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ABSTRACT

A method for the preparation of high-density ThO_2 of the correct particle size for vibratory compaction is under development. The method being developed appears easily adaptable to remote operation. Pure ThO_2 made by this method was readily compacted to 86% of theoretical density by pneumatic vibration. Preparations of high-density mixed ThO_2-UO_2 and UO_2 by this general method have been successfully carried out. Densification of arc-fused mixed oxide by a pneumatic vibrator compared favorably with results of tests with the same oxide using electronic vibrators at both Huntsville, Alabama, and Savannah River. In the limited tests with the electronic vibrators at the other sites, it appeared that a saw-tooth wave shape gave greater densification than a sine wave or random noise.

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DISCLAIMER

This report is the first bimonthly progress report of the ORNL Fuel Cycle Program. It covers experimental work by E. S. Bomar, P. A. Haas, O. C. Dean, J. W. Snider, W. S. Ernst, Jr., S. D. Clinton, K. H. McCorkle, and C. E. Schilling. An ORNL topical report covering this material, ORNL-2965, is in preparation.

Vibratory Compaction Studies

The most interesting observation made during this period was the behavior during vibratory loading of ThO_2 prepared by the hydrothermal process (see next section). This oxide, having a particle density greater than 99% of theoretical, vibrated to a bulk density of 8.66 g/cc, or 86% of theoretical, in a 3/8-in.-o.d. stainless steel tube. The powder used for the vibration experiment was sized and blended with a distribution which represented a variation on that blend found by Hanford to be optimum for fused UO_2.

Preparatory to selecting electronic equipment to supply vibratory energy for powder consolidation work, cursory vibration experiments were run on electronic equipment at the U. S. Army Ballistic Missile Agency, Huntsville, Alabama, and the AEC facilities at Savannah River. The experiments at Huntsville examined the effect of vibration wave form and acceleration level on fused 96.6 wt % ThO_2-3.4 wt % UO_2 powder. The tubes were preloaded at ORNL. Pure sine waves gave a decrease in density from the value of about 8.3 g/cc for as-loaded powder. This is at variance with Hanford's experience where sine-wave vibration is reported to yield densities greater than 90% of theoretical with fused UO_2 in Zircaloy-2 tubes. Random-noise and saw-tooth wave forms cause no significant change in density, while distorted-sine wave form resulted in a detectable density increase to a maximum value of 8.51 g/cc. These data are shown in Table 1. Acceleration values ranging from 10 to 50 G appeared to have little effect on the resulting bulk density. All of the above tubes were revibrated on the BH-1 1/4-in. pneumatic vibrator upon returning to ORNL. Increases of 2 to 5% were obtained. The tests at Savannah River were carried out by a procedure duplicating that used by Hanford. Four runs were made using sine-wave input and resulted in an average density of 8.57 g/cc. A single run with saw-tooth input yielded a density of 8.80 g/cc. The Hanford visit was made for a mutual exchange of information on experience to date with vibratory consolidation.

Moving pictures were made at a speed of 155 frames/sec to record the movement of powder contained in a 2-ft-long lucite tube. At certain locations along the length of the tube, the fine particles appeared to move rapidly. However, the coarse fractions were relatively motionless.

In the course of loading experiments, a temperature rise as a result of absorption of vibrational energy had been detected in the tubes. To obtain quantitative results, thermocouples were placed at 2-in. intervals along a 2-ft section of stainless steel tubing. A difference was found for the temperature rise of a tube loaded with oxide compared with an empty tube as

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Table 1. Huntsville Electronic-Vibrator Conditions and Results for ThO_2-UO_2 Powders

Run No.	Acceleration, G	Freq., cycles/sec	Wave Form	Loaded[a] Density, g/cc	Electronically[b] Vibrated Density, g/cc	Pneumatically[c] Vibrated Density, g/cc
2	60	2000	Sine	8.33	8.29	8.62
3	46	943	Sine	8.32	8.26	--
4	50	77	Sine	8.37	7.94	8.26
5	50	370	Saw-tooth	8.34	8.37	8.50
6	48	290	Saw-tooth	8.40	8.44	8.69
7	19	40	Saw-tooth	8.34	8.40	8.75
8	29	50-1000	Random-noise	--	8.37	--
9	10	50-1000	Random-noise	8.30	8.34	8.62
10	20	90	Distorted-sine	8.44	8.51	8.58
11	20	66	Distorted-sine	8.34	8.23	8.70

[a] All tubes 2 ft long. Vibrated for 1 min while powder was poured into the tubes.

[b] Vibrated for 10 min.

[c] Vibrated for 10 min on pneumatic unit after returning to ORNL.

follows: loaded tube 50°C warmer at the bottom, 5°C warmer at the center, 20°C warmer at three-quarters of the distance from bottom of the tube, and 10°C cooler at the top.

The Preparation of High-Density Fuel Materials

A method is being developed for the preparation of high-density thoria and thoria-urania fuel materials which appears to be easily adaptable to operation behind shielding. Much of the procedure is carried out in water, and the maximum temperature used for densification is 1300°C, within the capability of day-to-day operation of silicon carbide globar furnace elements in air. By making adjustments in procedure, the process promises to yield the desired particle sizes, making unnecessary a grinding and screening operation.

Dense, sized thoria fragments were prepared by dispersion of low-fired thorium oxide particles in thorium nitrate or nitric acid, followed by hydrothermal denitration to a dried gel state. On calcination to 1200°C, particles whose average size was as large as 1.4 mm, and whose density was 9.97 g/cc, >99% of theoretical, were prepared. A distribution of sizes of this material had a tap density of 7.95 g/cc and a vibrated bulk density of 8.66 g/cc. Particle size control is achieved through control of temperature and nitrate content in the denitration step.

Batches up to 300 g of urania-thoria whose U/Th atomic ratio was 5/95 were prepared by a slight modification of the procedure for pure thoria above. Particle density was 9.78 to 9.94 g/cc, 98.5% of theoretical. Control of size appears possible. The average size of the largest particle group was 2 mm. A 200-g sample was vibrated to a bulk density of 8.33 g/cc.

Urania was prepared by a procedure similar to that used for thoria and the mixed oxide. Uranous oxalate was thermally decomposed to UO_2, then peptized with HCl solution. Under argon atmosphere three successive evaporations produced a glassy gel similar to the dried denitrated thoria gels. Calcination in a hydrogen atmosphere produced strong fragments of ~1 mm size and having a particle density of 10.2 g/cc, 93% of theoretical.

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