Empirical Survey on the Sweetening Process For Sour Gas, A New Technology

Somayyeh Fazeli[1], Farshad Farahbod[2] (Corresponding Author)

[1.] Department of Chemical Engineering, Sirjan Branch, Islamic Azad University, Sirjan, Iran.

[2.] Department of Chemical Engineering, Firoozabad Branch, Islamic Azad University, Firoozabad, Iran.

E-mail: mf_fche@iauf.ac.ir

Abstract

Many different processes are used to treat raw natural gas to pipeline quality. Sulfur is commonly present as an impurity in fossil fuels. Magnetic field is applied in a fluidized bed which contains nano activated carbon to investigate hydrogen sulfide elimination. Sulfur removal in this way is presented experimentally and theoretically. The rate of mass transfer is introduced as function of gas temperature, amount of balls covered by nano carbon tubes, initial concentration of hydrogen sulfide, gas flow rate and magnetic field. The experimental data are presented and compared with the model results. The effect of hydrogen sulfide in the inlet sour gas on the mass flow rate in investigated in this paper. In addition, the effect of porosity percentage of catalytic bed on the hydrogen sulfide content of outlet gas is evaluated in this paper. The outlet concentration below 4

ppm is acceptable result due to commercial rules. The experimental data are in higher values of hydrogen sulfide comparing with the ones from modeling data.

1. Introduction

The sour gas is natural gas or any other gas containing significant amounts of hydrogen sulfide (H_2S). Natural gas is usually considered sour if there are more than 5.7 milligrams of H_2S per cubic meter of natural gas, which is equivalent to approximately 4 ppm by volume under standard temperature and pressure. However, this threshold varies by country, state, or even agency or application. For instance, the Texas Railroad Commission considers a sour gas pipeline one that carries gas over 100 ppm by volume of H_2S. However, the Texas Commission on Environmental Quality has historically defined sour gas for upstream operations – which requires permitting, reporting, and possibly additional emission controls – as gas that contains more than 24 ppm by volume. Natural gas that does not contain significant amounts of hydrogen sulfide is called "sweet gas." Although the terms acid gas and sour gas are sometimes used interchangeably, strictly speaking, a sour gas is any gas that specifically contains hydrogen sulfide in significant amounts, whereas an acid gas is any gas that contains significant amounts of acidic gases such as carbon dioxide (CO_2) or hydrogen sulfide. Thus, carbon dioxide by itself is an acid gas, not a sour gas. In addition to being toxic, hydrogen sulfide in the presence of water also damages piping and other equipment handling sour gas by sulfide stress cracking. Natural gas typically contains several ppm of volatile sulfur compounds, but gas from one well in Canada is known to contain 90% hydrogen sulfide and others may have H_2S contents in the tens of percent range. Burning fuels, the sulfur is released as sulfur dioxide—an air pollutant responsible for respiratory problems and acid rain [1-6]. Environmental regulations have increasingly restricted sulfur dioxide emissions, forcing fuel processors to remove the sulfur from both fuels and exhaust gases. The cost of removing sulfur from natural gas and petroleum in the United States was about $1.25 billion in 2008. In natural gas, sulfur is present mainly as hydrogen sulfide gas (H_2S), while in crude oil it is present in sulfur-containing organic compounds which are

converted into hydrocarbons and H_2S during the hydro desulphurization [7, 8 and 9]. In both cases, corrosive, highly-toxic H2 S gas must be converted into elemental sulfur and removed for sale or safe disposal [10, 11 and 12]. Formation fluids that contain Hydrogen Sulfide - By-product from anaerobic bacterial action on sulfur compounds present in the mud (i.e. Sodium Sulfite) - Thermal degradation of mud additives containing sulfur (i.e. Lignosulfonates) - Chemical reactions with tool joint lubricants containing sulfur [13, 14 and 15].

H2S is a weak acid that can go through the following 2 stages when dissolved in water or water based mud:

H2S ↔ +H+HS- -	1

both steps (1 and 2) can go back and forth depending on the pH.

HS- + OH- ↔ -S+H2O	2

2. Materials and Method

Sour gasses which contain different amounts of hydrogen sulfide are reactor bed feed. Two gray 20 lit of volume pressurized vessel contain sour gas can be joint to the experimental line. However, both feed vessel can not be used simultaneously. Plexiglas vessel with inside diameter of 8 cm and height of 60 cm is the reactor vessel. 1 cm from bottom and 1 cm from the top of inside height of vessel is without packing contains distributor and holders. In 4 cm of efficient volume of vessel plastic balls with 2.5 cm in diameter which are covered by nano activated carbon tubes are used as the packing.

2.1. Chemical reaction

Pre humidification is done to enhance the amount of adsorbed H2S on nano balls. Nano carbon tubes are contacted with moist air (RH=100%) for 20 min before using in the reactor. This time is longer for current activated carbon but nano carbon is applied in this study and humidity adsorption occurs in shorter contact time. This step is an exothermic process. Humidity helps oxidation of H2S and also increases the nano carbon capacity for H2S adsorption.

2.2. Mathematical modeling

For laminar flow, where only viscous drag forces come into play, NRe,p 20 , experimental data may be correlated by means of the Kozeny-Carman equation, the Equation 3 [1]:

$$f_b = \frac{150(1-\varepsilon)}{N_{\mathrm{Re},p}\phi_s}$$ | 3

Note: According to chemical engineering hand book [5], the factor of 150 was originally given by Carman as 180 for the case of laminar flow. Ergun later suggested a better value was 150 when the particles are greater than about 150 m in diameter.

In addition, for highly turbulent flow where inertial forces predominate, NRe,p 1000 , experimental results may instead be correlated in terms of the Blake-Plummer equation, Equation 4 [7]:

$$f_b = 1.75$$ | 4

3. Results and Discussion

Bed porosity, inlet gas temperature, inlet concentration of H_2S, magnetic field which influences the mass transfer area, mass transfer coefficient and rate of reaction are considered here.

The amounts of hydrogen sulfide catch from branch line defined the process performance. Inlet sour gas contains 148 ppm H2S, temperature of 30 C is fed into the catalytic bed with 0.48 m3/m3 porosity and magnetic field occurred with current of 1 A.

### 3.1. The effect of bed porosity on sulfur elimination

Three different amounts of porosity are achieved by three numbers of balls, 4, 5 and 6 balls. In this case higher porosity presents lower amount of nano carbon tube and also lower amount of mass transfer surface area. In the fluidized bed the channeling malfunction is not occurred and all packing's are moved with the gas velocity and gas is contacted to solid catalyst easily. There is not any unused surface area. So, the increase trend of C is predicted by increase in the amount of porosity. Outlet concentration below 4 ppm is acceptable result due to commercial rules. Experimental data are in higher values of hydrogen sulfide comparing with ones from modeling data. Figure 1 shows the trend of outlet hydrogen sulfide concentration with bed porosity, V_{nb}/V.
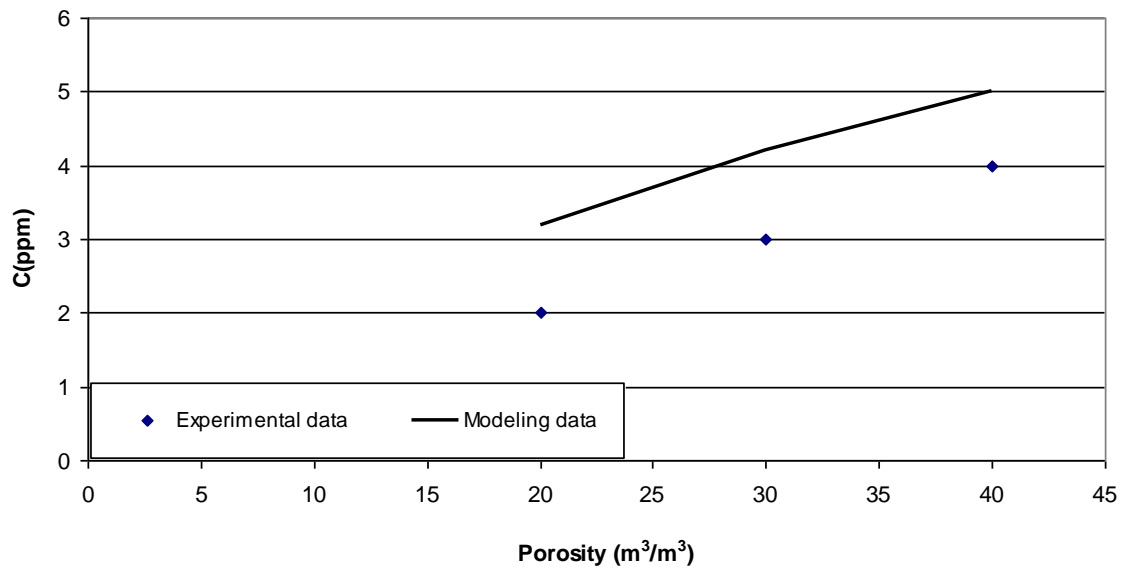
Figure 1. The effect of porosity on outlet hydrogen sulfide.

3.2. The effect of initial concentration and gas flow rate

Gas phase is not pure so gas, liquid and solid resistance should be considered in mass transfer rate. H2S adsorption on catalyst surface is considered physically and chemically. Both mechanisms are dependent on film of water on the nano carbon tubes. The effect of gas flow rate on mass transfer rate and mass transfer coefficient is studied here. The increase in initial amount of hydrogen sulfide decreases the gas phase resistance and facilitates the total mass transfer rate. The increase in flow rate with the fixed surface area increases the gas velocity and turbulence of gas phase. This also increases the solubility of H_2S on water film.
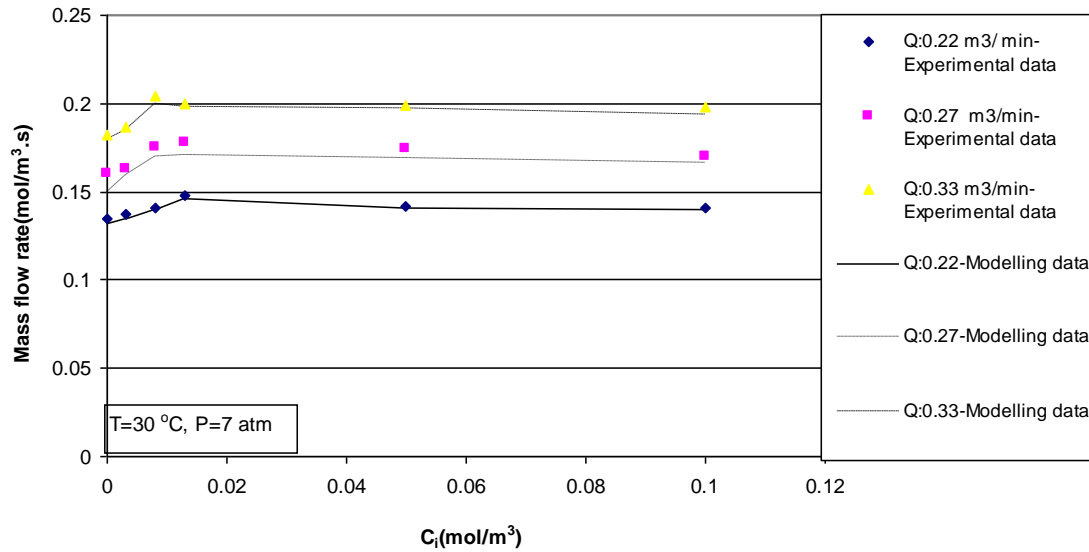
Figure 2. Mass flow rate versus inlet hydrogen sulfide and gas velocity.

Figure 2 shows the effect of gas flow rate and initial concentration of hydrogen sulfide on values of mass transfer flow rate. The increase in amount of gas flow rate increases the value of mass flow rate. This may indicate on the high adsorption capacity of nano carbon tubes and high surface area in fluidized bed. At 0.22 m3/min value of gas flow rate the increase in amount of initial hydrogen sulfur (0 to 0.013 mol/m^3) increases the flow rate initially and then the relatively decrease trend in flow rate is observed. This trend is similar for both 0.27 and 0.33 but the peak of graph happens in lower amount of initial hydrogen sulfide. This shows the limited value of carbon nano tube capacity. Also, the positive effect of turbulence on decreasing the gas phase resistance is observed. The higher value of driving force doesn't show the significant effect on increase of mass transfer rate when the capacity of nano carbon is limited. However, the theoretical values are predicted in lower ranges than values of experimental data. This difference may relate to calculation of mass transfer coefficient. The average mass transfer coefficient for gas phase is only considered in the relation while there is mass transfer coefficient for liquid film, actually.

4. Conclusion

Fluidized bed equipped with magnetic field which contains nano carbon tube is used to remove hydrogen sulfide from sour natural gas. Mass transfer rate and mass transfer coefficient is

measured and calculated experimentally and theoretically. The rate of mass transfer is introduced as function of gas temperature, amount of balls covered by nano carbon tubes, initial concentration of hydrogen sulfide, gas flow rate and magnetic field. Results show the hydrogen sulfide in outlet stream increases with increasing the porosity. In addition, the obtained results show the exponential correlation can defined the relation between the friction factor and pressure drop. The mass flow rate versus initial concentration of hydrogen sulfide is evaluated in this study. The experimental results show the initial concentration can be affected on the mass flow rate at the beginning.

References

[1] L. Carlos, G. Isabel, B. Irene, D. Luis I., R. Luis M. Experimental study of SO2 and NOx emissions in fluidized bed oxy-fuel combustion. Fuel Process Techno., 2013; 106: 587–594.

[2] M. de las Obras-Loscertales, A. Rufas, L.F. de Diego, F. Garcń-Labiano, P. Gayán, A. Abad, J. Adánez, Effects of Temperature and Flue Gas Recycle on the SO_2 and NO_x Emissions in an Oxy-fuel Fluidized Bed Combustor, Energy Procedia., 2013; 37: 1275-1282.

[3] W. Kaewboonsong, V.I. Kuprianov, N. Chovichien, Minimizing fuel and environmental costs for a variable-load power plant (co-)firing fuel oil and natural gas: Part 1. Modeling of gaseous emissions from boiler units, Fuel Processing Technology, 2006; 87: 1085-1094

[4] A.Irabien, Environmental and economic evaluation of SO_2 recovery in a ceramic hollow fibre membrane contactor. Chem Eng Process: Process Inten., 2012; 52: 151-154.

[5] H. Wang, Sh. Li, F. Lai, B. Wang, Computational Model of Greenhouse Gas Emissions of Power Station boiler Considering Desulphurization, Physics Procedia, 2012; 24: 44-49.

[6] D.L. Stern, K.E. Nariman, J.S Buchanan, N.A. Bhore, D.L. Johnson, R.K. Grasselli, The Mobil Oil SO_x Treatment Process (MOST). Catalytic removal of SO_x and H_2S from refinery tail gas, Catalysis Today, 2000; 55: 311-316.

[7] W. Zhou, C.S. Zhao, L.B. Duan, XP. Chen, C. Liang, Two-dimensional computational fluid dynamics simulation of nitrogen and sulfur oxides emissions in a circulating fluidized bed combustor. Chem. Eng. J., 2011; 173: 564-573.

[8] D. Eow, S. John, Recovery of sulfur from sour acid gas: A review of the technology Environmental Progress. Americ. Institut. Chem. Eng., 2002; 21: 143 - 162.

[9] D. Kunii, O. Levenspiel, Fluidization engineering. First edition. New York: Wiley; 1991.

[10] JF. Davidson, Fluidization. First edition. USA: Academic Press; 1991.

[11] D. Green, R. Perry. Perry's Chemical Engineers' Handbook. 8th edition, USA: Mc Graw Hill; 2007.

[12] W.Ch. Yang. Handbook of Fluidization and Fluid-Particle Systems. First edition. USA: Taylor & Francis; 2003.

[13] L.Davidson, Jr Amick, H.Erwin, Formation of gas bubbles at horizontal orifices. AIChE J 2004; 2: 337–342.

[14] M. Uzi. Principles of chemical reactor analysis and design. second edition. New York: Wiley; 2009.

[15] O. Levenspiel, Chemical Reaction Engineering. 3th Ed. New York: Wiley; 1999.