33. X High Flux Isotope Reactor Target Development

A. L. Lotts

The goal of the transuranium project is to produce gram quantities of the heavier transuranium elements for research purposes by successive neutron captures in ^{239}Pu . The first part of the production path, the conversion of ^{239}Pu to ^{242}Pu , ^{243}Am , and ^{244}Cm , was carried out in reactors at Savannah River. Target elements containing principally these three isotopes were fabricated at ORNL and are being irradiated in the High Flux Isotope Reactor (HFIR) at a flux of approximately 3×10^{15} neutrons $\text{cm}^{-2} \text{sec}^{-1}$. The target elements are removed periodically from the HFIR and reprocessed in the Transuranium Processing Facility (TRU). At the TRU, the product actinides are separated and the target actinides are recovered and fabricated into recycle target elements, which are returned to HFIR for further irradiation. Recycle target elements must be remotely manufactured because of the high gamma and neutron radiation associated with the contained isotopes.

Our tasks have included the design and validation of the original targets, the development of equipment and techniques for fabricating these elements both in a glove box and remotely, the monitoring of the performance of the target elements in HFIR, and the design and fabrication of modified targets that may withstand the HFIR service conditions.

At this point we have completed the irradiation program for confirmation of the HFIR target element design; we have fabricated the necessary ^{242}Pu targets; we have essentially completed the cold testing of the equipment in the transuranium facility; we have refurbished targets that were irradiated at the Savannah River Laboratory; and we have commenced an investigation of the mechanism of failure of targets that had been irradiated at Savannah River and in the HFIR to fission of approximately 25% of the plutonium. In addition, a

program for short-range and long-term solution of the limited burnup problem of the targets was initiated.

TRANSURANIUM TARGET FABRICATION EQUIPMENT STATUS AND OPERATION

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During irradiation in the HFIR, the target actinide oxides are encased in an assembly consisting of 31 individual elements. Each target element² consists of 35 individually jacketed pressed pellets, each composed of a mixture of 12 vol % actinide oxides dispersed in aluminum. The pellets are contained in a type X8001 aluminum tube with discontinuous fins. These fins are subsequently attached to the target rod sheath, which positions the element in the target array and through which cooling water flows during reactor operation. Manufacture of the targets consists of three parts, (1) pellet fabrication, (2) assembly and welding of the target element, and (3) inspection of the finished element.

The development of the equipment to remotely fabricate these targets in the TRU has been essentially completed, the equipment has been installed, and part of it has been used for the inspection and refurbishment of irradiated target elements. During remote checkout of the target fabrication process some of the components were modified to improve their reliability.

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²M. K. Preston, Jr., J. E. Van Cleve, J. D. Sease, and A. L. Lotts, *Metals and Ceramics Div. Ann. Progr. Rept. June 30, 1966*, ORNL-3970, pp. 179-81.

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After equipment installation was completed at the TRU, a detailed set of operation and maintenance procedures was completed. These procedures had been under development for some time but could not be completed until the equipment had been installed and operated in the TRU.

During the past year, 18 ^{242}Pu rods, fabricated at ORNL and irradiated in a Savannah River reactor for approximately one year, were inspected and refurbished before they were irradiated in the HFIR. Details of this work are covered below.

Cell 3 in the TRU contains the equipment necessary to fabricate and inspect pellets. The inability to maintain accurate calibration of the various weighing systems resulted in poor reliability of this equipment. A systematic examination of this problem is presently under way. Short periods of operation have allowed the examination of the remaining equipment, and it has operated satisfactorily.

Cell 2 contains the equipment required to load a target rod with inspected pellets, weld the top end plug, and helium leak test the welded target rod. Difficulties with the pellet inspection and loading equipment were traced to fasteners having been loosened by the vibratory action of the pellet feeders. Each of these pieces has been secured with a locking compound. The remaining auxiliary equipment appears to be satisfactory.

The assembly machine is being used out of the cell until a spare unit is fabricated. Several alignments and relocations of components have been performed to make remote maintenance easier. The construction of a spare unit is approximately 70% complete.

Final assembly, inspection, and cleaning of the target element are performed in cubicle 1. Most of the equipment in this cell was extensively used during inspection of the SRL target elements. Minor adjustments and modifications were made on some of the equipment. This confirmed the procedures envisioned for removal and maintenance of equipment components. All three of the inspection systems were used, and the dimensional inspection apparatus was removed for calibration and is the only piece of equipment not yet operational in cell 1. Final checkout of the device is awaiting the fabrication of redesigned components. The helium leak test station and the radiography station operated satisfactorily.

To eliminate the fretting discussed in the next section from future fabricated target elements, the

hex-can staking fixture has been redesigned to include one additional staking assembly for staking the three additional sets of fins. This subassembly has been fabricated and is being adjusted and checked out before being installed remotely on the in-cell unit.

The operation of the remainder of the equipment has in general been excellent, with very few malfunctions of equipment or operator errors. The procedures for this cubicle are in excellent condition, since this particular cell has been in use for an extended period of time, primarily in refurbishing SRL target elements.

A final report covering the engineering and developmental testing of the equipment is being prepared.

INSPECTION AND REFURBISHMENT OF TARGET ELEMENTS IRRADIATED AT SRL

E. J. Manthos

M. K. Preston, Jr.¹

Eighteen HFIR target elements that had been irradiated at Savannah River for approximately one year were inspected and refurbished at the TRU Facility during the period June 1 to September 1, 1966, before they were inserted in the HFIR for further irradiation.

Each SRL target element originally contained 10 g of ^{242}Pu , dispersed in 35 aluminum pellets. These rods were identical in design, except for ^{242}Pu loading, to the targets that were to be irradiated in HFIR only.³ Each target was centered in an outer hex-can sheath by four sets of target rod positioning fins. The upper three sets of fins each contained three fins spaced 120° apart, and the lowest set of fins contained six fins positioned 60° apart. The outer sheath was mechanically attached or staked to the target rod at the bottom six positioning fins only.

Each of the 18 SRL rods was transferred to TRU and examined visually with a telescope, dimensionally inspected, helium leak tested, and refurbished before it was further irradiated in HFIR.

Suspensions that the unattached fins had been damaged by fretting during reactor operations at Savannah River were confirmed. We observed fin

³J. D. Sease, *The Fabrication of Target Elements for the High Flux Isotope Reactor*, ORNL-TM-1712 (March 1967).

Fig. 33.1. Top of Hex-Can Sheath from HFIR Target Rod, Showing Hole Worn in Sheath by Positioning Fin. 1.4X.

wear on all of the unattached target rod fins. Half of the target elements examined also had a hole in the top of the hex-can sheath where a fin had worn through just below the hex portion of the sheath, as shown in Fig. 33.1. All of the damaged sheaths were removed, and new sheaths were installed and staked to the lower six positioning fins.

A spider-type spacer, a $\frac{3}{8}$ -in.-long split ring with either three or six legs to fit the inside of the hexagonal area, was placed in the hexagonal area of the refurbished rods to center the target in the sheath. Three-legged spiders were installed on the first two refurbished rods and six-legged spiders were installed on the remainder.

Except for the fin damage, the integrity of the elements was good. We saw no evidence of localized swelling, the appearance of the welds and cladding was normal, and none of the elements showed a leak during helium leak testing. On those rods where the hex-can sheath was removed, the positions of the pellets were visible because of differences in oxide coloring on the rod. We transferred 17 of the 18 target rods to the HFIR for further irradiation. One of the rods was retained at TRU for dissolution and chemical analysis.

The design of future target rods has been modified to reduce the possibility of fretting. All fin positions will contain six equally spaced fins, and the sheath will be staked at each fin location. We removed 29 previously manufactured unirradiated ^{242}Pu target rods of the three-fin design from the HFIR storage vault and staked them at each fin location.

IRRADIATION TESTING OF PROTOTYPES

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A. E. Richt	J. P. Moore

The postirradiation examination of the four prototype target elements in the ETR to validate the target element design was reported previously.^{4,5} The four rods, which had been loaded with ^{239}Pu , performed satisfactorily to burnup levels of 2.4×10^{21} fissions/cm³. However, the extent of reaction between the dispersed PuO_2 and the aluminum matrix was found to be strongly temperature dependent. In addition, preliminary work on the fabrication of the dispersion had indicated a strong influence of oxide particle size on the thermal conductivity of the dispersion. Consequently, a detailed investigation of the effects of actinide oxide particle-size distribution and volume percent loading on the thermal conductivity of the cermet was initiated.

This investigation is not completed, but some preliminary conclusions can be derived from the available data. Surprisingly, the thermal conductivity is essentially constant up to 120°C, the upper temperature limit of our comparative thermal conductivity apparatus. This behavior for those

⁴A. R. Olsen, J. D. Sease, A. E. Richt, J. W. Ullmann, and S. D. Clinton, *Trans. Am. Nucl. Soc.* 9(1), 66 (1966).

⁵A. R. Olsen, J. D. Sease, A. E. Richt, and J. W. Ullmann, *Metals and Ceramics Div. Ann. Progr. Rept.* June 30, 1966, ORNL-3970, pp. 182-84.

specimens where the oxide is the continuous phase requires further investigation. The volume percentage of the oxide has a significant effect but is secondary to the oxide size-distribution effect on the thermal conductivity. The addition of particle sizes less than 325 mesh ($44\ \mu$) greatly reduces the thermal conductivity. The use of ceramic grade powders, that is, particle sizes less than the $5\ \mu$ typical of powders precipitated from solutions, must be avoided, since they reduce the thermal conductivity to approximately 5% of that of a comparable volume loading of larger size oxide. With usable-size particles of oxide, the sintering associated with the thermal degreasing of the pressed cermet pellets (3 hr at 550°C) increases the cermet density 3% and approximately doubles its thermal conductivity. We have substituted UO_2 for the higher actinides to obtain a thermally induced reaction between the aluminum and the oxide. The effect of this reaction on the thermal conductivity has not yet been determined.

EXAMINATION OF FAILED TARGET ELEMENTS

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J. E. Van Cleve

Irradiation of 17 HFIR target rods that had been exposed in the Savannah River Reactors (SRL) for approximately 12 months and in the HFIR for approximately 5 months was terminated when curium activity was discovered in the HFIR primary water coolant system. Suspect target rods were isolated in HFIR, and inspection of these rods revealed that 5 of 17 SRL rods had failed.

Actually, two groups of target rods were being irradiated in the HFIR at the time of failures. In addition to the group of target rods irradiated in the SRL reactors for approximately 12 months and subsequently stored in water for 8 months, a second group of targets (14) had been irradiated only in HFIR. Before irradiation in HFIR, the SRL targets were inspected in the TRU Facility, and new outer sheaths were installed on 9 of the 17 rods, as described above.

Both groups of targets were of similar design, except that the SRL targets contained 10 g of ^{242}Pu each while the virgin targets contained 8 g. Each target contained 35 individually jacketed aluminum-matrix pellets, in which the ^{242}Pu was dispersed as PuO_2 . The target rod cladding and

outer hex-can sheath were fabricated from X8001 aluminum. Both the SRL and virgin targets were irradiated for approximately the same length of time in HFIR; however, the SRL targets were distributed in the outer circumference of the target array and consequently received a greater fast-neutron dose. We are presently investigating the mechanism of failure of these elements, and the results of our preliminary work are reported below.

Examination at TRU

E. J. Manthos J. E. Van Cleve

Nine suspect SRL target elements were inspected at TRU. We determined that five of these rods (56A, 34A, 6A, 10A, and 35A) had failed; we definitely saw cracks on the cladding of four of the rods. Element 56A was selected for detailed examination. We inspected it visually, obtained diameter measurements, and x-rayed, leak-tested, and pressure-tested the end plug welds at the TRU.

The end plugs, including the welds, were removed and tested to ascertain that the failure was not caused by a defective weld that had allowed water to enter the rod. Radiographs of both welds showed that they were normal. The welds were leak checked with 80-psi air while the end plugs were submerged under water, and we saw no evidence of a leak. They were also hydraulically tested with water at pressures exceeding 7000 psi; we saw no leakage of water through the welds. Since none of the above tests indicated that the welds were defective, subsequent rods were inspected only visually, dimensionally, and for fission gas activity buildup on a charcoal trap in a nitrogen sweep test. Heat generated by the targets prevented vacuum leak testing for helium.

Rod 56A contained the target tubing crack as shown in Fig. 33.2. Oriented 90° from the crack and slightly below it was a mechanically damaged region (a chatter mark), Fig. 33.3, which was covered with oxide similar in appearance to the oxide on the remainder of the rod. Two of the other rods each contained two cracks in the same line; one of these failures is shown in Fig. 33.4. A fourth visibly failed rod contained three cracks; two were in line and the third was 120° away.

All of the cracks occurred in the same general region, at the midpoint of the pellet column, which also coincided with the maximum neutron flux in the rod. The as-received target-rod tubing con-

Fig. 33.2. Appearance of Failure on SRL-HFIR Target Rod. 1×.

tained six full-length extruded fins, 60° apart; they were partially machined off to provide discontinuous fins, which supported the outer sheath. Some of these fins are visible in Figs. 33.2 and 33.4. Every crack occurred on one of the original fin lines. In rupture tests of unirradiated machined target-rod tubing, it ruptured along the original fin lines at pressures ranging from 4000 to 4700 psi. Full-length-finned tubing ruptured at pressures ranging from 5100 to 5400 psi, and it appears that the presence of full-length fins probably would not have prevented the failures but perhaps delayed them.

We attempted to remotely measure the outside diameter of all nine target rods with hand micrometers at points along the target rod corresponding to preirradiation measurements. However, we were not able to obtain accurate and reproducible measurements.

Several types of specimens were obtained from the failed rods and their sheaths and submitted for metallography and mechanical properties testing. Metallographic samples, including both the pellet and cladding, were sectioned from rods 56A and 34A. Sections of the sheath from the maximum flux region of three targets that had been exposed in both SRL and HFIR were obtained. A section from the maximum flux region of a sheath that had been exposed in HFIR only was also obtained.

The remaining sections of the failed rods are now being chemically processed. Before they were dissolved, all of the positioning fins were removed from the rods in the event that any further examinations are required. The hex-can sheaths from all nine of the rods that were inspected are also available for further examination if necessary.

Fig. 33.3. Appearance of Abraded Region on SRL-HFIR Target Rod 56A. 1×.

Fig. 33.4. Appearance of Failure on SRL-HFIR Target Rod 6A. 1X.

Metallographic Examination of a Failed SRL Target Element

E. L. Long, Jr.

Two specimens were sectioned from the failed region of HFIR target rod 56A at the TRU Facility and transferred to the HRLEL for metallographic examination. The specimens were transverse sections taken at the end and about midlength of the fracture in the cladding. The specimens included both cladding and target pellet; the specimen taken near the end of the fracture included a "chatter" mark that apparently had occurred prior to reactor service.

Figure 33.5 shows the specimen taken from near midlength of the cladding failure. The fracture was intergranular and extended through the cladding and pellet liner and well into the target dispersion. In the cladding we saw numerous microcracks that originated at the inner surface, and several extended over halfway through the wall thickness. The fracture in the cladding was of a brittle nature; there was no measureable reduction in wall thickness. Although the target material showed evidence of plastic flow and swelling in the fracture region, there was no evidence of melting or gross reaction between the target pellet material and the aluminum matrix. Numerous fission-gas bubbles were present in the target particles.

Examination of the section taken near the end of the cladding fracture showed that this crack was intergranular and extended only through the cladding; the pellet remained intact. However, the pellet diameter had increased by about 5 mils (approximately 2%) over the as-fabricated diameter. In contrast to the specimen described earlier, no other fractures or microcracks were noted that originated at the inner surface of the cladding. One microcrack that did not completely penetrate the cladding was noted; it occurred in the region of mechanical damage (described at the beginning of this section). No gross reaction had occurred between the fuel and the aluminum matrix. Numerous fission-gas bubbles were present in the fuel particles.

The cladding specimens examined were coated with about 2 mils of a dense tightly adherent aluminum oxide, and there was no evidence of localized corrosion or pitting. Microhardness measurements of the cladding showed over a 2.5-fold increase as a result of irradiation.

Effect of Neutron Irradiation on the Mechanical Properties of the X8001 Aluminum Sheath

R. T. King J. R. Weir

The mechanical properties of the irradiated X8001 aluminum sheath material were investigated to gain understanding of the failure of the HFIR target cladding. Since suitable tensile samples could not be prepared from the cladding, ring tensile samples were cut from the hex-can sheath. The sheath and cladding material neutron doses were 6.38×10^{22} neutrons/cm² thermal and 1.07×10^{22} neutrons/cm² fast (>0.82 Mev). However, the hex can had been

Fig. 33.5. Appearance of the Transverse Section Taken Through the Failed Region of Target Rod 56A. As polished 20× Reduced 20%

irradiated at approximately 130°F, while the cladding operated at approximately 275°F.

Samples from an unirradiated sheath and from the sheath exposed both in SRL and HFIR were tested in air at temperatures ranging from room temperature to 650°F. In testing ring samples, the ultimate stress and crosshead travel to fracture are

readily determined quantities measuring strength and ductility respectively.

An irradiation-induced increase in the ultimate strength existed at testing temperatures as high as 500°F. The ratio of ultimate strengths of irradiated material to unirradiated material ranged from 1.65 at room temperature to 2.3 at 500°F. The

ductility of the unirradiated sheath increased with increasing testing temperature. The ductilities of the irradiated and unirradiated hex cans were the same at room temperature, but the ductility of the irradiated hex can decreased to a minimum near 250 or 300°F. It is significant that the ductility minimum in the hex can occurred near the operating temperature of the cladding.

Because of differences in the metallurgical history and irradiation temperatures of the cladding and hex-can material, the in-reactor failure mode of the cladding may not have been duplicated in these tests. A program for a more detailed investigation of the effects of irradiation on aluminum alloys is being outlined.

HFIR Target Failure Analysis

R. E. Adams

Cladding failures have occurred in several HFIR target rods at maximum burnup equivalent to fission of about 25% of the plutonium. All of the failures were longitudinal cracks in the cladding at regions of highest burnup. The failures are typical of those expected from excessive internal fission gas pressures. However, similar prototype rods irradiated in the ETR did not fail at fission of 69% of the plutonium.

Data from both types of rods were analyzed. The ETR rods contained ^{239}Pu to permit high fission burnup in the test program. HFIR rods were loaded with ^{242}Pu . Exposure in HFIR resulted in a much higher fast neutron exposure for the HFIR target rods. Thus HFIR target cladding accrued a fast neutron dose (>0.82 Mev) of about 1×10^{22} neutrons/cm² while 25% of the plutonium fissioned, but the fast exposure of the ETR rods was 1/15 as much (0.7×10^{21} neutrons/cm²) during fission of 69% of the plutonium.

Pellets for each type of rod contained 10% porosity to accommodate fuel swelling and to facilitate release of fission gases to end plenums. In the ETR test, less than 5% of the fission gas was found in the end plenums,⁴ and the HFIR failures did not release significant amounts of fission gases to the reactor system.

An analysis of the ETR rod indicates that at fission of 69% of the plutonium, cladding stresses caused by fuel swelling and retained fission gases would remain below the failure stress only if the cladding yielded to allow the diameter to increase about 9 mils. Measured expansion ranged between about 5 and 13 mils.

We postulate that the HFIR rods failed at lower burnup because the high fast flux reduced the ductility of the cladding so that it could not yield to relieve fission-gas pressures. If no cladding expansion occurs, calculations suggest that excessive cladding stresses would develop in a standard pellet when about 35% of the plutonium has fissioned. Such calculated internal pressures are quite sensitive to initial void volume in the fuel pellet. Variations between individual pellets (within the fabrication tolerances used at the time the pellets were made) could result in 30% variation in initial void volume. Calculations based on the smaller void volumes indicate excessive internal pressures could occur at 25% fission.

We designed and fabricated HFIR target rods that contain 20 and 25% porosity in the pellets and 8 and 6 g of plutonium per rod (in contrast to the previous rods, which contained 10% porosity and 10 g of plutonium per rod). Calculations indicate that such rods would be expected to achieve 50 to 75% fission before excessive internal pressures develop. Six such experimental rods are now being irradiated in HFIR.