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**Pacific Northwest Laboratory
Quarterly Report to
Advanced Nuclear Energy
Systems, Space and Special
Purposes Division**

June 30, 1975

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PACIFIC NORTHWEST LABORATORY QUARTERLY REPORT
TO ADVANCED NUCLEAR ENERGY SYSTEMS,
SPACE AND SPECIAL PURPOSES DIVISION

J. H. Jarrett - Project Manager
K. M. Harmon - Principal Investigator

June 30, 1975

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SUMMARY

1. Preliminary conceptual and engineering analysis studies were made of a proposed U.S. Air Force 2kW(e) power system employing WESF $^{90}\text{SrF}_2$ capsules as a source of heat. The results showed the feasibility of such a system, but indicated that the SrF_2 capsule materials might have to be changed to satisfy operating life requirements. A draft RFP for the development and testing of a prototype 2kW(e) system was prepared and submitted to ERDA/DANES for review.
2. Dose rates from a WESF $^{90}\text{SrF}_2$ capsule were measured. Dose rates and radiation efficiencies for WESF $^{137}\text{CsCl}$ capsules of varying geometries were calculated.
3. A forecast of the potential supply of a number of beneficial reactor by-products was assembled.
4. A start was made on the definitions of a flow sheet for an integrated reactor by-product recovery plant.
5. Preliminary work was done on the methodology for forecasting long-term demand for reactor by-products.

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UTILIZATION OF NUCLEAR BY-PRODUCTS

J. H. Jarrett - Project Manager
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INTRODUCTION

The potential for beneficial use of the by-products of nuclear reactors has been studied in the United States under government and industrial sponsorship for many years. Early in FY-1975, the Pacific Northwest Laboratory (PNL) was commissioned by the USAEC to evaluate the potential use of radioactive isotopes to supply heat and power for various applications in the Cold Regions. In the spring of 1975, the scope of the PNL FY-1975 program was broadened to include the following additional activities:

1. Coordinate and perform work on the beneficial use of $^{90}\text{SrF}_2$ and $^{137}\text{CsCl}$ products from the Waste Encapsulation and Storage Facility (WESF) at Hanford. (WESF is operated for ERDA by the Atlantic Richfield Hanford Company, ARHCO.)
2. Estimate the potential supply of isotopes from power reactors through the year 2000, assess the potential demand for these isotopes, and evaluate the incentives for private industry to build and operate isotope recovery and encapsulation plants.

This report summarizes the work done during the fourth quarter of FY-1975 on the rest of the program. The Cold Regions work for FY-1975 is summarized in a report being prepared (BNWL-1935).

TECHNICAL PROGRESS

Beneficial Use of WESF $^{90}\text{SrF}_2$ Capsules: Air Force Power Source

The Air Force has a potential requirement for a highly reliable 2 kW(e) electrical power system that will operate unattended for extended periods in their North American air defense network. This need could probably be satisfied by a Stirling or organic Rankine engine powered by a radioisotope heat source. PNL has undertaken the correlation of a program to develop and test such a system, powered by WESF ^{90}Sr and using "off-the-shelf" components insofar as possible. PNL responsibilities include the following:

- Evaluate the feasibility of using WESF production $^{90}\text{SrF}_2$ capsules (Figure 1) in the heat source. If the capsule as presently designed will not meet heat source requirements, determine what changes need to be made in capsule design and work with ARHCO and ERDA to get them implemented
- Prepare and issue (through the Richland Operations Office) an RFP calling for design of an electrical power system; selection, development and testing of system components; and assembly and testing of prototype and field test systems
- Coordinate onsite (at Hanford) testing of prototype and field test assemblies

Power Systems - Heat Transfer Feasibility Study (D. H. Lester, Chemical Development)

Preliminary studies of the compatibility of SrF_2 with WESF capsule materials have indicated that system design should be such that SrF_2 -metal interface temperatures will not exceed 800°C (and may have to be significantly lower). This study was undertaken to determine if established coolant conditions (temperature and flow rate) for organic Rankine and Stirling engines were compatible with WESF capsule dimensions and with an 800°C temperature constraint. The study was performed by calculating temperature profiles from coolant to capsule centerline for system design concepts based on: 1) individually cooled capsules (organic Rankine system

FIGURE 1. Waste Encapsulation and Storage Facility Capsules

only) and 2) multiple capsules inserted in a nickel heat block (both systems). Coolant temperatures were assumed to be 338°C for the organic-cooled engine and 649°C for the helium-cooled engine. A thermal emissivity of 0.5 was assumed for all surfaces. Working fluid pressure drops were also calculated to determine percent thermal power used for pumping. No attempt was made in this early work to calculate thermal losses since the system design would be the major factor. Losses would be evaluated in future detailed designs.

Organic Rankine Cycle. A bare capsule, as fabricated, was compared with a finned capsule. The small heat transfer area of the bare capsule presents a difficult situation since the organic fluid offers film coefficients of about 1 Btu/hr-ft²-°F or less. Table 1 shows how using fins solves the area problem. The SrF₂-metal interface temperatures are well below the maximum (800°C) for both types of fins and well above the maximum for the bare capsule.

TABLE 1. Temperatures in Organic-Cooled, Single Capsules

Location	Bare Capsule		Longitudinal ^(a) Fins		Annular ^(b) Fins	
	(°F)	(°C)	(°F)	(°C)	(°F)	(°C)
Coolant	640	338	640	338	640	338
Capsule Surface	3796	2091	762	405	675	357
Liner Surface	3820	2104	1247	675	1218	659
SrF ₂ -Metal Interface	3824	2107	1251	677	1222	661
Centerline	4606	2441	2033	1112	2004	1095

(a) 10 fins, 0.15 in. thick, 2 in. high

(b) 40 fins/ft, 0.2 in. thick, 2 in. high

The capsules can be used without adding fins by placing them in a nickel block with coolant channels surrounding each capsule (Figure 2) and using some type of liquid metal bonding (Table 2). The high conductivity [32 Btu/hr-(ft²/ft)-°F] nickel serves the same purpose as fins. The liquid metal is used to fill the gap between the capsule and the block. In this concept, each capsule and its associated coolant channels occupy a section of solid nickel which is 7.5 in. in diameter and 20 in. long. The capsules for a 10 kW thermal source could be placed in a 60 x 20 x 20-in. block of nickel.

FIGURE 2. Nickel Block Concept

TABLE 2. Temperatures in Organic Cooled Nickel Block Concept^(a)

Location	Air Gap Clearance		Sodium Bonded	
	(°F)	(°C)	(°F)	(°C)
Coolant	640	338	640	338
Coolant Wall	1053	567	1053	567
Capsule Surface	1350	732	1062	572
Liner Surface	1572	856	1388	753
SrF ₂ -Metal Interface	1577	858	1392	755
Centerline	2358	1292	2174	1190

(a) 40 coolant channels, each 0.50-in. diam, arranged around capsule on a 6.5-in. diam.

Flow = 6 lb/hr per capsule

Stirling/Helium Cycle. The block concept was used in studying the feasibility of the Stirling/helium system. As evident in Table 3, liquid metal bonding is likely to be necessary to obtain sufficiently low temperature gradients. The interface temperature calculated for the design used

TABLE 3. Temperatures in Helium Cooled Nickel Block Concept^(a)

Location	Air Gap Clearance		Sodium Bonded	
	(°F)	(°C)	(°F)	(°C)
Coolant	1200	649	1200	649
Coolant Wall	1264	685	1264	685
Capsule Surface	1484	806	1267	686
Liner Surface	1671	911	1515	824
SrF ₂ -Metal Interface	1675	913	1519	826
Centerline	2457	1347	2301	1261

(a) 15 coolant channels, each 0.50-in. diam, arranged around the capsule on a 4.0-in. diam.

Flow = 5 lb/hr per capsule

Pressure Drop = 80 psi at 1200°F 200 atm

as a basis for the study are slightly above the prescribed maximum. Heat block design modifications would easily lead to the goal temperature. Each capsule would occupy an envelope about 4.5 in. in diameter by 20 in. long. Ten capsules (10 kW thermal) could be placed in a 20 x 14 x 20-in. nickel block.

Preparation of RFP (E. E. Warner, Consultant and D. Frieling, Battelle-Columbus)

A draft RFP was assembled and transmitted to DANES/ERDA for review. The work statement in the RFP includes the following major activities:

- Phase I - System definition
- Phase II - Design, development, and test (of components and of the prototype, P1)
- Phase III - WESF Prototype testing (1-year life test of P1 at Hanford)
- Phase IV - Qualification testing (second prototype and field units)
- Phase V - Production

WESF SrF_2 Capsule Dosimetry (R. L. Libby and F. N. Eichner)

Dose measurements were made on a current-production WESF SrF_2 waste capsule (No. 5-48) with a ^{90}Sr isotopic content of about 25%. Using TLD-700 (LiF) chips, 60-min exposures gave the following results:

<u>Distance from Capsule Midpoint</u>	<u>Dose, R/hr</u>
10 cm	12,100
20 cm	4,900
40 cm	1,600

Beneficial Use of WESF $^{137}\text{CsCl}$ Capsules: Gamma Sources for Sewage Sludge Irradiation

Sandia Laboratory is planning a large-scale demonstration of the beneficial treatment of sewage sludge by irradiation, hopefully using WESF $^{137}\text{CsCl}$ capsules for gamma sources. Since the WESF capsules were designed for long-term, underwater storage rather than for use as radiation sources, modifications in design may be required to make their use in the Sandia program attractive. Work is under way at PNL, in support of Sandia's demonstration, to optimize WESF capsule design.

Calculations of Dose Rates and Radiation Efficiencies (R. A. Libby, Criticality Safety and Shielding Analysis)

Calculations have been completed comparing dose rates and radiation efficiencies for $^{137}\text{CsCl}$ in capsules of various geometries. The calculations were made for solid cylinders ranging in diameter up to 2.06 in. (current WESF design), for annular cylinders with 2.06-in. outer diameters and varying inner diameters, and for a 1-in. cylinder with two thicknesses of clad.

Calculated dose rates in water and air for two thicknesses of clad are shown in Figures 3 and 4. Relative radiation efficiencies are shown in Table 4, and are given both in terms of: 1) an average dose rate/curie (averaged over the volume bounded by the cylinder surface and cylindrical surface which has a boundary 20 cm from the capsule surface) and 2) the fraction of the total source radiation absorbed in this same volume.

It is evident from these calculations that of the geometries examined, the 1-in. cylinder with 0.1-in. clad has the optimum radiation efficiency.

Isotope Availability

Estimation of Supplies of Isotopes through 2000 AD (C. M. Heeb, Engineering Systems Analysis)

Knowledge of the character and amounts of the potentially useful isotopes present in fuels discharged from nuclear power reactors is needed to estimate possible demands for these materials. The purpose of this study was to make a preliminary estimate of the cumulative amounts produced in the U.S. civilian nuclear power economy. The time period covered is from 1972 to the year 2000.

A large amount of detailed isotopic information for several reactor design classes has been generated previously at Battelle-Northwest (BNW) for other projects. The computer code ORIGEN was used to provide detailed information on 815 isotopes as a function of time since discharge. ORIGEN is essentially a nondimensional transmutation code equipped with several

FIGURE 4. Dose Rate from CsCl Capsule in Air

TABLE 4. Source Efficiencies

Annular Cylinders:

<u>Internal Radius (cm)</u>	<u>Clad Thickness (cm)</u>	<u>Average Dose Rate (R/hr/Ci)</u>	<u>Fraction Energy Absorbed</u>
0.0 ^(a)	0.5182	6.142	12.79%
1.27	0.5182	6.153	12.81%
1.81	0.5182	6.194	12.90%

Cylinders:

<u>Radius (cm)</u>	<u>Clad Thickness (cm)</u>	<u>Average Dose Rate (R/hr/Ci)</u>	<u>Fraction Energy Absorbed</u>
2.6162 ^(a)	0.5182	6.142	12.79
1.27	0.5182	7.974	15.07
1.27	0.2540	8.923	16.43
0.635	0.5182	8.985	16.04

(a) Present Capsule Configuration

massive nuclear property libraries. The constants (microscopic cross sections, decay constants, and fission yields) in each library are specific for a given reactor type. That is, the cross sections are modified somewhat to include the influence of the different neutronic environs for the various reactor types and designs.

To provide a forecast of cumulative isotopic availability, a small computer program was written to interrogate the isotopic files and to combine the retrieved specific weight (grams/metric tonne heavy metal) with a discharge scenario specified in tonnes per year for each of five reactor design types. The code does five things at each year time point:

- 1) Locates the specific isotope on the file
- 2) Converts the gram atoms per metric ton to grams per metric ton
- 3) Sums the amounts available from previous years by taking the decay into account from each previous year and the number of tonnes discharged that year
- 4) Steps 1 through 3 are repeated for each of three reactor types (LWR, HTGR, and LMFBFR) with two fuel variants in LWRs and two different LMFBFR designs
- 5) An availability summary is prepared by adding up the amount available from each of the five reactor fuel types.

Two projections were made using two discharge scenarios and the following mix of reactor types: LWRs fueled with uranium, LWRs fueled with mixed PuO_2 and UO_2 , HTGR on the ^{233}U -thorium cycle, and two LMFBFR design types. All of these reactor types were assumed to have reached equilibrium, i.e., constant feed composition and constant discharge exposure. One scenario represents an optimistic view (1972) of the nuclear power economy. The second scenario is based on the most pessimistic case of a set of three produced in 1974 by the Office of Planning and Analysis, AEC, and is currently judged to be the most probable. The results of this study have been summarized in Reference 1.

Integrated Recovery Flow Sheet Studies (H. H. Van Tuyl, Applied Chemistry)

Large-scale use of by-products from nuclear reactors is likely to be dependent on a commercial supply integrated with a commercial fuel reprocessing plant. Lead time for such a plant is 8 to 10 years, so the technology to be used must be developed and demonstrated long before plant startup. Processes for recovery of by-products have been developed through laboratory studies, and some processes have been demonstrated on a sufficiently large scale to permit plant design at any time. However, lower-cost processes might be possible using technology which has not been

developed this extensively. The principal effort in FY-1975 was devoted to examining new separations techniques which may represent a potential improvement over demonstrated technology. A computer-assisted literature search has been employed to get state-of-the-art references.

The most widely used method of chemical separation in the nuclear industry is solvent extraction. Its advantages include: high selectivity which assures product purity, continuous operation, versatility, speed of separation, and ease of remote operation. Solvent extraction has been the subject of extensive research and development.^(2,3) It has been thoroughly proven on an industrial scale at numerous nuclear facilities throughout the world. However, solvent extraction suffers from some disadvantages, including solvent flammability, toxicity of extractants, and radiation damage. Despite these disadvantages, this process has to be of primary consideration in the formulation of any nuclear product separation scheme.

Ion exchange has played a very useful role in the separation of fission products. The properties of this technique which make it especially suitable for work with radioactive substances include: comparative simplicity of operation and equipment, a high degree of flexibility, easy remote operation, and simplicity of multistage arrangements. Disadvantages associated with ion exchange processes include the large volume elutions and susceptibility to radiation damage. However, the usefulness of this process has been well demonstrated in the nuclear industry, especially in the purification of plutonium.

Refinements of ion exchange processes have involved the use of various complexing agents for elution.⁽⁴⁻⁸⁾ This method, commonly referred to as ion exchange chromatography, has been extensively used in the separation of americium and promethium from rare earths. Other methods for improving ion exchange techniques are under study. Inorganic ion exchangers have been used to overcome the problem of radiation damage to synthetic resins. Hydrous oxides, salts of acids with multivalent metals, and other exchangers have been studied. Zirconium phosphate has been extensively evaluated and titanium phosphate has been successfully used to separate cesium in a pilot plant.⁽⁹⁾

Several techniques are under consideration but need further study to determine the practicality of their incorporation into an industrial-scale operation. Foam separation offers the advantage of volume reduction. Also, low operating and installation cost is inherent with this process. However, the process is unstable and is not continuous. Extraction chromatography has been shown to be useful in the separation and isolation of rhodium, palladium, and technetium.⁽¹⁰⁻¹²⁾ However, most work with this technique has been done on a laboratory scale. Electrodialysis has been considered⁽¹³⁾ but because of the many problems associated with this technique, it is not considered to be a viable method. Molten salt electrolysis also has many problems and is not considered to be practical for large-scale operations.

Some newer methods usually associated with isotope separation have been investigated. Gas centrifugation, laser excitation, and diffusion separation all suffer from huge energy expenditures and are presently considered economically unacceptable to full-scale nuclear product separation.

Future efforts will be directed toward developing integrated flow sheets for by-product recovery using existing technology. No further consideration will be given to new technology at this time. A development program will be identified to translate existing laboratory technology into demonstrated plant processes.

In summary, we have reviewed the processes which show some promise for use in the separation of by-product isotopes from spent nuclear fuels.⁽¹⁴⁾ We feel that some combination of solvent extraction and ion exchange would be the most desirable method of separation in a full-scale industrial plant. While many other techniques show promise, they are not as economically or technically feasible as existing methods.

Demand Forecasting (A. M. Schneider, Contract Services)

The development of reliable forecasts of potential demand for various reactor by-products is an essential activity in any valid analysis of the

incentives for industrial recovery of isotopes. Limited effort was applied to development of a program for making such forecasts. The program contains the following:

- Develop the format for a comprehensive picture of overall, long-term potential demand for reactor by-products
- Review specific high-potential applications as they develop; provide or secure forecasts of related long-term demand; and incorporate these forecasts into the comprehensive demand picture
- Develop and maintain a catalog of potential uses of various isotopes and any available information concerning current or projected demand
- As funds and time permit, evaluate the lower priority uses and incorporate findings into the comprehensive demand picture.

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