Powder Technology 203 (2010) 211–222

Contents lists available at ScienceDirect

Powder Technology

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

Investigating the effect of shape on particle segregation using a Monte Carlo simulation

Stephen J. Roskilly [a], Elizabeth A. Colbourn [a], Oludolapo Alli [b], Daryl Williams [b], Kirsty A. Paul [c], Eloise H. Welfare [c], Paul A. Trusty [c,*]

[a] Intelligensys Ltd., Springboard Business Centre, Stokesley, North Yorkshire TS9 5JZ, UK
[b] Department of Chemical Engineering, Imperial College London, South Kensington Campus, London, SW7 2AZ, UK
[c] GlaxoSmithKline, Global Manufacturing & Supply, Ware, Hertfordshire, SG12 0DJ, UK

ARTICLE INFO

Article history:
Received 4 November 2009
Received in revised form 29 April 2010
Accepted 9 May 2010
Available online 17 May 2010

Keywords:
Monte Carlo
Segregation
Shape
Packing
Effective size

ABSTRACT

A Monte Carlo simulation has been used to investigate the segregation potential of a range of particulate systems under conditions in which the particles undergo high amplitude low frequency shaking. These systems involve a wide range of binary powder mixtures in which complex particle shapes have been investigated, including plates and rods which represent the real world materials encountered in pharmaceutical systems such as those which include crystalline components. Previous simulations on the segregation propensity of systems with different shapes were limited to spheres and spherocylinders, with relatively low vibrational amplitude drops. A commercial computer application for particle packing—called MacroPac—has been successfully employed here, as it has been able to model systems that are more complex where the shape variation is much wider. These simulations apply to the case of macroscopic particles, in the absence of air resistance and inter-particle forces. For non-spherical shapes, an 'effective size' which relates to the radius of gyration of the particles is determined. Our studies indicate that with high amplitude low frequency shaking, in a mixture of particles with different shapes but with equal volumes, the particles with the larger 'effective size', which tend to have a lower packing fraction, segregate to the top.

1. Introduction

Particle segregation is important in many industries which deal with granular material, including the important case of pharmaceutical formulations where segregation can cause poor content uniformity and result in subsequent batch rejection [1].

Computational modelling has become a mainstream research technique for many disciplines including materials science, yet its widespread use in the pharmaceutical industry is still relatively limited [2]. Computational fluid dynamics (CFD) tool sets are now well developed and many commercial packages are available. These have been deployed on a number of important problems including spray drying [3,4] and in the design of drug delivery devices such as dry powder inhalers [5,6].

The discrete element methods (DEM), first developed by Cundall and Strack [7], are probably the most widespread in their usage for pharmaceutical problems and have been applied to blending in rotating devices [8–10] and powder flow in hoppers [11–13] as well as to problems such as powder compaction [14]. The reader is referred to the extensive review of Zhu et al. [15] for a more detailed discussion of discrete particle simulations of particulate systems using especially the DEM approach.

The Monte Carlo method [16] is another modelling technique that has been applied to complex materials including particulates. This approach uses random numbers and probability to solve problems, so it is especially appropriate for disordered systems which inherently have a high degree of randomness. Because of their random nature, Monte Carlo simulations have been used to produce random starting points for subsequent DEM simulations, as has been done for example by Gethin et al. [17] for tablet compression.

Simulations using a Monte Carlo approach are simple to implement and computationally efficient. They have been used to model the packing of spheres [18,19], and are readily applicable to non-spherical entities [20]. Several aspects of known granular behaviour, including vibration-induced segregation, have been observed using these simulations [18,21,22]. The majority of systems modelled to date have used spherical particles and have systematically varied particle size. In such systems, size segregation is thought to occur as a result of voids opening up beneath larger particles during the shaking process. The smaller particles have a higher probability of filling these voids than have the larger particles. Over the course of time the larger particles move upwards as smaller particles fill the voids beneath, until distinct layers of different-sized particles exist.

* Corresponding author.
E-mail address: paul.a.trusty@gsk.com (P.A. Trusty).

0032-5910/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.powtec.2010.05.011

The effect of particle shape on segregation is less well documented and understood. Gan et al. [23] have reported on the packing of particles of arbitrary shape, but have not investigated their segregation behaviour. Segregation has been addressed by Abreu et al., who investigated the influence of particle shape on the packing of spherocylinders and on the segregation of spherocylinders mixed with spheres [20], for the case when the vibration amplitude is no more than twice the diameter of the spherical particles. Their systems were found to segregate such that the shape which, when pure, produced denser beds moved downward and concentrated at the bottom of the container. However, when they looked at systems in which the two types of objects had different volumes, they found that the size of the particles had an overriding effect. For example, spheres segregated above the lower packing fraction long spherocylinders.

Although a Monte Carlo simulation is used in this study, the work reported here differs from that of Abreu et al. [20] in two key aspects. First, a much larger amplitude was used for the vibration, so that our simulations correspond to high amplitude low frequency shaking rather than to small amplitude vibration. Secondly, the present work investigates a wider range of particle shapes, including some that are important in simulating pharmaceutical powder blends. Such crystalline particles, with their sharp corners and planar faces, will be poorly approximated by spheres and spherocylinders. Shapes like these can be described in terms of their aspect ratio, or alternatively using the radius of gyration, a term often used in colloid chemistry [24] to describe the spatial extension of a particle.

The study reported in this paper focuses on a series of particulate mixtures in which spherical particles are mixed with other objects whose dimensions are chosen so that the object has the same volume as the spheres. The shape and aspect ratio (and therefore the radius of gyration) of the other objects are varied systematically in order to assess the relationship between particle shape and the rate of segregation. The study is then extended to look at mixtures of non-spherical shaped particles, as well as systems in which the volumes of the particle shapes are not identical.

2. Methods

2.1. Computational methodology

A commercial software package, *MacroPac* (*version 6.2*), developed by Intelligensys Ltd, was used to perform the Monte Carlo simulations reported here. A basic assumption of the model is that there is a 'hard' interaction between particles. In this way, the 'pure' effects of particle shape can be investigated, with no additional complicating factors caused by inter-particle interactions or by features such as air resistance.

The particle shapes investigated consisted of spheres, cuboids and 'jacks' (a shape with six equivalent 'arms', an example of which is shown in Fig. 1(B)). The aim was to investigate the effect of the shape on the degree of segregation; for this reason, the objects were defined so that they had equal volumes so as to remove the overt influence of particle size. With the exception of two larger volume spheres used in some of the studies, all the objects had a nominal volume of 5 cubic units.

For each of the simulations reported here, two different particle shapes were mixed, using equal volumes of both types of particles. For the case where the particles had the same volume this meant that there were equal numbers of each type of particle. Simulations were undertaken using a Dell laptop computer with a Core2 Duo T5800 processor. Computer simulation times to pack and process the systems described in this paper varied from 60 min to 18 h.

2.2. Building particle shapes

Particle shapes were constructed using spheres, following the ideas introduced by Ferrar and Evans [25]. Spheres of different sizes can be used in constructing the same object, and spheres within the same object can be allowed to overlap in order to create a more accurate representation of the shape. For example, a cube may be represented by an array of 27 ($3\times3\times3$) spheres. Clearly with this construction there will be rounding of the cube at the corners, which, in an assessment of the role of shape in segregation, may have an effect on the results. To increase the accuracy of the shape it is possible to build the shapes with capped edges, where smaller spheres are placed along the edges and at the corners of the object so that it more accurately represents the required shape. This was undertaken for most of the cuboid shapes used, and is illustrated in Fig. 1(A) for the specific example of a cube. However, in the case of long rectangular rods (defined below in Table 1), the number of spheres required for capping led to extremely slow simulations; consequently the shape was not modified especially, since the representation without capped edges appeared to be adequate for this high aspect ratio shape. Capping was not applied in the case of jacks, where each of the arms, had a cross section of 4 spheres. Fig. 1(B) shows the component spheres of one of the jack shapes investigated.

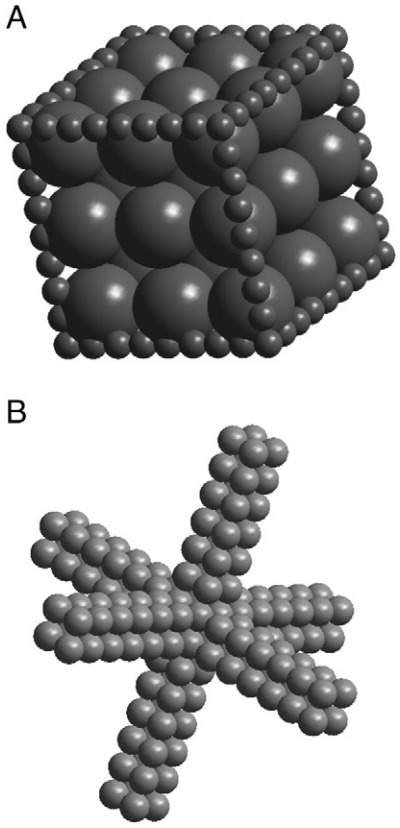

Fig. 1. (A) Cube, showing component spheres including capping spheres, (B) Jack shape showing component spheres.

The tables below outline the parameters that define each shape. The data for the cuboid shapes are summarised in Table 1. Table 2 gives the parameters which define the three 'jack' shapes, which have increasing relative arm length from 1 to 3. To maintain the same volume for each shape, the cross section of the 'jack' arm decreases as

Table 1
Parameters used for building cuboid objects.

Description	Length	Width	Height
Cube	1.71	1.71	1.71
Plate-like	2.5	2	1
Cubic rod	5	1	1
Long rectangular rod	10	0.5	1