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On the origin of non-linear breakage kinetics in dry milling[☆]

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ABSTRACT

While population balance models (PBMs) have described the impact of mechanical interparticle interactions on the specific breakage rate in dry milling processes through a phenomenological effectiveness factor, such models lack particle-scale information and thus a mechanistic basis. In this study, a mechanistic effectiveness factor of the non-linear PBM was derived and calculated by coupling a particle-scale breakage model with interparticle interactions obtained from discrete element method (DEM) simulations of a grinding ball impacting an unconfined particle bed. Mono, binary, ternary, and polydispersed particle beds were simulated to determine the effects of granular composition on breakage kinetics. The effectiveness factor obtained from the DEM simulations shows a reduction in the specific breakage rate for coarse particles in binary mixtures. The origin of this phenomenon, commonly known as cushioning or decelerated breakage in dry milling processes, was explained by the DEM simulations: fine particles in a particle bed increase mechanical energy loss, and reduce and distribute interparticle forces thereby inhibiting the breakage of the coarse component. On the other hand, the specific breakage rate of fine particles increased due to contacts associated with coarse particles. Such phenomenon, known as acceleration, was shown to be less significant, but should be considered in future attempts to accurately quantify non-linear breakage kinetics in the modeling of dry milling processes. The phenomenological effectiveness factor was also assessed and found to accurately describe the impact of mechanical interparticle interactions in binary particle beds as well as predict the non-linear breakage behavior in ternary and polydispersed particle beds. Aside from gaining particle-scale insight into non-linear breakage kinetics, the above findings provide the guidelines for the usage of non-linear PBM framework and are expected to improve the design, control, and optimization of dry milling processes that exhibit non-linear breakage kinetics.

1. Introduction

Investigation into micromechanical interactions of granular media is fundamental to understanding the complex behavior observed in particulate processes. Recent advances in computational approaches such as the discrete element method (DEM) have been directed toward this effort due to the inability of experimental methods to provide the necessary insight at the particle-scale [1,2]. DEM considers the translational and rotational motion as well as the mechanical interactions of discrete particles described by Newton's equation of motion and well-established contact mechanics models [3,4]. Among the major applications of DEM to model particulate processes (e.g. mixing, conveying, fluidization), there has been intensive work directed toward understanding the dynamic behavior of milling processes [5].

Milling, which entails the size reduction by breakage of particles, is a pervasive yet poorly understood process. Motivated by the necessity for precise control over product properties such as particle size distribution (PSD) and the need to improve poor energy utilization, DEM applied to milling has predominantly attempted to enhance the design, control, and optimization of these processes [5]. While initial studies focused on description of powder charge motion, more recent studies have focused on quantifying the specific breakage rate (related to breakage probability) or similar parameters which are of more practical use for modeling and prediction in particle breakage rates and the product PSD [6–16]. The specific breakage rate which describes the rate at which particles break is typically related to the stressing rate and energy which can be obtained from DEM.

In the modeling of milling process, it is generally assumed that the specific breakage rate for a given particle is dependent on its size, material properties, and the stressing conditions, but not by the presence of dissimilar sized particles in the milling environment [17]. An abundance of experimental evidence contradicts this assumption finding that the milling environment characterized by the instantaneous PSD may affect particle breakage behavior [18–32]. The occurrence of an environmental

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[☆] Dedicated to the memory of the late Professor Brian Scarlett on the occasion of the 10th anniversary of his passing.

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dependent specific breakage rate has been observed in particle bed compression tests [18–20] and various wet and dry milling devices including ball mills [21–32]. The phenomenon known as “cushioning”, hereafter referred to as “decelerated breakage”, is most commonly observed in which the specific breakage rate of coarse particle is reduced in the presence of fine particles resulting in a slower rate of breakage [21–24] although an increased rate of breakage known as acceleration has also been observed in some milling environments [25–27]. Experimental studies have also shown that fine particles in the presence of coarser particles may also be subjected to accelerated breakage [19,28]. Such breakage behavior can be attributed to mechanical interparticle interactions resulting in force and stress transmission dependent upon the granular composition; however, forces and stresses within powder beds cannot be easily measured. The phenomenon of environmental dependent breakage can understandably complicate the operation of milling processes due to the inability to accurately model and predict such complex breakage behavior.

Aside from purely empirical models and phenomenological models such as population balance models (PBMs) used to explain the breakage behavior discussed above [33–35], a limited number of studies using numerical approaches have sought to investigate mechanical interparticle interactions within sheared or compressed particle beds [36–43]. Tsoungui et al. [36] used the molecular dynamic (MD) method and Liu et al. [37] used a finite element method (FEM) to simulate the compression of 2D particle beds showing that fine particles create a hydrostatic effect around coarse particles thereby preventing their breakage. Wiacek and Molenda [38] investigated the compression of particle beds using 3D DEM and found that polydispersity affects the amount of mechanical energy lost due to friction. A host of DEM studies simulating the shear of particle beds have also found that the granular composition affects energy dissipation, coordination number, and interparticle forces and stresses [40–43]. While the results of the above simulations may be used to *infer* the influence of mechanical interparticle interactions on the breakage behavior in milling process, no DEM study has sought to *quantify* the effect of granular composition on the specific breakage rate and ensuing non-linear breakage kinetics. *As a major novelty*, this study adapts a multi-scale modeling approach and utilizes a particle-scale breakage model and DEM to obtain interparticle interactions at the ensemble scale (microdynamic information) combined with a non-linear PBM to explore the origin and influence of non-linear breakage kinetics in dry milling processes.

To investigate the phenomenon of non-linear or environment-dependent specific breakage rate, this study uses DEM to simulate impact tests of unconfined particle beds which are intended to reproduce the conditions likely to be found in a ball mill. Mono, binary, ternary, and polydispersed particle beds were simulated to elucidate the effects of their composition on the specific breakage rate. First, a non-linear PBM framework for batch milling processes, which accounts for mechanical interparticle interactions through a phenomenological effectiveness factor, is presented. The effectiveness factor is then defined mechanistically via a particle-scale breakage model for use in DEM simulations. Particle-scale dynamics obtained from DEM simulations combined with the mechanistic effectiveness factor show that vastly different mechanical interactions present in various mixtures of coarse–fines lead to marked deviations in the effectiveness factor, signifying a substantial deviation from the traditional first-order breakage hypothesis. The aforementioned experimentally observed deceleration and acceleration effects have been successfully delineated via the effectiveness factor determined from the DEM simulations. The analysis of impact energy rate, different types of collisions, and normal force distributions has enabled us to explain the origin of these prevailing non-linear effects. In addition, the phenomenological effectiveness factor was also assessed and found to accurately describe the influence of particle bed composition on non-linear effects in binary particle beds as well as predict the behavior in ternary and polydispersed particle beds. This insight supports the phenomenological effectiveness factor's use within the non-linear PBM framework for the simulation of dry milling processes. Overall, this study provides mechanistic insight and particle-scale understanding of the effects of the granular composition on the non-linear particle breakage in dry milling processes.

2. Theoretical

2.1. Non-linear population balance framework

Population balance models (PBMs) are extensively used to mathematically model and analyze milling processes [23,44]. The traditional size-discrete time-continuous PBM given in Eq. (1), otherwise known as the linear time-invariant PBM, can describe the temporal variation of the PSD for well-mixed batch milling processes.

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

Here, $m_i(t)$ is the mass fraction of particles with size x_i at milling time t. The first term on the right hand side of Eq. (1) is the disappearance or breakage rate at which particles of size x_i are broken into smaller particles. The second term 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 smallest particles usually in a geometric progression. In Eq. (1), k_i is the specific breakage rate parameter and b_{ij} is the breakage distribution parameter.

The specific breakage rate k_i is usually assumed time-independent and 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. In other words, it is assumed that the specific breakage rate is independent of the population mass density distribution. This assumption is restrictive for practical applications and mainly valid for short-time milling of narrowly distributed feeds [17,45]. Conversely, many milling processes exhibit a time-dependent specific breakage rate which results in non-first-order or non-linear breakage kinetics [18–32]. In order to phenomenologically model and quantify the effects of the evolving PSD on the breakage kinetics, Bilgili and Scarlett [34] decomposed the specific breakage rate of the traditional PBM of Eq. (1) into a first-order specific breakage rate S_i and a population-dependent functional $\digamma$ as shown in Eq. (2).

$$k_i \equiv S_i \digamma\left[W_{iq} m_q(t)\right] \tag{2}$$

When Eq. (2) is substituted into the PBM of Eq. (1), the model is renamed as the size-discrete time-continuous non-linear PBM. The functional, later referred to as the effectiveness factor [46], describes various non-linear breakage kinetics [33,34] and can be expressed via a sum of the weighted interactions of particles in size class i with other particles in generic size class q, i.e. $F_i[\sum_{q=1}^{N} W_{iq} m_q(t)]$. The weighting function W_{iq} expresses the contribution of any size class q to the specific breakage rate of particles in size class i during i–q size class interactions.

The use of the effectiveness factor to account for mechanical interparticle interactions in milling processes is critical to determine the true specific breakage rate and breakage kinetics. If particle interactions are neglected as is assumed in the linear time-invariant PBM, the breakage kinetics may be falsified and the PBM may become unusable in any descriptive or predictive capacity [46]. The definition of the