

Reactive Absorption of Hydrogen Sulfide in Aqueous Ferric Sulfate Solution

Z. Gholami, M. Torabi Angaji, F. Gholami, and S. A. Razavi Alavi

Abstract—Many commercial processes are available for the removal of H₂S from gaseous streams. The desulfurization of gas streams using aqueous ferric sulfate solution as washing liquor is studied. Apart from sulfur, only H₂O is generated in the process, and consequently, no waste treatment facilities are required. A distinct advantage of the process is that the reaction of H₂S with is so rapid and complete that there remains no danger of discharging toxic waste gas. In this study, the reactive absorption of hydrogen sulfide into aqueous ferric sulfate solution has been studied and design calculations for equipments have been done and effective operation parameters on this process considered. Results show that high temperature and low pressure are suitable for absorption reaction. Variation of hydrogen sulfide concentration and Fe³⁺ concentration with time in absorption reaction shown that the reaction of ferric sulfate and hydrogen sulfide is first order with respect to the both reactant. At low Fe₂(SO₄)₃ concentration the absorption rate of H₂S increase with increasing the Fe₂(SO₄)₃ concentration. At higher concentration a decrease in the absorption rate was found. At higher concentration of Fe₂(SO₄)₃, the ionic strength and viscosity of solution increase remarkably resulting in a decrease of solubility, diffusivity and hence absorption rate.

Keywords—Absorption, Fe₂(SO₄)₃, H₂S, Reactive Absorption.

I. INTRODUCTION

MANY natural and industrial gases contain H₂S. The presence of H₂S usually prohibits the direct use of these gases because of its toxic properties, the formation of SO₂ upon combustion (acid rain), and the problems it (usually) gives in downstream processing. This means that it is often necessary to remove H₂S from gas stream prior to use. Many processes have been developed to remove H₂S from gas streams [1]. Most of the process use gas-liquid contactors in which the H₂S is absorbed into a complexing reagent to give either another dissolved sulfide containing component and problems are the degradation of the solvent [2]. Iron (Fe) is an

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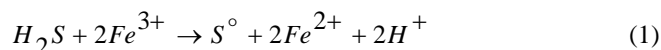
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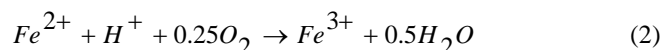
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excellent oxidizing agent to convert H₂S to elemental sulfur (S). During the 1960s, developmental work initiated in England focused on increasing the solubility of elemental Fe³⁺ in aqueous solutions [3].

In this study, the reactive absorption of hydrogen sulfide into aqueous ferric sulfate has been studied. In this process an aqueous Fe₂(SO₄)₃ solution is used as an absorbent. H₂S is absorbed and oxidized to elemental sulfur. At the same time, Fe³⁺ is reduced to Fe²⁺ according to



Elemental sulfur is removed from the solution by separator and the reactant Fe³⁺ is regenerated from Fe²⁺ by oxidation in reactor to the following reaction:



Depending on the gas flow rate and the efficiency required several types of absorbers are suitable, such as jet scrubbers, bubble-cap towers or packed towers. The sulfur separator can include filter press, settler and sulfur melters, Depending on quality of elemental sulfur required. Apart from sulfur, only H₂O is generated in the process, and consequently, no waste treatment facilities are required [2].

A distinct advantage of this process is that the reaction of H₂S with Fe₂(SO₄)₃, is so rapid and complete that there remains no danger of discharging toxic waste gas. The process is schematically depicted in Fig. 1.

II. PROCESS DESIGN CALCULATIONS

In this study assumed a packed tower used as absorber, continuous filter used as solid-liquid separator to separate sulfur from outlet liquid, and a back mixing reactor used as oxidizer to regenerate Fe²⁺ to Fe³⁺. According to these assumptions and for known gas flow rate (100000 kg/day sour gas, contain ≈ 4% v/v H₂S), design calculations have been done. Calculations have been done for a packed tower with 2 inches Raschig rings as packing and 0.1 molar Fe₂(SO₄)₃ concentration. Onda's method used to calculate k_G and k_L, gas and liquid side mass transfer coefficient.

$$\frac{a_w}{a} = 1 - \exp \left[-1.45 \left(\frac{\sigma_C}{\sigma_L} \right)^{0.75} \left(\frac{L_w^*}{a \cdot \mu_L} \right)^{0.1} \left(\frac{L_w^* a}{\rho_L \cdot g} \right)^{-0.05} \left(\frac{L_w^* a}{\rho_L \sigma_L a} \right)^{0.2} \right] \quad (3)$$

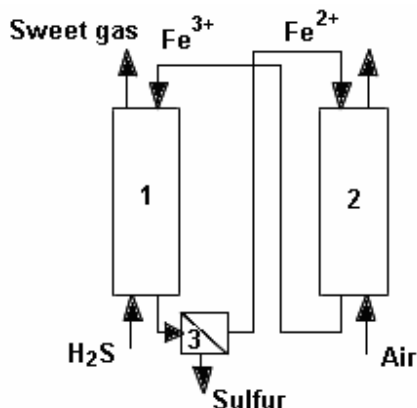


Fig. 1. Process scheme of process for H₂S removal, 1- Absorber, 2- Oxidizer, 3- Solid-liquid separator[2].

$$k_L \left(\frac{\rho_L}{\mu_L \cdot g} \right)^{1/3} = 0.0051 \left(\frac{L_w^*}{a_w \mu_L} \right)^{2/3} \left(\frac{\mu_L}{\rho_L \cdot D_L} \right)^{-1/2} (a.d.p)^{0.4} \quad (4)$$

$$\frac{k_G}{a} \cdot \frac{RT}{D_V} = 5.23 \left(\frac{V_w^*}{a \cdot \mu_v} \right)^{0.7} \left(\frac{\mu_v}{\rho_v D_v} \right)^{1/3} (a.d.p)^{-2.0} \quad (5)$$

The height of liquid and gas-film transfer units (H_G , H_L) and overall gas-phase transfer unit (H_{OG}) are obtained from (6) to (8).

$$H_G = \frac{G_m}{K_G \cdot a_w \cdot p} \quad (6)$$

$$H_L = \frac{L_m}{K_L \cdot a_w \cdot C_t} \quad (7)$$

$$H_{OG} = \frac{G_m}{K_G \cdot a \cdot P} \quad (8)$$

And after calculations, tower's height (Z) calculated. Subsequently, Diameter and height of absorption tower are 1.1 m and 3.8 m and pressure drop is 286 Pa/m packing [4]-[5]-[6].

In this process assumed that Oxidizer is a back mixing reactor. According to this assume, calculations have been done for oxidizer.

Fe³⁺ is regenerated from Fe²⁺ according to (2) and it is relatively slow reaction. The reaction is first order with respect to oxygen and is 0.536 with respect to Fe²⁺, as follow:

$$-r_{Fe^{2+}} = 4.39 \times 10^4 \exp \left(\frac{-23.3}{R.T} \right) \cdot C_{Fe^{2+}}^{0.536} \cdot C_{O_2} \quad (9)$$

Consequently, Oxidizer's height and diameter are 0.75 m and 1.5 m [7]-[8]-[9].

III. RESULTS AND DISCUSSION

A. Variations of Reactant Concentration

The absorption reaction of hydrogen sulfide by ferric sulfate solution appears to irreversibly and is first order with respect to both reactants [2]. Variations of concentration profile of the reactants calculated in recent studies. Absorption reaction kinetic is as follow:

$$r_{H_2S} = 16.815 C_{H_2S} \cdot C_{Fe^{3+}} \quad (10)$$

These profiles had shown in Fig. 2 and Fig. 3.

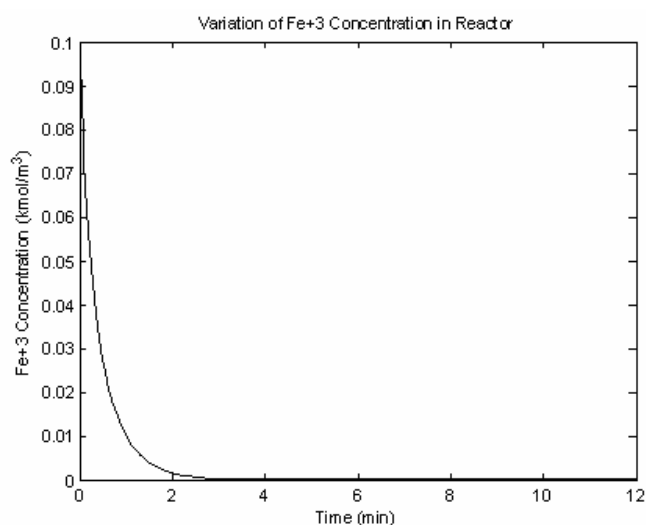


Fig. 2 Fe³⁺ Concentration profile

Recent studies have resulted at low Fe₂(SO₄)₃ concentrations (up to 0.3 M) the absorption rate of H₂S increase with total Fe₂(SO₄)₃ concentration. At higher Fe₂(SO₄)₃ concentrations a decrease in the absorption rate was found. At higher concentration of Fe₂(SO₄)₃ the ionic strength and viscosity of the solution increase remarkably resulting in a decrease of the solubility, diffusivity and liquid side mass transfer coefficient of H₂S and hence absorption rate, as shown in Fig. 4 [2].

B. Influence of Effective Parameters on the Process

It is important to have an exact control on effective parameters in reaction to have high efficiency and low cost

and energy consumption. Therefore, some effective parameters in this process are considered.

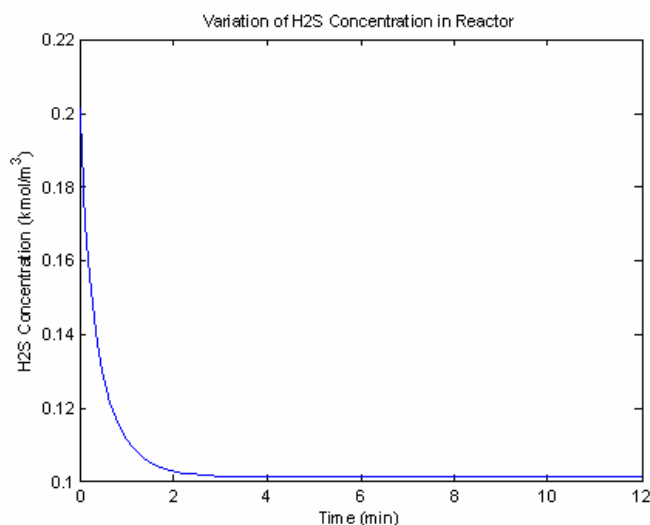


Fig. 3 H₂S Concentration profile

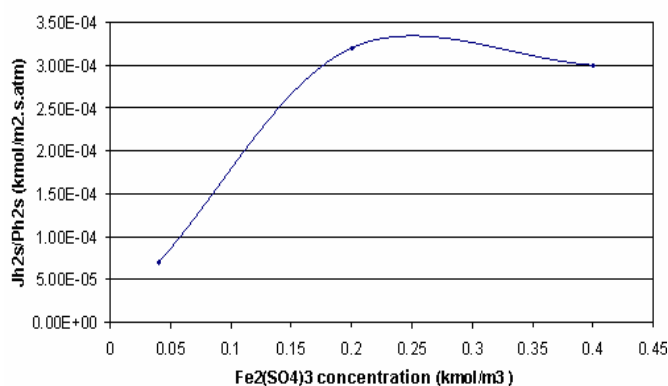


Fig. 4 Influence of Fe₂(SO₄)₃ concentration on the absorption rate of H₂S at 45°C [2]

IV. CONCLUSION

H₂S removal from gas streams using aqueous ferric solution as absorbent has been studied theoretically, calculations for absorber and oxidizer have been done, and their dimensions are obtained for a known sour gas flow rate.

The absorption reaction is first order with respect to both H₂S and ferric iron concentration. influence of temperature and pressure on H₂S absorption considered and shown that in low pressure and high temperature, the process should have higher efficiency. Optimum concentration of ferric sulfate should be used in the process due to increasing the ionic strength and viscosity of the solution at higher concentration of Fe₂(SO₄)₃, consequently, resulting in a decrease of the solubility, diffusivity and liquid side mass transfer coefficient of H₂S and hence absorption rate.

NOTATION

a Surface area of interface per unit volume of column, m²/m³
 σ Surface tension, J/m²

μ_L	Viscosity of liquid, Ns/m ²
μ_V	Viscosity of liquid, Ns/m ²
ρ_L	Density of liquid, kg/m ³
ρ_V	Density of gas, kg/m ³
D_L	Liquid phase diffusivity, m ² /s
D_V	vapour phase diffusivity, m ² /s
k_L	Liquid side mass transfer coefficient, m/s
k_G	Gas side mass transfer coefficient, kmol/m ² atm s
H_L	Height of transfer unit-liquid film, m
H_G	Height of transfer unit-gas film, m
H_{OG}	Height of transfer unit-overall (gas concentrations), m
K_L	Overall liquid-phase transfer coefficient, m/s
K_G	Overall gas-phase transfer coefficient, s/m
G_m	Molar rate of gas per unit cross-section, kmol/m ² s
L_m	Molar rate of liquid per unit cross-section, kmol/m ² s
L_W^*	Liquid flow rate (mass) per unit cross-section, kg/m ² s
V_W^*	Gas flow rate (mass) per unit cross-section, kg/m ² s
dp	Packing size, m
C_T	Total molar concentration, kmol/m ³
R	Universal gas constant, J/kmol K
Z	Height of packed column, m
g	Acceleration due to gravity, m/s ²
P	Total pressure, N/m ²
T	Absolute temperature, K

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