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VIBRATORY COMPACTION AND SWAGING OF URANIUM DIOXIDE TO HIGH DENSITY

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VIBRATORY COMPACTION AND SWAGING OF URANIUM DIOXIDE TO HIGH DENSITY

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ABSTRACT

Techniques for the fabrication of ceramic fuel elements, by vibratory compaction and swaging of powders inside a metal tube, have been investigated. Vibratory packing of powders containing three particle size fractions resulted in a maximum density of 9 gm/cm^3, or 82.2% of the theoretical density. It was possible to increase the packed density to 9.9 gm/cm^3, or approximately 90% of theoretical, by cold swaging, and to 10.5 gm/cm^3, or more than 95% of theoretical, by warm swaging at 600°C.

Fabrication of spherical UO_2 shot was investigated. Shot of the desired particle size ranges was produced with densities ranging from 10.2 to 10.5 gm/cm^3, or 93 to 96% of theoretical density.

I. INTRODUCTION

Since fissile ceramic materials appear promising as high temperature nuclear fuels, simple, economical methods of fuel element fabrication are of interest. Vibratory packing and swaging inside metal tubes may offer a possibility of reducing the number of manufacturing steps, eliminating the annulus between fuel and cladding, and reducing tolerances normally required for fuel elements loaded with stacked pellets.

To indicate the feasibility of compacted powders as nuclear fuel, techniques must be studied by which the desired fuel material can be effectively compacted in the desired geometry. The resultant fuel element must have a fuel density comparable to pellet-loaded fuel elements. In addition, the cost of producing such a fuel element must be significantly lower than that of a pellet-loaded element.

The purpose of this investigation is to determine maximum reproducible densities obtained by vibratory compaction of ceramic powders of selected particle sizes. Rotary swaging, as a method of increasing packed density by cladding deformation, is included in this study.

II. PREVIOUS INVESTIGATIONS

Vibratory packing studies conducted by Bell [1,2] on alumina and UO_2 powders indicate that packed densities, in the range of 70 to 80% of the theoretical density, are possible. The maximum densities obtained were 80% of theoretical with alumina powder, and 70% with UO_2. The highest packed densities were obtained under the following conditions:

a) coarse fraction greater than 50 wt %,

b) light pressure under vibration,

c) large amplitude for maximum impact,

d) 2 to 6 wt % water or alcohol used as a wetting agent.

Similar studies, by Kite and Smith [3] on UO_2 powders of two particle sizes, resulted in packed densities of approximately 73% of the theoretical density.

The coarse fraction used was 60/100 (passed through 60 mesh and retained by 100 mesh), while the fine fraction was -325 (passed through 325 mesh). Packed densities reported for mixtures ranging from 50 to 70 wt % coarse fraction and 30 to 50 wt % fine fraction were all between 7.8 and 8.06 gm/cm^3. The density obtained with 100/150 mesh particles as the coarse fraction was significantly lower.

III. CALCULATIONS

The closest possible packing of uniform spheres is described by Dalla Valle[4] as rhombohedral packing, with succeedingly smaller particles used to fill in the resultant voids. Assuming perfect spheres of uniform size, a system containing particles of five sizes having radii in the ratio of $1 : 0.414 : 0.225 : 0.175 : 0.152 = r_1 : r_2 : r_3 : r_4 : r_5$ will pack to 86.4% of the theoretical density. The screen fractions selected to provide particles with average radii in the desired ratio are given in Table I, with the wt % of each size fraction required.

TABLE I

CALCULATED COMPOSITION

Desired Particle Size (μ)	Screen Fraction	Size Range (μ)	% by wt
322.2(r_1)	42/48	350-297	74.5
133.0(r_2)	100/115	149-125	5.3
72.5(r_3)	200/250	74-62	1.7
56.4(r_4)	250/270	62-53	2.9
49.0(r_5)	270/325	53-44	2.0
($-r_5$)	-325	44-0	13.6

IV. PACKING

Sintered UO_2 pellets of 92 to 95% of the theoretical density were crushed and screened to provide the required particle size fractions. Samples of the screen fractions in Table I were prepared. Six compositions were used initially, starting with the coarse fraction 42/48 and adding one succeedingly smaller size fraction for each new mixture.

Each mixture was blended by rotating vigorously in a glass jar until thoroughly mixed, and then transferred to a steel graduate for vibrating. A weighted steel piston, graduated in tenths of cubic centimeters, functioned as the upper floating ram, as well as a volume measuring device. The powder was vibrated on a table vibrator connected to a 50-w amplifier and an audio signal generator until no further settling was apparent. Densities were determined from the weight and bulk volume of UO_2 in the container. A maximum density of 7.78 gm/cm^3 was obtained from a mixture approximating the calculated composition.

Subsequent tests were concerned with efforts to increase the packed density of the calculated composition by varying the vibrational amplitude, frequency, and time, and by adding water or a volatile alcohol.

The most effective type of vibration was determined by varying both amplitude and frequency. It was noted that a jolting effect of large amplitude at frequencies of 10 to 50 cps produced the most efficient packing.

To determine the optimum time of vibration, the coarse size fraction was vibrated for 10 min, with density determinations made at intervals of 10 sec for the first minute, 30 sec for the second minute, and 60 sec thereafter. Packing continued at a significant rate for 3 min and then tapered off. Slight additional density increases were attributed to the tendency of the particles to chip and produce fines (Figure 1).

Additions of either water or a volatile alcohol, in amounts from 1 to 4 wt %, effectively reduced particle size segregation while the powder mixtures were being transferred to the vibrating container. Dusting of the fine powder during vibration was eliminated, and packed densities were increased as much as 2 to 3% when approximately 3 wt % water or alcohol was used.

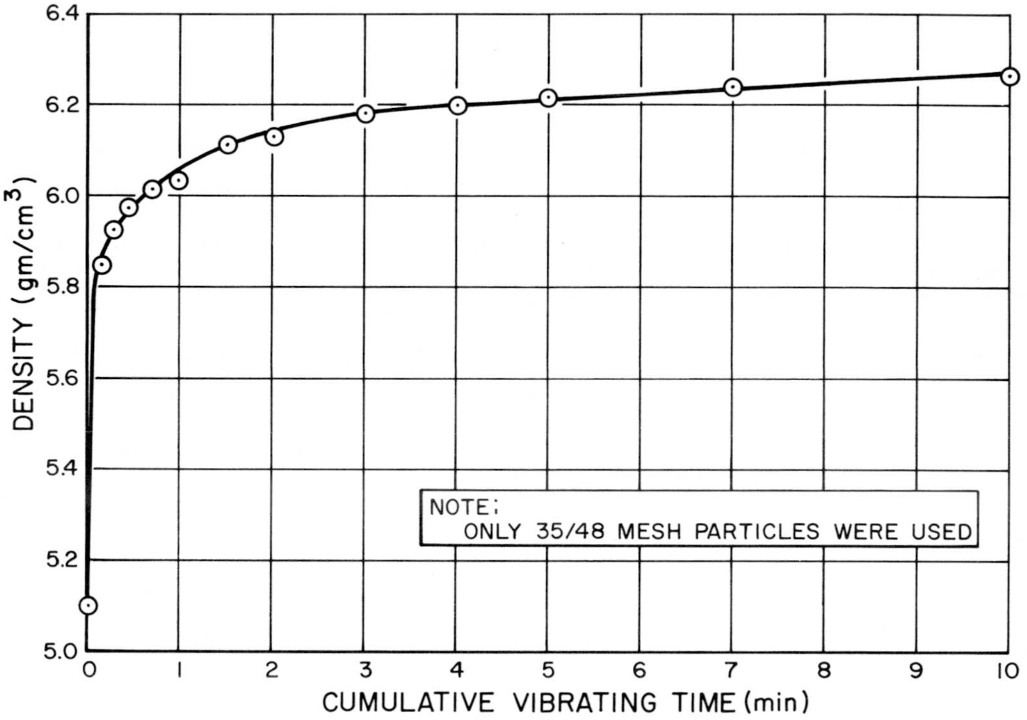


Figure 1. Effect of Time of Vibration on Density

Continued efforts to reproduce densities significantly greater than 8 gm/cm^3, by decreasing the amount of coarse fraction (35/48 and 35/100 mesh), and varying the proportions of intermediate and fine fractions, were unsuccessful. It can be seen, from Tables II and III, that variations in composition had little significant effect, for mixtures composed of 50 to 70 wt % coarse, 0 to 25 wt % intermediate, and 15 to 35 wt % fine fractions. Average densities were 72.8 ± 1.8% of the theoretical density (see Figure 2).

These results are probably due to the random particle size distribution and the corresponding large range of particle sizes within each size fraction. The irregular geometry of the individual particles tends to magnify these effects, since many variations in particle alignment are possible and resistance to packing varies accordingly.

From a geometrical viewpoint, the particle size should have no effect on packed density. In practice, however, fine particles always have lower packed

TABLE II

EFFECT OF SIZE DISTRIBUTION ON PACKED DENSITY

Coarse Fraction 35/100 mesh (wt %)	Medium Fraction 100/325 mesh (wt %)	Fine Fraction -400 mesh (wt %)	Bulk Density (gm/cm^3)	% Theoretical Density
100.0	----	----	6.01	55.0
86.2	13.8	----	6.37	58.1
74.5	11.9	13.6	7.21	65.9
75.0	11.6	13.4	7.78	71.0
67.0	----	33.0	7.94	72.5
66.5	8.5	25.0	7.94	72.5
66.4	9.1	24.5	7.84	71.5
66.2	9.7	24.2	8.09	73.9
65.0	9.9	25.1	7.94	72.5
65.0	9.8	25.2	7.88	72.0
64.0	10.0	26.0	8.00	73.0
63.5	8.1	28.5	8.10	73.9
63.5	8.1	28.5	8.30	75.9*
60.5	9.0	30.5	8.15	74.5
60.0	9.5	30.5	8.56	78.1
60.0	10.0	30.0	8.01	73.1
59.5	9.2	31.2	8.05	74.1
58.9	9.3	31.8	8.06	73.6
58.0	13.0	29.0	7.92	72.4
57.0	17.0	26.0	7.85	71.7
57.0	10.0	33.0	7.98	72.9
56.8	24.2	19.0	7.91	72.4
56.2	24.0	19.8	7.80	71.2
56.0	21.0	23.0	8.09	73.8
55.5	12.0	32.5	8.05	73.6
55.0	18.0	27.0	8.23	75.1
54.1	18.8	27.1	8.10	73.9
52.2	16.6	31.2	8.11	74.0
Coarse Fraction 6/20 mesh (wt %)	Medium Fraction 20/35 mesh (wt %)			
70.0	----	30.0	8.85	80.8
65.0	----	35.0	8.75	79.9
65.0	7.4	27.6	9.00	82.2
63.6	----	36.4	8.70	79.5

* 3 wt % water added

TABLE III

EFFECT OF COMPOSITION ON PACKED DENSITY

Weight (gm) of Specified Size Fraction											Bulk Density (gm/cm^3)	% Theoretical Density (TD = 10.95)
6/8	8/20	20/35	35/48	48/100	100/115	115/200	200/250	250/270	270/325	-400		
			75.4		5.3		1.7	2.9	2.0		6.37	58.1
			75.4		5.3		1.7	2.9	2.0	13.6	7.21	65.9
			61.2				2.8		6.5	23.7	7.94	72.5
			61.2				2.8		6.5	24.5	7.84	71.5
			61.2				1.8		6.5	22.7	7.88	72.0
			65.1		4.6		1.5	2.4	1.7	11.8	7.78	71.0
			65.1		4.6		1.5	2.4	1.7	26.3	8.00	73.0
			65.1		4.6		1.5	2.4	1.7	32.4	8.56	78.1
			39.0		2.6		0.8	1.3	1.0	14.3	8.09	73.9
			39.0		2.6		0.8	1.3	1.0	18.1	8.04	73.4
			60.0		4.0		1.0	1.7	2.0	30.0	8.15	74.5
			66.5		4.2			2.5	1.8	25.0	7.94	72.5
			66.5		4.2			2.5	1.8	30.0	8.10	73.9
			66.5		4.2			2.5	1.8	30.0	8.30	75.9*
			21.0	33.0		15.0		4.0	4.0	18.0	7.91	72.4
			21.0	33.0		15.0		4.0	4.0	20.0	7.80	71.2
			25.0	33.0		15.0		4.0	4.0	24.0	7.95	72.6
			30.0	33.0		15.0		4.0	4.0	30.0	8.09	73.8
			33.0	33.0		15.0		4.0	4.0	33.0	8.10	73.9
			36.0	33.0		15.0		4.0	4.0	33.0	8.23	75.1
			39.0	33.0		15.0		4.0	4.0	33.0	7.85	71.7
			39.0	33.0		15.0		4.0	4.0	38.0	8.11	74.0
			39.0	33.0		15.0		4.0	4.0	43.0	8.05	73.6
			30.0	28.0						29.0	7.94	72.5
			30.0	28.0		10.0				29.0	8.01	73.1
			30.0	28.0		13.0				29.0	7.92	72.4
			30.0	28.0		13.0				34.0	8.05	73.6
			30.0	28.0		10.0				34.0	7.98	72.9
			35.0	28.0		10.0				34.0	8.06	73.6
			37.0	28.0		10.0				34.0	8.05	74.1
70.0										40.0	8.70	79.5
65.0										35.0	8.75	79.9
57.5	12.5									30.0	8.85	80.8
57.5	12.5	8.0								30.0	9.00	82.2

*3% H_2O Added

$$\text{Bulk Density UO}_2 = \frac{\text{weight UO}_2\ (\text{gm})}{\text{bulk volume UO}_2\ (\text{cm}^3)}$$

$$\%\ \text{Theoretical Density} = \frac{\text{bulk density}}{\text{theoretical density (10.95)}} \times 100$$

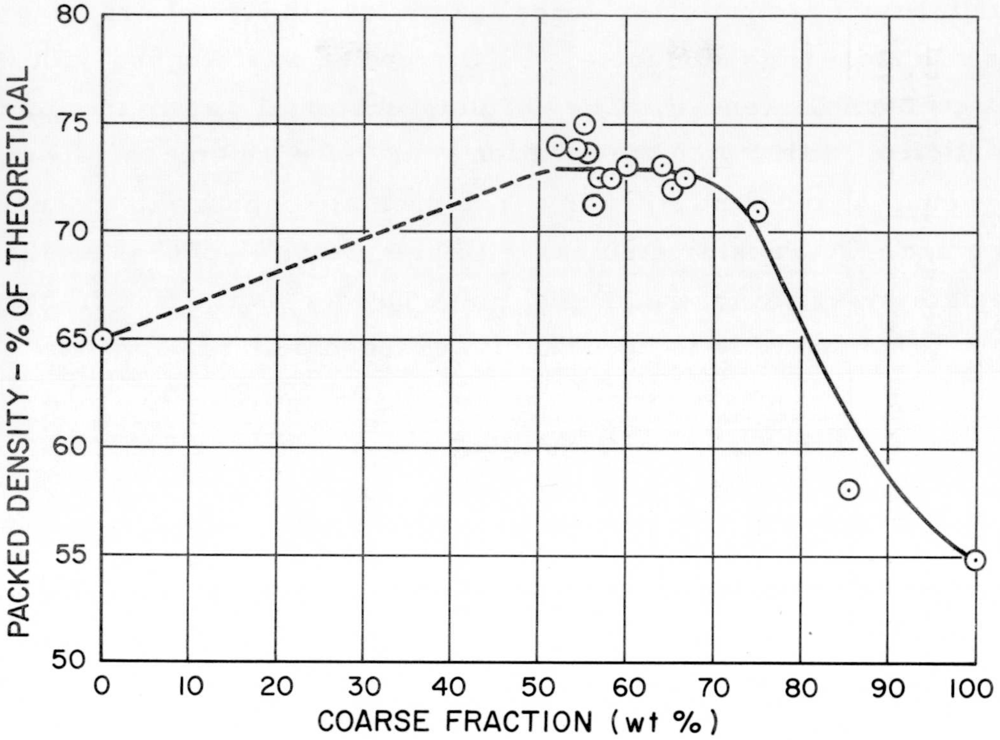


Figure 2. Composition vs Percent Theoretical Density

densities than coarser particles. A maximum reproducible density of approximately 8 gm/cm^3 was obtained for compositions based on 35/48 and 35/100 mesh as the coarse fractions. Since the coarse to fine particle diameter ratio for this system was only about 10 to 1, the possibility of producing higher packed densities by increasing this ratio was indicated. Therefore, additional sintered pellets were crushed to provide particle sizes in the range 6/8 and 6/20 mesh. The use of these size fractions increased the coarse to fine particle diameter ratio to about 50 to 1. As can be seen from Tables II and III, densities increased from 8 gm/cm^3, with 35/48 mesh particles as the coarse fraction, to 9.0 gm/cm^3, with 6/20 mesh particles as the coarse fraction. Compositions with 60 to 70 wt % coarse, 0 to 10 wt % intermediate, and 25 to 35 wt % fines packed to densities of 8.7 to 9.0 gm/cm^3.

The increased densities obtainable with greater coarse to fine particle diameter ratios are probably due to the ease with which fine particles can sift into voids during vibration. Also, there is a corresponding reduction in the number of voids where larger particles are used.

The feasibility of producing spherical particles, in lieu of crushing sintered pellets, was studied. High surface area UO_2 powder was blended with water and a binder in a twin-shell blender. The resultant material was in the form of spherical particles, with a size distribution range of 6/100 mesh. The green "shot" was vacuum dried, and fired in a hydrogen atmosphere to a density of 10.2 to 10.5 gm/cm^3. Packing studies, with high density spheres as the coarse and intermediate size fractions and -400 mesh powder as the fine fraction, produced results comparable to results with crushed pellets.

V. SWAGING

Since densities obtained by vibratory compaction techniques were below required values, rotary swaging was studied as a means of increasing packed density to an acceptable level.

Heavy wall (0.065-in.) stainless steel tubing was swaged in initial tests. Duplicate tubes, 10 in. long, were packed with UO_2 to a density of 8 gm/cm^3. End plugs were inserted and welded. The tubes were evacuated at 600°C. Figure 3 shows the cross section of an unswaged assembly.

The first tube was cold swaged in three passes from 0.750-in. diam to a final diameter of 0.565 in. The tubing wall thickness was decreased by 0.003 in., and density of the UO_2 was increased to 9.9 gm/cm^3. The density variation longitudinally was about 1% (Table IV). Some evidence of sintering at the UO_2-stainless steel interface was noted. The cross section of the stainless steel appeared uniform. No gross differences in cladding thickness were apparent. Figure 4a shows a sample of cold swaged UO_2 with the cladding removed.

The second specimen was hot swaged at 600°C to a final diameter of 0.503 in. in four passes. In

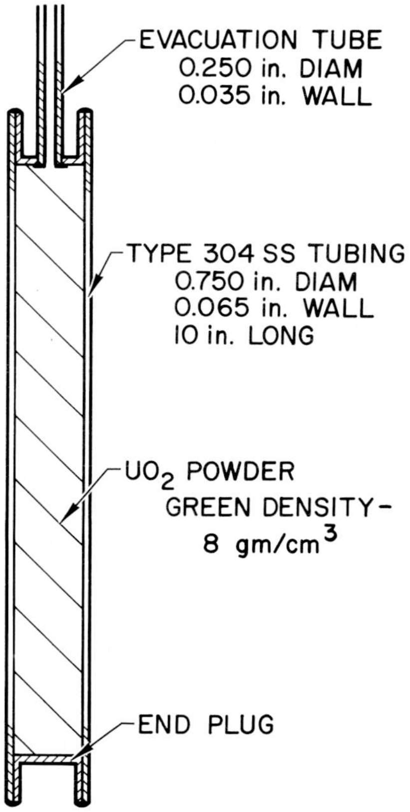
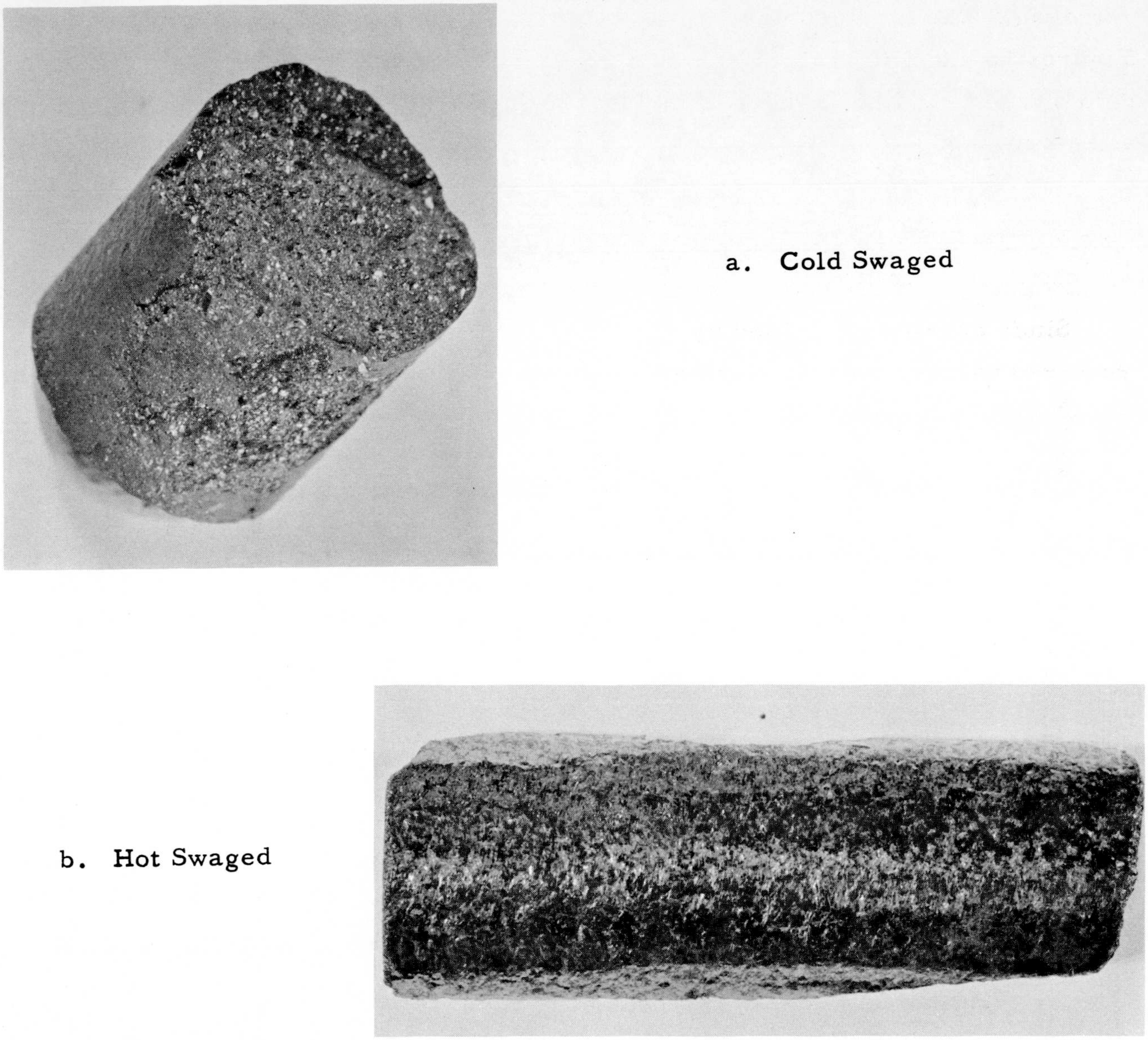


Figure 3. Swaging Assembly



a. Cold Swaged

b. Hot Swaged

Figure 4. Swaged UO_2 with Cladding Removed

this case, the cladding thickness was decreased 0.009 in. and the resulting UO_2 density was 10.4 gm/cm^3. There was definite evidence of sintering in this specimen, with more pronounced effects noted toward the cladding interface. This effect is evident in Figure 4b, showing a hot swaged specimen with cladding removed. The inner surface of the cladding was irregular, as can be seen in Figures 5a and 5b. Table V gives a comparison of the dimensions for unswaged, cold swaged, and hot swaged samples. Figure 6 shows a specimen before and after swaging.

TABLE IV

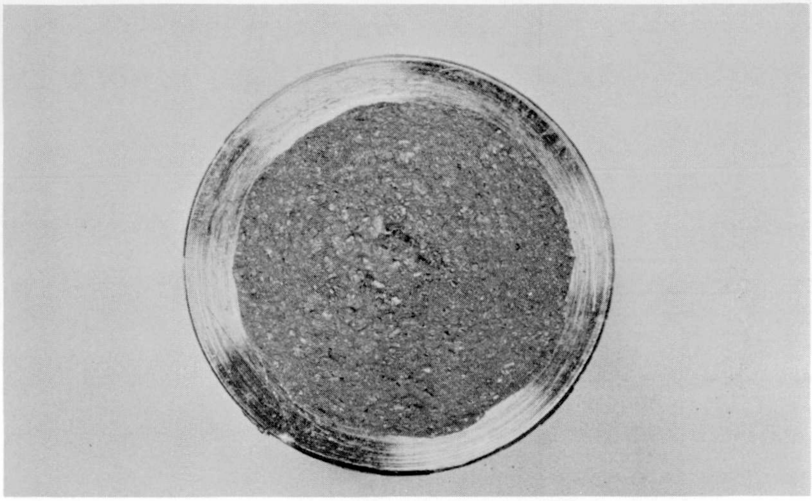
DENSITY OF COLD SWAGED ROD

Inches From End Of Rod	% Theoretical Density
1	90.8
2	91.2
3	90.4
4	90.2
5	90.6
6	91.0

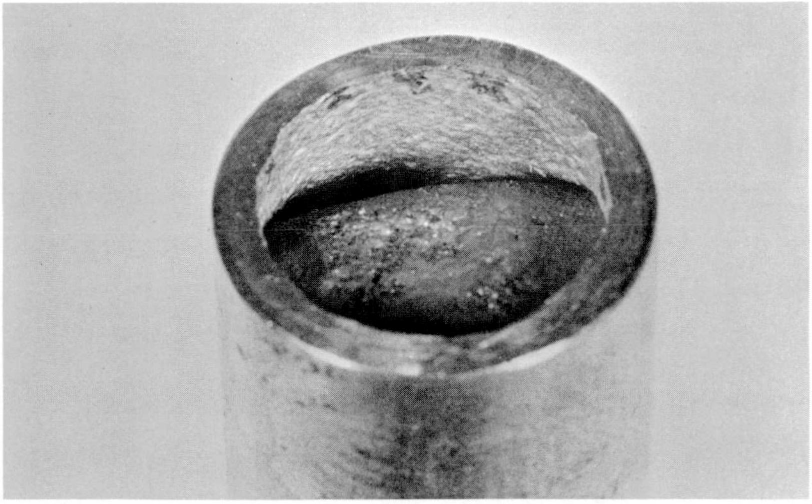
Densities were calculated from measured dimensions of each 1-in. segment and the density of the stainless steel. Segment No. 6 was at the center of the sample.

Later tests were concerned with thin-walled cladding. Tubing with wall thicknesses of 0.035 and 0.028 in. was packed to 80% of theoretical density with high density, crushed UO_2 mixed with 3 wt % alcohol. The material was vacuum dried at 80°C, and the tubes sealed. Only two swaging passes were required to reduce the diameter from 0.875 to 0.775 in. and increase the UO_2 density to 90% of theoretical.

Swaging of hollow assemblies was attempted. The outer cladding was 0.875 in. OD with 0.028 in. wall, and the inner tube was 0.312 in. OD with 0.028 in. wall. The annulus between the tubes was packed with UO_2 to a density of 81.5% of theoretical, using 3 wt % alcohol to prevent particle size segregation and to promote better packing. The assembly was swaged cold to a final diameter of 0.770 in. Swaging increased the UO_2 density to 90% of theoretical, with no



a. Machined End, Showing Cross Section



b. Nitric Acid Etched Section, Showing Inner Surface of Cladding

Figure 5. Hot Swaged UO_2

TABLE V

SWAGING DATA

	Unswaged	Cold Swaged (3 Passes)*	Hot Swaged (4 Passes)**
Length of Tube (in.)	10.0	14.55	17.75
Length of UO_2 (in.)	8.0	12.55	15.25
OD of Tube (in.)	0.750	0.565	0.503
Wall Thickness (in.)	0.065	0.062	0.056
Volume of UO_2 (cm^3)	39.60	31.8	30.20
Density of UO_2 (gm/cm^3)	8.0	9.9	10.4
Length of Tube (in.)	48.0	68.0	
Length of UO_2 (in.)	46.0	66.0	
OD of Tube (in.)	0.750	0.570	
Wall Thickness (in.)	0.035	0.032	
Volume of UO_2 (cm^3)	275.0	217.9	
Density of UO_2 (gm/cm^3)	7.6	9.64	

* Cold swaged through 0.687, 0.625, and 0.562 in. Dies.

** Hot swaged through 0.687, 0.625, 0.562, and 0.500 in. Dies.

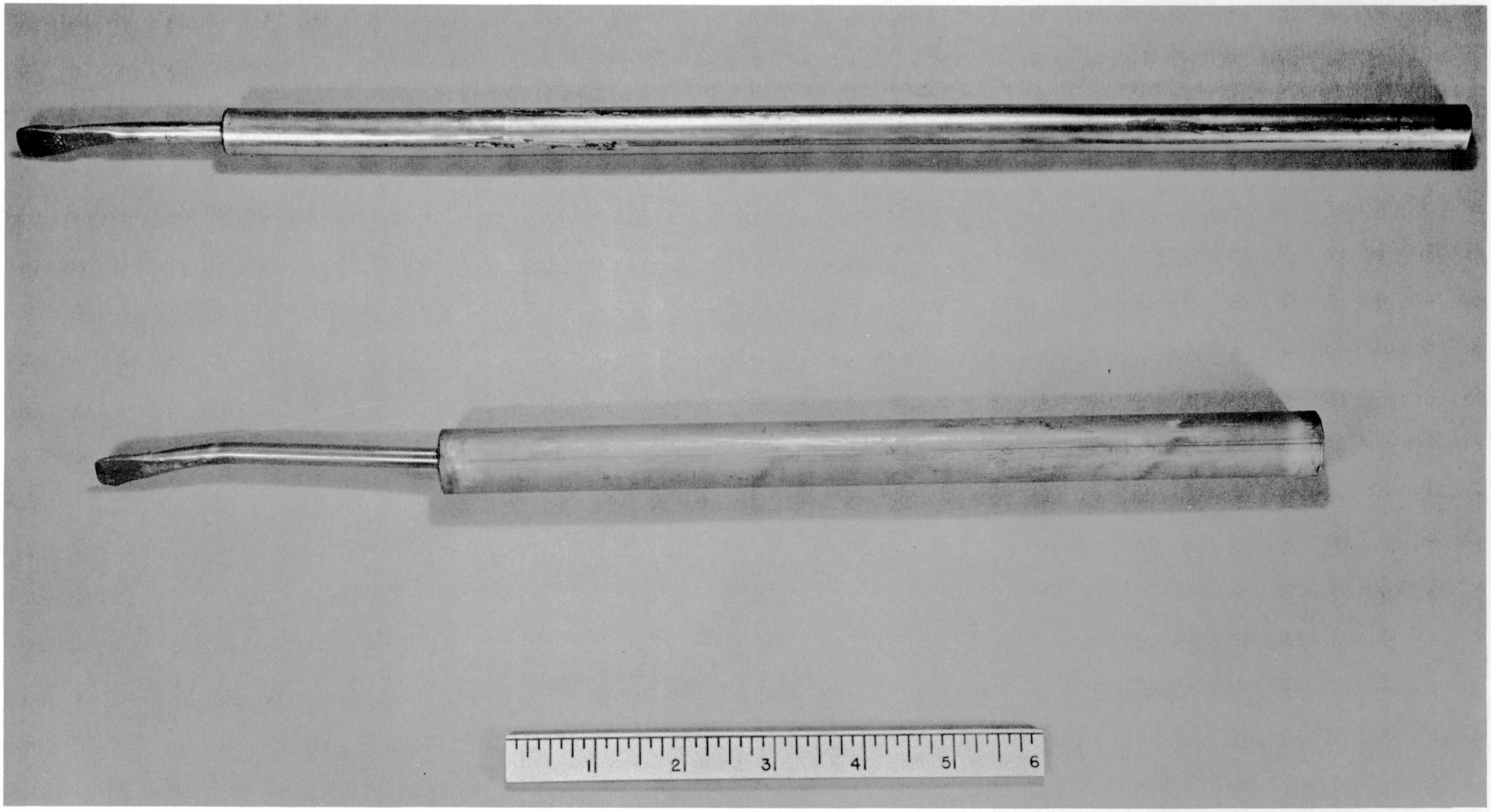


Figure 6. Sample Before and After Swaging

noticeable variation in wall thickness. Since a mandrel was not used, the resultant cross section of the inner tube was triangular, rather than circular. Larger hollow assemblies could not be swaged, as the maximum capacity of the swager was 15/16-in. diameter.

Several assemblies were made with 0.010 in. wall cladding. Tubes of 0.500 in. OD, with 0.010 in. wall thickness were packed with UO_2 shot to 78% of the theoretical density and cold swaged to a final diameter of 0.375 in. Resultant UO_2 density was 89 to 90% of theoretical. It was necessary to take small reductions on each swaging pass, to prevent cladding failures. Also, the starting UO_2 density had to be high to prevent wrinkling of the cladding on the initial swaging pass.

VI. CONCLUSIONS

The highest densities by vibratory compaction were obtained under the following conditions:

a) The particle diameter ratio of coarse to fine fractions is large (~50:1).

b) The coarse fraction is greater than 50 wt % of the composition.

c) UO_2 is presintered to near theoretical density.

d) A weighted piston is used as an upper floating ram.

e) Vibration or impact is of low frequency and large amplitude in the vertical direction.

f) Addition of 3 wt % water or alcohol is used to reduce particle size segregation and eliminate dusting.

Very little difference was noted between high density shot and crushed pellets as starting material. The maximum packed density was 82.2% of theoretical, although this value may be increased, under ideal conditions. High packed densities are desirable, since smaller reductions are necessary to achieve the desired swaged density.

It was possible to increase the packed UO_2 density to 9.9 gm/cm^3 by cold swaging and to 10.4 gm/cm^3 by hot swaging at 600°C. Results with thin wall cladding indicate the feasibility of swaging UO_2 packed in stainless steel tubing ranging from 0.065 to about 0.016 in. in wall thickness. Thicknesses less than about 0.016 in. are considered impractical, because of the anticipated high rejection rate due to cladding failures. Densities in excess of 91% of the theoretical value are difficult to achieve by cold swaging, since negligible sintering occurs. Higher densities are possible with hot swaging, but careful control is necessary to prevent thin spots in the cladding. The feasibility of swaging hollow elements is indicated, but a mandrel is necessary.

Vibratory compaction and subsequent swaging may be a useful and economical method for producing nuclear fuel elements from powdered and/or granular fuel materials. This process may be equally applicable to materials other than UO_2, such as carbides, nitrides, sulfides, phosphides, borides, and various cermets. In addition, control rod materials, such as Sm_2O_3 and Gd_2O_3, may be compacted into dense configurations by this method.

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