Particle Behavior in FBRs: A Comparison of the PBM–CFD, Multi-Scale CFD Simulation of Gas–Solid Catalytic Propylene Polymerization

Ya-Ping Zhu, Guo-Qiang Chen, Zheng-Hong Luo*

A multi-scale CFD model has been developed to describe the particle behavior in a polyolefin fluidized bed reactor (FBR). The model consists of a CFD model incorporating a single particle model and a population balance model (PBM). The main particle behavior in the FBR can be calculated using the multi-scale model. The multi-scale model is tested by comparing

simulation results with experimental data. Three cases including CFD coupled with PBM, CFD–PBM coupled with the single particle model without consideration of external diffusion, and multiscale CFD model under consideration of external diffusion are developed to further examine the model. The simulations demonstrate that both intraparticle mass and heat transfers, which are ignored by these conventional CFD–PBM models, have significant effects on the particle behavior.

1. Introduction

Many important polyolefin products are produced by solidcatalyzed olefin polymerization in continuous gas–solid fluidized bed reactors (FBRs). As the catalyst particle passes through the reactor system, monomer polymerizes causing polymer to encapsulate the catalyst, which expands and grows into the resultant polymer particle. It leads to the well known replication phenomenon.^[1] The modeling of this phenomenon is very complex, and requires a multiscale treatment.^[1,2]

Department of Chemical and Biochemical Engineering, College of Chemistry and Chemical Engineering, Xiamen University, Xiamen 361005, China

E-mail: luozh@sjtu.edu.cn

Department of Chemical Engineering, School of Chemistry and Chemical Engineering, Shanghai Jiao Tong University, Shanghai 200240, China





Generally, three scales, i.e. macro-, meso-/particle-, and micro-scale, are involved in the olefin polymerization process (see Figure 1).[3-5]

From Figure 1, one can associate different characteristic lengths and phenomena with these different levels. Among them, the meso-scale involves intraparticle, interparticle, and particle-wall interactions in terms of heat and mass transfer, which directly influence particle morphology evolution and monomer adsorption.^[1,6] It is also at this scale that agglomeration and/or sheeting can begin and that meltdown might occur. Thus, at this scale, particular attention should be paid to these particle variables, such as particle size, PSD, particle volume fraction and particle effectiveness factor etc.^[7] The particle scale is also the interface between the continuum approach used at the macro-scale and the discrete approach needed at the micro-scale.^[1] This work will concentrate on the particle behavior linked to the particle variables at this particle scale in a propylene polymerization FBR.

So far, most published reports on particle behavior in olefin polymerization FBRs were concerned with the transfer behavior within a single particle and interparticle

Y.-P. Zhu, G.-Q. Chen, Prof. Z.-H. Luo

Prof. Z.-H. Luo



Figure 1. The multi-scale phenomenon in olefin polymerization FBRs.

interaction behavior separately.^[2,5,6,8-20] Intraparticle transfer can affect particle growth while interparticle interaction may influence particle distribution in macroreactors. In practice, the mean particle size and PSD can be calculated via solving the single particle model coupled with a PBM,^[8–19] and the particle effectiveness factor and volume faction distributions can be obtained via solving the single particle model coupled with a CFD model.^[2,5,20] Nevertheless, few studies were implemented to calculate all the above four particle variables via integrated models simultaneously. For instance, Yiannoulakis et al.^[12] developed a steady-state PBM incorporating a single particle model to predict the mean particle size and PSD in ethylene copolymerization FBRs. Fan et al.^[21–23] suggested CFD–PBM coupled models to simulate gas-solid olefin polymerization FBRs. In their studies, PBM, chemical reaction engineering (CRE) model, and CFD were combined to investigate the roles of intrinsic kinetics and PSD of catalyst in a FBR. Polymer particle size, PSD and particle volume fraction were predicted simultaneously. However, the particle effectiveness factor was not considered and the single particle growth effect was not mentioned.^[21–23] Luo et al.^[16] presented a simplified single particle model coupled with a PBM for calculating the particle size and PSD of polypropylene produced in a loop reactor instead of FBR. More recently, the corresponding author's team^[5] developed a CFD-PBM-PMLM integrated model (polymeric multilayer model, PMLM, i.e. a single particle model) to predict the dynamic evolution of mean particle size, PSD and particle volume fraction. Herein, we pointed out that a single particle model, i.e. the PMLM, was incorporated into the CFD-PBM coupled model to construct a CFD-PBM-PMLM integrated model. Unfortunately, the particle effectiveness factor was still ignored and the single particle model adopted was an empirical model instead of a first-principle equation.^[5] Based on the above discussion, it becomes clear that the early efforts on modeling particle behavior in olefin polymerization FBRs were directed towards accounting for the detailed aspects of intraparticle transfer and/or gas–solid hydrodynamics without reaction and external diffusion (i.e. external transfer resistance of catalyst particle). It is also clear that comprehensive modeling of particle behavior encompassing simultaneous intraparticle transfer and reaction, external diffusion, particle kinetics, and gas–solid hydrodynamics in the olefin polymerization FBR has not been reported. In other words, multi-scale modeling of particle behavior in olefin polymerization FBR has not been attempted to date.

The main objective of this study is to achieve a more comprehensive understanding of particle behavior in propylene polymerization FBRs from the particle scale and in turn to provide new insights to polymerization FBRs. Therefore, a direct iterative multi-scale simulation approach suggested by Luo et al. at Shanghai Jiao Tong University and Xiamen University was adopted to construct a novel multi-scale model for describing simultaneous intraparticle transfer and reaction, external diffusion, gas-solid two-phase flow in polydisperse propylene polymerization FBRs.^[24] The multi-scale model developed here utilized a two-dimensional (2D) two-phase CFD model and incorporated a PBM and a single particle model based on first principles. The important particle variables (i.e. the mean particle size, PSD, particle volume fraction and particle effectiveness factor) in reactors could be calculated while accounting for intraparticle transfer and reaction, external diffusion as well as the solid PSD simultaneously via the multi-scale model.

2. Material and Simulation Methods

2.1. Simulated Reactor

To obtain particle behavior using the multi-scale CFD model, we selected an experiment-scale FBR as the reference reactor, which was studied in our previous works (see Figure 2).^[2,5,25] The selected reactor has an inner diameter of 0.33 m, height of 0.90 m, and initial bed height of 0.2 m (see Figure 2).

2.2. Concurrent Multi-Scale Model and Its Modeling Method

The multi-scale CFD model is analogous to that reported in our previous work^[5] with some improvements. These improvements are as follows: (1) the single particle model used in this work was directly deduced from first principles and is different from the PMLM.^[5] (2) The multi-scale





Figure 2. Geometrical features of the simulated FBR.

coupling method used in this work was a concurrent iterative method, while our previous iterative approach is not concurrent for the transportation of key information between different scales. For the other equations of the multi-scale model, the readers are encouraged to refer to Yan et al.'s work.^[5] Herein, only these differences above will be described.

2.2.1. Single Particle Model

The single particle model considers mass and heat transfer effects on chemical reaction both from the gas bulk to the particle surface (i.e. extra-diffusion) and further into the particle center (i.e. intra-diffusion). Based on first principles, the intraparticle governing equations are usually composed of mass, energy, and momentum balances as well as the auxiliary equations of state for gases, species reaction kinetics, multicomponent diffusion, etc.^[26,27] However, there is only a reactant component (i.e. propylene) in the propylene polymerization system. Therefore, the single particle model can be simplified at steady state operating condition and without intraparticle pressure gradient. Based on first principles,^[28] the following governing equations can be obtained:

Mass balance in a particle:

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 D_{\rm e}\frac{\partial c}{\partial r}\right) = \Re \tag{1}$$

Heat balance in a particle:

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$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\lambda_{\rm eff}\frac{\partial T}{\partial r}\right) = \Re\cdot\Delta H \tag{2}$$

Boundary conditions: at r = 0,

$$\frac{\partial c}{\partial r} = 0 \tag{3}$$

$$\frac{\partial T}{\partial r} = 0 \tag{4}$$

at $r = D_p/2$ and without the external diffusion,^[28]

$$c = c_{\rm s} \tag{5}$$

$$T = T_{\rm s} \tag{6}$$

at $r = D_p/2$ and with the external diffusion,

$$k_{\rm g}(c-c_{\rm s}) = D_e \frac{{\rm d}c}{{\rm d}r} \tag{7}$$

$$h_{\rm g}(T-T_{\rm s}) = \lambda_{\rm eff} \frac{dT}{dr}$$
(8)

where.

$$k_{\rm g} = \frac{D_{\rm e}Sh}{d_{\rm p}} \tag{9}$$

$$Sh = 2 + 0.6S_c^{1/3}Re_p^{0.5}$$
 (10)

$$S_{\rm c} = \frac{\mu_{\rm g}}{\rho_{\rm g} D_{\rm e}} \tag{11}$$

$$Re_{\rm p} = \frac{d_{\rm p} u_{\rm gs} \rho_{\rm g}}{\mu_{\rm g}} \tag{12}$$

$$u_{\rm gs} = \left| u_{\rm g} - u_{\rm s} \right| \tag{13}$$

$$h_{\rm g} = \frac{\lambda_{\rm g} N u}{d_{\rm p}} \tag{14}$$

$$Nu = 2 + 1.1Pr^{1/3}Re_p^{0.6}$$
(15)

$$Pr = \frac{C_{\rm p}\mu_{\rm g}}{\lambda_{\rm g}} \tag{16}$$

$$\lambda_{\rm eff} = \alpha_{\rm g} \cdot \lambda_{\rm g} + \alpha_{\rm s} \cdot \lambda_{\rm s} \tag{17}$$

Besides, to describe the kinetics of propylene polymerization on a Ziegler–Natta catalyst, a simple kinetics model is employed, which is the same as that used in our previous work.^[5] The applied kinetic model is as follows: Polymerization rate:

$$\Re = k_{\rm p} \cdot c \cdot \rho_{\rm cat} \cdot M \tag{18}$$



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$$k_{\rm p} = k_{\rm p}^{\rm 0} \exp\left(\frac{-E_{\rm A}}{R_{\rm gas}T}\right) \tag{19}$$

Furthermore, in order to use the single particle model accurately, the particle effectiveness factor is defined as the ratio of intraparticle reaction rate (i.e. macroscopic reaction rate) to the intrinsic reaction rate based on average volume (see Equation 20). If the intrinsic reaction rate is zero, the reaction effectiveness factor should be set to 1.0.

$$\eta = \begin{cases} \frac{\text{intraparticle reaction rate}}{\text{intrinsic reaction rate}} & (\text{at instrinsic reaction rate} > 0) \\ 1.0 & (\text{at instrinsic reaction rate} = 0) \\ (20) \end{cases}$$

2.2.2. Concurrent Coupling Mechanism

A concurrent coupling approach developed at Luo's group^[24] was adopted. It means that these key variables at different scales (i.e. the CFD model at the macro-scale for solving the flow field, the PBM at the particle scale for solving the particle diameter, and the single particle model at this micro-scale for solving the reaction rate) were solved using different models and the variable data obtained were transferred concurrently between different models.^[29–31] The detailed description for the coupling of different models is as follows:

After initialization, the pressure, temperature, species mass fraction distributions in the FBR at the initial stage are set and, the CFD model is solved to obtain the macro-scale flow field data (i.e. the particle volume fraction, temperature, velocity and particle size). Next, the flow field data obtained via solving the CFD model is transferred from the CFD model to the single particle model. Accordingly, the single particle model can be solved to obtain the reaction rate data, which are transferred from the single particle model to the PBM. Meanwhile, the PBM can be solved to obtain the new particle size and PSD data, which are transferred from the PBM to the CFD model. Based on the transferred particle size and PSD data, the next iterative solution of the CFD model can be performed. Finally, the three models are coupled with each other concurrently and iteratively. The solution program is executed in a loop with the above solution steps until the reaction time meets a given criterion.

2.2.3. Simulation Conditions and Modeling Method

The simulated results are dependent on the range of parameter values presented in the multi-scale model. Most of the parameters are linked to the properties of the gas and solid phases in the reactor. Some model parameters were reported in our previous works.^[2,5,25] The kinetic parameters were also reported in Yan et al.'s work.^[5] The previous parameter values^[2,5,25] were still used herein. Unless otherwise noted, the parameters used for the forthcoming simulations are those in Tables 1 and 2.

The 2D simulations based on the multi-scale model were performed with the industrial CFD code FLUENT 6.3.26 (Ansys Inc., US) in double precision mode. A commercial grid-generation tool, GAMBIT 2.3.16 (Ansys Inc., US) was used to generate the 2D geometries and the grids. Grid sensitivity was carried out initially and the results indicated that a total amount of 15 520 cells was adequate to conserve the mass of solid phase in the dynamics model.^[2,5,25] In addition, the governing equations in the single particle model were solved by the orthogonal collocation method coupled with the Newton method. The equations and the source terms of the single particle

Table 1. The boundary conditions and model parameters for the model.

Descriptions	Values	Descriptions	Values
Gas density	$21.56 \mathrm{kg} \mathrm{m}^{-3}$	m ⁻³ Granular temperature	
Polypropylene density	910 kg m $^{-3}$	Restitution coefficient	0.9
Gas viscosity	$1.081 \times 10^{-5} \operatorname{Pa} s$	Granular viscosity	[32,33]
Operating pressure	$1.4 imes 10^6$ Pa	Granular bulk viscosity	[34]
Drag law	Gidspow	Frictional viscosity	[35]
Inlet boundary condition	Velocity inlet	Angle of internal friction	30°
Outlet boundary condition	Pressure outlet	Initial bed height	0.2 m
Wall boundary condition	No slip for gas phase and	Initial solid packing	0.6
	free slip for solid phase		
Turbulent kinetic energy	$6.87 \times 10^{-4} \text{ m}^2 \text{ s}^{-2}$	Convergence criteria	$1 imes 10^{-3}$
Turbulent dissipation rate	$1.28 \times 10^{-4} \text{ m}^2 \text{ s}^{-3}$	Time step	1×10^{-3}
		Maximum iterations	50



Values	Descriptions	Values
$0.2 \mathrm{mol}\mathrm{kg}^{-1}$	De	10–10 $m^{-2} s^{-1}$
9 700 mol m^{-3}	λ_{g}	$0.0454 \mathrm{W}\mathrm{m}^{-1}\mathrm{K}^{-1}$
$2 840 \text{kg} \text{m}^{-3}$	λ_s	$0.12{\rm Wm^{-1}K^{-1}}$
85 830 J mol $^{-1}$		
	Values 0.2 mol kg ⁻¹ 9 700 mol m ⁻³ 2 840 kg m ⁻³ 85 830 J mol ⁻¹	Values Descriptions $0.2 \mod kg^{-1}$ De $9 700 \mod m^{-3}$ λ_g $2 840 \ kg \ m^{-3}$ λ_s $85 \ 830 \ J \mod^{-1}$ λ_s

Table 2. The parameters for propylene single particle model.

model and the PBM were incorporated into the CFD model via external user defined functions (UDFs). In additional, for the CFD simulation, the sub-relaxation iteration method was used to ensure the simulations converge and the simulations were executed on an Intel 2.83 GHz Xeon with 8 GB RAM platform.

3. Results and Discussion

3.1. Model Validation

In general, a model and approach should be validated with accurate and detailed experimental data. However,

Table 3. Main simulation and experiment conditions and results for model testing.^[5:37]

Particle property data used in simulation and experiment

	Small particles	Large particles			
Diameter	$1.52 \times 10^{-3} \text{ m}$	$2.49 imes 10^{-3}$ m			
Density	2523 kg m $^{-3}$	2526 kg m^{-3}			
Collision parameters for particle-particle collision data used in simulation and experiment					
Coefficient of normal restitution	0.97	0.97			
Coefficient of friction	0.15	0.10			
Collision parameters for particle-wall collision data used in simulation and experiment					
Coefficient of normal restitution	0.97	0.97			
Coefficient of friction	0.15	0.09			
FBR configuration data used in simulation and	1 experiment				
Bed height	0.7	m			
Bed width	0.15	5 m			
Initial bed height	0.15 m				

Comparison between simulation and experiment data

	Bed expansion height of the large particles (m)		Bed expansion height of the small particles (m)			
Time [s]	Simulation		Experiment	Simulation		Experiment
	Previous model	This model		Previous model	This model	
0-10	0.0853	0.0845	0.0825	0.0975	0.0973	0.0958
10-20	0.0810	0.0800	0.0789	0.1225	0.1200	0.1037
20-30	0.0802	0.0795	0.0771	0.1308	0.1312	0.1106
30-40	0.0766	0.0750	0.0738	0.1338	0.1300	0.1152
40-50	0.0732	0.0743	0.0717	0.1387	0.1403	0.1211
50-60	0.0719	0.0705	0.0702	0.1444	0.1412	0.1238





obtaining the experimental data is very difficult.^[36,37] Up to now, some hydrodynamic data, such as the flow field data in the polymerization FBR, could not be accurately obtained via experiment. Nevertheless, Goldschmidt et al.^[37] have obtained some flow field data experimentally in a cold-flow, pseudo 2D laboratory scale FBR. These experimental data under cold-flow condition were also used to verify our previous multi-scale CFD model,^[5] i.e. the CFD–PBM–PMLM model. As described above, the multi-scale CFD model is analogous to that reported in our previous work^[5] with some improvements. The key difference between the two models is the multi-scale coupling method. Therefore, Goldschmidt et al.'s experimental data^[37] were first used to test the new multi-scale model.

The main simulation conditions and quantitative data for the simulation and experiment are listed in Table 3. In addition, the simulation results obtained via our previous model are also listed in Table 3. As a whole, the simulation results from our previous and present models described in Table 3 are in qualitative agreement with the results obtained from Goldschmidt et al.'s experiments.^[37] In addition, the predicted results obtained from this work are slightly better than those from the previous.

3.2. Model Identification

Three cases including the CFD coupled with the PBM without the single particle model (Case 1), the CFD–PBM coupled with the single particle model without consideration of external diffusion (Case 2) and the multi-scale CFD model with consideration of external diffusion (Case 3) were developed to examine the model. The simulation results and their comparison in Cases 1–3 are described step by step as follows.

First, we recorded the total PSD changes in the FBR at different polymerization time points in Cases 1-3 (see Figure 3).

As shown in Figure 3, the widths of all total PSDs in Cases 1-3 increased with the extent of polymerization. During the initial polymerization period, both effects of external and intraparticle diffusions were not obvious. As the polymerization progressed, due to the existence of intraparticle heat diffusion limitation, the intraparticle temperature increased, which led to the increase of the effective reaction rate; namely, the single particle growth rate increased. This means that mean particle sizes with intraparticle diffusion (Cases 2 and 3) were larger than those without intraparticle diffusion (Case 1) in the later polymerization period (see Figure 3). According to the fragmentation and aggregation kernel equations (see ref.^[2]), the large particles were easy to fragment and in turn aggregate into larger particles, which also led to the increase of the final particle size. These different



Figure 3. The total PSD changes in the FBR at different time points.

particle kinetic behaviors (i.e. particle growth, particle fragmentation and aggregation) in Cases 1–3 led to different particle sizes and PSDs. Therefore, the resultant mean particle sizes and total PSDs in different cases are different due to the addition of the single particle model, which is further described in Figure 4.

Figure 4 shows that the mean particle sizes in the FBR change with polymerization time in Cases 1–3. Indeed, both the calculated mean particle sizes in Cases 2 and 3 are larger than that in Case 1, which further demonstrates that the addition of the single particle model altered the particle kinetics, especially particle growth, as shown in



Figure 4. The mean particle size changes in the FBR at different time points.



Figure 3. Nevertheless, the difference in Cases 1 and 2 or 3 is not obvious. On the other hand, the mean particle size in Case 3 is nearly equal to that in Case 2. This means that the simulated external diffusion limitation is negligible. In practice, there are breakage of large particles and aggregation of small particles in the FBR. Namely, particle growth must result from a "steady-state" between those two processes, which leads to the small differences in mean particle size in Cases 1–3. In addition, the absence of appropriate experimental parameter values may be one of the causes for the above small differences in particle size.

In order to further demonstrate the differences in particle behaviors in Cases 1–3 in the FBR, we recorded the particle size, volume fraction and effectiveness factor distribution profiles (see Figures 5–7).

Figure 5 shows the particle diameter distributions in the FBR at different times in Cases 1–3. When considering external and intraparticle diffusions, both intraparticle temperature and reaction rate increased, leading to an increase in particle effectiveness factor and particle size. This means that both total particle diameters in Cases 2 and 3 are larger than that in Case 1 (see Figure 5). In addition, from Figure 5, all particle distributions in the three cases were relatively uniform in the initial polymerization period. Due to different particle kinetics in Cases 1-3, both particle separations and particle diameter distribution differences increased with the extent of polymerization. For the particle separation phenomena, particles with small diameters accumulated in the upper part of the FBR and large particles tended to deposit at the bottom of the FBR in Cases 1–3. As a whole, Figure 5 demonstrates that intra-



Figure 5. The PSDs in the FBR at different time. a) Simulation results of Case 1, b) Simulation results of Case 2, c) Simulation results of Case 3.

particle polymerization kinetics had important effect on the flow field.

Figure 6 further describes the particle separation behavior using particle volume fraction distribution in the FBR in Cases 1–3. Indeed, although intraparticle polymerization kinetics had important effect on the flow field, the differences in particle volume fraction distribution in Cases 1–3 are still small. In addition, from Figure 6, both particle volume fraction distributions in Cases 2 and 3 are more uniform and concentrated than that in Case 1. As described earlier, the small differences in Cases 1-3 may be due to the absence of appropriate experimental parameter values.

Figure 7 shows the particle effectiveness factor distribution profiles in the FBR in Cases 2 and 3. According to the definition of effectiveness factor (see Equation 20), the effectiveness factor is set as 1.0 when the single particle model is neglected. In other words, the particle effectiveness factor is 1.0 at any position in the FBR in Case 1. Therefore, the factor distribution profile in







Figure 6. The particle volume fraction distributions in the FBR at different time points. a) Simulation results of Case 1, b) Simulation results of Case 2, c) Simulation results of Case 3.

Case 1 was not presented. From Figure 7, there is an obvious difference in effectiveness factor distribution profile in Cases 2 and 3. Since external diffusion limits mass and heat transfer, the limited heat and mass transfer will cause the effectiveness factor to deviate







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from 1.0. As described in Figure 7, both the maximum and minimum particle effectiveness factor values in Case 3 are indeed higher than those in Case 2 (see Figure 7), which demonstrates that external diffusion has important effect on particle reaction. This leads to the larger range of effectiveness factor in Case 3 than that in Case 2. This means that the non-uniform particle effectiveness factor distribution in Case 3 is more obvious than that in Case 2.

3.3. The Effect of Intraparticle Transfer on the Particle Behavior

It is well known that intraparticle transfer limitations are determined via the formulation of the transfer coefficients^[38–40] and the initial PSD in the FBR. Since the transfer coefficient value can be influenced by mean particle size, herein the multi-scale model in Case 3 was used to simulate the effect of initial particle size on particle behavior in the FBR.

Figures 8–12 compare the particle behavior in the FBR for cases with different initial mean particle sizes of nm respectively. Here we also point out

0.1, 0.2, and 0.3 mm, respectively. Here, we also point out that the initial PSDs are the same in the three cases.

Figure 8 illustrates the total PSD changes in the FBR with the polymerization proceeding with different initial particle sizes. There exists a big difference for the length

number density in the three cases with three initial mean particle sizes, which means that the initial particle size has a significant effect on the total PSD in the FBR (see Figure 8). In addition, as demonstrated in Figure 9, the particle growth rate increases with the increase of initial particle size, which further leads to the increase of mean particle size correspondingly. More detailed descriptions of the effect of initial particle sizes on particle behavior are further demonstrated in Figure 10–12.

Figure 10 shows the effect of initial mean particle size on the PSD in the whole FBR at different times. From Figure 10, with the increase of the initial mean particle size, the degree of uniformity of the PSD increases due to a weaker turbulence in the bigger particles. It also

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Figure 8. The effect of the initial mean particle size on the total PSD in the FBR. a) $D_p = 0.1 \text{ mm}$, b) $D_p = 0.2 \text{ mm}$, c) $D_p = 0.3 \text{ mm}$

shows that the bigger the initial particle size is, the faster the particle growth rate (also see Figure 9). In practice, this is consistent with the early description. Based on the single particle model, the intraparticle heat and mass transfer resistances increase with the increase of the initial particle size. Here, intraparticle heat transfer has the dominant role compared with intraparticle mass transfer. The higher reaction temperature within the particle leads to the increase of intraparticle polymerization rate, which further contributes to particle growth rate and the big particles



Figure 9. The effect of the initial mean particle size on the mean particle size in the FBR.

become bigger. On the other hand, the big particles tend to be deposited at the bottom of the bed, which promotes particle aggregation and leads to a direct increase of the particle size. In other words, the initial particle size can significantly influence the intraparticle polymerization rate and in turn change the particle growth rate.

Furthermore, the particle volume fraction and effectiveness factor were also computed to further demonstrate the effect of initial mean particle size on particle behavior (see Figure 11 and 12). As a whole, the initial mean particle size has a significant effect on the above two distributions. For instance, with the increase of initial mean particle size, the bed expansion decreases while effectiveness factor increases.

4. Conclusion

In this work, a multi-scale CFD model was proposed to describe the particle behavior in a propylene polymerization FBR. The multi-scale model consisted of a CFD model incorporating a single particle model and a PBM. Some open experimental data were first used to test this model. In addition, three cases including CFD coupled with PBM, CFD–PBM coupled the single particle model without consideration of external diffusion, and multi-scale CFD model with consideration of external diffusion were developed to test the multi-scale model. Based on the tested multi-scale model, the influences of intraparticle transfer limitations, which were described using initial particle size, on the particle behavior in the FBR were





Figure 10. The effect of the initial mean particle size on the PSD in the FBR. a) $D_p = 0.1 \text{ mm}$, b) $D_p = 0.2 \text{ mm}$, c) $D_p = 0.3 \text{ mm}$.



Figure 11. The effect of the initial mean particle size on the particle volume fraction distribution in the FBR. a) $D_p = 0.1 \text{ mm}$, b) $D_p = 0.2 \text{ mm}$, c) $D_p = 0.3 \text{ mm}$.





Figure 12. The effect of the initial mean particle size on the particle effectiveness factor distribution in the FBR. a) $D_p = 0.1 \text{ mm}$, b) $D_p = 0.2 \text{ mm}$, c) $D_p = 0.3 \text{ mm}$.





investigated numerically. Comparisons of the results gave the following conclusions:

- The model testing showed that the multi-scale model was appropriate for simulating particle behavior in propylene polymerization FBRs.
- (2) The simulations demonstrated that intraparticle mass transfer had significant effect on the particle behavior while the effect of intraparticle heat transfer could be neglected in the propylene polymerization FBR.
- (3) There existed a big difference in particle behavior with different initial mean particle sizes. In this regard, intraparticle heat transfer had the more dominant role compared with intraparticle mass transfer. The initial particle size had a significant effect on particle behavior in the polymerization FBR.

As a whole, the main contribution of this work was to demonstrate that a multi-scale model could better capture the phenomena of intraparticle transfer and reaction, particle kinetics, and gas-solid flow behavior in polymerization FBRs. This work also presented new insights into the fundamental mechanisms from the meso-scale viewpoint for gas-solid propylene polymerization. Therefore, the multi-scale model could produce more detailed and realistic reactor behaviors and would be very helpful towards multi-scale reactor modeling. In addition, we also point out that some things such as the dynamic sorption/desorption of monomer are important in a realistic situation and these have been neglected in the multi-scale CFD model. From the simulation viewpoint, up to now, no open model could integrate all transport and reaction phenomena involved in the particle behavior in FBRs due to its complexity. And the current work for modeling the particle behavior aim to explore the complexity forward step by step. The more realistic CFD model will be suggested in our future work.

5. Nomenclature

- c component concentration (kmol m⁻³)
- $C_{\rm p}$ mass heat capacity (J kg⁻¹ K⁻¹)
- $d_{\rm p}$ catalyst particle diameter (m)
- $D_{\rm e}$ Effective mass diffusion coefficient (m² s⁻¹)
- $h_{\rm g}$ heat transfer coefficient (W m⁻² K⁻¹)
- $h_{\rm mf}$ initial bed height (m)
- ΔH molar reaction heat (J mol⁻¹)
- $k_{\rm g}$ mass transfer coefficient (m s⁻¹)
- ΔP bed pressure drop (Pa)
- *r* coordinate inside single particle (m)
- \Re polymerization reaction rate (mol m⁻³ s⁻¹)
- T temperature (K)

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 u_{g} apparent gas velocity (m s⁻¹)

- $u_{\rm s}$ catalyst flow velocity (m s⁻¹)
- $u_{\rm gs}$ phases slip factor (m s⁻¹)
- $\rho_{\rm s}$ density of solid phase (kg m⁻³)
- $\rho_{\rm g}$ density of gas phase (kg m⁻³)
- $\alpha_{\rm g}$ volume fraction of gas phase
- $\alpha_{\rm s}$ volume fraction of solid phase
- Sh Sherwood number
- Sc Schmidt number
- *Re* Reynolds number
- Pr Prandtl number
- λ_{eff} effective thermal conductivity coefficient (W m⁻¹ K⁻¹)
- λ_g thermal conductivity coefficient of gas (W m⁻¹ K⁻¹)
- λ_s thermal conductivity coefficient of solid (W m⁻¹ K⁻¹)

5.1. Superscript

- p catalyst particle
- *s* the outer surface of catalyst particle

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