Multiple Scales Modelling of Chemistry and Mass Transfer in a SCR Process for Plate-Type Monolithic Catalysts

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Abstract. The modelling method of chemistry and mass transfer process was studied for plate-type SCR catalysts. Single- and multi-channel scale models were established for a monolithic plate-type SCR reactor, and the simulation results were compared with the experiments over a micro-scale reactor to verify the accuracy. It was found that good agreement are achieved between the results of multi-channel model and the experiments, whereas a lower NO_x reduction efficiency and a higher Sherwood number were calculated by single channel model. This illustrates that the symmetric boundary assumptions in single channel model might be not reliable, especially when the flow distribution at the reactor entrance is not uniform. The multi-channel model in consideration of channels interaction is more suitable for engineering applications.

1. Introduction

Selective Catalytic Reduction is a mature denitration technology, which is widely applied in coal-fired boilers. It uses ammonia to react with nitrogen oxides and transfer it into harmless water and nitrogen on catalyst surface. Currently, more and more researchers [1-2] have focused on the modeling of the detailed physical and chemical process within the catalysts, which help to evaluate the catalytic performance and make design decisions.

Monolithic reactor modeling can be simulated in consideration of fluid flow, gas diffusion, and intrinsic kinetic reaction. 3-D models will be established, and single channel scale modeling is most extensively applied at present. Researchers [3-5] assume that every channel in the monolithic reactor behaves exactly the same, thus the reactor can be simplified as a representative channel, then it can obtain the boundary condition that in the middle of the gas channel and the catalyst body the NO_x concentration gradient equals to zero, this conclusion can further deduce the expression of the effectiveness factor, which multiplies by the intrinsic kinetic rate to simulate the actual reaction process. However, Chen et al. [6] reviewed that if flow distribution at the entrance of the monolithic reactor is not uniform, the results from single channel model might be not reliable.

In this paper, models at multiple scales were established for plate-type SCR catalysts. Single- and multi-channel models were developed and compared for a monolithic plate-type SCR reactor. The simulation results were validated by the experiments over a micro-scale reactor.

2. Modeling of monolithic reactor

2.1. Experiments over a micro-scale reactor

The experiments over a micro-scale reactor were performed for numerical validation. Fig.1 (a) was the experimental setup, In the experiments, the inlet gas was composed of NO 135 ppm, NH$_3$ 135 ppm, O$_2$ 3.6% and the balance gas N$_2$, the gas flow rate was 12.4L/min, and reaction temperature was set from 230℃ to 400℃. As displayed in Fig.1 (b), the micro-scale reactor has 6 gas channels and 5 plate-type catalysts, the opening size of the channels is 3 mm × 22 mm, the thickness of the catalysts is 0.8mm, each catalyst has a 0.4 mm metal mesh inside it. As shown in Fig.1 (c), the catalyst plate is divided into 2 parts, the coated catalyst layer and the metal zone. The coated catalyst layer contains 100 percent catalyst and the metal support 60.87 percent catalyst and 39.13 percent metal mesh.
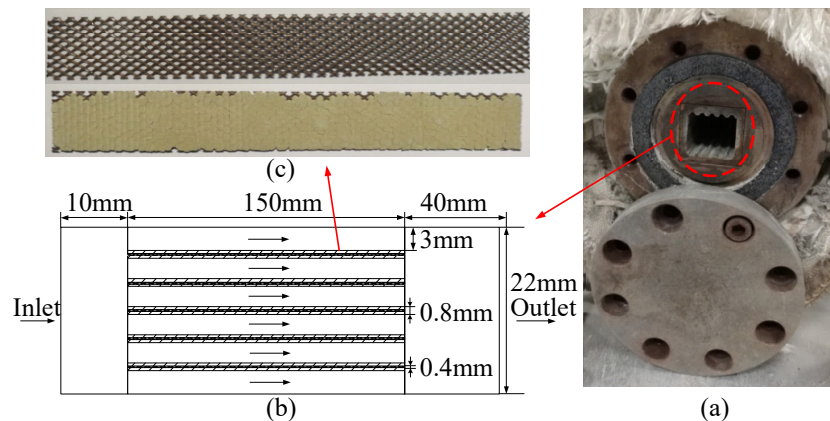

Figure 1. (a) the experimental setup; (b) the schematic of the reactor; (c) the plate-type catalyst.

2.2. Single channel and Multi-channel model

For the single channel model, it is assumed that all channels in the monolithic reactor have no interactions with each other, monolithic reactor can be modeled by calculating the gas channels one by one. According to this assumption, single channel model has the boundary condition that the NO concentration gradient equals to zero in the center of gas channel and catalyst body.
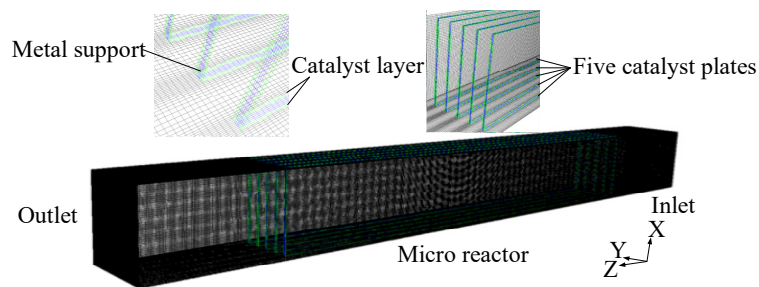

Figure 2. The 3D mesh of the multi-channel model.

Multi-channel model directly calculates the whole monolithic reactor with all channels coupled with each other. The 3D reactor model was built by using GAMBIT 2.4.6, as shown in Fig. 2. In both Single channel and Multi-channel model, the entrance was set as velocity inlet, the exit was pressure outlet, the coated catalyst layer and the inside metal support zone were separately set as two porous zones, in which mesh refinement was conducted. The inlet gas velocity and components concentration of single channel model were set as same as multi-channel modeling.

2.3. Single channel and Multi-channel model

The main mathematic models include continuity, momentum, energy and species transport equations, which can be described as follow:

$$\frac{\partial(\rho\phi)}{\partial t}+\mathrm{div}(\rho u\phi)=\mathrm{div}(\Gamma\, grad\,\phi)+\mathrm{S}_{\phi} \tag{1}$$

The species diffusion coefficients in gas channels can be calculated by Fuller equation:

$$De_{c,i} = \frac{1.43 \times 10^{-7} T^{1.75}}{P\left[(\Sigma V)_i^{1/3} + (\Sigma V)_{N_2}^{1/3} \right]^2} \left(\frac{1}{M_i} + \frac{1}{M_{N_2}} \right)^{1/2} \qquad i = NO, NH_3, O_2 \tag{2}$$

In the porous catalyst phase, including the coated catalyst layer and the metal support zone, viscous resistance and inertial resistance were added to the momentum source, which can be written as:

$$\Delta P/L = -(K\mu u) + C_2 \frac{1}{2} \rho u^2 \tag{3}$$

Because the catalysts consist of numerous tiny catalyst particles, K and C_2 can be calculated by the Ergun semi-empirical formula:

$$K = \frac{150}{D_p^2} \frac{(1-\gamma)^2}{\gamma^3} \tag{4}$$

$$C_2 = \frac{3.5}{D_p^2} \frac{1-\gamma}{\gamma^3} \tag{5}$$

The diffusion coefficient in the porous catalyst phase can be computed by Knudson formula:

$$De_r = (d/3) \times (8RTM/\pi)^{0.5} \times (\gamma/\tau) \tag{6}$$

The molecular weight of the fluid M can be written as:

$$M = \sum_i n_i M_i \qquad i = N_2, O_2, NO, NH_3 \tag{7}$$

In the coated catalyst layer, the source terms of NO and NH_3 can be calculated by Eq. (13) in consideration of the intrinsic kinetic reaction, the reaction rate r can be written as Eq. (3)-(5).

$$S_{w_i} = M_i r_i \qquad i = NO, NH_3 \tag{8}$$

In the metal support zone, the reaction rate of NO and NH_3 would multiply by a correction factor of catalyst volume fraction, thus the source terms of NO and NH_3 can be written as:

$$S_{w_i} = M_i (V_{cat} \cdot r_i) \qquad i = NO, NH_3 \tag{9}$$

The main parameters in Eq. (1)-(9) are given in Table. 1:

Table 1. Parameters of governing equations in Eq. (1)-(9).

D_p	γ	τ	d	$(\Sigma V)_{NO}$	$(\Sigma V)_{NH3}$	$(\Sigma V)_{N2}$	V_{cat}
1e-06	0.423	4	1.2e-08	23	20.7	17.9	0.609

Fluent 14.5 was used for simulation, the intra- and inter-diffusion coefficients and the reaction model were implemented into Fluent by using the diffusivity and source macros.

3. Comparisons of monolithic reactor models at different scales

3.1. Comparison of NO reduction efficiency

Fig.3 shows that there are good consistence between the results of multi-channel model and the experiments, which are obviously higher than the NO_x reduction efficiency in the case of single-channel model. As shown in Fig. 4(a), the gas velocity in 2 lateral channels (channel 1 and 6) is higher than in 4 internal channels (channel 2 to 4), this can attribute to the fact that the inlet gas flow contacts

only one catalyst plates before entrancing the lateral channels, whereas the flue gas before the internal channels are blocked by the catalyst plates in both two sides. In such non-uniform flow circumstances, the gas-phase NO concentration in the lateral channels becomes higher than in the internal channels, and this NO distinction can be more obvious in the results of single channel model.
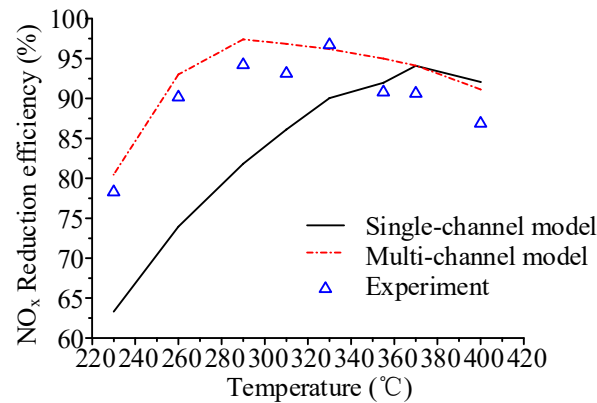

Figure 3. The comparison between the experiment and simulation results.
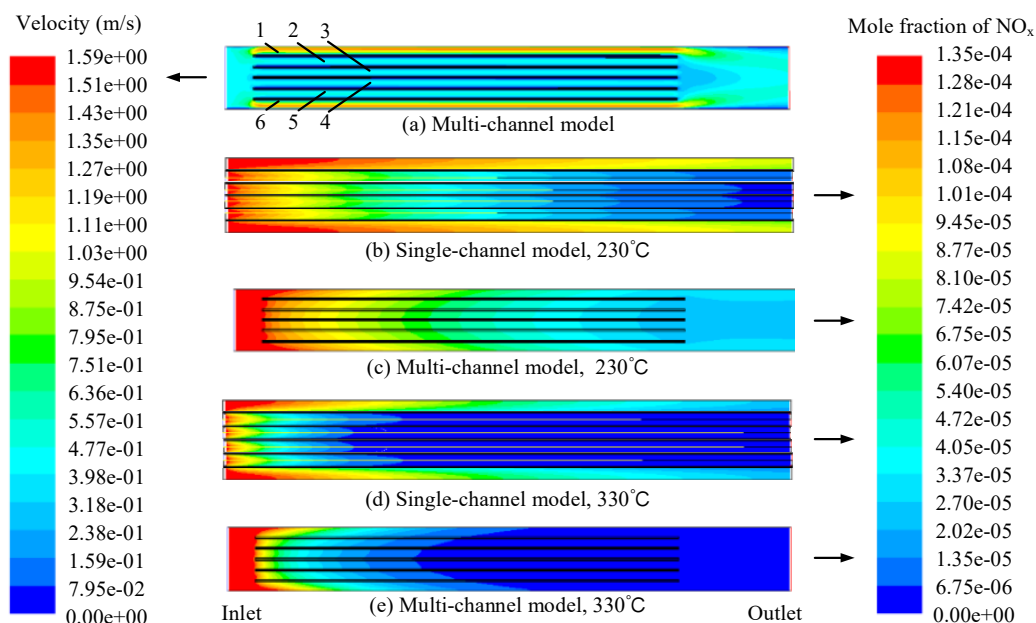

Figure 4. The distribution of gas velocity and NO concentration calculated by single- and multi-channel model.

As shown in Fig. 5, although the outlet dimensionless NO concentration in channel 2 and 3 are almost 0.07 and 0.04 according to single channel modeling, the outlet NO concentration in channel 1 is 0.65. On the other hand, the results of multi-channel model show that the outlet NO concentration in channel 1 to 3 are separately 0.2, 0.19 and 0.18, the differences of outlet NO concentration in different channels are much smaller. This illustrates that the NO reduction performance might be calculated lower by single channel model, due to the assumption that the interaction of different channels were ignored.
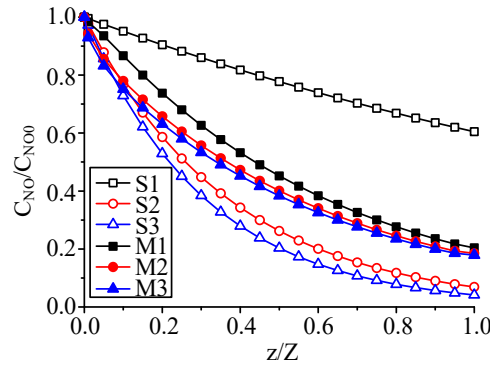
Figure 5. The dimensionless gas-phase NO concentration along Z-direction under 230℃. S1 to S3 are the results of channel 1 to 3 calculated by single channel model, M1 to M3 are the results of channel 1 to 3 calculated by multi-channel model.

3.2. Comparison of radial NO distribution

Fig. 6 gives the NO concentration along Y-direction. According to the single channel modeling, the NO concentration in the middle of the catalyst S is unequal, the reason is no coupling calculation for different channels. For the multi-channel modeling, the NO concentration at different channels can be interacted with each other by internal molecular diffusion within the catalyst body, thus the NO concentration in the middle of each catalyst body is continuous. It can be seen that unlike the assumption of single channel model, the NO distribution in bulk phase is also not symmetric, that is to say the NO concentration gradient in the middle of each gas channel is not zero.
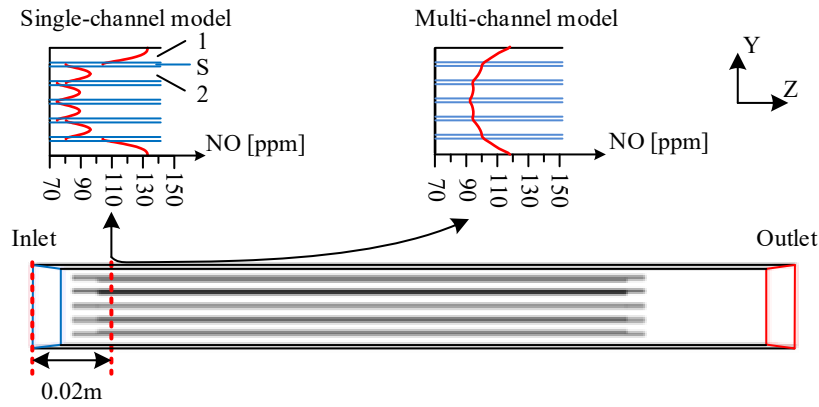

Figure 6. Calculated NO concentration along Y-direction by different model scales.

3.3. Comparison of radial external mass transfer

To evaluate the external mass transfer at the gas-solid interface of different channels, a local Sherwood number is defined as Eq. (10).

$$\text{Sh} = \frac{d_e}{\left(C_{NO}^w - C_{NO}^b\right)} \frac{\partial C_{NO}}{\partial y}\Big|y \to Y \tag{10}$$

where, d_e is the equivalent diameter of the channel, C_{NO}^w is the NO concentration between the bulk phase and the catalyst layer, the bulk average concentration C_{NO}^b can be written as Eq. (11).

$$C_{NO}^b = \left(\int_0^X \int_0^Y u(x,y) C_{NO}(x,y)\,dxdy\right)\Big/\left(\int_0^X \int_0^Y u(x,y)\,dxdy\right) \tag{11}$$

where, X and Y are the width and depth of gas channel.

As shown in Fig. 7, the external mass transfer in channel 1 is smaller than in channel 2 and 3, this is because the Sh decreased with the increase of the inlet gas velocity. On the other hand, the Sh calculated by single channel model is higher than multi-channel model, this may be attributed to the higher NO concentration gradient at gas-solid interface compared to multi-channel modeling.
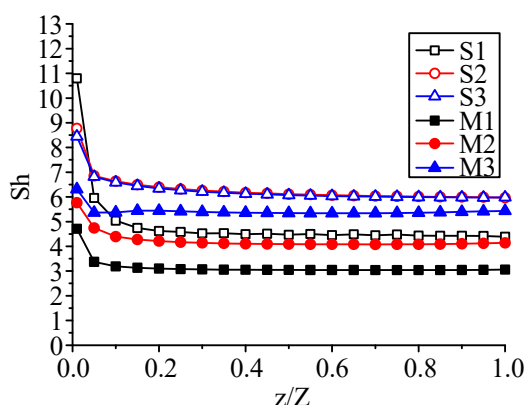

Figure 7. Sherwood number along Z-direction under 230℃. S1 to S3 are the results of channel 1 to 3 calculated by single channel model, M1 to M3 are the results of channel 1 to 3 calculated by multi-channel model.

4. Conclusion

Single- and multi-channel scale model were established for a plate-type monolithic SCR reactor, in which the flow distribution at the entrance were non-uniform. Good agreement was achieved between the results of multi-channel model and the experiments, which is obvious higher than the NO_x reduction efficiency calculated by single channel model. In consideration of the interaction between different channels, the radial NO distribution calculated by multi-channel model shows that the assumption of single channel model might not be reliable, that is to say the NO concentration gradient in the middle of each gas channel is not zero if the inlet flow distribution is not uniform. Based on this assumption, the NO concentration gradient and Sherwood number at gas-solid phase would be calculated higher by single channel model.

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