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Studies in ultrafine particle production

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ABSTRACT

Ultra fine grinding has attracted many industries for the better product quality. It has been seen that new developments in electronic industries have demand for the desired size distribution (narrow distribution) of fine sizes. Attrition mill has specific advantage over other conventional milling equipment such as Ball Mill and fluid energy mill due to the energy saving in the combination process. An attempt has been made to study the performance of grinding in a laboratory Attrition mill for the control of the product size distribution. The experiments were carried in the laboratory Attrition mill and the effect of ball size distribution on product size distribution was studied. The study was concentrated in the estimation of the comminution functions such as specific rate grinding by experimental techniques. The experimentally estimated breakage distribution functions were used to predict the rate of grinding, and the product size distribution.

INTRODUCTION

Fine powders are finding application in development of electronic and electrical products. Largest application is production of ultra fine powder of ferrite material. The process has been well developed but the quality product is being made only by a few established plants. The most important part of the product production is the development of grinding process for the powder of narrow size distribution. Most of the techniques of batch grinding results in production of over ground product as the batch mills are operated for long time to get the desired product size. The production of wide size distribution results in the inferior quality of final product. In this paper an attempt is made to study the batch grinding to use the comminution models for the prediction of product size distribution at different time of grinding.

Size reduction of particles in an Attrition mill takes place due to inter-particulate and particle-wall collision. Not much work is published in spite many commercial mills produced regarding the performance of the Attritor. Attrition of particles in fluidized bed[1] during pneumatic conveying[2] and due to their impact of on the solid surface[3] have been studied and particle degradation has been estimated. These observations encouraged the study size reduction in an Attrition mill[4,5]. Fine grinding of material in a Ball mill is compared[6] to that in an Attrition mill.

EXPERIMENTAL MILL AND PROCEDURE

Experimental Attrition mill is shown in Fig. 1. It consists of a two liter vessel with jacket for continuous water cooling. The impeller is a stainless steel shaft fitted with horizontal rods. The shaft is connected to a motor through a belt and pulley for speed control in the range of 300 rpm. to 500 rpm. The material and grinding media can be loaded into the mill by a movable platform. The grinding media used was stainless steel balls of uniform sizes. The Attrition mill has to be lifted to hold the rotating shaft inside and the desired amount of grinding media and material is added. The shaft was initially fixed before the addition of the grinding media and the material.

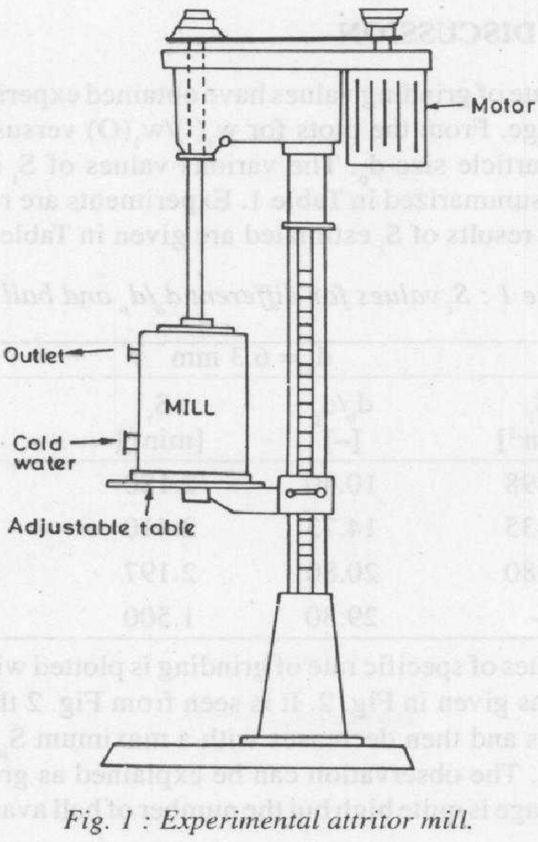

Fig. 1 : Experimental attritor mill.

Comminution Kinetics

The feed material of uniform size particles was ground at specific operating conditions and the rate of decrease of the top size is estimated experimentally. The method of experimentally estimating specific rate of breakages S_i is published elsewhere [7,8]. The specific rate of grinding S_i was obtained assuming first order breakage. In the plot of $w_i(t)/w_i(O)$ versus t on a semi-log graph for a uniform feed size, the slopes will give the specific rate of grinding. The details of the estimation are given by Ramian, *et.al.*[9]. The S_i values were estimated for different uniformed sized feed particles and the experiments were repeated for different ball sizes. The summary of estimated values of S_i and feed particle size, d_p is given in Table 1 for various mono-size balls used in the grinding process.

The top size $w_i(t)$ is related to specific rate of breakage S_i as shown in equation 1.

$$\frac{dw_i(t)}{dt} = -S_i w_i(t) \qquad \text{... (1)}$$

RESULTS AND DISCUSSION

The specific rate of grinding values have obtained experimentally by assuming first order breakage. From the plots for $w_1(t)/w_1(O)$ versus 't' the slope S_i is obtained for each particle size d_p. The various values of S_i obtained for different particle sizes are summarized in Table 1. Experiments are repeated with different ball sizes and the results of S_i estimated are given in Table – 1.

Table 1 : S_i values for different d_b/d_p and ball diameter d_b

d_b = 8.3 mm		d_b = 6.3 mm		d_b = 3.2 mm	
$d_b//d_p$ [–]	S_i [min^{-1}]	d_b/d_p [–]	S_i [min^{-1}]	d_b/d_p [–]	S_i [min^{-1}]
13.70	2.498	10.40	2.180	10.58	0.594
27.40	3.435	14.73	2.410	14.88	0.740
38.60	1.580	20.80	2.197	29.76	0.110
--	--	29.30	1.500	--	--

Estimated values of specific rate of grinding is plotted with ratio of the ball size and particle size as given in Fig. 2. It is seen from Fig. 2 that the specific rate of grinding increases and then decreases with a maximum S_{imax} corresponding to a particular d_b/d_{pmax}. The observation can be explained as grinding with the larger ball size the breakage is quite high but the number of ball available for the unit mass of the material is small. This results in reduction of selectivity of the grinding process. Thus the grinding rate is low. For the smaller ball diameter the selectivity is high due to large number of balls for the unit mass of the material. However due to the large size particle that are relatively bigger compared to the Ball size the particles are not sufficiently nipped and the force is not adequate enough for the particle to break. Hence the breakage rate is low. There is an optimum ratio of ball to particle diameter for which the rate of breakage is maximum. The estimated optimum ratio of ball diameter and particle size for maximum rate of breakage from Fig. 2 is given in Table 2.

Table 2 : S_{imax} and corresponding d_b/d_{pmax}

d_b [mm]	S_{imax} [min^{-1}]	d_b/d_{pmax} [–]
8.3	3.771	20.195
6.3	2.410	14.730
3.2	0.758	16.996

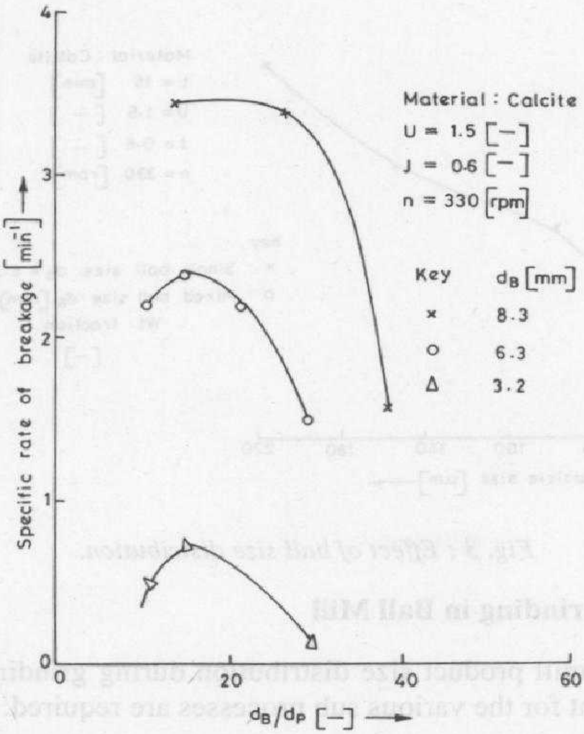

Fig. 2 : Variation of specific rate of breakage with d_B/d_p.

Effect of Ball Size Distribution on Product Size Distribution

The results of studies (from Table 2) that the average d_b/d_{pmax} is 17. A feed of average sizes 605, 427.5, 302.5, 215 and 152.5 μm with cumulative under size distribution of 100, 80, 60, 40 and 20 wt.% respectively were selected. The feed material was ground with mono size ball of 6.3 mm and the product size was analyzed after 5, 10 and 15 minutes of grinding in the Attrition mill. The result of the analysis is plotted in Fig. 2.

Ball sizes of 8.3 mm, 6.3 mm, 4.6 mm and 3.2 mm were selected corresponding to the feed sizes keeping d_b/d_p approximately 17. Experimental grinding was performed with a size distribution with a single size ball size. Experiment was performed at similar condition for same feed size distribution but with mixed ball size distribution. The product size distribution was measured at similar time grinding times and the results of the size analysis obtained is shown in Fig. 3. It is observed from Fig. 3 that the average size obtained with mixed ball size distribution is smaller than that obtained at any specific time of grinding and distribution is narrow.

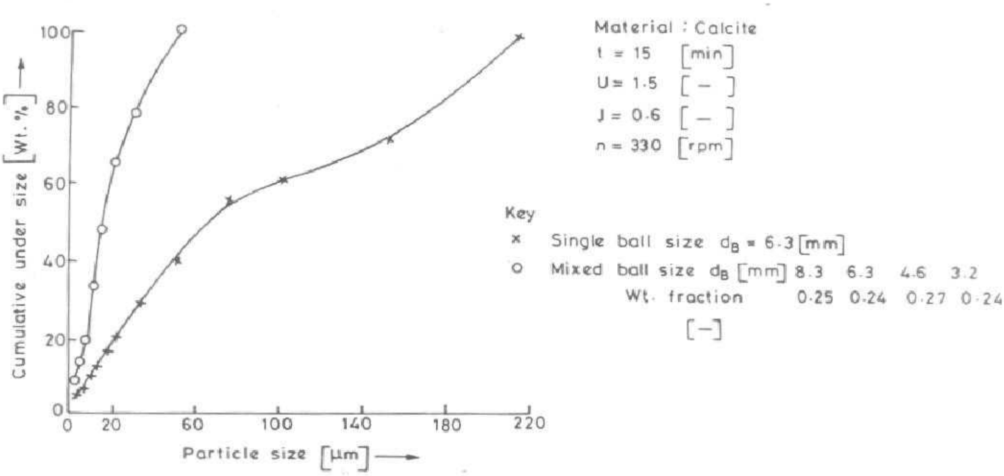

Fig. 3 : Effect of ball size distribution.

Studies on Fine Grinding in Ball Mill

To predict the mill product size distribution during grinding, mathematical models that account for the various sub processes are required.

That rate mass balance for bath grinding is given by equation 2.

$$\frac{dw_i(t)}{dt} = -S_i\, w_i(t) + \sum_{\substack{j=1 \\ i>1}}^{i-1} b_{i,j}\, S_j\, w_j(t) \quad n \geq i \geq j \geq 1 \qquad \ldots \quad (2)$$

where t is the time of grinding

Specific rate of breakage is given by a four parameter model by Austin *et.al.*[7] as in equation 3.

$$S_i = \frac{A\left(\dfrac{x_i}{x_1}\right)^{\alpha}}{\left(1 + \dfrac{x_i}{\mu x_1}\right)^{\lambda}} \qquad \ldots \quad (3)$$

Cumulative breakage distribution function is given by a three parameter model as in equation 4.

$$B_{i,j} = \begin{cases} \Phi\left[\dfrac{x_{i-1}}{x_j}\right]^{\lambda} + (1 - \Phi)\left[\dfrac{x_{i-1}}{x_j}\right]^{\beta} & i > j \\ 1 & i = j \\ 0 & i < j \end{cases} \qquad \ldots \quad (4)$$

where

$b_{i,j} = B_{i,j} - B_{i-1,j}$ for all $b_{i,j} > 0$

Solution of equation 2 with values of S_i and b_{ij} from the model and the initial feed size distribution gives a predicted set of product size distribution that can be verified with the experimental data for different time of grinding.

Experimental

Ball Mill :

The industrial Ball mill consists of a cylindrical ball mill (25 cm diameter and 25 cm height) made of mild steel with a 5 mm thick neoprene rubber lining. The ball mill is connected to a 0.3 H.P. motor through a shaft arrangement. The speed of mill is adjusted to 60 rpm. Six mills are operated simultaneously for grinding. Size reductions of metal oxide powder of size distribution given in Table 1 are ground in the mills. Grinding media used are zircon cylinders of 12.5 mm diameter.

Particle size distribution is determined using a Laser Particle Size Analyzer manufactured by Galai Inc. This device uses the diffraction technique for particle size analysis. It measures the particle size in the range of 150 μm to 1 μm.

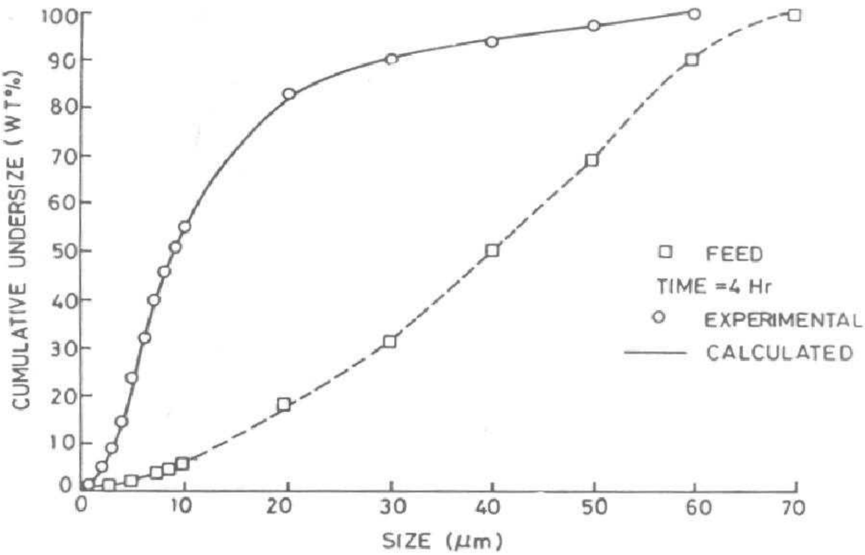

Fig. 4 : Comparison of calculated product size distribution with experimental data.

The Data Collection :

The Mills are operated with a ball loading of J = 0.1, material loading U=2.9 and slurry concentration of 77% (by weight). Corresponding to the ball loading, the weight of balls required is calculated from the volume of mill and density of balls. The ball bed porosity is 0.4. The mill is stopped after 4 hr, 6 hr and 12hr of grinding and product samples are taken from the middle of the mill and are analyzed for the complete size distribution using the laser particle size analyzer evident from Fig. 5 that the error in prediction is more for 12hr than 6 hr case.

The model predicts a higher rate of production of fines than the actual case. From Fig. 5, it is seen that the actual rate of production of fines decreases with time of grinding. This has not been accounted in the model, which results in a significant error in prediction of product size.

The slowing down of rate of grinding with time of grinding is due to the change in the selection function values and breakage function values. This can be verified by estimating the model parameters for 6 hr and 12 hr data and comparing it with the earlier estimated values of model parameters for 4hr data.

The value of parameter 'A' was changed for 6hr data from 0.0934 to 0.13 and

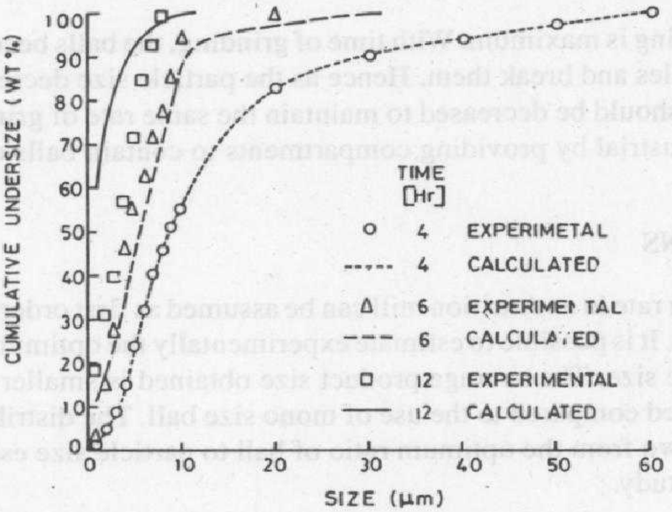

Fig. 5 : Prediction of product size distribution from parameters of 4 hr of grinding.

for 12 hr data was changed to 0.35 and the other parameters kept unchanged. The calculated product size distribution curve with the new values of parameters for 6 hr and 12 hr of grinding along with the experimental data is given in Fig. 6.

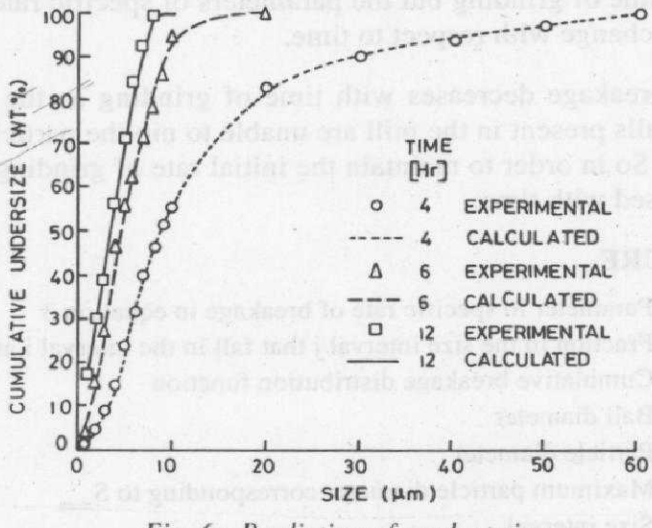

Fig. 6 : Prediction of product size distribution with parameters of selection function.

The calculated product size distribution curve with the new values of parameters fit well the experimental data for 6 hr and 12 hr grinding.

The balls added are unable to break the particles effectively as the particle size decreases. This is because there is an optimum ball size to particle size for which the rate of grinding is maximum. With time of grinding, the balls become too large to nip the particles and break them. Hence as the particle size decreases, the ball size in the mill should be decreased to maintain the same rate of grinding. This is achieved in industrial by providing compartments to contain balls of decreasing sizes.

CONCLUSIONS

The grinding rate in an Attrition mill can be assumed as first order rate for short time of grinding. It is possible to estimate experimentally the optimum ratio of ball size and particle size. The average product size obtained is smaller when mixed ball sizes are used compared to the use of mono size ball. The distribution of ball size can be known from the optimum ratio of ball to particle size estimated from the laboratory study.

The product size distribution in ball mills can be predicted with mathematical models that account for the various sub-processes. The method has been successfully demonstrated for the fine grinding of metal oxides in an industrial ball mill.

It is seen that the parameters of breakage distribution function are constant for small and large time of grinding but the parameters of specific rate of breakage show significant change with respect to time.

The rate of breakage decreases with time of grinding as the particle size decreases. The balls present in the mill are unable to nip the particles and break them effectively. So in order to maintain the initial rate of grinding the ball size should be decreased with time.

NOMENCLATURE

A	[min^{-1}]	Parameter in specific rate of breakage in equation 3
$b_{i,j}$	[–]	Fraction in the size interval j that fall in the interval i after breakage
$B_{i,j}$	[–]	Cumulative breakage distribution function
d_b	[mm]	Ball diameter
d_p	[mm]	Particle diameter
d_{pmax}	[mm]	Maximum particle diameter corresponding to S_{imax}
i,j	[–]	Size interval
J	[–]	Ball loading (fraction of mill filled by the ball bed at rest)
S_i	[min^{-1}]	Specific rate of breakage
S_{imax}	[min^{-1}]	Maximum specific rate of breakage
t	[min]	Time of grinding
U	[–]	Powder loading (fraction of spaces between the balls which is filled with powder)

$W_1(t)$	[gm]	Amount of size 1 present after time t
$W_i(t)$	[gm]	Amount of size i present after time t
$W_1(O)$	[gm]	Amount of size 1 present after time t
$W_1(t)$	[gm]	Amount of size 1 present after time t
$W_i(O)$	[gm]	Amount of size i present after time t=0
$W_i(t)$	[gm]	Amount of size i present after time t
x	[μm]	Particle size
x_1	[μm]	Particle of size 1

Greek

α	[–]	Parameter in equation 3
β	[–]	Parameter in equation 4
λ	[–]	Parameter in equation 3
Φ	[–]	Parameter in equation 4
μ	[–]	Parameter in equation 3
γ	[–]	Parameter in specific rate of breakage function

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