Refereed Proceedings

The 13th International Conference on Fluidization - New Paradigm in Fluidization Engineering

Engineering Conferences International *Year* 2010

INVESTIGATION OF CHANGE OF VOLUMETRIC FLOW IN FLUIDIZED-BED REACTORS

Andrés Mahecha-Botero* Franz Haseidl† Alexander Nguyen‡
Tingwen Li** John R. Grace††

*University of British Columbia, andresmb@gmail.com
†Technische Universität München
‡University of British Columbia
**National Energy Technology Laboratory
††University of British Columbia

INVESTIGATION OF CHANGE OF VOLUMETRIC FLOW IN FLUIDIZED-BED REACTORS

Andrés Mahecha-Botero [a], Franz Haseidl [b], Alexander Nguyen [a], Tingwen Li [c] and John R. Grace [a]

[a] Department of Chemical and Biological Engineering, University of British Columbia, 2360 East Mall, Vancouver, Canada V6T 1Z3.

[b] Department of Chemical Engineering, Technische Universität München, Arcisstrasse 21, 80333 München, Germany.

[c] National Energy Technology Laboratory, 3610 Collins Ferry Road, P.O. Box 880, Morgantown, WV, U.S.A., 26507-0880.

ABSTRACT

Volumetric flow can change due to such factors as variation in the number of moles due to reaction, feed distribution, change of phase, membranes and changes in temperature and pressure. This change in volumetric flow was investigated by varying the fluidized-bed pressure in a two-dimensional steel column. Pressure was impulsively adjusted, showing large transient voids during the transitions after rapid growth of the existing bubbles. The effects of sudden depressurization as well as elevated pressure fluidization were also investigated.

1. INTRODUCTION

Fluidized-bed reactors are the backbone of many industrial processes. Most industrial applications of fluidized-bed reactors contain small solid particles (catalysts or reactants) in contact with gas species. Given their rapid development in recent decades, modelling and simulation of such systems have become instrumental in design, scale-up and optimization. Because of the complexities of fluidized-bed reactors, leading modelling tools rely on a series of correlations and assumptions that need validation. Much work has been devoted to conventional modelling (CM) of fluidized beds (1), treating the interpenetrating solids and fluid using some variant of the classic two-phase theory of fluidization (2). Advanced models based on CM typically address fluidization phenomena by introducing correlations on mass-transfer and hydrodynamic phenomena and solving for the concentrations of species and bed temperature based on differential equations derived from conservation equations of mass and energy. On the other hand, computational fluid dynamics (CFD) model the fluidization behaviour at a higher degree of sophistication by additionally solving momentum balances to predict the fluid-bed hydrodynamic behaviour (3). Both types of reactor modelling implement a number of assumptions and require validation. In this work we present initial experimental results of tests on the ability of CFD modelling to predict changes in volumetric flow.

The variables in question are extremely difficult to measure non-intrusively in industrial reactors. As a result, we adopted an indirect approach to investigate the effects of change in volumetric flow. In this work the pressure of a fluidized-bed is varied to simulate a change in volumetric flow and observe the effects on the fluidization properties. By varying the pressure in the bed it is possible to study the effects on transient and steady state fluidization behaviour to mimic in a simple and observable way what is likely to occur in hot reacting units. In addition, sudden depressurization can occur in fluidized bed reactors, e.g. due to opening of a pressure relief valve, and pressure swing could also occur in some applications. This study is helpful in showing what could happen in such circumstances. The effects of volume change are likely to be most interesting in the bubbling flow regime, but they could also play a role in the other major fluidization flow regimes. In this work, we focus on the bubbling flow regime.

1.1 Change of Volumetric Flow in Gas Fluidization

Some of the most complex phenomena to model in gas fluidization are inter-phase mass transfer, axial and radial dispersion and gas-solid contacting. These phenomena are strongly dependent on the gas volumetric flow. For gases, the volumetric flow is given by:

$$v = v_f \cdot \left(\frac{F_T}{F_{T_f}}\right)\cdot\left(\frac{T}{T_f}\right)\cdot\left(\frac{P_f}{P}\right)\cdot\left(\frac{Z}{Z_f}\right) \text{ , where } \quad F_T = \sum_{i=1}^{N_C} F_i \qquad \textbf{(1)}$$

This equation shows that the volumetric flow ($v=A.U_g$) can change due to variations in molar flow rate (F_T), temperature (T), pressure (P) and compressibility factor (Z). Note that the subscript f denotes a feed condition. For a constant column cross section, Equation 1 can be written in terms of gas velocity rather than volumetric flow. In general, the gas volumetric flow can change due to:

1) Variations in molar gas flow due to chemical reaction: This occurs when the reaction network has a stoichiometry with increasing (e.g.: reforming of hydrocarbons and decomposition reactions such as calcination, cracking of hydrocarbons, etc.) or decreasing (e.g.: oxy-chlorination, polymerization and most synthesis reactions such as ammonia, methanol, Fischer Tropsch, etc.) molar gas flow;

2) Changes in temperature: due to exothermic or endothermic reactions, as well as heat transfer; this is seldom a major factor in fluidized beds due to the temperature uniformity;

3) Changes in bed pressure: due to the inevitable hydrodynamic pressure decrease or due to pressure swing;

4) Membranes: When selective membrane removal or input of species is implemented *in-situ* (e.g. fluidized-bed membrane reactors for steam methane reforming);

5) Distributed feed: Utilized to enhance yield to a particular product, to control highly exothermic/endothermic reactions or to maintain the reactor away from local flammability limits (e.g. in partial oxidation processes);

6) Change of phase: When the fluidized-bed experiences rapid drying (e.g. in food processing), sublimation or condensation.

Although the effects of change in volumetric flow in fluidized beds can be very significant, little attention has been paid to this issue due to the complexities associated with its quantification and study. At one extreme these could, for example, lead to defluidization or great expansion of the dense phase, or, at the opposite extreme, be accommodated by immediately transferring the “extra” or “missing” flow to the other phase with negligible influence on the local hydrodynamics. Since the changes in volumetric flow do not occur at the same rate in the fluidization phases (most extra moles and heat are generated in the high density phase), the effect of interphase flow is very important, but difficult, to quantify.

Despite the potential influence of changes of molar flow on hydrodynamics, most authors neglect the effects of change in volumetric flow. Some works have attempted to estimate its effects by using plausible, but untested methodologies. Irani et al. (4) modified the Kunii and Levenspiel model to account for volume change for single reactions and first order kinetics by assuming that all extra gas generated appears immediately as bubbles, while maintaining the mass transfer coefficient constant. Kai and Furusaki (5) modified the model of Irani et al. (4) to account for possible variations in mass transfer coefficients due to bulk flow for single reactions and first order kinetics. Mahecha-Botero et al. (6; 7) distributed gas between the phases depending on the increase/decrease of total volumetric flow, which may change due to chemical reaction, membrane permeation, changes in temperature or pressure, or some combination of the above. They defined the gas flow in the dilute phase according to the modified two-phase theory as $v_H = mU_{mf}A$ where m was taken as 1.0 by default (i.e. corresponding to the standard two-phase theory) and suggested that m be adjusted based on experimental measurements.

1.2 Influence of Pressure on Fluidization Properties

Depending on the effect of pressure on reaction equilibrium and kinetics, process performance can benefit from elevated pressures. Many industrial reaction processes take place at elevated pressures to achieve high throughput and reactant conversion. Fluidization at high pressure can result in reduced minimum fluidization velocity specifically for Geldart B particles, and has negligible effects for Geldart A particles (8; 9). Furthermore, increased pressure results in smaller bubbles as compared to lower pressure equivalents (10), which can be explained by a higher rate of bubble breake-up and by the decrease in volumetric flow caused by increased pressures. In addition, high-pressure fluidization has been reported to increase dense bed voidage (11; 9), decrease particle segregation (12), increase bed expansion (9), increase entrainment and increase heat transfer coefficients (13). Nevertheless, there is no consensus in the literature about some of these conclusions, and there is controversy with respect to the analysis of many high-pressure fluidization results (10; 11; 8).

2. EXPERIMENTAL HIGH PRESSURE FLUIDIZED-BED

In this work we used a two-dimensional column (13) with inner dimensions 310 × 10 × 510 mm, designed to operate at pressures up to 2170 kPa(a). A schematic representation of the experimental setup is given in Figure 1. The vessel is

constructed of 43 mm thick reinforced steel plates. To view the fluidization phenomena, four tempered glass windows of 152.4 mm diameter and 25.4 mm thickness are used. The windows are located in pairs on opposite faces of the column. A high pressure gasket is then placed outwards over the windows as well as steel plates to prevent the windows from sliding outwards. A fine metal mesh is used as the bottom gas distributor. Additional metal meshes are used to prevent backflow of the particles and to filter the off-gas streams. High-pressure o-rings sealed the top and bottom flanges. To monitor the pressure within the bed, two differential pressure transducers (Omega PX409-015DDU5V) and an absolute pressure transducer (Omega PX209-300A5V) measure the fluidized-bed pressure drop (i.e. between 8 mm and 45.9 mm above the distributor), the distributor pressure drop and the absolute bed pressure (at 8 mm above the distributor). Three pressure gauges above and below the bed and at the distributor are used in conjunction with the transducers to determine the pressure and pressure drop within and across the bed.

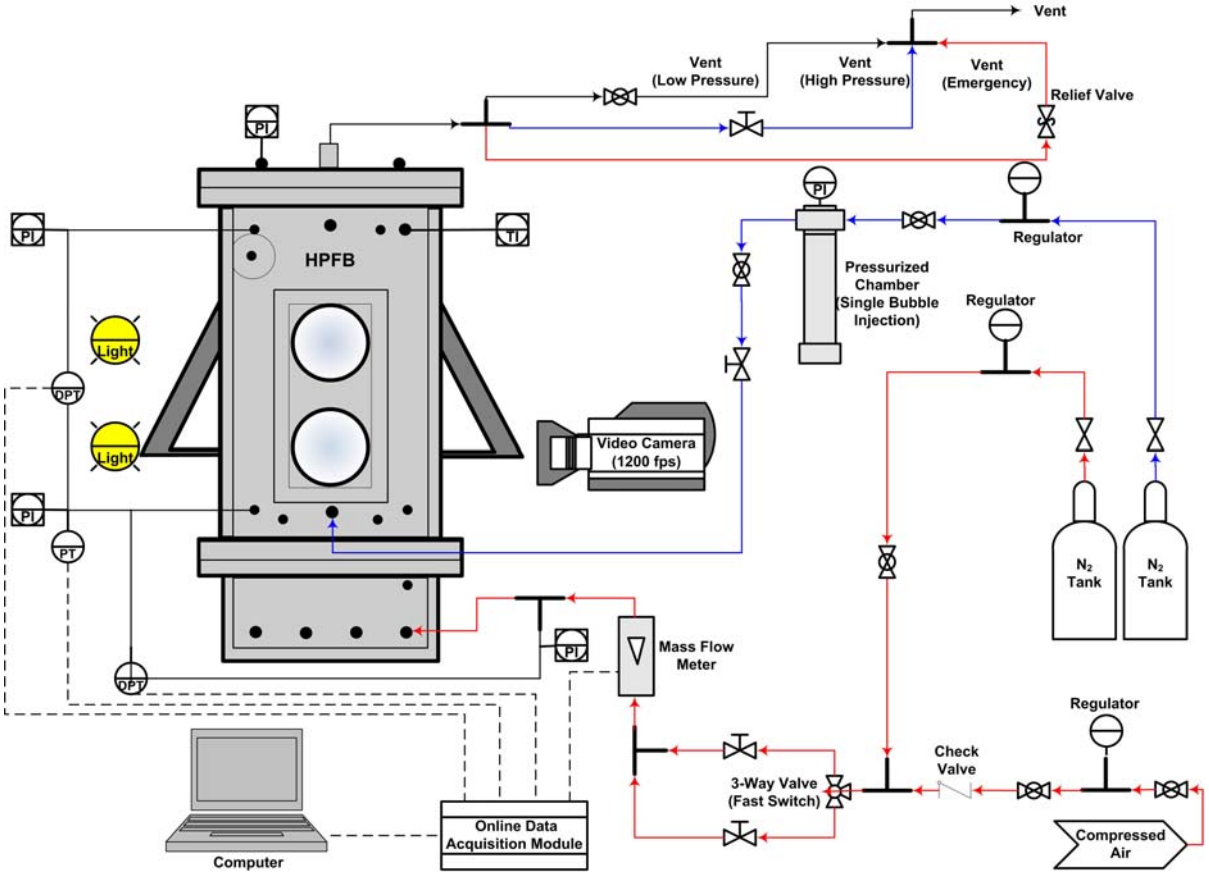

Figure 1. Flow diagram of high-pressure two-dimensional fluidized bed set-up.

There are two sources of gas for the fluidized bed: compressed air and nitrogen from gas tanks. Compressed air is used for intermediate pressure experiments, from 101-446 kPa(a), and nitrogen for high pressure experiments, from 446-2170 kPa(a). To control the gas flow into the fluidized bed, the input flow is adjusted by regulators and more precisely with needle valves. The input gas can be quickly switched from two preset flow conditions by a 3-way valve. The gas flow then travels through a high-range mass flow meter (Omega FMA1845) into the distributor and through the bed. To characterize the bubble properties in the bed, single bubbles can be

injected from a pressurized chamber into the base of the bed. The pressurized chamber can be charged with a predetermined amount of high-pressure nitrogen from a nitrogen tank, which is then released into the bed by a combination of ball and needle valves. The vessel off gas can be directed trough two flow lines (one at atmospheric pressure and the other one at high pressure) to rapidly increase/decrease the absolute pressure.

To obtain visual data, a Casio Exilim EX-F1 high speed camera records images at up to 1200 frames/s. The camera is placed at the front of the column while it is illuminated from behind to clearly show the bubbles. The bed was filled up to a bed height of 300 mm, utilizing 234 μm glass beads (supplied by Potter Industries). A data acquisition card (NI USB-6009) in conjunction with the Labview program (version 8.5) records the data from one absolute pressure transducer, two differential pressure transducers and a mass flowmeter. The recordings from the high-speed camera and data-acquisition system are synchronized during the experimental runs.

During the experimental runs, the bed pressure can be suddenly altered by redirecting the exit gas by means of synchronized action of a 3-way valve and a ball valve. The repeatability of the step changes of flow and pressure was monitored by transient recordings of bed pressure and gas input flow and by performing all tests in triplicate. It was found that the time scale of the valves action was always one to two orders of magnitude less than the time required to reach fluidization steady state.

3. RESULTS AND DISCUSSION

The minimum bubbling velocity (U_{mb}), was determined as the average of the values of U when the first bubbles appeared with increasing gas flow and when all bubbles disappeared with decreasing flow. The minimum fluidization velocity (U_{mf}) was obtained in the normal manner from the intersection point of the constant bed pressure drop line with the straight line for the packed bed region. To ensure equivalent fluidization conditions at all pressures, the two-dimensional bed was operated at a constant (U - U_{mf}) of 20 mm/s. For the current experimental runs, the system was operated at three different steady states with pressure set-points of 101.3, 273.6 and 446.0 kPa(a) as indicated in Table 1. Other experimental results are presented in Figures 2, 3 and 4.

Table 1. Experimental conditions.

Experiment	Parameter	Value	Parameter	Value
All Experiments	Column area, mm^2	310 x 10	Temperature, K	298
	Column height, mm	510	Particle density, kg/m^3	2500
	(U - U_{mf}), mm/s	20	Particle diameter, μm	234
	Variable	**Value**	**Variable**	**Value**
Steady State (1)	Pressure, kPa(a)	101.3	Experimental U_{mf}, mm/s	29.5
Steady State (2)	Pressure, kPa(a)	273.6	Experimental U_{mf}, mm/s	28.8
Steady State (3)	Pressure, kPa(a)	446.0	Experimental U_{mf}, mm/s	28.8

Transient pressure increase tests were performed from 101.3 kPa(a) to 273.6 and 446.0 kPa(a). The opposite changes in pressure were used to investigate the effect of pressure decrease. As the pressure increased, a slight reduction of the minimum fluidization velocity was observed. The prediction of Sidorenko and

Rhodes (8; 9) of a reduction in bubble size and smoother fluidization as pressure increases was also observed. Despite the smaller bubble size, an appreciable reduction in the expanded bed height was observed as the pressure increased.

Transient experimental tests were carried out to investigate the dynamic behaviour of fluidization properties as the bed pressure is suddenly varied. It was found that when increasing, as well as when decreasing, the pressure, the volumetric flow increased locally and temporarily inside the bed. This phenomenon is expected because for the cases of a sudden decrease in pressure, the gas expands rapidly increasing the bed volumetric flow until the system reaches a new steady state. On the other hand, for a sudden increase in pressure, there is a temporary increase in the bed volumetric flow (due to the larger opening of the input valve for the new high pressure condition) that dies out as the column gradually fills with gas. It is important to note that the volumetric flow at all steady states was almost constant (i.e. to compare fluidization conditions at the steady states) due to the very small variation of U_{mf} and the constant set-point of (U - U_{mf}).

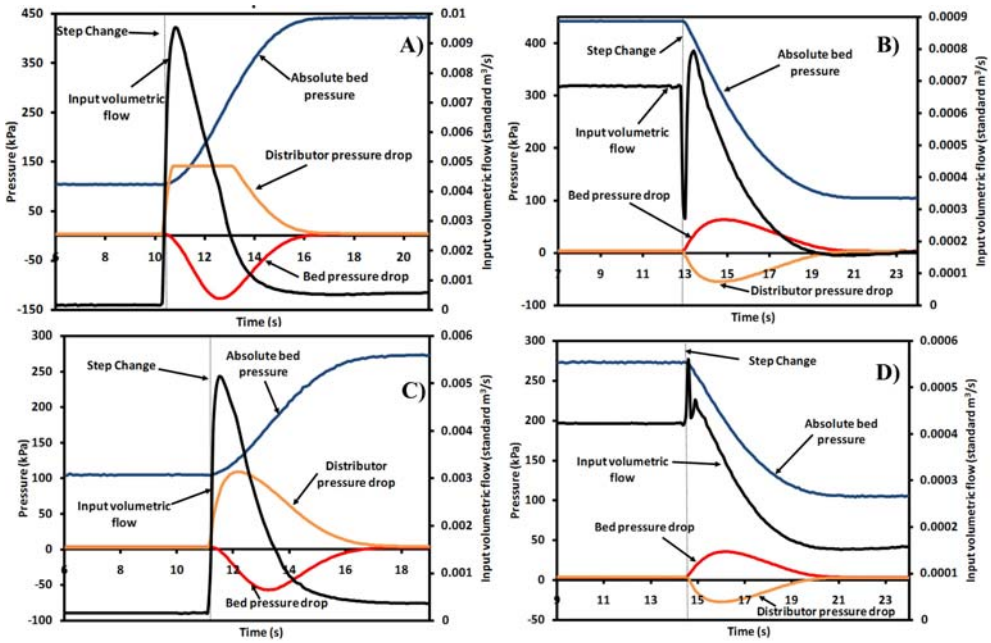

Figure 2. Data recorded after a step change in pressure: **A)** from 101.3 to 446.0 kPa(a). **B)** from 446.0 to 101.3 kPa(a). **C)** from 101.3 to 273.6 kPa(a). **D)** from 273.6 to 101.3 kPa(a). Input volumetric flow, absolute bed pressure, distributor pressure drop and bed pressure drop (signal at 1000 Hz, plotted at 10 Hz). For snapshots of transient behaviour in **A** and **D**, see Figures 3 and 4.

Figure 2 presents the variations of key variables when a step change in pressure takes place. At steady state, the fluidization pressure and input volumetric flow are constant. Additionally, the bed pressure drop as well as the pressure drop across the distributor are small during steady operation (i.e. less than 5 kPa). Pressure and input flow are then impulsively adjusted and the system's response is recorded in real time. A perturbation in the flow due to the sudden change in valve positioning is seen as peaks in the recordings of flow, which typically lasts less than 0.3 seconds. During the transient test, the bed pressure drop as well as the distributor pressure drop increase greatly due to an increase in local gas velocity

accompanied by a pressure wave. After the step change, the system rapidly adapts to a new steady state within 6 to 9 seconds.

The recorded transient behaviour of bubbles corresponding to a sudden increase (Figure 2A) and decrease (Figure 2D) in pressure are shown in Figures 3 and 4, respectively. As expected, bubbles expand for both circumstances because of the locally increased volumetric flow. However, differences can be visually observed from these figures that bubbles tend to shrink first when a sudden increase in pressure is imposed. Detailed analyses on effects of pressure change will be reported elsewhere due to space limitations here.

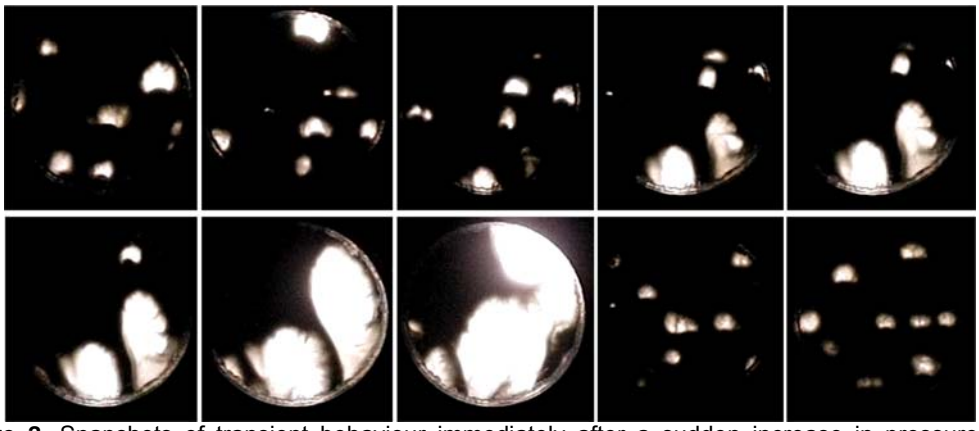

Figure 3. Snapshots of transient behaviour immediately after a sudden increase in pressure from 101.3 kPa(a) to 446.0 kPa(a). Video recorded at 1200 frames/s. Step change at t=10.3s. (Time from left to right: 9.7; 10.4; 10.45; 10.5; 10.52; 10.54; 10.56; 10.58; 18.95; 43.4 s.) For dynamic profiles see Figure 2A.

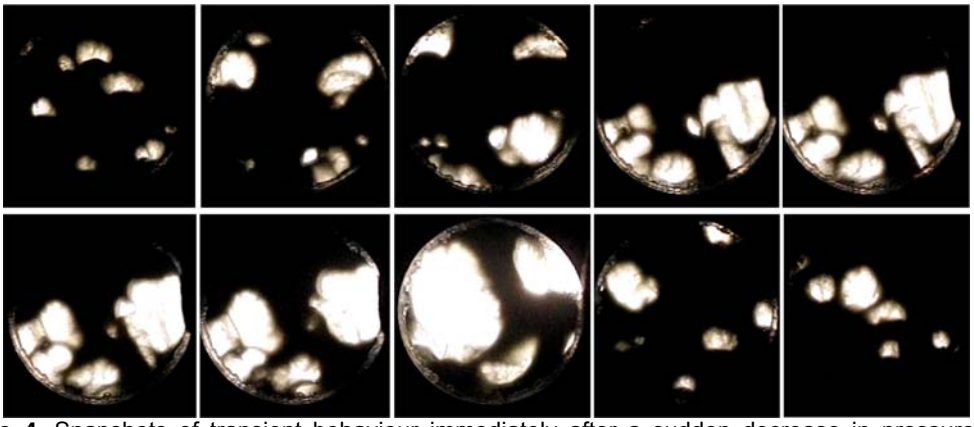

Figure 4. Snapshots of transient behaviour immediately after a sudden decrease in pressure from 273.6 to 101.3 kPa(a). Video recorded at 1200 frames/s. Step change at t=14.6s. (Time from left to right: 10; 14.7; 14.75; 14.8; 14.82; 14.84; 14.86; 14.9; 18.9; 45.6 s.) For dynamic flow and pressure profiles see Figure 2D.

As shown in Figures 3 and 4, impulsive changes in pressure, both upwards and downwards, were accompanied by transients where there were huge voids, before settling into the new steady state. The snapshots contain two frames of the initial state, then six frames are presented showing the huge changes in fluidization conditions that occur just after the step change, and finally two frames of the final state are shown several seconds later.

4. CONCLUSIONS

A high pressure, two-dimensional fluidized-bed using step changes in pressure was implemented to investigate the dynamic effects of change in pressure and volumetric flow in fluidized-bed reactors. As pressure increases, a reduction in the minimum fluidization velocity, a reduction in bubble size and smoother fluidization behaviour were observed. During the transients, huge voids appeared after a rapid growth of existing bubbles. The results shed light on the time scales of bubble expansion, as well as the variation of fluidization conditions due to variations in volumetric flow. These results provide information on sudden depressurization, and will be used in future work to test the ability of CFD simulations to predict the effects of variations in volumetric flow.

5. ACKNOWLEDGEMENTS

The authors are grateful to MITACS and to NSERC for providing funding for this project and to Pavitra Agarwal for assistance in preliminary planning of experiments.

6. NOTATION

A	reactor cross-sectional area, m^2	U_{mb}	minimum fluidization velocity, m/s
F_T	total molar flow, mol/s	U_{mf}	minimum fluidization velocity, m/s
N_c	number of gas species, (-)	v	volumetric flow , m^3/s
P	pressure, KPa(a)	Z	compressibility factor, (-)
T	temperature, K	**Subscripts**	
U_g	superficial gas velocity, m/s	f	feed condition

7. REFERENCES

1. Mahecha-Botero, A., Grace, J.R., Elnashaie, S.S.E.H. and Lim, C.J. (2009). Advances in modelling of fluidized-bed catalytic reactors: A comprehensive review. Chem. Eng. Commun., 196(11), 1375-1405.

2. Toomey, R.D. and Johnstone, H.F. (1952). Gaseous fluidization of solid particles. Chem. Eng. Prog., 48, 220-226.

3. Li, T., Mahecha-Botero, A. and Grace, J.R. (2009). Computational fluid dynamic investigation of change of volumetric flow in fluidized-bed reactors. Ind. Eng. Chem. Res., In Press.

4. Irani, K.R., Kurkarni, B.D. and Doraiswamy, L.K. (1980). Analysis of fluid bed reactors for reactions involving a change in volume. Ind. Eng. Chem. Fundam., 19, 424-428.

5. Kai, T. and Furusaki, S. (1983). Effect of volume change on conversions in fluidized catalyst beds. Chem. Eng. Sci., 39(1317-1319).

6. Mahecha-Botero, A., Chen, Z., Grace, J.R., Elnashaie, S.S.E.H., Lim, C.J., Rakib, M., Yasuda, I. and Shirasaki, Y. (2009). Comparison of fluidized bed flow regimes for steam methane reforming in membrane reactors: A simulation study. Chem. Eng. Sci., 64(16), 3598-3613.

7. Mahecha-Botero, A., Grace, J.R., Lim, C.J., Elnashaie, S.S.E.H., Boyd, T. and Gulamhusein, A. (2009). Pure hydrogen generation in a fluidized bed membrane reactor: Application of the generalized comprehensive reactor model. Chem. Eng. Sci., 64(17), 3826-3896.

8. Sidorenko, I. and Rhodes, M.J. (2003). Pressure effects on gas-solid fluidized bed behavior. Int. J. Chem. Reactor Eng. , 1(R5), 1-35.

9. Sidorenko, I. and Rhodes, M.J. (2004). Influence of pressure on fluidization properties. Powd. Technol., 141, 137-154.

10. Varadi, T. and Grace, J.R. (1978). High pressure fluidization in a two-dimensional bed. Fluidization - Proceedings of the 2nd Eng. Found. Conf. Davidson, J.F. and Keairns, D.L. Cambridge, Cambridge University Press**:** 55-58.

11. Yates, J.G. (1996). Effects of temperature and pressure on gas-solid fluidization. Chem. Eng. Sci., 51, 167-205.

12. Chen, J.L.P. and Keairns, D.L. (1975). Particle segregation in a fluidized bed. Can. J. Chem. Eng., 53, 395.

13. Botterill, J.S.M. and Desai, M. (1972). Limiting factors in gas-fluidized bed heat transfer. Powd. Technol., 6(4), 231-238.