226 (1966).

- 4) Boys, G. S. and W. H. McAdams: Ind. Eng. Chem., 29, 1240 (1937).
- 5) Harriott, P.: Chem. Eng. Prog. Symp. Ser., 29, 137 (1959).
- 6) Higbie, R.: Trans. AIChE, 31, 365 (1935).
- 7) Kern, D. Q. and H. J. Karakas: Chem. Eng. Prog. Symp. Ser., 29, 141 (1959).
- 8) Kool, J.: Trans. Inst. Chem. Engrs., 36, 259 (1958).
- 9) Lustenader, E. L. et al.: Trans. ASME J. Heat Transfer, 81C, 297 (1959).
- 10) Mizushina, T.: "Advances in Heat Transfer", Vol. 7, Academic Press (1971).
- 11) Skelland, A. H. P., D. R. Oliver and S. Tooke: Brit. Chem. Eng., 7, 346 (1962).

ABSORPTION OF DILUTE NITRIC MONOXIDE IN AQUEOUS SOLUTIONS OF Fe(II)-EDTA AND MIXED SOLUTIONS OF Fe(II)-EDTA AND Na_2SO_3

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Absorption rates of NO in aqueous solutions of Fe(II)-EDTA chelate as well as in mixed solutions of Fe(II)-EDTA and Na_2SO_3 were measured using a stirred vessel with a free flat gasliquid interface and a bubble column.

The rate constants of the complexing reaction of NO with Fe(II)-EDTA were determined on the basis of the theory of gas absorption accompanied by a reversible reaction. The chemical equilibrium constants were also determined at various pH values. It was found that the rate constant was of the order of $10^8 l/g$ -mol·sec and that the equilibrium constant was about $10^8 l/g$ -mol at 25°C. These values are much higher than the corresponding values of the reaction between NO and Fe(II) in the absence of EDTA.

The mechanism of the absorption of NO in mixed solutions of Fe(II)-EDTA and Na_2SO_3 was deduced from the observation that the absorption efficiency decreased in the early stage of absorption and then increased to some steady value. The absorption rates were satisfactorily explained on the assumption that NO coordinates to Fe(II) (EDTA) (SO_3^{2-}) irreversibly.

It was also found that the absorption rate of NO in the aqueous solution of Fe(II)-EDTA was much higher than those of other liquid absorbents so far investigated.

Introduction

Removal of NO_x in flue gases has recently become important preventing air pollution, and many processes to remove NO_x by liquid absorbents as well as solid-catalyzed reactions have been proposed. Recently it was shown that aqueous solutions of Fe(II) chelates, especially Fe(II)-EDTA chelate, are promising liquid absorbents of NO because of their very fast absorption rates and easy regeneration^{4,14}.

When NO is absorbed into Fe(II) chelate solutions,

it coordinates to Fe(II). As absorption proceeds, the absorption rate decreases because of the consumption of Fe(II)-EDTA and the reversible nature of the complexing reaction. However, if some reducing agents such as Na_2SO_3 , which can reduce NO coordinating to Fe(II), are added to the solution, high absorption rate can be maintained.

The purpose of this paper is to investigate the chemical equilibrium and the kinetics of the complexing reaction between NO and Fe(II)-EDTA and the absorption mechanism of NO both in the absence and in the presence of Na_2SO_3 , using a bubble column and a stirred vessel with a free flat gas-liquid interface. The effect of experimental conditions on the conversion of NO to N_2O was also examined.

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1. Experimental

Two types of gas absorbers were used, i.e., a bubble column and a stirred vessel. The bubble column was 2.5 cm in diameter. A nozzle (1.6 mm) or a ball filter (G1, 15 mm) was used as the gas distributor. The liquid volume was 80 ml and the gas flow rates were about 300 and 600 Nml/min.

The stirred vessel, 7.5 cm in diameter, was equipped with four vertical baffles and two stirrers, a Tefloncoated magnetic stirring bar (4.0 cm in length) in the liquid phase and a propeller-type stirrer (4.4 cm in diameter) in the gas phase. In most of the experiments, the stirring speed of the liquid phase, n_L , was adjusted at 450 rpm and that of the gas phase, n_{g} , at 1800 rpm. The vessel was filled with 300 ml of the solution and the mixture of NO and N₂ was introduced above the free liquid surface at a flow rate of 300 or 600 Nml/min. and NO was absorbed through the free gas-liquid interface. The concentration of NO was measured by a Shimadzu CLM-201 type NO_x monitor, and the absorption rate of NO was calculated from the gas flow rate and the difference between the concentrations of NO in the feed and effluent gases. The determination of N₂O in the effluent gas was carried out by a gas chromatograph (column packing: Porapack Q, 2m; carrier gas: helium; column temperature: 50°C). The chelate solutions were prepared by adding equimolar amounts of FeSO4 and EDTA-2Na to deionized water in an atmosphere of nitrogen, and pH of the solution was adjusted by NaOH or H₂SO₄.

The details of the experimental apparatus and procedure were described elsewhere15,17.

2. Predictions of Physical Properties and Mass Transfer Coefficients

The gas-phase and liquid-phase mass transfer coefficients at the free gas-liquid interface of the stirred vessel used in the present work were correlated with the stirring speeds of the impellers by the following equations at $25^{\circ}C^{15}$.

$$k_G = 4.33 \times 10^{-10} n_G^{0.71}$$
 (SO₂ in N₂) (1)

$$k_L = 3.19 \times 10^{-5} n_L^{0.756}$$
 (CO₂-water) (2)

The values of k_G and k_L for the transfer of NO under other conditions were estimated by the following empirical equations⁶.

$$k_G \infty (\rho_G / \mu_G)^{0,37} D_G^{2/3} \tag{3}$$

$$k_L \infty (\rho_L / \mu_L)^{0.37} D_L^{2/3} \tag{4}$$

The liquid-phase diffusivities of NO and Fe(II)-EDTA and the gas-phase diffusivity of NO were estimated by Wilke-Chang's equation²⁰⁾ and Chapman-Enskog's equation¹⁾, respectively. In estimating the molar

Table 1 Physical properties of liquid absorbents

Temp. [°C]	[Fe(II)- [EDTA] [M]	Na ₂ SO ₃] [M]	ρ $\begin{bmatrix} g \\ cm^3 \end{bmatrix}$	μ [cP]	$\frac{D_A \times 10^5}{\left[\frac{\mathrm{cm}^2}{\mathrm{sec}}\right]}$	$ \begin{bmatrix} \underline{\operatorname{atm}} \cdot l \\ \overline{\operatorname{g-mol}} \end{bmatrix} $
25	0.01	0	1.0	0.896	2.51	521
25	0.02	0	1.0	0.907	2.48	521
25	0.03	0	1.0	0.917	2.47	521
25	0.01	0.75	1.09	1.32	1.77	951
25	0.02	0.25	1.03	1.02	2.20	637
15	0.02	0.25	1.03	1.28	1.70	532
35	0.02	0.25	1.03	0.852	2.74	763



Fig. 1 Time-course of η in the absorption of NO in Fe(II)-EDTA solution (bubble column with ball filter)

volume of Fe chelate, the contribution of Fe was not taken into account because the atomic volume of Fe had not been reported. It was confirmed, however, that the error due to this approximation was at most 5% because of the very large molar volume of EDTA compared with the atomic volume of Fe. Solubility of NO in the liquid absorbent was evaluated by the method of van Krevelen and Hoftijzer¹⁹.

$$\alpha_w/\alpha = 10^{k_s t} \tag{5}$$

$$k_s = x_g(NO) + x_c(Na^+) + x_a(SO_3^{2-})$$
 (6)

where $x_g(NO)$ is -0.1931^{15} , $x_c(Na^+)$ is -0.0183^{11} , $x_a(SO_3^{2-})$ is 0.3275^{11} and α_w is $0.0430 \text{ m}l/\text{m}l^{9}$ at 25°C. Under the conditions studied the concentration of Fe(II)-EDTA was relatively low (<0.04 M), and therefore the contribution of Fe(II)-EDTA to the solubility was neglected. The physical properties of the absorbents are summarized in **Table 1**.

3. Results and Discussion

3.1 Absorption of NO in Fe(II)-EDTA solutions

1) Chemical equilibrium constant The equilibrium constant of the complexing reaction (a) was measured by means of a flow method.

 $NO + Fe(II) - EDTA \simeq Fe(II)(EDTA)(NO)$ (a)

The diluted NO was introduced into the bubble column containing 80 m/ of Fe(II)-EDTA solution, and the time-course of η , the absorption efficiency of



Fig. 2 Temperature dependence of K



Fig. 3 Effect of pH on N_A (stirred vessel, $[Na_2SO_3]=0$)



Fig. 4 N_A vs. A_i (stirred vessel, $[Na_2SO_3]=0$)

NO, was measured. A typical example of the data is shown in **Fig. 1**. The absorption efficiency gradually decreased and when chemical equilibrium was attained no more NO was absorbed. Based on this figure, the value of the equilibrium constant K was calculated from the following equations.

$$K = [Fe(II)(EDTA)(NO)]_{e}/([NO]_{e}[Fe(II)-EDTA]_{e})$$

$$[NO]_{e} = Py_{f}/H$$

$$[Fe(II)-EDTA]_{e} = [Fe(II)-EDTA]_{ini}$$

$$-(G_{M}y_{f}/L)\int_{0}^{\infty} \eta \, dt + [NO]_{e}$$

$$[Fe(II)(EDTA)(NO)]_{e} = (G_{M}y_{f}/L)\int_{0}^{\infty} \eta \, dt - [NO]_{e}$$

$$(8)$$

Figure 2 shows the temperature dependence of K at various pH values, where the data of Tanaka *et al.*¹⁴) are also shown. The value of K is in the order of 10^{6} l/g-mol and is about four orders of magnitude over that in the absence of EDTA⁸).

2) Reaction rate constant The effect of pH on absorption rate was investigated with the stirred vessel, and is shown in Fig. 3, where y_0 is the concentration of NO in the effluent gas. This figure indicates that N_A does not change with pH above 4.6, whereas at pH=2.43, N_A is quite low. This is explained as follows. In the pH range above approximately 4, Fe(II)-EDTA (normal chelate) is almost quantitatively formed from the equimolar amounts of FeSO4 and EDTA, whereas at pH=2.43, the concentrations of Fe(II)-EDTA and Fe(II)-HEDTA (protonated chelate) formed under the condition shown in Fig. 3 were calculated to be 0.0017 and 0.0040 M, respectively¹²⁾, and the total concentration of these chelates was only 0.0057 M. This low concentration is the cause of the low absorption rate.

The plots of N_A vs. A_i for various concentrations of Fe(II)-EDTA are shown in Fig. 4. Absorption rate was not increased by adding excess of EDTA. The reaction rate constant of reaction (a) was determined from the theory of gas absorption accompanied by a reversible chemical reaction. The approximate absorption rate based on the film model is expressed by

$$N_A = \beta k_L (A_i - A_0) \tag{9}$$

with

$$\begin{split} \beta &= [\gamma \xi / \{U \tanh(\gamma \xi)\}] [(\beta'-1) \{(\beta_{\infty}-\beta) + (\beta-1)(A_0/A_i)\} \\ &- r_c q_c (\beta'-\beta_{\infty})] - [\gamma \xi / \{U \sinh(\gamma \xi)\}] [(\beta_{\infty}-1) \{(\beta'-\beta) \\ &+ (\beta-1)(A_0/A_i)\} (A_0/A_i) - r_c q_c (\beta'-\beta_{\infty})] \end{split}$$
(10)

where

$$\gamma = \sqrt{kB_0 D_A} / k_L \tag{11}$$

$$\xi = \sqrt{\frac{\beta_{\infty}}{\beta_{\infty} - 1} - \frac{\beta}{\beta' - 1} + \frac{\beta - 1}{\beta' - 1} \left(\frac{A_0}{A_i}\right)} \tag{12}$$

$$U = (\beta_{\infty} - 1) \{ (\beta' - \beta) + (\beta - 1)(A_0/A_i) \} \{ 1 - (A_0/A_i) \}$$
(13)

Here β_{∞} is the reaction factor for the case when the reaction (a) is instantaneous and reversible¹⁰, and β' that corresponding to an instantaneous irreversible reaction⁵.

$$\beta_{\infty} = 1 + KB_0 r_C / (1 + KA_i r_C / r_B)$$
(14)

$$\beta' = 1 + r_B q_B \tag{15}$$

There absorption rate based on the penetration model was approximated by Eqs. (9)–(15) in which the terms representing diffusivity ratio are replaced with their square roots. Because absorption mechanism through a free gas-liquid interface follows the penetration model rather than the film model^{18,21)} the former model was adopted in this work. As initial reaction rates were measured, the value of A_0 in these equations

could be taken as zero. The value of k thus obtained at 25°C is $1.7 \times 10^8 l/g$ -mol·sec in the pH range 4.6-8.0. At pH=7.7, the values are 0.8×10^8 and $2.6 \times$ $10^{8} l/g$ -mol·sec at 15°C and 35°C, respectively. Under the conditions studied, q_B ranged from 5.6×10^4 to 6.5×10^5 , γ from 1800 to 3400, β from 1480 to 2970, β' from 1.7×10^4 to 2.2×10^5 , β_{∞} from 2900 to 20600, y_f from 75 to 300 ppm and y_0 from 29 to 160 ppm. The rate constant is markedly high and about 300 times higher than that of the complexing reaction between NO and Fe(II) in the absence of EDTA⁸, and about 50 times higher than that of the reaction between NO and KMnO₄¹⁵). The solid lines in Fig. 4 represent the absorption rates calculated by Eqs. (9)-(15) with $k=1.7\times10^8 l/g$ -mol·sec and $K=1.6\times10^6$ l/g-mol. The agreement between observed and calculated results is satisfactory.

3) Interpretation of η vs. time curves Simulation of the time-course of η , which was obtained with the bubble column and is shown in Fig. 1, was carried out based on the assumption that the effluent gas was in equilibrium with the liquid because of very fast chemical reaction rate. Based on this assumption, η at the start of absorption is unity because no NO complex exists in the liquid and therefore reaction (a) can be considered irreversible at t=0. This initial condition was approximately satisfied in the present absorption system, as shown in Fig. 1. The η vs. time relation is expressed as follows:

$$t = \frac{mn}{(m+1)^2} \ln \frac{m(1-\eta)+1}{\eta} + \frac{m^2n}{m+1} \frac{1-\eta}{m(1-\eta)+1}$$
(16)

where $m = KPy_f/H$ and $n = [Fe(II)-EDTA]_{ini}L/G_My_f$. It is seen from Fig. 1 that the calculated line based on Eq. (16) agrees fairly well with the experimental result. It must be noted that Eq. (16) cannot be applied to the case where the reaction rate is not so fast.

3. 2 Absorption of NO in mixed solutions of Fe(II)-EDTA and Na_2SO_3

1) Reaction mechanism When Na_2SO_3 is added to Fe(II)-EDTA solution, the colour of the solution changes from yellow to deep yellow. It was found from the UV and visible spectra of the mixed solutions of Na_2SO_3 and Fe(II)-EDTA that the presence of Na_2SO_3 markedly influences the spectra, indicating that there exists an interaction between Fe(II)-EDTA and SO_3^{2-} .

$$Fe(II)$$
-EDTA+ $SO_3^{2-} \rightleftharpoons Fe(II)(EDTA)(SO_3^{2-})$ (b)

When NO is absorbed into mixed solution of Fe(II)-EDTA and Na_2SO_3 , NO probably coordinates to Fe(II) according to reaction (c), and then it is supposedly reduced by Na_2SO_3 to produce N_2O , sodium sulfamate, sodium disulfamate and sodium dithionate¹³).

$Fe(II)(EDTA)(SO_3^{2-})$ +NO \rightleftharpoons Fe(II)(EDTA)(SO_3^{2-})(NO) (c)

The time-courses of η observed with the bubble column are shown in Figs. 5(a)-8(a). At the early stage of the absorption, η is quite high. However, as the absorption proceeds η decreases to a minimum value η_{\min} , and then increases to a value slightly lower than the initial value. As shown in Fig. 5(a), as [Na₂SO₃] increases, η_{\min} decreases and η is restored to the higher value. It is shown in Fig. 6(a) that η_{\min} and the time required to reach η_{\min} decrease with an increase in y_t , the concentration of NO in the feed gas. Based on these very interesting phenomena the following reaction mechanism may be deduced. The decrease of η in the initial stage of the absorption is probably due to the accumulation of NO complex and the consumption of Fe(II) chelate. In this stage the reduction rate of NO complex is not so fast that considerable amount of NO complex may accumulate in the liquid. The rate of elimination of NO by Na₂SO₃ from the NO complex which is formed in the early stage of absorption is probably slow. However, this complex gradually converts to some complex which can be easily regenerated by Na₂SO₃ to form NO free Fe(II)-EDTA, and this results in the increase in η with time. The supporting evidence that the fast reduction of NO complex proceeds in the second stage is the fact that the evolution of N₂O begins to increase in this stage as shown in Figs. 5(b)-8(b). Furthermore, Fig. 9 shows the η vs. time relation observed when Na₂SO₃ was added to the liquid in the course of absorption into Fe(II)-EDTA solution. It is seen that η increases rapidly just after the addition, and thus the reduction of Fe(II)(EDTA)(NO) which has been formed in the absence of Na₂SO₃ is considered to be rapid. Although the detailed mechanism is not clear, the following mechanism is presented here.

 $\begin{aligned} & \mathsf{Fe}(\mathrm{II})\text{-}\mathrm{EDTA} + \mathrm{SO}_3^{2-} \rightleftharpoons \mathsf{Fe}(\mathrm{II})(\mathrm{EDTA})(\mathrm{SO}_3^{2-}) \quad \text{(b)} \\ & \mathsf{Fe}(\mathrm{II})(\mathrm{EDTA})(\mathrm{SO}_3^{2-}) + \mathrm{NO} \rightleftharpoons \mathsf{Fe}(\mathrm{II})(\mathrm{EDTA})(\mathrm{SO}_3^{2-})(\mathrm{NO}) \quad \text{(c)} \end{aligned}$



When NO dissolves in the mixed solution of Fe(II)-EDTA and Na₂SO₃, both Fe(II)(EDTA)(NO) and Fe(II) (EDTA) (SO₃²⁻) (NO) are formed via reactions (a) and (c). The former is easily reduced by Na₂SO₃ via reaction (g), as shown in Fig. 9. The latter is considered to be produced much more than the former because the equilibrium of reaction (b) shifts to the







Fig. 8(b) Effect of temperature on y_{N_2O}

right. The complex $Fe(II)(EDTA)(SO_3^{2-})(NO)$ releases SO_3^{2-} to produce Fe(II)(EDTA)(NO), which can be easily reduced by Na_2SO_3 with the formation of $Fe(II)(EDTA)(SO_3^{2-})$ (reaction (g)). There is some

possibility of reactions (e) and (f). In the presence of NO, Fe(II)-EDTA probably does not react with SO_3^{2-} to a great extent because of marked reactivity of NO with Fe(II)EDTA. If it is assumed that reactions (d), (e) and (f) are slow compared with reaction

(g), and also that gradual conversion of Fe(II)- $(EDTA)(SO_3^{2-})$ to Fe(II)-EDTA through reactions (d) and (e) occurs during the reaction, the behavior shown in Figs. 5-8 can be satisfactorily explained in the following manner. When [Na₂SO₃] is high, the equilibrium of reaction (b) shifts to the right and η_{min} decreases. However, after most $Fe(II)(EDTA)(SO_3^{2-})$ -(NO) is converted to Fe(II)(EDTA)(NO), higher concentration of Na₂SO₃ makes reaction (g) faster, the steady-state value of [Fe(II)-EDTA] higher and therefore the restored value of η higher, as shown in Fig. 5(a). When y_f is high, relatively high concentration of Fe(II)(EDTA)(SO²⁻)(NO) accumulates in a short time, and η_{\min} as well as the time required to reach η_{\min} decrease, as shown in Fig. 6(a). Effect of [Fe(II)-EDTA] is shown in Fig. 7(a). It is a matter of course that η_{\min} increases with an increase in [Fe(II)-EDTA]. As shown in Fig. 8(a), a considerable temperature effect was observed. With increasing temperature, η_{\min} increases. This is probably due to the high activation energies of reactions (d) and (g) compared with those of reactions (a) and (c).

It was also confirmed by absorption of NO, diluted by helium and free from N_2 in a mixed solution of Fe(II)-EDTA and Na_2SO_3 , that N_2 was not formed in the reduction of NO-Fe(II) complex by Na_2SO_3 .

The molar ratio of NO absorbed to SO_3^{2-} consumed was determined under the following conditions; y_f : 2000, 5000 ppm; [Fe(II)-EDTA]: 0.01, 0.02, 0.03 M; [Na₂SO₃]: 0.1, 0.25 M; pH 7; temperature: 25°C. The concentration of SO₃²⁻ was determined by redox titration using a pH meter with a platinum electrode. The ratio thus obtained was about 1.4, irrespective of the reaction condition.

2) Absorption rate Figure 10 shows the effect of $[Na_2SO_3]$ on absorption rate in the stirred vessel. It is seen that N_A does not depend on $[Na_2SO_3]$ in the range 0.05–0.25 M. From the concentration dependence of N_A shown in Fig. 11, N_A can be empirically expressed by

$$N_A = \sqrt{k' [\text{Fe(II)-EDTA]} D_A} A_i$$
(17)

This equation corresponds to the gas absorption rate accompanied by an irreversible pseudo first order reaction in the range $\gamma > 3^{5_0}$. It was also found that N_A was not influenced by k_L . Thus it is concluded that the reaction (c) can be considered irreversible, probably owing to the interaction of NO with SO_3^{2-} which coordinates to Fe(II). The rate constant is $1.4 \times 10^8 \ l/g$ -mol·sec at 25°C, and this is a little smaller than the rate constant of reaction (a). The ranges of the values of the parameters in this experiment are as follows; γ and β : 1810–3430; β' : 1.8×10^4 – 2.3×10^5 ; y_0 : 36–160 ppm. It was found that the rate constant was little dependent on temperature in the range



Fig. 9 Effect of addition of Na_2SO_3 on η in the course of absorption of NO in Fe(II)-EDTA solution (bubble column)



Fig. 10 Effect of $[Na_2SO_3]$ on N_A (stirred vessel)



Fig. 11 N_A vs. A_i (stirred vessel)

from 15 to 35° C.

To study the effect of the experimental condition on the absorption rate at relatively high values of k_L , a series of experiments were carried out with the bubble column. The value of k_L for "large bubbles" (>2.5 mm in diameter) estimated by the equation of Calderbank *et al.*²⁾ was 0.036 cm/sec, and about 12 times larger than that at the free surface of the stirred vessel. The plots of R_A vs. $(y)_{lm}$ for various concentrations of the chelate are shown in **Fig. 12**. The following equation is derived by assuming that the flow pattern of the gas is plug flow and that Eq. (17) also holds in this case.

$$R_A = S\sqrt{k'[\text{Fe(II)-EDTA}]D_A}P(y)_{lm}/H$$
(18)



Fig. 12 R_A vs. $(y)_{lm}$ (bubble column with nozzle)



Fig. 13 Comparison of absorption rates of NO in various liquid absorbents

The ranges of the estimated values of the parameters are as follows; γ : 90-260; β' : 1.0×10^4 -2.9 $\times 10^5$. These values satisfy the relation $\beta' \gg \gamma \gg 3$, the condition for Eq. (17) to hold. It can be seen that the plots shown in Fig. 12 are in accordance with Eq. (18). The total gas-liquid interfacial area estimated from Eq. (18) with $k'=1.4\times10^8 l/g$ -mol·sec was about 40 cm².

3.3 Comparison of absorption rates into various liquid absorbents

Comparison of the absorption rates of NO into various liquid absorbents is shown in Fig. 13. All these data are absorption rates of NO through the free gas-liquid interfaces of stirred vessels under conditions where the gas-phase mass transfer resistances were negligible, absorption proceeded in the region of pseudo m-th order reaction and its rate was not influenced by k_L . It is clearly shown in this figure that the absorption rate of NO in the aqueous solution of Fe(II)-EDTA is markedly high in spite of quite low

concentration of Fe(II)-EDTA.

Conclusion

The equilibrium constant and the reaction rate constant of the complexing reaction of NO with Fe(II)-EDTA were determined. These values were very large, of the order of 10⁶ l/g-mol and 10⁸ l/g-mol·sec, respectively.

A plausible mechanism of the absorption of NO into mixed solution of Fe(II)-EDTA and Na₂SO₃, which can explain the experimentally observed timecourse of η , was presented. Based on the absorption spectra of the mixed solutions, the presence of the interaction between Fe(II)-EDTA and SO₃²⁻ was confirmed. This interaction was considered to play an important role in this absorption system.

The absorption rate of NO in Fe(II)-EDTA solution was found to be markedly high compared with those in other liquid absorbents in spite of very low concentration of Fe(II)-EDTA.

Nomenclature

A	= concentration of NO in liquid	[g-mol/cm ³]
В	= concentration of Fe(II)-EDTA	A [g-mol/cm ³]
С	= concentration of Fe(II) (EDT	A) (NO)
		[g-mol/cm ³]
D	= diffusivity	[cm ² /sec]
G_M	= total molar flow rate of gas	[g-mol/sec]
H	 Henry constant 	[atm·cm³/g-mol]
Ι	= ionic strength	[g-ion/ <i>l</i>]
Κ	= equilibrium constant of reacti	on (a) [cm ³ /g-mol]
k	= reaction rate constant of reac	tion (a)
		[cm³/g·mol·sec]
k'	= reaction rate constant of reac	tion (c)
		[cm ³ /g-mol·sec]
k_{G}	= gas-phase mass transfer coefficient	cient
	[8	-mol/cm ² · sec · atm]
k_L	= liquid-phase mass transfer co	oefficient [cm/sec]
k _s	= salting-out parameter	[<i>l</i> /g-ion]
L	= liquid volume in absorber	[cm ³]
N_A	= absorption rate of NO	[g-mol/cm ² · sec]
n_G	= rotational speed of gas-phase	impeller [rpm]
n_L	= rotational speed of liquid-pha	se impeller [rpm]
Р	= total pressure	[atm]
q_J	$= J_0/A_i (J=B, C)$	[—]
R_A	= absorption rate of NO	[g-mol/sec]
r_J	$= D_J/D_A (J=B, C)$	[—]
S	= total gas-liquid interfacial area	a [cm ²]
t	= time	[sec]
x_a, x_c, x_g	= contributions of anion, cation	and solute
	gas to k_s , respectively	[<i>l</i> /g-ion]
у	= concentration of NO in gas pl	hase
	[mo	le fraction or ppm]
y_{N_2O}	= concentration N ₂ O in gas pha	ise
	[mo	le fraction or ppm]
α	= Bunsen absorption coefficient	
	[N n	nl/ml solution]
β	= reaction factor	· · · · · ·

β = reaction factor for infinitely fast irreversible reaction [--]

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$$\beta_{\infty} = \text{reaction factor for infinitely fast}$$

$$\text{reversible reaction} \qquad [--]$$

$$\gamma = \sqrt{k[\text{Fe(II)} - \text{EDTA}]D_A/k_L} \text{ or}$$

$$\sqrt{k'[\text{Fe(II)} - \text{EDTA}]D_A/k_L} \qquad [--]$$

$$\gamma = \text{absorption efficiency (fraction of NO removed)} \qquad [--]$$

$$\xi = -\frac{\beta_{\infty}}{2} - \frac{\beta_{-1}}{2} - \frac{\beta_{-1$$

$$\mu = \text{viscosity} \qquad [poise] \\ \rho = \text{density} \qquad [g/cm^3]$$

<Subscripts>

× ··· · · ·	1 1	
A	=	NO
В		Fe(II)-EDTA
С	_	Fe(II)(EDTA)(NO)
e	=	equilibrium
f	—	feed
G	—	gas
i	-	gas-liquid interface
ini		initial
L	=	liquid
w	_	water
0		bulk liquid

Literature Cited

- 1) Bird, R. B., W. E. Stewart and E. N. Lightfoot: "Transport Phenomena", p. 510, John Wiley, New York (1960).
- Calderbank, P. H. and Moo Young, M. B.: Chem. Eng. Sci., 16, 39 (1961).
- 3) Danckwerts, P. V.: Trans. Faraday Soc., 46, 300 (1950).
- 4) Hasui, H. and H. Omichi: Nenryo Kyokaishi, 55, 588 (1976).
- 5) Hatta, S.: Kogyo Kagaku Zasshi, 35, 1389 (1932).

- 6) Hikita, H. and H. Ishikawa: Kagaku Kogaku Ronbunshu, 1, 45 (1975).
- 7) Hikita, H., S. Asai, H. Ishikawa, T. Okamoto, S. Sakamoto and M. Kitagawa: Preprint of the 42nd Annual Meeting of The Soc. of Chem. Engrs., Japan, Hiroshima, p. 457 (1977).
- Hikita, H., S. Asai, H. Ishikawa and S. Hirano: J. Chem. Eng. Japan, 10, 120 (1977).
- 9) "Kagaku Benran Kisohen", p. 770, Maruzen, Tokyo (1974).
- 10) Olander, D. R.: AIChE J., 6, 233 (1960).
- 11) Onda, K., E. Sada, T. Kobayashi, S. Kito and K. Ito: J. Chem. Eng. Japan, 3, 18 (1970).
- 12) Ringbom, A.: "Complexation in Analytical Chemistry", John Wiley, New York (1963).
- 13) Sawai, K. and T. Gorai: Ryusan to Kogyo, 187 (1976).
- 14) Tanaka, T., M. Koizumi and Y. Ishihara: Denryoku Chuokenkyusho Gijutsudaiichikenkyusho Kenkyuhokokusho 275017 (1976).
- 15) Teramoto, M., M. Ikeda and H. Teranishi: Kagaku Kogaku Ronbunshu, 2, 86 (1976).
- 16) Teramoto, M., M. Ikeda and H. Teranishi: *ibid.*, 2, 637 (1976).
- 17) Teramoto, M., T. Kitagawa and H. Teranishi: *ibid.*, 3, 99 (1977).
- 18) Teramoto, M., M. Nagamochi, S. Hiramine, N. Fujii and H. Teranishi: *ibid.*, 3, 34 (1977).
- van Krevelen, D. W. and P. J. Hoftijzer: Chim. Ind. XXI^{me} Congress Int. Chim. Ind., p. 168 (1948).
- 20) Wilke, C. R. and P. Chang: AIChE J., 1, 264 (1955).
- 21) Yano, T., T. Suetaka and T. Umehara: Nippon Kagaku Kaishi, No. 6, 1226 (1973).