29. High Flux Isotope Reactor Target Development

A. L. Lotts

The Transuranium Project is producing quantities of the heavier transuranium elements for research by successive neutron captures in ^{239}Pu . The program at ORNL started with primarily ^{242}Pu , ^{243}Am , and ^{244}Cm . Target elements containing principally these three isotopes are fabricated at ORNL and irradiated in the High Flux Isotope Reactor at a flux of approximately 3×10^{15} neutrons $\text{cm}^{-2} \text{sec}^{-1}$. In addition, special target elements of higher isotopes are fabricated periodically and irradiated in HFIR. The target elements are removed periodically from the HFIR and reprocessed in the Transuranium Processing Facility (TRU). At TRU the product actinides are separated and the target actinides are recovered and fabricated into recycle target elements. In addition, special targets for irradiation in the HFIR Hydraulic Rabbit Facility are fabricated and special encapsulations are made of the product actinides.

The program at TRU is carried out jointly by the Chemical Technology Division, the Metals and Ceramics Division, and the Analytical Chemistry Division. Our tasks in this program have included the design and evaluation of the original targets, the development of equipment and techniques for fabricating these elements, both in gloved boxes and remotely, the monitoring of the performance of the target elements in HFIR, and the design and fabrication of modified targets that may withstand HFIR service conditions. The principal activities during the past year have been the fabrication of recycle targets and special target elements and sources, improvement of the equipment in the TRU, further investigation of the target failures that were reported previously, and irradiation of targets modified to withstand the HFIR service conditions.

TRU TARGET FABRICATION

E. J. Manthos J. E. Van Cleve, Jr.

The target fabrication production line, used to fabricate target rods containing either $^{242}\text{PuO}_2$ or

$(\text{Am,Cm})\text{O}_2$ as the starting material, has operated as the feed was available. The feed material is transmuted to heavier isotopes and fissioned while in the HFIR core. The targets are removed, the fission products and heavier isotopes are separated, and the americium and curium are reconstituted into the oxide and finally assembled into new targets for additional irradiation.

The remote target fabrication equipment has been in use for approximately 18 months, and its operation has become almost routine. The only persistent trouble encountered was in the weighing systems. The scales used were modified top-loading balances. Because calibration of these scales was prone to drift, the balances were replaced with weigh cells of much sturdier construction and the same sensitivity. We have experienced no trouble with these units after approximately seven months in the hot cell. The target rods fabricated during this reporting period are listed in Table 29.1.

Small amounts of isotopes of special interest are most efficiently produced by exposing the starting material in the HFIR Hydraulic Rabbit Facility. We developed a rabbit design that is compatible with both our fabrication equipment and the requirements of the reactor. The rabbit is shown in Fig. 29.1. The encapsulation welder is used to make the end closure weld, and the unit is helium leak tested. The rabbits fabricated are listed in Table 29.2. The minute quantities of starting

Fig. 29.1. Design of HFIR Hydraulic Rabbit.

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Table 29.1. Targets Fabricated at TRU in FY 1969

Target	Oxide Form	Major Isotope	Actinide Weight (g)	Weight of Oxide (g)	Pellet Length Standard Deviation (in.)
					$\times 10^{-3}$
K26	Microspheres and shards	^{244}Cm	6.20	9.00	18
K17	Microspheres and shards	^{244}Cm	2.45	3.85	0.5
K16	Microspheres	^{244}Cm	5.37	8.03	6.3
K23	Shards	^{242}Pu	9.71	11.32	12.4
K24	Shards	^{242}Pu	9.71	11.32	17.8
D70	Shards	^{242}Pu	9.71	11.32	20.7
D75	Shards	^{242}Pu	9.71	11.32	17.8
D73	Shards	^{242}Pu	9.71	11.32	16.1
D74	Shards	^{242}Pu	9.71	11.32	15.2
D79	Shards	^{242}Pu	9.71	11.32	19.1
D77	Shards	^{242}Pu	9.71	11.32	26.4
D78	Shards	^{242}Pu	9.67	11.32	20.0

isotope are loaded into the rabbit by techniques developed jointly with Chemical Technology personnel. We prepare a pellet liner with a 90%-dense bottom cap pressed in place, slurry the isotope in solution with the aluminum core powder, vacuum the slurry through the

pellet liner using the bottom cap as a filter frit, and then press it into a pellet. The pellet is loaded into the rabbit, the end closure weld is made and inspected, and the rabbit is sent to the reactor.

Two neutron sources were assembled. We have developed a standard neutron source, Fig. 29.2, which we believe will suit most experimenters. The design requirements satisfied by the container were that it be of the physical dimensions and shape that we could handle it with our standard equipment. The two sources, shown in Table 29.2, were of the standard type made of stainless steel.

Table 29.2. Rabbits and Neutron Sources Fabricated in TRU in FY 1969

Identification	Major Isotope	Actinide Weight (μg)
HFIR Rabbits		
R02	^{253}Es	0.04
R03	^{252}Cf	10.00
R04	^{253}Es	0.50
R05	^{253}Es	0.50
R06	^{253}Es	0.50
12-68-1	^{252}Cf	10.00
R08	^{252}Cf	10.00
12-68-9	^{252}Cf	1.30
12-68-10	^{252}Cf	1.30
3-69-1	^{249}Bk	0.10
4-69-1	^{253}Es	0.50
5-69-2	^{253}Es	1.50
5-69-3	^{253}Es	0.20
5-69-6	^{253}Es	0.20
5-69-7	^{253}Es	2.50
5-69-8	^{252}Cf	800
5-69-9	^{252}Cf	1800
5-69-10	^{252}Cf	1900
5-69-11	^{252}Cf	1700
6-69-47	^{253}Es	4.00
Neutron Sources		
NS-2	^{252}Cf	500
NS-3	^{252}Cf	100

HFIR TARGET PERFORMANCE

E. J. Manthos

Status of Targets in HFIR

J. E. Van Cleve

Table 29.3 lists the target rods exposed in HFIR during the previous year. No sign of failure but an increase in the alpha activity of the primary coolant stream has been observed. The failures^{1,2} of targets irradiated at SRL and in HFIR occurred at approximately 40% FIMA, and, as may be seen in the table, several of the D series targets have exceeded this value.

¹A. L. Lotts, J. E. Van Cleve, E. J. Manthos, R. T. King, and E. L. Long, Jr., *Metals and Ceramics Div. Ann. Progr. Rept. June 30, 1968*, ORNL-4370, pp. 193-97.

²A. L. Lotts, E. J. Manthos, J. E. Van Cleve, E. L. Long, Jr., J. R. Weir, and R. E. Adams, *Metals and Ceramics Div. Ann. Progr. Rept. June 30, 1967*, ORNL-4170, pp. 187-91.

Fig. 29.2. Design of ^{252}Cf Neutron Sources Made in TRU.

Analysis of Target Performance

E. J. Manthos

Two groups of targets were removed from the HFIR and chemically processed at TRU to recover the contained transplutonium elements. One group consisted of 13 "virgin" plutonium targets that had been

Table 29.3. HFIR Target Element Performance

Rod	Burnup (% FIMA)	Fluence (neutrons/cm ²)	
		<0.41 ev	>0.81 Mev
		$\times 10^{22}$	$\times 10^{22}$
D67	27.1	6.02	1.85
D37	42.4	8.48	2.64
D35	51.9	10.13	2.59
D31	52.0	10.16	2.54
D14	58.1	11.28	3.08
D19	58.1	11.28	3.08
D21	58.1	11.28	3.08
E22	56.0	10.85	3.31
E70	62.0	12.09	2.98
E71	62.0	12.09	2.98
E72	62.0	12.09	2.98
E73	62.0	12.09	2.98
E74	62.0	12.09	2.98
E75	62.0	12.09	2.98
D12	70.5	14.07	3.54
K11 ^a	42.4	4.40	1.41
K12 ^a	42.4	4.40	1.41
K14 ^a	42.4	4.40	1.41
K19 ^a	42.4	4.40	1.41
K20 ^a	42.4	4.40	1.41
K21 ^a	42.4	4.40	1.41
D63	18.3	4.62	1.39
D68	33.0	7.03	2.17
E13	38.7	7.93	1.65
E30	38.7	7.93	1.65
E47	38.7	7.93	1.65
E49	38.7	7.93	1.65
E66	38.7	7.93	1.65
D69	38.7	7.93	1.65
D66	24.1	5.58	1.70
K16	25.5	2.63	0.76
K17	25.5	2.63	0.76
K23	8.57	2.63	0.76
K26	25.5	2.63	0.76

^aRemoved from HFIR and processed in TRU.

irradiated only in HFIR for 17 cycles. At the time of removal, June 12, 1968, they had accumulated fluences ranging from 8.19 to 9.09×10^{22} fast neutrons/cm² (>0.81 Mev) and 42 to 46% FIMA at the target midplane without any indications of failure. In comparison, the SRL-HFIR targets first failed at 39% FIMA and fluences of 6.36×10^{22} thermal neutrons/cm² and 1.04×10^{22} fast neutrons/cm² (>0.81 Mev).

There were at least two differences between the "virgin" and SRL targets. The "virgin" targets had been irradiated in HFIR only, whereas the SRL-HFIR targets

had been irradiated both at SRL and in HFIR. The plutonium loading was different: the "virgin" targets each contained 8 g ^{242}Pu and the SRL-HFIR targets contained 10 g of ^{242}Pu , both in pellets pressed to 90% of theoretical density.

According to a model developed, failure should have occurred in the virgin targets at 46% FIMA. Since none of these targets had failed at the scheduled removal time, one (D-12) was left in the reactor until failure. When D-12 fails, it will be sectioned and examined metallographically to see if the mode of failure is the same as for the SRL-HFIR targets. As of June 12, 1969, D-12 had been irradiated to 70.5% FIMA, 14.07×10^{22} thermal neutrons/cm², and 3.54×10^{22} fast neutrons/cm² (>0.81 Mev) without any indication of failure.

The 13 "virgin" targets were visually examined at TRU before they were processed, and we saw no evidences of failure on any of the targets. They were normal in appearance and covered with a light-colored oxide deposit. A section of the target tube and pellet was obtained from the midplane of target D-65 and submitted for metallographic examination. In addition, several cover sheath sections were also obtained and transferred to the HRLEL for further examinations.

The other group that was removed for processing comprised six curium recycle targets. They were removed from HFIR in February 1969 and had accumulated 4.40×10^{22} thermal neutrons/cm², 1.41×10^{22} fast neutrons/cm² (>0.81 Mev), and 44.4% FIMA. Each contained 4.5 g of curium, mainly ^{244}Cm , in pellets pressed to a density 80% of theoretical.

These six targets were the first curium recycle targets that had been fabricated in TRU. Each target was thoroughly examined after it was returned from HFIR. We saw no evidence of failure, and the targets were covered with a light-colored oxide deposit typical of targets that have been irradiated in HFIR.

As of June 1969, no target has failed for over two years since irradiations of the 10-g-Pu SRL-HFIR targets were terminated. At present seven targets in the HFIR have been irradiated for over two years. The six low-density targets^{1,2} have achieved a burnup of 62% FIMA and target D-12 70.5% FIMA. Planned exposure of future plutonium targets will be limited to twelve 23-day HFIR cycles and approximately 29% FIMA. Curium targets will be exposed for nine cycles and 40% FIMA.

All targets that have been fabricated since the SRL-HFIR target failures and all future targets will contain pellets pressed to 80% theoretical density instead of 90%. The failure model¹ predicts that target

life will be extended if more porosity is available in the pellets. Since all future targets will contain pellets with more porosity and irradiation time will be 12 cycles or less, we believe that the probability of target failures in HFIR has been reduced, if not eliminated. Surveillance of all HFIR targets will continue, and each target removed from HFIR will be carefully examined for any evidence of failure before it is chemically processed.

IRRADIATION EFFECTS ON MECHANICAL PROPERTIES OF CLADDING

R. T. King

The failure of the 8001 aluminum TRU target rod cladding has been investigated and ascribed to neutron irradiation effects, as described in Chap. 5 of this report. Our present work is directed toward understanding the causes of low-ductility failures in neutron-irradiated aluminum and determining which alloys are suitable for cladding use in the HFIR environment. We have found several neutron-irradiation effects that might increase the strength of irradiated aluminum and decrease its ductility: (1) the production of helium by $^{27}\text{Al}(n,\alpha)$ reactions with fast neutrons; (2) the production of hydrogen by $^{27}\text{Al}(n,p)$ reaction with fast neutrons and introduction of hydrogen from the aqueous reactor environment; (3) the production of silicon in supersaturated solid solution by thermal neutron reactions with aluminum, and the subsequent precipitation of silicon-rich precipitates; (4) the production of displacement damage and subsequent formation of high concentrations of voids in the material.

Cyclotron-injected uniform concentrations of hydrogen and helium have been used to discriminate between the effects of these gases and other irradiation effects. Above half the absolute melting point 5 ppm He reduces the short-time tensile ductility of 1100 and 8001 aluminum but not of 6061 and SAP alloys. The creep ductility of 1100 aluminum and high-purity aluminum was significantly lowered at 300°F by 3 ppm He in preliminary tests, but strains at failure still exceeded 30%. Up to 10 ppm of injected hydrogen had no significant effects on the creep properties of these alloys at 300°F.

Specimens of 6061, 1100, and 8001 aluminum alloys have been irradiated in special HFIR capsules, out of contact with the coolant. Irradiation for 3 and 5 cycles in HFIR produces fluences of 2 to 5×10^{21} neutrons/cm² (>0.81 Mev). These alloys exhibit reduced elevated-temperature tensile ductility and increased strength at 300°F and above, similar to the property changes observed in previous tests on ring and sheet

specimens irradiated in contact with the coolant. These results verify that irradiation damage is the source of the problem and have been used as a guide in designing reactor irradiation experiments aimed at developing damage-resistant aluminum cladding alloys.

DENSITY CHANGES IN ALUMINUM ALLOYS AS A RESULT OF IRRADIATION

E. L. Long, Jr.

Immersion densities have been measured on type 1100 aluminum specimens that were sectioned from an ORR N_f tray.³ The aluminum was irradiated at about 45°C and received a maximum fluence of 1.5×10^{22} neutrons/cm² (>1 Mev); a 1.5% decrease in density was measured at the region of maximum fluence. As expected, the measured decrease in density of the aluminum could be correlated with the flux profile along the N_f tray. Density changes were measured for type 1100 aluminum after postirradiation heat treatment between 90 and 620°C. In addition, densities have been determined by immersion on type 8001 aluminum specimens from target hex cans that had reached fluence levels up to 2.7×10^{22} neutrons/cm² (>0.81 Mev).

Specimens from the maximum-flux region of the ORR N_f tray were heat treated in air for 20 hr at various temperatures up to 620°C and air cooled; the results are shown in Fig. 29.3. Maxima in the decreases in density occurred at about 130 and 565°C; a minimum existed at 315°C (about $0.6T_m$). Transmission electron microscopy disclosed polyhedral voids about 800 Å across that had been formed during irradiation; a higher concentration of voids existed adjacent to the grain boundaries. A specimen heat treated at 200°C showed a decrease in void concentration and a coarsening of the silicon precipitate. A specimen heat treated at 315°C disclosed additional coarsening of the precipitate and no evidence of voids. The reversal of the slope of the curve from 315 to 565°C probably results from the formation of gas bubbles within the aluminum.

Density changes were determined on sections of hex cans that had received fluences ranging from 2.8×10^{21} to 2.7×10^{22} neutrons/cm² (>0.81 Mev). Specimens were taken from near maximum and minimum flux regions of several hex cans; the average operating temperatures of these sections were approximately 66 and 58°C respectively. The changes in density with fluence are shown in Fig. 29.4, along with

those of type 1100 aluminum from the ORR N_f tray. The type 1100 aluminum decreased 1.5% in density at a fast-fluence level of 1.5×10^{22} neutrons/cm²; at slightly less than twice that fluence the density of the type 8001 aluminum decreased about 5%. The two curves shown imply that a family of curves may exist for various aluminum alloys, but factors such as operating temperatures and neutron spectra also can influence the changes in density during irradiation.

Fig. 29.3. Density Changes in Specimens from the Maximum Fluence Region of an ORR N_f Tray After Heat Treatment at Various Temperatures for 20 hr. The fluence level of these specimens ranged from 1.4 to 1.5×10^{22} neutrons/cm² (>1 Mev).

Fig. 29.4. Density Changes in Irradiated Type 1100 and Type 8001 Aluminum Alloys from an ORR N_f Tray and HFIR Hex Cans. The type 1100 aluminum was irradiated at 45°C and the 8001 was irradiated at higher average temperatures from 58 to 66°C.

³S. S. Hurt, III, "Experience with Neutron Damage to Beryllium and Aluminum Lattice Components at the ORR," *Trans. Am. Nucl. Soc.* 10 (Supplement), 11-12 (July 1967).