ENERGY REQUIREMENTS FOR THE FINE
GRINDING OF COAL IN AN ATTRITOR

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ABSTRACT

Statistically designed experiments were carried out in a Union Process S-1 Batch Attritor involving a soft coal, Lower Freeport, and a hard coal, Montana Rosebud, in #6 and #2 fuel oil and water slurries. Energy requirements were determined from strain gauge measurements of the torque in the attritor impeller shaft and size distribution measurements in a Model TA Coulter Counter. It was found that the energy requirements for attrition milling depended strongly on the product size, the type of coal and the viscosity of oil. The data was fitted to a Charles type energy size reduction equation to allow the prediction of energy requirements for various operating conditions. Best estimates of the energy per ton of coal required to grind in #2 fuel oil slurries containing between 30% and 50% solids to a product with 10 μm median size is 26 and 106 Kwh/t for Lower Freeport and Montana Rosebud, while a product with a 1 μm mass median can be produced with energy input of 1500 and 6082 Kwh/t for the soft and hard coal, respectively.

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INTRODUCTION

ERDA personnel at the Pittsburgh Energy Research Center are currently evaluating the technical and economic feasibility of producing a "coal-in-oil" slurry which can be used as a commercial fuel. In order to produce a homogeneous non-settling suspension which burns at the required rate, the contained coal must be ground from "standard plant size" (70% minus 200-mesh) down to below 10 microns. A promising device for achieving this degree of size reduction efficiently is the Union Process Attritor. The technical feasibility of producing a fine coal suspension in fuel oil has been demonstrated in continuous Attritor tests in Akron, Ohio. A critical parameter required to evaluate the economic feasibility of producing such a fuel is the energy "cost" necessary to produce the required fineness of coal. This study has been concerned with the accurate determination of energy requirements for the attrition milling of standard plant size coal to a series of product sizes below 10 microns.

By working in conjunction with ERDA personnel the following specific research objectives were defined:

- (1) An accurate measurement of the energy required to produce a 10-micron, 3-micron and 1-micron coal in fuel oil slurry in a small Union Process Attritor was to be made
- (2) An assessment of the influence of coal pct-solids in the slurry on grinding efficiency was to be accomplished
- (3) An assessment of the range of energy requirements for different types of coal was to be made
- (4) A preliminary assessment of the ease with which scale-up from small batch experiments to large continuous commercial units was to be determined.

In order to accomplish these objectives a series of tasks was defined including: 1) development of an experimental design plan to test the effect of percent solids (30-50% by weight), coal type (Montana Rosebud and Lower Freeport), and oil type (#2 and #6 fuel oil) on energy requirements 2) develop and confirm a size analysis method for micron and sub-micron size coal products in oil 3) perform batch grinding experiments according to the design plan in S-1 Attritor and 4) use measured size distributions and energy inputs to construct a mathematical model which allows the prediction of the "specific energy input", E (kwh/ton), required to produce a coal product with a median size, d_{50} , as a function of percent solids, coal type and oil type. Detailed aspects of each of these tasks are described below.

EXPERIMENTAL PLAN

Experimental work was planned according to a 2^3 factorial experimental design.¹ Experimental design allows for rapid statistical determination of the relative importance of the different variables considered and at the same time detects any possible significant interactions between the variables. In this way the factors which are most important in controlling the grinding are determined with a minimum amount of experimentation. Furthermore, a mathematical empirical model can be fitted to the experimental results, which will represent the response of the system within the range of the variables considered. At each point of the experimental design information on the degree of fineness at different energy input was obtained and an equation of the form of the Charles equation:^{2,3}

$$E = A[d_{50,P}^{-\alpha} - d_{50,F}^{-\alpha}] \quad (1)$$

was fitted to the data. In this equation $\bar{E}$ represents the energy per unit mass (kwh/t) to produce a product with median diameter $d_{50,p}$. The dependence of the constants A and α on operating variables can then be determined from the data obtained from the experimental design. These results will provide the basis for predicting the energy required to produce a product of any specified size for a particular coal-oil combination and percent solids in a batch attritor.

Experimental Design

The variables considered in the experimental design were:

- 1) Hardness of coal
- 2) Viscosity of oil
- 3) Weight percent solids

The description of the experimental design appears in Table I. In addition to the experimental design itself, some additional grinding experiments were performed using water as the liquid phase. These experiments were performed in order to evaluate the behavior of attrition of grinding in a very low viscosity fluid. Finally, in one experiment the effect of using a dispersant, lecithin, on grinding efficiency was evaluated.

EXPERIMENTAL PROCEDURES

All grinding experiments were performed in the Union Process S-1 attritor of 1 gallon capacity. Grinding media used consisted of $\frac{1}{4}$ " S.S. balls. The impeller shaft rotational speed was fixed at 340 RPM for all the experiments. Other experimental conditions were varied according to the 2³ experimental design explained previously. The particular coal being ground was sampled during the experiments at different times in order to follow the kinetics of the grinding. Torque applied to the shaft

TABLE I
2³ Factorial Design for the Coal in Oil Mixture
Attritor Grinding

<u>Factors</u>	<u>Levels</u>	
	<u>-1</u>	<u>+1</u>
A: Hardness of coal* (Hardgrove Index)	96	50
B: Viscosity of oil† (cp)	4	3500
C: Weight percent solids (%)	30	50

<u>Experiment</u>	<u>Factorial Levels</u>			<u>Exp. No.</u>
	<u>A</u>	<u>B</u>	<u>C</u>	
1	96	4	30	8
a	50	4	30	10
b	96	3500	30	14
ab	50	3500	30	12
c	96	4	50	11
ac	50	4	50	3
bc	96	3500	50	13
abc	50	3500	50	2

* Two different kinds of coal were used: Montana Rosebud with a Hardgrove Index of 50 and Lower Freeport with a Hardgrove Index of 96

† Two different types of fuel were used: #6 Fuel oil with a viscosity of 3500 cp⁴ and #2 Fuel oil with a viscosity of 4 cp.⁴

of the attritor was monitored and recorded during the duration of the experiment in order to calculate the energy provided to the grind. The collected samples were later washed and analyzed for size distribution in the Coulter Counter.

Grinding Tests

A schematic of the S-1 attritor grinding chamber is shown in Figure 1. This grinding vessel has a nominal volume capacity of one gallon (3750 cc) and when filled with the $\frac{1}{4}$ " S.S. balls, with the impeller running at 340 RPM, can be filled with 2115 cc of slurry (void volume under these conditions). Depending on the percent solids, type of coal and type of oil to be used in a particular experiment, the necessary amount of oil and coal was calculated prior to the run as is shown in Appendix A.

In order to maintain constant temperature and to avoid the large temperature rises normally accompanying energy dissipation during grinding, the grinding chamber was refrigerated. Refrigeration of the chamber was achieved by running tap-water through the refrigeration jacket surrounding the grinding compartment. The temperature of the slurry was in this way held constant at around 65°F.

Sampling of the grinding vessel was achieved by stopping the attritor at certain times and extracting aliquots of about 20 cc of slurry through the upper ports of the grinding vessel by making use of a glass pipette with a plastic disposable tip which was introduced in different radial positions and at different depths in the grinding vessel in order to obtain a representative sample of the material at each energy level. After the sample was removed, the sampling device was carefully washed and the tip discarded in order to avoid contamination of subsequent samples.

At the end of the run, the attritor was discharged and carefully

Figure 1. Schematic drawing of the Union Process Attritor grinding compartment.

washed with oil of the same type as used in the experiment or when a change in oil was involved the mill was prewashed with acetone. Whenever acetone was used the mill was allowed to completely dry out before charging it for the next experiment.

Power Measurement

Accurate measurement of the power drawn by the mill during the experiments was accomplished by monitoring the torque applied to the central impeller shaft of the attritor. The schematic representation of the installation used is shown in Figure 2. The torque sensor unit used was a BLH type A-1 of 1000 in-lb capacity and a RCA WP-700A provided the necessary excitation voltage for its operation. The torque was recorded on a Soltec B-181 recorder. The angular velocity was determined by using a General Radio Type 1531 AB Strobotac electronic stroboscope. Once the torque and the angular velocity are known, the power applied to the rotating shaft can be calculated using the following formula⁵

$$P = 1.18 \times 10^{-5} \times T \times w \quad (2)$$

where

P = power delivered by the shaft (kw)

T = net torque applied to the shaft (in-lb)

w = angular velocity (RPM)

By using equation (2) the specific energy supplied to the mill for grinding can be calculated as follows:

$$\bar{E} = \frac{E}{M} = \frac{P \cdot t}{M} = \frac{1.18 \times 10^{-5} \times T \times w \times t}{M} \quad (3)$$

where

$\bar{E}$ = specific energy supplied to the material being ground
(kwh/ton of coal)

E = energy provided to the material being ground (kwh)

Figure 2. Schematic representation of modified S-1 Attritor including motor and torque measurement system.

M = amount of solid coal being ground (ton)

t = duration of the grind (hr).

Typical torque fluctuations during grinding experiments are illustrated in Figure 3 which shows the torque versus time curve for the grinding of Lower Freeport coal in No. 2 fuel oil at 50% solids by weight.

It can be observed from this figure that the torque changes as the grinding progresses, probably due to changes in particle size of the material being ground which in turn also result in changes in the effective viscosity of the slurry-grinding media combination. It is necessary then to evaluate $\bar{E}$ according to equation (3) using a torque T which is representative of the interval of time for which $\bar{E}$ is being evaluated. For this purpose an average value of T can be used or a more accurate integration of the torque curve can be carried out. For most purposes a linear interpolation for T is sufficient and allows easy and fast calculation. An example of specific energy evaluation for the experiment illustrated in Figure 3 is given in Appendix B.

Size Distribution Determination

Different sizing techniques were tried in order to establish an accurate size distribution determination methods for the ground coal products. First, the Micromeritics Sedigraph 5000-D, which is an X-ray sedimentometer, was used to analyze the ground products. This technique was unsuccessful due to the poor X-ray absorption characteristics of the coal and the method had to be eliminated as an alternative.

Microscopic observation of the samples provided a good measurement of the size of individual particles but this technique fails to give a good overall size distribution because it biases the distribution at the fine and coarse ends due to the inability of the operator to count a representative number of very fine particles (see number density function

Figure 3. Example of trace of net torque in attritor impeller shaft versus time (taken from Experiment 11)

shown in Table II) and to distinguish between single particles and clusters of 2, 3 or more particles agglomerated together. This fact, together with the problems associated with obtaining good particle dispersion and enough magnification for some of fine particles (0.01-0.1 μ) make microscopic observation a very useful technique to get a quick idea of the size range of the product but not to obtain an accurate overall size distribution.

Finally, the Coulter Counter Model TA of Coulter Electronics was used in an attempt to obtain good size distributions. This technique produced the best results of all the techniques examined with respect to reproducibility and agreement with average sizes determined by microscopic observation. So this technique was employed to analyze all the samples obtained along the research. Due to the special characteristic of the samples to be analyzed a special technique was developed to use the Coulter Counter. This technique is explained in detail below.

Coulter Counter Analytical Technique

The analytical technique developed for the Coulter Counter analysis consisted of three major steps: separating coal away from the oil of the samples, sample analysis through the Coulter Counter and data processing and analysis.

The first step in analyzing the coal in oil samples consisted of a careful oil extraction of the sample so as to obtain a clean coal particulate material free of oil. This step is necessary to ensure good dispersion of the coal, avoid agglomerate formation and avoid false counting of oil droplets as particles. The technique developed consists of washing each sample in toluene according to the following procedure:

Approximately 0.5 gram of sample is mixed with 20 ml of toluene, the mixture is shaken for about 3 hours with a wrist action shaker. The sample is then centrifuged at 3400 RPM for 10 minutes and supernatant containing toluene and oil is discarded. The solids and occluded solvent are treated with additional fresh toluene and the whole procedure is repeated until all the oil is removed. In most instances the samples were allowed to be in contact with the toluene for a period of over 24 hours especially for the samples in #6 fuel oil which require more washing steps to remove the oil. After the oil has been removed from the coal, 20 ml of methanol are added and the samples are ready for size analysis.

The second step in the analysis of the samples is the counting performed in the Coulter Counter. In order to analyze the coal samples in the Coulter Counter, an organic electrolyte was required to insure proper wetting of the coal. In this case, a 5% by weight LiCl in methanol solution was used as electrolyte. A few drops of the coal in methanol were added to the electrolyte until the desired concentration of solids was achieved and the slurry was subjected to ultrasonic field for dispersion. for 1 minute. A Branson Model 160 cell disruptor was used for this purpose.

The size analysis of the dispersed samples was carried out with the Coulter Counter Model TA and the 30 μ and/or the 100 μ aperture tube were used depending on the fineness of the product. In most instances samples were counted for 90 seconds and the electrolyte background counts were subtracted. Information was accumulated in 15 or 16 channels depending on the sample and aperture tube used, and the size distribution was plotted after instrument normalization, at the same time the gain

in each channel was recorded for further treatment of the data. In most cases the samples were counted twice and results were accepted when good agreement between one and the other was obtained.

The third step in obtaining the size distribution of the samples is the processing of the data obtained and later the analysis of the results. Generally the data processing consisted of renormalizing the data so that the addition of the percentages in each channel over the 16 channels counted resulted in 100% and the matching of the data obtained with the 30 μm and 100 μm aperture tube so that a single size distribution is obtained as a combination of the two analyses. In those cases where only the 100 μm aperture tube was used, an estimation of the percent of the material finer than the lower limit of detectability of the aperture, i.e., 1 μm , was performed by semi-log extrapolation of the information obtained for those cases in which both aperture tubes (30 μm and 100 μm) were used (samples corresponding to high energy input during grinding and consequently finer than those obtained at lower energy levels). An example of calculation comprising all the cases mentioned is given in Appendix C.

In interpreting the size distribution data obtained in this manner there are some other potential sources of error which should be kept in mind. It is well known that the Coulter Counter does not detect particles that go below its lower limit of detectability or over its upper limit of detectability for a given aperture tube. This problem is partially solved when two or more aperture tubes are used for the same sample since the size range of detectability is expanded when this is done. In some cases however, this is not enough, especially in the very fine products in the finer end, and there is still a significant fraction of the material that is finer than the lower limit of detectability

of the smaller aperture tube used. In this case the resulting size distribution obtained is biased to the coarser sizes and the density function obtained does not look normal. Under these conditions an extrapolation may be necessary. One way of doing the extrapolation would be to follow the shape of the size density function or to use a special technique like the one explained by J.W. Gentry⁶. The first technique was only used in a few cases in which extremely fine products were produced.

In this report all the data is reported in the form of mass size distributions since this kind of representation of size distribution is the most commonly used type for grinding studies. If number size distributions are used, then the products "appear" to be much finer since the number of particles increases very rapidly as the diameter decreases. A comparison of mass and number size distribution for the same sample is shown in Figure 4. In this case the mass based size distribution function and the number based size distribution function for the product obtained grinding Lower Freeport coal in No. 2 fuel oil at 30% solids after 80 kwh/t have been given to the mill are compared. It can be observed that the median size for the mass size distribution is greater than the median size of the number size distribution by a factor of 6. Table II summarizes the data employed to construct Figure 4. In examining this table note that the number of particles increase exponentially as particle size decreases. This points out the difficulty associated with obtaining a good size distribution when using a manual counting technique such as that used in microscopic analyses.

RESULTS AND DISCUSSION

Product size distribution obtained for various energy inputs from each point in the experimental design are shown in Figures 5 to 12.

Figure 4. Number and mass size distributions for a sample of Lower-Freeport coal ground in No. 2 fuel oil at 30% solids by weight after an energy input of 80 (Kwh/t).

TABLE II

Mass and Number Size Distribution for the
80 (kwh/t) Sample of the Experiment #8

Size Interval (μm)	Number of Particles Per Channel	Number Density Function (%)	Number Size Distribution Function (%)	Mass Density Function (%)	Mass Distribution Function (%)
40.3 x 32.0	7.7	0.0000773	100.00	0.73	99.27
32.0 x 25.4	10.6	0.000106	100.00	0.50	98.77
25.4 x 20.2	102	0.00102	100.00	2.41	96.36
20.2 x 16.0	251	0.00251	100.00	2.96	93.40
16.0 x 12.7	933	0.00933	100.00	5.51	87.89
12.7 x 10.08	2340	0.0234	100.00	6.92	80.96
10.08 x 8.00	5210	0.0521	100.00	7.70	73.26
8.00 x 6.35	11500	0.115	99.88	8.47	64.79
6.35 x 5.04	25300	0.253	99.63	9.34	55.45
5.04 x 4.00	53800	0.538	99.09	9.93	45.51
4.00 x 3.17	108000	1.08	98.01	9.93	35.58
3.17 x 2.52	186000	1.86	96.15	8.59	26.99
2.52 x 2.00	322000	3.22	92.93	7.42	19.57
2.00 x 1.59	572000	5.72	87.21	6.60	12.96
1.59 x 1.26	881000	8.81	78.40	5.08	7.88
1.26 x 1.00	1230000	12.30	66.10	3.55	4.33
1.00 x .794	1030000	10.30	55.80	1.48	2.85
.794 x .630	2330000	23.30	32.50	1.68	1.17
.630 x .500	3250000	32.50		1.17	
	10000000	100.00		100.00

Figure 5. Size distributions of the products obtained after grinding a sample Montana Rosebud coal in No. 2 fuel oil at 30% solids by weight at various specific energy levels in the S-1 Attritor.

Figure 6. Size distributions of the products obtained after grinding a sample Montana Rosebud coal in No. 2 fuel oil at 50% solids by weight at various specific energy levels in the S-1 Attritor.

Figure 7. Size distributions of the products obtained after grinding a sample Montana Rosebud coal in No. 6 fuel oil at 30% solids by weight at various specific energy levels in the S-1 Attritor.

Figure 8. Size distributions of the products obtained after grinding a sample Montana Rosebud coal in No. 6 fuel oil at 50% solids by weight at various specific energy levels in the S-1 Attritor.

Figure 9. Size distributions of the products obtained after grinding a sample of Lower Freeport coal in No. 2 fuel oil at 30% solids by weight at various specific energy levels in the S-1 Attritor.

Figure 10. Size distributions of the products obtained after grinding a sample of Lower Freeport coal in No. 2 fuel oil at 50% solids by weight at various specific energy levels in the S-1 Attritor.

Figure 11. Size distributions of the products obtained after grinding a sample of Lower Freeport coal in No. 6 fuel oil at 30% solids by weight at various specific energy levels in the S-1 Attritor.

Figure 12. Size distributions of the products obtained after grinding a sample of Lower Freeport coal in No. 6 fuel oil at 50% solids by weight at various specific energy levels in the S-1 Attritor.

Figures 5 through 8 are for the design points involving the Montana Rosebud coal while Figures 9 through 12 are for those involving Lower Freeport coal. In terms of general observations, it is apparent from these plots that for a given coal the "shape" of the size distribution is quite constant independent of operating conditions. In addition, for given set of operating conditions and energy input a much finer size distribution is produced from the softer, Lower Freeport Coal. Figure 9 shows the results of the single experiment designed to test for improved grinding efficiency in the presence of the dispersant Lecithin. Notice the size distributions for longer grind times with and without Lecithin are virtually superimposed.

As observed previously the data obtained from the size distributions of the experimental design were analyzed in the context of an energy size reduction relationship of the type which has been deemed appropriate for attrition milling of other minerals⁶ i.e.

$$\bar{E} = A d_{50,p}^{-\alpha} \quad (4)$$

where $\bar{E}$ is the specific energy input for grinding ($P \cdot t/H$, kwh/t of coal), and $d_{50,p} \ll d_{50,F}$ is the 50% passing point on the product size distribution produced after expending an energy $\bar{E}$. For each experiment values of $\bar{E}$ were plotted against $d_{50,p}$ on a log-log plot as shown in Figures 13-15. As is evident from equation 4 the slope of such a plot is α while the intercept at $d_{50,p} = 1 \mu m$ is A . The following points are clear from an examination of the data given in the figures:

- 1) An equation of the type given by (4) is appropriate for all coal-grinding fluid-percent solids combination examined.
- 2) Percent solids variations in the range of 30% to 50% by weight have no significant effect on the $\bar{E}$ required for a given product size $d_{50,p}$.

Figure 13. Energy-size reduction relationship (Charles Plot²) for attrition milling of Montana Rosebud coal in fluids of different viscosities for various experimental conditions.

Figure 14. Energy-size reduction relationship (Charles Plot²) for attrition milling of Montana Rosebud and Lower Freeport coals in No. 2 fuel oil at different percent solids.

Figure 15. Energy-size reduction relationship (Charles Plot²) for attrition milling of Montana Rosebud and Lower Freeport coals in No. 6 fuel oil at different percent solids.

- 3) For a given grinding fluid the slope of the energy size plot is approximately constant for different coals. The intercept varies approximately inversely with Hardgrove Index.
- 4) For a given coal type both the slope and the intercept increase with increasing fluid viscosity
- 5) The dispersant Lecithin has no significant effect on energy requirements at 3% by weight of coal in the size range examined.

An attempt at a mechanistic interpretation of these observations is given in the technical paper in Appendix D. Values of A and α determined from the data given in Figures 13 to 15 are reported in Table III. A variety of functional forms for the A and α dependence of fluid viscosity (μ), percent solids (S) and coal Hardgrove Index (H) were tested to represent these values with simple algebraic equations. The most effective relationships (determined by linear regression) using all the data in the experimental design are

$$A = \frac{\mu}{-1.5256 \times 10^{-3} + 3.4809 \times 10^{-5} H - 2.5664 \times 10^{-8} \mu^2 + 2.2163 \times 10^{-6} H \mu} \quad (5)$$

and

$$\alpha = 1.6589 \mu^{0.04364} \quad (6)$$

A comparison of experimental energy requirements with those calculated from Equation 4 with A and α given by Equations 2 and 3 are given in Table IV for three $d_{50,P}$ values for each operating condition in the experimental design. The agreement between predicted and experimental values is generally quite good.

As per the conditions of the contract, the predictive equations described above were used in conjunction with form of ground product size distributions depicted in Figures 5 to 12 to estimate energy

TABLE III

Charles Equation Parameters of the Data
Shown in Figures 13 to 15

Coal*	Hardness(H)	Fluid	Fluid Viscosity (cp)	A $(\frac{kwh}{t}(\mu m)^{\alpha})$	α
L.F.	96	No.6 F.O.	3500	8100	2.36
M.R.	50	No.6 F.O.	3500	47500	2.36
L.F.	96	No.2 F.O.	4	1500	1.8
M.R.	50	No.2 F.O.	4	6300	1.8
M.R.	50	H ₂ O	1	2870	1.63

* L.F. = Lower Freeport coal

* M.R. = Montana Rosebud coal

TABLE IV

Experimental and Calculated Energy Requirements
For Different Grinding Operating Conditions for the Montana
Rosebud and Lower Freeport Coals

(H _{coal} , (cp)	(H _{fluid} , (cp)	%S)	d _{Median,P} (μm)	E _{exp} ($\frac{kwh}{t}$)	E _{Calc.} ($\frac{kwh}{t}$)
50	3500	30	5.6	797	801
50	3500	30	7.5	388	401
50	3500	30	10.0	195	202
50	3500	50	5.9	635	708
50	3500	50	9.3	292	241
50	3500	50	12.0	166	132
50	4	30	3.1	802	830
50	4	30	4.5	428	431
50	4	30	6.1	223	252
50	4	50	3.1	842	830
50	4	50	4.5	402	431
50	4	50	6.2	199	245
50	1	30	2.2	822	830
50	1	30	2.8	456	556
50	1	30	2.4	680	718
50	1	50	2.6	664	629
50	1	50	4.1	319	295
50	1	50	5.8	159	166
96	4	30	1.5	794	735
96	4	30	2.1	409	413
96	4	30	2.8	214	253
96	4	30	1.6	727	694
96	4	30	2.1	379	417
96	4	30	2.9	198	230
96	4	50	1.5	823	780
96	4	50	2.1	444	424
96	3500	30	2.6	800	841
96	3500	30	3.6	388	389
96	3500	30	4.5	194	229
96	3500	50	2.9	800	650
96	3500	50	3.2	448	534
96	3500	50	5.0	207	183

requirements for producing coal products which 90% by weight finer than 4 μm . The estimates obtained in this way for the two coals in numbers 2 and 6 oil are given in Table V.

CONCLUSIONS

Data obtained in this study has confirmed the technical feasibility of grinding coal in an oil slurry within a stirred ball mill (Union Process S-1 Attritor) into the micron size range and has yielded accurate values of the energy requirements to achieve this size reduction. A carefully planned experimental design has revealed that the specific energy (kwh/Ton of coal) required to grind to a given fineness depends upon oil type (high viscosity oils requiring larger energy inputs) and coal type (smaller Hardgrove indices requiring larger energy inputs) but is essentially independent of the weight percent of coal in the slurry. An energy size reduction model of the form

$$\bar{E} = A d_{50,P}^{-\alpha} \quad (7)$$

has been fitted to the data obtained from the design and the dependence of A and α on oil type (viscosity) and coal hardness (Hardgrove index) was expressed in terms of simple algebraic equations. This energy size reduction relationship was shown to provide the basis for accurate predictions of the energy required to grind to any specified median size $d_{50,P}$ in the range from approximately 10 μm to 1 μm . Table VI summarizes the energy requirement predictions for 10, 3 and 1 micron median size coal expected from Montana Rosebud and Lower Freeport coals in #2 and #6 fuel oils. Based on an independent engineering study of the attrition milling of chalcopryrite for batch and continuous grinding carried out by the PI, it is expected that the energy requirements

TABLE V

Energy Required to Grind to
90% Finer than 4 μ m

	#2 Fuel Oil	#6 Fuel Oil
Montana Rosebud	1940(Kwh/t)	10,250(Kwh/t)
Lower Freeport	690(Kwh/t)	2,870(Kwh/t)

TABLE VI

Predicted Energy Requirements for
Producing 10, 3 and 1 Micron Products

		Median=10 μ m	Median=3 μ m	Median=1 μ m
Montana Rosebud	#2 Fuel Oil	106 (kwh/t)	880 (kwh/t)	6082 (kwh/t)
	#6 Fuel Oil	203 (kwh/t)	3515 (kwh/t)	47503 (kwh/t)
Lower Freeport	#2 Fuel Oil	26 (kwh/t)	217 (kwh/t)	1500 (Kwh/t)
	#6 Fuel Oil	35 (kwh/t)	599 (kwh/t)	8100 (kwh/t)

for coal grinding in larger continuous attritors would be essentially the same as those predicted from batch data as reported in Tables V and VI.

Future studies should be directed toward confirming scale-up relationships for coal ground in continuous attrition mills, developing a model which predicts the entire size distribution (rather than just a median size) for a given energy input with a specified coal and oil and optimizing the size reduction process with respect to operating variables including ball size and impeller speed and possible staging.

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APPENDIX A

EXAMPLE OF CHARGE CALCULATION
FOR THE S-1 ATTRITOR

EXPERIMENT NO. 12

Coal: Montana Rosebud

Fluid: No. 6. Fuel Oil

Percent solid of slurry: 30%

Physical Properties of the System:

Specific Gravity of coal: 1.558 gm/cc

Specific Gravity of No. 6. oil = 0.894 gm/cc

Void Volume of the attritor: 2115 cc.

Variables to be determined:

x: Amount of coal in grams to be used in the grinding.

y: Amount of No. 6. Fuel oil in grams to be used in the grinding.

Equations that provide a solution:

$$\text{Volume balance: } \frac{x}{1.558} + \frac{y}{0.894} = 2115$$

$$\% \text{ Solid expression: } \frac{x}{x+y} = 0.3$$

(1)

Solving the system of equation (1),

$$\begin{aligned} x &= 650.44 \text{ gms of coal} \\ y &= 1517.58 \text{ gms of oil} \end{aligned}$$

So, 650.44 gms of coal together with 1517.58 gms or 1697.52 cc of No. 6. fuel oil are introduced into the attritor before starting this experiment.

APPENDIX B

EXAMPLE OF ENERGY CONSUMPTION CALCULATION
FOR THE S-1 ATTRITOR

The torque versus time curve corresponding to experiment #11 has been shown in Figure 3. During the experiment, the grinding vessel was sampled after 2, 10, 21.5, 60, 129 and 231.6 minutes. In order to evaluate energy consumption values during these intervals of time, it was assumed that the torque changed approximately linearly with time during each interval.

In the case of experiment #11, the mill was charged with 1153 grams of coal and 1375 cc of No. 2 fuel oil. Using equation 3, the power is given as:

$$\bar{E} = \frac{1.18 \times 10^{-5} \times \text{torque(in-lb)} \times 340(\text{RPM}) \times \text{time(hr)}}{\frac{1153}{454} \times 2000} \text{ (kwh/t)}$$

$$\bar{E} = 3.16 \times \text{torque(in-lb)} \times \text{time (hr)} \quad [\text{kwh/t}] \quad (1B)$$

TABLE B1
ENERGY CALCULATION FOR EXPERIMENT #11

Time Interval (min)	Torque Variation (in-lb)	Average Torque (in-lb)	Energy given during this interval (kwh/t)	Cumulative energy given to the mill (kwh/t)
0-2	92 → 80	86.0	9	9
2-10	80 → 67.2	73.6	31	40.0
10-21.5	67.2 → 65.6	66.4	40.2	80.2
21.5-60	65.6 → 61.6	63.6	129.0	209.2
60-129	61.6 → 68.0	64.8	235.5	444.7
129-231.6	68.0 → 72.0	70.0	378.3	823.0

Table B1 summarizes the torque values read from Figure 3 at every time the mill was stopped and sampled. Then an average torque for the interval can be calculated and by using equation (1B) the energy supplied to the mill during that interval is also calculated. Finally a cumulative energy after a certain period of grinding can be calculated by adding the appropriate energy increments. The overall information is summarized in Table B1.

APPENDIX C

EXAMPLE OF SIZE ANALYSIS CALCULATION

An example of the way in which Coulter Counter data was treated to determine a final size distribution is given in this appendix. The case shown here corresponds to one of the products obtained from Experiment No. 2 in which Montana Rosebud coal was ground in No. 6 fuel oil at 30% solids by weight. Data given here correspond to the sample taken after 125 minutes of grinding when the mill had been supplied 797 kwh/t.

This particular sample was analyzed using both the 100 and 30 μ m aperture tubes. The raw data obtained from the Coulter Counter is shown in Table C-1. This data is first normalized so that sum of the percentages in each channel adds up to 100. This first normalization has also been included in Table C-1.

After the data obtained for each one of the aperture tubes has been normalized, then the information can be matched or combined into a single size distribution. This has been done in Table C-2, which is a worksheet provided by Coulter Electronics for such a purpose. Overlapping of 4 or 5 channels is necessary in order to carry out the calculation which is based on the fact that for a given channel equal percentages of the population should be detected by using either of the aperture tubes so long the sample is the same. Then a conversion factor is calculated as the average ratio between the percentages for the two tubes for the 3 or 4 overlapping channels. In this case, as is shown in Table C-2, the percentages for the 100 μ m aperture tube have been divided by those for the 30 μ m aperture. The three channels between 3.17 and 5.04 μ m have been considered and the average ratio or conversion factor calculated from these values was found to be 1.25. The combined size distribution

TABLE C-1

Coulter Counter Data for the 797 (kwh/t)
Product of Experiment #12

<u>Channel</u> μm	<u>Raw Data</u>		<u>Normalized Data</u>	
	100 μm ap. tube	30 μm ap. tube	100 μm ap. tube	30 μm ap. tube
0.500 x 0.630		2.8		1.0
0.630 x 0.794		3.2		1.1
0.794 x 1.00		2.3		0.8
1.00 x 1.26	7.2	5.0	3.6	1.7
1.26 x 1.59	7.6	7.3	3.8	2.5
1.59 x 2.00	7.8	9.5	3.9	3.3
2.00 x 2.52	7.5	11.7	3.7	4.0
2.52 x 3.17	13.3	15.2	6.6	5.2
3.17 x 4.00	18.8	22.0	9.3	7.6
4.00 x 5.04	24.8	27.8	12.2	9.6
5.04 x 6.35	28.5	35.5	14.1	12.2
6.35 x 8.00	29.4	34.5	14.5	11.9
8.00 x 10.08	26.5	47.0	13.1	16.2
10.08 x 12.7	18.1	37.1	8.9	12.8
12.7 x 16.0	8.2	30.0	4.0	10.3
16.0 x 20.2	2.6		1.3	
20.2 x 25.4	1.2		0.6	
25.4 x 32.0	0.0		0.0	
32.0 x 40.3	1.0		0.5

COULTER COUNTER® Model T & T_A Worksheet

SAMPLE 12-125																							
ELECTROLYTE LiCl in Methanol 5% by weight			DISPERSANT 1 min ultrasonic																				
EQUIPMENT Model TA	SERIAL	Aper. Dia.	Ser. No.	CALIBRATION DATA	Part. Dia.	W	± I A	A															
ORGANIZATION University of Utah		100μ	---	179 at 16-80 KΩ																			
OPERATOR	DATE Sept. 30, 1978	30μ	44784	183.4 at 32-160 KΩ																			
$k = d \sqrt{\frac{2w}{A}}$ <p>FOR MODEL T</p> <table border="1"> <tr> <td>± I A</td> <td></td> <td></td> <td></td> <td></td> </tr> <tr> <td>CALIB. A</td> <td></td> <td></td> <td></td> <td></td> </tr> <tr> <td>APERTURE DIA.</td> <td></td> <td></td> <td></td> <td></td> </tr> </table> <div style="float: right; text-align: right;"> $\frac{A_2}{A_1} = \left(\frac{d_2}{d_1}\right)^3 2 (W_2 - W_1)$ For Model T $\frac{A_2}{A_1} = \left(\frac{d_1}{d_2}\right)^3 2 (W_2 - W_1)$ For Model T_A </div>									± I A					CALIB. A					APERTURE DIA.				
± I A																							
CALIB. A																							
APERTURE DIA.																							

Geometric Mean μ ³	Volume μ ³	Diameter μ	Channel factors				SAMPLE DATA	
			100 μm tube	30 μm tube	conversion factors		Combined distribution	Normalized combined distribution
.00575	.004091	.198						
.0115	.008181	.250						
.0231	.01636	.315						
.0462	.03272	.397						
.0925	.06545	.500						
.1851	.1309	.630						
.3702	.2618	.794						
.7405	.5236	1.00						
1.481	1.047	1.26	3.6	1.7				
2.962	2.094	1.59	3.8	2.5				
5.924	4.189	2.00	3.9	3.3				
11.85	8.378	2.52	3.7	4.0				
23.70	16.76	3.17	6.6	5.2	1.27			
47.39	33.51	4.00	9.3	7.6	1.22 +	pivot		
94.78	67.02	5.04	12.2	9.6	1.27			
189.6	134.0	6.35	14.1	12.2				
379.1	268.1	8.00	14.5	11.9				
758.3	536.2	10.08	13.1	16.2				
1516.	1072.	12.7	8.9	12.8				
3033.	2145.	16.0	4.0	10.3				
6066.	4289.	20.2	1.3					
12.13 × 10 ³	8579.	25.4	.6					
24.27 × 10 ³	17.16 × 10 ³	32.0	0					
48.54 × 10 ³	34.31 × 10 ³	40.3	5					
97.18 × 10 ³	68.63 × 10 ³	50.8						
194.4 × 10 ³	137.3 × 10 ³	64.0						
388.7 × 10 ³	274.5 × 10 ³	80.6						
777.4 × 10 ³	549.0 × 10 ³	101.6						
1.555 × 10 ⁶	1.098 × 10 ⁶	128.						
3.109 × 10 ⁶	2.196 × 10 ⁶	161.						
6.219 × 10 ⁶	4.392 × 10 ⁶	203.						
12.44 × 10 ⁶	8.784 × 10 ⁶	256.						
24.88 × 10 ⁶	17.57 × 10 ⁶	322.						
49.75 × 10 ⁶	35.14 × 10 ⁶	406.						
99.50 × 10 ⁶	70.27 × 10 ⁶	512.						
199.0 × 10 ⁶	140.6 × 10 ⁶	645.						
398.0 × 10 ⁶	281.1 × 10 ⁶	812.						
796.0 × 10 ⁶	562.2 × 10 ⁶	1024.						

is constructed by using the 100 μm aperture tube data at the coarse end of the distribution until the pivot channel ($3.17 \times 4 \mu\text{m}$) is reached, and then by using the 30 μm aperture tube data multiplied by the conversion factor (in this case 1.25) until the finest channel is reached ($0.5 \times 0.63 \mu\text{m}$).

The size distribution obtained in this way is shown in the third column of Table C-2 and has to be normalized so that the channel percentages add to 100. The final normalized size distribution obtained in this way appears in the last column of Table C-2 and is plotted in Figure 7 for 797 (kwt/t).

When the finest channel ($0.5 \times 0.63 \mu\text{m}$) contains a large percentage it may be suspected that a significant portion of the material may have been below the lower limit of detectibility and passed through the aperture tube undetected. In these cases it may be desirable to make a correction for uncounted particles at the fine end of the distribution. Based on the information given in Table C-2 column 3 or 4 an estimate of the amount of material below $0.5 \mu\text{m}$ can be made either by plotting and extrapolating the size distribution assuming certain regular conditions concerning shape or by using another appropriate technique⁶. This was done in some analyses in this report just by following the shape of the size density function for 1 or 2 channels (extrapolating) and then by renormalizing the curve.

For those cases in which only the 100 μm aperture tube could be used, an estimate of the amount of undetected material was obtained in the following way. The authors have noticed that a plot of the logarithm of the fraction coarser than a certain size versus specific energy almost always results in a straight line. Similar behavior has been observed in tumbling mill grinding. The linearity of this plot is best for the

low energy end where the extrapolation is going to take place. Then based on the analysis of samples analyzed with both the 30 and 100 μm aperture tube (finer samples), the percent coarser than 1 μm can be plotted versus energy on semi-log paper, a straight line can be drawn and an estimate for lower energy inputs can be made.

Such a plot is shown in Figure 16 for experiment #13 corresponding to the grinding of Lower Freeport coal in No. 6 fuel oil at 50% solids by weight. Figure 16 was plotted using the data provided in Table C-3 which also includes the prediction for the lower energy input conditions.

After the extrapolation has been done, the values obtained can be added to the distribution and a new corrected distribution is obtained after renormalization.

Figure 16. Disappearance plot for the fraction coarser than 1 μ m from Experiment 13.

TABLE C-3

Disappearance Data for the Material
Coarser Than 1 μm in Experiment #13

Cumulative Specific Energy(kwh/t)	Fraction Coarser Than 1(μm)
800.0	91.39
448.2	92.37
207.3	94.24
80.0	97.00*
40.0	97.40*
7.1	98.00*

* Extrapolated value from Figure 16.

APPENDIX D

FUNDAMENTALS OF FINE AND ULTRAFINE GRINDING IN A STIRRED BALL MILL

J.A. Herbst and J.L. Sepulveda

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ABSTRACT

Stirred ball mill grinding appears to be an effective means for grinding commercial quantities of mineral materials into the fine (below 10 μm) and ultrafine (below 1 μm) size range. This paper presents the results of a study designed to develop relationships between energy input to an attritor and the resulting product size distributions. A variety of design and operating variables are examined including attritor dimensions, stirring speed, media filling, media density, media size, pulp density and fluid viscosity. A variety of materials are examined including coal, chalcopyrite, limestone, quartz and cupric oxide. Finally, a general population balance model is proposed for attrition milling.

FUNDAMENTALS OF FINE AND ULTRAFINE GRINDING IN A STIRRED BALL MILL

J. A. Herbst and J. L. Sepulveda

INTRODUCTION

Fine grinding of particulate materials can be achieved in many different ways. Among the most common techniques being used commercially are tumbling mill grinding, vibratory milling, impact milling and attrition milling. Products in the range of 10 to 1 microns are usually referred to as fine, and powders which are predominantly below 1 micron are referred to as ultrafine.

Tumbling mill grinding (conventional ball milling) is the most widely used technique for achieving ground products in the size range of 1/4" to a few microns. However, in order to obtain products below 2 micron in a conventional ball mill, long retention times and large energy inputs are necessary. Although impact milling is capable of producing submicron material, unit capacities restrict the application of this device to small tonnage operation. Vibratory milling⁽¹⁾ has been used to produce tonnage quantities of material in the range 10-20 microns, but its applicability to ultrafine grinding has not been demonstrated.

Recent studies have indicated that it may be possible to achieve ultrafine grinding of mineral materials on a large scale using stirred ball mills.⁽²⁾ This size reduction technique has been successfully used in the ceramic and paint industries for several years. A stirred ball mill consists of a vessel charged with grinding media that is agitated with an impeller. The material to be ground is usually in a slurry form filling the void spaces between the grinding media. As the impeller rotates the grinding media becomes fluidized and a gradient in the velocity of grinding

media is developed across the vessel diameter. Particle breakage in such a device undoubtedly occurs by a combination of rubbing impingement between adjacent balls, between the media and the impeller shaft and in some cases with the grinding chamber walls.

There are several types of stirred ball mills available commercially. One of the earliest devices is a shot mill⁽³⁾ in which fluidization of the grinding media is obtained by several discs located along the length of the impeller shaft. The shot mill uses a grinding media ranging from 0.5 mm to 2.00 mm and features a rotating filter mounted on and rotating with the shaft which protect the discharge area from grinding media and unground material.

In the late 30's the USBM⁽⁴⁾ developed an "attritor" with the purpose of conditioning minerals by removing surface contaminants for froth flotation. The attritor turned out to be a very efficient grinding device. It was later modified and patented by the USEM⁽⁵⁾ and by Union Process Inc. as the Szegvari attritor system⁽⁶⁾. The USBM attritor⁽⁷⁻¹¹⁾ consists of a cage-like rotor turning inside a cage-like stator, both of which are held in a cylindrical water-jacketed container. Simpler in design is the Szegvari attritor which consists of a cylindrical water-jacketed vessel containing the grinding media which is fluidized with a central branched shaft. Each of these devices can use various forms of grinding media, including steel balls, ceramic balls, sand and gravel.

A recent comparison of the effectiveness of various grinding devices for producing ultrafine products from a chalcopyrite concentrate⁽²⁾ has shown that the stirred ball mill is capable of producing a finer product with a smaller energy input than the other devices tested. These data are shown in Figure 1, where it is observed that for energy inputs exceeding 200 (kwh/t) the stirred ball mill continues to grind into the submicron

range when size reduction in the other devices has almost stopped. In addition, in the micron range the capacity of the stirred ball mill is high relative to the other devices because of the exceptionally high power input per unit volume of vessel. For example the power input per unit volume of the S-1 attritor operated under typical conditions is 0.33 kw/gallon compared to a value of 0.016 kw/gallons obtained for a 10 in. in diameter by 11.5 in. long tumbling ball mill.

Very little has been reported in the literature about attrition grinding and given its potential for ultrafine grinding of mineral materials the study of stirred ball milling fundamentals, engineering design and mill scale-up becomes necessary. With the exception of the research done by the USBM⁽¹²⁾ there have been no studies reported in the literature on these aspects of attrition milling. The purpose of the current paper is to report on studies of this type which are currently underway at the University of Utah.

EXPERIMENTAL

All of the experimental data reported here were obtained in Union Process attritors. Models S-1, C-3 and C-10 of 1, 3 and 10 gallons respectively nominal capacity were used. A schematic of one of these mills is shown in Figure 2. The S-1 and C-3 units have variable speed impeller drives. Modifications were made on each unit to allow the power input to the grinding compartment to be accurately monitored. All these features make the attritors employed at the University of Utah Metallurgy research labs very flexible devices for studying fundamental aspects of particle grinding kinetics and scale-up relationships for the stirred ball mill.

Although this research was primarily concerned with chalcopyrite grinding, other materials such as limestone, quartz, cupric oxide and coal were also ground in order to have a better understanding of attrition grinding as applied to different systems.

The model S-1 was run in batch mode and the C-3 and C-10 units were run in a continuous mode. Slurries between 30% and 70% solids were used and the agitator speed was varied between 100 and 500 RPM. Grinding media consisted on stainless steel and ceramic balls of 1/4, 1/8 and 3/32 in. in diameter used in various combinations.

Size analyses were performed in a Micromeritics SediGraph 5000D for all the materials with the exception of coal in which case the Coulter Counter Model TA was used. A 0.5% by volume solution of sodium silicate in H_2O was used as the sedimentation fluid for the SediGraph 5000D X-ray sedimentometer and samples were dispersed for one minute with an ultrasonic probe prior to being analyzed. A 5% by weight lithium chloride in methanol solution was used as the electrolyte for the Coulter Counter together with the 100 μm and 30 μm aperture tubes when analyzing the coal samples. Samples for the Coulter Counter were also subjected to ultrasonic treatment before the analysis was performed.

Energy expended during grinding was calculated by monitoring the net torque generated in the attritor shaft using a BLH torque transducer model A, measuring the rotational speed of the shaft and time of grinding.

RESULTS AND CONCLUSIONS

Figure 3 shows typical size distribution data obtained in the S-1 attritor for the grinding of Pima chalcopyrite concentrate. It can be observed that values of the fine size slope of the size distribution are large (≈ 1.5) compared to values normally obtained for conventional ball

milling ($\approx .5$ to 1), showing that attrition milling results in a smaller variance in product particle size than the conventional device. It is apparent from Figure 3 that the grinding progresses even after very large energy inputs, i.e. after 16 hours or 2000 kwh/t. This behavior is contrasted with conventional ball milling where the grinding seems to stop after extended periods of time⁽¹³⁾, and an apparent limit of comminution is obtained. The authors believe that the advantage of the stirred mill rests with the fact that intense shear stresses as well as normal stresses are applied to particles in the rotating ball mass and that a small mean free path for balls in the fluidized grinding media facilitates the capture of small particles.

The combined action of compressive and torsional stresses may result in an increased principal stress⁽¹⁴⁾, which may lead to fracture. It is generally accepted that as particles are reduced in size, large flaws are eliminated resulting in increased strength for small particles.

Experimental observations by Schönert et al.⁽¹⁵⁾ on the breakage of particles in the one micron size range, indicate that the only way to achieve fracture for these small particles is to apply a combination compressional and torsional stresses. In the conventional ball mill or vibratory mill particle loading is predominantly compressive. In contrast, the stirred mill results in the application of combined stresses required to break these fine particles as indicated in Figure 4.

The prediction of grinding behavior in a stirred ball mill and the subsequent development of scale up design relationships requires a good mathematical model relating energy input and the resulting product size distribution. An empirical relation which is often used to correlate energy input with some representative size of the distribution for coarse grinding, is the Charles Equation⁽¹⁶⁾. If the representative product size

is taken to be the median size, the Charles equation takes the form:

$$\bar{E} = A (d_{\text{Median},P}^{-\alpha} - d_{\text{Median},F}^{-\alpha}) \quad (1)$$

where $\bar{E}$ = energy input to the mill ($\frac{\text{kwh}}{t}$)

A = constant

$d_{\text{Median},P}$ = median size of the product (microns)

$d_{\text{Median},F}$ = median size of the feed (microns)

α = constant

When the products are much finer than the feed, equation (1) can be approximated by:

$$\bar{E} = A d_{\text{Median},P}^{-\alpha} \quad (2)$$

A logarithmic transformation of Equation 2 yields

$$\log \bar{E} = \log A - \alpha \log(d_{\text{Median},P}) \quad (3)$$

which reveals that if this form of the Charles Equation is appropriate for a given material-device combination then a plot of $\log \bar{E}$ against $\log(d_{\text{Median},P})$, should result in a straight line of slope $-\alpha$.

Figure 5 shows such a plot for the grinding of chalcopyrite. This plot includes data for two different concentrates, three different attritors (1, 3 and 10 gallon units), stirring speeds going from 100 RPM to 400 RPM, per cent solids from 30% to 70%, and grinding media sizes between 3/32 to 1/4 of an inch. Linear regression for all of the data shown yields the equation

$$\bar{E} = 460 d_{\text{Median},P}^{-1.79} \quad (4)$$

with a correlation coefficient of 0.95.

The fact that all the data is well described by a single straight line is very important from the standpoint of scale up since it suggests that it is possible to predict the median size of the product expected for a given energy input, independent of the attritor size and the operating conditions employed. Based on the success of this correlation for chalcopyrite, the same treatment was used for the other minerals examined in this study.

Figure 6 shows the size distributions obtained when limestone is ground in the S-1 attritor at various levels of energy input. As in the case of chalcopyrite, it may be observed that grinding progresses after retention times as long as 8 hours which corresponds to an energy input of 2155 kwh/t. Figure 7 shows the size distributions of products obtained for the grinding of a cupric oxide sample in the S-1 attritor. Similar conclusions can be drawn from this data and the size distribution data for coal. Figure 8 shows a test of Equation 2 for each of these minerals ground in water. Data for each of the minerals can be represented by a straight line and further, all of the straight lines are almost parallel to one another, indicating that α in Equation 2, has an approximately constant value of 1.8 for stirred ball milling. Values of the Charles constant A obtained for the different minerals are reported in Table 1. The values of A correlate roughly with mineral hardness with the possible exception of coal.

Results obtained with coal indicate that this mineral is more difficult to grind in the stirred mill than one would expect from the other minerals. This may be due to certain intrinsic properties of the coal-water system. First, the density of the coal is lower than that of the other minerals which may result in unfavorable hydrodynamical conditions for the capture of particles to be ground. The coal particles may move with the water, tending to be squeezed out of the grinding zone when two balls approach one another.

TABLE 1

Values of the Charles Constant A

Grinding of various minerals in water	$A[\frac{\text{kwh}}{t}(\mu\text{m})^{1.8}]$
Chalcopyrite	460
Limestone	500
Cupric Oxide	660
Quartz	920
Coal (Montana Rosebud)	3,000
Grinding of coal in fuel oil	
Montana Rosebud Coal in No. 2 Fuel Oil	5,900
Montana Rosebud Coal in No. 6 Fuel Oil	18,500
Lower Freeport Coal in No. 2 Fuel Oil	1,500

Second, there is some evidence that fine coal is more plastic in its deformation behavior than other common minerals, hence it may be that coal particles may be undergoing a substantial amount of plastic deformation rather than failing by brittle fracture when it is mechanically loaded in the mill.

Further evidence for the importance of particle hydrodynamics in stirred ball milling was obtained by examining data obtained for grinding in fluids of various viscosities. Two different coals were ground in No. 2 and No. 6. fuel oils. The viscosity of No. 2 oil at 65°F is about 4 centipoise, while the corresponding value for No.6 oil is about 3500 centipoise.

Figure 9 shows typical product size distributions obtained for the grinding of Montana Rosebud Coal in No. 2 fuel oil. Figure 10 shows the energy-size relationships obtained for the various grinding fluids used, including water. It can be concluded from Figure 10 that the grinding becomes more difficult as the viscosity of the grinding fluid increases.

This behavior can be explained based on the resultant increase in particle entrainment that will occur with increasing viscosity. As the degree of entrainment increases, the probability of capture of the particles between approaching balls is expected to decrease. In addition, high viscosity fluids may result in the formation of a thick boundary layer surrounding the balls which in turn may promote the passage of balls, one past another, at distances greater than that required to capture ultrafine particles. Finally, on a macroscopic scale high viscosity fluid may impede the circulation of slurry through high energy grinding zones in the mill. Differences observed between the Montana Rosebud and the Lower Freeport samples can be explained based on the hardness of the two coals. The Montana

coal has a Hardgrove index of 50 compared to one of 96 for the Lower Freeport coal. Again, it is apparent from Figure 10 that all the coals ground resulted in similar value of $\alpha \approx 1.8$ in the Charles Equation. The values of the Charles constant, A, obtained for the two coals with different viscosity liquids are reported in Table 1.

In order to use the information given in Figures 8 and 10 for scale up purposes, it is necessary to correlate the power drawn by the attritor with mill dimensions and operating variables. Sadler, Stanley and Brooks⁽¹²⁾ suggested that the power characteristics of an attritor can be represented by the same type of model used for turbine mixers. If such a correlation is used, the power drawn by the attritor can be written as a function of the attritor volume, angular velocity of impeller, and viscosity and density of the slurry. By correlating the power number versus the Reynolds number, three regions could be distinguished. A laminar behavior zone was observed to occur for Reynolds numbers smaller than about 200, a transition region between 200 and 8000, and a turbulent region for Reynolds number greater than 8000. Attritors are usually operated under experimental conditions which result in turbulent behavior. For a fully baffled vessel in the turbulent region the power drawn by shaft can be expressed by⁽¹²⁾:

$$P = K V^{1.67} N^3 \rho \quad (5)$$

where P = power to the impeller
 K = proportionality constant
 V = Volume of the vessel
 N = shaft angular velocity
 ρ = density of the fluid phase

A relationship similar to Equation 5 was developed for the attritor.

Based on more than 20 different experimental conditions, the following correlation was found:

$$P = 2.55 \times 10^{-5} V^{1.75} N^{1.37} d_B^{0.48} \rho_B^{1.09} \quad (6)$$

where

P = power on attritor shaft (kw)

V = attritor volume (gallons)

d_B = diameter of the grinding media balls (in)

ρ_B = density of the grinding media balls (g/cc)

N = shaft angular velocity (rpm)

It can be observed that the values of the exponents of Equation 6 compare well with the corresponding ones in Equation 5 with the exception of the speed dependence. The reduced dependence on speed has been interpreted as resulting from a reduction in the effective viscosity of the ball mass with increasing speed in the fluidized state. In gathering data for Equation 6, the attritor volume ranged from 1 to 10 gallons, the impeller speeds were varied between 100 RPM and 500 RPM, ball diameters between 3/32" and 1/4" in diameter, and density of the grinding media balls from 2.74 to 8.01 gm/cc. Multiple linear regression involving Equation (6) resulted in a root mean square of residuals of 0.042. Table 2 shows a sample of some of the experimental conditions used along with the experimental power measured and the power predicted by Equation 6. It can be observed that very good predictions are achieved over the entire range of mill dimensions and operating variables examined.

TABLE 2

Comparison of Experimental and Predicted Power*

<u>Operating Variables</u>				<u>Power</u>	
<u>V (gallons)</u>	<u>N (rpm)</u>	<u>d_B (in)</u>	<u>ρ_B (g/cc)</u>	<u>Experimental (kw)</u>	<u>Predicted (kw)</u>
1	400	0.13	8.01	0.33	0.34
1	100	0.19	8.01	0.07	0.06
1	300	0.25	2.74	0.10	0.10
3	160	0.25	8.01	0.85	0.92
10	175	0.094	8.01	5.25	5.31

*Predicted power was calculated using Equation 6.

When the power equation, Equation 6, is used in conjunction with the energy-size reduction relationship given by Equation 2 a reasonably accurate prediction of the product size distribution (median size) for any given stirred ball mill should be possible. As an example, the prediction of the median size for chalcopryrite being fed at a rate of $\dot{M}_F = 6$ lbs/hr to a C-10 attritor of 10 gallons capacity operated at a speed of 175 rpm with 3/32" steel balls can be made as follows. Introducing all this data in Equation 6, a predicted power of 5.31 kw is obtained. This value compares well with the observed experimental value of 5.25 kw. Then making use of the Charles relationship for chalcopryrite given in Equation 4, a prediction of the expected median size for this power input can be accomplished. According to Equation 4,

$$\bar{E} = \frac{P}{\dot{M}_F} = \frac{5.31}{6/2000} = 1770 \left(\frac{\text{kwh}}{t} \right) = 460 d_{\text{Median},P}^{-1.79} \quad (7)$$

Solving equation 7 for $d_{\text{Median},P}$ yields a value of $0.47 \mu\text{m}$. The predicted median size of $0.47 \mu\text{m}$ compares well with the observed experimental value of $0.48 \mu\text{m}$ reported in Figure 7.

Population Balance Description of Stirred Ball Mills

An alternative and more detailed mathematical representation of the stirred ball milling process can be achieved by adopting the population balance viewpoint for modeling⁽¹⁶⁾. This approach offers some advantages as compared with the Charles or Bond⁽¹⁸⁾ relationships in that it allows the prediction of the complete size distribution in the mill for different energy inputs. Basically, the population balance approach involves constructing a particle number balance for each particle size in the system. For the case of attrition milling in a batch mill this number balance for particles of size take the form:

$$\frac{\partial \bar{\Psi}}{\partial t} + \frac{\partial}{\partial d} (V_d \bar{\Psi}) + \bar{D} - \bar{B} = 0 \quad (8)$$

where $\bar{\Psi}d(d)$ = the number of particles of size d per unit volume of slurry at time t in the stirred mill

$\frac{\partial \bar{\Psi}}{\partial t}$ = represents time rate of change of the number density function

$\frac{\partial}{\partial d} (V_d \bar{\Psi})$ = change in the number of particles of size d per unit volume of slurry due to pure attrition

$\bar{D}$ = death of particles of size d per unit volume of slurry due to brittle fracture

$\bar{B}$ = birth of particles of size d per unit volume of slurry due to brittle fracture.

Equation 8 has been applied to stirred ball milling assuming that the amount of pure attrition is negligible compared to the amount of brittle fracture which occurs in the stirred mill. Under these conditions assuming that the rate of breakage of particles is proportional to the amount present, Equation 8 can be solved as is described in reference 19. This type of linear breakage behavior was observed for the grinding of coarse particles

by Sadler et al.⁽¹²⁾ in the USBM attrition. When the breakage process is linear over the entire size range, the parameters associated with the birth and death processes can be estimated from experimental data using a computer program called ESTIMILL⁽²⁰⁾. This program was used in conjunction with the limestone data given on Figure 6. The grinding kinetic parameters (selection functions and breakage functions) were estimated by fitting the model to the first three products ($\bar{E}$ ranging in between 68 and $255 \frac{\text{kwh}}{\text{t}}$) shown in Figure 11. Then, using these parameter estimates in conjunction with the population balance model, predictions were made for the next three products ($\bar{E}$ ranging between 493 and $2155 \frac{\text{kwh}}{\text{t}}$). Referring to Figure 11 it can be seen that fitted values and predictions are in good agreement with the experimental values for the majority of the products obtained. Deviation of the predictions from the experimental values are obtained only after extremely long grinding times (around 2000 kwh/t). For the extremely fine products produced under these conditions it is possible that pure attrition processes may begin to play an important role. Additional studies are currently underway to determine quantitatively the importance of pure attrition in these systems⁽²¹⁾. These studies hold considerable promise for helping to interpret stirred ball milling behavior mechanistically.

Summary and Conclusions

It has been demonstrated that stirred ball mills are effective devices for grinding mineral materials into the fine ($<10\mu\text{m}$) and ultrafine ($<1\mu\text{m}$) size range. Energy requirements correlate roughly with hardness for a variety of minerals. Product size for a given energy input can be predicted by using a Charles type energy-size reduction relationship and power requirements can be determined from a model similar in form to that used for turbine

mixers. It has been shown that these two relationships can be combined to provide a basis for scale up design of these mills. Finally, it was found that population balance models hold considerable promise for providing a more detailed description of stirred ball milling and perhaps for interpreting the grinding behavior of these mills mechanistically.

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Figure 1. Comparison of the effectiveness of various grinding devices for grinding devices for the ultrafine grinding of chalcopryite concentrate.

Figure 2. Schematic drawing of the Union Process Attritor (stirred ball mill).

Figure 3. Size distributions of the products obtained after grinding a sample of Pima chalcopyrite concentrate in water at various specific energy levels in the S-1 attritor.

CONVENTIONAL MILL

STIRRED MILL

Figure 4. Comparison of the grinding action in conventional and stirred ball mills

Figure 5. Energy-size reduction relationship for attrition milling of chalcopyrite concentrates in water.

Figure 6. Size distributions of the products obtained after grinding a sample of limestone in water at various specific energy levels in the S-1 attritor.

Figure 7. Size distributions of the products obtained after grinding a sample of cupric oxide in water at various specific energy levels in the S-1 attritor.

Figure 8. Energy-size reduction relationships for various minerals ground in water in the S-1 attritor.

Figure 9. Size distributions of the products obtained after grinding a sample of Montana Rosebud Coal in No. 2 fuel oil at various specific energy levels in the S-1 attritor.

Figure 10. Energy-size reduction relationship for different coals in various liquids ground in the S-1 attritor.

Figure 11. Population balance fitting and prediction of the data obtained for the grinding of limestone shown in Figure 6.