**NEPTUNIUM TARGET ELEMENT
FABRICATION**

C. R. BLOOMSTER

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NEPTUNIUM TARGET ELEMENT
FABRICATION

By

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C. H. Bloomster

ABSTRACT

A fabrication process was developed for preparing neptunium-aluminum alloy target elements for irradiation. Some of the target elements included high radiation level, "quick-turn-around" neptunium.

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NEPTUNIUM TARGET ELEMENT FABRICATION

C. H. Bloomster

INTRODUCTION

Neptunium target elements were required for irradiation in a K reactor by Douglas United Nuclear (DUN). Battelle-Northwest (BNW) was requested to develop and fabricate the target elements.

DUN was conducting a ^{238}Pu production demonstration program by irradiating neptunium. One of the unique features was the concept of quick-turn-around in which neptunium from a previous irradiation is recycled back into a reactor in a minimum period of time. This rapid recycle increases ^{238}Pu production and decreases costs. Fifty days, from reactor discharge to recharge, was set as the initial target for the quick-turn-around demonstration. The objectives of the target fabrication phase of the program were threefold: 1) development of a fabrication process, 2) demonstration of the fabrication process on quick-turn-around neptunium, and 3) fabrication of all other target elements required for the program.

To meet the quick-turn around objective, a process was required which would have a high yield and a short, dependable fabrication cycle. In previous work⁽¹⁾ BNW developed an aluminum-neptunium target element for irradiation in N reactor. The K reactor target elements, although smaller, were similar in design. Because the fabrication process for the N reactor target elements was highly successful, this process was adapted to the fabrication of the K reactor targets.

SUMMARY AND CONCLUSIONS

A casting process (Figure 1) was developed to fabricate neptunium target elements for irradiation in K reactor. Process

FIGURE 1. Flow sheet: Neptunium Target Element Fabrication Process

yields were good: once-through yields were 60%, and the continuous process neptunium yield, with recycle, was estimated at 98%.

Fifty targets were fabricated for a ^{238}Pu production demonstration program. Five of the targets, which were fabricated in two days, contained quick-turn-around neptunium which was discharged from a reactor only forty days previous. Contact radiation levels from the quick-turn-around neptunium were high, approximately 20 R beta plus gamma, including 5 R gamma.

Aluminum-neptunium alloys were prepared by reducing neptunium oxide with aluminum under a cryolite flux. The tubes were machined to specified lengths and diameters, and inserted into aluminum can assemblies. The completed elements were isostatically pressed to bring the can and core into contact. Scrap was recycled from the casting, machining, and assembly steps by remelting or reassembly. Overall, 2.3 Kg of neptunium entered the process; 2.0 Kg neptunium went into the targets, and 0.3 Kg of neptunium scrap remained for recovery at the end of the fabrication.

DISCUSSION

DESIGN CRITERIA

The following design criteria were established by DUN for the target elements (Figure 2).

1. For greater neutron efficiency, the neptunium target material will be tubular to concentrate the neptunium atoms on the periphery of the elements.
2. The target material will be an Al-10 wt% Np alloy.
3. The center of the tube will contain an aluminum mandrel to add mass to the element to prevent chattering or floating in the process tube.
4. The cladding will be aluminum.
5. The finished element will be a solid right cylinder, 1.5 in. diam by 9 in. long.

FIGURE 2. Design of Neptunium Target Element

PROCESS DESCRIPTION

Neptunium oxide was reduced by aluminum in the presence of cryolite to form an Al-Np master alloy. The master alloy was diluted with aluminum, if necessary, to 10 wt% Np and cast into thick wall tubes using graphite molds and mandrels. The graphite mandrels were removed from the castings by induction heating. The castings were inspected and machined to specified length and diameter. Each casting and a full-length aluminum rod were inserted into an aluminum can. The weld zone was wiped free of neptunium contamination, and an aluminum end cap was inserted. The elements were welded shut, helium leak-checked, and radio-graphed. The completely assembled element (Figure 3) was isostatically pressed at 500 psi to eliminate the assembly gaps.

REDUCTION OF NpO_2

Neptunium oxide was reduced by aluminum (in excess) in the presence of cryolite to form an Al-Np alloy. The reaction is:

The neptunium alloys with the excess aluminum, and the Al_2O_3 product was dissolved by the cryolite flux. The reductions were made at 1200 °C. The reaction was judged complete 30 min after disappearance of the green neptunium oxide color from the white cryolite. Reaction times usually ranged between 60 and 90 minutes. After the reduction was complete, the cryolite was skimmed from the melt, and the melt was cast into billets.

Three separate reductions of NpO_2 were made (Table I). High purity aluminum, 99.99%, was used for alloying. The reductions were made in induction melting furnaces housed in an air-atmosphere glove box. The melt charges were calculated to obtain a master alloy of 13 wt% Np in the first reduction, and 10 wt% Np in the last two reductions.

FIGURE 3. Neptunium Target Element and Component Parts

TABLE I. Reduction of NpO_2 with Aluminum

Melt Number	Charge, g				Yield, g
	NpO_2	Np-Al Recycle	Aluminum	Cryolite	Np-Al Alloy
1	554		3,342	1400	3,666
2	278		2,242	800	2,386
3	1784	1046	13,912	3500	16,004

Material balances were calculated for the last five NpO_2 reductions, including the last two reductions from the fabrication of the N reactor targets;⁽¹⁾ Np recovery was 98.8% in alloy form, with 0.7% into the waste cryolite flux, and 0.5% in losses. The last five reductions contained 5.26 Kg Neptunium. The sampling of the waste cryolite was not extensive; further work is required to verify the distribution of neptunium between the cryolite waste and the process losses.

CASTING

The master alloy was remelted and diluted, as necessary, to an Al-10 wt% Np alloy. The Al-Np alloy was cast into 1/4 in. thick wall tubes in a graphite mold around graphite mandrels (Figure 4). The graphite mandrels were tapered to facilitate later removal. To reduce scrap generation in the machining operation the casting was made 0.05 in. oversize on the OD, and the ID was cast to the desired size. The melt was made in an induction melting crucible and poured into a casting ladle, a No. 5 clay-graphite crucible preheated in a resistance furnace. The casting temperature was 735 °C to 740 °C; the mold and mandrels were preheated to 300 °C. A hand-ladling technique was used to produce castings having low porosity, good surfaces, uniform neptunium concentrations, and high yields. Proper casting technique was important. The metal was initially poured rapidly, slowing to a trickle near the end of the pour.

FIGURE 4. *Neptunium Target Casting*

The mold was supported on a copper block to promote directional solidification. The mold was rotated slowly by hand to prevent local overheating of the graphite mandrel. After casting, the mandrel was removed by induction reheating of the casting. The castings were inspected and radiographed to detect unsoundness and segregation.

The only casting defect encountered during fabrication was excessive solidification shrinkage near the inner top of the casting. This was caused by localized overheating at the point where the molten stream impinged on the mandrel. The defect was not serious, for the structural strength requirement of the castings was very low (~500 psi). The casting defect was eliminated by a combination of the proper pouring rate and mold rotation. The casting yield was 84%; the casting rejects were entirely recycled. With further experience, the casting yield should exceed 90%.

MACHINING

The cast tubes were cut to length. Turnings for chemical analysis were taken from each end. The outside diameter was machined to size in one pass by using live centers. No lubricants or cutting fluids were used in machining. The machined pieces were handled carefully with tongs to prevent surface contamination, inserted in plastic bags, and weighed before transfer to the assembly area. The yield from the machining operation was 77% of the cast alloy weight. Machining scrap [chips (19%) and rejects (4%) of the cast alloy weight] was recycled through the casting operation.

ASSEMBLY

Assembly was performed in a fume hood. Each casting and the aluminum rod were carefully inserted into an aluminum can.

Flux detectors, full-length Al-Co wires (bare and Cd clad), were assembled into four elements. Grooves were filed on the outside of two castings for the wires, and the other two wires were located inside the castings by a peening attachment to the aluminum rods. The weld zone was wiped free of contamination, and the end cap was inserted. During the assembly of the first fifteen target elements, considerable time was spent cleaning the weld zone. Small chips were detected clinging to the surface of the machined castings. Thereafter, before assembly, the machined castings were cleaned by soaking in perchlorethylene and dried by the natural air flow through the fume hood. Decontamination of the weld zone seemed easier after the solvent cleaning. Welding (TIG) was performed in a welding box by using direct current, a tungsten electrode, and a helium atmosphere.

The welded assemblies were free from contamination and were handled in the open laboratory. The elements were inspected for marred surfaces, helium leak-checked, and radiographed.

The assembly yield was 92%. The assembly rejects resulted entirely from incomplete weld penetration. The assembly rejects were simply recanned.

The target elements were finished by cold isostatic pressing in water at 500 psi to eliminate assembly gaps. Target element identification, weight, and analytical data are summarized in Table II.

COMPONENTS

Standard aluminum cladding components (99 + % Al) were supplied by DUN. The components were cleaned and etched using conventional techniques. The aluminum can was fully annealed to facilitate the isostatic pressing operation. A plug gage was used to test all aluminum cans prior to assembly.

TABLE II. *Target Element Data*

<u>Element No.</u>	<u>Casting No.</u>	<u>Weight of Machined Casting, g</u>	<u>Np Analysis, %</u>	<u>Np Content</u>
NEP 1 - 1	NPC 1 - B	386.5	10.6	41.0
2	2 - C	383.9	10.6	40.7
3	2 - D	386.3	10.6	40.9
4	1 - D	386.3	10.6	40.9
5	2 - B	388.5	10.6	41.2
6	3 - B	388.9	10.6	41.2
7	3 - A	387.3	10.6	41.1
8	1 - F	389.9	10.6	41.3
9	1 - E	382.1	10.6	40.5
10	2 - E	390.3	10.6	41.4
2 - 1	4 - A	384.1	10.0	38.4
2	B	385.7	10.0	38.6
3	C	384.0	10.0	38.4
4	D	382.8	10.0	38.3
5	E	384.0	10.0	38.4
3 - 1	5 - U	388.2	9.8	38.0
2	S	384.8	9.8	37.7
3	D	389.9	9.8	38.2
4	W	387.4	9.8	38.0
5	U	386.8	9.8	37.9
6	K	383.2	9.8	37.6
7	O	387.3	9.8	38.0
8	BB	386.6	9.8	37.9
9	L	383.7	9.8	37.6
10	B	383.9	9.8	37.6
11	I	387.5	9.8	38.0
12	J	386.8	9.8	37.9
13	AA	388.0	9.8	38.0

TABLE II. (Contd)

<u>Element No.</u>	<u>Casting No.</u>	<u>Weight of Machined Casting, g</u>	<u>Np Analysis, %</u>	<u>Np Content</u>
NEP 3 - 14	R	382.2	9.8	37.5
15	C	383.8	9.8	37.6
16	DD	385.2	9.8	37.7
17	H	385.9	9.8	37.7
18	N	389.7	9.8	38.2
19	E	382.7	9.8	37.5
20	Q	385.2	9.8	37.7
21	Y	388.2	9.8	38.0
22	X	385.9	9.8	37.7
23	M	386.2	9.8	37.8
24	CC	386.9	9.8	37.9
25	Z	387.6	9.8	38.0
26	F	385.7	9.8	37.7
28	A	387.5	9.8	38.0
4 - 1	6 - H	388.4	9.9	38.5
2	C	386.3	9.9	38.2
3	I	389.8	9.9	38.6
4	G	389.2	9.9	38.5
5	A	389.2	9.9	38.5
6	D	389.2	9.9	38.5
7	B	387.4	9.9	38.4
8	E	388.2	9.9	38.4
9	F	390.8	9.9	38.7

RADIATION EXPOSURE

One of the problems associated with quick-turn-around neptunium is the high radiation levels associated with the decay of

^{238}Np , the interim product of ^{237}Np irradiation. Neptunium-238 has a 2.1 day half-life, and beta decays to ^{238}Pu . Neptunium-237, itself, decays by alpha emission to ^{233}Pa . Protactinium-233 emits a 0.31 MeV photon which also produces an appreciable radiation field. Radiation levels from newly separated irradiated neptunium decrease rapidly as the ^{238}Np decays and then increases with the ^{233}Pa buildup. Equilibrium ^{233}Pa concentration is reached in approximately 160 days. Minimum radiation levels occur in approximately 50 days from reactor discharge.

Contact radiation levels from the 300 g batch of quick-turn-around NpO_2 available for target fabrication were ~ 20 R beta plus gamma and 5 R gamma.⁽²⁾ Radiation measurements of the finished target elements are shown in Table III.⁽³⁾

Personnel exposures during the target fabrication were quite high, considering the small quantities of neptunium involved, but only minor amounts of shielding were used. Insofar as practical, the work piece was separated from the working stock and unnecessary contact was avoided. Self-reading dosimeters were used to control personnel exposure. Larger scale fabrication may require installation of heavier shielding (particularly in the machining and assembly operations) and modification of some handling techniques to reduce personnel exposure. Adoption of remote handling techniques, however, should not become necessary.

TABLE III. Corrected Dose Rates from ^{237}Np Target Elements

Units of mrad/hr
Elements contain ~40 g ^{237}Np each

Distance*	Measured on 4/14/67		Measured on 4/26/67		Measured on 5/15/67 [†]	
	Element No. 1-8	Array** of 10 elements	Element No. 4-A	Array** 5 elements	Element No. 3-2	Array** of 10 elements
Contact	1635	2700	3350	5200	1970	4100
2 in.	---	---	310	2090	---	---
3 in.	100	815	---	---	135	1250
4 in.	---	---	210	1075	---	---
6 in.	50	560	130	680	70	640
9 in.	---	---	---	---	38	295
12 in.	20	200	50	250	27	215
15 in.	---	---	---	---	21	165
18 in.	10	100	22	140	17	120
24 in.	6	65	14	80	12	80
36 in.	3	31	8	40	7	36
48 in.	2	20	5	18	5	23

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* Distance was measured from the screen of the CP instrument to the surface of the elements(s).

** In the "array" geometry, the elements were placed side by side at approximately equal distances from the CP screen.

† The readings made on 5/15/67 may be slightly high because of background radiation in the area.

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