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Device for Sampling HTGR Recycle Fuel Particles

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Printed in the United States of America. Available from
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Price: Printed Copy $4.00; Microfiche $3.00

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Contract No. W-7405-eng-26

METALS AND CERAMICS DIVISION

THORIUM UTILIZATION PROGRAM (189a OH045)
Refabrication Development - Task 300

DEVICE FOR SAMPLING HTGR RECYCLE FUEL PARTICLES

R. R. Suchomel and W. J. Lackey

Date Published: March 1977

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DEVICE FOR SAMPLING HTGR RECYCLE FUEL PARTICLES

R. R. Suchomel and W. J. Lackey

ABSTRACT

Devices for sampling High-Temperature Gas-Cooled Reactor fuel microspheres were evaluated. Analysis of samples obtained with each of two specially designed passive samplers were compared with data generated by more common techniques. A ten-stage two-way sampler was found to produce a representative sample with a constant batch-to-sample ratio.

INTRODUCTION

High-Temperature Gas-Cooled Reactors (HTGRs) depend on fuel recycle for efficient fuel use. The fuel recycling process consists of separating ^{233}U, bred from ^{232}Th, out of spent fuel and refabricating it into new fuel elements. The total refabrication process is quite complex and is subdivided into a number of systems. In one of these systems, uranium-loaded, carbonized resin microspheres are converted and then coated with multiple layers of pyrolytic carbon and silicon carbide. Certain product attributes must be measured after conversion to the oxycarbide, after deposition of the porous buffer and the inner dense isotropic carbon layers, after application of silicon carbide layer, and again after deposition of the outer dense isotropic layer completes the Triso coating process. The particles are quite small; nominally only 360 μm in diameter before conversion and 660 μm in diameter after final coating. A commercial size coating furnace will process between 3 and 10 million microspheres per batch, but inspection tests are usually done on either one or, at most, a small number of particles. Thus, methods must be available to routinely obtain small representative particle samples from large batches. This report describes the development of the required sampler.

SAMPLER CRITERIA

The fundamental requirement for any sampling device is that it yield a sample representative of the parent batch. Sophisticated HTGR fuel inspection techniques have been developed to determine batch properties from a small sample.[1] For example, the mean particle diameter of a sample of fuel particles can be determined to within ±2 μm with 95% confidence. Such a number is meaningful only if there is equally high confidence that the sample is representative of the batch.

Although settling and segregation are common in batches of fuel particles, each particle of the batch must have an equal probability of being selected in a representative sample.[2] This requirement prohibits the use of top sampling, bottom sampling, or other such grab-type sampling techniques.

Quality control of refabricated HTGR fuel presents a unique problem because of the unavoidable presence of some ^{232}U. High-energy gamma radiation from the decay of radioactive daughter products of ^{232}U requires that fabrication and quality control testing be done in heavily shielded facilities. On-line inspection equipment must be located in the refabrication hot cell behind several feet of concrete. Thus, automated sampling equipment is required. Also, in some stages of the process, the fuel kernels are pyrophoric and must be contained in an inert atmosphere to maintain the desired chemical composition.

Since the same sampling device may be used during several steps of the coating process, the devices must also fulfill several physical requirements. For example, cross-batch contamination or particle breakage would not be permitted within the sampling device. Use in a hot cell and the added containment imposed by the inert enclosure mandates that the device be highly reliable and not subject to frequent maintenance or repair. Because of these restrictions it was decided that only passive samplers, that is, devices having no moving parts, would be considered.

SAMPLING DEVICES

Ten-Stage Sampler

Two passive sampling devices have been developed for HTGR fuel particle inspection. Both samplers use gravity to move the microspheres. The first sampler developed was a ten-stage conical splitter device (Fig. 1). The microsphere inlet tube, with feel funnel, is at the top of the sampler and both the sample and batch outlets are at the bottom.

The sampler consists of ten stages identical in function and more or less identical in appearance (Fig. 2). In each stage, batch division is done by passing the particles onto the apex of the cone, the cone having an included angle of 60°. The particles are directed to the apex of the cone either by a 6.4-mm-diam (1/4-in.) hole in the bottom of the preceeding stage, or in the case of the first stage, a 9.5-mm-diam (3/8-in.) hole in the inlet assembly. As the particles deflect off the cone, they are directed toward the base of the cone. The perimeter of the base is divided into eight sections, each of which subtends an angle of 45°. Four sections are sampling slits located 90° apart and four are ejection ports also 90° apart. The sampling slits include 180° of the circumference so half the batch falls through the slits to the next stage and half is rejected. The rejected particles pass through the exit windows (Figs. 1 and 2), reflect off the outer cylindrical shell, and fall to the batch outlet. The remaining particles, those emerging from the sampling slits, are directed toward the axial centerline of the next stage. After the first stage, half of the batch remains. The sample is cut inhalf by each succeeding stage and after the tenth stage, $(1/2)^{10}$ or 1/1024 (0.0977%) of the original batch falls through the slits.

To prevent any directional bias in the sampling path, each conical splitter is rotated 45° relative to the one above it. The four equally spaced sampling slits provided in each stage further prevent directional bias. Baffles radially surround all but the top stage to inhibit another kind of bias. Operating in conjunction with the deflectors built into the

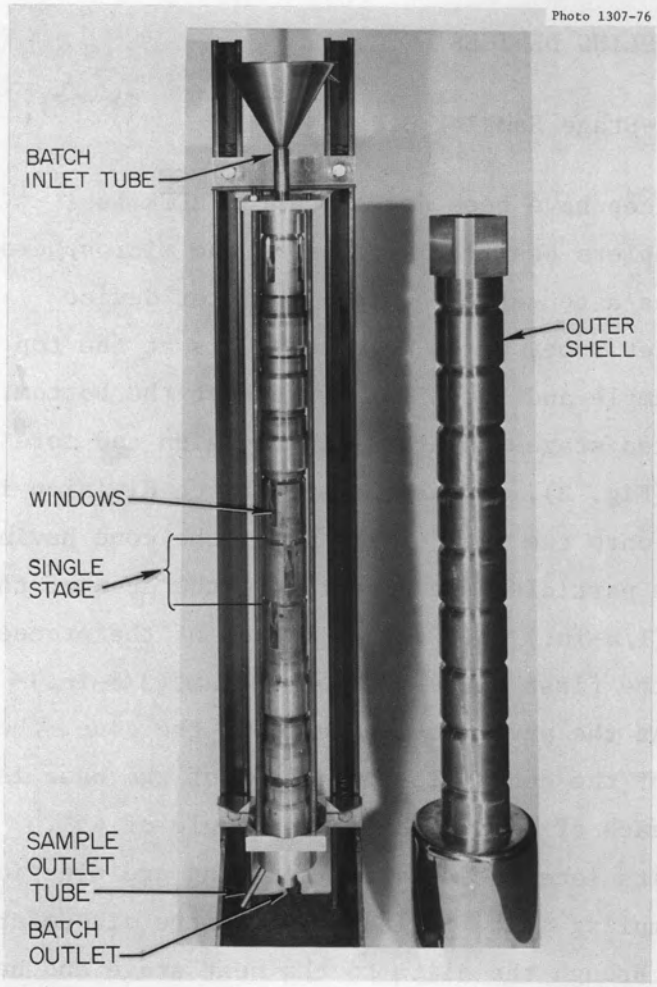

Fig. 1. Ten Stage Passive Sampler Assembled on Test Stand. The outer cylindrical shell and several baffles have been removed for illustration.

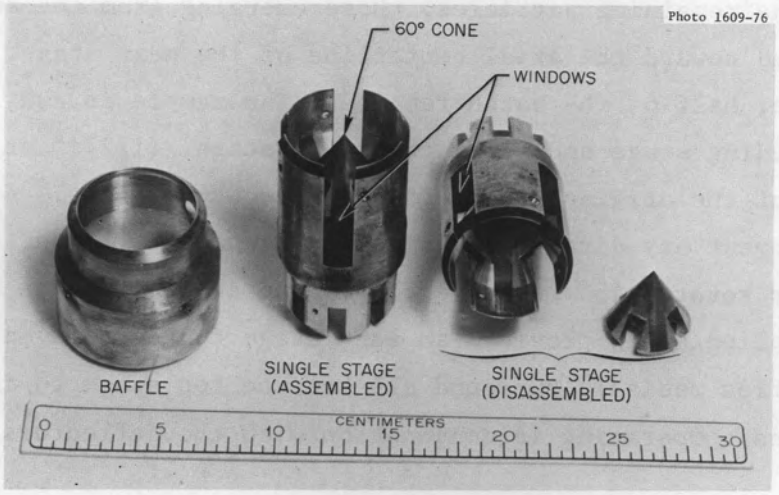

Fig. 2. Ten Stage Passive Sampler Components.

outer shell, these baffles prevent microspheres in the collecting annulus from finding a way back into any lower stage. Without these baffles in place, particle re-injection into the sampling stream was frequently observed which substantially altered the sampling ratio. The baffle and deflector system were also designed so that no substantial line of sight path existed from the exit port windows of any stage to the bottom collection funnel. The introduction of baffles reduced particle velocities and thereby eliminated a possible cause of particle breakage.

Since this sampler is composed of many identical stages, replacement parts could be easily and rather inexpensively stocked. In addition, if process or inspection changes required that the sample size be altered, one or more stages could be either added or removed to produce the desired sample size. The addition of one stage would halve the final sample size while the removal of a stage would double the sample size.

Three-Stage Sampler

The same conical splitting technique was employed in the design of a passive three-stage particle sampler (Fig. 3). As with the ten-stage sampler, gravity feed is used throughout the device. The inlet tube is atop the sampler and both the outlet and sample tubes are located on the bottom.

Splitting of the sample is done in a manner much like that used in the previously discussed unit. In each stage, particles are directed through a collimator onto the apex of a cone (included angle 120°) from which they fall and bounce toward either the sampling slits or exit windows (Fig. 4). The retained portion is comprised of that fraction of the particles which flow between knife edges; these particles subsequently fall through the slots in the cone and are directed toward the next stage. Three degrees of arc are subtended within each of the twelve gates for each stage; hence, a total of 36° out of 360° is subtended by the gates in each stage. Thus, 10% of the incoming particles are selected to continue on to the next stage while 90% are ejected to the outer collection annulus. One-tenth of the parent batch is retained after the first stage,

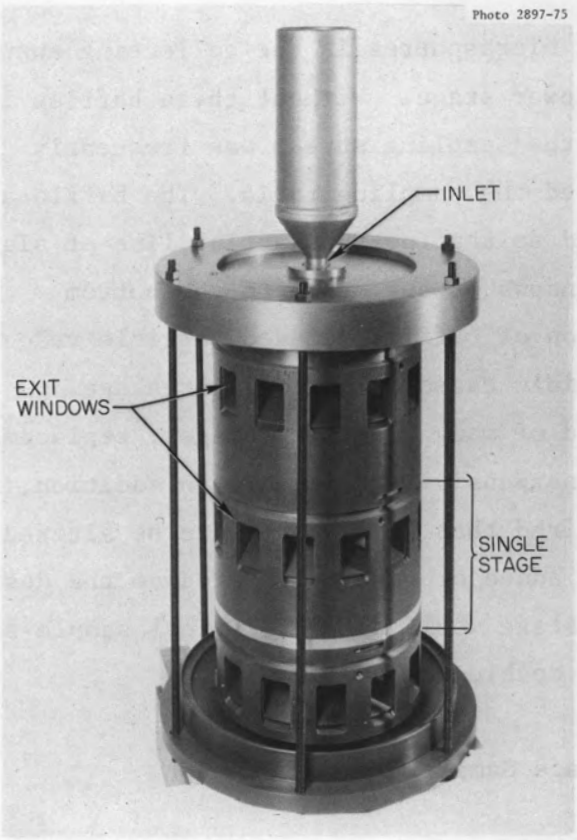

Fig. 3. Three-Stage Passive Sampler with Baffles for the Lower Two Stages and the Outer Cylindrical Containment Shell are Removed for Better Illustration.

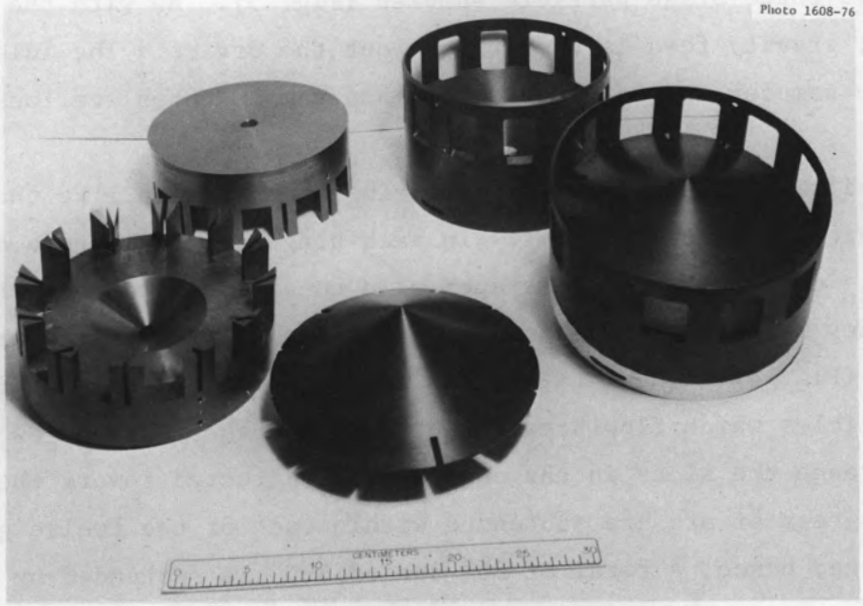

Fig. 4. Components Needed to Complete Two Stages of the Three-Stage Passive Sampler. The two splitter gate units (on the left) show the knife edges that separate the retained portion of the batch from the ejected portion.

1/100 is kept through the second stage, and 1/1000 of the batch is retained as a sample at the bottom of the sampler.

Baffles are located around the exit windows of the second and third stages to ensure that particles removed from the sampling stream do not re-enter it at a lower stage.

This sampler was designed to easily accept interchangeable collimators above each cone. Four orifice sizes of such collimators are available with which batch flow could be regulated to each separate stage (Table 1). The rate limiting flow is through the top collimator since only 10% of the batch reaches the second stage.

Table 1. Particle Feed Rates for Three-Stage Sampler

First Stage Orifice Diameter (cm)	Flow Rate (liter/min)
1.27	1.70
0.95	0.60
0.64	0.17
0.48	0.08

Other Sampling Techniques

Prior to the development of the multistage samplers, samples for fuel particle inspection were obtained using single-stage laboratory size conical splitters and chute rifflers. A single pass through either instrument reduced the sample to one-half its previous size. The conical splitter, used to reduce the sample size of large batches, is similar in principle to the two multistage samplers. The batch is poured onto the apex of a cone and the particles fall into gates located around the base of the cone to separate the initial batch into two segments. After sufficient passes have been made through the conical splitter to reduce the sample to about 50 g, further reduction is made using a chute riffler. This apparatus consists of a V-shaped trough containing 16 chutes feeding alternately two trays placed on either side of the trough. Again each

pass through the riffler reduces the sample size by one-half. When using either of these devices, care was taken to alternate portions selected for further sampling. This was done to prevent any systematic bias that might be inherent in the sampler. Naturally, because of the many cycles required to generate the correct sample size, these devices are not suited for remote particle sampling.

DATA FROM COMPARATIVE TESTING

The data in this section of the report are from comparisons of the performance of the various sampling devices. Sampler comparison and evaluation is not a simple task as no absolutes or standards are available. Since it is not feasible to determine the physical characteristics of every particle in a batch, a sample must inevitably be extracted using some device or technique. The testing reported here evaluated sampler performance on the basis of several easily measured characteristics. Sample quality with respect to these characteristics should be a fair indicator of overall sampler operation. Other microsphere characteristics could probably have been determined for the samples; however, it was felt that the inaccuracies associated in such measurements would mask any real sampler bias.

Testing was done using batches with different outer coatings and mean diameters. Both of the previously described passive samplers were tested. When appropriate, samples were also obtained using the laboratory riffler technique for comparative purposes.

Any type of grab sampling was expected to yield unrepresentative samples, since particle segregation should be increased by the many pneumatic transfers made during the coating operation. To verify that grab sampling would not yield satisfactory samples for product inspection, a number of samples were obtained by this method both by scooping particles from the top of a storage hopper and by intermittant sampling as particles flowed out of a hopper.

Mass Sampling Ratios

The first tests conducted with the as-fabricated samplers were determinations of batch-to-sample mass ratios. Such tests were used to observe whether experimental results approached design specifications. As a result of the initial tests, both devices were modified to include the baffles described previously. The baffles were necessary since in their absence particles bounced back into the sample stream causing the sample to be large. Widely flucuating batch-to-sample mass ratios of between 200 and 400 were observed for both devices without baffles in place. Also, without the baffles to slow the fall of the particles, some coatings cracked.

Following the modifications, further testing showed that both samplers were producing reasonable batch-to-sample ratios (Fig. 5). Confidence intervals were calculated since even if the devices were operating ideally one could not, statistically, expect to observe a constant ratio. A confidence interval shows tha range of batch-to-sample ratios that should statistically be expected from a propertly designed and operating sampler. These intervals were established based on an average test batch size of two million particles having an assumed normal distribution.

The ten-stage device produced excellent results; all but one of the 30 test runs lie within a 95% confidence interval of the design ratio value of 1024 (Fig. 5). Ratios were found to be independent of flow rate and particle type for this sampler.

However, the three-stage sampler produced ratios that were not independent of particle type and flow rate. It is assumed that various particle properties such as density, diameter, elasticity, and coating type influence internal flow within the sampler. For example, a very elastic particle should be more likely to bounce out of the sampling gate, or a very large particle should be more likely to strike the wall of such a gate and be deflected.

An obvious dependence on flow rate was observed for the three-stage sampler which results from the basic geometry of the sampling gates. As

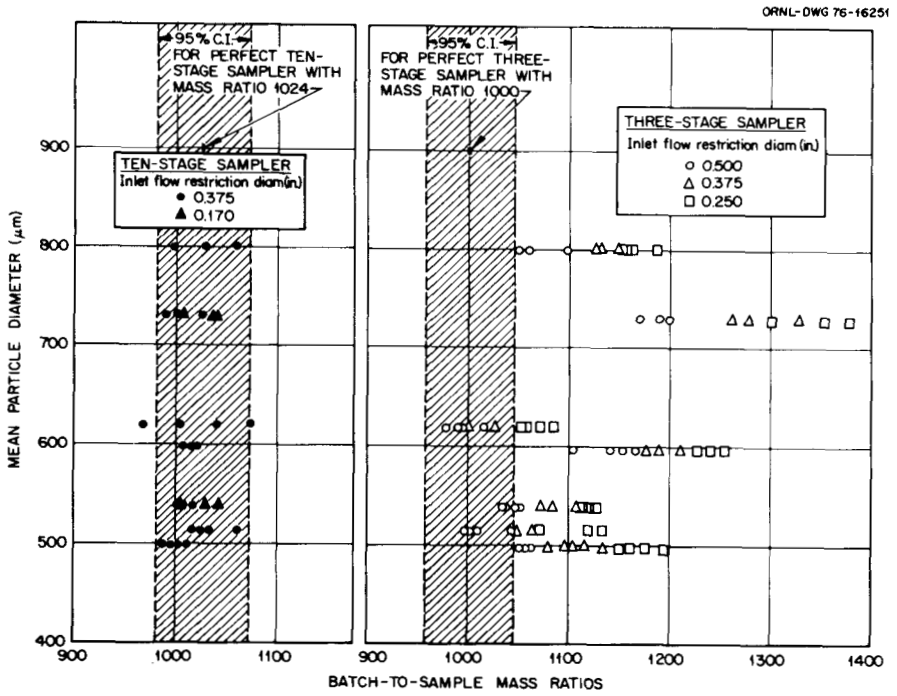

Fig. 5. Batch-to-Sample Mass Ratios Determined for Various Particle Batches Using Both Sampling Devices. Confidence intervals (C.I.) are included since even if the devices were operating ideally one could not, statistically, expect to observe a constant ratio.

the gates are quite narrow, about 10 particle diameters wide, a significant number of particles will strike the knife edges and bounce back upward. The mean free path of such recoils determines the fraction of such particles that escape from the sampling gate, and the path length in turn is determined by the flow rate. That is, a higher flow rate produces a shorter mean free path that generates a sampling ratio close to the design value. It was feared that with an excessively high flow rate (i.e., an inlet diameter greater than 1.27 cm) the homogeneity and randomness of the sampling process would be destroyed, especially for small batch sizes, so no such flows were investigated.

The consistency of batch-to-sample mass ratios is not an important criterion in itself, but it does indicate whether the sampler is working as expected. The true merit of a sampling device is measured by the representative nature of the sample with respect to sample properties such as mean particle diameter, particle size distribution, and particle density.

Mean Particle Diameter

An investigation was made of the ability of the two samplers to produce multiple samples having consistent mean particle diameters. For samples obtained with each device, the average diameter was determined with a particle size analyzer[3] (Figs. 6 and 7). Confidence intervals

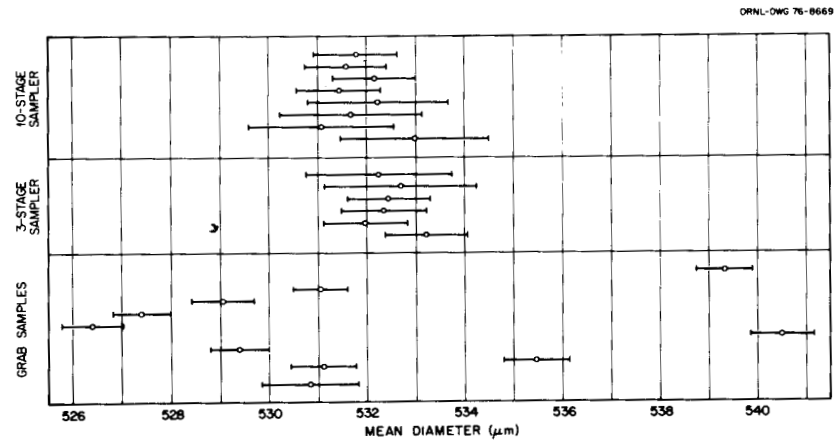

Fig. 6. Confidence Intervals for the Mean Particle Diameter of Inner Low-Temperature Isotropic Ninety-five Percent Coated Particles Sampled Using Three Different Techniques.

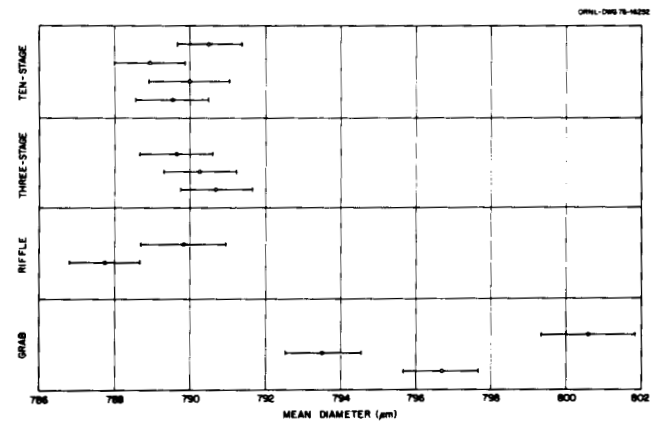

Fig. 7. Ninety-five Percent Confidence Intervals for the Mean Particle Diameter of Silicon Carbide-Coated Particles Sampled Using Four Different Techniques.

were calculated from the number of microspheres in each sample, and the standard deviaiton associated with the particle size analyzer data. There was close agreement between the samples collected with the three-stage and the ten-stage samplers (Fig. 6). This agreement contrasts greatly with the data obtained by grab sampling from the same parent batch. The grab sample data are not only different from that of the samplers but are also internally inconsistent. These grab sampling results are typical for this technique and show why such a technique is not suitable for sampling HTGR fuel particles.

Other batches of microspheres produced data similar to that already described. A batch of silicon carbide coated material was sampled several times with each of the two samplers, the laboratory riffler, and the grab sample technique. All of the samples, with the exception of the grab samples, showed good agreement (Fig. 7). Buffer-coated and Trico-coated batches were also used to test the sampling devices. The data showed no significant differences in mean particle size in samples obtained from either of the two samplers or the laboratory riffler.

An effort was made during this sampling to determine if a correlation existed for any of the devices between the flow rate and the mean particle size of the samples. No correlation like that found between flow rate and batch-to-sample mass ratio for the three-stage sampler was found. There was no evidence that mean particle diameter depended on flow rate.

Particle Size Distribution

The particle size distribution of many samples was also measured to find if the distribution was altered by changing sampling technique or flow rate. Cumulative distribution data (Fig. 8) show that the general shape of the curves was not affected by a change in sampling devices. For the case shown, the plot obtained from the grab sample is shifted relative to the other curves but has the same general shape with equivalent tails. The horizontal shift is consistent with the larger mean particle diameter of the grab sample. A close inspection of the data does resolve slight differences in the shape of the curves.

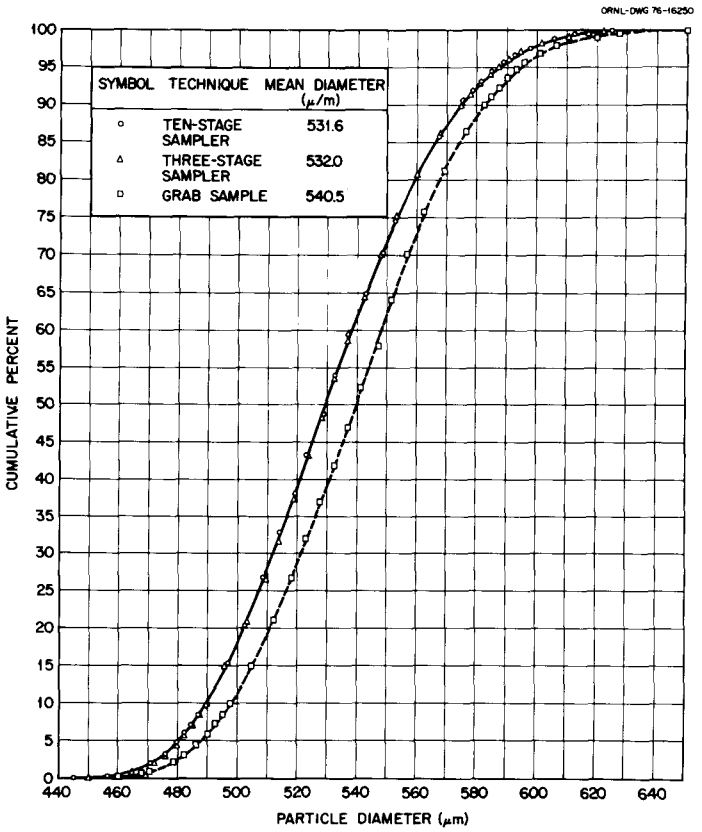

Fig. 8. Cumulative Size Distribution of ILTI-Coated Particles Samples Using Three Different Techniques.

Nevertheless, the overall similarity of the three curves might incorrectly justify the acceptance of the grab sample as a representative batch sample. The relative particle size distribution within a sample was not measurebly affected by flow rate for either sampling device.

Particle Density

As a final evaluation of the ability of the various sampling devices to produce a representative sample, density measurements were made on eight samples from the same buffer-coated parent batch. Nearly identical values were measured on samples from the two multistage samplers (Table 2). The laboratory riffler also provided similar data. However, measurements from the grab samples show greater dispersion but are insufficient for further conclusions.

Table 2. Particle Density as Measured by 34 MPa (5000 psi) Mercury Pycnometry for Biso-Coated Particle Samples Collected by Four Different Sampling Techniques

Sampling Technique	Particle Density (g/cm^3)
Ten-stage sampler	1.9912 1.9897
Three-stage sampler	1.9886 1.9884
Laboratory riffler	1.9886 1.9859
Grab sample	1.9918 1.9642

CONCLUSIONS

Two multistage sampling devices were developed that produce particle samples found to be representative of the parent batches. Both samplers fulfilled the design criteria in that they had no moving parts and they could be sealed to maintain an inert atmosphere. Experiments showed that both devices required baffles to attain appropriate batch-to-sample mass ratio values. With the baffles in place, both samplers demonstrated acceptably low particle holdup, abrasion, and breakage. Grab sampling was shown to be an unacceptable technique for particle inspection.

The only test for which the results from the two passive samplers varied significantly was the batch-to-sample mass ratio determination. While these test results were not indicative of the worth of the resultant samples, they do suggest that inspection requirements might be better met with the ten-stage device. A situation could conceivably develop in which the three-stage, 1000:1 ratio sampler would yield samples either too small for accurate analysis or too large. Such difficulties are not envisioned with the more uniform ten-stage, 1024:1 ratio sampler. The ten-stage device is also more versatile since stages may be added or removed as sample requirements change. Adding or removing one stage would

permit doubling or halving the sample size. When altering the three-stage device, the smallest allowable variation affects sample size by a factor of 10. The ten-stage unit is also lighter and hence easier to handle of the two units. For these reasons, devices similar to the ten-stage sampler are preferred for sampling coated fuel particles during the refabrication of HTGR fuel containing ^{233}U.

ACKNOWLEDGMENTS

The work of two Co-op students, J. S. Stewart and L. E. Davis, in doing the sampling experiments is greatly appreciated. Sample analysis was done with the help of A. G. Mason and L. J. Turner. The manuscript was edited by G. W. Griffith and prepared for publication by Susan Hanzelka of the Metals and Ceramics Reports Office.

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