Refereed Proceedings

The 12th International Conference on Fluidization - New Horizons in Fluidization Engineering

Engineering Conferences International *Year* 2007

Four Approaches to Structure Gas-Solid Fluidized Beds

J. Ruud van Ommen* John Nijenhuis†

Marc-Olivier Coppens‡

*Delft University of Technology, j.r.vanommen@tudelft.nl

†Delft University of Technology, j.nijenhuis@tudelft.nl

‡Delft University of Technology, M.O.Coppens@tudelft.nl

FOUR APPROACHES TO STRUCTURE GAS-SOLID FLUIDIZED BEDS

J. Ruud van Ommen, John Nijenhuis, and Marc-Olivier Coppens
Delft University of Technology - DelftChemTech
Julianalaan 136, 2628 BL Delft, The Netherlands
T: +3115 278 2133; F: +3115 278 5006; E: j.r.vanommen@tudelft.nl

ABSTRACT

Structuring fluidized beds can facilitate scale-up and increase conversion and selectivity by controlling the bubble size. We present four approaches to structure fluidized beds: oscillating the gas flow, distributing the gas injection, imposing an electric field to induce interparticle forces, and optimising distributed particle properties such as the size.

INTRODUCTION

Fluidized beds are used for several catalysed gas-phase reactions, especially for highly exothermal reactions and processes with regular catalyst replacement. However, they have serious drawbacks, the two most important being the difficult scale-up and the inefficient use of reactant gas due to the formation of bubbles. The rather small particles that are used in fluidized beds almost always ensure the absence of mass transfer limitations on the particle scale, but on a larger scale mass transfer limitations are present in most fluidized bed processes, especially in the bubbling regime but also in the turbulent regime. This leads to large differences between reactant concentrations in the dense and dilute phases, resulting in a lower conversion and selectivity.

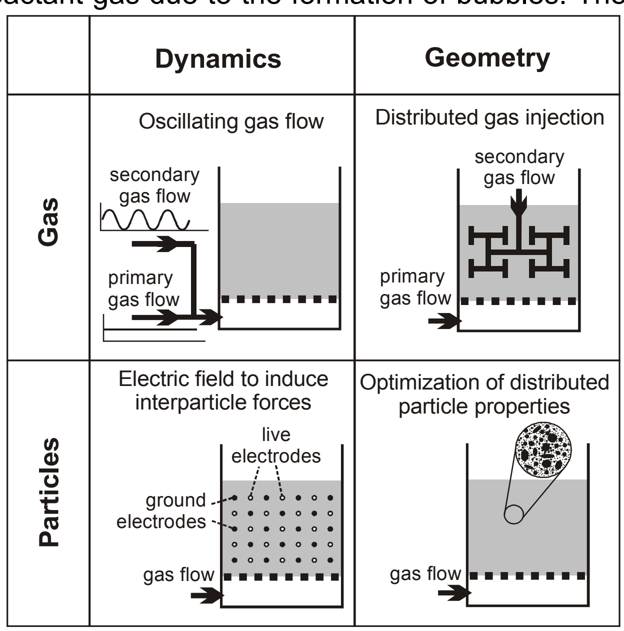


Figure 1. Overview of four approaches to introduce structure in gas-solid fluidized

In this paper, we present four ways to manipulate the hydrodynamics of fluidized bed in order to introduce more structure and to reduce the bubble size. This will lead to a simplified scale-up and an increased interphase mass transfer, typically leading to better conversion and selectivity. This can either be done by (1) modifying the gas supply or (2) interfering in the particle phase. In both cases, either the dynamics can be changed or the configuration can be altered. This gives in total four different possibilities (see Fig. 1), which will be illustrated in this paper:

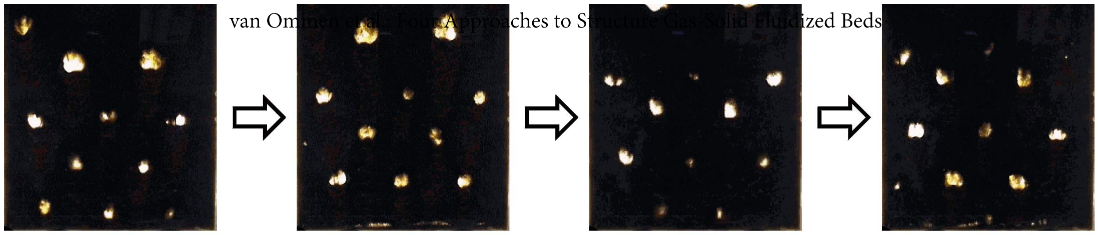
1.a. Oscillating the gas supply.
1.b. Distributing the gas supply over the height of the bed.
2.a. Varying the interparticle forces using electric fields.
2.b. Varying the particle size distribution and other distributed particle properties.

DYNAMIC OPERATION OF THE GAS SUPPLY

Conventionally, the gas flow to a fluidized bed is kept at a constant value; this value is only adjusted now and then, *e.g.*, to change the production level. However, by continuously varying the instantaneous gas flow rate around a given average value additional degrees of freedom are obtained. One approach is to apply feedback control: based on one or more measurements of the fluidized bed behaviour, the gas flow rate could be adapted in order to maintain a desired state. Some first steps in this direction have been made (1,2,3), but it seems to be a long way before this approach could be applied to large-scale installations. An alternative approach is to apply a periodically varying gas flow without feedback control. It is known that pulsing the gas (4,5) – as well as oscillating the distributor plate (6) – can cause considerable changes in the fluid bed hydrodynamics and significantly improve reactor performance (7). Furthermore, chaotic and other strongly non-linear systems may turn periodic by oscillating a characteristic (order) parameter. This explains ripples on sand beaches, and many other regular patterns seen in nature.

Coppens *et al.* (8) showed that by oscillating the gas flow introduced through the porous bottom distributor plate, bubble patterns in fluidized beds may indeed become ordered and periodic. Experiments were first carried out in a quasi-two-dimensional bed. A sinusoidally oscillating gas flow was added on to a constant primary gas flow above minimum fluidization, so that the total gas flow would remain above minimum fluidization. For an air-sand system, regular bubble patterns were observed within a broad range of frequencies (from 2.5 to 7 Hz) and amplitudes (an oscillating component of 0.2 to 0.7 times the gas flow required for minimum fluidization). These patterns are hexagonal: Bubbles rise in ordered rows with constant inter-bubble distance, with each row horizontally shifted with respect to the previous row by half the inter-bubble distance (Fig. 2). Above a certain height, the regularity of the patterns is destroyed, as fluctuations in the system start to dominate the hydrodynamics.

For the above quasi-2D air-fluidized bed of sand particles, the bubble pattern is regular to a height that is approximately equal to the bed width; the wider the bed, the less the left and right wall influence the pattern, and the greater the height over which the pattern formation persists. The large front and back wall help in stabilizing the patterns in quasi-2D fluidized beds. Wang and Rhodes (9) also observed regular bubble pattern in discrete particle simulations, but not as clear as in the experimental systems. Coppens *et al.* (8) observed regular bubble patterns in experiments in 3D cylindrical beds too, but only for bed heights of a few centimetres. These patterns are similar to the patterns seen in even shallower, vibrated granular layers (10). Up to now, we have not yet succeeded to stabilise such patterns for higher beds.



(a) (b) (c) (d)

Figure 2. Regular bubble patterns obtained in a quasi-2D bed of 40 cm wide and 43 cm high. The airflow is oscillated at a frequency *f* = 3.5 Hz. The constant component of the gas flow is equal to the minimum fluidization gas flow; the amplitude of the oscillating component is half of that. The sequence shows 4 snapshots out of one period of the bubble pattern (frequency 3.5/2=1.75 Hz).

It is important to note that both for 2D and 3D systems the waves are not a simple linear resonance phenomenon: the pattern wavelength is not inversely proportional to the driving frequency, while the pattern is formed in a range of frequencies and not at specific frequencies. Also, the ordered patterns in fluidized beds are propagated upwards via the rising gas flow, which differentiates these patterns from those observed in vibrated granular layers, where all the energy is transmitted to the particles via the moving bottom plate. As a result, dissipation is much stronger in vibrated granular matter than it is in gas-solid fluidized beds, where it is possible to influence the entire bed dynamics via a change in inlet gas dynamics. Further research is needed to apply this approach to deep 3D beds and to optimise the dynamic structuring such that a significant reduction of bubble size is reached.

DISTRIBUTING THE GAS SUPPLY OVER THE HEIGHT OF THE BED

An alternative way to obtain more degrees of freedom in operating a fluidized bed is to vary the gas injection in space instead of in time. In a fluidized bed, the gas is normally fed via the bottom, but examples of distributed gas supply do exist. In combustion, staged injection of the air can be used to reduce NO_x emission (*e.g.*, 11,12). Moreover, the application of membranes in fluidized beds to supply or remove gas has been studied (*e.g.*, 13,14).

To distribute the gas over the bed, Coppens (15) proposed to connect all injection points by a hierarchical, tree-like fractal structure. Gas flows from the stem of this tree to all branch tips, spread out over the reactor at optimised locations, where it exits. Via the bottom plate, enough gas is fed to ensure at least minimum fluidization throughout the bed. One important reason to use a fractal design is its intrinsic scalability, mimicking nature. When scaling up the tree-like injector, new branching generations are added to serve larger reactor volumes. The fractal injector branches in such a way that the length and diameter of branches of a given generation is the same. In this way, fluid leaves all outlets, however many there may be, at the same flow rate, because the hydraulic path lengths or pressure drops from the inlet to all outlets are all the same. For outlets lying in the same horizontal plane, this avoids radial non-uniformity. In the vertical direction, by spacing the outlets according to a designated pattern, one could compensate for the axial gradients in gas amounts and reactant concentrations. Moreover, the bubble size can be controlled, as less primary gas leads to smaller bubbles initially, while fresh feed blown into the reactor at various locations tends to break up existing bubbles or blow particles apart, leading to an emulsion phase of higher void fraction (16).

Experiments to study the effect of secondary injection using a fractal injector are carried out in a quasi-2D Plexiglas column, 20x1.5x80 cm. The column was operated within a cabinet controlled at 30ºC. The bed material consisted of mono-disperse glass beads, with a diameter of 550 µm and a density of 2400kg/m^3 (Geldart B; u_{mf}=0.21 m/s). The settled bed height was 40 cm. The fractal injector has 16 injection points (similar to the schematic in Fig. 1); the lowest row at 6 cm above the bottom plate and the highest row at 14 cm. Pressure fluctuations were measured using piezo-electric pressure transducers, Kistler type 7261, at 20 cm above the sintered metal porous distributor and in the plenum. The pressure fluctuations were measured with a frequency of 200 Hz. The bubble size was derived from the pressure fluctuations using a technique described by van der Schaaf *et al.* (17). The so-called 'incoherent variance' obtained in this way is a measure for the frontal area of the bubble - thus also for the volume of a bubble in a 2D column – and will be referred to as bubble size. The incoherent variance has been shown to be a good quantitative descriptor of the average bubble size at a certain height in a fluidised bed, although a calibration of this value (for example, using video analysis or optical probes) is required to determine absolute bubble sizes (18). In this study, we are interested in the reduction of average bubble size and therefore only consider the relative value of the incoherent variance.

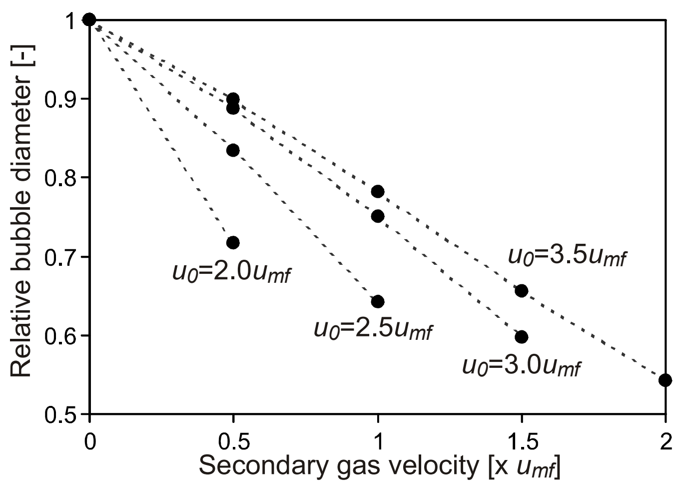


Figure 3. Relative bubble diameter, 20 cm above the distributor, as a function of the secondary gas velocity through the fractal injector. For every total gas velocity, the bubble diameter in the absence of secondary injection is defined as unity.

Figure 3 shows that for every total flow rate studied the bubble size is significantly reduced by feeding part of the gas (the secondary gas flow) via the fractal injector. The larger the fraction of the gas that is introduced via the fractal injector, the smaller the bubble size. At a higher measuring position the effect is somewhat smaller, since this position is further away from the upper level of the fractal injector. Further research is aimed at elucidating the mechanism behind the effects of distributed injection to allow a meaningful and directed approach to optimisation.

ELECTRIC FIELDS TO INDUCE INTERPARTICLE FORCES

By definition, a state of fluidization exists when the force of gravity on a set of particles is balanced by the drag arising from the flow of the fluidizing gas. It follows, therefore, that small interparticle forces, which may not be noticeable in other circumstances, may have a significant influence in a fluidized bed (19). Pandit *et al.* (20) recently showed via simulations that by introducing artificial interparticle forces, the behaviour in a fluidized can be moved from the Geldart B regime to the Geldart A regime. Since the bubble size in the Geldart A regime is smaller, imposing additional interparticle forces may be a way to reduce the bubble size.

By imposing an electric field on a fluidized bed of semi-insulating particles, the particles become polarised (*i.e.*, dipoles are induced on the particles); the net electric charge on each particle remains zero. These dipoles lead to an interparticle force, which strongly depends on the particle separation distance and the relative orientation of the particles in the electric field, both in magnitude and direction (21,22). Particles with the centre-to-centre axis aligned to the electric field will attract each other, while particles adjacent to each other in the field will repel one another. Particles at an angle to each other will experience a torque that attempts to align them to the field. Due to the electric field induced interparticle force, the particles in the bed will tend to form strings in the direction of the field. However, the electric field strength should be small enough to allow the free movement of particles; that is, the fluidity of the system must be preserved. By using an AC field instead of a constant (DC) electric field, this fluidity criterion is met.

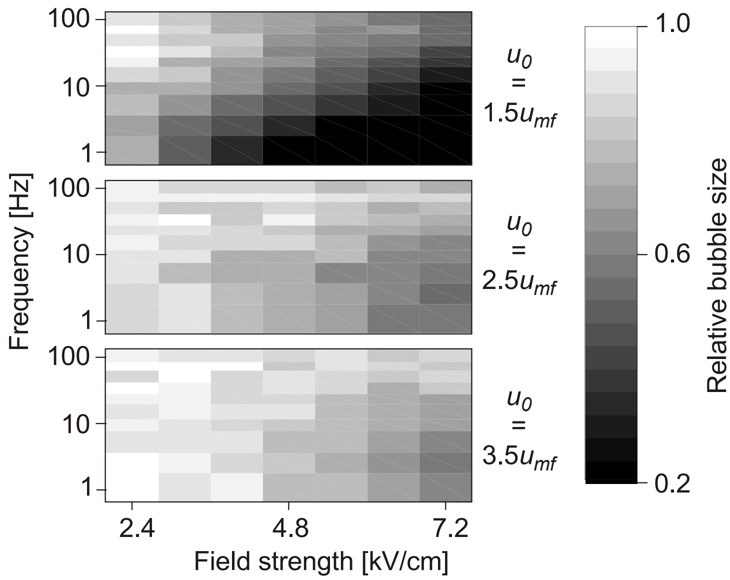


Figure 4. Ratio of bubble size with and without electric field, as determined by pressure fluctuation analysis at 20 cm above the distributor at 40% relative humidity, for three different gas velocities. Each subplot shows the bubble reduction ratio (*cf.* the grey scale) as a function of field strength (linear scale) and frequency (logarithmic scale).

In our experiments, the electric fields are introduced in the bed by stringing thin wires through the column, and alternately, both horizontally and vertically, driving these with an AC potential or grounding them (*cf.* Fig. 1). This creates a strongly inhomogeneous field in both the horizontal and vertical directions in the column. When no field is applied, the influence of these wires on the bubble behaviour is so small as to not be measurable. Oscillating (AC) electric fields with a frequency of 1 – 200 Hz and a strength up to 8 kV/cm were applied. The 2D set-up used is similar to the one used for the experiments with the fractal injector.

Figure 4 shows the ratio of the bubble size as a function of field strength and frequency to a base measurement without the presence of an electric field. The bubble size is derived from pressure fluctuations as described in the previous section; the pressure fluctuations are measured at 20 cm above the distributor plate. The figure shows that a large decrease in bubble size is obtained at higher field strengths (around 5 kV/cm and up). However, one should be careful: DC fields, or

The 12th International Conference on Fluidization - New Horizons in Fluidization Engineering, Art. 21 [2007]

very low frequency AC fields, may gridlock the particles and lead to defluidization. With increasing flow rate, the net effect of the electric fields becomes smaller. Further research will be aimed at, among other things, increasing the effect at higher gas velocities and developing acceptable electrode configurations for industrial applications.

OPTIMISING DISTRIBUTED PARTICLE PROPERTIES

It is a well-known fact in fluidization technology that the diameter and density of particles determine the type of fluidization: Geldart defined four particle types, each with a distinct fluidization behaviour (23). Not only the size of the particles, but also their size distribution strongly influences the fluidization behaviour. For example, the addition of fines (particles with a diameter < 45 µm (24)) improves the fluidization behaviour and leads to a better mass transfer. Sun and Grace (25) showed experimentally that a wider particle size distribution leads to higher conversions for ozone decomposition and suggested that this is due to the disproportionate amount of fines in the dilute phase (26). The current practice is, however, that fluidized bed particles used for catalysed gas-phase reactions are mainly optimised on the scale of a single particle. Most attention is given to their pore size distribution such that a high surface area is achieved and that the active sites are easily accessible by the gaseous components. Little attention is paid to the earlier mentioned mass transfer from gas in the dilute phase to the particles in the dense phase, essential to practical fluid bed operation.

We aim at improving the conversion and selectivity of gas-solid fluidized bed reactors by designing mixtures of particles with optimal properties (size distribution, density, shape, elasticity), aided by high-throughput experimentation, a novel approach for hydrodynamics research. The automated set-up we use allows access to quantitative information from large numbers of particle mixtures. Catalyst carrier materials such as silica and alumina are used as particles. Experiments are carried out in two industrially relevant fluidization regimes: bubbling fluidization and turbulent fluidization. In the experiments, pressure measurements, optical probes, and video analysis are used to assess the hydrodynamics.

Some preliminary results were carried out in a quasi-2D Plexiglas column (30x2.0x80 cm), operated within a cabinet controlled at 25ºC. The bed material consisted of alumina Geldart A particles, of which the fines concentration was increased from 0 to 30%; the median particle diameter is 75 μm at 0% fines. The increase in fines resulted in a decreased minimum fluidization velocity from 4.3 to 2.9 mm/s, and a decrease in voidage at minimum fluidization due to a better fit of small particles between the larger ones. Figure 5 shows the results for fluidization at 4.3 cm/s. An increasing amount of fines leads to an increased bed expansion. Moreover, the addition of fines results in a decrease of the bubble size and a smaller total amount of gas in the bubbles. This will enhance the mass transfer of components in the gas phase to the particle surface. Compared to the previous three approaches, our work on this fourth way of manipulating the fluidized bed hydrodynamics is still in the start-up phase. We will work on further development of the automated screening set-up, production of tailored particle mixtures, and scanning of a large number of these mixtures. Moreover, computational fluid dynamics will be used to obtain more insight into the interplay between particles with different properties.

CONCLUSIONS

We presented four approaches for controlling fluidized bed hydrodynamics. In this way, fluidized beds can be structured, which facilitates scaling-up. Moreover, the bubble size can be decreased which, for most catalytic fluidized bed processes, leads to increased conversion and selectivity. Better hydrodynamic control is achieved by introducing additional degrees of freedom. This can be done either by manipulating (1) the gas or (2) the particles:

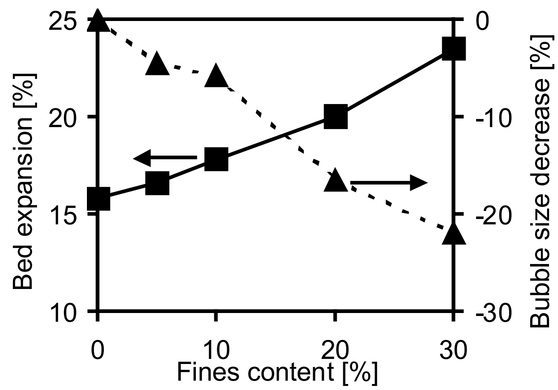


Figure 5. The influence of the fraction of fines in the bed on the bed expansion (compared to the bed height at minimum fluidization) and the decrease of the bubble size (compared to 0% fines). The bed is fluidized at 4.3 cm/s.

1.a. Varying the gas supply in time: we showed that an oscillating gas flow leads to regular bubble patterns.
1.b Distributing the gas supply over space: using a fractal injector, a much more even distribution of the gas is obtained leading to a strong reduction in bubble size.
2.a. Using an electric field, semi-insulating particles can be polarised. This induces interparticle forces, resulting in a significant decrease of the bubble size.
2.b. By tailoring the size distribution and other distributed properties of the particles, the bubble size can be decreased and the bed voidage increased.

Our group continues to investigate and optimise these four approaches.

ACKNOWLEDGEMENTS

We would like to thank D. Christensen, F. Kleijn van Willigen, and other current and former group members for their contributions to this research.

REFERENCES

1. Kaart, S., Schouten, J.C., van den Bleek, C.M., 'Improving conversion and selectivity of catalytic reactions in bubbling gas-solid fluidized bed reactors by control of the nonlinear bubble dynamics', Catal. Today 48, 185-194, 1999.
2. de Korte, R.J., Schouten, J.C., van den Bleek, C.M., 'Controlling bubble coalescence in a fluidized-bed model using bubble injection', AIChE J. 47, 851-860, 2001.
3. Harrison, A.J.L., Lim, C.N., Gilbertson, M.A., 'Modelling and control of the dynamics of bubbling fluidized beds', Proc. IMechE Part E: J. Process Mechanical Engineering, 220, 43-53, 2006.
4. Massimilla, L., Volpicelli, G., Raso, G., 'A study on pulsating gas fluidization of beds of particles', Chem. Eng. Progr. Symp. Ser. 62, 63-70, 1966.
5. Wong, H.W., Baird, M.H.I., 'Fluidization in a pulsed gas flow', Chem. Eng. J. 2, 104-113, 1971.
6. Köksal, M., Vural, H., 'Bubble size control in a two-dimensional fluidized bed using a moving double plate distributor', Powder Technol., 95, 205-213, 1998.

7. Pence, D.V., Beasley, D.E., 'Chaos suppression in gas-solid fluidization', Chaos 8, 514-519, 1998.
8. Coppens, M.-O., Regelink, M.A., van den Bleek, C.M., 'Pulsation induced transition from chaos to periodically ordered patterns in fluidised beds', in: Proceedings of the 4th World Conference on Particle Technology, Paper 355, 2002.
9. Wang, X.S., Rhodes, M.J., 'Pulsed fluidization - a DEM study of a fascinating phenomenon', Powder Technol., 159, 142-149, 2005.
10. Melo, F., Umbanhowar, P., Swinney, H.L., 'Transition to parametric wave patterns in a vertically oscillated granular layer', Phys. Rev. Lett. 72, 172-175, 1994.
11. Jensen, A., Johnsson, J.E., Andries, J., Laughlin, K., Read, G., Mayer, M., Baumann, H., Bonn, B., 'Formation and reduction of NOx in pressurized fluidized-bed combustion of coal', Fuel 74, 1555-1569, 1995.
12. Lyngfelt, A., Amand, L.E., Gustavsson, L., Leckner, B., 'Methods for reducing the emission of nitrous oxide from fluidized bed combustion', Energy Convers. Manage. 37, 1297-1302, 1996.
13. Adris, A.E.M., Grace, J.R., 'Characteristics of fluidized-bed membrane reactors: Scale-up and practical issues', Ind. Eng. Chem. Res., 36, 4549-4556, 1997.
14. Patil, C.S., van Sint Annaland, M., Kuipers, J.A.M., 'Experimental study of a membrane assisted fluidized bed reactor for H2 production by steam reforming of CH4', Trans. IChemE Part A Chem. Eng. Res. Des., 84, 399-404 MAY 2006
15. Coppens, M.-O., 'Structuring fluidized bed operation in a nature inspired way', in: Arena, U., Chirone, R., Miccio, M., Salatino, P. (eds.), Fluidization XI, Engineering Conferences International, New York, USA, 83-90, 2004.
16. Kleijn van Willigen, F., Christensen, D., van Ommen, J.R., Coppens, M.-O., 'Imposing dynamic structures on fluidised beds', Cat. Today 105, 560-568, 2005.
17. van der Schaaf, J., Schouten, J.C., Johnson, F., van den Bleek, C.M., 'Non-intrusive determination of bubble and slug length scales in fluidized beds by decomposition of the power spectral density of pressure time series', Int. J. of Multiphase Flow 28, 865-880, 2002.
18. Kleijn van Willigen, F., van Ommen, J.R., van Turnhout, J., van den Bleek, C.M., 'Bubble size reduction in a fluidized bed by electric fields', Int. J. of Chem. Reactor Eng. 1, paper A21, 2003. http://www.bepress.com/ijcre/vol1/A21
19. Seville, J.P.K., Willett, C.D., Knight, P.C., 'Inter-particle forces in fluidisation: a review', Powder Technol. 113, 261-268, 2000.
20. Wang, X.S., Rhodes, M.J., 'Pulsed fluidization - a DEM study of a fascinating phenomenon', Powder Technol., 159, 142-149, 2005.
21. Colver, G.M., 'An interparticle force model for ac-dc electric fields in powders', Powder Technol. 112, 126-136, 2000.
22. Kleijn van Willigen, F., van Ommen, J.R., van Turnhout, J., van den Bleek, C.M., 'Bubble size reduction in electric-field-enhanced fluidized beds', J. Electrostat. 63, 943-948, 2005.
23. Geldart , D., 'Types of gas fluidization', Powder Technol., 7, 285-292, 1973.
24. Yates, J.G., Newton, D., 'Fine particle effects in a fluidized-bed reactor', Chem. Eng. Sci., 41, 801-806, 1986.
25. Sun, G., Grace, J.R., 'The effect of particle size distribution on the performance of a catalytic fluidized bed reactor', Chem. Eng. Sci. 45, 2187-2194, 1990.
26. Sun, G., Grace, J.R., 'Experimental determination of particle dispersion in voids in a fluidized bed', Powder Technol. 80, 29-34, 1994.