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Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

AEC Research and Development Report

PACIFIC NORTHWEST LABORATORY
DIVISION OF ISOTOPES
DEVELOPMENT PROGRAMS
QUARTERLY REPORT:
AUGUST-OCTOBER 1971

November 1971

BNWL-1308-8

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Radioisotope and
Radiation Applications

PACIFIC NORTHWEST LABORATORY
DIVISION OF ISOTOPES DEVELOPMENT PROGRAMS
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CONTENTS

LIST OF FIGURES	iii
LIST OF TABLES	iii
INTRODUCTION	1
SUMMARY	2
LOW COST CESIUM AND STRONTIUM SOURCES	5
EVALUATION OF LARGE SCALE USES FOR PURIFIED FISSION PRODUCT XENON	6
ISOTOPE PRODUCTION EVALUATIONS	7
Target Irradiation in Power Reactors.	7
Isotope Availability Projections	9
NEUTRON RADIOGRAPHY USING ^{252}Cf	22
Imaging Techniques	22
Source Holder Development	25
ADVANCED SOURCE DEVELOPMENT	27
CIRCULATORY SUPPORT SYSTEMS	29
Dosimetry Studies	29
Radiological Engineering Studies	29
Phase I Heaters	29
Phase II Heaters	30
NUCLEAR TECHNIQUES FOR SEABED MINERAL EXPLORATION	31

LIST OF FIGURES

- 1 Radiograph Density Versus Exposure for Various Direct Imaging Techniques
- 2 Seabed Mineral Analysis System
- 3 Relative Gamma-Ray Detection Efficiency of a Simulated Ocean Mineral of 1.8 g/cm³ Density Covered by Layers of Varying Density and Neutron Absorption Characteristics
- 4 Relative Gamma-Ray Detection Efficiency of a Simulated Ocean Mineral of 4.0 g/cm³ Density Covered by Layers of Varying Density and Neutron Absorption Characteristics

LIST OF TABLES

- 1 Target Irradiation Capabilities of a Power Reactor (PWR)
- 2 Column Heading Identification for Table 3
- 3 Net annual Isotope Availability (with target irradiation, process losses and decay losses)
- 4 Comparison of Production Capabilities for Power Reactors with Savannah River Reactors for ²³⁸Pu from ²³⁷Np (base case ²³⁷Np availability)
- 5 Comparison of ²³⁸Pu Availability from ²³⁷Np Irradiation with ²³⁸Pu Availability from Americium Irradiation
- 6 Contrast Readings for ~~IC~~ With Various Converter-Film Combinations. (The films were exposed to a radiographic density of about 1.5.)
- 7 Comparison of Portable Source Holders (150 μ g ²⁵²Cf source)

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INTRODUCTION

This report, BNWL-1308-8, is the 20th 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 through 1308-7.

This report covers the three-month period, August through October 1971.

SUMMARYLOW COST CESIUM AND STRONTIUM SOURCES

About 8 kCi of cesium crude was obtained from Hanford Waste Management Program (HWMP) for testing prototype source fabrication. Equipment is being designed and assembled for this work.

EVALUATION OF LARGE SCALE USES FOR PURIFIED FISSION PRODUCT XENON

Contacts with industry have resulted in two firm expressions of interest in obtaining experimental quantities of purified fission product xenon. The major interest is in investigating xenon's application as a human surgical anesthetic.

ISOTOPE PRODUCTION EVALUATIONS

Target irradiation capabilities of a PWR (1975-startup design) were calculated for a selected group of isotopes using the 2DB and ALTHAEA computer codes. It was assumed that targets could be inserted in unused rod cluster control assembly locations.

Revised projections of isotope availability from power reactor spent fuel processing were developed following procedures outlined in BNWL-716 (July 1968). A slower rate of nuclear plant startups and lower capacity factors reduced projected quantities relative to those shown in BNWL-716. The difference was large in the early 1970's but amounted to about a one-year offset in the 1980's.

The revised projections including revised target irradiation cycles indicated that power reactors could convert neptunium to ^{238}Pu at approximately the same rate as the Savannah River reactors. However, the quality in terms of ^{236}Pu content would not be as good. High quality ^{238}Pu with zero ^{236}Pu content could be produced at about one-half the rate for the neptunium irradiation by irradiating recovered $^{241}\text{Am}/^{243}\text{Am}$ in power reactors.

NEUTRON RADIOGRAPHY USING ^{252}Cf

Exposure values and image quality indicator contrast values were generated for a number of converter-film combinations using thermalized ^{252}Cf neutrons. The use of calcium tungstate scintillation screens for enhancement of images for transfer foils proved to be a useful technique in trials with both dysprosium and europium metal foils. An evaluation of the new water-filled exposure facility was completed and a comparison was made with NC-1. Preliminary radiographs have been made with the new 1.7 mg ^{252}Cf source.

ADVANCED SOURCE DEVELOPMENT

Source preparation hardware for use in the up-dated glovebox sputtering system was developed. As a result, the sputter deposition of the source radioactive layer and the containment layer may be performed sequentially without breaking vacuum. A new $^{241}\text{AmO}_2$ sputtering target (the source of sputtered material) is being prepared. The present target gives a $^{241}\text{AmO}_2\text{-Cu}$ (cermet) active layer that is less than 5% americium oxide. The new target should give cermet films containing 50% americium oxide. A residual gas analysis (RGA) system and humidity control system are being procured. Work on the Advanced Source Development Program was presented at meetings in Denver and in Gatlinburg.

CIRCULATORY SUPPORT SYSTEMS

Measurements of the photon dose rates from an implanted Rankine prototype device containing a 32-W $^{238}\text{Pu}^{16}\text{O}_2$ (PNL30-6) source continue. The User's Manual for Dose Rate Calculations was written and is being edited for final preparation.

The first Phase I heater control sham was completed and implanted in a Hanford miniature swine. The Phase I heater bench model has been assembled. Shop fabrication drawings of the Phase II heater have been completed.

NUCLEAR TECHNIQUES FOR SEABED MINERAL EXPLORATION

Fabrication of nuclear probe components is nearly complete; however, final assembly and the initial testing has been delayed due to detector cryostat cooling problems. Programming of the electronic package has been initiated and evaluations are continuing on the effects of sediment composition and inhomogeneity on the gamma-ray counting rate from neutron activated seabed minerals. The evaluations are utilizing ^{252}Cf sources of 0.2 to 9 mg.

LOW COST CESIUM AND STRONTIUM SOURCES

H. H. Van Tuyl

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.

Equipment is being designed for fabricating prototype cesium sources using material recovered in the Hanford Waste Management Program (HWMP). Previously available feed contained 1324 Ci of ^{137}Cs per liter (about 44 g total Cs/liter), with equal weights of both sodium and potassium. A recent shipment of about 5 gal of cesium from HWMP contained only 465 Ci of ^{137}Cs per liter. If used at its present purity, this would make about two prototype sources at 1 in. diameter by 24 in. long. If purified to over 90% cesium, there would not be quite enough cesium to prepare even one full prototype source. The availability of more concentrated and higher purity feed is being investigated.

Most of the required operations of feed preparation can be accommodated in existing equipment with minor additions or modifications. Furnaces will be needed for melting the cesium chloride and filling the source container. The principal new equipment items will be for welding and decontamination. Design of a remotely operable system is not yet complete.

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 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.

An announcement was issued to major xenon users, producers, and nuclear fuel processors to advise them of the availability and quality of purified fission product xenon which could be made available in exchange for data on the advantages of the material. Two firm expressions of interest were obtained with more expected to respond. The major use will be in a resumption and continuation of work that began over 20 years ago concerning application as a human surgical anesthetic. It is probable that all of the presently available material could be requested for this one experimental application.

ISOTOPE PRODUCTION EVALUATIONS

R. W. McKee and C. E. Heeb

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.

TARGET IRRADIATION IN POWER REACTORS

The capabilities of 1975-startup-design PWR for target irradiations were calculated for a selected group of target materials and product isotopes. The operating mode of a power reactor with normal one-year refueling cycles is not attractive for production of short-half-lived isotopes and, for this reason, some of the short-half-lived product isotopes included in these calculations are not likely to be serious candidates for production in power reactors.

Because of the large epithermal neutron component in the neutron flux of a power reactor, resonance cross sections have a strong effect on yields and production rates. The resonance integrals used in these calculations were obtained from a compilation by M. K. Drake⁽¹⁾ of the Brookhaven Neutron Cross-Section Center. The reactor model for these calculations was the McGuire Nuclear Station,⁽²⁾ a PWR scheduled for startup in mid-1975. Targets were placed in unused rod cluster control assembly (RCCA) locations and irradiated at 82% of design power for one 11-month refueling cycle (equivalent to operation with an annual capacity factor of 75%). The calculation procedure was as follows:

- The 2DB computer code, a multigroup diffusion theory code which has been extensively used at PNL for reactivity and power distribution calculations, was used to construct a two-dimensional four-energy-group model of the fuel assembly, including the target locations. With this model, it was then possible to calculate the relationships between neutron flux in the target, reactivity, and infinite multiplication factor (k_{∞}) in the fuel assembly and target cross sections.
- Using this 2DB model, a calibration relationship between the collapsed one-energy-group target-region macroscopic cross section (Σ_a) and k_{∞} was developed based on calculations for cobalt target densities of 0, 10, 20, and 40 g/ft. The average neutron flux (ϕ_T) in the target region was also calculated.
- The neutron flux calculated with 2DB was converted to the Wescott flux (ϕ_w) and the target burnups and conversions to product isotopes were calculated using the ALTHAEA computer code. The ALTHAEA code is a high-speed burnup code for unit-cell or target-cell calculations. The Wescott cross section (σ_w) was also calculated in ALTHAEA.
- The enrichment required to maintain the design fuel exposure with targets inserted was calculated as follows:
 - a. The Σ_a for each target case was calculated for the relationship

$$\Sigma_a = \frac{\sigma_w \phi_w N}{\phi_T}$$

where N = region average nuclear density of the target isotope
in the target region.

- b. The effect of the target on k_{∞} was derived from 2DB calculated relationship between k_{∞} and Σ_a .
- c. The enrichment required to compensate for the reactivity loss due to the target and to produce the same burnup was calculated

utilizing a set of ALTHAEA calculated k_{∞} versus exposure calculations for a wide range of average fuel-cell enrichments.

- d. To check the accuracy of the above procedure for determining compensating enrichments, the enrichment requirements for the cobalt cases were calculated directly with 2DB and these results agreed within 0.5% with the ALTHAEA results.

Results of the calculations are summarized in Table 1.

For high cross section and high resonance integral materials, target density has a very strong effect on production rates because of the self-shielding and flux depression effects. Because target density had such a strong effect on conversion rates and enrichment requirements in the case of ^{237}Np , $^{241}\text{Am}/^{243}\text{Am}$, and cobalt, calculations were carried out at several target densities for each of these targets, but only one representative result is shown in Table 1. Determination of optimum density was outside the scope of this study. Previous studies of ^{237}Np irradiation have indicated the optimum density for this material to be in the range of 0.2 to 0.8 g/cm³, depending upon cost assumptions.

In the case of thulium and lithium targets, it was also necessary to reduce the target density to realize a reasonable enrichment requirement, but only one density case was calculated for these targets.

The quantities in Table 1 identified as "per 1000 MWe" represent the maximum possible utilization of RCCA vacancies. It may be that this much target handling would unreasonably complicate reactor refueling operations and the practical maximum may be somewhat less than the value in Table 1.

ISOTOPE AVAILABILITY PROJECTIONS

Revised projections of isotope availability from power reactor spent fuel processing were developed following procedures outlined in BNWL-716(3) (July 1968). The base case projection was based on presently scheduled nuclear plant startups delayed by six months, plus projected capacity

TABLE 1. Target Irradiation Capabilities of a Power Reactor PER

Target	Reaction	Product	Target Concentration g/cm ³	Enrichment Required in Assembly, (a) % ²³⁵ U	Product Yields (no loss basis)			Maximum kCi/ 1000 MWe/ Year	Product Activity: Ci/ gram of Element
					g/g	kg/Assembly	Maximum kW/1000 MWe/ Year		
²³⁷ Np (oxide)	n,γ	88y ²³⁸ Pu	0.5	4.09	0.193	0.5	32.1		
²⁴¹ Am & ²⁴³ Am (50/50) (oxide)	n,γ	98y ²³⁸ Pu 193d ²⁴² Cm 18y ²⁴⁴ Cm	0.25	4.24	0.106 0.115 0.113	0.138 0.150 0.147	8.8 2,050 47		
Cobalt	n,γ	5.3y ⁶⁰ Co	(20 g/ft)	3.99	0.028	0.134		17,300	31.5
Thulium (Tm ₂ O ₃)	n,γ	127d ¹⁷⁰ Tm	0.5	3.80	0.108	0.280	500	194,000	650
Bismuth	n,γ	138d ²¹⁰ Po	9.3	3.39	1.6 × 10 ⁻⁵	7.2 × 10 ⁻⁴	12	370	4,490
Nitrogen (Be ₃ N ₂)	n,p	5730y ¹² C	0.8	3.43	1.3 × 10 ⁻³	5.5 × 10 ⁻³	0.9 × 10 ⁻³	2.8	4.5
Lithium (Li ₂ O)	n,α	12.3y ³ H	0.1	3.87	0.262	0.136	5.6	150,000	9,690
Phosphorus (P ₂ O ₅)	n,γ	14.3d ³² P	1.0	3.40	6 × 10 ⁻³	8.0 × 10 ⁻³	875	260,000	440
Sulfur	n,γ	88d ³⁵ S	1.9	3.41	1.6 × 10 ⁻⁴	1.6 × 10 ⁻³	2.5	7,600	6.7
Telurium	n,γ	8.1d ¹³¹ I ¹²⁷ I ¹²⁹ I	5.7	4.14	2.6 × 10 ⁻⁶ 1.6 × 10 ⁻⁴ 1.4 × 10 ⁻⁴	7.6 × 10 ⁻⁵	3.6	1,100	1,040

a. Based on enrichment without target = 3.3% ²³⁵U

additions after 1977 to reach 150,000 MWe installed by the end of 1980 and 500,000 MWe installed by the end of 1990. Plutonium recycle was assumed to start in 1975.

Because of the recent Calvert Cliffs decision regarding AEC compliance with NEPA regulations, there is a large uncertainty in near-future projections of capacity additions. The six-month delay assumed for the base case is probably a minimum allowance.

The effect of fast reactors (LMFBR's) was not considered because it does not appear that significant numbers of these reactors can be in commercial operation much before the end of the period considered (1990).

Any spent fuel processed prior to 1971 was deleted from consideration in these projections.

A lower capacity factor for reactor operations was used in the present projection relative to the BNWL-716 projections; 75% compared to 85% in the early years of plant operations.

The items reported in the availability projection tables are identified by abbreviated titles which are defined in Table 2. The net annual availabilities for the base case projection are shown in Table 3.

The assumptions in the projections regarding target irradiation of neptunium and americium were as follows:

- ^{237}Np irradiation is based on ^{237}Np recovery starting in 1973.
- ^{237}Np is assumed to be sent to Savannah River reactors for irradiation through 1979 and sent to power reactors in and after 1980.
- $^{241}\text{Am}/^{243}\text{Am}$ recovery is assumed to start in 1975 and is sent to power reactors for irradiation.

It is of interest to compare the effect on ^{238}Pu availability of irradiating the ^{237}Np in power reactors rather than in the AEC Savannah River (SR) reactors. This comparison is made in Table 4. The base case assumption of SR reactors to 1979 and power reactors after 1980 is also

TABLE 2. Column Heading Identification for Table 3.
(Availability of the following defined isotopes are reported, allowing year for recovery after fuel discharge except as noted.)

Isotope	Description
Pu (reprocessed)	Total ^{239}Pu , ^{240}Pu , ^{241}Pu and ^{242}Pu reprocessed
Pu (stockpile)	Total ^{239}Pu , ^{241}Pu , ^{241}Pu and ^{242}Pu to stockpile (reduced by ^{241}Pu decay to ^{241}Am).
^{237}Np	^{237}Np from spent fuel
^{241}Am	^{241}Am from spent fuel (^{241}Am recovered includes 6-months accumulation of ^{241}Am from ^{241}Pu decay)
^{243}Am	^{243}Am from spent fuel
^{242}Cm	^{242}Cm from spent fuel present with ^{244}Cm aged 3 years
^{244}Cm	^{244}Cm from spent fuel aged 3 years from discharge
^{244}Cm	^{244}Cm produced from target irradiation of ^{243}Am
^{243}Am	
^{244}Cm Sum	Total ^{244}Cm from spent fuel and target irradiation
^{238}Pu Np	^{238}Pu produced from target irradiation of ^{237}Np

TABLE 2. (contd)

Isotope	Description
Xe Stable	Includes stable Xe isotopes, ^{131}Xe , ^{132}Xe , ^{134}Xe , ^{136}Xe
^{242}Cm ^{241}Am	^{242}Cm produced from target irradiation of ^{241}Am .
^{241}Am ^{241}Pu	^{241}Am produced from decay of ^{241}Pu in Pu stockpile (Pu-Sp)
Kr Stable	Includes stable Kr isotopes, ^{83}Kr , ^{84}Kr , ^{86}Kr .
Ru Stable	Includes stable Ru isotopes, ^{99}Ru , ^{100}Ru , ^{101}Ru , ^{102}Ru , ^{104}Ru
^3H	Grams of tritium produced as a fission product

TABLE 3. Net Annual Isotope Availability
(with target irradiation, process
losses and decay losses)
Case 1 Base Projection

During Period Ending	Total Installed Capacity GW (E)	Tonne	Isotope Production, kg/yr												
			Pu	Pu	237	241	243	242	244	244- 243	244	238Pu	238Pu	238Pu	238Pu
			(reprocessed)	(stockpile)	Np	Am	Am	Cm	Cm	Am	Sum	Np	Cm	Am	Sum
1969	2.3	0	0	0	0	0	0	0	0	0	0	0	0	0	0
1970	4.0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
1971	7.3	34	189	189	4	1	0	0	0	0	0	0	0	0	0
1972	18.1	45	309	308	10	2	1	0	0	0	0	0	0	0	0
1973	33.4	155	1,015	1,012	32	6	3	0	0	0	0	0	0	0	0
1974	46.9	392	2,596	2,586	82	16	9	0	0	0	0	6	0	0	6
1975	59.9	689	4,707	-1,062	155	29	18	0	1	0	1	20	0	0	20
1976	71.9	1,187	8,345	-120	288	54	33	0	2	0	2	43	0	0	44
1977	86.5	1,724	13,566	-1,492	482	97	75	0	4	0	4	84	1	11	96
1978	108.0	2,012	17,164	478	605	132	117	0	6	0	6	150	1	20	171
1979	128.0	2,403	21,857	1,021	734	178	173	0	15	5	20	218	3	42	263
1980	150.0	3,087	28,862	253	932	247	257	0	25	9	35	290	4	60	354
1981	173.0	3,870	36,666	1,020	1,133	325	347	0	41	25	65	200	6	88	294
1982	199.0	4,762	47,974	1,410	1,354	442	498	0	66	59	105	363	8	123	494
1983	229.9	5,548	58,804	491	1,514	574	688	0	95	66	161	368	11	165	544
1984	262.9	6,629	74,417	1,111	1,733	752	942	1	140	99	239	570	14	225	810
1985	299.8	7,880	89,408	179	1,978	911	1,171	1	203	143	346	626	19	295	940
1986	339.8	9,376	107,796	733	2,291	1,091	1,445	1	279	209	488	848	25	391	1,263
1987	379.8	10,886	129,900	496	2,541	1,329	1,836	1	346	292	639	958	30	484	1,472
1988	419.8	12,750	152,436	1,408	2,901	1,530	2,168	2	417	409	827	1,213	36	597	1,846
1989	459.8	14,268	175,075	829	3,138	1,776	2,588	2	519	532	1,051	1,363	44	729	2,136
1990	499.8	16,278	203,223	2,981	3,481	2,060	3,073	2	595	690	1,285	1,654	51	856	2,560

TABLE 3. (contd)

During Period Ending	GW-yr	Isotope Production, kg/yr													
		²³⁵ U	²³⁶ U	¹⁴⁷ Pm	¹⁴⁴ Ce	¹³⁷ Cs	⁹⁰ Sr	⁸⁵ Kr	⁹⁹ Tc	¹⁰³ Rh Sum	¹⁰⁶ Ru	Pd Stable	Xe Stable	²⁴² Cm ²⁴¹ Am	²⁴¹ Am ²⁴¹ Pu
1969	1.7	0	0	0	0	0	0	0	0	0	0	0	0	0	0
1970	3.0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
1971	5.4	396	60	0	3	14	7	0	10	6	1	8	62	0	0
1972	13.5	442	104	0	5	27	13	1	19	11	2	18	119	0	1
1973	25.0	1,500	340	2	16	89	41	2	62	34	6	62	391	0	3
1974	35.1	3,787	870	3	40	230	106	5	160	87	14	161	1,009	0	9
1975	44.8	6,506	1,596	9	73	428	198	9	297	160	27	300	1,874	0	23
1976	53.8	10,933	2,878	23	126	775	355	16	537	290	48	556	3,393	0	17
1977	64.7	14,462	4,480	41	186	1,259	568	25	868	460	76	947	5,519	3	17
1978	80.8	16,273	5,430	72	221	1,562	697	30	1,074	566	94	1,205	6,853	6	8
1979	95.7	19,415	6,526	108	265	1,904	838	37	1,307	695	118	1,503	8,350	12	11
1980	112.2	24,685	8,241	130	343	2,449	1,074	47	1,682	897	153	1,946	10,744	17	17
1981	129.4	31,695	10,164	157	430	3,031	1,322	58	2,082	1,123	193	2,419	13,284	24	18
1982	148.8	37,240	12,052	202	523	3,712	1,597	70	2,548	1,395	245	3,020	16,261	34	24
1983	172.0	42,877	13,548	254	606	4,279	1,814	79	2,935	1,634	293	3,540	18,725	45	32
1984	196.6	50,429	15,550	312	723	5,042	2,110	92	3,458	1,959	361	4,227	22,045	62	35
1985	224.1	59,852	17,970	363	856	5,857	2,443	107	4,019	2,302	429	4,905	25,588	80	41
1986	253.9	71,120	20,961	434	1,020	6,849	2,850	125	4,703	2,720	514	5,723	29,902	106	42
1987	283.4	81,463	23,383	513	1,177	7,773	3,205	140	5,338	3,137	606	6,537	33,906	130	47
1988	312.7	95,377	26,943	610	1,377	8,942	3,684	161	6,145	3,640	711	7,489	38,975	158	49
1989	341.6	105,864	29,313	705	1,538	9,864	4,032	177	6,779	4,066	806	8,311	42,967	193	57
1990	370.3	119,898	32,721	824	1,747	11,108	4,513	198	7,634	4,623	928	9,400	48,353	225	62

TABLE 3. (contd)

<u>During Period Ending</u>	<u>K_r Stable</u>	<u>R_u Stable</u>	<u>³H Grams</u>
1969	0.0	0	0
1970	-0.0	0	0
1971	4.4	23	0
1972	8.0	45	1
1973	26.2	150	3
1974	67.7	389	6
1975	125.7	721	12
1976	225.6	1,311	22
1977	361.9	2,149	35
1978	445.0	2,679	44
1979	534.5	3,285	54
1980	684.5	4,835	69
1981	841.0	5,251	85
1982	1,015.9	6,472	105
1983	1,152.9	7,501	120
1984	1,337.8	8,881	142
1985	1,547.2	10,323	164
1986	1,802.4	12,074	192
1987	2,023.9	13,746	218
1988	2,323.3	15,806	250
1989	2,539.6	17,483	276
1990	2,839.3	19,725	310

TABLE 4. Comparison of Production Capabilities of Power Reactors
with Savannah River Reactors for ^{238}Pu from ^{237}Np
(Base Case ^{237}Np Availability)

<u>Year</u>	<u>Np Irradiated in SR Reactors All Years</u>	<u>Np to Power Reactors After 1980</u>	<u>Np Irradiated in Power Reactors All Years</u>
1971	0	0	0
1972	0	0	0
1973	0	0	0
1974	6	6	0
1975	20	20	8
1976	43	43	20
1977	84	84	44
1978	150	150	85
1979	218	218	151
1980	290	290	213
1981	377	200	296
1982	476	363	393
1983	586	368	507
1984	692	570	637
1985	806	626	766
1986	932	848	922
1987	1078	958	1083
1988	1227	1213	1281
1989	1398	1363	1469
1990	1561	1654	1712
<u>Cumulative Totals</u>			
1980	804	804	515
1985	3673	2876	3065
1990	9645	8723	9330

shown in the middle column of Table 4 for reference. The power reactor conversion rate is slower in the early years because of a longer cycle, but catches up with and exceeds the SR reactor rate in the late 1980's because of more efficient conversion. Less neptunium is lost to ^{238}Np fission and to ^{238}Pu conversion to higher isotopes in the power reactors. The power reactor product, however, has the strong disadvantage of probable higher ^{236}Pu content [previously calculated⁽⁴⁾ at around 5 ppm as compared to about 1 ppm for the SR product].

The $^{241}\text{Am}/^{243}\text{Am}$ irradiation offers a promising prospect for large quantities of very high quality ^{238}Pu with no ^{236}Pu content. It would be diluted, however, to about 65% ^{238}Pu by ^{242}Pu from the branching decay of 16-hour ^{242}Am during irradiation. A two-step separation of the americium irradiation products was assumed in which the first separation at six months after irradiation would include about three-fourths of the ^{238}Pu and all of the ^{242}Pu . The last fourth of the ^{238}Pu would be recovered two years after irradiation after further decay of the ^{242}Cm . The last fourth of the ^{238}Pu would be both high quality and high purity ^{238}Pu . The base case quantities of ^{238}Pu from neptunium and americium irradiations are compared in Table 5 which shows that by the end of the period considered (1990), high quality ^{238}Pu with zero ^{236}Pu content could be produced from americium irradiation in power reactors at about half the rate for ^{238}Pu from neptunium irradiation.

TABLE 5. Comparison of ^{238}Pu Availability from ^{237}Np Irradiation
with ^{238}Pu Availability from Americium Irradiation

<u>Year</u>	<u>^{238}Pu Availability from ^{237}Np Irradiation</u>	<u>^{238}Pu Availability from Americium Irradiation</u>
1971	0	0
1972	0	0
1973	0	0
1974	6	0
1975	20	0
1976	43	0
1977	84	11
1978	150	20
1979	218	42
1980	290	60
1981	200	88
1982	363	123
1983	368	165
1984	570	225
1985	626	295
1986	848	391
1987	959	484
1988	1213	597
1989	1363	729
1990	1654	856
<u>Cumulative Totals</u>		
1980	804	132
1985	2876	1014
1990	8723	3993

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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.

IMAGING TECHNIQUES

Development of appropriate imaging techniques entails the greatest promise of widespread acceptance of neutron radiography with isotopic sources of neutrons. A number of direct imaging techniques have been evaluated and exposure versus radiographic densities are shown in Figure 1. Scintillators ($^6\text{LiF-ZnS}$, $\text{Gd}_2\text{O}_3\text{:Tb}$) produce the fastest images as can be seen from the figure. Table 6 compares contrasts obtained with the various converter-film combinations. Increasing cadmium contrast indicates a better thermal neutron image, while increasing lead contrast indicates a high ratio of gamma-to-neutron imaging. One wants the highest cadmium and the lowest lead contrasts possible. The cadmium contrast is a measure of the epithermal neutron imaging while the CH_2 contrast (plexi-glass) gives a measure of the contrast for a material which is a moderate attenuator of thermal neutrons. Although the use of $^6\text{LiF-ZnS}$ scintillators looks very promising, the image quality from these scintillators is poor with regard to resolution and uniformity. Fiber optics face plates have been used to increase the resolution; however, the time required to form an

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FIGURE 1. Radiograph Density Versus Exposure for Various Direct Imaging Techniques

TABLE 6. Contrast Readings for IQI with Various Converter-Film Combinations
(The films were exposed to a radiographic density of about 1.5.)

IMAGING TECHNIQUE*	CONTRAST (CHANGE IN FILM DENSITY " ΔD ")			
	Cd (0.5mm)	Pb (1.15mm)	CH ₂ (1.5mm)	In (0.76mm)
LiF-ZnS (NE421) + RP/R	0.65	0.03	0.16	0.37
LiF-ZnS (NE425) + RP/R	0.52	0.02	0.09	0.16
Gd ₂ O ₂ S:Tb + RP/R	0.34	0.13	0.24	0.18
Gd ₂ O ₂ S:Tb + TYPE O	0.23	0.12	0.26	0.23
Gd (2mils) + RP/R	0.31	0.23	0.17	0.16

* FILM DESIGNATIONS ARE FOR KODAK RADIOGRAPHIC FILMS

image is 1-1/2 to 5 times as long. The use of $\text{Gd}_2\text{O}_2\text{S:Tb}$ scintillators seems to be the most promising with regard to resolution, contrast and speed.

Evaluation of europium for indirect imaging in low fluxes is continuing. Calcium tungstate scintillation screens are being used to enhance the image density from the transfer foils. In a flux of $3 \times 10^3 \text{ n/cm}^2\text{-sec}$ the use of the screen increases the radiographic density by a factor of 3. Calcium tungstate screens have also been successfully used with dysprosium foils.

SOURCE HOLDER DEVELOPMENT

The portable source holder described in the previous quarterly report⁽¹⁾ (NC-2) was compared with the original portable unit (NC-1). Table 7 summarizes the data collected from the comparison. The imaging performance of NC-2 is obviously better than NC-1 and the unit is more portable as pointed out previously. ⁽¹⁾

A 1.7 mg source of ^{252}Cf was received and installed in our tank shield. ⁽²⁾ Surface dose rates with the source in the stored position are 10 mR/hr gamma and 4 mrem/hr neutrons. The same collimator used in NC-2 is being used in this facility resulting in images of comparable quality in about 1/10th of the time.

TABLE 7. Comparison of Portable Source Holders
(150 μ g ^{252}Cf source)

	NC-1	NC-2
Portable weight	350 lb	75 lb
Weight with moderator	350 lb	~320 lb
Moderator material	ZrH _x + others	H ₂ O
Typical collimation ratio	8:1	15:1
Source to object distance	69 cm	38 cm
Thermal flux	$2.8 \times 10^2 \text{ n/cm}^2\text{-sec}$	$9.4 \times 10^2 \text{ n/cm}^2\text{-sec}$
Cadmium ratio	1.4	1.8
Neutron-to-gamma ratio.	$6.3 \times 10^4 \text{ n/cm}^2\text{-mR}$	$4.2 \times 10^4 \text{ n/cm}^2\text{-mR}$
Gamma filter	1.8 in. ZrH ₂ + 0.18 in. depleted U	0.2 in. Pb
Surface dose rates neutron	150 mrem/hr	170 mrem/hr
gamma	50 mR/hr	100-180 mR/hr

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ADVANCED SOURCE DEVELOPMENT

J. H. Jarrett

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.

In the previous quarter a new tuning network assembly for the glovebox sputtering system was installed. The glovebox system is now capable of sequentially sputtering two different materials without releasing vacuum. This should result in better adherence between the two sputtered layers and, in the case of radiation sources, better containment of the sputtered radioactive layer by the sputtered containment layer. To take full advantage of the glovebox system's capability for sequential sputtering, some new source preparation hardware was developed in this quarter. This hardware will allow the sputter deposition of a 1/2 in. diameter radioactive layer in the center of ten, planar, 7/8 in. diameter substrates. In preparation for containment layer deposition the mask used to position the radioactive layer can be removed remotely without breaking vacuum. This will allow the containment layer to be deposited immediately over the full 7/8 in. diameter of the substrate. The hardware has been developed and evaluated using the cold (nonradioactive) sputtering system. Hardware will be installed and operated in the glovebox sputtering system during the next quarter.

In addition to the source preparation hardware development, work was started on a new ^{241}Am containing sputtering target (the source of sputtered $^{241}\text{AmO}_2$ for radiation sources). The existing $^{241}\text{AmO}_2\text{-Cu}$ (cermet) target gives sputtered layers that are less than 5% americium oxide. The next target should give sputtered layers that are over 80% americium oxide.

With the new target americium oxide-metal (cermet) sputtered layers containing on the order of 50% americium oxide should be possible with the flexibility to vary both the identity and the amount of the metal in the sputtered layer.

A residual gas analyzer (RGA) which will be used to monitor vapor phase components in the sputtering system before, during, and after a deposition run is being procured. System delivery and installation is anticipated after mid-FY-72. The anticipated advantages of RGA were discussed in the previous quarterly report in this series (BNWL-1308-7). Also under procurement is a humidity control system for the Advanced Source Development laboratory. This system should eliminate condensate formation on water cooled rf power leads and cooling lines, assuring safe and reliable sputtering system operation. Installation is expected after mid-FY-72.

Information on the Advanced Source Development Program was described at a meeting in Denver, Colorado, of source producers and source users. The meeting was held in conjunction with the 20th annual X-Ray Conference, August 10, 11, and 12. In October a paper⁽¹⁾ on the program was presented at the Third International Symposium on Research Materials for Nuclear Measurement in Gatlinburg, Tennessee.

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CIRCULATORY SUPPORT SYSTEMS

F. T. Cross, J. C. Sheppard
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 50-W plutonium heat sources.

DOSIMETRY STUDIES

Measurements of the photon dose rates from an implanted Rankine prototype device containing a 32W $^{238}\text{PuO}_2^{16}$ (PNL 30-6) source continue. The User's Manual for Dose Rate Calculations was written and is being edited for final preparation.

RADIOLOGICAL ENGINEERING STUDIESPhase I Heaters

The first control sham was completed and implanted in a Hanford miniature swine on September 28. The device is identical in geometry and weight to the actual blood heater, but excludes the telemetry system and isotope capsule. The presurgery techniques for cooling and transferring the device to the animal were simultaneously rehearsed with no major difficulties.

The bench model including telemetry system and electric heater has been assembled.

Phase II Heaters

Shop fabrication drawings of the heat conduit system have been completed and include the minor changes dictated by a previously completed computer thermal analysis.

NUCLEAR TECHNIQUES FOR SEABED MINERAL EXPLORATION

R. W. Perkins, H. G. Rieck, Jr. and N. A. Wogman

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 primary emphasis this past quarter has continued to be on the assembly of the underwater ^{252}Cf nuclear probe system, on the evaluation of the effect of various sediment layers on the gamma-ray counting rate from neutron activated seabed minerals, and on the effect of the ^{252}Cf source-to-Ge(Li) diode distance on the background and detection sensitivity of elements on the sea floor. In addition to the physical assembly of the probe, the electronics are being programmed, as shown in Figure 2, to report the elements, their concentrations, and their measurement precision based on standards and count rate information. This programming also allows the gain and zero of the amplifiers to be continually adjusted while the probe system is operating at depth.

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FIGURE 2. Seabed Mineral Analysis System

In the studies of the effect of various sediment layers on the gamma-ray counting rate from minerals of interest, the simulated seabed ^{252}Cf nuclear probe was used to irradiate large specimens of dunite, dunite- H_2O -In, dunite-Fe, dunite-Fe- H_2O , dunite-Fe- H_2O -Cd, and dunite-Fe- H_2O -In. These measurements (see Figures 3 and 4) are providing the necessary information to estimate the errors which could be introduced by variations in the water content, the matrix composition, the total neutron capture cross section, the density, and the elastic scattering cross sections of the matrix. The variation in counting rate associated with density changes of 1.8 to 4.0 have been well characterized.

The effect of increasing the cross section of overlying material has been carefully analyzed. For example, the addition of 1% of an extremely high cross section material, cadmium, to a 2.5 cm surface layer above the mineral being analyzed reduced the resulting gamma-ray counting rate about four-fold. However, in actual application, it appears the detection efficiency will not be impaired by a high cross section sample matrix. If the element of interest were in a matrix of density 4.0 g/cm^3 instead of the 1.8 g/cm^3 material typical of ocean sediments, its relative gamma-ray detection efficiency would be reduced about two-fold.

Experiments were conducted to determine the background of a Ge(Li) diode as a function of distance in fresh water from the ^{252}Cf source. These experiments indicated that a significant interference did occur at a source-to-detector separation distance of 6 ft. This background drops off very rapidly with distance and is insignificant at 10 ft. Because of this background contribution, the nuclear probe has been modified so that separation distances of 10 ft are provided during the counting period.

Fabrication of nuclear probe components for the first generation welding and assembly is nearly complete. Design modification of the irradiator for easy source removal is being established. All off-site equipment required for the ocean tests at Battelle's marine research laboratory at Sequim, with exception of the detector, has been received.

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FIGURE 3. Relative Gamma-Ray Detection Efficiency of a Simulated Ocean Mineral of 1.8 g/cm^3 Density Covered by Layers of Varying Density and Neutron Absorption Characteristics

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FIGURE 4. Relative Gamma-Ray Detection Efficiency of a Simulated Ocean Mineral of 4.0 g/cm^3 Density Covered by Layers of Varying Density and Neutron Absorption Characteristics

The detector manufacturer is having problems with the detector system, specifically with holding time on the cryostat. He reports a 17-hr holding time rather than the design value of 36 to 48 hr and consequently suspects a defect. He has not estimated a delivery date since testing of the planned changes in the system require considerable time to complete. Additional cooling or heat transfer fins are being installed in the cryostat heat exchanger.

The ND-4400 analyzer system which forms the heart of the electronic package has been used with a temporary diode to collect spectra from activation of several minerals. Operation appears to be satisfactory and the system can be used in our preliminary testing of the probe. Complete data processing cannot be incorporated into the present unit because of insufficient memory size. An additional 4K of memory is being obtained together with source programs which will allow us to make alterations in existing programs to accommodate our planned data processing. Spectrum recording on tape cassettes can be done with the present programs.

A brief description of the planned probe system is in order since several changes have been incorporated in the design. The detection-irradiation assembly is a triangular-shaped structure (included angle at axis is 57°) with 6-ft long legs (distance from axle to detector irradiator). This assembly is rotated or oscillated between irradiate and detect positions by a hydraulically driven rotary actuator. Power is supplied by a hydraulic pump located on the probe. Electric power is supplied from topside and direction of pump rotation controls the direction of oscillation. Switching of electric leads topside changes motor rotation. In the detect mode of operation, the irradiator assembly is extended away from the triangle leg by a hydraulic cylinder (also energized from the same hydraulic pump). Separation at extreme extension is approximately 10 ft (detector to irradiator). The detector-irradiator assembly will be protected by a lucite trough which extends 3 in. below the bottom of the framework. The detector-irradiator assembly will be at a fixed distance from the trough. A TV monitor-light assembly will be mounted in position that will allow observation of the probe operation from topside.

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