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PACIFIC NORTHWEST LABORATORY
MONTHLY REPORT ON ISOTOPES
DEVELOPMENT PROGRAMS
FOR JUNE 1972

JULY 17, 1972

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MONTHLY REPORT
ON ISOTOPES DEVELOPMENT PROGRAMS
FOR JUNE 1972

Compiled by J. H. Jarrett

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July 17, 1972

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08-01. RADIOISOTOPES PRODUCTION AND
MATERIALS DEVELOPMENT

08-01-03. FISSION PRODUCTS

I. LOW-COST CESIUM AND STRONTIUM SOURCES -

H. H. Van Tuyl and D. H. Siemens

The objective of this program is to provide technology for the production of inexpensive Cs and Sr sources by using the purified compounds generated in the Hanford Waste Management Program (HWMP). Large-scale use of these radioisotopes will require an assessment of the capabilities of HWMP for producing suitable radiation and heat sources, acceptance of the HWMP product by the customer, licensing of the sources, and market development activities.

The goal in FY-1972 is to develop equipment for fabricating smaller diameter sources in the HWMP facility. The incremental cost of producing smaller sources will be evaluated. Prototype sources will be fabricated with nonradioactive feeds, including loading capsules, welding, and NDT. The sources will be tested to determine mechanical integrity. Potential customers will be contacted to stimulate interest in the HWMP sources.

Approximately 60 kCi of ^{137}Cs was received as a partially purified crude from ARHCO. This material was purified in C-cell for use in fabricating two prototype sources. Purification was accomplished by absorption on

a zeolite ion exchanger, washing to remove sodium, and elution with ammonium carbonate. Because of the low purification equipment capacity, four batches were required. The cesium product is being concentrated and will be converted to the chloride for source fabrication.

II. UTILIZATION OF FISSION PRODUCTS Rh, Pd, Ru, and Tc -

R. H. Moore, F. P. Roberts and J. H. Jarrett

The traditional view of fission products as waste materials has been supplanted by their role as a national resource. Pursuant to this revised status, a resource strategy is now being formulated to direct optimum technological and economic application of the fission products Rh, Pd, Ru, and Tc created in operation of nuclear power reactors. This program seeks to evaluate the potential of these artifacts as supplements to the natural reserve of precious metals, and emphasizes the practical aspects of their safe use. The program comprises the following tasks:

- 1. Evaluation of rhodium and palladium produced in power reactors.*
- 2. Technology for recovery of Rh, Pd, Tc, and Ru.*
- 3. Market analysis of Rh, Pd, Tc, and Ru.*
- 4. Economic analysis of Rh, Pd, Tc, and Ru production from nuclear wastes.*
- 5. Fabrication process development for nuclear source (Rh, Pd, Tc, and Ru) products.*

Based on the purification process outlined in the May monthly report (BNWL-B-182), preliminary estimates were made of the capital equipment and operating costs for the purification of these fission product materials. As would be expected, the cost of the hot cell or canyon facility for the processing equipment is the major capital cost. The major operating expense is for the needed process materials. Storage costs and the effect of the cost of capital are being estimated.

III. EVALUATION OF LARGE-SCALE USES FOR PURIFIED FISSION PRODUCT

XENON - C. A. Rohrmann

Of all the fission products, Xe has the highest fission yield. Although many of the Xe isotopes are radioactive, they all have such short half-lives that their activity would be insignificant after allowing time for normal fuel reprocessing.

In the past, the extreme rarity of Xe in nature has clearly inhibited its consideration for specific large-scale uses even where its properties give it definite and unique advantages. But now, Xe could be made available in surprisingly large quantities. Perhaps the most important and beneficial use of Xe would be for human surgical anesthesia. Another of the potential uses is as a filler gas in incandescent light bulbs with significant increase in both brightness and efficiency.

The scope of the PNL program is to: (1) establish the quality of Xe which can be accepted or generally licensed for consumer uses, (2) determine what industries would be interested in evaluating fission product Xe for large-scale application, (3) distribute specimens to participating evaluators, (4) compile evaluation results, and (5) prepare a final report on the results of this total effort.

Investigators at the Lawrence Berkeley Laboratory have been provided information on purified fission product xenon as a possibility for use in the liquid xenon proportional chamber they are developing. Information on the use of xenon for human surgical anesthesia was forwarded to Dr. Julius Butler, Harborview Medical Center, Seattle, Washington.

V. STRONTIUM FLUORIDE COMPATIBILITY - H. T. Fullam

Lower cost strontium is possible by using the Hanford Waste Management Program (HWMP) produced fluoride as a fuel form instead of the currently used titanate. Many potential uses of strontium can be made more cost-effective if the HWMP material can be safely and efficiently utilized. The objective of this program is to determine the chemical compatibility and properties of strontium fluoride as produced by HWMP and develop suitable data for strontium fluoride licensing procedures.

The goals in FY-72 are to establish further contacts with regulatory agencies, detail the program work plan, and initiate the procurement of needed equipment and materials.

Efforts during the month have been limited principally to obtaining the materials and equipment needed for the experimental studies which will begin in July. All materials purchased, which will be used directly in the experimental studies, are being obtained using established quality assurance procedures. The laboratory in which the equipment for nonradioactive tests will be located has been obtained and is now being outfitted.

The capsule design which will be used in the initial nonradioactive compatibility tests has been established. The experimental program for FY-73 has been defined, and the program plan will be discussed in late July with Division of Applied Technology (DAT) personnel at AEC Headquarters and with other interested groups. Preliminary review of the proposed program by AEC Division of Materials Licensing was favorable, and a review by Naval Facilities Engineering Command is being made.

The initial laboratory study will involve the compatibility testing of very high purity inert SrF_2 with various containment materials. Most of the supplies needed for the tests have been ordered. Testing will begin as soon as the test capsules have been fabricated and the test conditions firmly established.

08-02. THERMAL APPLICATIONS

08-02-01 CIRCULATORY SUPPORT SYSTEMS -

F. T. Cross and W. K. Winegardner

The main objective of this program is to determine the dose from radioisotopic heat sources suitable for circulatory support systems. This is accomplished by measuring the dose rates within and surrounding various tissue equivalent phantoms.

Part of the program concerns the prediction of the measured dose rates by the development of computational codes. The computer codes are also used in parametric studies and for the prediction of the in-phantom dose rates from sources not available for measurement.

Another part of the program concerns the development of radioisotopic blood heaters for use in an animal study aimed at assessing the combined effects of heat and radiation in Hanford miniature swine implanted with 50W plutonium heat sources.

I. Dosimetry Studies

All measurement data on the photon and neutron dose rates from the $^{238}\text{PuO}_2^{16}$ Rankine prototypic heat source are being analyzed and compared with the dose rate calculations, in preparation for a topical report.

II. Radiological Engineering Studies

Phase I Heaters

Final assembly of the three blood-cooled heat exchangers will be completed upon receipt of the plutonium-238 heat sources.

Phase II Heaters

Work continues on the fabrication of a sham to simulate the Phase II heat source-heat exchanger and associated heat conducting cable.

08-03. RADIOISOTOPE SYSTEMS DEVELOPMENT

08-03-01. APPLIED INSTRUMENTATION DEVELOPMENT

I. Neutron Radiography Using ^{252}Cf -

K. L. Swinth

The objective of this program is to develop and investigate low-flux neutron radiography techniques and applications using ^{252}Cf . Previous efforts have led to the development of a portable shield and exposure facility for ^{252}Cf thermal neutron radiography. Evaluation of this facility is underway and will lead to recommendations for future designs. Applications of ^{252}Cf neutron radiography will be identified and efforts will be extended to fast neutron radiography.

The major effort in the program is development of techniques for low-flux imaging. A position-sensitive proportional counter is under development that provides for instantaneous readout of data representing the imaged object. Various converter foils, such as Eu and ^6LiF thermoluminescent foils, are being investigated as a means of radiographically recording the images.

A 189 in this program area was prepared and submitted to DAT. The program name was modified, at the suggestion of DAT staff, to Low Flux Neutron Imaging. This more accurately reflects the direction the program has taken in FY-72 and the direction anticipated for FY-73 and beyond.

The continuation of work in this program area will contribute directly to increased applications and utilization of ^{252}Cf by developing technology needed to reduce the time and cost associated with obtaining low-flux neutron radiographs.

Information and select publications resulting from this program were forwarded to Dr. L. G. Stang, BNL, at his request.

II. Advanced Source Development -

G. G. Culver and J. H. Jarrett

Advanced Alpha Photon Source Development

The objective of this program is to evaluate present and future heat and radiation source application requirements and to relate them to present and future encapsulation technology.

The goal of the current work is concerned primarily with the development of thin-film encapsulation and evaluation techniques for alpha photon sources. Emphasis will be directed toward the technique of sputtering as a means to obtain thin films of radioisotope and containment materials.

The vacuum station and sputtering chamber for the cold (nonradioactive) sputtering system have been cleaned and

component assembly has been started. Glovebox sputtering system cleaning was initiated. We anticipate having both systems cleaned, assembled and operational by the end of July.

System components for continuous, liquid nitrogen cooling of vacuum station cold traps have been ordered, with delivery and installation expected in July. This facility improvement will help minimize contaminants (such as vacuum pump oil) in the sputtering chambers. Plans are being made for sputtering laboratory modifications that will help control dust and other particulate material in working areas. The purpose of this effort is to minimize loosely adhering foreign particles on radiation source substrates and on radiation sources that are in preparation, thus leading to better thin film radiation sources.

08-03-03. OCEANOGRAPHIC AND NATURAL RESOURCE APPLICATIONS

I. Nuclear Techniques For Seabed Mineral Exploration -
N. A. Wogman and R. W. Perkins

In recent PNL studies of the composition of deep ocean sediments, it has been demonstrated that various mineral deposits and commercial-grade ores can readily be identified by neutron activation and instrumental analysis. From preliminary analysis, it is evident that the elemental composition of most mineral deposits can be easily identified at concentrations well below those of economic importance. In-place experience with ^{252}Cf as a neutron source has confirmed these conclusions. Two methods for in-place seabed mineral exploration are being investigated. The first involves placing a neutron source and detector system on the ocean floor. The second method involves bringing the samples on-board ship and employs a shipboard ^{252}Cf irradiator and a Ge(Li) spectrometer. The former technique

has the advantage of very rapid analyses, and the latter technique has the advantage that mineral depth profiles can be defined from core sampling. Primary emphasis of the program is presently directed toward the design construction, and the laboratory and sea floor calibration of the in-place seabed nuclear probe.

The major effort this past month has continued to center on testing and modifying the nuclear probe system prior to sea trials.

With the pressure housing over the nuclear probe detector and a 1000-ft submarine signal cable, the resolution of the solid cryogen cooled detector is better than 2.5 keV at ^{60}Co . Dry run tests have shown that this resolution is quite adequate for precise elemental determinations. Addition of a new amplifier section did not significantly improve the resolution.

All electronics and electrical controls, including the TV monitor, have been checked out in dry runs. The probe mount and positioning hardware have also been checked out in dry runs. The final procedure which is presently being conducted before sea trials is a dry run to check out the electronics and electrical controls with the positioning hardware as a complete system.

PROMETHIUM INVENTORY - June 30, 1972

Shipments - Through ORNL Isotope Sales	1,643,530 Ci
PNL Available Inventory	14,544,444
Crude Feed	11,227,575 Ci
Pre-Purified	3,175,824
Product - Run 59	53,553
Run 60	41,000
Run 61	<u>46,492</u>
(Subtotal)	14,544,444
Decay	13,039,913
Unaccounted For	1,497,701
Waste	<u>439,780</u>
Disposition of Promethium, Total	<u>31,165,368 Ci</u>
Received from ARHCO	<u>31,165,368 Ci</u>

Pm Inventory Account Activities - D. H. Siemens

Preparation for a promethium purification run, using prepurified feed material held in storage, was initiated on June 12th, and the purification was accomplished during the following week. Sufficient purified material is being converted to oxide to meet the requirements of two orders:

1. 50,000 Ci ¹⁴⁷Pm for Donald W. Douglas Laboratories
2. 25,000 Ci ¹⁴⁷Pm for ORNL.

The final conversion to oxide and the product analytical work will be completed prior to June 30. The shipments will be made during the first week of July.

The current status of the crude ^{147}Pm is as follows:

<u>Tank</u>	<u>Volume,</u> <u>gal</u>	<u>H+,</u> <u>M</u>	<u>Specific</u> <u>Gravity</u>	<u>Temp.,</u> <u>°C</u>	<u>Evaporation</u> <u>Rate, GPD</u>
PT-1	1730	0.50	1.127	59	4.5
PT-2	1925	0.50	1.125	59	4.5

The sump pump in tank T-5 (cell drain catch tank) has failed. The radiation level in Room 40A where the tank is located prevents personnel entry to inspect and/or replace the pump. Short-range plans are to install a positive displacement pump in the Small Transfer Hood and empty the tank through an existing 1/2 in. tubing line. Long-range plans are to reduce the radiation level in Room 40A and repair the existing pump or replace it with a new one.

Production and Shipments:

None.

ATLANTIC RICHFIELD HANFORD COMPANY FISSION PRODUCT ACTIVITIES-
G. L. Borsheim

A telephone request was received from a representative of the Division of Reactor Development and Technology (RDT), AEC Headquarters, in late May for dimensions, tolerances, and purchase specifications associated with the Waste Management ^{90}Sr and ^{137}Cs capsules. This information was to be transmitted to Oak Ridge National Laboratory (ORNL) for their

consideration in multiwatt generator studies. The following pertinent specification and drawing numbers were given to RDT by telephone with the recommendation that AEC-RL be contacted for copies of the information, since the Waste Encapsulation and Storage Facilities (WESF) project is AEC-managed.

HWS-8834, Rev. 1	Specification for Seamless Hastelloy C-276 Tubing for Strontium Fluoride Encapsulation, 225-B Encapsulation Facility, Project HAP-631.
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HWS-8835, Rev. 1	Specification for Seamless 316-L Austenitic Stainless Steel Tubing for Cesium Chloride Encapsulation, 225-B Encapsulation Facility, Project HAP-631.
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Drawing No. H-2-66758	Sr Inner Capsule Assembly and Details
Drawing No. H-2-66759	Sr Outer Capsule Assembly and Details
Drawing No. H-2-66760	Cs Inner Capsule Assembly and Details
Drawing No. H-2-66761	Cs Outer Capsule Assembly and Details

The two specifications show the present correct dimensions and tolerances. However, Drawings H-2-66759, and H-2-66760 require revision to correspond with the specifications. The two strontium capsule drawings will be revised shortly but design details associated with filling of the cesium inner capsule have not been confirmed.

During the last two years ARHCO has documented three studies pertaining to the production of special strontium and cesium sources in the WSEP. The documents and a brief

description of their contents are listed below. Note that these were preliminary studies and did not include integration with current encapsulation plant scheduling.

ARH-1897, Unclassified, "Production of Strontium Sources at ARHCO," G. L. Borsheim, December 11, 1970.

Includes an assessment of the capability and costs to encapsulate up to approximately 5 MCi per year of ^{90}Sr sources as SrF_2 , SrO , and SrTiO_3 in capsules ranging from 1 to 4 in. in diameter.

ARH-2121, Unclassified, "Navy Strontium-90 Information," G. L. Borsheim, M. J. Kirkman, J. D. Moore, June 11, 1971.

Includes cost estimates for the production of 20 MCi of ^{90}Sr as normal $^{90}\text{SrF}_2$ sources and a comparison of cost estimates for four alternative production schemes.

ARH-2101, Unclassified, "Production of Special Cesium Sources at ARHCO," J. D. Moore, May 14, 1971.

Includes cost estimates for the production of various quantities of 1-in. and 1-1/2-in. diameter CsCl capsules.

ARH-1897 was cleared for limited distribution to AEC contractors and AEC offices on August 2, 1971, as the result of a request from AEC Headquarters Library. The other two documents have not been cleared for off-site transmittal.

Preparation of a 189 submission outlining the recommended scope of ARHCO's efforts for FY-1973 and succeeding years was completed and forwarded to AEC-RL in early June.

Analyses of samples of the recent oxalate-carbonate purification test strontium nitrate product now held in the 36-1 tank and of the previously purified strontium nitrate solution held in the 36-3 tank are being reviewed and evaluated. It appears that the rare earth decontamination resulting from the test was slightly less than half of the factor of 6 that had been expected. Review of other isotopic and chemical purity data is continuing.

Discussions pertaining to ^{90}Sr and ^{137}Cs encapsulation were held by ARHCO and ORNL representatives at ORNL's Fission Product Development Laboratory on June 8 and 9, 1972. Representatives of ARHCO and PNL met on June 15, 1972, to discuss the overall DAT fission product program and to determine how ARHCO can best support PNL's current studies on licensing of $^{90}\text{SrF}_2$ capsules to be produced in the WESF. It appears that efforts should be increased to determine the concentrations of trace impurities Pb , $\text{SO}_4^{=}$, NO_3^- , Cl^- , and $\text{CO}_3^{=}$, and $\text{PO}_4^{=}$, which may cause capsule compatibility problems at the higher temperatures to which heat sources would be subjected.

DAT BUDGET STATUS - FY-1972 THROUGH JUNE 30, 1972

	<u>Current Month</u>	<u>FY-1972 CTD</u>	<u>FY-1972 Budget</u>
08-01 <u>Radioisotope Production and Materials Development</u>			
08-01-03 <u>Fission Products</u>			
I. Cesium/Strontium	4.9	60.0	60
II. Rh, Pd, Ru, and Tc Utilization	6.0	26.1	25
III. Xenon Utilization	1.0	11.0	10
IV. Isotope Production Evaluations	0.6	10.0	10
V. SrF ₂ Compatibility	10.4	13.5	17
Subtotal 08-01	22.9	120.6	122
08-02 <u>Thermal Applications Development</u>			
08-02-01 <u>Circulatory Support Systems</u>			
I. & II. Recipient Radiation Exposure and Radiological Engineering Studies	6.4	95.0	95
III. Population Radiation Exposure	20.1	94.9	95
Subtotal 08-02	26.5	189.9	190
08-03 <u>Radioisotope Systems Development</u>			
08-03-01 <u>Applied Instrumentation Development</u>			
I. Neutron Radiography	0.5	25.1	25
II. Advanced Source Development	5.7	34.5	35
III. Cost Benefit Study	20.4	29.7	30
Subtotal 08-03-01	26.6	89.3	90
08-03-03 <u>Oceanographic and Natural Resource Applications</u>			
I. Seabed Mineral Exploration	3.0	60.3	60
Subtotal 08-03-03	3.0	60.3	60
Subtotal 08-03	29.6	149.6	150
Total 08 Operating Budget	79.0	460.1	462
08 Equipment Budget	38.6	136.8	137*
Subtotal 08	117.6	596.9	599
Pm Inventory Account	24.6	282.1	285*
PROGRAM GRAND TOTAL	142.2	879.0	884

* Spent and outstanding commitments

** This includes \$75k authorization for ARHCO to ship new crude feed to PNL.

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