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Gadolinium-153 Production at the Oak Ridge National Laboratory

D. W. Ramey

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GADOLINIUM-153 PRODUCTION AT THE OAK RIDGE NATIONAL LABORATORY

Operations Division

D. W. Ramey

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Gadolinium-153 Production at the Oak Ridge National Laboratory

D. W. Ramey

Abstract

Gadolinium-153 production at the Oak Ridge National Laboratory (ORNL) involves the neutron irradiation of natural europium oxide (47.8% ^{151}Eu , 52.2% ^{153}Eu). This target material undergoes a series of neutron captures and radioactive decays to produce the desired ^{153}Gd product. Several undesirable europium isotopes (^{152}Eu , ^{154}Eu , and ^{156}Eu) are also produced during this irradiation process. Recent technical advances and other improvements in the radiochemical processing of this isotope have allowed ORNL to increase production by more than sevenfold. A newly developed electrochemical process separates the bulk of undesirable europium isotopes. The use of this process increases the efficiency of the high-pressure ion exchange step and effects a final product purity of >99.999%. Specific activities >60 Ci/g of gadolinium oxide and product specific yields >2.9 Ci/g of irradiated europium oxide have been produced. Use of unique glove box manipulators and special equipment designed at ORNL have allowed final source fabrication to keep pace with the increased production rate while minimizing the radiation exposure to operating personnel.

1. Introduction

In the late 1960s, a study (1) at ORNL funded by the National Aeronautical and Space Administration (NASA) was initiated to determine the most suitable radionuclide to generate a gamma backscatter signal for applications in an atmospheric density measuring device for the NASA's Mars probe. Gadolinium-153 ($t_{1/2} = 242$ d) was chosen as the most suitable in view of its energy characteristics, output, half-life, and cost. Two schemes were suggested for the production of ^{153}Gd : one involved the neutron irradiation of ^{152}Gd and the other the neutron irradiation of ^{151}Eu . Irradiation of ^{151}Eu was selected as the most favorable method of producing multicurie quantities of ^{153}Gd . Various purification methods such as coprecipitation with SrSO_4 (Jones reductant method), electrochemical purification with a lithium amalgam electrode, and high-pressure ion exchange were investigated. Early

production work used the electrochemical process but later gave way to high-pressure ion exchange (2). The more recent development of an electroreduction process (3) in conjunction with high-pressure ion exchange has combined to form a series of radiochemical processes which now consistently yields a product with only a few parts per million of radiochemical impurities.

At present, ^{153}Gd is one of the most important medical and industrial radioisotopes produced at ORNL. Industrial applications include mass unit absorption gauging, thickness monitoring, and X-ray fluorescent applications. This radionuclide is also extremely useful in the field of medicine for the diagnosis and monitoring of osteoporosis, a serious bone disease which affects an estimated 20 million people in the United States. In the medical field, ^{153}Gd is presently being used as the photon source in dual-photon absorptiometry (DPA) machines which are used to determine bone mineral content in osteoporosis patients. DPA diagnostics have caused a drastic increase in the demand for ^{153}Gd in the past several years. In the early 1980s (through 1983), the demand for ^{153}Gd had averaged approximately 30 Ci per year, but by April of 1986, ORNL had outstanding orders for 1,500 Ci. To meet the demand for this isotope, ORNL made 25% more irradiation positions available for ^{153}Gd production; redesigned the reactor target such that a 67% increase in Eu_2O_3 per target was achieved; cut radiochemical process losses in half by improving existing procedures and by using better production data tracking techniques; and, through several research and development efforts, produced an electroreduction process that had the effect of quadrupling the radiochemical processing capacity.

1. Gadolinium-153 Production Details

Irradiation of Europium-151. The method presently being used for the production of ^{153}Gd is through the neutron bombardment of natural europium oxide (47.8% ^{151}Eu and 52.2% ^{153}Eu) powder. During irradiation, ^{151}Eu captures a neutron (n, gamma reaction) and produces ^{152}Eu (see Fig. 1). A certain fraction of the ^{152}Eu radioactively

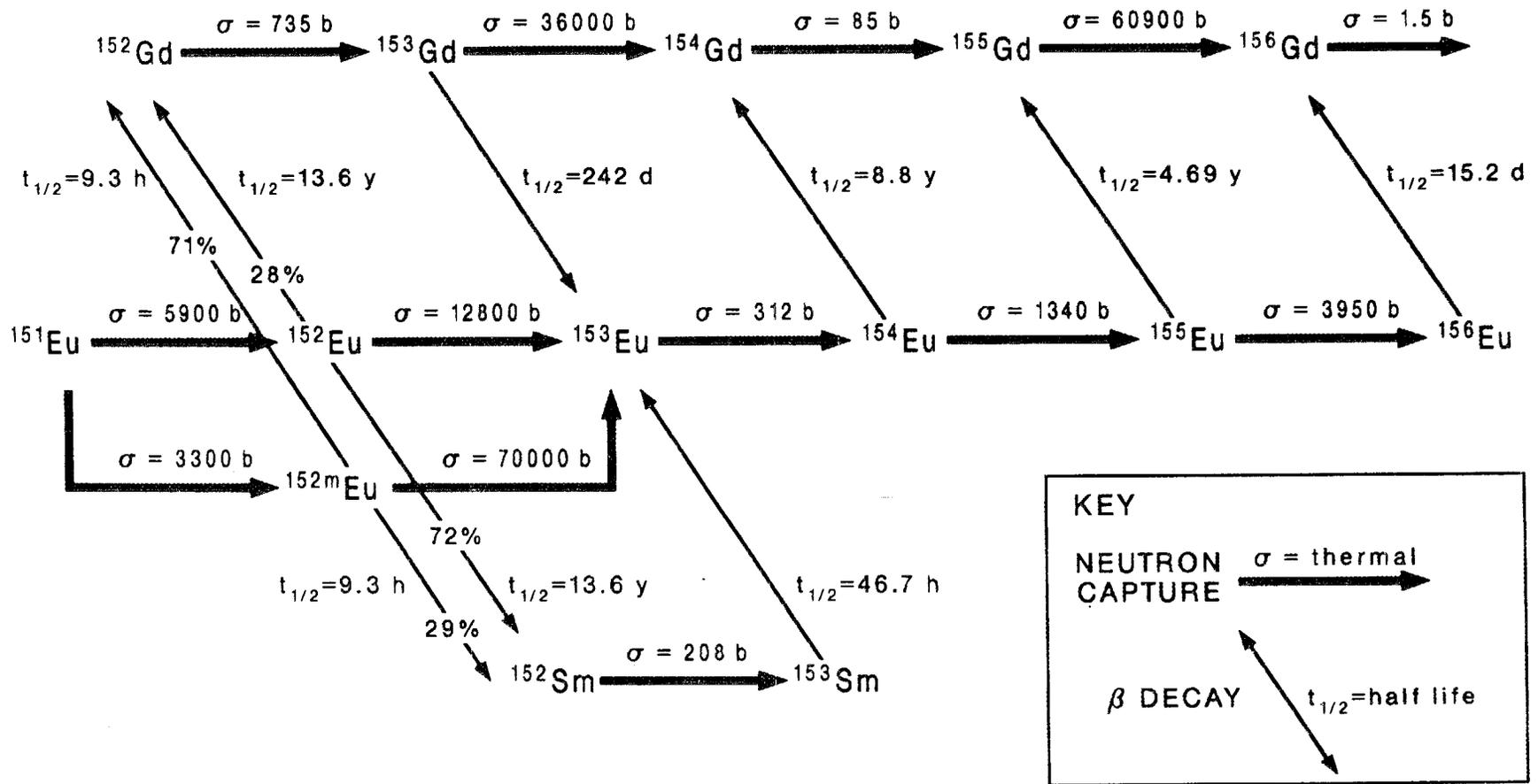
decays (by beta emission) to ^{152}Gd , which then captures a second neutron to produce the desired ^{153}Gd product. The remaining fraction of the ^{152}Eu undergoes further successive neutron captures to produce heavier europium isotopes. These heavier europium isotopes are undesirable byproducts of the irradiation process. Radiochemical separation processes must be used to separate the europium waste from the gadolinium product.

A standard target rod designed for isotope production in either the High Flux Isotope Reactor (HFIR) or the Oak Ridge Research Reactor (ORR) is used for the irradiation of the europium oxide. Target irradiation in the HFIR is performed either in the inner VXF^a position (for one cycle) or the outer VXF position (for two cycles). Oxide powder is loaded and flame sealed into quartz tubes 3 mm ID by 1 mm wall. Each quartz ampule contains 0.3 g of the oxide and is approximately 57 mm long. Five ampules are inserted and centered in each standard target rod which is then welded, examined and tested by several quality-control procedures. The total target length including spacers between each ampule is approximately 300 mm. Target rods are inserted in the HFIR in multiples of four or in the ORR in multiples of six per irradiation position. These targets experience an average neutron flux in the range of 2.5×10^{14} (ORR) to 3.5×10^{14} (HFIR outer VXF) or 8.0×10^{14} neutrons/cm²·s in the HFIR inner VXF position. Normal irradiation cycles for targets in the ORR, HFIR outer VXF, or the HFIR inner VXF positions are nine, six, or three weeks, respectively.

After irradiation, the series of targets are allowed to "cool-down" in the reactor pool from four to six weeks before being transferred to the hot cell facility for chemical processing. This cool-down period permits the decay of all short-life activation products generated during reactor irradiation. The decay of short-life isotopes facilitates safer target transport from the reactor to the hot

^a Vertical experiment facility

NUCLEAR REACTION AND DECAY PATHS IN THE PRODUCTION OF GADOLINIUM-153

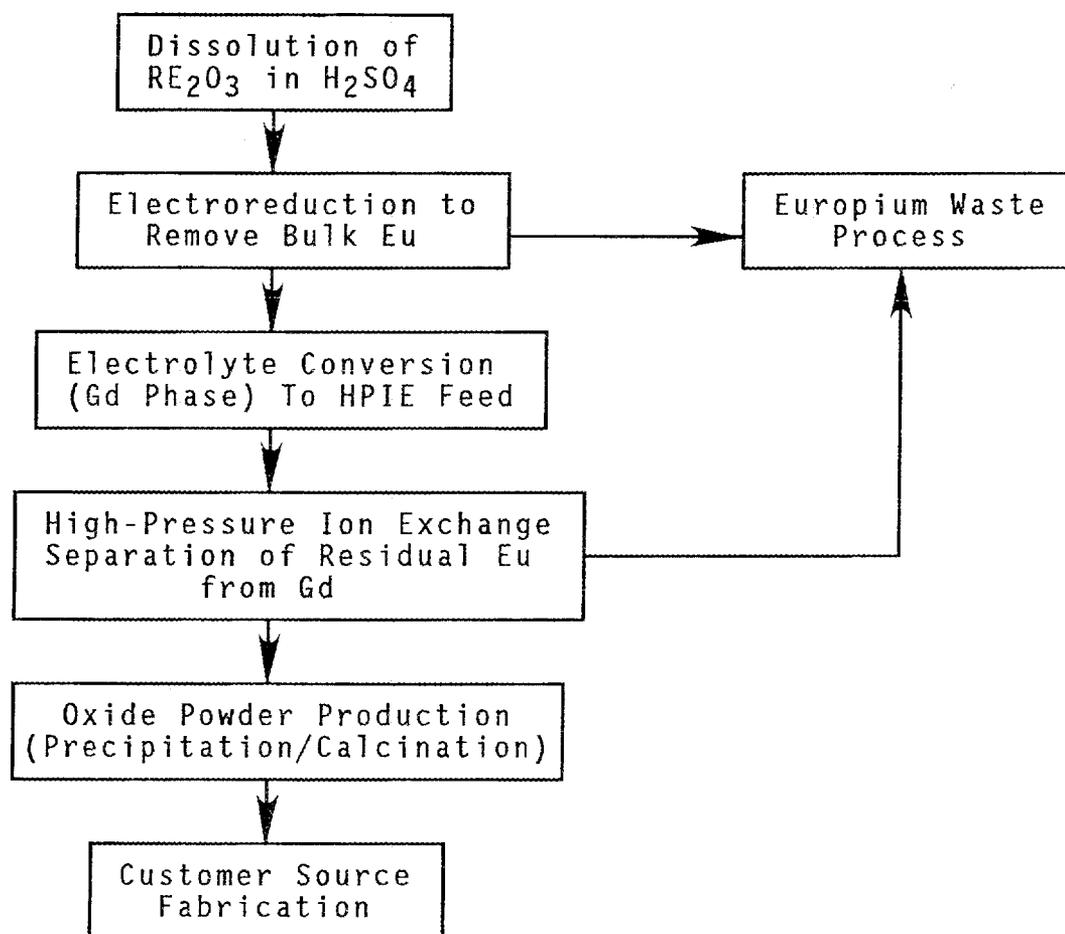


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1 Nuclear Reaction and Decay Paths in the Production of Gadolinium-153

cell facility and, in the case of ^{156}Eu ($t_{1/2} = 14.7$ d) and ^{153}Sm ($t_{1/2} = 46.7$ h), reduces radiation background and radiation damage in all phases of chemical processing.

Radiochemical Processing - Electroreduction. Once in the hot cell facility, the irradiated targets are disassembled, and their contents are processed by various wet chemistry techniques to separate the europium waste from the gadolinium product. An outline of all the radiochemical steps used in purifying the gadolinium product is presented below.

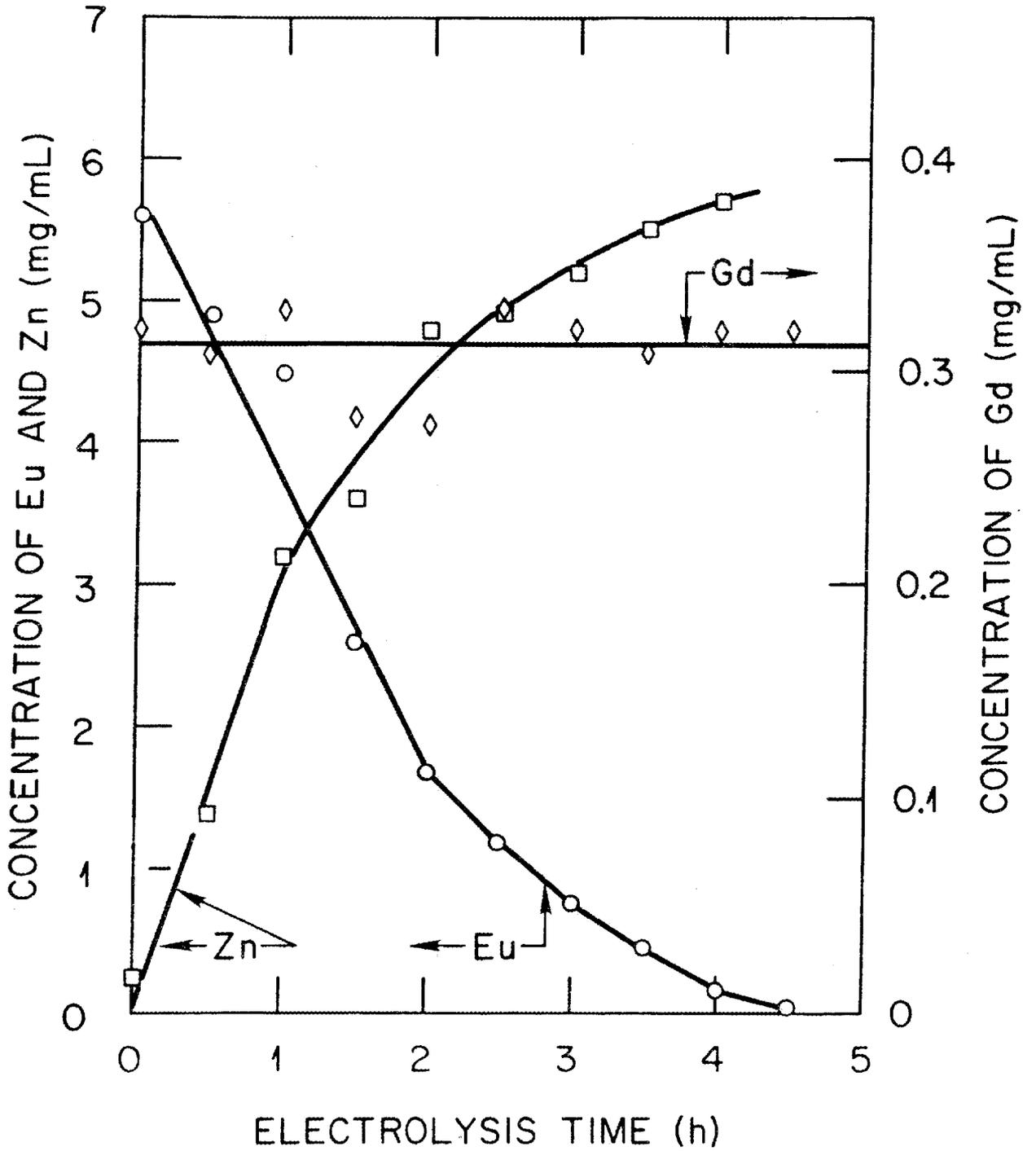


The quartz ampules from the target rods are crushed, and the rare earth oxides (Eu_2O_3 and Gd_2O_3) are dissolved in 1 N H_2SO_4 . This sulfuric acid solution is diluted to 0.25 N in preparation for the electroreduction process. The electroreduction separation technique

(developed at ORNL) utilizes the fact that the trivalent Eu(III) can be reduced to a lower oxidation state at a potential (cell voltage) at which Gd(III) remains unaffected. During electrolysis, the Eu(III) is reduced to the bivalent state, forms an insoluble sulfate, and is precipitated as EuSO_4 while leaving behind the Gd(III) still dissolved in the electrolyte. The europium-to-gadolinium ratio, initially at approximately 17 to 1, is typically reduced by more than two orders of magnitude (a decontamination factor of 10^2) in 4 to 5 h by the electroreduction process. The present electroreduction cell design will allow the processing of from 3 to 7 g of this europium-gadolinium oxide mixture, with a typical charge being 6 g. Each run is carried out at constant current with a current density of approximately 30 mA/cm^2 and an electrode wetted surface area of 83 cm^2 . The data in Fig. 2 illustrate the effect of this process on both the europium and gadolinium content of the electrolyte as a function of time. Note that the gadolinium content remains virtually unaffected by this process. The appearance of zinc in the electrolyte is from the electrolytic dissolution of the zinc electrodes used in this process. Further details of the electroreduction process can be found in Reference (3).

Radiochemical Processing - High-Pressure Ion Exchange. After the electroreduction step, the gadolinium product is removed from the electrolyte (and separated from the zinc) by a simple ammonium hydroxide precipitation. The resulting gadolinium hydroxide precipitate is dissolved in 8 N HNO_3 , evaporated to incipient dryness, and redissolved in 0.05 N HNO_3 in preparation as feed for the high-pressure ion exchange process.

With the bulk of the europium having been removed by the electroreduction process, a further separation is achieved by applying high-pressure ion exchange. Final purification is performed on a 1.7-cm-I.D. by 50.8-cm-long column with design features similar to those found in work reported by Brown and Callahan [Reference (2)]. Column features considered advantageous for hot cell operation include in-situ resin column loading from outside the hot cell, resin transfers within the column plumbing which allow backwash and repacking of the

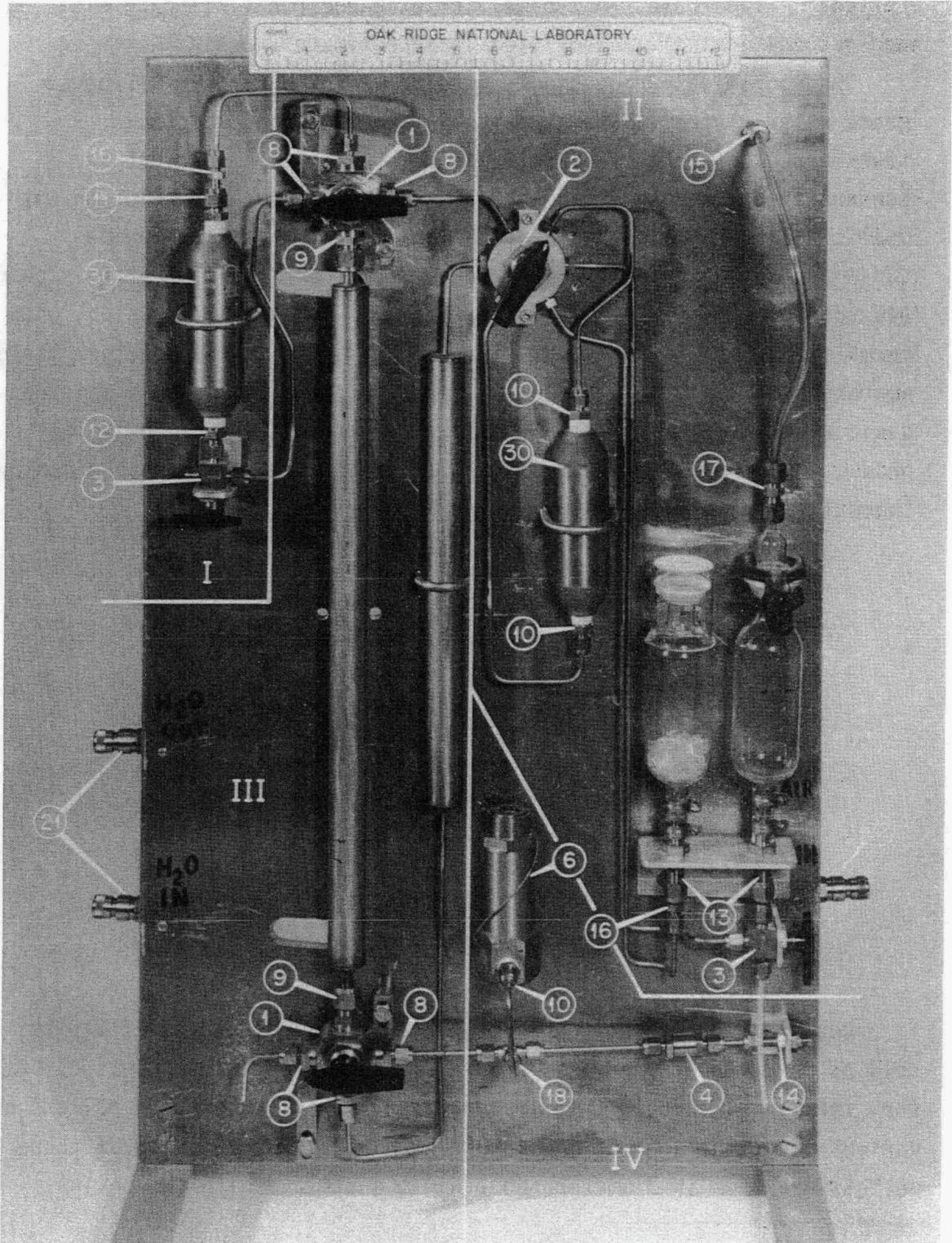


2 Performance Characteristics of an Electroreduction Cell

resin, and a contamination-free high-pressure feed pump located outside the hot cell environment (see Fig. 3). Details of the column's basic operational modes can be found in Reference (2). With the Dowex-50 type cation-exchange resin, the maximum capacity of this column size is 1.5 g of rare earth oxides. The column is eluted with 0.15 M alpha hydroxyisobutyric acid (AHIB) at a pH of 4.5. The eluent flow rate is 25 mL/m with a column temperature and pressure of 85°C and 700 psig, respectively. Real-time gross gamma radiation monitoring of the column effluent is used to track the separation of the gadolinium phase from the europium phase. The gadolinium phase will appear in the column effluent in 1 to 2 h from the time the AHIB is introduced. The gadolinium phase is collected in approximately ten cuts of 100 mL each. A sample of each 100-mL cut is removed from the hot cell and analyzed by gamma spectroscopy. Cuts containing more than the allowable europium are saved and consolidated for a recycle run. Product grade cuts are transferred to the precipitation/calcination facility for final processing. Starting with a 6-g charge for electroreduction, typically, an ion exchange run will recover approximately 16 Ci of ^{153}Gd with a radiochemical purity^b of >99.99% (routinely >99.999%) and an additional 1 to 2 Ci which must be recycled. Assuming a europium-to-gadolinium ratio of 0.17 in the feed, after ion exchange this ratio is reduced by approximately four orders of magnitude (a decontamination factor of 10^4).

When the end of the gadolinium phase is evident (by a sharp decrease in the radiation reading), product collection is stopped and the column eluent is switched to 0.5 M AHIB at a pH of 6.0 (eluted with approximately 25 column volumes) to hasten the elution of the europium phase. Column stripping of any residual ions is accomplished with 3 M HNO_3 to a point where the radiation probe returns to the baseline reading. The resin is then backwashed into a reservoir and repacked in the ion exchange column. The backwashing and repacking procedures are necessary to prevent resin packing with each successive run to the

^b Ci ^{153}Gd /Ci total radioactive rare earths



3 High-Pressure Ion Exchange Column: I - Resin Backwash Reservoir, II - Feed Loading Section, III - Ion Exchange Column, IV - High-Pressure Eluent Inlet

point where ever increasing pressures would be needed to achieve a reasonable flow rate. A typical ion exchange run (including cleaning and reconditioning) is completed in approximately 6 h.

Radiochemical Processing - Oxide Powder Production. Product grade material from the high-pressure ion exchange process is transferred to another hot cell where the gadolinium product is concentrated by evaporation, precipitated from the AHIB solution with oxalic acid, and calcined in air at 800°C for 5 h, to the oxide (Gd_2O_3). Typical specific activities of the oxide powder at this point (40 to 60 d out of the reactor) range from 45 to 55 Ci/g^c which, when normalized to the date the material was pushed from the reactor, yields approximately 60 Ci/g. The oxide powder is then stored in a shielded desiccator until ready for customer source fabrication. A typical product-grade gadolinium isotopic abundance analysis, as determined by thermal emission mass spectroscopy, is shown in Table 1.

Table 1. Typical ^{153}Gd product isotopic abundance

| Isotope | Weight percent |
|--------------------------|----------------|
| ^{152}Gd | 87.64 |
| ^{153}Gd | 1.76 |
| ^{154}Gd | 6.81 |
| ^{155}Gd | 0.19 |
| ^{156}Gd | 3.57 |
| $^{157},^{158},^{160}Gd$ | 0.03 |

Customer Source Fabrication. ORNL's final ^{153}Gd product may be sold as oxide powder, pressed oxide pellets, or chloride solution. By far, the largest product form requested has been for pressed pellets of various sizes for application in DPA machines. The majority of these pellets are 3 mm in diameter and 1 Ci in activity, with the pellet

^c Ci of $^{153}Gd/g$ of total rare earth oxide

with the pellet thickness varying inversely with specific activity. All pellet-pressing operations are performed in a glove box fitted with a set of special manipulators designed at ORNL (see Fig. 4). The oxide powder is weighed out according to the specific customer order, date of shipment, and specific activity of the particular batch of material. The required quantity of powder is loaded into a steel die and cold-pressed at 623,000 kPa (90,000 psi) for 10 s. With multiple-curie orders, the pellets are stack pressed in sets of three. With the aid of a vacuum handling tool, each pellet is either placed in a customer's metal source capsule or in a plastic vial for shipment to the customer. Before the design and installation of the pellet-pressing manipulator box, hand exposure, using lead-lined gloves, was averaging 1.7 rem/Ci pressed. Hand exposure now averages approximately 5 mrem/Ci ^{153}Gd pressed.

Europium Waste Processing. With every curie of ^{153}Gd produced, there are sizeable quantities of other radionuclides produced during the reactor irradiation (see Table 2). Those radionuclides with half-lives greater than one year will create a long-lived radioactive waste product (16 Ci Eu waste/Ci ^{153}Gd produced) that must be processed into a suitable form for proper disposal. For each curie of ^{153}Gd produced, 0.3 g of europium waste must be processed. Each gram of waste would produce an estimated radiation field of 450 rad/h at 1 ft.

Table 2. Radionuclide waste products of ^{153}Gd production at ORNL

| Radionuclide | Half-life | Ci Waste/Ci ^{153}Gd produced |
|-------------------|-----------|--|
| ^{153}Sm | 46.7 h | 37 |
| ^{152}Eu | 13.3 y | 12 |
| ^{154}Eu | 8.8 y | 4 |
| ^{155}Eu | 4.7 y | 0.6 |
| ^{156}Eu | 15.2 d | 10 |

Several approaches for eliminating the bulk of the europium from aqueous waste streams in the ^{153}Gd process were considered. The most



4 Gadolinium-153 Pellet-Pressing Box

desirable approach would be to remove the europium using a cation resin. Preliminary efforts have produced an ion exchange resin that can, after simple gravity flow contact, be dried and fired to an europium-aluminum titanate and hot pressed into a solid disk. This titanate waste form will offer the most leach-resistant package presently known for the europium waste. A short-term solution to the europium waste problem has been to use simple wet chemistry techniques to remove the waste from the aqueous streams of the gadolinium process and fire these rare earths to an oxide powder. Temporary storage in welded powder cans in a shielded facility has been proposed until the titanate waste form has been fully developed.

3. Production Optimization and Alternate Production Methods

Production Optimization. The computer code ORIGEN2 (4) was used to predict the gadolinium and europium isotopic weight fractions, the curies of each radioisotope, and the mass of each isotope as a function of time. Table 3 gives a summary of the isotopes of interest predicted by ORIGEN2^d that would be produced from 1 g of natural europium (47.8% ¹⁵¹Eu and 52.2% ¹⁵³Eu) irradiated for a 21-d cycle in a HFIR inner VXF position. A total of 3.14 Ci of ¹⁵³Gd is predicted to be produced from this 21-d cycle. The calculated specific activity of ¹⁵³Gd at the end of the cycle is 60.10 Ci/g of gadolinium oxide with a maximum specific activity of 60.22 Ci/g occurring at day 18. Irradiation past 21 days would generate more ¹⁵³Gd but would also produce more of the heavier gadolinium isotopes (in particular, ¹⁵⁴Gd and ¹⁵⁶Gd), while at the same time maintaining a relatively high level of ¹⁵²Gd. All these reactions will combine to reduce the specific activity,^e i.e., 51 Ci/g at the end of a 44-d irradiation (i.e., two HFIR cycles).

^d A discussion of the effective cross sections used for the gadolinium production can be found in Reference (5).

^e Maximizing specific activity is important in DPA applications; the higher the specific activity, the less self-absorption of photons and the longer the life of the source.

Table 3. Theoretical gadolinium and europium production from 1 g of natural europium irradiated for 21 d in a HFIR inner VXF position^a.

| Isotope | Gadolinium | | Europium | |
|---------|-----------------|-----------------------|-----------------------|---------------|
| | Wt. Percent (%) | Weight (g) | Weight (g) | Activity (Ci) |
| 151 | --- | --- | 1.06×10^{-1} | -- |
| 152 | 87.07 | 4.59×10^{-2} | 2.26×10^{-1} | 39.1 |
| 153 | 1.68 | 8.89×10^{-4} | 5.43×10^{-1} | -- |
| 154 | 6.95 | 3.67×10^{-3} | 5.07×10^{-2} | 13.7 |
| 155 | 0.19 | 1.01×10^{-4} | 4.10×10^{-3} | 1.9 |
| 156 | 2.91 | 1.54×10^{-3} | 5.97×10^{-4} | 32.9 |

^a Assumed average flux of 8.0×10^{14} neutrons/cm²·s

Actual production data compare quite well with the data predicted by ORIGEN2. Table 4 gives a summary comparison of average production data and those values predicted by the computer code. From a comparison of specific yield data, a recovery efficiency >90% for the overall gadolinium separation process is indicated. However, the ORIGEN2 computer code assumes an ideal target for irradiation. No allowance is taken for flux depression caused by target material self-shielding or target shadowing by other targets. In view of the large cross sections of some of the isotopes involved in the reaction (see Fig. 1), it is obvious that a less-than-ideal target (oxide powder, 3 mm in diameter) is being irradiated, and considerable flux depression more than likely takes place within each target. In all probability, a specific yield somewhat less than the 3.14 Ci/g Eu₂O₃ predicted by ORIGEN2 is produced during irradiation. Therefore, actual product losses are probably less than the 10% indicated by the calculated specific yield data.

Table 4. Average ^{153}Gd production data compared to data generated by the ORIGEN2 computer code

| Isotopic abundance | ORIGEN2 | Production |
|--------------------|---|------------|
| | <u>Wt %</u> | |
| ^{152}Gd | 87.07 | 87.64 |
| ^{153}Gd | 1.68 | 1.76 |
| ^{154}Gd | 6.59 | 6.81 |
| ^{155}Gd | 0.19 | 0.19 |
| ^{156}Gd | 2.91 | 3.57 |
| | <u>Ci/g total Gd</u> | |
| Specific Activity | 60.10 | 60.00 |
| | <u>Ci/g irradiated Eu_2O_3</u> | |
| Specific Yield | 3.14 | 2.90 |

In view of the data generated by ORIGEN2, a higher specific activity is theoretically possible by terminating the HFIR reactor cycle at day 18. However, the HFIR cycle is fixed at a nominal 21-d period. Also, the gain in specific activity would be hardly noticeable (0.2% increase) and would be more than offset by a drop in the specific yield (>6%). Fortunately, the present 21-d HFIR cycle is well suited for the type of target that is being used for ^{153}Gd production. Redesign of the target to reduce self-shielding may yield a more optimum irradiation process but, because of the 21-d irradiation cycle, would probably produce more of the heavier gadolinium isotopes and thereby reduce the specific activity.

Alternate Production Methods. The quantity of ^{153}Gd produced is directly proportional to the concentration of ^{151}Eu in the target. Experiments at ORNL have shown that normal europium (47.2% ^{151}Eu) when enriched to approximately 95% can essentially double the ^{153}Gd production. The cost of enriched europium is approximately \$3900/g compared to \$2.25/g for normal europium.

The irradiation of ^{152}Gd is an alternative production method which has several advantages. A shorter irradiation time would be required (2 d), and the generation of multicurie quantities of the

long-lived europium isotopes would be virtually eliminated. Shorter irradiations can be accomplished in the HFIR hydraulic tube facility, where target material will experience an effective flux of 3×10^{15} neutrons/cm²·s. Shorter target cool-down time before radiochemical processing would also result from a ¹⁵²Gd target. This might possibly allow the finished product to be shipped within two weeks from the date of irradiation rather than the six to eight weeks now required (in effect, a 10% increase in specific activity at date of shipment). The disadvantage of a ¹⁵²Gd target is the availability of the target material. As pointed out by Kohring (5), the extraction of gadolinium (>60% enriched in ¹⁵²Gd) from spent HFIR control plates could be a valuable source of raw material (29 g of gadolinium oxide per plate). It should be noted that with this production scheme the europium waste problem would be transferred to the HFIR control plate processing phase rather than to the final product radiochemical processing phase. An attractive alternative to the HFIR control plates as a source for ¹⁵²Gd would be the use of spent DPA sources. After a nominal 18-month life, these sources contain >87% ¹⁵²Gd, 0.3% ¹⁵³Gd, and approximately 1.3% ¹⁵³Eu^f. From 1984 through 1987, ORNL has shipped over 1400 Ci of DPA sources or approximately 35 g of gadolinium oxide. These spent sources contain approximately 30 g of ¹⁵²Gd oxide which, if recovered, reprocessed, reirradiated, and repurified, would represent a potential of over 2300 Ci of ¹⁵³Gd with very little europium waste products (2×10^{-3} Ci Eu waste/Ci ¹⁵³Gd produced). The specific yield predicted by ORIGEN2 is approximately 66 Ci ¹⁵³Gd/g of 87% ¹⁵²Gd oxide after a 2-d irradiation in the HFIR's hydraulic tube facility.

4. Conclusions

Gadolinium-153 production capacity at ORNL has surpassed the present demand for this radioisotope. Improved data management and interpretation, removal of bulk europium by a newly developed

^f Generated from the radioactive decay of ¹⁵³Gd

electroreduction process, efficient application of high-pressure ion exchange, and utilization of an ORNL-designed pellet-pressing manipulator box all contributed to the success of the gadolinium program. A realistic estimate of the radiochemical processing capacity at ORNL, assuming an average of three runs per week at 16 Ci/run, would be approximately 2300 Ci per year. Reactor capacity at the HFIR alone (assuming natural europium targets) could supply 1 to 1.2 kg of irradiated material for processing, which is approximately 14% more material than is needed for the maximum capacity of our present radiochemical processing facilities. Product specific activities at date of shipment are consistently in the range of 45 to 55 Ci $^{153}\text{Gd}/\text{g}$ of total rare earth oxide with a radiochemical purity of >99.999%. Specific yield of the gadolinium product has averaged 2.9 Ci/g of irradiated normal europium oxide. Data from the ORIGEN2 computer code show good agreement with production data and indicate that, even though self-shielding probably is occurring, redesign of the standard target is not warranted for the production of ^{153}Gd . The utilization of spent DPA sources as an europium-free source of ^{152}Gd is an attractive alternative to the irradiation of ^{151}Eu .

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