



Experimental and modeling study of selective catalytic reduction of NO_x with NH₃ over wire mesh honeycomb catalysts

Hong Sun, Yun Shu, Xie Quan*, Shuo Chen, Bo Pang, ZhaoYang Liu

Key Laboratory of Industrial Ecology and Environmental Engineering (Ministry of Education, China), School of Environmental Science and Technology, Dalian University of Technology, Dalian 116024, China

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ABSTRACT

The mass transfer of wire mesh honeycomb is investigated, and a new correlation $Sh = 1.108 Re^{0.5} Sc^{1/3}$ is proposed based on data for the mass transfer limited performance for selective catalytic reduction of NO_x by NH₃ over wire mesh honeycomb catalysts. The numerical model is developed to evaluate the performance of wire mesh honeycomb, which is verified by the experiments. By measuring the NO_x conversion over wire mesh honeycomb catalysts with different washcoat thickness and cell density, the intraporous diffusion and interphase diffusion are studied. The pressure drop of wire mesh honeycomb is low. Compared to ceramic monolith, wire mesh honeycomb catalysts possess high catalyst utilization relative to the geometric surface area and the catalyst weight.

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1. Introduction

Monolith or honeycomb catalysts have been widely applied for the treatment of exhaust gases such as afterburner for the combustion of hydrocarbons and the selective catalytic reduction of NO_x released from stationary or mobile sources. A typical monolith has a structure with parallel and usually straight channels or cells, and the catalysts are dispersed on the channels' wall. This reactor design provides a large gas–solid contact area and the low pressure drop (up to two orders of magnitude) [1]. Monolithic substrates are generally made of cordierite or metal alloys, but its impermeable walls suppress radial mixing. In monoliths, the interphase mass transfer was low [2]. Under certain circumstances the performance limit for pollutant removal of the monolith catalysts was determined by the rate of external mass transfer [3]. Therefore, the new structure catalysts should combine highly enhanced mass transfer and moderate flow resistances compared with monolith.

Recently, wire mesh catalysts had received much interest. Wire mesh catalysts are commonly used in the production of nitric acid from ammonia, and are made of pure noble metal (Pt/Rt catalyst). However, this kind of catalyst is very expensive. Currently, many non-noble wire mesh catalysts had been developed. Lyubovsky et al. reported the application of the Microlith® catalysts (consist of wire meshes) with supported Pd-based catalyst to catalytic

combustion and demonstrated this kind of catalyst had the stable performance at about 800 °C over a wide range of inlet conditions [4]. Ahlström-Silversand and Odenbrand compared the performance of wire mesh, monolith and pellets catalysts, and found that wire mesh catalysts had high mass and heat transfer numbers [5]. Mass transfer volumetric coefficients of wire gauzes were up to 90 times higher than that of standard monoliths [6]. However, wire mesh catalysts were structured by single gauzes stacked, which was inconvenient for being assembled in the reactor.

Wire mesh honeycomb that is manufactured by stacking alternatively corrugated and plain wire mesh sheets is new structure catalytic reactor, and its structure combines the characteristics of wire mesh and monolith catalyst. It had been used in catalytic oxidation of 1,2-dichlorobenzene [7], ethyl acetate [8,9], volatile organic compounds in air [10] and selective catalytic reduction (SCR) of NO_x [11].

In contrast to the study on catalytic process in the new structured catalytic reactor, little work was focused on the mass transfer characterization of wire mesh honeycomb catalysts. A perfect description of the mass transfer process is important to design the wire mesh honeycomb catalyst, and the physical size and other geometric parameters of the catalyst can be obtained by parameters of mass transfer. Jiang et al. applied the *Sh* expressions of honeycomb, gauze and packed bed to model the oxidation of ethyl acetate in wire mesh honeycomb modules, and found that the packed bed type *Sh* expression could predict the catalytic behavior satisfactorily [12].

Selective catalytic reduction of NO_x with NH₃ is used worldwide for NO_x abatement from stationary resources. The design

* Corresponding author. Tel.: +86 411 84706140; fax: +86 411 84706263.
E-mail address: quanxie@dlut.edu.cn (X. Quan).

Nomenclature

A	cross-section area of catalyst, m^2
a_{ext}	total external surface of catalyst, m^2
C_A	concentration of NO_x , mol/m^3
C_{A0}	inlet concentration of NO_x , mol/m^3
C_{As}	concentration at the external surface, mol/m^3
d_0	wire mesh diameter, cm
Da	Damköhler number
D_a	the effective Aris–Taylor dispersion coefficient, m^2/s
D_{AB}	diffusivity of pollutant, m^3/s
D_{eff}	effective diffusion coefficient, m^2/s
D_g	diffusion coefficient in the gas phase, m^2/s
D_h	hydraulic diameter, $m, D_h = (4V\varepsilon_b/a_{ext})$
D_k	Knudsen diffusion coefficient, m^2/s
E_a	activation energy, J/mol
f	the friction coefficient
J_D	j -factor for mass transfer
k_m	mass transfer coefficient, m/s
k_{ov}	overall effectiveness factor, $m^3/kg s$
k_r	intrinsic reaction rate constant, $m^3/kg s$
k_{ro}	pre-exponential constant, $m^3/kg s$
l	length coordinate, m
L	catalytic bed length, m
Pe_r	Peclet number
r_0	wire mesh semidiameter, m
r'	pore radius, m
Re	Reynolds number
S_a	specific surface area, m^2/kg
S_c	Schmidt number
Sh	Sherwood number
S_{wmh}	external surface area of wire mesh honeycomb substrate, m^2
T	temperature, K
U	the superficial velocity, m/s
v_0	feed rate, m^3/s
V	reactor volume, m^3
V_c	catalytic volume, m^3
V_p	the total pore volume of catalyst, m^3/kg
X	NO_x conversion
z	dimensionless length
ε_b	wire-mesh honeycomb void ratio
ν	kinematic viscosity, m^2/s
ρ	gas density at operating conditions, kg/m^3
ρ_c	density of catalyst, kg/m^3
ρ_{Fe}	density of the wire mesh, kg/m^3
η	the effectiveness factor
ϕ	Thiele modulus
τ	tortuosity factor

of monolith catalyst or reactors is a key technology for SCR. Constructing a model of monolith reactor for SCR of NO_x is beneficial for designing and controlling of integrated after-treatment systems.

The aim of this paper is to develop a numerical model to simulate the SCR of NO_x with NH_3 on wire mesh honeycomb catalyst so as to accurately describe the reactor performance. The modeling calculation is verified by the experimental data. The interphase and intraphase mass transfers are analyzed in order to investigate the effect of washcoat and size of the channel on the mass transfer. The performances of wire mesh honeycomb catalyst and ceramic monolith catalyst are compared in this study.

2. Experimental

2.1. Catalyst preparation

The plain and corrugated sheets were adopted, and they were stacked alternately to construct wire mesh honeycomb. It consisted of arrays of parallel channels with the porous wall, and the shape of the channels was sinusoidal. The wire mesh used for manufacturing wire mesh honeycomb had a wire diameter (d_0) of 0.13 mm.

Because the surface areas of metal wire are very low and the catalysts cannot be dispersed directly, it is necessary to deposit the ceramic oxide washcoat with a high surface area over the wire mesh substrate. Electrophoretic deposition (EPD) method had been employed to deposit alumina washcoat on the wire mesh substrates. The alumina suspension was prepared with γ -alumina ($\gamma-Al_2O_3$) powders gritted from γ -alumina pellets. Polycyclic acid and aluminum isopropoxide were used as additives. The detailed prepared approaches were described in previous work [13].

The wire mesh honeycomb was coated by an Al_2O_3 washcoat with a thickness of 30–70 μm , and the specific surface area of the Al_2O_3 layer (S_a) was about 87.5 m^2/g . The Al_2O_3 -coated wire mesh honeycomb was impregnated in titania sol which was prepared by hydrolysis of tetra- n -butyl titanate [$Ti(OC_4H_9)_4$]. The impregnated sample was dried at 110 °C for 3 h and calcined in air at 500 °C for 2 h. After preparing TiO_2 washcoat, the samples were impregnated into the solution of the vanadium oxalate, which was prepared from ammonium metavanadate and oxalic acid, and ammonium tungstate. Following the impregnation step, the catalysts were dried at 110 °C for 3 h and calcined at 350 °C for 2 h. The composition of catalyst was 3 wt.% V_2O_5 –7 wt.% WO_3 – TiO_2 .

2.2. Catalytic tests

The catalytic studies were carried out in a continuous tubular fixed-bed reactor which was a square-shape reactor (40 mm \times 40 mm in width) made of stainless steel with a total bed length of 1000 mm equipped with a temperature controller. The size of wire mesh honeycomb catalyst was 38 mm \times 38 mm \times 20 mm. The reaction gas mixture consisted of 900 ppm NO , 1000 ppm NH_3 and 3% O_2 in a N_2 balance. The reactant gases flowed into the reactor and the activity measurements were performed in the temperature range from room temperature to 450 °C. The experiments were normally performed in the mass transfer controlled region, where the observed reaction rate had exceedingly weak dependence on temperature.

On the basis of the Eley–Rideal mechanism between adsorbed NH_3 and gaseous (or weakly adsorbed) NO , the simple expression was adopted to describe the reaction rate of NO on the catalyst surface, in line with the literature on SCR monolith catalysts [14,15]:

$$r_{NO} = k_{NO} \frac{k_{NH_3} C_{NH_3}}{1 + k_{NH_3} C_{NH_3}} \quad (1)$$

Basing on the characteristics of our catalytic system and a preliminary analysis, the assumptions were made: (1) the reaction takes place under isothermal conditions due to very low concentration of NO and NH_3 . (2) The kinetic dependence on O_2 could be neglected for O_2 feed concentrations in excess of 2% (v/v) [15]. (3) For the overstoichiometric feed ratios ($NH_3/NO > 1$) and large amount of oxygen, the SCR reaction could be considered to a first-order with respect to NO and zero-order with respect to NH_3 [16,17].

2.3. Model description

2.3.1. Reactor model

The simulation of a wire-mesh honeycomb can be performed using models taking axial dispersion into consideration.

In this study one-dimensional catalyst model was selected. The assumption of a single channel model implied uniform catalyst distribution, a plug flow and an adiabatic reactor. In developing the model of wire mesh honeycomb reactor the axial dispersion was taken into account. Therefore, for the case of isothermal first-order reaction, one-dimensional two-phase model is

$$\frac{1}{Pe_r} \times \frac{d^2X}{dz^2} - \frac{dX}{dz} + Da(1 - X) = 0, \quad z = \frac{1}{L}, \quad X = \frac{C_A}{C_{A0}} \quad (2)$$

The conversion (X) can be determined by solving the second-order ordinary differential equation. Eq. (2) is solved numerically and the isothermal solution for a first-order reaction is given in Eq. (3):

$$X = 1 - \frac{4q \exp(Pe_r/2)}{(1+q)^2 \exp(Pe_r q/2) - (1-q)^2 \exp(-Pe_r q/2)},$$

$$q = \sqrt{1 + \frac{4Da}{Pe_r}} \quad (3)$$

Damköhler number (Da) is obtained according to $Da = (\omega_c k_{ov}/v_0)$, where w_c is the weight of catalyst, k_{ov} is the overall rate constant, and v_0 is the volume flow. Peclet number $Pe_r = (UL/\varepsilon_b Da)$ is dependent on the axial dispersion coefficient (effective Aris–Taylor dispersion coefficient) $D_a = D_{AB} + (U^2(D_h/2)^2)/48D_{AB}$, the superficial gas velocity (U) and the catalytic bed length (L).

2.3.2. The mass transfer coefficient and J_D (j -factor for mass transfer)

A simple balance of NO_x over wire mesh honeycomb catalyst was described with the boundary condition of zero NO_x concentration at the wall. The mass transfer coefficient (k_m) can be calculated from the conversion of NO_x obtained in the totally mass transfer controlled region according to Eq. (4):

$$k_m = -\frac{v_0}{a_{ext}} \ln(1 - X_{exp}) \quad (4)$$

j -factor for mass transfer was calculated from the conversion according to Ahlström–Silversand and Odenbrand [5]:

$$J_D = -\frac{\varepsilon_b A}{a_{ext}} \ln(1 - X_{exp}) \left(\frac{v}{D_g}\right)^{2/3} \quad (5)$$

where $J_D = Sh/Re Sc^{1/3}$, $Re = D_h U/\varepsilon_b v$.

2.3.3. Model parameters

For first-order isothermal system, the overall effectiveness factor, k_{ov} , which reflects the integrated effect of the pore diffusion, interphase mass transfer and intrinsic reaction, is defined as

$$\frac{1}{k_{ov}} = \frac{1}{k_m a_{ext}/w_c} + \frac{1}{\eta k_r} \quad (6)$$

where a_{ext} is the specific gas–solid interphase area, η is the effectiveness factor and k_r is intrinsic reaction rate constant. For first-order reaction, $k_r = k_{r0} \exp(-E_a/T)$. There are two important types of mass transfer limitation in monolith reactor. The first is diffusion limitation of reactants from the gas bulk into surface of the washcoat. Another is the diffusion in the porous washcoat. The effects of interphase mass transfer and intraporous diffusion are characterized by the express $1 - C_{As}/C_A$ and the effectiveness factor (η), respectively.

The effectiveness factor is calculated from Thiele modulus:

$$\eta = \frac{3}{\phi^2} (\phi \coth(\phi) - 1) \quad (7)$$

Thiele modulus of a first-order reaction is evaluated from the intrinsic reaction rate (k_r), the catalyst density (ρ_c), the effective

diffusivity (D_{eff}), the catalyst volume (V_c) and the exterior surface area (a_{ext}):

$$\phi = \frac{V_c}{a_{ext}} \sqrt{\frac{k_r \rho_c}{D_{eff}}} \quad (8)$$

where $D_{eff} = ((1/D_g) + (1/D_k))^{-1} (\varepsilon_c/\tau)$, $D_k = 97.0r' \sqrt{(T/M)} = 194(V_p/S_a) \sqrt{(T/M)}$.

The effectiveness factor and C_{As}/C_A usually have the value between 0 and 1. The value of 1 indicates that the diffusion is not important, whilst the value less than 1 indicates that there is diffusion limitation.

2.3.4. Properties of wire mesh honeycomb catalyst

When the external surface area of the catalyst is calculated, the catalyst layer is assumed to be evenly coated on the surface of wire mesh honeycomb substrate. The external surface is then calculated from the external surface area of wire mesh honeycomb substrate (S_{wmh}), the wire mesh radius (r_0), the catalyst weight (w_c) and the density of the catalyst layer (ρ_c):

$$a_{ext} = \frac{w_c 2\pi r}{\rho_c (\pi r^2 - \pi r_0^2)} = \sqrt{\frac{2w_c S_{wmh}}{\rho_c r_0} + S_{wmh}^2} \quad (9)$$

where the external surface area $S_{wmh} = (2w_m/\rho_{Fe} r_0)$.

2.3.5. Pressure drop

Wire mesh honeycomb can be described as a sinusoidal monolith with opening wall, so the pressure drop can be calculated with the Darcy–Weisbach equation that is well-known for predicting pressure drop of monolith [5]. For Darcy–Weisbach equation, pressure drop is calculated from the superficial gas velocity (U), the length of monolith (L), the hydraulic diameter (D_h), the friction coefficient (f) and the density of the gas (ρ):

$$\Delta P = \frac{fL\rho U^2}{2D_h} \quad (10)$$

where the friction coefficient $f = (64/Re)$, $Re = (D_h U/\varepsilon_b v_i)$.

On the other hand, wire mesh can be treated as stacked wire mesh catalyst with sinusoidal mesh holes. Ergun's model that usually described pressure drop through packed bed was employed to calculate pressure drop of wire mesh catalyst by Ahlström–Silversand and Odenbrand [5]. Furthermore, Ergun equation was adjusted to meet the new geometry of stacked wire mesh by Kołdzie and Łokewska [18]. The Ergun equation commonly is given as following, where the particle diameter is replaced with hydraulic diameter of wire mesh honeycomb:

$$\Delta P = 150 \frac{GhUL}{D_h^2} \frac{(1 - \varepsilon_b)^2}{\varepsilon_b^3} + 1.75 \frac{\rho U^2 L}{D_h} \frac{1 - \varepsilon_b}{\varepsilon_b^3} \quad (11)$$

3. Results and discussion

3.1. j -Factor and Sherwood numbers

The correlation of Sh could be regressed from the experimental data. Ahlström–Silversand and Odenbrand had demonstrated that Sh expression could be predicted according to the correlations between the j -factor for mass transfer and Reynolds number calculated from experimental data [5]. In the section, the mass transfer coefficient and J_D were calculated from the experimental data obtained in the mass transfer controlled region according to Eqs. (4) and (5), and the j -factor plotted versus the Reynolds number is shown in Fig. 1. By non-linear curve fitting, the expression of

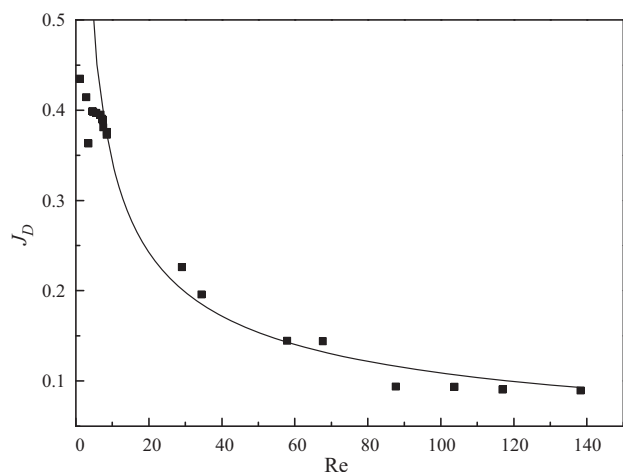


Fig. 1. Mass transfer experimental results: the j -factor versus Re for wire mesh honeycomb catalyst. Symbols presenting experimental data and the line a power-law fit to the experimental data.

a power-law fit to the experimental data is obtained:

$$j_D = \frac{1.08}{Re^{0.50}} \quad (1.1 < Re < 138) \quad (12)$$

According to $j_D = (Sh/Re Sc^{1/3})$, therefore, the Sherwood number was calculated as

$$Sh = 1.08 Re^{0.5} Sc^{1/3} \quad (13)$$

The Sh expression of wire mesh honeycomb is the same as the type of packed bed catalyst, which was consistent with the study by Jiang et al. [12].

3.2. Influence of the washcoat thickness

The catalytic activities of wire mesh honeycomb catalyst with different thickness were studied. A set of wire mesh honeycomb catalysts with different washcoat thickness was prepared with the catalytic active phase ($V_2O_5-WO_3$) kept at constant of 10 wt.%.

The experimentally obtained conversion and calculated data for NO_x catalytic reduction are presented in Fig. 2. As can be seen, the agreement between experimental and calculated data is excellent, which indicates that the model can predict well the influence of

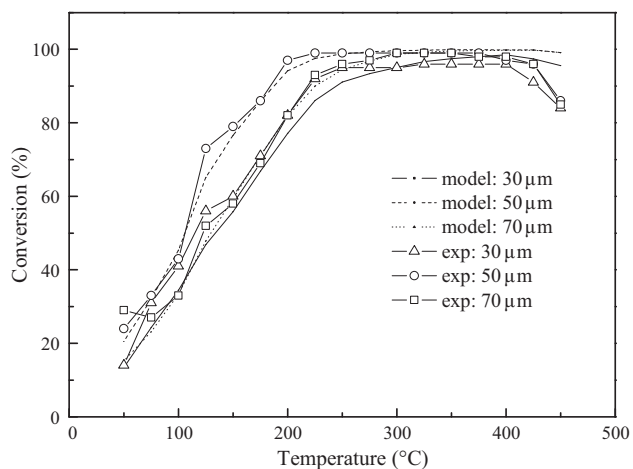


Fig. 2. Selective catalytic reduction of NO_x with NH_3 over wire mesh honeycomb catalyst with different washcoat thickness. Gas flow: $70 \text{ m}^3/\text{m}^2 \text{ h}$. The cell density: 50 cps.

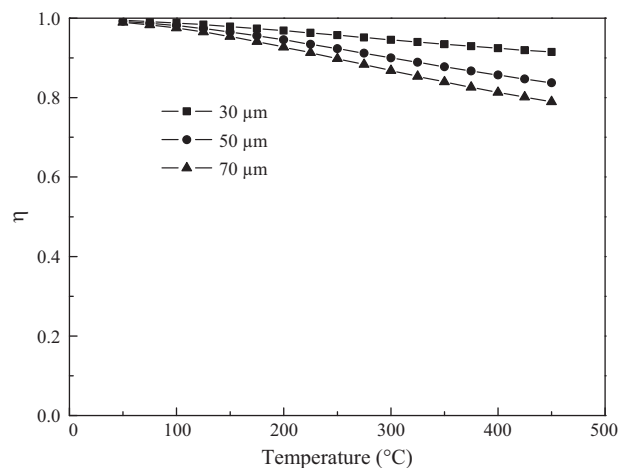


Fig. 3. Influence of intraporous diffusion in wire mesh honeycomb with different washcoat thickness. Gas flow: $70 \text{ m}^3/\text{m}^2 \text{ h}$. The cell density: 50 cps.

washcoat thickness on NO_x conversion over wire mesh honeycomb catalysts.

In Fig. 2, it is also found that NO_x conversion depends markedly on the washcoat thickness. The wire mesh catalyst with $50 \mu\text{m}$ exhibits the higher catalytic activity for NO_x than the catalysts with $70 \mu\text{m}$ and $30 \mu\text{m}$. The thinnest washcoat leads to the lowest NO_x conversion.

Washcoat thickness can have a significant effect on the catalyst activity [19]. The effect of washcoat on catalytic reaction has two parts: catalytic compounds loading and diffusion of reactants. For wire mesh honeycomb catalysts, it is common practice to maximize the number of active sites by dispersing the catalytic components onto the washcoat. Although the procedure maximizes the catalytic area, it introduces the mass transfer limit of the reactant to the catalytic sites. The diffusion in the catalyst/washcoat layer is significant factor influencing the catalyst activity [20].

The effectiveness factor (η) reflects intraporous diffusion, and the expression C_{As}/C_A represents the interphase mass transfer. Figs. 3 and 4 show that the effectiveness factor and C_{As}/C_A of the catalysts with different washcoat thickness, respectively. The results show that the effectiveness factors decrease with increasing the washcoat thickness, which indicates the dramatic influence of intraporous diffusion for thick washcoat, specially, at high temperature. The effectiveness factor of the washcoat of $30 \mu\text{m}$ is greater

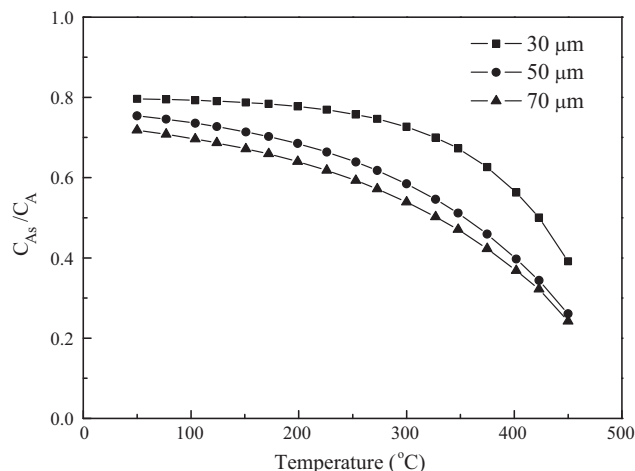


Fig. 4. Influence of interphase diffusion in wire mesh honeycomb with different washcoat thickness. Gas flow: $70 \text{ m}^3/\text{m}^2 \text{ h}$. The cell density: 50 cps.

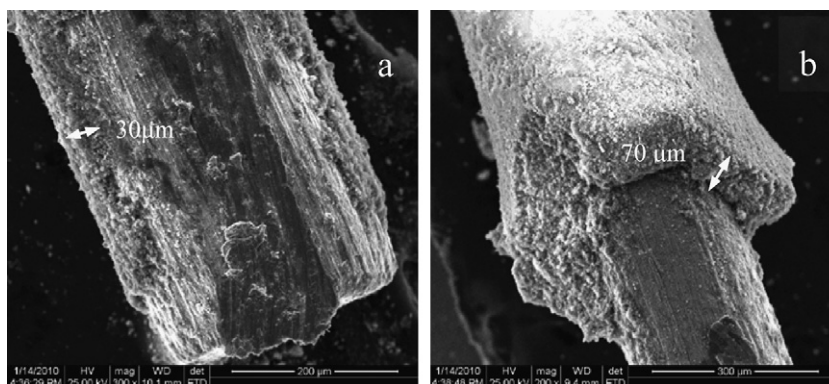


Fig. 5. SEM of washcoat over wire mesh honeycomb. (a) 30 μm ; (b) 70 μm .

than 0.9 at all temperatures. The influence of intraporous diffusion is small and can be ignored. Thus, for the wire mesh honeycomb catalysts with 30 μm , the low NO_x conversion could be associated with the catalytic sites. As shown in Fig. 5(a), SEM micrographs of wire mesh honeycomb coated Al_2O_3 washcoat with 30 μm presents that the thin washcoat is coated on the substrate, which could lead to the small loading of catalytic compounds.

In addition, the amount of catalytic active sites increases when the washcoat thickness increases. Therefore, the wire mesh honeycomb catalyst with 50 μm exhibits the higher NO_x conversion. But further increasing the washcoat thickness (in Fig. 5(b)), the effectiveness factor decreases, and the washcoat has a negative effect on NO_x conversion due to the intraporous diffusion resistance. Furthermore, for a thicker washcoat, although the amount of catalytic sites is sufficient, some sites are not accessible within the residence time of reactants [21].

On the other hand, the washcoat affects the surface concentration of reactants on the catalysts. The plots of the ratio of surface to bulk concentration (C_{As}/C_A) with temperature are shown in Fig. 4. It is apparent that the ratio decreases with increasing temperature. The catalyst with 70 μm washcoat thickness has the lowest ratio, which is in accordance with modeling results by Hayes et al. [22]. They reported that the washcoat could induce the larger variations in concentration around the surface, and the concentration on the washcoat in the corners was low where washcoat was thick. This is also one reason of low NO_x conversion for thick washcoat catalyst, and it indicates the effect of interphase diffusion resistance on catalytic activity.

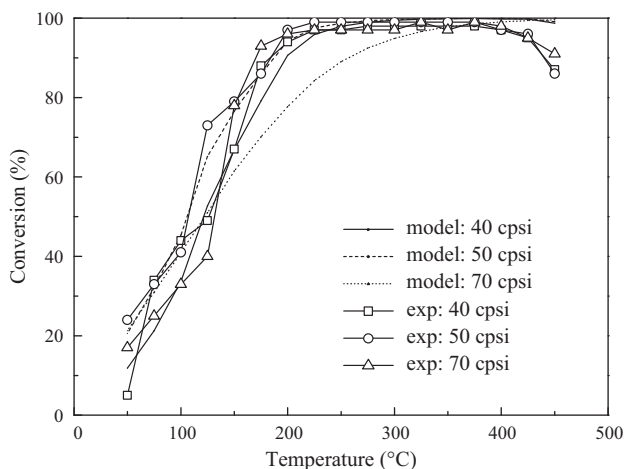


Fig. 6. Selective catalytic reduction of NO_x with NH_3 over wire mesh honeycomb catalyst with different cell density. Gas flow: $70 \text{ m}^3/\text{m}^2 \text{ h}$. Washcoat thickness: 50 μm .

3.3. Influence of the cell density

Catalytic activities of wire mesh honeycomb catalysts with cell density from 40 to 70 cpsi (the cells per square inch) are studied. All catalysts were coated with Al_2O_3 washcoat and active material was the same as that with 50 μm washcoat thickness in Section 3.2. Comparison of NO_x conversion between experimental data and the prediction obtained by one-dimensional model for wire mesh honeycomb catalyst with different cell density is shown in Fig. 6. It can be found that there is good agreement between the experimental data and theoretically predicted ones in the case of cell density 40 and 50 cpsi, whereas a deflection of calculation is observed for cell density 70 cpsi. The difference may be attributed to the assumption of a plug flow when one-dimensional catalyst model is adopted. Because the channel of wire mesh honeycomb is interconnected with each other through the openings in the wire mesh sheet, the gas feed entering the reactor can flow in three-dimensional directions, creating flow disturbance that increase mass transfer to the wall [23].

Furthermore, the different catalytic activity caused by the channel size of wire mesh honeycomb reflects the importance of interphase mass transfer. The ratio C_{As}/C_A of catalyst with different cell density is compared in Fig. 7. It shows that the surface concentration of reactant increases with increasing the cell density. The high cell density means the small cell space, and the interphase mass transfer path length for reactant from bulk gas to the surface of catalyst is short.

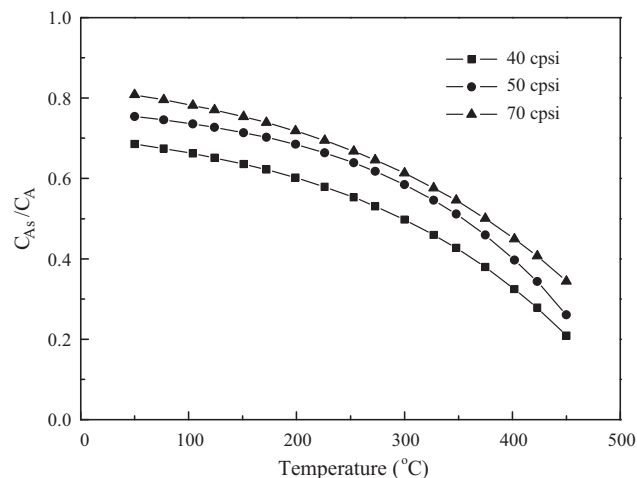


Fig. 7. Influence of interphase diffusion in wire mesh honeycomb with different cell density. Gas flow: $70 \text{ m}^3/\text{m}^2 \text{ h}$. Washcoat thickness: 50 μm .

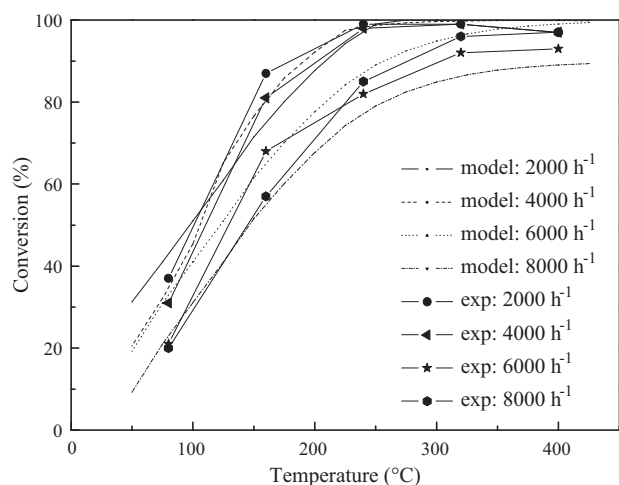


Fig. 8. Selective catalytic reduction of NO_x with NH_3 over wire mesh honeycomb catalyst at different space velocity. The cell density: 50 cpsi. Washcoat thickness: 50 μm .

3.4. Influence of space velocity (GHSV)

The effect of space velocity on the NO_x conversion is studied, and the NO_x conversion data measured were compared with the calculated data in Fig. 8. On the whole, the NO_x conversion decreases slightly with increasing GHSV. There is a good agreement between the experimental result and the predicted data at low GHSV. However, it is also important to note that there is a difference between calculation and practice at high GHSV because NO_x conversion at 8000 h^{-1} is higher than that at 6000 h^{-1} at the temperature range from 240 to 400°C .

The higher GHSV means the shorter residence time, therefore NO_x conversion decreased with GHSV. However, in the case of higher GHSV, high linear velocity could increase the gas flow disturbance caused by the porous wall to improve the mass transfer from the bulk gas to the catalyst surface.

3.5. Pressure drop

When the pollutant gas stream enters wire mesh honeycomb, a pressure drop usually develops along the reactor length. A higher pressure drop represents a loss in power. Therefore, maximum permitted pressure drops is an important design parameter for honeycomb as a part of the overall system engineering and economics.

The pressure drops are modeled with the modified Darcy–Weisbach equation and Ergun equation, respectively, and the results are compared with the experimental data. As shown in Fig. 9(a), it can be found that the Darcy–Weisbach equation is not fit for predicting the pressure drop of wire mesh honeycomb because the measure value is higher clearly than the model value. This indicated that wire mesh honeycomb could have the higher pressure drop than monolith with the same bed length and hydraulic diameter of channel.

For the porous wall of wire mesh honeycomb, it should be taken account of the presence of the convective flow across the bed diameter. The Ergun equation is often used to estimate the pressure drop of a packed bed, and the predicted value is shown in Fig. 9(b). At low flow rate, the experimentally obtained pressure drops and the calculated value by the Ergun equation had a good agreement. However, the calculated pressure drop is higher than the measured value at high flow rate. For high flow rate (high Re range), the flow through wire mesh honeycomb become turbulent flow from laminar, which could decrease the accuracy of model, as demonstrated

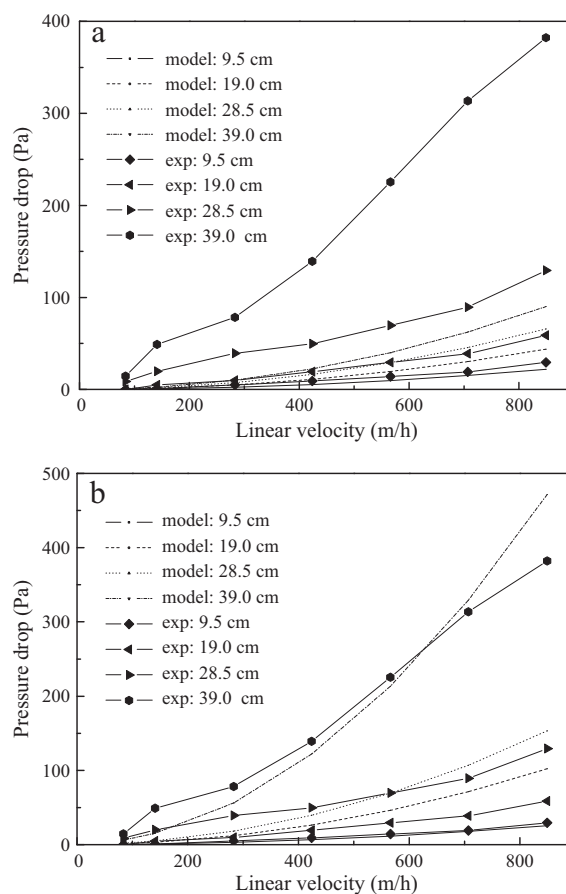


Fig. 9. Pressure drop versus flow rate. The theoretical value was calculated with Darcy–Weisbach equation (a) and with the Ergun equation (b).

by Kołdzie and Łokewska [18]. In addition, the result indicates that wire mesh honeycomb has a low pressure drop attributed to the high porosity.

3.6. Comparison of ceramic monolith and wire mesh honeycomb catalysts

In previous work, it was found that wire mesh honeycomb catalyst had higher catalytic ability than ceramic monolith with the same cell density [11]. Therefore, in this study, the catalytic activity of wire mesh honeycomb catalyst is compared with ceramic monolith catalyst with 400 cpsi, and the results are shown in Fig. 10. For both catalysts, the amount of the catalytic phase ($\text{V}_2\text{O}_5\text{--}\text{WO}_3$) was about 10 wt%. It is interesting that although wire mesh honeycomb catalyst has 50 cpsi, it exhibited the similar catalytic activity for NO_x to monolith catalyst with 400 cpsi.

It is important to evaluate the catalyst performance relative to the catalyst weight and the geometric surface area (GSA) in catalytic application. According to Ahlström–Silversand and Odenbrand [5], the $-\ln(1-X)/S_{\text{GSA}}$ and $-\ln(1-X)/w_c$ are employed to describe the catalytic performance relative to the geometric surface area and the catalyst weight, and the evaluation parameters for wire mesh honeycomb catalyst and ceramic monolith catalyst are presented in Fig. 11. The catalyst utilization for wire mesh honeycomb catalyst is obviously better than that for ceramic monolith catalyst, specially, relative to the geometric surface area. This fact is attributed to the excellent mass transfer and the high available surface area between the gas and the catalysts.

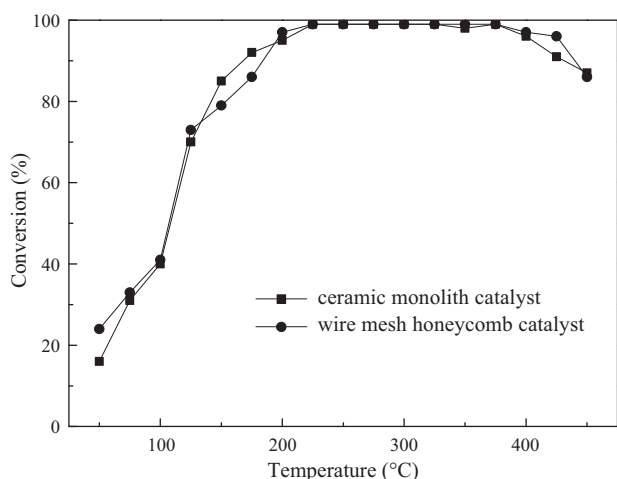


Fig. 10. Selective catalytic reduction of NO_x with NH_3 over wire mesh honeycomb and ceramic monolith catalysts. Gas flow: $70 \text{ m}^3/\text{m}^2 \text{ h}$. Wire mesh honeycomb: 50 cps; ceramic monolith: 400 cps.

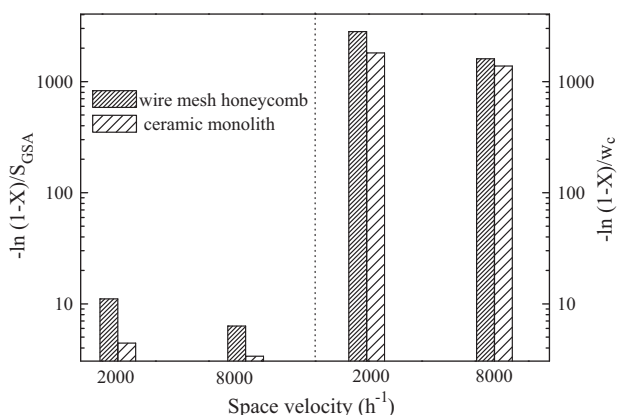


Fig. 11. The evaluation parameter $-\ln(1-X)/S_{\text{GSA}}$ and $-\ln(1-X)/w_c$ for wire mesh honeycomb catalyst and ceramic monolith catalyst. Reaction temperature: $T = 250^\circ \text{C}$. Wire mesh honeycomb: 50 cps. Ceramic monolith: 400 cps.

4. Conclusions

The mass transfer of wire mesh honeycomb catalyst is investigated experimentally by the selective catalytic reduction of NO_x with NH_3 . The expression of ($Sh = 1.08 Re^{0.5} Sc^{1/3}$) is obtained based on the measured data of NO_x conversion over wire mesh honeycomb, and it is similar to the packed bed type Sh expression. This Sh expression can predict satisfactorily the mass transfer behavior of wire mesh honeycomb, and the model predictions using a one-dimensional model agree well with the experimental data.

Both the experimental and model data confirm the optimum washcoat thickness is $50 \mu\text{m}$ in the range from $30 \mu\text{m}$ to $70 \mu\text{m}$, and the high cell density of wire mesh honeycomb benefits for NO_x removal, which are approved by the studies of the intraporous diffusion and interphase mass transfer. An optimum washcoat thickness for the wire mesh honeycomb catalyst can reduce the effect of diffusion resistance and save the catalysts. The high cell density means the short interphase mass transfer path for reactant from bulk gas to the surface of catalyst.

The pressure drop of wire mesh honeycomb is somewhat higher than that of monolith, but it is lower than packed bed, especially at high flow rate. The relatively low pressure drop is attributed to the

high porosity of wire mesh honeycomb. Furthermore, compared with ceramic monolith catalyst, wire mesh honeycomb catalyst exhibits the excellent catalytic activity for NO_x and high catalyst utilization due to high mass transfer characteristics. The combination of excellent mass transfer characteristics with a low pressure drop makes wire-mesh honeycomb catalysts perspective applied in automotive catalysts, catalytic combustion in energy production and VOCs removal.

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References

- [1] G. Groppi, E. Tronconi, Design of novel monolith catalyst supports for gas/solid reactions with heat exchange, *Chem. Eng. Sci.* 55 (2000) 2161–2171.
- [2] A. Cybulski, J.A. Moulijn, Monoliths in heterogeneous catalysis, *Catal. Rev.-Sci. Eng.* 36 (1994) 179–270.
- [3] M. Uberoi, C.J. Pereira, External mass transfer coefficients for monolith catalysts, *Ind. Eng. Chem. Res.* 35 (1996) 113–116.
- [4] M. Lyubovskiy, H. Karim, P. Menacherry, S. Boorse, R. LaPierre, W.C. Pfefferle, S. Roychoudhury, Complete and partial catalytic oxidation of methane over substrates with enhanced transport properties, *Catal. Today* 83 (2003) 183–197.
- [5] A.F. Ahlström-Silversand, C.U.I. Odenbrand, Modeling catalytic combustion of carbon monoxide and hydrocarbons over catalytically active wire meshes, *Chem. Eng. J.* 73 (1999) 205–216.
- [6] A. Kołodziej, J. Łokewska, Optimization of structured catalyst carriers for VOC combustion, *Catal. Today* 105 (2005) 378–384.
- [7] K.S. Yang, Z.D. Jiang, J.S. Chung, Electrophoretically Al-coated wire mesh and its application for catalytic oxidation of 1,2-dichlorobenzene, *Surf. Coat. Technol.* 168 (2003) 103–110.
- [8] K.S. Yang, G. Mul, J.S. Choi, J.A. Mouljin, J.S. Chung, Development of TiO_2/Ti wire-mesh honeycomb for catalytic combustion of ethyl acetate in air, *Appl. Catal. A: Gen.* 313 (2007) 86–93.
- [9] K.S. Yang, J.S. Choi, J.S. Chung, Evaluation of wire-mesh honeycomb containing porous $\text{Al}/\text{Al}_2\text{O}_3$ layer for catalytic combustion of ethyl acetate in air, *Catal. Today* 97 (2004) 159–165.
- [10] K.S. Yang, J.S. Choi, S.H. Lee, J.S. Chung, Development of $\text{Al}/\text{Al}_2\text{O}_3$ -coated wire-mesh honeycombs for catalytic combustion of volatile organic compounds in air, *Ind. Eng. Chem. Res.* 43 (2004) 907–912.
- [11] H. Sun, Y.B. Zhang, X. Quan, S. Chen, Z.P. Qu, Y.L. Zhou, Wire-mesh honeycomb catalyst for selective catalytic reduction of NO_x under lean-burn conditions, *Catal. Today* 139 (2008) 130–134.
- [12] Z.D. Jiang, K.S. Chung, G.R. Kim, J.S. Chung, Mass transfer characteristics of wire-mesh honeycomb reactors, *Chem. Eng. Sci.* 58 (2003) 1103–1111.
- [13] H. Sun, X. Quan, S. Chen, H.M. Zhao, Y.Z. Zhao, Preparation of well-adhered $\gamma\text{-Al}_2\text{O}_3$ washcoat on metallic wire mesh monoliths by electrophoretic deposition, *Appl. Surf. Sci.* 253 (2007) 3303–3310.
- [14] J.W. Beekman, L.L. Hegeud, Design of monolith catalysts for power plant NO_x emission control, *Ind. Eng. Chem. Res.* 30 (1991) 969–978.
- [15] A. Beretta, C. Orsenigo, N. Ferlazzo, E. Tronconi, P. Forzatti, F. Berti, Analysis of the performance of plate-type monolithic catalysts for selective catalytic reduction DeNO_x applications, *Ind. Eng. Chem. Res.* 37 (1998) 2623–2633.
- [16] J. Svachula, N. Ferlazzo, P. Forzatti, E. Tronconi, F. Bregani, Selective reduction of NO_x by NH_3 over honeycomb deNO_x catalysts, *Ind. Eng. Chem. Res.* 32 (1993) 1053–1060.
- [17] E. García-Bordejé, L. Calvillo, M.J. Lázaro, R. Moliner, Study of configuration and coating thickness of vanadium on carbon-coated monoliths in the SCR of NO at low temperature, *Ind. Eng. Chem. Res.* 43 (2004) 4073–4079.
- [18] A. Kołodziej, J. Łokewska, Experimental and modeling study on flow resistance of wire gauzes, *Chem. Eng. Process.* 48 (2009) 816–822.
- [19] N.J. Degenstein, R. Subramanian, L.D. Schmidt, Partial oxidation of n -hexadecane at short contact times: catalyst and washcoat loading and catalyst morphology, *Appl. Catal. A: Gen.* 305 (2006) 146–159.
- [20] S.T. Kolaczowski, Measurement of effective diffusivity in catalyst-coated monoliths, *Catal. Today* 83 (2003) 85–95.
- [21] M.J. Stutz, D. Poulikakos, Optimum washcoat thickness of a monolith reactor for syngas production by partial oxidation of methane, *Chem. Eng. Sci.* 63 (2008) 1761–1770.
- [22] R.E. Hayes, B. Liu, R. Moxom, M. Votsmeier, The effect of washcoat geometry on mass transfer in monolith reactors, *Chem. Eng. Sci.* 59 (2004) 3169–3181.
- [23] R.M. Heck, R.J. Farrauto, S.T. Gulati, *Catalytic Air Pollution Control*, John Wiley & Sons Inc., New York, 2002.