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URANIA BODIES: FIRED DENSITY VS. PARTICLE SIZE

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ABSTRACT

Urania bodies were prepared from batches of urania powder, as received and as ground for periods of 16, 56, and 140 hours. The particle size distribution of each batch was determined with a double pipet apparatus.

The prepared specimens were examined under a microscope to determine relative porosity, and their densities were measured and plotted.

A trend was noted whereby the density increased and porosity decreased as the particle size decreased. Calcined material yielded a body of higher density than uncalcined material of even finer particle size.

I. INTRODUCTION

Reliable physical property measurements on a substance such as urania require that the material be uniform and homogeneous throughout the entire specimen. The authors felt that a prepared specimen of high purity urania which had a density approaching the theoretical might be essentially a zero porosity body and therefore be best suited for physical property measurements.

II. EXPERIMENTAL PROCEDURES

A. Determination of Particle Size Distribution

1. Method: The particle size distribution was determined with a double pipet apparatus as described by Deshpande and Telang⁽¹⁾ (see Figure 1). The pipet method was chosen because it gives reliable results with a minimum expenditure of time and requires only a moderate amount of skill to operate.

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Figure 1

DOUBLE PIPET APPARATUS

The sample was prepared in the following manner: 50 grams of sample were weighed into a small jar along with 0.5 gram of Daxad #23 (the best dispersing agent found). Three hundred milliliters of distilled water were added, and the jar was sealed. The jar was then packed with sand into a ball mill so that it was rotated end over end when the ball mill was turned on. The sample was dispersed in this manner for 48 hours and then poured into the 1000 ml graduate. The graduate was filled to the 1000-ml mark with distilled water.

The suspension was agitated by hand for 5 minutes to insure homogeneity, and 10 ml were drawn off into a beaker of known weight. This sample determined the amount of urania in 10 ml of suspension at zero time.

The pipets were installed immediately, and 10 ml samples were drawn into them simultaneously at appropriate time intervals. The best time intervals vary with the type of material in suspension but are always taken more frequently at the beginning of the experiment.

The suspension in each of the pipets was drained into a previously weighed beaker, evaporated to dryness and the weight of urania determined. The weight of urania may also be determined by comparing the weight of the entire suspension drawn at any time to the weight of the zero time suspension, but complete evaporation eliminates error due to the liquid if there are slight discrepancies in the levels of the suspension in the pipet as successive determinations are made.

2. Calculations: A plot (Figure 2) of "Velocity versus Particle Size" was drawn. Particle sizes were assumed, and their velocities were calculated from the following relationship:

$$D = \frac{18 n V}{(S_1 - S_2) g}$$

where D = diameter of particle (cm)

V = velocity of particle (cm/sec)

n = viscosity of liquid = 0.008937 poises

S_1 = density of particle = 9.97 gm/cc

S_2 = density of liquid = 1 gm/cc

g = acceleration due to gravity = 980 cm/sec²

Then the time required for particles to fall through a constant distance was plotted (Time versus Particle Size) for four distances, as in Figure 3. Distance in this case is the distance from the top of the suspension to the bottom of the pipet and decreases each time some of the suspension is drawn off.

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Fig. 2 Velocity versus Particle Size

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Fig. 3 Time Required for Particles to Fall Constant Distance

The time required for a particle of any size to fall through any distance is obtained from the simple relationship:

$$d = V \times t$$

where d = distance (cm)

V = velocity (cm/sec)

t = time (secs)

and the velocity of the particle is obtained from Figure 2. The plot of time versus particle size is shown for only a small range of particle sizes. Another set of curves (not shown) was plotted for the larger particles and takes the same shape as Figure 3.

The particle size distribution was plotted as "Equivalent Spherical Diameter in Microns Versus Per cent Finer by Weight," (Figure 4).

By per cent finer by weight is meant $\frac{W_1}{W_2}$, where W_1 is the weight of urania in the zero time sample and W_2 is the weight of urania in a sample drawn off at some given time. The particle diameter at that given time is determined from the plot of Figure 3.

3. Grinding: The batches of urania having different particle sizes were obtained from "as received" urania and from urania ground in a ball mill for periods of 16, 56, and 140 hours.

4. Pressing and Firing: Earlier work conducted on the preparation of high density urania revealed that hydrostatic pressing at 35 tons/square inch and firing at 1750°C for 8 hours resulted in the highest density specimens made from any particular batch of urania. All samples made from batches of urania with a different particle size distribution were therefore prepared by this method.

5. Density Determination: The fired compacts of urania were machined into rectangular prisms, measured with a micrometer to determine the volume, and weighed on an analytical balance. Several prisms were obtained from a single fired compact.

III. EXPERIMENTAL RESULTS

The plot of Figure 4 could not be carried further into the larger particle range with the same accuracy as in the range shown, for in such case the time required to draw suspension into the pipets is appreciable when compared to the velocity of the particles and their relatively low abundance. In the case of as received urania, the distribution could be carried to 10 microns before the same difficulty was encountered.

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Fig. 4 Particle Size Distribution

Experimental data were not used for particles less than 1 micron in diameter, because the free fall of particles below this range becomes appreciably influenced by factors which this experimental procedure does not take into account.(2)

The particle range which is shown in Figure 4 is for that range where results from the two pipets are in complete agreement. For most purposes the plotted lines could be extrapolated in both directions and give satisfactory results.(3)

Figure 5 shows a trend toward increasing density with increasing grinding time. Reference should be made to Figure 4 for the particle size distribution for each of these grinding times.

Photomicrographs of two of the samples (Figures 6 and 7) reveal a marked difference in the number and sizes of the voids. This is indicative of all the samples in the series, i.e., the higher density samples exhibit less porosity which is as was expected.

The high density of the calcined sample is not at all surprising. All calcined batches of urania show an observable amount of shrinkage during calcining; it seems logical to assume that, starting with a batch of "pre-shrunk" urania, less shrinkage should be obtained in the final firing, so that the sample will not pull away from itself to form voids.

Little can be said concerning the form of most dense packing of the particles, as such studies would involve complete segregation of a few discrete particle sizes. The finer the particles, however, the less will be the chance for the formation of voids in the sample; for the small particles will tend to fill up vacancies between larger particles. Nevertheless even those samples of highest density contain many small voids.

IV. CONCLUSIONS

The density of urania specimens which have been hydrostatically pressed and fired to 1750°C in hydrogen increases with increase in the percentage of fine particles. There is a corresponding decrease in porosity even though an appreciable number of small voids exist in the samples of highest density. There is a practical limit to the elimination of voids in a sample of this type, however, and best results can be obtained by using calcined material having the smallest particle size.

Fig. 5 Grinding Time Versus Density

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Figure 6. Porosity of Fired Body

Micro #14447

500X

UO₂ ground 56 hours in ball mill, hydrostatically pressed at 35 tons/square inch and fired at 1750°C for 8 hours in hydrogen.

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Figure 7. Porosity of Fired Body.

Micro #14448

500X

UO₂ ground 140 hours in ball mill, hydrostatically pressed at 35 tons/square inch and fired at 1750°C for 8 hours in hydrogen.

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