PACIFIC NORTHWEST LABORATORY
MONTHLY REPORT TO THE
DIVISION OF ISOTOPES DEVELOPMENT
FOR SEPTEMBER 1970
Compiled by J. E. Hansen

October 15, 1970

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PACIFIC NORTHWEST LABORATORY
MONTHLY REPORT TO THE
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FOR SEPTEMBER 1970

08-01. RADIOISOTOPES PRODUCTION AND
MATERIALS DEVELOPMENT

08-01-03. FISSION PRODUCTS

I. LOW COST CESIUM AND STRONTIUM SOURCES - H. H. Van Tuyl,
H. C. Stidham, and W. E. Keder

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-1971 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.

A comment draft of the cesium report has been written and is being typed. The conclusions from the report are somewhat tentative since the price of HWMP cesium has not been defined.

(Cost to the AEC for the sources could be determined by engineering calculations, but price must be fixed by the AEC.) Assuming a price of 10 to 15 cents per curie for encapsulated cesium, there is a definite incentive to reduce container diameter and capsule wall thickness.

Most of the recent effort has been devoted to strontium. Currently, any alternative source geometry is limited by the present compactor design; it was designed to handle only waste management capsules. The compactor can be adjusted, however, to handle smaller than standard size capsule configurations if the appropriate shims, plugs, compactor rod and adaptors are fabricated. Loading a capsule that is larger in diameter than the standard size capsule will require redesign of the funnel mechanism, base support and compactor rod. With the larger capsule diameters, the compaction density will decrease to approximately 70% of theoretical density at a 4 in. diameter. A compaction density of 80% of theoretical density is being achieved with the standard sized capsules. Estimates are that the maximum possible density that can be achieved in the standard sized capsule is limited to 85% of theoretical density.

Rough estimates were obtained of the cost of building flexibility into the compactor design. The new compactor will cost in the neighborhood of \$15,000. If the compactor is redesigned at this time to accept diameters larger than the waste management capsules, each set of shims, adaptors and compactor rods will cost approximately \$1,500 for each capsule size. If the existing compactor design is used, a second compactor unit will be required for capsules of larger than the standard sized diameter.

The standard sized container of Haynes 25 metal was originally estimated by ARHCO at \$235 per complete capsule unit. A revised estimate by ARHCO is over \$400 per capsule, based on the precept that Haynes 25 metal tubing costs somewhere around \$100 per linear foot.

A study of alternative source geometries for strontium fluoride capsules has progressed. The centerline temperatures and outer skin temperatures for capsules of varying diameters and densities have been calculated. Calculations were made for capsules with densities that varied in 5% increments between 65% theoretical density and 90% theoretical density. The inner capsule diameter was varied in one-fifth in. increments between two and four in. Computations were based on using Hastelloy C metal, a wall thickness of 120 mils, and a separation of 130 mils between the inner and outer capsule. This large spacing between capsules is a "worst case," with the expected gap being considerably smaller. All calculations used a 12-in. inner capsule length. The thermal conductivity of Hastelloy C is very similar to that of the Haynes 25 metal.

The centerline temperature of the capsule in air will vary from about 800-°C to 1750 °C for 65% of theoretical density at diameters from two to four inches. Higher densities will result in lower centerline temperatures. Additional calculations are being made for other capsule geometries and environments.

The cost for higher density strontium fluoride is under investigation. The current compactor design has achieved slightly over 80% theoretical density using the waste management capsule. The current compactor design will handle only

the standard waste management capsule unit and will require extensive modifications and redesign for handling alternative source sizes.

Literature studies are being made which relate to the problem of effects of expected impurities on the properties of $^{90}\text{SrF}_2$ preparations. The desirable phase diagrams do not appear to exist, but some data with which to estimate solid solutions behavior have been located. The expected impurities appear to be the least troublesome ones. Much of the previous, fundamental work on SrF_2 and its mixtures is very old and not in English.

II. RADIATION CHARACTERISTICS OF FISSION PRODUCT Rh AND Pd

Nothing to report.

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 uses of Xe would be for human surgical anathesia. Another of the potential uses is 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.

Although further purification of the ^{85}Kr contaminated fission product xenon at ORNL is continuing, the use of the facilities there has yielded about 400 liters of material containing between 10 and 25 microcuries of ^{85}Kr contamination per liter of xenon. This is regarded as an encouraging result from equipment normally used for ^{85}Kr production. Further purification runs have been scheduled.

Formal contacts with potential users of the purified material are planned after the quantity and quality of the product can be determined. On the basis of interest developed in formal contacts made over the past year, expressions of interest in sharing in the evaluation of the product will be sought from an estimated twenty-four potential users. These include: (1) medical users, (2) producers of rare gases, (3) potential producers of fission product gas (power reactor fuel processors), and (4) users of rare gases. There also may be others who have rather specialized applications for this gas.

ISOTOPE PRODUCTION EVALUATIONS - R. W. McKee

This program is designed to evaluate major aspects of isotope production. Until recently, the principal source of isotopes has been the AEC

production reactors. Now, however, power reactors are rapidly becoming important sources of isotopes. Power reactors also offer attractive prospects for target irradiations. The future availability and cost of these unique isotopes depend on full and efficient utilization of the production potential of power reactors, and the inherent value of these materials can have a significant impact on nuclear power costs.

The scope of this program includes analysis of fast and thermal reactor fuel cycles within the framework of an expanding nuclear power industry to identify the sources, quantities, timing and cost of production of isotopes of potential importance.

A new task to develop an Isotope Data System was initiated in FY-1971. The objective is a computerized, quick-response system for answering queries on isotope properties, availability, applications, shielding requirements, decay compositions, toxicity, and other factors.

A. Fission Product Profitability Studies

A computer program identified as CAESR was completed and debugged to a workable status. An initial series of cases have been run to explore possible pricing requirements for profitable recovery of fission product cesium and strontium from power reactor fuel reprocessing. The capital cost estimate for the Cs/Sr recovery and encapsulation facilities was received from Vitro, Hanford Engineering Services and used as input for the pricing calculations.

The computer program provides for a market projection calculation and for the reprocessor to capture a specified fraction of the market, which can vary with time. There is a provision for looking at three different startup years

(1975, 1977, and 1980 were used) for each case and three plant sizes for each startup year to permit selection of a near optimum plant size for each set of conditions. Prices are calculated as present-worth levelized prices or the single price that could be charged over the life of the project to pay all costs and recover the initial investment with a specified profit or rate of return.

A basic assumption for the initial cases was that the market would first develop with AEC sales, but the AEC would then withdraw from the market as soon as a private facility was operating. Thus, this first plant would have 100% of the market initially. However, it was further postulated that a competitor would soon be operating and the first plant's share of the market would decline to 50% of the market in five years.

The reference plant capital cost estimate was as follows:

	<u>Capacity</u>	<u>Capital Cost</u>
Cesium only	29 MCi/yr	\$14.3 x 10 ⁶
Strontium only	28 MCi/yr	\$14.6 x 10 ⁶
Cs/Sr Co-Products	29/28 MCi/yr	\$20.5 x 10 ⁶

These estimates include a 20% escalation factor on a 1970 base estimate. This may be optimistic considering that construction costs are currently increasing at a rate of 10%/year.

Other basic input assumptions were:

Return on investment	15%/yr
Economic life	10 years
Income tax rate	50%
Depreciation method for taxes	Sum of years
Design and construction period	4 years

For market projections of 15 to 25 MCi/yr of Cs-137 in 1980 (current market is about 1 MCi/yr), the calculated price requirements ranged as follows:

Startup Year	1975	1977	1980
Price, ¢/Ci Cs-137	35-70	25-50	20-35

The results for Sr-90 were similar.

B. Isotope Data Compilations

Work continued on updating the "Nuclide Tables" data file.

Two requests from AEC laboratories were filled for copies of the existing isotope data file (computer tape).

08-01-04. SOURCE DEVELOPMENT

I. NONDESTRUCTIVE TESTING OF SOURCES - D. R. Newman

A. NDT Research and Development Support

The scope of this program is to develop methods of nondestructive examination of isotopic heat sources developed by DID contractors. Through a number of programs, DID is developing applications for various radioisotopes as heat sources for specific applications. The PNL program will concentrate on the development of methods to assure the long-term mechanical and thermal integrity of the as-manufactured sources.

Because it is important that NDT methods are compatible with capsule design, e.g., welds should be physically accessible for inspection,

the approach of this program is to become involved early with contractors who have design and development responsibilities. Continuing liaison will ensure that NDT methods are available for the source under development.

Design of a differential transducer was completed and construction will start soon. Work continued on the multi-element transducer system report. A paper, "Pulsed Laser Imaging System for Direct Visualization of Ultrasonic Waveforms," was accepted for presentation at the Acoustical Society of America's annual meeting in Houston, November 3-6.

II. TECHNOLOGY PUBLICATIONS - E. J. Wheelwright

The objective of this program is to provide technology for the use of Pm as a radioisotope heat source in aerospace or terrestrial applications. In the past, the program has included development of scientific and engineering data pertinent to: (1) the purification of Pm and conversion to a useful fuel form, (2) the design and fabrication of Pm heat sources, and (3) hazards analyses associated with radioisotopic heat sources. Presently, principal emphasis of the program is directed to the dissemination of Pm technology to the technical and scientific community.

The FY-1971 goal is to complete the metal compatibility studies and to make final arrangements for the publication of a book on Pm technology.

Most back chapters are completed. The remaining chapters are near completion with the exception of the analytical chemistry chapter, which is about one-half complete. The schedule will be met for the duplication and distribution of review copies to DID on all chapters with the possible exception of the analytical chemistry chapter.

III. ADVANCED SOURCE DEVELOPMENT - J. H. Jarrett

A. Advanced Alpha-Photon Source Development

The objective of this program is to evaluate present and future heat and radiatic 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 test results, reported by ORNL on PNL-prepared $^{241}\text{AmO}_2$ photon sources, were reviewed, and areas where PNL sources can be improved were identified. Several improvements in these areas are being incorporated into our source preparation work. The improvements include using mirror finish (7B) 304 stainless steel substrate material, minimizing the amount of oxygen in the system during sputtering, improving experimental techniques to minimize the effects of dusting, and sputter-etching thoroughly before the sputter-deposition of various source layers. These improvements should increase the adherence and integrity of the sputtered layers. Sputtered 304 stainless steel may possibly be susceptible to rusting when exposed to heated water or heated high humidity air. Apparent rusting was observed in the ORNL tests. A continuing evaluation of alternative material systems will help identify materials that are more satisfactory if this problem cannot be eliminated by the planned improvements in source fabrication.

An experiment was started to demonstrate the amount of rust resistance shown by 304 stainless steel layers sputtered under different system operating conditions. Using the cold (nonradioisotopic) sputtering system, stainless steel layers were deposited on polished copper and stainless steel materials. In one deposition run the residual air in the system before and during sputtering was minimized. In the second run, the initial amount of air in the system was not minimized and a controlled air leak was allowed during actual sputtering. The samples will be evaluated for their degree of rust resistance in October.

An experiment (described in the August 1970 monthly report, BNWL-B-13) designed to investigate the degree of cross-contamination that may be anticipated when sputtering different radioisotopes in a single glovebox sputtering system was started. Completion in October is anticipated.

A number of O-rings were replaced in the glovebox sputtering system. This resulted in a more leak tight system, and should help minimize air in the system during sputtering. The operational stability of the system has been improved by these and other maintenance efforts.

A paper, "Thin Film Radiation Sources Using RF Sputtering Techniques" by J. H. Jarrett has been accepted for presentation at the 1970 TMS Fall Meeting, October 19-22 in Cleveland, Ohio. The meeting is co-sponsored by AIME and ASM.

08-02. THERMAL APPLICATIONS08-02-01. CIRCULATORY SUPPORT SYSTEMS - F. T. Cross & J.C. SheppardA. Radiation Characteristics - Nonpromethium Systems

The 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.

The report, "In-phantom Dosimetry of Plutonium-238 Heat Sources in Relation to Circulatory Support Systems," which summarizes the measurements and computations made during FY-1970, has been released as BNWL-1489.

The development of computational techniques for determining organ dose rates from implanted ^{238}Pu sources is being continued.

The measurements of tissue dose rates from ^{252}Cf are being continued.

A comparison of recent radiographs of LASL-1 with the radiographic results of a year ago and the original radiographs taken at LASL indicates that the sealed source, now two years old, has not changed significantly.

08-03. RADIOISOTOPE SYSTEMS DEVELOPMENT08-03-03. OCEANOGRAPHIC AND NATURAL RESOURCE APPLICATIONSI. MARINE SYSTEMSA. Nuclear Techniques for Seabed Mineral Exploration -

R. W. Perkins and N. A. Wogman

Recent PNL studies of the composition of deep ocean sediments demonstrated that various mineral deposits and commercial-grade ores can readily be identified by neutron activation and instrument 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. Actual experience with Cf-252 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 samples on board ship and employs a shipboard Cf-252 irradiator and a Ge(Li) spectrometer. The former has the advantage of very rapid analyses, and the latter provides definition of mineral depth profiles from core samples.

The 1.8 mg Cf-252 source which was received last month has been used extensively in the calibration and testing program. With the Cf-252 source positioned at the bottom of a 1-in. diameter by 1-in. high paraffin cylinder, neutron flux characterizations have been made. A thermal neutron flux of 2.7×10^7 neutrons/cm²/sec is obtained at a distance of about 1 in. from the source. In paraffin this flux is only reduced twofold through the area of an 8-in. diameter hemisphere and threefold through a 14-in. diameter hemisphere. Similar flux measurements are being made in seawater and in seawater-saturated sediments and minerals. These latter measurements will provide the calibration needed for implementation of nuclear probe technology on the ocean floor. The essential design criteria for the nuclear probe have been completed and construction of some of the minor components of the probe is being initiated.

PROMETHIUM INVENTORY - SEPTEMBER 30, 1970

Received from ARHCO		16,361,900 Ci
Transfers and Shipments	1,448,500	
Concentrated Feed	5,406,170	
Product		
Run 22, 455 Ci/g, 8/31/70	19,425	
Run 40, 523 Ci/g, 8/31/70	35,495	
Run 55, 675 Ci/g, 8/31/70	79,890	
Waste	662,760	
Unaccounted	1,334,320	
Decay	7,375,340	
Total	16,361,900	

Production and Transfers

None.

DID BUDGET STATUS - FY-1971 Through September 30, 1970

	<u>Cost</u>	<u>Cost to Date</u>	<u>Budget</u>
08-01 <u>Radioisotope Production and Materials Development</u>			
08-01-03 Fission Products			
I. Low-Cost Cs/Sr Sources	6.5	13.3	80
II. Rhodium and Palladium	1.0	7.0	20
III. Xenon	0.1	0.4	20
IV. Evaluation of Isotopes Production	<u>15.2</u>	<u>27.7</u>	<u>80</u>
Subtotal 08-01-03	22.8	48.4	200
08-01-04 <u>Source Development</u>			
I. NDT R&D Support	1.5	6.2	25
II. Technology Publications	6.6	9.4	20
III. Advanced Source Development	<u>5.0</u>	<u>21.5</u>	<u>50</u>
Subtotal 08-01-04	13.1	37.1	95
Subtotal 08-01	35.9	85.5	295
08-02 <u>Thermal Applications Development</u>			
08-02-01 <u>Circulatory Support Systems</u>			
I. Circulatory Support	<u>9.6</u>	<u>23.1</u>	<u>85</u>
Subtotal 08-02	9.6	23.1	85

	<u>Cost</u>	<u>Cost to Date</u>	<u>Budget</u>
08-03 <u>Radioisotope Systems Development</u>			
08-03-03 <u>Oceanographic and Natural Resource Applications</u>			
I. Marine Systems	<u>4.4</u>	<u>17.0</u>	<u>60</u>
Subtotal 08-03	<u>4.4</u>	<u>17.0</u>	<u>60</u>
 Total 08 Operating Budget	 49.9	 125.6	 440
 08 Equipment Budget	 <u>0.8</u>	 <u>0.8</u>	 <u>17.0</u>
 Subtotal 08	 <u>50.7</u>	 <u>126.4</u>	 <u>457</u>
 Pm Inventory Account	 <u>3.6</u>	 <u>12.8</u>	 <u>210</u>
 PROGRAM GRAND TOTAL	 <u>54.3</u>	 <u>139.2</u>	 <u>667</u>