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Insight into first-order breakage kinetics using a particle-scale breakage rate constant

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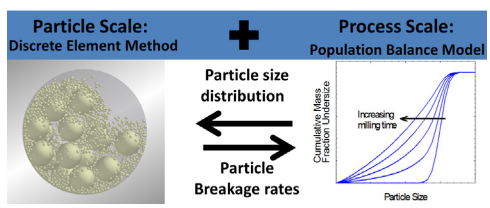
HIGHLIGHTS

- Formulated a particle-scale specific breakage rate of the PBM for dry milling.
- Validated the approach by quantifying specific breakage rates in ball milling.
- Demonstrated erroneous analysis of DEM results without a threshold impact energy.
- Elucidated impact of ball size and feed polydispersity on the breakage kinetics.
- Found no impact of evolving particle population on the specific breakage rate.

GRAPHICAL ABSTRACT



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ABSTRACT

This study formulates a particle-scale breakage rate constant of the linear time-variant population balance model (PBM) for batch dry-milling. The breakage rate constant separates material properties from the milling environment which is defined by the impact energy distribution obtained by the discrete element method (DEM). The breakage rate constant determined by experiments and DEM simulations of the ball milling of polydispersed silica glass were in close agreement confirming the validity of the proposed methods. The breakage rate constant formulated in this study also allowed a detailed and rigorous energy-based analysis of the milling environment which provided insight into the origin of first-order breakage kinetics and the time-invariance of the breakage rate constant. Feeds of varying polydispersity and the time-wise evolution of the particle size distribution (PSD) were simulated in DEM to show that the PSD does not affect the specific breakage rate constant as assumed in first-order breakage. The adherence to first-order breakage kinetics was attributed to the low feed loading in the mill and the absence of particle beds where mechanical multi-particle interactions can cause non-first-order breakage. It was found that a threshold impact energy must be accounted for in the analysis of DEM results because the majority of low energy impacts do not contribute to particle breakage. Without rigorous assessment of the impact energy distribution, DEM simulations may lead to an erroneous evaluation of milling performance. The findings of this study contribute to a unified DEM–PBM framework to model and analyze milling process and demonstrate that milling performance can be quantified from particle-scale interactions using DEM.

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1. Introduction

The need for greater control, efficiency, and product quality for milling processes in industries such as the ceramic, pharmaceutical, and mineral has motivated the steady development in the theory of grinding (Prasher, 1987). Due to the complexity associated with the grinding of particle ensembles, phenomenological modeling tools are used to simulate and understand milling performance. Among various approaches to mathematically model and analyze milling processes (see Bilgili and Capece, 2012), population balance modeling (PBM) is extensively used (Austin, 1971). The general size-discrete PBM given in Eq. (1) uses two functions known as the specific breakage rate function k which describes the rate at which particles break and the breakage distribution function b which describes the resultant mass-based particle size distribution (PSD) of broken particles.

$$\frac{dm_i(t)}{dt} = -k_i(t)m_i(t) + \sum_{j=1}^{i-1} b_{ij}k_j(t)m_j(t) \tag{1}$$

Solution of Eq. (1), otherwise known as the linear time-variant PBM, describes the temporal variation of the particle size distribution for well-mixed batch milling processes. Here, $m_i(t)$ is the mass fraction of particles with size x_i at milling time t. The first term on the r.h.s. is the disappearance rate at which particles of size x_i is broken into smaller particles. The second term on the r.h.s. represents the summed rate at which particles in all size classes $j<i$ are broken into size class i where i and j are size class indices extending from size-class 1 containing the coarsest particles to size class N containing the finest particles usually in a geometric progression. k_i is the size-discrete specific breakage rate constant and b_{ij} is the size-discrete breakage distribution parameter.

In Eq. (1), the specific breakage rate constant is often assumed time independent ($k_i(t)=k_i$) which is valid for processes wherein the breakage rate of particles can be described by first-order breakage kinetics, which states that the rate of breakage of a given particle size (or size class) is proportional to the weight of particles only of that size. First-order breakage kinetics has been observed for many materials and types of mills (Austin and Bhatia, 1972; Fuerstenau et al., 2004; Kotake et al., 2002). In contrast to first-order breakage kinetics, the specific breakage rate constant can vary with milling time due to changes of the PSD and effects associated with mechanical multi-particle interactions among feed particles resulting in non-linear or non-first-order breakage kinetics. Bilgili and Scarlett (2005) described and modeled such effects by decomposing $k(t)$ into an apparent specific breakage rate function and a population dependent functional. In this case, the specific breakage rate of a given particle size is not independent from the presence of other particles of the population as is assumed in the first-order breakage law.

While PBMs such as Eq. (1) are useful for description at the process scale, they are severely limited in the analysis and understanding of the milling environment at the particle-scale. The specific breakage rate constant, which is used as a model/fitting parameter, lacks any fundamental considerations relating to particle breakage such as particle strength or the stresses/strains acting on a particle due to mechanical interactions. Hence, a multi-scale modeling approach that incorporates PBM with consideration of mechanical particle interactions and breakage from particle-scale dynamics is needed for fundamental understanding of milling processes. Such an approach may improve the predictive capability of PBM while elucidating fundamental insight into particle breakage behavior resulting in either first-order or non-first-order breakage kinetics.

In order to study the particle-scale phenomena which produce the complex granular flow and particle fracture behavior in milling processes, the discrete element method (DEM) has recently been utilized (Mishra, 2003; Zhu et al., 2008). DEM is a numerical method which determines the trajectory and interaction of discrete particles using Newton's second law of motion combined with well-established contact mechanics models (Cundall and Strack, 1979). Recent studies have modeled milling processes to predict mill performance by analyzing particle interactions obtained from DEM as discussed below. The simplest approaches correlate an experimentally determined specific breakage rate constant to the total or average impact energy associated with collisions of the grinding media (Hoyer, 1999; Kano and Saito, 1998; Kwan et al., 2005; Mori et al., 2004). More advanced studies predict the specific breakage rate constant by correlating the impact energy obtained from DEM to a broken mass of particles which is experimentally determined from particle bed drop-ball tests (Bwalya et al., 2001; Datta and Rajamani, 2002; Wang et al., 2012). Despite these developments, they still do not include any fundamental considerations for particle breakage behavior or utilize any fracture mechanics models to predict the specific breakage rate constant from the micro-dynamic information. Consequently, these studies are limited in their predictive capacity and in their ability to fully analyze milling processes and the complex breakage behavior.

To address the limitations of the phenomenological PBM and current DEM approaches to predict the breakage rate of particles in milling processes, this study formulates a particle-scale specific breakage rate constant of the PBM based on a particle fracture model of Vogel and Peukert (2003). The formulation of the specific breakage rate constant is a *major novelty* as it separates material properties from the milling environment which is quantified based on the impact energy distribution obtained by DEM. Currently, there is no specific breakage rate constant that is explicitly expressed in terms of impact energy and material parameters that has been properly related to the linear time-variant PBM of Eq. (1). While a companion study (Capece et al., 2014), formulated the basic methodology to derive a particle-scale specific breakage rate constant valid for the initial breakage rate for mono-sized feeds milled for a short time, the same methodology is utilized and extended for polydispersed feeds milled for any length of time in Section 2 of this stand-alone study. The advancement of the specific breakage rate constant in Section 2 allows theoretical investigation into the effects of polydispersity and temporal evolution in the PSD with the objective to test the validity of first-order breakage kinetics. In relation to broader application and practical relevance, this study also contributes to the ongoing development of a unified DEM – PBM framework to model and analyze milling process.

To first confirm the validity of the specific breakage rate constant and the proposed methods, DEM simulations of silica glass processed in a rolling ball mill were performed. In contrast to most studies which only simulate a mono-sized feed (e.g. Capece et al., 2014) or omit the feed entirely (e.g. Datta and Rajamani, 2002), polydispersed feeds along with the grinding media were simulated to obtain the most accurate impact energy distribution. Section 4.1 compares the specific breakage rate constant determined experimentally and from DEM simulations. *As another major novelty*, the effects of various polydispersed feeds and the time-wise evolution of the PSD on the specific breakage rate constant were respectively investigated in subsequent DEM simulations in Sections 4.2 and 4.3. Analysis allowed a theoretical basis and particle-scale understanding of the milling environment that exhibits first-order breakage kinetics and confirmed the invariance of the specific breakage rate constant to changes in the PSD for milling processes with low feed loadings and devoid of dense particle beds.