# ABSORPTION OF NO$_x$ IN PACKED COLUMN (I)
## —MATHEMATICAL MODELLING—

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**Abstract**—A mathematical model for an absorption of nitrogen oxides into water in packed column was developed based on the mass-transfer coefficient in packed column and the chemical reaction accompanying NO$_x$ absorption produces HNO$_3$ and HNO$_2$ in the liquid phase. The subsequent dissociation of HNO$_2$ in the liquid-phase results in the formation of HNO$_3$ and NO gas, and then this NO gas follows to be oxidized by O$_2$ in air. The important factors influenced on the absorption of NO$_x$ are the oxidation state of NO gas and the partial pressure of nitrogen oxides in the gas. The efficiency of NO$_x$ absorption increases with the increase of the partial pressure of NO$_x$. The most critical value for using the model is the constant of $(\sqrt{DK_3}/H)_{N_2O_4}$. The selection of $11 \times 10^{-4}$ kg·mol/atm·m$^2$·sec for $(\sqrt{DK_3}/H)_{N_2O_4}$ is resonable for this model.

## INTRODUCTION

The scrubbing of NO$_x$ (nitrogen oxides) from gaseous mixtures of air and NO$_x$ is very important process in production of nitric acid as well as in air pollution abatement of industial process. Interest in this absorption process is increasing due to current problem in air pollution and resource recovery. The absorption of gaseous NO$_x$ into water and dilute HNO$_3$, and the gas-phase oxidation reaction of NO provide the basis for one method of NO$_x$ removals. Such an absorption process is accompanied by the reactions of NO$_x$ with water, resulting in the production of HNO$_3$. This scheme is attractive for the proposed application because the produced HNO$_3$ could be reused in the plant.
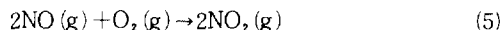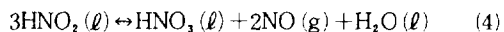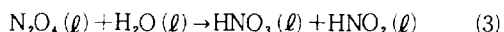
The most pronounced effect on the NO$_x$ conversion was dependent on the variation in the oxidation state of the gas. The second largest effect was produced by the variation in the partial pressure of NO$_x$ in the feed gas. In general, the NO gas has a much lower solubility and reactivity than the NO$_2^*$, defined as the sum of the NO$_2$ concentration and the product of two times its equilibrium polymer concentration, N$_2$O$_4$ [1,2]. The higher NO$_x$ partial pressure produces a larger proportional amount of higher molecular weight species, which are more absorbable and reactive.

The NO$_2$–N$_2$O$_4$ equilibrium and the absorption of gaseous N$_2$O$_4$ into water are simulated mathematically and combined into an equation for N$_2$O$_4$ absorption based on the mass-transfer in the packed column. The disappearance of liquid HNO$_2$ is simulated, resulting in the production of gaseous NO. NO is oxidized with O$_2$ in the gas phase. Then a mathematical expression was developed for NO$_x$ absorption in the packed column.

## ABSORPTION MECHANISM ASSOCIATED WITH CHEMICAL REACTION

The nitrogen oxides of interest in the gas-phase are NO, NO$_2$ and N$_2$O$_4$. The basic chemical reactions at the steady-state absorption of NO$_x$ compounds into water or dilute HNO$_3$ may be written as follows [3-6]:

$$2NO_2 (g) \leftrightarrow N_2O_4 (g) \tag{1}$$

$$N_2O_4 (g) \leftrightarrow N_2O_4 (\ell) \tag{2}$$

$$N_2O_4 (\ell) + H_2O (\ell) \rightarrow HNO_3 (\ell) + HNO_2 (\ell) \tag{3}$$

$$3HNO_2 (\ell) \leftrightarrow HNO_3 (\ell) + 2NO (g) + H_2O (\ell) \tag{4}$$

$$2NO (g) + O_2 (g) \rightarrow 2NO_2 (g) \tag{5}$$

where (g) and ($\ell$) indicate gas and liquid species, respectively. A review of the literature indicates that the following assumptions about the overall reactions may be made:
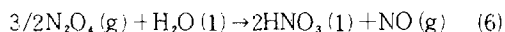
1) The NO$_2$ and N$_2$O$_4$ are in continuous gas-phase equilibrium[6-8],

2) Reaction of $N_2O_4$ with water proceeds by means of a fast pseudo first-order liquid-phase reaction[2,5,8-15]. When the partial pressure of $NO_2^*$ is higher than 0.01 atm, this is the predominant absorption reaction[2].

3) Liquid $HNO_2$ decomposes as described by reaction (4) [16], and

4) Oxidation of gaseous NO occures as an overall third-order gas-phase reaction [17-21].

Reaction (3) and (4) are sometimes combined as [9,10,13]

$$3/2N_2O_4 \, (g) + H_2O \, (1) \rightarrow 2HNO_3 \, (1) + NO \, (g) \quad (6)$$

Reaction (1) through (5) adequately describe the absorption process if the gaseous NO concentration is below 1,000 ppm or the gaseous $NO_x$ concentration is moderately high.

The gas phase is assumed to be saturated with $H_2O$,consistent with the temperature and liquid $HNO_3$ concentration. The partial pressure of $HNO_3$ is usually taken to be zero by comparing with the $NO_x$ partial pressures. The $HNO_2$ partial pressure is usually smaller than the $H_2O$ partial pressure and is always assumed to be equal or below the saturated partial pressure.

At $NO_2$-$N_2O_4$ equilibrium, $P_{N_2O_4}$ and $P_{NO_2}$ may be calculated using the quantity $\alpha$ that represents the degree of dissociation of $N_2O_4$. The partial pressure of $N_2O_4$ and $NO_2$ can be expressed in terms of $NO_2^*$ and $\alpha$[22].

The hydrolysis of $N_2O_4$ during the absorption of $N_2O_4$ into water or dilute nitric acid is essentially irreversible due to low concentrations of $HNO_2$ and $HNO_3$ in the liquid phase. The decomposition of aqueous nitrous acid in the liquid phase is generally expressed as a reaction (4). The NO generated by the decomposition of $HNO_2$ is oxidized with $O_2$ in the gas phase. The hydrolysis of $N_2O_4$, the decomposition of aqueous nitrous acid and the oxidation of NO in the gas phase were interpreted in the previous paper[22]. The values of $(\sqrt{DK_3}/H)_{N_2O_4}$ and reaction constants of $K_4$ and $K_5$ are used with the same values in the previous papers[22,23]. Where $K_4$ and $K_5$ are the reaction rate constant for reaction (4) and (5).

## THE CALCULATION OF THE FLUX OF NO, $NO_2$ AND $N_2O_4$

The steady-state transport of each absorbing component, j, across both the gas and liquid film is expressed by Danckwerts[24] as

$$I_j = -k_{G,j} \, (P_j^i - P_j) = (Ek_L)_j \, [C_j^i - C_j] \quad (7)$$

In the case of physical absorption or desorption, the enhancement factor, E, is taken to be unity. The absorbing species are $NO_2$ and $N_2O_4$ and the desorbing species are NO and $HNO_2$.

A flux of components, j, through the gas film may be represented by [6]

$$I_j = -\frac{D_{G,j}}{RT} \left(\frac{dP_j}{dz}\right) = k_{G,j} \, (P_j - P_j^i) \quad (8)$$

A flux of component $NO_2^*$ through the gas film in a steady-state process may be expressed by

$$I_{NO_2}^* = I_{NO_2} + 2I_{N_2O_4} \quad (9)$$

The local absorption rate of $NO_2^*$ and NO is expressed in terms of the gas-film mass-transfer coefficient as

$$-I_{NO_2}^* = \frac{1}{RT} \left(D_{NO_2}\frac{dP_{NO_2}}{dz} + 2D_{N_2O_4}\frac{dP_{N_2O_4}}{dz}\right) \quad (10)$$

and

$$-I_{NO} = \frac{D_{NO}}{RT} \frac{dP_{NO}}{dz} \quad (11)$$

The gradient of $N_2O_4$ in the gas film may be expressed as follows:

$$\frac{dP_{N_2O_4}}{dz} = 2K_1P_{NO_2}\frac{dP_{NO_2}}{dz} \quad (12)$$

Substituting Eq. (12) into Eq. (10) gives

$$-I_{NO_2}^* = \frac{1}{RT} \, (D_{NO_2} + 4D_{N_2O_4}K_1P_{NO_2})\frac{dP_{NO_2}}{dz} \quad (13)$$

Separating the variable and integration yields:

$$I_{NO_2}^* = k_{G,NO_2} \, (P_{NO_2} - P_{NO_2}^i)$$
$$+ 2k_{G,N_2O_4}K_1 \, \{P_{NO_2}^2 - (P_{NO_2}^i)^2\} \quad (14)$$

and

$$I_{NO} = k_{G,NO} \, (P_{NO} - P_{NO}^i) \quad (15)$$

The difference of $NO_2^*$ and NO can be expressed as:

$$I_m = I_{NO_2}^* - I_{NO} \quad (16)$$

Utilizing Eqs. (14) and (15), $I_m$ may be represented by

$$I_m = k_{G,NO_2} \, (P_{NO_2} - P_{NO_2}^i) + 2k_{G,N_2O_4}K_1 \, \{P_{NO_2}^2 - (P_{NO_2}^i)^2\}$$
$$- k_{G,NO} \, (P_{NO} - P_{NO}^i) \quad (17)$$

The absorption/desorption of the indicated gas species through the liquid film is represented by:

$$I_{NO_2}^* = (Ek_L)_{NO_2} \, [C_{NO_2}^i - C_{NO_2}]$$
$$+ 2 \, (Ek_L)_{N_2O_4} \, [C_{N_2O_4}^i - C_{N_2O_4}] \quad (18)$$

and

$$I_{NO} = (Ek_L)_{NO} \, [C_{NO}^i - C_{NO}] \quad (19)$$

Difining E to be 1 for physical absorption and assuming that the gas-phase resistance to the absorption rate of $N_2O_4$ is negligible and that the hydrolysis reaction of $N_2O_4$ is fast pseudo-first order, i.e. $C_{NO_2}$ and $C_{N_2O_4}$ are negligible, Eq. (18) and (19) become to be

$$I_{NO_2}^* = k_{L,NO_2}C_{NO_2}^i + 2 (\sqrt{DK_3})_{N_2O_4}C_{N_2O_4}^i \qquad (20)$$

and

$$I_{NO} = k_{L,NO} [C_{NO}^i - C_{NO}] \qquad (21)$$

Using the assumption that Henry's Law applies at the gas-liquid interface, $[C^i = H^iP^i]$

$$I_{NO_2}^* = (\frac{k_L}{H})_{NO_2}P_{NO_2}^i + 2 (\frac{\sqrt{DK_3}}{H})_{N_2O_4}P_{N_2O_4}^i \qquad (22)$$

or

$$I_{NO_2}^* = (\frac{k_L}{H})_{NO_2}P_{NO_2}^i + 2 (\frac{\sqrt{DK_3}}{H})_{N_2O_4}K_1 (P_{NO_2}^i)^2 \qquad (23)$$

$$I_{NO} = (\frac{k_L}{H})_{NO}P_{NO}^i - k_{L,NO}C_{NO} \qquad (24)$$

and

$$I_m = (\frac{k_L}{H})_{NO_2}P_{NO_2}^i + 2 (\frac{\sqrt{DK_3}}{H})_{N_2O_4}K_1 (P_{NO_2}^i)^2$$
$$- (\frac{k_L}{H})_{NO}P_{NO}^i + k_{L,NO}C_{NO} \qquad (25)$$

Utilizing Eq. (14) and (23), after some rearrangement,

$$\{2K_1 (\frac{\sqrt{DK_3}}{H})_{N_2O_4} + 2K_1k_{G,N_2O_4}\} (P_{NO_2}^i)^2 + \{(\frac{k_L}{H})_{NO_2}$$
$$+ k_{G,NO_2}\} P_{NO_2}^i - 2K_1k_{G,N_2O_4}P_{NO_2}^2 - k_{G,N_2O_4}P_{NO_2} = 0 \qquad (26)$$

Eq. (26) is a quadratic formula of $P_{NO_2}^i$, and then $P_{NO_2}^i$ may be obtained by taking the positive root.

The bulk-phase concentration of NO, $C_{NO}$, is calculated from a modified Abel-Schmid's expression[16],

$$C_{NO} = \frac{P_{NO}}{H_{NO}} = \frac{1}{H_{NO}} (\frac{C_{HNO2}^3}{K_4 C_H + C_{NO_3}})^{1/2} \qquad (27)$$

Substitution and rearrangement of Eq. (17) into Eq. (25) become to be

$$P_{NO}^i = \frac{1}{k_{G,NO} + (k_L/H)_{NO}} [\{(\frac{k_L}{H})_{NO_2} + k_{G,NO_2}\} P_{NO_2}^i$$
$$+ \{2 (\frac{\sqrt{DK_3}}{H})_{N_2O_4}K_1 + 2k_{G,N_2O_4}K_1\} (P_{NO_2}^i)^2 - k_{G,NO_2}P_{NO_2}$$
$$- 2k_{G,N_2O_4}K_1P_{NO_2}^2 + k_{G,NO} + k_{L,NO}C_{NO}] \qquad (28)$$

$P_{NO}^i$ and $P_{NO_2}^i$ are determined, and then the flux of $NO_2$, $N_2O_4$ and NO are calculated as:

$$I_{NO_2} = (\frac{k_L}{H})_{NO_2}P_{NO_2}^i \qquad (29)$$

$$I_{N_2O_4} = (\frac{\sqrt{DK_3}}{H})_{N_2O_4}P_{N_2O_4}^i \qquad (30)$$

and

$$I_{NO} = (\frac{k_L}{H})_{NO}P_{NO}^i - k_{L,NO}C_{NO} \qquad (31)$$

## THE CONVERSION OF NO TO $NO_2$ IN THE GAS-PHASE OF A PACKED COLUMN

Bodenstein studies the gas-phase oxidation of gaseous NO and confirmed the third-order kinetics consistent with the following rate equation[17].

$$- \gamma_{NO} = K_5 P_{NO}^2 P_{O_2} \qquad (32)$$

The gas flowing through a packed column is considered to be in plug-flow. At steady state the material balance for NO around a differential gas space is

$$\begin{array}{ll} \text{Input} & = \text{Output} + \text{Disappearance} \\ \text{of NO} & \text{of NO} \quad \text{by reaction} \end{array} \qquad (33)$$

The system is represented by the following equation such as $G(P_{NO})_{in}/(RT)$, $G(P_{NO})_{out}/(RT)$, and $[G(-\gamma_{NO})/RT]\tau$. Inserting these terms in Eq. (33), which, after rearrangement and division by RT, yields:

$$G (P_{NO})_{in}dX_{NO} = - \gamma_{NO}dV \qquad (34)$$

Further rearrangement yields an integral form,

$$\frac{1}{G (P_{NO})_{in}} \int_0^V dV = \int_0^{X_{NO}} \frac{dX_{NO}}{- \gamma_{NO}} \qquad (35)$$

The reaction rate may be expressed as:

$$- \gamma_{NO} = K_5 \{ (P_{NO})_{in} (1 - X_{NO}) \}^2 (P_{O_2})_{in}$$
$$(1 - \frac{(P_{NO})_{in}}{2 (P_{O_2})_{in}} X_{NO}) \qquad (36)$$

Combining terms and integration yields:

$$\frac{K_5 (P_{NO})_{in} (P_{O_2})_{in}V}{G} - \frac{1}{1 - (P_{NO})_{in}/2 (P_{O_2})_{in}} \{\frac{1}{1 - X_{NO}}$$
$$+ \frac{(P_{NO})_{in}/2 (P_{O_2})_{in}}{1 - \frac{(P_{NO})_{in}}{2 (P_{O_2})_{in}}} Ln \frac{1 - (P_{NO})_{in}/2 (P_{O_2})_{in}X_{NO}}{1 - X_{NO}} \} = 0 \qquad (37)$$

Solution of this polynomial involves finding the root, $X_{NO}$, between zero and 1. This solution is accomplished using a simple bisection routine.

## A MODEL FOR NO$_x$ ABSORPTION ON THE PACKED COLUMN

The absorption phenomena involved in the scrubbing of NO$_x$ from gas streams have been simulated for an incremental column volume as shown in Fig. 1. The change in component partial pressures and concentrations, as well as gas and liquid flow rates in the increment, may be neglected. The gas-phase is assumed to be ideal and isothermal conditions. The mathematical model is developed to be used in the computation of the partial pressure of gas species leaving the increment and concentration of liquid species entering the increment. The mathematical model developed is based on mass-transfer data for packed column and chemical reaction information for the NO$_x$-H$_2$O-HNO$_x$ systems.

The steady-state gas-phase performance equation for the incremental absorption phenomena of NO$_2$* and NO may be expressed as

input = output + disappearance + disappearance/
         due to gas-    appearance due
       phase reaction  to absorption
                   or desorption

                                  (38)

In the above equation, the disappearance terms are positive while the appearance terms are negative. The molar input of the component, j, to the column incre-
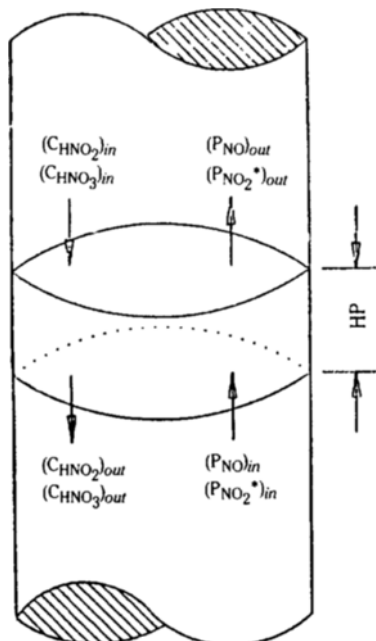


**Fig. 1. Representation of incremental volume in a packed column.**

ment may be represented as $G(P_j)_{in}/RT$, while the output is $G(P_j)_{out}/RT$.

The extent of the gas-phase oxidation of NO to NO$_2$ occurring in the increment, X$_{NO}$, is computed and used to adjust the molar rates of NO and NO$_2$ leaving the increment as $\pm \{G\,P_{NO}\,X_{NO}/(RT)\}$.

The molar rate of disappearance or appearance of NO$_2$* and NO in the incremental gas-phase is obtained as a $(I_{NO_2}^* a \cdot \Delta V)$ and $(I_{NO} a \cdot \Delta V)$. The term V is the incremental column volume. The developed mass balance equation in the gas-phase performance yields[23]:

$$\frac{G\,(P_{NO_2}^*)_{in}}{RT} = \frac{G\,(P_{NO_2}^*)_{out}}{RT} - (I_{NO_2} + 2I_{N_2O_4})\,a \cdot \Delta V$$

$$- \frac{GP_{NO}X_{NO}}{RT} \qquad (39)$$

$$\frac{G\,(P_{NO})_{in}}{RT} = \frac{G\,(P_{NO})_{out}}{RT} + (I_{NO})\,a \cdot \Delta V + \frac{GP_{NO}X_{NO}}{RT} \quad (40)$$

An overall steady-state performance equation for the liquid-phase of the incremental absorption reaction phenomena with respect to aqueous nitric and nitrous acids may be expressed as:

input of = output of − production from
HNO$_3$       HNO$_3$       reactions

         − production from
           decomposition of HNO$_3$    (41)

input of = output of − appearance
HNO$_2$       HNO$_2$   from reaction

         + decomposition
         of HNO$_2$      (42)

The production of nitric acid in the increment due to the decomposition may be represented by $\{1/2\,k_{L,NO}\,[C_{NO} - C_{NO}^i]\}$ and the appearance of nitrous acid by $\{-3/2\,k_{L,NO}\,(C_{NO} = C_{NO}^i)\}$. Incorporating these relationships into the liquid-phase performance equations yields:

$$L\,(C_{HNO_3})_{in} = L\,(C_{HNO_3})_{out} - (1/2I_{NO_2} + I_{N_2O_4})\,a \cdot \Delta V$$

$$+ 1/2I_{NO}\,a \cdot \Delta V \qquad (43)$$

$$L\,(C_{HNO_2})_{in} = L\,(C_{HNO_2})_{out} - (1/2I_{NO} + I_{N_2O_4})\,a \cdot \Delta V$$

$$- 3/2I_{NO}\,a \cdot \Delta V \qquad (44)$$

By knowing or assuming $(P_{NO_2})_{in}$, $(P_{NO})_{in}$, $(C_{HNO_2})_{out}$ and $(C_{HNO_3})_{out}$ the absorption efficiency of NO$_x$, X$_{NO_x}$, may be calculated at the increment from Eq. (39) and (40) provided I$_{NO_2}$, I$_{N_2O_4}$ and I$_{NO}$ may be calculated. By using multiple increments, the NO$_x$ removal performance of a packed column of any finite height can be modeled. An incremental height of 1 cm was used in these calculation.

For the mass-transfer coefficient in the packed column the values of $k_G$ and a were calculated from correlation by Onda et al.[25].

$$\frac{k_G RT}{a_t D_G} = 2.0 \left(\frac{G'}{a_t \mu_G}\right)^{0.7} \left(\frac{\mu_G}{\rho_G D_G}\right)^{0.33} (a_t d_p)^{-2.0} \quad (45)$$

$$\frac{a}{a_t} = 1 - \exp\left[-1.45 \left(\frac{\sigma_c}{\sigma_L}\right)^{0.75} \left(\frac{L'}{a_t \mu_L}\right)^{0.1}\right.$$

$$\left.\left(\frac{L'^2 a_t}{\rho_L^2 g}\right)^{-0.05} \left(\frac{L'^2}{\rho_L \sigma_L a_t}\right)^{0.2}\right] \quad (46)$$

Also, the value of $k_L$ was taken from experimental results by Mohunta et al.[26].

$$k_L a \left(\frac{a_t \mu_L}{g \rho_L}\right)^{2/3} \left(\frac{\mu_L}{g^2 \rho_L}\right)^{1/9} = 0.0025 \left(\frac{\mu_L L'^3 a_t^3}{g^2 \rho_L^4}\right)^{1/4}$$

$$\left(\frac{\mu_L}{\rho_L D_L}\right)^{-1/2} \quad (47)$$

## DISCUSSION

The program is designed to predict $NO_x$ absorption efficiencies. The program moves increment by increment up to the top of a column iteration. Column iterations that are at least equal to the number of increments are required, due to the gas-liquid equilibrium expressed in reaction (4). When a molar balance of acid produced to $NO_x$ absorbed is achieved, the program becomes to be converged to a solution.

The most effect on the absorption of $NO_x$ is the oxidation state of the gas because the solubility of NO gas is much lower than the $NO_2$ and $N_2O_4$. The next largest effect is the partial pressure of nitrogen oxides in the feed gas. The higher partial pressure of nitrogen oxides give more absorbable and reactive. This is explained by the increase of the driving force for the $NO_x$ absorption.

The most critical value is the constant of $(\sqrt{DK_3}/H)_{N_2O_4}$, ranging from $5.7 \times 10^{-4}$ to $11 \times 10^{-4}$ kg·mol/atm·m²sec. Model prediction of $X_{NO_x}$ were calcu- as the value of $(\sqrt{DK_3}/H)_{N_2O_4}$ is reasonable through the the various investigators. The $NO_x$ efficiencies predicted by this model are compared with the experimental data in the next paper (part II)[27]. As shown in figures in the paper the model prediction shows to be good agreement with the experimental data. In this study to select $11 \times 10^{-4}$ kg·mol/atm·m²·sec as the value of $(\sqrt{DK_3}/H)_{N_2O_4}$ is reasonable through the comparison with the experimental data.

## CONCLUSION

1. A mathematical $NO_x$ absorption model for

packed column was developed based on mass-transfer information, chemical reaction and mechanistic phenomena specific to the $NO_x$-$HNO_3$-$HNO_2$-$H_2O$ system.

2. This model agrees with the experimental results quite well. This model could be useful for $NO_x$ absorber design.

## NOMENCLATURE

a : gas-liquid interfacial area per unit volume [m²/m³]

$a_t$ : total surface area of packing per unit volume [m⁻¹]

$C_j$ : liquid-phase concentration of component j [kg·mol/m³]

$C'_j$ : interfacial concentration of component j [kg·mol/m³]

$D_j$ : diffusivity of component j [m²/s]

$D_G$ : diffusivity in the gas phase [m²/s]

$D_L$ : diffusivity in the liquid phase[m²/s]

E : enhancement factor [-]

G : gas flow rate [m³/s]

G' : superficial mass velocity of gas [kg/m²·s]

L' : superficial mass velocity of liquid [kg/m²·s]

g : gravitational constant [m²/s]

$H_j$ : Henry's law constant of component j [m³ atm/kg·mol]

$I_j$ : local absorption rate of component j [kg·mol/m³·s]

$K_j$ : reaction rate constant for reaction i [(kg·mol/m)$^{1-n}$/s]

$k_{Gj}$ : gas-phase mass-transfer coefficient for component j[kg·mol/m²·atm·s]

$k_{Lj}$ : liquid-phase mass-transfer coefficient for component j [kg·mol/m²·atm·s]

$K_{P,1}$ : pressure equilibrium constant for reaction (1) [atm]

L : liquid flow rate [m³/s]

$NO_2^*$ : $NO_2 + 2N_2O_4$

$NO_x$ : $NO + NO_2^*$

$P_j$ : partial pressure of gas compoent j [atm]

R : gas constant [m³·atm/kg·mol·K]

T : absolute temperature [K]

V : volume of column [m³]

$X_{NO}$ : conversion fraction of NO [-]

### Greek Letters

$\tau$ : residence time [S]

$\Delta V$ : incremental column volume [m³]

$\sigma_c$ : critical surface tension [kg/s]

$\sigma_L$ : surface tension [kg/s]

$\mu_G$ : viscosity of gas [kg/m·s]

$\tau_L$    :   viscosity of liquid [kg/m·s]

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