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IMPROVED SIZE UNIFORMITY OF SOL-GEL SPHERES BY IMPOSING A VIBRATION ON THE SOL IN DISPERSION NOZZLES

P. A. Haas
W. J. Lackey

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CHEMICAL TECHNOLOGY DIVISION
UNIT OPERATIONS SECTION

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A VIBRATION ON THE SOL IN DISPERSION NOZZLES

P. A. Haas
W. J. Lackey*

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ABSTRACT

A major part of the $\text{Th-}^{233}\text{U}$ fuel cycle program at ORNL has been concerned with the development of sol-gel processes to prepare ThO_2 and $\text{ThO}_2\text{-}^{233}\text{UO}_2$ spheres. The formation of sol drops having a uniform and controlled diameter is important to any sol-gel process for preparing oxide kernels for High Temperature Gas-Cooled Reactor fuels. A recently developed technique that incorporates use of a sol disperser with vibration has met the dispersion requirements for our ORNL sol-gel processes better than any of the previous techniques employing dispersers alone. With this new technique, the breakup of sol streams from orifices or capillaries is made more uniform and regular by imposing a vibration, at the natural frequency of drop formation, on the sol at the entrance to the orifices or capillaries. This technique has been applied to two-fluid nozzles to form 1,000 to 88,000 sol drops per minute. Batches consisting of 1 to 13 kg of fired ThO_2 spheres 370 to 500 μ in diameter had average diameters within 1% of the predicted values and standard deviations of 2.5 to 5.0 μ . Yields after both size and shape separation were greater than 95% and were usually greater than 98% in a 30- μ range. The same technique was applied to shear nozzles at rates up to 192,000 drops per minute. Nozzles of this type use multiple orifices and are more dependable for larger capacities than multiple two-fluid nozzles.

1. INTRODUCTION

The sol-gel processes developed at ORNL for preparing small thorium, uranium, and thorium-uranium oxide spheres are based on the conversion of

colloidal sol drops into gel spheres by extraction of water into an alcohol; usually 2-ethyl-1-hexanol (2EH).¹⁻³ The size uniformity of these drops is important to the overall yield of product since oversized and undersized spheres constitute the principal off-specification material. Also, size uniformity is essential to the continuous operation of fluidized-bed sphere-forming columns.¹ Scale-up to large capacities has been difficult for many of the previously used drop formation techniques. The average drop diameter must be predictable and controllable so that fired spheres of a specified size can be produced from various sols, each having different properties.

When ORNL sol-gel processes are used to supply the oxide kernels for pyrolytic-carbon-coated High-Temperature Gas-Cooled Reactor (HTGR) fuels, uniformity of sphere diameters has several important advantages. For example, variation of kernel diameter within batches is thought to be one of the main factors that determines why some particles of a given batch experience coating failure during irradiation while the other particles remain intact.⁴ The marked improvement in kernel size uniformity realized by using the new technique described in this report virtually eliminates kernel size variation as a source of variability in particle irradiation behavior. Further, the use of a particle size analyzer to determine coating thickness of HTGR fuel particles is anticipated in order to ensure coating process control. From this standpoint, improved uniformity in kernel size is also desirable since it will lead to a higher precision in determining the uniformity of coating thickness. In the case of buffer-coated HTGR fuel particles, there can be a correlation between kernel size and coating thickness such that the larger kernels, on the average, have a slightly thinner coating (which is undesirable). This effect is reduced as the kernel diameters become more uniform.

With most of the dispersion devices tested thus far, drops are formed from sol flowing through small (0.010- to 0.10-cm) orifices or capillaries.¹ When the sol flow rate and the drop size are specified, the

required rate of drop formation can be calculated from volume balances. Capacities of interest for current or future operations require 10,000 to 1,000,000 sol drops per minute.

A recently developed technique has met the above dispersion requirements better than any of the previously tested sol dispersers. In this new technique, the breakup of the sol stream from the capillaries or orifices of usual dispersion devices is made more regular and uniform by imposing a vibration on the sol at the entrance to the capillaries or orifices. The frequency of the vibration is set near the average natural frequency of drop formation for the particular disperser conditions so that the vibration promotes natural breakup. The effect of the vibration is to greatly improve size uniformity of the resulting gel spheres and to increase, to some extent, control of their mean diameter. This technique has important advantages over techniques employing dispersers that use vibration as the primary source of drop formation. A wide range of vibration power inputs or amplitudes is effective for the disperser-vibration combinations, while techniques using vibration alone generally have much narrower ranges of satisfactory power or amplitude.⁵

The equipment and procedures used to operate two types of dispersers, the two-fluid nozzle and the shear nozzle, in combination with vibration are described in this report. Typical results are given for the determination of the mean size and the uniformity of size for fired ThO_2 spheres 200 to 500 μ in mean diameter.

Acknowledgements -- The careful work of R. D. Arthur and D. A. McWhirter in operating the sphere preparation system and of J. B. Flynn in performing the screen analyses and making the radiographic measurements is appreciated.

2. TWO-FLUID DISPERSERS USED WITH VIBRATION

In the tests described here, a vibration was imposed on the sol entering the capillaries or orifices of the disperser in order to promote the natural frequency of drop formation. The ORNL sphere preparation processes require the formation of 10,000 to 1,000,000 drops of aqueous sol per minute in the 2EH process medium. A positive flow of 2EH is required to separate and prevent coalescence of the freshly prepared drops. After a few seconds of aging, these drops are covered by a surface film which is formed by the surfactants in the 2EH. This action minimizes coalescence.

Two of the disperser types previously tested appeared particularly suitable to the concept of promoting the natural frequency of breakup by imposing a vibration. In each, the sol drop size depends on the sol flow rate and the flow rate of the drive fluid. The natural frequency of breakup can be estimated, thus allowing selection of a suitable vibration frequency. The drive fluid flow removes and spaces the drops after formation. Other dispersion devices, with much smaller rates of drop formation from a larger number of orifices or capillaries, generally show flow variations between orifices and do not have a single natural frequency of drop formation.

2.1 Application of Vibration to the Sol

The procedure selected for applying the vibration was to vibrate a piston immersed in the sol at the entrance to the orifice or capillary from which the drops were formed. A typical equipment arrangement consisted of a 0.110-in.-diam piston in a 0.13-in.-ID sol feed channel (Fig. 1). The piston was coupled by a rod to a standard small electronic vibrator driven by a sine-wave power supply rated at 0 to 25 V-A and 50 to 10,000 cps. The coupling rod was sealed to the sol feed channel by a short section of thin-walled, 3/16- or 1/4-in.-ID rubber tubing. The difference between the piston diameter and the sol feed channel was kept small to reduce the loss

Fig. 1. Equipment Arrangement for Using the Two-Fluid Nozzle With Vibration.

of vibration input as a result of backflow by the piston. Results for our sol-gel preparation of spheres using vibration alone to disperse the sol were reported by Clinton.⁵ For these previous results, the vibrator was mechanically coupled to the capillary with the vibratory motion at right angles to the capillary axis.

The new technique for applying the vibration has advantages over arrangements which require mechanical coupling to the sol capillary or which use a diaphragm in contact with the sol. Some of these advantages are:

- (1) The mechanical arrangement is very simple and is easily combined with other disperser features.
- (2) The coupling rod is relatively stiff and there are no other movements of solid members; thus structural resonances at operating frequencies are eliminated.
- (3) No appreciable metal strains are present; consequently, the loss of vibration energy and the possibility of fatigue failures in metal components are minimized.
- (4) The vibration is applied near the point where dispersion occurs, thereby minimizing the loss of vibration energy.

As a result of the above factors, favorable results were obtained with low power inputs and with a complete range of high frequencies that were not practical for previous vibrating disperser arrangements. In techniques using vibration alone, the high frequencies are difficult to apply and are commonly usable only with careful "tuning" to make use of resonance effects.⁵

Some of the difficulties with using vibration alone arise from the damping effects if vibrating components are immersed in a liquid. In the ORNL sol-gel processes, the drops must be formed in the 2EH. If the drops are formed in a gas, the larger ones break up at the gas-2EH interface while the smaller ones accumulate and coalesce.

2.2 Operation of Two-Fluid Nozzles with Jet Breakup

Most of the vibration-promoted drops were formed via a single two-fluid nozzle with Rayleigh or varicose-type breakup of the sol after it left the feed capillary. The breakup of liquid jets at regular multiples of the jet diameter has been predicted theoretically and observed experimentally. For a two-fluid nozzle, the diameter of the sol jet may be varied by varying the flow rate of either the sol or the drive fluid. The sol is accelerated to the drive fluid velocity; then breakup into drops occurs in 2EH having a velocity equal to that of the sol stream.

Experimental results have shown that the diameters of the sol drops, D_I , are about 2.1 times the diameter of the jet. Thus, the sol flow rate, F , is easily related to the 2EH velocity, $V_{\max}$, by:

$$F = \frac{\pi}{4} \left(\frac{D_I}{2.1} \right)^2 V_{\max} \quad (1)$$

$$D_I = 2.1 \sqrt{\frac{4F}{V_{\max}}} \quad (2)$$

For a cylindrical flow channel and laminar flow, $V_{\max}$ at the center is twice the average velocity. Then

$$D_I = 1.70d \sqrt{\frac{F}{\bar{F}}} \quad (3)$$

where F and f are the sol and the drive fluid flow rates, respectively, and d is the inside diameter of the 2EH channel. From a simple volume balance, the rate of drop formation, n , is:

$$n = \left(\frac{6}{\pi D_I^3} \right) F. \quad (4)$$

As an example, we will calculate the conditions required to prepare 400- μ theoretically dense ThO_2 spheres from a ThO_2 sol containing 600 mg of thorium per milliliter, at a sol feed rate of 15 cc/min:

$$(600) \left(\frac{264}{232} \right) = 683 \text{ g of } \text{ThO}_2 \text{ per liter of sol.}$$

Fired spheres have 10 g of ThO_2 per cc = 10,000 g of ThO_2 per liter. Therefore,

$$D_I = (400) \left(\frac{10,000}{683} \right)^{1/3} = 982\mu = 0.0982 \text{ cm.}$$

$$\text{Drop volume} = \frac{\pi}{6} (0.0982)^3 = 0.495 \times 10^{-3} \text{ cm}^3/\text{drop},$$

$$n = \frac{15.0}{0.495 \times 10^{-3}} = 30,300 \text{ drops/min.}$$

Vibration frequency is 30,300 cpm or 505 cps.

For $d = 0.130 \text{ in.} = 0.330 \text{ cm}$,

$$\frac{f}{F} = \left[\frac{1.70d}{D_I} \right]^2 = \left[\frac{(1.70)(0.330)}{0.0982} \right]^2 = 32.5$$

$f = (32.5)(15.) = 490 \text{ cc of 2EH per minute.}$

The jet breakup was promoted and made more regular as shown by three observations. When observed with a stroboscopic light, operation at approximately the conditions given in the above calculations gives a jet about 3 to 4 in. long prior to breakup when vibration is not used, and a jet 1 to 2 in. long when a 500-cps vibration is applied. Without vibration, the drops are blurred and the formation of nodes in the jet is not sufficiently regular to be "stopped" by the stroboscopic light. With vibration, the breakup is easily and completely stopped by the stroboscopic light. The spheres made using vibration are more uniform as shown by size distribution data in Sect. 3.

When the two-fluid nozzle is matched to the vibration frequency, the beneficial effects of the vibration are obtained for a very wide range of power inputs or vibration amplitudes. Under the conditions of the example (above), any input available from the power supply (0.2 to 3.0 A) was effective with a small decrease in jet length before breakup when the power was varied over this range. The lower power inputs were not effective when frequencies were >600 cps, when the drive fluid flow was not matched to the frequency, or when the inside diameter of the sol feed capillary was very different from $D_I/2$. Any one of these three unfavorable factors can usually be overcome by increasing the power, but realization of effective operation is unlikely if two of them are unfavorable.

The capacity of a single nozzle is limited by the necessity for laminar flow of the drive fluid 2EH and by the excessive pressure drops required for high flow rates. The allowable $V_{\max}$ for 2EH is about 15,000 cm/min, since changes in cross section in the nozzle tend to cause turbulences. For large drops (1500 μ) and a sol jet of about 0.0750 cm, this corresponds to about 60 cc of sol per minute and about 35,000 drops per minute. For small drops (~ 500 μ) and a sol jet of 0.0250 cm, this $V_{\max}$ corresponds to about 7 cc of sol per minute and about 100,000 drops per minute. The uniformity of the drops was better for $V_{\max}$ values below 10,000 cm/min than for the 10,000- to 15,000-cm/min range.

One logical approach to scale-up to larger capacities is to use multiple sol feed capillaries with a single sol feed, single vibrator, and single 2EH drive fluid flow. One six-capillary nozzle was fabricated [Fig. 2(a)]. The vibration was effective for all the capillaries (the rate of drop formation per capillary was equal to the frequency), but the uniformity of drop size suffered from a nonuniform division of sol flows between the capillaries. This problem was not very noticeable when the ThO_2 sol flow was started with a clean nozzle; however, reduced flows of one or two capillaries tended to occur after less than 2 hr of operation, and partial or complete plugging of one or more capillaries was usually observed after 1 hr or overnight shutdowns. No difficulties of this type were experienced with single two-fluid nozzles, and full flow was easily restarted and maintained by the metered sol flow. The shear nozzles using orifices (Sect. 2.3) appear much more satisfactory for multiple sol-feed channels since the orifices of these nozzles suffer less from unequal divisions of flow.

Some operating conditions produced a very small satellite sol drop ($<0.1\%$ of the total volume). This could usually be eliminated by using a different vibration power input or by decreasing the 2EH drive fluid to about 80% of that calculated from Eq. (3). The lower drive fluid flow rate gave a closer drop spacing so that the "tail" between drops was pulled into the large drop instead of being separated as a satellite.

Operating conditions for which size distributions were determined (Sect. 3) are tabulated, along with conditions for some other tests, in Table 1. The two-fluid nozzle with vibration has been used routinely to prepare greater than 200 kg of 380- and 500- μ ThO_2 spheres with about 630 g of ThO_2 per hour as the typical flow rate. More than 400 hr of satisfactory operation has been experienced for this type of preparation, along with other types and experimental testing; and more than 100 startups have been made without significant equipment failures or problems.

Fig. 2. Equipment Arrangement for Using (a) the Six-Capillary Two-Fluid Nozzle and (b) the Twelve-Orifice Shear Nozzle with Vibration.

Table 1. Typical Test Conditions for Operation of Two-Fluid Nozzles with Vibration

Size measurement batch No.	J-145	J-147	J-148	J-149	J-154	—	—	J-172	J-192	J-157
Product batch No.	137-9-12	138-9-13	138-9-15	138-9-19	139-9-26	140-9-27 ^a	141-10-5	144 ^b	145 ^c	136 & 137
Sol volume, cc	3,450	6,400	4,050	4,000	4,800	3,770	1,600	—	—	—
Time, min	283	538	325	377	355	250	287	—	—	—
Sol flow rate F, cc/min	12.2	11.9	12.5	10.6	13.5	15.1 ^a	5.58	15.5	15.3	12
Sol concentration, g ThO ₂ /liter	670	670	670	670	615	678	676	676	668	670
Nozzle ID, d, cm	0.335	0.335	0.325	0.335	0.325	0.85	0.335	0.325	0.325	0.335
2EH flow rate through nozzle, f, cc/min	265	265	155	563	363	950	600	195	195	300
Sol capillary ID, μ	430	430	600	450	600	500	270	600	600	430
Vibration frequency, cps	480	480	230	1,000	500	90 ^a	1,400	265	260	None ^d
Drop formation rate, n, min ⁻¹	28,800	28,800	13,700	60,000	30,000	32,000 ^a	88,000	15,900	15,700	— ^d
Calculated drop diameter, μ	932	926	1,203	696	951	962 ^a	495	1,228	1,236	1,120 ^d
Calculated fired diameter, μ	378	376	490	283	375	392	202	501	501	450 ^d

^a Product batch 140-9-27 was obtained by use of six sol capillaries in parallel, with one vibrator and one 2EH flow [see Fig. 2(a)].

^b Four batches composited (144-10-26, 144-10-27, 144-10-30, 144-10-31).

^c Four batches composited (145-11-1, 145-11-2, 145-11-3, 145-11-6).

^d Two-fluid nozzle was used without vibrator; diameter calculated from Eq. (3).

2.3 Operation of Shear Nozzles with Multiple Orifices

In the shear nozzle type of disperser operation, sol drops are detached from an orifice. The drag or shear of a 2EH flow at right angles to the orifice axis, along with a smaller force from the weight of the drop, overcomes the interfacial tension to detach the drop. The force from the 2EH flow, and hence the drop size, is a complex function of the nozzle configuration, the 2EH flow rate, and the 2EH properties such as temperature or viscosity. Consequently, the sol drop size cannot be predicted by a simple equation as can be done for the Rayleigh breakup of the sol jet in the two-fluid nozzle. The orifices for a shear nozzle should generally have diameters of 0.025 to 0.060 cm.

The principal advantage of the shear nozzle over the two-fluid nozzle (described in Sect. 2.2) is that an equal division of a sol jet between multiple flow channels is much easier to obtain with orifices than with capillaries. Therefore, the shear nozzle (with multiple orifices) is practical for drop formation rates greater than 100,000 drops per minute, whereas two-fluid nozzles with multiple capillaries have been troublesome. The orifices can be freed of any plugs that occur during shutdown by resorting to a brief period of high-amplitude vibration before sol feed is resumed.

When the shear-nozzle-with-vibration technique was used, vibration was applied to the sol using the same arrangement as had been employed previously for the two-fluid nozzle. Of course, in this case, the sol feed channels were orifices in the tube wall and the 2EH flowed through the annulus between the sol tube and a larger outer tube [Fig. 2(b)]. The velocity of the sol through the orifices should be high enough to carry the sol away from the sol-tube wall but should not cause impingement on the outer tube walls. If the sol stream forms a jet instead of detaching drops near the orifice, then the jet breaks up according to the Rayleigh or two-

fluid nozzle mechanism as described by Eqs. (1) and (2). As a result of these limitations, the orifice diameter of the shear nozzle must be suitable for the sol flow rate and drop size required.

When the shear-nozzle-with-vibration technique is used, the sol flow through an orifice varies at the frequency of vibration. One type of effective operation is to discharge the sol in "slugs" with zero or negative flow rates for part of each cycle. Utilizing a stroboscopic light set to give a "slow motion" effect, the appearance is that of small spheres being discharged from the sol orifices with sufficient velocity to carry them to the middle of the 2EH stream. If the velocity of the sol is too high, the slugs are thrown to the opposite wall, where they tend to collect and coalesce. Collection and coalescence can be minimized by incorporation of one or more of the following: a larger orifice diameter, a higher 2EH velocity, a lower vibration power, a larger annulus for 2EH flow, or a lower sol flow rate. If the sol velocity is too low, the sol remains near the orifice and is likely to coalesce with the next slug. If the sol forms a jet with a two-fluid nozzle type of breakup, the jet may not be centered at the point of maximum 2EH velocity. Because of the above requirements, operation with the shear nozzle requires a trial-and-error selection of conditions while that of a two-fluid nozzle can be predicted with more certainty.

Equation (4) for the rate of drop formation also applies to the shear nozzles, where F is the sol flow rate per orifice. The 2EH flow rate may be estimated from experimental results for drop size from shear nozzles without vibration, or $V_{\max}$ from Eq. (2) can be used as a high estimate of the 2EH velocity. The preferred values for $V_{\max}$ are usually 50 to 100% of those calculated from Eq. (2). One upper-capacity limit for the shear nozzles would be the requirement for a laminar flow of 2EH to remove the sol drops, and this would be smaller than the capacities (35,000 to 100,000 drops per minute per orifice) as given for single two-fluid nozzles since the shear nozzles require larger 2EH flow channels and smaller $V_{\max}$. Maximum capacities of about one-tenth the above limits (from 1 cc per min per orifice for 500- μ sol drops to 10 cc per min per orifice for 1500- μ sol drops) are more satisfactory.

Table 2 tabulates the operating conditions for which size distributions were determined (Sect. 3), along with conditions for some other tests. The optimum results for operation of shear nozzles were not obtained in time to be included with the size distribution results reported in Sect. 3. No difficulties were experienced from plugging of orifices when the shear nozzles were restarted after overnight or weekend shutdowns. Poor distribution of the 2EH flow caused turbulences and breakup of drops into fines, as shown by the size distribution data for batch J-190; this problem was eliminated by making a minor alteration in the 2EH inlet.

3. SIZE DISTRIBUTION OF FIRED ThO_2 SPHERES

3.1 Size Measurement Procedures

Detailed size analyses of ten batches of ThO_2 microspheres were performed. Kernel diameters in each batch were determined by screen analysis and by radiographic measurements. In the case of the screen analyses the entire batch of spheres was analyzed. However, in order to prevent overloading of the screens, each batch was subdivided and only a 350-g quantity was analyzed at any given time. A screening time of 15 min was ample to allow good screening efficiency since a Syntron Test Sieve Shaker was used. New 8-in.-diam micromesh screens reported to be accurate to $\pm 2 \mu\text{m}$ were used. After screening, each size fraction was passed over a shape plate to remove any nonspherical or broken particles. The percentage of nonspherical material was determined for each size fraction by weighing. Adjustment of the shape plate to minimize rejection of material that is actually spherical is dependent on sphere size and flow rate, as well as on the operator. With one exception, the material was allowed to pass over the shape plate only once. The material initially rejected in Batch J-189 was passed over the plate a second time, and on the order of three-fourths of it was not rejected. Microscopic examination of the material that was initially rejected in this batch but which was accepted during the second shape separation showed it to be spherical. The rejected material consisted of doublets and

Table 2. Typical Test Conditions for Operation of Shear Nozzles with Vibration

Product batch No.	143-10-25A	143-10-25B	147-11-20B	150-12-7A	151-12-12	151-12-13
Size measurement batch No.	J-189	J-190	—	—	—	—
Sol volume, cc	3,400	1,550	1,950	2,26C	2,100	5,000
Time, min	218	100	35	8C	50	374
Sol flow rate, F, cc/min	15.6	15.5	55.7	28.3	42	13.4
Sol concentration, g ThO ₂ /liter	580	580	680	676	680	680
Nozzle ID, d, cm	0.95	0.95	0.95	1.27	1.27	1.27
2EH flow rate through nozzle, f, cc/min	530	720	740	56C	560	560
Sol orifice diameter, μ	460	460	530	53C	530	410
No. of sol orifices	8	8	12	12	12	12
Vibration frequency, cps	150	400	80	8C	40	140
Drop formation rate, n, min ⁻¹	72,000	192,000	57,600	57,60C	43,200	101,000
Calculated drop diameter, μ	744	535	1,230	98C	1,231	634
Calculated fired diameter, μ	303	218	500	399	500	258

nonspherical or broken particles. Thus, the values reported for the percentage of nonspherical material represent maximums.

Samples consisting of about 2000 spheres to be used for the radiographic measurement of diameter were riffled from each batch of spheres prior to screening or shape separation. For each resulting radiograph the diameters of 200 randomly selected spheres were measured at a magnification of 100X using a microscope equipped with a calibrated split-image eyepiece. The standard deviation of the radiographic measurement was determined to be 1.2 μm by repeatedly measuring the diameter of a single sphere. This error was removed from the reported particle diameter standard deviation, s , by an accepted statistical technique:

$$\text{Actual } s = \left[(\text{observed } s)^2 - (1.2)^2 \right]^{1/2} .$$

3.2 Size Measurement Results

Results of the screen analysis for one of the batches of spheres (J-146) are given in Table 3. Yields for each size fraction both before and after shape separation are reported; in each case, they were based on the initial batch weight. The excellent quality of the product is evidenced by the fact that 98.6 wt % of the as-fired material was spherical and in the size range 360 to 390 μm . Note that the amount of material rejected during shape separation was only 1.1 wt %.

A more precise measure of the quality of this material was obtained from the radiographic measurements. Histograms constructed from the radiographic data (see Fig. 3) indicate that the spheres made with the technique employing vibration were extremely uniform in size. The standard deviation of the particle diameter was 2.5 μm when vibration was used, compared with 27.8 μm for spheres made without vibration. Figure 4 shows the same data for the batch of spheres made with vibration, plotted again as a bar graph

Fig. 3. Comparison of Size Distribution of ThO_2 Microspheres Made With and Without Vibration.

Fig. 4. Particle Size Distribution Determined by Microradiography.

Table 3. Screen Size Analysis of ThO₂ Batch J-146

Total rejected by shape separation = 1.1%

Spherical material in size range 350-400 μm = 98.8%

Spherical material in size range 360-390 μm = 98.6%

Size (μm)	Yield Before Shape Separation (wt %)	Yield After Shape Separation (wt %)
< 350	0.09	0.03
350-360	0.05	0.05
360-370	0.79	0.78
370-380	91.42	90.58
380-390	7.26	7.22
390-400	0.14	0.13
> 400	0.24	0.14

but with an expanded scale. Such bar graphs were constructed to help determine the nature of the particle size distribution, that is, to determine whether the distributions were normal, skewed, or otherwise. Actually, the probability plot of Fig. 5 was of more value for this purpose. For such plots, a statistically normal distribution appears as a straight line. As can be seen, the observed distribution for Batch J-146 was not precisely normal. The dashed line in Fig. 5 represents a normal distribution with a mean value near 378 μm and a standard deviation only slightly less than that calculated from the radiographic measurements. Note that if about 5% of the smallest and 5% of the largest spheres in this batch are excluded, then the dashed line describes the observed size distribution. A study of Fig. 5 reveals that the percentage of spheres smaller than about 374 μm and larger than 381 μm was higher than expected based on the normal distribution represented by the dashed line. Probability plots for other batches showed some to follow a normal distribution, but most displayed a deviation. In many cases, the deviation was similar to that just described; however, in other cases, there were actually fewer of the smallest and/or largest spheres than would have been expected for a normal distribution.

Fig. 5. Particle Size Distribution Determined by Microradiography and Screening.

No particular significance is attached to this type of deviation, and it is probably of no practical consequence although it complicates calculation of tolerance intervals as described below,

Tolerance intervals for sphere diameter, that is, intervals having a stated probability of containing a specified percentage of the population, were calculated for most of the batches of spheres. One of the quantities needed in order to perform this calculation is the standard deviation of diameter for the batch of spheres of interest. If a standard deviation of $2.5\text{ }\mu\text{m}$, which is the value calculated from the radiographic measurements for Batch J-146, is used, the resulting diameter tolerance interval is not wide enough. For example, if one attempts to calculate a tolerance interval such that 95% of the spheres in 95% of future batches made by the process used to prepare Batch J-146 would have diameters that fell within the calculated interval, the resulting interval is 372.6 to $383.4\text{ }\mu\text{m}$. In other words, only 2.5% of the spheres should be smaller than $372.6\text{ }\mu\text{m}$ and only 2.5 % larger than $383.4\text{ }\mu\text{m}$. However, note in Fig. 5 that actually about 5% of the spheres were larger than $383.4\text{ }\mu\text{m}$ because of the departure of the size distribution from normality. To correct for this error, we resorted to an approximation, which is conservative in that it overestimates the width of the tolerance interval. Toward this end, we drew the solid line in Fig. 5 so that the radiographically determined data points for the smaller spheres were either on or above the line and the points for the larger spheres were either on or below the line. This line represents a normal distribution having a larger standard deviation (larger slope) than that represented by the dashed line. Use of this inflated standard deviation in calculating the tolerance interval results in a so-called "conservative tolerance interval." The 95%, 95% conservative tolerance interval for Batch J-146 was $378.0 \pm 8\text{ }\mu\text{m}$. Thus, 95% of the spheres in 95% of future batches made using the process used to prepare Batch J-146 would have diameters in the range $378.0 \pm 8.0\text{ }\mu\text{m}$. Consequently, one has a high degree of confidence that a very large percentage of the spheres in a batch will fall within an extremely narrow size range.

The closed data points in Fig. 5 show the results of the screen analysis. The screen analysis and radiographic results are in good agreement, considering that the uncertainty in the opening of the screens is $\pm 2 \mu\text{m}$. Strictly, the results of the screen analysis and radiographic measurements should not be expected to agree precisely since the screen analyses are based on weight percent and the radiographically determined values are based on the numbers of spheres rather than on weight. Since the spheres are nearly the same size, however, this difference is negligible except for one batch (J-190) which contained spheres not nearly as uniform in size as the other batches.

Screen analyses, histograms, and probability plots for the remaining batches of spheres are included in the Appendix. Pertinent parameters describing the size and size distribution of all the batches are summarized in Table 4. Except for the batch of smallest spheres (J-190), the results show that each batch made by employing sol vibration is of exceptionally high quality in terms of uniformity of size and sphericity. Results of the screen analyses typically show that the percentage of the original spheres found acceptable by shape separation and falling within $\pm 15 \mu\text{m}$ of the mean sphere diameter ranges from 95 to 99%. In the worst of the batches made with sol vibration (i.e., J-190), 93.4 wt % of the original batch was spherical and in the size range 200 to 250 μm .

The standard deviation of diameter for each of the batches made with vibration is plotted against the average sphere diameter for the batch in Fig. 6. For average diameters in the range 300 to 500 μm , the standard deviation of diameter varies from 2.1 to 5.9 μm . For the batch having an average diameter of 207.2 μm , the standard deviation is somewhat deceptively high from the standpoint of the weight percent of the material that falls within a fairly narrow size range. As previously stated, 93.4 wt % of this batch consisted of spherical particles in the size range 200 to 250 μm . It did contain 15 to 20% of undersized spheres on a number basis, as can be seen from the histogram and probability plots of Figs. 21 and 22 in the Appendix; however, on a weight basis, a rather large percentage of the

Table 4. Summary of Size Distribution Results for Sol-Gel ThO₂ Spheres Made with Vibration

Batch No.	Batch Weight (kg)	Average Sphere Diameter ^a (μm)	Standard Deviation of Sphere Diameter ^a (μm)	Conservative Estimate of Standard Deviation of Diameter (μm)	Tolerance Intervals (μm)		Rejected by Shape Plate (wt %)
					95%, 95% ^b	99%, 99% ^c	
J-146	2.2	378.0 ± 0.3	2.5 ⁺ _{-0.3}	3.7	378.0 ± 8.0	378.0 ± 10.9	1.1
J-147	4.2	379.2 ± 0.5	3.7 ⁺ _{-0.5}	4.6	379.2 ± 9.9	379.2 ± 13.5	1.4
J-148	2.6	489.4 ± 0.6	4.2 ⁺ _{-0.5}	4.2	489.4 ± 9.1	489.4 ± 12.4	1.1
J-149	2.6	282.7 ± 0.7	5.0 ⁺ _{-0.6}	7.7	282.7 ± 16.6	N.D. ^d	0.4
J-154	3.0	374.1 ± 0.3	2.4 ⁺ _{-0.3}	4.7	374.1 ± 10.2	374.1 ± 13.8	0.9
J-157 ^e	22.4	390.0 ± 3.9	27.8 ⁺ _{-3.4}	N.D.	N.D.	N.D.	0.9
J-172	13.2	499.8 ± 0.5	3.6 ⁺ _{-0.4}	7.5	499.8 ± 16.2	499.8 ± 22.0	1.4
J-189	2.2	302.5 ± 0.8	5.9 ⁺ _{-0.7}	5.9	302.5 ± 12.7	302.5 ± 17.3	0.15
J-190	0.9	207.2 ± 5.4	38.9 ⁺ _{-4.7}	N.D.	N.D. ^d	N.D. ^d	0.7
J-192	12.8	495.0 ± 0.3	2.1 ⁺ _{-0.3}	N.D.	N.D.	N.D.	0.6

^aThe uncertainty values give the 95% confidence interval, assuming that the distribution is normal.

^bThe probability is 0.95 that the reported range would include at least 95% of the kernels made by such a process.

^cThe probability is 0.99 that the reported range would include at least 99% of the kernels made by such a process.

^dNot determined because departure from normal distribution was large.

^eMade without vibration.

Fig. 6. Influence of Average Sphere Size on Uniformity.

material falls within a relatively small size range. The poorer quality of the spheres in this batch is explained by turbulence in the 2EH flow as a result of the inlet configuration, which caused breakup of drops into fines at one side of the nozzle. This effect was eliminated by a minor modification of the nozzle, but improved samples were not available in time for size distribution analysis.

The sphere densities for four of the batches (J-146, -147, -148, and -149) were determined by mercury pycnometry. In each of these batches, the spheres were found to be theoretically dense to within the accuracy of the technique (0.1 g/cm^3). Photographs of riffled samples from these batches appear in Figs. 7 and 8.

4. RESULTS AND CONCLUSIONS

The results obtained with the dispersion-with-vibration technique discussed in this report represent an important improvement in the sol-gel preparation of ThO_2 spheres. By using vibration to promote the natural frequency of drop formation, careful tuning becomes unnecessary. When imposed through a stiff rod immersed in the sol, the vibration is effective for a wide range of frequencies without effects from structural resonances. The mechanical arrangement is simple and has been trouble-free for more than 400 hr of operation and more than 100 startups and shutdowns. The allowable rate of drop formation is limited by our capability for removing freshly formed drops so they do not contact each other and coalesce.

Most of the vibration-promoted drops were formed with single two-fluid nozzles at the rate of 1,000 to 88,000 drops per minute. The vibration gives a controlled rate of drop formation for multiple two-fluid nozzles, but equal divisions of sol flow between parallel capillaries result in poorer uniformity of sol drops. Shear nozzles, which were also used with vibration, appear more satisfactory for multiple sol feed channels because the orifices suffer less from unequal divisions of flow. Shear nozzles

(a)

(b)

Fig. 7. Photomicrographs of Samples Riffled from (a) Batch J-146 and (b) Batch J-147. Magnification, 33X.

(a)

(b)

Fig. 8. Photomicrographs of Samples Riffled from (a) Batch J-148 and (b) Batch J-149. Magnification, 33X.

were operated to prepare 3×10^3 to 192×10^3 sol drops per minute and could easily be scaled up to larger capacities.

Detailed size analyses of ten batches of ThO_2 spheres show excellent uniformity and control of average diameter by use of two-fluid nozzles in combination with vibration. Data determined by radiographic measurements and screen analysis were treated by standard statistical methods. Batches consisting of 1 to 13 kg of fired, 370- to 500- μ -diam ThO_2 spheres had average diameters within 1% of the predicted values and standard deviations of 2.5 to 5.0 μ . Comparable samples prepared at rates greater than 10,000 drops per minute without vibration showed standard deviations of 28 μ for 390- μ ThO_2 , 16 to 19 μ for 160- μ UO_2 ,⁶ and 19 to 38 μ for 360- μ ThO_2 - UO_2 .² Use of the shear nozzle in combination with vibration can provide almost as good uniformity, as shown by a standard deviation of 5.9 μ for a batch of spheres having an average diameter of 303 μm .

The improved uniformity that is possible when these dispersers are used with vibration has important advantages for HTGR fuel kernels. Uniformity such as that reported here can virtually eliminate kernel size variations as a major source of variability in particle irradiation behavior. The improvement in kernel size uniformity will also result in greater precision in the determination of coating thickness uniformity. For buffer-coated HTGR fuel particles, the uniformity of kernel diameter is expected to minimize undesirable variations in coating thickness.

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6. APPENDIX

Screen size data for nine batches of ThO_2 spheres, and radiographic data for seven of the batches, are shown in Tables 5 - 13 and Figs. 9 - 22, respectively.

Table 5. Screen Size Analysis of ThO₂ Batch J-147

Total rejected by shape separation = 1.4%

Spherical material in size range 350-420 μm = 97.9%

Spherical material in size range 360-390 μm = 96.5%

Size (μm)	Yield Before Shape Separation (wt %)	Yield After Shape Separation (wt %)
< 350	0.14	0.13
350-360	0.32	0.31
360-370	1.52	1.50
370-380	82.51	81.40
380-390	13.65	13.59
390-400	0.90	0.89
400-410	{ 0.97	0.13
410-420		0.04
> 420		0.59

Table 6. Screen Size Analysis of ThO₂ Batch J-148

Total rejected by shape separation = 1.1%

Spherical material in size range 450-520 μm = 98.3%

Spherical material in size range 470-500 μm = 98.2%

Size (μm)	Yield Before Shape Separation (wt %)	Yield After Shape Separation (wt %)
< 450	{ 0.54	0.42
450-460		0.04
460-470		0.06
470-480	4.94	4.94
480-490	57.07	56.24
490-500	37.19	36.98
500-510	0.04	0.04
510-520	0.04	0.04
> 520	0.17	0.15

Table 7. Screen Size Analysis of ThO₂ Batch J-149

Total rejected by shape separation = 0.4%

Spherical material in size range 200-300 μm = 98.3%

Spherical material in size range 250-300 μm = 95.0%

Size (μm)	Yield Before Shape Separation (wt %)	Yield After Shape Separation (wt %)
< 200		1.28
200-220	3.86	0.91
220-230		0.66
230-240		0.99
240-250	0.88	0.68
250-300	95.19	95.03
300-310	0.03	0.03
310-320	0.01	0.01
320-330	0.01	0.01
> 330	0.02	0.02

Table 8. Screen Size Analysis of ThO₂ Batch J-154

Total rejected by shape separation = 0.9%

Spherical material in size range 350-400 μm = 98.1%

Spherical material in size range 360-390 μm = 98.0%

Size (μm)	Yield Before Shape Separation (wt %)	Yield After Shape Separation (wt %)
< 350	0.14	0.12
350-360	0.08	0.08
360-370	0.66	0.65
370-380	97.58	97.30
380-390	0.08	0.06
390-400	0.04	0.02
> 400	1.42	0.91

Table 9. Screen Size Analysis of ThO₂ Batch J-157^a

Total rejected by shape separation = 0.9%

Size (μm)	Yield Before Shape Separation (wt %)	Yield After Shape Separation (wt %)
< 300	N.D. ^b	0.89
300-350	N.D.	5.40
350-420	N.D.	75.75
420-500	N.D.	16.90
500-520	N.D.	0.10
> 520	N.D.	0.03

^a Made without vibration.^b N.D. = not determined.Table 10. Screen Size Analysis of ThO₂ Batch J-172 After Shape Separation

Total rejected by shape separation = 1.4%

Spherical material in size range 480-510 μm = 95.4%Spherical material in size range 470-520 μm = 96.2%

Size (μm)	Yield After Shape Separation (wt %)
< 470	1.74
470-480	0.40
480-490	0.78
490-500	86.34
500-510	8.27
510-520	0.39
> 520	0.69

Table 11. Screen Size Analysis of ThO₂ Batch J-189

Spherical material in size range 250 to 340 μm = 99.4%

Total rejected by shape separation = 0.15%

Initially rejected material was passed over shape plate
a second time

Size (μm)	Yield Before Shape Separation (wt %)	Yield After Shape Separation (wt %)
< 250	0.17	0.15
250-300 ^a	50.85	50.80
300-310	42.07	42.04
310-320	6.47	6.46
320-330	0.05	0.04
330-340	0.03	0.02
> 340	0.35	0.32

^a Intermediate screens were not available.

Table 12. Screen Size Analysis of ThO₂ Batch J-190

Total rejected by shape separation = 0.7%

Spherical material in size range 100-300 μm = 98.2%

Spherical material in size range 200-250 μm = 93.4%

Size (μm)	Yield Before Shape Separation (wt %)	Yield After Shape Separation (wt %)
< 100	0.59	0.49
100-180	2.11	2.00
180-200	0.74	0.72
200-220 ^a	58.23	58.15
220-250 ^a	35.38	35.23
250-300	2.22	2.06
> 300	0.73	0.69

^a Intermediate screens were not available.

Table 13. Screen Size Analysis of ThO₂ Batch J-192

Total rejected by shape separation = 0.6%

Spherical material in size range 470-520 μm = 98.8%

Spherical material in size range 480-510 μm = 98.7%

Size (μm)	Yield Before Shape Separation (wt %)	Yield After Shape Separation (wt %)
< 470	0.15	0.11
470-480	0.06	0.06
480-490	0.26	0.26
490-500	98.69	98.22
500-510	0.19	0.18
510-520	0.09	0.05
> 520	0.55	0.48

Fig. 9. Particle Size Distribution Determined by Microradiography.

Fig. 10. Particle Size Distribution Determined by Microradiography and Screening.

Fig. 11. Particle Size Distribution Determined by Microradiography.

Fig. 12. Particle Size Distribution Determined by Microradiography and Screening.

Fig. 13. Particle Size Distribution Determined by Microradiography.

Fig. 14. Particle Size Distribution Determined by Microradiography.

Fig. 15. Particle Size Distribution Determined by Microradiography.

Fig. 16. Particle Size Distribution Determined by Microradiography and Screening.

Fig. 18. Particle Size Distribution Determined by Microradiography and Screening.

Fig. 19. Particle Size Distribution Determined by Microradiography.

Fig. 20. Particle Size Distribution Determined by Microradiography and Screening.

Fig. 21. Particle Size Distribution Determined by Microradiography.

Fig. 22. Particle Size Distribution Determined by Microradiography and Screening.

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