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HIGH-TEMPERATURE IRRADIATION OF A
ZIRCONIUM-HYDRIDE-2 w/o
URANIUM ALLOY

by

Gerald E. Lamale
Alan W. Hare
Horatio H. Krause
Alan K. Hopkins
John H. Stang
Eugene M. Simons
Ronald F. Dickerson

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BATTELLE MEMORIAL INSTITUTE
505 King Avenue
Columbus 1, Ohio

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Enriched $ZrH_{1.65}$ -2 w/o uranium specimens were irradiated under 1 atm of hydrogen at center-line temperatures between 900 to 1400 F to uranium burnups of between 13 and 25 a/o. Specially designed irradiation capsules were used to provide the conditions of temperature and hydrogen atmosphere which were required. Each capsule was instrumented with five thermocouples so that ample temperature data could be obtained during the irradiation.

Almost negligible density changes were produced in the material by the irradiation. Changes in length and diameter were of a degree which could fall within experimental error in measurement. Metallographic examination showed no change in microstructure which could be attributed to the effect of irradiation.

INTRODUCTION

The prompt negative temperature coefficient of reactivity inherent in reactor systems making use of an intimate mixture of fuel and moderator is a valuable safety feature. One form of such a fuel-moderator combination that appears to have excellent potential for gas-cooled power reactors is the hydrided fuel alloy. The use of this solid material is an attractive method for introducing hydrogen moderation into these high-temperature reactor systems.

Zirconium hydride-uranium alloys are currently of interest for this application. The corrosion resistance, low cross section, and elevated-temperature strength of zirconium make it an excellent base for alloy fuels. Techniques for preparing zirconium hydride-uranium alloys that are stable at elevated temperatures have been developed. In irradiation tests, such alloys have performed satisfactorily at uranium burnups of up to 5 a/o at low temperatures. (1)

The objective of the present investigation was to evaluate the effects of exposing a delta-phase zirconium hydride-uranium alloy to high-temperature high-burnup irradiation. Delta zirconium hydride appears most useful for application in power reactors because it does not form low-melting eutectics with iron and nickel. A $ZrH_{1.65}$ -2 w/o uranium alloy was chosen for evaluation.

(1) References are at end.

Two separate programs were involved in the study. One phase involved the preparation and evaluation of the alloy. Techniques developed in an earlier study were employed in preparing the material. (2,3) The other phase of the study was concerned with the design and preparation of suitable capsules for the irradiation tests in the MTR. The necessity for surrounding the alloy specimens with a hydrogen atmosphere during the irradiation made an unusual capsule design necessary.

FABRICATION OF SPECIMENS

Two individual alloys of zirconium-2 w/o fully enriched uranium were made by repeated arc melting until the individual buttons were considered completely homogeneous. The material was then melted into shapes about 2-1/2 in. long and a 1/2 in. in diameter and these shapes were hot rolled and cold swaged until they were slightly over 3/16 in. in diameter and about 16 in. long. From this material individual specimens were machined to a size of 3/16 in. in diameter and either 1 in. or 1/2 in. long. Then a 1/16-in. hole was bored axially through each specimen. Similar specimens of unalloyed zirconium were melted, fabricated, and machined by the same techniques used for the fueled specimens.

All fueled specimens (designated RD) were made from the same casting, RD-1, except for one specimen, which was made from a second casting (designated RD-2). Both castings were 93 per cent enriched and were identical so far as fabrication techniques are concerned. Chemical analyses of the two rods were as follows:

	<u>Analyses, w/o</u>	
	<u>RD-1</u>	<u>RD-2</u>
Uranium	2.13	2.24
Carbon	0.025	0.031
Zirconium	97.84	97.73
(by Difference)		

To check the variation of density of zirconium-2 w/o uranium within a given rod, two 1/2-in. pieces were cut off opposite ends of the second rod. The density of the two specimens varied from their common mean by 1.2 parts per thousand.

The delta phase of zirconium hydride has a relatively large hydrogen content, with a hydrogen-to-zirconium ratio of 1.6:1.7. Previous X-ray diffraction data on the uranium-zirconium-hydrogen system showed that no uranium hydride forms when a uranium-zirconium alloy is hydrided. (3) Only X-ray patterns for alpha uranium and delta zirconium hydride were observed at high hydrogen contents, although these may also have been present in trace amounts of uranium-zirconium solid solution. On the basis that the hydrogen all combines with the zirconium, sufficient hydrogen was introduced into the alloy to reach the composition $\text{ZrH}_{1.65}$. The resulting atom ratios of the three elements were:

$$\text{U:Zr:H} = 0.008:1:1.65.$$

The nonfueled specimens were also hydrided to the composition $\text{ZrH}_{1.65}$. The immediate source of the hydrogen was uranium hydride, which was decomposed at 750 F to yield high-purity hydrogen. The uranium hydride was prepared previously from tank hydrogen from which oxygen, nitrogen, and water vapor were removed by passing the gas through a Deoxo unit and hot calcium hydride (1400 F).

The hydriding was accomplished⁽²⁾ in two steps: (1) hydrogen was reacted with the samples at 1700 F until a composition of $\text{ZrH}_{1.0}$ was reached, (2) the temperature was then lowered to 1500 F, and additional hydrogen was introduced over a 4-hr period until the composition $\text{ZrH}_{1.65}$ was achieved. The samples were then homogenized by maintaining the 1500 F temperature for 5 additional hr, after which the temperature was reduced to 800 F at a rate of 100 F per hr. The samples were then allowed to cool to room temperature with the furnace.

The hydrided fueled and nonfueled specimens are shown in Figures 1 and 2. A photomicrograph of a surface of freshly prepared zirconium hydride-2 w/o uranium is shown in Figure 3. As can be seen in Figure 3, the hydriding is more complete to a depth of about 0.002 in. from the outside surfaces than it is in the interior. The hydrogen is absorbed as an inverse exponential function of distance from the surface. It is interesting to note that this difference in hydrogen composition is not detectable by hardness determinations. Table 1 lists hardnesses of the unirradiated control material and the irradiated material.

ENCAPSULATION OF SPECIMENS

The capsule design for this particular irradiation was somewhat complicated because of the necessity of blanketing the specimens with hydrogen during irradiation and the need for dampening any vibration that might prove damaging to the specimens.

Figure 4 is an assembly drawing of the capsule design that was formulated. Figures 5 and 6 are views of the capsule components at various stages during assembly. Figure 7 is a view of Capsule BMI-20-1 just prior to final assembly. Capsule BMI-20-2 was identical in design and construction. Several of the important features of the design and the bases for their selection are described in the following paragraphs.

As indicated in Figure 5, each capsule was loaded with three fueled and two non-fueled hydride specimens. In addition, one-half of a fueled specimen was located at each end of the assembly to minimize axial heat flow and, thus, increase the uniformity of the axial temperature profile. Each of the fueled specimens was a hollow cylinder, nominally 3/16 in. in OD, 1/16 in. in ID, and 1 in. long, weighing 2.62 ± 0.03 g and containing a nominal loading of 0.048 g of uranium-235 per in.

A unique feature in the capsule design was the method employed to retain the hollow specimens within the stainless steel shell which served as the inner container of the capsule assembly. The specimens were encased in a close-fitting 5-mil-thick molybdenum sheath held axially by stainless steel end plugs slipfitted into this sheath and welded to the stainless steel. To minimize radial constraint on the specimens

4x

Specimen RD-1-6

RM9567

This specimen was cut in half, and the two pieces were used at top and bottom as fission-heat retainers.

4x

Specimen RD-1-9

RM9568

4x

Specimen S-3

RM9565

HX

Specimen RD-1-13

RM9569

4x

Specimen S-6

RM9565

4x

Specimen RD-1-11

RM9568

FIGURE 1. PREIRRADIATION VIEWS OF FUELED AND NONFUELED SPECIMENS AS LOADED FROM TOP TO BOTTOM IN CAPSULE BMI-20-1.

The fueled specimens are designated by the letters RD, the nonfueled specimens are indicated by the prefix S. The grooves and tapered ends of some of the specimens served as auxiliary identification marks.

4x

Specimen 1-15

RM9569

This specimen was cut in half, and the two pieces were used at top and bottom as fission-heat retainers.

4x

Specimen RD-1-12

RM9568

4x

Specimen S-10

RM9565

4x

Specimen RD-1-16

RM9569

4x

Specimen S-2

RM9565

4x

Specimen RD-1-14

RM9569

FIGURE 2. PREIRRADIATION VIEWS OF FUELED AND NONFUELED SPECIMENS AS LOADED FROM TOP TO BOTTOM IN CAPSULE BMI-20 -2

The prefix RD indicates fueled specimens, and the prefix S indicates nonfueled specimens. The circumferential grooves and tapered ends on some of the specimens served as auxiliary identification marks.

250X

RM13244

Etchant: 50 Cm³ Lactic Acid, 50 Cm³ HNO₃, and 5 Drops HF

a. Outer Diameter

250X

RM13245

Etchant: 50 Cm³ Lactic Acid, 50 Cm³ HNO₃, and 5 Drops HF

b. Inner Diameter

FIGURE 3. CROSS-SECTIONAL VIEW OF ETCHED
NONIRRADIATED CONTROL SPECIMEN

Note change in grain structure at edges suggesting
difference in hydrogen concentration.

TABLE 1. TABLE OF KNOOP HARDNESS OF CONTROL
AND IRRADIATED SPECIMENS

Knoop Hardness at Approximate 0.01-Cm Intervals From Outside Surface				
Control	RD-1-9	RD-1-11	RD-1-12	RD-1-14
236	269	223	246	279
236	269	239	261	249
230	283	226	252	258
236	291	231	272	239
227	283	218	300	291
233	275	296 (a)	255	288
241	237	239	272	279
233	261	249	317	220
233	314	322 (a)	309	239
239	206 (a)	246	326	350 (b)
239	336	252	296	
236	368	258	356	
239	356	291		
233				

(a) May be caused by indenter falling within grain boundary.

(b) Distorted impression.

FIGURE 4. ASSEMBLY OF CAPSULE FOR IRRADIATION OF $ZrH_{1.65-2}$ w/o URANIUM SPECIMENS

1. Stainless steel top plug
2. Nilstain spring
3. Top molybdenum end piece
4. One-half of Specimen RD-1-6
5. Specimen RD-1-9
6. Spacer S-3
7. Specimen RD-1-13
8. Spacer S-6
9. Specimen RD-1-11
10. One-half of Specimen RD-1-6
11. Bottom molybdenum end piece
12. Nilstain spring
13. Stainless steel bottom plug
14. Thermocouple: bent to approximate shape as an example of thermocouple already brazed into assembly wall (Thermocouple 5)
15. Molybdenum sheath
16. Thermocouple - after brazing into top plug (Thermocouple 1)
17. Hydrogen-loading assembly
18. Thermocouples 2, 3, and 4
19. Central assembly body
20. Bottom couple protector
21. Bottom capsule plug
22. Top portion of double-seal cap
23. Bottom portion of double-seal cap

N52078

FIGURE 5. EXPLODED VIEW OF CAPSULE BMI-20-1

This capsule is interchangeable with Capsule BMI-20-2.

FIGURE 6. CENTRAL CAPSULE ASSEMBLY OF BMI-20-1

Capsule BMI-20-2 was identical.

N52079

FIGURE 7. VIEW OF CAPSULE BMI-20-1 JUST PRIOR TO FINAL ASSEMBLY

Capsule BMI-20-2 was identical.

during irradiation, the seam of the sheath was not closed. Axial restraint on the specimens to preclude up-and-down movement, particularly during capsule-maneuvering operations, was created by Inconel X springs. These springs were separated from the fueled specimens by molybdenum end pieces. Molybdenum was chosen for this application because it does not form hydrides or low-melting eutectics with uranium.

The molybdenum sheath allowed accurate initial centering of the specimens and provided the specimen assembly with considerable shock resistance. This was an important consideration in view of the brittleness of the specimens and the constant pounding that a MTR capsule must withstand because of the rapid flow of cooling water past it.

Hydrogen was used as the atmosphere surrounding the hydride specimens. After the stainless steel plugs were welded into the inner container, the chamber was evacuated through the hypodermic tube shown in Figures 4, 5, and 7, and approximately 1 atm of hydrogen was introduced into it. Afterwards, the tube was sealed by crimping and welding.

During irradiation, the hydrogen pressure was estimated to be about 3 atm owing to thermal compression. This is significantly higher than the 1 atm of hydrogen required to maintain equilibrium with the hydride at the 1500 F level.

The inner and outer containers, which were to remain at fairly low temperatures during the irradiations, were fabricated of Type 347 stainless steel. These were sturdily constructed, having 1/8-in.-thick walls. All joints were made by the Heliarc welding technique and were carefully inspected with a helium leak detector before the hydrogen loading.

The 11/64-in. annulus separating these containers provided room for the thermocouples leading into the specimen zone. Because of its high conductivity, NaK was chosen as the annulus medium. The temperature gradient from the inner container outward had to be quite low to compensate for the steep temperature gradient across the hydrogen gap between the specimens and the wall of the inner container.

The design of the assembly is such that the specimens can be removed after irradiation without opening the NaK chamber. As can be seen in Figure 4, the specimens were made accessible merely by making a single cut near the capsule bottom.

Five Chromel-Alumel thermocouples (1/16-in.-OD stainless steel-sheathed units with MgO insulation) were provided in each capsule. These were positioned as follows:

- (1) One within the hollow of the top and the bottom fueled specimens to provide temperature data subject to a minimum of uncertainty; these are referred to as Thermocouples 1 and 5 in Figures 5, 6, and 7. The tips were protected by 1 mil of vapor-deposited molybdenum.
- (2) Three (Thermocouples 2, 3, and 4) along the inner container; these were inserted through the wall so that the tips emerged into the hydrogen space. Since in this capsule design the temperature gradient in this space is high, there must be gross uncertainty associated with the temperature. These three thermocouples were intended only to indicate general temperature fluctuations.

The thermocouple point-of-entry joints into the inner container were made by a furnace-brazing operation using a standard nickel-alloy-type braze which melts at approximately 1820 F; the remelting temperature is approximately 1850 F. These temperatures are considerably higher than those to which the brazes were to be exposed in service. At the operating temperatures, the braze materials were not to be affected by the hydrogen atmosphere.

As shown in Figure 7, all thermocouples were led from the capsule proper into the lead tube through a double-seal assembly. The seals here were also formed by furnace brazing, using the high-melting-point braze mentioned above. The leaktightness of all brazed joints in these capsule assemblies was confirmed by a careful inspection procedure in which a helium leak detector was employed.

The capsules were provided with 25-mil nickel-0.6 w/o cobalt dosimeters to obtain data on the integrated neutron flux during the irradiation. One dosimeter was placed along the length of the capsule on the molybdenum sheath parallel with the axis of the specimens and one along the length on the outside of the capsule.

In order to accomplish the desired burnup of uranium in the allotted time, it was necessary to design the capsule for operation at fluxes characteristic of the reactor positions generally available at the MTR. In order to learn something relating to capsule design, predictions of temperatures of the specimen for several conditions of irradiations were made. By making sensible comparisons with observed irradiation phenomena, it was planned that a great deal could be learned and used for other fueled-moderator capsule designs.

Pertinent specimen-temperature and heat-generation constants for the two capsules are presented in Table 2. Aside from the desirability of achieving the specimen temperatures listed, the designs were based on (1) a requirement to obtain a fission uranium-235 burnup of 20 per cent in the allotted irradiation period and (2) the necessity of locating the capsules in reactor positions that would be readily available, i. e., those having quoted fluxes in the neighborhood of 1×10^{14} nv.

The effective flux requirements were estimated by applying the following expression:

$$\phi_e = 0.7 \phi_u f e^{-(\Sigma_a)x},$$

where

ϕ_e is the effective flux

ϕ_u is the estimated unperturbed flux

f is the Brad Lewis perturbation factor (in this case, 0.93)

$e^{-(\Sigma_a)x}$, is the capsule attenuation based on Beer's law (in this case, 0.86)

where

Σ_a = total cross section in barns

TABLE 2. VALUES USED IN DESIGN STUDY

Capsule	Nominal Design Temperature, F		Design Flux, 10^{14} nv		Calculated Specimen Fission-Heat Generation ^(b) , Btu/(hr)(in.)	Irradiation Time Required to Produce 20 Per Cent Fission Burnup of Uranium-235, days
	Inner Surface of Specimen (Temperature Measured by Thermocouples)	Outer Surface of Specimen ^(a)	Unperturbed	Effective Specimen		
BMI-20-1	1500	1440	0.92	0.52	352	105
BMI-20-2	1200	1160	0.66	0.37	250	120

(a) The thermal conductivity of the specimens, approximately 5 Btu/(hr)(ft)(F), was taken to be that of nonfueled ZrH_{1.65}. The addition of the small amount of uranium to the specimens is assumed to have a negligible effect upon the conductivity.

(b) The gamma heating for these capsules was estimated to be approximately 120 Btu per in. of capsule length; this is based on a nominal gamma-heat production of 1.5 w per g.

x = thickness of capsule.

The value 0.7 is a correction factor based on Battelle experience with MTR-irradiated capsules.

While the Brad Lewis factor is not strictly applicable to hollow specimens, it might be applied satisfactorily in this case since the uranium loading of the specimens was small; consequently, the flux perturbation by the individual specimens was low. The Battelle factor of 0.7 is an average based on a study of a large group of experimental data from capsules irradiated in the MTR; its limits of uncertainty and its applicability to all capsule configurations have not been established.

Figure 8 shows the predicted radial temperature profile for Capsule BMI-20-1, assuming a center-line temperature of 1500 F, specimens accurately centered, and negligible end losses of heat. Axial fission-heat losses are minimized by fission heating in the fueled semipieces at each end of the specimen assembly. It may be seen from Figure 8 that the major temperature drop in the system occurs across the hydrogen gap. Heat transfer through this gap is by conduction from the molybdenum sheath to the inner container wall. Because of the high conductivity (for a gas) of hydrogen [a value of 0.20 Btu/(hr) (ft) (F) was used in the calculations for the 1500 F case and 0.18 Btu/(hr) (ft) (F) for the 1200 F case], the narrowness of the gap, and the low emissivity of molybdenum, radiative heat transfer was negligible.

The interpretation of temperatures measured by thermocouples located within the specimens in terms of fission-heat generation was subject to a number of factors other than the effect of the lateral displacement of the specimen with respect to the inner surface of the inner capsule. These other factors included the dimensions of the hydrogen gap, the conductivity of the hydrogen, and the gamma-heat generation. The latter factor was considered negligible in these calculations. It was assumed that the relative error was ± 2 per cent for the measurement of the outer radius of the gap and ± 5 per cent for the measurement of the inner radius of the gap and that the hydrogen-conductivity relative error was ± 10 per cent. The expression for the heat transfer for the concentric case at 1500 F was therefore

$$T_c = (60 \pm 10 \text{ F}) + \frac{352 \times 12}{2\pi} \left(\frac{\ln \frac{r_o \pm 2 \text{ per cent}}{r_i \pm 5 \text{ per cent}}}{k_{H_2} \pm 10 \text{ per cent}} \right) + (70 \pm 20 \text{ F}) + (120 \pm 10 \text{ F}),$$

where

Term I ($60 \pm 10 \text{ F}$) included an estimate of the absolute error of 10 F in the temperature difference across the specimen from uncertainty limits in the conductivity of the fueled material

Term II $\left[\frac{353 \times 12}{2\pi} \left(\frac{\ln \frac{r_o \pm 2 \text{ per cent}}{r_i \pm 5 \text{ per cent}}}{k_{H_2} \pm 10 \text{ per cent}} \right) \right]$ was the expression for the temperature difference across the hydrogen gap

Term III ($70 \pm 20 \text{ F}$) included an estimate of the uncertainty in the temperature difference from the edge of the hydrogen gap to the water

FIGURE 8. NOMINAL CALCULATED RADIAL TEMPERATURE FOR CAPSULE BMI-20-1

Term IV (120 ± 10 F) was the pool-water temperature and its associated absolute error.

Evaluation of the individual factors involved was made on the basis of a statistical interpretation of the linear absolute error terms. Applying a Taylor series of expansion* of three variables, r_i , r_o , and k , to Term II yielded a relative error of approximately $\pm 17\frac{1}{2}$ per cent and, for a 1250 F ΔT in Term II the over-all expression becomes

$$T_c = (60 \pm 10 \text{ F}) + (1250 \pm 220 \text{ F}) + (70 \pm 20 \text{ F}) + (120 \pm 10 \text{ F}) = 1500 \pm 225 \text{ F}.$$

The curves in Figure 9 were obtained by this procedure for the fission-heat range of interest.

Two types of calculations were made to estimate how the temperature level of a specimen was affected by a shift of the specimen to an off-center position in the annulus. In the first of these, the width of the entire hydrogen annulus was changed to the minimum distance occurring between the wall and the eccentrically positioned specimen. The results of calculations on this highly simplified basis are presented in Figure 10. For a 0.015-in. shift, the temperature drop was indicated to be about 1000 F on the side closest to the wall and about 1470 F on the opposite side. Any eccentricity would result in a reduction of the average ΔT across the gap, so the concentric case results in the maximum barrier for heat flow.

The second type of calculation made was more rigorous, utilizing a mathematical "shape factor"⁽⁴⁾ for cylinders, one inside the other and eccentrically positioned with respect to the outer wall. The surfaces of the eccentric cylinders are assumed isothermal. For the concentric case in Capsule BMI-20-1, the calculated nominal temperature drops are 60 F (center line to surface of inner cylinder), 1250 F (across the gas gap), and 70 F (inner surface of outer cylinder to ambient). As shown by the results given in Figure 11 for small departures from the concentric case (≈ 0.015 in.), the ΔT across the hydrogen annulus is subject to a reduction of approximately 50 F, which would also reduce the specimen center-line temperature by 50 F.

As a check on the accuracy of the above methods, a two-dimensional electrical analog was constructed, and tests were made. The radial cross section of the capsule was simulated using Teledeltos electrically conducting paper to which a voltage was applied. The thermal conductivities of the various materials were represented by various numbers of layers of the conducting paper. The results were as follows:

- (1) Simulating the case for the specimen centered in the hydrogen gap, the analog solution gave a ΔT across the hydrogen gap of 1240 F, with a specimen center-line temperature of 1500 F.
- (2) With an eccentricity of 15 mils, the maximum ΔT across the widest part of the hydrogen gap was 1210 F, the minimum was 1080 F, and

* This approach gives the relative uncertainty p_y in a function y of n variables, $x_1, x_2, \dots, x_n$, each of which has its own independent relative uncertainty, p_i , as

$$p_y = \frac{1}{y} \sqrt{\sum_{i=1}^n \left(x_i \frac{\partial y}{\partial x_i} p_i \right)^2}.$$

FIGURE 9. ESTIMATED UNCERTAINTY IN FISSION-HEAT GENERATION WITH CONCENTRIC SPECIMEN POSITIONING

FIGURE 10. ΔT ACROSS HYDROGEN GAP WITH SPECIMEN NONCONCENTRICITY BASED ON SIMPLIFIED VARIABLE CONCENTRIC-CYLINDER ANALYSIS

Plot assumes a specimen fission-heat generation of 352 Btu/(hr)(in.) and temperature dependency of k_{H_2} . Capsule BMI-20-1 is considered here.

FIGURE 11. ΔT ACROSS HYDROGEN GAP AS RADIAL POSITION IS CHANGED ACCORDING TO ANALYSIS EMPLOYING A SHAPE-FACTOR CONCEPT

Capsule BMI-20-1 is considered here.

the temperature at the center of the fuel was 1430 F. The drop of 70 F in the fuel center-line temperature for an eccentricity of 15 mils and a total heat flow corresponding to the concentric case was in good agreement with the 50 F drop obtained from the shape-factor solution which assumed an isothermal specimen surface.

The curves in Figure 12 illustrate the estimated effects of specimen shifting on thermocouple information in Figures 10 and 11 in the following way:

- (1) Values based on the simplified concentric-cylinder analysis were obtained by assuming that the thermocouple measures the arithmetic mean between the maximum and minimum temperatures at the inner surface of the specimen.
- (2) Values based on the shape-factor analysis were obtained in the same general way, using Figure 11 instead of Figure 10 to find the average ΔT across the hydrogen gap.
- (3) The value for the specimen center-line temperature corresponding to a 15-mil eccentricity as obtained from the analog experiment is also shown.

The analyses concerning eccentricity must be based on the questionable assumption of isothermal surfaces. Temperature data obtained directly from electrical-analog studies may have an inherent error of the order of ± 20 per cent as a result of anisotropy and inhomogeneity of conducting paper, inability to simulate conductances exactly, operational techniques, etc. In the reference heat-flow case, the maximum eccentricity envisioned, 0.015 in., would not result in a reduction of more than 50 F in either average or peak specimen temperature. Of course, as fission-heat generation is decreased from that assumed for the reference case, this effect decreases about proportionately. Figure 13 is a plot of fission-heat generation against thermocouple reading with the effect of eccentricity taken into account. The upper limit here is obtained by combining the temperature error introduced by the estimated maximum eccentricity of 15 mils with the upper curve in Figure 9; the lower curve in Figure 9 is not affected since eccentricity produces temperature errors in only one direction.

IRRADIATION HISTORY

Table 3 gives the irradiation history of the two capsules. The average cycle temperatures indicated in the table were derived from the recorded center-line temperature-time charts. Both general temperature levels and reported fluxes are lower than the design specifications (for BMI-20-1, 1500 F at $\phi_u = 0.92 \times 10^{14}$ nv and for BMI-20-2, 1200 F at $\phi_u = 0.66 \times 10^{14}$ nv). The relationship between the average cycle temperature and the corresponding quoted flux is not consistent with what would be expected analytically. This is not surprising, in view of the uncertainties in quoted flux, flux-perturbation factors, and capsule heat-transfer characteristics. However, when ϕ_u is plotted against T_c for each cycle, all but 2 of the 32 points fall within the

FIGURE 12. ESTIMATED THERMOCOUPLE TEMPERATURES FOR REFERENCE HEAT-FLOW CASE $q = 352 \text{ Btu}/(\text{hr})(\text{in.})$

CAPSULE BMI-20-1 is considered here.

FIGURE 13. ESTIMATED UNCERTAINTY IN FISSION-HEAT GENERATION WITH ECCENTRICITY INCLUDED

TABLE 3. IRRADIATION HISTORY FOR CAPSULES BMI-20-1 AND BMI-20-2

MTR Cycle	Irradiation			Location in Reactor	Position From Top of Reflector, in				MTR Estimated Flux (Center of Specimen), 10 ¹⁴ n _v		Core Temperatures, ^(a) F					
					Capsule		Center of Specimen		Top	Bottom	Top Specimen			Bottom Specimen		
	Top	Bottom	Top		Bottom	Maximum	Minimum	Mean			Maximum	Minimum	Mean			
BMI-20-1 ^(b)																
112	554	38.4	13.4	A-30-SE	2.0	15.0	9.0	13.0	0.26	0.47	1050	925	1000	1450	1250	1400
113	572	37.6	14.4	A-30-SE	2.0	15.0	9.0	13.0	0.26	0.47	1025	920	980	1320	1220	1290
114	568	21.8	14.7	A-30-SE	5.0	18.0	12.0	16.0	0.44	0.62	1175	1115	1140	1355	1325	1335
115	585	39.4	14.8	A-30-SE	7.0	20.0	14.0	18.0	0.53	0.71	1255	1190	1220	1300	1280	1290
116	673	38.6	16.7	A-30-SE	9.5	22.5	16.5	20.5	0.67	0.78	1215	1110	1160	1265	1215	1245
117	624	39.1	15.8	A-30-SE	12.5	25.5	19.5	23.5	0.77	0.83	1115	1005	1060	1275	1235	1250
118	557	38.5	14.2	A-30-SE	12.5	25.5	19.5	23.5	0.77	0.83	--	--	--	--	--	--
104.0 ^(c)																
BMI-20-2 ^(b)																
112	554	38.4	13.4	A-40-SW	13.5	25.5	19.5	23.5	0.68 ^(d)	0.73 ^(d)	1305	1275	1290	1260	1220	1240
113	572	37.6	14.4	A-40-SW	0	13.0	7.0	11.0	0.27	0.44	915	820	865	1170	1100	1150
114	568	21.8	14.7	A-40-SW	2.0	15.0	9.0	13.0	0.36	0.53	1015	905	950	1210	1190	1195
115	585	39.4	14.8	A-40-SW	2.0	15.0	9.0	13.0	0.36	0.53	1020	935	975	1200	1180	1190
116	673	38.6	16.7	A-40-SW	3.5	16.5	10.5	14.5	0.45	0.65	1095	955	1015	1220	1175	1195
117	624	39.1	15.8	A-40-SW	3.5	16.5	10.5	14.5	0.45	0.65	1085	950	1010	1160	1095	1135
118	557	38.5	14.2	A-40-SW	3.5	16.5	10.5	14.5	0.45	0.65	1035	930	970	1150	1055	1090
119	587	39.5	15.2	A-40-SW	3.5	16.5	10.5	14.5	0.37 ^(d)	0.53 ^(d)	890	740	845	960	865	935
120	633	37.7	14.0	A-43-SE ^(e)	5.0	18.0	12.0	16.0	0.37 ^(d)	0.59 ^(d)	--	--	--	--	--	--
133.2 ^(c)																

(a) No temperature charts are available for Capsule BMI-20-1 in Cycle 118 and for Capsule BMI-20-2 in Cycle 120

(b) Capsule BMI-20-1 was discharged at Cycle 119 shutdown (March 16, 1959), Capsule BMI-20-2 was discharged at Cycle 121 shutdown (April 6, 1959)

(c) Total irradiation based on temperature charts

(d) Fluxes were taken from map of A-39-NE for Cycle 112

(e) Transferred to A-43-SE during Cycle 120 shutdown

uncertainty band established from uncertainty assumptions of ± 20 per cent for ϕ_u and ± 20 per cent for the over-all perturbation factor, along with the heat-generation uncertainties indicated in Figure 13.

POSTIRRADIATION EXAMINATION

After the irradiations were completed, the capsules were removed from the test reactor and examined in the Battelle Hot-Cell Facility. As an initial step in this examination, the capsules were photographed as received from the reactor. These photographs are shown in Figure 14. It is interesting to note their excellent appearance. Both capsules were opened individually in a pipe fitter's vise by cutting off the lower end. This cutting made possible the easy removal of the end plug of the inner capsule and all specimens and spacers were easily removed and identified. The inside dosimeter wires of each capsule were removed and carefully sectioned. Each piece corresponded to a definite segment of the fueled material.

The fueled and nonfueled cylinders were photographed at magnifications of 4, 6, and 12 diameters. These are shown in Figures 15 and 16 and can be compared with the preirradiation photos shown in Figures 1 and 2. This comparison will show that there was no observable change in the fueled specimens. It must be remembered that Specimens RD-1-6 and RD-1-15 were cut and used as top and bottom fission-heat retainers in each capsule. The rough edge visible on the RD-1-6 top semipiece in Figure 15 was caused by this cutting operation and not by irradiation. Any surface imperfection noted on the fueled specimens was directly attributable to handling of the specimens either before or after irradiation. In the case of the nonfueled specimens, the only defect noted was in Specimen S-2. This defect was in the form of a hairline crack. An enlarged view of the cracked area is shown in Figure 16. It is believed that this crack resulted from handling rather than from exposure to the conditions of irradiation.

Tables 4 and 5 are summaries of the postirradiation data. Each length was measured twice, and the diameter was measured at eight different sections. The 1-in.-long fueled specimens decreased in length but expanded slightly diametrically. The net effect was a slight decrease in density for three of the six fueled specimens. The nonfueled spacers did not show any consistent increase or decrease.

It should be noted that the burnup data reported in Table 4 are the result of radiochemical analyses. In addition, estimates of the fission burnup of the top and bottom specimens in each capsule were derived from the estimated reactor flux and also the capsule-temperature data. The curves representing these data are reproduced in Figure 17. If a comparison of all three burnup values is made, it is interesting to note that in every case but the bottom specimen in Capsule BMI-20-2 (RD-1-14) the results of radiochemical analyses fit within the uncertainty bands established for each set of curves. Burnup was also obtained from dosimeter analyses and is plotted in Figure 17 as circled points. These points are not in good agreement with the results of radiochemical analysis. This fact raises the question as to which method gives the most accurate value and by so doing points out the need for more study in the area of burnup determinations.

Because it was impossible to obtain samples from the irradiation specimens for preirradiation metallographic examination, metallography was performed on samples of similar material as was discussed in the preceding section of this report devoted to fabrication. Typical microstructures of unirradiated fueled material are shown in Figure 18. Although this material was nominally $\text{ZrH}_{1.65}$ with 2 w/o enriched uranium, the structure appears to contain a second phase of another composition as well as some porosity.

Figure 19 illustrates the structure of the end of the fueled hydride specimen (RD-1-9) which was adjacent to Specimen RD-1-6 (the top semipiece in Capsule BMI-20-1). The burnup analysis reported in Table 4 for Specimen RD-1-9 was taken from Specimen RD-1-6, which was adjacent to the portion of Specimen RD-1-9 used for metallography. Figure 20 illustrates the structure of the bottom specimen in Capsule BMI-20-1 (Specimen RD-1-11) while Figures 21 and 22 illustrate the structures of the top and bottom specimens, respectively, from Capsule BMI-20-2 (Specimens RD-1-12 and RD-1-14). In every case, the structures are those of a single-phase material of high density. In no case was either the second phase or the porosity noted in the unirradiated material (Figure 18) present in the specimens. It is quite possible that the 100 to 134-day exposure, during irradiation, to hydrogen at temperatures ranging from 900 to 1400 F resulted in further hydriding and densification of the material. However, the hardness data of the irradiated specimens in Table 1 are too scattered to show the effect conclusively.

A close examination of the photomicrographs which make up Figure 22 will show the presence of microcracks concentrated near the outer edge and near the internal hole. However, the basic structure is the same as the other specimens examined. Although these cracks could have been caused by irradiation damage, a careful study of their location in the specimen and of the other specimens examined leads one to conclude that they are more likely the result of high stresses which were set up during fabrication. This being the case, either the thermal cycling during irradiation or handling could have caused the actual cracking.

HC2698, HC2699 and HC2700

Capsule BMI-20-1

HC2697, HC2696 and HC2695

Capsule BMI-20-2

FIGURE 14. APPEARANCE OF CAPSULE BMI-20-1 AND BMI-20-2 AFTER IRRADIATION

6X

Specimen RD-1-6
(Top Sempiece)

HC2706

4X

Specimen RD-1-9

HC2701

6X

Specimen S-3
(Unfueled Spacer)

HC2704

4X

Specimen RD-1-13

HC2702

FIGURE 15. SPECIMENS FROM CAPSULE BMI-20-1 AFTER IRRADIATION

6X

HC2707

Specimen S-6
(Unfueled Spacer)

4X

HC2703

Specimen RD-1-11

6X

HC2705

Specimen RD-1-6
(Bottom Semipiece)

FIGURE 15. (Continued)

4X

HC2709

Specimen RD-1-15
(Top Semipiece)

12X

HC2710

Specimen RD-1-12

6X

HC2714

Specimen S-10
(Unfueled Spacer)

4X

HC2713

Specimen RD-1-16

FIGURE 16. SPECIMENS FROM CAPSULE BMI-20-2 AFTER IRRADIATION

6X

HC2712

Speciman S-2
(Unfueled Spacer)

12X

HC2713

Crack in Specimen S-2

4X

HC2708

Specimen RD-1-14

6X

HC2716

Specimen RD-1-15
(Bottom Semipiece)

TABLE 4. SUMMARY OF POSTIRRADIATION DATA OF IRRADIATED
FUELED SPECIMENS OF $ZrH_{1.65-2}$ w/o URANIUM

Capsule	Specimen	Specimen Location in Capsule	Change, per cent			Average Center-Line Temperature ^(a) , F	Uranium Burnup, a/o	Irradiation Time, days
			Length	Diameter	Density			
BMI-20-1	RD-1-9 ^(b)	Top	0.0	0.60	-0.9	1140	25.0	100
	RD-1-13	Middle	0.0	0.30	-1.6	--	--	100
	RD-1-11 ^(b)	Bottom	-0.08	0.00	0.0	1200	23.2	100
BMI-20-2	RD-1-12 ^(b)	Top	-0.31	0.0	0.0	978	13.9	134
	RD-1-16	Middle	-0.13	0.40	0.0	--	--	134
	RD-1-14 ^(b)	Bottom	-0.18	0.0	-0.7	1153	13.6	134

(a) Averaged from Cycles 112 to 117 for Capsule BMI-20-1 and Cycles 112 to 119 for Capsule BMI-20-2.

(b) Burnup analysis taken from adjacent fueled end semipiece (RD-1-6).

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TABLE 5. POSTIRRADIATION DATA OF $ZrH_{1.65}$ SPECIMENS

Capsule	Specimen	Location	Change in Property Shown, per cent		
			Length	Diameter	Density
BMI-20-1	S-3	Top	+0.48	+0.45	0.0
	S-6	Bottom	-0.36	+0.15	+0.9
BMI-20-2	S-10	Top	0.0	+1.3	0.0
	S-2	Bottom	0.07	-0.49	0.0

Note: Solid curves denote nominal values; dotted curves define estimated uncertainty bands; circled points represent nominal values derived from dosimeter data

FIGURE 17. ESTIMATES OF THE FISSION BURNUP IN THE TEMPERATURE-MONITORED FUELED SPECIMENS

100X

RM13241

250X

RM13242

500X

RM13243

FIGURE 18. TYPICAL MICROSTRUCTURES OF THE UNIRRADIATED $\text{ZrH}_{1.65-2}$ w/o ENRICHED URANIUM

Some porosity and evidence of a second phase can be seen in these photomicrographs of the control specimens.

250X

Outer Edge at Top

HC2892

250X

Center Hole At Bottom

HC2889

500X

Interior

HC2891

FIGURE 19. TYPICAL MICROSTRUCTURES OF SPECIMEN RD-1-9 AFTER IRRADIATION

Note absence of porosity and second-phase structure.
This specimen was irradiated in Capsule BMI-20-1.

250X

Outer Edge At Top

HC2898

250X

Center Hole

HC2897

500X

Interior

HC2900

FIGURE 20. TYPICAL MICROSTRUCTURES OF SPECIMEN RD-1-11 AFTER IRRADIATION

This was the bottom full-size specimen (adjacent to Specimen RD-1-6, the bottom semipiece) in Capsule BMI-20-1.

250X

Center Hole

HC2902

250X

Interior of Specimen

HC2903

500X

Interior of Specimen 2

HC2904

FIGURE 21. TYPICAL MICROSTRUCTURES OF SPECIMEN RD-1-12 AFTER IRRADIATION

This was the top full-size specimen in Capsule BMI-20-2.

250X

Near Center Hole

HC2907

250X

Near Outer Edge

HC2910

500X

Interior of Specimen

HC2909

FIGURE 22. TYPICAL MICROSTRUCTURES OF SPECIMEN RD-1-14 AFTER IRRADIATION

This specimen was adjacent to the bottom semipiece in Capsule BMI-20-2. The cracks are believed to have occurred from stresses set up during fabrication rather than from irradiation damage.

CONCLUSIONS

The data obtained as a result of this research indicate that the $\text{ZrH}_{1.65-2}$ w/o enriched uranium alloy can withstand burnups on the order of 20 a/o uranium at center-line temperatures up to 1200 F. In the material examined, density and dimensional changes were negligible and specimen integrity was maintained. It can be concluded that the minor amount of cracking which was noted in one specimen was not the result of irradiation damage but rather of stresses caused by thermal cycling and/or handling. The metallographic studies and hardness data could be interpreted to indicate that hydriding continued during the irradiation test. The conditions of temperature and pressure which existed in this capsule were such that hydrides between $\text{ZrH}_{1.8}$ and $\text{ZrH}_{1.9}$ could be produced. It must be emphasized, however, that there are insufficient data to refute or establish the postulation that hydriding continues.

REFERENCES

- (1) Wallace, W. P., Simnad, M. T., and Turovlim, B., "Fabrication and Properties of Uranium-Zirconium Hydride Fuel Elements for Triza Reactors", Fuel Elements Conference Held at Gatlinburg, Tennessee, May 14-16, 1958, TID-7559 (Part I), (August, 1959).
- (2) Vetrano, J. B., "Delta-Phase Zirconium Hydride as a Solid Moderator", BMI-1243 (December 18, 1957).
- (3) Bigony, H. E., Doig, J. R., Jr., and Krause, H. H., Jr., "The Reaction of Hydrogen with Zirconium-1 and -25 w/o Uranium Alloys", BMI-1359 (July 6, 1959).
- (4) Andrews, R., "Solving Conductive Heat-Transfer Problems with Electric Analog Shape Factors", Chem. Eng. Progr., 51 (2), 67F-71F (February, 1955).

GEL/AWH/HHK/AKH/JHS/EMS/RFD:jch