ELSEVIER

Powder Technology 138 (2003) 39–45

POWDER TECHNOLOGY

www.elsevier.com/locate/powtec

Discrete element modelling of fluidised bed spray granulation

M.J.V. Goldschmidt[a,b], G.G.C. Weijers[a], R. Boerefijn[c], J.A.M. Kuipers[a,*]

[a] Twente University, P.O. Box 217, Enschede 7500 AE, The Netherlands
[b] Akzo Nobel Chemicals Research, P.O. Box 9300, Arnhem 6800 SB, The Netherlands
[c] Unilever Research, P.O. Box 114, Vlaardingen 3130 AC, The Netherlands

Abstract

A novel discrete element spray granulation model capturing the key features of fluidised bed hydrodynamics, liquid–solid contacting and agglomeration is presented. The model computes the motion of every individual particle and droplet in the system, considering the gas phase as a continuum. Microscale processes such as particle–particle collisions, droplet–particle coalescence and agglomeration are directly taken into account by simple closure models. Simulations of the hydrodynamic behaviour of a batch granulation process are presented to demonstrate the potential of the model for creating insight into the influence of several key process conditions such as fluidisation velocity, spray rate and spray pattern on powder product characteristics.

Keywords: Fluidised bed; Granulation; Agglomeration

1. Introduction

Fluidised bed granulation is an important powder production process with several key advantages compared to other powder production processes, such as spray drying or high-shear granulation. Reviews signifying the interest of this type of granulation process are given by Kristensen and Schaeffer [1], Banks and Aulton [2] and Nienow [3]. The most characteristic and essential part of spray granulation is the wetting of particles by an atomised liquid feed (solution, suspension or melt) and the induced growth of particles in a gas-fluidised bed. Thorough understanding of the mechanisms prevailing in the process is a prerequisite for obtaining proper control over powder properties. Mechanisms of granulation are often distinguished as wetting, progressive growth (agglomeration or layering), breakage and consolidation. It is virtually impossible to expect that these mechanisms will occur singly or sequentially and many granulator design and operation parameters will influence them. Population balance models have been used successfully to provide insight into the mechanisms by which particles grow [4–6]. However, since powder characteristics and essential hydrodynamic parameters regarding liquid–solid contacting, particle mixing and segregation are lumped into the kinetic rate constants, population balance models can not be applied for a-priori design and scale-up of fluid bed granulation processes.

An in depth discussion on hydrodynamic modelling of fluidised bed spray granulation processes has recently been presented by Goldschmidt [7]. Continuum models, in which all phases are considered as interpenetrating continua, constitute the most appropriate choice for hydrodynamic modelling of engineering scale systems. However, these models require constitutive relations to take particle–particle collisions, droplet–particle coalescence and granulation kinetics into account and little is known about the efficiencies of particle wetting and granule formation, required to describe the granulation process within a continuum framework. In this respect, more detailed discrete element models can be applied as a valuable research tool to gain more insight and test continuum theories describing these processes. In this paper, some of the first results obtained with a two-dimensional discrete element spray granulation model will be presented. The mechanism by which granules grow and the observed influences of process conditions on powder characteristics will be discussed.

2. Discrete element model

The discrete element spray granulation model is based on the hard-sphere discrete particle model for gas-fluidised

* Corresponding author. Twente University, P.O. Box 217, Enschede 7500 AE, The Netherlands.
E-mail address: j.a.m.kuipers@ct.utwente.nl (J.A.M. Kuipers).

0032-5910/$ - see front matter © 2003 Elsevier B.V. All rights reserved.
doi:10.1016/j.powtec.2003.08.045

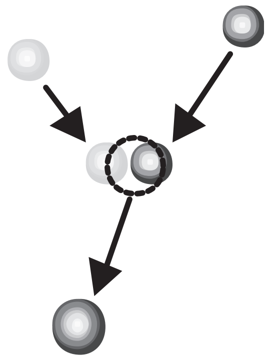

Fig. 1. Repositioning and merging of particles upon coalescence or agglomeration.

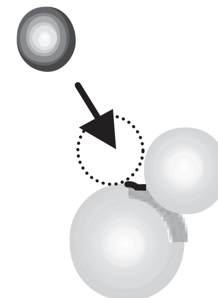

Fig. 3. Masking of wetted surface for subsequent agglomerations.

beds, originally developed by Hoomans [9] and Hoomans et al. [8]. The model computes the motion of every individual particle and droplet from the Newtonian equations of motion, whereby the gas phase is considered as a continuum. The gas flow field at sub-particle level is not resolved and empirical relations by Ergun [10] and Wen and Yu [11] are applied to describe gas–particle and gas–droplet drag. Particle–particle collisions, particle–droplet coalescence and agglomeration are directly taken into account by simple closure models. It should thereby be noted that particle and droplet rotation is not taken into account in the present version of the model. The discrete element model essentially processes a sequence of encounters in which all particles and droplets are moved under the influence of external forces until the next encounter occurs. A detailed description of the hard-sphere discrete particle model has been given by Hoomans [9] for gas-fluidised beds. In this paper, we will briefly comment on the extensions needed to describe fluidised bed spray granulation processes, whereas an in-depth discussion of the applied closure models is given by Goldschmidt [7].

The discrete element spray granulation model basically distinguishes between three different types of entities: dry particles, wetted particles and droplets. All three are assumed to be spherical and encounters are detected as soon as contact occurs at a point on the line joining the centres of two entities. The following types of encounters are distinguished:

1. Encounters among dry particles, described by hard-sphere collision laws;
2. Droplet–droplet encounters, described by hard-sphere collision laws since they are assumed to be repulsive for atomised liquid droplets with a typical radius of 50 μm, colliding with small mutual velocity differences;
3. Encounters between droplets and dry or wet particles, described as coalescense;
4. Encounters between a wetted particle and another particle, leading to either rebound described by hard-sphere collision laws or agglomeration. Which of the two occurs depends on the odds off the particles hitting each other on a wet spot;

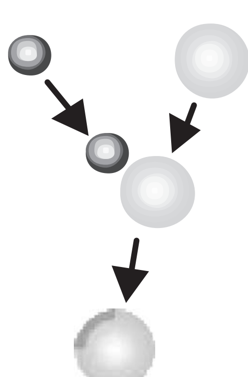

Fig. 2. Liquid layer formation upon coalescence.

Fig. 4. Initial situation for base case.

Table 1
Simulation settings for base case

System geometry (2D)		*Operation conditions*	
Granulator width	0.20 m	Number of primary particles	50,000
Granulator height	0.50 m	Fluidisation velocity	1.6 m/s
Number of grid cells in horizontal direction	41	Droplet spray rate	$2.77 \cdot 10^{-6}$ kg/s
Number of grid cells in vertical direction	101	Droplet injection velocity	−40 m/s
Horizontal position of spray nozzle	0.10 m	Standard deviation in radial direction	0.5 m/s
Vertical position of spray nozzle	0.25 m	Runtime	10 s
Width of nozzle	0.005 m	Final liquid/solid ratio	0.03
Gas phase properties		*Spray properties*	
Freeboard pressure	101,325 Pa	Mean droplet diameter	100 μm
Gas phase temperature	313 K	Standard deviation of size distribution	50 μm
Gas phase shear viscosity	$1.8 \cdot 10^{-5}$ Pa·s	Binder density	1228.0 kg/m^3
Heat conductivity	$2.883 \cdot 10^{-2}$ W/m·K	Coefficient of normal restitution	0.50
Particle properties		Minimum liquid layer thickness	20 μm
Mean diameter of primary particles	250 μm	Binder injection temperature	353.0 K
Standard deviation of size distribution	50 μm	Binder melting point	328.0 K
Particle density	2440 kg/m^3	Heat of fusion+cooling enthalpy	227.6 J/g
Coefficient of normal restitution	0.97		

5. Encounters between dry or wet particles and a wall, described by hard-sphere collision laws;
6. Encounters between droplets and walls, resulting in removal of the droplet from the simulation.

Upon coalescence or agglomeration, a new particle entity is formed at the position of the centre of mass of the original entities (Fig. 1). Mass, momentum and volume of the original entities are conserved and transferred to the newly formed particle, where after the original entities are excluded from the simulation. In case of coalescence, the area on the newly formed particle covered by liquid depends on the original particle size, the size of the droplet and a defined minimum liquid layer thickness (Fig. 2). In case of agglomeration, the wetted area available for subsequent agglomerations is reduced by the projected area of the smallest particle, to account for bridge formation and the masking of wetted surface, which cannot be reached anymore because the newly agglomerated particle is in the way (Fig. 3). Furthermore, for agglomerates containing more than three primary particles, inclusion of liquid and gas inside the pores is taken into account.

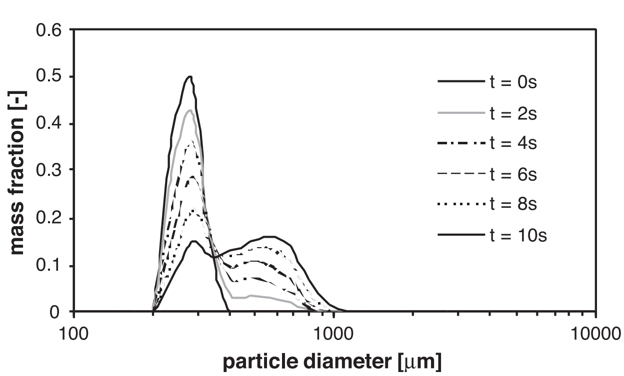

Fig. 5. Evolution of PSD for base case.

To simulate a single phase spray nozzle, one or more cells of the computational grid, applied to calculate the gas phase flow field, can be appointed as liquid injection points. New droplets are introduced at the bottom of these cells at regular intervals to maintain a specified liquid mass flow rate. All droplets are given the same initial axial velocity in the downward direction, whereas the radial velocity is imposed according to a Gaussian distribution. The standard deviation of this Gaussian distribution can be varied to modify the width of the spray cone. Furthermore, all droplets are randomly distributed over the bottom of the injection cells, whereby a log-normal droplet size distribution is applied.

The melted liquid binder is injected at a temperature just above its melting point, whereas the temperature of the fluidising gas is below the melting point of the binder. Therefore, the liquid will coagulate after a (short) while, either as a droplet, as a liquid layer on the outside of a particle or as a solid bridge between two or more particles within an agglomerate. At the time of injection the coagulation time (t_{coag}) is determined for every droplet from a

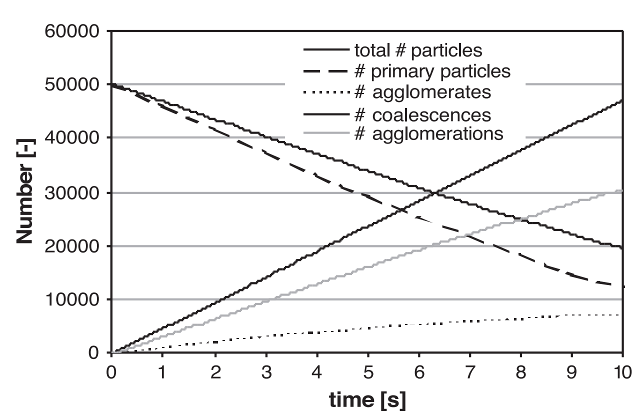

Fig. 6. Monitoring of key numbers for base case.

simple heat balance. From the moment of injection, t_{coag} is continuously counting down. When t_{coag} reaches zero, the droplet (or liquid layer) has fully coagulated and the droplet (or particle) is treated as a dry particle. When a droplet and a wetted particle coalesce or when two particles agglomerate, the coagulation time of the newly

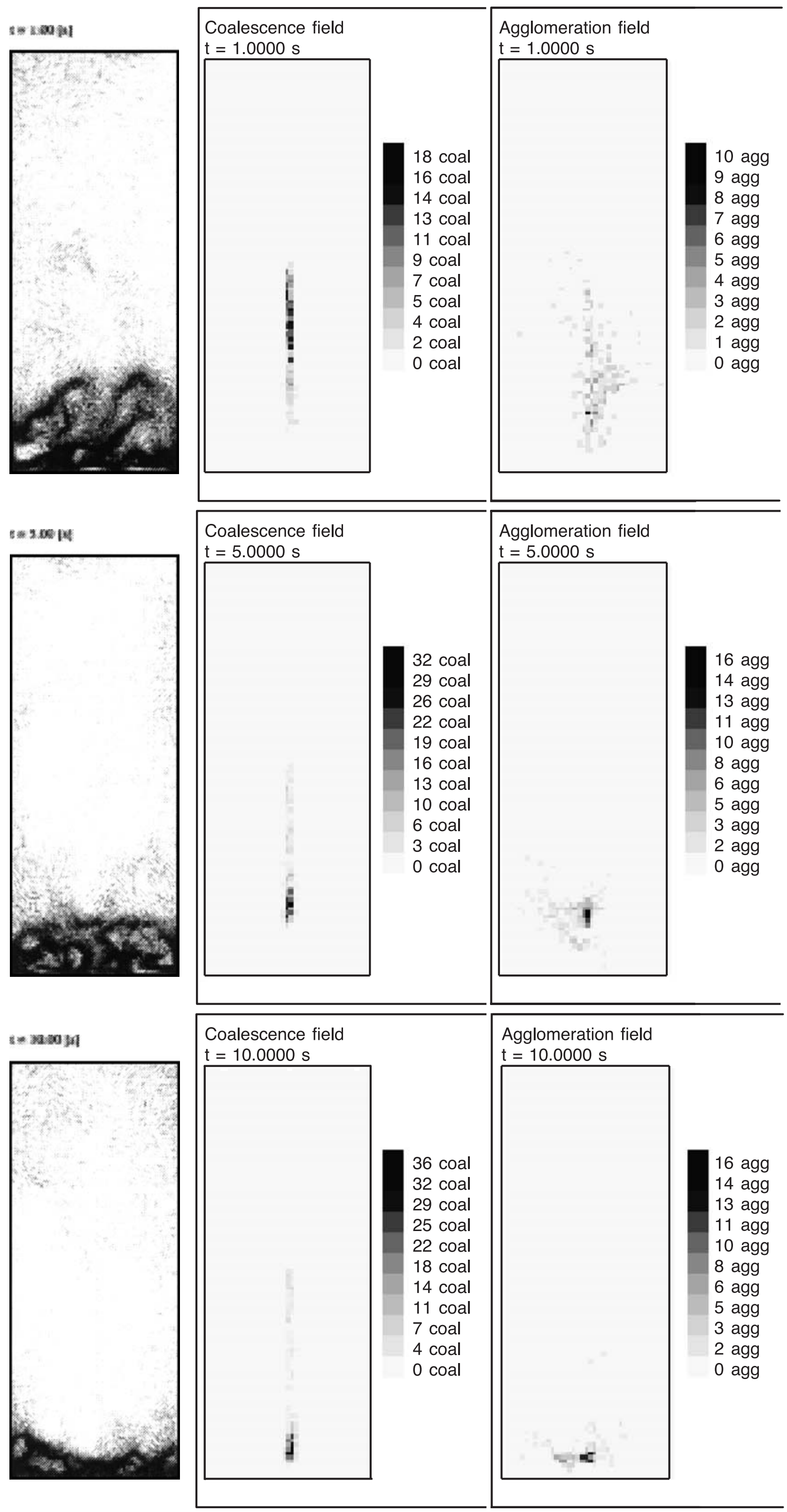

Fig. 7. Bed structure, number of coalescences (coal) and agglomerations (agg) for base case.

formed particle is set to the liquid volume weighted average coagulation time.

3. Spray granulation simulations

To study the influence of several key operating conditions on the spray granulation process, two-dimensional simulations of a small laboratory scale batch spray granulator have been performed. The initial turbulent fluidised bed for the base case is shown in Fig. 4. As the simulation starts, droplets of melted binder are sprayed onto the bed from a centrally positioned single phase nozzle. In the base case simulation, the spray pattern is relatively narrow to simulate a nozzle with a flat spray pattern. An extensive overview of all simulation conditions applied in this work for the base case simulation is presented in Table 1, the chosen properties essentially equal those of glass ballotini and PEG, for purpose of future validation.

3.1. Results of base case

The evolution of the particle size distribution (PSD) that results from the base case simulation described in the previous paragraph is presented in Fig. 5. To obtain this figure, the particle size distribution was divided into discrete size classes compliant with a $\sqrt[4]{2}$ sieve set. Fig. 6 shows the development of the number of primaries and granules during the simulation. Further, the total number of agglomerations and coalescences are shown. The smooth evolution of the particle size distribution and the straightness of the lines representing the decrease of the number of primary particles and the number of agglomerations indicate that layering is the prevailed granule growth mechanism in this simulation. The straight line for the number of coalescences is a result of the constant flow of droplets into the system, where the ratio of the number of coalescences to the number of agglomerations shows that typically 1.55 droplets are needed for every agglomeration.

To gain insight in the fluidised bed structure and the regions where droplet–particle coalescence and agglomeration take place, Fig. 7 shows some 'snapshots' of bed structures, coalescence and agglomeration fields. The coalescence and agglomeration fields represent the number of coalescences and agglomerations for each computational cell during a 0.1-s interval just before the presented snapshots of the bed structure were taken. These snapshots show how the bed height decreases during the simulation. As a consequence of the increase of the average particle diameter, the fluidised bed dynamics shifts from typical turbulent fluidisation behaviour to the behaviour of a violently bubbling shallow gas-fluidised bed. The coalescence fields show that droplet–particle coalescence initially takes place in the freeboard, whereas as the bed quiets down and the bed height decreases the area where most coalescence takes place shifts more towards the top of the dense bed. Although the spray influences the fluidised bed dynamics (it directs particles present in the spray region back into the bed and sometimes stifles eruption of bubbles), severe penetration of the fluidised bed by droplets is not observed. As shown in Fig. 7, agglomeration exclusively takes place in the freeboard near the top of the fluidised bed during the entire process.

The observed segregation pattern confirms a layered growth mechanism. Besides the fact that primary particles by far have the highest number density in the spray granulation system, coalescence of droplets with primaries is favoured by the segregation pattern. Therefore most binder droplets coalesce with primary particles in the spray zone. These primary particles subsequently agglomerate with the first particle that they have a successful encounter with on a wetted spot, which causes a gradual particle growth by layering. Since particles are directed back into the bed where the encounter frequency is high compared to the coagulation time of the binder, most binder is encapsulated inside granules by agglomeration before it solidifies. In experimental systems, granules will therefore deform and/or break before reaching their final composition and shape. These two effects are not taken into account in the (current) model, though they will certainly influence the particle size distribution and granule composition.

3.2. Influence of spray rate

The influence of the liquid spray rate on the powder characteristics was studied with two simulations. In the 'fast spray' simulation, the spray flux was doubled compared to the 'base case', whereas in the 'slow spray' simulation the spray rate was halved. In order to retain the same final liquid/solid ratio, the runtimes were respectively halved and doubled. Fig. 8 shows the particle size distribution of the granulation product. Interestingly, a monomodal sizes distribution results from the slow spray case, whereas a bimodal distribution is obtained in the fast spray case. The figure shows that a higher conversion of the primary particles into granules is obtained at low spray rates, whereas the granules that are formed are bigger at high spray rates. Thus, the modelling results are consistent with experimental results [1,6], showing that the granule size is proportional to the

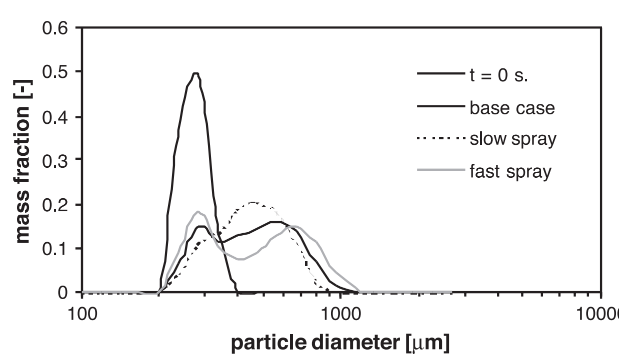

Fig. 8. Influence of spray rate on final PSD.

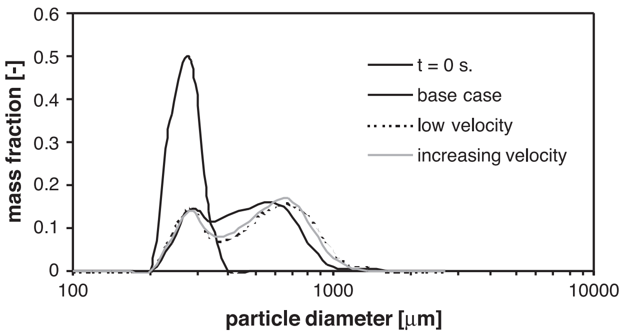

Fig. 9. Influence of fluidisation velocity on final PSD.

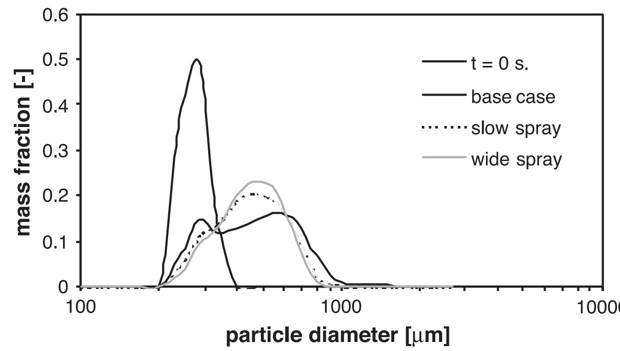

Fig. 11. Influence of spray pattern on final PSD.

spray rate. At low spray rates, particles pick up less liquid while they are in the wetting zone where coalescence and agglomeration mainly take place. Consequently, less agglomerations are required to cover all wetted particle area and agglomerates grow more gradually. Furthermore, the 'slow spray' simulation takes twice as long as the 'base case', so there is more opportunity for primary particles in the top of the bed to descend and participate in the granulation process.

3.3. Influence of fluidisation velocity

In a fluidised bed, the gas velocity has an pronounced influence on the fluidised bed behaviour and is therefore expected to influence the granulation process. To study the influence of the gas velocity on the granulation process, two simulations at different homogeneous fluidisation velocities were performed. In the 'low velocity' simulation, a constant fluidisation velocity of 1.0 m/s is applied (1.6 m/s in the 'base case'), whereas in the 'increasing velocity' simulation the gas velocity is linearly increased from 0.6 m/s at the beginning of the simulation to 1.6 m/s at the end. In both simulations, the bed was operated in the bubbling regime during the entire granulation process. The resulting particle size distributions are shown in Fig. 9. The particle size distributions obtained in both simulations are quite similar, but they differ significantly from the 'base case'. About the same amount of primary particles seems to be converted into granules, but the size of the produced granules is bigger. This can be explained by reduced mixing of the bed at lower gas velocities, which causes the granule residence time in the wetting zone to rise and consequently bigger granules to be formed.

3.4. Influence of spray pattern

The influence of the spray pattern on the agglomeration process is examined with a 'wide spray' simulation. In this simulation, the standard deviation of the initial radial droplet velocity is set to 5.0 m/s, 10 times higher than the standard deviation in the base case. Fig. 10 shows some typical snapshots of the bed structure, the spray cone and the droplet–particle coalescence field. Just as in the base case liquid–particle contacting and agglomeration basically take place in the freeboard, just above the top of the bed. The liquid however is spread over a much bigger area, which

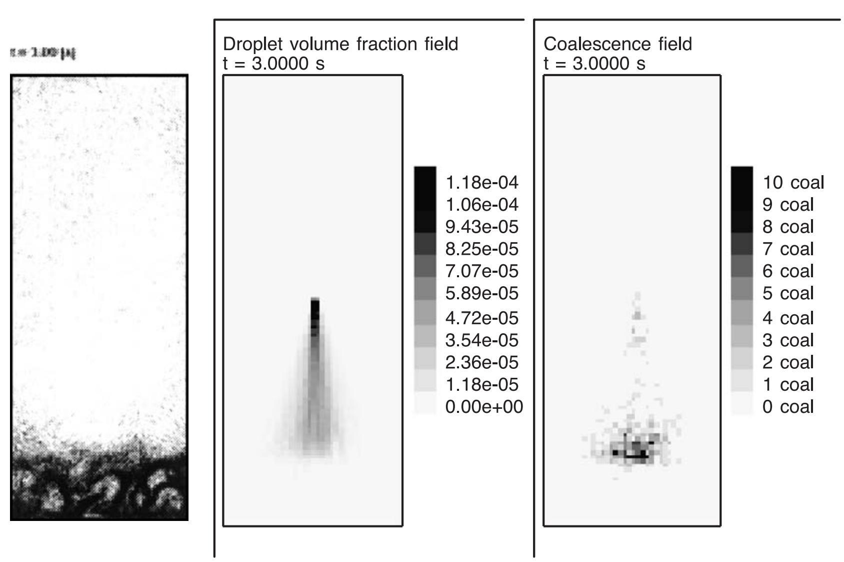

Fig. 10. Snapshot of bed structure, droplet vol. fraction and coalescence field for wide spray case.

makes that the particles pick up less liquid before they coalesce and causes a growth pattern that is quite similar to that obtained in the 'slow spray' simulation, as shown in Fig. 11. In the 'wide spray' case, the primary particles are more involved in the granulation process because elutriation is harder through the wider spray zone.

4. Discussion

The presented discrete element model contains simple closures to describe particle–droplet coalescence and agglomeration. Also the model is limited to a 2D Cartesian geometry and the number of particles is much lower than in systems of experimental relevance. Obviously, the presented model should be regarded as a proof of concept and further research is required to incorporate more detailed closure relations giving a more realistic description of coalescence, liquid spreading, agglomerate (de)formation, breakage of agglomerates, droplets, etc. Though expansion to 3D and more complex geometries is readily possible, the number of particles that can be handled by discrete element codes will limit the application of the model to small laboratory scale systems. Therefore, in respect of a multi-level modelling strategy, the discrete element model can be applied as a valuable learning tool to gain more insight into particle growth kernels and liquid–solid contacting efficiencies that are required to describe granulation processes in engineering scale models. The model can also be a useful tool to provide more detailed contact mechanical models with typical droplet–particle and particle–particle encounter characteristics, such as the impact angles and velocities, which are hard to obtain from experiments. However, validation of the discrete element spray granulation model with dedicated experiments is required in the near future, to gain more trust in the model predictions and indicate which improvements need to be made to the model.

5. Conclusion

A novel discrete element spray granulation model capturing the key features of fluidised bed hydrodynamics, liquid–solid contacting and agglomeration has been presented. Simulations of a batch granulation process containing 50,000 primary particles correctly predict the experimentally observed increase in granule size as the binder flow rate increases. Further, significant effects of the spray pattern and the fluidisation velocity are observed, which can be explained from the perspectives of liquid–solid contacting and bed mixing. In all simulations, droplet–particle coalescence and agglomeration took place in the freeboard and at the top of the bed throughout the batch runs. Particle growth was dominated by layering, whereas agglomeration of equally sized particles and bed penetration by droplets or wetted particles rarely occurred. Severe size segregation was observed. Big granules mainly remained at the bottom of the bed while above the spray nozzle mainly primary particles were detected. Furthermore, a low particle concentration was observed in the spray zone, because particles are directed back into the bed as soon as a droplet hits them. Clearly, the explanation of the different product characteristics from a hydrodynamic point of view can contribute to better understanding of spray granulation processes, where a mixture of apparatus design, operation conditions and physical properties of binder and primary particles determines the granular product characteristics.

References

[1] H.G. Kristensen, T. Schaeffer, Granulation—a review on pharmaceutical wet-granulation, Drug Dev. Ind. Pharm. 13 (1987) 803–872.

[2] M. Banks, M.E. Aulton, Fluidized bed granulation: a chronology, Drug Dev. Ind. Pharm. 17 (1991) 1437–1463.

[3] A.W. Nienow, Fluidised bed granulation and coating: applications to materials, agriculture and biotechnology, Chem. Eng. Commun. 139 (1995) 233–253.

[4] B. Waldie, D. Wilkinson, L. Zachra, Kinetics and mechanisms of growth in batch and continuous fluidized bed granulation, Chem. Eng. Sci. 42 (1987) 653–665.

[5] M.J. Hounslow, The population balance as a tool for understanding particle rate processes, KONA Powder Particle 16 (1998) 179–193.

[6] R. Boerefijn, M. Buscan, M.J. Hounslow, Effects of non-ideal powder properties on granulation kinetics, in: M. Kwauk, J. Li, W.-C. Yang (Eds.), Fluidization X, Engineering Foundation, New York, 2001, pp. 629–636.

[7] M.J.V. Goldschmidt, Hydrodynamic modelling of fluidised bed spray granulation, PhD Thesis, Twente University, Enschede, The Netherlands, 2001.

[8] B.P.B. Hoomans, J.A.M. Kuipers, W.J. Briels, W.P.M. Van Swaaij, Discrete particle simulation of bubble and slug formation in a two-dimensional gas-fluidised bed: a hard sphere approach, Chem. Eng. Sci. 51 (1996) 99–118.

[9] B.P.B. Hoomans, Granular dynamics of gas–solid two-phase flows, PhD Thesis, Twente University, The Netherlands, 2000.

[10] S. Ergun, Fluid flow through packed columns, Chem. Eng. Prog. 48 (1952) 89–94.

[11] Y.C Wen, Y.H Yu, Mechanics of fluidization, Chem. Eng. Prog. Symp. Ser. 62 (1966) 100–111.