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***Encapsulation of ThO_2
in Molybdenum Liners
Via Hot Pressing***

W. D. Pardieck

October 11, 1973

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MOUND LABORATORY

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Encapsulation of ThO₂ in Molybdenum Liners Via Hot Pressing

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ABSTRACT

Encapsulation of ThO_2 powder in molybdenum liners using hot pressing techniques was demonstrated. ThO_2 was used as a simulant for $^{238}\text{PuO}_2$. Five discs were hot pressed in an effort to establish die design and pressing parameters. Dies were designed to receive cups made of 0.005-in. or 0.031-in. molybdenum. Pressing temperatures of 1850°C and 1450°C were tried. A ThO_2 disc of 88% theoretical density was successfully hot pressed inside a cup fabricated from 0.031-in. molybdenum. The disc was pressed at 1850°C and 3800 psi.

INTRODUCTION

Mound Laboratory has been interested in developing a modular type of heat source* which would consist of capsulettes stacked together to form a module. Each capsulette, as a separate entity, would be capable of surviving post-impact oxidation. Schematics of a module and a capsulette, as conceptualized by J. E. Selle and J. R. Wysong, are shown in Figures 1 and 2. The inner section of the capsulette would consist of a fuel disc pressed into a tungsten liner. This concept has created interest in the encapsulation of $^{238}\text{PuO}_2$ inside tungsten liners via hot pressing techniques. Initial studies were made using ThO_2 as a simulant for $^{238}\text{PuO}_2$, and molybdenum instead of tungsten for fabricating the metal

FIGURE 1 - Preliminary conceptual design of Mound's modular type heat source unit.

*"Modular Heat Source Concepts" was a project in the Mound Laboratory Value Analysis/Engineering Seminar in May of 1969.

liners. These preliminary studies were made in an effort to define die design and to produce several encapsulated simulant fuel discs that could be used for impact testing of the capsulette.

METHOD OF HOT PRESSING

The discs were hot pressed using a Vacuum Industries hot press sintering furnace, capable of exerting up to 30 tons of force on compacts at temperatures up to 2000°C in a vacuum of 10^{-5} torr. A photograph of the hot press is presented in Figure 3. The double-acting ram, pedestal, heat zone, die, and high-frequency induction heating coil are contained in the horizontal water-cooled stainless steel chamber. The press structure supports the chamber and hydraulic pump, as well as the vacuum pumping system with its controls.

FIGURE 2 - Preliminary conceptual design of Mound's capsulette.

FIGURE 3 - Overall view of hot press manufactured by Vacuum Industries, Inc.

A photograph and schematic showing the hot press rams, chamber, and heat zone assembly are presented in Figure 4. The graphite die, 4 in. in diameter, 3.75 in. high, and fitted with graphite punches, rests on graphite spacers. The spacers are used to transfer the force from the water-cooled rams to the die punches. The punches have smaller diameters than the spacers. Therefore, the die punch lengths are sized so that the spacers come into contact with the die body preventing further densification when the desired compact densification has been accomplished. Grade AXF graphite purchased from POCO Graphite Inc. was used to fabricate the die body, punches, and spacers. The remainder of the heat zone assembly rests upon the water-cooled pedestal. The graphite susceptor is used to position the die. Temperature measurements are made with a disappearing filament pyrometer sighted on a hole through the top and inside of the susceptor. The susceptor is supported with a graphite stool piece. The stool piece and susceptor are machined from Union Carbides' grade ATJ graphite. Three layers of $\frac{1}{4}$ -in. thick graphite felt are used to insulate the top and sides of the assembly. A quartz cylinder around the outside of the heat zone prevents contact between the graphite felt and induction heating coil. Compact densification is measured with a dial indicator fastened to the top ram.

The charged die is loaded into the heat zone assembly. The top spacers and insulation are positioned. An initial clamping pressure of 1850 psi is applied to the die punches. After the chamber is evacuated to 5×10^{-5} torr, the die is heated to the proper temperature in approximately 1 hr. Die pressure is increased slowly as the temperature increases, reaching maximum pressure and temperature simultaneously. The temperature is maintained for 10 min after ram movement indicates the die is closed. The motor generator is turned off and the pressure is reduced to 1100 psi. The die cools in vacuo to ambient temperature in 3 hr. After the die is removed from the heat zone, three cuts, each 120° apart, are made in the die body to allow it to be separated from the punches and disc. The punches are then easily separated from the disc.

DISCUSSION AND RESULTS

Four discs were hot pressed using graphite dies, molybdenum cups, and die liner materials as shown in Figure 5. The molybdenum cups were hydroformed from a 0.005-in. thick molybdenum sheet. The molybdenum lids were machined to size using the same material. The dies, constructed with a step on the upper punch allowing the punch and cup lid to extend into the molybdenum cup as the ThO_2 powder is compacted, were designed to produce ThO_2 discs of 88% theoretical density upon die closure. A die charge of 140 g of 53-105 μm ThO_2 powder was required. The graphite dies were lined with tantalum, molybdenum, and graphite foils. Tantalum and molybdenum foils were used to cover the punch faces and die cavity walls. The molybdenum foils were placed next to the molybdenum cup. The tantalum foils in contact with the graphite dies were used as diffusion barriers to reduce graphite penetration of the molybdenum cups. Sticking of the compacts to the punches was prevented by placing grafoil 0.010-in. thick between the punch faces and tantalum foils.

FIGURE 4 - Photograph and schematic of hot press chamber, rams, and heat zone assembly.

FIGURE 5 - Schematic of die used to hot press ThO_2 powder inside 2.128-in. diameter cup. Cup was hydroformed using 0.005-in. thick molybdenum sheet.

The initial pressing (STO-1) was made using graphite die and punches lined with 0.001-in. tantalum and 0.001-in. molybdenum foils. The disc was pressed at 1850°C using 4000 psi pressure. Upon removal from the die, the side of the molybdenum cup broke into several pieces, leading to the assumption of graphite penetration. The lid and bottom of the cup remained intact.

In an effort to reduce the penetration of graphite, the second die was lined with 0.005-in. tantalum and 0.005-in. molybdenum foils. Also, the temperature and pressure were reduced. The pressing parameters of 1450°C and 2600 psi pressure did not close the die. A ThO_2 disc (STO-2) that was 70% of theoretical density was produced. The ThO_2 disc tended to flake and crumble not adhering to the molybdenum cup.

Two discs (STO-3 and STO-4) were pressed using die and foils similar to STO-2. These two discs were pressed at 1850°C and 3800 psi pressure. They appeared identical upon removal from the dies. Figure 6 is a photograph of STO-4 showing the side and top. The side of the molybdenum cup remains intact and extends above the lid. However, the side and bottom of the cup did not remain in one piece as shown in Figure 7. The molybdenum fractured around the edge of the cup at the 0.088-in. radius. The tantalum and molybdenum die liner foils adhered strongly to the molybdenum cup. Removal of liner foils, carbides, or graphite from the surfaces is desirable if the disc is to be encapsulated in another liner. Machining is a possible method of removing the outer surfaces of the molybdenum cup and obtaining dimensional accuracy. Also, the side of the cup extending above the lid could be removed during the machining operation.

FIGURE 6 - Top and side of molybdenum cup, fabricated from 0.005-in. thick molybdenum, in which is encapsulated a hot-pressed ThO_2 disc.

FIGURE 7 - Bottom and side of molybdenum cup, fabricated from 0.005-in. thick molybdenum, in which is encapsulated a hot-pressed ThO_2 disc. Note crack around the bottom edge of the cup.

In an effort to keep the bottom and side of the cup in one piece and to allow sufficient molybdenum metal in order that the outer surface could be removed by machining, the die assembly was redesigned. A schematic of this new die is shown in Figure 8. Three changes were made: first, a 0.031-in. thick molybdenum cup and lid were used; secondly, the radius around the bottom edge of the cup was changed from 0.088 in. to 0.187 in.; and finally, this radius was supported with the lower punch. The die was lined with 0.001-in. tantalum and molybdenum foils. To produce a ThO_2 disc of 88% theoretical density, 132.5 g of 53-105 μm ThO_2 powder was required. The disc was pressed at 1850°C and 3800 psi. Figure 9 is a photograph showing the top and side of the disc as it looked after removal from the die. The side of the molybdenum cup remains intact and extends above the ThO_2 disc and lid. Again, the tantalum and molybdenum foils adhered strongly to the cup. The side and bottom of the cup remained in one piece as shown in Figure 10. The results of this pressing were promising in that the ceramic was enclosed within the molybdenum cup after removal from the graphite die.

A summary of the data obtained on the five pressings is presented in Table 1.

CONCLUSION

Encapsulation of ThO_2 powder inside molybdenum liners via hot pressing was demonstrated. Dies were designed to encapsulate ThO_2 discs in cups made from either 0.005-in. or 0.031-in. molybdenum. Four cups made from the 0.005-in. molybdenum fractured around the bottom edge during the hot pressing operation. Similar fracturing was not observed when a 0.031-in. molybdenum cup was used. Due to the very limited scope of the study, only visual evaluation of the encapsulated discs was made. However, several observations can be stated. Surface scale and the resulting embrittlement of the molybdenum cups constitute a problem. The scale consists of the die liner foils and possibly graphite and carbides. Scale composition and depth of penetration need to be evaluated. Removal of the scale would be necessary to produce a usable product. Machining the cup is a possible method of removing scale and impurities from the surfaces as well as controlling cup dimensions. Use of dies or die liner materials that would not affect the cup should be evaluated. A study of cup reaction with die or die liner materials as a function of pressing parameters would also be of value. However, because the pressing characteristics of ThO_2 are different from $^{238}\text{PuO}_2$, this study would have to be performed with $^{238}\text{PuO}_2$.

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FIGURE 8 - Schematic of die used to hot press ThO_2 powder inside a molybdenum cup of 2.190-in. diam. Cup was hydroformed using 0.031-in. thick molybdenum sheet. Radius around bottom edge of cup was supported with lower punch.

FIGURE 9 - Top and side of cup, fabricated from 0.031-in. thick molybdenum, in which is encapsulated a hot-pressed ThO_2 disc.

FIGURE 10 - Bottom and side of cup, fabricated from 0.031-in. thick molybdenum, in which is encapsulated a hot-pressed ThO_2 disc. Bottom and side of cup remain in one piece.

Table 1

SUMMARY OF DATA OBTAINED ON ENCAPSULATION
OF ThO_2 POWDER IN MOLYBDENUM LINERS VIA HOT PRESSING

<u>Disc Number^a</u>	<u>Die Liner Foils Thickness (in.)</u>		<u>Mo Cup Thickness (in.)</u>	<u>Hot Pressing Parameters</u>		<u>Condition of Mo Cup</u>
	<u>Mo</u>	<u>Ta</u>		<u>Temp. (°C)</u>	<u>Pressure (psi)</u>	
STO-1	0.001	0.001	0.005	1850	4000	Very poor; side broke into pieces.
STO-2	0.005	0.005	0.005	1450	2600	Poor; cup did not adhere to ThO_2 disc; bottom and side did not remain in one piece.
STO-3	0.005	0.005	0.005	1850	3800	Fair; bottom and side did not remain in one piece.
STO-4	0.005	0.005	0.005	1850	3800	Fair; bottom and side did not remain in one piece.
STO-5	0.001	0.001	0.031	1850	3800	Good; bottom and side remained in one piece.

^a Density of ThO_2 discs produced was 88% of theoretical, except STO-2, which had a density of 70% of theoretical.