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Chemical Engineering Science 60 (2005) 2517–2531

Chemical Engineering Science

www.elsevier.com/locate/ces

DEM-based models for the mixing of granular materials

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Received in revised form 15 October 2004; accepted 1 November 2004

Abstract

This work discusses the current state of the art in the modeling of granular flows in mixing processes. A quick review of both continuum- and discrete-based models is first presented, and the focus is then put on the discrete element method (DEM), which has recently proven worthy of interest for the mixing of granular materials. Theoretical and practical aspects of this method are discussed, as well as new ideas to enable the simulation of more and more complex mixing systems. Numerous examples are presented throughout the paper to illustrate the applicability and the limitations of a DEM-based model.

Keywords: Granular materials; Powders; Mixing; Mathematical modeling; Simulation; Discrete element method

1. Introduction

The development of mathematical models to simulate and investigate the flow of granular materials is a topic of great relevance to process engineering, spanning a range of industries that includes pharmaceutical, chemical, agricultural, advanced materials and food. As an illustration, it is well known that the slightest changes in the ingredient properties or the process operating conditions for the manufacturing of a drug can have a huge impact on its quality. It is then of paramount importance to understand the physical phenomena that govern the flow and the mixing of the solid ingredients involved in the manufacturing of pharmaceutical products, to guarantee their quality and minimize the risks for health. Of particular relevance is the ability to mix tiny proportions of small particles in a matrix of much larger particles, a situation that is likely to prevail more and more with the formulation of new drugs based on small dosages of active ingredients (Muzzio et al., 2002).

Despite its importance, not much is known about the mixing of granular materials in comparison with the level of knowledge in the field of fluid mixing. It is admitted that we currently lack the fundamental knowledge and engineering expertise required to design reliable and scalable processes for the manufacturing of products involving granular ingredients. Two main reasons that can be put forward to explain this situation are the complex nature of granular materials and the multifaceted flow behavior of granular materials. The consequence of this state of affairs is that we only have a partial understanding of the mechanisms governing the processes involved in the development and manufacturing of granular products, which means that the design and scale-up of these processes is essentially achieved by empirical methods. As a result, it may be hazardous to change operating parameters or ingredient properties since it is known that a small change may result in particles behaving in a completely different way and, consequently, in a final product with unpredictable physiochemical properties. This is why, for instance, the pharmaceutical companies are reluctant to modify their manufacturing processes and why they rely upon monitoring methods to control as efficiently as possible the quality of the products they manufacture.

Granular flow modeling is experiencing rapid development thanks to worldwide research activities in physics, mathematics, computer science and engineering. It is a

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0009-2509/$ - see front matter © 2005 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ces.2004.11.048

highly multi-disciplinary field since it brings into play topics such as granular mechanics, solid–gas flows, solid–liquid flows, solid–solid flows, mixing and segregation, kinetic theory, rheology and, last but not least, parallel computing.

The objective of this work is to describe the current state of the art in the modeling of granular flows in mixing processes. After a brief review of methods that are currently available, emphasize will be put on the discrete element method (DEM), a deterministic method that has recently aroused interest for the mixing of granular materials. In particular, both theoretical and practical aspects of the method will be discussed, which includes new ideas to enable the simulation of more and more complex systems involving particles of different sizes and shapes, and mixers of all kinds. Numerous examples will be presented to illustrate the applicability of DEM but also to emphasize its current limitations and the challenges to be taken up in the future.

2. Modeling strategies for granular flow

There exist many modeling strategies that can be used to simulate granular flow. In the systems approach, one focuses on the operation of a complete mixing system, as opposed to the behavior of individual particles. One such method has aroused interest for the past 30 years and was devised by Inoue and Yamaguchi (1970) to study powder flow in a V-blender. In their work, the blender was described as a network of interconnecting cells and a Markov chain was used to predict the changes to the states (or particle concentrations) of the system. Markov chain models have since been used by many researchers to investigate powder flow in various chemical engineering processes such as, for instance, static mixers (e.g. Chen et al., 1972; Lai and Fan, 1975). This model has also been used to derive RTD curves in continuous processes (e.g. Fan et al., 1985). The systems approach has proven useful to describe the salient features of powder flows and provide trends that can help understand mixing systems or design new ones.

This paper focuses on approaches that are based on physical grounds and that can be applied for the simulation of powder flow in mixing systems. These include models that rely on continuum mechanics and that disregard the discrete nature of the flow, and the so-called particulate models that are based on individual force balances. We now consider these two types of model.

2.1. Models based on continuum mechanics

This category of models considers that the particles flow as a fluid and, as a result, is based on the solution of the underlying conservation equations using CFD techniques. Such an approach of course overlooks the local behavior of individual particles. Consequently, systems consisting of a small number of particles cannot be modeled with this approach. However, in systems comprising particles that are evenly distributed with the same bulk properties so that they can be thought of as forming a continuum, such an approach can yield results that are in qualitative agreement with experimental data. For instance, such an approach was used by Bertrand et al. (2001)[1] for the modeling of the transport of powders in a continuous polymerization reactor provided with a blade impeller. Following the so-called "Network-of-Zones" model of Mann and Hackett (1988), it consists of dividing the computational domain into cells and computing the displacement of the granular material by means of an unsteady mass balance that accounts for the gravity and the motion of the impeller. This can be viewed, up to a certain extent, as a finite volume method for the solution of the following mass conservation equation:

$$\frac{\partial C}{\partial t} + \mathrm{grad}(C\mathbf{v}) = S, \tag{1}$$

where C stands for the solid volume fraction, $\mathbf{v}$ for the solid bulk velocity and S for a source term that may be zero. In this model, $\mathbf{v}$ takes into account the gravitational force through the settling velocity given by

$$v_{\mathrm{settling}} = D^2 g(\rho_s - \rho_{\mathrm{air}})/18\mu_{\mathrm{air}}, \tag{2}$$

where D represents the particle diameter, g the gravitational acceleration, ρ_{air} and ρ_s the air and particle densities respectively, and μ_{air} the dynamic viscosity of air. The shape and the rotational speed of the impeller are accounted for by a series of control points in a manner that is reminiscent of the fictitious domain method (Bertrand et al., 1997). More precisely, the velocity of the impeller is added to $\mathbf{v}$ in the cells that contain at least one control point. To mimic powder cohesion, this velocity can also be imposed in the cells that are within a given distance from the impeller; the larger this "impeller working zone", the more cohesive the powder. Finally, values of $\mathbf{v}$ at the inlet (outlet) of the tank are determined from the inlet (outlet) flowrate and the size of the cells into which (from which) particles enter (exit).

As said before, this model was used to predict the motion of powders in continuous reactors. In particular, one simulation consisted in predicting the packing of polypropylene powder in a reactor provided with a blade impeller and initially filled at 35 wt%. Rotational speed of the impeller was 80 rpm. Length and diameter of the reactor were 9.5 and 4 in, respectively. Length, width and thickness of the blade were 4, 0.5 and 0.125 in, respectively. The blade was incurved and fixed with a 20° angle to a central shaft of 0.5− in diameter. Inlet and outlet flowrates were set to zero.

The mesh comprised 40 436 finite volumes and 7887 nodes (Fig. 1a). The impeller was represented by means of 3952 control points. The radius of the impeller working zone was set to 15 mm following experimental observations.

Fig. 1b shows the packing of the polypropylene powder in the tank at a given time, as predicted by the model. One

[1] Experimental part of the work was done by the group of C. Briens from University of Western Ontario.

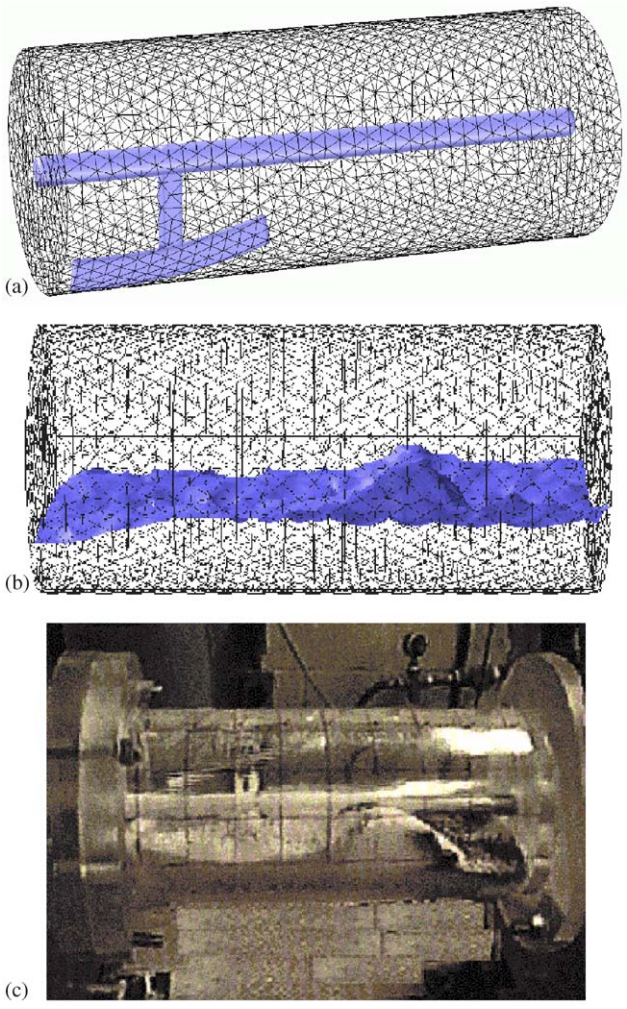

Fig. 1. Granular flow in the blade mixer (Bertrand et al., 2001): (a) finite volume mesh; (b) numerical packing of the polypropylene powder; and (c) packing of the polypropylene powder observed experimentally.

can readily see that this result is in good agreement, at least qualitatively, with the packing observed experimentally (Fig. 1c). In particular, the formation of a heap of powder downstream from the blade impeller was well predicted by the model.

Of course, this model based on continuum mechanics overlooks the behavior of the individual particles. These results however show that it can be a viable approach to predict bulk properties.

2.2. Particulate models

This second type of models does not rely upon continuum mechanics. It rather simulates the motion of each particle individually, with a special treatment for eventual collisions. Many such methods have been developed over the years and can be divided into two categories. The first category comprises the probabilistic methods and is based upon a random number generator to displace the particles in the simulation domain. For instance, Rosato et al. (1986) proposed a Monte-Carlo method to investigate the well-known "Brazil nuts" segregation phenomenon. Their method consists of displacing the particles of a system in a stepwise fashion and of resorting to a random walk procedure to either accept or reject a given displacement according to an energy principle. The main advantage of this method is that it is fast and can be used to simulate the sedimentation of non-spherical particles (Vidal et al., 2001). Its main limitation is linked to the fact that it does not allow for particle collisions, which no doubt play an important role in mixing processes. In practice, it has been used mainly to simulate the settling of particles of all shapes and sizes. In particular, it has proven efficient to provide approximations for the porosity of polydispersed packings with a tendency to overestimate its value, a phenomenon that is due to the fact that inter-particle collisions are not accounted for, as explained by Bertrand et al. (2004). The second type of models comprises deterministic methods such as the Stokesian Dynamics method (or simply Stokesian Dynamics (SD)) of Brady and Bossis (1985) and the DEM first introduced by Cundall and Strack (1979). SD is nothing but a wide-ranging method for computing the trajectories of interacting particles in a suspending fluid. It is based on the N-body Langevin equation of motion

$$M\frac{\mathrm{d}\mathbf{v}}{\mathrm{d}t} = \mathbf{F}_{\mathrm{total}}. \tag{3}$$

In this equation, M is a matrix that contains the mass of the particles, $\mathbf{v}$ is a vector that represents the velocity of the interacting particles and $\mathbf{F}_{\mathrm{total}}$ is a net force that can contain hydrodynamic terms as well as non-hydrodynamic terms to account for colloidal forces and Brownian motion. The main drawback of this method lies in the fact that it requires the solution of a full matrix system, which restricts to the order of 1000 the size of the suspensions that can be studied. To generate sparser matrices and thus improve on the computational efficiency, approximations such as that proposed by Ball and Melrose (1997) only consider short-range forces and neglect the far-field interactions. SD has been developed to better understand the link between the rheology of a suspension and its microstructure. For instance, Catherall et al. (2000) used SD to study the shear-thickening behavior of high solids content suspensions. Other applications of this method include the sedimentation of non-spherical particles (Hase and Bousfield, 1994), the migration of particles in non-Newtonian fluids (Phillips, 1996) and the pressure-driven flow in a channel (Nott and Brady, 1994). To our knowledge, SD has never been used in solid mixing. The three-dimensional (3D) simulation of granular flow in a mixing system generally implies the tracking of a large quantity of particles. Simulations of even the smallest systems (10^4 particles) could hardly be achieved with SD within reasonable time. Fortunately, another deterministic particulate method, the DEM, appears to be more suitable for this kind of simulations. This model has indeed been used

recently to simulate with qualitatively good accuracy the flow of granular materials in a wide variety of mixing systems (e.g. Moakher et al., 2000; Stewart et al., 2001). The subsequent sections of this paper will focus on this method and its applicability to solid mixing.

3. Presentation of the DEM

The DEM bears a resemblance to molecular dynamics methods since the position of each particle in a system is obtained by integrating twice with respect to time Newton's second law of motion. DEM is in fact a time-driven soft-particle method (e.g. Mehta, 1994; Duran, 1999) that allows two particles to interpenetrate so as to mimic particle deformation. In other words, collisions are not instantaneous with DEM and the level of particle interpenetration is linked to the nature of these collisions. As a result, DEM is well suited for contact-dominated flows such as those prevailing during the mixing of powders.

After computing the total force acting on each particle, Newton's equation of motion can be integrated numerically to yield the velocity and the position of all particles at the current time:

$$m_i \frac{\mathrm{d}^2\mathbf{x}_i}{\mathrm{d}t^2} = \mathbf{F}_{\mathrm{total},i}, \tag{4}$$

where m_i represents the mass of particle i, $\mathbf{x}_i = (x_i, y_i, z_i)$ the coordinates of its center of gravity, and $\mathbf{F}_{\mathrm{total},i}$ the total force acting on this particle. The rotation of each particle can be obtained in a similar fashion through an angular momentum balance. In the context of mixing, the total force acting on particle i is given by

$$\begin{aligned}\mathbf{F}_{\mathrm{total},i} &= \mathbf{F}_{\mathrm{hydrodynamic},i} + \mathbf{F}_{\mathrm{non\text{-}hydrodynamic},i} + \mathbf{F}_{\mathrm{contact},i} \\ &= \mathbf{F}_{\mathrm{hydrodynamic},i} + \mathbf{F}_{\mathrm{non\text{-}hydrodynamic},i} + \sum_{j\neq i} \mathbf{F}_{\mathrm{contact},ij}.\end{aligned} \tag{5}$$

In this expression, the hydrodynamic force term can take into account the gravitational force as well as the drag and buoyant forces exerted by the fluid phase (e.g. air in a dry solid mixing process). The non-hydrodynamic force term plays a role for instance in cohesive systems and can allow for colloidal and capillary forces. We will come back to this point later.

A model is required to evaluate contact force term $\mathbf{F}_{\mathrm{contact},ij}$ in force balance (5). Generally, collisions involving two particles or one particle and the wall of the simulation domain are considered to be inelastic and are accounted for using the following *generalized spring-dashpot* model:

$$\mathbf{F}_{\mathrm{contact},ij} = \mathbf{F}_{\mathrm{contact},n,ij} + \mathbf{F}_{\mathrm{contact},t,ij}, \tag{6}$$

where

$$\begin{aligned}\mathbf{F}_{\mathrm{contact},n,ij} &= k_n \boldsymbol{\delta}_{n,ij}^{\alpha} + C_n \dot{\boldsymbol{\delta}}_{n,ij}, \\ \mathbf{F}_{\mathrm{contact},t,ij} &= k_t \boldsymbol{\delta}_{t,ij}^{\beta} + C_t \dot{\boldsymbol{\delta}}_{t,ij},\end{aligned} \tag{7}$$

which is an extension of an original model proposed by Cundall and Strack (1979). In (6), the contact force is equal to the sum of normal (head-on) and tangential (shear) contributions. The first and second terms in the right-hand sides of (7) represent repulsion and dissipation forces, respectively, $\boldsymbol{\delta}_{n,ij}$ and $\boldsymbol{\delta}_{t,ij}$ the normal and tangential components of a small overlap between contacting particles i and j, k_n and k_t stiffness coefficients, C_n and C_t damping coefficients, and α and β parameters. The tangential force $\mathbf{F}_{\mathrm{contact},t,ij}$ is generally bounded above by $\mu_s |k_n \mathbf{F}_{\mathrm{contact},n,ij}|$ (or a similar expression) following the Coulomb law of friction, where μ_s is the friction coefficient.

As mentioned at the beginning of this section, the DEM described above belongs to the family of soft-particle methods. In this case, particle overlaps are allowed and used to express the non-instantaneous contact between two colliding particles. There are particulate methods, based upon hard-sphere models, for which the collisions are instantaneous and no interpenetration is allowed (e.g. Mehta, 1994). Note that these latter models are also often referred to as DEMs.

Many models based on (6) and (7) can be found in the literature and the reader is referred to Bertrand et al. (2004) and Schäfer et al. (1996) for more details. The most familiar of these models is undoubtedly the *linear spring-dashpot model* ($\alpha = \beta = 1$ in (7)) of Cundall and Strack (1979):

$$\begin{aligned}\mathbf{F}_{\mathrm{contact},n,ij} &= k_n \boldsymbol{\delta}_{n,ij} + C_n \dot{\boldsymbol{\delta}}_{n,ij}, \\ \mathbf{F}_{\mathrm{contact},t,ij} &= k_t \boldsymbol{\delta}_{t,ij} + C_t \dot{\boldsymbol{\delta}}_{t,ij},\end{aligned} \tag{8}$$

in which a spring is used to provide a repulsive force and a dashpot to dissipate a proportion of the kinetic energy. In this model, particles are allowed to overlap to an extent determined by the value of stiffness parameters k_n and k_t. It is possible to avoid the explicit use of a damping coefficient for the normal force by resorting to the hysteretic *partially latched spring model* of Walton and Braun (1986), which consists of employing different values of stiffness coefficient k_n for the loading and unloading parts of a particle collision:

$$\mathbf{F}_{\mathrm{contact},n,ij} = \begin{cases} k_{n,in} \boldsymbol{\delta}_{n,ij}, & \text{if } \dot{\boldsymbol{\delta}}_{n,ij} \geqslant 0, \\ k_{n,out} \boldsymbol{\delta}_{n,ij}, & \text{if } \dot{\boldsymbol{\delta}}_{n,ij} \leqslant 0. \end{cases} \tag{9}$$

It can be shown that this corresponds to a dissipative system since the coefficient of normal energy restitution $e_n = \sqrt{k_{n,\mathrm{in}}/k_{n,\mathrm{out}}} \neq 1$. In this model, the tangential force is calculated as

$$\mathbf{F}_{\mathrm{contact},t,ij} = k_t \boldsymbol{\delta}_{t,ij}.$$

Another DEM-based model relies upon Hertz theory and considers that the contacts are elastic and non dissipative. Such a model was proposed for instance by Johnson (1989) to simulate normal impacts ($\alpha = 3/2$, $C_n = 0$, $k_t = 0$, $C_t = 0$ in (7)):

$$\begin{aligned}\mathbf{F}_{\mathrm{contact},n,ij} &= k_n \boldsymbol{\delta}_{n,ij}^{3/2}, \\ \mathbf{F}_{\mathrm{contact},t,ij} &= 0,\end{aligned} \tag{10}$$

where the stiffness coefficient, k_n, can be expressed as a function of the Young's modulus E and Poisson ratio υ, and thus depends on the particle elastic properties.

Recently, Zhou et al. (1999) proposed a model that can be viewed as a combination of the model proposed by Cundall and Strack (1979) and the Hertz law of contact given by (10):

$$\mathbf{F}_{\text{contact},n,ij} = k_n \boldsymbol{\delta}_{ij}^{3/2} + C_n \dot{\boldsymbol{\delta}}_{ij},$$
$$\mathbf{F}_{\text{contact},t,ij} = k_t \boldsymbol{\delta}_{ij} + C_t \dot{\boldsymbol{\delta}}_{ij}, \tag{11}$$

where

$$k_n = k_n(E, \upsilon),$$
$$c_n = c_n(E, \upsilon),$$
$$k_t = k_t(\mu_s |k_n \boldsymbol{\delta}_{n,ij}^{3/2}|),$$
$$c_t = c_t(\mu_s |k_n \boldsymbol{\delta}_{n,ij}^{3/2}|). \tag{12}$$

The term $\mu_s |k_n \boldsymbol{\delta}_{n,ij}^{3/2}|$ is used to connect the shear force to the normal force by means of the Coulomb law of friction. Model (11)–(12) was used by Stewart et al. (2001) to simulate the flow of non-cohesive granules in a blade mixer. In particular, these authors studied the effect of the friction coefficient on the quality of the results and showed that a value of 0.4 in the case of glass beads lead to dispersion indices that were comparable to those obtained from experimental results based on positron emission particle tracking (PEPT). Since they also obtained rather good values for the angle of repose of this type of particles using the same friction coefficient, they concluded that this parameter is in effect a physical parameter that can be used for any DEM simulation involving glass beads.

DEMs have been used to simulate the flow of granular materials in various applications (Cleary and Sawley, 1999a). In the field of mixing, most of the works from the literature have focused on the dynamics of non-cohesive powders in common mixers and tumbling devices. In particular, the following mixing systems have been investigated in 3D:

- Rotating drum (Ristow, 1998);
- Double-cone blender to mix monodisperse and bidisperse powders (Moakher et al., 2000);
- V-blender to mix monodisperse and bidisperse powders (Moakher et al., 2000);
- Tote blender (from Moakher's web page at http://lcvmsun9.epfl.ch/~moakher/mixing/);
- Tumbling (ball) mills for polydisperse powders (Cleary and Sawley, 1999a; Rajamani et al., 2000);
- Helical ribbon mixer (Kaneko et al., 2000; Bertrand et al., 2003);
- Flat blade mixer (Stewart et al., 2001);
- Kenics static mixer (Szépvölgyi, 2001).

All these investigations have pertained to non-cohesive spherical grains. To our knowledge, a limited number of studies have considered the case of either cohesive or non-spherical particles. We can cite the work of Szépvölgyi et al. (1999) who developed a DEM-based model to predict the flow of cohesive spherical particles in a high-shear mixer. Also of interest is the recent work of Pelessone (2003) who proposed a strategy for modeling the flow of non-spherical particles in tumbling mills. We will come back to the treatment of cohesive forces and non-spherical particles later in separate sections.

As was just seen, there is more than one DEM-based model since particle/particle and particle/wall collision dynamics can be described in many ways. A thorough analysis of the contact force schemes of this section as well as other ones to describe viscoelastic behavior can be found in Schäfer et al. (1996), Ristow (1998) and in the recent work of Di Renzo and Di Maio (2004). The models used for the applications of DEM to granular mixing reported above are:

- Linear spring-dashpot model (Ristow, 1998; Cleary and Sawley, 1999a; Kaneko et al., 2000; Rajamani et al., 2000; Pelessone, 2003);
- Hysteretic partially latched spring model (Moakher et al., 2000);
- Generalized spring-dashpot model with $\alpha = 3/2$ (Hertz law) (Szépvölgyi et al., 1999);
- Model of Zhou et al. (1999) (Stewart et al., 2001; Bertrand et al., 2003).

All these models depend on parameters, the value of which must be set. Moreover, they are not equivalent since they deal with particle contacts differently. For instance, model (10) is based on Hertz theory and considers that the contact between two colliding particles lead to an elastic deformation, whereas the linear spring-dashpot model (8) considers that this deformation is inelastic. Moreover, a DEM simulation generally requires millions of time iterations involving numerical algorithms such as the leapfrog or Verlet schemes (Allen and Tildesley, 1987). Even if a high-order (and costly) predictor–corrector scheme is used, this large quantity of iterations may lead to a significant error on the position of the particles in the simulation domain. Consequently, it is very important to validate any DEM-based model prior to using it, a step that is often dodged by researchers.

4. Validation of the DEM

A careful validation strategy should always include as an ultimate test a comparison between the numerical results and experimental data for a relevant application. For instance, we used DEM to simulate the sedimentation of monodisperse and polydispersed pigments on a paper substrate in paper coating applications; the accuracy of the simulations were assessed by comparing the porosity of the numerical packings obtained with DEM to mercury porosimetry data (Bertrand et al., 2004). In the case of mixing applications, comparisons between DEM results and different types of

experimental data can be found in the literature:

- Calculation of dispersion indices using PEPT measurements. With this technique, a radioactive tracer is tracked by a positron camera as it moves in the vessel and the behavior of the entire particulate system can be reconstructed from the motion of this single tracer over a long period of time. This approach was used for example by Stewart et al. (2001) in the case of a blade mixer to show the influence of the friction coefficient on the dispersion index.
- Qualitative observation using a video camera system
  - McCarthy et al. (1996) used observation to develop a DEM-based model and qualitatively study mixing in rotating containers;
  - Kaneko et al. (2000) used a colored particle to measure the circulation time in a vessel provided with a helical ribbon impeller;
  - Moakher et al. (2000) studied segregation and mixing mechanisms in double-cone and V-blenders;
  - Venugopal and Rajamani (2001) compared actual and simulated particle profiles in a tumbling mill.
- Measurement of the power draw. This approach was proposed by Venugopal and Rajamani (2001) in the case of a tumbling mill. The numerical power draw can be evaluated by summing over time the work done by the particles when they collide with one another and the wall of the tumbling mill.
- Estimation of the mixing time. For instance, Kaneko et al. (2000) evaluated the time needed for tracer particles to mix with polypropylene particles. To do so, they collected samples of particles at regular time intervals and plotted the concentration of the tracer particles with respect to time. The estimated mixing time was then the time at which the concentration curve levels off.

Before testing out a DEM-based model on a relevant application, it certainly makes sense to assess its validity using a benchmark test such as the one proposed by Beverloo et al. (1961). It consists of the flow of grains through an orifice at the base of a cylindrical storage silo or container, with the objective of relating the discharge rate of these grains to the size of the orifice. In fact, Beverloo et al. (1961) and later Nedderman et al. (1982) showed that the discharge rate W of mono-sized grains of diameter d and density ρ through a circular orifice of diameter D_0 is given by

$$W = C\rho\sqrt{g}(D_0 - kd)^{5/2} \tag{13}$$

if the following hypotheses

$$\begin{aligned} &H > 2D_0, \\ &D > 2D_0, \\ &400\,\mu\text{m} < d < D_0/6, \end{aligned} \tag{14}$$

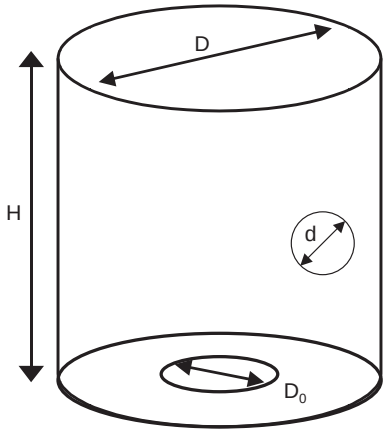

Fig. 2. Dimensions of the container used for the Beverloo's correlation.

are satisfied, where D and H stand for the diameter and the height of the container (Fig. 2). Constants k and C are two dimensionless parameters to be determined experimentally. In the case of particles that are not too small ($d > 1$ mm), values of $C = 0.61 \pm 0.3$ and $k = 1.4 \pm 0.1$ were reported by Beverloo et al. (1961).

We routinely use the Beverloo's correlation to validate our DEM-based models. Actually, the structure of our DEM code is such that any of the models from the previous section can be activated. So far, we have mainly used the linear spring-dashpot model (8) and a modified version of the model proposed by Zhou et al. (1999). As an illustration, we show next the results of the validation of the latter model in the case of a container filled with spherical particles, the dimensions of which are as follows (Fig. 2):

- $H = 0.5$ m;
- $D = 0.3$ m;
- $D_0 = 0.1$, 0.12 and 0.14 m.

Two particle diameters were considered for this example: 10 and 16 mm.

Even though the values of C and k in Beverloo's correlation do not depend much on the nature of a particle, it was decided to vary the value of the friction coefficient μ_s in the simulations to see if such a variation would have an impact on the value of these regression parameters. More precisely, the following values were considered:

$$\mu_s = \begin{cases} 0.2 < \mu_{s,\text{static}} < 1.5 & \text{if } v_{\text{particle}} \leqslant \varepsilon, \\ 0.05 < \mu_{s,\text{dynamic}} < 0.15 & \text{if } v_{\text{particle}} > \varepsilon, \end{cases} \tag{15}$$

where v_{particle} is a threshold velocity below which a particle is considered to be at rest, and $\mu_{s,\text{static}}$ and $\mu_{s,\text{dynamic}}$ represent the static and dynamic friction coefficients respectively, following the Coulomb law of friction. The value of the other parameters of the DEM-based model can be found in Table 1. In particular, the values chosen for the Young's modulus, the Poisson ratio and the density are those for typical glass beads.

Table 1
Values of the parameters in the DEM-based model for the simulation of the flow of spherical particles through an orifice

Parameter	Symbol	Value
Particle diameter	d	10 and 16 mm
Density	ρ	2500 kg/m^3
Young's modulus	E	2.16×10^6 Pa
Poisson ratio	v	0.3
Static friction coefficient	$\mu_{s,\text{static}}$	[0.2–1.5]
Dynamic friction coefficient	$\mu_{s,\text{dynamic}}$	[0.05–0.15]

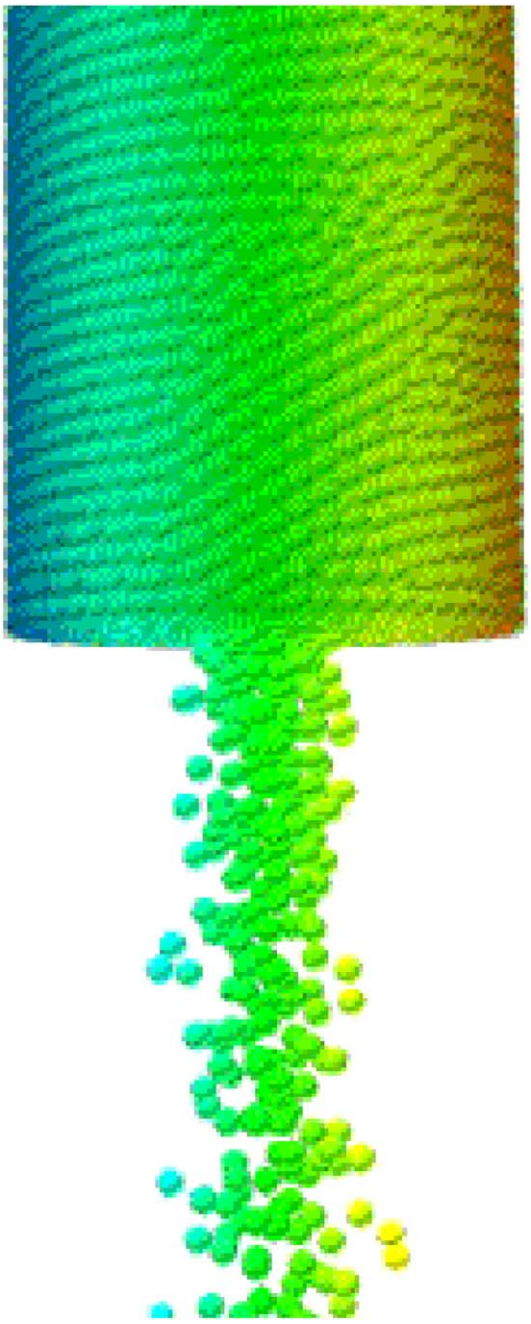

Fig. 3. Flow of spherical particles through the orifice of the container. A color is initially assigned to each particle depending on its position (x-coordinate) in the container.

Table 2
Values of parameter k for different values of the static and dynamic friction coefficients

Friction coefficients		Parameter	Orifice diameter	Discharge rate	Discharge rate
$\mu_{s,\text{dynamic}}$	$\mu_{s,\text{static}}$	k	D_0 (mm)	DEM (kg/s)	Beverloo's correlation (kg/s)
0.05	0.2	1.404	100	7.545	7.601
			120	13.41	13.49
			140	21.35	21.51
0.02	0.4	1.414	100	7.774	7.562
			120	14.03	13.44
			140	22.08	21.43
0.04	0.4	1.445	100	7.447	7.442
			120	13.24	13.27
			140	21.22	21.21
0.05	0.5	1.468	100	7.269	7.353
			120	13.06	13.14
			140	20.81	21.04
0.08	0.4	1.481	100	7.010	7.303
			120	12.44	13.07
			140	20.08	20.95
0.1	0.4	1.504	100	6.906	7.215
			120	12.32	12.94
			140	19.88	20.78
0.05	0.8	1.509	100	7.307	7.208
			120	13.15	12.93
			140	21.07	20.77
0.05	1	1.525	100	7.453	7.136
			120	13.36	12.83
			140	21.54	20.63
0.1	1	1.538	100	6.834	7.087
			120	12.39	12.76
			140	19.83	20.54
0.15	1.5	1.542	100	6.357	6.799
			120	11.49	12.25
			140	18.49	19.72

Particle diameter is 16 mm.

Simulations of the flow of 9294 16 mm particles were first carried out (Fig. 3). These simulations were performed over a period of time long enough (less than one second) for the discharge rate to stabilize. The discharge rate was calculated by dividing the number of particles going through the orifice by the elapsed time. For a given value of the static and dynamic friction coefficients, the value of k was obtained by linear regression assuming that $C = 0.58$ in Eq. (13). Three data points were used each time, which correspond to the discharge rate obtained for three orifice diameters. As can be seen in Table 2, k is between 1.404 and 1.542, which complies with the results reported by Beverloo et al. (1961) and Nedderman et al. (1982). More precisely, the discrepancy between the numerical discharge rates and the ones predicted by correlation (13) varies from 1% to 6%, depending on the value of the friction coefficients. Moreover, one may observe that when friction increases, the value of k increases and the discharge rate decreases. This is to be expected since friction then generates more resistance to the flow.

Table 3
Values of parameter k for different particle sizes and different values of the static and dynamic friction coefficients

Friction coefficients		k	
$\mu_{s,\text{dynamic}}$	$\mu_{s,\text{static}}$	$d = 10$ mm	$d = 16$ mm
0.04	0.4	1.434	1.445
0.05	0.5	1.467	1.468
0.08	0.4	1.475	1.481

A few other simulations involving 38 878 10-mm particles were carried out to assess the influence of the particle size on the value of k. These results are presented in Table 3. As can be seen, the effect of the particle size is negligible and the discharge rate is indeed proportional to the so-called "empty annulus" $D_0 - kd$ in Eq. (13).

These results confirm that the correlation

$$W = 0.6\rho\sqrt{g}(D_0 - 1.4d)^{5/2} \tag{16}$$

can be used in the validation process of any DEM-based model. As a matter of fact, this test has allowed us to detect, on more than one occasion, faults in our codes, sometimes in portions that we thought were bug free.

For the sake of completeness, let us mention that a few numerical investigations of the Beverloo's correlation can be found in the literature. For instance, Hirshfeld and Rapaport (2001) used a 3D molecular dynamics method to study the effect of the orifice diameter on the flowrate of particles. Also of interest is the work of Kano et al. (1998) who revisited the Beverloo's correlation using DEM in the case of conical hoppers.

5. Techniques to describe the motion of impellers

Simulating the flow of particles in a mixing system can be quite challenging because one must cope with a complex computational domain comprising tank walls and a moving impeller. In such simulations, the interactions between the particles and these boundaries are critical to the flow behavior and, as a result, efficient methods for representing the boundary surfaces must be developed and integrated into the DEM model. It appears that this aspect of discrete element modeling has not drawn much interest so far, a fact that is likely to change with the broadening of the range of applications for this method.

Three strategies can be used to take into account the shape and the kinematics of boundaries in a DEM simulation. The first one is based on a continuous formulation whereby a boundary surface is approximated by a mathematical expression. This technique can be effective when the geometry of the domain is simple as in the case, for instance, of a rotating cylinder without internal parts (e.g. Yamane et al., 1998). It is however not suitable in the case of complex boundary shapes or in the case of a computational domain that contains a moving part.

The second strategy is based on the discretization of the boundary surfaces by means of a finite-element mesh (a mesh of triangles, for instance) that can be generated with a mesh generator such as IDEAS. This technique, called the multi-wall method by Kremmer and Favier (2001), is very flexible and can easily handle moving boundaries such as those of an impeller. In this latter case, the position of the corresponding mesh nodes can be determined in a straightforward manner from the impeller kinematics. The main drawback of this technique is that it entails modifications to an existing DEM code to take into account particle/triangle interactions in addition to particle/particle interactions. Moreover, these interactions are much more expensive computationally since they can take place in different ways: contact within a triangle, along one of its edges, on one of its vertices. As an illustration of this technique, we show in Fig. 4a a triangular discretization of a vessel provided with a helical ribbon impeller. The characteristics of this helical ribbon blender are given in Table 4. A total of 4070 and 9022 triangles were used to discretize the vessel walls and the helical ribbon, respectively. A simulation was carried out to predict the flow of 6440 10-mm spherical particles in this blender at an impeller speed of 4 rad/s or 38 rpm. Fig. 4b shows the filling step before the impeller is set in motion; each particle is assigned a color according to its axial position (z-coordinate) in the vessel at $t = 0$ s. Figs. 4c–f display the position of the particles at $t = 1, 2, 5$ and 6 s, respectively. In particular, one may observe that, from a qualitative point of view, the helical ribbon generates rather good axial and radial dispersions, Fig. 4f corresponding to the situation just before the impeller has completed its fourth revolution.

To gain more insight into the accuracy of this second technique, we simulated the flow of 16 000 5-mm glass beads in a system consisting of a vertical 249-mm diameter cylindrical mixer equipped with two flat and aligned 40-mm high blades, following up on a recent work by Stewart et al. (2001). Simulations were performed using parameters similar to those of these authors. The rotational speed of the blade impeller was 60 rpm. We present in Figs. 5 and 6 the position of the particles in the blender after 0.1, 0.4, 0.7, 1, 2, 3, 4, 5 s, respectively. As in the previous simulations, the color of a particle denotes its position in the vessel at $t = 0$ s. As evidenced by the yellowish wake in Fig. 5d, a good convective dispersion owing to the motion of the blade impeller may be observed. A rather efficient although more progressive (diffusive) radial dispersion may also be noted. In fact, the dispersion indices that were calculated from these results are comparable to the numerical and PEPT-based experimental dispersion indices reported by Stewart et al. (2001). In particular, we detected a similar influence of the friction coefficient on the value of these indices.

The third strategy consists of resorting to granular boundaries built from an assemblage of particles the size and the position of which are chosen to mimic as accurately as

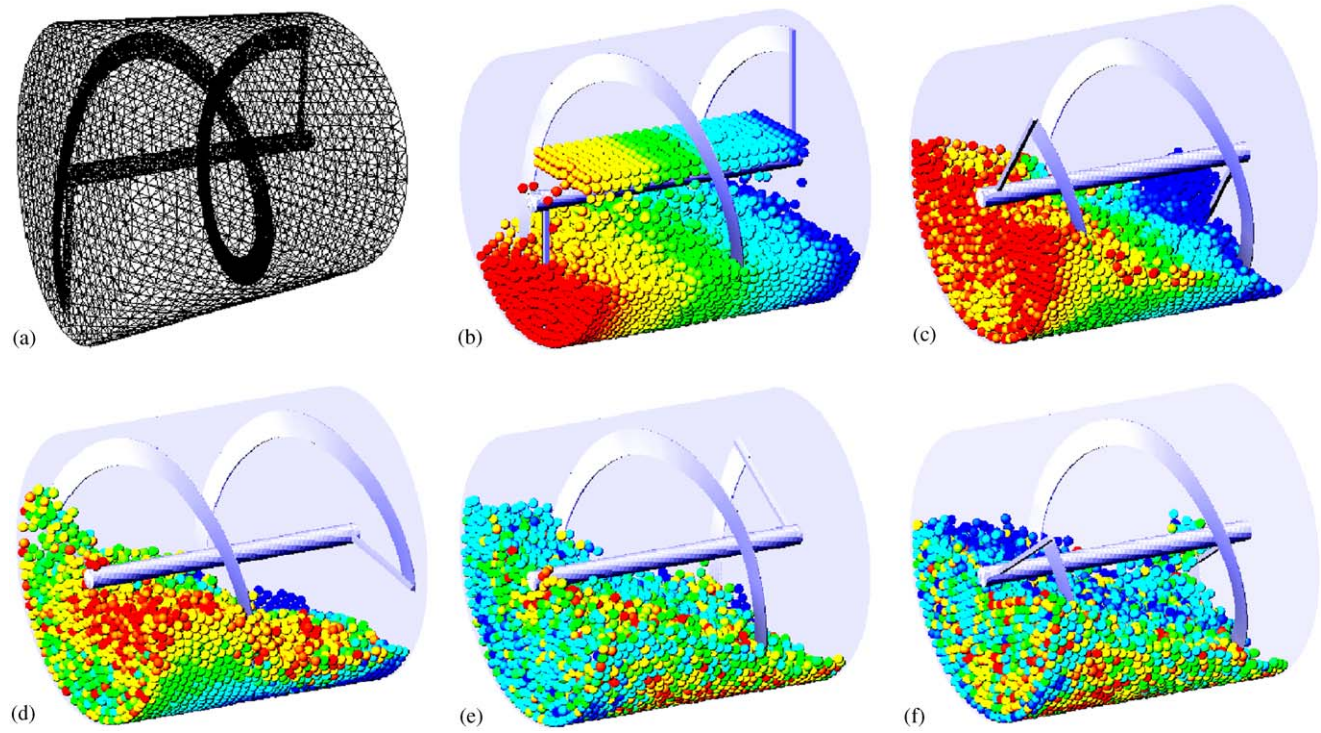

Fig. 4. DEM simulation of the flow of spherical particles in the helical ribbon blender: (a) triangular discretization of the boundary surfaces; (b) filling step at $t = 0$ s; and (c–f) position of the particles after 1, 2, 5 and 6 s.

Table 4
Characteristics of the helical ribbon mixer

Vessel diameter (m)	0.292
Ribbon diameter (m)	0.263
Vessel length (m)	0.299
Ribbon length (m)	0.276
Ribbon width (m)	0.0255

possible the roughness of the domain boundaries. In particular, the spacing between these boundary particles can be large to mimic a rough wall or small with an overlap to reproduce a smooth surface. Such a technique was used by Hirshfeld et al. (1997) to describe the walls of a silo and by Cleary et al. (1998) in the case of a rotating drum. We show in Fig. 7 the application of this technique to the case of a tumbling mill. The walls of the tumbling mill were first discretized into 5748 triangles using IDEAS. Each triangle was then covered by a set of overlapping spheres as depicted in Fig. 7a. Fig. 7b shows a portion of the resulting granular boundary that comprises a total of 41 000 spherical particles, overlaid on the triangular mesh. Let us mention that the properties of these wall particles (e.g. diameter, friction coefficient) need not be identical to those of the true particles. This technique is very appealing since it relies on particle/particle interactions. Therefore, no major modifications to an existing code are needed. The biggest drawback a priori seems to be the amount of wall particles that results from the covering of all the boundary triangles but, as we will see next, this method is not so costly in practice.

A simulation was carried out for 4096 8-mm spherical particles and a rotational speed of 40 rpm (Figs. 7c and d). It required 15 h of CPU time per revolution on an IBM P690 computer in serial mode. A similar simulation performed using just the mesh of 5748 triangles to account for the tumbling mill walls (second strategy) required about the same time. At first sight, it would seem that this latter simulation is more accurate since it does not rely upon a granular boundary. But considering that there is a relationship between the roughness of a wall and its friction properties, it would make sense to suggest that similar flow behaviors can be obtained by both techniques if proper parameters are used, that is the wall particle diameter, overlap size and friction coefficient for the third technique, and the triangle friction coefficient for the second technique. Quite clearly, a more thorough investigation is needed to compare the accuracy of these two techniques and propose guidelines for the setting of the corresponding parameters.

6. Techniques to handle non-spherical particles

Flow of non-spherical particles is of great importance in mixing applications and typical examples can be found for instance in the manufacturing of pharmaceutical products. It appears that no reports can be found in the

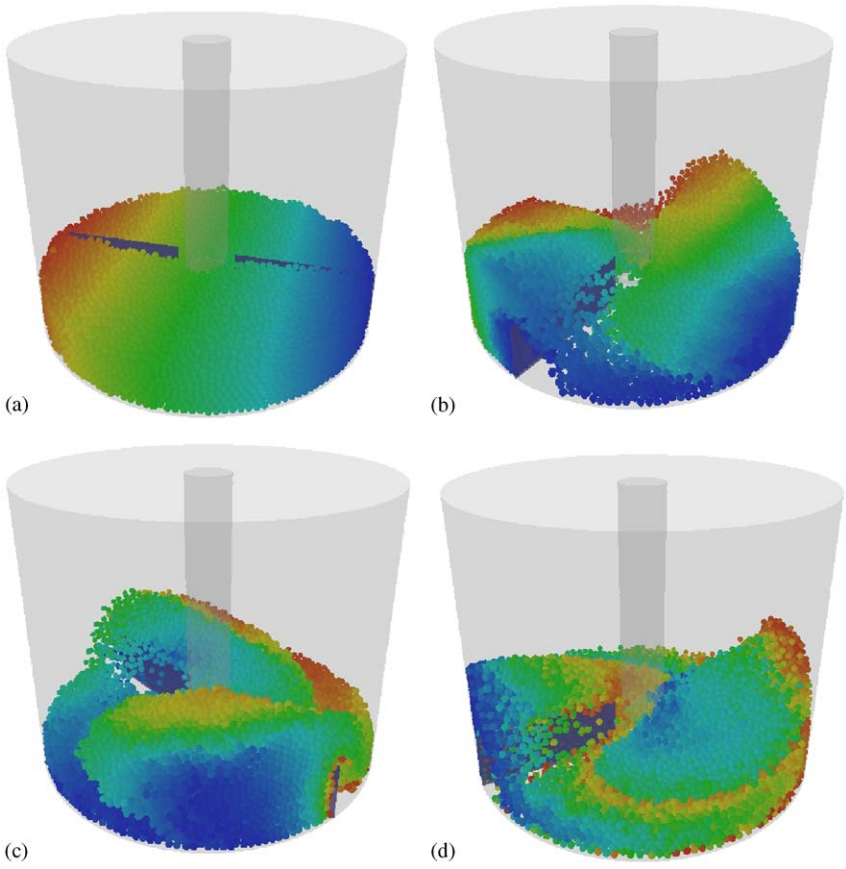

Fig. 5. DEM simulation of the flow of spherical particles in the blade mixer after (a) 0.1 s, (b) 0.4 s, (c) 0.7 s and (d) 1 s.

literature as regards the modeling of the mixing of irregularly shaped particles, with the exception of the recent work of Pelessone (2003) who simulated the flow in a tumbling mill of macro-particles composed of four overlapping spheres. In other fields of application, the number of numerical works involving non-spherical particles remains limited, most of them relating to flow simulations in silos and hoppers (e.g. Hirshfeld et al., 1997) with only a few in 3D (e.g. Langston et al., 2004).

Contact detection is far more intricate in the case of non-spherical particles, which explains why most of the numerical investigations with DEM have concerned spherical particles. Several techniques have been devised over the years to cope with particles of irregular shapes and good reviews can be found in Dziugys and Peters (2001) and Langston et al. (2004). Currently, two approaches seem to stand out and arouse interest. The first one considers that the particles are of a given shape, for instance an ellipsoid or a super-ellipsoid, and determines whether or not there is a contact between two such neighboring particles by solving the underlying mathematical equations. Such a method was developed in 2D by Wait (2001) and Cleary and Sawley (2002), and developed and used in 3D by Munjiza et al. (2003) to predict the packing of pill-shaped particles. If this approach is very accurate, it is however computationally greedy and difficult to implement efficiently in 3D, although recent developments proposed by Munjiza et al. (2003) show promise. To our knowledge, it has never been used in the context of granular mixing.

The second class of methods can be viewed as a clustering technique, following along the lines of Gallas and Sokolowski (1993) and Pelessone (2003). The idea is to model an irregularly shaped particle as a collection of spherical particles, and to enforce a rigid body motion of this composite particle using an appropriate value of a fictitious force between the atomic particles. This strategy is advantageous from the point of view of contact detection; only a standard contact detection algorithm for spheres is required. As a result, it is quite flexible and can be used to handle particles of very complex shapes and sizes. On the downside, it is less accurate than the first technique and increases significantly the number of spheres in the system. Note that the spheres making up a macro-particle may overlap and vary in diameter. This clustering technique was implemented in our code and used to simulate the flow of non-spherical particles in various systems, including a tumbling mill rotating at 40 rpm with 1024 tetrahedral particles each composed of four 8-mm spheres (Fig. 8). The influence of the number of

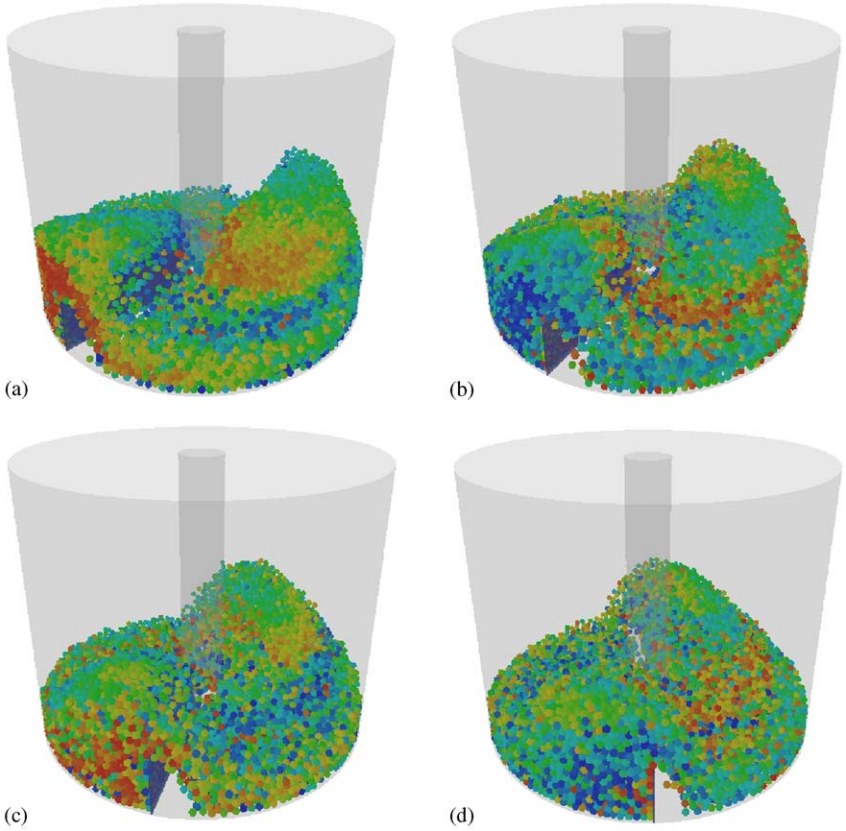

Fig. 6. DEM simulation of the flow of spherical particles in the blade mixer after (a) 2 s, (b) 3 s, (c) 4 s and (d) 5 s.

atomic particles composing a macro-particle on the overall accuracy of the method has never been studied. It is currently under investigation by our group.

7. Algorithmic aspects

Three-dimensional simulations done with DEM are known to be extremely CPU-intensive, requiring from a few hours to many days in the case of systems involving a large number of particles. To alleviate this problem and enable the simulation of realistic systems (10^6 particles and more, depending on the particle diameter and the size of the vessel), DEM must be parallelized. This task can be achieved through domain decomposition techniques since the particles are assumed to interact only with their immediate neighbors. The idea is to split up the computational domain into sub-domains and use overlapping regions called halos to enable the treatment of particle contacts across the border of these sub-domains. We refer the reader to Cleary and Sawley (1999b) for more information on this issue. Suffice it to say here that, in a parallel implementation of DEM, each sub-domain is assigned a different processor and is given the mandate to calculate the position of each of its particles and transmit to the neighboring sub-domains (processors) the position of the particles in its halos.

The expected speedup with parallel DEM depends on the problem and, in particular, on the shape of the computational domain and the number of particle interactions. It also depends on the type of parallel computer and on the domain decomposition technique used (decomposition in slices, boxes or cubes). This issue has been addressed so far by a few researchers either theoretically (e.g. Henty, 2000) or in the case of 2D packing problems (e.g. Ferrez et al., 1996). To our knowledge, no such investigation has been reported in the case of granular mixing simulations. As a matter of fact, we are currently examining the potential of the parallel version of DEM that we implemented recently on Beowulf clusters (Leclaire, 2004). To this end, we have considered as a benchmark problem the flow of 8-mm spherical particles in a tumbling mill rotating at 40 rpm, as previously discussed in Section 5. A fairly large amount of simulations have been carried out so far using a varying number of processors on 24-processor (866 MHz Pentium-III) and 32-processor (700 MHz Xeon) clusters. From the CPU times and speedup values that have been collected, we built Table 5 that gives the expected CPU time per impeller revolution with respect to the number of particles in the tumbling mill and the number of processors used for the simulation. As can be seen, DEM simulations are extremely time consuming, so much that a parallel computer is recommended in the case of systems containing fewer than 10^4 particles, and required when

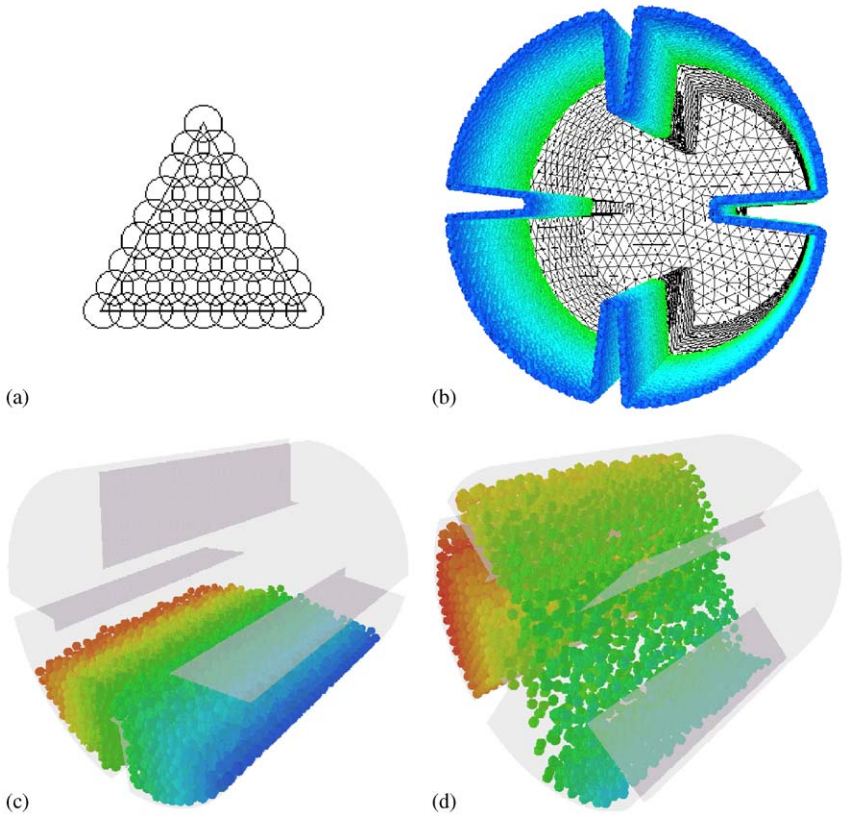

Fig. 7. DEM simulation of the flow of spherical particles in the tumbling mill: (a) filling of a triangle with spheres; (b) use of granular boundaries; and (c and d) position of the particles at 0 and 0.6 s.

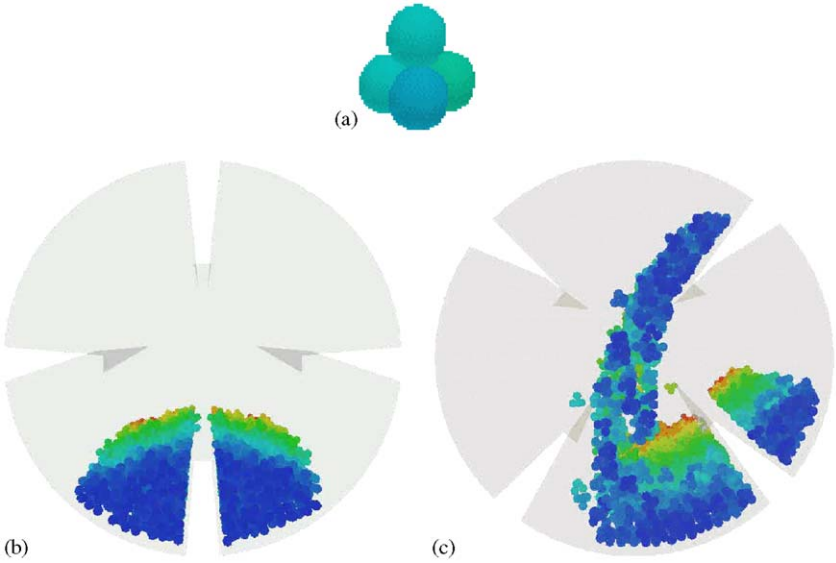

Fig. 8. DEM simulation of the flow of tetrahedral particles in the tumbling mill: (a) tetrahedral particle composed of four 8-mm spheres; and (b and c) position of the particles at 0 and 4 s.

the number of particles is larger. In particular, the expected CPU time per impeller revolution for 10^6 particles is of the order of a half week on a 64-processor Beowulf cluster! The question that then comes to mind is: how can we simulate larger granular systems involving smaller particles and possibly non-hydrodynamic cohesive forces? We will address

Table 5
CPU time with DEM for the simulation of the flow of spherical particles in a tumbling mill

Number of particles	CPU time/impeller revolution		
	Serial computer	32 procs (speedup = 16)	64 procs* (speedup = 32)
10^3	3 h	10 min	5 min
10^4	1.25 days	2 h	1 h
10^6	4 months	1 week	3.5 days

*Indicates extrapolated values.

this question in the concluding remarks after we have discussed the treatment of cohesive forces in DEM.

8. Cohesive systems

In granular mixing or in other processes involving solid particles, the powder is no longer free-flowing and cohesion becomes a factor when the size of the particles is so small (smaller than of the order of 10 μm) that colloidal forces (electrostatic or Van der Waals) are significant, or when moisture is such that particle agglomeration may take place. If a fairly large amount of progress has been made in the last 25 years about the understanding of the phenomena characterizing the flow of non-cohesive particles, the level of knowledge is much lower in the case of the flow and mixing of small interacting cohesive particles (Bridgwater, 2003). For instance, as mentioned in the introduction, the mixing of tiny proportions of small cohesive particles in a matrix of much larger particles is difficult to achieve in pharmaceutical applications (Muzzio et al., 2002). A better understanding of the phenomena governing the flow of cohesive systems is definitely required.

Theoretically, in DEM, cohesive forces can be added to the force balance for each pair of neighboring particles. As far as we know, the only report concerning the use of DEM to simulate the mixing of cohesive particles is that of Szépvölgyi et al. (1999). More precisely, they investigated in 3D the dynamic behavior of a bed of spherical particles in an elliptical rotor-type high shear mixer. The number of particles was of the order of 5000 and their diameter was rather large, 200 mm. Adhesion in such a system resulted from the coating of these core particles by much finer particles. To model the resulting Van der Waals forces, these authors resorted to the so-called JKR theory after the work of Johnson et al. (1971). In this theory, the attractive force between two neighboring particles is a function of their intrinsic surface energies.

The DLVO theory represents a milestone for the evaluation of colloidal forces (electrostatic and Van der Waals forces) between two particles. It was combined by Toivakka and Eklund (1994) with the SD method of Brady and Bossis (1985) to simulate in 2D the packing of fine cohesive particles. It could be combined with DEM to investigate the flow behavior in systems of cohesive particles.

Another approach was proposed recently by Deiva et al. (1998) to take into account powder cohesion. Called the powder chemistry model, it considers the mechanisms of agglomeration/deagglomeration in a system of fine particles as reversible processes similar to chemical reactions. The overall strategy hinges around molecular dynamics and a hydrodynamic force balance for each particle, following Gallas et al. (1992). At the end of each time iteration, the occurrence of agglomeration/deagglomeration of two neighboring particles is decided on the basis of their kinetic energy and the values of the so-called bond and minimum collisional energies.

The occurrence of liquid bridging between particles can be studied by resorting to the model of Lian et al. (1993) wherein the corresponding (capillary) force is a function of the particle radius, the so-called half-filling angle of the liquid bridge and the surface tension of the fluid. McCarthy (2003) used this model recently to shed light on the effect of liquid bridging on the mixing efficiency of standard tumblers. When investigating the effect of liquid bridging on granular flow in a centrifugal tumbling granulator, Muguruma et al. (2001) showed the efficiency of a similar model by comparing their numerical results with velocity profiles obtained using a particle velocimetry technique.

As was just shown, rather simple models are available to describe the interaction of two neighboring particles subject to cohesion forces (Van der Waals forces or capillary forces due to liquid bridging) and their implementation in a DEM code can be done in a straightforward manner. The use of such models will no doubt lead in a very near future to a better understanding of the effects of these cohesion forces on the flow behavior of particles in mixing systems.

9. Concluding remarks

The goal of this work was to portray the current state of the art concerning the modeling of granular flows in mixing processes. Emphasize was put on DEM, a method that is used more and more to study the flow behavior of granular materials in mixing systems. Theoretical as well as practical aspects of the method were covered including the treatment of non-spherical grains, cohesion and complex impeller systems. The greediness of DEM was also discussed and illustrated with CPU times in the case of a tumbling mill with a varying number of particles. These CPU times clearly stress, on the one hand, the importance of running a DEM code in parallel using for instance commodity clusters of Intel processors. They also point out on the other hand that, even with the use of such powerful computers, the number of particles that can be dealt with at the moment is limited. So the question of the end of Section 7 remains: how can we simulate the flow of a huge number ($> 10^6$ or even 10^{10} in

the case of industrial pharmaceutical blenders) of fine (cohesive) particles in a mixing system? One partial answer to this question consists of combining CFD techniques and DEM, as suggested recently by Christakis et al. (2002). In this computational framework, a finite element (or finite volume) method is used to compute a bulk velocity $\mathbf{v}_{\text{bulk}}$ and each species concentration C_i is obtained by integrating the transport equation

$$\frac{\partial C_i}{\partial t} + \text{grad}(C_i(\mathbf{v}_{\text{bulk}} + \mathbf{v}_{\text{drift}})) = S, \tag{17}$$

where $\mathbf{v}_{\text{drift}}$ stands for a drift velocity the value of which comes from a micro-mechanical model. The form of this model can be derived from the behavior of the particles in relevant flow systems as predicted by DEM.

The above strategy is reminiscent of a hybrid technique proposed by McCarthy and Ottino (1998), which consists of calculating the individual motion of particles (drift velocity) only where it is relevant, and to move the other particles using a bulk velocity the value of which comes from geometrical considerations. They applied this idea with success to the mixing of particles in a tumbler.

As mentioned recently in Michaels (2003), DEM simulations should be used to investigate the mesoscale properties of powders. These simulations, which would involve much fewer particles than in the case of a whole system, could lead to the development of constitutive models to be passed along to less computationally greedy and more practicable process-scale models such as the ones described above and the ones based on Markov chains as discussed in Section 2. The development of such hybrid strategies represents without a doubt one the challenges that will need to be taken up in the future.

Acknowledgements

The financial contribution of NSERC is gratefully acknowledged.

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