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Pacific Northwest Laboratory Quarterly Report to USERDA Nuclear Research and Applications Division for October-December 1975

January 1976

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PACIFIC NORTHWEST LABORATORY QUARTERLY REPORT TO USERDA NUCLEAR RESEARCH AND APPLICATIONS DIVISION FOR OCTOBER-DECEMBER 1975

By

H. T. Fullam and K. M. Harmon

January 1976

Battelle
Pacific Northwest Laboratories
Richland, Washington 99352

EXECUTIVE SUMMARY

STRONTIUM HEAT SOURCE DEVELOPMENT PROGRAM

The fuel-grade $^{90}SrF_2$ has been obtained from ARHCO, and fabrication of the long-term $^{90}SrF_2$ compatibility test couples is now underway. The non-radioactive test couples have been fabricated and testing will begin in January.

Tests are underway to determine the effects of thermal aging on the impact strength (Charpy) of Hastelloy C-4 over the temperature range of 600 to 1000°C. Results of the 1000 hr tests show that aging at 600°C causes a sharp reduction in the impact strength of the alloy.

BENEFICIAL ISOTOPES UTILIZATION PROGRAM

Calculations of potential supplies of useful by-products through the year 2000 and projecting the effect of removing HTGR contributions were issued in an attachment to BNWL-B-435.(1)

Experimental dose measurements on a WESF $^{137}CsCl$ capsule were found to agree well with previous theoretical calculations.

Radiation efficiencies of a $^{137}CsCl$ plate source were calculated with plate thicknesses of 0.5, 1.0, and 2.0 cm (0.254 cm SS cladding) and plate area of 40 cm^2. The efficiencies are larger than those for cylindrical geometry (1 and 2 in. diameter); overall plate efficiency is estimated at 60 to 70% where cylindrical efficiencies are 30 to 50%.

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STRONTIUM HEAT SOURCE DEVELOPMENT PROGRAM

H. T. Fullam

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 $^{90}SrF_2$ heat sources and specifically the WESF $^{90}SrF_2$ capsules. The information needed for licensing can be divided into three general areas:

1. *Long-term SrF_2 compatibility data.*
2. *Chemical and physical property data on $^{90}SrF_2$.*
3. *Capsule property data such as external corrosion resistance, crush strength, etc.*

The current program is designed to provide the required information.

LONG-TERM COMPATIBILITY TESTS

The fuel-grade $^{90}SrF_2$, required for the long-term compatibility tests, has been prepared by ARHCO at WESF. The fluoride was shipped to PNL in mid-December. The $^{90}SrF_2$ powder was loose-packed in standard Hastelloy C-276 inner capsules which were then sealed in 316L stainless steel overpacks for the shipment. Approximately 7.5 kg of $^{90}SrF_2$ in five capsules were transferred to PNL.

The $^{90}SrF_2$ was analyzed by ARHCO, and the results obtained are given in Table 1. Additional analytical data are being obtained by PNL and will be reported at a later date.

The first fluoride capsule has been opened at PNL; no difficulties were encountered in removing the fluoride from the capsule. Fabrication of the $^{90}SrF_2$ test couples is now underway in the 325A Building hot cells.

TABLE 1. Preliminary Analysis of WESF $^{90}SrF_2$

^{90}Sr Isotopic Content - 50.6%

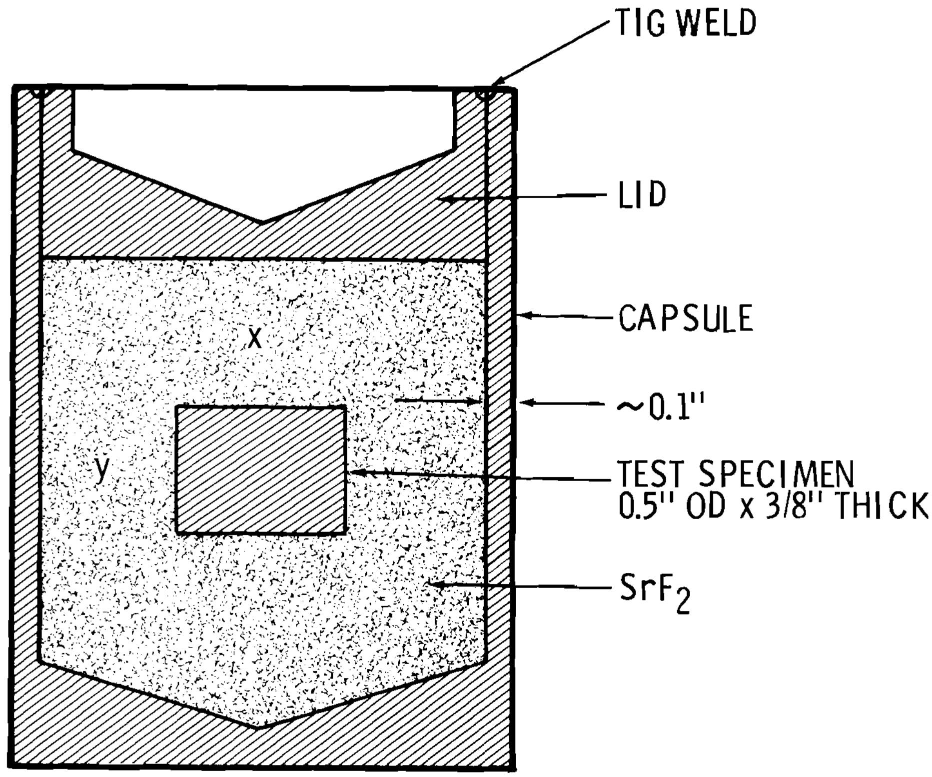
Element	wt%
Sr	67.50
Na	2.17
Al	0.31
Ca	0.45
Mg	0.048
Ba	0.91
Cd	0.017
Fe	0.39
Cr	0.45
Ni	---
Pb	0.016
Mn	0.014
Zr	1.55

Based on calorimetry and Sr isotopic analysis the WESF SrF_2 contains ~96% SrF_2

The fabrication of the nonradioactive test couples has been completed, and testing will begin in January. The nonradioactive SrF_2 used in the tests has approximately the same composition as the $^{90}SrF_2$ obtained from ARCHO (Table 1). The nonradioactive SrF_2 was prepared by the WESF flowsheet using reagent-grade chemicals.

The couple designs used for the nonradioactive and radioactive tests are shown in Figures 1 through 3. Figure 1 shows the couple design used with metallographic test specimens. Three different capsule sizes are used to permit testing of different metal surfaces to SrF_2 volume ratios (S/V). The smallest ratio being tested (S/V = 0.9 cm^{-1}) approximates the S/V ratio for the WESF capsule. Figure 2 shows the design used with tensile specimens, and Figure 3 the design used with Charpy test specimens. In addition, reference metallographic, tensile and Charpy specimens will be tested without fluoride present.

Testing of two full-size WESF $^{90}SrF_2$ capsules is planned. Each capsule will be placed in a special container and allowed to self-heat to approximately 800°C. One capsule will be tested for 6 months and the other for 12 months. At the completion of the tests the capsules will be sectioned and examined to determine the extent of fluoride-metal interaction. A sketch of the capsule container is shown in Figure 4. The container is equipped with three movable thermocouples which measure the surface temperature of the capsules at various locations. Tests with an electrically heated dummy capsule indicated a significant temperature gradient between the center and ends of the capsule due to uneven heat losses. At a power input of 1000 watts, equivalent to a fuel-grade WESF $^{90}SrF_2$ capsule, the outer surface temperature of the capsule varied from 676 to 652°C between the center and ends of the capsule. This corresponded to capsule-SrF_2 interface temperatures of 816 and 787°C. Since the desired interface temperature is 800°C, the container is adequate for the required tests. Testing of the full-size WESF capsules will start in January.

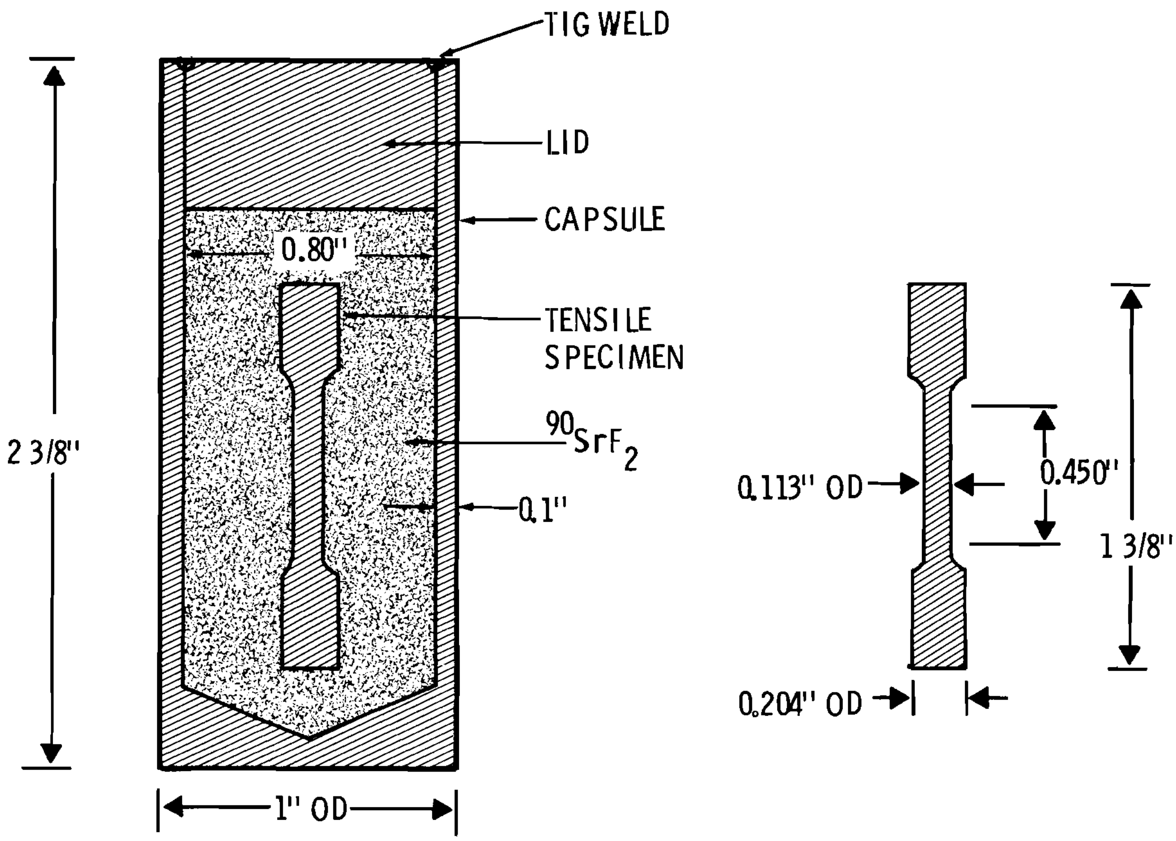


NOTES:

1. CAPSULE, LID AND TEST SPECIMEN ARE SAME MATERIAL
2. SrF_2 COMPACTED TO ~75% OF THEORETICAL DENSITY
3. THREE DIFFERENT SIZE CAPSULES WERE USED TO GIVE THREE DIFFERENT METAL SURFACE/FUEL VOLUME RATIOS (S/V) THE CAPSULE DIMENSIONS WERE AS FOLLOWS:

CAPSULE	S/V, cm^{-1}	x	y
A	4.5	0.8 in.	0.7
B	2.5	1.3 in.	0.7
C	0.9	2.4 in.	3.0

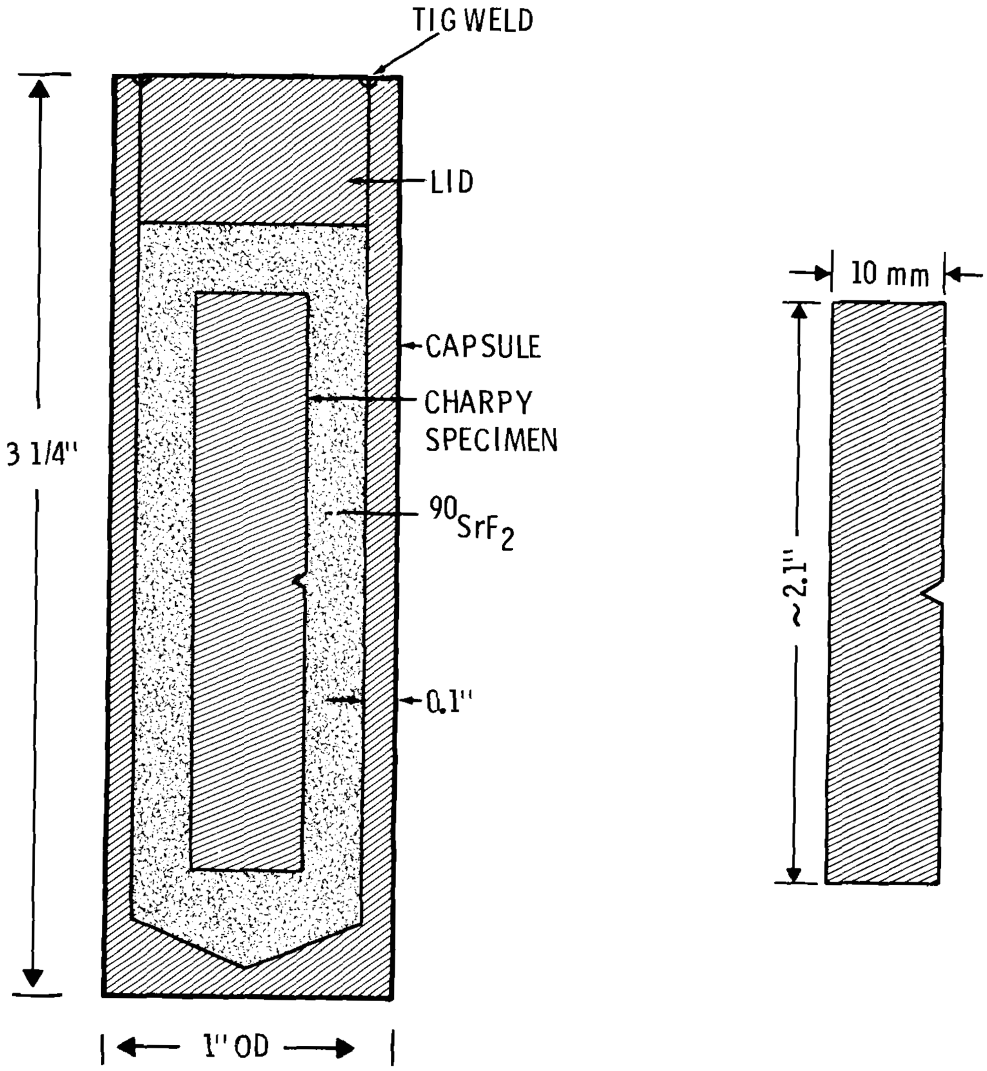
FIGURE 1. Couple Design Used with Metallographic Test Specimens



NOTES:

1. CAPSULE, LID AND TENSILE SPECIMEN ARE SAME MATERIAL
2. TENSILE SPECIMEN DESIGN CORRESPONDS TO ASTM SPECIFICATION E8-69 (0.113 in. DIAMETER ROUND SPECIMEN - FIGURE 8)
3. SrF_2 IS COMPACTED TO APPROXIMATELY 70%
4. COUPLE METAL SURFACE TO FUEL VOLUME RATIO (S/V) $\cong$ 2.8 cm^{-1}

FIGURE 2. Couple Design Used with Tensile Specimens



NOTES:

1. CAPSULE, LID AND CHARPY SPECIMEN ARE SAME MATERIAL
2. CHARPY SPECIMEN DESIGN CORRESPONDS TO ASTM SPECIFICATION E23(10mm x 2.5mm SUBSIZE IMPACT SPECIMEN -- FIGURE 7)
3. SrF_2 IS COMPACTED TO ~70% OF THEORETICAL DENSITY
4. COUPLE METAL SURFACE TO FUEL VOLUME RATIO (S/V) ~2.8 cm^{-1}

FIGURE 3. Couple Design Used with Charpy Test Specimens

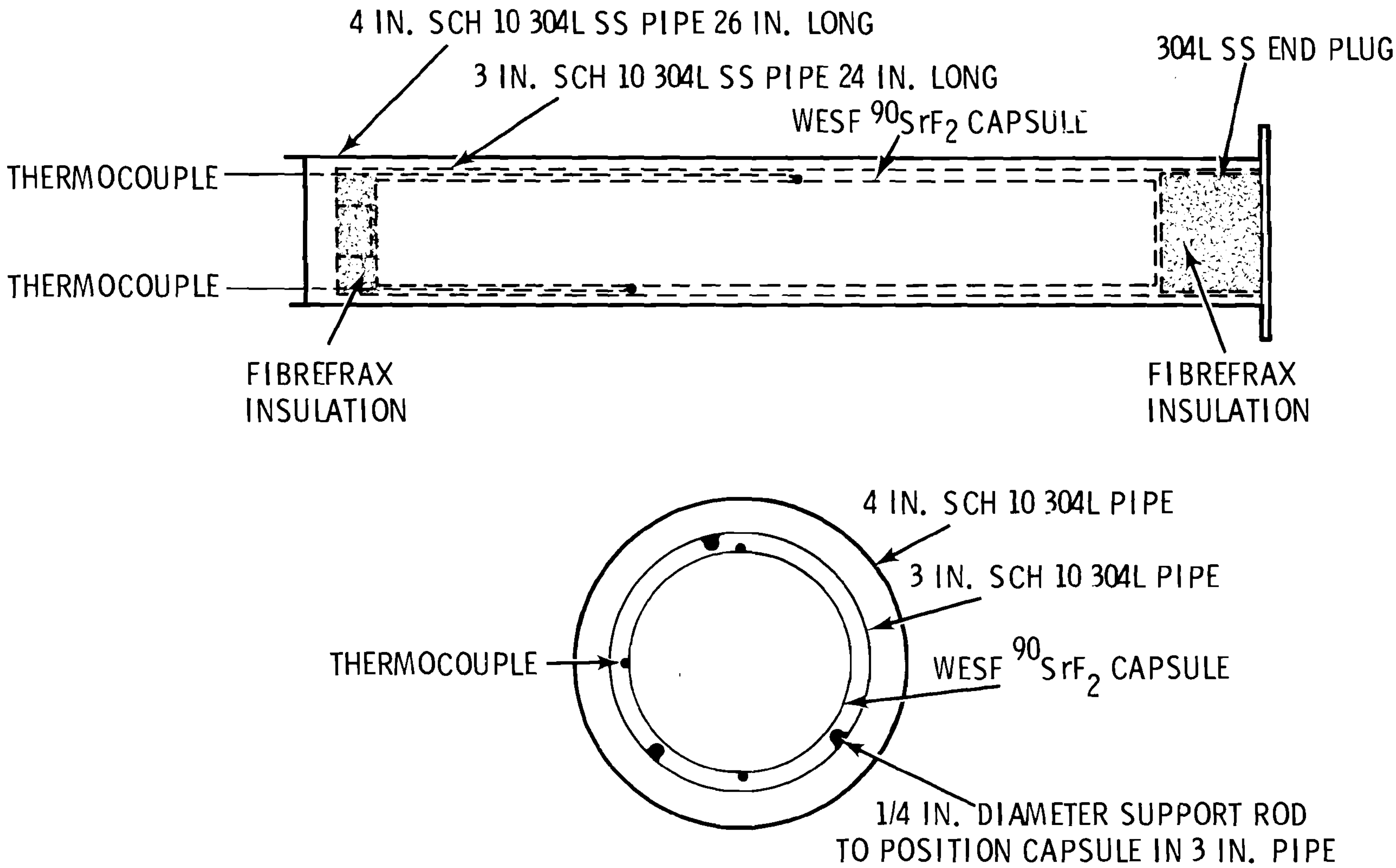


FIGURE 4. Container to Hold Full Size WESF $^{90}SrF_2$ Capsule

The topical report summarizing the results of the short-term compatibility tests is now at printing and should be ready for distribution in January.

THERMAL AGING OF HASTELLOY C-4

Tests are underway to measure the effects of thermal aging on the impact strength of Hastelloy C-4. Charpy V notch specimens are being tested at temperatures of 600, 800, 900 and 1000°C for times of 1000, 5000, 10,000 and 30,000 hr. The 1000-hr tests have been completed and the specimens examined using a Baldwin Charpy impact machine. The specimens, which were 0.145 in. thick, were tested at 25 and 250°C, and the results obtained are shown in Table 2.

TABLE 2. The Effect of Thermal Aging for 1000 hr on the Impact Strength of Hastelloy C-4

Aging Temperature °C	Charpy Impact Strength, ft-lb	
	Room Temperature	250°C
As-Received	61.8	64.2
600	38	41
800	56.1	59.0
900	76.3	78.5
1000	81.5	83.0

The results are unusual in that the specimens aged at 900 and 1000°C show a higher impact strength than the "as-received" alloy, which had been solution heat treated at 1950°F and rapid quenched. Photomicrographs were obtained of the specimens to determine the effect of thermal aging on the microstructure of the alloy (see Figure 5). Microstructure of the "as-received" material was typical of the solution heat treated alloy. The

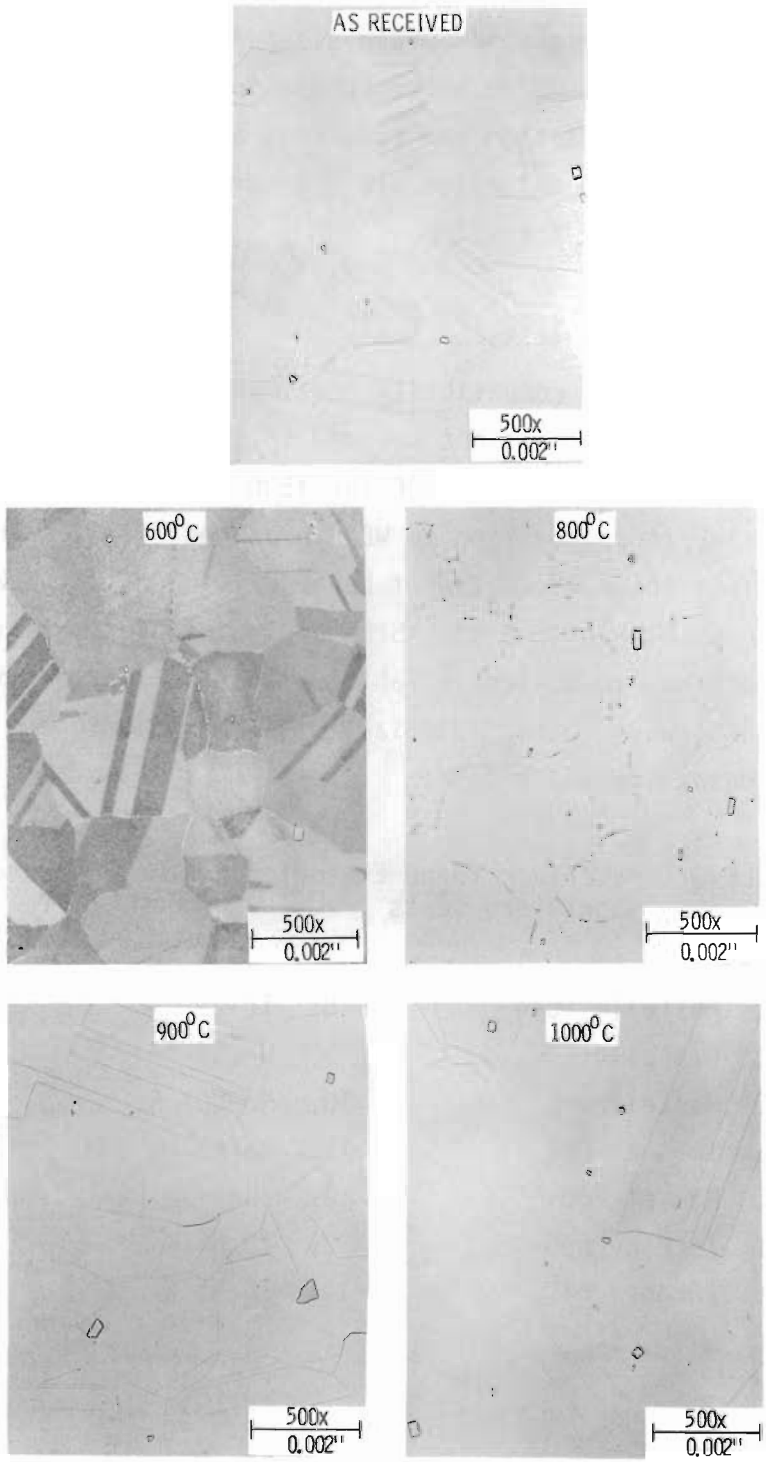


FIGURE 5. Micrographs of Hastelloy C-4 Specimens Aged 1000 hr

specimen aged at 600°C exhibited some intergranular precipitation and extensive second phase formation. Grain size was similar to the "as-received" material. At 800°C no second phase formation was apparent, but some intergranular precipitation had occurred, as well as slight grain growth. At 900 and 1000°C extensive grain growth occurred, but there was no evidence of precipitate formation.

ADDITIONAL SHORT-TERM COMPATIBILITY TESTS

Additional short-term compatiblity tests are planned to evaluate potential containment materials not covered in the initial short-term tests. The tests will be carried out at 800°C for 1500 and 4400 hr using nonradioactive SrF_2 similar in composition to WESF-produced $^{90}SrF_2$. The materials tested will include those which cannot be readily used in the WESF process but which may be usable in special $^{90}SrF_2$ applications. Materials currently scheduled for testing are tested in Table 3, and fabrication of the test couples is now underway. Other materials will be added to the test as potential candidates are identified.

TABLE 3. Materials to be Evaluated in the Advanced Short-Term Tests

1.	Hastelloy C-4	8.	Ir-2% W
2.	Hastelloy S	9.	W-25% Re
3.	Hastelloy B	10.	Mo-50% Re
4.	Inconel 671	11.	Hafnaloy
5.	Nickel 200	12.	Ductile Cast Iron
6.	Incoloy 800	13.	TD Ni
7.	Inconel 617	14.	TD Ni Cr

BENEFICIAL ISOTOPES UTILIZATION PROGRAM

K. M. Harmon

The objectives of the program for FY-1976 are to identify and develop beneficial uses of nuclear reactor by-products through: 1) estimation of long-term availability and cost of useful isotopes from commercial suppliers; 2) identification and development of beneficial applications of isotopes, including their use in remote regions of the world and 3) identification and evaluation of the actions required to optimize the $^{90}SrF_2$ and $^{137}CsCl$ products from the Hanford Waste Encapsulation and Storage Facility (WESF) for beneficial use.

The program is divided into three tasks, as follows:

Task I - Isotopes Availability
Task II - Cold Regions Applications
Task III - WESF Product Utilization

TECHNICAL PROGRESS

Task I - Isotopes Availability

The objectives are to: 1) provide an estimate of the quantities of useful isotopes which could be available from commercial nuclear power reactor operations through the year 2000, 2) define chemical flowsheets for the recovery of these isotopes, and 3) estimate capital and operating costs for an isotope recovery plant.

Calculations of Potential Supplies (C. M. Heeb)

An attachment to BNWL-B-435[1] was issued. The attachment provides: 1) additional calculations of potential supplies of useful by-products through the year 2000, and 2) projections of the effect of removing contributions from commercial HTGR's.

Task III - WESF Product Utilization

The objective is to foster the beneficial use of WESF $^{137}CsCl$ and $^{90}SrF_2$ by investigation of concepts for potential applications and by coordination of efforts to optimize capsule design.

WESF ^{137}CsCl Capsule Dosimetry (R. A. Libby and F. N. Eichner)

Dose measurements were made on WESF capsule no. 68, which contained 2.741 kg CsCl, at distances in air of 20, 30 and 50 cm from the capsule midpoint. The measured results agree well with those calculated earlier. The comparison is as follows:

Position (cm)	Experimental	Calculated
20	302,000 R/hr	300,000 R/hr
30	169,000 R/hr	160,000 R/hr
50	55,000 R/hr	68,000 R/hr

A fairly large error (10%) may be present in the calculated dose rate at 50 cm because the value was taken from a curve interpolated between points at 25 cm and 100 cm. The potential error in experimental dose rates is estimated at 10%.

WESF ^{137}CsCl Capsule Radiation Efficiencies (R. A. Libby)

A series of calculations of the efficiency of a ^{137}CsCl plate source has been completed. The source was assumed to be a plate 40 cm^2 with the dose point in the center (in the calculations, only 1/4 of the source was modeled, and symmetry was applied). Source thicknesses of 0.5, 1.0 and 2.0 cm with 0.254 cm (0.1 in.) stainless steel clad were used. Three different points were calculated for each thickness; these were located at the source-clad boundary, the surface of the clad and 1.0 cm outside the clad surface. The efficiency is defined as the ratio of the dose rate with attenuation and buildup to the dose rate with no attenuation and no buildup. The results are as follows:

Source Thickness	Location	Efficiency
0.5 cm	Source-clad	81%
0.5 cm	Clad surface	58%
0.5 cm	1.0 cm outside	79%
1.0 cm	Source-clad	78%
1.0 cm	Clad surface	59%
1.0 cm	1.0 cm outside	73%
2.0 cm	Source-clad	74%
2.0 cm	Clad surface	58%
2.0	1.0 cm outside	69%

As with cylindrical sources, the efficiency increases with smaller source thickness, due to a reduction in self-shielding. The efficiency at the clad surface is indicative of a plate source infinite in height and width, while the larger value (at 1.0 cm) would apply to the smaller plate size of 40 cm^2. As the plate size is reduced, the source more closely approaches a point source and its efficiency improves since less SS needs to be traversed to reach the detector.

The 40 cm^2 plate source thus has a radiation efficiency of between 70 and 80% at the center of the plate, but drops off towards the edges. The overall efficiency may be in the 60 to 70% range. This may be compared with the following calculated efficiencies for cylindrical sources.

1-in. diameter clad in 0.1-in. stainless steel	75%
2-in. diameter clad in 0.1-in. stainless steel	69%
2-in. diameter clad in 0.2-in. stainless steel	60%

From the above, it appears that a flat plate configuration up to 2 cm thick will not give a significantly higher efficiency than the standard WESF capsule.

REFERENCES

1. C. M. Heeb, The Availability of Useful Isotopes from Civilian Power Reactors to the Year 2000, BNWL-B-435, July 1975.

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