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OF
SNAP 8 CLADDING MATERIAL
IRRADIATION EXPERIMENT NAA 77-4
(Title Unclassified)

AEC Research and Development Report

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M-3679 (40th Ed.)

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POSTIRRADIATION EXAMINATION
OF
SNAP 8 CLADDING MATERIAL
IRRADIATION EXPERIMENT NAA 77-4
(Title Unclassified)

By:

C.J. ENGBERG
K.J. MILLER

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M. E. Nathan	Initial concept
A. C. Crawford	Initial design
A. F. Lillie	Capsule design and assembly
B. J. Thomas	Capsule irradiation
S. L. Englebert	Hot cell metallography
D. D. McAfee	Hot cell metallography
J. R. Miller	Heat transfer analysis
R. Roberts	Computer codes
G. Ervin, III	Data analysis

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

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ABSTRACT

NAA 77-4 was the fourth capsule in an irradiation series designed to measure the radiation stability of SNAP 8 hydrogen barrier materials. The capsule contained a closed-end cladding tube, coated internally with the advanced SNAP 8 hydrogen barrier material, SCB-1. Irradiation of NAA 77-4 was begun in June of 1964, and the capsule failed when the temperature was increased to 1700°F. Postirradiation examination disclosed that a tensile force had been exerted on the capsule membrane during the rapid cooling of a reactor scram, resulting in a circumferential rupture. Metallographic examination showed the fracture to be intergranular, with many intergranular voids in the area near the fracture. The cladding material, Hastelloy N, exhibited low ductility in the area of failure, since none of the grains showed evidence of plastic deformation.

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I. INTRODUCTION

Irradiation experiments NAA 77-1, -2, -3, and -4 were performed to establish the radiation stability of the SNAP 8 hydrogen permeation barrier material. NAA 77-1 was built to verify the capsule design, as well as to evaluate the neutron irradiation behavior of the hydrogen barrier material. The capsule operated, as designed, for 4343 hr at temperatures ranging from 1200 to 1520°F, receiving a peak neutron dosage of 1.54×10^{21} nvt ($E_n > 1$ Mev). NAA 77-2 and -3 were built and irradiated simultaneously to evaluate the combined effects of neutron and fission fragment damage, if any, on the hydrogen barrier material at temperatures of 1300 and 1500°F, respectively. An estimated neutron irradiation of 1.4×10^{21} nvt ($E_n > 1$ mev) and a fission fragment bombardment of 6.9×10^{16} fragments/cm² were selected as the minimum irradiation requirements for NAA 77-2 and -3. These values correspond to the peak exposure expected for a 600-kwt, 10,000-hr, SNAP 8 core.

The NAA 77-4 capsule, which is the subject of this report, was designed to operate at 1700°F to study the effects of crystallization on the performance of the hydrogen barrier material during irradiation. X-ray diffraction studies have shown that SCB-1* devitrified⁽¹⁾ during certain thermal treatments. At temperatures above 1600°F, a barium silicate crystal structure develops rapidly. Initial permeation studies of SCB-1 showed that membranes that were heat treated to induce crystallization had higher permeation rates than similar membranes that were not so heat treated. The crystallized membranes also showed increases in the permeation rates during long-time anneals. Since fused quartz also devitrifies on thermal treatment and the process is accelerated during neutron irradiation, it was logical that SCB-1 material should behave in a similar manner. Therefore, neutron and fission fragments could accelerate the devitrification process to such an extent that increases in the permeation rates would be inevitable, thereby possibly limiting the life of the SNAP 8 reactor. A test temperature of 1700°F was selected for NAA 77-4 to accelerate the thermal devitrification process, in order to obtain this information as rapidly as possible.

*SCB-1 Ceramic coating is a baria-alumina-titania silicate glass, developed at Atomics International

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Figure 1. Cutaway Drawing of NAA 77-4 Capsule, Showing Capsule Design
(Figure Unclassified)

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II. CAPSULE DESCRIPTION

The NAA 77-4 capsule contained one chromized Hastelloy N SNAP 8 cladding tube, approximately 0.560 in. in diameter by 15 in. long with a 0.010-in. wall thickness. The tube was coated on the inside with two, 1-mil thick layers of SCB-1 hydrogen barrier material. The test membrane was centered in a Hastelloy N intermediate chamber, called the gamma heater, which in turn was centered inside the capsule outer jacket, as shown in Figure 1. The capsule outer jacket was made of Type 304 stainless steel. Inside the membrane was a "fission tube" which provided a fission fragment bombardment similar to a fuel rod. This fission tube consisted of an aluminum oxide tube coated on the outside with a UO_2 -doped glass. In order for the SCB-1 barrier material to receive the same fission fragment bombardment from the fission tube as a SNAP 8 fuel rod, the glass coating contained about 4 wt % fully enriched UO_2 , and was only 0.0007 in. thick.

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Figure 2. Engineering Test Reactor, Showing Relative Locations of
NAA 77 Capsule and Equipment
(Figure Unclassified)

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III. CAPSULE IRRADIATION

The capsule was lowered into the N-14 northwest irradiation facility of the Engineering Test Reactor (ETR) during June 1964, for a planned 4000-hr irradiation. An artist's conception (Figure 2) of the ETR shows the relative locations of the capsule, associated piping, and equipment necessary for the experiment. The NAA 77-4 capsule membrane operated only 100 hr. Failure of the membrane occurred when the capsule temperature was increased from 1500 to 1700°F. The operating plan called for the capsule to operate for 3 days at 1500°F with an internal hydrogen pressure of 1 atm. After the capsule successfully completed this phase of the operating sequence, the capsule temperature was to be increased from 1500 to 1700°F. Before the temperature increased to 1600°F, a control rod dropped, causing the reactor to scram. After restart, and upon reaching 165 Mw, the radiation detectors for the NAA 77-4 capsule sounded, indicating that the NAA 77-4 membrane had ruptured. The maximum membrane temperature recorded was 1735°F. This temperature was in the 1700°F operating control range, and should not have caused the membrane to rupture. The helium-nitrogen temperature control system was operated on pure helium for the remaining half of the ETR cycle, to maintain the membrane temperature at the low end of the temperature control range. The capsule was removed from the ETR during the normal mid-cycle refueling shutdown, and was shipped to Atomics International Hot Laboratory for postirradiation examination in June 1964. This report describes the results of the investigation, which set out to determine the cause for the premature capsule failure.

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Figure 3. External View of NAA-77 Capsule After Irradiation
(Figure Unclassified)

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Figure 4. Fast Neutron Flux Profile of ETR N-14NW Facility, as
Determined From NAA 77-4 Dosimeters
(Figure Unclassified)

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IV. CAPSULE DISASSEMBLY AND EXAMINATION

The capsule was unloaded from the shipping cask on June 30, 1964. The appearance of the capsule was good, as illustrated in Figure 3. The dosimeter wires attached to the outside of the capsule had been removed at the National Reactor Testing Station (NRTS) for analysis by Phillips Petroleum Company. Figure 4 is the flux profile obtained from the nickel dosimeter. The capsule received an estimated dosage of 4×10^{19} nvt ($E_n > 1$ Mev) at the time of removal from the ETR.

An Elox machine (electrical discharge machine) was used to make the first disassembly cut, which involved cutting through the capsule just below the upper end cap of the membrane. This cut was made 1.77 in. from the top of the upper bulkhead. Figure 5 is a view of the top and bottom sections of the capsule after completing the cross-section cut. The capsule was turned upside down, in an unsuccessful attempt to remove the fission tube. A small amount of water was poured from the capsule during this operation. It is believed that the water entered the capsule during the hot-knifing operation performed in the ETR canal to remove the capsule from the lead tube.

Next, approximately 1 in. of the capsule was cut off, in order to remove the chill block, exposing the top end of the membrane, as illustrated in Figure 6. Two of the thermocouples were easily removed with manipulators at this time, but the membrane could not be removed from the gamma heater. The bottom end of the membrane was then exposed by cutting through the capsule in the space between the bottom end of the membrane and the bottom of the well in the gamma heater. The membrane was removed by holding the gamma heater in the General Mills manipulator and pushing down on a dowel pin at the base of the membrane. A tap on the gamma heater was necessary to dislodge the membrane. A visual examination of the membrane disclosed no apparent defects. Figure 7 is a composite photograph of the membrane.

The thermocouple wires and banding straps were removed; and the membrane was visually examined, using a stereomicroscope. During this inspection, a transverse crack in the cladding was located under the fifth thermocouple band, 6.47 in. from the bottom of the membrane. This transverse crack, shown in Figure 8, extended around $3/4$ of the circumference of the membrane. The fact that the crack was transverse meant that the membrane had probably experienced a large axial tensile force during the irradiation.

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Figure 5. Cross-Sectional View
of Membrane After Cut Through
Chill Block
(Figure Unclassified)

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Figure 6. View of Top End of
Membrane After Chill Block
Removed
(Figure Unclassified)

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77-4-10, -11, -12, -13, -14, -15

Figure 7. Composite Photograph of Permeation Membrane After Removal From Gamma Heater

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Figure 8. Location of Failure in Membrane

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The diameter and straightness of the membrane were then determined, using a profilometer measuring instrument. Diameter measurements were made at 0, 60, and 120° orientations. This instrument consisted of a modified lathe bed, with two end stocks to support the membrane being measured. Two highly sensitive linear displacement transducers, which travel axially along the membrane, transmit a signal to a dual-pen recorder. Each profile scan of the membrane was repeated to check reproducibility. From the scans, diameter measurements were determined every 1/2 in. along the length of the membrane. Results of these measurements are recorded in Tables 1, 2, and 3.

Comparison of the pre- and postirradiation measurements in Table 1 indicates that the diameter of the tube decreased slightly and became more uniform during irradiation. This is the second piece of evidence which indicates that an axial tensile force was exerted upon the membrane during the irradiation. The profilometer measurements also revealed that the membrane was bowed slightly (about 0.011 in. TIR). It is believed that this bowing may have caused the difficulty encountered in removing the fission tube from the membrane. In addition to the bowing of the membrane, it was found that circumferential indentations, ranging in depth from 0.0003 to 0.0007 in., occurred at several locations along the length of the membrane, as noted in Tables 1 through 3. The maximum indentation and the membrane failure occurred under the fifth thermocouple band, located approximately 6.47 in. from the bottom of the membrane. Figure 9 is a plot of the average depth of indentation vs thermocouple band location. The indentations appeared to be typical of the necking that occurs in short-time tensile failure. This is the third piece of evidence which indicates that an axial tensile force must have been imposed upon the membrane, causing the premature failure.

A search for methods by which a tensile force could have been applied to the membrane revealed three possible means. The first part of the search was concentrated on the fission tube. Alumina had been selected as the material for the fission tube, because the thermal expansion of alumina is about half that of Hastelloy N (e.g., 8.3×10^{-6} in./in.-°F for Hastelloy N, and 3.9×10^{-6} in./in.-°F for alumina). Therefore, the thermal expansion of the alumina tube should not have caused a tensile force on the membrane under design conditions. A calculation showed that the temperature of the fission tube would have to be above 3100°F, in order to apply a tensile force to the membrane operating at 1700°F.

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TABLE 1
NAA 77-4 POSTIRRADIATION DIAMETER MEASUREMENTS

Distance From Bottom of Membrane (in.)	Diameter (in.)		
	Scan Orientation		
	0°	60°	120°
0.5	0.5632	0.5618	0.5629
1.0	0.5629	0.5618	0.5625
1.5	0.5629	0.5622	0.5625
2.0	0.5622	0.5622	0.5625
2.5	0.5618*	0.5625	0.5622
3.0	0.5618	0.5622	0.5622
3.5	0.5612*	0.5620	0.5620
4.0	0.5622	0.5622	0.5622
4.5	0.5616*	0.5614	0.5625
5.0	0.5625	0.5611*	0.5618
5.5	0.5622	0.5618	0.5625
6.0	0.5610*	0.5622	0.5622
6.5	0.5622	0.5625	0.5620
7.0	0.5618	0.5629	0.5622
7.5	0.5604*	0.5623	0.5622
8.0	0.5616	0.5618	0.5620
8.5	0.5618	0.5615*	0.5622
9.0	0.5622	0.5625	0.5629
9.5	0.5625	0.5629	0.5629
10.0	0.5624	0.5625	0.5630
10.5	0.5627	0.5629	0.5632
11.0	0.5630	0.5629	0.5632
11.5	0.5632	0.5622	0.5636
12.0	0.5632	0.5622	0.5640
12.5	0.5632	0.5622	0.5640
13.0	0.5623	0.5629	0.5643
13.5	0.5618	0.5629	0.5641
14.0	0.5622	0.5636	0.5636
14.5	0.5629	0.5632	-
Postirradiation average diameter	0.5624	0.5623	0.5624
Standard deviation	±0.0005	±0.0005	±0.0005
Pre-irradiation average diameter (4)	0.5624	0.5628	0.5630
Standard deviation	±0.0012	±0.0007	±0.0010

*Values are not included in averages, as the measurements were located in depressed areas of the membrane under the thermocouple bands.

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TABLE 2
DIAMETER MEASUREMENTS OF MEMBRANE
AT THERMOCOUPLE BANDS

Thermocouple Band, Numbered From Bottom End of Membrane	Diameter (in.)		
	Scan Orientation		
	0°	60°	120°
1	0.5629	0.5622	0.5625
2	0.5618	0.5618	0.5622
3	0.5612	0.5618	0.5614
4	0.5611	0.5607	0.5614
5	0.5607	0.5622	0.5607
6	0.5600	0.5622	0.5614
7	0.5614	0.5614	0.5622
8	0.5624	0.5625	0.5629
9	0.5633	0.5618	0.5640
10	0.5622	0.5629	-
11	0.5629	0.5628	-
Average	0.5618	0.5620	0.5620

TABLE 3
DEPTH OF INDENTATION IN MEMBRANE
UNDER THERMOCOUPLE BANDS

Thermocouple Band		Depth (in.)		
Number	Distance From Bottom of Membrane (in.)	Scan Orientation		
		0°	60°	120°
1	1.50	0.0000	0.0000	0.0000
2	2.75	0.0000	0.0002	0.0000
3	4.00	0.0010	0.0002	0.0008
4	5.00	0.0011	0.0011	0.0009
5	6.50	0.0015	0.0007	0.0015
6	7.75	0.0009	0.0002	0.0008
7	8.75	0.0006	0.0006	0.0004
8	10.25	0.0000	0.0000	0.0000
9	12.25	0.0000	0.0000	0.0000
10	14.25	0.0000	0.0000	0.0000
11	14.50	0.0000	0.0000	0.0000

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Figure 9. Depth of Necking in Membrane vs Thermocouple Band Location

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Since it would be impossible to develop a temperature differential of this magnitude, it was concluded that the fission tube could not have been the cause for failure.

The second part of the search was directed to the thermocouples and the thermocouple straps. Figure 7 shows the method used to attach the thermocouples to the membrane. The thermocouple straps were adjusted finger tight, to allow for thermocouple expansion. The tip end of the longest thermocouple was expected to move 0.045 in. relative to the membrane when heated to 1500°F [e.g.,

$$\Delta \alpha \cdot \Delta T \cdot L ,$$

where:

$\Delta \alpha$ = difference in thermal expansion between Hastelloy N and Type 304 stainless steel

ΔT = temperature rise (°F)

L = length of the thermocouple].

The straps were Type 304 stainless steel, which should have expanded 0.002 in. more than the membrane upon heating. Thus, the finger tight straps would become loose at 1500°F. During design of the capsule, it was thought that the thermocouples would slide along the membrane during a temperature change. If, for some reason, the thermocouples did not slide in the bands during a temperature rise, then an axial tensile force could be applied to the membrane. Since the straps were about 1 in. apart, the maximum stress that could be applied to the membrane by the thermocouples was 860 psi, as calculated using Euler's formula and assuming the thermocouples behaved as long slender columns. A second calculation resulted in a tensile force of 450 psi, when it is assumed that the membrane and thermocouples behaved as a bimetallic strip with the same corresponding cross-sectional areas. Since the tensile strength of Hastelloy N at 1700°F is 34,000 psi, it must be concluded that thermal expansion of the thermocouples could not produce sufficient tensile force to cause the membrane to fail.

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Figure 10. Axial Temperature Distribution in Membrane and Gamma Heater of NAA 77-4 Capsule During Irradiation

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A third possibility for the application of a tensile force to the membrane requires seizure of the slip joint between the bottom end of the membrane and the gamma heater while the capsule is at high temperature. During each reactor startup, the membrane temperature was increased to 1500°F and the gamma heater to 1100°F, for a temperature difference of 400°, as shown in Figure 10. This temperature difference caused the membrane to increase in length 0.033 in. more than the gamma heater. If the membrane seized in the thermal expansion joint during the 1500°F operation, a strain of 0.003 in./in. (assuming that 10 in. of the membrane was at temperature) would be imposed upon the membrane during cooling. Since the modulus of elasticity for Hastelloy N is 24×10^6 psi at 1500°F and 31×10^6 psi at room temperature, this strain would result in a stress of at least 72,000 psi, assuming completely elastic behavior. The yield strength at 0.2% offset varies from 50,000 psi at room temperature to 24,000 psi at 1500°F. Thus, it was concluded that the stress imposed under the stated condition is sufficient to cause the tube to yield. The yielding of the tube is evident in both the decreased diameter and the necking under the thermocouple bands. In fact, the necking must have occurred when the membrane was at an elevated temperature, because it is only then that a small temperature increase exists under the thermocouple bands. One method for a tensile force to develop when the membrane is at an elevated temperature is for the membrane to cool faster than the gamma heater. For example, in the first few seconds after a reactor scram, the membrane might cool 200°F. In the same time, the gamma heater might cool 100°F. Then a stress of about 20,000 psi could be applied to the membrane when it is still at a temperature of 1300°F. Such a stress should cause the tube to either yield or fracture, if the yielding was localized, as under a thermocouple band.

From this analysis, it was concluded that failure occurred because the membrane seized in the thermal expansion joint sometime during the 1500°F operation. Then, on a reactor scram, a sufficient tensile force was applied to the membrane to cause the membrane to rupture. Since the reactor experienced eight major scrams during the 80-hr operation at 1500°F, it is conceivable that one or all of the eight scrams may have contributed to the membrane failure.

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Figure 11. Locations Where Metallography and Adherence Test Sample Were Taken From Membrane 7568-51584

P774-19

a. Sample Taken From Top End of Membrane (350°F Irradiation Temperature)

P774-20

b. Sample Taken From Center of Membrane (1500°F Irradiation Temperature)

Figure 12. Adherence Test Samples After Bending Cladding Open

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V. METALLOGRAPHIC EXAMINATION AND ADHERENCE TESTS

Upon completion of the physical measurements, the membrane was cut, as shown in Figure 11, to provide two samples for metallographic examination and two samples for adherence testing. The 1/2-in. long rings were slit longitudinally to permit the ring to be opened with the manipulators for adherence testing. The metallographic samples were cut in half to provide four metallographic samples, comprising two transverse and two longitudinal sections. One of the longitudinal samples was selected to contain the failure, so that the grain structure adjacent to the crack could be observed. The remaining two pieces of the membrane were slit longitudinally to expose the ceramic coating for visual observation.

The adherence examination consisted of a qualitative flexure test to evaluate the bond between the ceramic and the base metal. For this purpose, the sample was spread open, gripped in two manipulators, and bent back and forth to test the flaking and spalling characteristics of the ceramic coating. Upon opening the adherence samples, however, the ceramic spalled off quite easily, indicating very poor adherence of the ceramic to the metal. Figure 12 shows the two samples after being spread open. The glass coating appeared glossy, indicating that it apparently had not devitrified.

It should be noted that adherence in the hot zones of NAA 77-1, -2, and -3 membranes has proved to be excellent after irradiation.⁽⁶⁾ The poor adherence exhibited by 77-4 may have been due either to the fact that it was one of the first production-line membranes ever made, or to the fact that it spent a relatively short time on test.

The two remaining sections of the membrane were slit longitudinally to facilitate visual examination of the ceramic coating, using a Kollmorgen periscope and a stereomicroscope. Figure 13 is a photograph showing the coating of the two sections of the membrane. The coating appeared to be in the same condition as before irradiation, except for the one spot where the ceramic had spalled. A handling accident probably caused the ceramic to spall, since the tube was found to be dented at this spot. Examination of the end cap (see Figure 14) showed no reaction between the ceramic and the alumina fission tube. It was suspected that radiation-induced diffusion might have caused the ceramic to fuse to the alumina, but it is apparent that this did not occur.

a. Upper Section

P774-21, -22

b. Lower Section

P774-23, -24, -25

Figure 13. Condition of Ceramic Coating After Irradiation

Figure 14. Membrane Bottom End Cap, Showing Lack of Reaction Between Fission Tube and Ceramic Coating

774-27

P774-18

Figure 15. Upper End of Fission Tube After Removal From Membrane

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Appearance of the fission tube, shown in Figures 13 and 15, was the same as before irradiation, except for a color change. The color changed from a light green to a very dark green. The upper section of the fission tube was removed after the membrane was cut in half. The lower section could not be removed until after the first longitudinal slice of the membrane. Since there was no evidence of bonding between the ceramics on the fission tube and that on the membrane, it was concluded that bowing of the membrane caused an interference between the fission tube and the membrane which resulted in the disassembly problem.

Metallographic samples were cold mounted in epoxy resin, ground on 240-, 320-, and 600-grit cloths, and polished with 6- and 1- μ diamond dust. Samples were etched with a solution consisting of 3 parts concentrated HCl, 2 parts H₂O, and 1 part 10% chromic acid. Difficulty was encountered in etching the sample with the crack, using this etch, but etching was successful with aqua regia saturated with CuSO₄. The photomicrograph of the fracture, Figure 16, shows the failure to be intergranular in nature. Out-of-pile SNAP 8 cladding tube creep failures have been intergranular, while short-time burst tests have been transgranular. The postirradiation failure of the NAA 68-1 membrane also was intergranular.⁽²⁾ Figure 17, which was taken about 0.1 in. away from the fracture, also shows the intergranular cracks that were present in regions of high stress near the fracture. These figures show that the SNAP 8 Hastelloy N cladding material does exhibit low ductility at high temperature, in agreement with observations by T. T. Claudson and H. F. Pessl⁽³⁾ of Hanford Laboratories and W. R. Martin and J. R. Weir⁽⁴⁾ of Oak Ridge National Laboratory. Figure 18 shows the general condition of the Hastelloy N and ceramic near the top end of the membrane. The temperature of this section was below 400°F during irradiation. The ASTM grain size varied from 5 to 7 for the complete membrane. From this observation, it is concluded that negligible grain growth occurred during irradiation. The black spots in the coating in Figure 18 are the normal randomly distributed spherical gas bubbles which develop during application of the coating.

a. Top Side of Fracture

01-2-4, -5

b. Bottom Side of Fracture

01-2-1, -2

Figure 16. Intergranular Character of Fracture

Figure 17. Hastelloy N
Grain Structure,
Approximately
0.1 in. From Failure,
Showing Intergranular
Cracks Extending
Almost Through
Cladding

01-2-6

a. Grain Structure

01-1-1

b. Coating Structure

01-1-2

Figure 18. Typical Structure of Hastelloy N and Ceramic Coating After
Irradiation at 520°F to a Dosage of 1.01×10^{20} nvt ($E_n > 1$ Mev)

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VI. CONCLUSIONS

Failure of irradiation test membrane NAA 77-4 occurred prematurely, following a reactor scram and after the operating temperature reached $\sim 1700^{\circ}\text{F}$. Postirradiation examination disclosed that the failure of NAA 77-4 was probably caused by the membrane becoming lodged in the thermal expansion joint between the membrane and the gamma heater while operating at 1500°F . Then, upon a reactor scram, a tensile force developed in the membrane as the capsule cooled, causing the membrane to fail. The evidence supporting this hypothesis is:

- 1) The membrane was stuck in the gamma heater and had to be forced out during capsule disassembly.
- 2) The failure was a transverse crack. A large tensile force is required to produce such a failure.
- 3) Additional evidence for a tensile force is that the diameter of the membrane decreased during irradiation, and necking occurred under the thermocouple bands. The maximum necking occurred at the band which covered the failure.
- 4) The necking under the bands means that a tensile force was applied while the membrane was at an elevated temperature.

Metallographic examination showed the fracture to be intergranular, with many intergranular voids in the region of the fracture. The cladding material exhibited low fracture ductility in the area of failure, since none of the grains showed evidence of deformation. Examination of the SCB-1 ceramic coating revealed poor adherence to the Hastelloy N. The coating had the same structural appearance as before irradiation. No evidence of radiation damage to the hydrogen barrier material was observed.

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