World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:4, No:8, 2010

Experimental Investigation on Solid Concentration in Gas-Solid Circulating Fluidized Bed for Methanol-to-Olefins Process

Biao Wang, Tao Li, Qi-Wen Sun, Wei-Yong Ying, Ding-Ye Fang

Abstract**—**Methanol-to-olefins coupled with transformation of coal or natural gas to methanol gives an interesting and promising way to produce ethylene and propylene. To investigate solid concentration in gas-solid fluidized bed for methanol-to-olefins process catalyzed by SAPO-34, a cold model experiment system is established in this paper. The system comprises a gas distributor in a 300mm internal diameter and 5000mm height acrylic column, the fiber optic probe system and series of cyclones. The experiments are carried out at ambient conditions and under different superficial gas velocity ranging from 0.3930m/s to 0.7860m/s and different initial bed height ranging from 600mm to 1200mm. The effects of radial distance, axial distance, superficial gas velocity, initial bed height on solid concentration in the bed are discussed. The effects of distributor shape and porosity on solid concentration are also discussed. The time-averaged solid concentration profiles under different conditions are obtained.

Keywords**—**Branched pipe distributor, distributor porosity, gas-solid fluidized bed, solid concentration.

I. Introduction

METHANOL conversion to light olefins has received much attention to date and is an important process from different aspects including C1 chemistry and reaction mechanism, production of non oil-based chemicals, and as an economical solution for the increasing demand of propylene [1]. Methanol-to-olefins process provides an additional route for production of ethylene and propylene for the chemical industry. This process has some advantages over the current ethylene and propylene production process, the steam cracking of NGL, naphtha or other light fractions of petroleum, because methanol-to-olefins can provide a wider and more flexible range of ethylene to propylene ratio than stream cracking to meet market demand. Also, methanol can be produced from synthesis gas, which can be made from any source of carbon-containing materials such coal, petroleum residue, biomass and natural gas, which are practically impossible to be directly converted into ethylene and propylene [2], [3]. SAPO-34 is a silicoaluminophosphate with the CHA topology and a small pore opening that is 0.38 nm in diameter [4]. It is an effective catalyst for the conversion of methanol-to-olefins. Since the methanol-to-olefins reactions are fast and exothermic, and the SAPO-34 catalyst is easily deactivated, a circulating fluidized bed reactor containing reaction-regeneration system is the preferred reactor.

Solid concentration in circulating fluidized bed is of great interest not only to scholars in academic circles but also to engineers in practical industry [5]-[7]. In order to obtain deeper insight into a highly complex Solid concentration profiles, detailed and accurate experimental works are obviously important. Comparing with the pressure fluctuation measurements, which have long been used in fluidization studies and more often provide global behaviors of the gas–solids suspension, optical fiber probes provide more local flow information because of their smaller measurement volumes [8], [9].

According to Taghipour et al[10], the hydrodynamics of a two-dimensional gas–solid fluidized bed reactor are studied experimentally and computationally. Experiments are conducted in a 2D Plexiglas column of 1.0m height, 0.28m width and 0.025m thickness .The reflective optical fiber probe, PC-4 Powder Voidmeter, supplied by the Institute of process engineering of the Chinese Academy of Science in Beijing is used for voidage measurements in the Experiments. Zhang et al[11], [12] study bubble rising and descending velocity and Rising and descending bubble size distributions simultaneously, using a BVW-2 four-channel conductivity probe bubble parameters apparatus and its analysis in gas-liquid and gas-liquid-solid bubble columns. According to Qi et al [13], in order to investigate solids concentration in the fully developed region of co-current downward gas–solid flow, actual solids concentrations are measured in a circulating

The financial support by the National Key Technology R&D Program of China (No. 2006 BAE 02B02) and Shanghai Yankuang Energy R&D Co., Ltd.

B. Wang is with Engineering Research Center of Large Scale Reactor Engineering and Technology, Ministry of Education, State Key Laboratory of Chemical Engineering, East China University of Science and Technology, Shanghai, 200237, PR China. (e-mail: jameswang@mail.ecust.edu.cn).

T. Li is with Engineering Research Center of Large Scale Reactor Engineering and Technology, Ministry of Education, State Key Laboratory of Chemical Engineering, East China University of Science and Technology, Shanghai, 200237, PR China.

Q. W. Sun is with State Key Laboratory of Coal Liquefaction and Coal Chemical Technology, Shanghai, 201203, PR China.

W. Y. Ying is with Engineering Research Center of Large Scale Reactor Engineering and Technology, Ministry of Education, State Key Laboratory of Chemical Engineering, East China University of Science and Technology, Shanghai, 200237, PR China. (Corresponding author to provide phone: +86 21 64252192; fax: +86 21 64252192; e-mail: wying@ecust.edu.cn).

D. Y. Fang is with Engineering Research Center of Large Scale Reactor Engineering and Technology, Ministry of Education, State Key Laboratory of Chemical Engineering, East China University of Science and Technology, Shanghai, 200237, PR China.

fluidized bed (CFB) downer with 9.3 m in height and 0.1 m in diameter using a fiber optical probe. The results obtained from this work and in the literature show that the average solids concentration in the fully developed region of the CFB downers is not only a function of the corresponding terminal solids concentration, but the operating conditions and particle properties also have influences on the average solids concentration in the fully developed region of the CFB downers.

In order to develop reaction-regeneration system for methanol-to-olefins process, to investigate solid concentration profile and particle velocity profile of fluidized bed catalyst based SAP0-34 in circulating fluidized bed is indispensable. Cold model experiment system used solid particle which simulates fluidized bed catalyst based SAP0-34 is established to predict solid concentration profile of fluidized bed. Reflective-type optical fiber probes are effective tools for measuring the local solid concentration in fluidized bed reactors, and were widely used by many investigators [14]. They yield high signal-to-noise ratios and, if properly designed, they create a minimum disturbance to the overall flow structure [15]. More importantly, they are nearly free of interference by temperature, humidity, electrostatics and electromagnetic fields [16]. A PC-6D Solid Concentration Analyzer made by Institute of process engineering, Chinese Academy of Sciences, are employed to investigate solid concentration profile d in gas-solid fluidized bed.

I. Experimental

Figure 1 shows a schematic diagram of the experimental equipment, which consists of (A)Air system; (B) Branched pipe distributor; (C) circle pipe distributor; two fiber optic probe system; Circulating fluidized bed system and a series of cyclone. The distributors are made of stainless steel pipe and equipped with nozzles. The fiber optical probe system used for our tests is produced by the Institute of Process Engineering, Chinese Academy of Sciences, Beijing, China. The probe tip of PC-6D solid Concentration Analyzer is 4mm in diameter and 2*2mm in cross-sectional area. The precise calibration procedure of the probe and other details can be found in Zhang et al [15]. Local solid concentrations under 10 operating conditions are measured at 9 radial positions (r/R=0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9) on 10 axial levels (500, 600,700, 800, 900, 1000, 1100, 1200, 1400, 1600mm) and 4 initial bed height (600, 800, 1000, 1200mm). For each level, 20 measurements are taken for every one of 9 radial positions. The relative standard deviation is found to be within 5%. Particle circulation rate can be measured by volumetric method. The butterfly valve 11 in figure 1 can be closed transiently, the volume of particle and the duration should be measured simultaneously. According to (1), Particle circulation rate can be calculated. The Material properties and experimental conditions are listed in Table 1.

$$G_S = \frac{\rho_b V}{S_r t} \quad (1)$$

TABLE I
Material Properties and Experimental Conditions

Property	Value
Classification of Geldart	B
Particle diameter (mm)	$154\sim180 \times 10^{-3}$
Solid density(Kg/m^3)	2550
Minimum fluidization velocity (m/s)	0.09
Maximum solids concentration	0.5703
minimum solid voidage	0.4297
Gas density	1.225
Gas viscosity	1.789×10^{-5}
Superficial gas velocity (m/s)	0.3930, 0.4912, 0.5895,0.6877,0.7860
Gas distributor shape	Branched pipe and circle pipe
Distributor porosity	5‰ and 2.5‰
Nozzle diameter (mm)	2

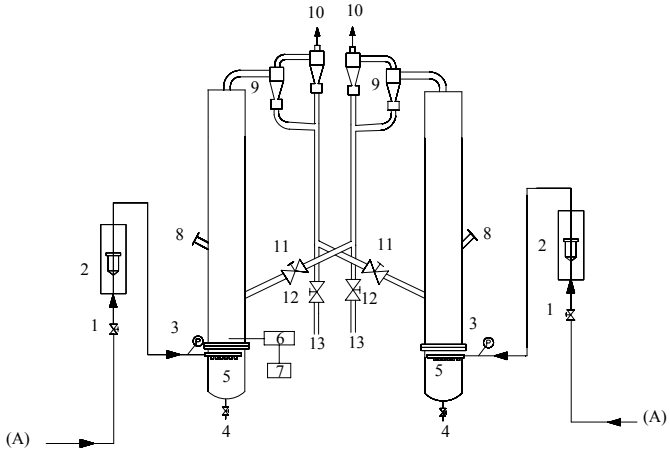


Fig. 1 Process flow chart
1. Valve; 2. Rotamater; 3. Pressure gauge; 4. Unloading valve; 5. Distributor; 6. Fiber optic probe system; 7. Computer; 8. Material inlet; 9. Cyclone; 10. Gas outlet; 11. Particle circulating valve; 12. Particle circulating unloading valve; 13. Particle circulating outlet;

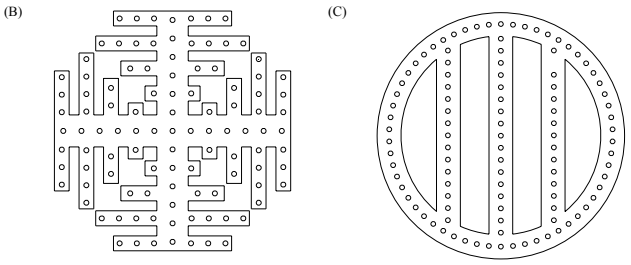


Fig. 2 (B) Branched pipe distributor and (C) circle pipe distributor

II. Results and Discussion

A. Effect of radical distance on solid concentration

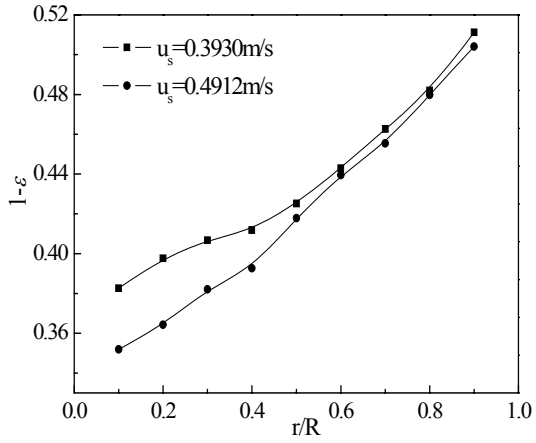


Fig. 3 H_0=600mm, H=500mm, Branched pipe distributor (2.5‰)

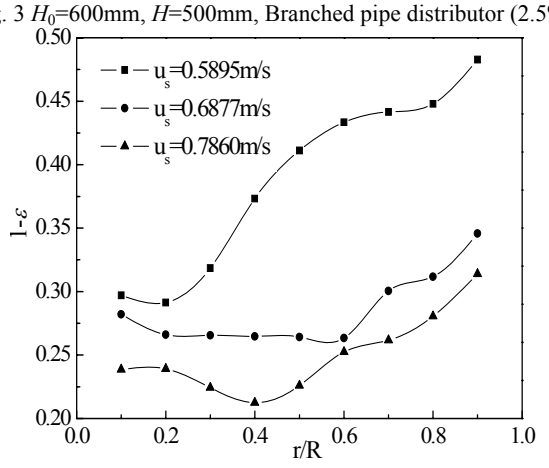


Fig. 4 H_0=600mm, H=700mm, Branched pipe distributor (5‰)

Figure 3 illustrates typical radial distributions of the time-average solid concentration at initial bed height of 600mm and axial distance of 500mm while branched pipe distributor (2.5‰) is used; figure 4 illustrates radial distributions of the time-average solid concentration at initial bed height of 600mm and axial distance of 700mm while Branched pipe distributor (5‰) is used. It is clearly that the solid concentration increases with the increase of *r/R*. This is probably due to the wall effect of the bed, which makes the gas tend to flow in the central region, thus causing the solid concentration smaller in the central region and bigger near the wall region. In the center region where gas velocity is relatively high, the drag force acting on the particles is larger. This makes the particles tend to move away from the central region to the wall region in order to consume less energy. The results show good agreement with former researchers [16], [17].

B. Effect of axial location on solid concentration

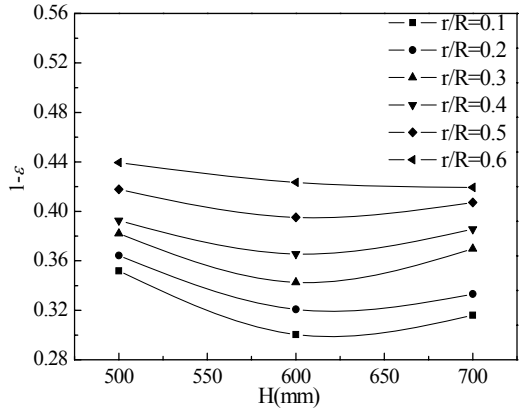


Fig. 5 H_0=600mm, U_S=0.3930m/s, Branched pipe distributor (2.5‰)

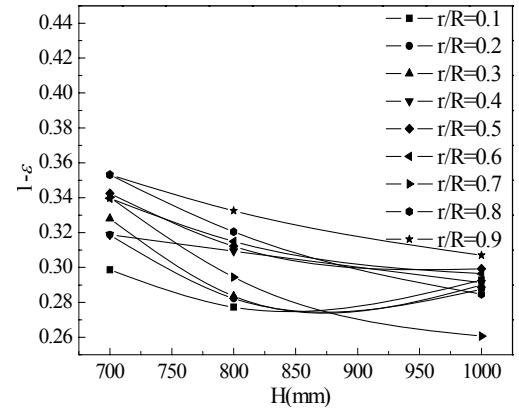


Fig. 6 H_0=800mm, U_S=0.6877m/s, Branched pipe distributor (5‰)

The effects of axial location on solid concentration are given in figure 5 and figure6. It can be seen that with the increase of axial location the solid concentration decreases. But it should be pointed out that the solid concentration does not decrease monotonously with axial distance. The experimental data show that there is some fluctuation with the increase of axial location. Gravitation attributes to one of the reasons of the solid concentration distribution in the axial location. It is known that in the bed the gas need to overcome the gravitation of the solid particle and the solid particle tends to fall down without any other external force. Therefore, it is easy to understand why the solid concentration is much higher in the bottom and getting lower and lower with the increase of axial location. As for the fluctuation, when the gas penetrates out from the nozzle on the distributor, it usually leads to a vibration of pressure and generates a large number of bubbles. So it is always a rather unstable region, thus causing the fluctuation of solid concentration.

C. Effect of superficial gas velocity on solid concentration

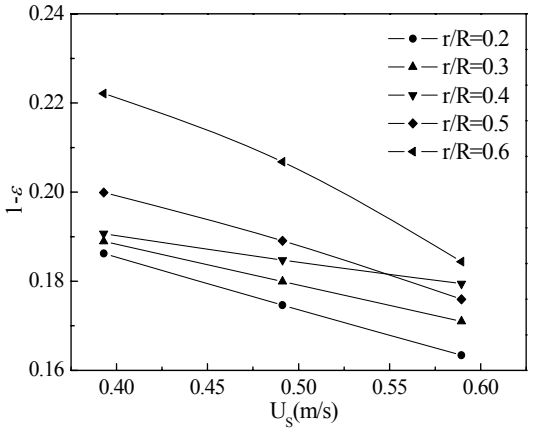


Fig. 7 H_0=1000mm, H=1100mm Branched pipe distributor (5‰)

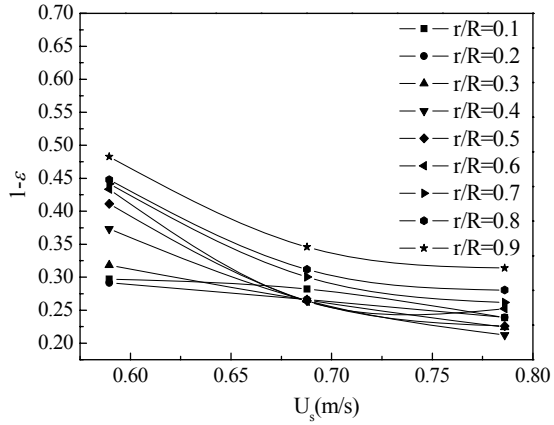


Fig. 8 H_0=600mm, H=700mm Branched pipe distributor (5‰)

As the results in figure 7 show, the solid concentration decreases with the increase of the superficial gas velocity in the detecting area. With 5‰ branched pipe distributor employed and the initial bed height H_0=1000mm, at the position of r/R=0.2, H=1100mm, the solid concentration decreased from 0.1863 to 0.1634; at the position of r/R=0.6, H=1100mm, the solid concentration decreased from 0.2221 to 0.1844. This can be explained that the bigger gas velocity provides more kinetic energy for the system and makes the solid particles easier to move upwards in the bed. So the bed height rises and the solid concentration decreases consequently.

D. Effect of initial bed height on solid concentration

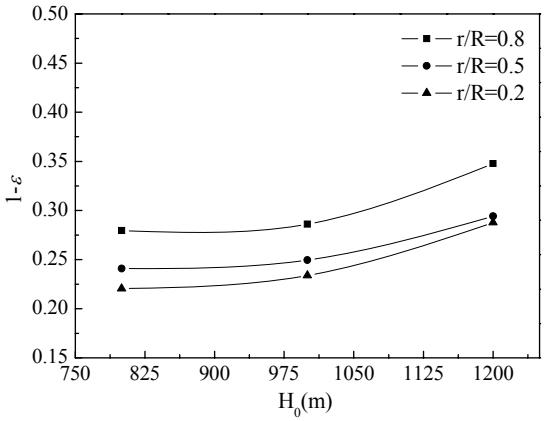


Fig. 9 U_S=0.5895m/s, H=1100mm Branched pipe distributor (5‰)

Figure 9 illustrates initial bed height on the time-average solid concentration at superficial gas velocity of 0.5895m/s while axial distance is 1000mm. The results indicate that the solid concentration increases with the increase of initial bed height in the detecting area. With other conditions unchanged, it is more difficult for the gas to blow the solid particles up when there are more particles in the bed.

E. Effects of distributor shape and porosity on solid concentration

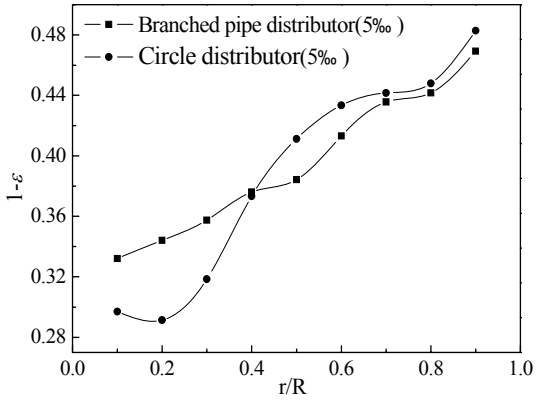


Fig. 10 H_0=600mm, H=700mm, U_S=0.5895m/s

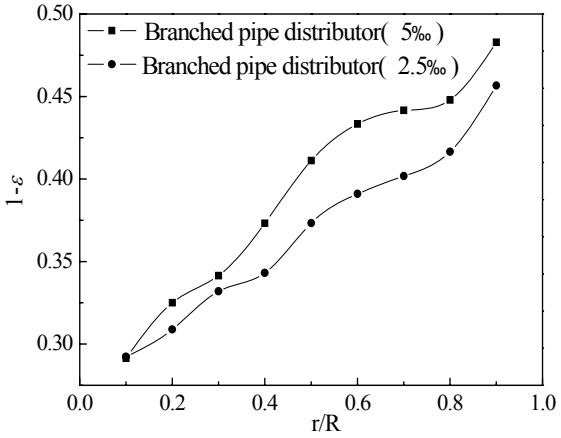
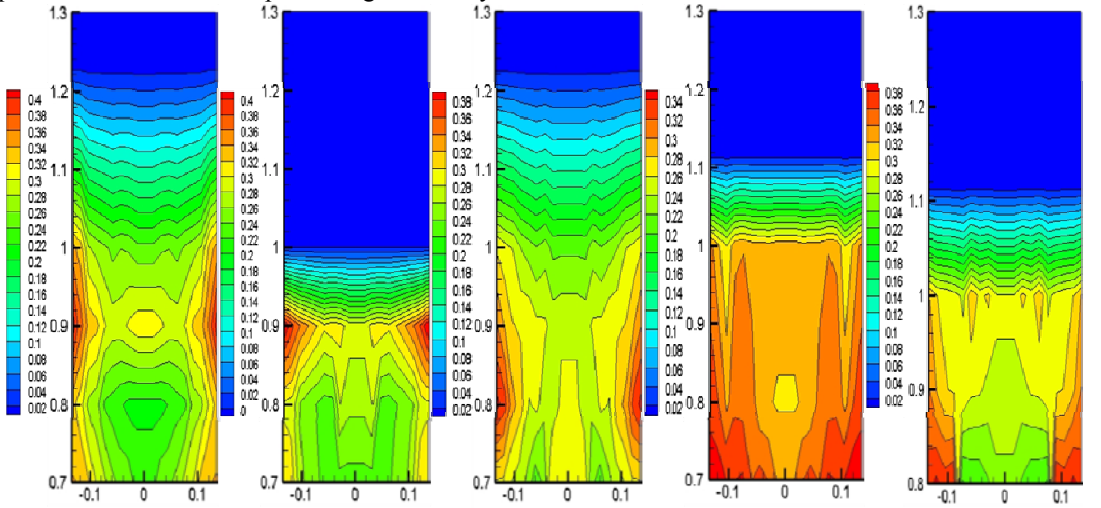


Fig. 11 H_0=600mm, H=700mm , U_S=0.4912m/s

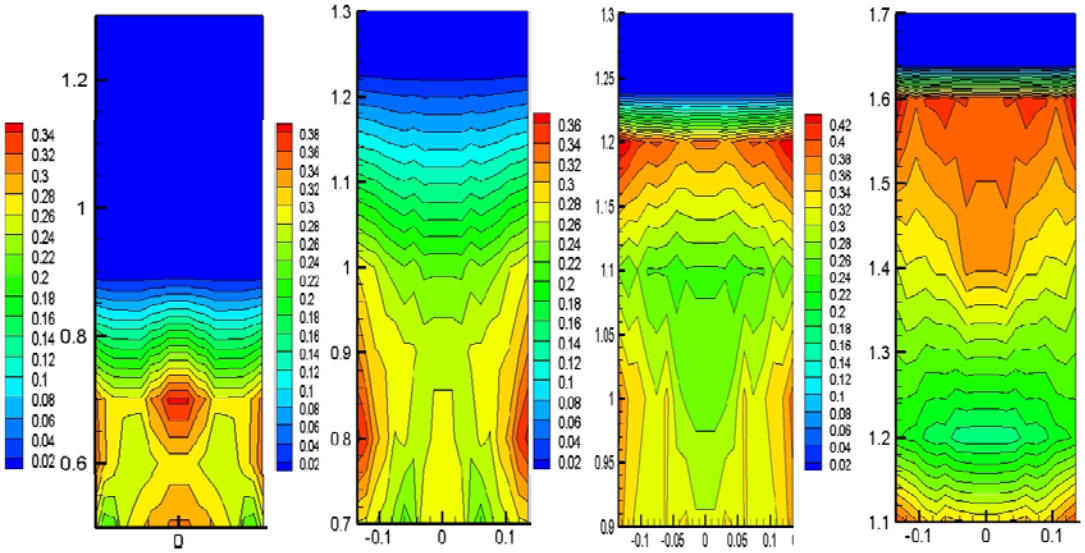
Figure 10 illustrates distributor shape on the time-average solid concentration at initial bed height of 600mm and superficial gas velocity of 0.5895m/s while axial distance is 700mm and the distributor porosity is 5‰; figure 11 illustrates distributor porosity on the time-average solid concentration at initial bed height of 600mm and superficial gas velocity of 0.4912m/s while axial distance is 700mm and the is used. The results indicate that the solid concentration does not change significantly while the branched pipe distributor and the circle pipe distributor is used. The pressure drop of branched pipe distributor is larger than circle pipe distributor with the same superficial gas velocity, on account of complicated structure of branched pipe distributor. So the superficial gas velocity of branched pipe distributor is smaller than circle pipe distributor, according to figure 10, variation trends are similar to the effects of superficial gas velocity on solid concentration. According to figure11, the solid concentration of lower distributor porosity is smaller than the higher distributor porosity at the same operated conditions. This is due to that with the same superficial gas velocity, the gas velocity from nozzle of lower distributor porosity distributor is larger; larger gas velocity provides more kinetic energy for the system and makes the solid particles easier to move upwards in the bed.

F. Time-averaged solid concentration profiles



U_S=0.3930m/s U_S=0.4912m/s U_S=0.5895m/s U_S=0.6877 m/s U_S=0.7860 m/s

Fig. 12 Time-averaged solid concentration profiles under different superficial gas velocities



H_0=600mm H_0=800mm H_0=1000mm H_0=1200mm

Fig. 13 Time-averaged solid concentration profiles under different initial bed height

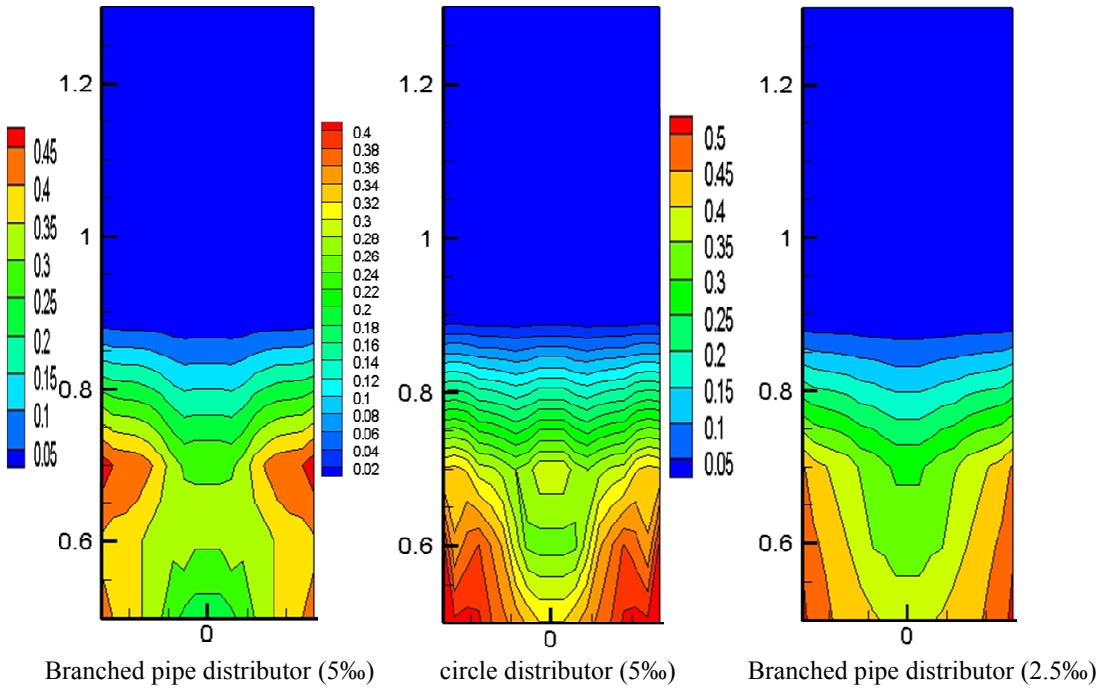


Fig. 14 Time-averaged solid concentration profiles under different distributor shape and porosity

The solid concentration profiles under different conditions have been calculated according to the experimental data obtained. Figure 12 shows the profiles using branched pipe distributor (5‰) under different superficial gas velocity ranging from 0.3930~0.7860m/s when the initial bed height is 800mm. The elliptic contours show similarity with experimental and calculated results of researchers Gidaspow, Ettehadieh[18] and Kuipers et al [19]. From the figure we can see there is a small region near the wall of in the lower part of the bed where the contours approximately equal to the solid concentration at minimum fluidization state. At the center in the lower part of the bed the solid concentration is much smaller than elsewhere. Figure 12 illustrates that as superficial gas velocities increase, the annular region rise and get more and more obvious. Figure 13 shows the profiles using branched pipe distributor (5‰) under different initial bed height ranging from 600~1200mm when the superficial gas velocities is 0.6877m/s. According to figure 13, we can find that the height of bed cubical expansion increases as initial bed height increases from 600mm to 1200mm. Figure 14 shows the profiles at the same operated conditions of initial bed height 600mm and superficial gas velocities 0.4912 m/s using different distributor shape and porosity. According to figure 14, we can find that the solid particles are well distributed while the branched pipe distributor (5‰) is used.

G. Effects of superficial gas velocity and initial bed height on particle circulation rate

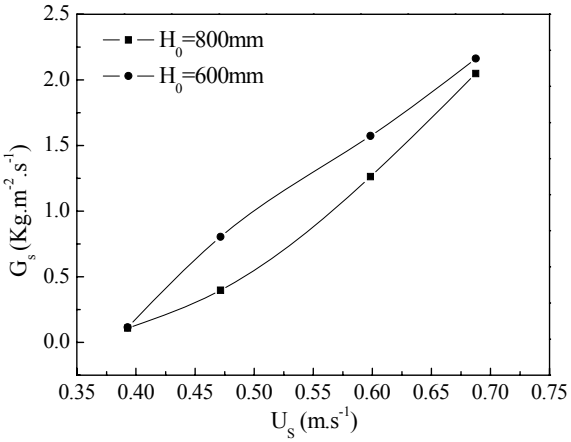


Fig. 15 Effects of superficial gas velocity and initial bed height on particle circulation rate

Figure 15 shows the particle circulation rate distribution using 5‰ branched pipe distributor under different superficial gas velocity ranging from 0.3930~0.6877m/s when the initial bed heights are 600mm and 800mm. According to the figure, it is found that particle circulation rate increases as superficial gas velocity increases and higher initial bed height has smaller particle circulation rate. This can be explained that the bigger gas velocity provides more kinetic energy for the system and makes the solid particles easier to move upwards in the bed. With other conditions unchanged, it is more difficult for the gas to blow the solid particles up when there are more particles in the bed.

III. Conclusions

In conclusion, from the results obtained up till now, in the similar detecting area and with other operating conditions unchanged, the following can be stated for the similar gas-solid fluidized bed:

(1) The solid concentration in the dense phase area increases with the increase of *r/R*, initial bed height, while decreases with the increase of axial distance and superficial gas velocity.

(2) Effects of distributor shape and porosity on time-averaged solid concentration are discussed, the time-averaged solid concentration profiles under different conditions are investigated systematically using PC-6D fiber optical probe system, according to solid concentration profiles under different distributor shape and porosity, we can find that the solid particles are well distributed while the branched pipe distributor (5‰) is used.

(3) Effects of different superficial gas velocity ranging from 0.3930~0.6877m/s, initial bed heights of 600mm and 800mm, on particle circulation rate are also discussed. The results can provide basic data for similar industrial operation.

Symbols Used

G_S	[kg/(m^2.s)]	Particle circulation rate
H	[mm]	Axial location
H_0	[mm]	Initial bed height
r/R	[-]	Dimensionless radius
S_r	[m^2]	Column sectional area
t	[s]	Duration
U_S	[m/s]	Superficial gas velocity
V	[m^3]	Circulation particle volume
1-ε	[-]	Solid concentration
ρ_b	[kg/m^3]	Circulation particle density

Acknowledgment

The authors gratefully acknowledge the financial support of the National Key Technology R&D Program of China (No. 2006 BAE 02B02) and Shanghai Yankuang Energy R&D Co., Ltd.

References

[1] A. Izadbakhsh, F. Farhadi, F. Khorashe, "Effect of SAPO-34's composition on its physico-chemical properties and deactivation in MTO process", Appl. Catal., A, vol. 364, pp. 48-56, 2009.

[2] X. C. Wu, M. G. Abraha, R. G. Anthony, "Methanol conversion on SAPO-34: reaction condition for fixed-bed reactor", Appl. Catal., A, vol. 260, pp. 63-73, 2004.

[3] B. V. Vora, T. L. Marker, P. T. Barger, H. E. Fullerton, H. P. Nilson, S. Kvisle, T. Fuglerud, "Economic route for natural gas conversion to ethylene and propene", Stud. Surf. Sci. Catal., vol. 107, pp. 87-92, 1997.

[4] H. Q. Zhou, Y. Wang, F. Wei, D. Z. Wang, Z. W. Wang, "In situ synthesis of SAPO-34 crystals grown onto a-Al2O3 sphere supports as the catalyst for the fluidized bed conversion of dimethyl ether to olefins", Appl. Catal., A, vol. 341, pp. 112-118, 2008.

[5] H. Y. Zhu, J. Zhu, "Gas-Solids Flow Structures in a Novel Circulating Turbulent Fluidized Bed", AIChE. J, vol. 54, no. 5, pp. 1213-1221, 2008.

[6] Y. P. Li, B. J. Ma, J. B. Hu, K. Zhao, "Numerical Simulation of the Hydrodynamics of Gas/Solid Two-Phase Flow in a Circulating Fluidized Bed with Different Inlet Configurations", Chem. Eng. Tech., vol. 32, no. 6, pp. 964-965, 2009.

[7] W. Q. Zhong, Y. Zhang, B. S. Jin, M. Y. Zhang, "Discrete Element Method Simulation of Cylinder-Shaped Particle Flow in a Gas-Solid Fluidized Bed", Chem. Eng. Tech., vol. 32, pp. 386-396, 2009.

[8] J. J. Nieuwland, R. Meijer, J. A. M. Kuipers, W. P. M. van Swaaij, "Measurements of solids concentration and axial solids velocity in gas–solid two-phase flows", Powder Technol., vol. 87, pp. 127-137, 1996.

[9] D. Tayebi, H. F. Svendsen, A. Grislingås, T. Majdell, K. Johannessen, "Dynamics of luidized-bed reactors: development and application of a new multi-optical fiber probe", Chem. Eng. Sci., vol. 54, pp. 2113-2123, 1999.

[10] E. Taghipou, N. Ellis, C. Wong, "Experimental and Computational Study of Gas-solid Fluidized Bed Hydrodynamics", Chem. Eng. Sci., vol. 60, pp. 6857-6867, 2005.

[11] L. J. Zhang, T. Li, W. Y. Ying, D. Y. Fang, "Experimental Study on Bubble Rising an Descending Velocity Distribution in a Slurry Bubble Column Reactor", Chem. Eng. Tech., vol. 31, no. 9, pp. 1362-1372, 2008.

[12] L. J. Zhang, T. Li, W. Y. Ying, D. Y. Fang, "Rising and descending bubble size distributions in gas–liquid and gas–liquid–solid slurry bubble column reactor", Chem. Eng. Res. Des., vol. 86, pp. 1143-1146, 2008.

[13] X. B. Qi, H. Zhang, J. Zhu, "Solids concentration in the fully developed region of circulating fluidized bed downers", Powder Technol., vol. 183, pp. 417-427, 2008.

[14] J.J. Nieuwland, R. Meijer, J.A.M. Kuipers, W.P.M. van Swaaij, "Measurements of solids concentration and axial solids velocity in gas–solids two-phase flows", Powder Technol., vol. 87, pp. 127-137, 1996.

[15] H. Zhang, P.M. Johnston, J. X. Zhu, H. I. D. e. Lasa, M. A. Bergougnou, "A novel calibration procedure for a fiber optic solids concentration probe", Powder Technol., vol. 100, pp. 260-270,1998.

[16] H. Johnsson, F. Johnsson, "Measurements of local solids volume-fraction in fluidized bed boilers", Powder Technol., vol. 115, pp. 13-18, 2001.

[17] F Wei, H Lin, Y Cheng, Y Jin, Z Yu, "Profiles of Particle Velocity and Solids Fraction in a High Density Riser", Powder Technol., vol. 100, pp. 183-193, 1998.

[18] D. Gidspow, B. Ettehadieh, " Fluidization in two dimensional beds with a jet: Part Ⅱ. Hydrodynamic modeling" , Ind. Eng. Chem. Fundam., vol. 22, pp. 193-201, 1983.

[19] J. A. M. Kupiers, H. Tammes, W. Princs, W. P. M. van Swaaij, "Experimental and theoretical porosity profiles in two dimensional gas-fluidized bed with a central jet", Powder Technol., vol. 71, pp. 87-92, 1992.