14

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Quarterly Report on the Strontium Heat Source Development Program, Advanced Systems and Materials Production Division for April-June 1978

H. T. Fullam

July 1978

Prepared for the U.S. Department of Energy
under Contract EY-76-C-06-1830

Pacific Northwest Laboratory
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QUARTERLY REPORT ON THE STRONTIUM HEAT SOURCE DEVELOPMENT PROGRAM, ADVANCED SYSTEMS AND MATERIALS PRODUCTION DIVISION FOR APRIL-JUNE 1978

H. T. Fullam

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Prepared for
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under Contract EY-76-C-06-1830

Pacific Northwest Laboratory
Richland, Washington 99352

SUMMARY

All of the remaining compatibility tests are proceeding as scheduled. Results of the 20,000-hr compatibility tests with nonradioactive SrF_2 show that, with one exception, attack of the specimens by the SrF_2 after 20,000 hr is no greater than after 6000 and 12,000 hr. The one exception involved Haynes Alloy 25 specimens tested at 1000°C. These test specimens exhibited isolated subsurface voids to depths of 0.025 in., although the general surface attack was only about 0.001 inches. The cause of the void formation has not yet been determined.

Tests with Hastelloy S and Hastelloy C-4 at 600 to 800°C for 2500 hr show that oxidation of the two alloys adheres to the parabolic rate law. Both alloys form adherent oxide layers over the temperature range of interest. Intergranular attack, pitting, and subsurface void formation increase sharply with increased oxidation temperature with both alloys, and at 800°C the depth of metal exhibiting oxidation damage is several times greater than the thickness of the oxide layer.

CONTENTS

FIGURES

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 $^{90}SrF_2$ 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 (H. T. Fullam)

Completion of the theoretical analysis of the consequences of a $^{90}SrF_2$ capsule rupture in an ocean environment has been delayed until FY 1979. Unexpected costs relating to maintenance of the hot cell in which the long-term $^{90}SrF_2$ compatibility tests are being carried out necessitated the delay.

TASK 2 - $^{90}SrF_2$ COMPATIBILITY STUDIES (H. T. Fullam)

All of the remaining compatibility tests are proceeding as scheduled; completion dates are as follows:(a)

- The 20,000-hr test with $^{90}SrF_2$ will be completed by July 15, 1978.
- The 30,000-hr test with $^{90}SrF_2$ will be completed by September 6, 1979.
- The 30,000-hr test with nonradioactive SrF_2 will be completed by June 19, 1979.

(a) Completion dates assuming no additional delays due to equipment problems.

The Oak Ridge National Laboratory has completed sectioning of the test capsules used in the 12,000-hr tests with $^{90}SrF_2$. The metal test specimens were easily removed from the capsules, and very little of the $^{90}SrF_2$ adhered to the specimens' surfaces. Conversely, in the 1000- and 6000-hr tests the $^{90}SrF_2$ adhered strongly to the specimens' surfaces. The physical differences observed among the different sets of capsules may be due to additional sintering of the $^{90}SrF_2$ in the 12,000-hr capsules. Oak Ridge National Laboratory has begun metallographic examination of the compatibility specimens from the 12,000-hr tests, as well as testing of the tensile and Charpy specimens from the tests. These activities should be completed by the end of the fiscal year.

The 20,000-hr tests with nonradioactive SrF_2 have been completed, and the specimens examined. Estimates of metal attack, based on photomicrographs of the specimens, are presented in Table 1. The results obtained in the shorter-duration tests are also included for comparison purposes. Evaluation of the data presented in Table 1 shows that, except for two specimens, very little increase in chemical attack of the specimens occurred after 6000-hr exposure. The two specimens which suffered increased attack were Haynes Alloy 25 specimens tested at 1000°C. These specimens exhibited isolated subsurface voids to a considerable depth, although the general surface attack was limited. Changes in alloy microstructure, due to SrF_2 attack, remained essentially constant after 6000-hr exposure, and in the case of the Haynes Alloy 25 may have decreased with exposures of more than 6000 hr.

The Hastelloy C-276 specimens exposed to nonradioactive SrF_2 for 20,000 hr suffered a limited chemical attack at 600°C that consisted primarily of a general surface attack with formation of an adherent reaction layer. Some intergranular attack and subsurface void formation

TABLE 1. Attack of Metal Specimens Exposed to Nonradioactive SrF_2[a,b]

Metal Tested	Test Temperature °C	Depth of Metal Affected, mils[c]							
		Chemical Attack				Change in Microstructure			
		1000 hr	6000 hr	12,000 hr	20,000 hr	1000 hr	6000 hr	12,000 hr	20,000 hr
Hast C-276	600	<1	3	2	1	0	4	8	6
	600	1	4	2	1	5	5	7	4
	800	<1	2	2	3	<1	3	3	4
	800	1	2	2	2	3	3	2	2
	1000	2	2	4	3	4	6	7	6
	1000	3	3	2	3	4	7	8	5
Haynes 25	600	<1	2	2	2	5	3	2	0
	600	<1	1	2	2	3	2	<1	0
	800	<1	3	3	2	3	6	4	2
	800	<1	3	3	3	1	4	5	2
	1000	2	3	4	25[d]	1	7	4	4
	1000	2	3	3	18[d]	1	4	12	5
TZM	600	0	<1	0	<1	0	0	0	0
	600	<1	<1	-	<1	0	0	-	0
	800	<1	<1	<1	<1	0	0	0	0
	800	<1	<1	<1	<1	0	0	0	0
	1000	<1	<1	<1	<1	0	0	0	1
	1000	<1	<1	<1	<1	0	0	0	1

(a) The SrF_2 contained impurities approximating those found in WESF-produced $^{90}SrF_2$.
(b) Each test couple had a metal surface to fuel volume ratio (S/V) of 4.5 cm^{-1}.
(c) Estimated from specimen photomicrographs.
(d) Specimen exhibited isolated voids to a considerable depth.

was also indicated (see Figure 1). Microstructural changes consisted of an extensive surface layer containing intergranular precipitates not found in the body of the specimens. The chemical attack increased at 800°C, and the attack mechanism appeared to change from that observed at 600°C. At 800°C the attack consisted of a general surface dissolution with some pitting, isolated grain boundary attack, and subsurface void formation. Microstructure changes consisted of a surface layer in which normal alloy precipitates were partially depleted. At 1000°C the chemical attack consisted of general surface dissolution with some grain boundary attack and extensive void formation. Microstructural changes consisted of a narrow surface layer containing finely dispersed precipitates not found in the body of the specimen, and a broad inner zone depleted of normal alloy precipitates.

Overall, the attack of the 20,000-hr Hastelloy C-276 specimens was very similar to that observed with specimens tested for shorter time periods. This can be seen by referring to Figures 2 and 3, which show specimens that had been tested at 600°C and 800°C for 1000 to 20,000 hours. At 600°C all of the specimens exhibit the same types of attack, and the extent of attack remains relatively constant after 6000 hr exposure. Likewise, the 800°C specimens all exhibit the same type of attack with little increase in the level of attack after 6000-hr exposure (Figure 3). Similar results were obtained with the 1000°C tests.

The Haynes Alloy 25 specimens exposed to nonradioactive SrF_2 at 600°C suffered limited chemical attack, but no changes in alloy microstructure due to SrF_2 attack were observed (see Figure 4). The chemical attack consisted of a general surface attack with formation of an adherent reaction layer. At 800°C the chemical attack consisted of a general dissolution of the specimen surface with some pitting. Microstructural changes consisted of a narrow surface layer depleted of normal alloy precipitates and a thin inner zone containing irregular discrete precipitates not found in the body of the specimen. The 1000°C Haynes Alloy 25 specimens exhibited isolated subsurface voids to a depth of 0.025 in., but except for the void formation, they exhibited only a limited general surface attack (about 0.001 in.). Microstructural changes consisted of a broad surface layer depleted of normal alloy precipitates.

In general, the Haynes Alloy 25 specimens exposed to SrF_2 for 20,000 hr suffered the same types of attack as corresponding specimens tested for shorter time periods. Except for the specimens tested at 1000°C for 20,000 hr, the attack of the Haynes Alloy 25 specimens remained relatively constant after 6000-hr exposure.

The TZM specimens exposed to nonradioactive SrF_2 for 20,000 hr suffered limited chemical attack (see Figure 5) which increased slightly with increased exposure temperature. The type of attack appears to be the same at all three temperatures and apparently involves surface dissolution. The alloy microstructure seems to be unaffected at the lower test temperatures, but at 1000°C it appears that some recrystallization of molybdenum has occurred at the specimens' surfaces. Overall, the TZM specimens exposed to SrF_2 for 20,000 hr exhibit the same types and levels of attack as the specimens tested for 6000 to 12,000 hr.

Based on the results obtained to date, it appears that attack of test specimens exposed to SrF_2 at 600 to 1000°C proceeds quite rapidly during the first few thousand hours of exposure and then remains relatively constant thereafter. The results indicate that the attack observed is due primarily to impurities in the SrF_2 that are consumed in a relatively short period of time.

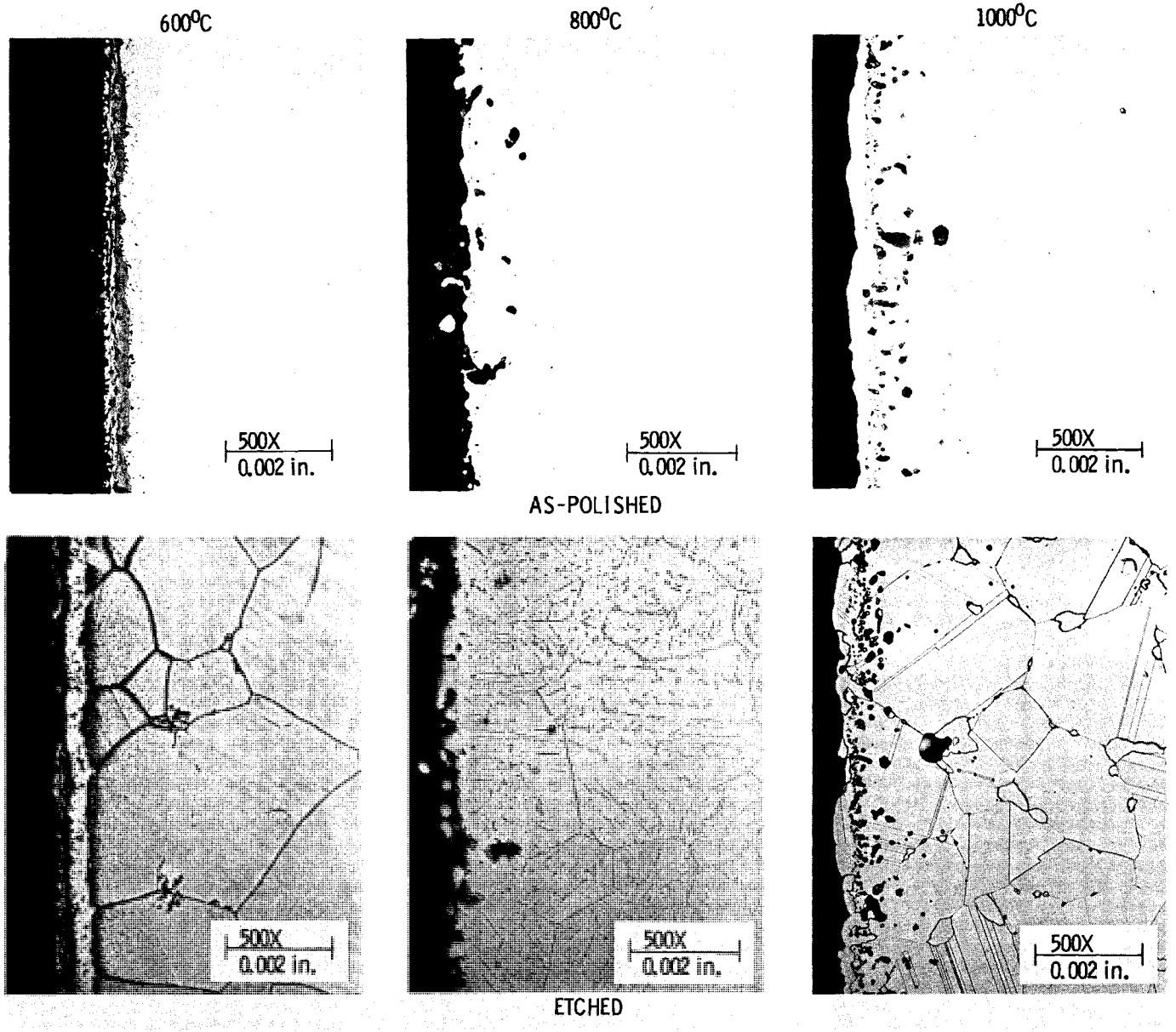

FIGURE 1. Hastelloy C-276 Specimens Exposed to Nonradioactive SrF_2 for 20,000 hr

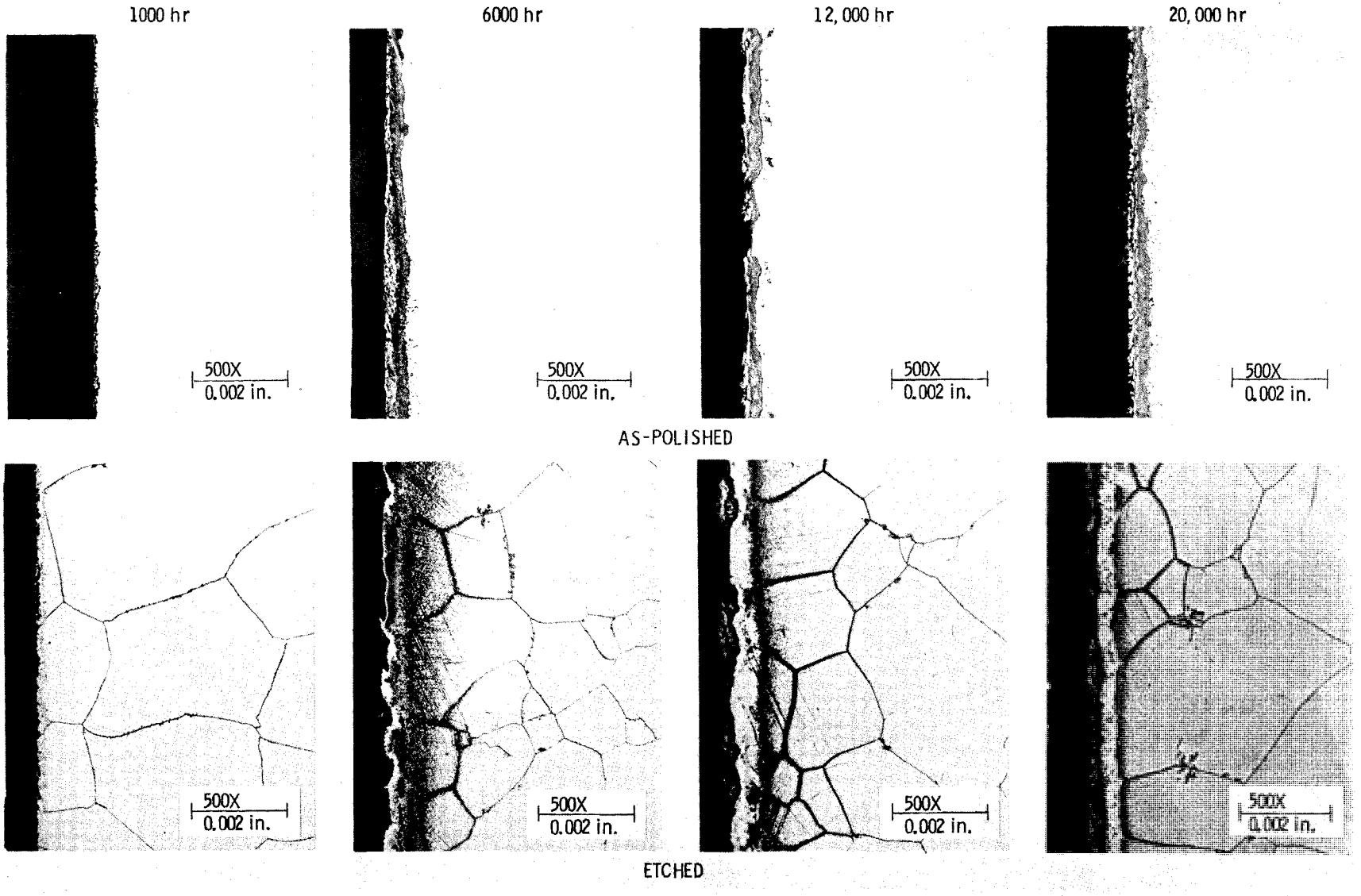

FIGURE 2. Hastelloy C-276 Specimens Exposed to Nonradioactive SrF_2 at 600°C

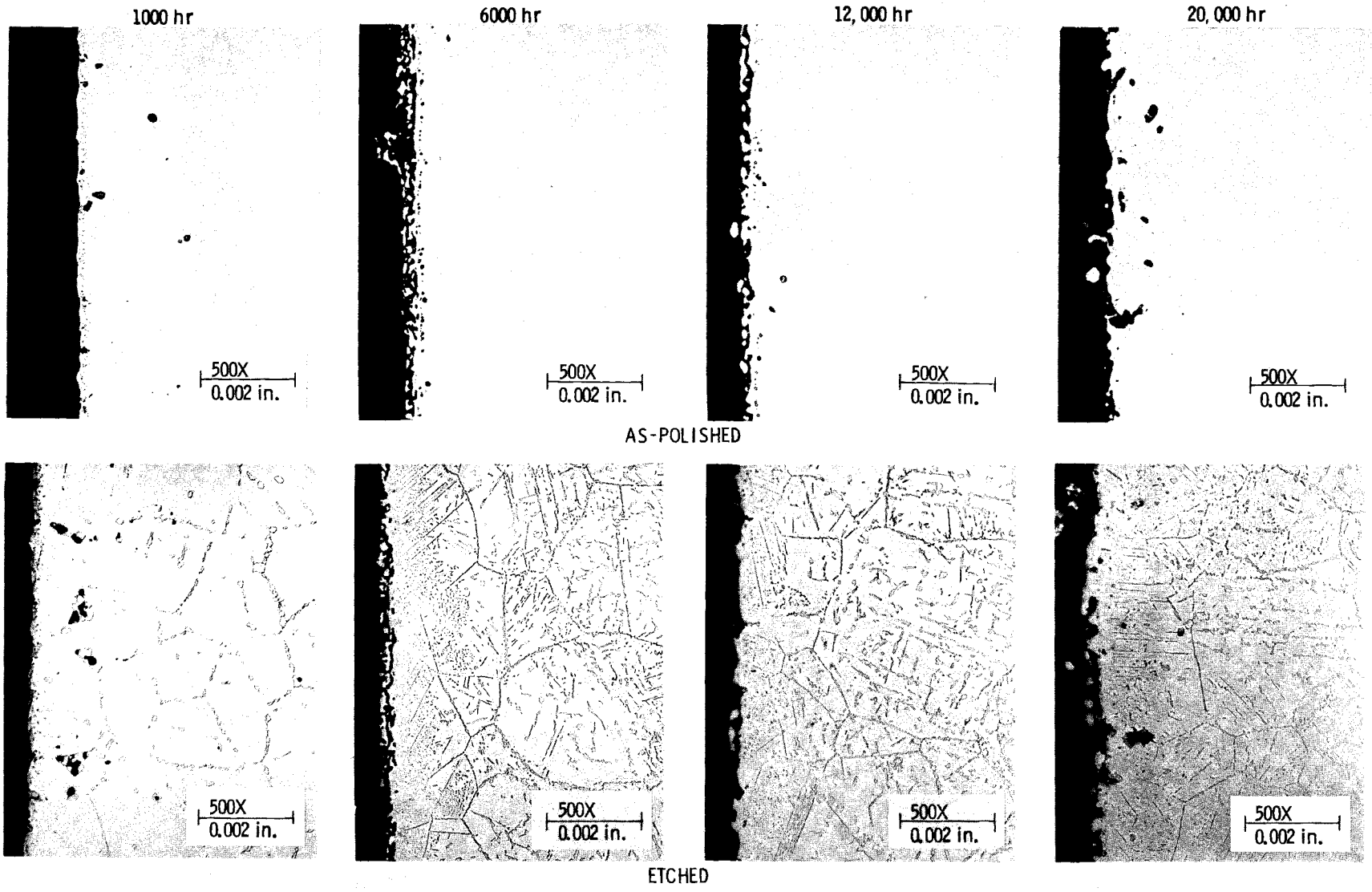

FIGURE 3. Hastelloy C-276 Specimens Exposed to Nonradioactive SrF_2 at 800°C

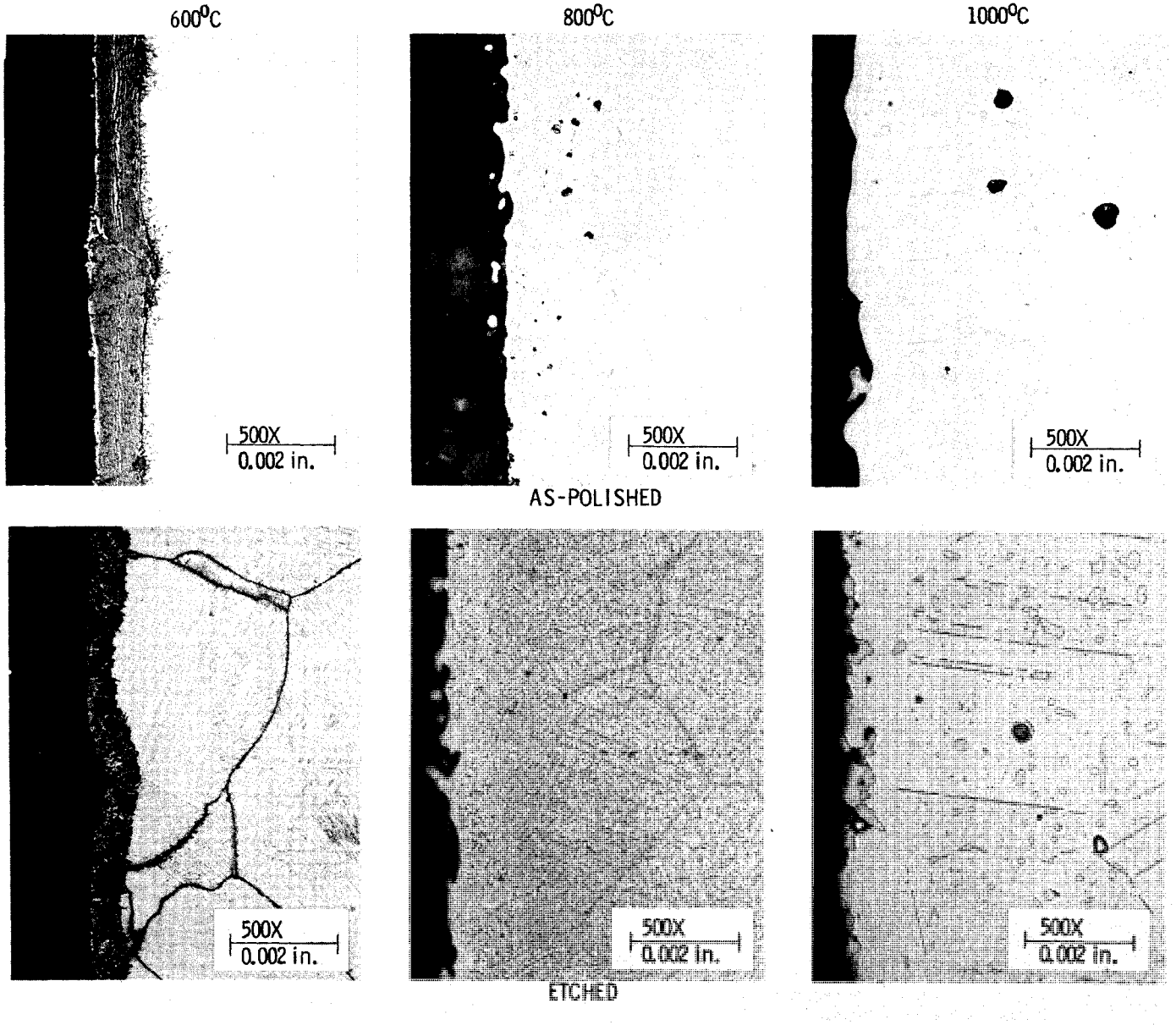

FIGURE 4. Haynes Alloy 25 Specimens Exposed to Nonradioactive SrF_2 for 20,000 hr

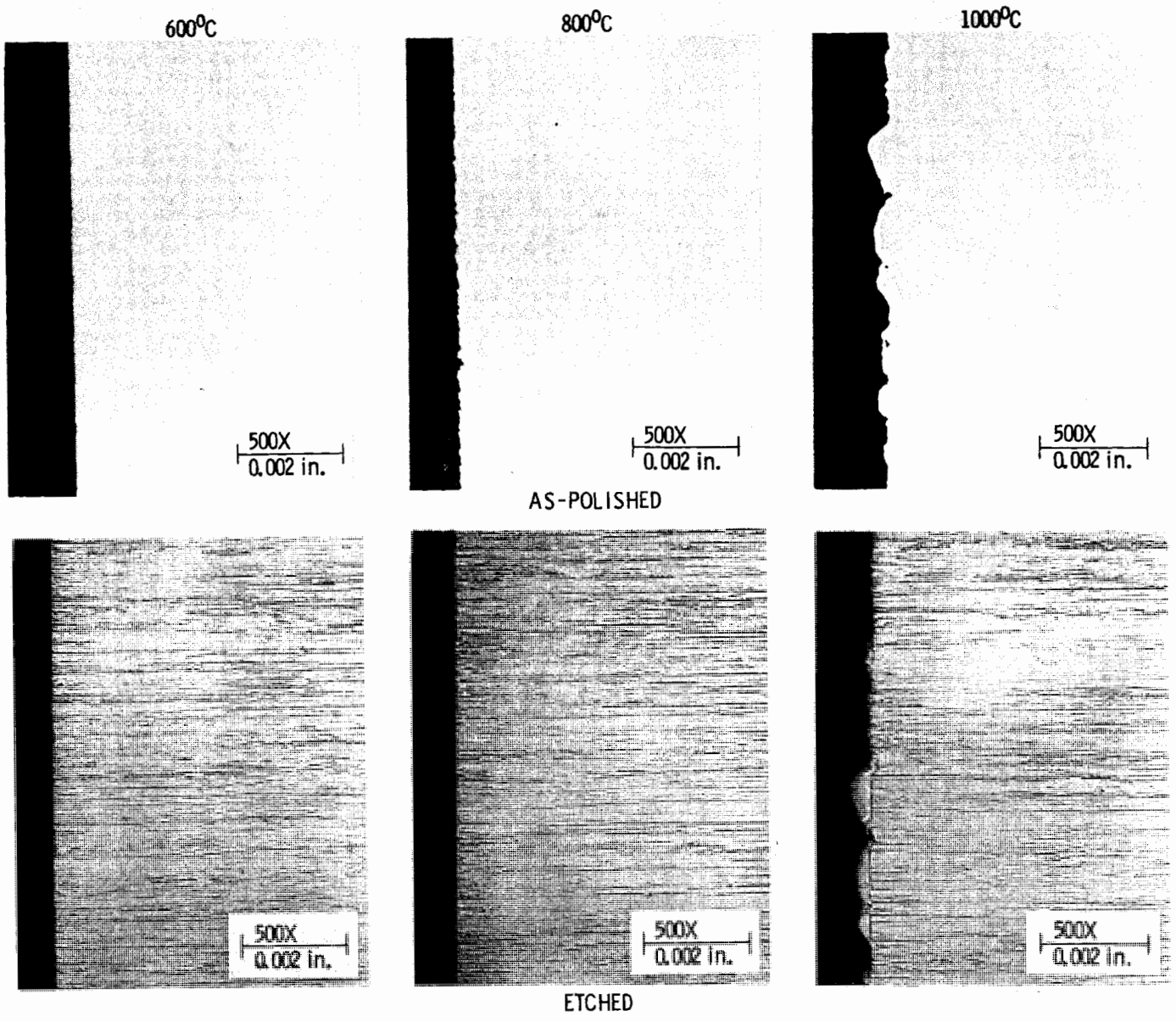

FIGURE 5. TZM Specimens Exposed to Nonradioactive SrF_2 for 20,000 hr

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

Capsule Design

Work is continuing on the evaluation of the Pacific Northwest Laboratory (PNL) outer capsule design. Several AISI 1018 steel prototype test capsules are being fabricated in order to allow experimental testing of the preliminary design. The steel capsules will be tested as stand-ins for thermally-aged Hastelloy S, and will be subjected to the external pressure test (1000 bar), the drop test (30 ft), and the puncture test.

A steel capsule welding parameter study was initiated this quarter and has progressed to the stage where test specimen welding is imminent. The main difficulty encountered to date has been the need for a welding fixture that could hold and rotate large diameter thick-wall tubing. We were able to modify an existing tube-welding fixture and now have a device that will hold and rotate the capsule at a controlled rate under a controlled positioning TIG welding torch. The mechanical property specimens needed to characterize the AISI 1018 steel were also fabricated this quarter.

A site has been selected for the 30-ft-drop test, and the required concrete and steel pad is now being installed. It is currently planned that all testing of the AISI 1018 steel prototype outer capsules will be completed by the end of the fiscal year.

The report presenting the outer capsule design rationale and the results of the computer simulations used to evaluate the outer capsule design has been submitted to Advanced Systems and Materials Production Division for review.

Corrosion Resistance of Hastelloy S and Hastelloy C-4 (H. T. Fullam)

The long-term tests to evaluate the oxidation resistance of Hastelloy S and Hastelloy C-4 at 600 to 800°C are continuing. Over the temperature range studied, both alloys form adherent oxide layers. The results of tests with both alloys lasting up to 2500 hr show that increases in specimen weight due to oxidation follow the parabolic rate law (see Figures 6 and 7).

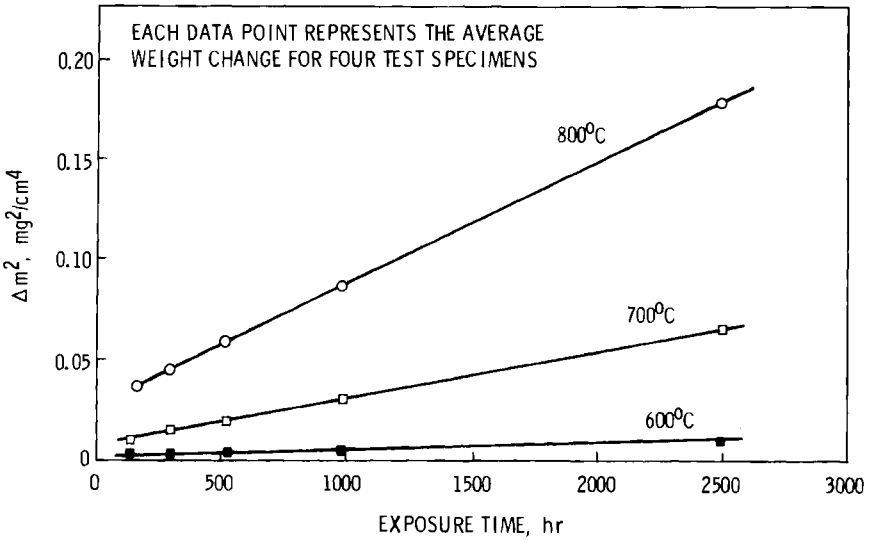

FIGURE 6. Weight Gain of Hastelloy S Specimens Exposed to Air in a Muffle Furnace at 600 to 800°C

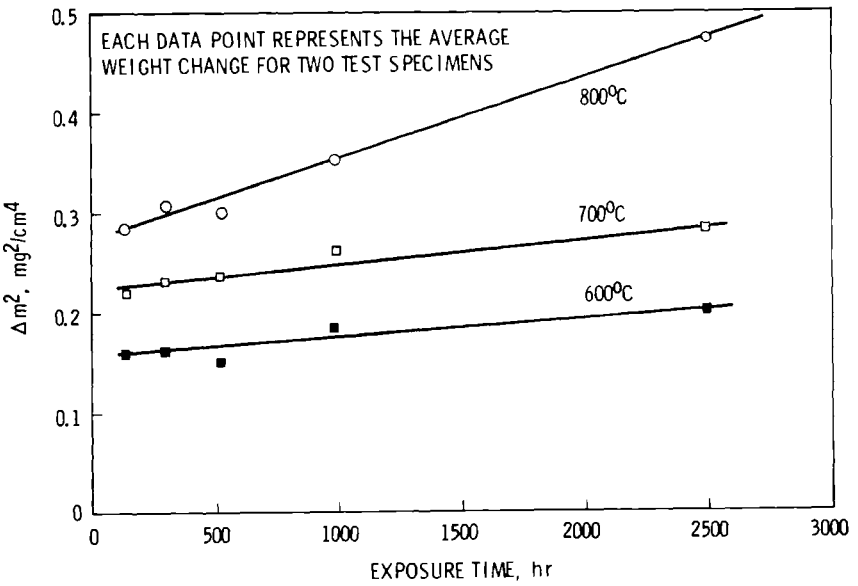

FIGURE 7. Weight Gain of Hastelloy C-4 Specimens Exposed to Air in a Muffle Furnace at 600 to 800°C

However, metallographic examination of the various 2500-hr test specimens shows that, while the oxide layers formed are relatively uniform across the specimens' surfaces, intergranular attack, pitting, and subsurface void formation increases sharply with increased exposure temperature (Figures 8 and 9). At 800°C each alloy exhibited oxidation damage to a depth that was several times greater than the thickness of the oxide layer. The grain boundary attack and void formation at 800°C was much greater in the 2500-hr specimens than in the 1000-hr specimens. Tensile specimens of each alloy, which were oxidized for 1000 or 2500 hr at each temperature, are now being evaluated to determine the effects of oxidation on the tensile properties of the alloys.

Long-term tests are underway to measure the resistance of Hastelloy S and Hastelloy C-4 to corrosion by seawater. The tests are being carried out in flowing natural seawater at ambient temperature. Tensile and prestressed specimens are being tested in addition to the standard corrosion coupons. Initial results of the tests, up to 1000-hr exposure, indicate very little attack of the two alloys. The maximum weight loss observed with the Hastelloy S specimens was only 0.007 mg/cm^2, while the Hastelloy C-4 specimens suffered a maximum weight loss of 0.06 mg/cm^2. Metallographic examination of the Hastelloy S specimens provided no visual evidence of attack, while the Hastelloy C-4 specimens exhibited very slight attack (see Figure 10).

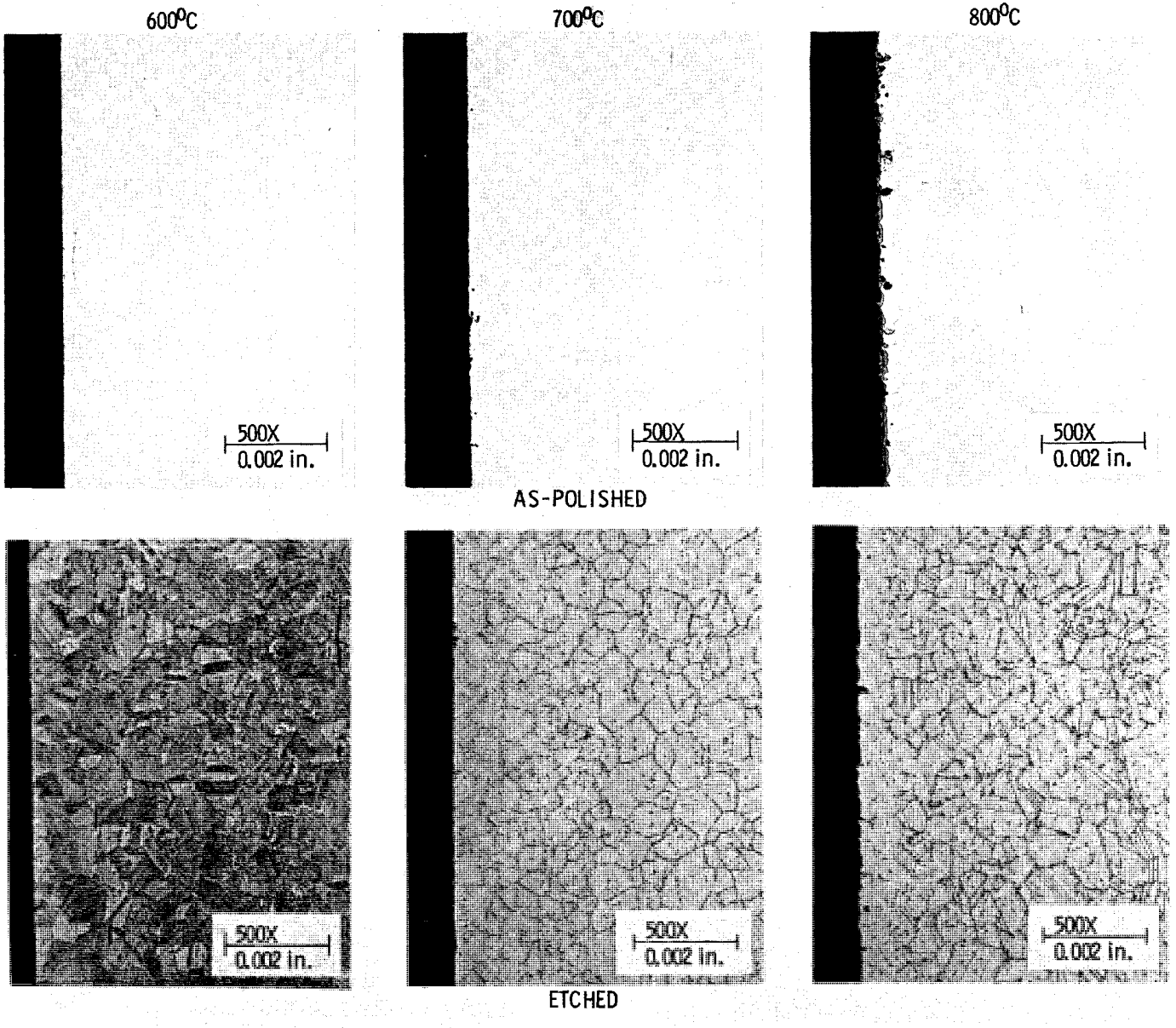

FIGURE 8. Hastelloy S Specimens Heated in Air for 2500 hr

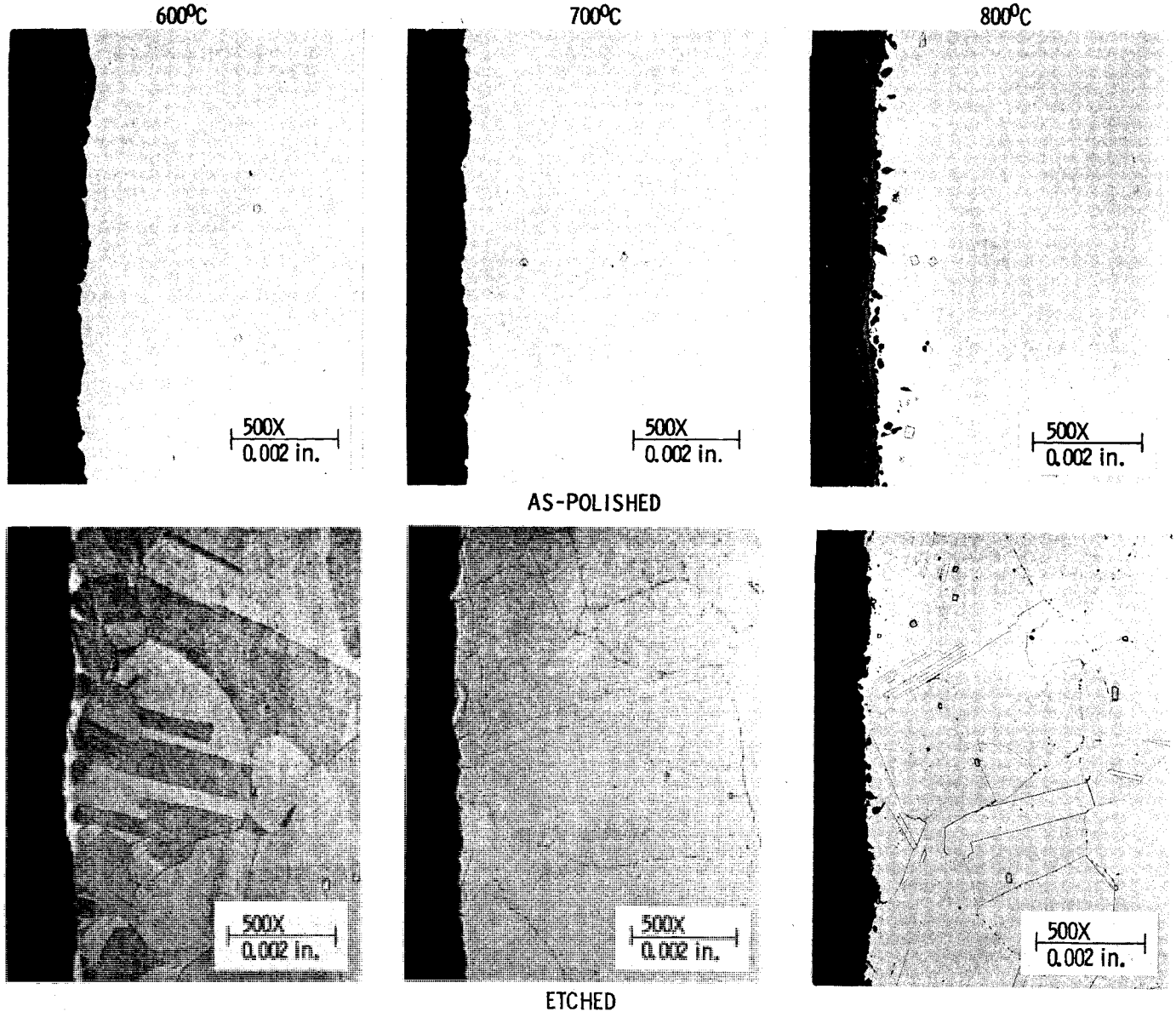

FIGURE 9. Hastelloy C-4 Specimens Heated in Air for 2500 hr

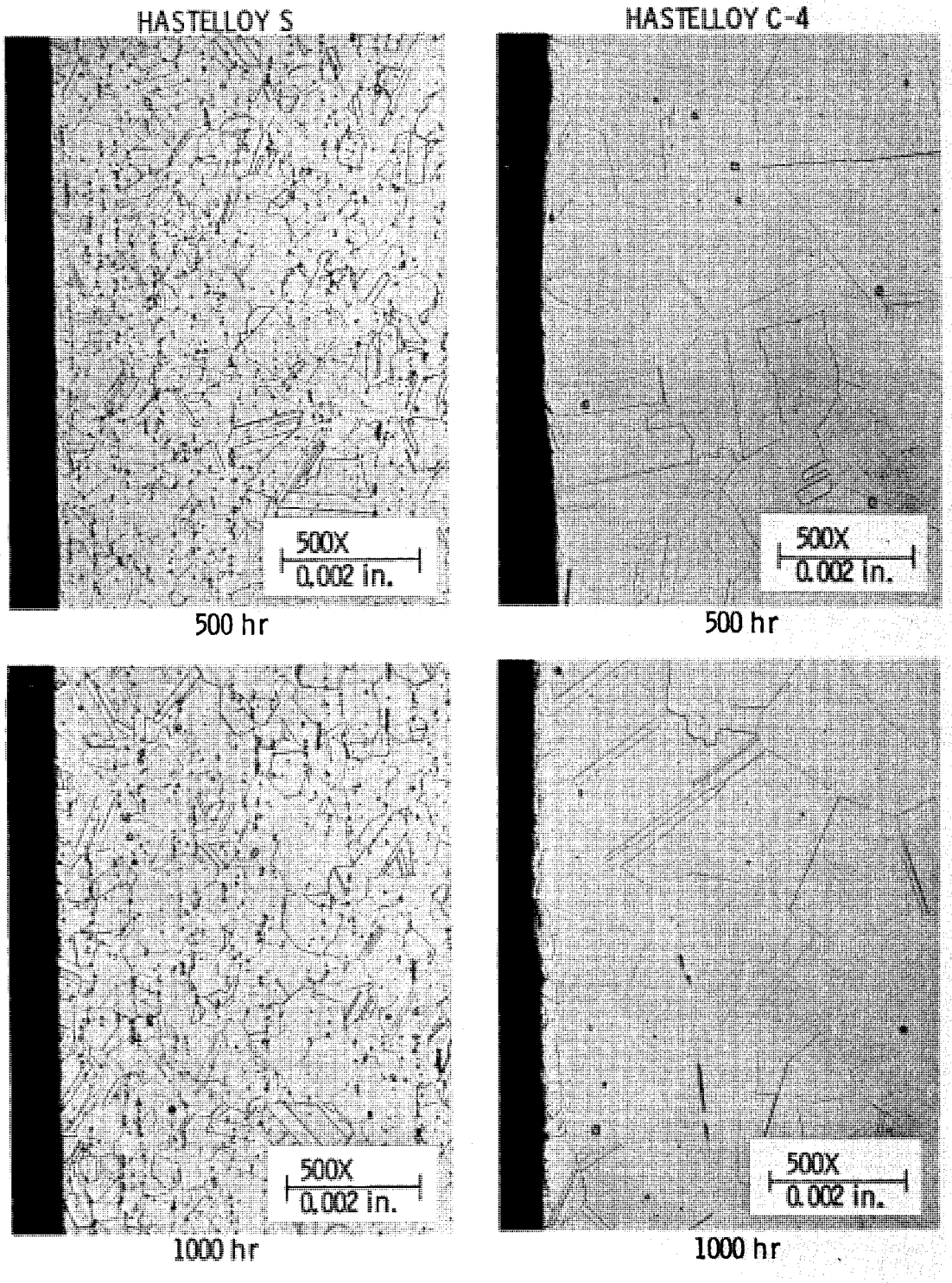

FIGURE 10. Hastelloy S and Hastelloy C-4 Test Coupons Exposed to Flowing Natural Seawater at Ambient Temperatures

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