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FIRST-ORDER AND SECOND-ORDER BREAKAGE RATE OF COARSE PARTICLES IN BALL MILL GRINDING

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Abstract: It has been observed by many authors that the breakage rates of coarse particles in a ball mill slow down with increasing grinding time and deviate from the first order. In this paper it is intended to find out whether the breakage rates of coarse particles obey second-order kinetics or not. For this purpose, quartz, limestone, iron ore and a mixture of quartz/limestone (weight ratio of 1:1) were selected as a ball mill feed. The first-order breakage rate was determined for the four particle sizes of quartz, limestone, iron ore and the mixture of quartz/limestone. Results indicating good first-order kinetics were obtained with the fine-sized particles (-1.2+1 mm, -0.6+0.42 mm). However, the coarse-sized particles (-5+4 mm, -3.15+2.5 mm) showed deviations from the first order. These coarse particles were in the abnormal breakage region. The second-order breakage rate was determined for the coarse particles (-5+4 mm, -3.15+2.5 mm). It can be seen that, for both sizes and all the materials, the second-order plot had better fit than the first-order plot. Also, it can be concluded that the second-order kinetics could model the breakage of coarse particles better than the first-order kinetics, and the validity of the second-order breakage rate was increased with increasing particle size. However, it is suggested to examine the validity of the second-order breakage rate kinetics for other materials and particle sizes.

Keywords: ball mill, grinding kinetic, particle breakage

Introduction

The breakage rate equation, similar to a chemical reaction, can be expressed in a general way as follows:

$$-\frac{dm}{dt} = Sm^n \quad (1)$$

where m is the total mass, t is time, S is the breakage rate constant or selection function, and n is the order of equation.

Loveday (1967) and Austin (1971) have found experimentally that, in the case of batch grinding, the breakage rate obeys a first-order law or $n=1$. However, there is no valid reason for this.

If the total mass load in the mill is at particle size $i=l$ (single-sized particles), then for a first-order breakage process (Austin, 1971) is:

$$-\frac{dm_1}{dt} = S_1 m_1 \quad (2)$$

It is usually adequate to assume that S_l does not vary with time. Then, with integration from $t = 0$ to t :

$$m_1(t) = m_1(0) \exp(-S_1 t) \quad (3)$$

or taking logs

$$\log(m_1(t)) = \log(m_1(0)) - \frac{S_1 t}{2.303} \quad (4)$$

where $m_l(0)$ and $m_l(t)$ are respectively the weight of particle size $i=l$ at time 0 and t and S_l is a proportionality constant called specific rate of breakage with unit of time^{-1} . A plot of $\log\left(\frac{m_1(t)}{m_1(0)}\right)$ versus t should give a straight line with slope $\left(\frac{S}{2.033}\right)$.

The kinetics of breakage has been studied by many researchers, the majority of whom have classified breakage as a first-order reaction. However, it has been reported by various researchers that breakage rate deviates from the first order; this breakage is called non-first order (Austin et al., 1971; Austin et al., 1972; Austin et al., 1977; Gardner et al., 1975; Mankosa et al., 1989; Tuzun et al., 1995; Cho et al., 1996; Gao et al., 1993; Fuerstenau et al., 2004).

It has been observed by many authors that the breakage rates of coarse particles in the ball mill slow down with increasing grinding time and deviate from the first order (Tavares et al., 2009; Austin et al., 1982; 2006). This behaviour has been often encountered for the sizes above those corresponding to the maximum breakage rate (Tavares et al., 2009) and has been called by Austin "abnormal breakage region". The deviation from the first-order grinding kinetics is resulting from either mill conditions or material properties and has been explained in the literature with two points. First, as grinding time increases, fine products accumulated in the mill cover the coarse material and prevent further grinding, which is usually encountered in fine-dry grinding and high-viscosity wet grinding and is called the "medium effect". The second point could be that the size of the material is much bigger than the grinding ball diameter. In this case, coarse particles cannot be nipped by the balls and some amount of the material behaves as a relatively weaker material, while some behaves as a relatively stronger material. It is called by Austin the "material effect", which is in contrast with an "environmental effect", by which breakage rates of all sizes slow down as fines accumulate in the charge in dry grinding (Austin and Bagga, 1981). However, it is an oversimplification of the physical phenomenon, since materials are known to have a range of fracture energies (Tavares and King, 1998; Tavares, 2007). In addition, some researchers believe machines operating prior to grinding stage effect

on the grinding breakage rate and this may cause to deviation from first order breakage rate. This can be called the ‘device effect’ (Tumidajski et al., 2010; Gawenda, 2013; Saramak, 2013).

Gardner and Rogers (1975) presented the mathematical formulation for milling a material which behaved as if it consisted of a mixture of soft and hard components, each breaking in a first-order manner. They noted that attaching physical significance to the two hypothesized components was avoided.

Second-order breakage rate

The kinetics of some mineral processing processes is second-order rate or between first- and second-order rates. For example, the kinetics of froth flotation obeys the second order for low-grade ores or more concentrated pulps.

In this paper, it is tried to find out whether the breakage rates of coarse particles in the abnormal breakage region obey the second order or not. If the breakage material is assumed as one reactant and the breakage rate obeys the second order ($n=2$), then Eq. 1 can be written as follows:

$$-\frac{d[m_1]}{dt} = S_1[m_1]^2 \quad (5)$$

and integration from $t = 0$ to t gives

$$\frac{m_1(0)}{m_1(t)} = 1 + m_1(0)S_1t \quad (6)$$

where $m_i(0)$ and $m_i(t)$ are the weight of particle size $i=l$ at time 0 and t , respectively, and S_l is a proportionality constant called specific rate of breakage with the unit of $\text{mass}^{-1} \cdot \text{time}^{-1}$. A plot of $\frac{m_1(t)}{m_1(0)}$ versus t should give a straight line with slope $(m_1(0)S_1)$ and y-intercept (1).

Materials, equipment, and procedure

Materials

The materials used in this research were quartz, limestone and iron ore. Quartz is a hard material (7 on the Mohs scale) and limestone is a soft material (3 on the Mohs scale). A mixture of quartz/limestone (weight ratio 1:1) was also prepared as the medium-soft material. Large chunks of each material (quartz, limestone and iron ore) were broken in a jaw and roll crusher. The crushed materials were first screened and then carefully sieved to obtain the single-sized particles. Four single-sized particles sizes (-5+4 mm, -3.15+2.5 mm, -1.2+1 mm, -0.6+0.42 mm) were prepared from quartz, limestone, iron ore and the quartz/limestone mixture.

Mill

Grinding tests were performed in a 20 cm×20 cm stainless steel laboratory mill. It was operated at the constant speed of 85 rpm (84 % of the critical speed). The mill charge consisted of stainless steel balls with 16 and 42 mm diameter and the total ball load weighed 8.79 kg. The volume of the ball charge with voids was 25% of the total volume of the mill.

Procedure

Dry grinding experiments were carried out using the single-sized particles of quartz, limestone, iron ore and the quartz/limestone mixture. The material weight in each experiment was 350 g. Each grinding experiment was continued for short time periods until about 90% of the material passed through the sieve (fraction). After each grinding period, the mill content was discharged and sieved for 20 min and the remaining materials were determined. Then, the whole material was returned to the mill for the next grinding period. By fitting Eqs. 4 and 6 to the experimental grinding data, the first- and second-order breakage rates of the particles sizes were determined, respectively.

Results and discussion

Figs. 1-4 show the first-order plots for various feed sizes of quartz, limestone, and the mixture of quartz/limestone and iron ore. The coefficient of determination, denoted by R^2 , which indicates how well data fit a breakage rate kinetic model was calculated and reported in Fig. 5. The results showed that R^2 decreased with increasing the particle size. The results indicated that good first-order kinetics was obtained with fine-sized particles (-1.2+1 mm, -0.6+0.42 mm). However, the coarse particle sizes (-5+4 mm, -3.15+2.5 mm) showed deviations from the first order. Figure 6 demonstrated the variation of the first-order specific rate of breakage values (S_1) against feed particle sizes. It can be seen that the coarse-sized particles were in the abnormal breakage region. As mentioned before, in the abnormal breakage region, deviation from the first-order breakage occurred, because the particles were too large to be properly nipped by the balls.

For the evaluation, the second-order breakage rate, Eq. (6), was fitted to the grinding data of the coarse particles. Figs. 7-10 show the second-order plots for the coarse feed sizes (-5+4 mm, -3.15+2.5 mm) of quartz, limestone, iron ore and the mixture of quartz/limestone. Table 1 demonstrates the comparison between the R^2 of the first- and second-order plots for the coarse particle sizes of quartz, limestone, iron ore and the mixture of quartz/limestone. It can be seen that, for both particle sizes and all the materials, the R^2 of the second-order plot was higher than that of the first order; i.e. the second-order plot had a better fit than the first-order plot, which was especially true for -5+4 mm particles and harder materials. It can be concluded that, in the

abnormal breakage region, with increasing particle size, the validity of the second-order breakage rate would be increased.

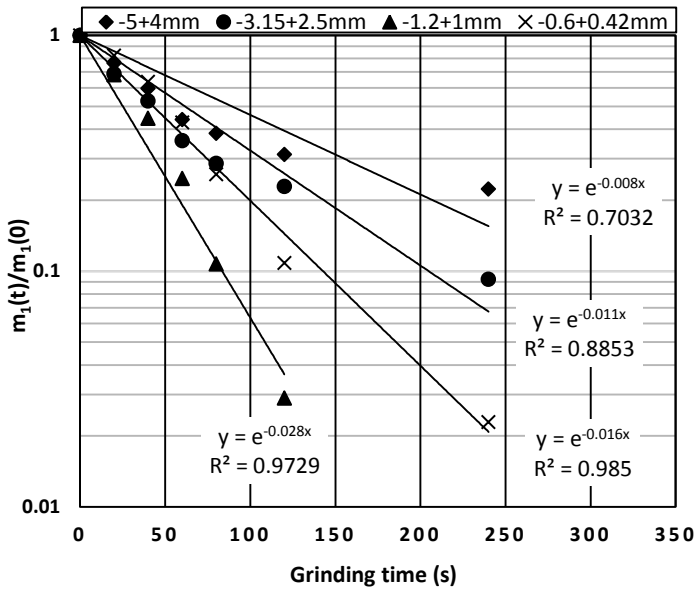


Fig. 1. First-order plots for various feed sizes of quartz

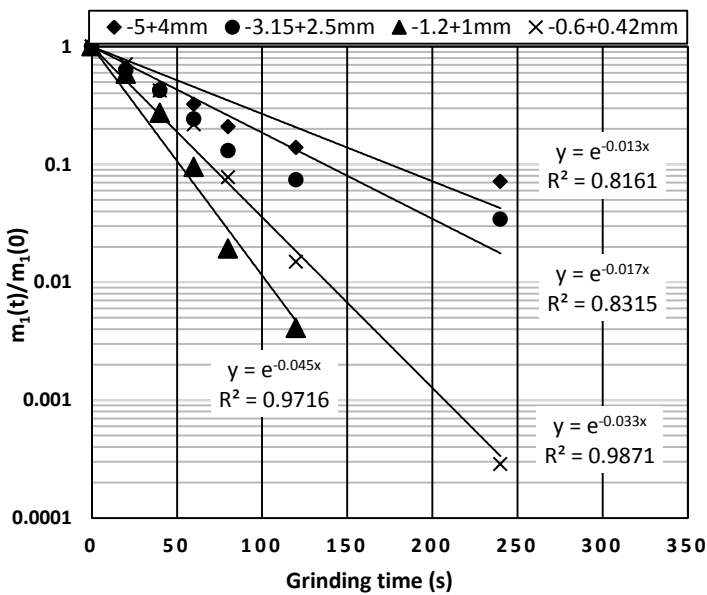


Fig. 2. First-order plots for various feed sizes of limestone

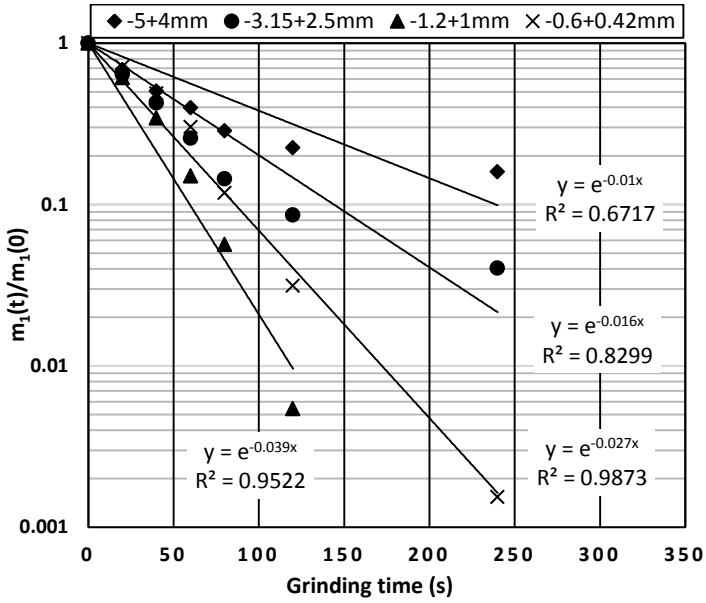


Fig. 3. First-order plots for various feed sizes of the mixture of quartz/ limestone (weight ratio of 1:1)

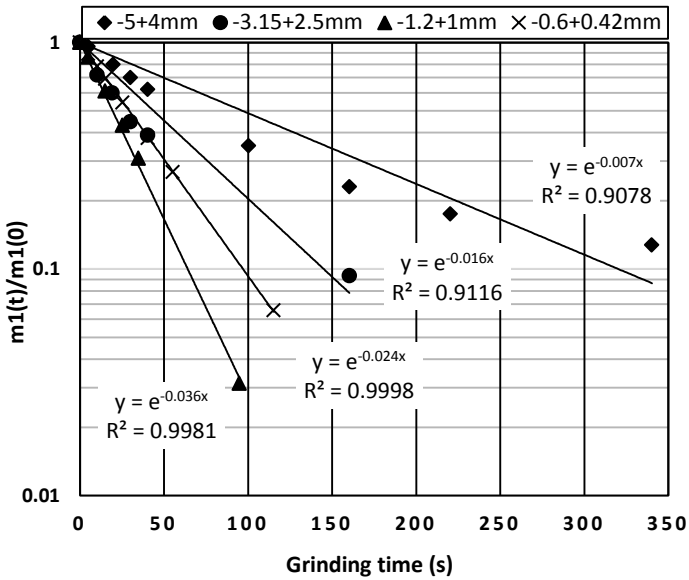


Fig. 4. First-order plots for various feed sizes of iron ore

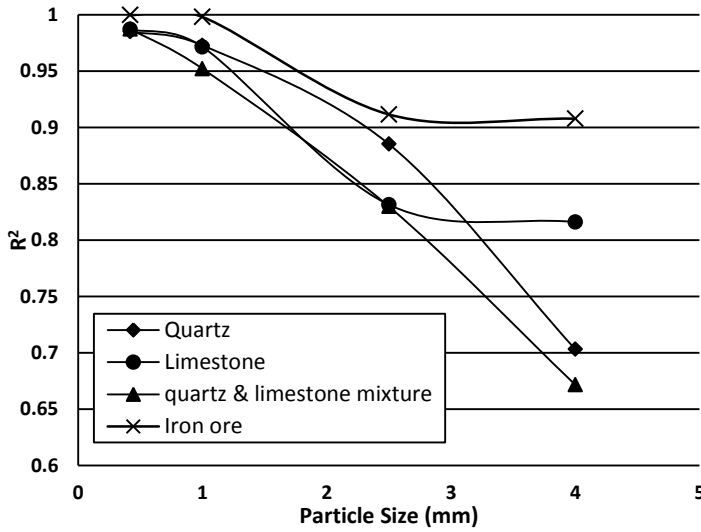


Fig. 5. R^2 of the first order kinetics model versus particle size

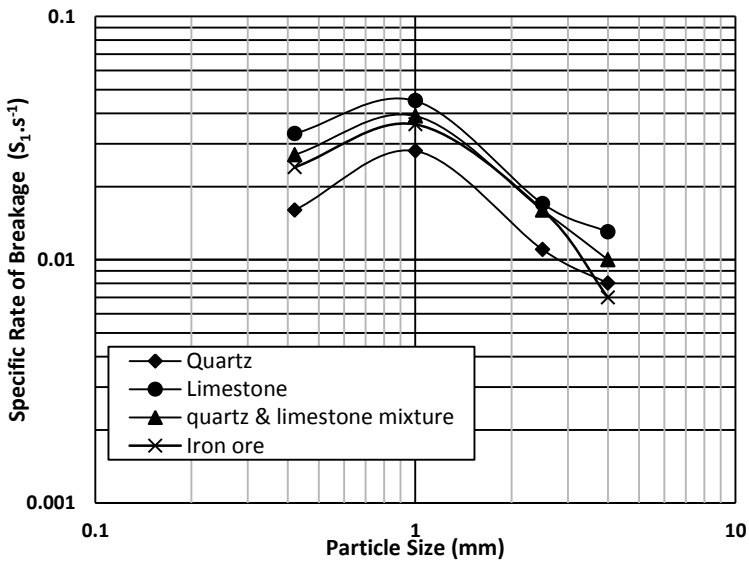


Fig. 6. Specific rates of breakage as function of particle size

Tavares and Carvalho (2009) showed that breakage does not necessarily follow a simple power law time, but is rather related to the complex micro-behavior of individual particles and fundamental fracture mechanics theory. The Tavares and Carvalho model is a very complex one with many parameters that are difficult to measure or nigh on impossible to back-calculate because of parameter sensitivity issues. This model is also suspect in that the breakage distribution function is based on

single particle impact tests, rather than the statistical approach advocated by Austin (1971) where the particle bed in the ball mill is treated as a statistical ensemble composing many particles.

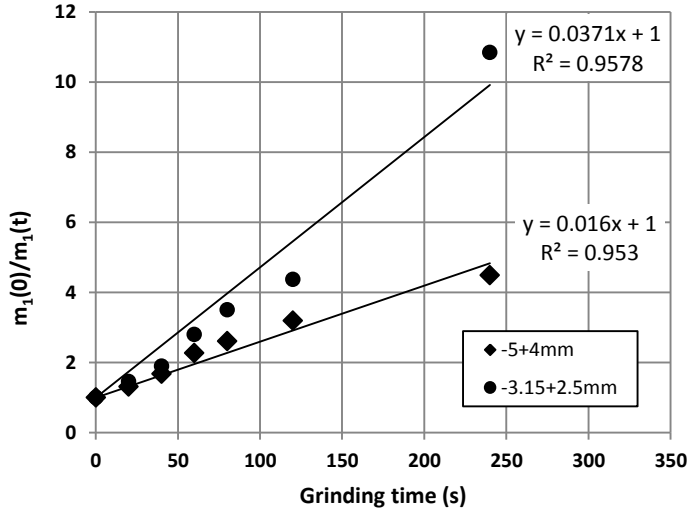


Fig. 7. Second-order plots for the coarse feed sizes of quartz

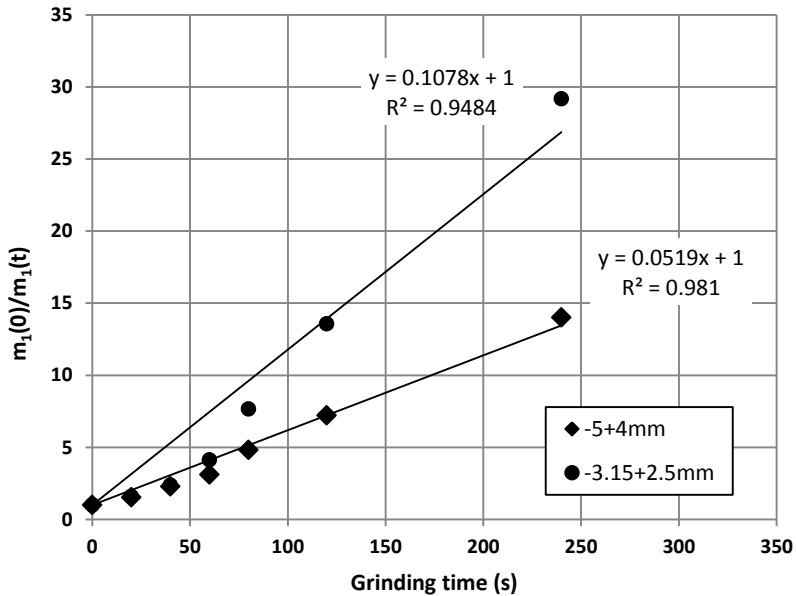


Fig. 8. Second-order plots for the coarse feed sizes of limestone

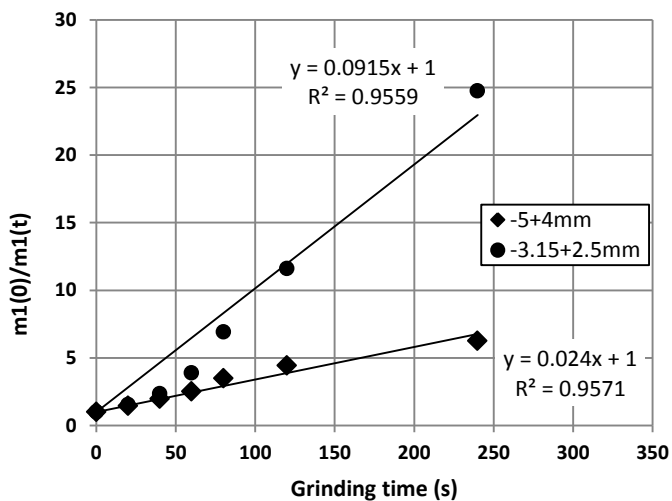


Fig. 9. Second-order plots for the coarse feed sizes of the mixture of quartz/limestone (weight ratio of 1:1)

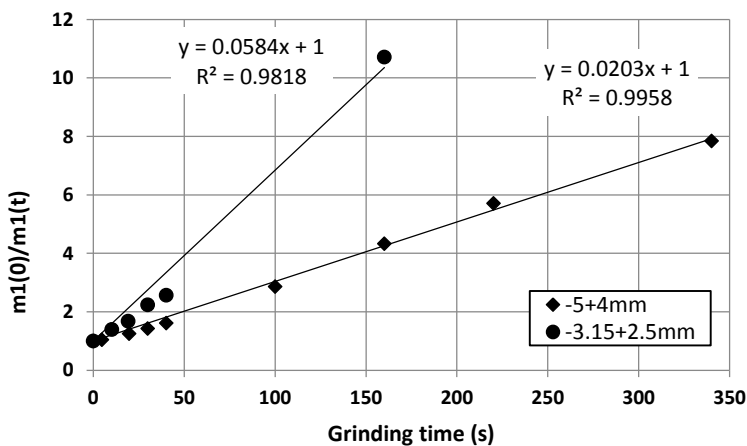


Fig. 10. Second-order plots for the coarse feed sizes of iron ore

Table 1. R-squared of first and second order for the coarse particles sizes

kinetic order	-5+4mm				-3.15+2.5mm			
	quartz	limestone	q&l mixture	iron ore	quartz	limestone	q&l mixture	iron ore
First-order	0.7032	0.8161	0.6717	0.9078	0.8853	0.8315	0.8299	0.9116
Second-order	0.9531	0.9810	0.9571	0.9958	0.9578	0.9484	0.9559	0.9818

Summary and conclusion

The first-order breakage rate was determined for four particle sizes of quartz, limestone, iron ore and a mixture of quartz/limestone (weight ratio of 1:1). The results indicated good first-order kinetics for fine particle sizes (-1.2+1 mm, -0.6+0.42 mm). However, the coarse particle sizes (-5+4 mm, -3.15+2.5 mm) showed deviations from the first order. These coarse particle sizes were in the abnormal breakage region. In the abnormal breakage region, deviation from first-order breakage occurred, because the particles were too large to be properly nipped by the balls.

The second-order breakage rate was determined for the coarse particles (-5+4 mm, -3.15+2.5 mm). It can be seen that, for both particles sizes and all the materials, the second-order plot had a better fit than the first-order plot, which was especially true for -5+4 mm particles and harder materials. It can be concluded that, in the abnormal breakage region, with increasing particle size, the validity of the second-order breakage rate was increased.

The results of this research showed that the second-order kinetics can model the breakage of coarse particles better than the first-order kinetics. This phenomenon was also observed by the authors of this paper for different iron ore samples. However, it is suggested to examine the validity of the second-order breakage rate kinetics for other materials and particle sizes as well.

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