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PACIFIC NORTHWEST LABORATORY
ISOTOPES DEVELOPMENT PROGRAMS
QUARTERLY REPORT:
FEBRUARY - APRIL 1972

May 1972

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Radiation Applications

PACIFIC NORTHWEST LABORATORY
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INTRODUCTION

This report, BNWL-1308-10, is the 22nd 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 1308-9.

This report covers the three-month period, February through April 1972.

SUMMARY

LOW-COST CESIUM AND STRONTIUM

Design, fabrication, procurement and testing of equipment are being completed so that prototype cesium capsules (1 in. diameter by 24 in. long) can be fabricated this fiscal year. Satisfactory conditions for remote welding of Type 316 stainless steel have been developed.

Extensive thermodynamic calculations were made to determine the interaction of zirconium metal or fluoride with impurities which might be present in strontium fluoride heat sources. Zirconium would be an excellent getter for oxygen, nitrate, carbonate, water, sulfate, and phosphate. The oxygenated anions are expected to be decomposed to form zirconium oxide. Zirconium, either added as a getter or formed by decay of ^{90}Sr , would protect Hastelloy-C from attack by the impurities studied thus far.

EVALUATION OF LARGE SCALE USES FOR PURIFIED FISSION PRODUCT XENON

The first shipment of fission-product xenon purified under this program was completed to the Linde Division of Union Carbide. Other potential uses continue to develop but firm requests have not yet been received.

FISSION PRODUCT RHODIUM, PALLADIUM, RUTHENIUM AND TECHNETIUM UTILIZATION

Study on the separations chemistry of these fission product materials was continued. A sulfide precipitation process was evaluated using a synthetic high-level waste solution. Yields from 43 to 89% were obtained for the various fission product elements. The projected supply/demand situation and a general market analysis related to these materials are being investigated.

NEUTRON RADIOGRAPHY USING ^{252}Cf

The current effort on ^{252}Cf neutron radiography is nearing completion and this report summarizes developments over the past 18 months. Programs

concerning the development of exposure facilities, film-converter imaging techniques and electronically aided imaging techniques are summarized. Recent progress with Polaroid film and an image intensifier has shown that neutron radiographs can be produced with low thermal fluxes ($<10^3$ n/cm²-sec) with reasonable exposure times (2 to 30 min). Although the image with the image intensifier (2 min exposures at 7×10^2 n/cm²-sec) is not as high quality as is desirable, it is expected that further efforts could substantially improve the image quality.

ADVANCED SOURCE DEVELOPMENT

The investigation of alpha photon source preparation using the ²⁴¹AmO₂-Cu sputtering target has been completed. Facilities improvements and experimental work of previous months are summarized and areas for future efforts are indicated. A new ²⁴¹AmO₂ sputtering target has been fabricated and it should permit the preparation of higher activity alpha photon sources having excellent radioisotope containment properties.

CIRCULATORY SUPPORT SYSTEMS

Neutron dose rate measurements with PNL 30-6 in the Rankine prototypic heat source system were terminated and the source was returned to LASL. IAEA sources were placed in the prototypic device for determination of the average photon absorption coefficient of the device materials. Work continues on modifying the blood-cooled heat exchangers which will be implanted in Hanford miniature swine.

NUCLEAR TECHNIQUES FOR SEABED MINERAL EXPLORATION

The seabed probe system fabrication has continued. In addition, an analysis program has been developed for the elements, S, Ca, Cl, Mo, Mg, Al, F, K, Na, In, Ga, Nd, Ni, Co, Cu, Ba, Sn, V, **I**, Eu, Ti, Nb, W, Sb, Dy, Ag, Os, Br, Rh, Pt, La, **Ir**, and As.

LOW-COST CESIUM AND STRONTIUM SOURCES

J. L. Ryan, D. H. Siemens, and 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 (MWHP). 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.

Plans for final purification of cesium by ARHCO have changed so that purified material will not be available until next fiscal year. Design, fabrication and procurement of equipment is being completed so that prototype capsules can be produced. Since purified cesium will not be available from ARHCO, the cost of prototype capsules will have to include cesium purification costs.

Seal welding of Type 304-L stainless steel (SS) test capsules made from 1 in. $\varnothing$ x 0.035 in. wall tubing and 1/16 in. thick end caps has been successfully demonstrated. Welding development was performed on 304-L SS while 316 SS was being procured. To perform the seal weld, the capsule is chucked in a horizontal turntable that has a fixed position guide 1 in. from the open end of the capsule to prevent runout. A torch holder is permanently attached to this guide to assure that the welding torch is correctly positioned. A standard gas-cooled TIG pencil torch is used. The following conditions have been established for welding the 304-L SS capsules:

welding speed, 0.037 in./sec; 16 amps, dc; gap, 1/8 in.; argon flow; 10 SCFH. Satisfactory seal welds were also made on capsules fabricated from Type 316 SS using the equipment and procedures that were developed for 304-L SS capsules.

The balance of the fiscal year will be devoted primarily to fabricating one or more demonstration sources, 1 in. diameter x 24 in. long, doubly encapsulated in 316 SS. After preliminary evaluations, the sources will be shipped to Brookhaven National Laboratory for more extensive use-testing. A summary document on the program to date is also to be completed this fiscal year.

The effect of impurities and the effect of decay of $^{90}\text{SrF}_2$ on the long-term integrity of containment of a $^{90}\text{SrF}_2$ heat source is being studied. The decay of ^{90}Sr produces stable ^{90}Zr . In a sealed system, decay of $^{90}\text{SrF}_2$ will produce $^{90}\text{ZrF}_2$ which, since Zr(IV) is the normal zirconium oxidation state, will either be stable as such or disproportionate to zirconium metal and ZrF_4 . The preparation of ZrF_2 has been reported by F. K. McTaggart and A. G. Turnbull, in Australian J. Chem., vol. 17, p. 727, 1964. They estimated a free energy of formation $\Delta G_{298^\circ\text{K}} = -213 \text{ kcal/mole}$ for this compound on the basis of lattice parameter calculations based on a reasonable estimate of 0.80 Å ionic radius for Zr(II). For ZrF_2 to be stable at 298°K, $\Delta G_{298^\circ\text{K}}$ must be at least as negative as -216 kcal/mole. Experimentally, they prepared ZrF_2 by reduction with atomic hydrogen and found it to be an extremely strong reducing agent, decomposing to ZrF_4 and zirconium at 800°C.

It is difficult to predict the physical effects of the grow-in of ^{90}Zr . The ionic radius of Zr(II) is much less than that of Sr(II), thus swelling of the SrF_2 would not be expected if Zr(II) is capable of simply substituting for strontium in SrF_2 . Because of the instability of ZrF_2 and the lack of closely related (4 d-group) difluorides, it would be difficult to determine or predict a possible phase diagram for SrF_2 - ZrF_2 . Since ZrF_2 apparently dissociates at 800°C, it seems likely that this would occur to a large extent in a $^{90}\text{SrF}_2$ source at elevated temperatures. As noted in

earlier monthly reports, ZrF_4 would be expected to produce a rather complex phase diagram with SrF_2 (although this phase diagram has not been measured). Compound formation and low melting eutectics are possible. The ionic radius of Zr(IV) is much less than that of Sr(II) but the metallic radius of zirconium is somewhat greater. If the Zr(IV) ions and zirconium metal atoms simply occupied Sr(II) sites, the net volume change due to ^{90}Sr decay would be negative. It seems almost certain that this would not be the case, however, and the defect structures produced by radioactive decay might or might not produce swelling of the SrF_2 .

The chemical effects of the formation of ZrF_2 on the corrosion of containment material (Hastelloy-C) are probably easier to predict. Since ZrF_2 and zirconium metal are both very powerful reducing agents, they may be expected to react with oxidizing impurities and thus act as a getter for these impurities. It would appear that the grow-in of ^{90}Zr may actually improve the containment of SrF_2 . Table 1 lists free energies of formation at 300° and 1100°K of fluorides and oxides of several elements including strontium and zirconium, elements present in Hastelloy-C, and possible impurities in the original $^{90}\text{SrF}_2$. These are given as kcal/g atom of fluorine or oxygen ion and are arranged in increasing order. Essentially, these are an electromotive series and any given compound will react with the free element (or sometimes lower oxidation compound) to produce any compound below it in the series. Thus, PbF_2 (a likely impurity in the $^{90}\text{SrF}_2$) can be expected to react with several of the metals present in Hastelloy-C. Also, H_2O and HF can be expected to react with several of these metals and if reaction with hydrogen or diffusion out of hydrogen also occurs, even metals above HF or H_2O may react. Thus, as shown in Table 1, zirconium occupies a desirable position. In Table 1 the data for ZrF_2 are estimated from the calculations and observed stability reported by McTaggart and Turnbull. Either ZrF_2 or zirconium metal is capable of reacting with all fluorides and oxides that could attack the containment material. On the other hand, they will not react with the alkali or alkaline earth fluorides (including strontium) to produce free metals. Although the data indicate

TABLE 1. Free Energy of Oxide and Fluoride Formation,

 ΔG° in kcal/g Atom of Anion

Fluorides				Oxides			
1100°K (827°C)		300 °K (27°C)		1100°K (827°C)		300°K (27°C)	
HgF ₂	-24	HgF ₂	-37	Cu ₂ O	-21	Ag ₂ O	-3
AgF	-36	AgF	-44	CrO ₃	-27	Co	-33
CuF ₂	-46	PF ₃	-54	PbO	-27	Cu ₂ O	-35
WF ₄	-47	WF ₄	-57	Cs ₂ O	-29	CrO ₃	-40
PF ₃	-48	CuF ₂	-58	As ₂ O ₃	-33	PbO	-45
SbF ₃	-54	AsF ₃	-63	CdO	-34	As ₂ O ₃	-45
MoF ₆	-57	MoF ₆	-64	Sb ₂ O ₃	-34	CO ₂	-47
SnF ₂	-58	HF	-65	NiO	-35	Sb ₂ O ₃	-49
PbF ₂	-58	SbF ₃	-66	CoO	-35	NiO	-51
NiF ₂	-58	SnF ₂	-72	MoO ₃	-43	CoO	-51
AsF ₃	-61	PbF ₂	-72	H ₂ O	-44	CdO	-55
CdF ₂	-61	BiF ₃	-72	Fe ₃ O ₄	-46	H ₂ O	-57
CoF ₂	-61	NiF ₂	-73	CO ₂	-47	MoO ₂	-59
BiF ₃	-64	CdF ₂	-73	WO ₂	-47	Fe ₃ O ₄	-61
ZnF ₂	-65	CoF ₂	-74	P ₂ O ₅	-48	WO ₂	-61
HF	-66	FeF ₂	-79	K ₂ O	-49	Cs ₂ O	-63
FeF ₂	-66	ZnF ₂	-82	CO	-50	P ₂ O ₅	-65
VF ₂	-71	CrF ₂	-86	ZnO	-56	Mn ₃ O ₄	-76
CrF ₂	-73	MnF ₂	-90	Mn ₃ O ₄	-60	ZnO	-76
MnF ₂	-77	VF ₂	-93	Na ₂ O	-62	K ₂ O	-77
BF ₃	-86	TiF ₃	-99	Cr ₂ O ₃	-67	Cr ₂ O ₃	-83
TiF ₃	-86	AlF ₃	-102	MnO	-73	MnO	-86
BeF ₂	-86	BeF ₂	-104	V ₂ O ₃	-77	Na ₂ O	-90
AlF ₃	-87	ZrF ₄	-106	B ₂ O ₃	-79	V ₂ O ₃	-93
ZrF ₂	a-91	UF ₃	-107	SiO ₂	-81	B ₂ O ₃	-94
ZrF ₄	-92	ZrF ₂	a-108	TiO	-99	SiO ₂	-99
UF ₃	-93	ThF ₃	-112	Al ₂ O ₃	-106	TiO	-116
ThF ₃	-98	CsF	-124	UO ₂	-107	UO ₂	-123
CsF	-102	MgF ₂	-125	ZrO ₂	-108	ZrO ₂	-124
KF	-104	KF	-127	BaO	-109	Al ₂ O ₃	-125
NaF	-108	NaF	-129	SrO	-115	BaO	-126
MgF ₂	-109	SmF ₃	-129	MgO	-115	SrO	-134
BaF ₂	-122	LaF ₃	-134	Ba ₂ O	-115	MgO	-136
SrF ₂	-123	BaF ₂	-136	BeO	-118	BeO	-136
CaF ₂	-123	SrF ₂	-138	La ₂ O ₃	-120	La ₂ O ₃	-136
		CaF ₂	-138	Sm ₂ O ₃	-120	Ba ₂ O ₃	-137
				ThO ₂	-122	Sm ₂ O ₃	-127
				CaO	-126	ThO ₂	-140
						CaO	-144

that zirconium will react with alkali oxides, oxide will only be an impurity in the system and reactions such as $\text{ZrF}_4 + 2\text{Na}_2\text{O} \rightarrow \text{ZrO}_2 + 4\text{NaF}$, $\Delta G_{1100^\circ\text{K}} = -156 \text{ kcal/mole}$, will prevent the existence of Na_2O . Thus, it appears that zirconium, being approximately intermediate between SrF_2 and the fluorides of the containment alloy, is nearly an ideal getter.

Thermodynamic data are also being accumulated for compounds such as hydrides and sulfides to determine how well zirconium will act as a getter, since hydrogen and sulfur, for instance, will result from decomposition of H_2O , SO_4^{2-} . The data indicate that zirconium will also be an effective getter for Cl^- impurity. The corrosiveness of free elements such as lead (Pb) that are produced by the reduction of impurities will also be investigated by a metallurgist. Since ZrF_2 is so unstable relative to ZrF_4 (thus having a ΔG per gram atom F^- about the same as ZrF_4) and since these are both well removed from the Hastelloy component, it appears that fluoride free energies would prevent any cyclic carrying process whereby one fluoride of zirconium would attack the metal at one temperature to produce a zirconium fluoride which would migrate and decompose at another temperature.

Thermodynamic calculations for reactions of the zirconium decay product of $^{90}\text{SrF}_2$ with impurities in the SrF_2 were extended to include anionic and neutral impurities that might be present, such as nitrate, carbonate, water, sulfate, and phosphate.

Any nitrate present would be decomposed by zirconium metal or the very unstable ZrF_2 . Based on the thermochemical data for oxides given in Table 1, zirconium would act as a getter for the oxide ions produced and prevent their interaction with containment materials. The nitrogen of the nitrate ion will be converted to nitride and this again will be gettered by the zirconium as shown from the free energies of formation of nitrides of zirconium and Hastelloy-C components listed in Table 2.

TABLE 2. Free Energy of Nitride Formation,
 ΔG° in kcal/g Atom of Nitrogen

	<u>298°K</u>	<u>800°K</u>	<u>1000°K</u>	<u>1100°K</u>
Fe ₄ N	+0.9		+9.5	
W ₂ N	-11.0			
Mo ₂ N	-11.97	-4.4		
Mn ₅ N ₂	-18.7	-10.3		
CrN	-23.65	-14.25		
ZrN	-80.5			-62.5

Carbonate will also be reduced by zirconium to oxide and carbide. The reaction:

has a free energy at 1100°K of about -143 kcal/mole of carbonate. Zirconium will also getter the carbide relative to the components of Hastelloy-C as shown in Table 3.

TABLE 3. Free Energy of Carbide Formation,
 ΔG° in kcal/g Atom of Carbon

	<u>298°K</u>	<u>1100°K</u>
Co ₃ C	+9.0	
Ni ₃ C	+8.9	+5
Fe ₃ C	+4.8	+0.45
Mo ₂ C	+2.97	-1.10
Mn ₃ C	-3.5	-3.6
WC	-8.8	-7.8
Cr ₄ C	-16.75	-18.15
ZrC	-43.45	-41

The reaction of zirconium with water:

has a free energy at 1100°K of -128 kcal/mole zirconium. The zirconium can be expected to be an effective getter for the hydrogen. The free energy of formation of ZrH_2 is -29.32 kcal/mole at 298°K and is -6.77 kcal/mole at 1100°K. The free energy of formation of Ni_2H is +5.64 kcal/mole H_2 at 298°K and is about +26.5 kcal/mole H_2 at 1100°K. Although data are not available for other constituents of Hastelloy-C, the literature indicates that their hydrides are no more stable than that of nickel. However, some of these metals in their pure state do show slight solubility of hydrogen at 1100°K. This amounts to from 0.0001 to 0.0004 atoms H/atom of metal.

Sulfate will also be reduced by zirconium. In the presence of excess ZrF_4 (or ZrF_2 which decomposes to Zr and ZrF_4), the net reaction will be:

having a free energy at 1100°K of about -284 kcal/mole SrSO_4 . In the absence of ZrF_4 the reaction would be:

having a free energy at 1100°K of about -260 kcal/mole SrSO_4 . Although it is certain that neither ZrS_2 nor SrS can act as a sulfiding agent on components of Hastelloy-C, the free energies of formation of their sulfides are not as yet tabulated.

The reaction of zirconium with $\text{Ca}_3(\text{PO}_4)_2$

has a free energy at 1100°K of about -114 kcal/mole $\text{Ca}_3(\text{PO}_4)_2$. Although zirconium phosphides are known to form, no thermodynamic data for them have been found. Data are not readily available for $\text{Sr}_3(\text{PO}_4)_2$.

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.

The Linde Division of the Union Carbide Corporation received the first delivery of xenon purified under this project. Linde will evaluate the material for large scale uses such as long-lived incandescent lamps. Airco has also expressed interest but has not found a large-scale use that would justify a request for material.

University of California personnel have shown increasing interest but would require material of much less krypton-85 content. Material meeting their purity requirement may be easier to produce than initially expected. The requirements, which their development indicates, would be very large for widespread use in specialized radiation detectors.

FISSION PRODUCT RHODIUM, PALLADIUM,
RUTHENIUM, AND TECHNETIUM UTILIZATION

F. P. Roberts

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.*

TECHNOLOGY FOR RECOVERY OF Rh, Pd, Ru and Tc

The search was continued for a process to isolate Rh, Pd, Ru, and Tc, as a group, from the other fission products in the high-level wastes from nuclear fuels reprocessing. Laboratory experiments were carried out to evaluate the precipitation of these elements as sulfides from a synthetic high-level waste expected from processing spent nuclear fuels. In these experiments the aqueous waste was saturated with gaseous hydrogen sulfide. The resulting precipitate was dissolved and analyzed to determine the yield of Rh, Pd, Ru, and Tc. Radioactive tracers were used for following the Rh, Ru, and Tc. Palladium was analyzed gravimetrically using dimethylglyoxime as a precipitant.

In these experiments, normal concentrations of nitric acid in the aqueous waste (4 to 5M HNO_3) caused the formation of massive amounts of elemental sulfur which made it difficult to recover the precipitate by

filtration. However, this problem was overcome by removing most of the nitric acid which was accomplished by treating the waste with formaldehyde. When the residual nitric acid was 1.0M or below, a filterable precipitate was obtained for Ru. See results in Table 1.

TABLE 1. Recovery Yields of Rh, Pd, Ru, and Tc by Sulfide Precipitation

<u>Element</u>	<u>% Recovered</u>
Rh	82
Pd	89
Tc	77
Ru	43

The use of sulfide precipitation to isolate the platinum metal group presents an obvious drawback in that some of the sulfide is oxidized to sulfate. The sulfate which is formed causes the precipitation of strontium along with the platinum metals. A second separation step to remove strontium would be desirable if long-term storage of the platinum metals to allow decay of the radioactive isotopes, Rh and Ru, is included as part of the recovery process.

MARKET ANALYSIS FOR Rh, Pd, Ru, AND Tc

Study of the economic factors associated with the use of the fission products Rh, Pd, Ru, and Tc has been initiated. This study will attempt to estimate the costs related to recovery, interior storage to allow decay of the radioactive isotopes of Rh and Ru, separation and purification of the individual platinum metals and the special handling operations required because of the radioactivity associated with the fission-product platinum metals. The results of this study will be included in this report on the market analysis for Rh, Pd, Ru, and Tc.

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.

The current effort on neutron radiography at Battelle-Northwest is drawing to a close and this report summarizes the progress during the past 18 months. Technical details of the efforts can be found in past monthly and quarterly reports and in open literature publications which are referenced in this report. The effort has been divided into three areas of development and evaluation: exposure facilities, film-converter imaging, and electronically aided imaging.

EXPOSURE FACILITIES

To utilize isotopic sources for thermal neutron radiography requires a means of moderating the source neutrons and a means of extracting a beam of thermalized neutrons. The same facility is generally expected to provide the necessary radiation shielding. This implies a fairly massive shield and a means of blocking the thermal neutron beam during film changes. Three such facilities have been developed at Battelle-Northwest for use with ^{252}Cf .

Since neutron radiography requires extraction of a beam of neutrons from the exposure facility, a means of isolating this beam from occupied areas must be provided during exposure by either shielding or distance. To meet these requirements, all of our experiments are performed in an underground room which provides adequate shielding to occupied areas.

The first neutron radiography facility (NC-1) was described in Californium-252 Progress⁽¹⁾ and in a Battelle report.⁽²⁾ It was noted in previous program reports that the collimation and the gamma filter or moderator can be changed in the facility. Table 1 shows the effect of changing the materials in the beam tube. Comparison of the flux levels and neutron-to-gamma ratio shows that it would be preferable to operate without added materials for transfer radiography (highest flux) and that a ZrH_2 moderator with a uranium gamma filter would be preferred for direct radiography (best neutron-gamma ratio). Figure 1 shows measured flux levels and calculated unsharpness when collimators of the same inlet diameter, but different lengths, are used with NC-1. It is apparent that improved resolution requires a reduction in the thermal flux and a consequent increase in the time needed to form an image.

The second exposure facility (NC-2)⁽³⁾ uses the same external fiberglass shell used for NC-1; however, it uses a fixed collimator and a water moderator. Figure 2 shows the internal arrangement of NC-2 with its fixed collimator of 15:1. An off-center source geometry with an air-gap collimator is used to take advantage of the improved beam extraction offered by this technique.⁽⁴⁾ With a water moderator it is possible to position the fiberglass shell without the moderator and then fill it with water and install the source. To permit the handling of large sources, the source could be transferred remotely from a storage shield to the pre-positioned NC-2. When small sources are used, the imaging beam is blocked by filling the collimator with water. This is done remotely and permits handling of objects in the exposure position. Table 2 summarizes and compares the features of NC-1 and NC-2.

TABLE 1. Effect of Various Moderators and Gamma Filters on the Beam Flux and Gamma Ratio

Moderator	Gamma Filter(a)	Flux (n/cm ² -sec) (x 10 ³)	Neutron-Gamma Ratio (n/cm ² /mR) (x 10 ⁴)	Cadmium Ratio(c)
None	None	1.48	3.6	2.00
Paraffin	Pb-1(b)	1.43	5.15	2.60
Paraffin	U-1	~1.4	6.28	----
ZrH ₂	None	1.19	5.95	2.32
ZrH ₂	Pb-1	0.941	5.46	2.0
ZrH ₂	Pb-2	0.959	6.28	2.10
ZrH ₂	U-1	1.05	6.30	2.15
ZrH ₂	U-2	1.19	5.95	2.10
ZrH ₂	U-1	1.22	7.30	2.43
TiH ₂	None	0.542	2.38	1.56
TiH ₂	Pb-1	0.562	2.80	1.65

a. Gamma filter on film side of moderator, others on source side

b. Pb-1 = 0.5 cm; Pb-2 = 1.0 cm; U-1 = 0.44 cm; U-2 = 0.88 cm

c. Ratio of bare indium response to cadmium covered response.

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FIGURE 1. Thermal Flux and Calculated Geometrical Unsharpness for the Radiography Beam from NC-1; Operation with a Paraffin Moderator and 0.1 in. Lead Gamma Filter

TABLE 2. 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_2O
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 to 180 mR/hr

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FIGURE 2. Internal Configuration of a Source Holder and Exposure Facility (NC-2) Designed for Ease of Positioning. The unit uses a liquid moderator (H_2O) which can be removed during positioning.

Unlike NC-1 and NC-2, the third unit⁽³⁾ is stationary and employs a large source (1.7 mg) of ^{252}Cf . The facility, shown in Figure 3, is simply a large water tank 4 ft x 4 ft x 3 1/2 ft deep. Plywood with a fiberglass liner was used to construct the tank and additional shielding in the form of concrete blocks is positioned around the outside of the tank. A stepping motor is used to position the source remotely at any level up to 18 in. above the surface of the water. The unit is an extremely versatile source of neutrons. When the source is in the stored position, the surface dose rates are 4 mrem/hr neutrons and 10 mrem/hr of gamma. With a collimation ratio of 15:1, a flux of $9.5 \times 10^3 \text{ n/cm}^2\text{-sec}$ is obtained. This is with a collimator positioned vertically in the tank of water and a 0.200 in. lead gamma filter. Collimation ratios of up to 40:1 ($1 \times 10^3 \text{ n/cm}^2\text{-sec}$) are used with this facility.

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FIGURE 3. Large Water Tank Containing a 1.7 mg Source of ^{252}Cf . The unit is designed for development of neutron radiography with ^{252}Cf , but is also a versatile source of neutrons.

FILM-CONVERTER IMAGING TECHNIQUES

The second portion of the program at Battelle-Northwest involves the development of imaging techniques. Since most of this information has been presented elsewhere,^(3,4) only a short summary is included here.

Figure 4 shows density versus exposure for various film-converter combinations exposed in a flux of $2.85 \times 10^2 \text{ n/cm}^2\text{-sec.}$ ^(3,4) The rare earth scintillator, gadolinium oxysulfide ($\text{Gd}_2\text{O}_2\text{S:Tb}$), which was developed as an X-ray phosphor, because of the high gadolinium content has been found useful for neutron radiography. Although $\text{Gd}_2\text{O}_2\text{S:Tb}$ is intermediate in speed between the gadolinium metal foil and $^6\text{LiF-ZnS(Ag)}$, the resolution obtainable is significantly better than $^6\text{LiF-ZnS(Ag)}$ and with low collimation ratios it is comparable to the gadolinium foils. (4)

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FIGURE 4. Radiograph Density Versus Exposure for Various Imaging Techniques

Recent experiments with Polaroid Type 57 film have resulted in an increase in speed of $\sim 2\frac{1}{2}$ times when compared with blue sensitive X-ray film. The Polaroid film is a high-speed pancromatic film providing improved response to the spectrum of the emission from the gadolinium oxy-sulfide compared to blue sensitive film.

The application of indirect radiography (transfer imaging) has been limited by the low-fluxes available from ^{252}Cf , since the induced activity in a transfer screen saturates after a few half-lives and longer exposures cannot be used to compensate for low beam intensities. Europium metal foils have been developed for transfer imaging along with the use of calcium tungstate screens to enhance transfer images from both europium and dysprosium. (5) Figure 5 shows exposure curves for europium foils with and without a calcium tungstate intensifying screen. The use of the intensifying screen with a dysprosium foil allows one to obtain a radiographic density of 1.5 in a flux of $9.5 \times 10^3 \text{ n/cm}^2\text{-sec}$ compared to 0.95 without the screen. With the low collimation ratios employed, improved contrast is obtained without any detectable loss in resolution.

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FIGURE 5. Radiograph Density Versus Flux Level for Europium Transfer Imaging Using Kodak RP/R Film with and without Calcium Tungstate (CaWO_4) Intensifying Screen

ELECTRONICALLY AIDED IMAGING

A prototype position-sensitive proportional counter was investigated for use in neutron imaging applications.⁽⁶⁾ The counter uses a converter foil of ^6LiF and a collimation grid to achieve improved resolution compared to counters filled with a neutron-sensitive counting gas. The proportional counter consists of a grid of 36 1-mil pyrolytic carbon coated quartz fibers on 2.5 mm centers. Because of the high resistance of the collector wires and the distributed capacitance to ground, the rise time of a pulse will increase with distance along the collector. The rise-time information is used to determine the position of an event to accuracies less than 1 mm along the wire. Standard nuclear instrumentation techniques are used with readout on a multichannel pulse height analyzer or on a storage scope. The advantages of an imaging counter are in the digital storage of information (multi-channel pulse height analyzers) and in the ease of adapting existing hardware for computer analysis.

Tests performed with alpha particles showed that a geometry using collector wires off-center by 0.5 mm produced the best resolution. The best resolution achieved was 0.59 mm FWHM, but the off-center geometry was not used for the neutron imaging experiments because of difficulties in achieving stable operation. Neutron images produced by this counter showed an unsharpness of 5.5 mm (3.3 mm FWHM) and a contrast of 55%; this was with a ^6LiF converter foil (100 μ). Images with ^{10}B and Gd converter foils showed unsatisfactory contrasts. Efforts have shown that an unsharpness of about 1 mm is possible. The prototype counter has an efficiency of 0.25% which is about one-half the calculated efficiency. Figure 6 shows an image produced with the counter in a thermal neutron flux of $1.6 \times 10^3 \text{ n/cm}^2\text{-sec}$. The object is a cadmium test piece with 1 cm and 0.5 cm diameter holes which are visible in the photograph.

Resolution with the imaging counter is better than could be obtained with a gas-filled detector (BF_3) at 2 atm pressure (6.2 mm FWHM); however, the efficiency is lower by a factor of 25. The counter will not meet the

imaging speed or resolution of an image intensifier system (0.25 mm FWHM; 4×10^5 n/cm²); however, most intensifiers are not useful at low fluxes.

FIGURE 6. Photograph of the Thermal Neutron Image of a Cadmium Test Strip with 1 cm and 0.5 cm Diameter Holes

A special image intensifier with integrating capability was evaluated with a $^6\text{LiF-ZnS}$ conversion screen. It took an exposure of 120 sec in a flux of $7 \times 10^2 \text{ n/cm}^2\text{-sec}$ to produce a suitable image that could resolve 1 mm diameter holes in a cadmium test strip. The system was not optimized in this work; however, better resolution and higher speed could be obtained.

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ADVANCED SOURCE DEVELOPMENT PROGRAM

G. G. Culver and J. H. Jarrett

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 activities of this program in past months are summarized, and recommendations are made for future effort. The discussion is divided into a section on source preparation facilities and a section on completed and planned experimental work. Mr. G. G. Culver, Assistant Professor of Mechanical Engineering Technology, Oregon Technical Institute, has been a NORCUS appointee assigned primarily to this program for the past nine months, with assignment completion expected in August 1972. He has contributed significantly to the program and because of the NORCUS program arrangement he has incurred costs to DAT only for materials and supplies.

FACILITIES

Improvements in our experimental capabilities for preparing thin film radiation sources by rf sputtering are being made on a continuing basis. The major improvements are listed below:

<u>Improvement</u>	<u>Benefit</u>
<ul style="list-style-type: none"> • Residual Gas Analysis (RGA) System 	<p>The RGA system permits analysis of gaseous impurities in the system before, during, and after a sputtering run, allowing better quality control and offering potential for system automation.</p>

- Humidity Control System
(Installation 90% Complete)

Complete installation is expected in late May 1972. The system will maintain low humidity (below 30%) and constant laboratory temperature ($72 \pm 2^\circ\text{F}$). This will help minimize introduction of water vapor into the vacuum systems and eliminate condensation on water-cooled rf power leads.

- Network Cooling for Glovebox Sputtering System

This improvement has increased the reliability of the glove system, allowing operation without constant surveillance and minimizing system down-time.

- High Activity $^{241}\text{AmO}_2$ Sputtering Target (The Source of Sputtered Material)

The new target allows the deposition of layers containing over 80% ^{241}Am and also allows the preparation of metal- ^{241}Am oxide cermet layers in which the metal and its concentration may be varied. This target replaces a $^{241}\text{AmO}_2$ -Cu target of fixed composition that gave sputtered radioactive layers of less than 5% ^{241}Am .

Several facilities improvements are planned, based on the results of experimental work to date and on planned work. These should lead to better and more reproducible source preparation runs. The following improvements are projected:

Planned Improvement	Benefit
<ul style="list-style-type: none"> ● Clean Vacuum Systems; Provide Cold Traps 	<p>Based on RGA data and discussions with vacuum technology experts, it is apparent that amounts of vacuum pump oil that may have a significant effect on experimental results are present in the sputtering chamber. Completion of this improvement will minimize the chamber contaminants originating from diffusion pump oil and mechanical pump oil.</p>

- $^{241}\text{AmO}_2$ Target Handling and Storage Facility

The quality and outgassing characteristics of the $^{241}\text{AmO}_2$ target will be maintained by minimizing its contact with moisture and air (storage in an inert gas dessicator or in vacuum), and appropriate radiation protection devices for target handling will be fabricated.
- System Automation

Operator time will be reduced and system operating conditions will be recorded automatically, providing a permanent record. Improvements in this area will be accomplished in phases, with evaluation of each degree of automation.
- Laminar Flow Assembly Area

The cleaning and assembly of hardware to be placed in the sputtering system should be done under carefully controlled conditions. Using a laminar flow of filtered air across the work area provides the necessary conditions and should contribute to an increased proportion of high integrity sources in each run.
- Miscellaneous System Improvements - Assure dryness of glovebox air, improve rf grounding.

These improvements will increase experimental quality.

EXPERIMENTAL WORK

In activities oriented toward the preparation by rf sputtering of high integrity thin-film radiation sources having a $^{241}\text{AmO}_2$ -Cu active layer, several closely related areas have been investigated. The results of this work are summarized below:

<u>Experimental Area</u>	<u>Results, Recommendations</u>
<ul style="list-style-type: none"> Selection of Substrate and Containment Layer Materials for Sources Designed to Withstand Mechanical Abuse and Heating at 1700°F in Air. 	<p>The oxidation resistance, mechanical properties and cost of 304 stainless-steel make it suitable for use as the 0.060 in. thick substrate material. Other, more costly alloys (Inconels,</p>

(Continued-Selection of Substrate and Containment Layer Materials for Sources Designed to Withstand Mechanical Abuse and Heating at 1700°F in Air.)

- Development of a Mask for Positioning the Active Layer on the Source Substrate

- Source Preparation Conditions and Procedures

Hastelloys and stainless steels) and elements (such as platinum) were considered and have appropriate properties, but are generally more costly than 304 stainless steel. Similar materials were considered for the containment layer which is less than 0.0002 in. (5 microns) thick. Platinum was chosen because of its excellent oxidation resistance. Inconel 750 has good oxidation resistance and is recommended along with ceramic materials for further investigations.

Several mask materials (mild steel, stainless steel, quartz and aluminum) and mask designs have been investigated. Charge build-up and insufficient grounding were indicated in the tests. The necessary modification of the sputtering systems is planned. A 304 stainless steel mask assembly is presently recommended with provision for handling and outgassing the mask remotely in the sputtering chamber during a source preparation run to allow the sequential deposition of the active layer and the containment layer without breaking vacuum. The operation of such an assembly is shown in Figures 1, 2, and 3.

Operational data are being compiled on the large number of variables that affect the preparation of high integrity, high activity radiation sources by rf sputtering, and high quality sources have been prepared. The following general procedures should be followed:

1. Clean substrates and masking hardware thoroughly.
2. Clean the sputtering chamber periodically and maintain chamber cleanliness by minimizing backstreamed pump oil, and contact of chamber components with air and water.

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FIGURE 1. Mask Assembly in Place for Sputter Deposition of Radioactive Layer. Ten radiation ,sources are prepared per run.

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FIGURE 2. Removal of Mask in Preparation for Containment Layer Deposition

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FIGURE 3. Mask Fully Removed for Deposition of Containment Layer

Experimental Area (contd)
(Continued-Source Preparation Conditions and Procedures)

- Results, Recommendations (contd)
3. Minimize dust on substrates, hardware and chamber components.
 4. Pump system down for several hours prior to sputtering to a base-⁻⁷ pressure near or below 4×10^{-7} torr. Backfill with Ar and re-evacuate at least two times.
 5. Outgas the mask hardware, sputter-clean the 5 in. diameter ²⁴¹AmO₂-Cu target and sputter etch the substrates at 200 W for 5 min.
 6. Deposit the radioactive layer at 500 W (25 W/in.²), remove the mask and then deposit the Pt containment layer.
 7. Backfill the chamber with Ar and remove the sources without adding glovebox contamination to them.

Experimental Area	Results, Recommendations
<ul style="list-style-type: none"> • Preparation of Sources Having a Sputtered $^{241}\text{AmO}_2$-Cu Active Layer 	<p>Early in this program a $^{241}\text{AmO}_2$-Cu cermet sputtering target was fabricated for the production of alpha photon and photon radiation sources. Several of these sources of different designs have been tested by ORNL in their DAT Source Safety Testing Program. Some high quality thin-film sources were prepared, but even better sources should be possible by making the facilities improvements, and by following the source preparation procedures already discussed; and by using the new $^{241}\text{AmO}_2$ sputtering target. Detail on these three areas has been given in the preceding sections. Our results have indicated that the $^{241}\text{AmO}_2$-Cu system is very sensitive to impurities such as air, water and probably backstreamed pump oil. It is also noted that the $^{241}\text{AmO}_2$-Cu sputtering target should near theoretical density to minimize outgassing problems.</p>

Experimental work planned for future effort will emphasize the preparation of alpha photon sources using the new $^{241}\text{AmO}_2$ sputtering target. Higher activity sources with excellent ^{241}Am containment are expected. Different materials systems, including ceramic materials and different $^{241}\text{AmO}_2$ -metal cermet combinations, will be investigated. It is also anticipated that a continued awareness of potential source applications will be maintained, and that work will be started on the economics of preparing sources using rf sputtering.

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 50-W plutonium heat sources.

DOSIMETRY STUDIES

Neutron dose rate measurements with the plutonium oxide (PNL 30-6) source in the Hittman device (Rankine prototypic heat source system) were terminated. PNL 30-6 was packaged in container DOT-SP-6286 for return shipment to LASL.

IAEA photon calibration standards were placed in the Hittman device for determination of the average photon absorption coefficient of the device materials. The "homogenized" device material determined from these measurements will be used to simplify future dose rate computations.

RADIOLOGICAL ENGINEERING STUDIES

Work continues on modifying the blood-cooled heat exchangers in order to improve their reliability and simplify their implantation in Hanford miniature swine.

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.

Major emphasis this past quarter has continued on the preparation of basic software for the seabed analyzer probe and on the preparation of the analyzer for its sea trials. A set of programs has been designed to allow testing of the software and hardware systems. These versions allow the operator to determine elemental concentrations on the seafloor. The programs also allow storage of data on magnetic tape for further study by the investigator. The set of basic software programs is 60% written and allows the analysis of S, Ca, Cl, Mo, Mg, Al, F, K, Na, In, Ga, Nd, Ni, Co, Cu, Ba, Sn, V, **■**, Eu, Ti, Nb, W, Sb, Dy, Ag, Os, Br, Rh, Pt, La, **Ir**, and As. This analysis scheme utilizes a 2-min irradiation, a 1-min decay and a 2-min count followed by a 3-min count with the gamma-ray analysis unit.

The resolution of the delivered Ge(Li) detector package is 2.8 keV which is less than the specified resolution of 2.5 keV. Methods such as matching amplifier DC levels and time constant variations are being evaluated to improve the detector performance at the end of its 1000-ft undersea cable. The closed circuit television system has also been evaluated. It allows visual monitoring of a 20 in. by 30 in. area from its position, 40 in. above the sea floor. The system's remote focus allows the seabed to be visually monitored while the probe is being lowered to and is maneuvering on the sea floor. This video system has not yet been tested to its designed depth of 1000 ft nor through its 1000-ft cable linkage to the surface. The final hardware for coupling the Ge(Li) diode and ^{252}Cf assembly to the seabed probe are being fabricated. The 1000 ft of undersea cable, the reel, and the ^{252}Cf shipping cask have been received.

In addition to preparation of the electronic package and continuing fabrication of the in situ nuclear probe components, sets of ocean sediment samples, phosphorite, and sea sand were neutron irradiated and analyzed for the previously mentioned elements as well as for those that might be detected by varying the neutron irradiation conditions. Ocean sediments that contain Na, Fe, La, Sm, Br, Co, Sc, Cr, Eu, Hg, Th, Tb, Ta, Cs, Sn, Sb, and Zn do not mask detection of some of the more important elements such as Mn, Co, and Cu from deposits such as manganese nodules. Sea sands were studied for their elemental content to determine which elements might interfere in the in situ activation analysis of inert tracers which were added to study sand movement in a littoral system or through a continental shelf region. For example, Dy could be added to sand as a rare earth ore such as gadolinite and the seabed probe used to activate and follow its movement in tideland or deeper ocean areas. Although the rare earths La, Eu, Sm, and Ba were detected with long irradiations, $^{165\text{m}}\text{Dy}$ (74 sec) could not be detected in the sand and in the short irradiation and counting procedures which would be practical for tracer studies on the ocean floor. The phosphate mineral,

phosphorite, was also analyzed. Although phosphorus cannot be detected directly, **it** may be possible to detect **it** from its characteristic trace elements which can be easily measured. Therefore, a correlation study of its minor elements has been undertaken to investigate the possibility of using both actual concentrations and element ratios as an identification methods.

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