Conf-9209265--1

CHARACTERIZATION OF COMMERCIALLY PURE ALUMINUM POWDER FOR RESEARCH REACTOR FUEL PLATES*

V. D. Downs
Babcock & Wilcox
Lynchburg, VA USA

and

T. C. Wiencek
Argonne National Laboratory
Argonne, IL USA

ANL/CP--77513

DE93 003001

To be presented at the
1992 International Meeting on
Reduced Enrichment for
Research and Test Reactors

September 27-October 1, 1992
Roskilde, Denmark

*Work supported by the U.S. Department of Energy, Office of International Affairs and Energy Emergencies, under Contract No. W-31-109-ENG-38

CHARACTERIZATION OF COMMERCIALLY PURE ALUMINUM POWDER FOR RESEARCH REACTOR FUEL PLATES*

V. D. Downs
Babcock & Wilcox
Lynchburg, VA USA

and

T. C. Wiencek
Argonne National Laboratory
Argonne, IL USA

ABSTRACT

Aluminum powder is used as the matrix material in the production of uranium aluminide, oxide, and silicide dispersion fuel plates for research and test reactors. Variability in the characteristics of the aluminum powder, such as moisture content and particle-size distribution, influences blending and compacting of the aluminum/fuel powder.

A detailed study was performed to characterize the physical properties of three aluminum powder lots. An angle-of-shear test was devised to characterize the cohesiveness of the aluminum powder. Flow-rate measurements, apparent density determination, subsieve analysis, surface area measurements, and scanning electron microscopy were also used in the study.

It was found that because of the various types of commercially available powders, proper specification of powder variables will ensure the receipt of consistent raw materials. Improved control of the initial powder will reduce the variability of fuel-plate production and will improve overall plate reproducibility. It is recommended that a standard specification be written for the aluminum powder and silicide fuel.

1 INTRODUCTION

The success of fuel-plate production depends to a great extent on the characterization and control of the initial raw materials. Reproducibility of the fabrication processes is best achieved by the use of consistent raw materials. Often the problem is not so much how to control the raw material, but rather knowing what to control. Powders have many characteristics, some interrelated, that must be considered. Among these are shape, surface area, particle structure, composition, bulk density, apparent density, size

*Work supported by the U.S. Department of Energy, Office of International Affairs and Energy Emergencies, under Contract No. W-31-109-ENG-38

distribution, compressibility, flow-rate, and average size. Only a few may be critical for a given application, but the final selection can be made only if all of the major characteristics are understood. Standard testing methods allow the quantitative measurement of certain of these physical properties; some of these methods are reviewed in this paper and, in addition, nonstandard test methods are also presented.

The properties of powders can change over time between manufacture and actual use. The powders studied were stored in "sealed" (not airtight) containers that either contained air or were under a vacuum. Specific trends due to age of the powder were not considered unless powder conditions, such as "degassed and 24 hr exposure to air", were specified. Due to differences in production facilities, a comprehensive study of powder characteristics under all environmental conditions was not possible. Therefore, the goal of this paper is to show general trends in powder characteristics.

2 BACKGROUND

Aluminum powder, in particular Alcan 101, is used extensively in the Research and Test Reactor program. Aluminum powder is the matrix material used in the picture-frame method for manufacturing aluminum fuel plates. In this process, aluminum powder is blended with fuel particles, the mixture is poured into a die cavity and pressed to form a compact, and the compact and aluminum alloy frame is assembled and then hot rolled into a fuel plate.

Discussions with one major powder supplier, Alcan Toyo America, Inc., revealed two classifications of aluminum powder: conventional and spherical. Conventional is molten aluminum atomized in air; the result is a globular powder form. This powder's nonspherical shape is due to the formation, during cooling, of surface oxide films that distort the internal liquid aluminum as it solidifies. Alcan lists 13 types of conventional powders, with percentage of powder passing through a 325-mesh (44 μm) sieve ranging from 35-99%. Spherical powder is produced by atomization in an argon atmosphere containing 3-4% oxygen for controlled oxidation. This powder is used primarily for armament. The heat generated by the powerful exothermic reaction of oxidizing aluminum, combined with an explosive, has a devastating effect. The low percentage of oxygen produces a thin protective oxide layer that permits safe handling of the powder when exposed to atmosphere. Alcan lists 14 different types of spherical powder, with the average particle size ranging from 3 to 35 μm. Particle-size distributions for selected types are plotted in Figures 1 and 2. The graphs show particle-size distribution based on the percentage that is finer than a certain size. For instance, in Figure 1, approximately 90% of the Type 101 particles are smaller than 82 μm. For spherical powder, however (Figure 2), 90% of the type X-86 particles are smaller than 65 μm.

3 SAMPLING

Given the large population of particles in most powdered metals, a sampling method must be used to achieve statistically meaningful results. Careful attention must be given to collecting unbiased samples. Recommended techniques[1] provide samples that are representative of the total powder population. However, the science of sampling is beyond the scope of this paper. The key here is the use of a good sampling method.

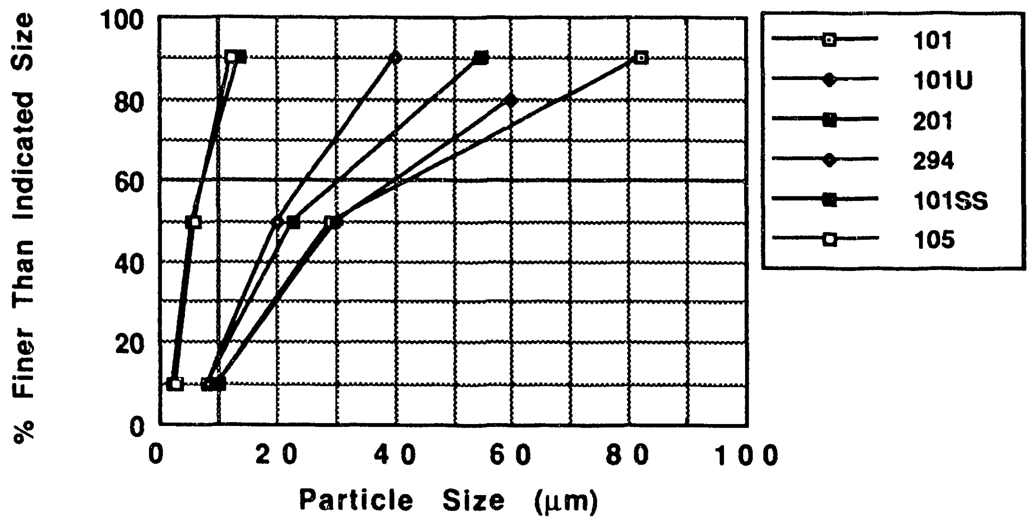

Figure 1. Particle-Size Distributions of Conventional Aluminum Powders

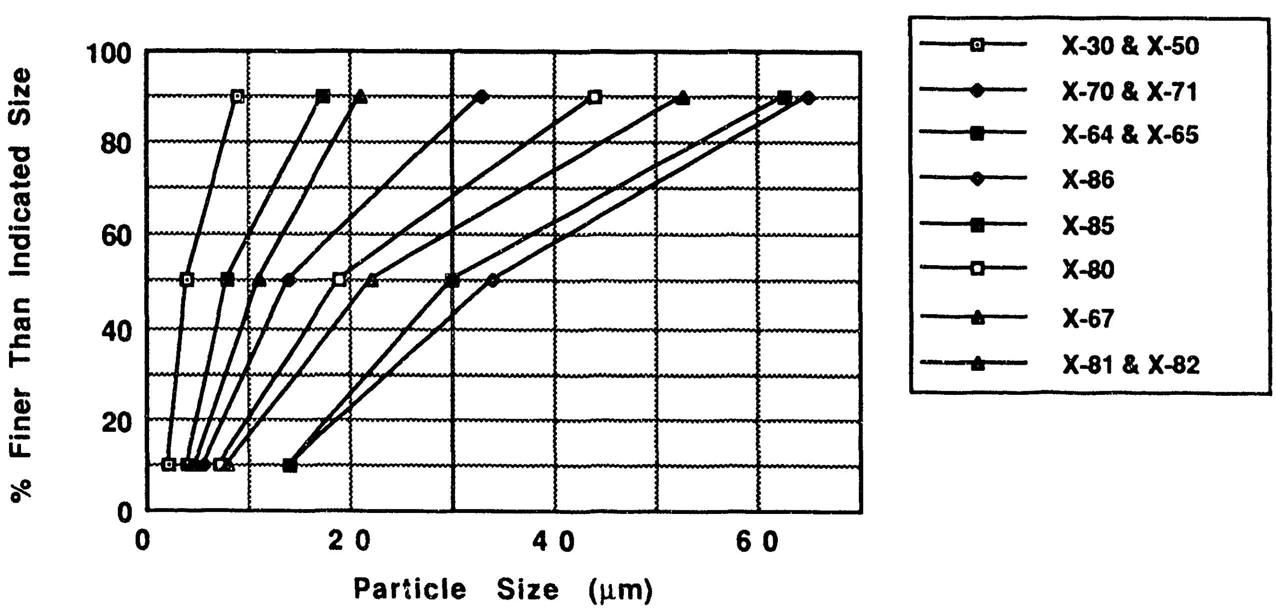

Figure 2. Particle-Size Distributions of Spherical Aluminum Powders

4 CHARACTERISTICS OF STANDARD POWDERED METAL

4.1 Analysis of Powder Size by Sieve Tests

Particle-size analysis[2] is a common, rapid, and easy test for powdered metals. Data are usually generated by sieving the material through a series of screens. Results are given by the weight percentage of material above a certain diameter that will not pass through the screen. The screens range from large >0.3 cm (6-mesh) to the smallest commonly used standard sieve size of 37 μm (400-mesh).

The majority of the aluminum powder used for fuel plate production is smaller than 44 μm. A common powder is Alcan Type 101. This powder has a range of 75-85 wt.% <44 μm particles. Thus, a common specification is the requirement of a minimum amount of <44 μm-size powder. However, this can be a misleading specification because sieve-test results are greatly influenced by the time of sieving and the intensity of vibration. Sieve tests are only valid if the forces that tend to agglomerate powders are overcome by the mechanical action of the sieve shaker. Forces that bind particles together result from (a) friction due to the physical shape of particles, (b) Van der Waals forces acting on the powder itself, and (c) adsorbed surface-water molecules.

Small particles are more influenced by Van der Waals forces than are larger particles due to the smaller mass per surface area. This force is further enhanced by water molecules attached to small particles. To prevent these forces from influencing powder-particle-size determination, wet sieving is recommended. A low-viscosity liquid, (e.g., acetone) can be used as a carrier vehicle to counteract the forces acting on the dry powder. This general test is very useful for describing aluminum powder, but should not be the sole specification for production powder.

Three lots of aluminum powder were extensively studied in this review. Table 1 summarizes analytical results of the as-certified Alcan powder.

Powders from the three different batches of as-received aluminum powder were individually blended and then dry sieved at Argonne National Laboratory (ANL) for comparison with the manufacturer's data. The results are given in Figure 3 for the coarse (>44 μm) particles. The fines (<44 μm)) were 78, 84, and 77% for Lots 10003, 1770 and CO-713, respectively. The data agree well with those in Table 1. Two characteristics are of major concern in controlling the feedstock powder. First, a significant amount of powder is present in the 53-125 μm sizes. Control of this powder distribution may be necessary to improve fuel core reproducibility. Second, the range of allowable <44 μm powder for Alcan 101 aluminum is 75-85%. Although all three lots fall within this range, the differences between lots are significant and may affect the final powder properties. Overall, Figure 3 shows significant differences in powder-size distributions. These differences may lead to variance in final fuel-plate homogeneity.

Table 1. Manufacturer's Certified Analytical Results for the Three Lots of Alcan Type 101 Aluminum Powder

Characteristic	Lot 10003	Lot 1770	Lot CO-713
Free Aluminum (wt.%)	99.5	99.3	99.4
Volatiles (wt.%)	0.01	0.03	0.02
Oil and Grease (wt.%)	0.01	0.10	0.01
Screen Analysis			
-100 Mesh (<150 μm,%)	100	100	100
-200 Mesh (<74 μm, %)	94.0	96.8	94.8
-325 Mesh (<44 μm, %)	81.8	85.0	77.0
+325 Mesh (>44 μm, %)	18.2	15.0	23.0

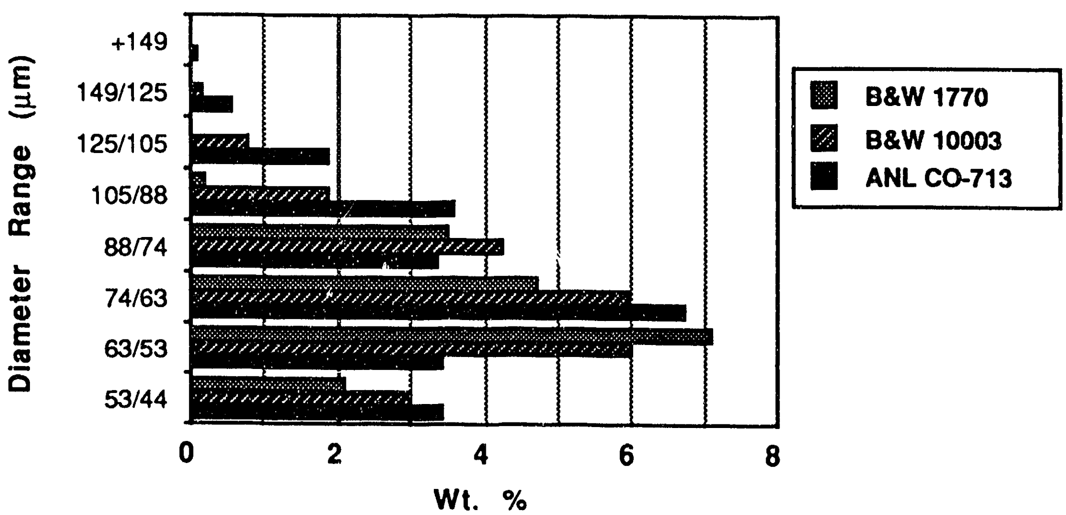

Figure 3. Distribution of 149-44 μm As-received, Blended Powder from Lots 1770, 10003, and CO-713 (B&W = Babcock & Wilcox)

Another related powder specification is average particle diameter. This value is not very informative without any knowledge of the specific particle size distribution. Specifying both a minimum of <44 μm percentage and an average particle diameter is recommended as a method to decrease powder variability.

4.2 Analysis of Subsieve Particle Size

Because the majority of the aluminum powder is <44 μm (-325 mesh), a technique other than standard mesh sieving must be used. Various methods can be used to determine the distribution of subsieve-size particles in a metal powder.[3,4] Results were generated on a Leeds and Northrup Microtrac particle-size analyzer, which can analyze powder distributions with sizes <44 μm. This machine uses a laser beam to measure 0.2 to 42 μm particles dispersed in water. Figure 4 gives the results of these tests for Lots 10003 and 1770.

4.3 Measurement of Apparent Density

Standard devices are readily available for determining the flow-rate and apparent density of free-flowing and non-free-flowing powders.[5-7] The most commonly used instrument, the Hall flowmeter, is an accurately machined funnel. For non-free-flowing powders, a Carney flowmeter (a larger-hole version of the Hall flowmeter) is used. The time required for a weighed sample of powder (usually 50 g) to flow from the funnel into a cup is determined and the flow rate is expressed in seconds, or g/min if a nonstandard weight of the sample is used.

The Hall flowmeter can also be used to measure another commonly reported characteristic for metal powders, i.e., apparent density. The powder is allowed to flow through the flowmeter into a container of known volume (usually 25 cm^3). After the powder is struck level, it is weighed and the result is given in g/cm^3.

The three as-received lots of aluminum powder were sampled at the top, center, and bottom of the shipping container and the results are given in Figure 5. Significant differences between the three lots were measured. Apparent density decreased from the top to the bottom of the can, possibly because of segregation of the powder during shipping and handling. The effect of particle size on apparent density was also measured. In general, coarser particles led to reduced apparent densities.

Due to poor flow of the powders, no Hall or Carney flow times were measured on the three lots of powder. It is recommended that flow time not be used as an aluminum powder specification.

Another related characteristic is tap density, a measure of the density of a powder sample after a specified number of taps or controlled vibrations. Because there are no established standards, this method was not used during the characterization and is mentioned here only to make others aware of it.

4.4 Measurement of Surface Area

A standard method used to determine the surface area of powders is the Brunnauer, Emmett, and Teller (BET) test. This nondestructive test determines

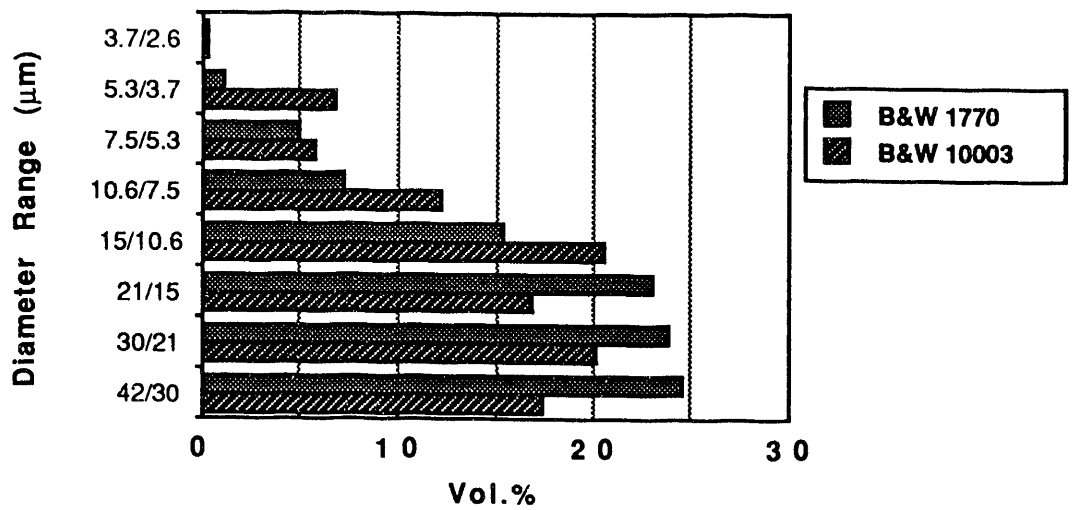

Figure 4. Results of Subsieve Analysis

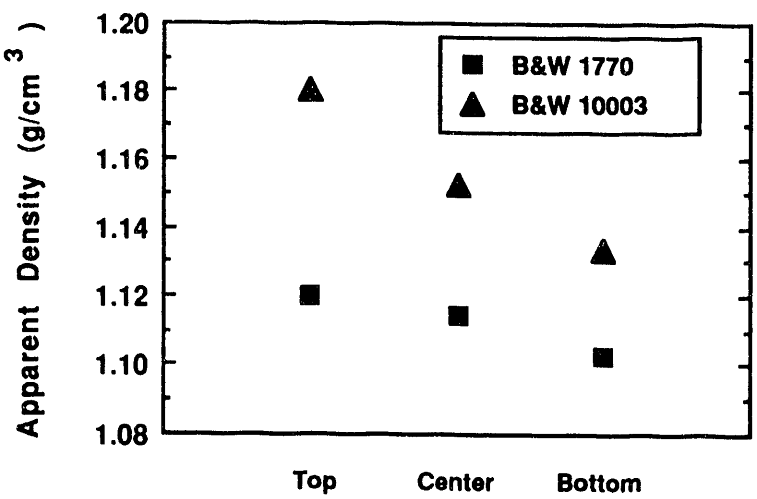

Figure 5. Comparison of Apparent Density of B&W Aluminum Powders

the surface area by measuring the amount of absorbed gas on a known weight of powder. This sorption method relies on the fact that under certain circumstances molecules may be adsorbed onto the surface of powder particles to form monomolecular layers. To obtain accurate results from the standardized methods[8] that have been developed, the proper gas must be used.

Surface areas in the range of 0.02 to 5.0 m^2/g area (111-0.44 μm average particle size) are measured with krypton gas as the adsorbate. In the range from 3 to 100 m^2/g (0.74-0.02 μm average particle size), nitrogen gas is used.

For surface-area measurements, a Micromeritics Instrument Corporation FlowSorb II 2300 was available. However, due to scheduling and equipment problems only limited measurements were taken. Because of the preliminary nature of the information, the results are not included in this report.

5 NONSTANDARD TESTS FOR POWDER METAL CHARACTERISTICS

5.1 Angle of Shear

Cohesive strength is a critical parameter in predicting blending and die-loading characteristics of powders. Both blending and die loading directly affect the homogeneity of the final fuel core. The angle of shear (which is similar to angle of repose) determines the relative cohesive strength of the aluminum powder. No simple and inexpensive standardized test was found that could provide the necessary information. A simple test was developed that uses a bottom-sieve testing catch pan. A fine-grit abrasive paper was placed in the bottom of the pan to ensure that the relative shear forces within the powder will be determined, rather than sliding friction between powder and pan. Powder was shaken onto the abrasive paper to form a rectangular section. One edge of the pan was placed against a reference mark, while the opposite edge was slowly tilted upward alongside a vertical ruler. When the powder moved, trigonometric principles were used to calculate the angle of shear. Results of testing at ANL and B&W are shown in Table 2.

Annealed powder was maintained in vacuum storage until needed. The tests at ANL were performed in the winter with typically 25-35% relative humidity. Testing at B&W took place during the summer in a humidity-controlled room with less than 64% relative humidity. Humidity was found to be an important factor in determining angle of shear. The powder adsorbed moisture immediately upon exposure to air. Moisture adsorption continued to affect the angle of shear until, in the case of Lot 10003, the pan was raised to the maximum possible angle vertically and the powder remained intact as a pile. It has not been determined why one powder lot was affected more by moisture than the others.

5.2 Surface Morphology

Three center samples were examined by scanning electron microscopy (SEM). Two different size ranges, >63 μm (+230 mesh) and <44 μm (-325 mesh), from each lot were studied. No significant differences in shape or surface were noted after visual inspection at magnifications of 75 to 800X for all six samples. All three lots were granular or complex-nodular (not spherical), which is characteristic of air-atomized powder. A typical SEM photograph of the aluminum powder is shown in Figure 6. For production-powder analysis, an imaging program such as Image 1.44 from the USA National Institutes for Health could be used to analyze for the average particle size and distribution.

Table 2. Comparison of Angle of Shear for B&W and ANL Aluminum Powder

Sample	B&W Lot 10003	B&W Lot 1770	ANL Lot CO-713
ANL Tests			
Top[1]	45°	45°	Not Measured
Center[1]	45° (46.0°)[2]	48°	45°
Bottom[1]	44°	47°	Not Measured
B&W Tests			
Center[1]	71°	49°	Not Measured
Center[2]	72°	46°	Not Measured
Center[3]	85°+	59°	Not Measured

[1]As-received powder.
[2]As-degassed powder.
[3]After 10-day exposure to room air.

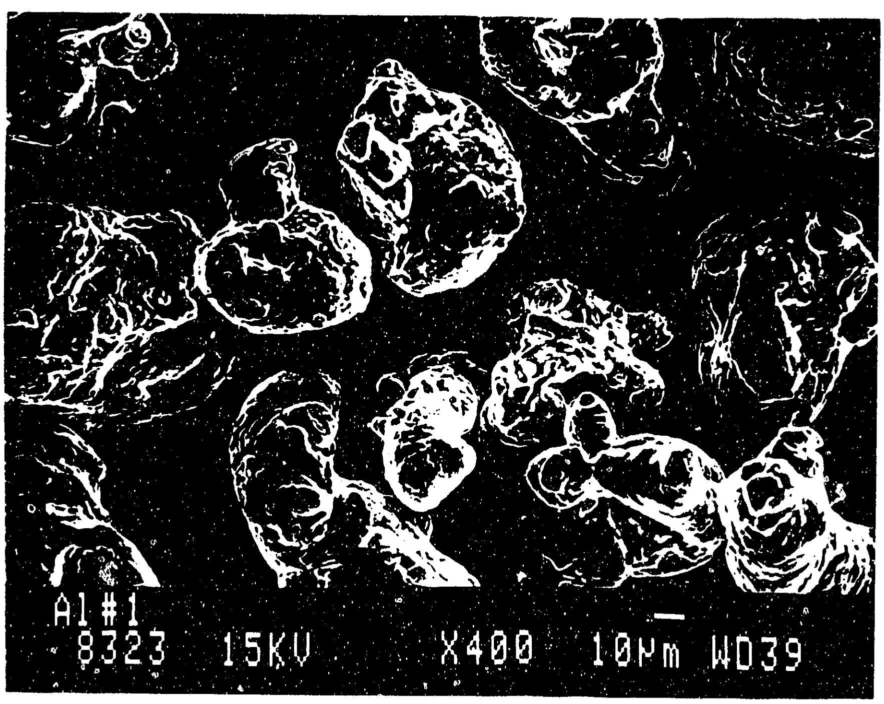

Figure 6. SEM Photomicrograph of >63 μm Lot CO-713 Aluminum Powder

6 SUMMARY AND RECOMMENDATIONS

A precise aluminum powder specification is a key step in consistently manufacturing acceptable powder-metallurgy-type fuel plates. ANL and B&W are working jointly (in conjunction with Oak Ridge National Laboratory and EG&G Idaho National Engineering Laboratory) to establish a standard specification for aluminum powder. It may be beneficial to the Research and Test Reactor Fuel Elements (RTRFE) community to establish an international standard for: powder type (atomized, flake, or granule), particle shape (globular, spherical, acicular, irregular, etc.), range limits for average particle size, particle size distribution, and surface area. An international standard would ensure uniformity in the materials used to manufacture fuel plates.

Receipt inspection and testing of aluminum powder lots are necessary before releasing the powder to the fabrication process. Because the original powder costs make up only a low percentage of the final finished fuel-plate cost, any lot not meeting the desired specifications should be rejected. In addition, a standard specification such as that for plutonium dioxide powder[9] may also need to be developed for the fuel powder.

REFERENCES

1. Standard Test Methods of Sampling Finished Lots of Metal Powders, American Society for Testing and Materials, Standard No. B 215, Vol. 02.05, 1982.

2. Standard Test Method for Sieve Analysis of Granular Metal Powders, American Society for Testing and Materials, Standard No. B 214, Vol. 02.05, 1986.

3. Standard Test Method for Average Particle Size of Powders of Refractory Metals and Their Compounds By the Fisher Sub-Sieve Sizer, American Society for Testing and Materials, Standard No. B 330, Vol. 02.05, 1982.

4. Standard Test Method for Particle Size Distribution of Refractory Metal-Type Powders by Turbidimetry, American Society for Testing and Materials, Standard No. B 430, Vol. 02.05, 1984.

5. Standard Test Method for Apparent Density of Free-Flowing Metal Powders, American Society for Testing and Materials, Standard No. B 212, Vol. 02.05, 1982.

6. Standard Test Method for Flow Rate of Metal Powders, American Society for Testing and Materials, Standard Number B 213, Volume 02.05, 1990.

7. Standard Test Method for Apparent Density of Non-Flowing Metal Powders, American Society for Testing and Materials, Standard No. B 417, Vol. 02.05, 1989.

8 Standard Methods for Chemical, Mass Spectrometric and Spectrochemical Analysis of Nuclear-Grade Uranium Dioxide Powders and Pellets, American Society for Testing and Materials, Standard No. C 696, Vol. 12.01, 1980.

9. Standard Specification for Nuclear-Grade Plutonium Dioxide Powder, Sinterable, American Society for Testing and Materials, Standard No. C 757, Vol. 12.01, 1983.

END

DATE
FILMED
2/3/93