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The simultaneous effect of graphene oxide and sodium dodecyl sulphate nanoparticles on the kinetics of CO_2 absorption in amine solvent

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ABSTRACT

Amine solvents are extensively used on an industrial scale for removing carbon dioxide (CO_2). The presence of some additives in amine solvents has a desirable effect on CO_2 absorption kinetics and also improves the absorption process. In this study, graphene oxide (GO) nanoparticles and the anionic surfactant sodium dodecyl sulphate (SDS) were used as additives to the amine solvent. The number of CO_2 moles that were used (n_g), the values of the diffusion coefficient (D_{AB}), and the mass transfer coefficients of CO_2 gas absorption in the amine solvent (K_c) were determined. Furthermore, the effect of the additives on the kinetics of CO_2 gas absorption in the amine solvent was investigated. The results showed that mass transfer coefficients increased with a decrease in pressure and an increase in temperature as well as in the SDS and GO concentrations. The values of the mass transfer coefficient under different conditions varied between 0.0311 and 0.0587 *cm/s*. The molecular diffusion coefficient of CO_2 in the amine solvent increased from 0.000025 to 0.000287 cm^2/s with decreases in the pressure and with increases in the temperature and increases in concentrations of additives. The laboratory data were statistically analyzed via Design-Expert software using response surface experiment design and a historical method. A mathematical relation was proposed to estimate the mass transfer coefficients. Moreover, a mathematical relation was introduced to predict the molecular diffusion coefficient of CO_2 in the amine solvent.

1. Introduction

With the development of human communities and their ever-increasing needs, various industries have greatly expanded causing global CO_2 emissions to increase day by day. The produced CO_2 accumulates in the atmosphere, giving rise to adverse effects on global weather. This has prompted many scientists and researchers to conduct numerous studies on CO_2 removal and absorption. Consequently, many studies have been conducted on the removal and absorption of CO_2 by liquid solvents. The use of amine solvents for CO_2 absorption are extensively used in various industries for CO_2 absorption. Jostein Gabrielsen *et al.* [1], Haji-Sulaiman *et al.* [2], D.P. Hagewiesche *et al.* [3], Boumedine *et al.* [4], and Cheng *et al.* [5] investigated CO_2 absorption by amine solvents in their studies. Such studies proved that the fine particles increased solubility; then, nanoparticles were introduced, and it was found that the use of surfactants had desirable effects. Therefore, various nanoparticles and surfactants have been employed to optimize and increase the efficiency of CO_2 gas absorption in amine solvents. Some studies have researched the solubility of CO_2 in the presence of nanoparticles and surfactants. Taheri *et al.* (2016) examined the simultaneous absorption of carbon dioxide (CO_2) and hydrogen sulfide (H_2S) from a CO_2-H_2S-CH_4 gas mixture in an ethanolamine solvent using 0.005-0.5 wt% SiO_2 and Al_2O_3 nanoparticles at 0.005-0.5 wt% in a wetted wall column (WWC). The results showed thatCO_2 absorption was improved by 33% in the presence of Al_2O_3 nanoparticles at 0.05 wt% and by 40% in

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the presence of SiO_2 nanoparticles at 0.05 wt%, respectively [6]. Rahmatm and *et al.* (2016) investigated the increase in CO_2 absorption by SiO_2, Al_2O_3, Fe_3O_4nanoparticles,and carbon nanotubes (CNTs)in aqueous amine solutions.The results indicated that SiO_2 and Al_2O_3 nanoparticles were more effective at high concentrations (0.1 wt%) and were able to increase absorption capacity by up to 21 and 18%, respectively; whereas, the Fe_3O_4 nanoparticles and CNTs were more effective at lower concentrations (0.02 wt%) and improved absorption capacity by up to 24 and 34%, respectively [7]. Tao Wang *et al.* (2016) used three different types of nanoparticles (SiO_2, TiO_2, and Al_2O_3) to increase CO_2 absorption via the mono ethanolamine (MEA) solvent. They observed that the overall mass transfer increased with increases in the loading rates of the nanoparticles under the influence of the bubble burst mechanism; the CO_2 absorption rate also improved by up to 10% [8]. Jiazong Jiang *et al.* (2014) studied both the effect of adding various nanoparticles (TiO_2, MgO, Al_2O_3 and SiO_2) and the nanoparticle size on CO_2 absorption in MEA and MDEA solutions [9]. Ratnesh *et al.* (2015) conducted experiments and performed modelling to investigate the increases in the mass transfer rates of CO_2 absorption in water in a capillary tube by using SiO_2 and TiO_2.Theynoticed that mass transfer coefficients increased by more than 165% at the low volume percentages of the nanoparticles [10]. Wang *et al.* (1997) studied the improvement in the kinetics of CO_2 absorption in a wetted wall column and the synergistic effect of nanoparticles on this improvement. The suspended nanoparticles in the solutions have the potential power to increase gas-liquid mass transfer, and they have been studied for gas separation. Diffusion and reaction processes have been carefully investigated to reveal the kinetic enhancement mechanism. Experiments and the study of models showed that micro-convection induced by the Brownian motion of particles played an important role in mass transfer enhancement. MEA, MDEA and PZ solutions, which have different kinetics, were selected to study the effect of nanoparticles on the absorption kinetics of solutions with various reaction rates. The increase in the absorption kinetics in the presence of nanoparticles followed the same order observed for the increased reaction rate between the solvent with CO_2, which was MDEA<MEA<PZ. According to the kinetic model for the WWC, kinetics-limited diffusion in PZ solution could lead to much more significant kinetics enhancement by adding nanoparticles. The mass transfer rate of the liquid fraction in the MEA solution could increase by more than 15% under conditions of optimization operations [11]. Farajzadeh *et al.* (2007) examined the mass transfer rate of CO_2 gas to water and to the aqueous solutions of sodium dodecyl sulfate (SDS). According to their results, natural convection in the early stages increased the mass transfer rate. Furthermore, adding pure SDS did not have a significant effect on the mass transfer rate of CO_2 gas in water under experimental conditions [12]. S.K. Verma et al. investigated the properties of nanoparticles in 2017 as an advantage in increasing heat and mass transfer. They stated that even adding a small amount of nanoparticles in the solutions could significantly improve the thermal efficiency, stability, and mass and heat transfer efficiency. [13] Zhang et al. (2018) reviewed the latest progresses in the use of nanofluids for CO_2 absorption and described the mechanisms presented in various research works [14]].The present study aims to investigate the CO_2 absorption kinetics and diffusion coefficient in monoethanolamine (MEA) in the presence of GO nanoparticles and at different concentrations of the surfactant SDS.

2. Calculations

2.1. Determine the amount of moles consumed of CO_2

The present study used the Peng-Robinson equation proposed by Peng-Robinson in 1976, in which the parameters of the state equation were obtained using the critical properties and a centric factor.

$$P = \frac{RT}{V-b} - \frac{a}{V(V+b)+b(V-b)}$$

$$\alpha = 0.45724\frac{R^2T_c^2}{P_c}[1+m(1-T_r^{0.5})]^2$$

$$b = 0.0778\frac{RT_c}{P_c} \quad (1)$$

$$T_r = \frac{T}{T_c}$$

$$P_r = \frac{P}{P_c}$$

$$m = 0.37464 + 1.54226\omega - 0.26992\omega^2$$

In the above relations, V is the molar volume, T_c and $P_c$are the temperature and pressure at the critical point, respectively, R the gas constant, and ω the acentric factor. The Peng-Robinson equation can be formulated in terms of the z-compressibility factor as follows:

$$Z^3 - (1-B)Z^2 + (A-3B^2-2B)Z - (AB - B^2 - B^3) = 0 \quad (2)$$

$$A = \frac{a\alpha P}{R^2T^2}B = \frac{bP}{RT}$$

The Z value obtained at the initial and final (equilibrium) temperature and pressure was used to calculate the number of initial and final (equilibrium) moles by employing Equations (3) and (4).

$$n_0 = \frac{VP_0}{Z_0RT_0} \quad (3)$$

$$n_e = \frac{VP_e}{Z_e RT_e} \quad (4)$$

where V is the volume of the loading chamber (V_{Lc}).
V is obtained from Equation (5).

$$V_{Solvent} - V_{Ac} + V_{Conectors} + V = V_{Lc} \quad (5)$$

The number of absorbed gas moles is obtained from Equation (6).

$$\Delta n = n_0 - n_e \quad (6)$$

2.2. Calculate the mass transfer coefficient

The Seidel and Linke equation was used to calculate the mass transfer and diffusion coefficients. This equation was published by Atherton Seidel and William F. Linke in 1952. The following relations can be used to calculate the mass transfer coefficient (K_c). The mass transfer coefficient, K_C, is obtained by plotting Q versus $(P_i-P_{eq})/RT$, and by calculating the slope of the line.

$$\frac{dn}{dt} = K_c * A * \Delta_P \quad (7)$$

$$\frac{dn}{dt} = K_c * A * \frac{(P_i - P_{eq})}{RT} \quad (8)$$

$$\frac{dn}{dt * A} = K_c * \frac{(P_i - P_{eq})}{RT} \quad (9)$$

$$Q = \frac{dn}{dt\,.\,A} = K_C * \frac{(P_i - P_{eq})}{RT} \quad (10)$$

$$A = \frac{\pi}{4} ab \quad (11)$$

In the above relations, P_i is the Initial pressure; P_{eq} is the equilibrium pressure; R is the gas constant; T is the temperature; a and b are the width and length of the absorption cell; A is the contact area of the liquid and gas equal to$47.1 cm^2$; and Q is the Molar Flux Absorption. By plotting the Q diagram in terms of $\frac{(P_i-P_{eq})}{RT}$and calculating the gradient of the line, the mass transfer coefficient K_c is determined.

2.3. Calculate the diffusion coefficient:

Equation (12) expresses the relationship between the molar flux of mass transfer and the intermolecular diffusion coefficient.

$$Q = 2C^* \sqrt{\frac{Dt}{\pi}} \quad (12)$$

D_{AB} *is the* CO_2 diffusion coefficient in solution in $\frac{cm^2}{s}$, *and* C^* is the amount of CO_2 dissolved in the solution in$\frac{mol}{cm^3}$.
By plotting Q versus $\sqrt{t}$ and obtaining the slope of the line, the following relations can be used to determine the diffusion coefficient of CO_2 in the amine solvent.

$$Slope = 2C^* \sqrt{\frac{D_{AB}}{\pi}} \quad (13)$$

$$D_{AB} = \frac{Slpoe^2 * \pi}{4C^{*2}} \quad (14)$$

3. Materials and methods

3.1. Laboratory device

Different parameters affect the absorption of a gas in a liquid and the gas solubility varies under different conditions. The solubility of a gas in a liquid varies depending on the type of gas, type of solvent, and contact time of both phases as well as the temperature and pressure conditions. To calculate the solubility, the number of gas moles must be determined at the beginning and end of the gas absorption process. An experimental setup was designed for this purpose. As the operation starts, CO_2 gas and MEA are brought into contact, and the two phases remain in contact until equilibrium is reached. The temperature and volume are constant; the number of CO_2 moles at the beginning, during, and at the end of gas absorption can be calculated with pressure changes during the process. The experimental setup is schematically shown in Figure. 1.

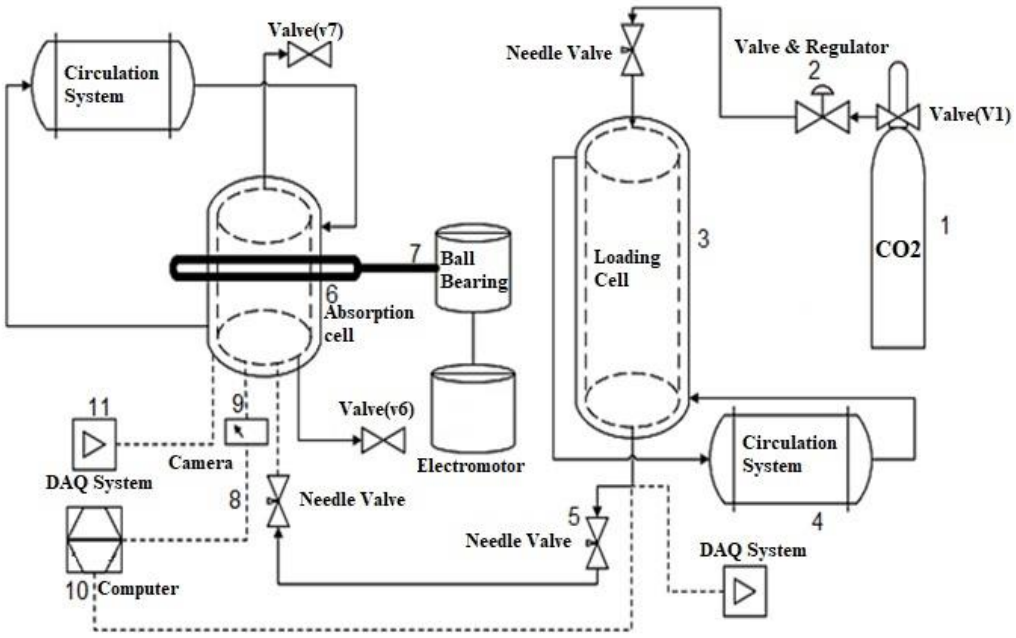

Fig. 1. Schematic view of the laboratory device used in this study.1-Co_2Save Capsule; 2-Crank valve regulator pressure regulator and pressure gauge; 3-loading cell; 4-Bathroom circulation system for temperature and circulation; 5-Needle valve; 6-absorption cell; 7-Electromotor and agitator absorption cell; 8-Temperature sensor; 9-pressure sensor; 10-Power box, Temperature and Pressure display; 11-Data Acquisition, Data Transfer and Registration (DAQ).

3.2. Materials

The carbon dioxide gas (MW=44.0095 g/mol) used in the experiments with a 99.99% purity was purchased from the Arvand Industrial Gases Company. Its solubility in water was 1.45 g/l. Monoethanolamine (MEA) with the chemical formula C_2H_7NO, molecular weight of 61.08 g/mol and 99% purity was purchased from the LobaChemie Company (India). Sodium dodecyl sulfate (SDS) was used as the surfactant in all the experiments. Graphene oxide (GO) nanoparticles with a 95% purity and a particle size of smaller than 60 nm were purchased from Nanotech. (India).

3.3. Experiments

The solution was first prepared to carry out the experiments. The following three solutions were tested:
a) MEA+ Distilled Water
b) MEA + Distilled Water + Surfactant
c) MEA + Distilled Water + Nanoparticles
First, the MEA solution with the concentration of 25 wt% was prepared using pure MEA and distilled water (base fluid). Then, the base fluid was mixed with a certain amount of surfactant and/or nanoparticles using an ultrasonic bath; it was stirred with a magnetic stirrer for 20 min. For each experiment, 100 ml of the above solution was prepared and injected into the absorption chamber using a syringe. The stirrer speed of 5 rpm was used in all the experiments. After setting this parameter, the data logging software was started, and the CO_2 was transferred from the loading chamber to the absorption chamber at the same time that the valve was opened. When the valve was opened, the CO_2 was absorbed by the solution, and the process continued until the system reached equilibrium. During this process, the data logger software recorded the pressure and temperature. The number of CO_2 moles consumed at each moment in the absorption process and the constants of the kinetics parameters of CO_2 absorption in amine solvent were calculated.

Table 1. Experimental tests conditions

Run	P in (bar)	T in (K)	SDS	GO
1	18.6	295.15	0	0
2	18.6	306.25	0	0.0375
3	19	297.35	0	0.05
4	20	298.75	0	0.075
5	18.7	298.55	0	0.1
6	18.9	299.05	0.0375	0
7	18.9	300.75	0.05	0
8	18.9	294.45	0.075	0
9	18.8|	299.45	0.1	0
10	18.9	297.35	0.075	0.05
11	18.6	297.15	0.05	0.05

4. Results and discussion

The experimental data were analyzed by Design-Expert software for design of experiments. The response surface design methodology and a historical method were used for statistical analysis. In this case, the four parameters affecting CO_2 absorption in the amine solution in the presence of the SDS surfactant and GO nanoparticles

(Tables 2 and 3) were randomly entered into the software. The parameters A, B, C and D represent reactor pressure, reactor temperature, SDS concentration and amount of GO, respectively. The effects of the changes in the above-mentioned parameters on the mass transfer coefficient of CO_2 absorption in the amine solvent (R_1), the intermolecular diffusion coefficient of CO_2 absorption in MEA in the presence of GO nanoparticles, and the SDS surfactant were investigated in the laboratory.

4.1. Statistical analysis of the parameters influencing the mass transfer coefficient of CO_2 absorption in the MEA solvent

Table 2 shows mass transfer coefficient calculated at different temperatures, pressures and GO and SDS concentrations in all the experiments. The statistical analysis yielded relation (15) for estimating the mass transfer coefficient of CO_2 absorption in the amine solvent:

$$K_c = 0.16 - 0.029A + 0.24B + 0.052C + 0.069D - 0.027AB - 0.17AC - 0.3AD + 0.12BC + 0.3BD + 0.06\,CD \quad (15)$$

Figure 2 shows the changes in the mass transfer coefficient with changes in reactor pressure. The mass transfer coefficients decreased with an increase in reactor pressure.

Table 2. Experimental results used for calculating the mass transfer coefficients

Run	Factor 1 A:P in (bar)	Factor 2 B:T in (K)	Factor 3 C: SDS (g)	Factor 4 D : GO (g)	Response 1 Kc
1	18.6	295.15	0	0	0.0311393
2	18.6	306.25	0	0.0375	0.044358
3	19	297.35	0	0.05	0.0327934
4	20	298.75	0	0.075	0.0320159
5	18.7	298.55	0	0.1	0.0396961
6	18.9	299.05	0.0375	0	0.0587312
7	18.9	300.75	0.05	0	0.0413235
8	18.9	294.45	0.075	0	0.0502357
9	18.8	299.45	0.1	0	0.0432016
10	18.9	297.35	0.075	0.05	0.0452878
11	18.6	297.15	0.05	0.05	0.0319444

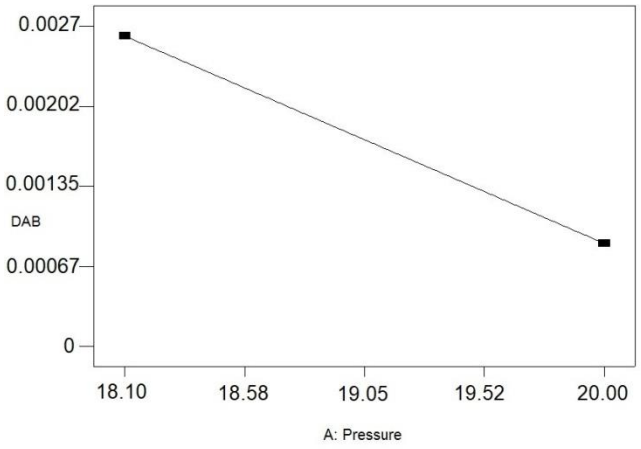

Fig. 2. The changes in the mass transfer coefficient of CO_2 absorption in MEA with the reactor pressure.

Figure 3 shows the changes in the mass transfer coefficient of CO_2 absorption with changes in the reactor temperature. The mass transfer coefficient of CO_2 absorption in the amine solvent significantly increased with an increase in reactor temperature.

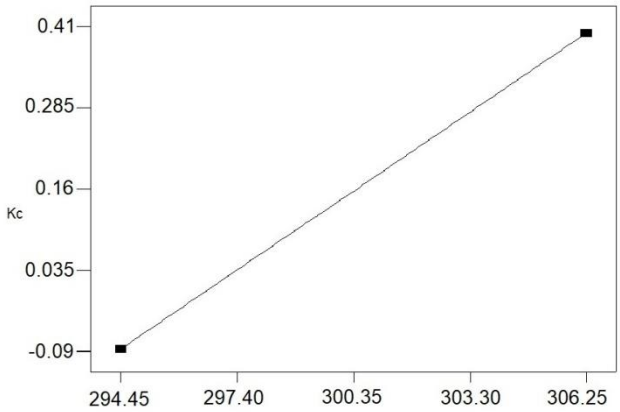

Fig. 3. The changes in the mass transfer coefficient of CO_2 absorption in MEA with the reactor temperature.

Figure 4 shows the changes in the mass transfer coefficient of CO_2 absorption in the amine solvent with changes in SDS concentration. The mass transfer coefficients of CO_2 absorption in the amine solvent increased with an increase in SDS concentration.

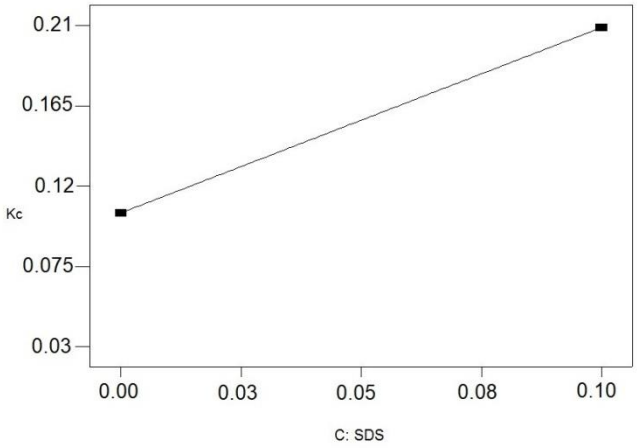

Fig. 4. The changes in the mass transfer coefficient of CO_2 absorption in MEA with SDS concentration.

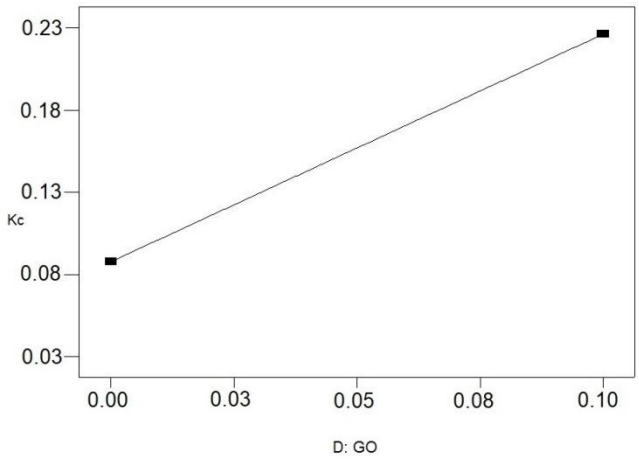

Fig. 5. The changes in the mass transfer coefficient of CO_2 absorption in MEA with GO concentration.

Figure 5 shows the changes in the mass transfer coefficient of CO_2 absorption in the amine solvent with changes in GO concentration. The mass transfer coefficient of CO_2 absorption in the amine solvent increased with an increase in GO concentration.

Figures 2 to 5 show the analysis of the individual effects of the parameters influencing mass transfer coefficient in CO_2 absorption in the amine solvent regardless of their interactions. The interaction effects of the parameters are discussed below.

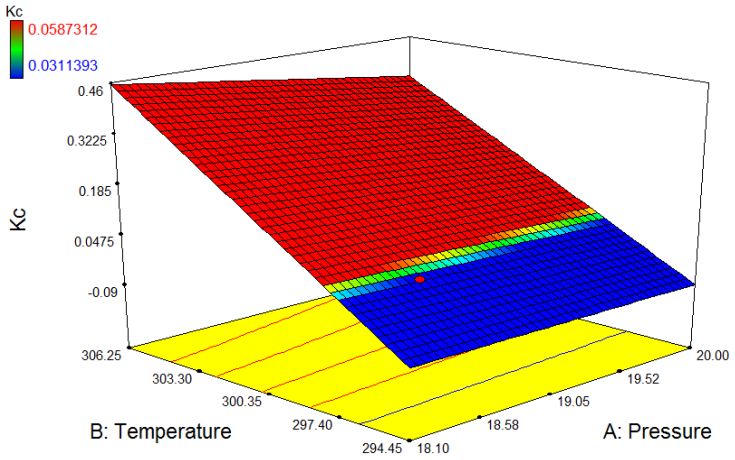

Fig. 6. The simultaneous interaction effects of reactor pressure and temperature parameters on mass transfer coefficient of CO_2 absorption in the amine solvent.

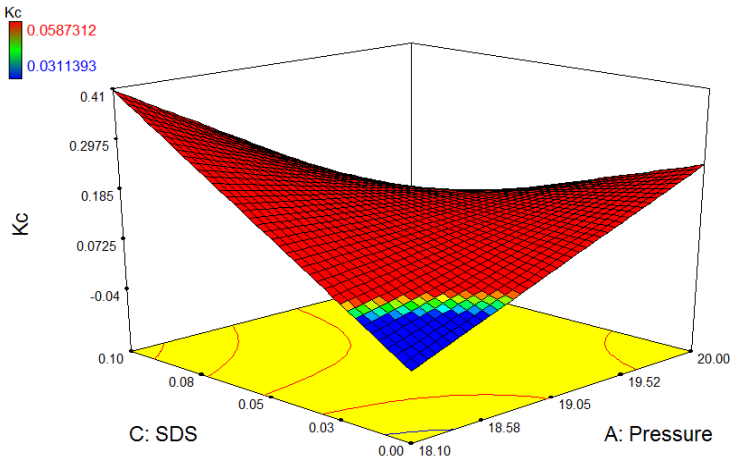

Fig. 7. The simultaneous interaction effects of the pressure and SDS concentration parameters on mass transfer coefficient of CO_2 absorption in the amine solvent.

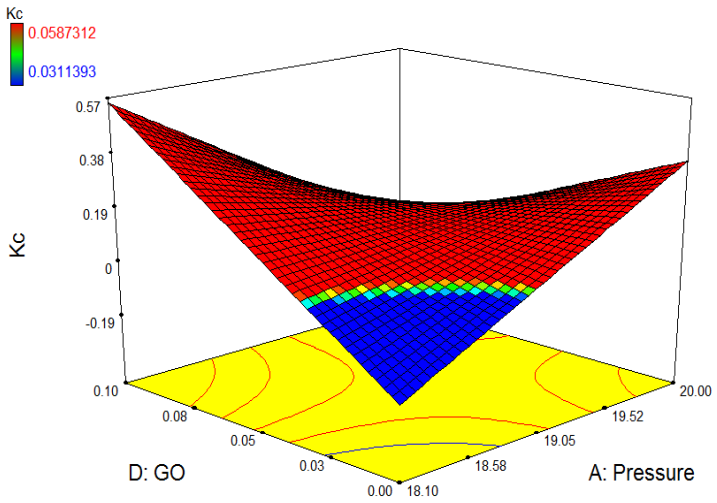

Fig. 8. The simultaneous interaction effects of the pressure and GO concentration parameters on mass transfer coefficient of CO_2 absorption in the amine solvent.

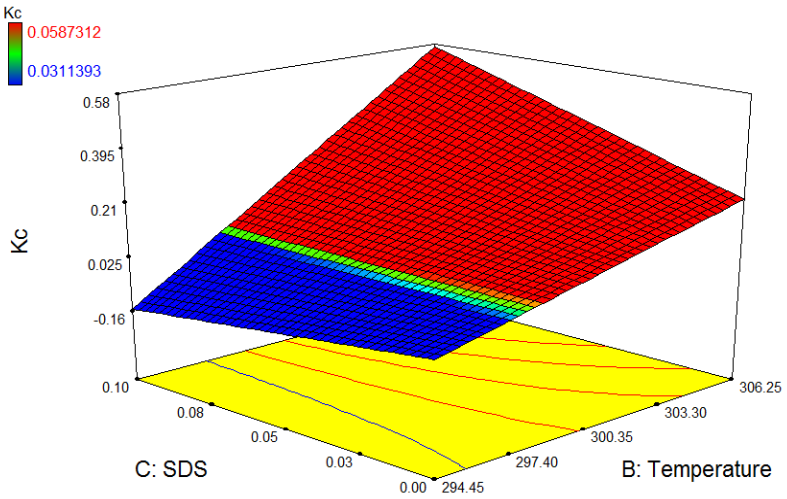

Fig. 9. The simultaneous interaction effects of the temperature and SDS concentration parameters on mass transfer coefficient of CO_2 absorption in the amine solvent.

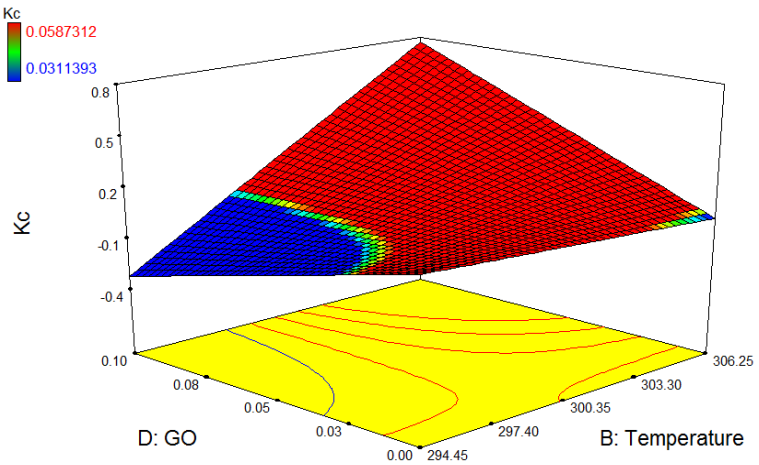

Fig. 10. The simultaneous interaction effects of the temperature and GO concentration parameters on mass transfer coefficient of CO_2 absorption in the amine solvent.

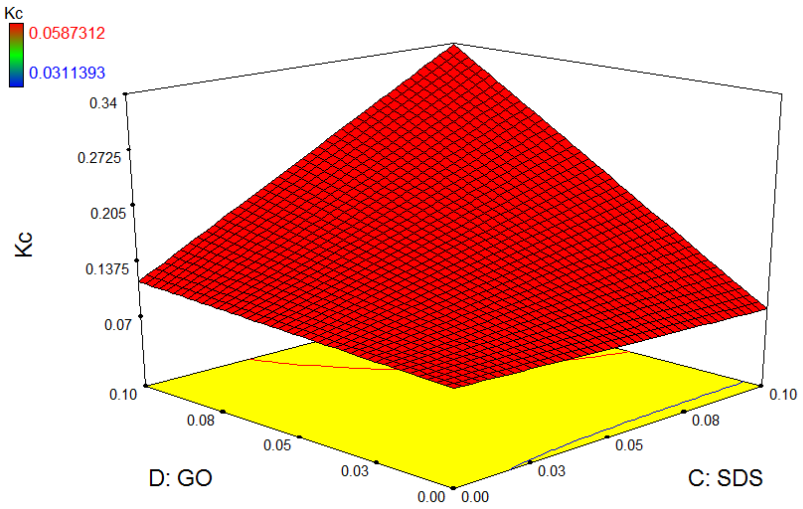

Fig. 11. The simultaneous interaction effects of the SDS concentration and GO concentration parameters on mass transfer coefficient of CO_2 absorption in the amine solvent.

4.2. Statistical analysis of the parameters influencing the molecular diffusion coefficient of carbon dioxide in the amine solvent

Table 3 shows the intermolecular diffusion coefficient calculated at different temperatures, pressures and concentrations of nanoparticles and surfactant for all of the experiments.

Table 3. Experimental results used to determine the molecular diffusion coefficient

Run	Factor 1 A:P in (bar)	Factor 2 B:T in (K)	Factor 3 C: SDS (g)	Factor 4 D : GO (g)	Response 2 D_{AB} (Cm^2/s)
1	18.6	295.15	0	0	2.5E-05
2	18.6	306.25	0	0.0375	0.000124
3	19	297.35	0	0.05	3.74E-05
4	20	298.75	0	0.075	0.000288
5	18.7	298.55	0	0.1	0.000143
6	18.9	299.05	0.0375	0	0.000272
7	18.9	300.75	0.05	0	4.78E-05
8	18.9	294.45	0.075	0	0.000106
9	18.8	299.45	0.1	0	0.000282
10	18.9	297.35	0.075	0.05	0.000185
11	18.6	297.15	0.05	0.05	9.51E-05

The statistical analysis yielded relation (15) for estimating the molecular diffusion coefficient of CO_2 in the amine solvent:

$$D_{AB}=1.743*10^{-3}-8.753*10^{-4}A+3.48*10^{-3}B+7.74*10^{-4}C+1.17*10^{-3}D-6.48*10^{-4}AB-2.7*10^{-3}AC-3.85*10^{-3}AD+1.88*10^{-3}BC+4.34*10^{-3}BD+8.27*10^{-4}CD \quad (16)$$

Figure 12 shows the changes in the molecular diffusion coefficient with changes in the reactor pressure. The molecular diffusion coefficient decreased with increased pressure.

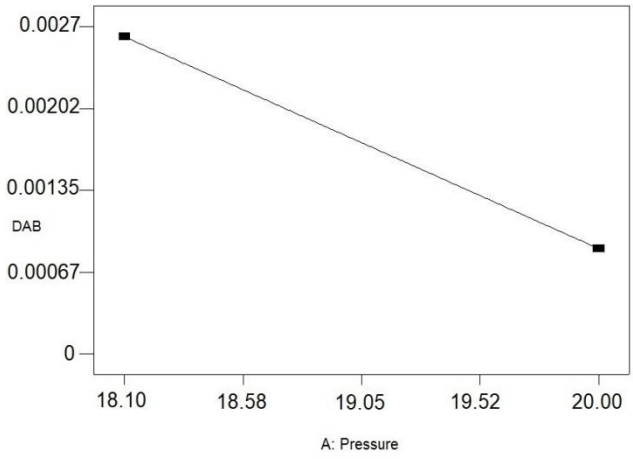

Fig. 12. The changes in the molecular diffusion coefficient with the reactor pressure.

Figure 13 shows the changes in the molecular diffusion coefficient of CO_2 in the amine solvent with changes in reactor temperature. The molecular diffusion coefficient significantly increased with an in temperature.

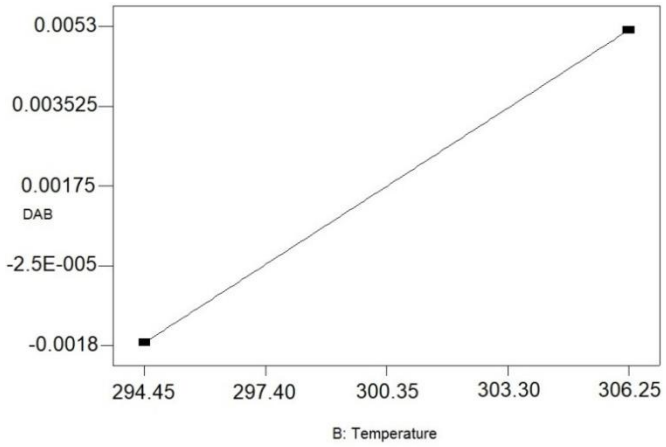

Fig. 13. The changes in the molecular diffusion coefficient with the reactor temperature.

Figure 14 shows the changes in the molecular diffusion coefficient of CO_2 in the amine solvent with changes in SDS concentration. The molecular diffusion coefficient increased with an increase in SDS concentration.

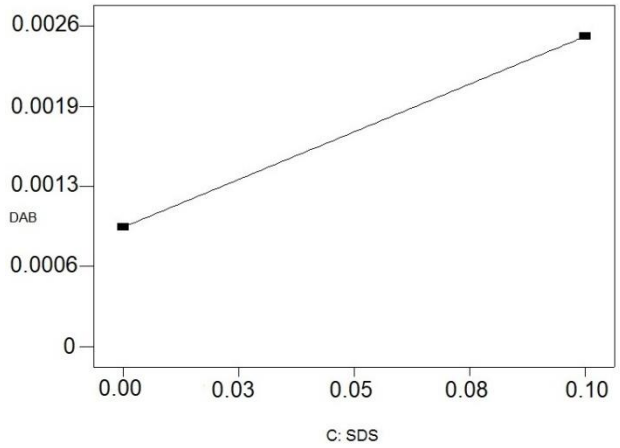

Fig. 14. The change in the molecular diffusion coefficient with SDS concentration.

Figure 15 shows the changes in the molecular diffusion coefficient of CO_2 in the amine solvent with changes in GO concentration. The molecular diffusion coefficient increased with an increase in the GO concentration.

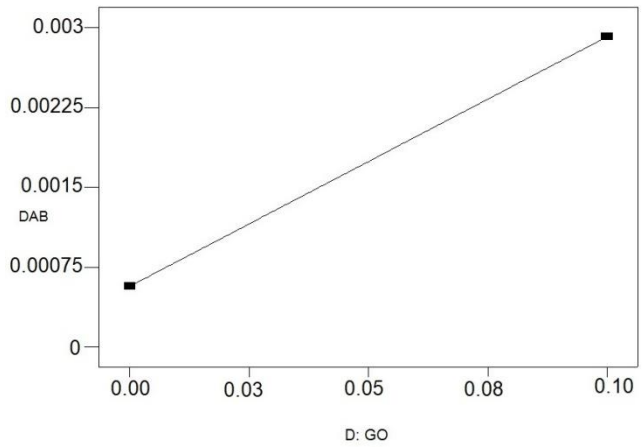

Fig. 15. The change in the molecular diffusion coefficient with GO concentration.

Figures 12 to 15 show the individual effects of the parameters influencing the molecular diffusion coefficient of CO_2 in the amine solvent regardless of their interactions. The interaction effects of the parameters are discussed below.

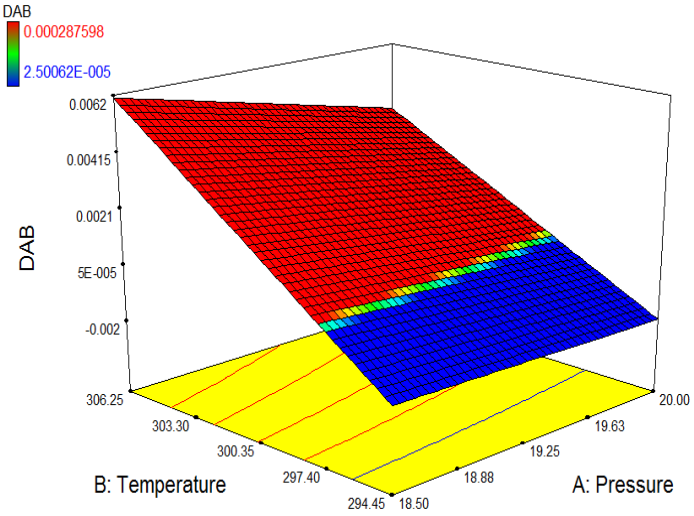

Fig. 16. The simultaneous interaction effects of reactor pressure and temperature parameters on the molecular diffusion coefficient of CO_2 in the amine solvent.

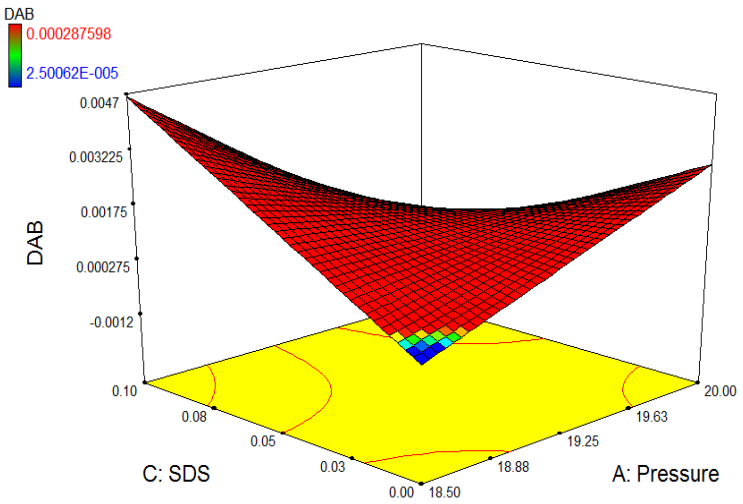

Fig. 17. The simultaneous interaction effects of the pressure and SDS concentration parameters on the molecular diffusion coefficient of CO_2 in the amine solvent.

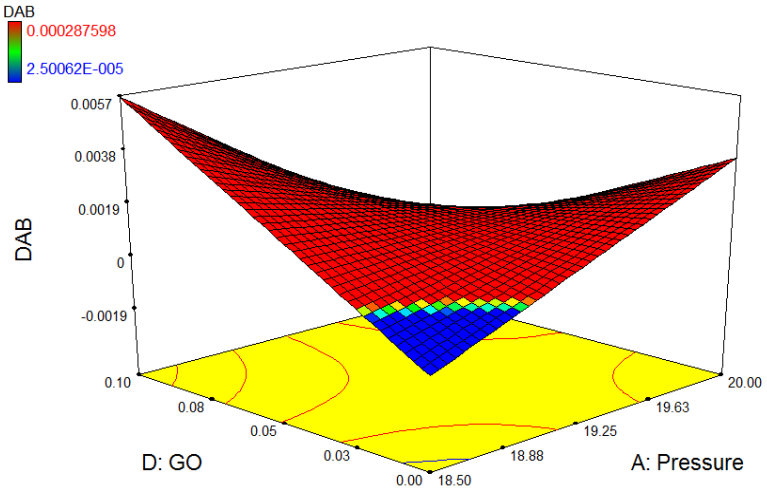

Fig. 18. The simultaneous interaction effects of the pressure and GO concentration parameters on the molecular diffusion coefficient of CO_2 in the amine solvent.

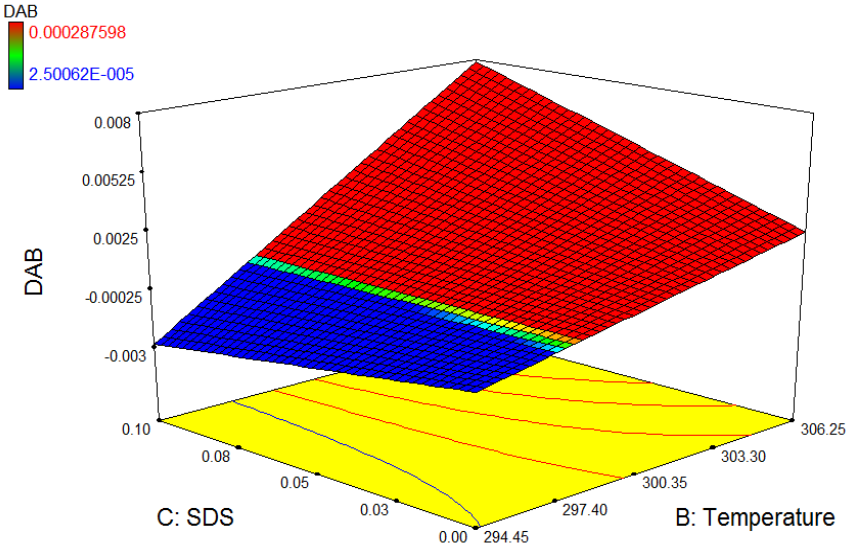

Fig. 19. The simultaneous interaction effects of the temperature and SDS concentration parameters on the molecular diffusion coefficient of CO_2 in the amine solvent.

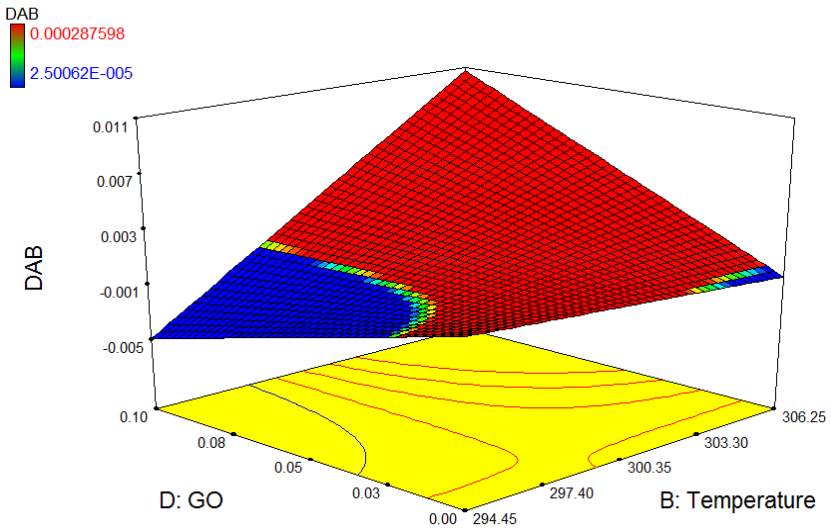

Fig. 20. The simultaneous interaction effects of the temperature and GO concentration parameters on the molecular diffusion coefficient of CO_2 in the amine solvent.

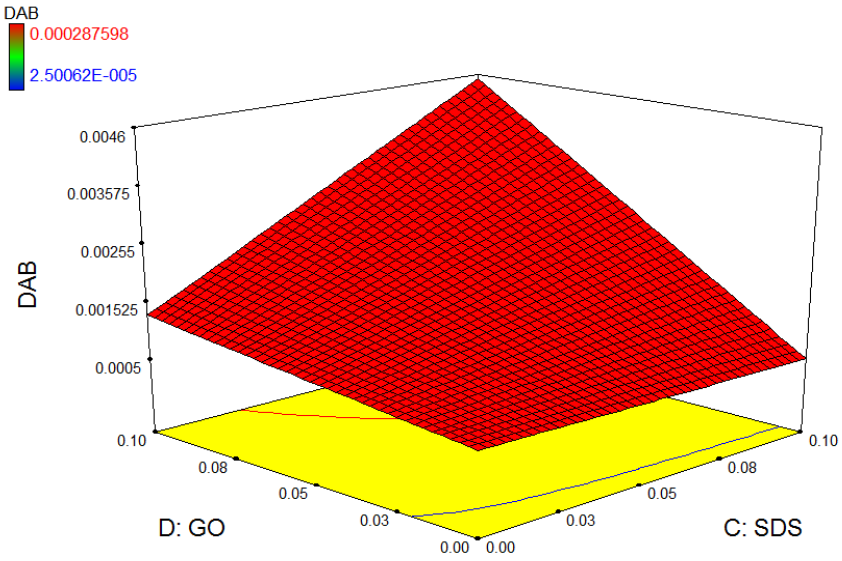

Fig. 21. The simultaneous interaction effects of the SDS and GO concentration parameters on the molecular diffusion coefficient of CO_2 in the amine solvent.

5. Conclusions

The results showed that the mass transfer coefficient of CO_2 absorption in the amine solvent increased with decreases in pressure and increases in temperature. The mass transfer coefficient of CO_2 absorption in the MEA solvent improved with increased SDS and GO concentrations. Among the effective parameters, pressure had the greatest effect on the values of the mass transfer coefficient. The laboratory results showed an increase in the molecular diffusion coefficient of CO_2 in the amine solvent with decreases in pressure and increases in temperature. The molecular diffusion of CO_2 in the amine solvent increased with an increase in SDS and GO concentrations. Among the effective parameters, pressure had the greatest effect on the molecular diffusion coefficient of CO_2 in the amine solvent. Using the experimental data, a statistical relationship was introduced to estimate the molecular diffusion coefficient. This relation was presented to predict the mass transfer coefficient of CO_2 absorption in the amine solvent.

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