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Chemical Absorption of H_2S in Aqueous $Fe_2(SO_4)_3$ Solution

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Abstract— Many commercial processes are available for the removal of H_2S from gaseous streams. In this study chemical absorption of H_2S into aqueous $Fe_2(SO_4)_3$ solutions, was studied in a sieve trays column 70 cm in diameter, number of trays 8, the height of 450 cm. with assumed steady state and isothermal condition. The gas flow rate was $G = 75114.4763 \text{ cm}^3/\text{s}$, liquid flow rate was $10000 \text{ cm}^3/\text{s}$, the concentration of aqueous $Fe_2(SO_4)_3$ solution is 0.002 mol/cm^3 . Mass transfer was carried out theoretically by developing the mathematical model of mass transfer phenomena. Various operating parameters were tested to determine the effect of these parameters on the H_2S absorption efficiency.

It was that percentage of absorbed H_2S can be enhanced by increasing temperature, liquid flow rate, $Fe_2(SO_4)_3$ concentration, and decreasing sour gas flow rate. Gas side resistance actually is more reins than that solution; it means that the absorption process is dominated by gas side resistance than liquid side resistance.

Keywords: Sour gas purification, H_2S removal, $Fe_2(SO_4)_3$

I. INTRODUCTION

Hydrogen sulfide is being produced by many industrial activities such as petroleum refining, natural gas and petrochemical plants, viscose rayon manufacturing craft, pulp manufacturing, food processing, aerobic and anaerobic wastewater treatments and many other industries [1]. Sour gas contains hydrogen sulfide gas and ammonia. The existence of hydrogen sulfide in sour gas is very disturbing, besides it smells bad, on combustion it will evolve (SO_2) gas which pollutes environment, in global, it will cause acid rain. So that, it also needs to be eliminated from sour gas.

Therefore, it is necessary to purify the sour gas first before combustion. The research is executed at Khartoum Refinery Company (KRC). Because hydrogen sulfide in the (KRC) is controlled by combustion process and hydrogen sulfide (2.401t/h) is converted to sulfur dioxide, which is also an air pollutant, in combustion process, it is not the better method to remove hydrogen sulfide by combustion. To operate a successful refinery you need to have a clean production, clean technology and be environmentally friendly.

Sour gas is split into two streams to enter the burner of sour gas incinerator. One stream comes from sour water stripping (SWS) unit (1670 t/h), the other path coming from Resid Fluid Catalytic Cracking RFCC (594t/h) and Delayed Coking DCU (137t/h) units passes through sour gas knockout drum for liquid separation, then each stream enters the burner separately.

Sour gas emissions can cause damage to the environment due to the presence of the pollutant hydrogen sulphide, which is harmful to human beings and animals. At lower concentrations, this gas has an unpleasant odour; at higher concentrations, it can be life-threatening. The recommended industrial exposure limits are from 8 to 10 ppm for 8 hours a day per week [2].

However, direct combustion of sour gas without removal of H_2S leads to the formation of sulphur dioxide (SO_2), which is another toxic pollutant and a major contributor to acid rain in the atmosphere. Gas must be very clean to reach pipeline quality, and must be of the correct composition for the local distribution network such as Carbon dioxide, Water, hydrogen sulfide and particulates must be removed if present.

There are many kinds of process that stated in literature to removing H_2S gas [2]. These can be divided into two methods; those are chemical absorption and adsorption. Chemical absorption is an absorption which is followed by chemical reaction when the absorbed gas is reacted with the reactant in liquid phase, while adsorption is absorption on the surface of solid particle which is called adsorbent. In using H_2S gas, the adsorbent in which commonly used



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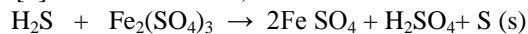
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is ZnO that reacts H₂S into ZnS. Some researchers have learnt about the removing process of H₂S gas in biogas flowing by changing it that it becomes more stable sulfur [2], [3] stated that gas desulphurization with Ferric Chelate is more stable than EDTA and HEDTA. Al stated that the H₂S changing process becomes sulfur is by reaction with zinc natrium ferric (Asai, 1990). While Hix and Lynn stated that the absorption process of H₂S gas into SO₂ solution in polyglycol ether (Horikawa, 2004) proposes removing process of gas H₂S from biogas with changing it to become sulfur (S) applies absorption process into soluble chelate iron from at EDTA[2].

In this study, the chemical absorption of hydrogen sulfide into aqueous ferric sulfate has been studied on a sieve trays column has not done yet. In this process an aqueous Fe₂(SO₄)₃ solution is used an absorbent. H₂S is absorbed and oxidized to elemental sulfur [4]. At the same time, Fe³⁺ is reduced to Fe²⁺ according to



Depending on the gas flow rate and the efficiency required several types of absorbers are suitable, such as plate towers or packed towers. The sulfur separator can include filter pres, settler and sulfur melters, Depending on quality of elemental sulfur required [5]. The theoretical study is important for column design and column performance analysis [5].

II. MATERIALS AND METHODS

This study is done in simulation to determine sieve trays column performance in absorption of H₂S gas into Fe₂(SO₄)₃ solution. Theoretical study is divided into parts. The first part developed transfer model between phases to estimate the value of enhancement factor (E) that yielding the ordinary differential equation system in second order and it is solved by the orthogonal collocation method. Estimation of the value of (E) required several data; there are reaction kinetics data, solubility data, and mass transfer data, after that the value of (E) is applied for sieve trays column model of chemical absorption of H₂S yielding of ordinary differential equation system of first order solved by using Runge Kutta method. Output of sieve trays column model is % Recovery H₂S, concentration distribution of A in gas phase, concentration distribution of B in liquid phase. Ferric sulfate solution is used for H₂S absorption the reaction rate can be expressed as [4]

$$r = k C_{\text{H}_2\text{S}} C_{\text{Fe}^{3+}} \quad (1)$$

By assuming that the kinetic constant varies with temperature according to an Arrhenius function, the activation energy can be calculated. Therefore the reaction rate constant values were obtained as a function of temperature [4]

$$\log k = 9.3855 - \frac{2636.6}{T} \quad (2)$$

Mathematical Model Expansion

Development of Mass transfer Model between Phases to Estimate the Enhancement Factor

The expansion of mass transfer model between Phases to estimate the value of Enhancement with reaction that is represented below by irreversible second order reaction.

Mass balance for component A(H₂S) (Film Model)

$$\frac{d^2 A}{dX^2} - MAB = 0 \quad (3)$$

Mass balance for component B (Fe₂(SO₄)₃)

$$\frac{d^2 B}{dX^2} - \frac{MAB}{S} = 0 \quad (4)$$

With orthogonal collocation method, the collocation equation made an A_i, B_i, explicit to be solved by successive approximation method

For component A

$$A_i = \frac{\sum_{j=1}^{N+2} D_{ij} A_j - D_{ii} A_i}{(M B_i - D_{ii})} \quad (5)$$

For component B

$$B_i = \frac{\sum_{j=1}^{N+2} D_{ij} B_j - D_{ii} B_i}{\frac{M A_i}{S} - D_{ii}} \quad (6)$$



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The Development of Sieve Trays Column Model

The develop meet of model is begun by forming mass balance for component A (H₂S) in gas phase and B Fe₂(SO₄)₃ in liquid phase, which following by the equations:

$$\frac{dY_A}{dz} = \frac{Ek_L a}{G_m} (C_{Ai} - C_A^0) \tag{7}$$

$$\frac{dC_B^0}{dz} = -2 \frac{Ek_L a}{U} (C_{Ai} - C_A^0) \tag{8}$$

The system of non linear differential equations was solved using orthogonal collocation method and collocated equations described as non linear algebraic equations was solved using orthogonal collocation method. Then, the isothermal enhancement factor can be predicted in equation (9), where the value $[dC_A/dx]_{x=0}$ of can be estimated using orthogonal collocation method. [6]

$$E = \frac{\bar{R}}{k_L C_{Ai}} = \frac{-D_A [dC_A/dx]_{x=0}}{k_L C_{Ai}} \tag{9}$$

Sherwood and Holloway (1940) in Danckwerts (1970) give the following expression for k_L and a [4]

$$k_L = 0.42 \left(\frac{g\mu}{\rho} \right)^{\frac{1}{3}} \left(\frac{D_{AB}\rho}{\mu_L} \right)^{\frac{1}{2}} \tag{10}$$

$$a = 0.38 \left(\frac{u}{u_t} \right)^{0.775} \left(\frac{u\rho}{nd\mu_L} \right)^{0.125} \left(\frac{g\rho}{d\sigma} \right)^{\frac{1}{3}} \tag{11}$$

Here ut is the velocity of rise of the bubbles in the froth (taken to be 26.5cm/s under most conditions), n the number of holes per unit area of plate. For sieve trays, Chan and Fair (1983) give the following expression for gas-phase mass-transfer coefficient k_G a [7].

$$k_G a = \frac{316D_G^{0.2} (1030f + 867f^2)}{h_L^{1/2}} \tag{12}$$

Where: f = approach to flood, fractional

The interfacial Concentration of H₂S gas dissolved at interface estimated from solubility data following Henry's law and by considering gas side resistance that is,

$$C_{Ai} = \frac{k_G P_A + E.k_L C_A^0}{E.k_L + k_G H_e} \tag{13}$$

III. RESULT AND DISCUSSION

In this study steady state and isothermal condition are assumed and this study of simulation comprises sieve trays column 70 cm in diameter filled with sieve trays 0.5 m spacing to the height of 450 cm. Simulation the absorption of H₂S from sour gas available at 30°C, and column is operated at atmospheric pressure, the feed sour gas flow rate is 2401 kg/h with mole fraction of H₂S in inlet sour gas is 0.28887, the liquid flow rate 10000 cm³/s, the concentration of Fe₂(SO₄)₃ in inlet absorbent 0.002 mol/cm³. The simulation result in the form of mole fraction distribution of H₂S in gas phase, y_A, in sieve trays column, and concentration distribution of Fe₂(SO₄)₃ in liquid phase, C_B, in sieve trays column and plot graphically influence various process variables versus % recovery H₂S showed at Fig. (1) to Fig. (6).

Distribution of Mole Fraction and Concentration Distribution in tray Column

Figure.1 depicts the prediction results of the distribution of mole fraction H₂S in gas phase, y_A, and concentration distribution of Fe₂(SO₄)₃ in liquid phase as a function of axial position in sieve tray column. Simulation conditions: y_A in 0.28875, G = 751.144.763 liter/s, L= 10 liter/s C_(Fe₂SO₄) = 0.002 mol/cm³. and temperatures 30°C. and pressure = 1 atm.

In this figure, $Z = 0$ indicates the top of column and $Z = 4.5$ m indicates bottom of column. Figure.1 shows that the mole fraction of H_2S in gas phase increases rapidly from top to bottom while concentration of $Fe_2(SO_4)_3$ in liquid phase decreases slowly due to the amount of B as in excess to that required for theoretical reaction.

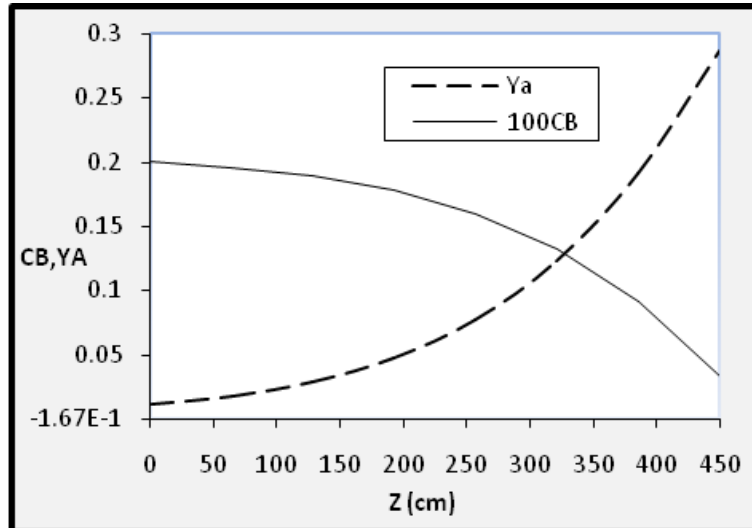


Fig. 1: Distribution of concentration $Fe_2(SO_4)_3$ liquid phase and distribution of fraction H_2S in gas phase in sieve trays column

Effect of Temperature and Concentration of $Fe_2(SO_4)_3$ Solution on H_2S Removal Efficiency

Figure.2 shows; An increase in the value of the temperature also produces an increase on the %removal of H_2S . According to the principle of molecular dynamics, the rate constant of reaction and the diffusion coefficient increase with the increase in the temperature, and this is beneficial to improve the absorption rate. In an absorber, the transfer of H_2S from sour gas into $Fe_2(SO_4)_3$ liquid brings about a heating effect.

Also Figure.3 shows indicates that the effect of temperature but with difference concentration of $Fe_2(SO_4)_3$ (2, 2.5, and 3M). For temperature the %removal of H_2S is given the same result in Figure.2 For that the increase in the value of $Fe_2(SO_4)_3$ concentration is still give influence to increase of H_2S removal efficiency. This is because when the concentration of $Fe_2(SO_4)_3$ is high, more H_2S must be introduced into the system to react with $Fe_2(SO_4)_3$, and this thing of course is caused because at higher concentration of $Fe_2(SO_4)_3$ in solution, the ferric ion would be more reactive with sulphur ion, so that will increase absorption rate of H_2S from sour gas.

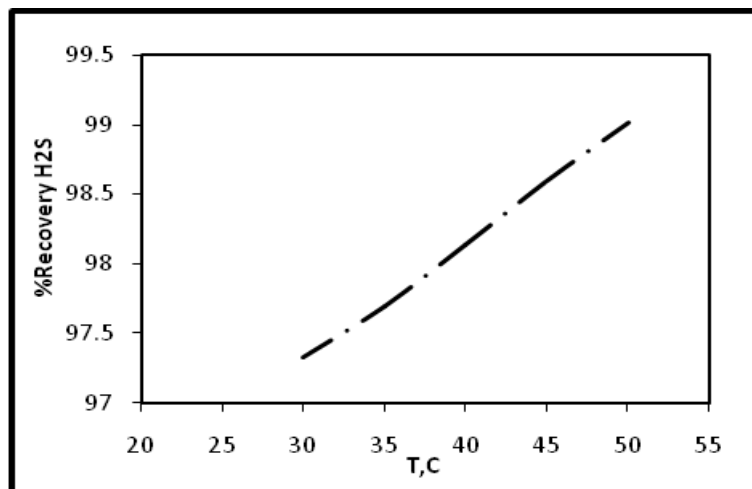


Fig.2: Temperature Influence on the % Removal

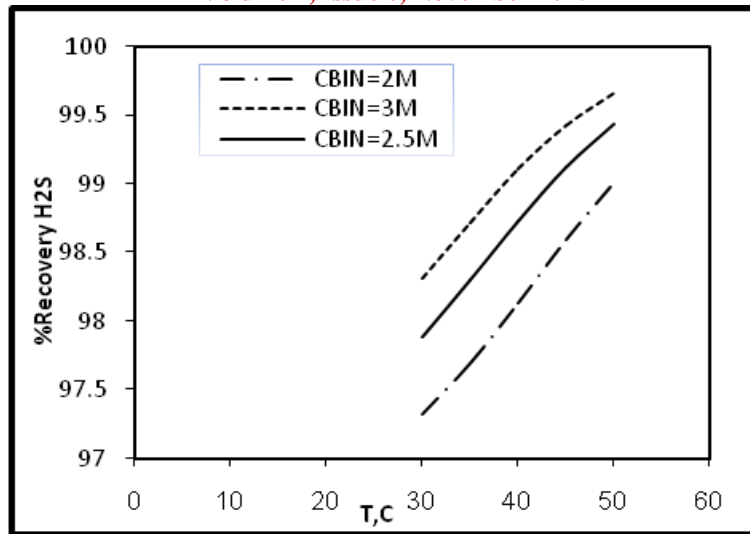


Fig 3: Temperature Influence on the % Removal with different solvent concentration of $(Fe_2SO_4)_3$

The Effect of Flow Rate of $Fe_2(SO_4)_3$ Solution on H_2S Removal Efficiency:

Figure.4; shows indicates that the increase in the value of $Fe_2(SO_4)_3$ flow rate is still give influence to increase of H_2S removal efficiency. Liquid flow rate determines how readily Component H_2S will absorb into the liquid phase. When the liquid flow rate is large, component H_2S will be absorbed more readily into the liquid phase.

This increase of H_2S removal efficiency occurred because the increasing of absorbent flow rate the liquid disturbance is enhanced, increase of driving force concentration that caused as result of decrease of concentration of H_2S in liquid bulk, which results in a higher speed of H_2S diffusing into the liquid. The consumed absorbents at the boundary layer could diffuse into the liquid phase at a higher speed due to the increase of liquid flow rate.

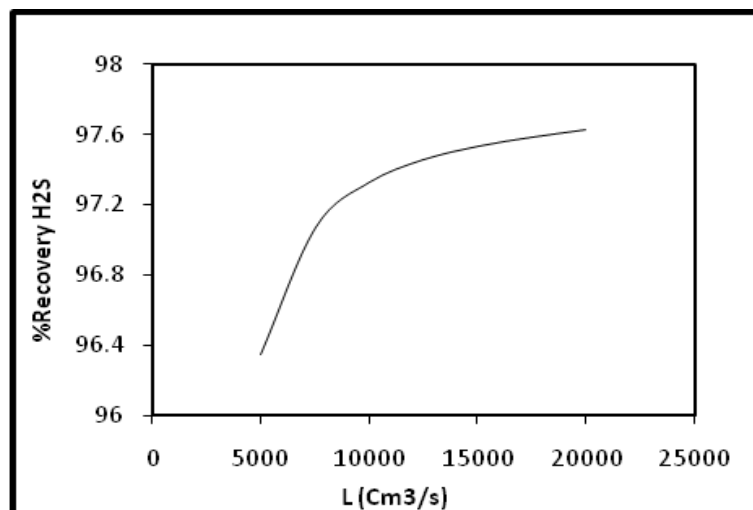


Fig 4: Flow Rate of $(Fe_2SO_4)_3$ Influence on the % Removal

Also Figure.5. Shows indicates that the effect of $Fe_2(SO_4)_3$ flow rate but with difference temperature (30-50°C). For %removal of H_2S is given the same result in Figure .4

In addition, as the increase of liquid flow rate, the thicknesses of gaseous and liquid-phase boundary layers decrease, leading to enhancement of the mass transfer rate, also it can be explained when the flow rate increased, the interfacial area between gas-liquid places increased. Therefore, the result is to enhance gas-liquid mass transfer rate and improving the H_2S absorption efficiency.

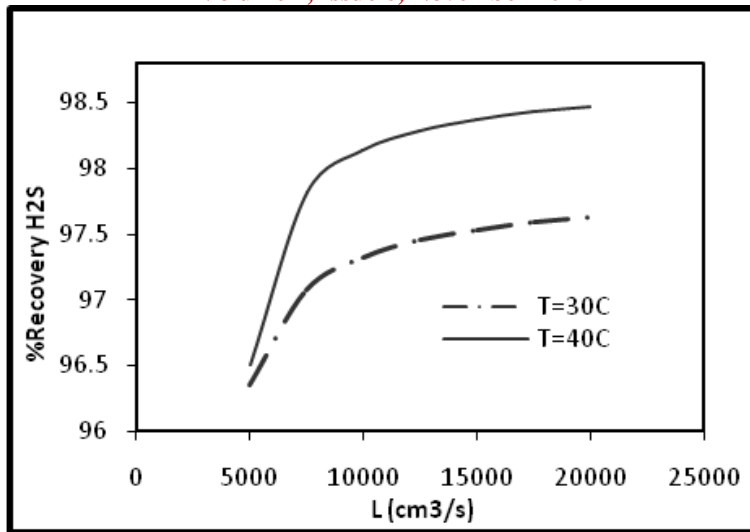


Fig .5: Flow Rate of (Fe₂SO₄)₃ Influence on the % Removal with different Temperature

The Effect of gas Flow Rate of Sour gas on H₂S Removal Efficiency:

Figures .6. shows indicates that the increase in the value of inlet sour gas flow rate is give to declines and main negative effects on removal efficiency H₂S removal efficiency. This is because when the flow rate of sour gas is high would a decrease the gas - liquid two phase contact time in the absorption column so the reaction rate of H₂S absorption slowed down. It was also that the increasing sour gas flow rate carried spray absorbent out from the tower, and led to the loss of the absorbent. Therefore, be concluded that the gas flow rate condition tends to become unfavorable to H₂S absorption when the gas flow rate increases within the operation range investigated.

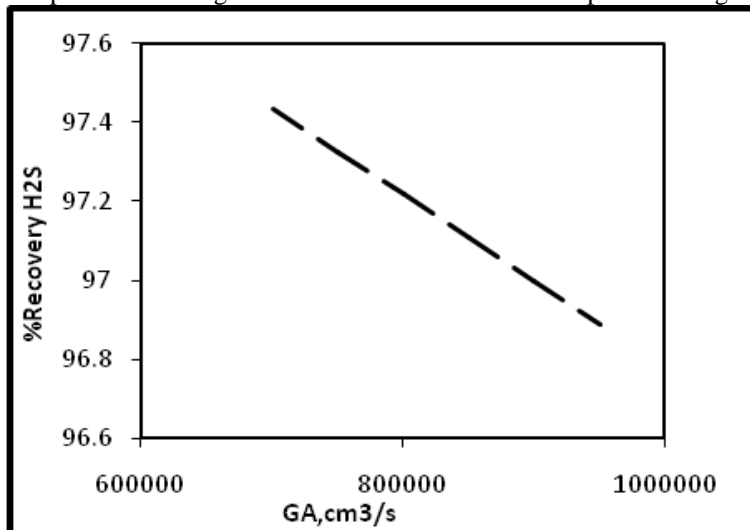


Fig.6: Flow Rate of sour gas Influence on the % Removal

IV. CONCLUSION

H₂S removal from sour gas using aqueous ferric solution Fe₂(SO₄)₃ needs an advanced air pollution control device to reduce greenhouse gas emissions. And as absorbent has been studied theoretically, calculations for absorber have been done, and their dimensions are obtained for a known sour gas flow rate.



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Absorption of H_2S by $Fe_2(SO_4)_3$ solutions was performed in this study to clarify the H_2S absorption efficiency. The absorption of H_2S into aqueous solutions of $Fe_2(SO_4)_3$ as encountered in sour gas was carried out using sieve plate column.

Various important operating factors affecting the H_2S absorption efficiency were discussed in this research. Various operating parameters, including liquid flow rate, $Fe_2(SO_4)_3$ concentration, temperature, and gas flow rate, were tested to determine the effect of these variables on the H_2S absorption efficiency. The simulation results showed that the percentage of absorbed H_2S can be enhanced and should have higher efficiency by increasing temperature, liquid flow rate, and $Fe_2(SO_4)_3$ concentration and decreasing flow rate of sour gas.

The methodology proposed, whereby only one parameter in each column is used to be employed to develop simulation models of industrial plants where measurements of operating variables are available and possible.

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