**Quarterly Report on the Strontium
Heat Source Development Program and
the Terrestrial Radioisotope
Applications Program, Advanced
Systems and Materials Production
Division for October-December 1977**

H. T. Fullam

January 1978

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QUARTERLY REPORT ON THE STRONTIUM HEAT SOURCE
DEVELOPMENT PROGRAM AND THE TERRESTRIAL
RADIOISOTOPE APPLICATIONS PROGRAM, ADVANCED
SYSTEMS AND MATERIALS PRODUCTION DIVISION
FOR OCTOBER-DECEMBER 1977

H. T. Fullam

January 1978

BATTELLE
Pacific Northwest Laboratories
Richland, Washington 99352

SUMMARY

STRONTIUM HEAT SOURCE DEVELOPMENT PROGRAM

WESF-grade nonradioactive SrF_2 was hot-pressed in graphite dies at 825°C and 4950 psi to a density of 4.19 g/cm^3 - approximately 99% of theoretical density. The pellets were thermally stable and could be placed directly into or removed from an 800°C furnace without cracking.

Hastelloy C-276 specimens exposed to WESF produced $^{90}\text{SrF}_2$ for 6000 hours exhibited approximately the same level of chemical attack as specimens tested for 1000 hours. However, microstructural changes due to fluoride attack were significantly greater in the 6000 hour specimens than in the 1000 hour specimens.

Hastelloy C-276 specimens from a full-size WESF $^{90}\text{SrF}_2$ capsule held at approximately 800°C for 5000 hours showed much less chemical attack than one would predict from the other compatibility tests with Hastelloy C-276 and WESF $^{90}\text{SrF}_2$. In addition, the Hastelloy C-276 specimens showed no evidence of microstructural changes due to fluoride attack. This is very unusual since the Hastelloy C-276 specimens from the other $^{90}\text{SrF}_2$ tests all exhibited extensive microstructural changes.

Work has started on design of the $^{90}\text{SrF}_2$ heat source. Hastelloy S has been selected as the outer capsule material. Heat transfer calculations indicate the temperature drop between the inner and outer capsule can be reduced as low as 32°F by a combination of factors, including filling the gap between capsules with helium and increasing the emissivities of the capsule surfaces.

TERRESTRIAL RADIOISOTOPE APPLICATIONS

The Terrestrial Radioisotope Applications Program has the objectives of evaluating the long-term availability and cost of useful isotopes, beneficial applications of isotopes, and actions needed to optimize the WESF products

for beneficial uses. Accomplishments during this quarter were the completion of two studies; one compared the economics of sludge irradiation with other alternative municipal sludge treatment methods and the second compared the economics of radioisotope (WESF ^{90}Sr) fueled heaters with fuel oil systems for use in remote cold regions.

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STRONTIUM HEAT SOURCE DEVELOPMENT PROGRAM

H. H. Van Tuyl, Program Manager
H. T. Fullam, Principal Investigator
D. G. Atteridge
F. A. Simonen

At Hanford, strontium is separated from the high-level waste, converted to the fluoride, and doubly encapsulated in small, high-integrity containers for subsequent long-term storage. The fluoride conversion, encapsulation and storage take place in the Waste Encapsulation and Storage Facilities (WESF). The encapsulated strontium fluoride represents an economical source of ^{90}Sr if the WESF capsule can be licensed for heat source applications under anticipated use conditions. The objectives of this program are to obtain the data needed to license $^{90}\text{SrF}_2$ heat sources and specifically the WESF $^{90}\text{SrF}_2$ capsules. The information needed for licensing can be divided into three general task areas:

- Task 1 - Chemical and Physical Properties of $^{90}\text{SrF}_2$*
- Task 2 - $^{90}\text{SrF}_2$ Compatibility Studies*
- Task 3 - Capsule Qualification and Licensing*

Efforts are proceeding concurrently on all three tasks to obtain the required information.

TASK 1 - CHEMICAL AND PHYSICAL PROPERTIES OF $^{90}\text{SrF}_2$ (H. T. Fullam)

A study was carried out to evaluate the hot pressing characteristics of strontium fluoride. The nonradioactive SrF_2 used in the study contained impurities similar to those found in WESF-produced $^{90}\text{SrF}_2$. The SrF_2 was pressed in an air atmosphere using a graphite die and plunger. A sketch of the equipment is shown in Figure 1. The graphite die had an inner diameter of 1.00 in., and sufficient SrF_2 was used in each pressing to give a compacted pellet having a length-to-diameter (L/D) ratio of approximately one. A thermocouple placed in the side wall of the die was used to monitor the pressing temperature. The pressing temperature was varied from 700 to 850°C, and the applied pressure from 2250 to 4950 psi. Since the hot pressing was carried out in air, the outer surface of the die and plunger slowly oxidized and they had to be replaced after 8-10 pressings.

FIGURE 1. Equipment for Hot Pressing Nonradioactive Strontium Fluoride

The procedure used in the hot pressing experiment follows:

A weighted amount of SrF_2 was placed in the die. The plunger was inserted in the die, the die was placed inside the heating element, and the thermocouple was positioned in the die. The heating element and die were then covered with blanket insulation. The die was heated to the desired temperature and pressure applied to the plunger. The pressure was maintained for 10 min while holding the temperature constant. After the pressure was released, the die was cooled to room temperature and the pellet ejected from the die. The pellet was then weighed and its dimensions obtained to determine the density.

Pellets pressed at 825°C or less were easily removed from the die, and were free from visible flaws or cracks. The pellets were covered with a thin layer of graphite which was easily removed by heating the pellets in air at about 800°C for 30 min. Figure 2 shows some typical pressed pellets before and after heating in air to remove the graphite layer. The firing in air at 800°C had no measurable effect on the density of the pellets. Thermal stability of the SrF_2 pellets was very good and they could be placed directly into or removed from an 800°C furnace without fracturing. Examination of the inner surface of the die indicated very little reaction between the fluoride and the graphite.

Pellets pressed at 850°C adhered to the graphite plunger and plug. It was possible to eject the pellets from the die, but they could not be separated from the plug and plunger without fracturing. Examination of the inner surface of the die and the plug and plunger indicated some diffusion of fluoride into the graphite. This apparently results from the small quantity of molten phase present in WESF-grade SrF_2 at 850°C (thermal analysis of WESF-grade SrF_2 indicates the existence of a low melting phase having a melting point of $851 \pm 5^\circ\text{C}$ - the quantity of molten phase present at 850-860°C is estimated to be less than 0.1 wt%).

The results obtained show that the WESF-grade nonradioactive SrF_2 is easily compacted to high densities using hot pressing techniques. Figure 3 shows the pellet densities obtained as a function of pressing temperature and applied pressure. The results show that pellet densities approaching

FIGURE 2. Typical Hot Pressed Pellets of SrF_2 Before and After Firing at 800°C to Remove the Graphite Layer

FIGURE 3. The Effects of Temperature and Applied Pressure on the Density of Hot-Pressed Pellets of Nonradioactive Strontium Fluoride

99% of theoretical density can be obtained by hot pressing in graphite at 825°C (the theoretical density of WESF-grade SrF_2 is calculated to be 4.24g/cm³ versus 4.29g/cm³ for pure SrF_2).

The strontium-90 Fluoride Data Sheet^{*} has been issued.

TASK 2 $^{90}\text{SrF}_2$ COMPATIBILITY STUDIES (H. T. Fullam)

Long Term Tests

All of the remaining compatibility tests are proceeding on schedule. No additional operating problems have been encountered with the furnaces and controllers since the additional over-temperature protection circuits were installed.

The 12,000 hr tests with WESF $^{90}\text{SrF}_2$ were completed in mid-September. However, it has not been possible to section the test capsules because the hot cell normally used for the sectioning operation has not been available for use. No other acceptable hot cell space has been identified at Hanford, and arrangements have been made to ship the capsules to ORNL for sectioning. It is currently planned to make the shipment in January 1978.

ORNL has completed the metallographic examination of the Hastelloy C-276 specimens exposed to WESF $^{90}\text{SrF}_2$ for 6000 hrs. Estimates of metal attack, based on the micrographs obtained, are given in Table 1. The data from the 1000 hr tests are also included for comparison purposes.

The data show that chemical attack of the Hastelloy C-276 did not increase appreciably with increasing exposure time except for the couple having a $S/V=2.5 \text{ cm}^{-1}$, which was tested at 1,000°C. The extent of microstructural changes did increase somewhat with increasing exposure time especially at the higher temperatures. As was the case with the 1000 hr tests, the addition of Zr and ZrF_4 to the $^{90}\text{SrF}_2$, to simulate decay product buildup, produced a marked decrease in the chemical attack of the Hastelloy C-276 at all temperatures.

^{*}H. T. Fullam, Strontium-90 Fluoride Data Sheet, BWNL-2284, Battelle, Pacific Northwest Laboratories, Richland, WA, October 1977.

TABLE 1. Estimated Attack of Hastelloy C-276 Specimens Exposed to WESF $^{90}\text{SrF}_2$

Temp. °C	S/V Ratio ^(b) Cm ⁻¹	Depth of Metal Affected, ^(a) mils			
		Chemical Attack		Change in Microstructure	
		1,000 Hr	6,000 Hr	1,000 Hr	6,000 Hr
600	4.5	3	3	4	4
600	4.5	3	3	5	5
600	2.5	10	7	12	10
600 (c)	4.5	1	<1	2	2
800	4.5	7	8	12	20
800	4.5	6		11	
800	2.5	30	24	35	>30
800 (c)	4.5	2	1	5	5
1000	4.5	7	8	15	24
1000	4.5	10	12	15	>30
1000	2.5	12	25	15	>35
1000 (c)	4.5	3	4	7	>30

(a) Estimated from photomicrographs

(b) S/F - Exposed metal surface to $^{90}\text{SrF}_2$ volume ratio of test couple

(c) Zr and ZrF₄ added to $^{90}\text{SrF}_2$ to simulate decay product buildup equivalent to 10 yr decay.

The Hastelloy C-276 specimens exposed to $^{90}\text{SrF}_2$ at 600°C for 6000 hr suffered a uniform attack of the surface with formation of an adherent reaction layer (see Figure 4). Etching of the specimen leached away most of the reaction layer. There was only slight indication of grain boundary attack. Microstructural changes consisted of a narrow zone adjacent to the reaction layer which showed a slight depletion of normal alloy precipitates. Similar types of attack were observed in the 1000 hr-600°C test specimens.

At 800°C the attack was also uniform across the specimen surface but appeared to proceed along grain boundaries leaving particles of unreacted metal in the reaction layer (see Figure 5). Etching of the specimen dissolved away most of the reaction products, leaving free grains of metal. The concentration of metal particles increased from the outer edge of the reaction layer to the unreacted specimen surface indicating that the metal

FIGURE 4. Hastelloy C-276 Specimen Exposed to WESF $^{90}\text{SrF}_2$ for 6000 hr at 600°C - S/V = 4.5 cm⁻¹

FIGURE 5. Hastelloy C-276 Specimen Exposed to WESF $^{90}\text{SrF}_2$ for 6000 hr at 800°C -S/V = 2.5 cm^{-1}

particles are reacting with the $^{90}\text{SrF}_2$ or contained impurities. Microstructural changes occurred to considerable depths and consisted primarily of heavy intragranular precipitates not found in the main body of the specimens. Overall, the attack mechanisms were very similar to those observed in the 1000 hr-800°C specimens.

At 1000°C attack of the Hastelloy C-276 was quite different from that observed at the lower temperatures, and from that observed in the 1000 hr tests at 1000°C. The attack apparently involved general dissolution of the specimen surface with extensive pitting and grain boundary attack (see Figure 6). No reaction layer was observed on any of the specimen surfaces; however, free granules of unreacted metal were evident adjacent to the surfaces. These particles were apparently held in place at the surface by a reaction layer which was leached away during the grinding-polishing operation prior to etching of the specimen. Because of the presence of the free grains of metal, it was difficult to accurately assess the extent of metal attack in the Hastelloy C-276 specimens exposed to $^{90}\text{SrF}_2$ at 1,000°C. Data presented in Table 1 for the 1,000°C-6,000 hr specimens are, at best, rough approximations of the chemical attack. In some of the 1,000°C specimens microstructural changes occurred to greater depths than could be determined from the micrographs (>30 mils). In general, the microstructural changes consisted of: a surface zone containing irregularly shaped intergranular precipitates, a central zone largely depleted of precipitates, and an inner zone in which the average grain size was much smaller than in the unaffected areas of the specimens. The width of the inner zone could not be determined from the micrographs. Overall, the attack mechanisms, both chemical and microstructural, observed in the specimens tested at 1000°C for 6000 hr were significantly different from those observed in the 1000-hr specimens.

As mentioned previously, the addition of Zr and ZrF_4 to the $^{90}\text{SrF}_2$ produced a marked decrease in Hastelloy C-276 attack at all temperatures (see Figure 7). The chemical attack mechanisms were similar at the three test temperatures and consisted primarily of general surface dissolution with some grain boundary attack and subsurface void formation. Microstructural

FIGURE 6. Hastelloy C-276 Specimen Exposed to WESF $^{90}\text{SrF}_2$ for 6000 hr at 1000°C - $\text{S/V} = 4.5 \text{ cm}^{-1}$

FIGURE 7. Hastelloy C-276 Specimens Exposed for 6000 hr to WESF $^{90}\text{SrF}_2$ Containing Zr + ZrF_4 - S/V = 4.5 cm^{-1}

changes were much greater at 1000°C than at the lower temperatures. At 600° and 800°C the microstructural changes consisted of a narrow surface layer in which normal alloy precipitates were partially depleted. At 1000°C the microstructural changes consisted of: 1) a narrow surface layer in which the grain size was much smaller than normal and which contained heavy continuous grain boundary precipitates and some intragranular precipitates, and 2) a broad inner zone containing abnormal intragranular and grain boundary precipitates.

⁹⁰SrF₂ Capsule Demonstration Test

ORNL has completed the examination of the test specimens from inner capsule of the full-size WESF storage capsule that had been held at temperature for 5000 hr. The specimens had been taken from four locations along the length of the capsule. Figure 8 shows the calculated temperature profile for the metal-⁹⁰SrF₂ interface of the test capsule. It was not possible to measure the metal-⁹⁰SrF₂ interface temperature directly and the temperature profile was calculated from measured outer capsule surface temperatures using standard heat transfer techniques. Temperature variations of up to ±6°C occurred during the tests and the temperatures given in Figure 8 are average values. Figure 8 also shows the four locations where the capsule was sectioned and the approximate temperatures at each location. Estimates of metal attacks at the four locations, based on the photomicrographs obtained, are given in Table 2. The extent of the chemical attack varied significantly between the four locations. The chemical attack mechanisms also appeared to vary between locations (see Figure 9). Variations in the extent of the chemical attack cannot be attributed to temperature differences since the maximum attack occurred at an intermediate temperature. However, the basic microstructure of the Hastelloy C-276 did vary with temperature, and this could possibly account for the apparent differences in attack mechanisms at the different locations. The micrographs show that the specimens heated at 762° and 790°C contained very little intragranular precipitation, whereas the specimens heated at 813° and 821°C contained extensive bulk precipitation which is probably *mu* phase (Ni₇Mo₆).

FIGURE 8. Temperature Profile of the WESF $^{90}\text{SrF}_2$ Capsule Held at Temperature for 5000 hr

TABLE 2. Estimated Metal Attack at Various Locations in the Full-Size WESF $^{90}\text{SrF}_2$ Capsule Held at Temperature for 5000 hr - S/V = 0.86 cm^{-1} .

Section No.	Approximate Temperature °C	Depth of Metal Affected, mils	
		Chemical Attack	Change in Microstructure
2	762	3	0
4	790	11	0
3	813	8	0
1	821	9	0

FIGURE 9. Micrographs of the Hastelloy C-276 Specimens from the Full-Size WESF $^{90}\text{SrF}_2$ Capsule Held at Temperature for 5000 hr

At 762° and 790°C the attack mechanisms were similar although the extent of attack was about four times greater at 790°C. The chemical attack appeared to consist primarily of general surface dissolution with some grain boundary attack. At the higher temperatures (813 and 821°C), the chemical attack appeared to consist primarily of grain boundary attack and pitting, leaving free grains of unreacted metal near the specimen surface.

None of the specimens from the full-size capsule exhibited any microstructural changes. This is surprising since in all of the other compatibility tests at 800°C involving Hastelloy C-276 and $^{90}\text{SrF}_2$ or nonradioactive SrF_2 the alloy suffered extensive microstructural changes.

Overall the Hastelloy C-276 specimens from the full-size WESF $^{90}\text{SrF}_2$ capsule suffered significantly less attack than one would predict from the initial results of the long-term tests with Hastelloy C-276 and WESF $^{90}\text{SrF}_2$. Metal attack in the full-size capsule, which has an S/V of about 0.85 cm^{-1} , was about the same as that observed in Hastelloy C-276/ $^{90}\text{SrF}_2$ capsules having an S/V of 4.5 cm^{-1} and much less than that observed in capsules having an S/V of 2.5 cm^{-1} . The reason for the reduced attack has not been identified but may be due to lower concentrations of critical impurities in the $^{90}\text{SrF}_2$ in the full-size capsule.

Operations at WESF have been shut down for several months while new equipment is being installed. Therefore, Rockwell International has been unable to section the full-size WESF $^{90}\text{SrF}_2$ capsule which was held at temperature for one yr. Latest estimates are that Rockwell will be able to section the capsule in March or April. This should allow sufficient time to analyze the specimens from the capsule by the end of FY 1978.

TASK 3 - CAPSULE QUALIFICATION AND LICENSING (D. G. ATTERIDGE)

Work is underway on the design of the $^{90}\text{SrF}_2$ heat source. The design criteria for the heat source were provided in the previous quarterly report. The heat source is to consist of the existing WESF Hastelloy

C-276 inner capsule and a new PNL-designed outer capsule. The new outer capsule must be capable of meeting the qualification test requirements* in both the as-fabricated and thermally aged conditions.

F. A. Simonen, of the PNL Engineering Physics Section, has been added to the staff involved in the capsule design activity. Mr. Simonen has had previous experience in radioisotope heat source design for SNS at Battelle-Columbus and is currently involved in developing canister designs for the high-level waste vitrification program.

Generic outer capsule design studies in such areas as the effect of design on heat transfer and the impact resistance of the closure weld have been initiated, and, in addition, material-specific design and design support studies are underway. These latter studies are based on the selection of Hastelloy S as the primary outer capsule material. Four principal candidates were considered for use as the outer capsule material: Hastelloy S, Hastelloy C-4, Inconel 625 and Inconel 617. Hastelloy S was selected for use, based on its preferred high temperature aging characteristics, with Hastelloy C-4 as a back-up material.

Elevated Temperature Testing of Aged Hastelloy S and Hastelloy C-4 Alloys

The testing of the second set of aged Hastelloy S and Hastelloy C-4 specimens was finished during the quarter. Several specimens were tested at room temperature to determine whether there were any inherent size effects between the PNL-subsized tensile specimens and the Cabot Corp. full-sized tensile specimens. Preliminary analysis of the test results indicates that the room-temperature tensile property results are essentially identical for the two sizes of specimens.

A series of selected tensile specimens from the various aging temperatures was tested at 800°C to determine whether low-temperature exposure resulted in degradation of high-temperature properties. The tensile specimens were soaked for 1/2 hr at 800°C before testing. The results indicate that even this short pretest soak was enough to anneal

*H. T. Fullam, Quarterly Report on the Strontium Heat Source Development Program and the Beneficial Isotopes Utilization Program, Division of Nuclear Research and Applications for July-September, 1977, BNWL 1845-36, Battelle, Pacific Northwest Laboratories, Richland, WA, 1977.

out the low-temperature strengthening and/or embrittling reaction products. The tests demonstrated mechanical properties essentially the same as those for specimens aged for comparably short times at 800°C.

The remaining aged specimens were tested at their respective aging temperatures as duplicates of the original specimens in order to determine the average data scatter, thus the relevance of the previously reported elevated-temperature data trends. Preliminary analysis of the data indicates that the data trends for the specimens tested at 1,200°F and below fall very close to the original test data. The data for 1,400°F and 1,600°F, however, exhibit more data scatter than was expected, particularly in the uniform elongation values. The results from both sets of tests will be presented in the next report.

Heat Transfer Considerations

Heat transfer calculations were carried out to determine the temperature drops to be expected in the PNL designed $^{90}\text{SrF}_2$ heat source. The purpose of these calculations is two-fold:

- to establish the maximum wall temperature for the outer capsule so that metallurgical aging effects can be predicted and/or experimentally determined, and
- to establish the measures required in design and fabrication of the heat source to minimize the temperature drop across the gap between the inner and outer capsules.

The results of these calculations and the general conclusions reached regarding capsule heat transfer agree with previous heat transfer studies carried out by PNL and Teledyne Energy Systems with regard to the WESF $^{90}\text{SrF}_2$ storage capsule.

Capsule Parameters

The capsule interface configuration is shown in Figure 10. Compatibility considerations relative to SrF_2 and Hastelloy C-276 require that the inner capsule-fuel interface temperature not exceed 800°C (1472°F). In the present capsule design, the gap between the inner and outer capsules

is nominally 0.078 in. In the redesign of the outer capsule, it is believed that this gap can be reduced to 0.025 in. Presently the gap is filled with air. In a redesigned system, the use of helium with a higher thermal conductivity will be desirable. In the present system, the surface emissivities of the inner and outer capsules are reported to be 0.25 and 0.20, respectively. By surface treatments, these emissivities could be increased to 0.55 and 0.80. Nominal power output per capsule is 1000 watts. Excluding capsule end caps, the effective length of the capsule for heat transfer purposes is about 18.0 in.

FIGURE 10. Configuration of Interface Between Inner and Outer Capsules

Heat Transfer Mechanisms

Calculations included heat transfer by the mechanisms of radiation across the gap and conduction through the air within the gap. Heat transfer rates by convection were estimated and found to be negligible. The governing equations are:

$$Q = A\sigma(T_1^4 - T_2^4)/(1/\epsilon_1 + 1/\epsilon_2 - 1), \text{ radiation}$$

$$Q = Ak(T_1 - T_2)/L, \text{ conduction,}$$

where,

Q = heat flux, BTU/sec-in.²

A = area, in.²

σ = Stefan-Boltzman constant

$$= 3.31 \times 10^{-15} \text{ BTU/sec-in.}^2 - ^\circ\text{R}^4$$

T_1 = temperature of outer surface of inner capsule, $^\circ\text{R}$

T_2 = temperature of inner surface of outer capsule, $^\circ\text{R}$

ϵ_1 = emissivity of outer surface of inner capsule

ϵ_2 = emissivity of inner surface of outer capsule

k = conductivity of gas within gap between capsules,
BTU/in.-sec- $^\circ\text{F}$

L = width of gap, in.

Conductivities of the materials of interest are given in Table 3.

In the present context, the heat flux is known, while the temperature gradients due to the heat flux are to be determined. The total capsule heat output of 1000 watts is equivalent to 0.948 BTU/sec.

TABLE 3. Conductivity Data Used in Heat Transfer Calculations

<u>Material</u>	<u>Temp., °F</u>	<u>Thermal Conductivity BTU/in.-sec-°F</u>
Hastelloy C-276	1200	2.64×10^{-4}
	1600	3.11×10^{-4}
Air	1000	7.80×10^{-7}
	1300	9.14×10^{-7}
	1600	10.07×10^{-7}
Helium	980	41.08×10^{-7}
	1520	53.08×10^{-7}
	3140	82.82×10^{-7}

Temperature Drop - Wall of Inner Capsule

At the inner surface of the inner capsule, the heat flux is equal to 0.948 BTU/sec divided by the surface area of 36.18 in.² ($\pi \times 2.01 \times 18.0$). The temperature drop is:

$$T_1 - T_2 = QL/k = 0.00834 \times 0.125 / 2.96 \times 10^{-4} \\ = 3.49^\circ\text{F.}$$

Thus, the outside surface of the inner capsule is 1468.5°F. This temperature drop is relatively insignificant.

Temperature Drop - Across Gap

The heat transfer equation for radiation is nonlinear in T_2 (i.e., T_2^4). An iterative solution was required to determine values of T_2 which would give a combined heat flux due to radiation and conduction that would correspond to the 1000 watt heat output of the capsule. Solutions for three combinations of heat transfer parameters are given in Table 4.

TABLE 4. Temperature Drop Across Gap Between the Inner and Outer Capsules

	<u>Existing Design</u>	<u>Existing Design with Helium Fill</u>	<u>Optimized Design</u>
Gas within gap	Air	Helium	Helium
Gap width	0.078 in.	0.078 in.	0.025 in.
Emissivity of inner capsule	0.25	0.25	0.55
Emissivity of outer capsule	0.20	0.20	0.80
Temperature drop across gap	379°F	96°F	32°F
Temperature of ID outer capsule	1089°F	1373°F	1436°F

The first solution is for the capsule assembly as it presently exists. A relatively large ΔT of 379°F was calculated. Heat transfer across the gap is relatively difficult due to the large gap width, poor thermal conductivity of air, and the lower surface emissivities. Heat transfer rates by conduction and radiation are of about equal significance. A temperature drop of 379°F is unacceptable from the standpoint of system performance.

The second case given in Table 4 indicates the improvement in heat transfer resulting from replacing air within the gap with helium. A temperature drop of 96°F was calculated which is probably tolerable. Heat transfer by conduction through the helium dominates over heat transfer by radiation.

The final case in Table 4 corresponds to a redesigned capsule for enhanced heat transfer. The gap width has been reduced to 0.025 in., and helium has replaced air in the gap. Also, surface emissivities have been increased (e.g., by oxidizing the surfaces) to increase transfer by radiation. The calculated temperature drop has been reduced to 32°F, which perhaps represents a practical limit for the existing design restraints.

Heat Transfer Conclusions

The heat transfer calculations presented above indicate that the heat transfer characteristics of the gap between the inner and outer capsules will essentially determine the magnitude of the temperature drop across the heat source capsules. They also show that the gas contained in the gap is the largest single component controlling the temperature drop across the gap (see Table 4).

The results of the heat transfer calculations lead PNL to recommend that the gap between capsules be filled with helium gas. We are in the process of determining the feasibility and desirability of increasing the emissivity of the capsule surfaces by a pre-fabrication oxidation treatment and/or decreasing the temperature drop across the gap by requiring a close tolerance fit between capsules.

The close tolerance fit option, as well as weld cap design and hot cell fabrication philosophy, was recently discussed with WESF personnel. It was decided that experimental measurements on production WESF inner capsules will be made to help determine the feasibility of decreasing the gap size without requiring inner capsule machine cleanup or straightening. Such things as prefilled inner capsule out-of-roundness and maximum longitudinal bending as well as maximum weld bead buildup will be measured to ascertain which one will be the minimum gap controlling factor.

Metal-Metal Compatibility (H. T. Fullam)

The $^{90}\text{SrF}_2$ heat source, as currently visualized, will consist of a Hastelloy C-276 inner capsule and a Hastelloy S, or possibly Hastelloy C-4, outer capsule. At heat source operating temperature (700-800°C) it is possible that some interaction may occur between the inner and outer capsules. Therefore, a series of tests are planned to measure the compatibility of Hastelloy C-276 with Hastelloy S and Hastelloy C-4. The couple design being used in the tests is shown in Figure 11. The test capsule was machined from 1 in. dia. Hastelloy C-276 bar stock. One-half in. dia. discs of Hastelloy C-276 and Hastelloy S, or Hastelloy C-4, were stacked in the capsule as shown. An interference fit lid was pressed into the capsule, using a

hydraulic press, to provide intimate contact between the discs. The capsule was sealed by TIG welding the lid in place, then leak checked.

The flat faces of the discs were polished to a metallographic finish using 0.3 micron alumina for the final polish. Some of the polished discs were oxidized in air at 800°C for 24 hr prior to insertion in the capsules to determine the effects of high emissivity oxide coatings on metal-metal compatibility.

The tests will be carried out at 700°C and 800°C and will last for 1500, 4400, and 8800 hr. At the conclusions of the tests the capsules will be sectioned and each disc subjected to metallographic examination to determine the extent of metal-metal interaction. The tests will begin in January 1978.

- NOTES:
- CAPSULE MACHINED FROM 1" DIA. BAR STOCK
 - DISCS SECTIONED FROM 1/2" DIA. BAR STOCK
 - FLAT FACES OF DISCS POLISHED TO METALLOGRAPHIC FINISH USING 0.3 μ ALUMINA FOR FINAL POLISH
 - SOME DISCS OXIDIZED IN AIR AT 800°C FOR 24 HOURS TO FORM OXIDE COATING ON SURFACE
 - INTERFERENCE FIT LID PRESSED INTO PLACE USING HYDRAULIC PRESS TO PROVIDE INTIMATE CONTACT BETWEEN DISCS

FIGURE 11. Couple Design Used for Metal-Metal Compatibility Tests

Corrosion Testing (H. T. Fullam)

The PNL-established qualification tests for the $^{90}\text{SrF}_2$ heat source specify that the outer capsule of the heat source must be able to meet the various test requirements after: 1) exposure to seawater for 1 yr, and 2) exposure to air for 10 yr at heat source operating temperatures ($<800^\circ\text{C}$). Tests are planned to show that the Hastelloy S (and Hastelloy C-4) is sufficiently resistant to seawater corrosion and air oxidation to meet the qualification requirements.

Equipment and materials are being assembled for seawater corrosion testing of both Hastelloy S and Hastelloy C-4. The tests will last up to 10,000 hr. The tests will be conducted in a covered polyethylene tank using seawater from the strait of Juan de Fuca at the Battelle-Northwest facility at Sequim, WA. The seawater will be pumped through the tank on a once-through basis. All system parts contacting the seawater, except for the test specimens, will be fabricated from nonmetallic materials. Welded specimens, tensile specimens and pre-stressed specimens will be tested in addition to standard corrosion strips. Multiple specimens will be tested for each of several time periods. The tests will begin in the second quarter of FY 78.

Tests lasting up to 10,000 hr are planned to measure the oxidation resistance of Hastelloy S and Hastelloy C-4 at 600° , 700° and 800°C . Tensile specimens will be included in the tests to determine the effects of oxidation on the tensile properties of the two alloys. Sections of bar stock will also be thermally aged at each set of conditions, then machined to provide control tensile specimens. Multiple specimens will be tested at each temperature for each of seven time periods. The tests will begin in January 1978.

TERRESTRIAL RADIOISOTOPE APPLICATIONS PROGRAM

H. H. Van Tuyl, Program Manager

W. E. Sande, Principal Investigator

The major objective of this program is to identify potential beneficial uses of nuclear reactor by-product through three principal tasks: 1) evaluation of long-term availability and cost of useful isotopes from commercial suppliers or military wastes; 2) evaluation of beneficial applications of isotopes; and 3) identification and evaluation of the actions required to optimize the $^{90}\text{SrF}_2$ and $^{137}\text{CsCl}$ products from the Hanford Waste Encapsulation and Storage Facility (WESF) for beneficial uses.

The program is divided into three tasks:

Task I - Isotope Availability

Task II - Beneficial Applications

Task III - WESF Product Utilization

TASK 1 - ISOTOPES AVAILABILITY

Flowsheet Calculations

The document, Selection and Evaluation of Processes for Recovery of Beneficial Isotopes from Commercial High-Level Nuclear Wastes, BNWL-2063, by D. K. Davis, J. A. Partridge, and O. H. Koski is being reviewed internally at PNL. This document has undergone major revision since the first draft was completed over one year ago. Hopefully, it will be issued next quarter.

TASK 2 - BENEFICIAL APPLICATIONS

Cold Regions Applications

The Federal Aviation Administration (FAA) is using five radioisotope thermoelectric generators (RTGs) to power remote aircraft navigation aid systems. The systems are located in Lake Clark Pass, Alaska, between Kenai and Bristol Bay. Previous use of propane thermoelectric generators did not provide the required reliability. After about one year of operating experience (the RTGs were installed in September 1977), a cost comparison between RTGs and other power systems will be made.

The study entitled Economic Feasibility of Strontium-90 Fueled Heaters for Use in Cold Regions, PNL-2476, by W. E. Sande and R. L. Aaberg is being cleared for distribution. The objectives of this study are to conceptually design and obtain cost estimates for various radioisotope-fueled heating systems for use in potential cold regions applications, and to determine the economic feasibility of radioisotope fueled heaters by comparing them to fuel oil systems. The applications have been identified and grouped according to thermal output, as: 1) 1 kWt septic tank heaters, well head heaters, heaters to assist operation of sewage lagoons, and 2) 7 or 12 kWt potable water system heaters. Economical locations for the heaters are in remote Alaskan communities of 50 to 200 residents where fuel oil prices and reliability requirements are high.

Strontium-90 is the radioisotope heat source used in the heater conceptual designs and is presently available from the Hanford Waste Encapsulation and Storage Facility (operated for the Department of Energy by the Rockwell Hanford International Company) through the Isotope Sales Division at Oak Ridge National Laboratory, Oak Ridge, Tennessee. The heat generating material is contained in a double-walled stainless steel-Hastelloy C-276 capsule which is approximately 20-in. long and 2.6-in. diameter. The heat generation rate is about 1 kWt per capsule. One capsule is equivalent to 150,000 curies of strontium-90 (^{90}Sr). The present government price for this encapsulated ^{90}Sr is 10¢/curie fob Hanford, Washington; thus, one capsule would cost \$15,000.

To assist in cost analysis, the costs can be reduced to overall equivalent fuel oil price, i.e., the price of fuel oil that would make radioisotope systems compete equally with fuel oil systems. These are summarized below. Isotope fuel costs have been factored out to show their contribution.

Radioisotope System Size	Equivalent Fuel Oil Price, \$/gal	
	Overall Costs	Radioisotope Fuel
1 kW	7-16	6-8
7 kW	11-12	9
12 kW	12-15	11

A representative fuel oil price for remote locations in 1976 can be taken as \$1.50/gal; thus, fuel oil prices must increase dramatically before radioisotope systems can be economically competitive.

As shown above, the radioisotope fuel costs contribute 50% or more to the overall cost. Thus, an effective method to make a substantial reduction in radioisotope system cost would be to lower the strontium price from 10¢/curie. To make the radioisotope system economically competitive with \$1.50/gal fuel oil, the price would need to be lowered to 0¢/curie. Since the primary reason for encapsulating the strontium-90 is for nuclear waste management and not for providing radioisotope heat sources, a price reduction may not be entirely out of the question, but this must be decided by various government agencies who are in charge of the strontium, and potential user agencies or potential private user groups.

In conclusion, radioisotope heaters will not be economically competitive with fuel oil systems unless one or more of the following situations occur:

- the price of strontium-90 is reduced below 10¢/curie,
- fuel oil prices increase significantly, or
- for a particular application, the reliability or adaptability of a fuel oil system is unacceptable.

TASK 3 - WESF PRODUCT UTILIZATION

Sandia Support - Alternatives for Sludge Handling

During this quarter, the report entitled An Alternative Economic Comparison of Sludge Irradiation and Alternative Methods of Municipal Sludge Treatment, PNL-2431, by S. B. Ahlstrom and H. E. McGuire was issued.

One method to handle the increasing volume of sludge produced in the United States is through beneficial reuse (e.g. as a soil conditioner). However, sludge should not be reused unless it is made safe for those who may come in contact with it. For example, pathogens must be inactivated and toxic materials must be removed from the sludge. One method to minimize

the pathogen level is through radiation treatment. This study reports the relative economics of radiation treatment and other sludge treatment processes. The desirability of radiation treatment is assessed in terms of cost and the quality of the treated sludge product.

The major conclusions of this study are:

- radiation treatment is a high-level disinfection process. Therefore, it should only be considered if high levels of disinfection are required for widespread reuse of the sludge.
- the handling, transporting and pathogen growback problems associated with disinfected wet sludge makes it less attractive for reuse than dry sludge.
- radiation of composted sludge produces a product of similar quality at less cost than any thermal treatment and/or flash drying treatment option for situations where a high degree of disinfection is required.
- heavy metal concerns, especially cadmium, may limit the reuse of sludge despite high disinfection levels.

Radiation treatment of sludge, particularly dry sludge, merits additional research. A sensitivity analysis investigating the optimal conditions under which sludge irradiation operates should be instigated. Furthermore, costs of adding sludge irradiation to existing sludge treatment schemes should be determined. In order to assess the use potential of radiation treatment, a scenario should be prepared accounting for land use parameters, regulations, and existing treatment methods.

A decision on further work by PNL to assist Sandia's sludge irradiation program will be made in the near future.

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