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BNWL-1845-32

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Pacific Northwest Laboratory Monthly Report on the Strontium Heat Source Development Program, Division of Nuclear Research and Applications for January, 1977

February 1977

Prepared for the Energy Research and Development Administration under Contract EY-76-C-06-1830

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BNWL-1845-32

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PACIFIC NORTHWEST LABORATORY MONTHLY REPORT ON THE STRONTIUM HEAT SOURCE DEVELOPMENT PROGRAM, DIVISION OF NUCLEAR RESEARCH AND APPLICATIONS FOR JANUARY 1977

H. T. Fullam

February 1977

BATTELLE
Pacific Northwest Laboratories
Richland, Washington 99352

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STRONTIUM HEAT SOURCE DEVELOPMENT PROGRAM

by

J. H. Jarrett, Program Manager
H. T. Fullam, Principal Investigator
D. G. Atteridge

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 task areas:

Task 1 - Chemical and Physical Properties of $^{90}SrF_2$
Task 2 - $^{90}SrF_2$ 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 $^{90}SrF_2$

No activity this month.

TASK 2 - $^{90}SrF_2$ COMPATIBILITY TESTS

Long-Term Compatibility Tests

Work was started at the end of January on sectioning the couples from the 6000-hr tests with WESF $^{90}SrF_2$. The tests were completed early in December, but sectioning of the test couples was delayed for several weeks because of a lack of hot cell space. As reported previously, a number of the test couples which had been held in one furnace at 800°C had failed for unknown reasons. All of the compatibility couples used in the long-term tests are sealed in protective jackets of Inconel 600 to prevent external oxidation of the couples during testing. The jackets were fabricated from 1 1/2-in. Schedule 40 Inconel 600 pipe. The couples were sealed in the jackets by TIG welding, and each jacket was leak checked prior to testing. When the failed capsules were removed from the furnace, it was found that both the Inconel 600 jackets and the test couples were badly corroded. Metallographic examination of sections from the failed capsules is now underway in an attempt to identify the cause(s) of the failures.

The 6000 hr couples which had been held at 600 and 1000°C, plus some which had been held at 800°C in a second furnace, were removed from their Inconel 600 protective jackets. None of the Inconel 600 jackets or test couples showed any visual evidence of excessive corrosion. Sectioning of the couples is now underway.

Thermal Gradient Test

In the thermal gradient test a Hastelloy C-276 capsule filled with non-radioactive strontium fluoride was subjected to a temperature gradient of about 560°C, with a maximum temperature of 920°C, for a period of 4400 hr. At the conclusion of the test the capsule was opened and SrF_2 and metal samples from various locations were analyzed to determine metal-fluoride interaction. The Hastelloy C-276 capsule was fabricated from a 26-in. long section of 1 1/2 in. Schedule 40 seamless pipe using 1/8-in. thick end caps. The strontium fluoride was cold-pressed into the capsule to a final density of approximately 70% of the theoretical density. The filled capsule was closed by TIG welding the end cap in place. The nonradioactive strontium fluoride contained impurities similar to those found in WESF-produced $^{90}SrF_2$. The composition of the fluoride is given in Table 1. The exposed metal surface-to-fuel volume ratio (S/V) for the capsule was 1.1 which was slightly larger than that of the WESF $^{90}SrF_2$ capsule (0.9).

Chromel-Alumel thermocouples (5) were welded to the surface of the capsule at various locations to monitor the capsule temperature gradient throughout the test. The capsule was placed in a heavy wall 316L stainless steel jacket which in turn was placed in a horizontal three-zone tube furnace. Each zone of the furnace was controlled independently allowing a temperature gradient to be maintained along the length of the capsule. Figure 1 shows the capsule surface temperature as a function of location. The temperature varied from 360 to 920°C giving a thermal gradient of 560°C over the length of the capsule. Temperature control was quite stable and the maximum temperature fluctuation observed at any location during the test was ±6°C.

TABLE 1. Composition of the SrF_2 Used in the Thermal Gradient Test

Component	Wt %	Component	Wt %
Sr	65.9	Nd	1.0
Al	0.26	Ni	0.02
Ba	0.70	Pb	0.06
Ca	0.21	Si	0.02
Cr	0.050	F	29.5
Fe	0.022	H_2O	<0.01
K	0.010	NO_3^-	<0.01
Mg	0.13	O	<0.01
Na	2.5		

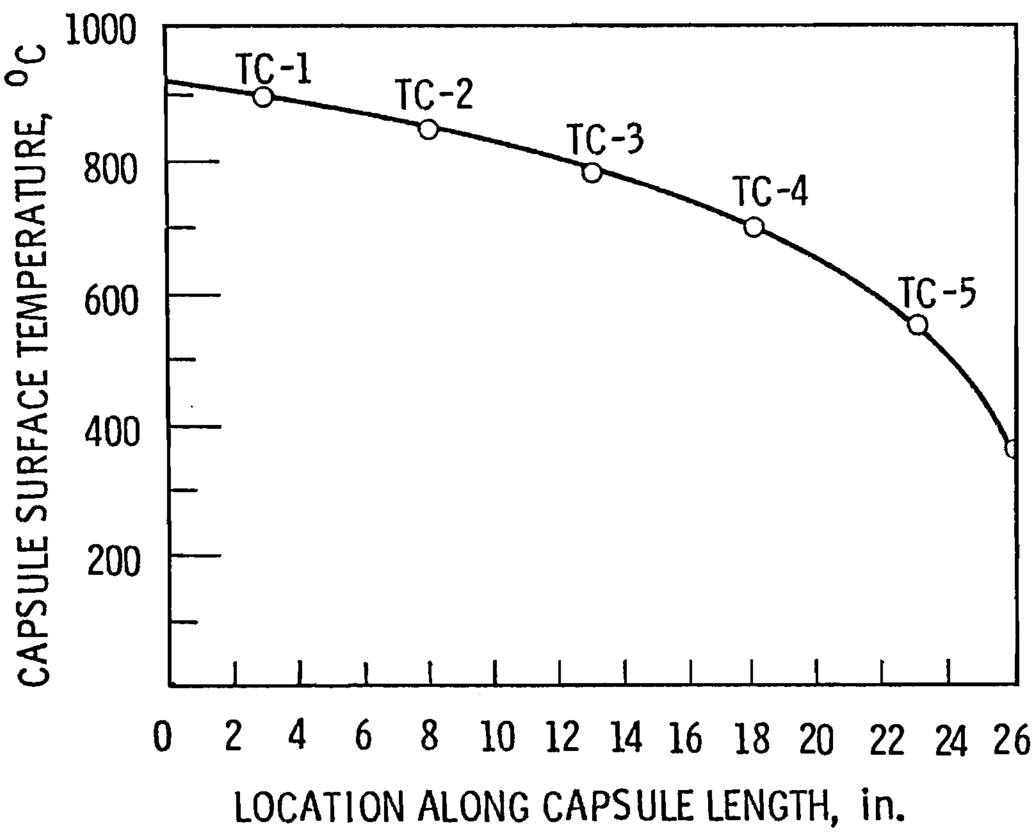


FIGURE 1. Capsule Surface Temperature at Various Locations

At the conclusion of the test the capsule was cut into several sections using an abrasive saw. The sectioned capsule is shown in Figure 2. The temperatures indicated in the photograph are the approximate test temperatures at the locations where the cuts were made. The strontium fluoride in the capsule varied from a compacted powder at the cold end of the capsule (<680°C) to a highly sintered porous mass at the hot end. The SrF_2 which had been held above about 850°C adhered strongly to the inner capsule surface. The SrF_2 which had been held between approximately 750 and 850°C appeared to have sintered and shrank from the capsule wall and was easily removed from the capsule. The SrF_2 in the cold end had not sintered to any appreciable degree and was easily removed from the capsule. The color of the SrF_2 varied from white at the cold end of the capsule to pale blue at the hot end.

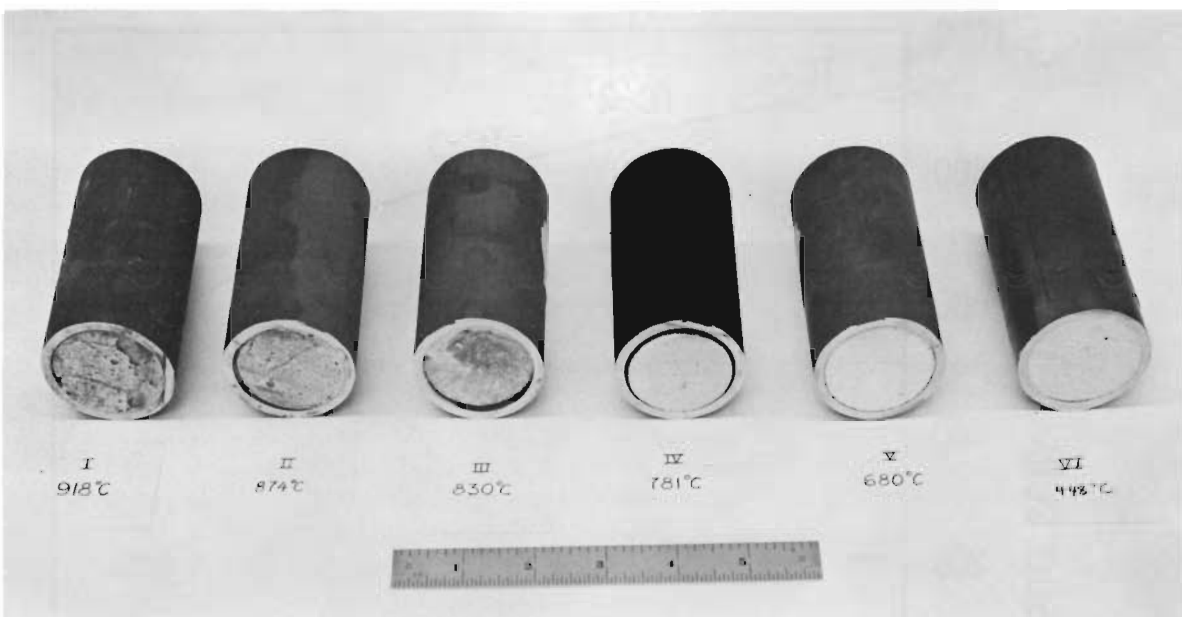
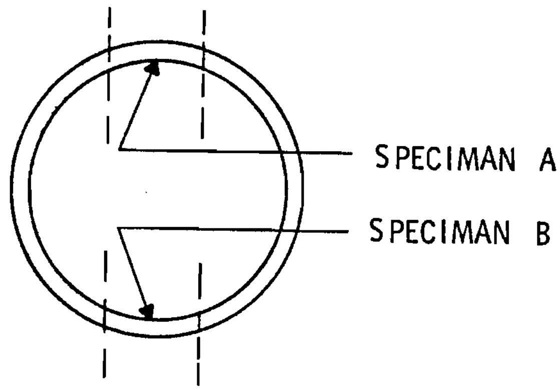


FIGURE 2. Sectioned Capsule from the Thermal Gradient Test

Specimens were taken from various locations in the capsule and examined using metallographic techniques to determine the extent of alloy-fluoride interaction. At each location where a specimen was to be taken, a ring approximately 3/4-in. wide was cut from the capsule. The ring was then split as shown below to give two diametrically opposed specimens. Each pair of specimens was examined and photomicrographs obtained. Figure 3 shows micrographs of the specimen surfaces which had been in contact with the fluoride. Micrographs showing changes in the alloy microstructure as a function of temperature are presented in Figure 4.



Estimates of metal attack as a function of temperature based on the micrographs are given in Table 2. The attack observed with each pair of specimens was very similar; and the estimates provided in Table 2 represent the maximum attack observed with each pair. Chemical attack of the Hastelloy C-276 increased sharply with increasing exposure temperature. The type of attack observed also varied with the temperature. At the lower temperatures the attack was relatively uniform across the surface with only slight indication of grain boundary attack. At the higher temperatures (≥830°C) extensive grain boundary attack and subsurface void formation was observed, and the reaction layer which formed contained free grains of unreacted metal.

Contact with the fluoride also produced some marked changes in the alloy microstructure. The depth of metal affected by microstructural changes increased with increasing exposure temperature up to about 870°C and then apparently decreased at higher temperatures. The microstructural changes consisted primarily of the disappearance of normal alloy precipitates.

Samples of the strontium fluoride were taken from various locations in the capsule and analyzed to determine if changes in the impurity levels had occurred due to the thermal gradient imposed on the capsule or due to reactions with the Hastelloy C-276. The analytical results indicate that the

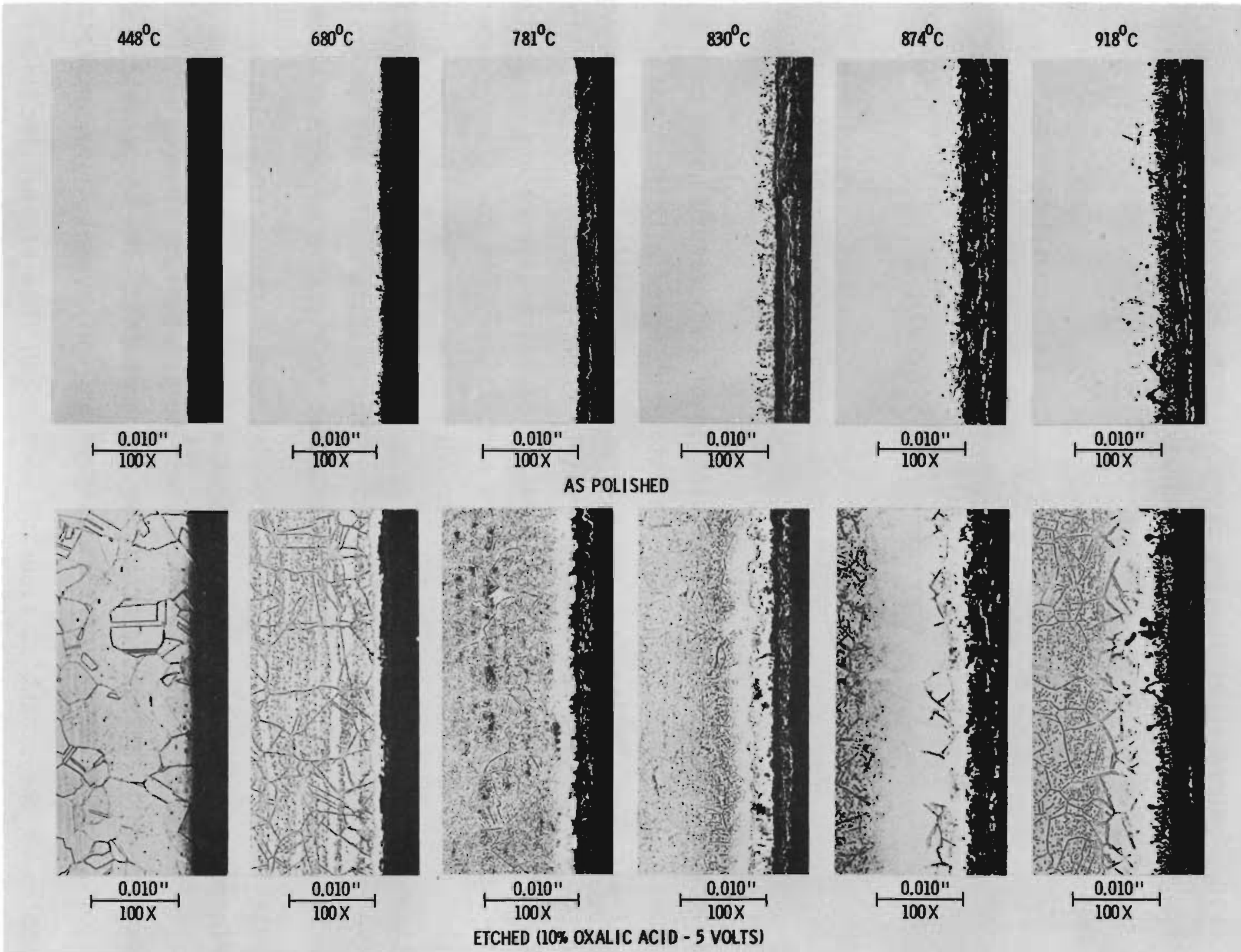


FIGURE 3. Hastelloy C-276 Specimens Exposed to Nonradioactive SrF_2 for 4400 hr

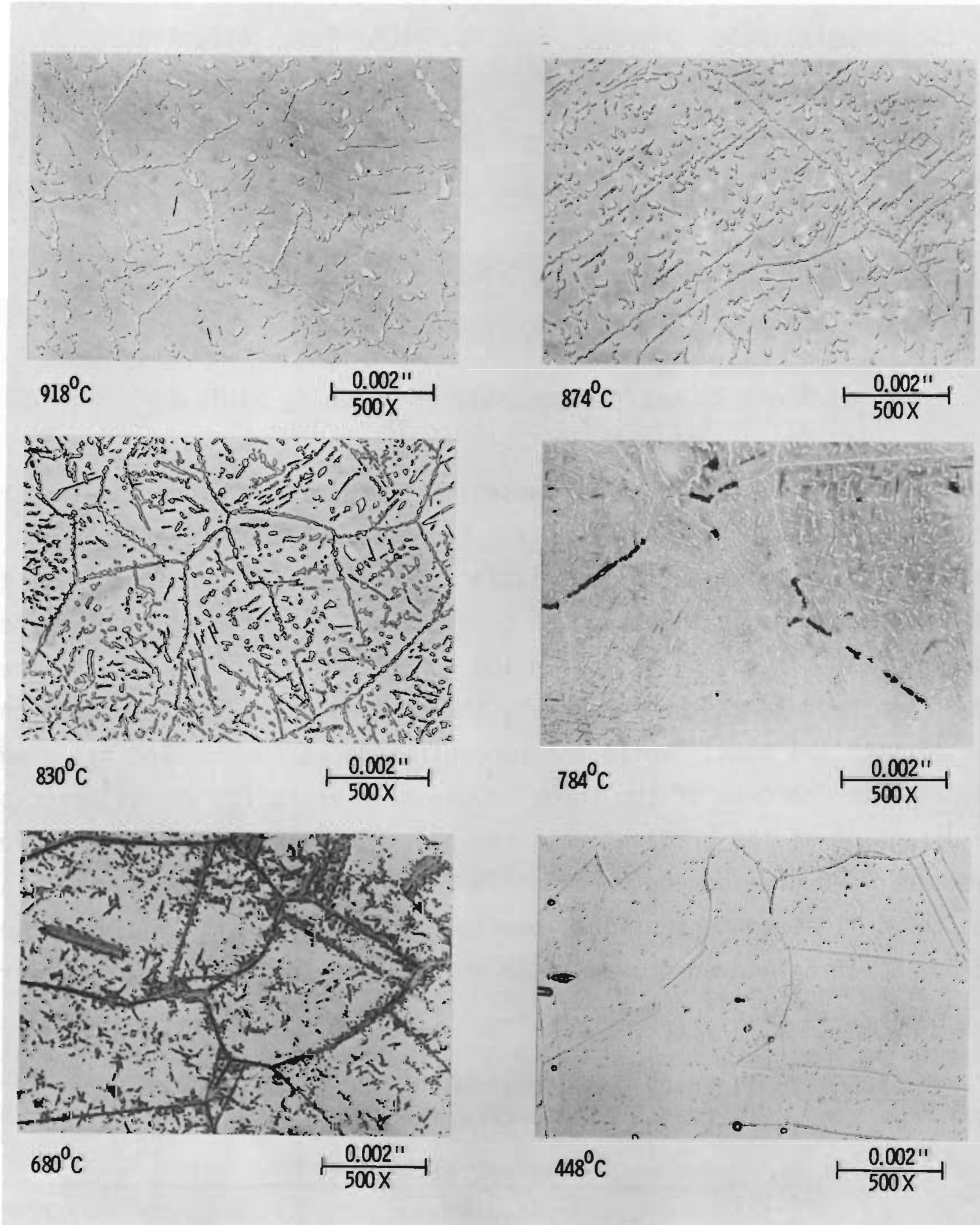


FIGURE 4. The Microstructure of Hastelloy C-276 Held at Various Temperatures for 4400 hr

TABLE 2. The Effect of Temperature on the Attack of Hastelloy C-276 by Nonradioactive Strontium Fluoride[a]

Approximate Interface Temperature, °C	Exposure Time, hr	Depth of Metal Affected, mils[b] Chemical Attack	Change in Microstructure
918	4400	8	9
874	4400	7	15
830	4400	6	11
781	4400	4	6
680	4400	2	3
448	4400	<<1	<1

(a) The SrF_2 had the composition shown in Table 1.
(b) Attack estimated from photomicrographs.

impurity levels in the SrF_2 were essentially unchanged along the length of the capsule except for two elements: iron and chromium (see Table 3). Both the iron and chromium exhibited distinct concentration gradients over the length of the capsule although the gradient directions were reversed for the two elements. In the case of iron the concentration in the SrF_2 at the cold end of the capsule was approximately the same as the bulk concentration at the start of the test, while the concentration at the hot end was greatly reduced. In the case of the chromium the same variation was observed but the direction of the gradient was reversed. The data show that the overall concentrations of iron and chromium in the SrF_2 were reduced substantially from the initial concentrations, probably by reaction with the Hastelloy C-276. Electron microprobe analysis of the test specimens is now underway to identify the reaction mechanisms.

TABLE 3. Impurity Concentrations in SrF_2 Samples from the Thermal Gradient Experiment

Element	Concentration, wt% Initial SrF_2	448°C	680°C	781°C	830°C	874°C	918°C
Na	2.5	2.7	2.5	2.9	2.4	2.4	2.4
Fe	0.022	0.016	0.019	0.020	0.013	0.006	0.002
Cr	0.055	0.006	0.015	0.014	0.025	0.059	0.055
Si	0.02	0.02	0.01	0.02	0.03	0.02	0.02
Ca	0.21	0.27	0.19	0.21	0.24	0.20	0.22

TASK 3 - CAPSULE QUALIFICATION AND LICENSING

Heat Source Capsule Qualification Requirements

A list of capsule qualification requirements is currently being compiled for resubmission to DNRA. A study into the vibration testing aspect of these qualifications by the Structures and Mechanics Section is nearing completion. This study will determine the need for WESF capsule vibration testing and, if it is established that vibration testing is needed, the test will also determine what kind of test matrix should be required.

Capsule Design Philosophy

The study of determining the critical design-limiting outer capsule component is continuing, along with a study of capsule design using failure criterion versus elastic strain criterion.

The basic design of the outer capsule will be a right circular cylinder large enough to enclose the present WESF Hastelloy C-276 inner capsule. No concentrated effort will be made in the capsule design area until the recommendations for the Capsule Design Philosophy have been made to DNRA and they are accepted.

Outer Capsule Material Selection

Outer capsule material options are being explored through a continuing literature review and manufacturing contacts as well as the mechanical and corrosion resistant property tests being run at PNL. Thermally-aged and tested tensile specimens of Hastelloy C-4 and Hastelloy S have been received from the Cabot Corporation in Kokomo, Indiana and are currently being prepared to be remachined into subsize tensile specimens for elevated temperature testing. The received specimens are tensile specimens that were aged at various times and temperatures and then tested at room temperature. The aging temperatures varied from 800 to 1600°F in 200°F increments while the aging times were 1000, 4000, 8000 and 16000 hr.

PNL received one tested specimen for each test condition. Two standard ASTM subsize tensile specimens, currently used in the compatibility section of this program, will be machined out of the broken ends of the original tensile specimens. The gage section of the subsize specimens will come from the no-plastic-strain shoulder region of the old specimens, as illustrated in Figure 5. It is currently planned that both of the new specimens will be tested at elevated temperatures: one at the aging temperature of the original sample, and the other at 800°C, the currently predicted maximum allowable use temperature of the WESF capsule.

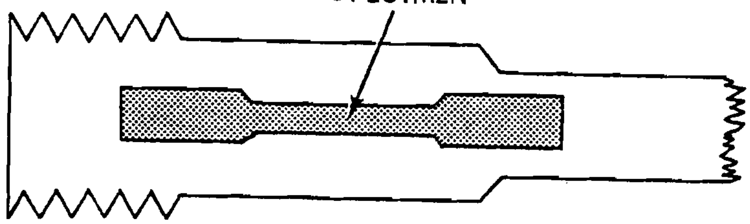


FIGURE 5. Subsized Tensile Specimens to be Machined from Sections of Tested Specimens

Oxidation Resistance of Hastelloy C-4 and Hastelloy S

Experiments are underway to determine the effects of thermal aging on the oxidation resistance of Hastelloy C-4 and Hastelloy S. When the solution heat-treated forms of the two alloys are heated at temperatures between approximately 500 and 900°C, they undergo long-range ordering reactions and precipitate formation which affect their mechanical properties.[1] The reactions involved proceed relatively slow and occur over a several thousand hour period when the alloys are heated. It is anticipated that the microstructural changes which occur when the alloys are heated between 500 and 900°C could affect their resistance to oxidation.

Specimens of the two alloys, which had been held at 600, 700, 800 or 900°C for 1000 hr, were heated in air and the rate of oxidation determined as a function of time. At 1000°C or less the oxidation rates of the thermally aged specimens were similar to those previously reported for the solution heat treated alloys. At 1100°C, however, there were marked differences in the oxidation rates of the aged and unaged specimens. Figure 6 shows the results obtained with Hastelloy C-4. When Hastelloy C-4 is heated in air at 1100°C for extended periods of time spalling of the oxide layer occurs. Metallographic examination of test specimens indicates that a very thin uniform reaction is retained on the specimen surface and most of the oxide layer is lost by spalling. Once spalling begins, with the mill-annealed (solution heat treated) alloy, the sample weight loss exhibits a linear time dependence. Thermal aging at 600 to 900°C apparently increases the rate of oxidation (and weight loss) of the alloy initially, but after approximately 250 hr exposure the oxidation rate decreased sharply. The reduced rate was less than that observed with the mill-annealed alloy. Metallographic examination of the test specimens is now underway to determine what causes the changes in the oxidation rates of the aged specimens after 250 hr exposure.

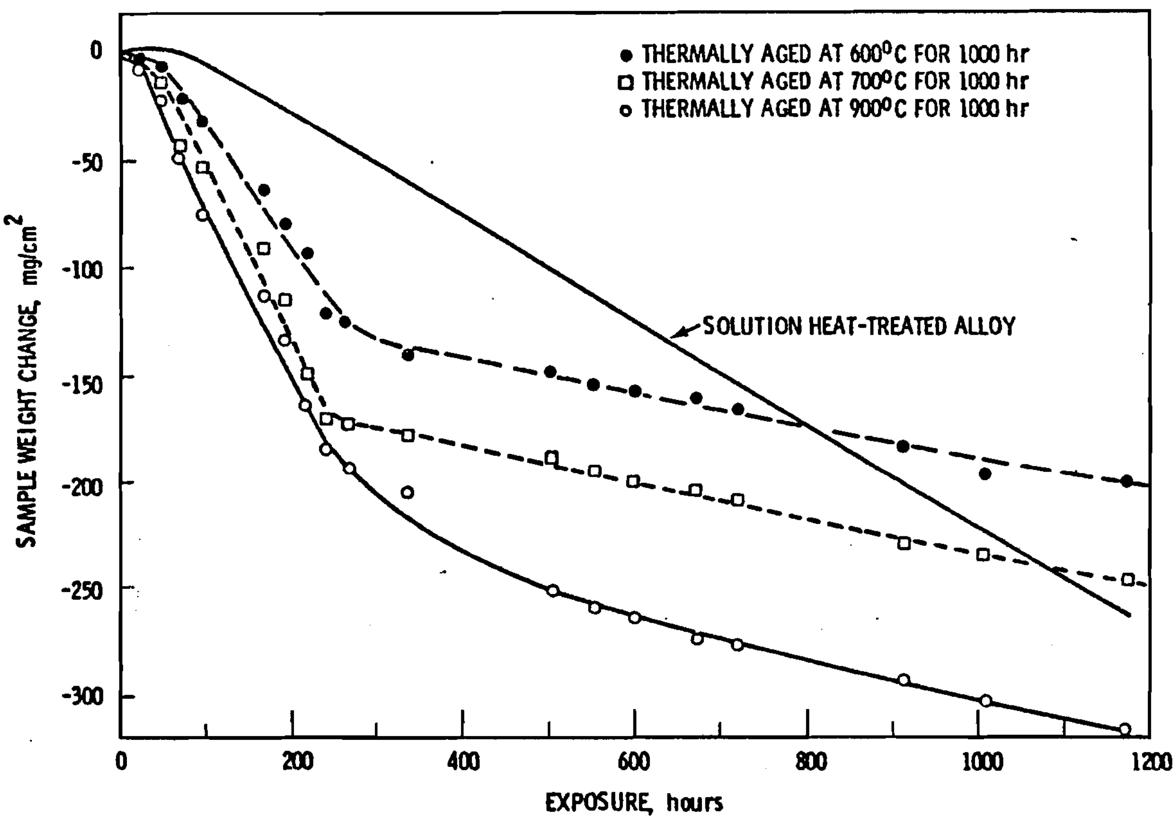


FIGURE 6. The Effect of Thermal Aging on the Oxidation Resistance of Hastelloy C-4 at 1100°C

The results obtained with Hastelloy S are presented in Figure 7. At 1100°C an adherent oxide layer formed on the mill-annealed alloy initially, but after approximately 50 hr exposure slight spalling of the oxide layer began. After approximately 550 hr exposure the spalling increased drastically and the specimens lost weight very rapidly. The alloy specimens which had been thermally aged at 600°C exhibited a slow continuous weight loss, due to oxide spalling, which was linearly time dependent. Examination of the test specimens indicated oxide spalling was uniform across the specimen surface and only a thin reaction layer was retained on the surface. Similar results were obtained with the Hastelloy S specimens which had been aged at 700 and 800°C. The specimens aged at 900°C showed a marked difference in oxidation resistance from those aged at the lower temperature; they suffered a very rapid weight loss initially but after approximately 200 hr exposure the rate of weight loss decreased to about the level observed with the specimens aged at 600 to 800°C. Metallographic examination of the specimens is underway to identify the reason(s) for the differences in the oxidation rates.

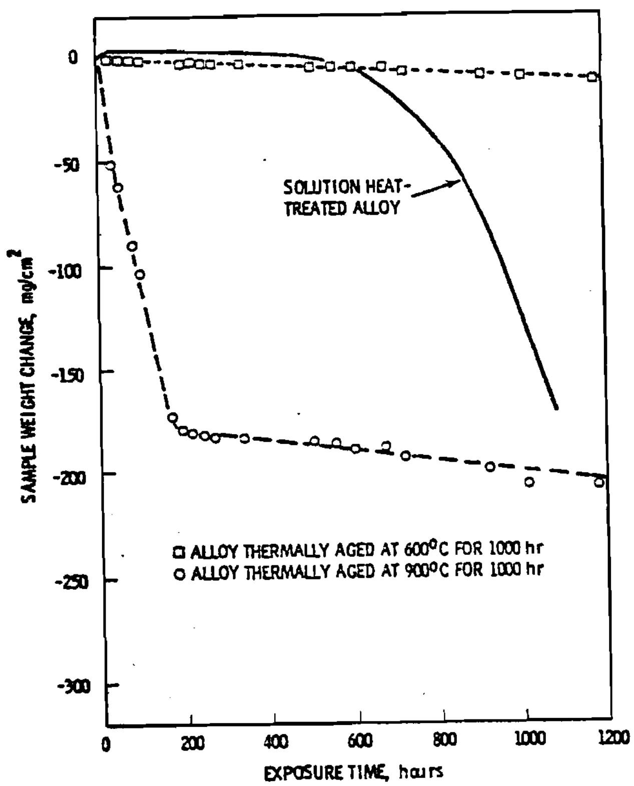


FIGURE 7. The Effect of Thermal Aging on the Oxidation Resistance of Hastelloy S at 1100°C

Additional oxidation experiments are planned using Hastelloy C-4 and Hastelloy S specimens which have been aged at 600 to 900°C for periods up to 10,000 hr.

Prototype Capsule Fabrication and Testing

The cart for the helium leak detector, which will be used to leak check prototype capsules during fabrication and testing, has arrived. Both the leak detector system and the capsule leak detection chamber are currently being fitted to the cart.

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1. S. J. Matthews, Thermal Stability of Solid Solution Strengthened High Performance Alloys, Report No. 8979, Stellite Division, Cabot Corp., February 1976.

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