International Journal of Pharmaceutics 503 (2016) 68–77

Contents lists available at ScienceDirect

International Journal of Pharmaceutics

journal homepage: www.elsevier.com/locate/ijpharm

Evolution of the microstructure during the process of consolidation and bonding in soft granular solids

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ARTICLE INFO

Article history:
Received 19 July 2015
Received in revised form 13 February 2016
Accepted 18 February 2016
Available online 21 February 2016

Keywords:
Powder compaction
Discrete particle modeling
Microstructure anisotropy
Tablets
Tensile strength
Force distribution

ABSTRACT

The evolution of microstructure during powder compaction process was investigated using a discrete particle modeling, which accounts for particle size distribution and material properties, such as plasticity, elasticity, and inter-particle bonding. The material properties were calibrated based on powder compaction experiments and validated based on tensile strength test experiments for lactose monohydrate and microcrystalline cellulose, which are commonly used excipient in pharmaceutical industry. The probability distribution function and the orientation of contact forces were used to study the evolution of the microstructure during the application of compaction pressure, unloading, and ejection of the compact from the die. The probability distribution function reveals that the compression contact forces increase as the compaction force increases (or the relative density increases), while the maximum value of the tensile contact forces remains the same. During unloading of the compaction pressure, the distribution approaches a normal distribution with a mean value of zero. As the contact forces evolve, the anisotropy of the powder bed also changes. Particularly, during loading, the compression contact forces are aligned along the direction of the compaction pressure, whereas the tensile contact forces are oriented perpendicular to direction of the compaction pressure. After ejection, the contact forces become isotropic.

1. Introduction

Powder compaction is widely used in manufacturing, such as pharmaceutical industries, to produce tablets of high relative density and strength. To get high quality design and product, understanding the mechanics of the compaction process is essential. During compaction, the particles deform significantly and usually develop bonding at contact surfaces. The amount of deformation of each particle and the bond strength created between contacting particles depends on the properties of the particles and the applied compaction pressure.

A lot has been learned from prior experimental studies on tablet compaction, including the effects of particle size on powder compaction (Adolfsson et al., 1997; Almaya and Aburub, 2008; Eichie and Kudehinbu, 2009; Fichtner et al., 2005; Herting and Kleinebudde, 2007; Khomane and Bansal, 2013; Mckenna and Mccafferty, 1982; Patel et al., 2007; Sun and Himmelspach, 2006; Sun, 2008; Yohannes et al., 2015), the role of lubricants on compaction and tensile strength of tablets (Almaya and Aburub, 2008; Sun and Himmelspach, 2006), relationship between porosity and compaction pressure (Heckel, 1961) to mention a few. However, experiments alone cannot provide full description of the characteristics in the bulk of the compacted powders. To understand and investigate the compaction process in detail, particularly the interactions between individual particles and the heterogeneity in the bulk, computational modeling of the compaction process is required. Various methods have been used to model powder compaction in the past. These methods are based on Finite Element Method (FEM) (Klinzing et al., 2010), Discrete Element Method (DEM) (Cundall and Strack, 1979; Koynov et al., 2011; Martin and Bouvard, 2004; Olsson and Larsson, 2013; Sheng et al., 2004; Skrinjar and Larsson, 2004), or combined Discrete-Finite

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http://dx.doi.org/10.1016/j.ijpharm.2016.02.032
0378-5173/

Element Method (DFEM) (Zavaliangos, 2002; Zhang, 2009; Zhang and Zavaliangos, 2011; Zheng and Cuitiño, 2002). In FEM, usually, only the global values of the physical properties are assumed, while in DEM the interaction between individual particles are accounted for, which enables modeling the heterogeneity of powders at the microscopic/particle scale. Hence, DEM gives more accurate results than continuum methods, which consider only the global physical properties of the material. In DEM, the inter-particle forces are computed based on the physical properties of the particles. Originally, DEM was developed for application pertaining to very small elastic deformations (Cundall and Strack, 1979). In recent years, DEM has been used to model plastic deformation (Martin and Bouvard, 2004; Olsson and Larsson, 2013; Sheng et al., 2004) and bonding between particles (Potyondy and Cundall, 2004).

In DEM, an explicit integration method is usually used to track the displacement and deformation of individual particles at every time step. Since DEM accounts for the interaction of individual particles directly, it requires a significantly large computational resources, especially for large number of particles. To reduce the computational time requirement, Sheng et al. (2004) increased the time step in the numerical integration by increasing the mass of individual particles by a factor of 10,000. Using this approach, the same authors showed that for a given compaction pressure, the strain decreases as the coefficient of friction between the particles is increased. In addition, they showed that lower coefficient of friction of particle results in higher coordination number (number of contact between particles). Similarly, Olsson and Larsson (2013), using DEM simulations of binary mixtures, showed that the particle size distribution has a very small influence on the compaction pressure and elastic unloading for narrow particle size distribution (Martin and Bouvard, 2004).

At larger deformations, the shape of particles deviates significantly from a spherical shape and the inter-particle contact forces depend on each other, in addition to the material property, effectively exhibiting a non-local effect (Gonzalez and Cuitiño, 2012). Further, Mesarovic and Johnson (2000) used FEM models to study details of deformation mechanisms of particles. To account for these complicated particle deformation and contact forces, Zavaliangos (2002) has used DFEM, which can be considered as an advanced DEM, because in addition to computing the contact force between two particles, the deformation of the particles is further modeled using a finite element method. Specially, for compaction of powder bed to a very large relative densities, the particles deform significantly and only considering the assumptions of the DEM will not be sufficient. However, DFEM requires huge computational resource compared to DEM and is mostly applied to 2D simulations with limited number of particles, though it can be applied to 3D simulations as well (Güner et al., 2015).

In this paper, a 3D numerical compaction of granular materials was used (Gonzalez and Cuitiño, 2012, 2016; Martin and Bouvard, 2004; Mesarovic and Johnson, 2000; Storåkers et al., 1997, 1999). This method has a reduced computational time compared to DEM, while accounting for the interaction of particles directly as in the case of DEM. Based on this method and a compaction experiment, the material properties of two commonly used pharmaceutical powders were calibrated. The simulations were validated by compacting the powders at various compaction pressures and measuring the diametrical tensile strength. Then, the evolution of the microstructure during compaction, unloading, and ejection of the compacts were analyzed. Specifically, the evolution of the probability distribution of the contact forces, the anisotropy, and the inter-particle bond strength during the compaction process were investigated.

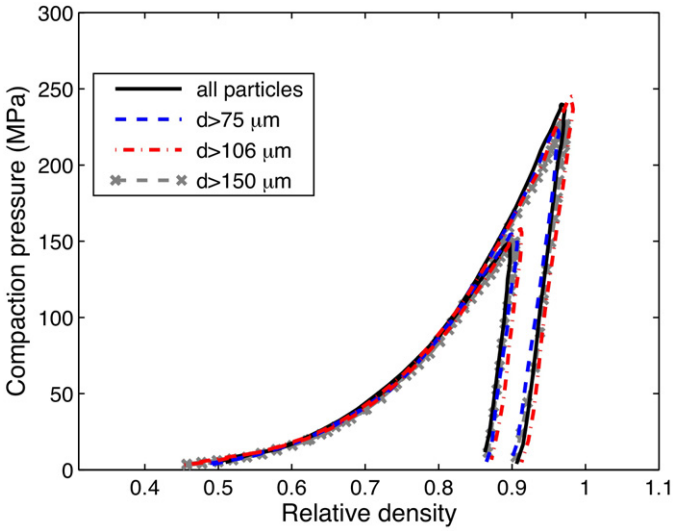

Fig. 1. Compaction pressure vs. relative density for lactose samples with different particle size distribution.

2. Numerical simulation

The first step in all kinds of discrete particle modeling schemes is forming a packing of particles with the target particle size distribution. For this study, the particle size distribution of two pharmaceutical powders, lactose monohydrate (Foremost Farms, Baraboo, WI) and microcrystalline cellulose (MCC-Avicel PH102, FMC Biopolymer, Newark, DE) were used. Since discrete particle modeling methods require the representation of individual particles in the material, the computational efficiency is constrained by the number of particles in the system. State-of-the-art discrete particle models are capable of modeling about a million particles. However, even a very small volume of powder bed consists of a vast number of particles. For example, a 500 mg of MCC-Avicel PH-102 (MCC) powder consists of about 9 billion particles. In terms of number of particles, the majority of these particles (about 91%, calculated based on measured particle size distribution (Yohannes et al., 2015) and assuming all particles are spherical) are fine particles of less than 1 μm size. In terms of volume, these fine particles account for about only 3% of the total volume. On the other hand, if the particle sizes less than 100 μm are removed, a 500 mg of MCC powder will have only about 500,000 particles. Our previous experiments (Yohannes et al., 2015) have indicated that removing the fine particles does not affect the compaction profile and tensile strength of the compacts. Fig. 1 shows the compaction profiles, during unloading and loading of the compaction pressure, for four samples of lactose powder with differing amount of fine particles. The four samples were compacted to in-die maximum relative densities of 0.90 and 0.97, which correspond to maximum compaction pressures of 150 MPa and 230 MPa, respectively. The four samples have very similar compaction profiles indicating that the fine particles have little or no effect on the compaction of the samples. Based on these results, it is possible to use particle sizes greater than 100 μm to model the powder compaction, which will significantly reduce the number of particles used in the simulations.(These compaction profiles are later used to calibrate the mechanical properties of the powders). Fig. 2 shows the particle size distribution used in the simulations and experiments in terms of number of particles (f_o(d)). The procedures for the preparation of the sieved powder samples are discussed in detail in Yohannes et al. (2015). The sizes of particles range between 100 and 240 μm for lactose and between 100 and 750 μm for MCC.