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MULTISPECIMEN HOT PRESSING OF UO_2 *

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ASTRACT

A procedure for multispecimen hot pressing of urania disks was developed as a preliminary step in manufacturing $(U_{0.8}Pu_{0.2})O_2$ fuel plates for the Zero Power Plutonium Reactor (ZPPR). Hot pressing is a technique which is generally used to produce a small number of pieces, and is feasible for large production only if several specimens can be pressed at one time. The technique is amenable to close dimensional control and produces specimens having a fine grained structure.

Pressings of five specimens each were made at 1400, 1500, and 1625°C, and one pressing of eleven specimens was made at 1625°C. Density variation of specimens within one pressing was approximately 0.5%. Specimen densities over the range of temperatures were from 76.2 to 88.9% of theoretical. Plate thickness was maintained within ± 0.005 of an inch, and the diameters were maintained within a smaller tolerance. Carbon pickup was from 165 to 508 ppm on the as pressed specimens, which, after a hydrogen anneal at 1550°C, contained from 25 to 44 ppm carbon. Photomicrographs are presented to show the difference in reaction of the UO_2 with two grades of graphite mold.

Hot pressing is a fabrication technique used to simultaneously press and sinter powders. It produces specimens uniformly fine grained and provides dimensional control, not easily attained by the cold pressing and sintering method. Normally, the hot pressing technique is used when very few specimens are required. Production of a large number of pieces is feasible only if several specimens can be produced in one pressing. The objective of this work was to develop a technique for multispecimen hot pressing.

Hot pressing of uranium dioxide has been studied by other investigators. Murray, et al.[1], found that hyperstoichiometric oxide could be hot pressed to densities equivalent to those of stoichiometric oxide at lower temperatures. A titanium carbide nickel-bonded die was used in another investigation[2] to press UO_2 at 800°C at 10 tsi. This procedure gave somewhat better dimensional

* Work performed under the auspices of the U.S. Atomic Energy Comission.
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control than the conventional graphite dies at 1400 to 1800°C. These investigations were not concerned with producing large numbers of specimens.

The hot press apparatus used consisted of an induction heated furnace mounted in a hydraulic press. A cross section of the induction coil, insulation, and die assembly is shown in Figure 1. Pressure and power were regulated manually, and the temperature was measured by means of an optical pyrometer. Argon was used to blanket the die assembly in some runs.

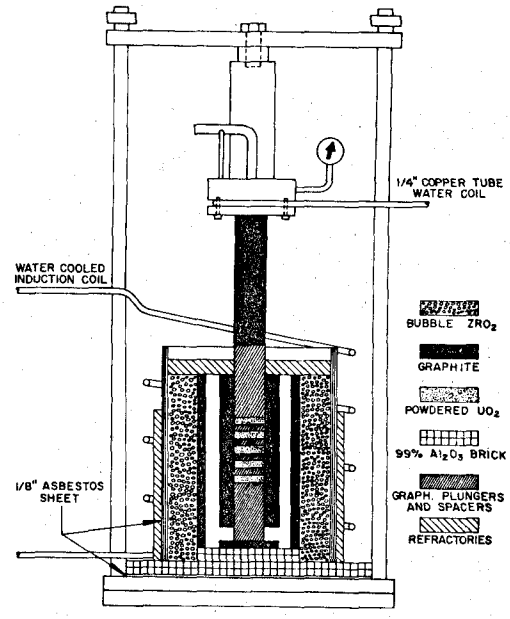

Figure 1. Cross Section of Hot Pressing Assembly.

The die-susceptor assembly, punches and spacers were machined from both CS and ATJ grades of graphite. Of the two grades, CS is softer, coarser grained, and has lower strength than the ATJ. Liners made from filter paper were used on the punch and the spacer faces to reduce sticking.

Five specimens per pressing was chosen because this number would provide a specimen at the center of the stack. Thus the

specimens could be evaluated in relation to position, and the results would have meaning if the number of specimens per pressing were increased.

The ceramic grade UO_2 was supplied by a commercial vendor. Oxygen-to-uranium was 2.13, determined by oxidizing to U_3O_8; fluorine content was 100 ppm; and Fisher average particle size was 1.3μ.

The procedure for loading the die was as follows. The bottom plunger was inserted into the mold leaving a cavity depth of one inch. The UO_2 powder was charged into the cavity over a paper liner. After the powder was leveled, a paper liner and a graphite spacer were inserted and a 550 kg load was applied to force the composite down to a level so another composite could be formed. This procedure was repeated until sufficient powder had been charged for the number of specimens desired. A final load of 820 kg was applied to the assembly before placing it in the hot-press furnace. The initial pressure in the furnace was slight, but it was increased with time and temperature, as shown in Figure 2 for a typical pressing cycle to 1625°C. Three pressings of five specimens each were made at temperatures of 1400, 1500, and 1625°C; the relationships of time, temperature, pressure, and subsidence were similar in each to those shown in Figure 2.

Geometric densities were determined on each of the specimens. Thicknesses were the averages of five determinations and the diameters of two. Table I lists densities as a function of stack position for a pressing at 1625°C. Average densities for pressings at the three temperatures are shown in Table II.

The density profile of specimens in the stack is skewed toward the top, as a result of using a single action press. The profile probably would be symmetrical if a double action press were used. Possibly this would also reduce the spread among the densities. Other pressings at 1625°C had a density spread and pro

file similar to the one listed. A single pressing of eleven specimens at 1625°C produced a density spread similar to the spread observed in one of the five specimen stacks. Although the densities of pressings at 1400 and 1500°C were below the range of interest, they were within a stack similar to those at 1625°C.

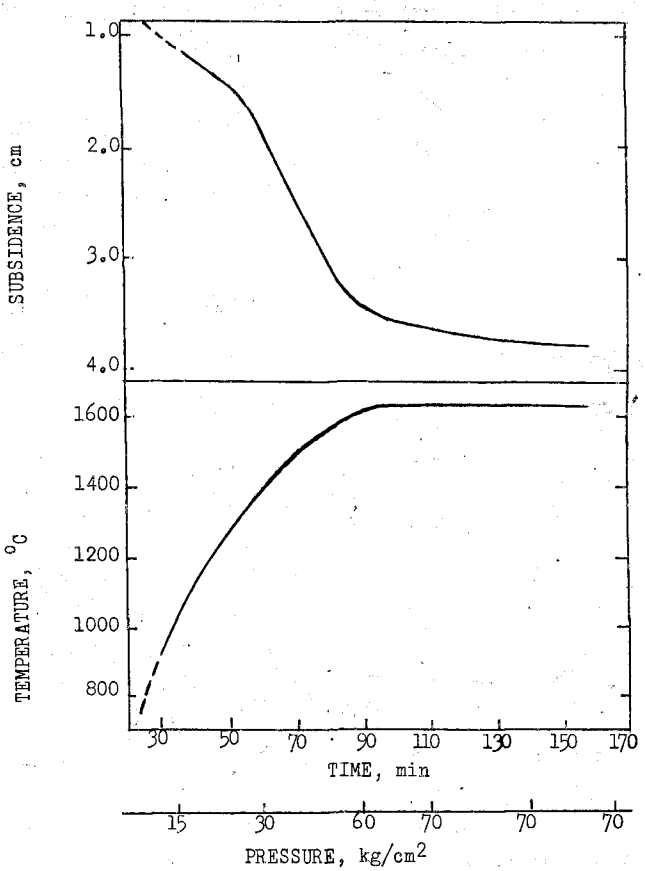

Figure 2. Typical Hot Pressing Cycle.

TABLE I DENSITY VERSUS POSITION IN STACK

Position	Density (% Theoretical)
Bottom 1	87.97
2	88.61
3	88.97
4	89.61
Top 5	89.24

TABLE II AVERAGE DENSITY VERSUS TEMPERATURE

Temp. (°C)	Density (% Theoretical)
1400	76.21
1500	81.40
1625	88.88

The diameter of specimens was determined by the die, consequently, very little variation of this dimension was observed. The thickness, however, was a function of powder weight, uniformity of loading, and ultimate density achieved. The standard deviations of thicknesses for specimens pressed at the three temperatures are listed in Table III.

TABLE III THICKNESS STANDARD DEVIATIONS

Temp. (°C)	Standard Deviation (cm)	Relative Standard Deviation (%)
1400	0.0085	1.2
1500	0.0095	1.8
1625	0.008	1.4

If the ultimate density at 1625°C is known, the thickness can be controlled within 1.4%.

Since carbon and UO_2 react in the temperature range of the pressings, carbon pickup was a concern. Consequently, both metallographic examination and chemical analyses were made on selected specimens. Portions of the same specimens were annealed in H_2 at 1550°C for one hour to evaluate this method for removing the carbon. Visually, as-pressed specimens appeared blacker on the edges and near the circumference of the faces. After annealing in H_2, this effect was absent and the specimens appeared uniformly dark, reddish brown. The apparent carbon pickup was more prenounced on specimens pressed in CS-grade graphite than those pressed in ATJ grade.

Photomicrographs of fragments of a specimen pressed in a CS-grade graphite die at 1625°C are shown in Figure 3, as-pressed, and Figure 4, H_2-annealed. The small, rounded, opaque inclusions which are uranium carbide are distributed uniformly throughout the as-pressed specimen. The H_2-annealed fragment contained only a few nonuniformly distributed inclusions of the carbide. A photo

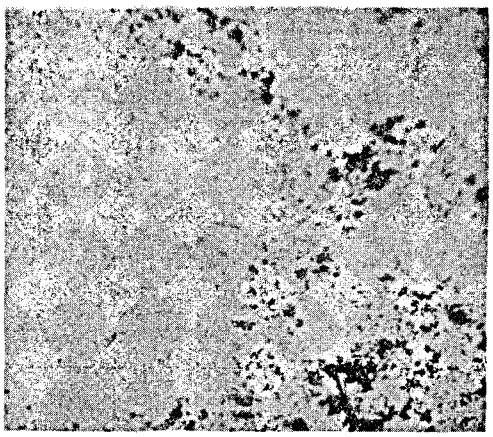

Figure 3. Uranium Dioxide Pressed at 1625°C in CS-Grade Graphite Die. As-polished. White inclusions are uranium carbide. (800X)

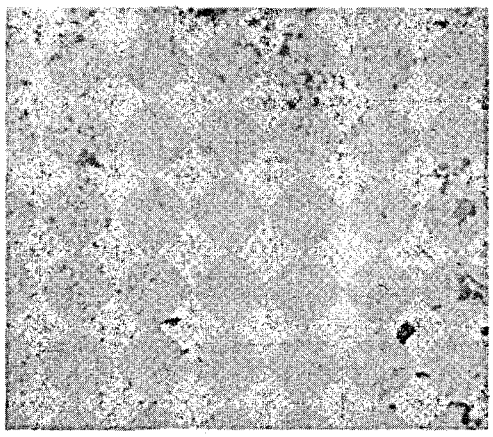

Figure 4. Uranium Dioxide Pressed at 1625°C in CS-Grade Graphite Die and Annealed in H_2 at 1550°C. Taken from same specimen shown in Figure 3. (800X)

micrograph of a specimen pressed in ATJ-grade graphite at 1625°C is shown in Figure 5. Again, carbide inclusions are present, but fewer in number and less uniformly distributed. The distribution had no consistent relation to the UO_2-graphite interface. Specimens pressed at 1500 and 1400°C had no carbide inclusions. There was no microscopically detectable effect of an argon blanket on specimens pressed at 1625°C in ATJ-grade graphite.

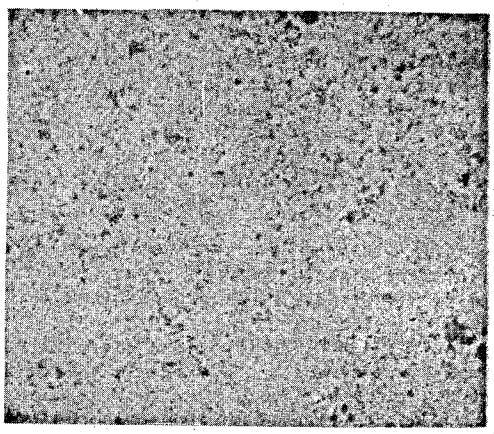

Figure 5. Uranium Dioxide Pressed in ATJ-Grade Graphite at 1625°C. White inclusions are uranium carbide. As-polished. (800X)

Chemical analyses for carbon were made on specimens pressed in CS-grade graphite at 1625 and 1500°C. Specimens, as-pressed and H_2-annealed, from the top and middle of the stacks were sampled and the results are listed in Table IV.

TABLE IV CARBON ANALYSES OF UO_2 SPECIMENS

Designation (°C)		Carbon Content (ppm)	
		As-Pressed	H_2-Annealed
Top	1625	193	25
	1625	221	26
	1500	508	35
Middle	1625	212	44
	1625	165	32
	1500	357	30

The results indicate no correlation between specimen position in stack and carbon pickup. The higher carbon content of the 1500°C specimens is surprising in view of the absence of carbide observed microscopically. Carbon content was reduced substantially by the H_2-anneal in each specimen. The reduction results from the presence of a small amount of H_2O in the H_2. At this temperature, uranium monocarbide does not react with pure H_2.

RESUMO

Foi desenvolvido um processo para a compactação simultânea, à quente, de múltiplos discos de urânia, como uma fase preliminar de fabricação de placas combustíveis de $(U_{0,8}Pu_{0,2})O_2$ para o Reator de Plutônio de Potência Nula ("Zero Power Plutonium Reactor" - ZPPR). Compactação à quente é uma técnica geralmente usada para produzir pequeno número de peças e torna-se praticável para produção em grande escala sòmente se vários espécimes podem ser prensados ao mesmo tempo. Tal técnica possibilita rigoroso contrôle dimensional e produz espécimes tendo estrutura de granulação fina.

Compactações de 5 espécimes cada, foram feitas a 1400, 1500 e 1625°C, tendo sido realizada a 1625°C uma prensagem de onze espécimes. A variação de densidade dos espécimes em cada operação de compactação foi de aproximadamente 0,5%. A densidade dos espécimes no intervalo de temperatura considerado variou entre 76,2 a 88,9% do valor teórico. A espessura das peças foi mantida com variações dentro de ± 0,005 de polegada, sendo que os diâmetros satisfizeram tolerâncias ainda menores. Contaminação por carbono variou entre 165 e 508 ppm nos espécimes analisados após a compactação, passando para 25 a 44 ppm depois de recozimento a 1550°C, em hidrogênio. Apresentam-se micrografias para mostrar a diferença de reatividade do UO_2 com duas qualidades de matrizes de grafita.

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