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Formulation and Comparison of Alternative Multiscale Models for Drum Granulation

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Abstract
Continuous granulation of powders is a unit operation that poses both operational and modelling challenges. We discuss the formulation of several alternative multiscale models for a drum granulator. All combine a vessel scale population balance model with a granule scale discrete element method (DEM) model, but they are linked using different *multiscale integration frameworks*. The inter-scale flow of information and the potential applications of the alternative models are outlined.

Keywords: multiscale modelling, granulation, population balance model, discrete element method

1. Introduction

Ennis (1997) reported that around 60% of chemical industry products are particulate materials and that a further 20% use powder intermediates. Even as long as ten years ago, particulates contributed to products worth \$US1 trillion per annum in the US economy alone. Particulate materials pose modelling challenges for the CAPE community over and above those for purely fluid systems. Not only is an additional phase present—and several solid phases may co-exist—but they have a persistent structure, possibly at the nano or micro level, that is often essential to their end-use properties. Particulate processes use unit operations not found in gas-liquid systems. Mathematically, models that use the population balance, which describes the conservation of particle number, are sets of integro-partial differential algebraic equations. Particulate systems can prompt advances in multiscale thinking and mathematical methods by providing a rich variety of important problems to solve.

Granulation is a key, if problematical, particulate unit operation. It is the process of agglomerating a fine powder into semi-permanent granules. In drum granulation, a liquid binder or slurry is sprayed onto a bed of powder tumbling in a rotating drum. Continuous granulation circuits usually operate with high recycle ratios and often suffer surging and off-specification product. Granulation can be viewed as a multiscale process; we identified several characteristic length scales earlier (Ingram and Cameron, 2004).

Sections 2 and 3 of this paper outline vessel scale and granule scale granulation models. In Section 4 we compare different ways of linking them.

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2. Vessel Scale Modelling: the Population Balance Equation

2.1 Conservation equations

The general modelling aim is to predict dynamic changes in the particle size distribution (PSD) along the length of the drum. The PSD is represented by the number density function n, where $n(v,z,t)\mathrm{d}v$ is the number of particles per unit bed volume with size in the range v to $v+\mathrm{d}v$ at axial position z at time t. Note that a particle's volume v is used as the measure of its size. The key assumptions at the vessel scale are

- the active mechanisms in the drum are agglomeration and solids transport;
- radial variations in the PSD are unimportant compared to axial changes in the drum.
- we neglect axial dispersion in the vessel for this study

Under these assumptions, the population balance for $n(v,z,t)$ is

$$\begin{aligned}\frac{\partial n(v)}{\partial t} &= -\frac{\partial}{\partial z}\big(n(v)u(v)\big) + \frac{1}{2}\int_0^v \beta(w,v-w)n(w)n(v-w)dw \\ &\qquad - \int_0^\infty \beta(v,w)n(v)n(w)dw\,,\end{aligned} \tag{1}$$

where u is the solids axial velocity and β is the agglomeration kernel (Iveson, 2002). We simplify eq. (1) by discretising both v and z. The particle size range is discretised by the popular method of Hounslow et al. (1988). This results in S geometrical size intervals such that $v_{i+1} = 2v_i$; v_1 is the lower bound of particle size in the first size interval. The drum is divided axially into M equally-spaced, well-mixed volumes. The discretised version of eq. (1) is

$$\begin{aligned}\frac{\mathrm{d}N_{i,k}}{\mathrm{d}t} &= N_{i-1,k}\sum_{j=1}^{i-2} 2^{j-i+1}\beta_{i-1,j,k}N_{j,k} + \frac{1}{2}\beta_{i-1,i-1,k}N_{i-1,k}^2 \\ &\quad - N_{i,k}\sum_{j=1}^{i-1} 2^{j-i}\beta_{i,j,k}N_{j,k} - N_{i,k}\sum_{j=i}^{S}\beta_{i,j,k}N_{j,k} \\ &\quad + \frac{Q_{i,k-1}}{V_k}\big(N_{i,k-1} - N_{i,k}\big), \qquad i = 1,\ldots,S;\ k = 1,\ldots,M.\end{aligned} \tag{2}$$

The number of particles per unit bed volume in size class i and axial position k is $N_{i,k}$, the fictitious quantity $N_{0,k} = 0$ and $Q_{i,k}$ is the volumetric flowrate of solids of size i leaving position k. The volumetric holdup of the kth solids bed V_k needs to be defined:

$$\frac{\mathrm{d}V_k}{\mathrm{d}t} = \sum_{i=1}^{S}\big(Q_{i,k-1} - Q_{i,k}\big), \qquad k = 1,\ldots,M. \tag{3}$$

Eqs (2)-(3) comprise the vessel scale conservation equations. Closure requires equations to relate the solids flowrate $Q_{i,k}$ and the agglomeration kernel $\beta_{i,j,k}$ to the granulator design, operating conditions, particle properties and the current PSD. Validated models on world-scale plants using this approach have been recently developed (Balliu 2004).

2.2 Constitutive equations

At this point, we have a choice: use existing constitutive relations for Q and β, or take a multiscale approach and obtain them from a suitable granule scale model.

There are reliable relations for the transport of solids through a rotating drum (Li et al., 2002), although the differential flow of differently-sized particles is not usually considered. Most often, the solids transport relations are simple functions of the local solids bed depth and other system parameters.

The agglomeration kernel is more troublesome. A variety of empirical and semi-theoretical relations are available (Iveson, 2002), but they all require fitting to the system of interest. Some progress can be made by factoring β into two terms:

$$\beta = C\psi. \tag{4}$$

Here, C is the collision rate coefficient and ψ is the probability that a collision results in agglomeration. Iveson (2002) lists several models for predicting whether a given granule pair will agglomerate or rebound on collision. Generally, they are functions of the sizes of the colliding granules, their relative impact speed and their mechanical properties. The key missing information is the frequency and relative velocity of granule collisions for the equipment design, operating conditions and PSD of interest. It can be provided by a model that predicts the granular motion inside the drum, for example a discrete element method (DEM) simulation.

Several choices for calculating Q, ψ and C are considered in Table 1. We selected option II because it uses a minimal DEM simulation—and DEM is very computationally intensive—to supply only that information not reliably available from existing constitutive relations.

Table 1. Options for supplying constitutive information to the population balance model.

Option	Information source [a]			Implications for DEM model
	Q	ψ	C	
I	CON	CON	CON	No DEM. PSD data required for parameter fitting.
II	CON	CON	DEM	DEM possibly 2D; simple contact force scheme ok to predict collision frequency and initial impact velocity.
III	CON	DEM	DEM	DEM possibly 2D; complex contact forces to account for rebound/agglomeration; must allow particle coalescence.
IV	DEM	DEM	DEM	As for III, but 3D DEM required for axial solids velocity.

[a] CON: vessel scale constitutive equation; DEM: granule scale DEM model.

3. Granule Scale Modelling: the Discrete Element Method

DEM simulations solve Newton's second law of motion to predict the trajectories of every particle in a particulate assembly (Mishra, 2003; Tijskens et al., 2003). The linear and angular momentum equations for granule ι are

$$m_\iota \frac{\mathrm{d}\mathbf{v}_\iota}{\mathrm{d}t} = \sum \mathbf{F}_\iota , \qquad I_\iota \frac{\mathrm{d}\omega_\iota}{\mathrm{d}t} = \sum T_\iota , \qquad \iota = 1,\ldots,P ; \tag{5}$$

where m_ι, $\mathbf{v}_\iota$, I_ι and ω_ι are respectively the granule's mass, velocity vector, moment of inertia and angular velocity. The sum of applied forces $\Sigma\mathbf{F}_\iota$ includes contributions from gravity, fluid drag and particle-particle and particle-wall contact forces, and so on; ΣT_ι is the associated moment sum. There are two basic DEM algorithms: the 'hard sphere' event driven method and the 'soft sphere' fixed time step approach. In practice, DEM implementation requires an effective integration scheme for eqs (5), efficient contact detection and the use of neighbour lists to reduce unnecessary contact detection. DEM simulations are computationally expensive with typical running times of hours to days, even weeks, to simulate seconds of real time.

In modelling granule scale behaviour in the granulator, our main assumptions are

- spherical granules, constrained to 2D motion in the plane of the drum cross section;
- the system is dense with multiple, lasting contacts, so we use the soft sphere method;
- only gravity and contact forces are important, fluid drag is negligible.

The contact force scheme recommended by Brendel and Dippel (1998) was used—a 'safe' linear spring and dashpot with limiting friction. The contact force parameters are estimated from data on the mechanical properties of wet granules.

4. Multiscale Integration and its Implications

Multiscale models can be classified according to how the constituent models at different scales are linked. Five broad *multiscale integration frameworks* may be distinguished: simultaneous, serial, multidomain, embedded and parallel (Ingram et al., 2004). No matter which framework is chosen, the same kind of information is transferred between the scales. Essentially, the granule scale model needs to supply the agglomeration kernel $\beta_{i,j,k}$ to the vessel scale model. To do so, the granule scale model requires the current PSD $N_{i,k}$ and the granule bed depth or V_k from the vessel scale. We discuss next how the multiscale integration framework influences the details of this information flow. Figure 1 depicts the various multiscale models.

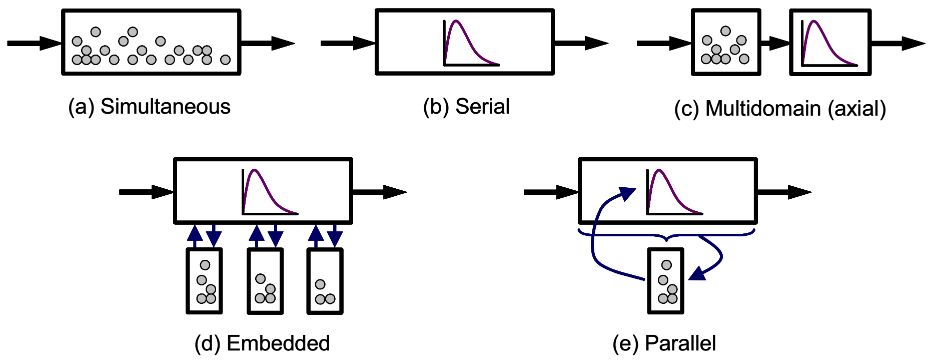

Figure 1. Alternative multiscale models for a drum granulator.

4.1 Simultaneous

Simultaneous integration corresponds to modelling the granulator entirely with DEM: the population balance is not used (Figure 1a). The PSD inside the granulator is calculated directly from granule scale data. Our choice of option II in Table 1 precludes simultaneous integration because no axial movement or agglomeration is taken into account. In practice, this option is currently infeasible because of the high computational burden of DEM. In the future, it could be used for detailed design work and to test the accuracy of other integration frameworks. Morton and Cleary (2002) modelled particle motion, but not agglomeration, in a variety of granulators using DEM.

4.2 Serial

One form of serial integration is to parameterise the DEM simulations—that is, to reduce them to a simple functional form that can be evaluated quickly. The DEM model becomes a constitutive equation for the population balance model (Figure 1b).

Since the PSD is often approximately log-normal, we calculate the geometric mean μ and standard deviation σ from $N_{i,k}$ and the bed fill fraction f from V_k, thereby reducing the number of inputs to the serial model from (S+1) to 3. The required number of granules P in the DEM simulation is estimated from $N_{i,k}$, f, the drum diameter and the expected bed porosity. The granule sizes D_l used in DEM are drawn from a log-normal distribution with parameters μ and σ. A series of DEM simulations is run offline. Statistics are collected to correlate the collision rate coefficient $C_{i,j}$, using the same size discretisation as in eq. (2), and the distribution of impact speeds f_{coll}. The kernel $\beta_{i,j}$ is calculated from eq. (4) using a weighted average agglomeration probability,

$$\psi_{i,j} = \int_0^{\max(v_{\mathrm{coll}})} \eta_{\mathrm{coll}}(v_{\mathrm{coll}}, i, j).f_{\mathrm{coll}}(v_{\mathrm{coll}}, i, j).\mathrm{d}\,v_{\mathrm{coll}} \;, \qquad (6)$$

where η_{coll} is the output (0 for rebound or 1 for coalescence) of a granule agglomeration model, such as one of those listed by Iveson (2002).

Serial integration offers the fastest simulations by far, but at the expense of flexibility. It relies heavily on the quality and scope of the parameterisation. Serial integration is the most feasible option for online control and fault diagnosis applications. Tan et al. (2004) used another form of serial integration of their DEM results to develop an agglomeration kernel for fluid bed granulators.

4.3 Multidomain

In a multidomain simulation, part of the drum is modelled using DEM while the population balance alone describes the remainder. Two possibilities for dividing the drum are axial division and division across the cross-section. The former may reflect the change in dominant processes through the granulator: nucleation, growth and agglomeration (Figure 1c). The latter reflects the presence of stagnant zones. McCarthy and Ottino (1998) report a hybrid DEM technique that took advantage of a stagnant zone to study powder mixing in a rotating drum. Option II does not allow multidomain integration for the same reasons as mentioned in Section 4.1. If option IV were used with axial division, DEM could be used in the first part of the drum. At the end of the DEM section, the PSD, solids flowrate and agglomeration kernel—$N_{i,0}$, Q_0 and $\beta_{i,j,k}$—are calculated from the particle trajectories for use in the population balance section.

Multidomain integration for granulator modelling relies strongly on insight into the solids flow pattern and the granulation regime. It is a useful bridge between full DEM (Section 4.1) and using the population balance exclusively (Section 4.2). Still, the computational load remains high. Multidomain integration could be used in design and optimisation where some local detail is needed, perhaps near sprays or drum internals.

4.4 Embedded

In the embedded framework the DEM model is invoked by the population balance at each position k to supply the agglomeration kernel using eqs (4) and (6) as in serial integration, but this time in an online manner. Option II (Table 1) works well here. Instead of parameterising the size distribution into μ and σ, the full PSD $N_{i,k}$ is supplied by the population balance to generate the DEM particle sizes D_ι (Figure 1d). It is analogous to the method of Laso et al. (1997), which invokes a molecular level model to supply the shear stress–shear rate relationship on the fly to a continuum fluid mechanics model of polymer flow.

Embedded integration is potentially suitable for design and optimisation applications. It can use a minimal DEM model, but is still computationally intensive.

4.5 Parallel

One implementation of parallel integration involves using a single instance of the DEM model to supply the agglomeration kernel for every position k in the population balance (Figure 1e). Clearly, this is an approximate approach. The population balance could provide the DEM with the size distribution and fill fraction averaged over the length of the drum, $N_{i,\mathrm{avg}}$ and f_{avg}. Another variation on parallel integration was discussed by Bauer and Eigenberger (1999) for modelling bubble column reactors by combining computational fluid dynamics (CFD) with a more conventional reactor model that includes a total number balance for the bubbles.

Parallel integration makes the smallest demands of a coupled DEM model. Consequently, it runs faster than the other methods, aside from serial integration.

5. Conclusions

Particulate systems present a diversity of challenges in modelling and simulation for the CAPE community. We have explored the formulation of alternative multiscale models for drum granulation. The chosen *multiscale integration framework* influences the inter-scale flow of information and the suitability of the multiscale model for different applications, such as design, optimisation and control. We have not touched on the further refinements, combinations and analysis of the frameworks.

References

Balliu, N., 2004, An object oriented approach to the modelling and dynamics of granulation circuits, PhD Thesis, School of Engineering, The University of Queensland, Australia.

Bauer, M. and G. Eigenberger, 1999, Chem. Eng. Sci., 54, 5109.

Brendel, L. and S. Dippel, 1998, NATO ASI Series, Series E, 350, 313.

Ennis, B.J., 1997, in Powders & Grains 97, R.P. Behringer and J.T. Jenkins Eds., A.A. Balkema, Rotterdam, 13–23.

Hounslow, M.J., R.L. Ryall and V.R. Marshall, AIChE J., 34, 1821.

Ingram, G.D. and I.T. Cameron, 2004, Dev. Chem. Eng. Mineral Process., 12, 293.

Ingram, G.D., I.T. Cameron and K.M. Hangos, 2004, Chem. Eng. Sci., 59, 2171.

Iveson, S.M., 2002, Powder Technol., 124, 219.

Laso, M., M. Picasso and H.C. Öttinger, 1997, AIChE J., 43, 877.

Li, S.-Q., Y. Chi, R.-D. Li, J.-H. Yan and K.-F. Cen, 2002, Powder Technol., 126, 228.

McCarthy, J.J. and J.M. Ottino, 1998, Powder Technol., 97, 91.

Morton, D. and P.W. Cleary, 2002, Geotechnical Special Publication, 117, 397.

Tan, H.S., M.J.V. Goldschmidt, R. Boerefijn, M.J. Hounslow, A.D. Salman and J.A.M. Kuipers, 2004, Powder Technol., 142, 103.

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