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PACIFIC NORTHWEST LABORATORY
DIVISION OF ISOTOPE
DEVELOPMENT PROGRAMS
QUARTERLY REPORT: MAY - JULY 1969

September 1969

AEC RESEARCH &
DEVELOPMENT REPORT

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BNWL-1177

UC-23, Radioisotope and
Radiation Applications

PACIFIC NORTHWEST LABORATORY
DIVISION OF ISOTOPE DEVELOPMENT PROGRAMS

QUARTERLY REPORT: MAY - JULY 1969

Edited by W. F. Sheely

September 1969

BATTELLE MEMORIAL INSTITUTE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

Printed in the United States of America
Available from
Clearinghouse for Federal Scientific and Technical Information
National Bureau of Standards, U.S. Department of Commerce
Springfield, Virginia 22151
Price: Printed Copy \$3.00; Microfiche \$0.65

DIVISION OF ISOTOPE DEVELOPMENT PROGRAMS

QUARTERLY REPORT: MAY-JULY 1969

ABSTRACTPm₂O₃ AND Sm₂O₃ CHARACTERISTICSPhase Studies

X-ray diffraction studies and differential thermal analyses to define phase relationships and transformation temperatures in the Pm₂O₃ - Sm₂O₃ system have been completed. The phase diagram resulting from these studies shows complete, solid solubility between all of the allotropes of Pm₂O₃ and Sm₂O₃.

Stability Studies

Thermogravimetric studies on Pm₂O₃ in reducing atmospheres have been completed. The results indicate little deviation from stoichiometry under an oxygen potential of -100 kcal/mole O₂.

Property Measurements

The surface tension of liquid Sm₂O₃ was determined at the melting point using the pendant drop technique. The average surface tension obtained from experiments in vacuum and argon is 962 ± 53 dynes/cm. In addition, the liquid density of Sm₂O₃ at the melting point was found to be 5.90 g/cm³.

The heat capacity of Pm₂O₃ was estimated from existing literature data on the heat capacities of other lanthanide sesquioxides. The heat capacities of cubic and monoclinic Pm₂O₃ are tabulated over the temperature intervals 300 to 900 °K and 300 to 1600 °K, respectively.

The vapor pressure of Pm₂O₃ was determined over the temperature interval 2050 to 2250 °C using the Knudsen effusion technique. The vaporization process $\text{Pm}_2\text{O}_3(\text{s}) = 2 \text{ PmO}(\text{g}) + \text{O}(\text{g})$ was assumed in analyzing the data. A second-law treatment gives $\Delta H_s = 266$ kcal for this reaction.

RADIATION CHARACTERISTICS OF PROMETHIUM METAL

The attenuation of radiation from a promethium metal sample by lead and aluminum shielding was measured, and the relative contributions of primary gammas and bremsstrahlung to measured dose rate was determined.

RADIATION CHARACTERISTICS OF FISSION PRODUCT Rh AND Pd

The isotopic compositions in rhodium from samples of spent fuel from the Yankee Reactor and from Hanford reactors dose rate calculations have been made for three cases of hypothetical fission product rhodium application.

No gamma peaks were found in a sample of purified palladium obtained from spent Yankee fuel.

ENCAPSULATED WASTE MANAGEMENT CESIUM AS A RADIATION SOURCE

Evaluation of radiation source users interest in waste management encapsulated cesium has shown that the capsules as presently planned could be incorporated into industrial processes, and that they could be made more attractive by some modification in capsule geometry.

NONDESTRUCTIVE TESTING TECHNIQUES FOR RADIATION SOURCESNDT for Irradiation Sources

Completed studies on NDT techniques for SrSiO_3 and ^{60}Co irradiation sources have shown that high testing rates can be accomplished using ultrasonic techniques which employ multi-element transducers and thermal techniques which use thermal transducers.

Neutron Radiography with ^{252}Cf

An analytical study has been completed of the moderation and detection by foil activation of neutrons from a ^{252}Cf source. The results of this analysis will be used in the design of a

portable neutron source for radiography. A ^{252}Cf source has been obtained and an experimental camera built.

SOURCE FABRICATION TECHNOLOGY

Preliminary experiments are underway to evaluate the sputter-etch process for fabricating alpha sources.

EVALUATION OF ISOTOPE PRODUCTION IN POWER REACTORS

The effect of target density on the cost of producing ^{238}Pu by neutron irradiation of ^{237}Np . The variations of neutron flux and fabrication costs with target density were taken into consideration. The effect of purity on the cost of producing ^{238}Pu and the effect of increasing exposure on ^{238}Pu purity were also evaluated.

RADIATION CHARACTERISTICS OF ISOTOPES

The QAD code has been used to calculate in-phantom dose-rates for an 8.42μ ^{252}Cf needle source and to calculate neutron and photon dose rates from sources simulating a ^{238}Pu source. Considerations which must be applied in mixing isotopes to simulate a ^{238}Pu are discussed.

In-phantom dose-rate measurements with ^{252}Cf and ^{238}Pu sources are in acceptable agreement with the predictions made by the modified QAD code.

^{146}Pm BURNOUT BY NEUTRON IRRADIATION

Two ^{147}Pm targets which originally contained 3.5 ppm of ^{146}Pm have been irradiated in a Savannah River reactor to burn-out the ^{146}Pm and have now been returned to BNW. They will be allowed to cool for another 5 or 6 months before the $^{146}\text{Pm}/^{148\text{m}}\text{Pm}$ ratios will be measured.

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INTRODUCTION

This is the eleventh report of a series initiated in 1966 to provide detailed and timely information on the results of the isotopic fuels program and related research being conducted at the Pacific Northwest Laboratory by Battelle-Northwest for the U.S. Atomic Energy Commission. The previous reports of the series are BNWL-385, 466, 570, 680, 681, 781, 877, 924, 1010 and 1085.

This report covers the 3-month period, May 1969 through July 1969.

Pm₂O₃ AND Sm₂O₃ CHARACTERISTICSPHASE STUDIES

C. E. McNeilly and P. G. Pallmer

Phase studies have been carried out on promethium sesquioxide by X-ray diffraction to determine the phase stability and changes accompanying the replacement of promethium by samarium due to the decay process. After separation of ¹⁴⁷Pm from other fission products and precipitation as an oxalate, X-ray samples were prepared by calcination at various temperatures in air. The cubic C-phase could be obtained at temperatures up to 700 °C. Heating to 700 °C or above results in the retention of the monoclinic B-phase upon cooling to room temperature. Retention of the hexagonal A-phase could not be definitely confirmed because of interference with the B-phase X-ray pattern.

Several attempts were made to quench either the A or H forms of Pm₂O₃ by rapid cooling of a sample block while it underwent differential thermal analysis. Even at apparent cooling rates of about 5000 °C/min, however, the transformation peaks showed up on the DTA curves, indicating a change back to the B form. Only in the case of samples melted in tungsten or rhenium filaments did a slightly different X-ray pattern result with a possible hexagonal indexing of four reflections coincident with the B-phase. Although the precision of assignments was not as good as for the B-phase calculations, a cell volume of 75.80 Å³ based on a = 3.81 Å and c = 6.03 Å, agrees well with a 6 times larger volume of 455.3 Å³ calculated for the B-phase.

Because the monoclinic B-phase is stable at lower temperatures after it is once formed, it is present over the widest temperature range for heat source applications and was the

subject of the majority of the X-ray diffraction studies. The isomorphous replacement of promethium by samarium in B-phase Pm_2O_3 produced a slight linear reduction in lattice parameters (Figure 2.1), corresponding to the fractional decrease in going from Pm_2O_3 to the Sm_2O_3 parameters. Because of a half-life of 2.6 years for ^{147}Pm , the higher concentrations were prepared by addition of samarium or by coprecipitation prior to calcining,

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FIGURE 2.1. *Lattice Parameters of Solid Solutions of Monoclinic Promethium, Samarium Sesquioxides. Compositions are Related to In-Growth of Samarium After Separation of Promethium on the Upper Scale*

Diffraction patterns were obtained by the Debye-Scherrer method. The effects of the high sample radiation on film background was minimized by use of small samples, high-intensity X-ray tubes, and most important, by placing a $K\beta$ radiation filter between the sample and film. Frequently, nearly coincident reflections could be resolved, such as the (310), $(20\bar{3})$, and (003). The expected position of all possible reflections, excluding systematic extinctions, was computed. This computation indicated the correct assignment of the $(11\bar{2})$ reflection which was confirmed by the occasional observation of the weaker (112) reflection and by the resulting calculation of the angle β in better agreement with adjacent rare earth sesquioxides.

The precision of calculation of the monoclinic parameter was typically ± 0.02 Å for a and c, ± 0.01 Å for b, and 0.02° for β . Since a computer program was used to obtain the best fit, the varying standard deviations reflect the uneven representation of the parameters in the observed reflections. The lattice parameters for pure $^{147}\text{Pm}_2\text{O}_3$ were estimated by graphic extrapolation of the parameters as a function of composition. A significant difference from the parameters expected by comparison to the other rare earth oxides is the smaller value of a at $14.22 \overset{\circ}{\text{Å}}$ as compared to a value of 14.26 intermediate between those for neodymium and samarium. The unit cell volumes for both B-phase and C-phase are larger than the Sm_2O_3 volumes by 40% of the increase between Sm_2O_3 and Nd_2O_3 . The lattice constant values obtained are summarized in Table 2.1.

The solid-state phase transformations and melting points in the Pm_2O_3 - Sm_2O_3 system were examined by differential thermal analysis (DTA). Temperatures were measured for the B $\rightleftharpoons$ A, A $\rightleftharpoons$ H, H $\rightleftharpoons$ X, and X $\rightleftharpoons$ melting on compositions containing 98, 53, 26, 9, and 0 wt% Pm_2O_3 . The results are given in

Table 2.2 and a proposed phase diagram based on these data and the X-ray results shown in Figure 2.2, The phase designation is based on the work of Foëx⁽³⁾ on other lanthanide oxide systems.

TABLE 2.1. *Lattice Parameters of Nd, Pm, and Sm Sesquioxides*

Parameter	B-Phase					C-Phase	
	a (Å)	b (Å)	c (Å)	β (°)	V (Å ³)	a (Å)	V (Å ³)
Nd ₂ O ₃ ⁽¹⁾	14.35	3.666	8.99	100.34	465.3	11.08	1360
Pm ₂ O ₃	14.22	3.65	8.91	100.1	455.3	10.99	1327
Sm ₂ O ₃ ⁽²⁾	14.18	3.63	8.85	99.96	448.8	10.93	1306
% Change in 2.6 yr for 147Pm ₂ O ₃	-0.14	-0.28	-0.34	-0.07	-0.7	-0.27	-0.8

TABLE 2.2. *Temperatures for B $\rightleftharpoons$ A, A $\rightleftharpoons$ H, H $\rightleftharpoons$ X, and X $\rightleftharpoons$ Melting on Compositions Containing 98, 53, 26, 9, and 0 wt% Pm₂O₃.*

Reaction	(Pm _{0.98} Sm _{0.02}) ₂ O ₃	(Pm _{0.53} Sm _{0.47}) ₂ O ₃	(Pm _{0.26} Sm _{0.74}) ₂ O ₃	(Pm _{0.09} Sm _{0.91}) ₂ O ₃	Sm ₂ O ₃
B $\rightleftharpoons$ A	1740	1740	1780 ^(c)	1880	1930
A $\rightleftharpoons$ H	2135 ^(a)	2115	2065 ^(d)	2112	2160
H $\rightleftharpoons$ X	2225	2250 ^(b)	2245	2252	
Melting Point	2320	2360	2330	2325	2365

a. Originally peak at 1930 which changed to 2135 °C after a few heating - cooling cycles.

b. Originally peak at 2035 which disappeared after a few heating - cooling cycles.

c. Originally peak at 1825 which changed to 1780 °C after a few heating - cooling cycles.

d. Originally peak at 2140 which changed to 2065 °C after a few heating - cooling cycles.

FIGURE 2.2. The Pm_2O_3 - Sm_2O_3 Phase Diagram. (The two phase regions are extremely narrow and are represented by a single line)

There is no ready explanation for the appearance of some unexpected peaks as noted in Table 2.2 and their subsequent disappearance or shifting to other temperatures. They may have been due to selective evaporation of impurities or one of the components, or reaction with the tungsten crucible.

The phase diagram (Figure 2.2) shows the system to be one of complete solid solubility with the second order, irreversible $C \rightarrow B$ transformation occurring over the temperature range of about 650 to 900 °C. The higher temperature transformations are all reversible and occur over extremely narrow temperature ranges, as evidenced by the sharpness of the DTA peaks. The liquidus-solidus region must also be quite narrow and extends only from 2320 °C at Pm_2O_3 to 2350 °C at Sm_2O_3 .

STABILITY STUDIES

C. E. McNeilly

The results from the previously reported stability experiments have been analyzed. A 600 mg sample of Pm_2O_3 , held at 950 °C, was alternately cycled (24-hr periods) in CO/CO_2 and $\text{H}_2/\text{H}_2\text{O}$ atmospheres whose ratios were adjusted to give a constant $\Delta\bar{G}(\text{O}_2) = -100$ kcal/mole O_2 . The results showed a large initial decrease in weight due to release of adsorbed water and decomposition of residual oxalate. Cycling between the two types of atmospheres did not result in any significant weight changes. This result indicates that the stoichiometry remained essentially constant during the experiments. It also indicates that anomalous weight changes noted in earlier work are not due to adsorption of OH- radicals from the $\text{H}_2/\text{H}_2\text{O}$ atmosphere under these conditions.

If a particle size of 20 μ is assumed, the Pm_2O_3 powder would have a surface area of about 4 μ^2/g or about 2.4 μ^2 for the sample used. This amount of surface would accommodate $\sim 11 \times 10^{-6}$ g of OH- per monolayer which is about two orders of

magnitude less than the weight changes observed in changing from a CO/CO₂ to an H₂/H₂O atmosphere. This also supports the conclusion that OH⁻ is not surface sorbed on Pm₂O₃.

PROPERTY MEASUREMENTS

Surface Tension of Liquid Sm₂O₃

J. J. Rasmussen

The value of the surface tension of liquid Pm₂O₃ is required in order to make an adequate safety analysis for the use of this isotopic fuel in space missions. Sm₂O₃ was used as a stand-in for Pm₂O₃ since a radiographic furnace facility for working with radioactive materials was not available. A previous quarterly report⁽⁴⁾ describes the technique used to determine liquid surface tension from the shape of pendant drops radiographed in closed refractory metal capsules.

To calculate a liquid surface tension value, one must know the density of the liquid. Because the density of liquid Sm₂O₃ was not available in literature, it was necessary to measure a value. To accomplish this, a molten pendant drop was radiographed. The system was then allowed to cool to room temperature, and the drop radiographed again. The drop was removed from the end of the Sm₂O₃ rod, and a liquid displacement technique was used to determine the density. The maximum diameter of the molten and solidified drops was measured from the radiographs. Assuming the drop volume and the density to be proportional to the cube of the drop radius, the liquid density was determined from the relation

$$\rho_{\ell} = \rho_s \frac{r_s^3}{r_{\ell}^3} \quad , \quad (1)$$

where ρ_{ℓ} , ρ_s and r_{ℓ} , r_s are the densities and radii of the liquid and the solid drops, respectively. This technique leads

to a value for the density of liquid Sm_2O_3 at the melting point of 5.90 g/cm^3 . Using this technique, we found the density of liquid Al_2O_3 to be 3.38 g/cm^3 compared with a measured value of $2.97 - 3.05$.⁽⁵⁾ This result indicates that the uncertainty in the value of the liquid density of Sm_2O_3 limits the accuracy of the surface tension value calculated. To remove this uncertainty the liquid density should be measured directly.

Sm_2O_3 rods were prepared by hydrostatic pressing without any binder. They were sintered at 1730°C for 4 hr in air to 83% of theoretical density. The rods were machined to size and suspended in a molybdenum capsule which was electron-beam welded in either vacuum or argon. The capsule walls were thinned slightly in a taper from each end to the center. This allowed a temperature gradient to form when the capsules were brought to temperature by resistance heating. With the temperature gradient imposed, several drops of Sm_2O_3 could be formed on each rod. The surface tension value of the liquid Sm_2O_3 at the melting temperature calculated from enlargements of five different pendant drops in vacuum was $956 \pm 42 \text{ dynes/cm}$. The surface tension value calculated from enlargements of two different pendant drops in argon was $985 \pm 89 \text{ dynes/cm}$. The overall average from both runs is $962 \pm 53 \text{ dynes/cm}$.

The precision of these results is reasonable but the accuracy is limited by the uncertainty in the liquid density. An attempt was also made to determine the surface tension by a drop-weight technique, but the results were questionable because of the uncertainty in determining the weight of each drop.

Heat Capacity of $^{147}\text{Pm}_2\text{O}_3$

R. L. Gibby

During this quarter an extensive analysis was completed of the heat capacities of the rare earth sesquioxides. This

analysis confirmed our previous conclusions that the heat capacity of Pm_2O_3 should lie between the heat capacities of Nd_2O_3 and Sm_2O_3 , at least at temperatures less than approximately 1100 °K. As an approximation for the heat capacity of Pm_2O_3 , the data for either of these oxides should be good to within $\pm 5\%$, assuming of course that the accuracy of these data are better than $\pm 1\%$. Values were determined, however, for the heat capacity of both cubic and monoclinic Pm_2O_3 based on the systematic trends observed for heat capacity in the rare earth oxides.

The thermodynamic properties of the rare earths and their oxides have been assessed and tabulated by Holly, Huber, and Baker.⁽⁶⁾ Their heat capacity data for the cubic and hexagonal sesquioxides are plotted in Figure 2.3 at constant temperatures as a function of the atomic number of the rare earth cation. In the temperature interval of these data, the sesquioxides of lanthanum through neodymium crystallize only in the hexagonal form, while all those above neodymium crystallize in the cubic structure. At approximately 900 °K Pm_2O_3 begins to transform irreversibly to the monoclinic structure while Sm_2O_3 through Dy_2O_3 transform to the monoclinic structure at increasingly higher temperatures. The heat capacity data for hexagonal and monoclinic sesquioxides are shown in Figure 2.4 as a function of atomic number.

The oxides of the end members of the series, lanthanum and lutetium, have $^1\text{S}_0$ ground states. Consequently, their heat capacities are due entirely to lattice vibrations.⁽⁷⁾ Although La_2O_3 and Lu_2O_3 exist in different crystalline forms (hexagonal and cubic, respectively), their heat capacities are not too different, particularly above 400 °K. Thus, the dashed lines connecting their heat capacities in Figure 2.3 are assumed to correspond to the lattice heat capacities of the other rare earth sesquioxides. The heat capacity in excess

of the lattice heat capacity is due primarily to electronic transitions.⁽⁷⁾ The heat capacity of cubic and monoclinic Gd_2O_3 primarily appears to be due to lattice vibrations above 500 °K. Moreover, it is interesting to note in Figure 2.3 that the variation of the electronic heat capacity with atomic number is somewhat symmetrical about Gd_2O_3 (which lies in the middle of the rare earth series), although the electronic contribution to heat capacity decreases at high temperatures near the end of the series as the 4f shell is completed.

FIGURE 2.3. *The Heat Capacities of the Hexagonal and Cubic Rare Earth Sesquioxide as a Function of the Atomic Number of the Cation.*

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FIGURE 2.4. *The Heat Capacities of the Hexagonal and Monoclinic Rare Earth Sesquioxides as a Function of the Atomic Number of the Cation.*

The observed systematic variation of heat capacity with atomic number indicates that the heat capacity of cubic Pm_2O_3 should lie about midway between the data reported for Nd_2O_3 and cubic Sm_2O_3 . These values, which have been taken from Figure 2.3 are tabulated in Table 2.3 and plotted in Figure 2.5. In addition, Figure 2.5 shows these values along with data for the neighboring sesquioxides, as well as Gd_2O_3 which has essentially no electronic heat capacity. The heat capacity for monoclinic Pm_2O_3 was determined in the

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FIGURE 2.6. *The Heat Capacity of Monoclinic Pm₂O₃ Estimated from the Data Shown for Hexagonal Nd₂O₃ and Monoclinic Sm₂O₃*

Vapor Pressure of Pm_2O_3

C. A. Hinman

An important property in the high temperature application of isotopic heat source materials is the vaporization rate. Both from design and safety aspects, one must be able to predict mass redistribution within a closed system and mass loss in the case of a system failure.

The standard Knudsen effusion technique was used to measure the rate of effusion as a function of temperature for promethium sesquioxide. The vapor pressure is calculated from this data by using the equation

$$P_i = \frac{m_i}{44.33 \text{ at}} \sqrt{\frac{T}{M_i}} \quad , \quad (2)$$

where

P_i = vapor pressure (atmospheres) of the i th vaporizing species

m_i = mass of the i th species which has effused through the effusion hole (g)

a = area of the effusion hole (cm^2) corrected for thermal expansion

t = time during which the effusion took place (sec)

T = temperature of the effusion cell ($^{\circ}\text{K}$)

M_i = molecular weight of the i th species

The quantity m_i/at is called the rate of effusion and is given in $\text{g}/\text{cm}^2\text{-sec}$.

Rhenium was chosen as the cell material and the Knudsen cell machined from a polycrystalline 1.27 cm diameter Re rod. The effusion hole was prepared as a knife edge in order to eliminate the necessity for a Clausing correction. Only one cell was used in the present work so that the effect of hole diameter upon the rate of effusion was not investigated. The

area of the effusion hole was determined by microphotography at a known magnification and calculation of the average diameter. The calculated area was then corrected for thermal expansion using $\Delta l/l_0 = 9.79 \times 10^{-5} + 5.89 \times 10^{-8}T + 8.57 \times 10^{-10}T^2$ for the expansion of rhenium.⁽⁸⁾ The weight loss due to vaporization of the Re from the empty cell was measured at 2280 °C. It was found that the experimental data agreed extremely well with that calculated from published vapor pressure data for Re.⁽⁹⁾ Therefore, no other empty cell weight losses were measured, and the published data were used to correct the effusion data.

The cell was supported on a three-point contact tungsten tripod which was in turn supported on the end of a tungsten rod. The cell was positioned within a water-cooled copper current concentrator used in conjunction with an external induction coil. The vacuum enclosure was made from a double-walled, water-cooled pyrex column and was sealed on the top with a water-cooled, stainless steel lid with a shutter and sight glass for temperature measurement. The bottom of the envelope was mated to the pumping system flange. The vacuum pumping system consisted of a standard 4-in. oil diffusion pump and liquid nitrogen-cooled baffle. Pressures in the 10^{-6} to 10^{-7} Torr range were routinely obtainable.

A 450 kHz induction generator was used as the power supply. The temperature was monitored using a micro-optical brightness pyrometer which was sighted through a sight glass mounted in the glove box wall to a prism located on top of the vacuum enclosure window. The temperature was read through the effusion hole into the cell where black-body conditions existed. The micro-optical pyrometer was calibrated using an NBS tungsten strip lamp while sighting through both glass windows and the glass prism. This temperature calibration extended only to 1880 °C and was extrapolated beyond this

limit into the 2000 °C range. The accuracy of the temperature measurement in the cell is estimated to be ± 8 °C. During the effusion experiments, the temperature was read at 1 to 5 minute intervals. At the higher temperatures some fogging of the sight glass was observed even though the window shutter was opened only during the actual temperature measurement. This may account for some inaccuracy in temperature measurement and control. In addition to the fogging, it appeared that some radiation-induced coloring of the sight glass may have occurred due to the β - γ emission of the Pm-147. This effect was barely noticeable to the eye.

The effusion cell temperature was controlled manually by the operator. It was found that with practice the operating temperature could be obtained within approximately 1 min from room temperature. Both the heating and cooling rates were sufficiently fast so that no correction was applied to the data for losses incurred during these periods. The precision of the temperature control during a typical 30-min run at about 2200 °C was ± 3 °C. The run temperature was modified taking the activation enthalpy into account to obtain an effective run temperature due to the fluctuations. The effective temperature is given by the relationship

$$T_{\text{eff}} = - \frac{\Delta H_s}{3R} / \ln \left[\frac{1}{t_T} \sum_i e^{-\frac{\Delta H_s}{3RT_i} \Delta t_i} \right] , \quad (3)$$

where

- $\frac{\Delta H_s}{3}$ = one-third of the heat of sublimation for Pm_2O_3 as determined from a plot of $\frac{m}{at} \sqrt{T}$ versus $1/T$
- t_T = the total time for the effusion experiment
- T_i = the measured temperature during the time interval Δt_i where Δt_i varied from 1 to 5 minutes.

The experimental apparatus and technique were tested by measuring the rate of effusion for Sm_2O_3 at temperatures of 2285 and 2096 °C. First, the empty cell was outgassed at ~1000 °C in vacuum and then weighed on an analytical balance to an accuracy of ± 0.025 mg (the balance has an ultimate sensitivity of ± 0.005 mg but vibrations within the glove box prevented the attainment of this accuracy). About 230 mg of Sm_2O_3 (Trona Chemical Company, Code 82392) was loaded into the effusion cell. The cell plus the oxide powder was outgassed at ~1200 °C for 10 min and a starting weight obtained. The system was then heated to the desired temperature for the effusion experiment and held there for a definite period of time (~30 min), quenched, and the cell weighed. Several weight loss experiments were run at the higher temperatures in order to ascertain that a constant rate of weight loss had been attained. Only one experimental point was measured at the lower temperature.

Upon completion of the Sm_2O_3 runs, the effusion hole was rephotographed. The cell was then thoroughly cleaned and reheated to about 2300 °C for 15 min to vaporize the remaining oxide from the cell.

Approximately 150 mg of recently purified air calcined Pm_2O_3 was then loaded into the cleaned cell. The same general experimental technique as cited above the Sm_2O_3 was followed to obtain the effusion rates for Pm_2O_3 . Effusion rates were obtained at 50 °C intervals from 2050 to 2300 °C. It proved necessary to recharge the cell once during the run series with new material.

The rate of effusion for Sm_2O_3 was measured at 2285 and 2096 °C. It was found that the oxide powder required "conditioning" before repeatable values of weight losses could be obtained. At 2285 °C the weight losses were reproducible after one 10-min run. The Sm_2O_3 exhibited a

color change after the first 10-min run at 2285 °C changing from light yellow to a medium to dark gray. This effect was noted in previous work⁽¹⁰⁾ and was taken as an indication of a loss of a slight amount of oxygen. This slight change in stoichiometry may be associated with the "conditioning" effect mentioned above. The rate of effusion of the Sm_2O_3 at 2285 °C was found to be 0.622 $\text{mg}/\text{cm}^2\text{-sec}$ and at 2096 °C a value of 0.0985 $\text{mg}/\text{cm}^2\text{-sec}$ was obtained. A second-law technique was used to determine the heat of sublimation from these two data points. The assumption is made that:

$$P = A e^{-\frac{\Delta H_s}{RT}}, \quad (4)$$

where

P = vapor pressure

A = pre-exponential constant

ΔH_s = heat of sublimation

T = absolute temperature

The pressure is obtained from equation (2), although the vaporizing species and their relative compositions must be known to calculate the vapor pressures. It has been found by other experimenters⁽¹⁰⁾ that the vapor species in equilibrium with $\text{Sm}_2\text{O}_3(s)$ are SmO , Sm , and O and that the pressure ratio of $\text{SmO}/\text{Sm} \approx 16$. The accuracy of this value has not been determined. Therefore, it is not possible to calculate the partial pressures. For comparison purposes, the reaction

can be assumed, and the heat of sublimation can be calculated for this process. The value for ΔH_s obtained by this method is 360 kcal/mole of Sm_2O_3 in the temperature range 2100 to 2278 °C. Since the lower temperature point was determined from only one effusion experiment, it is not certain how reliable this number

may be. A second-law ΔH_S calculated from published rate-of-effusion data for $\text{Sm}_2\text{O}_3^{(10)}$ is 423 kcal/mole of Sm_2O_3 , and the third-law value is 443 kcal/mole at 0 °K. The effusion data for the Sm_2O_3 runs are shown in Table 2.5 and Figure 2.7.

TABLE 2.5. *Rates of Effusion of Pm_2O_3 Obtained at Temperatures of 2050, 2100, 2150, 2200, and 2250 °C.*

Temperature, °C	Rate of Effusion, mg/cm ² -sec	Vapor Pressure, atm*	
<u>Sm_2O_3</u>		<u>SmO</u>	<u>O</u>
2095	0.0985	8.00×10^{-6}	1.24×10^{-6}
2285	0.622	5.25×10^{-5}	8.14×10^{-6}
<u>Pm_2O_3</u>		<u>PmO</u>	<u>O</u>
2050	0.0516	4.19×10^{-6}	6.56×10^{-7}
2100	0.0932	7.65×10^{-6}	1.20×10^{-6}
2150	0.144	1.19×10^{-5}	1.87×10^{-6}
2200	0.202	1.69×10^{-5}	2.65×10^{-6}
2250	0.301	2.55×10^{-5}	3.99×10^{-6}

* Based upon the reaction $\text{M}_2\text{O}_3 \rightarrow 2 \text{MO}_{(g)} + \text{O}_{(g)}$ (see text).

The Pm_2O_3 powder used was purified 50 days before the experimental effusion studies were started which allowed approximately 4% of the decay product samarium to build up. Rates of effusion of Pm_2O_3 were obtained at temperatures of 2050, 2100, 2150, 2200, and 2250 °C. These data are tabulated in Table 2.5. At 2250 °C, there was evidence of a slight amount of effusion hole clogging after a number of runs had been made. The data obtained at this temperature indicated the effects of this clogging to be negligible. However, two effusion runs were attempted at 2300 °C and a gross amount of clogging occurred. The effusion data at this temperature were far out of line with data obtained at lower temperatures and were not used in the results.

FIGURE 2.7. Vapor Pressures of Sm_2O_3 and Pm_2O_3 Based Upon the Reaction $\text{M}_2\text{O}_3(\text{s}) \rightarrow 2 \text{MO}(\text{g}) + \text{O}(\text{g})$ (See text for discussion)

The Pm_2O_3 , as was indicated by the experiments with Sm_2O_3 , required a high temperature "conditioning" before repeatable effusion data could be obtained. At 2150 °C it took 2 hr of effusion before the rate-of-effusion stabilized, with the initial rates being lower than the final steady-state value. During the course of the experiments the Pm_2O_3 changed from the normal light purple to a dark purple and finally to a brownish-black color. In the following analysis it is assumed that the deviation from stoichiometry, as indicated by the color change, is insignificant.

The calculated second-law value for the heat of sublimation, assuming the vaporization reaction

is 266 kcal/mole of Pm_2O_3 . The vapor pressures due to this reaction are shown in Figure 2.7.

As mentioned earlier, a mass spectrometric study of both Sm_2O_3 and Nd_2O_3 ⁽¹⁰⁾ has shown that the gaseous monoxide of samarium decomposes to a certain extent to the metal vapor and oxygen gas. However, the NdO gas decomposes only to a very small extent since the ratio of NdO/Nd was found to be greater than 100. Since promethium lies between these two elements, it is impossible to predict whether or not the PmO gas decomposes to the elemental forms. Additional studies would be necessary to determine this.

In the referenced work on vapor pressures of the rare earth oxides⁽¹⁰⁾ tungsten effusion cells were used. The authors stated that an additional weight loss amounting to 5-10% of the observed weight losses were due to a reaction between the tungsten cell and the rare earth oxide. They were not able to correct their data for this effect because the mechanism of the reaction was unknown. The authors further stated the inclusion of this tungsten loss in their data would

introduce a change in their calculated values for the heat of the reaction of less than 1 kcal/mole. In the melting point studies carried out in this laboratory⁽¹¹⁾ it was found that there was a severe reaction between a tungsten filament and molten Pm_2O_3 . However, metallographic examination of a rhenium filament upon which Pm_2O_3 had been melted showed apparent compatibility between the liquid Pm_2O_3 and rhenium. The question of compatibility between the oxide and the cell material and its effect upon the data obtained requires more study.

A comparison of the rate-of-effusion data obtained in this work for Sm_2O_3 and Pm_2O_3 with published data for Sm_2O_3 ⁽¹⁰⁾ (and other rare earth oxides, excluding Pm_2O_3) indicates a significant difference. The second-law heats of sublimation for Sm_2O_3 show a difference of 63 kcal (360 versus 423 kcal/mole). Thermodynamic data for Pm_2O_3 are not available for a third-law treatment and a interpolation of the high temperature free-energy functions for Sm_2O_3 and Nd_2O_3 does not yield consistent results for a third-law treatment of our Pm_2O_3 data. This suggests that more detailed work is needed to obtain more refined thermodynamic data for the vaporization of Pm_2O_3 .

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RADIATION CHARACTERISTICS OF PROMETHIUM METAL FISSION PRODUCTS

H. H. Van Tuyl

For heat source applications, promethium is encapsulated in a material chosen partly because of its effectiveness in shielding out bremsstrahlung radiation. Thus, shielding for heat sources is primarily concerned with higher energy bremsstrahlung (above 0.1 MeV) and gamma radiation. Promethium also shows promise as a radiation source, particularly as metallic promethium. For this application, lower atomic number shielding would be used so that maximum gamma energy would be transmitted.

Dose rate measurements were made on a promethium metal sample embedded in a stainless steel block. The promethium was 0.122 in. thick by 0.436 in. diam, and weighed about 2 g. Measurements were made with a CP dose rate meter with an effective chamber distance of 6 in. from the sample. The sample was enclosed in a polyvinyl chloride plastic bag 12 mils thick and dose rate measurements were made through a series of aluminum and lead absorbers. The contribution of electrons to the dose rate was evaluated by making a series of measurements with two of the lead absorbers along with varying thicknesses of aluminum either above or below the lead absorber.

Results with pure absorbers are shown in Figure 3.1 as dose rate versus weight of absorber. With no absorber, the photon energy is mostly about 0.035 MeV, but the effective photon energy increases as shielding increases. With only 150 mg per cm² lead absorber, the dose rate is mostly from 0.06 to 0.08 MeV photons, while above 2 g/cm² lead much of the dose rate is due to the ¹⁴⁶Pm gammas. An effective gamma radiation source using promethium would require low atomic number shielding, preferably even lower than aluminum. A 30 mil aluminum absorber attenuated the radiation from the promethium metal source used in these experiments by about 40%. Even greater attenuation would occur with a smaller source since the photon energy would be lower due to the reduced self absorption in the promethium.

FIGURE 3.1. Attenuation of ^{147}Pm Radiation by Pure Shields

The component of dose rate due to electrons is shown in Figure 3.2. Attenuation of primary gammas is the same whether aluminum or lead is nearer the detector in a laminated shield. The difference between the curves at constant lead thickness is due to secondary radiations produced in the lead but absorbed in the aluminum. The observed attenuation, about 10% by 10 mg per cm² of aluminum, would correspond to a photon energy of below 15 keV. However, about 90% of the radiation is not attenuated appreciably by the aluminum. Therefore, the decrease in dose rate by covering the lead with about 10 mg/cm² of aluminum must be caused by a very large attenuation of radiation which is present to a small extent. The only photons which might satisfy this requirement are the lead M X-rays, which are expected to be a very minor constituent of the dose rate. Electrons of about 0.1 MeV or less would be absorbed in this manner. The electrons would be formed in lead as the result of the photoelectric process.

FIGURE 3.2. Attenuation of ^{147}Pm Radiation by Laminated Shields

RADIATION CHARACTERISTICS OF FISSION
PRODUCT RHODIUM AND PALLADIUM

F. P. Roberts

In the study to determine the nature and the amounts of radioactivity associated with fission product rhodium and palladium, efforts were concentrated on improving the techniques for measuring the isotopic composition of rhodium and calculating dose rates which will be observed with massive sources of fission product rhodium. Also, a 22 mg sample of fission product palladium was isolated from the irradiated Yankee fuel and its radiation characteristics evaluated.

The isotopic composition of rhodium has been determined on three samples of spent fuel from Yankee Reactor and two samples from Hanford reactors. The results given in Table 4.1 are corrected for decay back to reactor discharge. The values obtained for the Yankee samples are in good agreement, but the large difference between the two Hanford samples is much greater than the expected experimental error.

TABLE 4.1. *composition of Fission Product Rhodium*

No.	Source	Burn-up, MWd/T	Composition $\mu\text{Ci/g Rh}$	
			^{102}Rh	$^{102\text{m}}\text{Rh}$
1	Yankee	33,000	1900 $\pm$ 700	280 $\pm$ 60
2	Yankee	33,000	1900 $\pm$ 700	300 $\pm$ 60
3	Yankee	37,000	2600 $\pm$ 1000	420 $\pm$ 90
4	Hanford - 1 yr old			9.6 $\pm$ 3
5	Hanford - 12 yr old			30 $\pm$ 9

Dose rate calculations based on the data in Table 4.1 were made for the three following cases:

- Case 1: A 1 kg source of rhodium in the form of a right cylinder.
- Case 2: A 1 kg source of 10% Rh - 90% Pt in the form of a right cylinder.
- Case 3: A catalyst bed for a 300 tonne/day nitric acid plant containing 20 kg 10% Rh - 90% Pt.

The calculated dose rates are given in Table 4.2 for both Hanford and Yankee rhodium. Data is presented at various cooling times and for source to detector distances of 1 ft and 1 m.

TABLE 4.2. *Calculated Dose Rates for Massive Sources of Fission Product Rh*

<u>Cooling Time</u>		<u>Dose Rates, mR/hr</u>			
		<u>Yankee</u>		<u>Hanford</u>	
		<u>1 ft.</u>	<u>1 M</u>	<u>1 ft.</u>	<u>1 M</u>
Case 1	At discharge	4500	400	250	22
	10 Y	160	15	11	1
	20 Y	15	1	1	0.1
	30 Y	1	0.1	0.1	0.01
Case 2	At discharge	170	15	10	0.9
	10 Y	15	1.4	0.9	0.08
	20 Y	1	0.1	0.08	0.007
	30 Y	0.1	0.01	0.008	0.0003
Case 3	At discharge	8200	1401	450	77
	10 Y	300	51	20	3
	20 Y	27	5	2	0.3
	30 Y	2.5	0.4	0.2	0.03

A 22 mg source of palladium isolated from the spent Yankee fuel was purified to a high degree and the gamma spectrum was observed. No gamma peaks were detected.

ENCAPSULATED WASTE MANAGEMENT CESIUM
AS A PROCESS RADIATION SOURCE

H. H. Van Tuyl

A status report has been written on use of Hanford Waste Management cesium product as radiation sources. That report concludes that the cesium containers which will be fabricated in the Hanford Waste Management Program will be suitable for use in almost any gamma irradiation process. Container sizes will be large enough and container walls heavy enough to cause appreciable loss of gamma radiation to the source and container. Also, the proposed source size cannot be accommodated in many existing or proposed irradiation facilities without modification of the facility. In spite of these disadvantages, radiation source users are interested in obtaining low-cost cesium sources, even if the source geometry is not ideal. Users would be more satisfied if source geometry could be improved (probably to 1 in. OD $\times$ 18 in. long) at low incremental cost.

NONDESTRUCTIVE TESTING TECHNIQUESNDT FOR IRRADIATION SOURCES

R. W. Steffens

Studies to investigate nondestructive techniques for SrSiO_3 and ^{60}Co irradiation sources were completed and the results are being prepared for publication. Ultrasonic techniques using multi-element transducers and thermal techniques using special thermal transducers were shown to provide high speed testing. These methods are particularly applicable for quick inspection of irradiation sources before expensive and time-consuming fabrication or test procedures are performed on the sources.

Advances in the fabrication of the multi-element ultrasonic transducers permitted construction of transducers with 32 elements per 0.6 in. Figure 6.1A shows the transducer active element before fabrication into the transducer housing, and Figure 6.1B shows the element with the individual transducer wires connected and the transducer housing. Each transducer element is individually monitored through cyclic scanning.

The multi-element transducer has been used as a receiver in a pitch-catch method with a paint brush transmitter as shown in Figure 6.2. The object to be tested is passed between the transducers and loss of signal due to dispersion of the ultrasonic energy by an anomaly is detected by one or more of the multi-element elements. An irradiation source such as a Mark III ^{60}Co source can be inspected in a matter of seconds. Development of electronic circuitry to permit practical use of the multi-element transducer is in progress.

The thermal transducers, shown in Figure 6.3, consist of a metal foil having a temperature sensitive coating and a flexible backing. The outside of the foil is covered with a thin dielectric and placed in dry contact with the surface of the test specimen. Heat is generated in the foil by passing an

a. Active element

b. Transducer Components

FIGURE 6.1. *Multi-element Ultrasonic Transducer Description*

FIGURE 6.2. *Multi-element-Paint Brush Ultrasonic Transducer Pair*

Neg 0692785-8

Flat Thermal Transducer

FIGURE 6.3

electrical current through it. Defects in the test specimen are observed as temperature differences in the foil that result from differences in thermal surface impedance caused by the defects. The temperature differences are observed as differences in the brightness of the temperature sensitive coating. In this way, the test specimen can be "scanned" in a few seconds using either an ordinary camera or a video system. The thermal transducers easily detected clad-core unbonds and core anomalies in nonirradiated ^{60}Co sources.

NEUTRON RADIOGRAPHY WITH ^{252}Cf

J. L. Cason

Neutron radiography has been used to provide otherwise hard-to-obtain radiographic interrogation of objects such as radioactive samples, thick heavy metal sections, light element samples, and composites. Numerous other specific problems have been solved with neutron radiography dealing with nuclear reactor control materials, homogeneity, control material burn up, neutron shielding, rocket propellants, aircraft components, electronic hardware and biological specimens. The rapid growth of applications for neutron radiography has inevitably led to the need for a portable unit to provide neutron radiographic services. Californium-252 is being evaluated for the AEC Division of Isotope Development for use as a portable neutron radiographic source.

The first two phases of work projected for FY 69-70 for developing ^{252}Cf as a transportable neutron source camera or apparatus for neutron radiography have been completed. The third and fourth phases have been initiated (i.e., 'shielding etc.). The first two phases involved mathematical analysis of the use of ^{252}Cf as a source for neutron radiography and obtaining a source. The third phase provides for the experimental evaluation of the analytical results and design work for an experimental apparatus. The fourth phase is concerned with the final design and development of a transportable camera followed by extensive testing, evaluating, and development of imaging techniques.

The analysis was performed in the following manner:

A computer-assisted analysis was performed using codes ANISN, HRG, and THERMOS with a UNIVAC 1108 computer to provide interpretative data regarding bare source spectra, moderated spectra for a given moderator thickness, resultant flux and relative image foil response. Moderators evaluated were Paraffin, C, Be, ZrH_2 , TiH_2 , BeH_2 and H_2O . Image response foils considered were Gd, Dy, Cd, Au, and In.

The ANISN code was used to solve numerically the multi-group form of the neutron transport equation for the spherical geometry case. The general Boltzman neutron transport equation is written in terms of the neutron flux $\Psi(rv\Omega)$ as

$$\vec{\nabla} \cdot (\Omega \Psi) + \sigma \Psi = S(r, v, \Omega), \quad (1)$$

where Ψ/v has dimensions of neutrons/unit volume per unit velocity (v, Ω).

The equation is a statement of the fact that in a time-independent situation, losses from leakage and collisions are equal to gains from source S . The loss rate from collisions is the product of the macroscopic total cross section σ and the flux (nv) on Ψ . In general, S depends upon Ψ through scattering collisions and fission reactions. To program the physics of equation (1) into a useful tool, the coordinate system and average of Ψ are defined, and then equation (1) is derived for a finite cell in phase space, and S is specified in detail.

The results indicate solutions for distinct boundary conditions, thus satisfying the requirements for developing a transportable neutron source. The boundary conditions imposed on the analysis were small source volume, steady-state intensity, optimum moderation and high resolution collimation.

The small source condition was satisfied by a zone calculation in the ANISN code with an associated volume of about $4.2 \times 10^{-3} \text{ cm}^3$. Optimum moderator conditions were dependent upon simultaneously maximizing the thermal flux and image foil activity response. Metal hydrides were selected because of the high hydrogen content per cm^3 as compared to an equal volume of H_2O and Paraffin. Metal hydrides required about 50% less material to moderate the source as compared to H_2O and Paraffin. C and Be were selected as possible alternatives because of their unique moderation characteristics. Collimation was considered by providing a vacuum path to reduce neutron scattering. Efforts were made to provide the data needed to permit an evaluation of the feasibility of neutron spectra tailoring by providing various differential neutron plots.

Analysis of the results determined the feasibility of using ^{252}Cf for neutron radiography by obtaining well collimated thermal fluxes of about 10^6 nv , (based on a 268 μg source). As expected, the metal hydrides offered the greatest efficiency for moderating the neutrons requiring an average of about 3.2 cm of material with a thermal flux of about $1.2 \times 10^6 \text{ nv}$ as well as providing an effective gamma filter.

Water and paraffin were the second best moderators, needing about 5 cm with a thermal flux of $7.0 \times 10^5 \text{ nv}$ and Be and C the least efficient, requiring an average of about 25 cm of material with a flux of about $1 \times 10^5 \text{ nv}$. Results also indicated the practicality of shifting the neutron spectra by varying the moderator material and/or thickness. The shift in the neutron spectra leads to a corresponding shift in image foil response. This selective moderation would allow one to select neutrons [$\phi(E)$ versus E, finite thermal band] most appropriate for a given material thickness material neutron cross section, and image technique.

A summary of moderation data for various materials is plotted in Figure 6.4. All fluxes should be corrected by multiplying the tabular value by the actual source strength, calculated to be 10^9 . Optimum conditions can be determined for selecting the peak thermal flux (Rt boundary flux) versus thickness of moderator.

A typical activity response versus moderator thickness plot for a given moderator for the various image foils is given in Figure 6.2.

Image foil responses are plotted in Figure 6.3 for Gd which gives the optimum thickness of a given moderator to obtain peak response for a given image foil. (In this Figure Gd is chosen as a typical image foil.)

The data plotted in Figures 6.4, 6.5, and 6.6 allows one to select the ideal moderator-image foil combinations, thus optimizing the radiographic results.

A plot of the unmoderated neutron spectra for ^{252}Cf is given in Figure 6.7. Plots of relative flux versus lethargy for a given moderator thickness are given in Figures 6.7 and 6.8.

The value for the thermal flux as determined from the data plotted in Figure 6.4 allows one to calculate the required exposure time for a given object material. Another factor influencing the selection of moderator material and thickness is the total flux (thermal-fast) for a given moderator thickness.

Phase II of the program has been completed by the acquisition of a 268 μg ^{252}Cf source from ORNL for experimental evaluation.

Phase III and IV is proceeding in the following manner:

An experimental neutron camera has been fabricated (Figure 6.9). Experimental evidence has been produced with this unit confirming the analytical results indicating the feasibility of performing neutron radiography with ^{252}Cf (Figure 6.10). Underway at present is a complete, experimental evaluation and design work for a prototype camera.

FIGURE 6.4. *Right Boundary Thermal Flux Versus Moderator Thickness (Function of Moderator Material)*

FIGURE 6.5. *Relative Foil Activity Versus Moderator Thickness (Function of image foil)*

FIGURE 6.6. *Relative Activity Versus Moderator Thickness
(Function of Moderator Material)*

^{252}Cf Neutron Source Spectrum
Pure Spectrum (Unmoderated)

^{252}Cf Neutron Source Spectrum
Moderated by 30cm of Graphite

^{252}Cf Neutron Source Spectrum
Moderated by 4.0cm of H_2O

^{252}Cf Neutron Source Spectrum
Moderated by 5.5cm of Paraffin

FIGURE 6.7. *Plot of Relative Flux Versus Lethargy for Moderator Materials. Graphite, H_2O and Paraffin*

^{252}Cf Neutron Source Spectrum
Moderated by 20cm of Beryllium

^{252}Cf Neutron Source Spectrum
Moderated by 3.5cm of BeH_2

^{252}Cf Neutron Source Spectrum
Moderated by 2.5cm of TiH_2

^{252}Cf Neutron Source Spectrum
Moderated by 4.5cm of ZrH_2

FIGURE 6.8. *Plot of Relative Flux Versus Lethargy for Moderator Materials. Be, BeH_2 , TiH_2 and ZrH_2*

SPECIFICATIONS: 10 in. × 10 in. × 12 in. Long
 Weight Loaded Approximately 20 Pounds
 With Lead Shielding 50 Pounds

FIGURE 6.9. *Neutron Radiography Source Camera*

FIGURE 6.10. *Neutron Radiograph Taken with ^{252}Cf , Moderated with 3.8 cm Paraffin, Thermal Flux $1 \times 10^5 \text{ nv}$, Image Technique: Gd. Direct, AA Film, Exposure Time: 10 min.*

*Objects: A-Borated Polyethylene 0.050 in. holes
End A 1/2 in. thick, tapered*

*B-Cd plate: 0.020 in. thick
0.020 in. holes - small*

*C-Teflon block: large hole 0.200 in.
small hole 0.070 in., 0.250 in. thick*

The complete, experimental evaluation will involve radiographic testing under optimum moderator and image conditions. Special attention will be focused on the metal hydrides. Approximately 100 in.³ of ZrH_2 , and TiH_2 in metal plate form have been obtained for evaluation.

SOURCE FABRICATION TECHNOLOGYSOURCE MATERIAL PROCESSING*

N. C. Davis

High Energy Rate Forming (HERF) has been developed as a fabrication method for the simultaneous densification and encapsulation of promethium sesquioxide. The process may be considered as a simple closed die forging operation to densify materials or as an extremely complex process with many stress factors affecting the final shape and integrity of a heat source. The isotope is subjected to high impact pressure and temperature in a closed die arrangement. The radioisotope fuel compound, completely contained in a primary or compatibility cladding, is encased in a secondary mold serving as an energy transfer medium. This mold, a sacrificial billet component, also serves as ductile safety containment during impaction. The mold is readily removed by machining or chemical dissolution after HERF. The HERF process has been used in the fabrication of promethium compatibility capsules both in single and multi-capsule impactions. The high density and the intimate interface contact between the fuel core and the cladding are desirable for optimum compatibility conditions.

Although the HERF process produces maximum fuel power density, it does not have the dimensional control capabilities of more conventional fabrication processes. To maintain the best possible dimensional control of a capsule and uniformity of the cladding, some of the HERF parameters to be considered are:

- Fuel core initial density
- Cladding thickness
- Cladding ductility at impaction temperature

* This is a summary of *Fabrication of Promethium Sesquioxide Heat Sources by High Energy Rate Forming*, N. C. Davis and D. W. Brite. BNWL-1041, May 1969.

- Capsule shape
- Tooling arrangement
- Impaction pressure and temperature.

These major parameters and their associated characteristics must be evaluated for any heat source encapsulation design.

For many conventional right cylinder shapes requiring relatively ductile thick-walled cladding, the HERF process will yield a high integrity, dense, and intimately bonded heat source capsule with minimal developmental effort. Minor surface machining of the HERF processed shape may be necessary, depending on the final application. With thin-walled or more complex shapes the extent of required developmental effort must be compared to the need for end product advantages. HERF in some cases may be the most attractive fabrication method.

PROTOTYPE ALPHA SOURCE DEVELOPMENT

W. B. Hall

Efforts were initiated for preparation of advanced alpha radiation sources. ^{241}Am was selected as the initial isotope of interest, and a supply of $^{241}\text{Am}_2\text{O}_3$ was obtained internally at PNL. The objective of present efforts is to prepare $^{241}\text{Am}_2\text{O}_3$ sources suitable for testing at ORNL to evaluate sources prepared by the rf-sputtering process.

Work in this area is being divided into four technical areas:

- Determination of substrate criteria for optimum deposit-substrate bonding
- Determination of window deposit fabrication parameters for optimum deposit bonding and integrity
- Determination of preferred techniques for depositing the isotope source material on the substrate
- Theoretical analyses to determine preferred material types and structures for specific applications, to provide conceptual source designs offering maximum isotope containment, and to provide a continuing basis for experimentation. Work is proceeding in all of these technical areas.

About twenty substrate-deposit samples have been prepared for evaluation of bonding strength as related to substrate surface finish and deposition parameters. The deposition process being used is the rf-sputtering process with sputter etch of the substrate during deposition. Initial specimens consist of copper on stainless steel and silicon on stainless steel substrates. Substrate surface finishes used include a range of machined surfaces, polished surfaces, acid etched, cathodic etched, and pressed and sintered surfaces. Figures 7.1 and 7.2 show a polished stainless steel substrate without and then with copper deposit.

Initial evaluations of deposit-substrate bonding strength have included tensile testing, bend tests, dye penetrant testing and thermal testing. Tensile testing using epoxy cements has been unable to remove thin ($<10\mu$) deposits. Severe bend testing has been able to fracture deposit and substrate materials but has not been able to unbond the ductile copper deposits. The more brittle silicon deposits have been unbonded by severe bend testing.

Both deposit materials have been thermal tested as follows:

- 1 hr. at 750 °F
- 35 min at 300 °F
- Quenching to 32 °F
- Quenching to -70 °F

In addition two silicon deposit specimens were cycled between 750 °F and -70 °F five times. Examination by dye penetrant and microscopic means found all of these specimens to be free of cracks and porosity, both before and after the thermal testing.

Efforts planned for next quarter include installation of the rf-sputtering system in a glovebox, preparation of substrate

Neg 2039

1000 X

FIGURE 7.1. Polished Stainless Steel Substrate

Neg 4C397

400 X

FIGURE 7.2. Copper Deposited on Polished Stainless Steel Substrate by rf-Sputtering Process ($\sim 10\mu$)

with the ^{241}Am source material, and further investigation into the source design concepts for optimum alpha sources. Preparation of hot sources will commence immediately upon completion of the rf-sputtering system installation.

EVALUATION OF ISOTOPE PRODUCTION IN POWER REACTORS

R. W. McKee and S. E. Nunn

 ^{238}Pu COST STUDIESTarget Density Effects

The effect of target density on the cost of producing ^{238}Pu from ^{237}Np has been evaluated. The TAIRCOST program which optimizes target irradiation time was used for the cost calculation; and the target irradiation yield data employed were obtained with the BURN program.

A range of costs, representing estimated maximum and minimum target irradiation costs in a PWR, were used as in the previously reported⁽¹⁾ results for ^{238}Pu costs at a fixed target density, but with two important exceptions:

- Target fabrication (encapsulation) costs were assumed to be inversely proportional to the density
- The neutron flux at the targets was varied inversely with density based on ^{237}Np ALTHAEA⁽²⁾ calculations for target densities of 1.5 g/cm³ and 0 g/cm³ with linear variation in between.

The input data that did not change with densities are shown in Table 8.1 for the high- and low-base-cost cases. Table 8.2 shows the input data for neutron flux and fabrication costs as a function of target density for the same two base cases.

Target density changes affect product costs in several ways. The fabrication cost per unit of target increases as density is reduced, resulting in an increase in product cost. The neutron flux increases as density is reduced, resulting in increased target conversion rates and lower product cost due to the reduction in inventory charges. Self-shielding is reduced as target density is reduced, resulting in a higher proportion of resonance absorptions which increases both conversion rate and yield which in turn result in lower product costs.

The cumulative effects of density variation on the cost of ^{238}Pu are shown in Table 8.3 for ^{237}Np valued at \$100/g. An

optimum or minimum-cost density phenomenon is indicated. In the low cost case the optimum occurs between 0.2 and 0.4 g/cm³. An 18% reduction in cost indicated when the density is reduced from 1.6 g/cm³ to 0.4 g/cm³. This reduction resulted from the increased yield and production rate even though the fabrication cost increased four-fold.

TABLE 8.1. *Base Cost Sets*

	<u>Low Base Costs</u>	<u>High Base Costs</u>
Inventory Charge, %/yr	10	10
Encapsulation and Reprocessing Losses, % each	2	2
Reprocessing Cost, \$/kg	2000	15,000
Irradiation Costs		
Handling, \$/ cm ³	0.1	0.30
Neutrons, \$/g	2700	2700
Revenue loss, \$/cm ³ -yr	0	3.50
Reprocessing Time, months	4	4

TABLE 8.2. *Density Dependent Variables*

<u>Density of 237Np, g/cm³</u>	<u>Average flux, n/cm²/sec</u>	<u>Low Base Fabrication Cost, \$/kg Np</u>	<u>High Base Fabrication Cost \$/kg Np</u>
1.6	1.52×10^{13}	380	2,380
0.8	2.1×10^{13}	760	4,750
0.4	2.38×10^{13}	1,520	9,500
0.2	2.53×10^{13}	3,040	19,000
0.02	2.67×10^{13}	30,400	190,000

TABLE 8.3. *The Effect of Density on ^{238}Pu Production Cost with ^{237}Np at \$100/g*

Target Density, —g/cm ³ —	Low-Base Costs		High-Base Costs	
	Optimum Irradiation Time, months	^{238}Pu Cost. \$/g	Optimum Irradiation Time, months	^{238}Pu Cost. \$/g
1.6	17.4	327	19.5	467
0.8	12.8	284	20.3	430
0.4	11.3	270	18.3	456
0.2	11.0	270	16.9	527
0.02	27.8	354	13.8	2100

For the high cost case the advantage for reduced density is not as large and the optimum density is about 0.8 g/cm³ with a cost reduction of 8% relative to the cost at 1.6 g/cm³. When the density is reduced to very low values in the high cost case, the effect of fabrication and revenue loss costs results in a very steep increase in product cost. For example, between 0.2 g/cm³ and 0.02 g/cm³, product cost increases four-fold.

The revenue-loss component of product cost in the high cost case is relatively not too important at the higher densities but becomes the controlling cost at low densities. This cost represents the cost for displacing power producing fuel rods with target rods and would not be incurred when target rods are inserted just in unused control rod locations. When this cost is deleted from the high cost case, the minimum product cost is approximately \$390/g at a density of 0.6 g/cm³.

In either the low or high cost case the ^{238}Pu product cost is not very sensitive to small displacements from the optimum irradiation time. The irradiation time could be adjusted from the values shown in Table 8.3 to coincide with a normal annual refueling without any significant change in product costs,

Effect of Purity

The effect of purity on the cost of producing ^{238}Pu from ^{237}Np target irradiation in a power reactor was also examined using results of the TAIRCOST program and target irradiation yield data from the BURN program. The cost inputs were the same for the density effect evaluations (Tables 8.1 and 8.2).

Figure 8.1 shows the effect of increasing exposure on ^{238}Pu purity. To obtain purities of greater than 90%, relatively short exposures are required. For example, with a flux of $2.5 \times 10^{13} \text{ n/cm}^2/\text{sec}$, the irradiation time would be only 9 months. The amount of ^{241}Pu in the product is also dependent on exposure and increases rapidly at higher exposures due to the buildup of its parent, ^{240}Pu .

In Figure 8.2, both purity and cost are plotted as a function of exposure. This allows one to compare the cost for a specific purity against the minimum or optimum cost. Three cases are shown:

- Neptunium at \$100/g with high costs and optimum target density
- Neptunium at \$100/g with low costs and optimum target density
- Neptunium at \$50/g with low costs and optimum target density

If the product purity is restricted to no less than 80%, the minimum cost product could always be realized. However, with a requirement of 90% purity, a cost premium is indicated. For example, in the high cost case with Np at \$100/g, the ^{238}Pu cost would increase from the minimum cost of \$430 at a purity of 84% to a cost of \$460 at a purity of 90%.

If purity requirements are no more stringent than 90% ^{238}Pu , the cost premium would be no more than 5 to 10% of the minimum cost. However, the cost premium increases sharply at purities above 90%.

Exposure, 10^{20} n/cm^2 *

* Irradiation Time, days =

$$\frac{(\text{Exposure, n/cm}^2)}{(8.64 \times 10^4 \text{ sec/day}) (\text{flux, n/cm}^2/\text{sec})}$$

FIGURE 8.1. Effect of Exposure on ^{238}Pu Purity

FIGURE 8.2. Cost of ^{238}Pu as a Function Exposure and Purity

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RADIATION CHARACTERISTICS OF ISOTOPES FOR
APPLICATIONS IN CIRCULATORY SUPPORT

A. SHIELDING AND IN-PHANTOM DOSE PREDICTION

F. T. Cross

Bach and Caswell's neutron fluence-to-dose conversion factors have been incorporated into the QAD library. In addition, the format of QAD had been changed for greater convenience in reading.

In-phantom dose-rates have been calculated with the QAD code for an 8.42 μg ^{252}Cf needle source. These dose-rates are in acceptable agreement with the experimental values reported in the literature and the values measured at PNL

The QAD code was also used to calculate the neutron and photon dose-rates from radiobiological sources simulating ^{238}Pu (table below). The ^{90}Sr - ^{90}Y and ^{241}Am -Be sources were assumed to be of the same geometry and shielding as LASL-1, the 30 watt ^{238}Pu source presently in use at PNL. The ^{252}Cf dose-rates are those previously calculated for the SRL radiotherapy needle which contains a comparable thickness of platinum shielding to the tantalum of the ^{238}Pu source. The tabulated photon dose-rates are normalized at 1 cm into tissue from the source sides. The quantities of isotopes are based on equal neutron emission rates to a 30 W ^{238}Pu source.

The data also indicated that for an equivalent neutron emission rate, the neutron dose rate at 10 cm into tissue would be 1.5 times greater from the ^{241}Am -Be source. This is due to the higher effective energy from the ^{241}Am -Be spectrum. For an equivalent neutron dose-rate, therefore, the emission rate would need to be reduced an equal amount. Once this quantity is determined, the total photon dose-rates can be simulated by adding the correct amount of bremsstrahlung from a ^{90}Sr - ^{90}Y source to the photon dose-rates from the ^{241}Am . Similar considerations apply to mixtures of ^{252}Cf and ^{90}Sr - ^{90}Y .

TABLE 9.1. *Photon Dose-Rates in Phantom*

Tissue Thickness	^{238}Pu (a)		^{252}Cf (b)		^{90}Sr - ^{90}Y (c)		^{241}Am (d)	
	mrad/hr	Rel.	mrad/hr	Rel.	mrad/hr	Rel.	mrad/hr	Rel.
1 cm	185.5	1.00	82.1	1.00	185.5	1.00	10.7	1.00
5 cm	16.1	0.087	4.0	0.049	18.0	0.097	1.0	0.093
10 cm	3.3	0.018	0.9	0.011	5.2	0.028	0.2	0.019

(a) *LASL-1 Source at beginning of mission life (no ^{236}Pu)*

(b) *0.071 μg ^{252}Cf source with 1.67×10^5 n/sec at beginning of life*

(c) *0.53 Ci ^{90}Sr - ^{90}Y source (quantity varies with source configuration)*

(d) *^{241}Am -Be source with 1.67×10^5 n/sec*

Monte-Carlo calculations have been made on various types of phantoms to determine the effect of phantom size and shape on the neutron dose-rates. These data have not yet been analyzed.

B. IN-PHANTOM DOSIMETRY MEASUREMENTS

In-Phantom measurements with ^{252}Cf and ^{238}Pu continue. The dose-rate values found thus far are in acceptable agreement with the predictions by the modified QAD code. There is generally better agreement with the Bach-Caswell fluence to dose conversion factors than with those by Snyder-Neufeld. On the average, the Snyder-Neufeld neutron dose-rate predictions are about 25% greater than the Bach-Caswell predictions. Since the former conversion factors include multiple scattering whereas the latter are more applicable to "first collision dose-rates", the in-phantom measurements should be in closer agreement with the Snyder-Neufeld predictions. Any discrepancies will be clarified as the experimental error is reduced and/or the values of the various parameters in the code libraries are improved.

C. ^{146}Pm BURNOUT BY NEUTRON IRRADIATION

J. C. Sheppard

Two ^{147}Pm targets, originally containing about 3.5 ppm of ^{146}Pm and irradiated in a Savannah River reactor to "burn-out" the ^{146}Pm , were returned to BNW in last May. These targets, which had cooled about 5 to 6 months to allow a substantial amount of $^{148\text{m}}\text{Pm}$ to decay away, were opened in the hot-cells and samples were taken for analysis in mid June. The $^{148\text{m}}\text{Pm}/^{147}\text{Pm}$ ratios for these samples were determined. As expected, the $^{148\text{m}}\text{Pm}/^{147}\text{Pm}$ ratios for these samples were about ten times those found for Hanford irradiated promethium which had cooled about 1 year. An attempt to determine the $^{146}\text{Pm}/^{148\text{m}}\text{Pm}$ ratios for these Savannah River irradiated promethium targets, even though the very high $^{148\text{m}}\text{Pm}$ was expected to interfere with the detection of ^{146}Pm , was planned for late June using J. Cooper's high-sensitivity Ge(Li) gamma detecting system. This system was inoperative during this time, so the samples were not counted. The Savannah River target material will be allowed to cool for another 5 or 6 months to reach the $^{148\text{m}}\text{Pm}/^{147}\text{Pm}$ ratios found for the Hanford target material and the $^{146}\text{Pm}/^{148\text{m}}\text{Pm}$ will be determined at that time.

A report on the Hanford ^{146}Pm "burn-out" experiments is in the final stages of preparation and will be published in early September.

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