# Abstract

The processing of particulate materials is crucial in various branches of engineering and science. During these processes, one can observe important micro-mechanisms, such as aggregation of smaller particles to form larger aggregates and breakage (or shattering) of particles into smaller fragments. These micro-mechanisms eventually play a critical role in changing the macroscopic behavior (e.g., dissolution rate, system permeability, and flow characteristic) of the particle population. Thus, modeling and simulation of particulate processes are essential in predicting their macroscopic behavior virtually. This thesis is devoted to the modeling and simulation of different particulate processes consisting of aggregation and breakage mechanisms. The modeling techniques used in this thesis are population balance modeling (PBM), Monte Carlo (MC) modeling, and discrete element modeling (DEM). The MC technique is used in this work as a tool to mimic experimental systems and generate (or extract) alternative experimental outcomes. The main achievements of this thesis are summarized in the following paragraphs.

The use of population balance equations (PBE) in predicting particulate processes is curbed by the unavailability of physically motivated macroscopic mathematical kernels. The present thesis deals with the development of mathematical models of the size- and time-dependent PBE kernels corresponding to processes involving aggregation and breakage. The simultaneous influence of particle size, process time, and probability of occurrence of events are included successfully. The developed models use the population balance modeling approach and have the potential to predict results on a large scale. The particulate processes chosen in this thesis are general aggregation and breakage (linear and nonlinear) mechanisms, spray fluidized bed aggregation and ultrasound-assisted sonofragmentation.

A series of sonofragmentation experiments were performed by varying the ultrasonic amplitude and sonication time to investigate the breakage of rectangular-shaped pyrazinamide crystals. An MC framework has been developed to understand and replicate the breakage behavior of the crystals and to estimate the breakage frequency parameters corresponding to different ultrasonic amplitudes. It was observed that the pyrazinamide crystals break across the width of the particles, and the fractures occur at some random point in a confined region along the length axis only. Depending on the breakage criteria, a comprehensive bivariate PBM, consisting of the mathematical models of bivariate breakage selection function and bivariate breakage distribution

function, is developed, which predicted the experimental observations with great accuracy and efficiency.

Furthermore, the spatially homogeneous spray fluidized bed aggregation (SFBA) process has been extensively modeled using the MC simulation and the one-dimensional PBM approach. A constant number MC framework is used, which simulated several important micro-mechanisms of the SFBA process (binder droplet addition, droplet drying, volume-dependent particle collisions, aggregation, and rebound). The developed PBM incorporated all these micro-mechanisms to formulate the mathematical models of the volume- and time-dependent aggregation kernel, the size distribution of wet and dry particles, and the total number of available droplets, respectively. The accuracy of the developed PBM is verified using the observations of MC simulation.

In order to work with the developed PBMs, we need to have some prerequisite parameters in advance, such as volume dependency in particle selection, average frequency of associated events. If these necessary process parameters are not provided in advance, one needs to model or extract these input parameters. For this, a multiscale bi-directional PBM-DEM coupling framework is successfully developed to predict the aggregation process outcomes. The PBM and DEM take place periodically to update the particle size distribution and extract the collision frequency. Additionally, the developed coupling framework explained the correlation between the size distribution of particle classes and the corresponding frequency of collisions.

Moreover, due to the unavailability of numerical schemes in the literature, we have also developed a weighted finite volume scheme to solve the collision-induced nonlinear breakage PBE numerically. The scheme is designed to conserve the total mass of the system while preserving the total number of particles in the system.

**Keywords:** Population balance model; Aggregation kernel; Breakage kernel; Collisional breakage; Monte Carlo simulation; DEM simulation; Spray fluidized bed; Sonofragmentation; Ultrasound; PBM-DEM coupling.