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EFFECTS OF IRRADIATION ON STAINLESS STEEL-CLAD UO_2 PELLETS IN HELIUM OR CARBON DIOXIDE

by

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EFFECTS OF IRRADIATION ON STAINLESS STEEL-CLAD UO_2 PELLETS IN HELIUM OR CARBON DIOXIDE

Gerald E. Lamale, John E. Gates, and Ronald F. Dickerson

Uranium dioxide pellets sealed in Type 316 stainless steel containers with a helium gas were irradiated in helium and in CO_2 in thermal fluxes of the order of 1×10^{13} n/(cm^2)(sec). Cladding-surface temperatures were reportedly between 1200 and about 1800 F. The irradiation was performed at BNL by the Ford Instrument Company.

The hot-cell examination performed by BMI showed that there were no obvious effects of the irradiation on the specimen tested in helium. However, the specimen irradiated in the presence of CO_2 exhibited severe cladding-CO_2 reaction and possible central melting of the UO_2.

Although comparisons between pre- and postirradiation data were difficult because of involved fabrication history of the specimens, the tests did further establish the fact that helium is a satisfactory coolant gas for stainless steel cladding material at a temperature of 1200 F. The data obtained from the specimen tested in the presence of CO_2 indicate that at temperatures in the range of 1600 to 1800 F Type 316 stainless steel is not compatible with CO_2.

INTRODUCTION

To obtain engineering data for the design of a closed-cycle gas-cooled-reactor gas-turbine power plant, the Ford Instrument Company irradiated stainless steel-clad UO_2 pellets in both helium and CO_2 atmospheres. After the irradiation in the Brookhaven National Laboratory reactor, the specimens were given a cursory examination in the BNL hot-cells. These examinations indicated that a more detailed study of the specimens was in order, and other facilities were required. General Atomic shared the interest of the Ford Instrument Company in the data to be obtained from the test specimens and requested an examination of the material in the Battelle Hot-Cell Facility.

This report reviews briefly the preirradiation and irradiation history of these specimens, using data reported in some detail by Ford Instrument Company(1,2), and discusses the results of the postirradiation examination performed on Specimen 2, irradiated in helium, and Specimen 8, irradiated in CO_2.

(1) References at end of report.

PREPARATION OF THE FUEL SAMPLES AND ENCAPSULATION

The 5.4 per cent enriched UO_2 was fabricated by the Norton Company into cylindrical pellets varying in diameter between 0.420 and 0.424[3] in. at a nominal length of 0.5 in. The pellets were formed by cold compacting followed by sintering to a density greater than 92 per cent of theoretical. The cladding consisted of tubes of Type 316 stainless steel having either a 5- or 10-mil wall thickness. The inside diameter of the cladding varied from 0.425 to 0.430 in. Comparison of the UO_2 pellet diameters and the inside diameters of the tubing shows that the diametrical clearance of the pellets within the tubing could vary from 0.001 to 0.010 in.

Specimen 2, irradiated in helium, consisted of two oxide pellets in a stainless steel tube having a wall thickness of 0.005 in. End caps were welded to the tubing in such a way that there was a minimum of space in which the pellets could move. Specimen 8, irradiated in a CO_2 atmosphere, also consisted of two UO_2 pellets loaded into a properly sized Type 316 stainless steel tube. The wall thickness of this tubing was 0.010 in. End caps were attached in the same manner as in Specimen 2. In each case, the atmosphere inside the tube was helium. After welding, the specimens were leak tested by a helium mass spectrometer.

The irradiation capsules were made from extruded aluminum tubing about 1 in. in diameter with a 30-mil wall thickness. Fins 1/2 in. high ran longitudinally along the outside of the tubing. One end of the 5-in. capsule was plugged with a 2-in. slug of 2S aluminum which was welded in place. The other end was terminated by an aluminum nipple with a shouldered hole to which 30 ft of 0.25-in. aluminum tubing was brazed. The irradiation specimen was suspended inside the capsule between two conical ceramic centers. The attached aluminum tubing provided a means for both the desired gas and a Chromel-Alumel stainless steel-sheathed magnesium oxide-insulated thermocouple to enter the capsule. The thermocouple bead was fixed to the Type 316 stainless steel cladding by means of a steel clip. See Figure 1 for a cutaway view of the capsule assembly with the contained UO_2 pellets.

The capsules were loaded into the reactor and evacuated. Helium was charged into the capsule containing Specimen 2 and dry CO_2 was charged into the capsule containing Specimen 8. This process was repeated five times to minimize the presence of contaminants. The helium capsule was finally charged to 4 psig of helium and the CO_2 capsule to 3.5 psig of CO_2. Both capsules were sealed by the control valve so that gas samples could be obtained during irradiation. After this was done, irradiation was initiated.

IRRADIATION

The irradiation of the capsules was conducted as shown in Figure 2. A Brown recorder was used to monitor temperatures and the sample wagon was used to collect aliquots of gas which had been allowed to circulate freely through the tubing and around the outside surface of the stainless steel-clad specimen in the capsule.

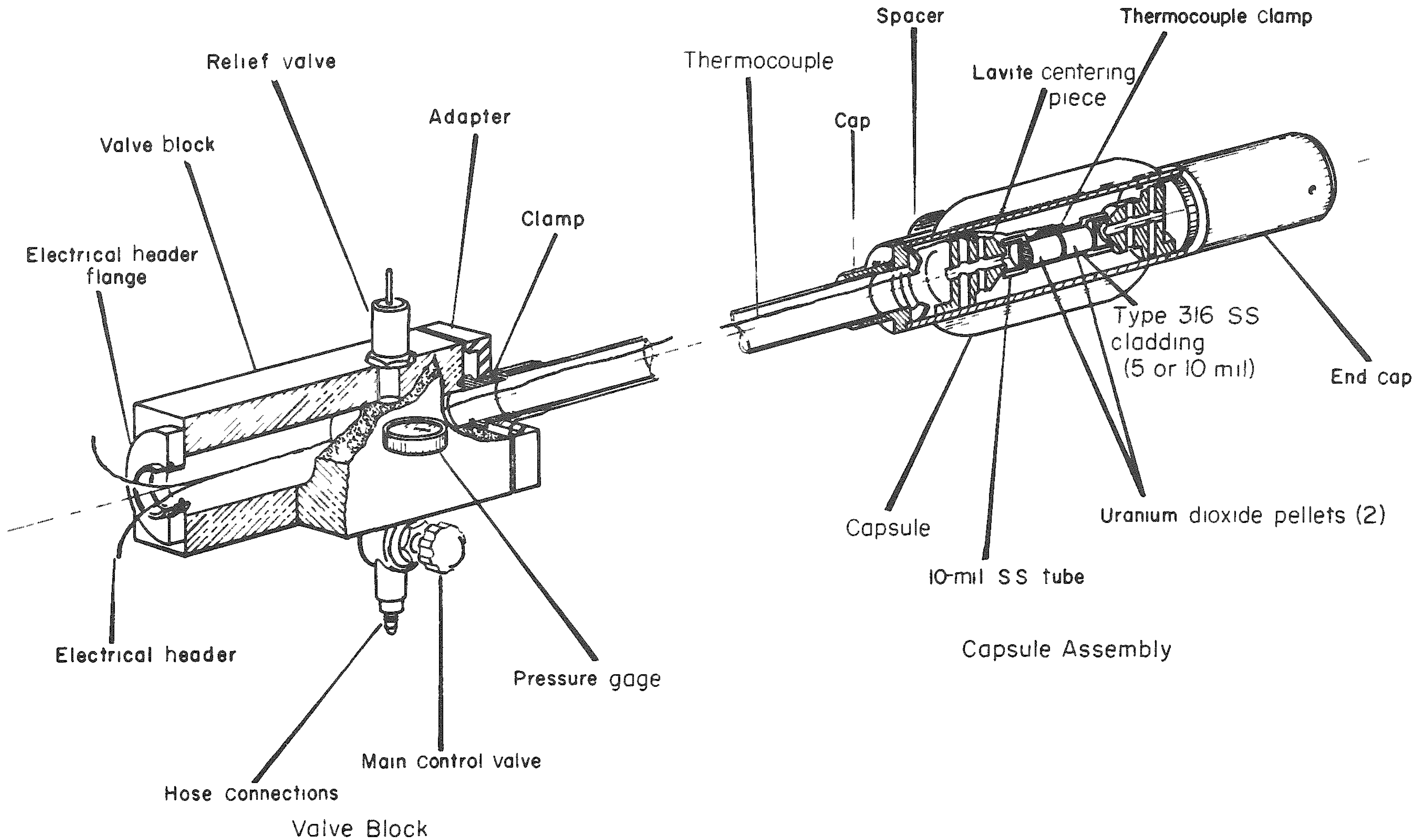


FIGURE 1. CROSS SECTION CAPSULE ASSEMBLY AND VALVE BLOCK

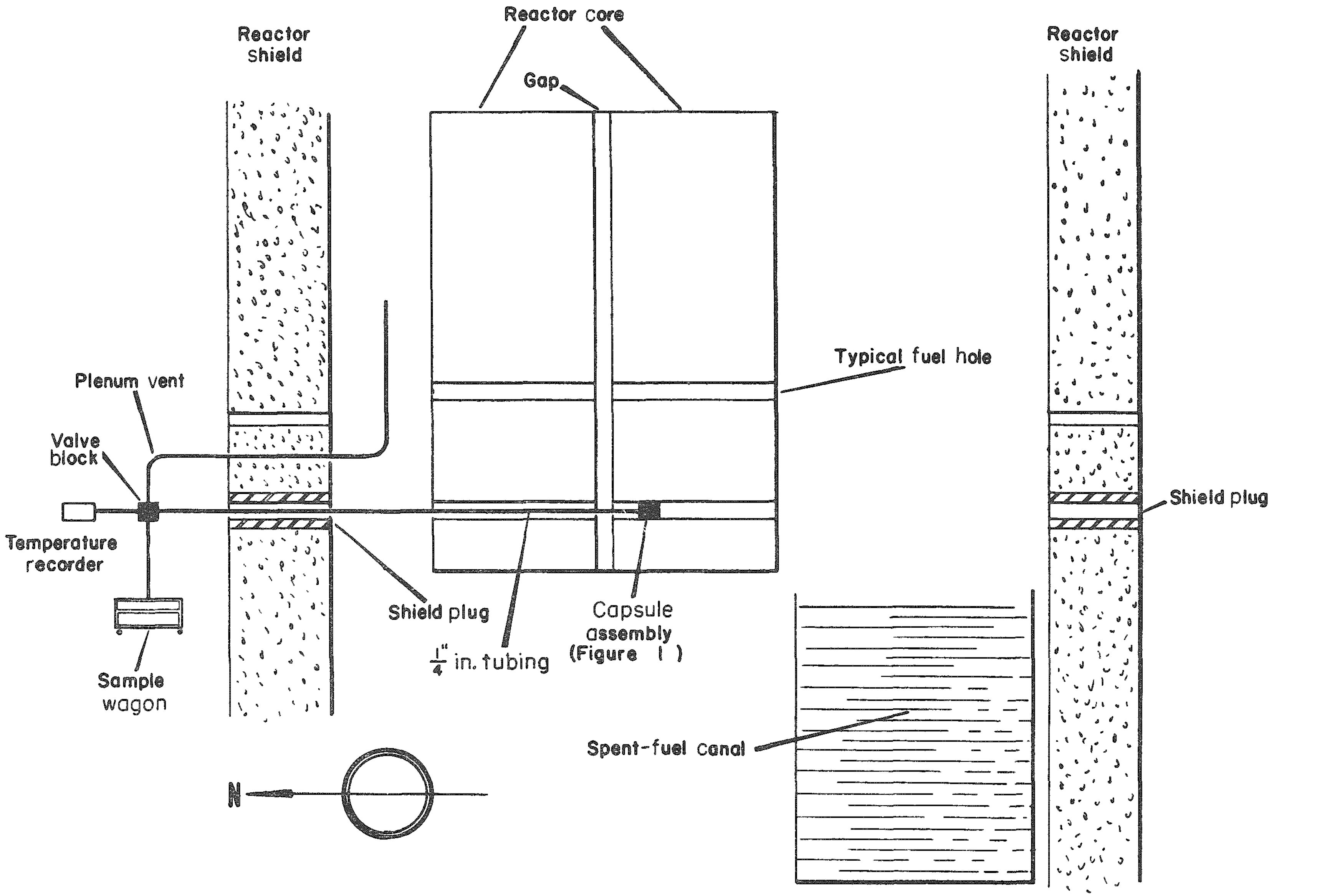


FIGURE 2. ORIENTATION OF PELLET IN REACTOR

Specimen 2 was irradiated in an unperturbed thermal flux of about 6×10^{12} n/(cm^2)(sec) and exposed to an unperturbed integrated flux of 4.7×10^{19} n per cm^2 without any fission products being found in the helium gas surrounding the clad UO_2. The cladding surface temperature during the irradiation was 1200 F.(2)

Specimen 8, encapsulated in CO_2, performed in a different manner. Four hours after startup, the temperature of the cladding was 1800 F and the pressure inside the capsule was 6.5 psig (3 psig higher than at startup). The increased pressure was probably due to the increase in temperature of the CO_2. Fourteen hours after startup, the temperature had decreased to 1700 F and the CO_2 pressure had dropped to the original 3.5 psig. The next day the absolute pressure in the capsule was down to 28 in. of mercury and the temperature was about 1630 F. The capsule was recharged with CO_2 to a pressure of 3.5 psig. Two days later the temperature was about 1580 F and the CO_2 pressure was 0 psig. The next day the absolute pressure had dropped to 22 in. of mercury, a pressure considerably below the pile cooling-channel pressure, and the temperature had decreased to 1500 F. The capsule was again recharged to 3.5 psig.

A sample of the CO_2 was then withdrawn from the capsule and it was found that the gas was highly contaminated. Two days later the capsule was removed from the reactor. Six days had elapsed since startup.

A summary of the data obtained during irradiation of the two specimens of interest as reported by Ford Instrument is presented in Table 1.(2) (In each case the enrichment was 5.4 per cent.)

TABLE 1. IRRADIATION HISTORY OF SPECIMENS 2 AND 8

Specimen	Reactor Channel	Unperturbed Thermal Flux(a), n/(cm^2)(sec)	Unperturbed Integrated Flux, n per cm^2	Estimated Burnup of Fuel(a), MWD/T	Fission Products Detected in Gas	Cladding Temperature, F	Remarks
2	C-11-1	6×10^{12}	4.7×10^{19}	960	None	1200	Helium atmosphere, 5-mil cladding
8	C-6-1-1/2	1×10^{13}	5.2×10^{18}	110	Gross activity after 4 days	1600-1800	CO_2 atmosphere, 10-mil cladding

(a) The burnups as analyzed and the effective fluxes calculated from these results are listed in the section entitled "Evaluation of Data and Conclusions".

POSTIRRADIATION EXAMINATION

A cursory examination in the BNL hot cells showed that the specimen which was irradiated in the CO_2 atmosphere (Specimen 8) had swollen and that a ridge had formed around the specimen approximately in the center adjacent to one edge of the thermocouple clip. The stainless steel appeared spongy and the ridge was cracked in several places, indicating reaction with the CO_2. On the other hand, the specimen irradiated in a helium atmosphere (Specimen 2) showed no apparent swelling or other deleterious effect of the irradiation.

Since further examination could not be accomplished at the time in the BNL facilities, the specimens were shipped to BMI for more detailed study. Upon arrival at Battelle, the specimens were examined visually, measured, and then cut open. The pellets were also examined visually and measured. The results of the subsequent detailed examinations of the specimens are discussed below.

Specimen 8

In the process of attempting to remove the thermocouple clip from Specimen 8, the cladding broke completely through at the peripheral ridge (see Figure 3a). The flare in the cladding can be noted in the portion of the specimen at the right of the photograph. One of the UO_2 pellets can be seen protruding from the broken cladding. It is interesting to note that the area of exaggerated swelling, which provided the plane of weakness for fracture of the cladding wall, occurred at the interface between the two fuel pellets.

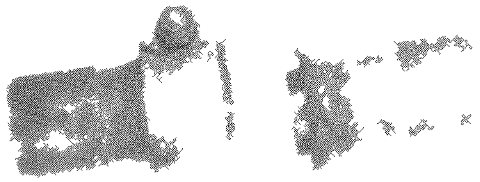
The two halves of the specimen were stood on end and photographed. Figure 3b shows the end of the specimen to which the thermocouple clip was still attached. The end surface of the UO_2 pellet observed in this photomacrograph is that surface which was adjacent to the other pellet in the specimen. Figure 3c illustrates the mating surface of the adjacent UO_2 pellet which remained in the other half of the broken specimen. No serious cracking can be noted.

As the pellet illustrated in Figure 3c was loose in the cladding and easily removable, it was removed, turned over, and replaced in the cladding. The opposite end of the pellet thus exposed is shown in Figure 3d. Aside from slight edge chipping which could well have resulted from handling during any of the experimental steps, the surface is comparable to an as-fabricated specimen of UO_2. A comparison of the two photographs (Figures 3c and 3d) indicates some change occurred in the UO_2 in the plane of the swollen cladding.

Both of the pellets were removed from the specimen and measured (Table 2). One pellet was cut transversely into two halves with a dry cutoff wheel. The half which appeared unaffected by the irradiation was dissolved and analyzed radiochemically for cesium-137. The other half was longitudinally sectioned and the longitudinally cut face was examined by metallographic techniques. The other pellet was removed, measured, and cut. However, it crumbled while cutting and it was not used for metallography because of the irregularity of its pieces.

The results of the radiochemical analyses indicated that the burnup of the pellets in Specimen 8 was about 0.009 per cent of the total uranium. The results are based on the separation and identification of cesium-137 by pulse-height analysis and by the determination of the residual uranium content by spectrophotometric techniques.

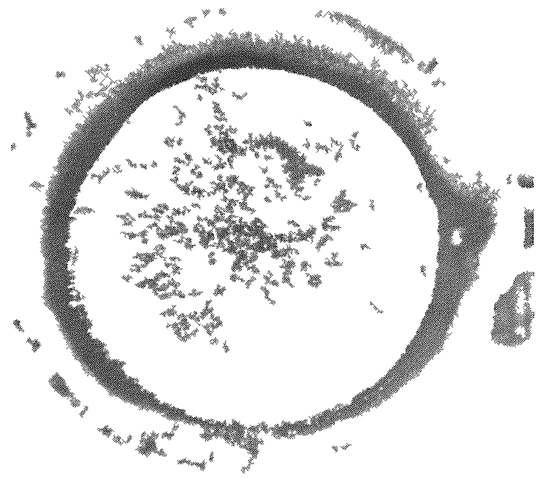
The metallographic section was polished and examined. The central portion of this section was porous, having the appearance of a shrunken pipe. Immediately adjacent to this area was a band of dense material having large columnar grains. This band phased into a UO_2 structure which had some porosity and a grain size considerably larger than that found in UO_2 compacts. It appeared to be a thoroughly sintered structure. Figure 4a illustrates a section of the specimen showing the central pipe, the columnar-grain region, and the sintered area near the outer edge of the pipe of the specimen.



1X HC2007

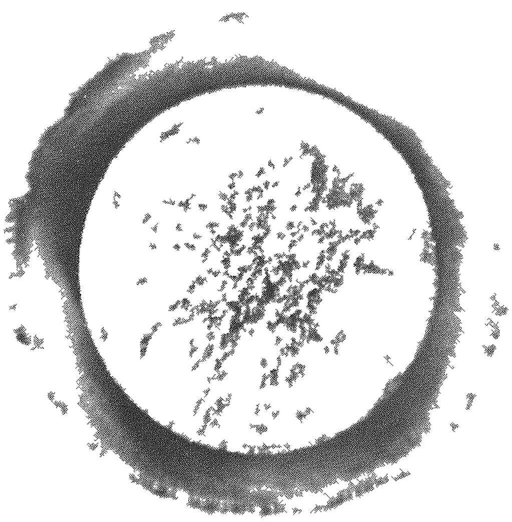
a Specimen 8 With the Attached Thermocouple Clip

Note flare in cladding and protruding UO_2 pellet



6X HC2009

b Surface of the UO_2 Pellet in Section With Clip Attached



6X HC2010

c End Surface of UO_2 Pellet in Section Opposite Thermocouple Clip

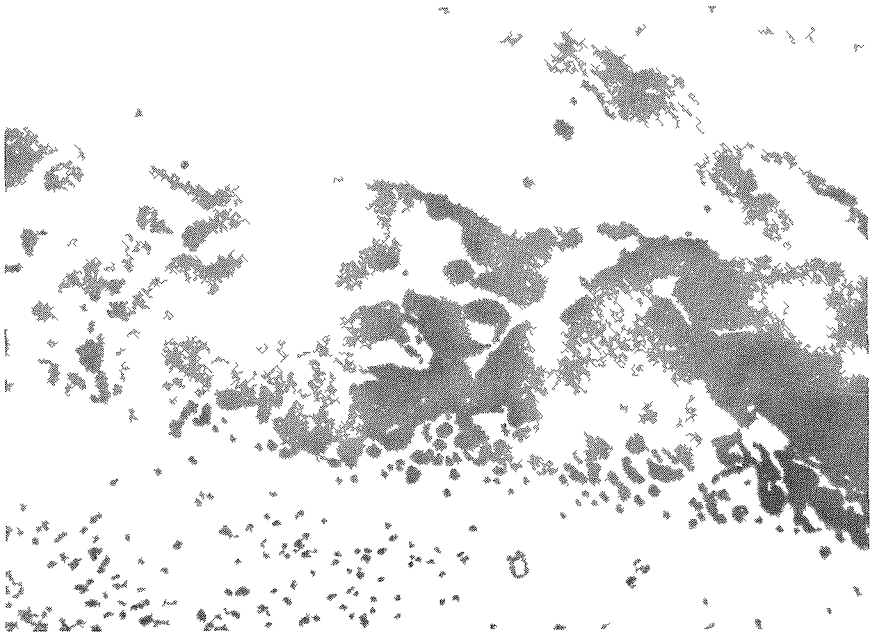
6X HC2011

d Opposite End of UO_2 Pellet Illustrated in Figure 3c

FIGURE 3. SPECIMEN 8 AFTER IRRADIATION IN CO_2 ATMOSPHERE

TABLE 2. POSTIRRADIATION MEASUREMENTS OF SPECIMENS 2 AND 8 AND THE UO_2 PELLETS CONTAINED

Specimen	Cladding Thickness, mils	Component Measured	Dimensions, in. Diameter	Dimensions, in. Length	Remarks
2	5	Clad specimen, as received	0.4520	1.9732	--
		UO_2 pellets	--	--	Pellets fractured during attempts to remove cladding
8	10	Clad specimen, as received	--	--	Severe localized swelling made measurement useless
		UO_2 pellet	0.4254	0.5300	Pellet from end with thermocouple clip
		UO_2 pellet	0.4250	0.5294	--



50X HC2184

a Central Pipe Columnar-Grain Region and Sintered Area

250X HC216[illegible]

c. Typical Finer Grained Edge Area

250X HC2185

b. Typical Area Within Columnar-Grain Region

FIGURE 4. MICROSTRUCTURE OF UO_2 PELLET FROM SPECIMEN 8

Figure 4b illustrates a typical area within this columnar region. The shape of the voids nearest the center region further suggests that this region may have been plastic. It is interesting to note that for the most part the material appears quite dense.

Figure 4c illustrates the structure of the outer region illustrated in Figure 4a. The porosity present is for the most part porosity that probably existed in the material prior to irradiation. This grain size is larger than the preirradiated grain size. To obtain a qualitative idea of the extent of grain growth which occurred, Figures 4b and 4c should be compared with Figure 6b. Although the structure shown in Figure 6b is typical of the UO_2 specimen irradiated in helium, it also represents the grain size one might expect to exist in UO_2 fabricated by the techniques used for the test specimens.

The macroscopic examination of Specimen 8 showed the porous nature of the Type 316 stainless cladding in the region of the cladding break. Microscopic study showed that the steel had been oxidized severely by the exposure to the CO_2 atmosphere. Figure 5 illustrates a typical area.

Specimen 2

The cladding of Specimen 2 showed no visible effect of irradiation in the helium atmosphere. Figure 6a is a photograph of the clad specimen. There was no evidence of gross swelling, and no difficulty was experienced in removing the thermocouple clip. The clad specimen was measured (Table 2) and then was cut to remove the UO_2 pellets. In opening the specimen, one of the UO_2 pellets was cut transversely near the outer end and random cracking observed. The pellet fragments would not drop out of the cladding, however, and further cutting operations were necessary to furnish material for burnup analyses and metallography. After the portions for metallography and burnup were cut, it was noted that they were cracked. This cracking may have resulted from handling. Figure 6b illustrates the structure of the UO_2 from Specimen 2, and shows the end of one of the cracks typical of those observed in this pellet.

The results of the radiochemical burnup analysis showed that 0.05 per cent of the total uranium in Specimen 2 was fissioned.

Metallographic examination of the 5-mil stainless steel cladding of Specimen 2 showed no effect of irradiation in the helium atmosphere. The structure was typical of unirradiated Type 316 stainless steel and because of this is not illustrated in the report.

EVALUATION OF DATA AND CONCLUSIONS

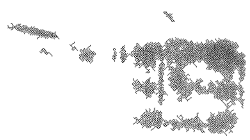
Dimensional Measurements

Evaluation of the dimensional data of Table 2 is difficult because the only pre-irradiation measurements are in the form of manufacturing tolerances. Based on the diameter variations of manufacturing tolerances, and comparing the diameter measurement after the cladding thickness is subtracted, the diameter of the pellets from Specimen 2 increased from 4 to 5 per cent. On the other hand, actual measurements of

250X HC2157

FIGURE 5. AN AREA OF THE 10-MIL-THICK TYPE 316 STAINLESS STEEL CLADDING OF SPECIMEN 8 NEAR THE RUPTURED REGION

Porosity of region is clearly evident.



1X HC2008

a. (Above). Specimen 2 After Removal of Thermocouple

Paper clips illustrate size of specimen.

b. (Right). Typical Structure of Pellet From Specimen 2

The cracks are typical of those observed in this pellet.

250X HC2186

FIGURE 6 FULL-SIZE VIEW OF SPECIMEN 2 PRIOR TO SECTIONING AND ILLUSTRATION OF TYPICAL UO_2 STRUCTURE the pellets from Specimen 8 indicate the diameter increased from 0.3 to 1.3 per cent for one pellet and from 0.2 to 1.2 per cent for the other. The data presented are far too few for use in drawing any conclusions on the possible swelling of UO_2 during irradiation.

Radiochemical Burnup Analyses

The results of an evaluation of the burnup data are shown in Table 3. The calculated effective flux was about one-half or one-third that of the unperturbed flux listed in Table 1. The heat-generation rate shown in Table 3 was calculated from the known number of fissions and the known length of time of irradiation. The application of the burnup data to each specimen is discussed below.

TABLE 3. IRRADIATION CONDITIONS CALCULATED FROM RADIOCHEMICAL ANALYSES

Specimen	Burnup, total a/o	Burnup of Fuel, MWD/T	Calculated Effective Flux: Incident, $n/(cm^2)(sec)$	Calculated Effective Flux: Integrated, n per cm^2	Heat-Generation Rate[(a)], $Btu/(hr)(ft^2)$
2	0.0164	396	2.05×10^{12}	1.61×10^{19}	35,950
8	0.003	71	5.48×10^{12}	2.85×10^{18}	97,700

(a) Heat flux emerging from the lateral surface of the pellets.

Specimen 8

Using the calculated heat-generation rate and the recorded cladding temperature, possible temperatures for Specimen 8 have been calculated at the surfaces of the inside of the cladding and of the UO_2 pellets. Similar calculations were made of the temperatures at the center line of the UO_2 pellets to determine whether central melting could have possibly occurred. The thermocouple junction was attached to the clip and was about 10 or 20 mils removed from the cladding surface and about 1/4 in. off the center transverse plane. The temperature of the cladding at the midplane position may have been 50 to 100 F higher than the record shows. Possible temperature differences resulting from any corrosive effects of the gases on the thermocouple itself were neglected in the calculations which are detailed below.

The general equation of temperature distribution in a long cylinder when the steady state has been reached is

$$\frac{k}{r}\left(\frac{d}{dr}\left[r\,\frac{dt}{dr}\right]\right) = -q \quad ,$$

where

k = thermal conductivity, Btu/(ft)(hr)

r = distance from center to point in question

q = heat-generation rate, $Btu/(ft^2)(hr)$

t = temperature.

After integrating the above differential equation and evaluating the constants of integration in terms of the physical problem, the resulting equation for the temperature within the specimen is

$$T = T_a + \frac{q}{4k}(a^2 - r^2) \quad ,$$

where

T_a = the temperature at the outer surface

a = distance to the outer surface.

The temperature difference from the center line to the outer surface is simply

$$T - T_a = \frac{q}{4k} a^2 \quad .$$

For this particular problem, the temperature difference was calculated as 2040 F, using a thermal conductivity of 0.85 Btu/(hr)(ft) for UO_2.

The other formula for conduction of heat through cylindrical annuli is the familiar

$$\Delta T = \frac{H \ln\left(\frac{r_o}{r_i}\right)}{2\pi k} \quad ,$$

where

H = the heat flux, Btu/(hr)(ft^2)

r_o = the outer radius

r_i = the inner radius

k = the thermal conductivity of the particular medium, Btu/(hr)(ft).

The heat per unit length transferred by radiation is

$$\frac{H}{\ell} = \sigma \epsilon_{eff} A_1 (T_1^4 - T_2^4) \quad ,$$

where

ℓ = unit length

ϵ_{eff} = the effective emissivity, $\frac{1}{\frac{1}{\epsilon_1} + \frac{1}{\epsilon_2} - 1}$, where ϵ_1 is the emissivity of the UO_2 and ϵ_2 is the emissivity of stainless steel

A_1 = unit area

T_1 = surface temperature of UO_2

T_2 = surface temperature of stainless steel

σ = Stefan-Boltzmann constant, 0.174×10^{-8} Btu/(ft^2)(hr).

An assumed value of 0.4 for the emissivity of UO_2 at a temperature of 1000 F was used. Because of the corroded surface of the stainless steel, and the uncertainty as to just when in the 6-day irradiation period the stainless steel surface actually became corroded, two different emissivities, 1 and 0.1, corresponding to the emissivities of a highly polished surface and a highly corroded surface at 1800 F, were used. Table 4 is a summary of the calculations and is meant only to indicate possible center-line temperatures.

The center-line temperatures were calculated for the situation existing before and after failure of the cladding. From the irradiation history, the highest cladding temperature was recorded immediately after reactor startup. Helium was more than likely still present within the specimen at this time, and the calculation shows that for a highly polished surface the center-line temperature was 3680 F, well below the melting temperature of UO_2. By the fourth irradiation day fission gases were detected in the coolant, indicating a break in the cladding with the possibility of CO_2 replacing helium in the gas bond region. By this time the material was certainly thoroughly sintered in the 2600 to 4000 F range and densified, allowing a pipe to form. After the CO_2 replaced the helium around the pellets, the center-line temperature of the pellets probably increased due to the lower thermal conductivity of CO_2 and could possibly have exceeded the melting temperature. The microstructures illustrated in Figures 4a and 4b seem to indicate melting; however, the structure might occur as a result of special sintering conditions as noted above.

Obviously, all of the necessary parameters are not sufficiently known to place complete confidence in these calculated temperatures. Nevertheless, these calculations coupled with the metallography do indicate that the center of the specimen was in a range where melting could have occurred.

From the experience gained with Specimen 8, it can be concluded that Type 316 stainless steel is not compatible with CO_2 in an irradiation field at temperatures in the range of 1800 F. Under these conditions, the stainless steel offers no appreciable resistance to reaction with the CO_2 gas or its decomposition products. Its strength is obviously drastically reduced, and gas may pass easily through the porous structure resulting from the reaction. It was calculated that there was sufficient iron present in the highly corroded area of the cladding to combine chemically with all of the CO_2 (about 0.0093 mole) contained in the system. Nickel oxides are also known to form at this temperature and in the same gaseous environment. The net effect would be observed as a decrease in carbon dioxide pressure, and was thus observed during the experiment.

Specimen 8 probably failed in the following manner: It appears that as the cladding material at temperatures of 1800 F or above reacted with the oxygen available from the decomposition of CO_2, the cladding was weakened, and the sealed static helium gas and accumulated fission gas under a pressure of about 5 atm pushed the cladding outward. This progressed until cracks formed in the cladding and allowed the helium and fission gases inside to escape and the carbon dioxide to diffuse in around the UO_2 pellets. The diameter of the bulged portion of Specimen 8 increased to about 0.60 in., an increase of about 40 per cent, and metallographic examination showed that the wall thickness in this midplane area increased to about twice its original size.

TABLE 4. CALCULATED CENTER-LINE TEMPERATURES FOR SPECIMEN 8 AT VARIOUS STAGES IN IRRADIATION PERIOD

Estimates are based on a cladding temperature of 1800 F and a heat-generation rate of 97,700 Btu/(hr)(ft^2).

Assumed Condition of Stainless Steel Surface	Assumed Emissivity of Component Surface		Estimated Specimen Center-Line Temperature at Indicated Condition(a, b), F			
				Cladding Broken and All Components of Specimen Exposed to CO_2		
	Stainless	UO_2	Cladding Still Intact; Minimum Pellet Diameter and Maximum Cladding ID	Minimum Pellet Diameter and Maximum Cladding ID	Minimum Pellet Diameter and Minimum Cladding ID	Maximum Pellet Diameter and Maximum Cladding ID
Bright	1.0	0.4	3680	4040	3960	3980
Corroded	0.1	0.4	3960	5040	4540	4640

(a) Size differentials were obtained by using a coefficient of linear expansion of 11.1×10^{-6} per F for stainless steel and the extrapolated range of reported [Reference (4)] minimum values of linear expansion for UO_2. These data indicate that UO_2 increases in diameter by 1.43 and 2.19 per cent t calculated average temperatures corresponding to 3145 F and 3660 F, respectively.

(b) Dimensional values:

	Maximum	Minimum
Pellet Diameter, in.	0.424	0.420
Cladding ID, in.	0.430	0.425

Specimen 2

In the case of the helium-atmosphere specimen, there was no evidence that central core melting occurred and there was no apparent effect of the irradiation on the UO_2 structure. Because of the difficulties encountered in removing the pellets from the cladding, it was difficult to determine the extent of the cracking which resulted from the test and that which resulted from handling. The fact that the pellets stuck in the cladding tube raises some question about the dimensional stability of the UO_2 pellets; however, it must be emphasized that the lack of preirradiation data makes it impossible to reach any definite conclusion of this point.

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GEL:JEG:RFD/mmk