

Kinetics of Absorption of Hydrogen Sulfide into Aqueous Ferric Sulfate Solutions

Absorption experiments of H_2S into aqueous $\text{Fe}_2(\text{SO}_4)_3$ solutions, concerned with a unique process capable of regenerating by biological oxidation the reactant from the products, were performed at 20–40°C in an agitated vessel with a flat interface. The absorbents were aqueous solutions of $\text{Fe}_2(\text{SO}_4)_3$ with and without the addition of K_2SO_4 , KOH or H_2SO_4 of various concentrations, which was added to change the ionic strength or pH of the solutions. The experimental results revealed that the species which reacts with H_2S is FeOH^{2+} . While comparing the observed absorption rates with the theoretical predictions, it was found that the reaction is irreversible and first order with respect to H_2S and FeOH^{2+} . The reaction rate constants were measured and correlated.

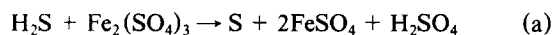
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Introduction

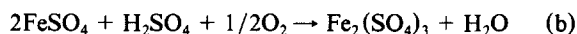
Removal of H_2S from gas streams is a familiar industrial requirement, whose economical importance will grow with the increasing utilization of fuels with a higher sulfur content. Among the many available removal processes of H_2S (Astarita et al., 1983; Kohl and Riesenfeld, 1985), conversion to elemental sulfur is advantageous because it can be used for treatment in an environmentally permissible procedure and applied even to the treatment of gases with relatively low concentrations of H_2S in the presence of CO_2 in substantial concentration, unless the amount of sulfur to be removed from the gas streams exceeds about 10 ton/d. The established procedures, however, often have some drawbacks, such as deactivation and loss of absorbent or catalyst due to poisoning or side reaction, unfavorable selectivity, corrosiveness, toxicity, and the need to operate at a high pressure or high temperature.

Recently Dowa Mining Co. in Japan developed a unique new process of H_2S removal (Imaizumi, 1986). In this process, aqueous $\text{Fe}_2(\text{SO}_4)_3$ solution is used as an absorbent. H_2S is oxidized to elemental sulfur, and $\text{Fe}_2(\text{SO}_4)_3$ is reduced to FeSO_4 . The reaction is:

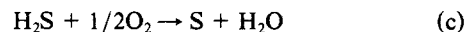


The sulfur formed is separated with a filter, and the reactant $\text{Fe}_2(\text{SO}_4)_3$ is regenerated from the products FeSO_4 and H_2SO_4

by biological oxidation using the iron-oxidizing bacteria, *Thiobacillus ferrooxidans*:



Then, the overall reaction of reactions a and b is:



Therefore, any drawbacks mentioned above are not encountered in this environmentally favorable closed system.

In this paper, the kinetics and mechanism of reaction were investigated by absorption experiments of H_2S into aqueous $\text{Fe}_2(\text{SO}_4)_3$ solutions with various values of ionic strength and pH.

Experimental Method

An agitated vessel with a flat interface, similar to that used in a previous work (Hikita et al., 1975), was used as an absorber. The vessel made of glass was 13.2-cm-id and 23.0-cm-high. Five equally spaced vertical baffles, each one tenth of the vessel diameter in width, were attached to the internal wall of the vessel. The liquid level was kept equal to the vessel diameter. The vessel was jacketed to maintain a constant temperature. The liquid stirrer was a paddle agitator with 5.3-cm-dia. flat blades placed into the center of the liquid and driven at constant speeds of 1.67 s^{-1} . The gas stirrer was a turbine agitator with six 8.5-cm-dia. flat blades placed in the center of the gas phase and driven at 8.33 s^{-1} .

The gas phase was mixtures of H_2S and N_2 saturated with water vapor at the experimental temperature. The mole fraction

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of H_2S in the influent gas was varied from 0.019–0.0019. The absorbents were aqueous solutions of $\text{Fe}_2(\text{SO}_4)_3$ of concentrations from 0.01–1.0 kmol/ m^3 ; and K_2SO_4 , KOH or H_2SO_4 of various concentrations was added to the absorbents to change the ionic strength or pH of the solutions. The 9K culture (Silverman and Lundgren, 1959), to which $\text{Fe}_2(\text{SO}_4)_3$ and KOH were added to give the $\text{Fe}_2(\text{SO}_4)_3$ concentrations of 0.0176 or 0.0620 kmol/ m^3 and $\text{pH} = 2$, was also used as an absorbent, allowing for the future biological application. The liquid compositions used in the analysis are shown in Table 1. All the experiments were carried out in batch operation with respect to the liquid. A mixed gas of H_2S and N_2 (1.92%), and pure N_2 were taken from individual pressure cylinders, metered by gas flowmeters, mixed, and saturated with water vapor. After that, the resulting gas mixture was passed into an agitated vessel and then vented into the exhaust line through a gas bubbler holding aqueous NaOH solution. After a fixed volume of the absorbent was introduced into the agitated vessel from the bottom, the gas mixture was fed into the vessel, the stirring of the gas phase was started, and then the gas space in the vessel was completely replaced with influent gas. Subsequently, the stirring of the liquid was started. Liquid samples were taken from the bottom with a sampling pipette at regular intervals. The absorption rates of H_2S were calculated from the graphical differentiation of the curve representing variation in the concentration of FeSO_4 formed as time elapsed. The experimental temperatures were 20, 30 and 40°C.

In this reaction system, sulfur formed during the chemical absorption processes deposits in the liquid, and part of the gas-liquid interface is covered by the sulfur film. Since this interferes with the liquid flow near the interface, it is impossible to predict the liquid-phase mass transfer coefficients from a previous empirical correlation (Hikita et al., 1975). Therefore, a very small amount of benzene was previously dissolved as a tracer in the absorbents, and the physical desorption rates were measured simultaneously with the chemical absorption rates of H_2S . The liquid-phase mass transfer coefficients of H_2S were

estimated from the observed coefficients for benzene by correcting for diffusivity.

The Fe^{2+} in the solution was determined by oxidation titration with standard $\text{K}_2\text{Cr}_2\text{O}_7$ solution in the presence of excess HCl , using diphenylamine in concentrated H_2SO_4 solution as an indicator. When the color of the coexisting Fe^{3+} made it difficult to confirm the end point of titration, aqueous phosphoric acid solution was added to eliminate the color. The concentration of Fe^{3+} in the solution was analyzed in the same manner as that of Fe^{2+} , after the conventional reduction of Fe^{3+} to Fe^{2+} by SnCl_2 . For analysis of the benzene in the solution, the liquid sample was treated with KMnO_4 and then KOH to eliminate interference from the coexisting species. Then, benzene was extracted into isooctane and measured by a spectrophotometer.

Physical Properties

When the dissolved H_2S undergoes an irreversible pseudofirst-order reaction, the concentration of the reactant is constant through the liquid phase at the values in the bulk of the liquid. In addition, when the experimental data are analyzed using the observed initial absorption rates of H_2S , the contribution of the concentrations of the products to the relevant physical properties may be regarded as negligible. Therefore, the physical solubility and the liquid-phase diffusivity of H_2S in the solution of the same composition as those of the initial bulk liquid should be used for the theoretical prediction of the absorption rates or reaction factor of H_2S .

The Henry constant H_i of H_2S in the absorbent was calculated from the correlation of Danckwerts and Gillham (1966) for mixed electrolyte solutions:

$$\log \frac{H_i}{H_w} = - \sum_j k_{sj} I_j \quad (1)$$

The Henry constant H_w for the H_2S –water system was taken from the *Handbook of Chemistry* (1984). The salting-out parameter k_{sj} is expressed as the sum of the contributions due to

Table 1. Experimental Conditions and Physical Properties of Liquids Used

Run No.	$[\text{Fe}_2(\text{SO}_4)_3]$ (kmol/ m^3)	$[\text{K}_2\text{SO}_4]$ (kmol/ m^3)	$[\text{KOH}]$ (kmol/ m^3)	$[\text{H}_2\text{SO}_4]$ (kmol/ m^3)	y_A	T (K)	$A_i \times 10^3$ (kmol/ m^3)	$D_A \times 10^9$ (m^2/s)	$k_{LA}^* \times 10^5$ (m/s)
1	0.00997	0	0	0	0.0192	303	1.50	2.10	1.48
2	0.0171	0	0	0	0.0192	303	1.48	2.09	1.29
3	0.0338	0	0	0	0.0192	303	1.47	2.06	1.11
4	0.0698	0	0	0	0.0192	303	1.40	2.00	1.05
5	0.160	0	0	0	0.0192	303	1.38	1.85	1.04
6	0.0183	0.0833	0	0	0.0192	303	1.44	2.06	1.23
7	0.0180	0.199	0	0	0.0192	303	1.36	2.02	1.16
8	0.0177	0.319	0	0	0.0192	303	1.28	1.98	1.05
9*	0.0195	0	0.0206	0	0.0192	303	1.45	2.08	1.30
10*	0.0344	0	0.0618	0	0.0192	303	1.31	2.05	1.19
11*	0.0658	0	0.148	0	0.0192	303	1.18	1.98	1.09
12**	0.0178	0	0	0.137	0.0192	303	1.62	2.06	1.54
13†	0.0184	0	0.0372	0	0.0192	303	1.40	2.08	1.04
14‡	0.0176	0	0.0214	0	0.0192	303	1.45	2.09	1.38
15‡	0.0620	0	0.149	0	0.0192	303	1.22	1.99	1.23
16	0.0684	0	0	0	0.00189	303	0.161	2.01	1.25
17	0.0644	0	0	0	0.00494	303	0.410	2.01	1.18
18	0.0648	0	0	0	0.0192	293	1.91	1.54	1.20
19	0.0693	0	0	0	0.0192	313	1.16	2.53	1.49

*pH = 2.0; **pH = 0.98; †pH = 2.23; ‡9K culture adjusted to pH = 2.0

the positive and negative ions of the single electrolyte j present and the dissolved gas:

$$k_{sj} = i_{j+} + i_{j-} + i_g \quad (2)$$

The values of i for the relevant ions and gas were taken from the data of Danckwerts (1970) and Hikita et al. (1975). The physical solubility A_i was calculated using the partial pressure of H_2S in the effluent gas and the Henry constant H_i , since the gas phase was mixed almost perfectly.

The liquid-phase diffusivities D_A of H_2S and D_C of benzene were estimated from the following expression (Hikita et al., 1979):

$$\frac{D_j}{D_{jw}} = 1 - \sum_j \kappa_j C_{ej} \quad (J = A \text{ for } H_2S \text{ and } C \text{ for benzene}) \quad (3)$$

where

$$\kappa_j = 0.03 + 0.55\omega_j \quad (4)$$

ω_j is a coefficient defined by

$$\frac{\mu_w}{\mu_j} = 1 - \omega_j C_{ej} \quad (5)$$

and ω_j is obtainable from the data of viscosity μ_j for the solution of single electrolyte j .

The viscosities of the aqueous $Fe_2(SO_4)_3$ solutions were measured at 20, 30 and 40°C, and the values of ω_j were determined to be 1.53, 1.37 and 1.31 m³/kmol, respectively. The ω_j values of the aqueous solutions of KOH and K_2SO_4 were evaluated to be 0.0973 and 0.251 m³/kmol at 30°C, respectively, using the published data (Hitchcock and McIlhenny, 1935; *Handbook of Chemistry*, 1984). The diffusivity D_{Aw} of H_2S in water was predicted from the value of 1.64×10^{-9} m²/s at 20°C (Arnold, 1930), by correcting for temperature according to the Stokes-Einstein relation. The diffusivity D_{Cw} of benzene in water was evaluated analogously using the values of 1.02×10^{-9} m²/s at 20°C (Witherspoon and Bonoli, 1969).

The physical properties of the solutions for the 9K culture were approximated with the values in the absence of the culture because the concentrations of the relevant electrolytes were substantially low.

The gas-phase mass transfer coefficients of H_2S were predicted from the empirical correlation (Hikita et al., 1975).

The predicted values of the physical properties are listed in Table 1.

Results and Discussion

Liquid-phase mass transfer coefficient

The liquid-phase mass transfer coefficients k_{LC}^* of benzene were determined in the experiments of the desorption of benzene, which was coupled with the chemical absorption of H_2S and were evaluated by using the relation

$$\ln \frac{C_0 - C_i}{C - C_i} = k_{LC}^* S \int_0^t \frac{dt}{V} \quad (6)$$

assuming complete mixing of the bulk liquid. The interfacial concentration C_i of benzene was taken as zero, since the gas flow

rates were large enough. Thus, the mass transfer coefficients were obtained from the slope of the plot of $\ln C$ vs. the integral values in the righthand side of Eq. 6.

Figure 1 shows the ratio of the observed mass transfer coefficients k_{LC}^* of benzene to the predicted ones, k_{LC}^{*p} . The latter values were predicted from the previous correlation (Hikita et al., 1975):

$$Sh_{LC} = 0.322 Re_L^{0.7} Sc_{LC}^{1/3} \quad (7)$$

where

$$Sh_{LC} = \frac{k_{LC}^* l}{D_C}, \quad Re_L = \frac{d_L^2 N_L \rho_L}{\mu_L}, \quad Sc_{LC} = \frac{\mu_L}{\rho_L D_C} \quad (8)$$

The abscissa was taken conveniently as the ionic strength I of the solution. As will be shown later, the increase in the ionic strength reflects roughly the increase in the reactant concentration with some exceptions, including run 12 to which H_2SO_4 had been added. From the figure, it may be seen that the ratio of the mass transfer coefficients decreases with an increase in the ionic strength I , amounting to a maximum deviation of 30%. This may be attributed to the increased fraction of the interface being covered by sulfur film formed with an increase in the reactant concentration, finally attaining a given maximum coverage. The values of the liquid-phase mass transfer coefficients k_{LA}^* of H_2S were estimated from the observed values for the benzene by correcting for diffusivity according to the following equation:

$$k_{LA}^* = k_{LC}^* (D_A/D_C)^{2/3} \quad (9)$$

The evaluated values of k_{LA}^* are shown in Table 1.

Absorption rates of H_2S

Figure 2 presents the effect of the concentration of $Fe_2(SO_4)_3$ on the measured absorption rates N_A of H_2S with and without

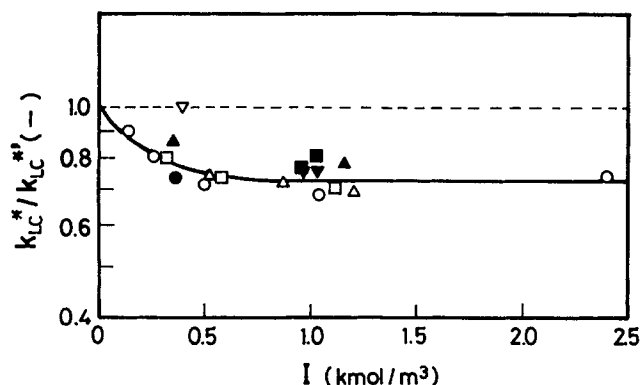


Figure 1. Effect of deposition of elemental sulfur on liquid-phase mass transfer coefficient of benzene.

$y_A = 0.02$, $T = 303$ K
 ○ $Fe_2(SO_4)_3$, △ $Fe_2(SO_4)_3 + K_2SO_4$
 □ $Fe_2(SO_4)_3 + KOH$ (pH = 2.0)
 ▽ $Fe_2(SO_4)_3 + H_2SO_4$ (pH = 0.98)
 ● $Fe_2(SO_4)_3 + KOH$ (pH = 2.23)
 ▲ $Fe_2(SO_4)_3 + KOH + 9K$ medium (pH = 2.0)
 ■ $Fe_2(SO_4)_3$, $y_A = 0.002$, 0.005
 ▼ $Fe_2(SO_4)_3$, $T = 293, 313$ K

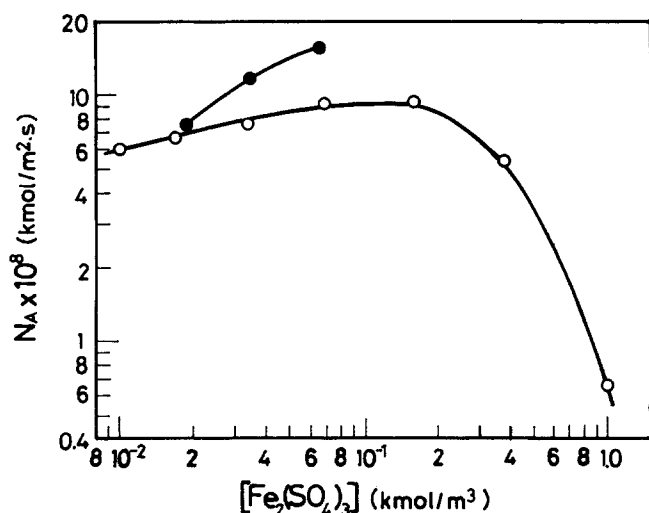


Figure 2. Effect of $\text{Fe}_2(\text{SO}_4)_3$ concentration on absorption rate of H_2S .

$y_A = 0.02$, $T = 303 \text{ K}$
 $\circ \text{Fe}_2(\text{SO}_4)_3$, $\bullet \text{Fe}_2(\text{SO}_4)_3 + \text{KOH}$ ($\text{pH} = 2.0$)

adjusting the pH of the absorbent to 2 by the addition of KOH. It may be noted that the absorption rates N_A without adjustment of the pH increase with the $\text{Fe}_2(\text{SO}_4)_3$ concentration in the region of relatively low concentrations, while N_A decreases remarkably in the region of higher concentrations. On the other hand, the observed absorption rates for the absorbents adjusted to $\text{pH} = 2$ increase substantially compared with those with no adjustment of pH; and the difference between the absorption rates with and without pH adjustment increases with the $\text{Fe}_2(\text{SO}_4)_3$ concentration. Figure 3 shows the dependency of the absorption rates on pH, which was adjusted by the addition of H_2SO_4 or KOH to the absorbent. As can be seen, the absorption rates increase significantly with pH.

If one regards $\text{Fe}_2(\text{SO}_4)_3$ as a chemical species reacting directly with H_2S , the effects of the $\text{Fe}_2(\text{SO}_4)_3$ concentration and pH on the absorption rates, which were referred to in Figures 2 and 3, cannot be explained reasonably only by allowing for the

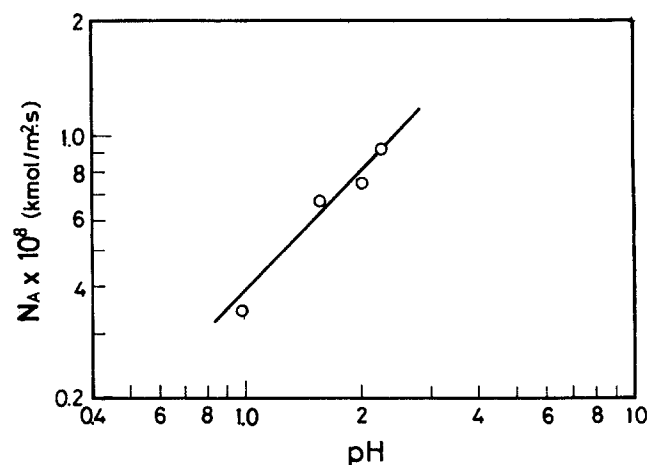


Figure 3. Effect of pH on absorption rate of H_2S .

$y_A = 0.02$, $T = 303 \text{ K}$, $[\text{Fe}_2(\text{SO}_4)_3] = 0.018 \text{ kmol/m}^3$

weak variation in the values of the relevant physical properties with the composition of the absorbent.

Chemical species reacting with H_2S

In what follows, the equilibria of the reactions in the solution are considered to identify the species likely to react with H_2S . The relevant reactions are shown in Table 2.

The concentrations of each species were evaluated from the chemical equilibrium constants of the reactions listed in Table 2, mass balance for iron, sulfate and potassium, and the measured values of pH. The chemical equilibrium constants were taken as a function of the ionic strength of the solution and temperature from the literature (Smith and Martell, 1976). The concentrations of some species at 25°C , which exist in the substantial concentration, were plotted as a function of the $\text{Fe}_2(\text{SO}_4)_3$ concentration and pH of the solution in Figures 4 and 5, respectively.

The calculations were limited to ionic strengths of less than 3.0 kmol/m^3 , owing to the unknown ionic strength dependency of the relevant chemical equilibrium constants for the larger values of the ionic strength. The concentration of FeOH^{2+} increases with the concentration of $\text{Fe}_2(\text{SO}_4)_3$ in the region of low concentrations of $\text{Fe}_2(\text{SO}_4)_3$, but the rate of increase in the FeOH^{2+} concentration decreases with an increase in the $\text{Fe}_2(\text{SO}_4)_3$ concentration. The addition of KOH to the $\text{Fe}_2(\text{SO}_4)_3$ solutions or an increase of pH greatly causes enhancement of the FeOH^{2+} concentration. These behaviors are consistent with that observed, as described above, concerning the absorption rates of H_2S . The tendency of all other species existing in lower concentration, not shown in Figures 4 and 5, were found not to be in line with that expected from the observed absorption rates. Thus, the chemical species reacting with H_2S is likely to be FeOH^{2+} .

Observed reaction factor vs. theoretical prediction

By choosing FeOH^{2+} to be the actual reactant, the experimental data of the absorption rates were compared with the theoretical predictions. The gas-phase resistance to mass transfer was always found to be negligible (below 1.4%) compared with the total resistance under the experimental conditions studied. Furthermore, the relation $[\text{Fe}_2(\text{SO}_4)_3] \gg A_i$ was satisfied to analyze the experimental data based on the theory of absorption with an irreversible pseudo- m th-order reaction. When

Table 2. Chemical Equilibria in Aqueous $\text{Fe}_2(\text{SO}_4)_3$ Solutions Containing KOH, H_2SO_4 or K_2SO_4

$\text{Fe}^{3+} + \text{OH}^- = \text{FeOH}^{2+}$	(d)
$\text{Fe}^{3+} + 2\text{OH}^- = \text{Fe}(\text{OH})_2^+$	(e)
$\text{Fe}^{3+} + 3\text{OH}^- = \text{Fe}(\text{OH})_3$	(f)
$\text{Fe}^{3+} + 4\text{OH}^- = \text{Fe}(\text{OH})_4^-$	(g)
$2\text{Fe}^{3+} + 2\text{OH}^- = \text{Fe}_2(\text{OH})_2^{4+}$	(h)
$3\text{Fe}^{3+} + 4\text{OH}^- = \text{Fe}_3(\text{OH})_4^{5+}$	(i)
$\text{Fe}^{3+} + \text{SO}_4^{2-} = \text{FeSO}_4^+$	(j)
$\text{Fe}^{3+} + 2\text{SO}_4^{2-} = \text{Fe}(\text{SO}_4)_2^-$	(k)
$\text{H}^+ + \text{SO}_4^{2-} = \text{HSO}_4^-$	(l)
$\text{K}^+ + \text{SO}_4^{2-} = \text{KSO}_4^-$	(m)
$\text{H}^+ + \text{OH}^- = \text{H}_2\text{O}$	(n)

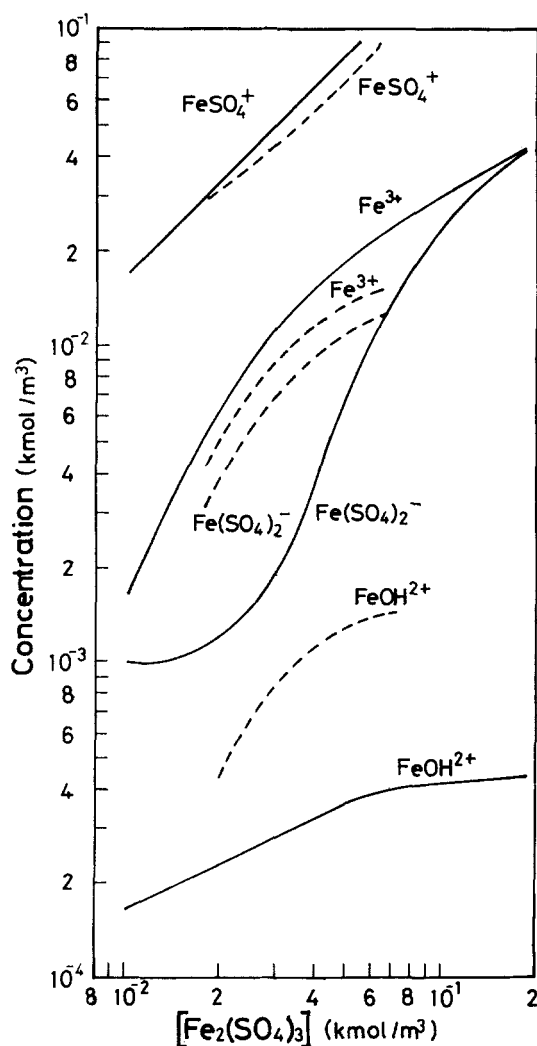


Figure 4. Variation in concentrations of relevant ions with $\text{Fe}_2(\text{SO}_4)_3$ concentration.

$T = 298 \text{ K}$
 — no adjustment of pH; ---- pH = 2.0

this reaction is fast, the absorption rate is given by (Hikita and Asai, 1963)

$$N_A = \sqrt{\frac{2}{m+1}} k D_A A_i^{m+1} B_o^n \quad (10)$$

Figure 6 presents the effect of the gas-phase mole fraction y_A of H_2S on the absorption rates. It is found that the absorption rates are proportional to the mole fraction, i.e., the physical solubility A_i . This suggests that the reaction order m with respect to H_2S may be taken as unity. Based on Eq. 10, the experimental data for runs 1–5 are illustrated in Figure 7 as a plot of $N_A/A_i\sqrt{D_A}$ vs. the bulk concentration B_o of the reactant FeOH^{2+} . It may be seen that the slope of the representative line of experimental data is $1/2$. This indicates $n = 1$ (first order with respect to FeOH^{2+}), if the reaction rate constant k is substantially independent of the ionic strength of the solution. Figure 8 shows the dependency of the reaction rate constant k on the ionic

strength, which was varied by the addition of K_2SO_4 to aqueous $\text{Fe}_2(\text{SO}_4)_3$ solutions. The data were plotted on the basis of Eq. 10 with $m = n = 1$. This figure shows that the values of the ordinate corresponding to \sqrt{k} are constant, and therefore the reaction rate constant is independent of the ionic strength. Thus, this reaction may be regarded as second order (first order in H_2S and FeOH^{2+}). The experimental data for various liquid compositions at 30°C were compared with the theoretical predictions in the form of the reaction factor β . However, the comparison was confined to the data corresponding to the ionic strength $I < 3 \text{ kmol/m}^3$, in which the concentration of FeOH^{2+} could be evaluated from the known values of the chemical equilibrium constants. The reaction factors β were evaluated from the measured absorption rates N_A using the following expression:

$$N_A = \beta k_L^* A_i \quad (11)$$

The solution of the reaction factor β for an irreversible pseudofirst-order reaction on the basis of the L  v  que model, which

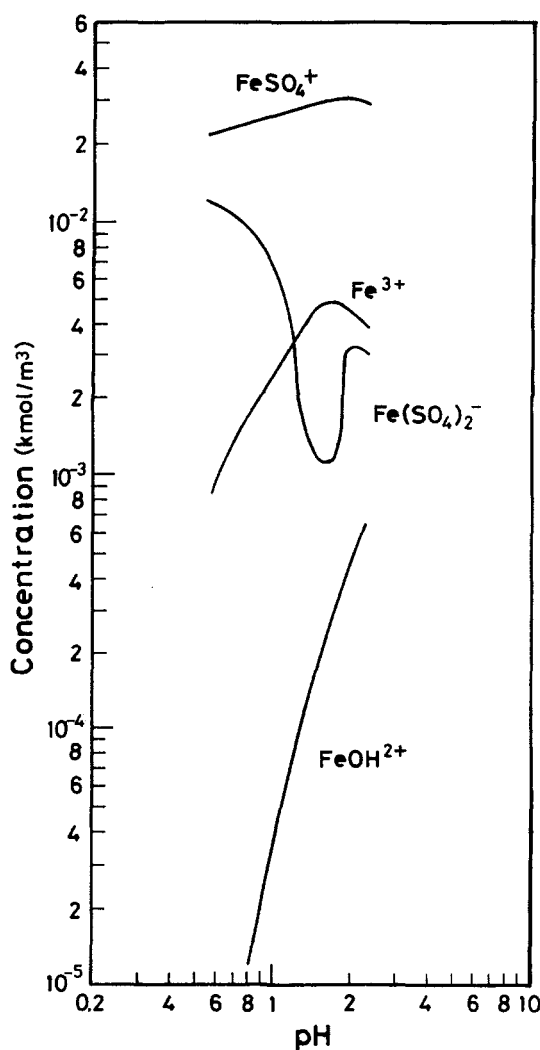


Figure 5. Variation in concentrations of relevant ions with pH.

$T = 298 \text{ K}$, $[\text{Fe}_2(\text{SO}_4)_3] = 0.018 \text{ kmol/m}^3$

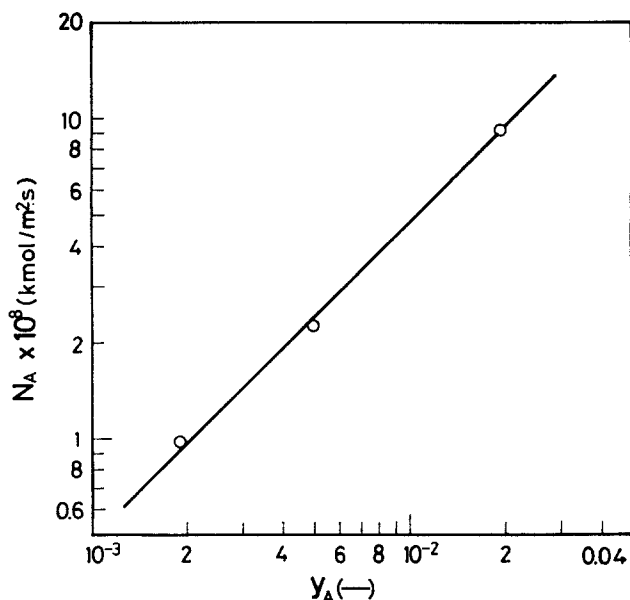


Figure 6. Effect of gas-phase concentration on absorption rate of H_2S .

$[Fe_2(SO_4)_3] = 0.068 \text{ kmol/m}^3$, $T = 303 \text{ K}$

is applicable to mass transfer in the present agitated vessel with a flat interface, can be closely approximated by (Hikita et al., 1973, 1975)

$$\beta = \gamma + \frac{0.474715}{\gamma} \quad (\gamma \geq 2.4) \quad (12)$$

$$\beta = 1 + \sum_p b_p \gamma^{2p} \quad (\gamma \leq 2.4) \quad (13)$$

where the coefficients b_p are the numerical constants as given by Hikita et al. (1973, 1975), and γ is the dimensionless parameter defined by

$$\gamma = \frac{\sqrt{k B_0 D_A}}{k_{LA}^*} \quad (14)$$

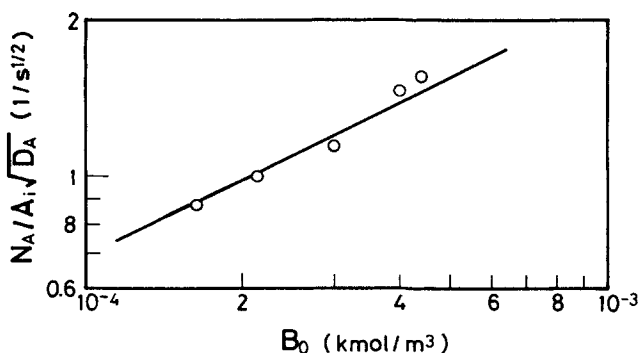


Figure 7. Effect of $FeOH^{2+}$ concentration on absorption rate of H_2S .

$y_A = 0.02$, $T = 303 \text{ K}$

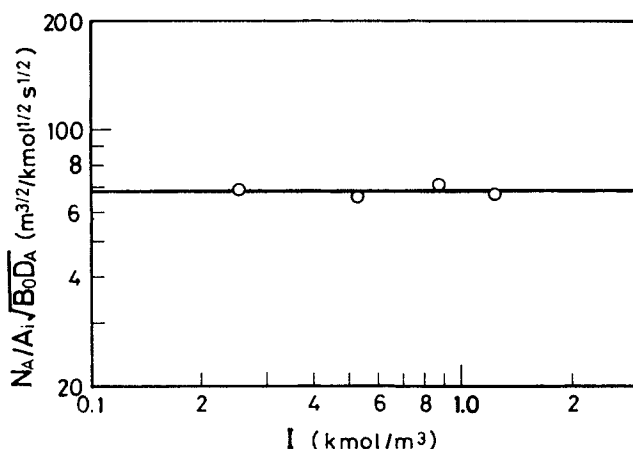


Figure 8. Effect of ionic strength on absorption rate of H_2S .

$y_A = 0.02$, $[Fe_2(SO_4)_3] = 0.018 \text{ kmol/m}^3$, $T = 303 \text{ K}$

The observed reaction factors β are compared with Eqs. 12 and 13 in Figure 9. The reaction rate constant k was evaluated to be $4,100 \text{ m}^3/\text{kmol} \cdot \text{s}$ at 30°C , by best fitting the experimental data to the theoretical line. As seen from this figure, the agreement between the theoretical predictions and the measured reaction factors for the various compositions of gas and liquid is remarkable. Thus, it may be inferred that the species which reacts with H_2S is $FeOH^{2+}$ and that the reaction is irreversible and first order with respect to both H_2S and $FeOH^{2+}$.

Correlation of reaction rate constant and reaction mechanism

The data measured at 20 and 40°C were also analyzed in the same manner. Figure 10 shows the Arrhenius plot of the

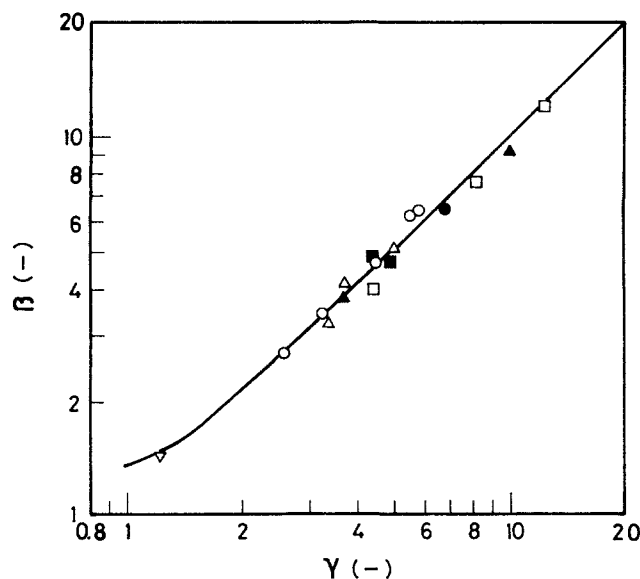


Figure 9. Comparison of observed reaction factors with theoretical line, Eqs. 12 and 13.

$k = 4,100 \text{ m}^3/\text{kmol} \cdot \text{s}$, $T = 303 \text{ K}$

Keys same as in Figure 1.

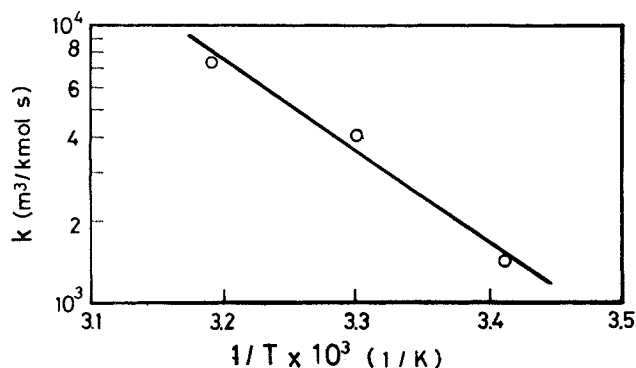


Figure 10. Arrhenius plot of observed reaction rate constant.

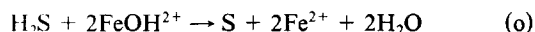
$$y_A = 0.02, [\text{Fe}_2(\text{SO}_4)_3] = 0.068 \text{ kmol/m}^3$$

reaction rate constant k . The plot is found to be linear, and the reaction rate constants can be correlated by

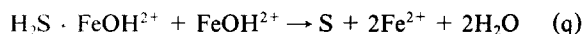
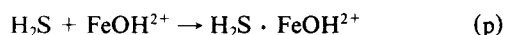
$$\ln k = 32.98 - \frac{7,520}{T} \quad (15)$$

corresponding to the activation energy of 62.5 kJ/mol · K.

Since FeOH^{2+} , which is formed by the hydrolysis reaction of $\text{Fe}_2(\text{SO}_4)_3$, is a chemical species reacting with H_2S , it may be considered that the overall reaction is:



which may consist of the following two steps:



The first step, reaction p, is an irreversible second-order reaction. Thus, if one assumes this reaction to be the rate-controlling step of the overall reaction (Eq. o), this reaction mechanism is in line with the present experimental results.

Conclusions

Absorption of H_2S into aqueous $\text{Fe}_2(\text{SO}_4)_3$ solution is accompanied by the reaction between H_2S and FeOH^{2+} , which is first order with respect to the individual species. The relevant reaction rate constants were correlated by Eq. 15 independent of the ionic strength, and the overall reaction was supposed to consist of reactions p and q.

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Notation

- A = concentration of dissolved gas, H_2S , kmol/ m^3
- B_0 = bulk concentration of reactant, FeOH^{2+} , kmol/ m^3
- b_p = coefficient in Eq. 13
- C = concentration of tracer, benzene, kmol/ m^3

- C_{ej} = concentration of electrolyte j , kmol/ m^3
- D = liquid-phase diffusivity, m^2/s
- d = diameter of stirrer, m
- H = Henry's law constant, kmol/ $\text{m}^3 \cdot \text{Pa}$
- I = ionic strength, kmol/ m^3
- i = contribution to salting-out parameter, m^3/kmol
- k_L^* = liquid-phase mass transfer coefficient, m/s
- k_s = salting-out parameter, m^3/kmol
- k = reaction rate constant, $(\text{m}^3/\text{kmol})^{m+n-1}/\text{s}$
- l = diameter of agitated vessel, m
- m = reaction order with respect to dissolved gas, H_2S
- N = stirring speed, 1/s
- N_A = absorption rate of H_2S , kmol/ $\text{m}^2 \cdot \text{s}$
- n = reaction order with respect to reactant, FeOH^{2+}
- Re = Reynolds number
- S = gas-liquid interfacial area, m^2
- Sc = Schmidt number
- Sh = Sherwood number
- T = thermodynamic temperature, K
- t = time, s
- V = volume of solution, m^3
- y = mole fraction in gas phase

Greek letters

- β = reaction factor
- γ = dimensionless number defined by Eq. 14
- κ = coefficient given by Eq. 4, m^3/kmol
- μ = viscosity, Pa · s
- ρ = density, kg/ m^3
- ω_j = coefficient in Eq. 5, m^3/kmol

Subscripts

- A = dissolved gas, H_2S
- C = tracer, benzene
- i = gas-liquid interface
- j = electrolyte j
- j^+ = positive ion of electrolyte j
- j^- = negative ion of electrolyte j
- L = liquid phase
- O = initial
- w = water

Superscript

- ' = calculated value

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