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HYDROGEN PERMEATION THROUGH SNAP-8 FUEL ELEMENT CLADDING ASSEMBLIES

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STATEMENT OF PROBLEM Review hydrogen permeability of SCB-1 and AI-8763D. Paper to be presented at the 69th Annual Meeting of the American Ceramic Society, New York, April 29 - May 4, 1967.			
ABSTRACT: The hydrogen permeability of SCB-1 and AI-8763D glass-enamels applied to Hastelloy N is shown to fit the simplified permeation equation, $F = K'A \Delta P \exp(-Q/RT)$ for the temperature range of 1200 to 1600°F. Thermal endurance permeation rates for ten SCB-1 coated cladding assemblies at 1500°F and 1 atm hydrogen pressure are presented. The simplified permeation equation was applied to endurance test data to show an expected hydrogen loss range of 1.75 to 2.5% in 10,000 hours for a typical SNAP reactor core. The calculated loss was about 1/3 to 1/2 the design goal of 5% hydrogen loss in 12,000 hours for SNAP-8 reactors. This work was supported by the U.S. Atomic Energy commission under contract: AT(04-3)-701B.			

HYDROGEN PERMEATION THROUGH SNAP-8 FUEL ELEMENT CLADDING ASSEMBLIES

by C. J. Engberg

Hydrogen retention is a unique design requirement of the U-Zr-H nuclear reactors. In the SNAP-8 reactors, more than 95% of the hydrogen must remain in the fuel after 12,000 hours of operation. The fuel element cladding assemblies became the reactor components which prevented excessive hydrogen losses in the evolution of the SNAP reactors. The SNAP reactor fuel elements consist of solid cylinders of hydrided zirconium - 10% uranium alloy contained in glass-enameled Hastelloy N cladding assemblies. In operation, a differential hydrogen pressure is developed across the cladding assembly as a result of thermally-induced dissociation of the hydrided fuel. The glass enamel provides a barrier which prevents excessive losses of hydrogen from the fuel. Two high temperature glass enamels have been developed by Atomics International to meet the stringent compatibility and hydrogen retention requirements of the SNAP fuel element assemblies. The glass-enamels are called AI-8763D and SCB-1. This paper discusses the hydrogen retention properties of these coatings when applied to the SNAP reference fuel cladding material, Hastelloy N.

A diagram of the equipment for measuring hydrogen permeation rates is shown in Slide 1. Pressure, temperature, and hydrogen flow are recorded for each experiment. Hydrogen flow measurements are continuously recorded by use of a mass spectrometer. The hydrogen conduction area is determined by the physical size of the tube sample, and the length of the tube in the isothermal zone of the furnace. The glass-enameled tube extends out of the isothermal zone of the furnace.

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A sharp temperature gradient at the mouth of the furnace is required to insure that the flow from this portion of the membrane is always less than 10% of the total flow. Four furnaces can easily be connected to a single mass spectrometer as shown in Slide 2.

Glass-enameled reduces the hydrogen permeability of a metal by a factor of one thousand. Slide 3 compares the permeation rates of bare Hastelloy N with those of SCB-1 and AI-8763D coated Hastelloy N. In addition, the permeation rates for some of the common metals are also included. The difference between the permeation rates for the four coated samples is the normal scatter found between identically processed samples.

An interesting fact about hydrogen permeation through homogeneous metal and glass membranes is revealed in the pressure dependence shown in Slide 4. Hydrogen permeates as atoms in metals, resulting in a dependence on the square root of pressure; whereas, hydrogen permeates as molecules in glasses, resulting in a linear pressure dependence. The total flow through enameled membranes is made up of the flow through bare metal and the flow through the glass-enameled portion of the membrane. Slide 5 shows the two flow paths for hydrogen in an enameled membrane. Of course, if the enamel were free of holes and cracks, the amount of flow through bare metal only (F_m) would be zero. Substituting the values for F_c and F_m from the previous slide yields the complete permeation equation for glass-enameled membranes. This equation has too many variables to solve without some simplifying approximations. We assumed that the simplified permeation equation shown in Slide 5 could be used in the limited temperature range of interest (1200 to 1500°F).

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Slide 6 shows how the hydrogen permeation rates for a typical Hastelloy N membrane coated with AI-8763D fit the simplified permeation equation. Note that a linear pressure dependence was found for this membrane except at 1500°F. The 18 kcal/mole activation energy for this sample is on the low end of the activation energy range. Activation energies of 18 to 33 kcal/mole have been calculated from permeation data on Hastelloy N coated with AI-8763D. The two data points marked 'A' are the only two points which do not fall on the 18 kcal/mole line; and they are the same data points which cause the curvature in the 1500°F pressure dependence relationship. The difference between the experimental and the extrapolated value, at three atmospheres hydrogen pressure and 1500°F, is much too large not to be a real difference. A possible explanation for this effect is that at 1500°F, the enamel is soft enough to flow under higher pressures and will seal cracks or pores in the enamel.

Slide 7 shows that SCB-1 enamel has a linear pressure dependence and two activation energies in the temperature range of interest. Above 1400°F, the activation energy is approximately 27 kcal/mole and below 1400°F, it is only 12 kcal/mole. Large differences in activation energy for permeation through metal oxides and enamels are reported in the literature. T. Smith, in a study of hydrogen diffusion through the zirconium oxide on zirconium, reports activation energies ranging from 8 to 40 kcal/mole, and was unable to explain the reason for the large range. This problem is one which needs additional attention in order to be properly explained.

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Until now, we have discussed instantaneous hydrogen permeation rates. In order to use these permeation rates to predict the hydrogen retention properties of fuel element cladding assemblies, the permeation rate as a function of time must be known. The last slide shows the permeation rates of SCB-1 as a function of time at 1500°F and 1 atm hydrogen pressure. The gradual increase in permeability with time indicates some coating degradation occurs. The maximum and minimum endurance test data were used with the simplified permeation equation developed for SCB-1 to calculate the % of hydrogen content lost by the fuel in a SNAP reactor as a function of time. To make the calculations, the following conditions were assumed for all elements in the reactor core:

a. Fuel hydrogen content	68,000 cc(STP) hydrogen
b. Inlet coolant temperature	1100°F
c. Outlet coolant temperature	1300°F
d. Maximum cladding temperature	1400°F
e. Cladding assembly size	0.571 O.D. x 17.325 in.

For these fuel element conditions, hydrogen losses ranging from 1.75 to 2.5% in 12,000 hours should be expected. The calculated H_2 loss is between 1/3 and 1/2 the design goal of 5% loss in 12,000 hours for the SNAP-8 reactor core.

PERMEATION EQUIPMENT SETUP

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SLIDE 3

HYDROGEN PERMEATION THROUGH METALS AND COATINGS

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HOMOGENEOUS MEMBRANES

METALS

$$F_m = K_m \frac{A}{d} \Delta p^{1/2} \exp -Q_M/RT$$

GLASSES

$$F_c = K_c \frac{A}{d} \Delta p \exp -Q_c/RT$$

F = TOTAL GAS FLOW (cc(STP)/hr)

K = PERMEATION CONSTANT (cc(STP)/atmⁿ-cm hr)

Δp = PRESSURE DIFFERENTIAL ACROSS MEMBRANE (atm)

A = CONDUCTING AREA (cm²)

d = THICKNESS OF HOMOGENEOUS MEMBRANE (cm)

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SLIDE 4

HETEROGENEOUS MEMBRANES

$$F_T = K_m \frac{A_m}{d_m} \Delta p^{1/2} \exp(-Q_M/RT) + K_c \frac{A_c}{d_c} \Delta p \exp(-Q_c/RT)$$

$$F_T \approx K'A \Delta p^n \exp(-Q/RT)$$

SLIDE 5

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SLIDE 6

AI 8763D COATED HASTELLOY N

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SLIDE 7

SCB-I COATED HASTELLOY N

7-7696-105-7

SLIDE 8

HYDROGEN PERMEABILITY vs TIME

7-7696-105- A