Self-Similar Distributions of Fine Particles Produced in Non-Linear Batch Grinding

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Abstract

A parametric approach is described for tracking the evolution of particle size distribution with time in batch grinding in micron and sub-micron range. The approach is based on an asymptotic self-similarity solution to population balance equation of grinding. The possible presence of non-linearity in the fine grinding regime is captured by a modification to the model formulation scheme. It is assumed that the breakage rate is a function of the grind time and therefore, indirectly of the time-dependent mean, median or a percentile size. It is shown that the self-similar character is preserved under a wide class of functions that can in principle describe the non-linear characteristics of the comminution process. The resulting parametric model describes the evolution of particle spectra. Several sets of published data are employed to validate the model.

Keywords: *Self-similarity, non-linear grinding, fine grinding*

INTRODUCTION

Preparation of fine particles of subsieve size by comminution is an energy intensive unit operation that is nevertheless widely employed in a variety of material processing industries. Because typical tumbling mills are quite inefficient and uneconomical for subsieve grinding, alternate technologies which operate with higher energy density, such as, jet mill, circular fluid energy mill, stirred media mill, centrifugal mill, pin mill etc. are preferred. Stehr (1988) provides a list of various products which are commercially processed in fine grinding mills, including minerals, pharmaceutical powders, printing inks, paints and pigments, fertilizers, ceramics, food products and others.

Quantitative understanding of the evolution of particle size distribution is a necessary step for proper process control and optimization of the grinding operation. For conventional tumbling mill type of grinding operations, various schemes of Particle Population Balance (PPB) are used to track the grinding kinetics. A continuous-size continuous-time population balance model of size reduction in a batch process over the entire particulate mass is described by the following integro-differential equation (Kapur 1972; Austin et al 1984).

$$\frac{\partial m(x,t)}{\partial t} = -k(x,t)\, m(x,t) + \int_{x}^{\infty} k(v,t)\left[\frac{\partial B(x,v)}{\partial x}\right] m(v,t)\,dv \tag{1}$$

Where *m(x, t)* is the weight fraction of particles in the size range *x* to *x+dx* at grind time *t*. The two major parameters of the model are –

1. Breakage rate function, *k(x,t)* which is the specific rate of breakage of particles of size *x* at grind time *t* (refer Equation (2)).
2. Cumulative breakage distribution function, *B(x,v)* which is the fraction of particles of size *x* and finer formed when a unit mass of particles of size *v* are broken (refer Equation (3)).

In theory, solution to Equation (1) provides the evolution of particle size distribution in the form of a density function. In practice, however, a close form solution is feasible only for highly specialized and simplified forms for $k(x,t)$ and $B(x,v)$ functions, as discussed later.

Subsequently, PPB approach has been extended to fine grinding applications. Kapur et al (1996) while studying the role of dispersants in kinetics of stirred mill grinding showed that product distributions of stirred media mills follow self-similarity and the evolution of product size distributions could be described by the well-known G-H scheme (Kapur 1987). Nair (1999a and 1999b) and Berthiaux and Dodds (1999) showed that G-H scheme could be used to characterize grinding in fluid energy type mills. Tuzun et al (1995), Berthiaux et al (1996) and Shinohara et al (1999) demonstrated that different forms of Rosin-Rammler-Bennet distribution can represent the particle size distribution in stirred media milling.

A majority of the published work assume that the specific breakage rate function is time invariant and is given by $k(x)$. This is also denoted as linear or first order grinding (Austin 1984). Deviation from first order grinding kinetics, as defined above, could be due to either time invariant but more complex grinding parameters than those discussed in Equations (2) and (3) or inherent non-linear grinding characteristics of the system. In the latter case, it is argued that in many instances the breakage characteristics are influenced by the surrounding particle population (Austin et al 1984; Tangsathikulchai 2003; Fuerstenau et al 2004; Bilgili and Scarlett 2005). The breakage rate can either accelerate or decelerate depending on the material type, size distribution and fineness, type of milling technology, and operating conditions.

In this communication, we present a model based on the extension of the asymptotic self-similarity solution to population balance model (Equation (1)). It is assumed that the breakage rate is a function of the instantaneous size distribution which, in turn, is captured by the time-dependent mean, median or a percentile size of the particle mass. The resulting parametric model describes the evolution of particle size distribution. Several sets of published data from various research groups are employed to test the model.

MATHEMATICAL MODEL

Kapur (1972) showed that for the following functional form of the grinding parameters

$$k(x) = Ax^a \tag{2}$$

and

$$B(x,v) = B\left(\frac{x}{v}\right) \tag{3}$$

Equation (1) exhibits a similarity solution -

$$m(x,t) = \left(\frac{1}{u_1(t)}\right)Z\left(\frac{x}{u_1(t)}\right) \tag{4}$$

Where $u_1(t)$ is the characteristic size of the distribution denoted by the first moment of the mass-diameter density function. $Z(x/u_1(t))$ is a similarity function of scaled particle size $x/u_1(t)$. Kapur (1972) showed that when the breakage distribution function is further specialized in following manner

$$B\left(\frac{x}{v}\right) = \left(\frac{x}{v}\right)^b \tag{5}$$

Equation (4) has an explicit close form similarity solution given by

$$m(x,t) = \left(\frac{K_1}{u_1(t)}\right)\left(\frac{x}{u_1(t)}\right)^{b-1} \exp\left[-\frac{1}{aK_0}\left(\frac{x}{u_1(t)}\right)^a\right] \tag{6}$$

Equation (6) defines the particle size distributions of comminuted particles in most general way in a density distribution form. For example, Rosin-Rammler-Bennet equation is readily obtained from Equation (6) by setting $K_1 = b = a$, $K_0 = 1/a$, and the characteristic size $u_1(t) \propto x_{0.632}(t)$.

By imposing the following boundary conditions on Equation (5) -

$$\int_0^\infty m(x,t)dx = 1 \tag{7}$$

and considering the first moment of the size distribution as -

$$\int_0^\infty x\, m(x,t)dx = u_1(t) \tag{8}$$

Kapur (1972) showed that

$$K_1 = \frac{a}{(aK_0)^{b/a}\Gamma(b/a)} \tag{9}$$

and

$$K_0 = \frac{1}{a}\left[\frac{\Gamma(b/a)}{\Gamma((b+1)/a)}\right]^a \tag{10}$$

Kapur (1972) transformed the similarity function $Z(x/u_1(t))$ into $\overline{Z}(x/u_1(t))$ in order to illustrate the similarity solution in terms of cumulative size distributions, $F(x,t)$, as

$$F(x,t) = \overline{Z}(x/u_1(t)) \tag{11}$$

Thus self-similar or self-preserving distributions at different grind times collapse on to a single curve when cumulative size distributions are plotted as a function of scaled particle size. The self-similarity solution is an asymptotic solution, which becomes increasingly valid with grinding time as the influence of feed size distribution is progressively smoothened out with time due to repetitive breakage. Equation (11) suggests that the self-similar distribution is driven forward in time solely by the mean size of the distribution. For a given percentile, P, Equation (11) also suggests that $x_P(t) \propto u_1(t)$.

Kapur (1971; 1972) emphasized that the similarity solution in Equation (6) is not affected by a time dependent breakage rate function of following kind that could arise when coarse particles are ground preferentially or protected by the surrounding fines,

$$k(x,t) = Ax^a\phi(t) \tag{12}$$

The function $\phi(t)$ may represent a wide-class of functions that in principle describe the non-linear characteristics of the comminution process. In the following discussions we restrict ourselves to one such functional form given below.

$$\phi(t) = u_1(t)^{-c} \tag{13}$$

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Where c is a constant. Kapur (1971; 1972) further showed that the rate of change of mean size of the distributions in similarity solution is given by

$$\frac{\partial u_1(t)}{\partial t} = -AK_0(u_1(t))^{a-c+1} \tag{14}$$

Substituting the value of K_0 from Equation (10) into Equation (14) and integrating Equation (14) from start of grinding to a grind time, t results in

$$\tag{15}$$

$$u_1(t) = \frac{u_1(0)}{\left[1 + A\left(1 - \frac{c}{a}\right)\left(\frac{\Gamma(b/a)}{\Gamma((b+1)/a)}\right)^a (u_1(0))^{a-c} t\right]^{1/(a-c)}}$$

Where $u_1(0)$ represents mean size of the virtual feed size distribution that is self-similar with the asymptotic self-similar size distribution at longer grind times. The concept of virtual feed is rather important here since the idea of self-similarity may not be applicable for real feed size distributions, which are far removed from the eventual self-similar distributions.

Equation (15) can be approximated at higher grind times as

$$u_1(t) = \frac{1}{\left[A\left(1 - \frac{c}{a}\right)\left(\frac{\Gamma(b/a)}{\Gamma((b+1)/a)}\right)^a t\right]^{1/(a-c)}} \tag{16}$$

The cumulative finer size distribution, $F(x,t)$, which gives the mass fraction of material passing size, x at grind time, t is given by

$$F(x,t) = \int_0^x m(x,t)dx \tag{17}$$

Substituting Equation (4) into Equation (17) gives

$$F(x,t) = \int_0^p Z(p)dp \tag{18}$$

Where the dimensionless size, p is given by

$$p = x/u_1(t) \tag{19}$$

Further, substituting the value of similarity function, $Z(p)$ from Equation (6) yields

$$F(x,t) = \left(\frac{K_1(aK_0)^{b/a}}{a}\right)\int_0^{\left(\frac{p^a}{aK_0}\right)} s^{\frac{b}{a}-1} \exp(-s)ds \tag{20}$$

Where s is a transformed dimensionless particle size defined by

$$s = \frac{p^a}{aK_0} \tag{21}$$

Substituting the values of K_1, K_0 and p from Equations. (9), (10) and (19) into Equation (20), after simplification gives

$$F(x,t) = \frac{\gamma\left(\frac{b}{a}, \left(\frac{x}{u_1(t)}\right)^a \left(\frac{\Gamma((b+1)/a)}{\Gamma(b/a)}\right)^a\right)}{\Gamma(b/a)} \qquad (22)$$

Equation (22) suggests that the evolution of particle size distributions in self-similar non-linear batch grinding systems is defined by a regularized gamma distribution. A combination of Equations (22) and (16) yields a parametric model with four parameters namely A, a, b and c, which can be estimated from a given set of experimental data through optimization routines by minimizing the sum of squares of errors between model and experimental values.

In many instances grinding time and specific energy expended are interchangeable (Kapur 1996; Austin 1997). For the experiments that are conducted at various specific energy inputs, E instead of the grind time, t, the grind time in Equation (22) can be replaced by specific energy input to the mill in order to optimize the model parameters.

Equation (11) suggests that under self-similarity

$$u_1(t) = g\, x_{50}(t) \qquad (23)$$

Where g scales the 50th percentile size of the distribution into the mean size of the distribution. Now, substituting Equation (23) into Equation (22) results in

$$F(x,t) = \frac{\gamma\left(\frac{b}{a}, \left(\frac{x}{x_{50}(t)}\right)^a \left(\frac{\Gamma((b+1)/a)}{g\,\Gamma(b/a)}\right)^a\right)}{\Gamma(b/a)} \qquad (24)$$

Equation (24) expresses the evolving self-similar distributions in terms of the median size. At 50th percentile, Equation (24) reduces to

$$\frac{\gamma\left(\frac{b}{a}, \left(\frac{\Gamma((b+1)/a)}{g\,\Gamma(b/a)}\right)^a\right)}{\Gamma(b/a)} = 0.5 \qquad (25)$$

The value of scaling factor g can be obtained by solving Equation (25) numerically which when substituted in Equation (23) gives the mean size of the self-similar distributions for corresponding measured median size.

RESULTS AND DISCUSSIONS

Estimation of Particle Size Distribution

The proposed scheme was validated with eight sets of published data for various materials ground in stirred ball mills. Table 1 gives the parameters of model for these data sets as estimated by a non-linear fit algorithm. Table 1 also includes the sum of least squared errors (SSE) achieved for the best fit as well as the mean size to median size scaling factor, g. It should be noted that the feed distribution was not included in the estimation of model parameters. The parameter estimation scheme proposed in this paper is reasonably less cumbersome. Unlike the G-H solution for the size discretized batch-grinding equation (Kapur 1987), which has two parameters G_i and H_i for each ith size-class particles, the self-similarity solution proposed in this paper has only four parameters to describe the entire grinding process.

Figure 1 shows a comparison of the model predictions with experimental data for parameter sets #3 for limestone and #6 for chrome sand in Table 1. The model simulations are in good agreement with experimental data. Figure 2 illustrates the self-similarity nature of the experimental data on a scaled size, $x/x_{50}(t)$, for the data depicted in Figure 1.

Table 1: Estimated Model Parameters for the Stirred Mill Grinding Data Taken From Literature

| Sl No	Material ground	Size distributions measured at various units of	Model Parameters				SSE	g	Reference
			a	b	c	A			
1	Chalcopyrite concentrate	Specific energy, kWh/t	0.34384	3.42057	-1.62473	2.103E-03	0.0235	1.4294	Fig 3 of Herbst and Sepulveda, 1978
2	Cupric Oxide	Specific energy, kWh/t	0.52270	2.80548	-1.43455	1.298E-03	0.0333	1.3114	Fig 7 of Herbst and Sepulveda, 1978
3	Limestone	Specific energy, kWh/t	0.78843	1.32202	-0.72864	1.036E-03	0.0103	1.4271	Fig 11 of Herbst and Sepulveda, 1978
4	Diamond	Grind time, hrs	1.66501	1.10230	-0.33247	1.236E-01	0.0568	1.2009	Shinohara et al, 1999
5	Hydragillite	Grind time, mins	0.69318	1.34666	-2.02930	7.803E-05	0.0263	1.4957	Berthiaux et al, 1996
6	Chrome Sand	Specific energy, kWh/t	0.93335	1.11292	-0.57936	5.338E-04	0.0118	1.4224	Tuzun et al, 1995
7	Base metal ore	Specific energy, kWh/t	1.26584	1.17441	-0.48806	1.382E-03	0.0067	1.2651	Fig 9 of Kapur et al, 1996
8	Base metal ore	Specific energy, kWh/t	2.69208	0.84274	0.74970	4.099E-04	0.0070	1.1730	Fig 10 of Kapur et al, 1996
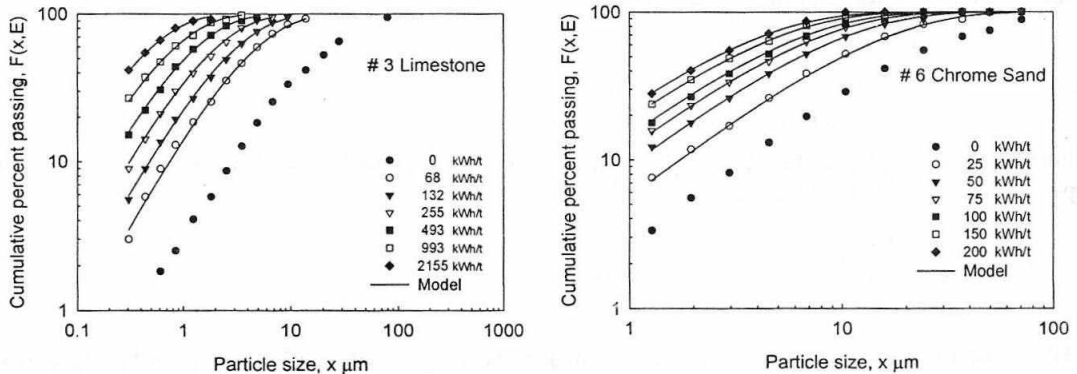
Fig. 1: Comparison of Experimental and Calculated Data for the Data Sets #3 for and #6 of Table 1
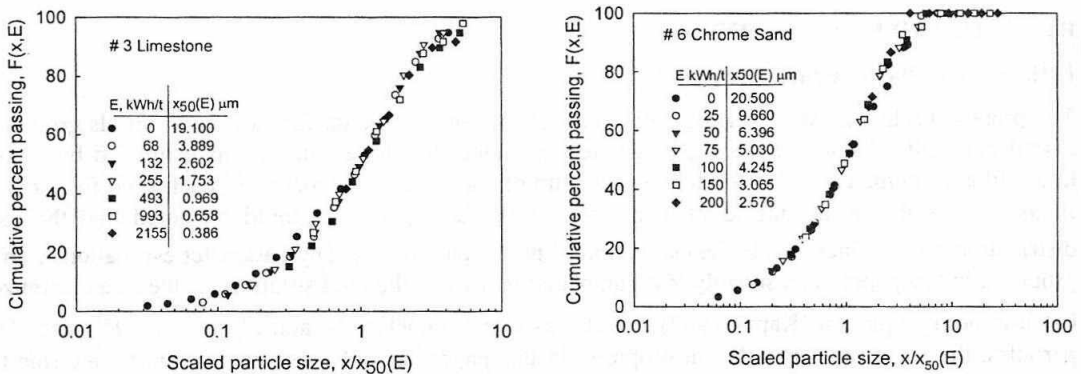

Fig. 2: Self-Similarity in the Experimental Data of Figure 1

Estimation of Breakage Rate

Figure 3 presents the variation of breakage rates of different size particles with the grinding energy input as represented for data sets #2 for cupric oxide and #8 for a base metal ore of Table 1 (refer Equations (12) and (13)). Essentially, the plot depicts that the rate function can increase or decrease monotonically as grinding progresses. The experimental data for cumulative percent retained on a size as a function of grinding energy input shown in Figure 4. For data set #2, the breakage rate decelerates whereas for data set #8, breakage rate accelerates. The different forms of non-linearity encountered in breakage rates have been discussed in literature (Austin et al 1984; Biligili and Scarlett 2005). Furthermore, the model calculations in Figure 3 suggest that the non-linearity in grinding starts right from the beginning of the process, unlike the proposed mechanism of "false time" to describe non-linearity in the later stages of grinding (Yekeler et al 2001; Tangsathitkulchai 2003).
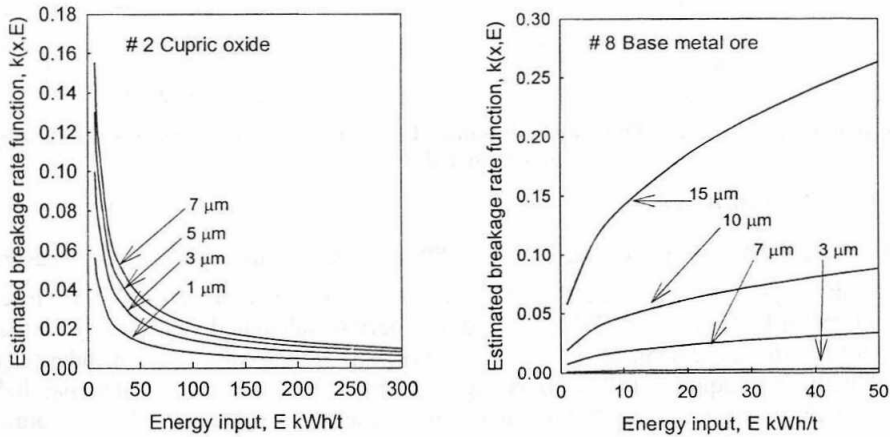

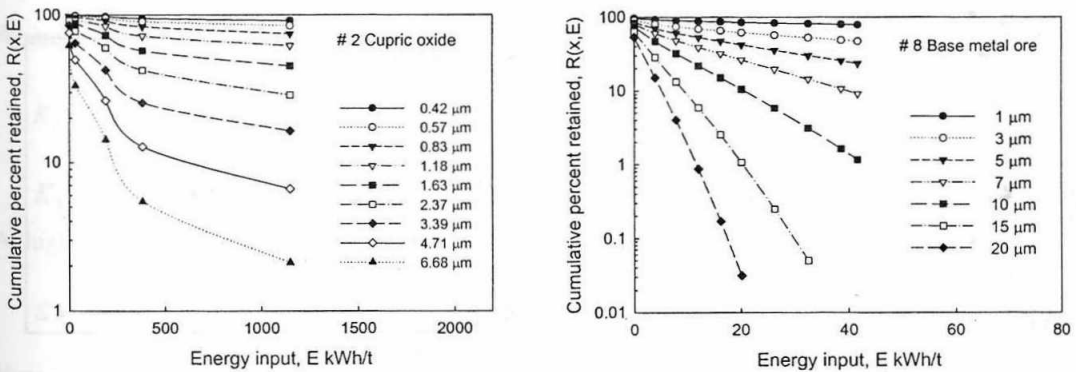**Fig. 3: Estimated Non-Linear Breakage Rate Function as a Function of Input Energy for the Data Sets #2 and #8 of Table 1**

Fig. 4: Non-Linearity of Experimental Data As Depicted on a Semi-Log Plot for the Data Sets of Figure 3

Estimation of Mean Size in Fine Range

It is difficult to get a complete measurement of the particle distribution in the fine size range, which poses problems in the estimation of the mean size of the distribution. The proposed model overcomes this difficulty. Since the model tracks the evolution of particle distributions in the submicron range well, Equation (16) can be used to characterize the mean size of the evolving distributions. Alternatively

Equation (23) can be used to estimate the mean sizes of the evolving self-similar distributions. Figure 5 shows the decrease in the mean size of evolving self-similar product size distributions for the data sets #5 for hydragillite and #8 for a base metal ore of Table 1 as grinding proceeds.
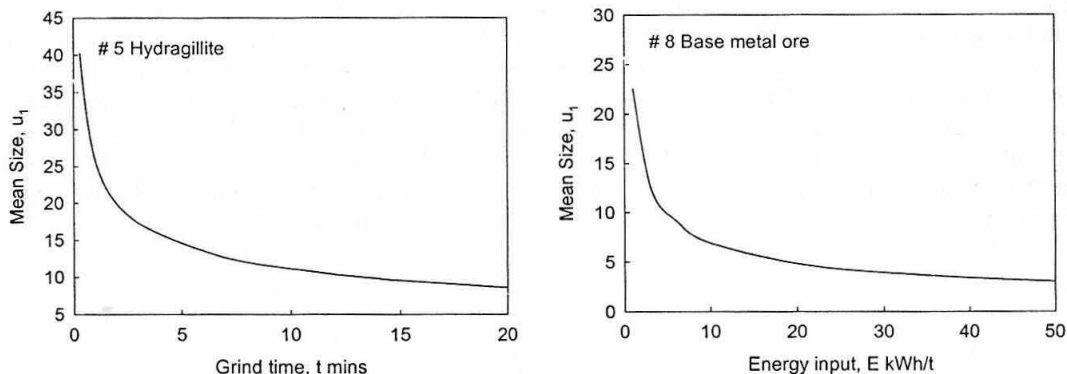

Fig. 5: Estimated Mean Size of the Evolving Self-Similar Product Size Distributions for the Data Sets #5 and #8 of Table 1

Proportionality of Percentile Sizes

Figure 6 shows the decrease in median, x_{50} and 80th percentile size, x_{80} of the measured size distributions with respect to input energy or grind time for the data sets #1 for chalcopyrite concentrate and #5 for hydragillite of Table 1. Also in Figure 6, calculated value of u_1 for these data sets using Equation (16) are shown. For t or $\bar{E} >> 0$, the plots show power law dependence (refer to Equation (16)) with slope equal to $(-1/(a-c))$ as suggested by Table 1 parameters. The parallel trends in the presented values in Figure 6 indicate that x_{50} and x_{80}, and u_1 values are indeed proportional to each other. Figure 7 shows a linear relationship for $(a-c+1)$ values calculated from the model with those obtained by linear-regression of x_{50} and x_{80} values of the measured size distributions, which also shows that the different percentile sizes of self-similar distribution are proportional to each other.
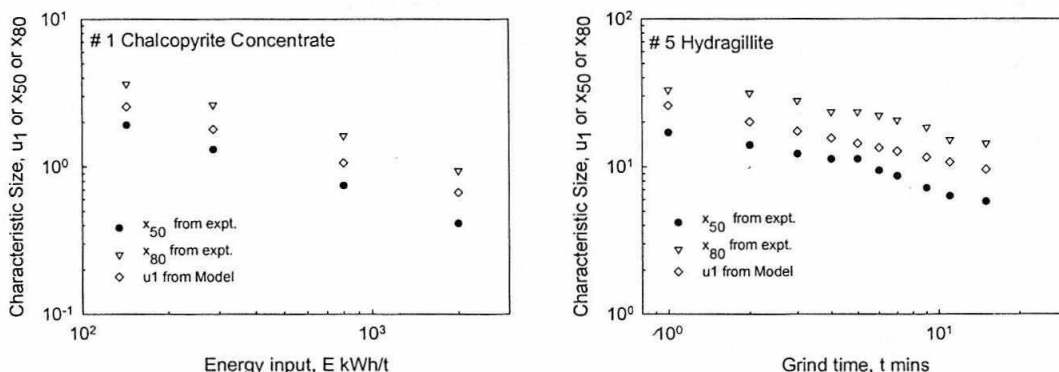

Fig. 6: The Proportionality between x_{50} and x_{80} of Measured Distributions with Model Estimated Mean Size, u_1
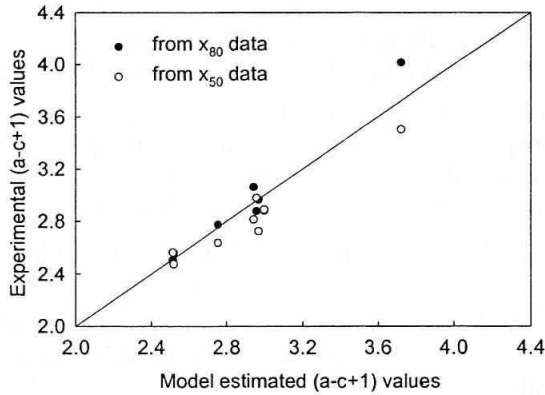
Fig. 7: The Correlation between Model $(a - c + 1)$ Values with Estimated $(a - c + 1)$ Values From Regression of x_{50} and x_{80} Values of Measured Size Distributions

Energy-Size Relationships

Equation (14) when expressed in terms of input grinding energy, E, gives the general relationship between the mean size and input grinding energy. Since the mean size is proportional to any percentile of the distribution, Equation (14) represents the general energy law of grinding (Kapur 1987). Kick, Bond and Rittinger have proposed (Austin, 1984) the value of exponent to be 1, 1.5 and 2 respectively. However, the current model reveals that for fine grinding range this exponent is more than 2.5 $((a-c+1)$ values from Table 1). Therefore it is inferred that conventional energy-size relationships do not hold good in the fine grinding range.

Furthermore, Equation (15) can be written in terms of grinding energy expended, E as

$$E = \frac{1}{K_2}\left(\frac{K_3}{(u_1(E))^{(a-c)}} - 1\right) \tag{26}$$

Where,

$$K_2 = A\left(1 - \frac{c}{a}\right)\left(\frac{\Gamma(b/a)}{\Gamma((b+1)/a)}\right)^a (u_1(0))^{a-c} \tag{27}$$

$$K_3 = (u_1(0))^{(a-c)} \tag{28}$$

At higher reduction ratio, Equation (26) reduces to a familiar form of classical energy law

$$E = \frac{K_3}{K_2}\left((u_1(E))^{-(a-c)}\right) \tag{29}$$

Here $u_1(E)$ can be replaced by a percentile size by invoking the proportionality relation between them (similar to Equation (23)). Hence it is verified that Charles law holds good for fine grinding in stirred mills for different operating conditions. This finding is in the same line of experimental results reported in literature (Sepulveda and Herbst 1987; Gao et al 1995).

CONCLUSIONS

A parametric approach of modeling fine particle grinding data using similarity solution of population balance equation is presented in this article. The model has several prominent features as listed below

1. The model incorporates non-linear breakage rate parameter and it is successfully tested on 8 sets of stirred media milling data obtained from published work involving different kinds of material and milling conditions. The model is also simple where numbers of estimated parameters are only four.

2. The parametric model is quite general. Popular Rosin-Rammler-Bennet distribution can be derived from it under special conditions. Furthermore, a general particle size-energy relationship can be deduced from it. It also reveals that Kick, Rittinger and Bond's law cannot characterize the grinding operation in very fine range, as the size-energy exponent is more than 2.5.

3. Breakage rate estimated from the model parameters shows that non-linear breakage rate prevails right from the beginning of the grinding.

4. Lastly, a major advantage of the model is that it allows one to estimate the mean size from the measured data in the fine size range, which otherwise is difficult to characterize in the absence of knowledge of complete distribution.

NOMENCLATURE

A	parameter in Equation (2)
a	exponent in Equation (2)
b	exponent in Equation (5)
$B(x,v)$	cumulative breakage distribution function
c	parameter accounting non-linearity in Equation (13)
E	specific energy input to the mill, kWh/t
F	cumulative percent passing
g	scaling factor for 50th percentile defined in Equation (23)
G_i	G-H model parameter
H_i	G-H model parameter
K_0	constant defined in Equation (10)
K_1	constant defined in Equation (9)
K_2	constant defined in Equation (27)
K_3	constant defined in Equation (28)
$k(x)$	time independent breakage rate function
$k(x,t)$	time dependent breakage rate function
$m(x, t)$	particle size distribution in density form at grind time t
p	reduced particle size defined by Equation (19)
P	percentile
R	cumulative percent retained
s	transformed dimensionless particle size defined by Equation (21)
t	grinding time
u_1	mean size of the particle distribution
v, x	particle size, microns
x_{50}, x_{80}	passing sizes for 50th percentile (median) and 80th percentile
$Z, \overline{Z}$	similarity functions

GREEK SYMBOLS

γ, Γ	gamma functions
ϕ	non-linearity function defined in Equation (12)

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