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BALL-MILLING OF GRAPHITE-BASE REACTOR FUEL

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ABSTRACT

The practicality of using ball milling for fine grinding of graphite reactor fuels was evaluated briefly. The results of the study indicate that ball milling is not a feasible approach primarily because the grinding efficiency was low. Consequently, very long grinding times would probably be required to grind rough-crushed coated particle fuels fine enough to rupture the particle coatings (approximately to 100 mesh).

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INTRODUCTION

A grind-leach process for recovering uranium and thorium from graphite-base reactor fuels is being evaluated at Oak Ridge National Laboratory (ORNL). This process, as applied to fuels containing coated particles, consists in grinding the fuel fine enough to rupture the particle coatings and then leaching the residue with a nitric acid solution to recover uranium and thorium. The leach solution, and subsequent washes, are separated from the graphite residue by filtration. The grinding procedure that currently appears most attractive is rough crushing in a hammer mill followed by fine grinding with a roll mill.[1] Grinding the fuel to -140 mesh is required to ensure rupture of the particle coatings. Use of a roll mill to achieve material that is totally -140 mesh results in a significant amount of very fine (-325 mesh) material that could present a problem during filtration. It would, therefore, be desirable to have a grinding procedure that results in a narrow range of particle size distribution at about 140 mesh.

Under special circumstances, ball milling can yield a product having a narrow range of particle size distribution. Consequently, tests were made with type AGOT reactor grade graphite to determine the feasibility of utilizing ball milling instead of roll crushing for fine grinding of graphite fuels.

RESULTS

Initially, tests were made in a 4-in.-long, 4-in.-ID ball mill to determine the effect of milling time on the average particle size. In each experiment, the mill contained 32 ceramic balls (1-in.-diam); the mill was rotated at 80 rpm. Type AGOT graphite was first crushed in a hammer mill and, then, 150-g samples were ball milled for times up to 2 hr. After milling, the particle size distributions of the products were determined by sieve size analysis. The results of these experiments are shown as Rosin-Rammler[2] distribution diagrams in Fig. 1. Under the conditions of the experiments, the grinding efficiency was low as illustrated by the fact that the average particle size (determined from the points corresponding to 36.79% on the diagram) was decreased only from about 14 mesh to 30 mesh in each case. Grinding was practically complete

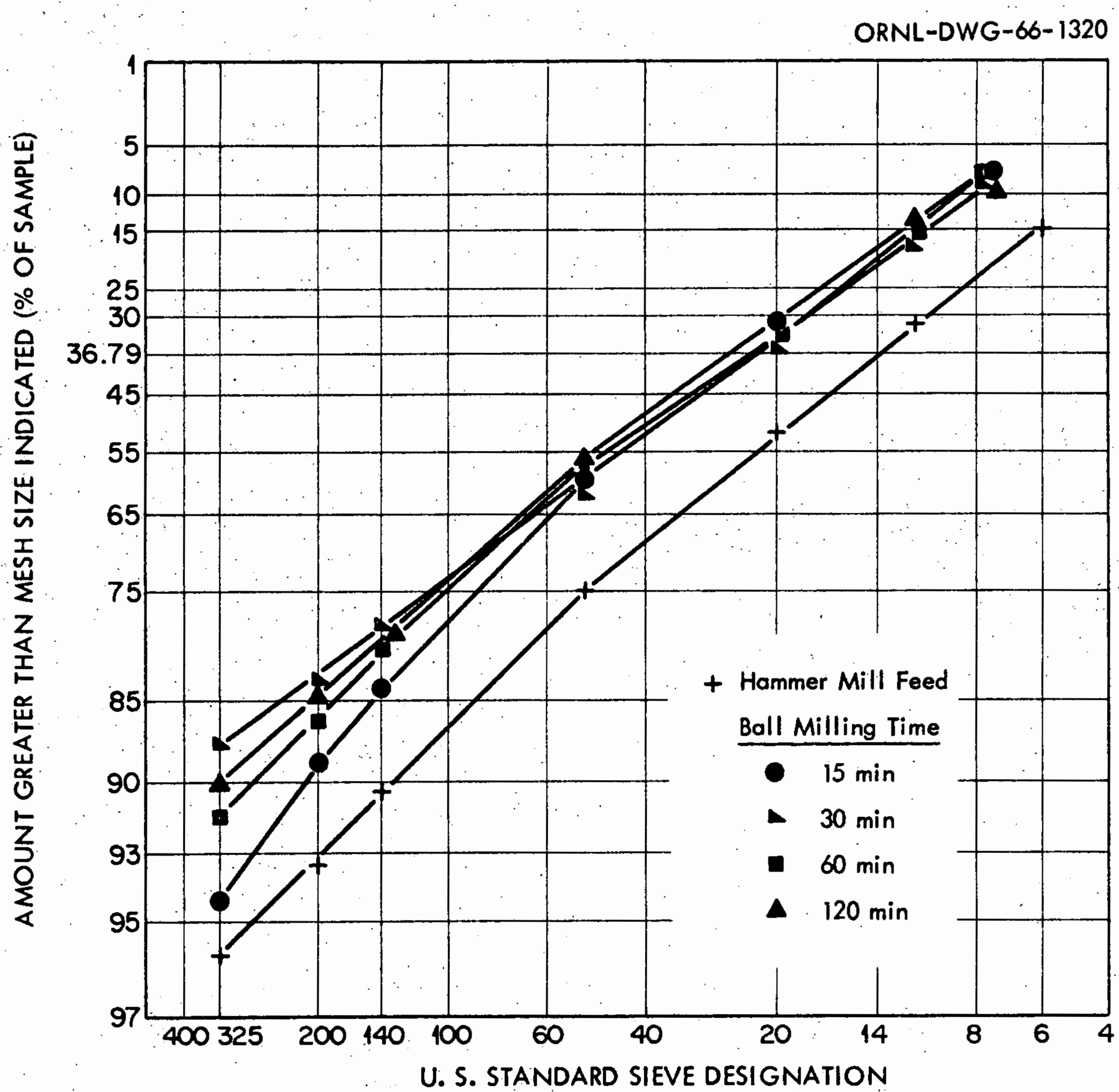

Fig. 1. Particle Size Distribution of Products Obtained by Ball Milling Type AGOT Graphite.

in about 15 min; increasing the grinding time to 2 hr did not significantly change the average particle size or size distribution obtained after 15 min of grinding. As noted, the particle size distributions were practically the same irrespective of grinding time. Furthermore, the distribution coefficients, n, (the slope of the lines in Fig. 1) of the products were about the same as that of the feed. This shows that there was little tendency to form a product having a narrow range of particle size distribution. Obviously, as the range of particle sizes becomes narrower, the value of n increases and would be infinite for a sample having particles of the same diameter.

It can be concluded from the above series of experiments that ball milling of graphite using large diameter balls does not reduce the average particle size to as low a value as desired and does not result in a narrowing of the particle size distribution. Grinding so that the sample is totally −140 mesh would be required to ensure rupture of particle coatings in Peach Bottom reactor fuel.

Another series of experiments was conducted using smaller balls (so that finer grinding could be achieved) to determine the effect of graphite inventory in the mill on size reduction. For these experiments, the mill was loaded with 190 steel balls of 10 mm diam. Charges of 100 and 380 g of graphite (pre-crushed to −10 mesh in a hammer mill) were each ground for 30 min in the ball mill. The 100-g charge represented about 25% of the entire ball volume. The average particle size was decreased most in the experiment where the graphite inventory was the lowest (Fig. 2). However, even in this experiment, an appreciable amount of +100 mesh material remained. A similar effect with fueled graphite would mean that a significant amount of coated particles would remain intact. The general conclusion, then, is that even with small balls and a low graphite inventory the grinding efficiency is too low to be useful.

The efficiency of grinding samples of type AGOT graphite having different initial particle diameters was determined in tests using 190, 10-mm-diam balls in the mill. Samples (150 g) of various sieve fractions (Table 1) were each ground for 30 min, and the distribution coefficients of the products were determined by sieve analysis. The distribution coefficients, n, decreased with increasing initial average particle diameter (Fig. 3), but the grinding efficiency was greatest with particles in the intermediate size range (1- to 2-mm diam). This latter effect is illustrated in Fig. 3 by

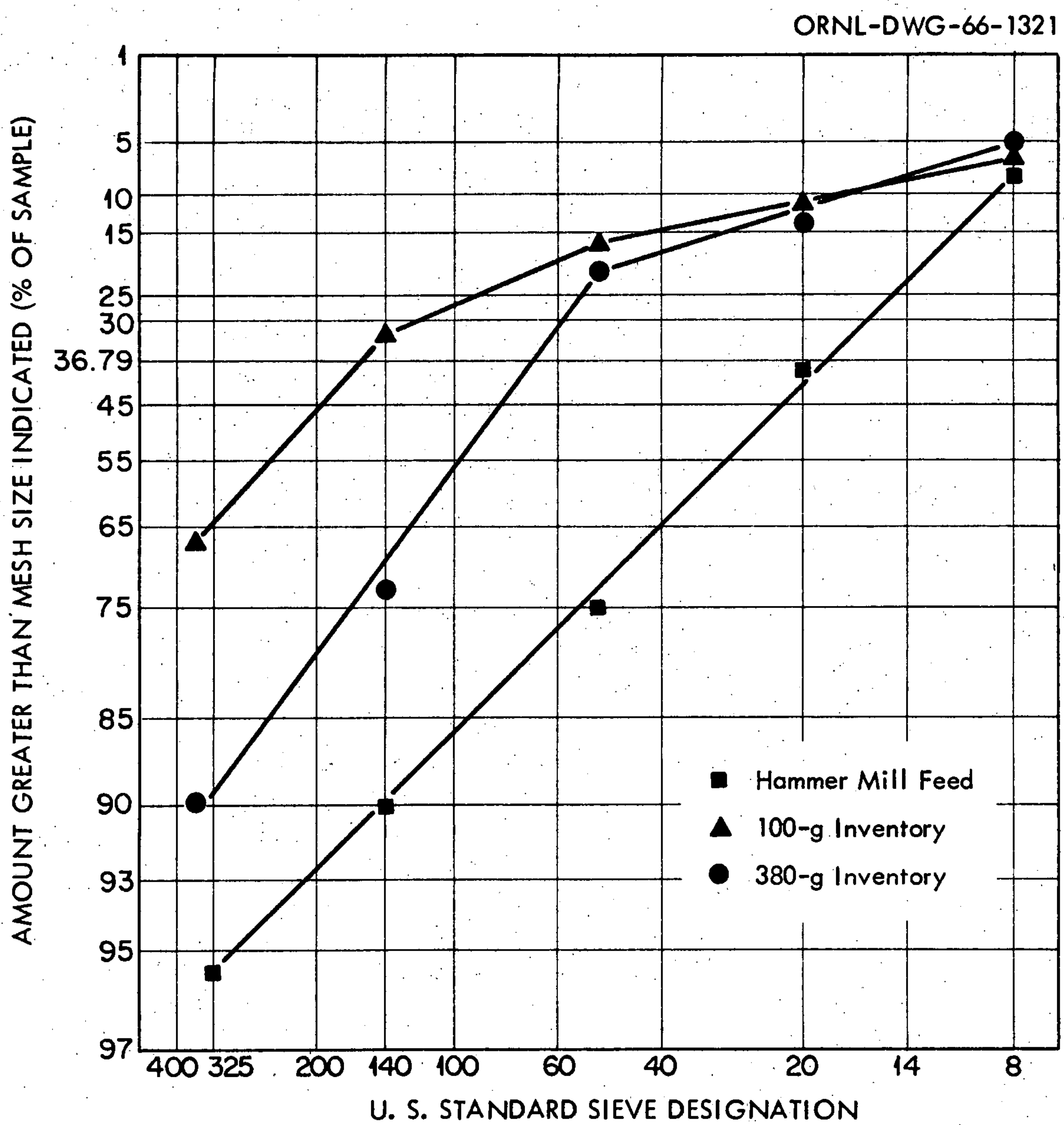

Fig. 2. Effect of Graphite Inventory on Particle Size Distribution of Products Obtained by Ball Milling Type AGOT Graphite.

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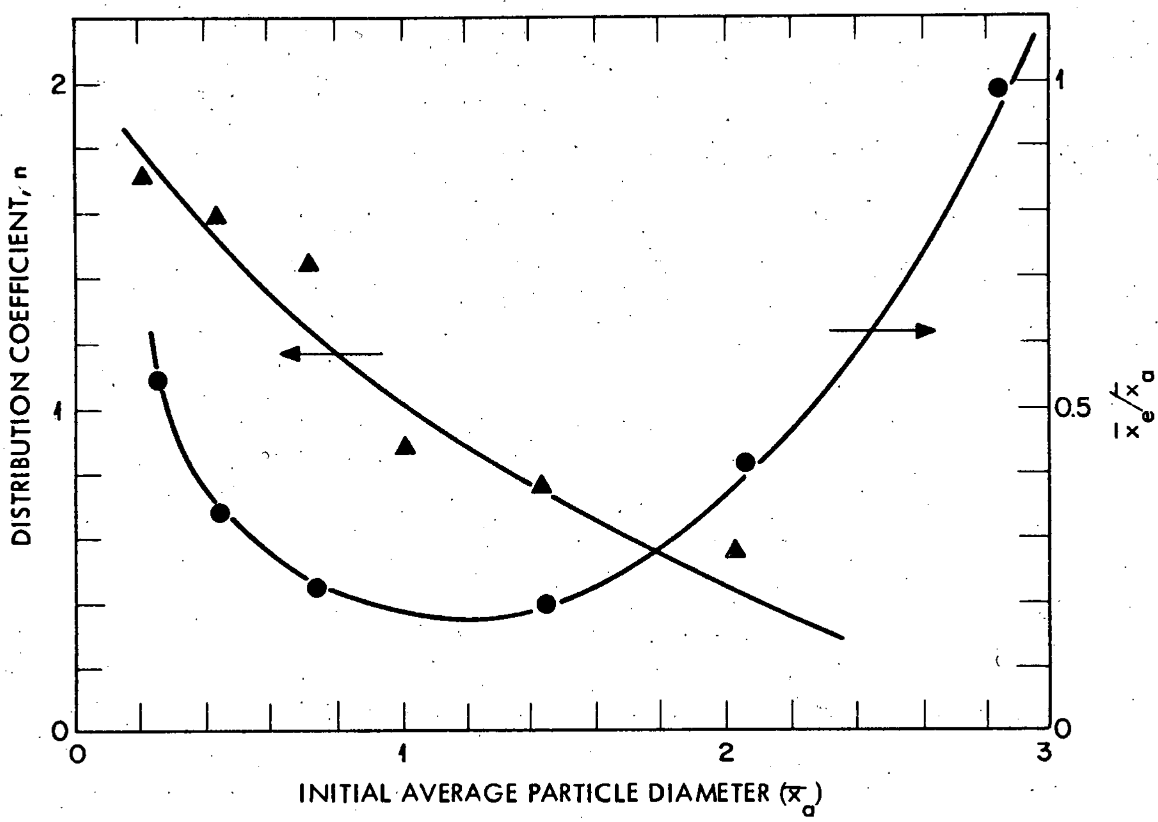

Fig. 3. Distribution Coefficients and Relative Average Particle Diameters of Graphite after Ball Milling of Various Sieve Fractions.

the minimum in the plot of the ratio $\overline{x}_e/\overline{x}_a$ (the average particle diameter of the product divided by the average particle diameter of the feed) versus the average diameter of the feed. Ratios near unity indicate that little or no grinding has been effected. Thus, under the conditions used in these experiments the maximum grinding effect could be achieved only with a feed initially ground to about 2-mm-diam (approximately 8- to 12-mesh). These results also show that a significant amount of +100 mesh material remains after 30 min of grinding, irrespective of the average particle size of the feed, and that a narrow range of particle size was not achieved even when the diameter of the feed particles was small.

Table 1. Initial Average Particle Diameters of Sieve Fractions Used in Ball Milling Studies

Sieve Size (U.S. Standard)	Average Particle Diameter (mm)
6/8	2.87
8/12	2.03
12/16	1.44
16/20	1.02
20/30	0.72
30/50	0.44
50/100	0.22

CONCLUSIONS

The results of this brief study indicate that ball milling is an inefficient method for grinding graphite. Even with relatively small balls (10-mm-diam) and a low graphite inventory in the mill, very long grinding times will probably be required to reduce rough-crushed graphite to about 100 mesh due to the poor efficiency of grinding coarse material. Use of balls of different diameters might be helpful, but it appears that very small balls would be required to achieve the fineness of grinding desired in a reasonable length of time. Assuming that the hollow space between close-packed balls defines the limit of grinding, balls of only about 0.4-mm-diam would be necessary to produce

0.1-mm-diam (100 mesh) particles. Because of the low weight of such little balls, the grinding time still would undoubtedly be quite long. It must be concluded, then, that, at this point in development, roll crushing is the most promising method for the fine grinding of graphite fuels.

ACKNOWLEDGMENT

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