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Original Research Paper

Dependencies between internal structure and mechanical properties of spray dried granules – Experimental study and DEM simulation

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ABSTRACT

The mechanical properties of spray dried granules are decisive with regard to further applications and can be modified via internal granule structure. To obtain the correlations between structural and mechanical properties, necessary experiments are often time and resource consuming. The simulation of varied granule structures and their effect on resulting mechanical properties seems to be a promising approach.

In this paper, a model of the particulate internal structure of a spray dried granule was generated with the Discrete Element Method (DEM) based on real structure parameters. The model considers real primary particle number, particle size distribution and radial granule inhomogeneity, what results in the implementation of granule shell thickness and macro void. The internal structure of simulated granules showed significant influence on their mechanical properties. An increase of granule shell thickness and packing density of the primary particles within the shell results in fracture strength increase accompanied by decreasing fracture strain. The simulated reduction of the solid bridge bond size between the primary particles representing the decreasing binder amount leads to decreasing fracture strength and strain as previously determined experimentally (Eckhard et al., 2014). Consequently, the DEM is appropriate for evaluating the effect of changed real internal structure parameters on resulting mechanical granule properties.

1. Introduction

The mechanical properties of spray dried granules, like fracture strength and strain, have a significant influence on the processing of the whole granule batch. Depending on the applications the granules have to meet specific requirements: For the generation of homogeneous green compacts during die pressing low fracture strengths of the spray dried granules are desired to guarantee a complete destruction of the granules during compression step. Parallel a sufficient granule strength is needed to survive previous handling, transport and dosage possesses without breakage as destroyed granules would influence the flow behavior of the batch negatively and may cause inhomogeneities within the compact structures [2]. If the spray dried granules shall be used, e.g. as catalyst carrier structures, high fracture strengths are needed to achieve a high stability for surviving handling and regeneration cycles. The granules have to be stable to guarantee a constant flow velocity of gas through the catalyst bulk. For an optimized reaction efficiency a high granule surface and porosity is desired [3].

The relation between internal structure and mechanical properties of spray dried granules is still a field of research. The mechanical granule properties can be tailored by the specific selection of type and amount of additives to the suspensions previous to spray drying or by the modification of the internal granule structure itself [1,4–6]. Rumpf [7] and Kendall [8,9] described the agglomerate strength as function of porosity, size and properties of the primary particles as well as of the applied additive system. Binder additives increase granule fracture strength [10–12]. At the same time the internal structure is changed but the structural impact on the mechanical properties is overlaid by additive influence [13]. Especially if the required additives are expensive or the suitable additive selection is limited because of further processing steps or applications, a desired change of the mechanical granule properties can only be achieved via internal granule structure modification. For particular agglomerates Subero et al. and others

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Nomenclature

A	area (image analysis) [m^2]
A_{PB}	solid bridge bond area (DEM) [m^2]
d	diameter or respectively size [m]
F_{break}	granule fracture force [N]
F_C	contact force (DEM) [N]
F_D	damping force (DEM) [N]
F_{grav}	gravitational force (DEM) [N]
F_{PB}	solid bridge bond force (DEM) [N]
F_{tot}	total contact force (DEM) [N]
H	granule classification parameter (image analysis) [–]
J	moment of inertia (DEM) [kg/m^2]
k	contact stiffness (DEM) [N/m]
k_{PB}	solid bridge bond stiffness (DEM) [N/m^3]
m	mass [kg]
M_C	contact moment (DEM) [N m]
M_{homo}	amount of homogeneous granules per batch (image analysis) [%]
M_{hollow}	amount of hollow granules per batch (image analysis) [%]
M_{PB}	resulting moment from applied solid bridge bond (DEM) [N m]
M_{tot}	total applied moment at a contact (DEM) [N m]
S_{gran}	shell thickness of a single granule (image analysis) [%]
T	temperature [°C]
t_{crit}	critical time step (DEM) [s]
U	overlap between two contacting entities (DEM) [m]
ε_{break}	granule fracture strain [%]
ε_{micro}	micro porosity (image analysis) [%]
ε_{macro}	macro porosity (image analysis) [%]
λ_{PB}	solid bridge bond size radius multiplier (DEM) [–]
η	damping coefficient (DEM) [kg/s]
μ	friction coefficient (DEM) [–]
σ_{PB}	solid bridge bond strength (DEM) [N/m^2]
σ_{break}	granule fracture strength [MPa]
v	contact velocity (DEM) [m/s]

showed an increasing fracture strength with decreasing porosity [10,14–16]. Also the reduction of the primary particle size leads to increasing fracture strength because of increased contact numbers and packing density. To have a more detailed look on the effect of different internal structure parameters of spray dried granules on fracture strength and strain, a new method for structure preparation and quantification was developed [17,18]. In our previous study granules with increased shell thickness (macro structural change) as well as reduced micro porosity (micro structural change) showed an increased fracture strength [1]. Moreover the fracture strain can be decreased reducing the micro porosity and vice versa. Both effects, micro and macro structural change, can overlay each other and result in higher pronunciation or neutralization of the influencing structure changes. For the investigated granules, a dominating influence of the micro structure on the resulting mechanical properties was identified [1,19].

For the verification and expansion of the experimentally derived correlations regarding the specific change of mechanical properties via internal granule structure parameters more experimental investigation and systematic structure modifications are necessary. As the experimental possibilities are limited and very time consuming, the development of a simulation tool is a promising technique to review and expand the experimentally derived correlations.

For the actual study a simulation model based on DEM is chosen. The suitability of the discrete element method for the realization of tasks concerning the mechanical properties of particular systems during compression or impact is already described by several authors [20–28]. The compression of agglomerate structures using DEM was performed e.g. by Thornton et al., Antonyuk et al. and others [29–33]. State of the art is the simulation of porous systems as homogeneous particular structures over the whole system (granule) volume as done e.g. by Müller [34,35], Antonyuk [21,22], Khanal [36] and others [32,37–39]. The particles forming the agglomerate are hold together by adhesion forces or implemented bonds [22,32,33]. These studies did not focus on real internal spray dried granule structures. For the aspired DEM investigation of the effect of changed internal granule structures with regard to micro and macro structure variations, the established structure simulation has to be improved. Radial structure differences have to be implemented into the DEM granule structure model to be able to simulate the effect of changed structure composition and therewith porosity distribution within the granule on resulting mechanical properties.

Aim of this study is the reproduction of real internal structures of porous spray dried granules with defined internal macro void and shell thickness and the appropriate simulation of the deformation behavior using DEM simulation. For this actual study of the uniaxial compression test of single spray dried granules the PFC3D simulation code by Itasca is used.

2. Contact model for the simulation with the discrete element method (DEM)

Basis of the DEM simulation is a discrete numerical model firstly introduced by Cundall and Strack [40] describing the mechanical behavior of solid disks or spheres. Therewith the behavior of a complex system consisting of a defined amount of single objects can be modeled. Single objects represent rigid and non-deformable primary particles moving independently from each other only interacting at contact points. Particle elements can be modeled as spheres or as random combination of spherical components. Besides particle elements wall elements, representing machine equipment or surrounding boundaries, can be modeled. Wall elements interact with the particles but not with each other. Forces developing from contacts between individual particles and walls only influence the behavior of each particle object. To model a complex system behavior, as it is the case for the particular granule structures of this study, single particles within the simulation can be bonded together at contact points representing solid bridges. Besides shear and compression forces, solid bridge bond elements are able to transmit tensile forces, which are calculated according to linear-elastic beam theory (Euler–Bernoulli beam), as introduced into DEM by Potyondy and Cundall [41]. For the actual studied effect of changed internal granule structure on resulting mechanical granule properties, the DEM software PFC3D (Itasca Consulting Group) was used.

By applying a suitable contact model with specific particle and solid bridge bond properties a realistic system behavior can be modeled. As the system behavior is an effect of forces between contacting entities and resulting movement acting on the objects, one calculation cycle contains the repeated quasi simultaneous application of 'Newton's Second Law' of motion for every particle as well as the application of the 'Force Displacement Law' at every