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PACIFIC NORTHWEST LABORATORY
DIVISION OF ISOTOPE DEVELOPMENT
PROGRAMS QUARTERLY REPORT:
JULY - SEPTEMBER, 1967

JANUARY 1968

AEC RESEARCH &
DEVELOPMENT REPORT

DA/D Labs 60009
for A.P. Lyden

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DIVISION OF ISOTOPE DEVELOPMENT PROGRAMS
QUARTERLY REPORT: JULY - SEPTEMBER, 1967

Edited by R. L. Moore
Applied Chemistry Section
Chemistry Department

January, 1968

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

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QUARTERLY REPORT: JULY - SEPTEMBER, 1967

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INTRODUCTION

This is the fourth report of a series initiated in 1966 to provide potential users with detailed and timely information on the results of the promethium isotopic fuels program and other radioisotope research being conducted at the Pacific Northwest Laboratory by Battelle for the U.S. Atomic Energy Commission. The previous reports of the series are BNWL-385, BNWL-466, and BNWL-570.*

The adopted editorial policy is to cover only work having either been recently completed or having reached some reportable milestone rather than to report each quarter on all investigations in progress (as is done in the Monthly Record Reports on this program). It is felt that this will provide a report of maximum utility to users and that the reports will have continuing reference value. Although this policy will be continued, the scope and title of the reports are being changed. At the request of the sponsors, work performed for the Division of Isotope Development (DID) and for the Division of Space Nuclear Systems (SNS) will no longer be reported together. The DID reports will continue under the present editorship while those for SNS will be authored by Mr. Kirk Drumheller, Manager, Materials Development Section.

The first quarterly report (BNWL-385) was devoted primarily to the chemistry of promethium; the second report, to fabrication technology of promethium and the fueling of the Goddard Microthruster; and the third covered the first

* Available from the AEC Division of Technical Information Extension, Oak Ridge, Tennessee.

phase of the AMSA heat source program (plus other selected topics). This report completes the story of the fabrication and certification of the AMSA heat source.

SUMMARY

Fabrication of the 60 W AMSA heat source has been successfully completed. Problems encountered in the fabrication of the ca. 240 g Pm_2O_3 pellet are described in some detail.

AMSA Pm_2O_3 HEAT SOURCE

The AMSA heat source is a 60 thermal watt promethium heater designed to serve a temperature control function in an experimental guidance system proposed for use on the Advanced Manned Strategic Aircraft (AMSA). Initial work on this project was described in the last Quarterly Progress Report.⁽¹⁾ Fabrication of the heat source has now been completed, and details are presented in the following paragraphs.

IMPACT TESTS - N. C. Davis

Final impact testing of Phase II AMSA simulated heat source capsules was accomplished at Sandia with the impaction tests of BNW capsules No. 39 and 40. The capsules were impacted at 675 ft/sec velocity at the critical 45° impact attitude and a temperature of 300 °F. The target was 6000 psi (compressive strength) concrete. Unlike those from the previous granite impactions, these capsules exhibited very minor deformation. Capsule No. 39, typical of those impacted, is shown in Figure 1 after impaction. Capsule No. 39 was sectioned and compared to Capsule No. 33, which was previously impacted at 392 ft/sec against concrete. No discernible difference exists (Figure 2).

PELLET FABRICATION - N. C. Davis, D. W. Brite

Cold pressing and sintering of the Pm_2O_3 pellet for the AMSA capsule were carried out in a lead-shielded glovebox modified with an extension which fitted between the platens of a 200 ton press. A stainless steel bellows flanged to the bottom of the extension made it possible to apply pressure to a die inside while maintaining complete containment of the Pm_2O_3 within the glovebox and its extension. The modified glovebox and press are shown in Figure 3.

1. Pacific Northwest Laboratory Radioisotope Technology Programs Quarterly Report, April - June 1967, BNWL-570, pp. 26-45. Pacific Northwest Laboratory, Richland, Washington.

Neg 0672736-4

FIGURE 1. *Capsule No. 39*

BNW Capsule No. 33

*Impact Velocity, 392 ft/sec
Attitude, 45° - Temp., 285 °F
Target, Concrete*

BNW Capsule No. 39

*Impact Velocity, 678 ft/sec
Attitude, 45° - Temp., 300 °F
Target, Concrete*

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FIGURE 2. *Sectioned Capsule Comparison*

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FIGURE 3. Modified Glovebox and Press

A platinum resistance furnace was used for sintering the Pm_2O_3 pellet. Dimensions of the furnace are 2 in. ID x 10 in. OD x 11 in. long. The furnace has been operated successfully at 1540 °C using 1500 W (13 A at 117 V).

After the press extension was installed on the shielded glovebox and the furnace was in place, a shake-down run was made for the pressing and sintering processes, using Sm_2O_3 as a stand-in for Pm_2O_3 . Three Sm_2O_3 pellets were pressed and sintered. A special powder container (Figure 4) designed to (1) transport a preweighed quantity of Pm_2O_3 from the powder preparation facility to the pressing glovebox, and (2) load the powder into the die, was tested during these experiments and found to work satisfactorily. Pressing and sintering data for these three pellets are shown in Table I.

TABLE I. Pressing and Sintering Data, Trial Sm_2O_3 AMSA Pellets (246 g pellets, sintered for 12 hr at 1450 °C)

Pellet	Pressure, TSI	Pressed Dimensions, In.		Sintered Dimensions, In.	
		Diameter	Length	Diameter	Length
1	63	1.570	1.477	1.443	1.400
2	58	1.570	1.483	1.448	1.425
3	53	1.570	1.507	1.440	1.440

These three pellets were sintered in a single furnace run, with all pellets located in the central 4.5 in. of the furnace tube. Pellet No. 1, located in the center of the furnace, had a higher density than the other pellets. This observation indicated that the central location in the furnace was a more favored position; in subsequent sintering runs of actual Pm_2O_3 pellets, a Sm_2O_3 pellet was placed on each side of the Pm_2O_3 pellet to simulate the trial run.

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FIGURE 4. *Special Powder Container*

Microcracks were noted in the three Sm_2O_3 pellets in the as-pressed condition where the straight cylinder walls blended into the radiused ends. Observation of these cracks after sintering indicated no enlargement.

Following the trial pressing- and sintering-run, an unsuccessful attempt was made to press and sinter a one-piece 240 g Pm_2O_3 pellet. The upper dome of the pressed pellet broke off--resulting in a two-piece pellet. Evidence of a circular crack and several radial cracks was noted in the large portion when the broken dome was removed. The sintering operation (12 hr to a temperature of 1450 °C, 30 hr at temperature, and 12 hr cooling) increased the severity of these cracks, particularly at the broken end surface (Figure 5). The maximum diameter at the break in the pellet was 1.511 in. The lower portion of the pellet, measuring an average of 1.445 in. in diameter, was less than the 1.480 in. required for assembly into the capsule.

An attempt was made to assemble the badly distorted two-part pellet into a modified tantalum container by squeezing the cracks closed in a three-jaw chuck. Sufficient force to close the cracks could not be developed, and an attempt was made to press a second Pm_2O_3 pellet.

The radial and circumferential cracks observed in the first Pm_2O_3 pellet were probably caused by differential thermal expansion across the radius of the pellet after pressing due to the thermal gradient. During pressing of the second pellet, an attempt was made to overcome this effect by applying approximately 1000 psi pressure to partially consolidate the powder in the die. A wait of 1 hr permitted the powder mass to increase in temperature and attain an equilibrium temperature gradient. Final pressing pressure of 63 TSI was then applied.

Neg 0672642-6CN

FIGURE 5. A 240 g Pm_2O_3 Pellet with Broken
Dome and Circular-Radial Cracks

This procedure was effective in eliminating radial cracks (Figure 6) in the as-pressed pellets but the problem of broken domes remained. Two die modifications were made to eliminate possible sticking in the concave die plugs, and a colloidal graphite die lubricant was substituted for the volatile Sterotex[®] die lubricant previously used. Two die modifications evaluated with Sm_2O_3 powder were (1) 120° pie-shaped segmented die plugs, and (2) split radiused rings with solid die plugs.

The pie-shaped segmented die plugs caused broken domes, which, with light handling, subsequently broke into pie-shaped segments. The split-radiused rings produced single pellets in which faint crack lines could be detected at the intersection of the dome ends with the straight walls. Rough handling separated the dome ends from the pellet body. Good separation with both types of die plugs occurred between the pellet and the die as a result of the graphite coating on the die. The graphite was completely removed during sintering. Surface samples and internal samples of a Sm_2O_3 pellet pressed in a graphite-coated die and sintered for 16 hr at 1450 °C were analyzed for carbon content. Results were 65 ppm and <50 ppm, respectively.

One additional scheme was tried in an attempt to press uncracked Sm_2O_3 pellets. This consisted of using die plugs machined from teflon, thus eliminating the need for die lubricant. However, the teflon was too soft, and Sm_2O_3 particles became imbedded in the teflon surface. All of these Sm_2O_3 experiments were performed by using heating tape wrapped around the die to heat the powder to 125 °C before pressing, thus partially simulating the self-heating effect of the Pm_2O_3 powder.

The third Pm_2O_3 pellet was pressed by using a graphite-coated die with a split-radiused end ring. A further

Neg 0672936-3

FIGURE 6. Pm_2O_3 Pellet with Broken Dome and No Cracks

improvement was incorporated by using a mechanical stop to assure that the pellet was pressed to the desired length-to-diameter ratio. It was noted that the pressing force (95 tons, or 50 TSI) required to reach the mechanical press stop was less than that required for the two previous Pm_2O_3 pellets (120 tons, or 63 TSI).

The pellet produced by the third attempt was fractured into three parts: an upper radiused end cap, a cylindrical body, and a lower radiused end cap. After sintering for 30 hr at 1475 °C and smoothing the laminarly fractured surfaces by sanding, the three pieces could be assembled into the AMSA inner tantalum cladding. The three-piece pellet assembly prior to loading into the capsule is shown in Figure 7.

Weight of Pm_2O_3 loaded in the assembly was 227 g. Process design parameters were based on the use of 235 g of Pm_2O_3 , corresponding to 65 W thirty days after purification of the promethium. Heat output of the 65 W source was to be reduced to 62.4 W on October 1, 1967, due to aging of the promethium. The use of the three-piece pellet, which suffered a slight weight loss due to chipping of the fracture surfaces, caused a further reduction of 2.1 W in heat output of the capsule.*

CAPSULE ASSEMBLY - N. C. Davis, R. F. Boolean

As expected, only the inner clad assembly offered significant problems in contamination control. After decontamination was effected and the first weld was made, the capsule was nonsmearable for the remainder of the assembly operations. Electron beam welding of the inner tantalum capsule and the outer L-605 alloy capsule was

* *Calorimetry of the final AMSA capsule (see later section) determined that it had a heat output of 60.32 ± 0.07 W as of October 1, 1967.*

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FIGURE 7. *Three-Piece Pellet Prior to Loading*

accomplished by using the Carl-Zeiss welder shown in Figure 8. Figures 9 and 10 show the capsule following welding of the inner and outer clads, respectively. The final capsule, with some handling tools, is shown in Figure 11.

The quality control procedures used in the fabrication are shown in Table II.

RADIATION MAPPING OF AMSA SOURCE - F. P. Roberts

The dose rate of the encapsulated AMSA source was measured at 12 in. from its center with a carefully calibrated C.P. dose rate meter. The source was placed on a turntable which enabled us to make measurements at various angles and attitudes. The measured dose rate was 70 ± 2 mR/hr at all points, in good agreement with calculation. Dose rates at greater distances may be estimated adequately from the inverse square relation. Thus, the dose rate at 1 m would be ca. 6 mR/hr, etc.

CALORIMETRY OF AMSA SOURCE - U. L. Upson

The AMSA capsule was measured for thermal output for a 48 hr period ending at noon, Sunday October 8. The power output of the capsule, corrected to 1200 October 1, was 60.32 ± 0.07 W at the 95% confidence level. The stated precision includes the 4-sigma deviation in the calorimeter calibration ($\pm 0.10\%$), but does not include possible bias error in the calibration. The half-life assumed for these calculations, 957 days (2.62 yr), was roughly confirmed (± 102 days) by these measurements.

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FIGURE 8. Carl-Zeiss Welder

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FIGURE 9.

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FIGURE 10.

FIGURE 11.

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TABLE II. Quality Assurance System

Quality Control and Inspection of Pm_2O_3 AMSA Radioisotope Heat Source and Components				
Procedure	Location of Step in Process Flow	Type of Test	Reason for Test or Inspection	Design Specs or Limits
Istopic Purity	After Pm_2O_3 purification	Radiochemical	To verify isotopic purity	Less than 0.25 ppm ^{146}Pm ; curie ratio of $^{148}\text{Pm}/^{146}\text{Pm}$ less than 0.1 ^{152}Eu , ^{144}Ce none detectable
Analysis of $^{147}\text{Pm}_2\text{O}_3$	After oxide conversion and heat treatment	Emission spectrography	To verify oxide is within spec limits	Less than 1% other than Sm
Analysis of $^{147}\text{Pm}_2\text{O}_3$	After oxide conversion and calcination	X-ray diffraction analysis	To confirm Pm_2O_3 used is in the monoclinic crystallographic form	Monoclinic form
Pm_2O_3 Pellet Inspection	After cold pressing	Dimension checks, density calculation	To determine that pellets are proper size to obtain L to D ratio needed after sintering	75-80% of theoretical density
Pm_2O_3 Pellet Inspection	After sintering	Dimension checks, density calculation	To verify final density to assembly in cladding and meet wattage requirements	OD 1.450 in. max.; length 1.450 in. max.; dome 0.36 in. R
Ta Inner Cladding Evaluation	Raw material	Impurity analysis	To verify purity of inner cladding	99.9% minimum purity
Ta Inner Cladding Evaluation	After capsule fabrication	Dimension checks and dye penetrant test	To verify dimensions and capsule integrity	All irregularities will be reported, dimensions in accordance with Dwg. H-3-27668. Bleed back from penetrant will require further investigation.
Ta Inner Cladding Evaluation	After pellet loading	Decontamination smear test	To verify that outer surface of cladding is free of smearable Pm_2O_3 , prevent weld contamination	Nonsmearable
Ta Inner Cladding Evaluation Test Weld	After test weld	Helium leak test, ultrasonic weld evaluation, metallography	To verify welding parameters and weld penetration	Leak rate less than 10^{-6} std atm cm^3/sec ; 50% minimum weld penetration as determined by ultrasonic test and confirmed by metallography
Ta Inner Cladding Evaluation	After inner capsule welding	Decontamination, dimensions, helium leak test, ultrasonic, radiation mapping	Pm_2O_3 free; to make outer capsule assembly; insure weld closure; determine weld penetration; determine radiation field	Nonsmearable; to meet Dwg. H-3-27668; leak rate less than 10^{-6} std atm cm^3/sec ; 50% min. weld penetration; less than 30 mR/hr at 1 m
Ta Inner Cladding Evaluation - Post Test Weld	After welding second test weld	Helium leak test, ultrasonic, metallography	To verify welding parameters and weld penetration	Leak rate less than 10^{-6} std atm cm^3/sec ; 50% min. weld penetration as determined by ultrasonic test and confirmed by metallography
L-605 Alloy Outer Cladding Evaluation	Raw material	Chemical impurity analysis	To confirm chemical content of alloy	To be from same heat as stock used for corrosion and physical test
L-605 Alloy Outer Cladding Evaluation	After outer cladding fabrication	Dimension checks and dye penetrant	To verify dimensions and capsule integrity	Dimensions in accordance with Dwg. H-3-27669; any bleed back indication will require additional investigation or rejection
L-605 Alloy Outer Cladding Evaluation Test Weld	After test weld	Helium leak test, ultrasonic weld evaluation, metallography	To verify welding conditions	Leak rate less than 10^{-6} std atm cm^3/sec ; 85% min. weld penetration; weld voids 0.015 in. max. magnitude limited to one per quadrant, confirmed by metallography
L-605 Alloy Outer Cladding Evaluation	After assembly and final welding	Decontamination smear test	To verify outer capsule as welded, free of smearable Pm_2O_3	Nonsmearable
L-605 Alloy Outer Cladding Evaluation Test Weld	After test weld	Helium leak test, ultrasonic weld evaluation, metallography	To verify welding conditions	Leak rate less than 10^{-6} std atm cm^3/sec ; 85% min. weld penetration; detect weld voids, 0.015 in. max. diameter, limited to one per quadrant, confirmed by metallography
Final Inspection	After final finishing	Decontamination, helium leak test, dimensions, ultrasonic flaw test, radiation mapping, gamma spectrometry, calorimetry	Pm_2O_3 free; confirm weld integrity; to fit preconditioner; radiation integrity; radiation spectrum; to establish actual heat output	Nonsmearable; leak rate less than 10^{-6} std atm cm^3/sec ; 85% min. weld penetration, detect weld voids, one per quadrant of 0.015 in. diam permitted; less than 30 mR/hr at 1 m; 65 W on July 1, 1967
		ultrasonic raw test, radiation mapping, gamma spectrometry, calorimetry	tioner; radiation integrity; radiation spectrum; to establish actual heat output	

APPENDIX A

ADDITIONS TO THE PROMETHIUM ISOTOPIC POWER DATA SHEETS -

H. T. Fullam

No new information has appeared since issuance of the last Quarterly Report (BNWL-570).

APPENDIX BERRATA

Figures 1 and 8 of the previous report of this series, BNWL-570, were accidentally interchanged during printing. Holders of copies of BNWL-570 are asked to make this correction.

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