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Selectivity study of H_2S and CO_2 absorption from gaseous mixtures by MEA in packed beds

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ABSTRACT

In this work, an experimental and theoretical investigation of selectivity was performed for simultaneous absorption of H_2S and CO_2 from gaseous streams into an aqueous mono-ethanol amine (MEA) solution in a packed bed. The operating parameters playing a vital role and have been considered here, are amine concentration and pressure, while the structural and hydrodynamic parameters such as packing size and *L*/*G* ratio were found to be important as well. The qualitative effects of these parameters on absorption performance were analyzed via evaluating of absolute and relative removal efficiency. In the proposed mathematical model for theoretical investigation of the absorption performance, the two-film theory of mass transfer was used. In order to check the validity of the proposed model, in experimental section, a pilot-scale plant was constructed and variety of experimental data were obtained in a wide range of operating conditions. In this plant, construction of packed beds in several sections of the column, gives the possibility of changing the structural and hydrodynamic parameters in a flexible way. Also the proposed mathematical model is able to consider the effects of vital parameters on the selectivity of absorption. The predicted results by the proposed mathematical model were compared to the experimental data. This comparison shows a good agreement between these data and so verifies the validity of the proposed model. The results reveal that the effects of pressure, amine concentration, packing size and *L*/*G* ratio on the selectivity performance of H_2S absorption into an MEA solution are significant. Results show that, the relative efficiency could be increased near 20% when *L*/*G* is manipulated properly. As a side effect it reduces the absolute efficiency. Using the packing size 1 in. instead of 0.5 in., relative efficiency could be increased more than 5% while absolute efficiency is reduced. Then the experimental results testified the benefit of any operating procedure appropriately involving these parameters for selective H_2S absorption.

In general, since CO_2 is a weak competitor against H_2S in being absorbed by MEA solution, under difficult conditions for absorption process, the weakness of CO_2 is more pronounced. Therefore, the selectivity of absorption is increased, while the absolute efficiency is decreased. In particular, increasing the gas flowrate and decreasing the *L*/*G* ratio, increases the selectivity. Also when higher packing size is used, selectivity is increased. In contrast, when large amount of amine are contacted with a special quantity of acid gas in the unit volume of packed bed (generally lower packing size and higher *L*/*G* ratio), due to the strength of MEA, the amine solution absorbs nearly all gaseous acidic compounds and reduces the selectivity.

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Nomenclature

a	specific surface area of the column (m^2/m^3)
A	cross-sectional area of the tower (m^2)
C	liquid concentration ($kmol/m^3$)
C_i	liquid concentration of component i ($kmol/m^3$)
C_p	specific heat of mixture at constant pressure (kJ/(kmol °C))
d_p	packing diameter (m)
dv	volume of each volume element (m^3)
dz	height of each volume element
D_i	diffusivity of component i in liquid phase (m^2/s)
G	gas phase flow rate (m^3/s)
G'	mass flux of gas (kg/(m^2 s))
H_i	Henry's constant of component i in aqueous MEA solution
j	volume element counter
k_{CO_2}	kinetic reaction constant of CO_2 with MEA (m^3/(kmol s))
k_G	gas phase mass transfer coefficient (kg/(m^2 s))
k_L	liquid phase mass transfer coefficient (m/s)
K	equilibrium constant
L	liquid flow rate (m^3/s)
L'	mass flux of liquid (kg/(m^2 s))
L/G	MEA solution–gas ratio (mol MEA/mol gas)
n	number of data in statistical analysis
N_i	mass transfer flux of component i (kmol/(m^2 s))
P_i	partial pressure of component i in gas phase (kPa)
q_i	heat of absorption of component i (kJ/kmol)
r_i	consumption (reaction) rate of component i (kmol/s)
Sc	Schmidt number
Ss	standard deviation of statistical data for population
SE	standard deviation of statistical data for samples
t	confidence coefficient in statistical analysis
T	temperature (K)
T_L	liquid bulk temperature (K)
V_i	molecular volume (m^3/kmol)
x	direction toward depth of liquid phase
x_i	mole fraction of component i in liquid phase (mol i/mol liquid)
y_i	mole fraction of component i in gas phase (mol i/mol gas)

Greek letters

α	confidence limit in statistical analysis
δ_c	thickness for zone of liquid phase which there, the reaction of CO_2 with MEA takes place (m)
ε	packed bed void fraction (m^3/m^3)
η_i	absolute efficiency of absorption of component i
η_{rel}	relative efficiency which is equal to (η_{H_2S}/η_{CO_2})
μ	viscosity (kg/ms)
μ_1	normal population mean of each data set
Φ	association factor for solvent
Φ_0	degree of freedom in two way ANOVA test

1. Introduction

Removal of H_2S from gaseous mixtures by use of amine solutions is a conventional technique (variety of processes such as SCOT and natural gas sweetening are using this technique). Usually these gaseous mixtures contain 1–5% CO_2 which is a close competitor of H_2S (less than 1.5%) in being absorbed by amine solutions. So the behavior of selective H_2S absorption by amine solution is very interesting from economical and operational points of view. It should be noted that the targets for modeling of this system in different situations, might be completely different. For example in SCOT process which the inlet gas contains considerable amount of CO_2, even many times as existing H_2S, the purpose of modeling is to study the process with emphasis on the selectivity of absorption. This subject has been studied in different aspects by different investigators (Zheng et al., 2006; Lyddon and Nguyen, 1999; Yasavage et al., 2001; Rascol et al., 1997; Pant and Srivastava, 2007; Glasscock and Rochelle, 1993). The basic idea in majority of these studies is the use of the sterically hindered type of amines as a chemical factor toward improving the absorption selectivity. In some researches, physical parameters have been investigated to show that they have such a potential for developing the new practical parameters toward achieving more selectivity in chemical absorption. In this manner, it has been verified that, although MEA is not a sterically hindered amine, but it could be a selective chemical absorber (Pant and Srivastava, 2007).

For a long time MEA has been the widest industrially used amine (especially in oil refineries). Therefore, any successful effort toward optimum usage of this amine (even a little progress) leads to considerable improvement. MEA is a strong amine and under well prepared conditions in absorption process, it absorbs almost all available gaseous acidic compounds without serious competition (alkalinity potential). By use of secondary and tertiary amines, difference potential between absorption of H_2S and CO_2 will be more pronounced due to the weakness of these amines. These types of amines are naturally selective and when they are involved in the liquid mixture, the observed improvements followed from the key parameters in this article are not noticeable beside the effects of the intrinsic selectivity of these amines.

The main orientation of this study is the controllable employing the existing potential of selectivity for absorption of H_2S and CO_2 into aqueous MEA solutions. In order to examine the pure effects of proposed parameters on the observed selectivity, the unselective amine like MEA was chosen as a case study here.

In spite of numerous researches in this field, little considerations have been given to the mechanisms and related parameters of selective amine absorption. For example, the structural parameters such as packing size and the hydrodynamic parameters such as L/G ratio have not been considered widely as effective parameters on mechanisms and selectivity of absorption for these compounds. It is the purpose of this work to consider these parameters in details. To do this a combination of chemistry and transport phenomena is considered as a basis of the mathematical model. The resulted trends let us conclude useful guidelines for appropriate use of these parameters to achieve more selective performances.

The pilot-scale plant used in the experimental study was constructed in sulfur recovery unit (SRU) in Shiraz oil refinery. It should remained as close as possible to the industrial condi-