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THE FABRICATION OF DENSE URANIUM DIOXIDE
CYLINDRICAL COMPACTS BY COLD PRESSING AND SINTERING

J. Glatter
H. R. Hoge
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Contract AT-11-1-GEN-114

October 14, 1955

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METALLURGY AND CERAMICS

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BY COLD PRESSING AND SINTERING

J. Glatter
H. R. Hoge
B. E. Schaner

CONTRACT AT-11-1-GEN-14

October 14, 1955

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DATE MAR 12 1957 *pet*

For The Atomic Energy Commission

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Chief, Declassification Branch

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ABSTRACT

Pressing and sintering characteristics of Mallinckrodt hydrogen reduced UO_2 powder are presented. Punch and die materials and factors influencing punch and die design are described. Density and shrinkage data for sintered UO_2 are given. A graphite resistor sintering furnace is described.

INTRODUCTION

The purpose of this paper is to present empirical information and data which were acquired in the course of fabricating dense UO_2 compacts. The method of fabrication employed was cold pressing UO_2 powder and subsequently sintering in vacuo or argon. This work was undertaken because of the strong consideration accorded to UO_2 as a potential fuel material for nuclear power reactors.

The source of the raw material used in this work was the Mallinckrodt Chemical Works, St. Louis, Missouri. The uranium dioxide powder was produced by the hydrogen reduction of UO_3 . The text reference to this oxide is MCW powder. The suffixes 1-2 and 5-1/5-10 refer to 1951 and 1954 lots of MCW powder respectively.

The theoretical density (T.D.) of UO_2 is taken as 10.95 grams per cc. UO_2 compacts possessing a bulk density in excess of 90% of theoretical density are considered dense.

PRESSING CHARACTERISTICS OF UO_2 POWDER

UO_2 powder can be formed into a coherent mass by compression under very high pressures in a mold of a desired contour. The compaction technique is similar to dry pressing in the ceramic industry and is identical to that employed in the powder metallurgy field.

Fig. No. 1 shows the "as pressed" or green compact density as a function of the compacting pressure. The data for this plot were obtained by pressing a series of compacts of MCW 5-10 powder using a fixed charge of 5.5 grams. The pressure on a 0.400 in. diameter ram was increased from 25 tsi to 162.5 tsi in increments of 12.5 tsi.

UO_2 powder is brittle, friable, and somewhat abrasive in nature. These characteristics account for the high compaction and die ejection pressures which

necessarily had to be employed to overcome the high wall frictional forces encountered in the cold pressing operation. Early experiments revealed that relatively uniform dense compacts could be made by pressing from two directions and by maintaining a length to diameter ratio less than 1.0. Die wall frictional forces play an important part in limiting the L/D ratio to less than 1.0. For example, if a compact is pressed with an L/D ratio exceeding 1.0, there will be a noticeable density gradient between the middle and ends because the high wall friction forces inhibit movement of the powder towards the center of the compact. Sintering revealed evidence of slight density gradients even in the compacts pressed to an L/D ratio of less than 1.0. Without exception, there was always a diametral shrinkage differential of 0.001 in. to 0.005 in. between the middle and ends of the sintered compacts.

The early work also showed that during the pressing operation the compacts commonly exhibited two types of shear failures. One type appeared as a 45° angle cracking at the ends of the compact, extending inward for a small distance and resulting in the lifting off of a thin annulus from both ends of the compact. The employment of very high pressures accentuated the inward 45° cracking and often resulted in cones of material falling out of either one or both ends of the compact. (See Fig. No. 2). The addition of an internal lubricant such as polyvinyl alcohol or camphor to the oxide powder was successful in reducing this type of shear failure. The second type occurred as transverse laminations or fissures (fractures). The elastic expansion of the compact as it was ejected from the die caused this failure. A die with a 0.400 in. bore will produce an expanded compact of 0.405 in. diameter when pressed at 100 tsi. A slight taper on the exit side of the die for a distance equivalent to the compact length provided a more gradual expansion of the compact upon ejection and eliminated the occurrence of fissuring.

Experiments performed using various particle size fractions and combinations thereof, indicated that the "as pressed" density as a function of compacting pressure was unaffected by particle size distribution. Because of the friable nature of MCW powder, fragmentation of all the particles to a basic crystallite size apparently occurred under the high pressures used.

PUNCH AND DIE DESIGN FACTORS AND MATERIALS

In cold pressing UO_2 powder, care must be exercised in the design and selection of materials for punches and dies because of the high load required for densification (i.e., 70%-75% T.D.) in the "as pressed" state.

Dies of high carbon, high chrome-tool steel hardened to R_c 60-65 were used initially. The life of these dies was extremely short because of the drastic scoring of the die wall upon ejection of the compact. During compaction the die expands and when the load is removed contraction of the die around the compact occurs. This action coupled with the abrasive nature of the MCW powder explains the necessity of employing high loads for ejection and the resultant wear of the die wall.

The use of a tungsten carbide lined die reduced the aforementioned wear considerably. Shrink rings were used to place the carbide liners under compression and thus fracture of the liners during compaction was prevented. An added benefit derived from the shrink rings was that the amount of expansion of the die bore during compaction was reduced. This resulted in reduction of the elastic expansion of the compact with the attendant minimization of fissuring. Experience showed that a highly polished surface on the tungsten carbide liner resulted in less wear and reduced both the compacting and ejecting pressures.

The most suitable punch material was found to be tungsten carbide because of its extreme stiffness and high compressive strength. The application of high pressure to hardened tool steel punches produced distortion by expansion. In

using carbide punches, care had to be exercised to avoid sharp corners or cross sectional changes which would result in chipping and breakage.

COMPACTING PRESS

A Stokes 280-G, 100 ton capacity mechanical toggle press equipped with a hydraulic pressure equalizer was used in this work (Fig. 3). The purpose of the hydraulic pressure equalizer was to insure uniform density of the pressed compacts. The press exerts pressure on the compact from above and below by means of a moving die-table which descends during compression at one-half the speed of the upper punch. Handwheel control of the clutch and drive allows the press to be operated manually. The rated output of the press ranges from 7 to 26 compacts per minute.

Fig. 4 shows a carbide lined die, solid carbide punches with steel shoulders shrunk on, and a compact made in the die. The diametral clearance between the punch and die wall is $0.001'' \pm 0.0005''$

The MCW powder as received was not free flowing because it was composed of 50% to 60% of minus 325 mesh particles whose average particle diameter was 4-6 microns. It was converted into free flowing granules by means of a mechanical agglomeration procedure involving the use of one (1) w/o polyvinyl-alcohol.

The availability of free flowing UO_2 granules enabled automatic operational data to be obtained. The following are typical examples of such data:

1. Compacts 0.433" diameter and 67% T.D. were produced at a rate of 8 per minute.
2. Compacts 0.405" diameter and 71% T.D. were produced at a rate of 15 per minute.

SINTERING APPARATUS

Sintering of the cold pressed compacts was performed in a graphite resistor furnace designed and built at the Bettis Laboratory. A photograph of

the furnace and attached accessories is shown in Fig. 5. A schematic diagram of the furnace proper is shown in Fig. 6. Two D. C. plating type generators connected in series served as the power supply. The current was introduced to the graphite resistor by means of water cooled copper rings which were shrunk into place. The resistor is insulated by a layer of granular graphite which is held in place by a thin cylinder of solid graphite. The outer graphite cylinder is insulated from the power leads by a disc of fired lava. There is no insulation between the outer graphite cylinder and the outside metal shell. The outside shell is a section of 12 in. diameter copper pipe which has turns of 3/8 in. copper tubing soldered in place. The 3/8 in. tubing provides water cooling for the furnace. The furnace has a usable hot zone of 2 in. in diameter by 6 in. long. Although there is a great deal of heat lost through the water cooled electrodes, a uniform temperature distribution can be maintained throughout the six (6) inch hot zone.

The furnace can be operated either in vacuo or in a protective atmosphere. A temperature of 1800°C can be obtained in this furnace within 20 minutes.

A graphite boat was employed to contain the cold pressed UO_2 compacts during sintering. Thin compacts of UO_2 served as setting tiles and prevented contamination of the cold pressed compacts by the graphite.

DENSITY AND SHRINKAGE DATA

The first sintering experiments were conducted with MCW 1-2 powder cold pressed to 70% T.D. Compacts from this powder were sintered for 2, 4 and 6 hours at 1500, 1650 and 1800°C in vacuo. The data obtained from these experiments are plotted in Fig. 7. It can be seen that the temperature of sintering has a greater effect on the sintered density than the length of time.

of sintering. For example, at 1500°C, the density increased only 3% as the heating time of the compact increased from 2 to 6 hours, whereas during a two-hour sintering time, the density of the compacts increased 8% when the temperature was increased from 1500 to 1800°C. There is, however, a variation in the sintering characteristics of different lots of MCW powder. UO₂ powder designated as MCW-5-1 was used for a second set of experiments. These results, shown in Fig. 7, indicate that 93% T.D. can be obtained for this powder by heating for 6 hours at 1800°C. All of the compacts used for the experiments which gave the data for Fig. 7 were cold pressed to 70% T.D. using the same dies and punches. Similar results were obtained when the above experiments were performed in an argon atmosphere.

Final or sintered density of UO₂ compacts is also a function of the cold pressed density. Fig. 8 graphically illustrates this point. The data upon which the graph is based were obtained by cold pressing compacts at various pressures to produce the range of initial densities shown. The compacts were then sintered at 1750°C for 6 hours in an argon atmosphere and final densities were measured.

Data pertaining to axial, diametral, and volume shrinkage were obtained on sintered compacts made from MCW 1-2 powder. Fig. 9 shows that a density increase from 70% to 90% results in an 8% axial, 10% diametral, and 26% volume shrinkage of the compacts. Fig. 10 is a photographic illustration of the shrinkage effect in a cold pressed compact which was subsequently sintered.

CONCLUSIONS

Cold pressing and sintering is a usable technique for fabricating dense UO₂ compacts provided that attention is paid to the proper design of

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tungsten carbide punches and dies. Initial densities ranging from 67% T.D. to 71% T.D. in the Mallinckrodt hydrogen reduced UO_2 cold pressed compacts yield high densities (i.e., greater than 90% T.D.) in the sintered compacts.

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Fig. 1 Graph of the "as pressed" or green compact density vs. the compacting pressure.

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Fig. 2 Illustration of conical shear failure in a cold pressed compact.

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Fig. 3 Photograph of Stokes 280-G press

Fig. 4 Photograph of carbide lined die, solid carbide punches (steel shoulders shrunk on), and a pressed compact.

Fig. 5 Photograph of graphite resistor furnace and accessories.

GRAPHITE RESISTOR FURNACE

① WATER COOLING FOR ELECTRODE	⑥ TEFLON	⑪ COPPER POWER CLAMP
② POWER LEAD	⑦ MICARTA	⑫ GRANULATED GRAPHITE
③ COPPER ELECTRODE	⑧ GRAPHITE	⑬ VACUUM CONNECTION
④ WATER COOLED BRASS FLANGES	⑨ COPPER PIPE	⑭ SUPPORT
⑤ SIGHT GLASS	⑩ LAVA	

DWG. NO. 60-A-9875

Fig. 6 Schematic diagram of graphite resistor furnace.

Fig. 7 Graph showing effect of time and temperature on sintered density of compacted UO₂.

Fig. 8 Graph showing effect of cold pressed density on sintered density of UO₂ powder

Fig. 9 Graph showing effect of sintering on shrinkage of UO₂ compacts.

Fig. 10 Photograph of a cold pressed and sintered compact showing the effect of shrinkage.