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Identification of the breakage rate and distribution parameters in a non-linear population balance model for batch milling

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ABSTRACT

Non-linear population balance models (PBMs), which have been recently introduced due to the limitations of the classical linear time-invariant (LTINV) model, account for multi-particle interactions and thus are capable of predicting many types of complex non-first order breakage kinetics during size reduction operations. No attempt has been made in the literature to estimate the non-linear model parameters by fitting the model to experimental data and to discriminate various models based on statistical analysis. In this study, a fully numerical back-calculation method was developed in the Matlab environment to determine the model parameters of the non-linear PBM. Not only does the back-calculation method identify the parameters of complicated non-linear PBMs, but also it gives the goodness of fit and certainty of the parameters. The performance of the back-calculation method was first assessed on computer-generated batch milling data with and without random error. The back-calculation method was then applied to experimental batch milling data exhibiting non-first order effects using both the LTINV model and two separate non-linear models. The back-calculation method was able to correctly determine the model parameters of relatively small sets of batch milling data with random errors. Applied to experimental batch milling data, the back-calculation method with a two-parameter non-linear model yielded parameters with reasonable certainty and accurately predicted the slowing-down phenomenon during dry batch milling. This study encourages experimenters to use advanced non-linear population balance models along with the back-calculation method toward estimating the breakage rate and distribution parameters from dense batch milling data sets.

1. Introduction and mathematical formulation

Population balance models (PBMs) as mathematical description of size reduction have been used extensively in literature [1–6]. Early PBMs for milling describe size reduction as a first-order rate process as developed by Sedlatschek and Bass [7]. Their first-order breakage hypothesis simply stated that the disappearance of particles of a given size due to breakage is proportional to the weight of particles of that size present. Later, concepts of breakage distribution and probability of selection-for-breakage [8,9] were included by Broadbent and Callcott [10–13], but they treated particle breakage as occurring in stages. In order to apply the PBM to time-continuous milling processes, Gaudin and Meloy [14] derived the time- and size-continuous mass density form of the equation for a well-mixed batch milling process. More commonly, the time-continuous size-discrete form seen in Eq. (1) is preferred as experimental data is inevitably in discrete form [2].

$$\frac{dM_i(t)}{dt} = -S_iM_i(t) + \sum_{j=1}^{i-1} b_{ij}S_jM_j(t) \qquad N \geq i \geq j \geq 1 \text{ with } M_i(0) = M_{ini} \tag{1}$$

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In Eq. (1), i and j are the size-class indices and extend from size-class 1 containing the coarsest particles to size class N containing the finest particles usually in a geometric progression. M_i represents the mass fraction of particles in size-class i. S_i is the specific breakage rate parameter and b_{ij} is the breakage distribution parameter, which describes the distribution of particles formed when a particle of size class j is broken. This equation is also known as the linear time-invariant (LTINV) model because the specific breakage rate does not vary with time and its discretized value is only dependent on particle size of given size class. The following constraints also apply to Eq. (1) due to the conservation of mass:

$$S_N = 0, \quad \sum_{i=j+1}^{N} b_{ij} = 1, b_{ii} = 0. \tag{2}$$

Population balance models have the ability to simulate the evolution of the particle size distribution of a milling process, but also to elucidate the breakage mechanisms (e.g. fracture, cleavage, attrition) [6,15–17]. Numerous methods to determine S_i and b_{ij} from experimental milling data for the LTINV model have been proposed using both direct measurements and back-calculation [18–24]. The so-called "direct measurement method" demands tedious breakage experiments on numerous mono-sized feeds to determine the parameters without resorting to complex non-linear optimization methods. Austin and Bhatia [18] outlined the experimental procedure for determining the breakage rate parameter and breakage distribution parameter assuming first-order breakage. Austin and Luckie [19] also detailed multiple methods, known as the BI, BII, and BIII methods, to determine the breakage distribution parameters. Aside from obvious complications of these methods such as time-consuming preparation of numerous mono-sized feeds, they also require certain assumptions such as negligible re-breakage of particles which may cause error in the calculation. Back-calculation, a technique which calculates the model parameters that best fit the model to the experimental data, is also widely employed and has significant advantages over the direct measurements. It allows the milling of a natural-sized feed instead of multiple mono-sized feeds and reduces the need for laborious preparation of material. Both linear [22] and non-linear [21,23] optimization techniques have been used with either Reid's analytical solution [25] to the batch milling equation or other approximate solutions [26] to back-calculate the model parameters.

Despite the varying success of the above methods and application of the LTINV model, non-random deviation between experimental milling data and modeled predictions of Eq. (1) were noticed and became a source of some criticism [4,27–29]. Such deviations became pronounced at long milling times or when the presence of fines became significant. Austin and Bagga [4] investigated such deviation and observed that the specific rate of breakage decreased as fines accumulated and determined that the source of non-first order effects originated from a cushioning action provided by fine particles. To account for this, Austin and Bagga [4] and Austin et al. [27] introduced a time-dependence to the specific breakage rate parameter. They subsequently assumed that the specific rates of breakage for all particle sizes varied in the same way as the milling environment changed according to an acceleration–deceleration function $\kappa(t)$. They solved the resulting linear time-variant (LTVAR) model by invoking the concept of a false time or equivalent first-order grind time, θ, as seen in Eq. (3).

$$\begin{aligned} &\frac{dM_i(\theta)}{d\theta} = -S_i(0)M_i(\theta) + \sum_{j=1}^{i-1} b_{ij}S_j(0)M_j(\theta) \\ &N \geq i \geq j \geq 1 \text{ with } M_i(0) = M_{ini} \\ &d\theta = \kappa(t)dt \text{ with } \theta(0) = 0 \end{aligned} \quad (3)$$

False time may be correlated to true grind time through a series of mono-sized feed milling experiments. The acceleration–deceleration function may be determined in the same manner. While this method can correctly predict a decrease (or increase) in specific breakage rate as milling progresses, it still lacked the ability to account for the source of the non-first order effects (i.e. cushioning of coarse particles by fine particles) in an explicit way. Similarly, other time-variant models to account for non-first order effects based on Kapur's method [22] do not either [28,30].

Bilgili and Scarlett [31] introduced a population balance framework to mathematically explain non-first order effects arising from multi-particle interactions for rate processes. In their model, the specific breakage rate is decomposed into an apparent breakage rate and a population dependent functional where the functional describes different types of non-first order breakage kinetics. Their non-linear population balance model in Eq. (4) is shown in size-discrete form.

$$\begin{aligned} &\frac{dM_i(t)}{dt} = -k_iF_i\left[\sum_{q=1}^{N} P_{iq}M_q(t)\right]M_i(t) + \sum_{j=1}^{i-1} b_{ij}k_jF_j\left[\sum_{q=1}^{N} P_{jq}M_q(t)\right]M_j(t) \\ &N \geq i \geq j \geq 1 \text{ with } M_i(0) = M_{ini} \end{aligned} \quad (4)$$

In Eq. (4), k_i is the apparent specific breakage rate parameter, F_i is a functional of the weighted distribution of the mass fraction where P_{iq} expresses the contribution of the generic size q to the disappearance rate of particles of size i due to multi-particle interactions, and all other terms are identical to Eq. (1). The following constraints also apply to Eq. (4):

$$\begin{aligned} &k_i \geq 0, k_N = 0, \sum_{i=j+1}^{N} b_{ij} = 1, b_{ii} = 0 \\ &F_i[\,] \geq 0,\ F_i[\,] \rightarrow 1\ as\ \forall M_q(q \neq i, t) \rightarrow 0. \end{aligned} \quad (5)$$

The choice of functional, as discussed by Bilgili and Scarlett [31] and Bilgili et al. [32], is partly empirical and depends on mill type, design variables, operation mode, operating variables, and material properties. However, it is well-established that short-time milling of a mono-sized feed is first-order [1]. It has also been shown in many milling studies [4,30,33,34] that non-first order kinetics is significantly contributed by multi-particle interactions. In other words, the breakage of a particle is affected by the surrounding population. Specifically, for dry milling in a ball mill, finer particles exert a “cushioning action” on the coarser particles, thus reducing the specific breakage rate of the coarser particles [4]. The functional chosen (e.g. $F \equiv \exp[\,], (1+[\,]^{-1})$, etc.) and the weighting function P must accurately reflect these considerations and physical interactions. Finally, whatever functional chosen, it should explain Type I and Types II or III deviations from the LTINV model [31], while yielding parameters with higher statistical certainty than the LTINV model, as being demonstrated in this paper. Therefore, if a functional is chosen completely arbitrarily without taking into account any of the above considerations, the non-linear model will probably fail, resulting in inferior predictions and parameters as compared with those of the LTINV model.

Numerical simulations using a simplified version of Eq. (4), known as Model B and shown in Eq. (6) in the sequel, were performed by Bilgili and Scarlett [31] and Bilgili et al. [32]. In their work, two specific forms of the functional (Eqs. (7) and (11) in this study) were considered and found to successfully describe different types of non-linear kinetics. Furthermore, they successfully predicted the three types of milling behavior observed in literature including the decrease in specific breakage rate observed by Austin [27] that could not be explained by the LTINV model, i.e., Eq. (1). Further novelty of their framework extends from the fact that the non-linear model reduces to the linear model in the absence of multi-particle influence or in other words when $F[\,] \rightarrow 1$. Due to the ability of this non-linear population balance model to encompass many different types of particle breakage kinetics phenomenologically, we argue that it can lead to better design, control, and optimization of size reduction processes.

Because no analytical solution exists for the non-linear model, calculation of the breakage rate parameters, breakage distribution parameters, and the non-linear functional parameters remains a formidable challenge. Determining the model parameters is of upmost importance in order to use the PBM for process simulation, design, scaling, control, and optimization [1–3,15,35]. Having estimated the model parameters for a given set of operating conditions and material in a mill, one can predict the temporal evolution of the size distribution and final product size distribution for any given feed (initial) particle size distribution. This is especially important when non-first-order effects are significant, which result in a pronounced impact of the initial condition on the temporal evolution [31]. In fact, multiple feed size distributions can also be used to generate more dense data sets for determining PBM parameters with more statistical certainty. In addition, one can fit the same model to size distributions obtained from different operating conditions (e.g. volume percent media loading in ball milling) and assess the impact on the model parameters for a given material. This can allow a numerical optimization of the process parameters. Similarly, for the same operating conditions in the mill, one can determine the specific rate of breakage of different materials and assess their relative particle strengths. Finally, fitting the non-linear PBM