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PACIFIC NORTHWEST LABORATORY
DIVISION OF ISOTOPE
DEVELOPMENT PROGRAMS
QUARTERLY REPORT: FEBRUARY - APRIL 1970
May 1970

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Radiation Applications

PACIFIC NORTHWEST LABORATORY
DIVISION OF ISOTOPE DEVELOPMENT PROGRAMS
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INTRODUCTION

This report, BNWL-1308-2, is the 14th 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, 1085, 1177, 1256 and 1308-1.

This report covers the 3-month period, February through April 1970.

SUMMARY

LASER APPLICATIONS

Analysis of candidate isotopes for the γ -ray laser concept has produced four isotopes worthy of further study. They are ^{193}Ir , ^{191}Os , ^{165}Dy and ^{192}Ir . Problems associated with laser-activated separation of isotopes of lithium and carbon have been considered.

PROMETHIUM COMPATIBILITY

The final draft of an unclassified document detailing the results of the entire $^{147}\text{Pm}_2\text{O}_3$ compatibility program has been completed. The compatibility testing of various containment materials with samarium metal (stand-in for promethium metal) is continuing and will be completed in FY-1970. The results of tests completed to date are presented.

ENCAPSULATED WASTE MANAGEMENT CESIUM AS A PROCESS RADIATION SOURCE

The incentives for reducing cesium source diameters or container thicknesses were evaluated semiquantitatively. Any reduction in container thickness will be beneficial, and for cesium costs of more than 5¢ per curie, a reduction in container diameter will be beneficial. Strontium fluoride offers good promise of being an acceptable fuel form for most existing or proposed heat source applications. Encapsulation of both strontium and cesium at PNL was found to offer significant advantages under some demand conditions, but the advantage will not be great unless demands increase.

RADIATION CHARACTERISTICS OF FISSION PRODUCT RHODIUM AND PALLADIUM

Work is continuing to determine the half-life of $^{102\text{m}}\text{Rh}$.

EVALUATION OF LARGE SCALE USES FOR PURIFIED FISSION PRODUCED XENON

The Xenon study was authorized with partial funding for the remainder of this fiscal year. Arrangements were completed for shipment of the crude gas from NRTS, ICCP and the purification at ORNL. Advice and guidance on licensing or approval for test uses was requested from the Division of Radiation Protection Standards. Radiochemical purification to remove ^{85}Kr to acceptable levels appears to be attainable in existing facilities. The most promising large scale application may be for use as the principal agent in a human anesthesia.

NONDESTRUCTIVE TESTING TECHNIQUES

NDT for Irradiation Sources

Development was continued on a multielement, ultrasonic transducer which will permit scanning wide paths on large, flat surfaces for internal defects.

Neutron Radiography with ^{252}Cf

A first generation portable neutron device (NC-1) has been developed and evaluated. An optimum condition for producing neutron radiographs has been initiated and is presently being used. The engineering and material characteristics of the neutron camera have been summarized and assessed. Areas where a portable device would have the greatest potential for applications have been established. The materials and material problems associated with potential applications have been classed and the experimental and theoretical data for a thorough study are being acquired.

ADVANCED SOURCE DEVELOPMENT

Select alpha-photon sources prepared in the previous quarter were evaluated. Twenty-five $^{241}\text{AmO}_2$ photon sources

were prepared in a single run using rF-sputtering techniques. Five of these sources were sent to Oak Ridge National Laboratory for testing and evaluation. The preparation of 25 alpha-photon sources in a single run is in progress.

EVALUATION OF ISOTOPE PRODUCTION IN POWER REACTORS

A revised series of parametric cases have been prepared to show the range of costs that can be anticipated for ^{238}Pu produced by neptunium target irradiation in a pressurized water power reactor. The results show the effects of varying losses, inventory charges and reprocessing times on ^{238}Pu costs as a function of neptunium cost.

A series of isotopic property compilations is being prepared using data stored in the Battelle-Northwest Isotopic Data Tape system. The various tables will arrange the isotopes in decreasing order of such important properties as half-life and decay energy.

RADIATION CHARACTERISTICS OF CIRCULATORY SUPPORT ISOTOPIC HEAT SOURCES

The shielding and in-phantom dose predictions for LASL-1 continue. Calorimetric measurements indicate the heat output of LASL-1 to be 29.034 ± 0.063 (2σ) W on 1-30-70. Measurements of the neutron dose rates from LASL-1 placed within the homogeneous phantom have been started. Total ($n+\gamma$) dose rates within and surrounding the Remab phantom from LASL-1 are presented.

Analysis of the Savannah River promethium targets indicated that the ^{146}Pm concentration was reduced by almost a factor of two for the longest irradiation. This reduction was less than expected. A suggestion was made to use graphite targets to reduce the $^{147}\text{Pm}(\gamma, n)^{146}\text{Pm}$ reaction and thereby achieve a substantial reduction in the ^{146}Pm concentration.

NUCLEAR TECHNIQUES FOR SEABED MINERAL EXPLORATION

A laboratory seabed mineral analyzer prototype which will use a 2-mg ^{252}Cf source has been designed and is under construction. A similar mock-up system was used to evaluate the precision with which the elements in minerals could be determined. The results of this latter investigation, "Laboratory and Environmental Mineral Analysis Using a Californium-252 Neutron Source," were submitted for publication in Nuclear Applications.

LASER APPLICATIONS

W. Vali

INDUCED γ -RAY EMISSION

An analysis of induced γ -ray emission on 12 isotopes was initiated in March 1970. The object was to determine the length of crystal required to reach the threshold of laser action. The following basic assumptions were made:

- Pumping of isomeric states is done by thermal neutron flux of $10^{15} \text{ sec}^{-1} \text{ cm}^{-2}$.
- The minimum number of coherent γ 's was taken to be 10^8 .
- Since detailed data on absorptivity and second-order line broadening are unavailable, these effects were disregarded.

Of the 12 isotopes considered, only ^{193}Ir , ^{191}Os , ^{165}Dy , and ^{192}Ir were projected to require crystals of reasonable length to be of further interest, i.e., no more than a few decimeters. It was concluded that the next step to be taken is the measurement of absorptivity of the γ -s along the Bragg angle with Mossbauer recoilless γ -rays in a good single crystal. Other means of pumping should also be considered.

LASER ISOTOPE SEPARATION

The literature work on isotope separation has revealed the following items of interest:

The isotope shift between the $6707 \text{ \AA}$ lines ($2 \text{ P}_{1/2, 3/2} \rightarrow 1 \text{ S}_{1/2}$) of ^6Li and ^7Li is $150 \text{ M \AA}$. This separation is sufficient to allow excitation of only one isotope in lithium vapor. A chemical reaction is now being sought to take advantage of this circumstance.

Initial ideas have been formulated on a possible process of separating ^{13}C from ^{12}C in natural CO_2 gas. One approach would employ a CO_2 laser containing only one isotope of carbon.

A candidate reaction is

The natural CO_2 must be held at an elevated temperature inasmuch as a workable percentage of CO_2 molecules will be in the first symmetric vibrational state. The laser light would then serve to excite these molecules further, to the first asymmetric vibrational state. Yet to be determined is the magnitude of the change in reaction rate induced by the presence of asymmetric vibrational excitation.

PROMETHIUM COMPATIBILITY

H. T. Fullam and J. H. Jarrett

The final draft of an unclassified document detailing the results of the entire $^{147}\text{Pm}_2\text{O}_3$ compatibility program has been completed. The report is now in the process of being cleared for publication.

The first draft of the report on the reactions of Pm_2O_3 with water has been completed (coauthored by F. P. Roberts).

A paper on the "Reactions of Pm_2O_3 , Sm_2O_3 and Nd_2O_3 with Water," by H. T. Fullam and F. P. Roberts, was presented at the Eighth Rare Earth Research Conference in Reno, Nevada in April, 1970. H. T. Fullam presented the paper.

The compatibility testing of various containment materials with samarium metal (stand-in for ^{147}Pm metal) was continued. The metallographic examination of the specimens removed from the furnaces in November has been completed. Summary compatibility information is presented in Table 1. The data shown in Columns 3, 6, and 8 are from the specimens taken out of the furnaces in November. The data in Columns 1, 2, 4, 5, and 7 were presented in the May-July 1969 DID Quarterly Report, BNWL-1177 Sup. The information is again presented for purposes of comparison.

It is important to note that containment materials have been identified which show less than one mil penetration of the samarium-containment material interaction zone into the containment material wall. This is shown quite clearly in Table 1. In each case the penetration was determined visually by the metallographic examination of a cross-section showing the containment material-interaction zone interface. Materials that show the least interaction with samarium are tantalum, Ta-10W, titanium, zirconium, and TZM.

TABLE 1. Metal-Containment Material Compatibility Using Samarium Metal as a Stand-in for ^{147}Pm Metal

Containment Material	Compatibility ^(a)							
	1000 hr 400 °C	4400 hr 400 °C	8800 hr 400 °C	1000 hr 700 °C	4400 hr 700 °C	8800 hr 700 °C	1000 hr 1000 °C	4400 hr 1000 °C
Tantalum	<1	<1	<1	<1	<1	<1	<1	<1
Ta-10W	<1	<1	<1	<1	<1	<1	<1	<1 ^(c)
TZM	<1	<1	<1	<1	<1	10	<1	<1 ^(b,c)
Titanium	<1	<1	<1	<1	<1	<1	<1	<1 ^(c)
Zirconium	<1	<1	<1	<1	<1	<1	<1	(d)
Inconel	<1	<1	80	125	125	125	75	(e)
Haynes-25	<1	<1	40	60	50	50 ^(c)	75	(e)
Hastelloy-X	<1	<1	75	65	63	65 ^(c)	100	(e)
Type 304L SS	<1	<1	20	40	53	50	125	(e)
Type 316L SS	<1	<1	30	60	61	55	125	(e)

- a. Compatibility - The maximum penetration of the interaction zone into the 125 mil containment material wall, mil. The penetration was determined by the metallographic examination of a cross-section showing the containment material-interaction zone interface.
- b. 3400 hr test.
- c. There was evidence that samarium metal may have been released from the capsule - probably in the weld area. The samarium release appeared to result from lack of weld integrity since the interaction between the capsule wall material and samarium metal was in most cases minimal.
- d. The quartz ampoule containing the capsule broke, allowing the capsule and the samarium metal to oxidize.
- e. Because of the severe interaction between samarium and Inconel, Haynes-25, Hastelloy-X, Type 304L SS and Type 316L SS in the 1000 hr, 1000 °C tests, these materials were not tested further at 1000 °C.

The 8800 hr, 1000 °C tests are scheduled to be completed in June, 1970. This will be the final phase of the samarium metal (stand-in for ^{147}Pm metal)--containment material compatibility testing.

ENCAPSULATED WASTE MANAGEMENT CESIUM AS A
PROCESS RADIATION SOURCE

H. H. Van Tuyl

The scope of the work was changed during the reporting period to include the use of strontium from the Hanford Waste Management Program (HWMP). Work on cesium has been limited to paper studies, with experimental work being deferred until next year. A brief study was made of the feasibility and cost of producing cesium and strontium sources at PNL until sources are available from HWMP. Progress in each of the three areas is subsequently reported in separate sections.

CESIUM SOURCES

H. H. Van Tuyl and L. K. Mudge

Cost estimates for fabricating cesium sources in the Hanford Waste Management Plant have been studied as a function of source size. The standard storage container, which holds about 60 kCi of ^{137}Cs , will require about six hours of operator time per container at the nominal production rate of 30 MCi per year. Estimates for smaller source sizes are dependent upon a capability to process more containers in the same work station.

HWMP is designed to optimize production at about 500 cesium containers per year, with reasonable allowance for maintenance and unscheduled down-time. Some increase in number of containers is possible with the same basic equipment and procedures, but with an increase in work force. Additional increases can be made by further refinements in work procedures, automation of operations wherever possible, and equipment development for simultaneous handling of a larger number of containers.

One major limitation in producing more containers will be the capability to fill the containers in the melt-cast operation.

At present, plans call for filling four containers simultaneously. Engineering work is in progress at HWMP to solve the problem of adequate vacuum seals with only four containers. A large increase in throughput would require simultaneous vacuum seals for a larger number of containers, or a very rapid procedure for changing containers. Cost estimates and production capabilities are highly dependent upon the optimism used in predicting results of development efforts.

Optimistic extrapolation of capabilities would indicate a potential for a four or fivefold increase in the total number of containers, along with a cost reduction per container of perhaps as much as a factor of two. At 15 kCi per capsule, this production rate is large enough to accommodate the full HWMP production. Such a large increase would require very high operating efficiency, and additional research and development which is neither planned by HWMP nor currently included in the DID program. A more pessimistic estimate of a twofold increase in capacity would permit encapsulation of 10 MCi per year in 15 kCi containers, and the balance in 60 kCi containers. Also, the cost per capsule may be essentially unchanged in spite of the higher production rate.

Although the actual production capacity and cost per container is difficult to evaluate, it appears reasonable to evaluate the incentives for changing source size on the basis of \$200 per container regardless of size. The evaluation was made on the basis of effective source strength. The actual number of curies in a capsule was corrected for attenuation in the source and in the cladding to obtain the number of curies of an unshielded and non-absorbing source which would produce an equivalent radiation field. This reduced number of curies is the effective source strength. For a preliminary evaluation, the effect of self shielding was assumed to be represented by energy absorption in the source, and the effect of

external shielding was assumed to be dependent upon absorption of primary radiation. Attenuation factors for these effects were described in the last report in this series (BNWL-1308-1). Additional evaluations need to be made, based upon irradiation of slabs of varying thicknesses, to obtain the desired dose rate at the center of the slab.

The evaluation shown in Table 2 was based on two outside diameters of the inner cesium container, 2.0 and 1.0 in. Inner container wall thicknesses of 95 or 25 mils were used, along with outer container wall thicknesses of 109 or 25 mils. Cesium chloride costs of 12.5¢, 5¢, and 2¢ per curie were used. For each configuration evaluated, the actual and effective curies per capsule was determined. The number of capsules required to yield an effective megacurie of ^{137}Cs was calculated, along with the actual megacuries of cesium required to produce this amount of radiation. The decrease in cesium requirement (as compared to the HWMP design of 2.0-in. diameter inner container, wall thicknesses of 95 and 109 mils) resulted in a savings in cesium cost. The change in the number of capsules resulted in a savings or penalty for capsule fabrication. The net savings were then calculated as cents per effective curie of ^{137}Cs . This shows the incentives for changing cesium diameter, inner wall thickness, outer wall thickness, and price of cesium based upon the assumed effectiveness of shielding.

On the basis of this analysis, a decrease in wall thickness is strongly favored. A decrease in container diameter is favored so long as the price for cesium exceeds 5¢ per curie. If deep penetration into a target is desired, the effect of self shielding will be much greater, and a large incentive will exist for a decrease in container diameter.

TABLE 2. Incentives for Changing Cesium Source Size

OD of Inner Container, in.	Wall Thickness, mils		¹³⁷ Cs Content, kCi		Net Savings, ¢/eff.Ci		
	Inner	Outer	Actual	Effective	12.5¢/C	5¢/Ci	2¢/Ci
2.0	95	109	60	33	---	---	---
2.0	25	109	70	40	0.9	0.4	0.2
2.0	25	25	70	46	3.8	1.6	0.7
1.0	95	109	12	8	0.9	-0.8	-1.5
1.0	25	109	17	11	2.8	0.4	-0.5
1.0	25	25	17	13	5.5	1.6	0.1

Cesium source optimization was considered with respect to licensing for use and safety in shipping. Some reduction in wall thickness could be achieved and still meet the 2R classification. Further reduction in wall thickness is not expected to cause appreciable problems in either shipping or licensing, so long as corrosion will not weaken the wall unduly. During shipment, the protection afforded by the cask can be adequate to withstand accident conditions. The possible accident conditions during use can be controlled, and the consequences of accidents can be minimized. Thus, the source itself does not necessarily have to withstand severe accidents. Some additional encapsulation may eventually be required for ultimate disposal. The principal considerations in source optimization are price of cesium, incremental cost of fabrication and encapsulation in special sources, and dose rate uniformity from the assembled irradiator. A report is being written on cesium source optimization.

STRONTIUM SOURCES

H. H. Van Tuyl, J. H. Jarrett, and E. J. Wheelwright

Strontium is currently being removed from Purex wastes and stored as an impure concentrate of the approximate composition shown in Table 3. By FY 1974, equipment will be

installed to permit further purification of the strontium, conversion to the fluoride, and encapsulation for long-term storage. At present the container is expected to be fabricated from Hastelloy C-276, but other materials are also being considered. The inner container will be about 2-1/2 in. OD by 15.4 in. long, with a wall thickness of 120 mils. This will be inserted into an outer container, about 2-7/8 in. OD by 16.5 in. long, with a 120 mil wall thickness. The flexibility of the plant for fabricating alternative container geometries is being studied.

TABLE 3. Crude Strontium Composition

<u>Constituent</u>	<u>Concentration</u>	
	<u>Molarity</u>	<u>g/l</u>
H	2.85	2.8
Sr	0.096	8.4
Na	0.51	11.7
Ca	0.158	6.3
Mg	0.183	4.5
Ba	0.0016	0.22
Mn	0.201	11.0
Ni	0.0024	0.14
Al	0.041	1.1
Cr	0.0055	0.29
Fe	0.011	0.61
Pb	0.010	2.1
¹⁴⁴ Ce-Pr	(232 Ci/l)	
⁹⁰ Sr	(705 Ci/l)	

Strontium has been used primarily as the titanate, although some strontium oxide sources have also been prepared. HWMP has selected the fluoride for storage of strontium since it is more easily prepared and has adequate properties for their needs. Some properties of strontium compounds are

compared in Table 4. Visits were made to DRDT Isotopic Power Branch, Naval Facilities Engineering Command, Division of Materials Licensing, and Teledyne-Isotopes to discuss the potential substitution of strontium fluoride for strontium titanate in heat sources. The principal concern of users is cost of the completed heat source in an approved and licensed manner. For most applications foreseen at this time, fluoride is either directly usable instead of the titanate, or design changes could accommodate the fluoride with no loss of performance and little or no economic penalty. Acceptance of the fluoride is contingent upon licensing. The known properties of the fluoride seem to offer no appreciable obstacles to licensing. However, additional data need to be collected concerning the fluoride. One area in which more data are required is the effect of probable impurities on melting point and other physical properties. Published data are being reviewed to determine which properties still need to be measured.

The flowsheets for purification of strontium were reviewed briefly. Solvent extraction, ion exchange, and precipitation flowsheets are available from laboratory studies, but have not been demonstrated on a plant scale. Current solvent extraction processes are not expected to yield a purity as high as is desired, and the impurities (principally magnesium) will probably be high enough to make application of existing compatibility data uncertain. Ion exchange flowsheets offer good promise of meeting the required purity, both chemical and radiochemical, but no suitable equipment is available in B-Plant. Precipitation flowsheets offer good promise for adequate chemical purity, but little separation from cerium is expected in the sulfate precipitation. A combination of solvent extraction followed by hydroxide precipitation to remove magnesium is expected to yield adequate purity.

TABLE 4. Properties of Selected Strontium Compounds

Property	<u>SrTiO₃</u>	<u>Sr Metal</u>	<u>SrF₂</u>	<u>Sr₂TiO₄</u>	<u>SrO</u>
Theoretical Power Density, ^(a) W/cm ³	1.26	1.33	1.54	1.57	2.17
Theoretical Density, g/cm ³	5.11	2.60	4.29	4.99	4.98
Melting Point, °C	2040	722	1463	1860	2457
Thermal Conductivity at 700 °C cal/cm-sec-°C	0.0082	0.206	0.0031	0.0073	0.0050
<u>Potential As-Produced Properties</u>					
Purity, %	97		97	97	
% TD	90		80	90	
Power Density, W/cm ³	1.10		1.19	1.37	

a. Theoretical Power Density - 100% Purity Fuel Forms at 100% TD with ⁹⁰Sr Equal to 55% of Total Strontium.

ENCAPSULATION AT PNL

H. H. Van Tuyl, V. L. Hammond, and F. L. Mourich

Firm orders for both cesium and strontium have been increasing in recent months and are expected to total two megacuries of each isotope in FY 1971. At present, the supplies of both cesium and strontium on hand at ORNL are limited. Because of changes in shipping regulations, no suitable casks are available or approved at present for shipping very large amounts of either cesium or strontium to ORNL. The facilities at Hanford which were previously used for strontium purification have now been deactivated. Unless other arrangements are made for strontium purification, and for either shipping to ORNL or encapsulation at PNL, neither cesium nor strontium will be available much longer.

Several alternatives are available to the AEC. Purification can be accomplished with greatest assurance of success by using the PNL ion exchange facility now being used for promethium purification. The purified material could be converted to the desired chemical form and encapsulated at PNL using existing hot cells. Alternatively, the purified material could be shipped to ORNL for conversion and encapsulation.

A brief study was made of the feasibility and cost of producing encapsulated sources at PNL. Many advantages would be realized by performing the purification, conversion, and encapsulation at PNL. Improved safety and lower cost at high production rates are probably the greatest advantages, but other significant benefits would also be realized. However, review by DID concluded that the advantages of encapsulation at PNL were not sufficiently large to outweigh the initial set-up costs in view of the limited expected lifetime of the facility, and uncertain future demand for both cesium and strontium. Both cesium and strontium will continue to be purified at Hanford and shipped to ORNL for encapsulation,

until HWMP begins to produce sources in FY 1974. Licensing for suitable casks has been requested.

The availability of adequate strontium purification capability was also reviewed. ARHCO is currently holding 700,000 curies of Semiworks purified ⁹⁰Sr at B-Plant. The purity is probably satisfactory for direct conversion to heat source grade fuel, but the material has been transferred through several tanks of uncertain contamination status. Two cask transfers would be required to move this material to ORNL as a solid carbonate. ARHCO operations has permission from the AEC to blend this material with current B-Plant strontium product if and when tank space becomes a problem. ARHCO intends to keep it isolated as long as they can.

HWMP expects to employ a second solvent extraction cycle employing DTPA and citric acid complexing agents. The citric acid is used to remove calcium. This operation will yield satisfactory DFs from all impurities except magnesium, which can be removed as a precipitate by adding caustic to pH 10. Equipment costing in excess of \$500,000 will be required before the second solvent extraction cycle could be adopted routinely. Because of the need for a new concentrator, the actual incremental cost would be around \$25,000. These facilities are included in HWMP as part of encapsulation and are scheduled for operation in FY 1974.

The first-cycle strontium product could be purified in the interim by a double sulfate precipitation. This would require a follow-on treatment to remove the cerium. ARHCO is considering a plant test late in CY 1970. Such a test would require \$30,000 to \$50,000 for equipment. The major problem associated with a double sulfate precipitation is that it must be performed in the equipment now being used to recover strontium from the Purex waste tank acid sludge. An interruption

in the Waste Management Program schedule would be required as well as some risk of contamination with overaged strontium. Cerium removal is also uncertain.

Experiments are currently in progress to evaluate the purification of first-cycle strontium product by absorption on the zeolite exchanger, Linde 13X. If the hot cell experiments confirm this method as a useful plant process, it would be possible to use the cesium ion exchange equipment for strontium on a campaign basis. Unfortunately, the equipment will not be ready for operation before FY 1974.

Suitable facilities exist at PNL for purifying strontium on Dowex resins, using a demonstrated flowsheet. Since this offers the best assurance of adequate strontium purity, cost estimates for this operation are being generated.

RADIATION CHARACTERISTICS OF FISSION PRODUCT
RHODIUM AND PALLADIUM

F. P. Roberts

^{102m}Rh Half-Life

The exploitation of fission product rhodium for some industrial applications may be limited by the amount of radioactive rhodium present. From earlier work it is known that ^{102}Rh and ^{102m}Rh are the principle radioactive isotopes present in power-reactor produced rhodium. The ^{102m}Rh has a half-life in excess of three years and will be the limiting factor for determining time needed for decay of the radioactivity to acceptable levels. At present the value of the half-life of ^{102m}Rh is not known with sufficient accuracy for this purpose. A study is in progress to establish an accurate value for the half-life.

Samples of rhodium have been recovered from 13-year old Hanford waste. Because of the great age of this material it contains an insignificant amount of the short-lived ^{102}Rh and is ideal for following the decay of ^{102m}Rh .

After extensive purification of the rhodium and packaging in sealed vials, the decay of ^{102m}Rh is being followed by gamma counting using a germanium diode. The decay has been measured over a 9-month period and will be continued until sufficient precision is obtained to give a half-life value within 10% of the true value.

EVALUATION OF LARGE SCALE USES FOR PURIFIED
FISSION PRODUCED XENON

C. A. Rohrmann

Authorization to initiate the fission product xenon large scale applications study was received in this period. Contacts were made with Division of Radiation Protection Standards to obtain details of licensing and/or approvals for the test use of the material. Also arrangements were made with ORNL for the purification of the crude material. This material was also scheduled for shipment to ORNL from NRTS early in May. It appears that ORNL should be able to purify to the desired 1 to 10 microcuries per liter (STP) at reasonable yield in existing facilities. It is uncertain as to the separating that may be achievable from other contamination. Such contaminants include oxygen, water, hydrocarbons, carbon oxides, and nitrogen oxides. Before steps are taken to obtain customers for this unique element, a fairly complete analysis (along with advice from the Division of Radiation Protection Standards) will have to be obtained.

By way of review, the background, bases, reasons and plans for the study are summarized in the following paragraph.

The rare and inert gas xenon has several unique properties. However, the rarity of it in nature makes it essentially impractical for consideration for major large scale uses. On a weight basis in the fission reaction (nuclear power) xenon occurs as the most prevalent fission product. The growth of nuclear power and accompanying fuel processing ensures that xenon need no longer be considered rare and from this source can indeed be considered available for beneficial large scale uses. In addition with minimized air in-leakage the off-gases from the fuel processing dissolution step should be very rich in xenon, conceivably well above a few percent by volume.

Furthermore, from typical fuel cycles the residual fission-produced isotopes are all stable. All radioactive ones that exist have such short half lives that in the anticipated time between creation and use the activity would be reduced to insignificant levels (10^{-10} curies per liter). The most promising use is in human anesthesia. Its anesthetic properties are unique among the rare gases as well as among conventional anesthetics (krypton is ineffective). In addition, man among all mammals is uniquely specific to its anesthetic effect at atmospheric pressure. Other mammals respond only when the gas is at super-atmospheric pressures.

The anticipated off-gas concentration plus the relationship of fission product xenon to the highly radioactive but less prevalent (by a factor of about ten) fission product krypton provide reasonable expectations for low cost production--less than one dollar per liter. At prices near such levels other uses such as more widely applied high intensity plasma arc lighting and replacement of presently used argon and krypton for specialty type, long-lived, brighter, incandescent filament lamps can be anticipated. However, with satisfactory low cost separation from radiokrypton, which appears to be readily achievable (and the possible future requirement for ^{85}Kr removal and secure storage for pollution abatement reasons), the anesthetic uses would probably consume all xenon that could be made available in the next 30 years.

A program managed by Battelle-Northwest is underway to have the substantial stock of crude xenon accumulated at NRTS purified at ORNL to remove the ^{85}Kr activity to acceptable levels (based on Division of Radiation Protection Standards recommendations) so that the gas can then be distributed without charge to a variety of users (producers, users of nuclear power reactor fuel processors) who would evaluate it for certain potentially large-scale applications and report the

results of their evaluations. Battelle-Northwest would assemble and report on these evaluations. By this means the future available resources of fission product xenon may be most effectively promoted for beneficial applications.

NONDESTRUCTIVE TESTINGNDT FOR IRRADIATION SOURCES

R. W. Steffens

The NDT methods and device development continued with the fabrication of detector gate circuits for integration into the multielement ultrasonic instrumentation. The addition of this gate completes the development of the tester circuits and development of analog circuits will be initiated to implement the CRT readout. Performance of general type ultrasonic test circuitry associated with the multielement instrument was evaluated for use in inspecting closure type welds encountered on isotope heat sources and excellent results were obtained.

A topical report is being prepared on this phase of the program.

NEUTRON RADIOGRAPHY WITH ^{252}Cf

J. L. Cason

Research, development, and utilization of portable isotope sources for neutron radiography requires that several conditions be fulfilled. These conditions include needs to:

- Develop a first generation portable neutron device capable of performing neutron radiography.
- Resolve the optimum conditions for operation and use of a portable source.
- Evaluate the ability of a portable device to withstand adverse environmental (natural and artificial) conditions.
- Establish these areas of application where a portable neutron source is most useful.
- Define the material and material problems associated with a given area of application.
- Define the capability and characteristics of a radiographic system.

- Obtain the experimental data necessary to evaluate and establish the feasibility of using neutron radiography for specific applications.
- Evaluate specific applications.

Once these conditions are met, the use of portable sources for thermal neutron radiography will be ensured for future applications.

DISCUSSION

The development of a portable neutron camera⁽¹⁾ has been followed by an evaluation of optimum operation conditions. These operation conditions include the establishment of neutron radiographic techniques for use with a portable device. Included is an evaluation of techniques for use with small non-reactor sources where the number of neutrons is limited. Conditions included flux measurements⁽²⁾ and neutron radiographs.⁽³⁾ Once the feasibility of using such a device for thermal neutron radiography had been demonstrated, the device was characterized as to usability. Usability in this case is defined to mean evaluation of requirements for shipping, withstanding adverse environmental conditions, and trouble-free operation. An evaluation of the usability has been initiated and is near completion. The results of those portions completed will be covered in this report. The establishment of a sound engineering design for a portable neutron device is just as important as ensuring that the device perform radiographically as determined by the analytical and experimental evaluation.

Application studies require incorporation of Items 4 through 7 of the introduction. This development stage is underway and initial results include use of special imaging and collimation techniques for small source radiography. An outline of proposed material studies and related applications is provided as a guide for future research and development efforts.

Research leading to the development of the neutron device, preliminary flux measurements, and neutron radiographs have been discussed and reported previously.⁽¹⁻³⁾

Imaging with Small Sources

Recent results include an improved imaging technique using a $\text{Li}^6\text{F ZnS(Ag)}$ neutron scintillation screen with a special RAR Kodak film. Accessory hardware for the use of this direct imaging technique includes a vacuum cassette. The results provide film images with high contrast, resolution, and speed characteristics with a minimum of geometrical unsharpness (collimator technique was 8:1 λ/d).

The film (Kodak 2496 RAR) is especially sensitive to the artificial light produced by the P20 phosphor in the neutron scintillation screen. The film has a resolving power of 136 to 225 lines/mm, with a relative CRT speed of 320. The photo recording sensitivity is 125 with a speed value of 80. Figure 1 presents initial data regarding integrated neutron dose versus film density. A copy of a typical radiograph taken with this technique appears in Figure 2 with a description of the object.

At this point a measure of the inherent lack of sharpness is given by the imaging of a 0.010-in. hole in a 0.020-in. thick cadmium object placed 0.100-in. away from the image screen. A characterization of "unsharpness" as a function of collimator technique and object situation will be dealt with in the application studies.

This type of imaging technique is considered a pre-requisite to application studies because the exposure time is reasonably short (<1 hr) for high resolution work. A comparative speed for this technique can be judged by a comparison to another direct image technique such as double gadolinium (Gd) screens. The scintillation technique requires less than

FIGURE 1. Exposure Curve for Li⁶F ZnS(Ag) Neutron Scintillation Screen with Special RAR Kodak Film

1. Cadmium strip 0.250 in. wide, 0.020 in. thick, 0.100 in. from film.
 - a. Hole ~0.130 in.
 - b. Hole ~0.070 in.
 - c. Hole ~0.020 in.
 - d. 0.010 in. hole, not visible because of poor reproduction.
2. Same object as No. 1, located 0.200 in. from film.
3. Same object as No. 1, located 0.500 in. from film.
4. Same object as No. 1, located 0.750 in. from film.
5. Same object as No. 1, located 1.200 in. from film.

FIGURE 2. Neutron Radiograph of Cadmium Test Object

0.02 of the exposure that a double Gd screen technique would require for the same film density.

Although the Gd produced image would be sharper, the $\text{Li}^6\text{F ZnS(Ag)}$ produced image is considered sufficient for most applications.

The vacuum cassette reduces film fogging induced by the scattered light resulting from an air space between screen and film. By cooling the lithium screen with CO₂, reciprocity law failure could be avoided and image quality could be improved.

Neutron Camera Containment Vessel

General

The camera is constructed in a modular manner so that modification can be made for shielding, moderation, and collimation. The camera consists of an egg shaped laminated fiberglass outershell, two internal metal drums, shielding and moderator materials, adjustable metal holder for collimators and moderators, and an electric motor drive/controller unit. (See Figure 3)

The camera is designed so that a ²⁵²Cf source,* when positioned within the inner drum, may be rotated, by remote control or manually, to two lockable positions. One position will allow a neutron beam to be selectively moderated and radiographs produced (Pos. 1, Figure 3), the other position is for storage of the source in a shielded condition (Pos. 2, Figure 3).

Fiberglass Characteristics

The fiberglass material used for the construction of the camera case has several advantages over typical materials such as Type 302 SS. This material was selected in part because of its light weight, self extinguishing fire characteristics, ease of molding, cheap cost, and durability.

Table 5 outlines many of the advantages of the use of fiberglass over conventional materials which summarizes the

* See asterick on Figure 3.

FIGURE 3. Sketch of Neutron Photography Camera

general reasons for the choice of this particular material. Other considerations were design cost/performance comparison between standard materials for radioactive containment vessels and fiberglass. These results are summarized in Tables 6 and 7.

TABLE 5. Physical Properties of Fiberglass

<u>Mechanical or Physical Property</u>	<u>Data Calculated</u>	<u>Measured (NC-1)</u>
Tensile Strength	30×10^3 psi	32×10^3 psi
Compressive Strength	30 to 50×10^3 psi	
Flexural Modulus	1.2×10^6 psi	
Specific Gravity	1.5 g/cm^3	
Flammability	Self Extinguishing	

A comparison of the fiberglass material to an equal thickness of Type 302 SS alloy is given in Table 8.

TABLE 6. Fabrication and Handling Advantages of Fiberglass

<u>Fiberglass Advantages</u>	<u>Trans- portation</u>	<u>Materials Handling</u>	<u>Protective Covers and Housings</u>	<u>Aerospace and Mili- tary Uses</u>
Non-corrosive	X	X	X	X
Less Finishing	X		X	
Electrical Properties		X	X	X
Accurate Detail				X
Molded-in-color			X	
Molded in Attach- ments	X	X	X	X
Molded in One Piece	X	X		X
Dimensional Stability				X
Light Weight	X	X	X	X
Low Maintenance	X	X	X	X

TABLE 7. Comparative Cost of Fiberglass and Stainless Steel

<u>Material</u>	<u>Thick- ness, in.</u>	<u>Tensile Strength</u>	<u>Material Cost, ¢/lb</u>	<u>Density, lb/in.³</u>	<u>Space Filling, ¢/in.³</u>
Fiberglass	0.036	28×10^3 psi	31	0.055	1.7
Type 302 SS	0.033	30×10^3 psi	57	0.290	16.5

The slight disadvantage in physical characteristics of the fiberglass was countered by using a laminate 0.500 in. thick. Table 8 presents calculated equivalent thicknesses for Type 302 SS and fiberglass for various normalized physical properties.

TABLE 8. Equivalent Thicknesses of Fiberglass and Stainless Steel

<u>Physical Property, normalized</u>	<u>Thickness of Fiberglass, in.</u>	<u>Equivalent Thickness of Type 302 SS, in.</u>
Tensile strength	0.500	0.458
Bending strength	0.500	0.494
Stiffness	0.500	0.199
Tensile elongation	0.500	0.299

The directional strength properties of glass cloth is another of the important characteristics of fiberglass. The charts in Figure 4 illustrate this. Note that strengths are greatest at 90 and 180 degrees, dropping to the lowest point at 45 degrees. This indicates that in design, reinforcement should be parallel to maximum stress.

In the design of the NC-1 camera, an egg shape (Figure 3) was considered the most practical approach so as to approximate conditions where the strengths would always be the greatest during compression and tension; i.e., impact forces generated by a fall or collision.

FIGURE 4. Properties of Laminated Epoxy Glass

The fatigue nature of epoxy glass cloth laminate is satisfactory for most applications. A laminate such as used for NC-1 should withstand 10^7 cycles at a load not exceeding 1/5 of its ultimate strength, providing that the laminate was not crazed on the initial application of the load. Table 9 presents comparative data on specific fatigue strength at 10^7 cycles.

TABLE 9. Fatigue Properties

	Specific Fatigue Strength, <u>lb/in.² per lb/in.³</u>
Epoxy-glass cloth laminate	240,000
Steel 18-8	180,000
Titanium 150-A	70,000

The neutron device (NC-1) can be shipped under normal ICC shipping regulations as outlined by the Code of Federal Regulations (10CFR). The device meets the requirements for surface radiation dose rates.

As can be interpreted from the above data, the device is a soundly engineered piece of hardware that has good physical characteristics. To date, use of the camera for radiography has resulted in no malfunctions in more than 300 exposures or trials.

The device will continue to be evaluated for reliability as well as conformance for use as a Special Permit Container, 10CFR. The neutron camera will be even more versatile if it conforms to the Special Permit Container class of requirements.

Areas of Applications - NC-1 Camera

There are numerous problem areas where a portable source for neutron radiography would be useful. These can best be summarized in tabular form as given in Table 10.

TABLE 10. Application Areas

<u>Area</u>	<u>Conditions of Use</u>
• Reactor Fuel Radiography	Hot-Cell, In-Service Use
• Forensic Science	Laboratory and Field Use
• Radioactive: Heat and Radiation Sources	Hot-Cell, In-Service and Laboratory
• Radiation Shielding Materials	In-Service, Field Use
• Rocket and Missile Technology (Aerospace)	On-Line Inspection, In-Service
• Physics and Chemistry Studies	Laboratory

A few of the material problems associated with the above areas include enrichment determination, material identification, material quantity measurement, homogeneity, voids, cracks or other material defects and intermaterial diffusion states.

Classes of materials that are used in a few of the above applications include: fuels (uranium and plutonium), poisons (gadolinium and boron), radioactive sources (e.g., americium), fiber composites (high H^* materials with loaded filaments), and light Z^{**} materials in heavy Z containers.

Studies are in progress using the NC-1 camera for evaluating some of the above materials and others in determining exposure curves, resolution capability, as a function of material thickness and shape as well as material identification and

* H relates to hydrogen content of materials.

** Z relates to atomic number of materials.

quantity measurement. The data obtained will be plotted for use with any neutron source provided the thermal to epithermal ratio, "unsharpness" and contrast limits of the radiographic system as well as the flux is a known quantity.

The feasibility of defining (experimentally and theoretically) the modulation transfer function for U_g (radiographic unsharpness) and contrast for the optimum use of NC-1 is being considered. This quantity (MTF) could then be used in determining the radiographic feasibility for evaluating a given application prior to actual radiography.

Specific Applications for Use with Concrete Polymer Evaluations

Neutron radiographs have been made of concrete polymer samples supplied by Brookhaven National Laboratory. These radiographs are being evaluated with a scanning densitometer to obtain integrated density measurements. Preliminary results have been obtained on two samples; one dried at 100 °C with no polymer and the other containing 5% polymer. The results indicated that the sample containing a minimum hydration level with no polymer versus 5% polymer resulted in a 0.20 (H&D units) change in film density.

A complete evaluation is underway at present on these samples.

Conclusion

Several conditions as outlined in the introduction have been fulfilled. Those areas relating to the actual development and evaluation of NC-1 as a portable neutron source have been completed.

The areas associated with application studies are under evaluation. The concrete polymer evaluation indicates that the results on the differences in neutron attenuation as

imaged radiographically can provide specific data as to polymer content and distribution within a given sample. A separate and final report will be prepared regarding this study.

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ADVANCED SOURCE DEVELOPMENT

J. H. Jarrett

Select alpha-photon sources prepared in the previous quarter were evaluated. A photon source preparation run was completed. In this run 25 sources were made at one time. Deposition rate information and information on $^{241}\text{AmO}_2$ deposition uniformity from source to source was obtained. Five of these photon sources were sent to Oak Ridge National Laboratory for testing. The preparation of 25 alpha-photon sources in a single run was started and work on determining source activity of MCi amounts of $^{241}\text{Am}/\text{cm}^2$ was initiated. A review and evaluation of literature on the effects of alpha-radiation on candidate radiation source materials was started.

EVALUATION OF ALPHA-PHOTON SOURCES PREPARED IN THE PREVIOUS QUARTER

In the report for the previous quarter (BNWL-1308-1) several methods of preparing radiation sources by using sputtering techniques were described; and the experimental work conducted in this area at PNL was reviewed. One of the most promising source fabrication methods demonstrated involved sputtering a copper- $^{241}\text{AmO}_2$ cermet layer onto a copper substrate, then sputtering a layer of copper over the cermet, and finally sputtering a stainless steel protective layer on the top and bottom surface of the resulting radiation source. Several alpha-photon sources of this description were prepared last quarter in a single run. The sources showed both alpha and photon activity.

One of the sources has been examined metallographically. The source cross-section was examined at 500X magnification. All layers appeared to be well-bonded to one another, and each layer was quite uniform in thickness. Layer thicknesses

measured from the 500X cross-sections, and rf-sputtering deposition times were combined to give deposition rates. This information is summarized in Table 11.

TABLE 11. Metallographic Examination of $^{241}\text{AmO}_2$ Alpha-Photon Source

<u>Layer Identification</u>	<u>Deposition Time, hr</u>	<u>Layer Thickness (from 500X picture)</u>	<u>Deposition Rate, μ/hr</u>	<u>Forward rf-Sputtering Power, W</u>
Cu substrate	--	~ 80 mil	--	--
$^{241}\text{AmO}_2$ -Cu	2.25	5.0 μ	2.2	800
Copper	4.0	8.1 μ	2.0	800
Stainless Steel	3.0	1.5 μ	0.5	800

The effect of soaking one of these sources in water was investigated. A source was sectioned, half of it was placed in tap water at ambient temperature, and the other half was stored in air at ambient temperature. After thirty days each section, upon visual examination, looked the same, and had an appearance quite similar to that at the start of the test. The water soak did not appear to degrade the source section.

Another one of the sources was placed in tap water in an ultra-sonic cleaner for 10 min and did not appear to be affected adversely. This source has been stored in tap water at ambient temperature since early February and it still appears to possess good integrity.

PREPARATION OF PHOTON SOURCES

A photon source preparation run in which 25 sources were made at one time has been completed. The source design was generally similar to the previously discussed successful alpha-photon sources, except that the layers covering the $^{241}\text{AmO}_2$ -copper layer were made thick enough to shield out the

alpha radiation. The properties of one of the sources that was sectioned and examined metallographically are summarized in Table 12. The deposition rate information was determined from the layer thicknesses and layer deposition times.

TABLE 12. Metallographic Examination of $^{241}\text{AmO}_2$ Photon Source

<u>Layer Identification</u>	<u>Deposition Time, hr</u>	<u>Layer Thickness (from 500X picture)</u>	<u>Deposition Rate, μ/hr</u>	<u>Forward rf-Sputtering Power, W</u>
Cu substrate	--	30 mil	--	--
$^{241}\text{AmO}_2$ -Cu	16.0	25 μ	1.6	400
Copper	9.25	24 μ	2.6	800
Stainless Steel	6.0	5.3 μ	0.9	800

The use of the rf-sputtering system to prepare 25 photon radiation sources at one time was demonstrated in this run. In the rf-sputtering system material is deposited from a 5-in. diameter target onto a 5-in. diameter substrate. Uniform ($\pm 5\%$) deposition may be expected in an area of the substrate 3-1/2-in. in diameter, leaving a 3/4-in. wide band of less uniform deposit around the circumference of the 3-1/2-in. diameter uniform deposit. In order to make maximum use of this area of uniform deposition in the photon source preparation run, 25 numbered copper substrate plates, 5/8-in. on a side, were placed on the 5-in. diameter substrate holder in a square grid 3-1/8-in. on a side. A square mask containing 25 1/2-in. diameter holes was placed on the grid with each hole centered over one of the 25 square copper substrate plates. Subsequent deposition of the copper- $^{241}\text{AmO}_2$ cermet layer gave 25 squares of copper substrate, each with a 1/2-in. diameter area of cermet centered on its surface. The mask was next removed and the copper containment layer was deposited. The stainless steel protective layer was deposited on the

copper layer. A stainless steel layer was also deposited on the bottom of each source. The hardware used for preparing 25 sources in a single rf-sputtering run is shown in Figures 5 and 6.

Gamma radiation measurements were made on each of the 25 sources to get an indication of $^{241}\text{AmO}_2$ uniformity. This was done with the sources in place in the substrate holder (Figures 5 and 6), after the $^{241}\text{AmO}_2$ -copper layer had been deposited, but before the next (copper) layer was deposited. The measurements were made through a 1/8-in. diameter hole in a 3/8-in. thick copper plate so that radiation from adjacent sources would not significantly affect the radiation readings. The readings are shown in Table 13. The factors that influence uniformity readings between sources are rf-sputtering deposition uniformity across the substrate, uniformity of $^{241}\text{AmO}_2$ as mixed in the copper- $^{241}\text{AmO}_2$ cermet sputtering target, and possible errors involved in the radiation measurement system. Good $^{241}\text{AmO}_2$ uniformity from source to source is demonstrated. This is especially true for the middle block of nine sources where all source readings range from 39 to 42 k counts/min with an average of 40.3 k counts/min. Seventeen out of the 25 sources have readings from 35 k counts/min to 42 k counts/min with an average reading of 38.9 k counts/min.

Five of the photon sources from this run (Sources 8, 9, 14, 18, 19, Table 13) have been sent to Oak Ridge National Laboratory for testing and evaluation in their Source Safety Testing Program. The general intent of the testing is to determine the source behavior under increasingly strenuous test conditions until a significant amount of $^{241}\text{AmO}_2$ is released from the source. Americium release will be monitored as the sources are subjected to tests in three different areas: heating in flowing air, leeching in hot water, and heating in humid air. As-prepared and purposely damaged sources will be tested.

Neg 701117-3

FIGURE 5. Rf-Sputtering Hardware for Preparing 25 Radiation Sources in a Single Run

Neg 701117-2

FIGURE 6. Assembled rf-Sputtering Hardware for Preparing 25 Radiation Sources
in a Single Run

TABLE 13. $^{241}\text{AmO}_2$ Uniformity

Background Reading	0.7				
Source Number	1	2	3	4	5
Instrument Reading	30	28	34	36	32
Source Number	6	7	8	9	10
Instrument Reading	24	39	39	41	36
Source Number	11	12	13	14	15
Instrument Reading	37	40	41	42	38
Source Number	16	17	18	19	20
Instrument Reading	34	39	42	40	38
Source Number	21	22	23	24	25
Instrument Reading	32	37	39	38	33

NOTE: The instrument readings are in k counts/min, as measured with an Eberline count rate meter, model BNW-1, having a mica window GM probe. They represent direct readings and are uncorrected. The source to probe distance was about 1/2-in. and the radiation was collimated by a 1/8-in. diameter hole in a 3/8-in. thick copper plate.

EVALUATION OF ISOTOPE PRODUCTION IN POWER REACTORS

R. W. McKee and S. E. Nunn

PLUTONIUM-238 COST STUDIES - THE EFFECT OF NEUTRON FLUX AND COST

A revised series of parametric cases have been prepared to show the range of costs that can be anticipated for ^{238}Pu produced by neptunium target irradiation in a pressurized water power reactor. The calculations were made using the TAIRCOST program and, as in previous comparisons, costs were calculated for high and low processing cost cases to represent the range of uncertainties. Cost is used here in the sense that it represents cost to a user of ^{238}Pu and is calculated as the price required by a producer of ^{238}Pu to recover his costs and realize a normal profit. In all cases, the cost calculations represent costs optimized with respect to both irradiation time and target densities.

In previously reported results, a revenue-loss cost factor was included in the high-cost cases to represent the cost of displacing a fuel rod with a target element. Since it is unlikely that this would be done, the revenue-loss component has been deleted from these calculations. The target elements are assumed to be inserted in either unused control rod locations or some other location that does not displace a fuel rod. The result has been to reduce the spread between the high- and low-cost cases from around \$150/g ^{238}Pu to about \$100/g ^{238}Pu .

Results are summarized in Figures 7, 8, and 9 with ^{238}Pu costs shown as a function of neptunium cost. The low- and high-base processing costs are shown as solid lines and are the same for all three figures. The basic cost assumptions are tabulated on Figure 7.

Figure 7 illustrates the effect of process losses over a range from 1% to 5%. The losses refer to a percentage of both

FIGURE 7. Cost Ranges with Effect of Target and Product Losses for Optimized ^{238}Pu Production in a Pressurized Water Power Reactor

FIGURE 8. Cost Ranges with Effect of Inventory Changes for Optimized ^{238}Pu Production in a Pressurized Water Power Reactor

FIGURE 9. Cost Ranges with Effect of Reprocessing Time for Optimized ^{238}Pu Production on a Pressurized Water Power Reactor

the neptunium target and the ^{238}Pu product. It is anticipated that process losses should not be greater than 1 to 2%.

Figure 8 shows the effect of inventory charges (rate of return) over a range from 5% to 15%/year. This is a before tax value and the base 10% value is on the low side of current electric utility requirements.

Figure 9 shows the effect of reprocessing time over a range from four months to 12 months. The base 4-month value is about the minimum feasible time and would require using at least three reactors with staggered reload schedules for recycling the neptunium targets. A 12-month reprocessing time is inherent if only a single reactor operating on an annual refueling schedule is used.

Isotope Data Compilations

R. W. McKee, S. E. Nunn, and D. R. Haffner

A series of isotopic property compilations is being prepared using data stored in the Battelle-Northwest Isotopic Data tape system. This system contains selected data on the properties of approximately 1600 isotopes and was designed to permit surveying for isotopes with specified properties, but could be used for a general ordering of isotopes by properties.

The Isotopic Data Tape system is stored on several magnetic tapes and in order to provide the required compilations as complete lists, a method for consolidating the data into a single system was required. After reviewing the requirements for data sorting and output formats, it was decided to use Computer Sciences Corporation's general purpose 1108 DRUM SORT routine for sorting the data from the Isotopic Data Tape rather than using the sort routine in the Data Tape system itself. Additional programming was required either way, but use of the DRUM SORT routine appeared the most expeditious method.

The various tables being prepared include all nuclides by Z and N numbers; all nuclides by decreasing half lives; alpha, beta, positron, K-capture and isometric transition isotopes by decreasing half lives; all nuclides by decreasing alpha, beta, positron and photon decay energies; all nuclides by decreasing fission yeilds, and all nuclides by decreasing thermal neutron absorption cross-section.

RADIATION CHARACTERISTICS OF CIRCULATORY
SUPPORT ISOTOPIC HEAT SOURCES

F. T. Cross and J. C. Sheppard

SHIELDING AND IN-PHANTOM DOSE PREDICTION

The shielding and in-phantom dose predictions for LASL-1 continue. Also, the description of the computer codes and ^{238}Pu in-phantom dose prediction program was begun for eventual inclusion in the annual report.

IN-PHANTOM DOSIMETRY MEASUREMENTS

Calorimetric measurements indicate the heat output of LASL-1 to be 29.034 ± 0.063 (2σ) watts on 1-30-70.

Measurements of the neutron dose rates from LASL-1 placed within the homogeneous phantom were started. These will be compared with, and are expected to corroborate, the previous measurements with ^{252}Cf . Figures 10 and 11 represent the total ($n+\gamma$) dose rates measured within and surrounding the Remab phantom from LASL-1 placed in the indicated position. The external dose rates were measured in the bisecting radial plane of the source and, therefore, represent maximum values. The internal dose rates were measured along the central axis of the Remab phantom. The regions of various internal organs are indicated for reference only. Their dose rates may be inferred, especially for those organ systems juxtaposed to the central axis of the body.

^{146}Pm BURNOUT BY NEUTRON RADIATION

J. C. Sheppard

The $^{147}\text{Pm}_2\text{O}_3\text{-Al}$ targets, prepared from Shippingport promethium, were irradiated in a Savannah River reactor in an effort to reduce the ^{146}Pm concentration by taking advantage of the $^{146}\text{Pm}(n,\gamma)^{147}\text{Pm}$ reaction. After about 13 months of

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FIGURE 10. Total (n+γ) Dose-Rates in mrad/hr in Bisecting Radial Plane of Source*

* Excluding ^{236}Pu Contribution

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FIGURE 11. Total (n+γ) Dose-Rates in mrad/hr Along Central Axis of Remab Phantom*

* Excluding ^{236}Pu Contribution

cooling to allow most of the ^{148}mPm to decay away the targets have been analyzed for their ^{146}Pm , ^{148}mPm , and ^{147}Pm content to determine the $^{146}\text{Pm}/^{147}\text{Pm}$ ratio. The results are summarized below.

Results of Savannah River Irradiation

<u>Sample</u>	<u>Initial</u>	<u>A-1</u>	<u>A-2</u>
Integrated flux, n/cm^2	0	7.7×10^{20}	14.7×10^{20}
$^{146}\text{Pm}/^{147}\text{Pm}$ (a)	3.90×10^{-6} (b)	3.40×10^{-6} (c)	2.21×10^{-6} (c)

- a. $(\text{d/s of } ^{146}\text{Pm})/(\text{d/s of } ^{147}\text{Pm})$
- b. Value at start of irradiation.
- c. Value at time of discharge from reactor.

The $^{146}\text{Pm}/^{147}\text{Pm}$ ratios are much higher than expected and indicate, once again, the importance of the $^{147}\text{Pm}(\text{n},2\text{n})^{146}\text{Pm}$ and $^{147}\text{Pm}(\gamma,\text{n})^{146}\text{Pm}$ back reactions. The former reaction is a function of the hardness of the neutron flux and the latter is related to the cladding material used. It is not known which of these contributions is dominant, but the Hanford experiments indicated that the $^{147}\text{Pm}(\gamma,\text{n})^{146}\text{Pm}$ was important because of the massive aluminum targets used. The Savannah River targets were a $\text{Pm}_2\text{O}_3\text{-Al}$ matrix clad in aluminum so the $^{147}\text{Pm}(\gamma,\text{n})^{146}\text{Pm}$ reaction was probably important again. Graphite targets could probably eliminate this contribution. The $^{147}\text{Pm}(\text{n},2\text{n})^{146}\text{Pm}$ reaction will still be important and can only be minimized by performing the irradiation in a well moderated reactor.

A report of the Savannah River ^{146}Pm burnout experiments is being written.

NUCLEAR TECHNIQUES FOR SEABED MINERAL EXPLORATION

R. W. Perkins and N. A. Wogman

Rapid nondestructive mineral analyses can be performed by the combined use of a ^{252}Cf neutron source and a Ge(Li) diode gamma-ray spectrometer. ^{252}Cf , which has only recently become available,⁽¹⁾ decays by spontaneous fission with a half-life of 2.5 years and emits 2.34×10^9 neutrons/sec-mg.⁽²⁾ This quantity also provides a thermal neutron flux of about 10^7 neutrons/cm²-sec at a distance of a few centimeters in a water moderator. Although this flux is about one millionfold below that available from nuclear reactors, it is sufficient for laboratory mineral analyses. This potential is being evaluated for in situ mineral exploration through development of a laboratory seabed mineral analyzer and a prototype underwater mineral exploration system.

The final design of a laboratory seabed mineral analyzer prototype has been completed (see Figure 12). This system had a scheduled delivery date of April 15, 1970. Equipment is being fabricated for transfer of the source (2 mg of ^{252}Cf) from its Oak Ridge National Laboratory shipping capsule for placement in the Battelle-Northwest assembly. The prototype system will determine the efficiency of many possible irradiator-detector arrangements which could be used for actual seabed mineral analyses. The ^{252}Cf source will be located in paraffin or polyethylene moderators approaching 24-in.-diameters and heights. This system is located in the bottom of a 24-in.-diameter by 48-in.-high polyethylene tank filled with seawater. This tank system is surrounded by a 48-in.-diameter by 48-in.-high tank of seawater which is subsequently located in a 69-in.-diameter by 72-in.-high biological shield. Mineral specimens to be analyzed are held in a 22-in.-diameter by 10-in.-thick container for a specific irradiation. Following the irradiation

exposure, the sample is removed rapidly from the irradiator and placed in a Ge(Li) mineral analyzer system located in a similar seawater assembly. The diode system provides a final gamma-ray analysis.

In Table 14 the elemental concentrations are listed which are required to permit their measurement in various minerals with a precision of $\pm 3\%$ to $\pm 15\%$. The radionuclides which serve to identify and quantitatively measure each element are listed, along with their half-lives, with the element they identify. The sensitivities are based on the use of a 1 mg ^{252}Cf source in the prototype system previously discussed utilizing a 5 kg mineral sample, an 80 cm^3 Ge(Li) detector, and irradiation and analysis times of 2 min each. If a tenfold larger ^{252}Cf source were employed, these sensitivities could be obtained with a sample ten- to twentyfold smaller.

An irradiation and detector assembly for seabed mineral exploration evaluation has also been designed (see Figure 13). The operation of the system envisions the irradiation of a specific ocean floor area with neutrons, followed by direct analysis of the field area with a Ge(Li) detector system. A procedure for cooling the Ge(Li) detector assembly at depth has been developed. This procedure, which uses a high-pressure Dewar and solid propane, allows remote use for approximately 24 hr.

In situ mineral exploration of the ocean floor is feasible and can provide a rapid and practical means of establishing the elemental content of the ocean and freshwater sediments. Actual in situ analyses for terrestrial mineral exploration are also possible by use of a probe consisting of a ^{252}Cf source irradiation, followed by a Ge(Li) detector analysis. A similar approach could be employed on a roving vehicle for unmanned planetary mineral analyses.

**TABLE 14. Sensitivities for Measurement of Elements
in Representative Minerals by a Rapid ^{252}Cf
Irradiation and Direct Counting**

Element	Mineral	Radionuclide Daughter(s)	Half-Life	Detectable Concentrations ^(a)				
				ppm		%		
				1-10	10-100	0.01-0.1	0.1-1	1-10
Aluminum	all minerals	^{28}Al	2.30 min				X	
Antimony	stibnite	^{122}Sb	2.8 day					
		$^{122\text{m}}\text{Sb}$	4.2 min					X
		$^{124\text{m}}\text{Sb}$	21 sec					
Arsenic	arsenopyrite	^{76}As	26.5 hr				X	
Barium	witherite	$^{133\text{m}}\text{Ba}$	38.9 hr					
		^{139}Ba	83 min				X	
Cobalt	cobalite	$^{60\text{m}}\text{Co}$	10.5 min				X	
Copper	chalcopyrite	^{64}Cu	12.8 hr				X	
		^{66}Cu	5.1 min					
Dysprosium	gadolinite	^{165}Dy	2.35 hr		X			
		$^{165\text{m}}\text{Dy}$	1.26 min					
Fluorine	gadolinite	^{20}F	11 sec			X		
Gold	quartz	^{198}Au	64.8 hr		X			
Indium	chalcopyrite	$^{116\text{m}}\text{In}$	54 min	X				
Magnesium	magnesite	^{27}Mg	9.5 min				X	
Manganese	all minerals	^{56}Mn	2.58 hr			X		
Mercury	cinnabar	^{205}Hg	5.2 min					X
Niobium	columbite	$^{94\text{m}}\text{Nb}$	6.3 min					X
Molybdenum	molybdenite	^{101}Mo	14.6 min			X		
Scandium	scheelite	$^{46\text{m}}\text{Sc}$	20 sec	X				
Selenium	chalcopyrite	$^{77\text{m}}\text{Se}$	18 sec	X				
Silver	argentite	^{108}Ag	2.4 min		X			
		^{110}Ag	24 sec					
Strontium	witherite	$^{87\text{m}}\text{Sr}$	2.8 hr				X	
Titanium	rutile	^{51}Ti	5.80 min				X	
Tungsten	sheelite	$^{183\text{m}}\text{W}$	5.3 sec				X	
		^{187}W	24 hr					
Vanadium	any mineral	^{52}V	3.77 min		X			

a. ^{252}Cf source of 1 mg
Sample weight of 5 kg
Irradiation time of 2 min
Counting time of 2 min
Ge(Li) diode size of 80 cm^3 and resolution of 3 keV
Expected precision at the stated detectable concentrations is ± 3 to 15%.

^{252}Cf IRRADIATOR Ge (Li) MINERAL ANALYZER

FIGURE 12. ^{252}Cf Neutron Irradiation Facility (Laboratory Prototype for in situ Seabed Mineral Analysis).

OPERATION

1. SUB OPERATOR POSITIONS IRRADIATOR-DETECTOR ASSEMBLY ON OCEAN FLOOR.
2. OPERATOR INITIATES START COMMAND.
3. STEPPING MOTOR ROTATES IRRADIATOR-DETECTOR ARM TO POSITION OPPOSITE OF THAT SHOWN.
4. FLOOR AREA IS IRRADIATED FOR ONE MINUTE.
5. STEPPING MOTOR ROTATES ARM TO POSITION SHOWN.
6. A SPECTRUM IS COLLECTED FOR ONE MINUTE IN A COMPUTER MEMORY.
7. THE COMPUTER COMPARES THE SPECTRUM OBTAINED WITH CHARACTERISTIC ORE SPECTRA ON FILE AND READS OUT TYPE AND GRADE OF ORE PRESENT.
8. OPERATOR PROCEEDS TO NEXT STATION.

Neg. 700335-1

FIGURE 13. Proposed Experimental Arrangement of ^{252}Cf Source and Ge(Li) Detector for Seabed Mineral Exploration

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