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IRRADIATION STABILITY
OF
SCB-1 COATED HASTELLOY N
SNAP 8 FUEL CLADDING
EXPERIMENT NAA 77-1
(Title Unclassified)

AEC Research and Development Report

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IRRADIATION STABILITY
OF
SCB-1 COATED HASTELLOY N
SNAP 8 FUEL CLADDING
EXPERIMENT NAA 77-1
(Title Unclassified)

By

C. J. ENGBERG
K. J. MILLER
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- M. E. Nathan Initial concept and computer
codes for data analysis
- A. C. Crawford Capsule design, assembly, and
ETR installation
- B. J. Thomas Capsule irradiation and in-pile
permeation measurements

Without the assistance of these people, this irradiation program would not have succeeded.

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ABSTRACT

NAA 77-1 was the first of a series of irradiation experiments designed to determine the radiation stability of SCB-1 hydrogen barrier material. The capsule contained a single SCB-1 coated cladding assembly, and had the design objectives of:

- 1) Determining the feasibility of performing an in-pile hydrogen permeation experiment
- 2) Evaluating the neutron irradiation behavior of the hydrogen barrier material.

NAA 77-1 was irradiated in an Engineering Test Reactor (ETR) facility (N-14 NW) from August 1963 to June 1964, during which time it received a fast neutron dosage of 1.54×10^{21} nvt ($E_n > 1$ Mev). The capsule operated for 4343 hr at a temperature between 1200 and 1500°F, and a hydrogen pressure of 3 atm abs. The hydrogen permeation rate at 1500°F averaged 0.37 cc/hr at 3 atm hydrogen pressure, and 0.17 cc/hr at 1 atm hydrogen pressure; these rates remained essentially constant throughout the test. These permeation rates were obtained by use of a mathematical partition function, to separate the hydrogen flow coming from a hot spot in the hydrogen supply line from the flow from the test membrane.

The 3 atm pressure across the membrane resulted in a hoop stress of ~1200 psi. Measurements showed that the diameter of the Hastelloy N cladding tube had increased 0.5% during the irradiation. An additional 0.1% increase, making a total of 0.6% diameter increase, was measured at the locations of thermocouple strapping bands. The higher temperature under the thermocouple bands is believed to have caused this higher creep deformation, even though the temperature increase due to the bands was estimated at only 8°F. The average creep rate for NAA 77-1 was 10^{-6} in./in.-hr, which is higher than predicted from extrapolated rates from higher stress short-time tests. Long-time low stress tests performed on nonirradiated samples also show a similar accelerated creep rate.

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Examination of the ceramic coating revealed good to excellent adherence to the Hastelloy N base material. The SCB coating was in excellent condition after irradiation, even though some unusual bubbles or blisters were observed. The Hastelloy N cladding material experienced a small grain growth during the irradiation, as did nonirradiated samples subjected to the same thermal treatment. Grains with diameters of 0.005 in. or greater covered a major fraction of the membrane cross-sectional area. The Hastelloy N grain boundaries became decorated with precipitated particles, in both irradiated and unirradiated samples, during long-time annealing.

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I. INTRODUCTION

The NAA 77 irradiation experiment was conceived as the second phase of a study to evaluate the effect of reactor irradiation on the hydrogen barrier materials used in the SNAP reactors. The first phase, Irradiation Experiment NAA 68, undertook to determine the irradiation stability of hydrogen barrier material AI 8763D, by comparing pre- and postirradiation hydrogen permeation rates.⁽¹⁾ The second phase of the study determined the irradiation stability of an advanced hydrogen barrier material, SCB-1, by measuring the permeation rate during reactor operation. The advantages of the in-pile permeation study are:

- 1) The effect of the ionizing radiation field during the permeation process can be determined
- 2) The rate of radiation damage to the hydrogen barrier material, if any, can be measured.

The irradiation study included four separate capsule irradiations. This report covers only the first irradiation, Experiment NAA 77-1.

The objectives of Experiment NAA 77-1 were to determine the feasibility of performing an in-pile hydrogen permeation experiment and to evaluate the neutron irradiation behavior of the hydrogen barrier material. Experiments NAA 77-2, -3, and -4 had the design objectives of determining the combined effects of fission fragment and neutron radiation damage on the hydrogen barrier materials at temperatures of 1300, 1500, and 1700°F, respectively. The NAA 77-4 experiment failed prematurely; the postirradiation examination⁽²⁾ disclosed that radiation-induced diffusion caused the thermal expansion joint to freeze, resulting in rupturing of the membrane during a reactor scram. A report will soon be published on Experiments NAA 77-2 and -3, when the post-irradiation examinations have been completed.

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Figure 1. NAA 77-1 Irradiation Experiment Capsule Design
(Figure Unclassified)

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II. DESCRIPTION OF THE EXPERIMENT

The design of the NAA 77 irradiation experiments was based upon Darcy's law for gas flow through membranes. Darcy's law, stated mathematically, is:

$$F = \phi P \frac{A}{d} \quad , \quad \dots (1)$$

where;

F = total gas flow [cc(STP)/hr]

ϕ = permeation constant [cc(STP)-cm/hr-cm²-atm]

P = pressure differential across membrane (atm)

d = thickness of membrane (cm)

A = conducting area (cm²)

In order to obtain the permeation constant, we must measure the gas flow, the pressure across the membrane, the gas conducting area of the membrane, and the membrane thickness. In this experiment, the gas flow was measured by a mass spectrometer, the pressure by a strain gage transducer, the conducting area was determined from the membrane dimensions and temperature profile, and the thickness was measured during fabrication of the membrane. The use of a mass spectrometer to measure hydrogen flows requires the membrane to be in a vacuum chamber.

The NAA 77 capsule design comprised a series of concentric tubes. The membrane consisted of a standard SNAP 8 cladding assembly, surrounded by an intermediate shell which served as the vacuum chamber and gamma heater, and a Type 304 stainless steel outer capsule cladding tube, as shown in Figure 1. The vacuum chamber between the membrane and gamma heater was connected to a high vacuum pumping station and two mass spectrometers. Two hydrogen mass spectrometers were employed, to assure that one of these delicate instruments was operating properly at all times. The spectrometers were calibrated, using glass capillary tubes with known hydrogen flow rates. The instrumentation and piping diagram, Figure 2, shows the piping and valves necessary for three simultaneous irradiations. Figure 3 is an artist's conception of the Engineering Test Reactor, showing the relative locations of the capsule and the associated

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Figure 2. Instrumentation Flow Diagram (Figure Unclassified)

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Figure 3. Engineering Test Reactor, Showing Installed NAA 77 Capsule and Instrumentation
(Figure Unclassified)

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Figure 4. Control Console, NAA 77-1 Experiment
(Figure Unclassified)

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pipng and equipment necessary for the experiment. Five stainless steel sheathed Chromel-Alumel thermocouples were strapped to the membrane to measure the membrane temperature during the irradiation. The annulus between the gamma heater and the outer capsule cladding was filled with a variable gas mixture of helium and nitrogen. Changes in this gas composition could be introduced to the heat transfer characteristics of the capsule. A helium-rich mixture would result in high heat transfer, while a nitrogen-rich mixture would produce low heat transfer properties. The heat generation rate in the capsule was proportional to the gamma flux being attenuated by the capsule materials. Thus, to maintain a prescribed membrane temperature, the gas composition was adjusted to balance the heat generation rate and the heat transfer properties. A helium-rich mixture was used during periods when high heating rates were available, while a nitrogen-rich mixture was used during periods of low heating rates. Figure 4 shows the temperature control console for NAA 77-1 before the second mass spectrometer was installed.

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TABLE 1

SCB-1 COATING DATA FOR THE NAA 77-1 CAPSULE MEMBRANE

Coating Layer	Slip Number	Coating Weight (Dry)(g)	Firing Temperature (°F)	Firing Time (min)
1	SCB-1-25H	1.1	2100 ± 25	15
2	SCB-1-25G (Sm ₂ O ₃)	0.77	2100 ± 25	15
3	SCB-1-25G (Sm ₂ O ₃)	0.91	2100 ± 25	15

TABLE 2

COMPOSITION OF
SCB-1-25 FRIT

Oxide	wt %
SiO	46.7
BaO	32.7
TiO ₂	13.1
Al ₂ O ₃	7.5

TABLE 3

COMPOSITION OF COATING
SLIPS NUMBER SCB-1-25G
AND -25H

	Parts
SCB-1-25 Frit	100
Black Label Clay*	3
Bentonite	1/4
Cr ₂ O ₃	1
Distilled Water	75
Sm ₂ O ₃ [†]	4

*Ferro Corporation Trademark

[†]Sm₂O₃ was only included on the outer two layers of coating

TABLE 4

PRE-IRRADIATION PERMEATION TEST DATA
FOR THE NAA 77-1 MEMBRANE

Total Time (hr)	Temperature (°F)	H ₂ Pressure (atm)	Total H ₂ Flow (cc/hr)
41.1	1500	1	0.26
5.0	1500	1	0.25
5.4	1500	1	0.24
23.0	1500	1	0.15

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III. CAPSULE FABRICATION AND IRRADIATION

A. MEMBRANE FABRICATION

The membrane used in NAA 77-1 was a standard production SNAP 8 cladding tube, No. E-807. The cladding assembly was chromized twice, in order to achieve the chromized layer thickness specified for production material. The tube was part of Hastelloy N Heat No. Y-8437, which had the following chemical analysis:

	<u>wt %</u>		<u>wt %</u>
Manganese	0.28	Nickel	71.62
Silicon	0.21	Molybdenum	16.04
Iron	4.49	Hydrogen	0.0017
Tantalum	0.024	Oxygen	0.0094
Chromium	7.33	Nitrogen	0.0028
Carbon	0.04	Boron	0.004

The membrane had three coats of SCB-1, with the second and third coats containing about 4% Sm_2O_3 , as noted in Table 1. The chemical composition of SCB-1-25H frit is noted in Table 2, and the composition of SCB-1-25G slip is given in Table 3. The outside surface was coated with Turco Pretreat to prevent oxidation during the 2100°F air firing of the coatings. Each coat of SCB-1 was dried at 400°F for 1-1/2 hr, prior to firing. The membrane was fired in the vertical position to minimize bowing. Pitting of the SCB-1 coating was evident throughout the length of the membrane after the application of the second layer. The coating covering the 4 in. near the blind end was very rough and blistered prior to the third coat. After the third coat, these areas were visible as brown spots, and the rough spots were eliminated. Pitting was also reduced after the third coating.

The pre-irradiation permeation test results are given in Table 4. The conducting area of the membrane during permeation testing at 1500°F was 130 cm^2 , which resulted in a permeation rate of $1.11 \times 10^{-3} \text{ cc(STP)/hr-cm}^2$.

The capsule was inserted into the core of the ETR on August 5, 1963. During the insertion, the capsule was accidentally dropped, about 4 ft, to the concrete floor. No apparent damage to the capsule was noted; and, since no method was

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Figure 5. NAA 77-1 Outer Capsule Thermal Flux Dosimeter Analysis (Figure Unclassified)

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Figure 6. NAA 77-1 Outer Capsule Fast Neutron Flux Dosimeter Analysis (Figure Unclassified)

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available to determine whether the glass hydrogen barrier had been damaged, other than complete disassembly of the capsule, the experiment was continued on the assumption that no damage had occurred as a result of the fall.

B. IRRADIATION HISTORY

NAA 77-1 was irradiated in ETR position N-14 NW from August 1963 to June 1964, for a total time at temperature of 4343 hr. While the capsule was being designed, the peak fast neutron flux of 1.65×10^{14} n/cm²-sec ($E_n > 1$ Mev) was measured in the ETR N-14 NW facility. This fast neutron flux was used to predict the expected neutron dose for the irradiation of 2.58×10^{21} nvt ($E_n > 1$ Mev).

Two sets of dosimeters were placed on the capsule to measure the actual neutron dosage received by the capsule. One set consisted of five dosimeter wires extending along the length of the capsule on the outside of the cladding. These wires were removed from the capsule after it was withdrawn from the reactor, and were retained by Reactor Operations for analysis. The five wires included one titanium, one 0.1% Co-Al, two chrome-plated iron, and one nickel wire. Only the titanium and the Co-Al wires were analyzed, because of problems encountered with the iron and nickel wires due to corrosion by the ETR coolant water. The titanium and Co-Al wires were analyzed at 1-in. intervals along their lengths, with the results of this analysis shown in Figures 5 and 6. The average fast neutron flux received at the outside of the capsule (based on the titanium wire analysis) was 0.76×10^{14} n/cm²-sec, for a total fast neutron dosage of 1.19×10^{21} nvt ($E_n > 1$ Mev).

The remaining dosimeters were located at the top, center, and bottom of the membrane. These dosimeters were removed from the membrane, placed in sample bottles, and sent to Edgerton, Germeshausen, and Grier, Inc., for analysis. The dosimeters consisted of small nickel, cobalt, and iron foils encapsulated in stainless steel tubes. These foils were analyzed after irradiation for Co⁶⁰ and Mn⁵⁴ from the cobalt and iron foils, respectively. The nickel foils were not analyzed. The results are reported in Table 5.

The $\pm$ values on the dosimeter counts signify counting statistics only, and are standard errors. The overall accuracy of the Mn⁵⁴ measurements are $\pm 20\%$, and the Co⁶⁰ measurements are $\pm 10\%$.

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TABLE 5
E.G. & G. DOSIMETER ANALYSIS

Dosimeter Number	Position on Membrane	Isotope	Dosimeter Count (dpm/g of Foil)
13	Top	Co ⁶⁰	$1.53 \pm 0.02 \times 10^{15}$
		Mn ⁵⁴	Not recovered
14	Center	Co ⁶⁰	$1.64 \pm 0.02 \times 10^{14}$
		Mn ⁵⁴	$2.87 \pm 0.01 \times 10^{10}$
15	Bottom	Co ⁶⁰	$8.30 \pm 0.10 \times 10^{14*}$
		Mn ⁵⁴	$9.70 \pm 0.01 \times 10^{10}$

*Accuracy of this analysis is $\pm 50\%$, due to a damaged foil

From the data reported in Table 5, an average neutron flux of 0.23×10^{14} n/cm²-sec was calculated, for a total fast neutron dosage of 3.60×10^{20} nvt ($E_n > 1$ Mev). This neutron dosage does not agree with the calculated dosage of 2.56×10^{21} nvt or with the analysis reported by the reactor operators of 1.19×10^{21} nvt. Therefore, to check these results, four samples of the membrane were selected for analysis. The results of the analyses are noted in the following list.

Position on Membrane	Total Neutron Dosage [nvt ($E_n > 1$ Mev)]
Top	1.25×10^{21}
Center	1.54×10^{21}
	1.44×10^{21}
Bottom	1.34×10^{21}

The analysis of these samples was based on a Hastelloy N iron content of 4.49% and 4343 hr of exposure. No corrections were made for intermittent reactor operation. These results agree with the analyses performed at the reactor on the titanium wire. Because of agreement between the dosimeter analyses of Hastelloy N samples and those for the external dosimeter wires, it was concluded that the membrane received a peak dosage of 1.54×10^{21} nvt ($E_n > 1$ Mev), rather than the design estimated dosage of 2.56×10^{21} nvt ($E_n > 1$ Mev).

C. IN-PILE HYDROGEN PERMEATION MEASUREMENT

The performance of the NAA 77-1 capsule demonstrated the feasibility of making reliable in-pile permeation tests. The membrane temperature, as determined from the five thermocouples, is given in Figure 7. The effective conducting area for permeation at 1500°F was calculated, from the membrane temperature profile, to be 95 cm².

The initial measured flow rate of 3 cc(STP)/hr for the capsule was found to be approximately an order of magnitude higher than expected, as illustrated by comparing the pre-irradiation data listed in Table 4 with the initial in-pile data shown in Figure 8. It was found that these high permeation rates were caused by overheating of the hydrogen supply line, which was inside the vacuum line for about 3 in. just above the capsule. During the design heat transfer calculation, it was assumed that the gamma flux was sufficiently low in this section of the capsule that the temperature of the hydrogen supply line would not be higher than 700°F. This assumption proved to be incorrect. Figure 9 shows the temperature distribution of the hydrogen supply line as a function of gamma flux. Calculations showed that a 1 w/g increase in gamma heating rate would cause a 300°F rise in peak temperature of the hydrogen supply line, which in turn would cause the hydrogen flow from the tube to increase from 0.6 to 3.8 cc/hr. Figure 8 shows that the hydrogen flow from the capsule decreased to about 1 cc/hr at 1500°F and 1 atm H₂ pressure during the first 700 hr of the irradiation. This decrease in hydrogen flow was probably caused by the supply line becoming oxidized. After this initial period of hydrogen flow rate decrease, the hydrogen flow rates remained constant for the remainder of the irradiation.

Various operational methods were considered, in order to separate the membrane flow from the total hydrogen flow from the capsule. The method which proved successful was based on the assumption that the hydrogen permeation through the supply line was constant at constant pressure, while the permeation through the membrane changed with membrane temperature. This assumption is justified, since the membrane temperature is adjusted by changing the He/N₂ ratio in the gas annulus, which does not affect the supply line temperature. Therefore:

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Figure 7. Longitudinal Temperature Distribution for NAA 77-1 Membrane During Irradiation (Figure Unclassified)

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Figure 8. Initial Hydrogen Flow Rates From NAA 77-1 Capsule

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Figure 9. Temperature Distribution of Hydrogen Supply Line as a Function of Gamma Flux (Figure Unclassified)

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$$F_c(t) = F_L + F_m(t) \quad , \quad \dots(2)$$

where:

$F_c(t)$ = total hydrogen flow from the capsule as a function of temperature

F_L = hydrogen flow from the supply line

$F_m(t)$ = hydrogen flow from the membrane as a function of temperature.

Thus, at two different temperatures, $T_1 > T_2$:

$$\Delta F_c(T_1 - T_2) = \Delta F_m(T_1 - T_2) \quad . \quad \dots(3)$$

In order to gather data for use in Equation 2, during the second ETR cycle (after 700 hr irradiation), the total hydrogen flow from the capsule was measured each day in accordance with the sequence shown in Table 6.

TABLE 6
DAILY PERMEATION RATE MEASURING SEQUENCE

Measurement Step	Membrane Temperature (°F)	H ₂ Pressure (atm abs)
1	1500	3
2	1200	3
3	1200	2
4	1500	2
5	1500	1
6	1200	1
7	1500	3

The rate of hydrogen flow through the membrane at a given hydrogen pressure was calculated from the data obtained in this operational sequence by subtracting the flow rate measured at a membrane temperature of 1200°F from that measured at 1500°F (Equation 2). For this calculation, since the flow rate through the membrane at a temperature of 1500°F is a factor of eight higher than the rate at 1200°F, then as a first approximation, $\Delta F_m(1500^\circ\text{F} - 1200^\circ\text{F}) \cong F_m(1500^\circ\text{F})$.

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Figure 10. Hydrogen Flow Rates and Neutron Dosages as a Function of Irradiation Time for NAA 77-1 Capsule

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Figure 11. Flow Rates for Irradiated and Unirradiated SCB-1 Coated Membranes at 1500°F and 1 atm H₂ Pressure

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The calculated hydrogen flow rates as a function of time for the membrane with an internal hydrogen pressure of 3 atm are summarized in Figure 10. For convenience, the accumulated neutron dosages are also shown.

The calculated permeation rate data for the capsule are compared with the averaged permeation rate data for 22 unirradiated SCB-1 coated tubes, both crystallized and uncrystallized, at conditions of 1500°F and 1 atm hydrogen pressure, in Figure 11. The similarity between the two curves indicates that the mechanism of hydrogen permeation, and therefore the rate of permeation, is not grossly affected by the presence of the intense ionizing radiation and fast neutron flux in a nuclear reactor core.

The SCB-1 hydrogen barrier material was designed to be a "nucleated" glass that could be used in either the vitreous or devitrified condition. A "nucleated" glass is defined as one that contains one or more constituents that tend to promote devitrification under appropriate heat treatment conditions. Work on out-of-pile tests in another part of the SNAP 8 program⁽³⁾ indicated that devitrification of the SCB-1 coating results in an increase in the average hydrogen permeation rate from 1.6×10^{-3} cc (STP)/hr-cm² for vitreous coatings, to an average rate of 2.5×10^{-3} cc (STP)/hr-cm² for devitrified coatings. The membrane coating in the NAA 77-1 experiment was initially in the vitreous condition, and it was not heat treated to promote devitrification.

IV. POSTIRRADIATION EXAMINATION

A. CAPSULE DISASSEMBLY

The appearance of the capsule after irradiation was good, and is illustrated in Figure 12. The capsule was disassembled with the intention of permeation testing the membrane in-cell. Thus, the outer cladding of the capsule was first removed to expose the gamma heater, and the cladding at the top of the capsule was removed to expose the vacuum and hydrogen supply lines. During this operation, the hydrogen supply line was found to be broken in two places. The location of the fractures were 1/2-in. above the top chill block and 1/2-in. below the top bulkhead which formed a vacuum seal. The fractured end of the hydrogen supply line is shown in Figure 13. A brown corrosion product, presumably due to the ETR process water, was observed part way around the circumference of the fracture, indicating that the initial fracture had existed for some time. It is believed that the fracture occurred during the knifing operation at the ETR when the capsule was severed from its lead tube. The occurrence of the fracture indicates brittle behavior of the stainless steel, and it is believed that further strains may have been imposed on the hydrogen supply line during remote handling in the hot cell. The fracture of the hydrogen supply line made postirradiation permeation testing of the membrane impracticable, and this part of the experiment was therefore abandoned.

The membrane was removed from the gamma heater without difficulty, and is shown in Figure 14. No evidence was found of any reaction between the gamma heater and the membrane in the thermal expansion joint at the bottom of the membrane. Upon removal of the thermocouple bands, marks were found on the membrane at each thermocouple band position. These marks, seen in Figure 14b, are associated with the difference in oxidation of the membrane under the bands and in the exposed areas during the irradiation. Visual examination of the membrane surface revealed some localized pitting, as shown in Figure 15. This pitting is believed to have occurred during the three 15-min air firing cycles of the SCB-1 coatings at 2100°F. In more recent work, reduction of this firing cycle to a total of 5 min for each coating layer has been shown to eliminate this pitting.

Figure 12. NAA 77-1 Capsule After 4343 hr Irradiation in the ETR
(Figure Unclassified)

771-1

Figure 13. Fracture Surface
of Hydrogen Supply Line
(Figure Unclassified)

P771-4

P771-5, -6, -7, -8, -9

a. Before Thermocouple Bands and Dosimeters Were Removed

Figure 14. NAA 77-1
Membrane After
Removal From
Gamma Heater

P771-12, -13, -14, -15, -16

b. After Thermocouple Bands and Dosimeters Were Removed

771-10

Figure 15. Pitting of External
Surface of Hastelloy N
Membrane

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B. CREEP MEASUREMENTS

The diameter of the membrane was measured at 0, 60, and 120° angular orientations, using a profilometer. This device consists of a modified lathe bed with two end stocks for support of the object, and two highly sensitive linear displacement transducers which are translated along the object to be measured. The transducers provide signals to a dual-pen recorder, which prints the continuous output from the transducers. From the recorder data, the dimensions of the capsule membrane were determined at 1-/2-in. intervals along the length of the membrane. Table 7 shows that the diameter of the membrane increased by approximately 0.002 in. during the irradiation. In addition, it was found that distortion of the membrane had increased from a TIR of 0.008 to 0.030 in. during the irradiation. Examination of the profilometer data revealed local swelling that coincided with the positions of the thermocouple bands. The diameter data for these thermocouple band locations are listed in Table 8. Although only four of the values tabulated in Table 8 fall outside the 90% significance level, meaning that these values are statistically significantly higher than the average post-irradiation diameter, one can best judge the swelling characteristics of the membrane by comparing the local diameter to the diameter on both sides of the bulges and adjacent to them. Figure 16 shows the average swelling as a function of position along the membrane. It was found that the maximum swelling occurred in the lower half of the membrane, where the temperature was highest during irradiation. These areas appear to be due to creep effects in the membrane during exposure to internal hydrogen pressure and elevated temperature for a period of 4343 hr. The temperature of the membrane underneath the thermocouple bands was apparently increased sufficiently by the presence of the bands to cause an increase in the total creep at these locations, even though the temperature increment was calculated to be $< 8^{\circ}\text{F}$.

Figure 17 compares the short- and long-time creep rates for unirradiated samples with the average creep rate for the irradiated NAA 77-1 membrane. E808, E822, and the NAA 77-1 membranes were sister membranes. The E808 and E822 membranes were placed on permeation endurance test, which was continued for 14,000 hr (5000 hr with H_2 internal pressurization, and 9000 hr with air internal pressurization). The average creep rates for the low-stressed membranes were higher than would have been predicted from extrapolation of

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TABLE 7
POSTIRRADIATION MEASUREMENTS OF
MEMBRANE DIAMETER

Distance from Bottom of Membrane (in.)	Membrane Diameter (in.)		
	Scan Orientation		
	0°	60°	120°
0.5	0.5610	0.5618	0.5618
1.0	0.5626	0.5626	0.5618
1.5	0.5633	0.5641	0.5618
2.0	0.5637	0.5637	0.5626
2.5	0.5637	0.5637	0.5633
3.0	0.5641	0.5637	0.5637
3.5	0.5637	0.5633	0.5637
4.0	0.5637	0.5637	0.5641
4.5	0.5641	0.5641	0.5638
5.0	0.5641	0.5641	0.5640
5.5	0.5633	0.5641	0.5641
6.0	0.5633	0.5641	0.5641
6.5	0.5626	0.5641	0.5641
7.0	0.5629	0.5641	0.5641
7.5	0.5633	0.5637	0.5633
8.0	0.5629	0.5633	0.5633
8.5	0.5633	0.5637	0.5633
9.0	0.5633	0.5633	0.5629
9.5	0.5633	0.5637	0.5633
10.0	0.5629	0.5633	0.5633
10.5	0.5629	0.5633	0.5629
11.0	0.5626	0.5633	0.5626
11.5	0.5626	0.5633	0.5622
12.0	0.5629	0.5637	0.5618
12.5	0.5629	0.5641	0.5614
13.0	0.5633	0.5633	0.5618
13.5	0.5633	0.5626	0.5626
14.0	0.5633	0.5618	0.5629
14.5	0.5626	0.5618	0.5631
Postirradiation average	0.5632	0.5634	0.5630
	±0.0006	±0.0007	±0.0008
Pre-irradiation average	0.5613	0.5623	0.5612
	±0.0009	±0.0012	±0.0011
Diameter increase	0.0019	0.0011	0.0018

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TABLE 8
DIAMETER MEASUREMENTS OF MEMBRANE
AT THERMOCOUPLE BANDS

Thermocouple Band, Numbered From Bottom End Of Membrane	Distance From Bottom (in.)	Membrane Diameter (in.)		
		Scan Orientation		
		0°	60°	120°
1	1.50	0.5645*	0.5633	0.5618
2	1.75	0.5641	0.5641	0.5618
3	3.00	0.5641	0.5645	0.5641
4	4.50	0.5645*	0.5643	0.5643
5	6.00	0.5641	0.5648*	0.5645*
6	7.50	0.5637	0.5642	0.5642
7	9.00	0.5633	0.5637	0.5634
8	10.0	0.5633	0.5637	0.5634
9	10.75	0.5633	0.5641	0.5633
10	12.50	0.5629	0.5637	0.5620
11	13.25	0.5633	0.5637	0.5616
12	14.00	0.5637	0.5633	0.5629
13	14.75	0.5618	0.5626	0.5631
Average		0.5636	0.5638	0.5631
Table 7 Average		0.5632	0.5634	0.5630

*Values fall outside the 90% significance level based on the postirradiation average diameter reported in Table 7.

TABLE 9
COMPARISON OF CREEP RATES OF IRRADIATED AND
UNIRRADIATED SNAP 8 MEMBRANES

	Experimental Creep Rate (%/hr)	Predicted* Creep Rate (%/hr)
NAA 77-1	1×10^{-4}	5×10^{-6}
E-808 and E-822 unirradiated tubes	3.3×10^{-5}	5.0×10^{-8}

*Predicted by extrapolation of data from References 4 and 5

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Figure 16. Location and Height of Bulging Observed on Surface of NAA 77-1 Membrane (Figure Unclassified)

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Figure 17. Low Stress Creep Studies of Irradiated and Unirradiated SNAP 8 Cladding Assemblies (Figure Unclassified)

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high-stress data. Table 9 summarizes the average experimental and extrapolated creep rate data for Membranes E808 and E822 and for the NAA 77-1 membrane, and shows the extent that the creep rates increased from the extrapolated values.

C. DESTRUCTIVE EXAMINATION OF NAA 77-1 CAPSULE

Following the visual and dimensional inspection, the membrane from the capsule was destructively analyzed to evaluate the effect of high temperature irradiation on (1) the microstructure and ductility of the Hastelloy N membrane, and (2) the microstructure, appearance, and adherence of the SCB-1 coating. Five metallographic samples and three adherence samples were cut from the membrane, as shown in Figure 18.

For the evaluation of the adherence of the SCB-1 coating to the Hastelloy N substrate, a qualitative test was performed, where each sample was longitudinally slit along one side and the sample was then spread open and bent back and forth with the manipulators to test the flaking and spalling behavior of the ceramic. The appearance of three of the samples after the bend tests is shown in Figure 19. The sample from the top of the membrane, which was irradiated at approximately 700°F, exhibited poor adherence, while the remaining two samples, which were irradiated at 1400 to 1500°F, exhibited good to excellent adherence; the sample with excellent adherence exhibited no chipping of the ceramic after 30 bends.

The general condition of the ceramic appeared to be excellent, as shown in Figure 20, although the color of the ceramic changed from green to bluish grey during the irradiation. Some abnormalities were found in the coating after the irradiation, and included spalling, bubble growth, and blistering. Some of these are shown in Figure 21. An estimate of the effect of the defect shown in Figure 21b on the hydrogen permeation rate was made, assuming that the blister was formed during fabrication of the membrane and that base metal was exposed as a result of the blister. Under these conditions, such a defect could account for an increase in total hydrogen flow of about 0.1 cc/hr at 1500°F and 3 atm H₂ pressure, as compared with a defect-free coating. The series of blisters shown in Figure 21c could result in an increase in the hydrogen flow rate of about 0.05 cc/hr under the same conditions. However, since the metal surface was oxidized at those defects, as illustrated by a comparison of Figures 21a and 21b, the permeation rate through either defect was probably <0.01 cc/hr. Changes

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Figure 18. Location of Metallography and Adherence Test Samples From NAA 77-1 Membrane (Figure Unclassified)

Figure 19. Adherence Test Samples After Testing

Figure 20. Ceramic Coating on Inside of NAA 77-1 Membrane After Irradiation

P77-1-17, -18, -19

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P77-1-21

a. Lack of Oxidation on Metal Surface Exposed by Ceramic Chipping Which Occurred During Postirradiation Examination

P77-1-22

b. Oxidation of Metal Surface Caused by Ceramic Chipping (Blistering assumed to have occurred during fabrication)

P77-1-23

c. Series of Bubbles or Blisters (Assumed to have been formed during fabrication)

P77-1-25

d. Rough Ceramic Caused During Fabrication

Figure 21. Abnormal Surface Effects Found in SCB-1 Coating After Irradiation (Irradiation temperature = 1500°F, time = 4343 hr)

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a. Top (Cracks are assumed to have formed during metallographic mounting)

10-2-5

b. Center

10-3-4

c. Weld Area

10-5-5

Figure 22. Bubble Density in SCB-1 Coating After Irradiation
(Irradiation temperature = 1500°F, time = 4343 hr)

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of this magnitude in the total hydrogen flow rate could not be detected during irradiation, because of the sensitivity limitations of the instrumentation. For reference purposes, the average hydrogen flow rate from the capsule membrane at 3 atm H_2 pressure and 1500°F was 0.37 cc(STP)/hr.

Figure 22 shows the normal bubble density found in SCB-1 coated tubes, which was unchanged during the irradiation. The cracks shown in the ceramic were assumed to have formed during the metallographic mounting process.

D. METALLOGRAPHIC EXAMINATION OF HASTELLOY N

Five metallographic samples were taken from the NAA 77-1 capsule membrane, as shown in Figure 18. In addition, reference samples were taken from two unirradiated membranes, numbered E808 and E822, as illustrated in Figure 23. These membranes had been annealed for 14,000 hr at 1500°F, with the detailed temperature profile shown in Figure 23, as a part of long-time endurance tests of a cluster of membranes. The atmosphere outside these membranes during annealing was vacuum ($\sim 5 \times 10^{-4}$ mm Hg); while they were internally pressurized at 1 atm H_2 pressure for the first 5000 hr, and at 1 atm air pressure for the last 9000 hr of the annealing treatment. All samples were mounted, roughground with 240-, 320-, and 600-grit abrasive, and finally polished with 6- μ diamond dust and then with 1- μ diamond dust. The samples were chemically etched with a solution of three parts concentrated HCl, two parts H_2O , and one part 10% chromic acid. The resulting microstructures are illustrated in Figures 24, 25, and 26.

A comparison of the grain sizes in the irradiated and unirradiated membranes may be made from Figure 24. In each case, the long-time annealing temperature for the unirradiated material was approximately 700°F. Since this is below the temperature at which grain growth or precipitates occur in Hastelloy N, no significant change in the microstructure of the material was expected or observed. However, it was noted that the irradiated specimen was more easily etched.

Figure 25 shows the microstructures of samples taken from the mid-lengths of both irradiated and unirradiated membranes. For the NAA 77-1 membrane, this location also corresponded to the region of maximum creep strain. It may be observed, in Figures 24 and 25, that the grain structure is the same in the areas of maximum strain as in the remainder of the membranes. However, the

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Figure 23. Location of Metallography Samples From the Out-of-Pile Permeation Endurance Test Membranes (Figure Unclassified)

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a. Irradiated for 4343 hr
(Transverse section,
largest grain size)

10-1-4

5958-6-2

1) Longitudinal Section,
Typical Grain Size

5958-5-3

2) Transverse Section,
Largest Grain Size

b. Unirradiated, Annealed for 14,000 hr

Figure 24. Comparison of Grain Size in Hastelloy N Irradiated at
700°F and in Unirradiated Hastelloy N Annealed at 700°F
(Figures Unclassified)

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10-2-2

1) Largest Grain Size

10-2-4

2) Typical Grain Size

a. Irradiated for 4343 hr

5958-3-3

1) Largest Grain Size

5958-3-2

2) Typical Grain Size

b. Unirradiated, Annealed for 14,000 hr

Figure 25. Comparison of Grain Size in Hastelloy N Irradiated at 1500°F and in Unirradiated Hastelloy N Annealed at 1500°F (Transverse sections) (Figures Unclassified)

10-5-2

- 1) Irradiated at $<500^{\circ}\text{F}$ for 4343 hr
- 2) Unirradiated, Annealed at 1500°F for 14,000 hr

5958-4-5

a. Flat Area of End Cap

10-5-3

- 1) Irradiated at $\sim 1400^{\circ}\text{F}$ for 4343 hr
- 2) Unirradiated, Annealed at 1500°F for 14,000 hr

5958-4-3

b. TIG Welded Zone

10-5-4

- 1) Irradiated at $\sim 1400^{\circ}\text{F}$ for 4343 hr
- 2) Unirradiated, Annealed at 1500°F for 14,000 hr

5958-4-2

c. Just Above TIG Weld

Figure 26. Comparison of Grain Size at Weld Area of Cladding Tube in Irradiated Hastelloy N and in Unirradiated, Annealed Hastelloy N (Longitudinal sections) (Figures Unclassified)

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irradiated samples appear over etched, and the detailed structure of the precipitates was therefore lost. The oxidation layer, shown at the outside surface of the unirradiated samples, is believed to be due to a 20-hr annealing treatment at 1900°F in air that was used to promote devitrification of the SCB-1 coating.

The microstructures of the tubing, bar stock, and weld areas for both the irradiated and unirradiated Hastelloy N cladding samples are shown in Figure 26. These samples were taken from the closed-end portion of the tubes. Since this part of the NAA 77-1 membrane was located in the expansion joint of the capsule, its temperature during irradiation was estimated to be 500°F, while the annealing temperature for the comparable unirradiated samples was 1500°F. In general, it was found that the Hastelloy N bar stock exhibited the largest grain size and the highest population density of precipitates, both within the grains and in the grain boundaries. The largest single grain found in the samples studied occurred in "as-received" bar stock, and measured approximately 0.020 in. by 0.016 in. across the two maximum diameters. Thus, it is believed to be coincidental, rather than an irradiation effect, that the grain size observed in the irradiated bar stock sample from the NAA 77-1 membrane was larger than that in the corresponding unirradiated sample. The weld areas, in general, have a typical cast structure; while the area in the tube wall adjacent to the weld has a small grain size, as shown in Figure 26. The reason for this apparent refinement in grain size in this localized area is unknown.

The observations on grain size effects in Hastelloy N membranes that were made in this and other related studies may be summarized as follows:

- 1) The grain size of "as-received" Hastelloy N tubing is usually in the range of ASTM 6 to 8 (0.0018 to 0.0009 in. average grain diameter).
- 2) The Hastelloy N tubing grain size, after the 45 min firing time at 2100°F that was used for curing of the SCB-1 coating, was approximately ASTM 3 to 5 (0.005 to 0.0025 in. average grain diameter).
- 3) The Hastelloy N tubing grain size after long-time annealing, with or without irradiation, was in the range of ASTM 2 to 4 (0.007 to 0.0035 in. average grain diameter).

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4) The largest grain size found occurred in "as-received" Hastelloy N bar stock. Further processing did not reduce or increase this grain size.

With respect to the precipitates that were found in the Hastelloy N samples, it was determined by electron microprobe analyses that the precipitates were rich in molybdenum; the relative intensities of the nickel and molybdenum emissions corresponded approximately to the β -phase composition in the nickel-molybdenum system (72 Ni - 28 Mo).

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V. CONCLUSIONS

Based on the results of the NAA 77-1 experiment and related work, the following conclusions were drawn:

- 1) The capsule operated at 1200 to 1500°F for 4343 hr with 3 atm internal hydrogen pressure in the membrane without any indication of failure.
- 2) Within the accuracy of the experiment, it was found that the hydrogen barrier material was little affected by neutron irradiation to a dosage of 1.4×10^{21} nvt ($E_n > 1$ Mev) at a temperature of 1500°F. The average permeation rate throughout the 4343-hr test was $\sim 1.8 \times 10^{-3}$ cc(STP)/hr-cm² at a permeation test temperature of 1500°F and an internal hydrogen pressure of 1 atm.
- 3) Metallographic examination of the SCB-1 hydrogen barrier material did not reveal evidence of deterioration of the coating due to neutron irradiation. The adherence of the ceramic to the Hastelloy N ranged from good to excellent in the high-temperature portion of the membrane.
- 4) The low-stress creep deformation of both irradiated and unirradiated membranes was found to be significantly greater than would be predicted from high stress test results.
- 5) The Hastelloy N grain boundaries became decorated with a precipitate, as a result of long time thermal annealing.
- 6) No evidence was found that indicated acceleration of grain growth in the Hastelloy N due to irradiation. However, the bulk of the membranes contained grains with diameters of 0.005 in. or greater after the SCB-1 coating firing cycle.

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