locations for the experimental control plate, based on the effective flux estimates, are 7.24×10^{20} and 1.9×10^{19} neutrons/cm² respectively. The axial locations for the three additional control plates corresponding to these fluences were determined, and the estimated fertile region of each plate was established. Using the ORIGEN2 output for total grams of gadolinium per gram of europium feed material, a curve similar to Fig. 15 was generated for each of the three cases. Integration of the curves over the established usable region provided an estimate for the total concentration of gadolinium residing in each 20-cm portion of the control plates. Table 7 summarizes the results of these calculations.

Table 7. Comparison of calculated gadolinium concentration in selected control plates

Control plate	Accumulated MWd	Lower boundary (mm)	Upper boundary (mm)	Gd concentration in fertile region (g)
7-1 ^a	92178	150	350	33.3
7-1 ^b				18.4
8-4	96533	155	355	16.5
6-3	69952	135	335	17.9
4-2	41622	105	305	14.8

^aMass balance calculations

^bORIGEN2 calculations

The derived relationships between the axial control plate locations and the effective neutron fluxes and exposures (fluence) are recognized as incomplete and open for significant improvement. However, for the purpose of estimating the location of usable gadolinium concentrations within a spent HFIR control component, this correlation can be used to supply adequate information. Comparison of the computational efforts thus far to the experimental results indicates that the current ORIGEN2 model for the HFIR control plates conservatively estimates the total gadolinium concentrations and overestimates the ¹⁵²Gd assays. It is hoped that future experimentation using the recovered material will assist in determining better gadolinium cross section information and allow some tailoring of the ORIGEN2 inputs to better simulate actual flux conditions.

CONCLUSION

In summary, the research which investigated the feasibility of extracting enriched gadolinium from spent HFIR control plates was considered successful even though it could not be carried out to its desired completion. The confidence in the data obtained through the mass spectrographic analyses has inspired an investigation into the isotopic composition of the gadolinium powder that had been shelved as surplus from the europium target irradiation process. It was determined that over 85% of the gadolinium in the processed material awaiting shipment was ^{152}Gd , considerably higher than that of the small amounts of control plate analyzed. Plans are currently being studied for the re-irradiation of these targets as well as the material recovered from the 150-mm location of control plate 7-1 in the HFIR hydraulic tube. It has been calculated that a short irradiation will not adversely increase the radiochemical composition of the europium radionuclides and that marketable specific activities of ^{153}Gd can be produced from other depleted gadolinium sources without further chemical separation.

The confidence placed in the calculated quantities of gadolinium recoverable from the irradiated control plates and the lessons learned in the chemical separation process have encouraged researchers to continue with this program as soon as operations at ORNL permit. By applying the expected 70% recoverability of the gadolinium fraction in the "fertile" (>60% enriched) region of the control plate and a 40 Ci/g specific activity, a yield of up to 700

curies of ^{153}Gd per control plate can be achieved. The spent HFIR control plates can be considered an excellent resource for the supplemental production of an extremely useful and reliable radioisotope.

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