

# DESIGN MODEL FOR SELECTIVE CATALYTIC REDUCTION PROCESS: SOLUTION OF CONVECTIVE DIFFUSION AND NO<sub>x</sub> REACTION

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**ABSTRACT:** A simple model for the design of selective catalytic reduction (SCR) process is presented. The two-dimensional mass transfer equation shown in Bai and Chwu (1997) is simplified and the analytic solution is obtained. The analytic solution is verified by the solution of the numerical two-dimensional model as well as published experimental data. Then it is applied for the design of the SCR reactor. Effects of operating temperature, inlet molar ratio of NH<sub>3</sub>/NO, channel pitch, and gas flow rate on the SCR reactor volume are evaluated based on a designed NO removal efficiency. The NH<sub>3</sub> slip problem of a given inlet ratio of NH<sub>3</sub>/NO also is discussed. It is shown that the injection ratio of NH<sub>3</sub>/NO must be slightly higher than the desired NO removal efficiency so that the reactor volume can be reduced and the problem of NH<sub>3</sub> slip is not a major concern.

## INTRODUCTION

After the first patent of the selective catalytic reduction (SCR) process in 1959 in the United States, it has been employed worldwide for the NO<sub>x</sub> removal from stack gases. The SCR process converts NO<sub>x</sub> to nitrogen gas and water vapor by using ammonia as a reducing agent. It is the only commercially proven NO<sub>x</sub> control technology capable of removing more than 60% of NO<sub>x</sub> emissions. Typical operating temperatures are in the range of 300–400°C, and gas hourly space velocities (GHSV) are in the range of 2,000–7,000 h<sup>-1</sup>. For the most widely employed ceramic honeycomb catalyst cells, cell sizes vary typically from 3 to 10 mm (Beeckman and Hegedus 1991). The smaller cells normally are used for “clean” gas applications and larger cells for “dirty” gas applications.

Because the SCR system is a commercialized process, the operating experiences and performance data of the SCR reactor with different catalysts can be found easily in the literature (Kittrell and Eldridge 1985; Nishimoto et al. 1987; Stiles et al. 1994). But there are only a few theoretical analyses of the SCR process. Binder-Begesteiger et al. (1990) presented an analytical treatment on the SCR performance. Because of the assumption of fast chemical reaction at the catalytic wall, their model is valid only for a purely diffusion-controlled operation. On the other hand, Buzanowski and Yang (1990) neglected the diffusion limitation in the interphase transport and derived a simple one-dimensional (1D) analytic solution for the SCR monolith reactor. Therefore their analysis applies only to the SCR reactor with very small channel pitch where the SCR process is controlled by chemical kinetics.

The other models (Beeckman and Hegedus 1991; Lefers et al. 1991; Tronconi et al. 1992) were 1D lumped parameter analyses. They have modified the theoretical analysis of Buzanowski and Yang (1990). However, although the adequacy of an 1D lumped parameter model has been analyzed (Tronconi and Forzatti 1992), the parameters used in 1D models are unknown and needed to be justified. The unknown parameters include mass transfer coefficient ( $k_m$ ), the effective diffusion coefficient, the catalytic reaction rate, and the NH<sub>3</sub> adsorption constant. Take the mass transfer coefficient as an example;

Lefers et al. (1991) did not address it on the determination of  $k_m$ . Beeckman and Hegedus (1991) used a Hawthorn equation to determine  $k_m$ , but the Hawthorn equation is valid only in the pure diffusion region. Tronconi et al. (1992) and Tronconi and Forzatti (1992) employed an analogue between heat and mass transfer and obtained the value of  $k_m$  based on similarity between the Nusselt number and the Sherwood number. They found that the value of the Damkohler number significantly alters the Sherwood number; thereby it may lead to a large deviation of  $k_m$ .

A two-dimensional (2D) numerical model was employed by the authors (Bai and Chwu 1997) to evaluate the performance of various SCR catalysts in terms of NO<sub>x</sub> removal. The model accounted for simultaneous effects of external diffusion and chemical kinetic limitations in the SCR process. It is accurate but extensive computational effort is required. In the practical application of SCR design, the design model should be as simple as possible provided the accuracy of the design is not biased. However, it seems that available SCR design models in the literature were either too complicated to be applied to the field or too simple to retain their accuracy.

In this paper the 2D equations are further simplified using appropriate assumptions, and an analytical solution is obtained that greatly reduces the computation time. The accuracy of the analytical solution is tested by comparing the results with those of the 2D numerical model as well as experimental observations in the literature. Then it is applied for the design of a SCR reactor. Therefore, once a new catalyst is developed in a laboratory study, its SCR performance can be predicted easily using the simple analytic solution for the 2D model.

## THEORY

In the SCR process the NO<sub>x</sub> species are reduced by injection of NH<sub>3</sub>, ultimately, to N<sub>2</sub> gas over a heterogeneous catalyst in the presence of O<sub>2</sub>. The results of SCR performance tests in actual applications indicated that the following reaction is dominant (Cho 1994; Bai and Chwu 1997):



The presence of O<sub>2</sub> is essential for the SCR reaction to occur. For O<sub>2</sub> concentration of lower than 0.5%, the NO conversion increases with an increase in the O<sub>2</sub> concentration and it reaches a stable NO conversion rate at an O<sub>2</sub> concentration greater than 1% (Bosch and Janssen 1987). The SCR reaction is a heterogeneous catalytic reaction. The rate of the reaction is controlled by external and internal diffusion as well as chemical kinetic limitations. Bai and Chwu (1997) discussed physical and chemical mechanisms of the SCR process. The adsorption and chemical behaviors over catalytic surfaces also were discussed.

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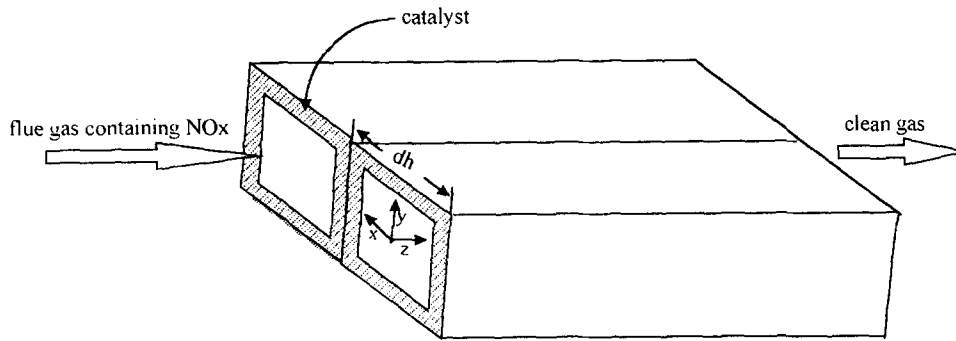


FIG. 1. Schematic Diagram of SCR Design Model

## 2D Model Equation

A schematic diagram of the SCR reactor is shown in Fig. 1. The geometry of the catalytic support is made of square ducts. With the use of appropriate model assumptions (Bai and Chwu 1997), the steady-state 2D mass transfer model can be made as

$$U \frac{\partial C_i}{\partial Z} = D_i \left( \frac{\partial^2 C_i}{\partial X^2} + \frac{\partial^2 C_i}{\partial Y^2} \right) \quad (2)$$

where  $C_i$  = gas concentration ( $i = \text{NO}$  or  $\text{NH}_3$ );  $U$  = flow velocity in the ducts;  $D_i$  = diffusion coefficient of reagents;  $Z$  = main flow direction; and  $X$  and  $Y$  = directions perpendicular to the main gas stream. The left-hand-side term is the convection term whereas the two terms on the right side denote the external diffusional deposition of NO and  $\text{NH}_3$  molecules onto the catalytic wall. Eq. (2) can be normalized using the non-dimensional parameters  $C_i^* = C_i/C_i^0$ ,  $X^* = 2X/d_h$ ,  $Y^* = 2Y/d_h$ ,  $U^* = U/U_m$ , and  $Z^* = 4ZD_{\text{NO}}/(U_m d_h^2)$ , where  $C_i^0$  is the reagent inlet concentration,  $d_h$  is the channel pitch, and  $U_m$  is the average flow velocity. As a result the nondimensional equations for the evolution of NO and  $\text{NH}_3$  concentrations in the SCR reactor are

$$U^* \frac{\partial C_{\text{NO}}^*}{\partial Z^*} = \frac{\partial^2 C_{\text{NO}}^*}{\partial X^{*2}} + \frac{\partial^2 C_{\text{NO}}^*}{\partial Y^{*2}} \quad (3)$$

$$U^* \frac{\partial C_{\text{NH}_3}^*}{\partial Z^*} = \frac{D_{\text{NH}_3}}{D_{\text{NO}}} \left( \frac{\partial^2 C_{\text{NH}_3}^*}{\partial X^{*2}} + \frac{\partial^2 C_{\text{NH}_3}^*}{\partial Y^{*2}} \right) \quad (4)$$

The velocity profile usually is assumed to be fully developed laminar flow in the SCR system (Lefers et al. 1991; Tronconi and Forzatti 1992). The fully developed velocity profile in a square duct is obtained from Shah and London (1978). The initial condition is set at the inlet of the SCR system such that

$$\text{at } Z^* = 0, \quad C_i^* = 1, \quad (5)$$

where the boundary conditions are that the reagent concentrations are symmetric at the center line of a square duct; and the deposition flux is controlled by the effective rate of catalytic reaction between NO and  $\text{NH}_3$  on the catalytic surface

$$\text{at } X^* = 0, \quad \frac{\partial C_i^*}{\partial X^*} = 0 \quad (6)$$

$$\text{at } Y^* = 0, \quad \frac{\partial C_i^*}{\partial Y^*} = 0 \quad (7)$$

$$\text{at } X^* = 1, \quad -\frac{\partial C_i^*}{\partial X^*} = \frac{\text{Da}_i}{2} R^* \quad (8)$$

$$\text{at } Y^* = 1, \quad -\frac{\partial C_i^*}{\partial Y^*} = \frac{\text{Da}_i}{2} R^*, \quad (9)$$

where  $\text{Da}_i (=K_C d_h/2D_i)$  = Damkohler number for species  $i$ ;  $K_C$  = effective rate constant; and  $R_{\text{NO}}^* (=R_{\text{NO}}/K_C C_{\text{NO}}^0)$  = dimensionless reaction rate for NO.

## Analytic Solution of Model Equation

The model equations in (3) and (4) can be further simplified with the following assumptions:

1. The flow pattern in the SCR reactor can be represented by plug-flow field instead of fully developed laminar-flow field.
2. The diffusivities of  $\text{NH}_3$  gas and NO gas are the same in the external as well as the internal (pore) regions. Therefore (3) and (4) are identical and only one equation is needed for the SCR design that

$$U^* \frac{\partial C_i^*}{\partial Z^*} = \frac{\partial^2 C_i^*}{\partial X^{*2}} + \frac{\partial^2 C_i^*}{\partial Y^{*2}} \quad (10)$$

Therefore using the method of separation of variables, (10) is solved for the evolution of NO concentration in the SCR reactor as

$$C_{\text{NO}}^*(X^*, Y^*, Z^*) = \frac{\beta^2}{\sin^2 \beta} \exp(-2\beta Z^*) \int_0^1 \int_0^1 \cos(\beta X^*) \cdot \cos(\beta Y^*) dX^* dY^* \quad (11)$$

where  $\beta$  is defined by that

$$\beta \tan(\beta) = \frac{\text{Da}_{\text{NO}}}{2} \theta_{\text{NH}_3} = \frac{\text{Da}_{\text{NO}}}{2} \frac{K_{\text{NH}_3}^* C_{\text{NH}_3}^*}{1 + K_{\text{NH}_3}^* C_{\text{NH}_3}^*} \quad (12)$$

where  $\theta_{\text{NH}_3}$  = fractional surface coverage of ammonia adsorbed on an active sites of catalyst; and  $K_{\text{NH}_3}^*$  = dimensionless adsorption constant ( $K_{\text{NH}_3}^* = K_{\text{NH}_3} C_{\text{NO}}^0$ ). The NO removal rate  $\eta_{\text{NO}}$  is

$$\eta_{\text{NO}} = 1 - C_{\text{NO}}^*(X^*, Y^*, Z^*) \quad (13)$$

The same equations can be made for the evolution of ammonia concentration in the SCR reactor. Detailed solution procedures of (10) are shown in Appendix I. The determination of  $\text{NH}_3$  adsorption constant and effective rate constant is referred to Bai and Chwu (1997). Eq. (11) can be further written as

$$C_{\text{NO}}^*(X^*, Y^*, Z^*) = \frac{\beta}{\sin^2 \beta} \exp(-2\beta^2 Z^*) \sum_{i=0}^N \sum_{j=0}^N \cos\left(\beta \frac{i}{N}\right) \cdot \cos\left(\beta \frac{j}{N}\right) \quad (14)$$

The computation is easier using (14) instead of (11). The error induced by using (14) is less than 0.5% for  $N > 50$ , and approximately 2% for  $N = 20$ . It would be larger than 20% for  $N < 10$ .

## RESULTS AND ANALYSIS

### Analysis of Model Accuracy

The assumptions for deriving the analytic solution are evaluated. When different diffusivities are used for  $\text{NH}_3$  and  $\text{NO}$  gases, two model equations are needed for the behaviors of the two gas species. Therefore an analytic solution could not be derived, and the numerical method of forward-time centered-space was used for solving the coupled equations in (3) and (4). When assuming identical diffusivities, the analytic solution equation in (14) can be used for predicting the  $\text{NO}$  conversion rates. Results of the two cases are compared in Fig. 2 as a function of dimensionless axial distance ( $Z^*$ ) under various Damkohler numbers. It is seen that similar results are obtained for the two cases at Damkohler numbers ranging from 0.01 to 10. Therefore, assuming identical diffusion coefficients for the two gases does not yield significant differences between the model outputs.

The assumption of fully developed flow pattern is more appropriate than the plug-flow field assumption for the velocity profiles in the SCR reactor. However, with that the analytical solution is difficult. Therefore, the validity of plug-flow field assumption was tested and shown in Fig. 3. The results from the numerical model were obtained using the fully developed flow pattern, and those from the analytic model were obtained using the plug-flow pattern. It is observed that the value of the Damkohler number significantly affects the model results at smaller  $Z^*$ . For the typical range of the Damkohler number on the order of unity in an SCR reactor, the deviation may be as large as 10% of  $\text{NO}$  conversion.

The discrepancy induced by the assumption of plug-flow pattern can be minimized. The differences in the results from the two flow patterns are compared using the 2D numerical model, and a correction formula is obtained. A new Damkohler number is defined with the correction factor  $W$

$$Da_{\text{new}} = W \times Da_{\text{original}} \quad (15)$$

where values of  $W$  can be found using Fig. 4. The regression equation of  $W$  as a function of Damkohler number also is obtained as

$$W = 0.8631 - 0.2176 \times \ln(Da_{\text{original}}) \quad (16)$$

Results from the regression equation in (16) also are shown in Fig. 4. It is seen that the value of  $W$  decreases as the Damkohler number increases. This denotes that the discrepancy between plug-flow pattern and fully developed laminar-

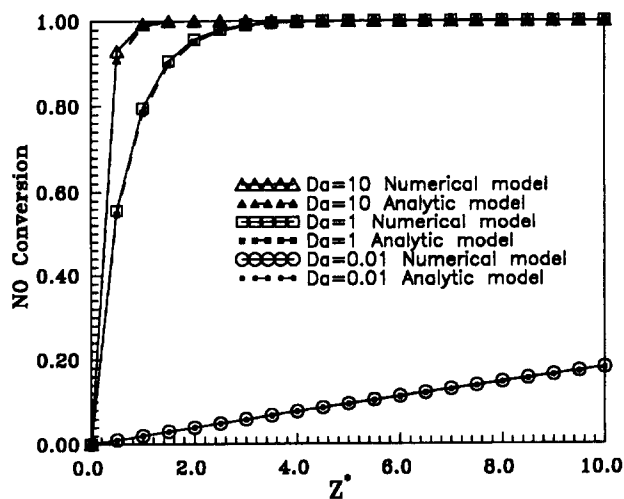


FIG. 2. Comparison of Results Using Different Diffusion Coefficients for  $\text{NH}_3$  and  $\text{NO}$  (Numerical Model) and Using Identical Diffusion Coefficient for  $\text{NH}_3$  and  $\text{NO}$  (Analytic Model)

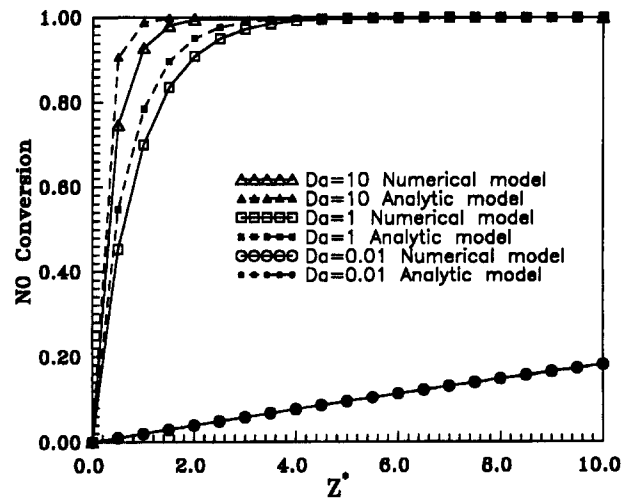


FIG. 3. Comparison of Results Using Fully Developed Laminar-Flow Pattern (Numerical Model) and Using Plug-Flow Pattern (Analytic Model)

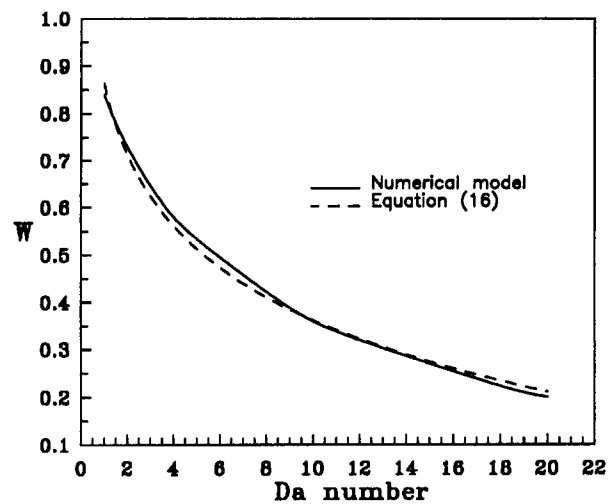


FIG. 4. Correction factor  $W$  as a Function of Original Damkohler Number (Solid Line); Results from Regression Equation of Correction Factor Also is Shown (Dashed Line)

flow pattern increases with an increase in the Damkohler number. The new Damkohler number can be used in the analytic solution to minimize the error induced by the assumption of plug-flow pattern in an SCR reactor.

### Model Prediction

The results of the analytic solution developed in this study are compared with those of Buzanowski and Yang (1990) in terms of  $\text{NO}$  conversion. The model of Buzanowski and Yang (1990) was developed assuming that diffusion limitation is negligible. The  $\text{NO}$  removal efficiency  $\eta_{\text{NO}}$  in their model is

$$\eta_{\text{NO}} = 1 - \exp \left[ - \left( \frac{\sigma L}{U_m A} \right) \frac{1}{\frac{1}{K_m} + \frac{1}{(D_e K_i)^{1/2} \frac{1 + e^{-2\phi}}{1 - e^{-2\phi}}}} \right] \quad (17)$$

where  $\sigma$  = perimeter length of a cell in the honeycomb;  $L$  = total length of the reactor;  $A$  = cross-sectional area of a cell;  $D_e$  = effective diffusion coefficient;  $K_i$  = intrinsic rate of reaction; and  $\phi$  = Thiele modulus defined as  $\phi = L(K_i/D_e)^{0.5}$ .

Fig. 5 depicts the comparison of results of the two models. The experimental data also were obtained from Buzanowski and Yang (1990). One can see that both models well predict

the experimental observations. However, the experimental data of Buzanowski and Yang (1990) were conducted at very narrow channel pitch ( $d_h = 0.14$  cm). Therefore the assumption of neglecting the diffusion limitation in their model analysis is valid in this case. But the model of Buzanowski and Yang (1990) may not be applicable for larger channel pitch, which is more practical in the SCR design.

Fig. 6 shows the comparison results of the two analytic solutions with the SCR data of Beekman and Hegedus (1991). The experiments were conducted with a typical channel pitch of 0.6 cm where diffusion limitation cannot be neglected. It is observed that the analytic solution of Buzanowski and Yang (1990) underpredicts the experimental data by 10–15% of NO conversion rates. And the analytic solution developed in this study overpredicts by 5–8% before correction of the Damkohler number. This is because although the analytic solution developed in this study has the ability of accounting for the effects of both diffusion and chemical reaction limitations, the assumption of plug-flow field results in deviations at large channel pitch (small  $Z^*$ ). After correction of the Damkohler number with the factor  $W$ , the results of the analytic solution developed in this study are in good agreement with the experimental data.

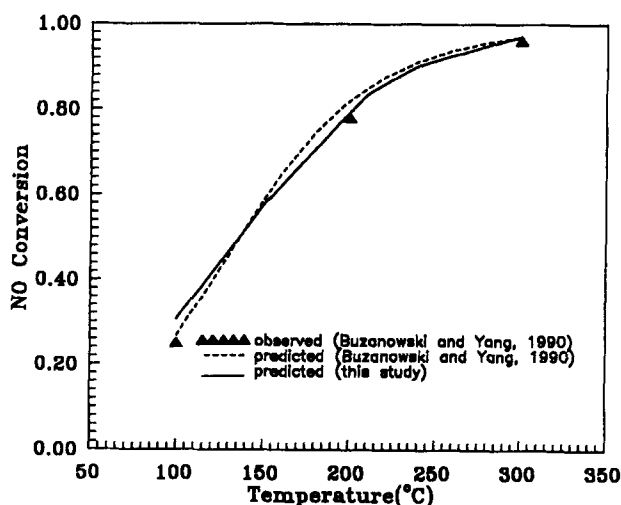


FIG. 5. Comparison of Model Prediction Result with Theoretical Result as Well as Experimental Observations of Buzanowski and Yang (1990)

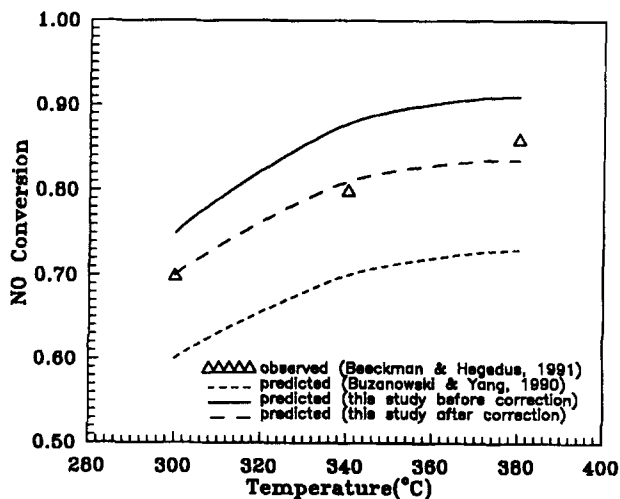
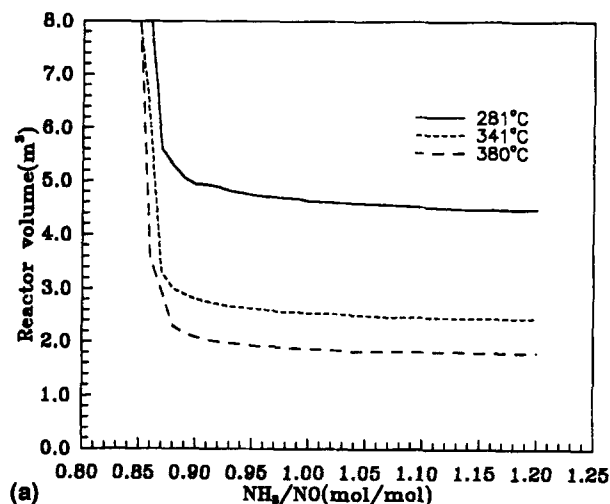


FIG. 6. Comparison of Prediction Results from this Study and Buzanowski and Yang (1990) with Experimental Observations of Beekman and Hegedus (1991)

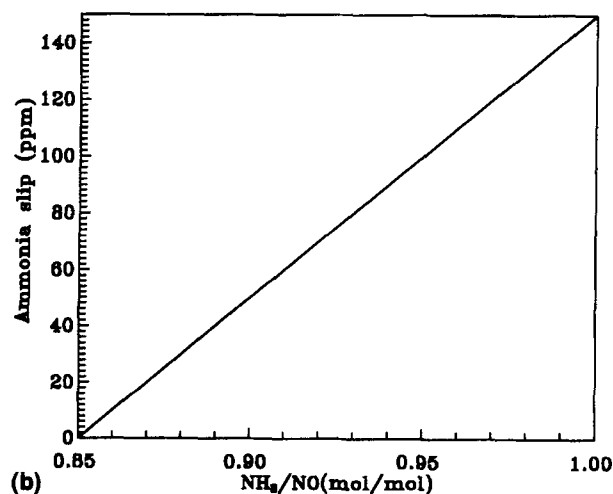
## Practical Application

The analytic solution of the 2D model is easier to use in the design of an SCR system. Fig. 7(a) shows the effect of inlet molar ratio of  $\text{NH}_3/\text{NO}$  on the reactor volume. The catalyst was obtained from the Catalyst Research Center, China Technical Consultants, Inc., Taiwan. Characteristics of the catalyst are referred to in Bai and Chwu (1997). The designed NO removal efficiency was 85%; the NO inlet concentration was 1,000 ppm and the flue gas flow rate was  $5 \text{ m}^3/\text{s}$  at standard temperature of  $0^\circ\text{C}$   $1.01 \times 10^5 \text{ Pa}$ . The channel pitch of the SCR reactor was 0.6 cm. It is seen that if the inlet ratio of  $\text{NH}_3/\text{NO}$  is at 0.85, then the reactor volume would have to be infinitely large to achieve 85% of NO removal. Therefore the injection ratio of  $\text{NH}_3/\text{NO}$  must be higher than the desired NO removal efficiency. The reactor volume reduces quickly as inlet ratio of  $\text{NH}_3/\text{NO}$  increases from 0.85 to 0.90. It gradually reaches a stable size as inlet ratio of  $\text{NH}_3/\text{NO}$  is larger than 0.9. Besides, an increase in the operating temperature significantly reduces the reactor volume. This is because the catalytic reaction rate increases with increasing temperature.

The ammonia slip problem is another major concern in the design of an SCR reactor. Fig. 7(b) depicts the amount of ammonia slip as a function of inlet ratio of  $\text{NH}_3/\text{NO}$ . It is seen that they are linearly proportional. Because inlet ratio of  $\text{NH}_3/\text{NO}$  is greater than 0.90, the ammonia slip is higher than 50 ppm. This would violate the regulations for ammonia emission either in the United States or in Taiwan. Therefore in this



(a)



(b)

FIG. 7. Design of SCR as Function of Inlet Molar Ratios of  $\text{NH}_3/\text{NO}$ : (a) Reactor Volume; (b) Ammonia Slip

case, it is out of the question to design the injection ratio of  $\text{NH}_3/\text{NO}$  to be greater than 0.90. But it also is seen that as the inlet ratio of  $\text{NH}_3/\text{NO}$  is less than 0.86, the required volume of the reactor is very large. Therefore the optimal injection ratio of  $\text{NH}_3/\text{NO}$  falls between 0.86 and 0.88, and the corresponding amount of ammonia slip is 10–30 ppm. It must be noted that the ammonia slip shown in Figure 7(b) is a conservative estimate of the amount of ammonia slip from an SCR reactor. Under field operation the injected ammonia also would be consumed by other gas species such as  $\text{SO}_2$  and  $\text{CO}_2$  in the flue gas. This is not accounted for in this study.

Fig. 8 shows the effect of channel pitch on the reactor volume. The designed NO removal efficiency was 85%; inlet NO concentration was 1,000 ppm; inlet ratio of  $\text{NH}_3/\text{NO}$  was 0.88; gas flow rate was  $5 \text{ Nm}^3/\text{s}$ . It is observed that the reactor volume increases with an increase in the channel pitch. This is because larger channel pitch indicates that the SCR process is controlled by diffusion limitation. Therefore the SCR process is more economically applicable to the gas-firing system where narrow channel pitch can be used.

Fig. 9 depicts the effect of flue gas flow rate on the reactor volume. The other parameter values were the same as those for Fig. 8. It is seen that the reactor volume is linearly proportional to the flue gas flow rate. One of the design parameters used in the SCR reactor, the GHSV, is defined as  $Q/V$ , where  $Q$  is the gas flow rate and  $V$  is the reactor volume.

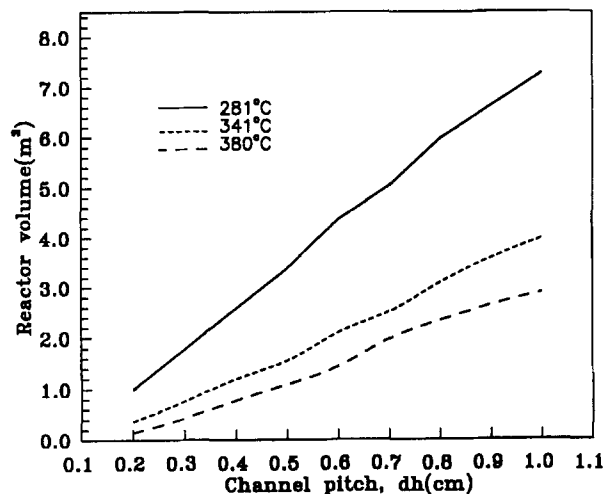


FIG. 8. Design of Reactor Volume as Function of Channel Pitch

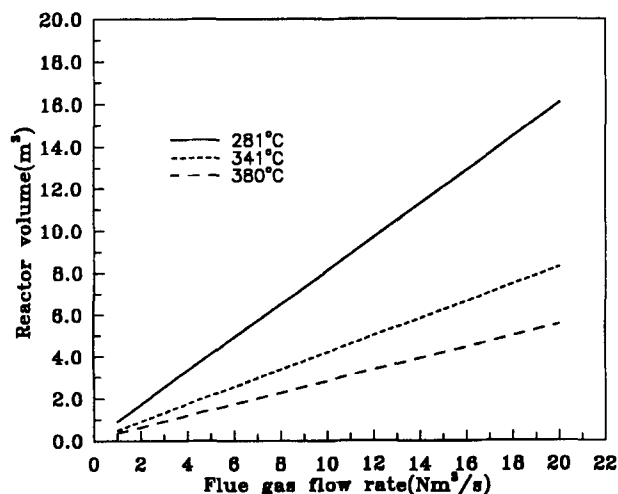


FIG. 9. Design of Reactor Volume as Function of Flue Gas Flow Rate

Given the cases shown in Fig. 9, the values of GHSV are 3,910, 7,200, and 9,730  $\text{h}^{-1}$ , respectively, for SCR temperatures of 281, 341, and 380°C for flue gas flow rate of  $1 \text{ Nm}^3/\text{s}$ . For flue gas flow rate of  $20 \text{ Nm}^3/\text{s}$  the GHSV values are 4,480, 8,690, and 12,970  $\text{h}^{-1}$ , respectively, for SCR temperatures of 281, 341, and 380°C. Typical range of GHSV in the SCR are 2,000–7,000  $\text{h}^{-1}$ . Therefore, for the catalyst used in the design study, the required SCR temperature can be in the lower range of typical design value (300–400°C). An SCR reactor with operating temperature of less than 340°C would satisfy the typical range of GHSV. On the other hand, if the SCR reactor can be operated at a higher temperature (such as 380°C), then the required reactor size can be reduced. A comparison of the energy cost for reheating the flue gas to a desired temperature and the installation cost of the SCR reactor will decide which method is economical.

## CONCLUSIONS

A model is presented to assist in the design of an SCR reactor. The model is an analytic derivation of the 2D mass transfer equation. The advantages of the model are that it is not only simple but it also takes into account of the simultaneous effects of diffusion and chemical kinetic limitations in the SCR process. After tests of the model accuracy, it was found that the assumption plug-flow field in the analytic solution may lead to some deviations from the numerical solution. But with correction of the Damkohler number, the analytic solution compares well with experimental observations. The analytic model presented in this study is helpful in the proper design of the SCR reactor. The design parameters such as operating temperature, injection ratio of  $\text{NH}_3/\text{NO}$ , channel pitch, and reactor volume can be determined easily.

## APPENDIX I. DERIVATION OF ANALYTIC SOLUTION

With the use of the plug-flow assumption and identical diffusivities assumption for the  $\text{NH}_3$  and NO gases, (10) becomes

$$\frac{\partial C^*}{\partial Z^*} = \frac{\partial^2 C^*}{\partial X^{*2}} + \frac{\partial^2 C^*}{\partial Y^{*2}} \quad (18)$$

where  $C^*$  = concentration profiles of  $\text{NH}_3$  or NO in the SCR system. By applying the method of separation of variables, we obtain the solution of  $C^*$  of the form

$$C^* = F(X^*)G(Y^*)H(Z^*) \quad (19)$$

Inserting (19) into (18) we have

$$FG \frac{\partial H}{\partial Z^*} = GH \frac{\partial^2 F}{\partial X^{*2}} + FH \frac{\partial^2 G}{\partial Y^{*2}} \quad (20)$$

Dividing both sides of (20) by  $FGH$ , we find

$$\frac{1}{H} \frac{\partial H}{\partial Z^*} = \frac{1}{F} \frac{\partial^2 F}{\partial X^{*2}} + \frac{1}{G} \frac{\partial^2 G}{\partial Y^{*2}} \quad (21)$$

Each side of (21) must be equal to a constant, say  $(-\mu)$ , thus

$$\frac{1}{H} \frac{\partial H}{\partial Z^*} = -\mu \quad (22)$$

$$\frac{1}{F} \frac{\partial^2 F}{\partial X^{*2}} + \frac{1}{G} \frac{\partial^2 G}{\partial Y^{*2}} = -\mu \quad (23)$$

The solution of equation (22) can be obtained readily as

$$H = H_0 \exp(-\mu Z^*) \quad (24)$$

where  $H_0 = \text{constant}$ , and (23) is written separately as

$$\frac{1}{F} \frac{\partial^2 F}{\partial X^{*2}} = \mu_1 \quad (25)$$

$$\frac{1}{G} \frac{\partial^2 G}{\partial Y^{*2}} = -\mu_2 \quad (26)$$

where

$$\mu_1 + \mu_2 = \mu \quad (27)$$

The general solutions for (25) and (26) are

$$F = F_1 \cos(\mu_1 X^*) + F_2 \sin(\mu_1 X^*) \quad (28)$$

$$G = G_1 \cos(\mu_1 Y^*) + G_2 \sin(\mu_1 Y^*) \quad (29)$$

where  $F_1, F_2, G_1,$  and  $G_2 = \text{constants}$ . With the use of the boundary condition equation in (6) it is found that  $F_2 = 0$ , therefore (28) takes the form that

$$F = F_1 \cos(\mu_1 X^*) \quad (30)$$

and substitute the dimensionless reaction rate  $R^* = \theta^* C^*$  into (8) to obtain that

$$X^* = 1, \quad -\frac{\partial C^*}{\partial X^*} = \frac{\text{Da}}{2} \theta C^* \quad (31)$$

Substituting (30) into (31) we have

$$\frac{\partial C^*}{\partial X^*} + \frac{\text{Da}}{2} \theta C^* = -\mu \sin(\mu_1) + \frac{\text{Da}}{2} \theta \cos(\mu_1) = 0 \quad (32)$$

Therefore the constant  $\mu_1$  is defined as

$$\mu_1 \tan(\mu_1) = \frac{\text{Da}}{2} \theta \quad (33)$$

The solution procedures for  $Y$ -direction are similar to the foregoing equations and therefore

$$\mu_2 \tan(\mu_2) = \frac{\text{Da}}{2} \theta \quad (34)$$

and from (33) and (34) we know that

$$\mu_1 = \mu_2 \quad (35)$$

Therefore the general solution for  $C^*$  is

$$C^*(X^*, Y^*, Z^*) = A_0 \exp(-\mu Z^*) \int_0^1 \int_0^1 \cos(\mu_1 X^*) \cdot \cos(\mu_1 Y^*) dX^* dY^* \quad (36)$$

where  $A_0 = \text{a constant}$ , and we know from (27) and (35) that

$$\mu = 2\mu_1 \quad (37)$$

With the use of the initial condition that at  $Z^* = 0, C^* = 1$ , we define the relationship between constant  $A_0$  and  $\mu_1$  as

$$A_0 = \frac{\mu_1^2}{\sin^2(\mu_1)} \quad (38)$$

Hence we derive the analytic solution of  $C^*$  as

$$C^*(X^*, Y^*, Z^*) = \frac{\mu_1^2}{\sin^2(\mu_1)} \exp(-2\mu_1 Z^*) \int_0^1 \int_0^1 \cos(\mu_1 X^*) \cdot \cos(\mu_1 Y^*) dX^* dY^* \quad (39)$$

where  $\mu_1 = \text{a constant}$ , which is defined by equation (33).

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## APPENDIX II. REFERENCES

- Bai, H., and Chwu, J.-W. (1997). "A theoretical analysis of selective catalytic reduction catalysts." *J. Envir. Engrg.*, ASCE, 123(5), 431–436.
- Beeckman, J. W., and Hegedus, L. L. (1991). "Design of monolith catalysts for power plant NOx emission control." *Ind. Engrg. Chem. Res.*, 30(5), 969–978.
- Binder-Begesteiger, I., Herzog, G. W., Megla, E., and Tomann-Rosos, M. (1990). "Zur kinetic der denox-reaktion an TiO<sub>2</sub>/WO<sub>3</sub>-Waben-Katalysatoren." *Chem. Ing. Tech.*, 62(1), 60.
- Bosch, H., and Janssen, F. (1987). "Catalytic reduction of nitrogen oxides—a review of the fundamentals and technology." *Catal. Today*, 2(4), 369–532.
- Buzanowski, M. A., and Yang, R. T. (1990). "Simple design of monolith reactor for selective catalytic reduction of NO for power plant emission control." *Ind. Engrg. Chem. Res.*, 29(10), 2074–2078.
- Cho, S. M. (1994). "Properly apply selective catalytic reduction for NOx removal." *Chem. Engrg. Progress*, (Jan.), 39–45.
- Kittrell, J. A., and Eldridge, J. W. (1985). "Novel catalyst preparation method for NOx control." *Envir. Progress*, 4(2), 78–84.
- Lefers, J. B., Lodder, P., and Enoch, G. D. (1991). "Modelling of selective catalytic denox reactors—strategy for replacing deactivated catalyst elements." *Chem. Engrg. Technol.*, 14, 192–200.
- Nishimoto, Y., Seto, T., Iida, K., Mitsuoka, S., and Iwata, K. (1987). "Update technology of selective catalytic flue gas NOx removal system." *Tech. Rev.*, 24(1), 54–61.
- Shah, R. K., and London, A. L. (1978). *Laminar flow forced convection in ducts*. Academic Press, New York.
- Stiles, A. B., Klein, M. T., Gauthier, P., Schwarz, S., and Wang, J. (1994). "Selective catalytic reduction of NOx in the presence of oxygen." *Ind. Engrg. Chem. Res.*, 33(10), 2259–2264.
- Tronconi, E., and Forzatti, P. (1992). "Adequacy of lumped parameter models for SCR reactors with monolith structure." *AIChE J.*, 38(2), 201–210.
- Tronconi, E., Forzatti, P., Gomez Martin, J. P., and Malloggi, S. (1992). "Selective catalytic removal of NOx: a mathematical model for design of catalyst and reactor." *Chemical Engrg. Sci.*, 47(9-11), 2401–2406.

## APPENDIX III. NOTATION

The following symbols are used in this paper:

- $A$  = cross-sectional area of a cell (cm<sup>2</sup>);  
 $C_i$  = gas concentration,  $i = \text{NH}_3$  or NO (mol/cm<sup>3</sup>);  
 $C^*$  = dimensionless gas concentration,  $C_{\text{NO}}/C_{\text{NO}}^0$ ;  
 $\text{Da}$  = Damkohler number,  $K_c d_h / 2D_e$ ;  
 $D_e$  = effective diffusion coefficient (cm<sup>2</sup>/s);  
 $D_i$  = diffusion coefficient,  $i = \text{NH}_3$  or NO (cm<sup>2</sup>/s);  
 $d_h$  = channel pitch (cm) (defined in Fig. 1);  
 $K_C$  = effective chemical reaction rate constant (cm/s);  
 $K_i$  = intrinsic rate of reaction (1/s);  
 $K_m$  = mass transfer coefficient (cm/s);  
 $K_{\text{NH}_3}$  = adsorption constant of NH<sub>3</sub> (cm<sup>3</sup>/mol);  
 $K_{\text{NH}_3}^*$  = dimensionless adsorption constant,  $K_{\text{NH}_3} C_{\text{NO}}^0$ ;  
 $L$  = total length of the SCR reactor (cm);  
 $Q$  = gas flow rate (cm<sup>3</sup>/s);  
 $R_{\text{NO}}$  = reaction rate of NO (mol/cm<sup>2</sup> s);  
 $R_{\text{NO}}^*$  = dimensionless reaction rate of NO,  $R_{\text{NO}}/(C_{\text{NO}}^0 K_C)$ ;  
 $U$  = flow velocity (cm/s);  
 $U_m$  = average flow velocity (cm/s);  
 $U^*$  = dimensionless flow velocity  $U/U_m$ ;  
 $V$  = volume of the SCR system (cm<sup>3</sup>);  
 $W$  = correction factor of the Da number for the analytic solution;  
 $X$  = direction perpendicular to the flow (cm);  
 $X^*$  = dimensionless parameter of  $X$ ,  $2X/d_h$ ;

$Y$  = direction perpendicular to the flow (cm);  
 $Y^*$  = dimensionless parameter of  $Y$ ,  $2Y/d_h$ ;  
 $Z$  = direction of main flow (cm);  
 $Z^*$  = dimensionless parameter of  $Z$ ,  $4ZD_{NO}/U_m D_h^2$ ;  
 $\eta_{NO}$  = removal efficiency of NO;  
 $\theta_{NH_3}$  = fractional surface coverage of ammonia absorbed on the active sites of catalyst;

$\sigma$  = perimeter length of a cell in honeycomb (cm); and  
 $\phi$  = Thiele modulus,  $L(K_i/D_c)^{0.5}$ .

### Subscripts and Superscripts

0 = initial condition; and  
\* = dimensionless parameter.