

**CO₂ CAPTURE BY WATER-BASED Al₂O₃ AND Al₂O₃-SiO₂ MIXTURE
NANOFLUIDS IN AN ABSORPTION PACKED COLUMN**

**CAPTURA DE CO₂ MEDIANTE NANOFLUIDOS BASADOS EN MEZCLAS DE Al₂O₃
Y Al₂O₃-SiO₂ EN UNA COLUMNA DE ABSORCIÓN EMPACADA**

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Abstract

In the present work, the influence of mixture of Al₂O₃ and Al₂O₃ - SiO₂ nanoparticles in base fluid water (nanofluid) on absorption of CO₂ in a packed column was investigated. The effects of concentration of nanoparticles and types of nanoparticles on the absorption rate were investigated. The packed column was used to study gas liquid countercurrent flow. The results indicated that the optimum value was equal to 0.05 vol% for both nanofluids, and also addition of nanoparticles increased the mass transfer rate. Besides, the maximum enhancement of mass transfer rate for nanoparticles of Al₂O₃ and mixture of Al₂O₃ - SiO₂ compared to the pure water was 14% and 10% at 0.05 vol% of the nanofluid, respectively.

Keywords: absorption, carbon dioxide, nanofluids, packed column, Al₂O₃.

Resumen

En este trabajo, estudiamos el efecto de las nanopartículas Al₂O₃ y compuestos de Al₂O₃- SiO₂ en el fluido a base de agua, es decir nanofluido, a la absorción de CO₂ dentro de una columna empacada. También se ha investigado el efecto de la concentración así como el tipo de la nanopartícula en la tasa de la absorción. Se ha utilizado una columna empacada para investigar el flujo de gas líquido contracorriente. Los resultados indicaron que la cantidad óptima para ambos nanofluidos es 0.05 por ciento del volumen y el aumento de la concentración de las nanopartículas, causa el aumento de la tasa de la transferencia de masa. Además la mayor tasa de la transferencia de masa para nanopartículas Al₂O₃ y compuestos de Al₂O₃- SiO₂ en comparación del agua destilada son 14 y 10 por ciento respectivamente, lo cual sucede en concentración de 0.05 por ciento de volumen.

Palabras clave: absorción, dióxido de carbono, nanofluido, columna empacada, Al₂O₃.

1 Introduction

Population growth and development industry and technology are factors that increase environmental pollution such as production of wastewater and greenhouse gases, therefore investment must be done in treatment and disposal of that. That is why a lot of studies and methods have been done for wastewater treatment and removal of dangerous materials like Biological methods (Gan *et al.*, 2013, Gupta *et al.*, 2015), use of adsorbents (Bello-Huitle *et al.*, 2010, Martín-González., 2013) and other methods (Lopez-Ojeda *et al.*, 2015, Staehelin., 1984, Shen *et al.*, 2006). Gas absorption such as CO₂ capture is one of the most important processes in the chemical industry. Reducing the emission of CO₂, which considered a

greenhouse gas, has found great importance in recent years. Lopez *et al* (Lopez *et al.*, 2014) studied the absorption of carbon dioxide in aqueous solutions of Triethanolamine using a bubble-column reactor. The effects of gas flow rate and concentration of TEA on the absorption rate were investigated. They found that the absorption rate increases at the beginning of the experiments with low TEA concentration but it decreases with increasing of TEA concentration. To enhance the physical absorption of CO₂, an ionic liquid (IL), 1-butylimidazole hexafluoride phosphate [bmim] PF₄, was dispersed in surfactant solution to form an IL-in-water emulsion by Liu *et al* (Liu *et al.*, 2013). Their results indicated that the dispersed IL droplets can significantly increase the absorption rate of CO₂ at the gas-liquid interface.

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One of the general goals of scientists and engineers is to increase efficiency and miniaturize the systems used in the industry. Nanotechnology has enabled us to make nanoparticles with different types and sizes. Heat transfer nanofluids were first reported by Choi (Choi, 1995). Nanofluid is a new fluid, in which particles with diameter smaller than 100 nm are suspended in the base fluid. Recently, many studies have been carried out on increasing mass transfer by adding nanoparticles. However, the number of these studies is still less than the ones performed on heat transfer and thermal conductivity. Increasing the mass transfer coefficient increases efficiency and reduces size and cost of the equipment. In general, there are three methods for enhancement of mass transfer; mechanical treatment, chemical treatment, and nanotechnology approach (Kang *et al.*, 2003). In mechanical methods, the absorption rate is improved by modifying the shape, surface, and structure of the system (Isshiki *et al.*, 1996; Kim *et al.*, 2003). Chemical methods consist of adding surfactants to the base fluid (Kim *et al.*, 2005), while in the nanotechnology approach; the mass transfer is enhanced by addition of nanoparticles.

The use of nanoparticles as additives in the absorption gas to improve the mass transfer fluid has attracted many researchers. Kim *et al.* carried out a study to investigate the effect of binary nanofluids on the absorption of gas in a bubble column. It was observed that the size of the bubbles generated in nanofluids is smaller than that in the base fluid; therefore, their absorption in nanofluids is fast. It is found that the absorption rate in nanofluids is greater than that in the base fluid and the absorption rate increases with increasing the concentration of nanoparticles. The results have shown that the absorption rate increased to 3.21 times by adding the nanoparticle (Kim *et al.*, 2006) and this increase can be up to 5.32 times when surfactant and nanoparticle are used together (Kim *et al.*, 2007). Kim *et al.* (Kim *et al.*, 2008) added silica nanoparticle to water (base fluid) to absorb CO₂ gas in a bubble column and observed that the absorption rate increases up to 76% during the first minute and total amount of absorption up to 24%. The effect of carbon nanotube on gas absorption is reported by Ma *et al.* (Ma *et al.*, 2009). They concluded that addition of nanoparticles enhances mass transfer. Yang *et al.* (Yang *et al.*, 2011) studied the effect of nanoparticles of Al₂O₃, Fe₂O₃, and ZnFe₂O₄ in a binary nanofluid of ammonia-water on gas absorption in a wetted-wall column. They obtained the optimal values for

each of nanoparticles and surfactants. For Fe₂O₃ and ZnFe₂O₄ nanoparticles, the absorption rate increased 70% and 50% at 15% of the mass concentration of ammonia, respectively. Lee *et al.* (Lee *et al.*, 2011) studied the effect of Al₂O₃ and SiO₂ nanoparticles on CO₂ absorption in a bubble column with methanol-based fluids. They found that there are optimal values for nanoparticles and the maximum absorption of CO₂ for Al₂O₃ and SiO₂ nanoparticles (compared to the pure methanol) are 4.5% and 5.6% at 0.01 vol%, respectively. Pineda *et al.* reported CO₂ capture in a tray column with Al₂O₃ and SiO₂ nanoparticles using methanol-based fluids (Pineda *et al.*, 2012). They showed that maximum enhancement in the absorption rate (compared to the base fluid) are 9.4% at 0.05 vol% of Al₂O₃ and 9.7% at 0.05 vol% of SiO₂. Jung *et al.* studied the effect of Al₂O₃ nanoparticle in methanol (base fluid) on absorption (Yung *et al.*, 2012) and they found that the absorption rate of nanofluid is 8.3% (at 0.005 vol%) higher than the base fluid. The Effect of magnetic nanoparticle was studied by Wu *et al.* (2013) and Komati *et al.* (2008). They reported that addition of magnetic nanoparticles increase the absorption rate.

The effect of magnetic nanoparticles NiO and Fe₃O₄ and water as base fluid on CO₂ absorption in a packed was reported by Salimi *et al.* (2015). They found that addition of nanoparticles into the solvent enhances mass transfer characteristics and the magnetic field showed positive effect on the CO₂ absorption performance.

In the current study, the effect of nanofluids water/Al₂O₃ and water/Al₂O₃ - SiO₂ mixture as a solvent has been investigated on CO₂ absorption in a packed column. The concentration of nanofluids and the type of mixture nanoparticle are considered as key parameters influencing the absorption rate. Moreover, for the first time, the effect of mixing the nanoparticles on the absorption is investigated. The aim of the present work is to obtain optimal concentration, at which the maximum absorption rate occurs for nanoparticles.

2 Preparation of nanofluids

Preparation of nanofluids is the first step in the application of this concept. Nanofluids with more stability have more effective in enhancing mass transfer. Because some of the specific characteristics of nanofluid, such as high heat and mass transfer coefficient and the microconvection, have more effects in nanofluid with more stability than poor (Yang *et al.*,

2011). In this research, a two-step method was used to prepare nanofluids. At first, Al₂O₃ (dp=15-20 nm) and SiO₂ (dp=10-15 nm) nanoparticles were used at the concentrations of 0.005, 0.01, 0.05, 0.1 and 0.2%vol. Then, ultrasonic and mechanical stirrers were used to stabilize nanoparticles in the fluid. Finally, the solution was placed under ultrasonic oscillations for one hour. In order to avoid the deposition of nanoparticles in the base fluid, the prepared nanofluid immediately was used to the absorption CO₂.

3 Experimental apparatus

Fig. 1 shows the schematic view of the apparatus used to measure the gas absorption. The column was made of glass and was filled with Raschig ring packing. A round Lexan flow meter was used to measure the gas flow rate. GR flow meter was used to measure the liquid flow rate. Two thermometers (accuracy ±1 oC) were used at the outlet and inlet of the liquid to measure the liquid temperature. Glass wool was used to insulate the glass column and the operating conditions kept near isothermal conditions. The device properties and operational inlet conditions are shown in Tables 1 and 2, respectively.

CO₂ gas enters from the bottom of the instrument and the fluid enters from the top of the tower as shown in Fig. 1. After steady state condition is achieved, CO₂ concentration in the liquid outlet is measured by the titration method. The mass transfer rate (N_A) and the mass transfer coefficients ($K_L a$) are calculated by the following equations:

Table 1. Properties of the instrument.

Parameters	
Length of column (cm)	90
Diameter of column (cm)	3
Diameter of packing (mm)	4
Void fraction	0.89

Table 2. Operational inlet conditions for the packed column absorber.

	Inlet flux (l/min)	Inlet Pressure (bar)	Temperature (°C)	Purity (%)
CO ₂	1	1	-	99.99
Water	0.3	-	25	-

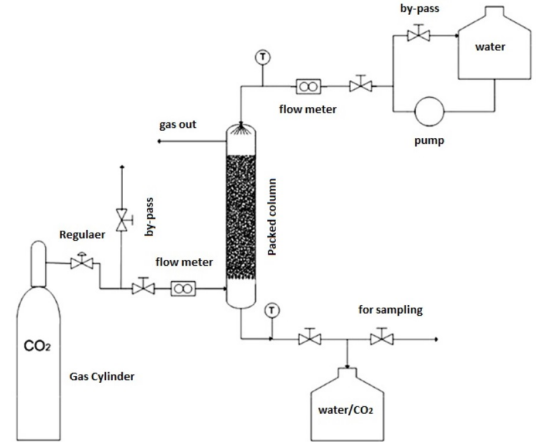


Fig. 1. Schematic view of the apparatus.

$$N_A = \frac{Q}{S.Z}(C_0 - C_i) \quad (\text{mol/m}^3\text{s}) \quad (1)$$

$$(K_L a)_{ave} = \frac{Q}{S.Z} \ln \frac{C^* - C_i}{C^* - C_o} \quad (2)$$

where Q is the fluid flow rate, S the tower cross section, Z the tower height, C_i the feed concentration, C_o the outlet concentration, and C^* is the saturation concentration.

For validation, the results of experiments obtained for the mass transfer coefficients in pure water were compared to a model proposed by Billet and Schultes (Billet *et al.*, 1999). They have offered the model base on 3500 experiments in packed columns for 46 different systems. The model is based on following equations:

$$\frac{a_w}{a} = 1.5(a.d_h)^{-0.5} Re_l^{-0.2} Fr_l^{-0.45} We_l^{0.75} \quad (3)$$

$$Re_l = \frac{u_l d_h}{\nu_l} \quad (4)$$

$$Fr_l = \frac{u_l^2 \rho_l d_h}{\sigma_l} \quad (5)$$

$$We_l = \frac{u_l^2}{g d_h} \quad (6)$$

$$K_L a_w = C_l 12^{1/6} u_l^{-0.5} \left(\frac{D_l}{d_h}\right)^{0.5} a \left(\frac{a_w}{a}\right) \quad (7)$$

$$\bar{u}_l = \frac{u_l}{h_l} \quad (8)$$

$$h_l = \left(12 \frac{\eta_l}{\rho_l g} u_l a^2\right)^{1/3} \quad (9)$$

All the experimental data have errors. Even if the experimental design is highly accurate, the presence of random errors will affect the results. If

$$N_A = N_A(Q, V, C_o)$$

And if $Y = \ln \frac{C^* - C_o}{C^*}$ Then $Kla = Kla(Q, V, Y)$

To obtain the experimental error for final results, the following equations will be determined.

$$E_{N_A} = \frac{\sqrt{\left(\frac{\partial N_A}{\partial Q} E_Q\right)^2 + \left(\frac{\partial N_A}{\partial V} E_V\right)^2 + \left(\frac{\partial N_A}{\partial C_o} E_{C_o}\right)^2}}{N_A} \quad (10)$$

$$E_{Kla} = \frac{\sqrt{\left(\frac{\partial Kla}{\partial Q} E_Q\right)^2 + \left(\frac{\partial Kla}{\partial V} E_V\right)^2 + \left(\frac{\partial Kla}{\partial Y} E_Y\right)^2}}{Kla} \quad (11)$$

E_Q = flow rate error (l/m)

E_V = volume error (m3)

E_{C_o} = concentration error (mol/m3)

E_Y = error of Y (-)

E_{N_A} = mass transfer rate error (mol/m2s)

E_{Kla} = mass transfer coefficient error (1/s)

According to equations 3 and 4, the average errors for mass transfer rate and mass transfer coefficient are about 2.6% and 4%, respectively.

4 Discussion and conclusions

Fig. 2 shows a comparison between the experimental results obtained and Billet and Schultes model (Billet *et al.*, 1999). It is clear that the trends are same and the differences might be due to the operating conditions and packing type.

Figs. 3 and 4 show the mass transfer rate based on the concentration of nanoparticles.

It is found that the mass transfer rate increases with increasing the concentration of nanoparticles, and the maximum enhancement in the mass transfer rate is at 0.05 vol%. Therefore, the concentration of 0.05 vol% is an optimal value for CO₂ absorption in nanofluids of water/Al₂O₃ and water/mixture of Al₂O₃ - SiO₂. There is no mechanism that completely justifies the increased mass transfer rate with nanofluids; thus, such mass transfer enhancement is still speculative. The Brownian motion of nanoparticles cannot directly enhance the mass transfer (Krishnamurthy *et al.*, 2006), but it is one of the main factors creating micro convection in nanofluids (Krishnamurthy *et al.*, 2006; Kim *et al.*, 2012; Sara *et al.*, 2011). The grazing effect of nanoparticles is another parameter that increases the mass transfer (Kars *et al.*, 2011; Kim *et al.*, 2005).

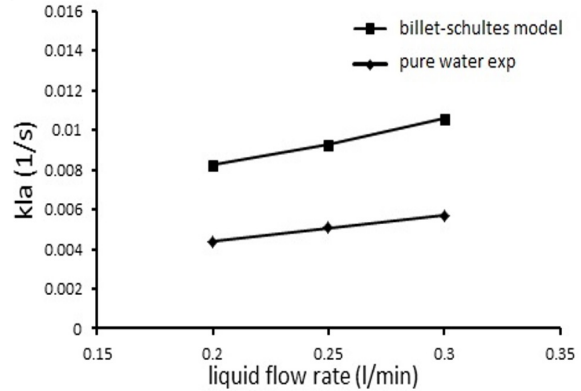


Fig. 2. Comparison of the experimental data and Billet and Schultes model.

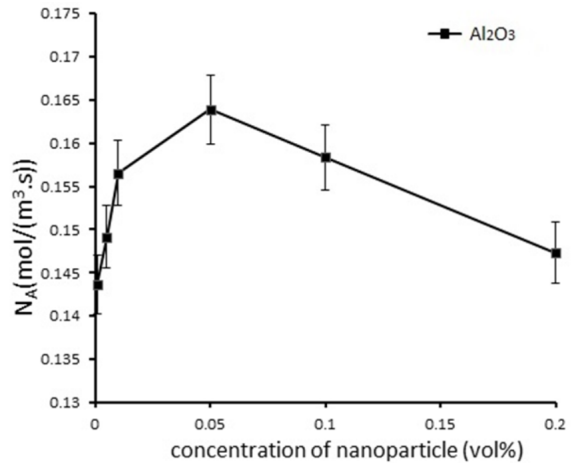


Fig. 3. Mass transfer rate at the different concentration of nanoparticles for Al₂O₃ nanoparticle.

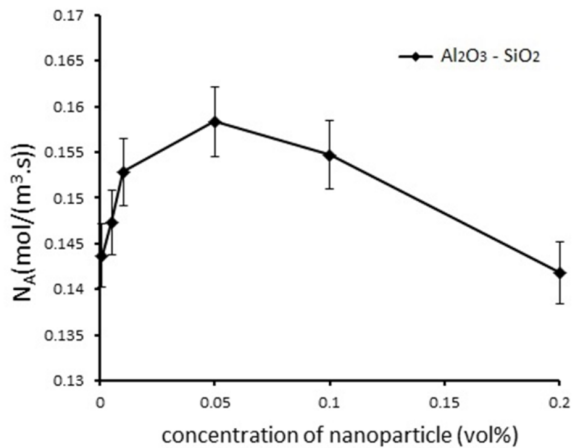


Fig. 4. Mass transfer rate at the different concentration of nanoparticles for mixture of Al₂O₃-SiO₂ nanoparticles

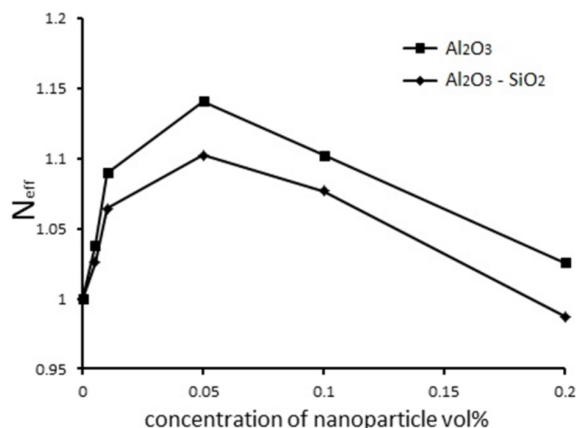


Fig. 5. Effective mass transfer ratio at the different concentration of nanoparticles.

Due to this grazing effect, the gas molecules are adsorbed on the particles in the nanofluid and moved to the bulk liquid where they desorb.

Fig. 5 shows the effective mass transfer ratio (N_{eff}) obtained from following equation:

$$N_{eff} = \frac{N_{nf}}{N_{bf}} \quad (12)$$

where N_{nf} and N_{bf} are the mass transfer rates of nanofluids and base fluids, respectively.

Fig. 5 shows that the maximum enhancement in the mass transfer rate for Al_2O_3 and mixture of $Al_2O_3 - SiO_2$ are about 14% and 10% respectively, which occur at the concentration of 0.05 vol%. When the concentration of nanoparticles is greater than 0.05 vol%, the mass transfer rate will decrease. It can be due to decreased self-diffusion coefficient of the fluid (Pineda *et al.*, 2012). Gerardi *et al.* (Gerardi *et al.*, 2009) showed a reduction trend in the diffusion coefficient for Al_2O_3 / water and proposed two reasons for this phenomenon; first, the water molecules collide the nanoparticles and the curvature of their diffusion path are enhanced, second, the water molecules stick on the surface of particles and move with them, which have a diffusion coefficient smaller than the free molecules.

Another possible factor for reducing the mass transfer rate is that the nanoparticles at high concentrations stick together and they become larger and their Brownian motion is reduced (Kim *et al.*, 2012; Lee *et al.*, 2011). Also, viscosity increases with increasing the concentration of nanoparticles that is a negative factor in increased mass transfer rate (Yang *et al.*, 2011).

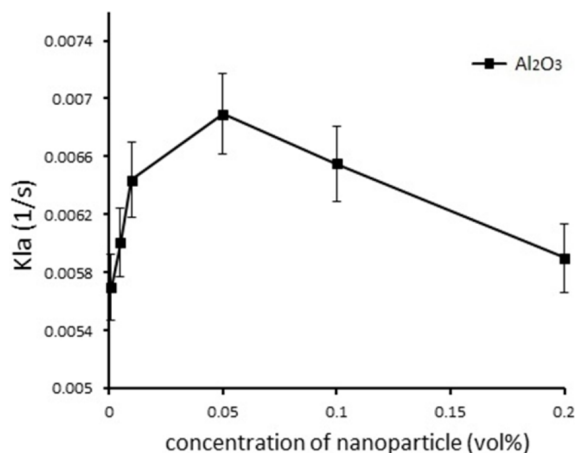


Fig. 6. Mass transfer coefficient at the different concentration of nanoparticles for Al_2O_3 nanoparticle.

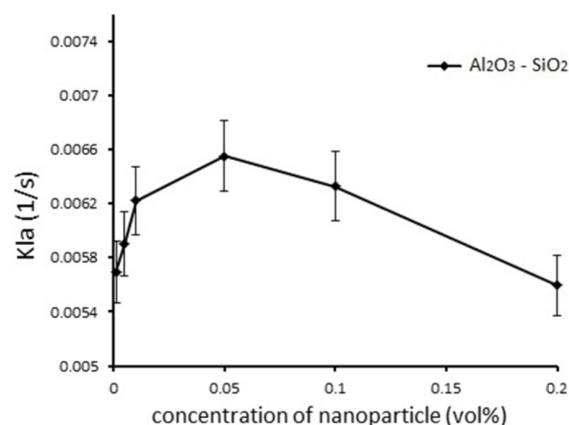


Fig. 7. Mass transfer coefficient at the different concentration of nanoparticles for mixture of $Al_2O_3 - SiO_2$ nanoparticle.

Effects of nanoparticles volume fraction on mass transfer coefficient are shown in Figs. 6 and 7.

It is clear that the mass transfer coefficient increases with increasing the concentration of nanoparticles and the maximum enhancement in mass transfer coefficient is at 0.05 vol%. As noted previously, the mass transfer coefficient is a function of several factors such as the Brownian motion of nanoparticles, as well as diffusivity and viscosity of the fluid. It is found that beyond 0.05 vol%, reduced diffusion coefficient and increased viscosity are more effective than the micro convection created by the Brownian motion of nanoparticles; therefore, the mass transfer coefficient is reduced.

When the nanoparticle concentration is upper a critical concentration, the amount of nanoparticles become too dense, so that they have a negative

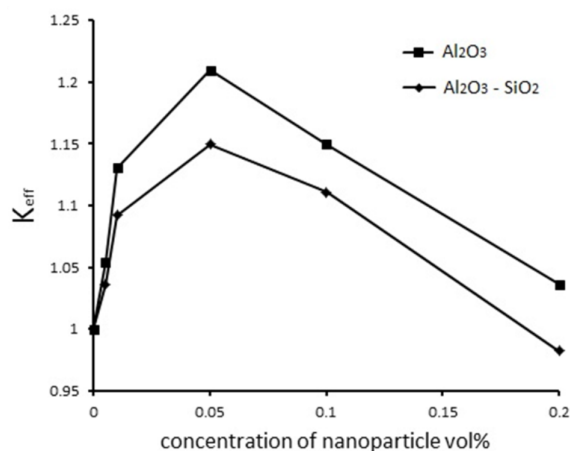


Fig. 8. Effective mass transfer coefficient at the different concentration of nanoparticle.

effect and reduce the absorption process; therefore, the viscosity is increased highly with increasing nanoparticle concentration (Lee *et al.*, 2015). On the other hand, the thickness of diffusion layer is directly proportional with the viscosity. Enhancement of the nanofluid viscosity increases the thickness of the diffusion boundary layer; furthermore, the mass transfer coefficient decreases (Syam Sundar *et al.*, 2013, Samadi *et al.*, 2014).

Fig. 8 shows the effective mass transfer coefficient (K_{eff}) obtained from equation 3:

$$K_{eff} = \frac{K_{nf}}{K_{bf}} \quad (13)$$

where K_{nf} and K_{bf} are the mass transfer coefficient of nanofluids and the mass transfer coefficient of base fluids, respectively.

The maximum enhancement in mass transfer coefficient for Al_2O_3 and mixture of $Al_2O_3 - SiO_2$ are about 20% and 15%, respectively, which occurred at the concentration of 0.05 vol%. Fig 8 also shows a comparison between the nanoparticles of Al_2O_3 and mixture of $Al_2O_3 - SiO_2$ in the CO_2 absorption process that shows, Al_2O_3 nanoparticles have performance better than that observed for combined $Al_2O_3 - SiO_2$. This may be due to the different structure of Al_2O_3 and SiO_2 . Yang (Yang *et al.*, 2010) studied the effect of α - Al_2O_3 and γ - Al_2O_3 nanoparticles on gas absorption and concluded that γ - Al_2O_3 has better performance than α - Al_2O_3 . The two types of Al_2O_3 have different structures because of different properties of their surface, which leads to different ability to increase the mass transfer rate.

Conclusions

In this study, the nanofluids of water/ Al_2O_3 and water/mixture of $Al_2O_3 - SiO_2$ were used for gas absorption in a packed column and the following results were obtained:

1. Both the mass transfer rate and the mass transfer coefficient increased with adding the nanoparticles to the base fluid.
2. The maximum enhancement in mass transfer and the mass transfer coefficient for Al_2O_3 nanoparticle was around 14% and 20%, respectively, which occurred at the concentration of 0.05 vol%.
3. The maximum enhancement in mass transfer and the mass transfer coefficient for mixture of $Al_2O_3 - SiO_2$ nanoparticle was around 10% and 15%, respectively, which is related to the concentration of 0.05 vol%.

Nomenclature

N_A	mass transfer rate, $\text{mol s}^{-1} \text{m}^3$
Q	volume flow rate, $\text{m}^3 \text{s}^{-1}$
S	cross-sectional area, m^2
Z	column length, m
C	concentration, mol m^{-3}
Kla	mass transfer coefficient, s^{-1}
Y	parameter
E	error
a	specific surface area of packing, $\text{m}^2 \text{m}^{-3}$
C	constant in Eq. 7
d	diameter, m
D	diffusion coefficient, $\text{m}^2 \text{s}^{-1}$
Fr	Froude number
g	gravitational acceleration, ms^{-2}
h_l	column holdup, $\text{m}^3 \text{m}^{-3}$
Re	Reynolds number
\bar{U}	mean effective velocity, ms^{-1}
U	velocity, ms^{-1}
We	Weber number
d_h	hydraulic diameter, m
σ_L	surface tension, kgs^{-2}

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