Experimental and Modeling of CO_2 Absorption in a Bubble Column Using a Water-Based Nanofluid Containing Co-Doped SiO_2 Nanoparticles

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Abstract

In this study, we investigated the effect of Co/SiO_2 NPs on CO_2 absorption in the form of single bubbles rising through a bubble column (20 °C and 1 atm). Co-doped SiO_2 nanoparticles were first synthesized through the chemical vapor deposition (CVD) method, then nanofluids with different concentrations of the synthesized NPs (0.001, 0.01, 0.02, 0.05, and 0.1 wt.%) were prepared. Through comprehensive experimental studies, the effects of NPs concentration and nanofluid volume on the CO_2 absorption rate were examined. The stability of nanofluids, as a key factor in nanofluid efficiency, was investigated over a period of 10 days. Based on the experimental data, mass transfer operations were analyzed through dimensionless numbers (Sherwood (*Sh*), and Schmidt (*Sc*) numbers) to elaborate the rate of CO_2 diffusivity into the Co/SiO_2 nanofluid. Results showed that increasing NPs concentration from 0.001 to 0.02 wt.% caused the CO_2 absorption rate to reach a maximum value followed by a downward trend.

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Increasing nanofluid volume did not affect the gas absorption rate, which is attributed to the fact that the predominant mechanism of CO_2 absorption was the Brownian motion of NPs. Results confirmed that the prepared nanofluids had acceptable stability over the test period of 10 days, and the optimum nanofluid (80 mL with 0.02 wt.% of NPs) had the maximum CO_2 absorption, 28% more than the base fluid. The magnitude of the CO2 mass transfer coefficient in the nanofluid was found to be 1.953×10^{-4} ($m.s^{-1}$), which was 1.89 times higher than the base fluid. Finally, a comprehensive correlation (R^2= 0.99) was introduced to predict the CO_2 mass transfer coefficient in the Co/SiO_2 nanofluid.

Keywords: CO_2 absorption, Nanoparticle, Nanofluid, Bubble column

1. Introduction

Carbon dioxide (CO_2) absorption refers to capturing and storing carbon dioxide from the atmosphere or industrial processes. It is a crucial component of mitigating climate change, as CO_2 is a potent greenhouse gas that contributes to global warming (Åhlén et al. 2023; Zarepisheh, Binazadeh, and Esmaeilzadeh 2023; Heidari et al. 2023; Ayatollahi, Esmaeilzadeh, and Mowla 2021). During the last decades, many ways have been introduced to remove and absorb CO_2 from gas streams like using membranes (Jung, Lee, and Lee 2023; Hamalová et al. 2023; Fu et al. 2023), cryogenics (Y. Kim, Lee, et al. 2023; He et al. 2023; Meng et al. 2023), absorption (Gautam and Mondal 2023; Huhe, King, and Chuang 2023; Sharif et al. 2023; Zandahvifard et al. 2021), and adsorption (Skjervold et al. 2023; Al-Absi et al. 2023). Notably, the field of CO_2 capture has seen significant advancements, and absorption and adsorption methods have been developed to a practical stage. However, current literature indicates that absorption methods are more commonly employed in practical applications (Zhien Zhang, Borhani, and Olabi 2020; Ochedi et al. 2021; Fang et al. 2020; Rath et al. 2023; Gautam and Mondal 2023; Dziejarski, Krzyżyńska, and Andersson 2023; Zarepisheh, Binazadeh, and Esmaeilzadeh 2023). These references provide up-to-date insights into the prevailing trends in CO_2 capture, demonstrating the preference for absorption techniques due to their efficiency, cost-effectiveness, and widespread implementation. The absorption methods are divided into physical and chemical types of absorbents. Chemical absorption using amine series solutions is a mature and well-developed technology (Dutcher, Fan, and Russell 2015; Koytsoumpa, Bergins, and Kakaras 2018), but enormous costs for energy supply to regenerate solvent is a major drawback of this method (M. Wang et al. 2015; Raynal et al. 2011; Khan et al. 2023; Liu, Lu, and Wang 2023; Dziejarski, Krzyżyńska, and Andersson 2023). Degradation of amine absorbents, side reactions, and corrosion are some other problems (Ünveren et al. 2017; Wu et al. 2020; Elhambakhsh, Heidari, and Keshavarz 2022). While the physical absorption exhibits

a relatively low CO_2 recovery capacity, it proves to be more suitable than chemical methods for high-pressure systems like the integrated gasification combined cycle (IGCC) (Ban, Keong, and Mohd Shariff 2014; Zhien Zhang, Borhani, and Olabi 2020).

In the context of physical absorption for CO_2 capture, two vital factors come into play when selecting an ideal solvent: a high reaction rate and a high CO_2 absorption capacity (Mota-Martinez, Hallett, and Mac Dowell 2017). To enhance the physical absorption process, researchers have explored the use of nanofluids, stable colloidal solutions formed by dispersing nanoparticles homogeneously in a base fluid, typically water or another liquid. These nanoparticles, which can be a metallic, metal oxide, or carbon-based with dimensions ranging from 1 to 100 nanometers, significantly impact the properties of the base fluid, including thermal conductivity, viscosity, and absorption characteristics (Tavakoli et al. 2022; Zhien Zhang et al. 2018; J.S. Lee, Lee, and Kang 2015). Numerous attempts have been made to harness the potential of nanofluids as a suitable candidate for enhancing the physical absorption of CO_2 through mass transfer enhancement (Mohd Rozaiddin and Lau 2022; Zhien Zhang et al. 2018; Hussin et al. 2023). Adding nanoparticles to a base fluid offers several advantageous properties, including high surface area, reactivity, and the ability to form stable dispersions. Consequently, nanofluids have the potential to enhance the absorption of CO_2, ultimately improving the overall efficiency of the CO_2 absorption process and potentially reducing the energy requirements for CO_2 separation from other gases.

Over the past decades, many studies have been carried out to improve the mass transfer characteristics of nanofluids. For instance, a study by Kim et al. (W.-g. Kim et al. 2008) investigated CO_2 absorption experiments in a bubble-type absorber utilizing water-based nanofluids with SiO_2 nanoparticles. The results indicated that the inclusion of nanoparticles resulted in a notable 24% increase in the total CO_2 absorption. According to Jiang et al. (J.

Jiang et al. 2013), the introduction of nanoparticles in monoethanolamine (MEA) solution resulted in an improvement of up to 8% in the CO_2 absorption rate compared to the pure MEA solution. Pang et al. (Pang et al. 2012) conducted a study on NH_3/H_2O bubble absorption performance using Ag nanoparticles and observed that the inclusion of 0.02 wt.% Ag NPs in the nanofluid led to a significant enhancement of 55% in the absorption rate compared to the base fluid. In a separate investigation, Pineda et al. (Pineda et al. 2012) examined CO_2 absorption in a tray column absorber and reported maximum absorption rate enhancements of 9.4% for Al_2O_3 nanoparticles and 9.7% for SiO_2 nanoparticles. Researchers (Zarei and Keshavarz ; Lashgarinejad et al. 2023) have made a significant discovery that silica nanoparticles exhibit a remarkable increase in CO_2 absorption efficiency at low temperatures. Amaris et al. (Amaris, Bourouis, and Vallès 2014) conducted a study investigating the impact of carbon nanotubes (CNTs) on the $NH_3/LiNO_3$ absorber's performance. They reported that the presence of CNTs resulted in a maximum enhancement of absorption performance, reaching 1.64 times at 40°C and 1.48 times at 35°C for the cooling water. Lee et al. (J.W. Lee et al. 2016) examined a bubble absorber's CO_2 absorption performance enhancement. According to their findings, the addition of 0.01 vol.% of Al_2O_3 nanofluid at 20 °C resulted in an enhancement of up to 4.5% in the CO_2 absorption rate. Similarly, the inclusion of 0.01 vol.% of SiO_2 nanofluid at 20 °C led to a 5.6% increase in the CO_2 absorption rate. Lee and Kang (J.W. Lee and Kang 2013) investigated enhancement in CO_2 absorption of NaCl aqueous solution-based Al_2O_3 nanofluid. They measured the CO_2 solubility in Al_2O_3/NaCl nanofluid for different Al_2O_3 concentrations and solvent temperatures. The results showed that the CO_2 solubility enhancement ratios at 0.01 vol.% of Al_2O_3 nanoparticle concentration, were 11.0% at 30 °C, 12.5% at 20 °C, and 8.7% at 10 °C. Conversely, other studies (Valeh-e-Sheyda and Afshari 2019; J.W. Lee and Kang 2013; H. Xu et al. 2013) have highlighted that high temperatures can lead to the agglomeration of silica nanoparticles. As a result, a decision was

made to maintain the absorber column temperature at 20 ºC to strike the right balance and optimize CO_2 absorption while mitigating agglomeration issues.

Although several studies have shown that nanofluids can improve the performance of CO_2 absorption compared to conventional fluids, the use of nanofluids for CO_2 absorption is still in the experimental stage, and more research is needed to fully understand their potential. One advantage of the nanofluids is attributed to the high surface area of the nanoparticles, which provides more contact area between the CO_2 and the fluid. This leads to faster reaction rates and higher CO_2 absorption capacity. Furthermore, nanoparticles have the potential to augment the mass transfer coefficient, facilitating a higher rate of CO_2 transfer from the gas phase to the liquid phase. But nanoparticle concentration, nanoparticle size and shape, nanoparticle surface chemistry, fluid properties, temperature, and pressure are deciding factors in using nanofluids for CO_2 absorption. The effectiveness of nanoparticles can be limited by issues such as aggregation and sedimentation.

Computational models can be used to simulate the absorption of CO_2 using nanofluids. The computational models for simulating CO_2 absorption using nanofluids typically involve solving the mass and energy balances for the system. The equations are solved numerically using a variety of techniques such as finite element analysis, finite difference methods, or computational fluid dynamics. These models offer the capability to forecast the performance of various nanofluids concerning CO_2 absorption under diverse conditions, encompassing temperature, pressure, and concentration. Moreover, the models provide valuable insights for optimizing the design and operation of absorption systems that utilize nanofluids. Several crucial factors must be taken into account in computational models for CO_2 absorption using nanofluids. These factors include the nanoparticle concentration and size, the characteristics of the base fluid, system temperature, pressure, and the mass transfer coefficient between the gas and liquid phases. Until now, many modeling studies have been accomplished on CO_2

absorption (J.-z. Jiang, Liu, and Sun 2017; Ansarian and Beiki 2022; Sodeifian and Niazi 2021). Jamali et al. (Jamali and Azari 2023), reviewed the numerical computational fluid dynamics (CFD)simulation of CO_2 absorption columns and investigated the application of different nanoparticles in various amine-based solutions and the effect of different packings in the packed bed absorption columns. Rashidi et al. (Rashidi and Mamivand 2022), examined the effect of temperature, volume fraction of NPs, and flow rate of nanofluids on the mass transfer coefficient for CO_2 absorption using Al_2O_3-water nanofluid and concluded that the rising temperature, nanoparticle concentration, and fluid flow rate favors the mass transfer coefficient. Generally, the modeling of carbon dioxide absorption can suffer from a few weaknesses such as assumptions, incomplete data, simplifications, and model validation, which may lead to inaccurate predictions or results.

In this study, SiO_2 nanoparticles were first synthesized, followed by the doping of Co onto them. Next, their performance on CO_2 absorption was investigated at 20 °C, which was a low and challenging temperature. In this regard, several concentrations of nanoparticles and volumes of the nanofluid were prepared and finally, the nanoparticle dispersion stability was evaluated by measuring the total CO_2 absorption over a period of ten days. It also tried to predict the absorption rate and mass transfer coefficient by presenting a new comprehensive correlation. To the best of our knowledge, this is the first investigation of the absorption rate, molar flux, mass transfer coefficient, and diffusivity coefficient of CO_2 into a Co/SiO_2 water-based nanofluid in a single bubbles column through a set of comprehensive experiments and precious correlation.

2. Experiments

2.1 Materials and instrumentations

Tetra epoxy silane (TEOS, 95%) and Bis(cyclopentadienyl)cobalt(II) (Cobaltocene, 98%) were purchased from Merck Company, Germany, and used to synthesize Co/SiO_2 NPs. Deionized water was used to prepare and dilute nanofluids. All chemical materials were used without more purification.

The transmission electron microcopy (TEM) was performed to assess the size distribution of dry nanoparticles while the dynamic light scattering (DLS) was accomplished to investigate the size distribution of dispersed nanoparticles in deionized water. The size of nanoparticles and their agglomeration were characterized using TEM images, taken from Hitachi, 9000 NA, Japan (Andrade et al. 2012). To conduct TEM tests, a dispersed suspension of NPs in ethanol (0.001 wt. %) was sonicated using an ultrasonic bath, specifically the Parsonic 30S-400W operating at 28 kHz, for a duration of 20 minutes, followed by placing on the graphite surface. Then, to remove ethanol from the samples, the samples were placed into a vacuum oven for 4 h. DLS measurements were performed by Malvern, Zeta Sizer Nano ZS, United Kingdom (G. Xu, Zhang, and Song 2003; Pham, Fullston, and Sagoe-Crentsil 2007). The zeta potential (ξ-potential) tests were accomplished using ELSZ-2000 (Otsuka Electronics Co., Osaka, Japan) to measure the stability and surficial electrostatic charges of NPs (Darvanjooghi and Esfahany 2016). The ξ-potential was considered to explain the electrostatic charges of NPs, which resulted in repulsive forces among the dispersed particles. NPs' stability is known based on the positive and negative high ξ-potential, whereas low ξ-potentials indicate the tendency of NPS to agglomeration (Davoodi et al. 2016). A mass flow controller instrument (MFC, Brooks model, 1-888-554-flow, USA) was employed to inject CO_2 into the nanofluids through the absorption setup. A CO_2 sensor (Testo 535, Germany) was used to measure CO_2 concentration in the outlet gases. To prepare nanofluids, a certain amount of the synthesized Co/SiO_2 NPs were measured using a precise electric balance (TR 120 SNOWREX, Taiwan) and added to the water. The pH of the solutions was measured using a pH meter (PCE-PHD 1, UK). To

prevent agglomeration of NPs, an ultrasonic processor (QSONICA-Q700, NY, USA) was utilized. A mechanical ball-mill (YKM-2L, China) was used to grind the clustered NPs. A magnetic stirrer (IKA-10038, Germany) was used to stirrer the solutions.

2.2 Methods

2.2.1 Synthesis of Co/SiO_2 NPs

The Co/SiO_2 nanoparticles were synthesized using the chemical vapor deposition (CVD) method, following the methodology outlined by Dev et al. (Dev, Sardoiwala, and Karmakar 2021). Tetra epoxy silane was chosen as the precursor for SiO_2 nanoparticles. Initially, 0.05 g of the tetra epoxy silane reagent was completely dispersed in 50 mL of acetone. To enhance CO_2 absorption and facilitate the formation of Co/SiO_2 NPs, 12.0 wt.% of cobaltocene was introduced into the acetone medium. The activation energy required to initiate the process was found to be achieved at a high temperature, typically ranging from ~600 to 1100 °C, as reported by Demirci (Demirci 2022). Therefore, the prepared solution was transferred into the tubing furnace and set to 800 °C. This temperature enables the decomposition and subsequent deposition of the precursor, allowing the nanoparticles to be generated effectively. To ensure the success of the CVD process, the reaction furnace was heated gradually at a rate of 5 °C·min^{-1}, a temperature profile commonly used in such syntheses (Gulino et al. 2005; H. Wang et al. 2022; Zailei Zhang et al. 2013; H. Kim, Shin, et al. 2023). The nano-sized particles were allowed to grow and form within the furnace for 2 hours, as determined by preliminary experimentation and optimization studies (Dev, Sardoiwala, and Karmakar 2021; Soltys et al. 2023; Duc Vu Quyen et al. 2019; Podrezova et al. 2013). The chosen duration allowed for the desired reaction kinetics and the appropriate growth of Co/SiO_2 nanoparticles.

2.2.2 Preparation of nanofluid

First, the nanoparticles were placed (4 h) in a ball mill device for the separation of the accumulated NPs. Then, nanofluids were prepared by dispersing a specific amount of the synthesized Co/SiO_2 NPs in 80 mL of deionized water. A vast range of NPs concentrations (0.001, 0.01, 0.02, 0.05, and 0.1 wt.%) were examined in this study. Next, the prepared suspensions were stirred at 800 rpm for 5 h. Finally, the sonication process dispersed NPs in the nanofluid through three series of 20 min. the cycle time and amplitude of the sonication process were set on 0.5 s and 70%, respectively.

2.3 Experimental setup

The experimental setup was consisted of a bubble absorber column, filled with the synthesized Co/SiO_2 NPs loaded in the nanofluid. In the experimentation process, the nanofluid absorption column was subjected to sequential injections of N_2 and CO_2, using specific volumes (20, 25, 30, 35, 40, 45, and 50 mL). The setup for this involved a 170 mm height and 60 mm diameter Poly(methyl methacrylate) tube, depicted in Figure 1, which served as a semi-batch device to investigate CO_2 absorption through the nanofluid. To control the rate of feed gases, two regulators were placed in the outlet of the CO_2 and N_2 accumulators. In each experiment, CO_2 was introduced into the nanofluids within the absorber column using a constant flow rate of 80 standard cubic centimeters per minute (SCCM). To monitor the gas flow rates accurately, two mass flow controllers (MFC) were employed at the inlets of the absorber column. The duration of bubble rise was recorded using an LTD digital chronometer, which boasted an accuracy of ±0.01 seconds. The rising time of the bubbles was almost 2.3 s. The diffuser was meticulously designed and fabricated using cylindrical acrylic material. The top plate of the spherical diffuser featured multiple punched orifices, each with a diameter of 7 mm. The top plate itself had a thickness of 0.3 cm and a diameter of 6 cm. In total, there were 70 orifices strategically distributed across the top plate. The designed spherical gas diffuser was embedded at the bottom of the absorber column to produce bubble sizes ranging from 6.9 to 7 mm. The

concentration of CO_2 is accurately measured and recorded by the CO_2 sensor at the outlet of the absorber column in equal time intervals. The temperature of the absorbent was automatically monitored and controlled during the absorption process using a set of thermocouples and thermostats.

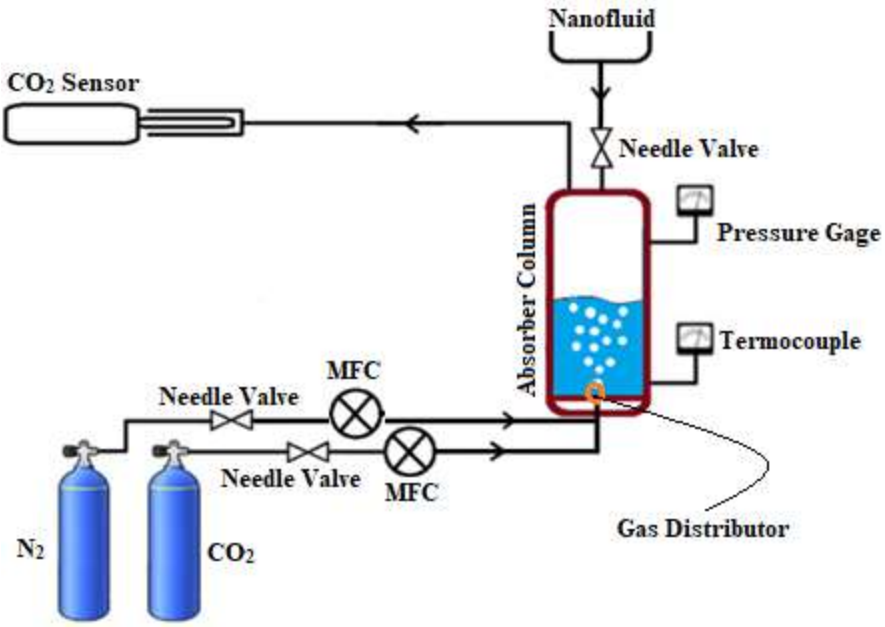


Fig. 1. Schematic diagram of the experimental setup.

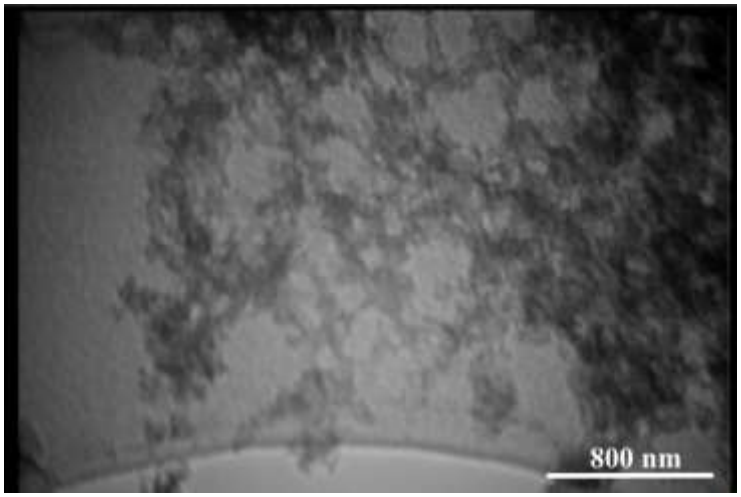
2.4 Experimental procedure

To measure the CO_2 absorption in the prepared nanofluid, first, pure N_2 was injected into the absorber column for 1 min. Next, 80 mL of the prepared nanofluid was injected to the absorber column. The temperature of the absorber column was adjusted to 20 °C (atmospheric pressure). N_2 was reinjected again for 1 min to purge CO_2 from the nanofluid. CO_2 was injected into the nanofluid at 80 SCCM. CO_2 concentration in the outlet gas stream was recorded (ppm) every 3 s for 27 minutes. By knowing the concentration of injected carbon dioxide and applying the ideal gas equation, the number of moles of absorbed carbon dioxide was calculated.

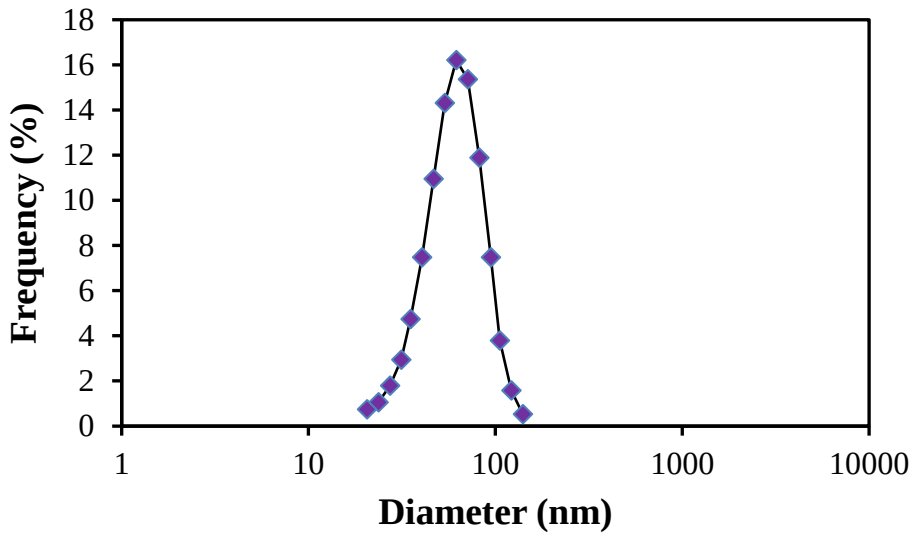
3. Results and discussion

3.1. Nanofluid characterization

The average size of NPs was determined using dynamic light scattering (DLS) and transmission electron microscopy (TEM). Figures 2 (a & b) illustrate the TEM (a) and DLS (b) images of Co/SiO_2 NPs used for the nanofluid preparation. The analysis revealed that the average size of the synthesized nanostructures was 70 nm with a semi-spherical morphology without any special agglomeration (Shi et al. 2008).



(a)



(b)

Fig.2. (a)Transmission electron microcopy (TEM) and (b) dynamic light scattering (DLS) images of Co/SiO_2 nanoparticles.

DLS is a technique that measures the hydrodynamic size of particles in a liquid suspension by analyzing the fluctuations in scattered light caused by their Brownian motion. It provides

valuable insights into the size distribution and stability of nanoparticles in solution. In our study, the DLS test confirmed that the average size of the NPs (70 nm) was consistent with the size obtained from the TEM test. This finding suggests that there was no significant agglomeration or clustering of NPs during their dispersion in the base fluid (deionized water). Therefore, the DLS results support the conclusion that the NPs remained well-dispersed and did not form larger aggregates, which is crucial for ensuring their effective performance in the intended application. Therefore, results confirmed that the method used in this study for NPs dispersion led to a well-dispersion of NPs.

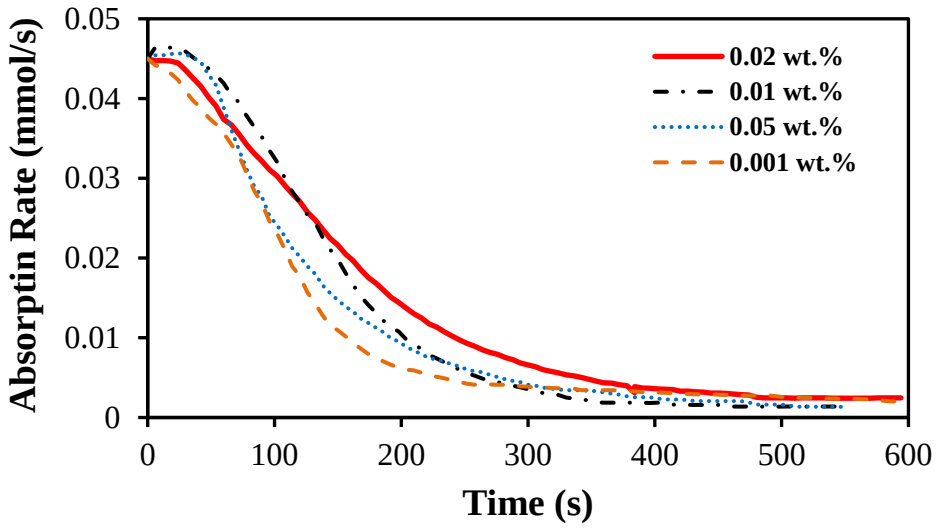
Zeta-potential analysis was accomplished to evaluate the NPs' stability in the nanofluid (áO'Brien 1990). Since the value of the ξ-Potential indicates the magnitude of the electrostatic repulsion among particles with similar charges and the synthesized Co/SiO_2 NPs had a high ξ-Potential value of –98.7 mV. Since the ξ-Potential of the synthesized NPs was lower than –45 mV, it was confirmed that the synthesized NPs benefited from high stability (W.-g. Kim et al. 2008).

3.2 Absorption

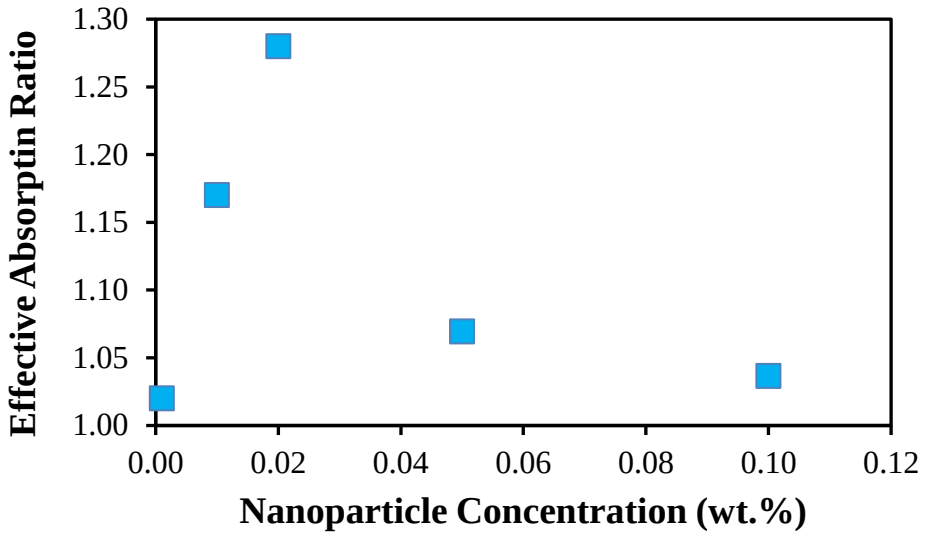
3.2.1 Maximum absorption

Nanofluids were prepared with different NPs concentrations including 0.001, 0.01, 0.02, 0.05, and 0.1wt.% of NPs. The CO_2 absorption was measured over a 27 min period. The experiments were repeated three times at each volume fraction of the synthesized NPs and the standard deviations were shown as the error bars. In Figures 3(a) and 3(b), the CO_2 absorption rate into the Co/SiO_2 nanofluid and the corresponding effective absorption ratio were presented, respectively. To determine the CO_2 absorption rate, the calculated absorbed CO_2 (in mol) was divided by the time interval (3 seconds). For the effective absorption ratio, the equilibrium CO_2 absorption in each nanofluid was divided by its equilibrium absorption in the base fluid. Based on the results presented in Figures 3 (a & b), the average CO_2 absorption increased with

increasing Co/SiO_2 NPs from 0.001 to 0.02 wt.% while the absorption decreased for higher NPs loadings (0.02 to 0.1 wt.%). It can be concluded that the CO_2 absorption molar flux reached a peak value of 0.02 wt.% of the synthesized NPs. The Co/SiO2 NPs were observed to enhance micro-convection and improve the mass transfer rate compared to the base fluid. This explained the initial increase in CO_2 absorption with the aforementioned NPs mass fraction. However, as the number of NPs increased, the viscosity of the nanofluids also increased, leading to a dominant effect of NPs on micro-convection, which, in turn, reduced CO_2 absorption within the nanofluid (Esmaeili Faraj et al. 2014; Darvanjooghi and Esfahany 2016).



(a)



(b)

Fig. 3. (a) Absorption rate versus time and, (b) Effective absorption ratio of CO_2 versus mass fraction of Co/SiO_2 NPs.

Furthermore, Figure 4 affirms the higher amount of CO_2 absorption in nanofluids including 0.02 wt.% of Co/SiO_2 NPs than in deionized water. It was found that the maximum CO_2 absorption enhancement in comparison to pure water is 28% with nanofluid containing 0.02 wt.% of Co/SiO_2 NPs. The high surface area and porous structure of Co/SiO_2 nanoparticles provide ample sites for CO_2 molecules to adsorb onto the nanoparticle surface physically. This physical absorption increases the concentration of CO_2 near the liquid phase, facilitating the dissolution of CO_2 into the nanofluid. The surface functional groups or active sites of Co/SiO_2 nanoparticles chemically interact with CO_2. These chemical interactions could lead to the formation of soluble carbonate or bicarbonate species, increasing the overall CO_2 absorption capacity of the nanofluid. Besides, the nanoscale confinement between Co/SiO_2 nanoparticles in the nanofluid might alter the physical properties of the fluid, affecting the solubility of CO_2 within this confined space. This confinement effect could enhance CO_2 absorption compared to bulk water. The presence of nanoparticles can improve mass transfer rates between the CO_2 gas phase and the nanofluid's liquid phase. This enhancement accelerates the absorption

process and results in a more efficient uptake of CO_2 by the nanofluid. Moreover, the combination of physical and chemical interactions between CO_2 and Co/SiO_2 nanoparticles, along with the presence of water as the base fluid, creates synergistic effects that contribute to the overall enhanced CO_2 absorption observed in the nanofluid.

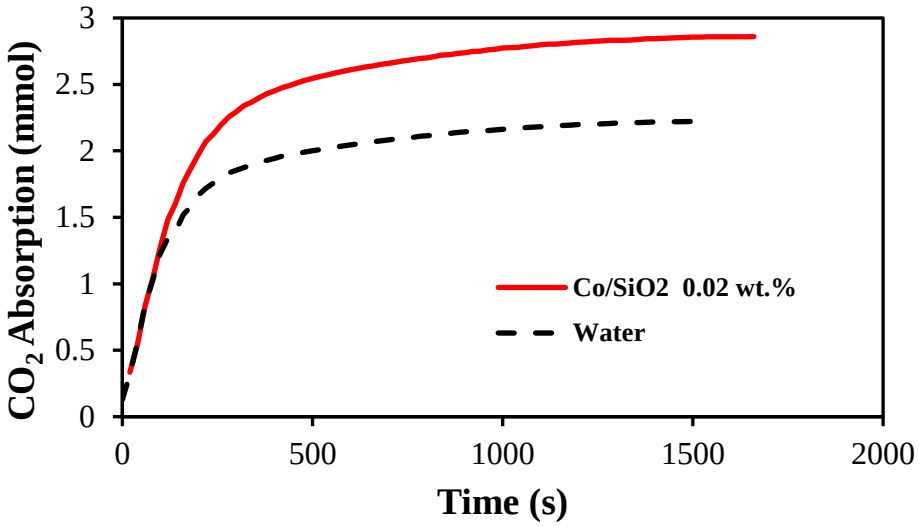
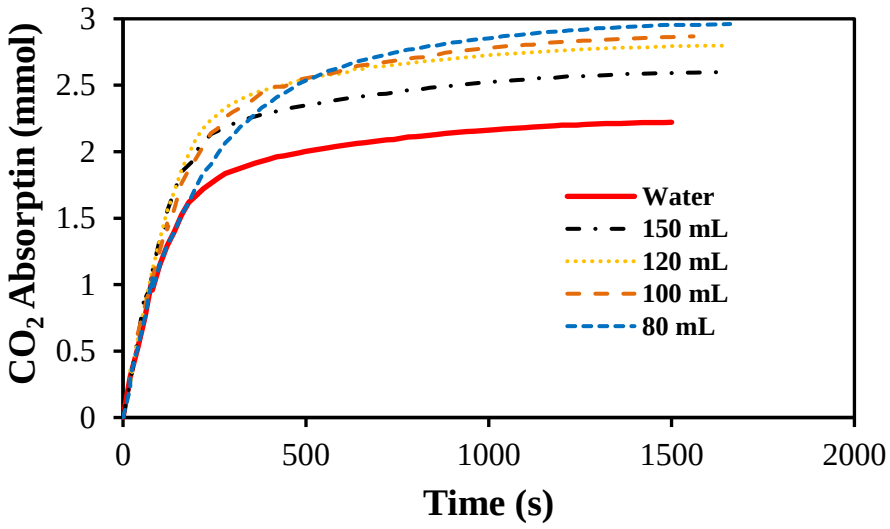


Fig. 4. CO_2 absorption for the nanofluid and the base fluid (deionized water) versus time.

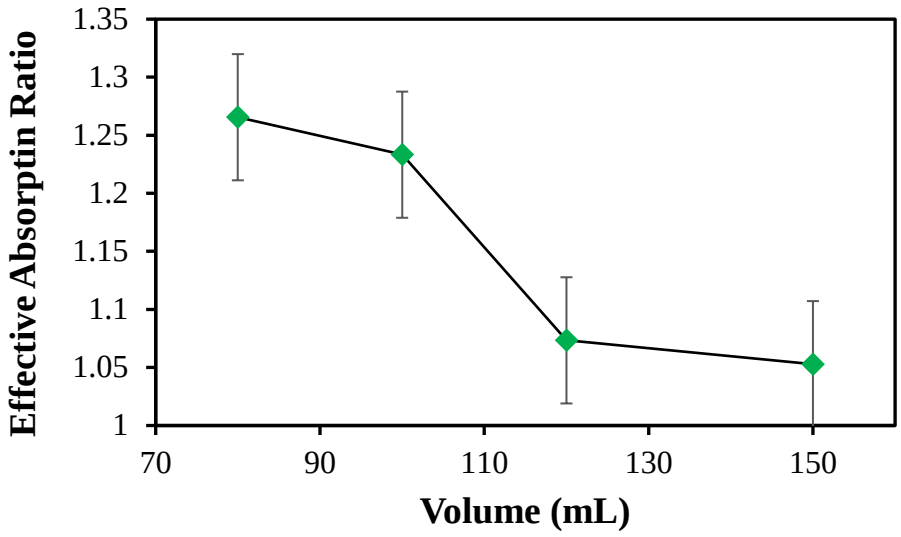
3.2.2 Probing the rate of mass transfer

To investigate the impact of absorbent volume on the CO_2 absorption rate, a single nanofluid solution with a fixed concentration of 0.02 wt.% was utilized. This nanofluid solution, containing the synthesized nanoparticles, was prepared at the optimum mass fraction and maintained at a temperature of 25 °C. Subsequently, four different volumes (80, 100, 120, and 150 mL) of this nanofluid solution were evaluated. By varying the volume while maintaining the concentration constant, it was tried to examine how changes in the absorbent volume affected the rate of CO_2 absorption. Findings (Figure 5) revealed that the absorption rate and mass transfer flux decreased with the enhancement in the volume loading of nanofluid. It can be attributed to the high height of absorbent in the vessel at a higher volume of nanofluid. The presence of forced and natural convection in the nanofluid within the specified vessel geometry

results in a broader diffusion regime when the height of the absorbent is increased, compared to a smaller height. In other words, a higher absorbent height facilitates enhanced diffusion of the nanofluid, leading to a wider region of dispersal. Since the Brownian motion of NPs and CO_2/water mixing flow are two important factors affecting CO_2 absorption, it was found that the effect of nanoparticles becomes weaker by forced or natural convection in a good mixing condition. As a result, the effect of the Brownian motion of NPs is in the reverse relationship with the height of absorbent, which is in good agreement with other findings (Samadi, Haghshenasfard, and Moheb 2014).



(a)



(b)

Fig. 5. (a) CO_2 absorption versus time (b) Effective absorption ratio versus volume of nanofluid.

3.2.3 Absorption stability

The stability of nanofluid suspension is an important parameter, in determining the success of the absorption process. The suspended nanoparticles in the adsorbent experience both intermolecular repulsive forces due to the Coulomb force and attractive forces resulting from the van der Waals interactions. Under this condition, nanoparticles may agglomerate, grow in size, and form greater clusters. It is highly likely that heavy clusters sediment due to gravitational force. Since the main reason for the mass transfer enhancement of nanofluid is the presence of NPs, their sedimentation causes the absorption efficiency to reduce. To assess the stability of the Co/SiO_2 nanofluid in this investigation, nanofluid was formulated using the optimal conditions, i.e., 80 mL of nanofluid with 0.02 wt.% of nanoparticles (NPs) at an absorbent temperature of 20 °C. The stability of the nanofluid was evaluated by monitoring the variations in equilibrium CO_2 absorption over a span of 10 days. Each day, the nanofluid was subjected to a 5-minute sonication and then measured the equilibrium CO_2 absorption over a 27-minute duration. Following this, the setup was left undisturbed for 24 hours, and the subsequent day, the equilibrium CO_2 absorption was calculated once more after 27 minutes.

Based on the results shown in Figure 6, the cumulative absorption of CO_2 versus time decreases with almost a constant slope. The constant slope confirms the saturation of the nanofluid with CO_2 with no agglomeration process inside the nanofluid over the period of 10 days. Therefore, it can be concluded that the prepared Co/SiO_2 nanofluid is an acceptably stable absorbent.

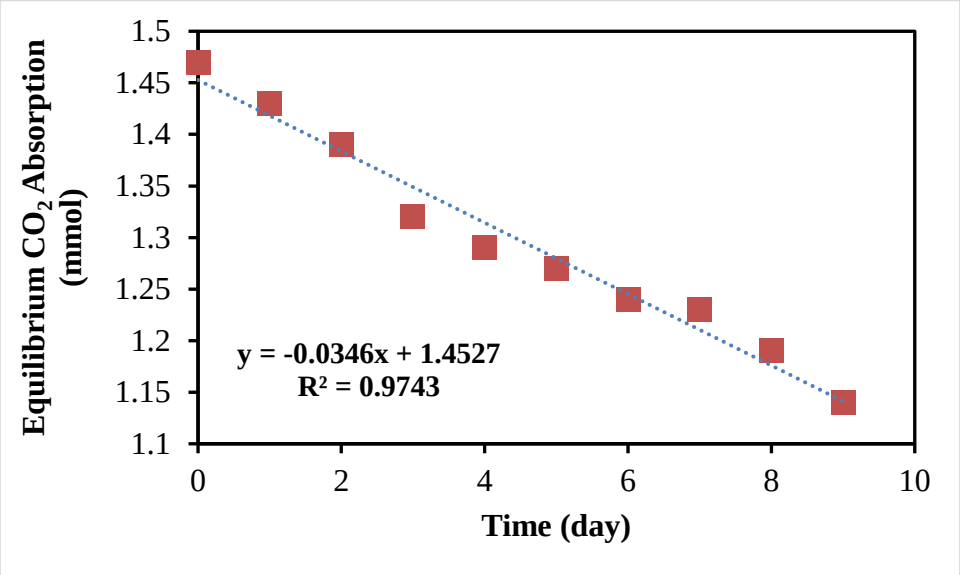


Fig. 6. Equilibrium CO_2 absorption over time.

3.2.4 Mass transfer coefficient

To determine the mass transfer coefficient, experiments were conducted by injecting different volumes of CO_2 gas (20, 25, 30, 35, 40, 45, and 50 mL) separately into the absorber column. Subsequently, the CO_2 concentration and its molar flux were measured. Figure 7 depicts the relationship between the average molar flux and the dissolved concentration of CO_2 in the liquid bulk. The results clearly indicated that increasing the CO_2 bulk concentration led to a reduction in the average molar flux due to a decrease in the mass transfer driving force. This decrease followed a linear pattern. To assess this linear trend, the principal mass transfer equation (Equation 1) was utilized and the experimental values were fitted to it (Darvanjooghi, Esfahany, and Esmaeili-Faraj 2018). This equation does not pertain to an adsorption model specifically; rather, it serves as a mass transfer equation for CO_2 absorption in a silica-water

nanofluid. It establishes a relationship between the average mass transfer rate (N_{avg}) and the mass transfer coefficient (K_l) in the liquid phase, considering the disparity between the bulk concentration of CO_2 (C_{CO2}) and the concentration of CO_2 at the gas-liquid interface (C^*_{CO2}).

The equation is grounded on certain assumptions: (1) The mass transfer process adheres to first-order kinetics, assuming that the concentration of CO_2 in the bulk phase (C_{CO2}) surpasses that at the gas-liquid interface (C^*_{CO2}). (2) The mass transfer coefficient (K_l) governs the rate at which CO_2 is transferred from the gas phase to the liquid phase and is influenced by various factors such as nanoparticle size, temperature, and fluid properties. (3) The model assumes steady-state conditions and disregards any potential reactions or intricate interactions between CO_2 and the silica-water nanofluid (Darvanjooghi, Esfahany, and Esmaeili-Faraj 2018).

$$N_{avg} = K_l(C^*_{CO2} - C_{CO2}) \quad (1)$$

where K_l represents the mass transfer coefficient ($m.s^{-1}$) at the liquid phase. C_{CO2} ($mol.m^{-3}$) and C^*_{CO2} ($mol.m^{-3}$) show the bulk and gas-liquid interface concentration of CO_2, respectively. It is worth mentioning that the observed CO_2 concentration in the interface was accounted for by extrapolating the line fitted on the obtained experimental data. It raises from the assumption of linear pattern for gas concentration and molar flux. The results from Figure 7 demonstrated that the proposed model exhibited a satisfactory fit to the experimental data, with an R^2 value of 0.9855. This high R^2 value indicates the regression's excellent accuracy and the model's minimal deviation from the experimental data.

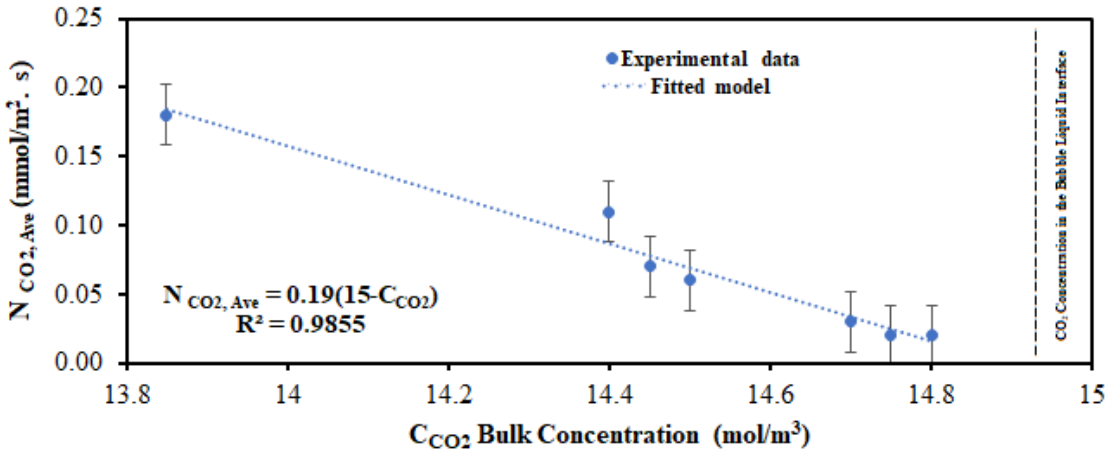


Fig. 7. Average molar flux versus CO_2 bulk concentration.

It can be concluded that the dashed line diagram (vertical plot) illustrates the observed concentration of CO_2 at the liquid-bubble interface. Additionally, the diagonal plot displaying the average molar flux versus the bulk concentration of CO_2 depicts the gas absorption operating line. According to the slope of the operating line in Figure 7, the relative mass transfer coefficient for CO_2 absorption (K_l) using Co/SiO_2 nanofluid was found to be $1.953 * 10^{-4}$ ($m.s^{-1}$), which was 1.89 times more than that for the water alone (relative mass transfer coefficient). The CO_2 absorption using nanofluid indicated higher values for relative gas concentration and relative mass transfer coefficient at the liquid-bubble interface.

3.3 Diffusivity Coefficient

Generally, the diffusivity of gases into a liquid has a significant impact on both the rate of gas absorption and the magnitude of the mass transfer coefficient. In this study, it was tried to benefit from Equation 2 to obtain CO_2 diffusivity into the Co/SiO_2 nanofluid. Equation 2 presents rising single bubbles within a fluid according to Dankwert`s theory (Esmaeili-Faraj and Nasr Esfahany 2016; Zhao et al. 2003).

$$N_{ave} = \frac{D \sinh\left(\delta\sqrt{\frac{s}{D}}\right) + D\, r_0\sqrt{\frac{s}{D}}\cosh\left(\delta\sqrt{\frac{s}{D}}\right)}{r_0 \sinh\left(\delta\sqrt{\frac{s}{D}}\right)} \left(C_{CO2,i} - C_{CO2}\right) \quad (2)$$

where the main factors of the model affecting the rate of mass transfer are the diffusivity of gas within a liquid (D), the thickness of the diffusion layer (δ), the radius of bubbles (r_0), and the rate of surface renewal (s). N_{ave} represents the CO_2 molar flux ($mol.m^{-2}s^{-1}$) while C_{CO2} and $C_{CO2,i}$ are the CO_2 concentration through the liquid bulk and at the interface of liquid-bubble ($mol.m^{-3}$), respectively.

Comparing Equations 1 and 2 results in Equation 3, showing the mass transfer coefficient of a gas within a liquid based on a single-bubbles model:

$$K_l = \frac{D \sinh\left(\delta\sqrt{\frac{s}{D}}\right) + D\, r_0\sqrt{\frac{s}{D}}\cosh\left(\delta\sqrt{\frac{s}{D}}\right)}{r_0 \sinh\left(\delta\sqrt{\frac{s}{D}}\right)} \quad (3)$$

In this study, Equation 3 was used to estimate the CO_2 diffusivity through the nanofluid. Based on Darvanjooghi et al. report, important factors of Equation 3 like D, s, and δ violently depend on the size of NPs in the nanofluid (Darvanjooghi, Esfahany, and Esmaeili-Faraj 2018). They mentioned that the range of NPs` size was between 50 to 70 nm, the rate of surface renewal (s) was 6.85 $mm.s^{-1}$, and the thickness of the diffusion layer was 0.201 mm. In this study, on the one hand, the average size of nanoparticles was 70 nm, and on the other hand, the values of s and δ were assumed to be constant during the CO_2 absorption, depending on only the mean diameter of NPs. Besides, the mass transfer coefficient of CO_2 within the nanofluid was previously calculated. Therefore, Equation 3 can be simplified to Equation 4 as follows:

$$F(s,\delta,D) = \exp\left(2\delta\sqrt{\frac{s}{D}}\right) \pm \frac{D - r_0\sqrt{s.D} - r_0 K_l}{r_0\sqrt{s.D} - r_0 K_l} = 0 \quad (4)$$

where s and δ were considered to be 6.85 and 0.201, respectively (Darvanjooghi, Esfahany, and Esmaeili-Faraj 2018). Equation 4 was solved according to the Newton-Raphson method as follows:

$$D_{n+1} = D_n - \frac{F\ (s, \delta, D_n)}{\frac{\partial F(s, \delta, D_n)}{\partial D_n}} \quad n = 0, 1, 2, \ldots \tag{5}$$

where $\frac{\partial F(s,\delta,D_n)}{\partial D_n}$ can be calculated based on the partial derivation of Equation 4. Besides, the initial value of D_0 was considered to be 10^{-10}.

Results illustrated that the diffusion values of CO_2 into Co/SiO_2 nanofluid and base fluid were $5.86 * 10^{-9}$ and $2.12 * 10^{-9}$ $m^2.s^{-1}$, respectively.

Previous studies confirmed that only Brownian micro-convection and grazing effect are predominate mechanism for gas absorption into a nanofluid (Darvanjooghi, Esfahany, and Esmaeili-Faraj 2018; Esmaeili-Faraj and Nasr Esfahany 2016; Ashrafmansouri and Nasr Esfahany 2016; Ullah et al. 2023; Koo and Kleinstreuer 2005). In this study, it was found that Brownian mechanism serves an important role for the CO_2 absorption into Co/SiO_2 nanofluid. It is due to the lack of a strongly polar structure and an asymmetric molecular configuration in CO_2 molecules, which results in the absence of significant molecular charges (O=C=O) that can be absorbed on the surface charge of nanoparticles. Therefore, increasing volume of nanofluid is not in the favor of more CO_2 absorption because of lower micro-convections. It is worth mentioning that since Co/SiO_2 NPs have a high magnitude of surface charge (Darvanjooghi and Esfahany 2016), attributing to the silanol bond formation (Si-O-H), the grazing effect could be another affecting mechanism with a low-intensity.

3.4 Correlation to Predict CO_2 Absorption

The ratio of convective mass transfer to diffusive mass transfer from bubbles into a liquid in a bubble column is represented by the Sherwood number, Equation 6 (Vasconcelos, Orvalho, and Alves 2002). Equation 6 has been used as a precious correlation to predict gas absorptions into a vast range of liquids using single bubbles in an absorber column (Calderbank and Lochiel 1964).

$$Sh = 0.6\, Re^{\frac{1}{2}} Sc^{\frac{1}{3}} \tag{6}$$

To use Equation 6 for the estimation of *Sh* number for the CO_2 absorption into a nanofluid, some other physical properties are needed to obtain like kinematic viscosity, dynamic viscosity, and density of Co/SiO_2 nanofluid (Mishra et al. 2014). These physical properties can be calculated according to Equations 7 to 9 as follow:

$$\rho_{nanofluid} = \varphi\rho_p + (1-\varphi)\rho_{basefluid} \tag{7}$$

$$\mu_{nanofluid} = \mu_{basefluid}(1-\varphi)^{2.5} \tag{8}$$

$$\vartheta_{nanofluid} = \frac{\mu_{nanofluid}}{\rho_{nanofluid}} \tag{9}$$

where φ is the volume fraction of Co/SiO_2 NPs in the base fluid and can be calculated from Equation 10. $\mu_{nanofluid}$ and $\mu_{basefluid}$ are dynamic viscosities of the nanofluid and base fluid, respectively. ρ_p and $\rho_{basefluid}$ present the bulk density of NPs (2.196 $kg.m^{-3}$) and density of the base fluid (1000 $kg.m^{-3}$), respectively.

$$S\varphi\ (vol.\%) = \frac{w\ (wt.\%)}{w\ (wt.\%) + \frac{\rho_p}{\rho_{basefluid}}\left(100 - w\ (wt.\%)\right)} \tag{10}$$

Re, *Sh*, and *Sc* numbers were calculated from Equations 11 to 13, respectively.

$$Re_b = \frac{U_b d_b}{\vartheta_{nanofluid}} \tag{11}$$

$$Sh_{nanofluid} = \frac{k_{l,nanofluid}.d_b}{D_{nanofluid}} \quad (12)$$

$$Sc_{nanofluid} = \frac{\vartheta_{nanofluid}}{D_{nanofluid}} \quad (13)$$

In these Equations, U_b is the rising velocity of the bubbles in the absorber column, which was considered to be 0.21 $m.s^{-1}$. Besides, d_b means the diameter of the bubbles, which was measured at almost 7 mm.

Table 1 shows the physical properties of Co_2 absorption into Co/SiO_2 nanofluid and base fluid.

Table 1. Diffusion coefficient, Reynolds (*Re*), Sherwood (*Sh.*), and Schmidt (*Sc*) numbers for CO_2 absorption into Co/SiO_2 nanofluid.

Absorbent	ν ($m.s^{-1}$)	*Re*	*Sh.*	*Sc*
Nanofluid	$8.88 * 10^{-7}$	1300	243	146
Basefluid	$8.9 * 10^{-7}$	1298	316	420

Based on the values presented in Table 1, there were no significant changes in *Re* number in the nanofluid or base fluid during the absorption process i.e., $\nu_{nanofluid} \approx \nu_{basefluid}$. Therefore, it can be concluded that *Re* did not have a significant effect on the *Sc* number and followed a relative function according to below:

$$\frac{Sh_{nanofluid}}{Sh_{basefluid}} = K\left(\frac{Sc_{nanofluid}}{Sc_{basefluid}}\right)^m \quad (14)$$

m and K were considered by conducting a two-dimensional regression analysis on the experimental data presented in Figure 8. From the obtained results, Equation 15 was proposed to describe the physical parameters, achieving an R^2 value of 0.99. Based on the results K and m values are 1.34 and 0.53, respectively. Therefore, using Equation 15, one can predict the *Sh* number for CO_2 absorption into a nanofluid at Re_b~1300.

$$\frac{Sh_{nanofluid}}{Sh_{basefluid}} = 1.34\left(\frac{Sc_{nanofluid}}{Sc_{basefluid}}\right)^{0.53} \quad (15)$$

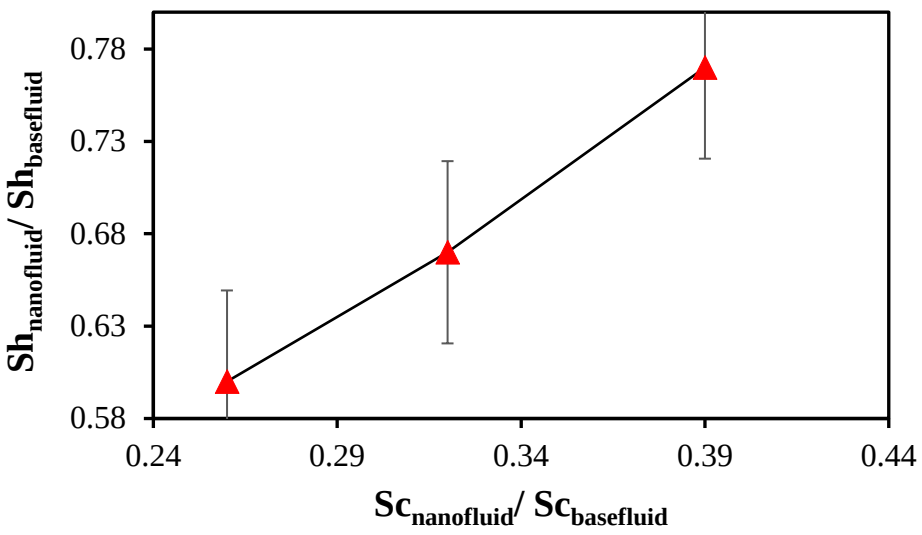


Fig. 8. The effect of relative *Sh* number on relative *Sc* number.

4. Conclusions

In this research, Co-doped SiO_2 nanoparticles were synthesized and used to enhance CO_2 absorption in a bubble column at 20 °C and 1 atm. Results confirmed that the prepared nanofluid had high stability with the ξ-potential lower than –45 mV. TEM and DLS analyses represented the average size of the synthesized NPs was 70 nm. The results also confirmed that the NPs' weight percentage and the volume of the nanofluid served important roles in the CO_2 absorption rate in such a way that the optimum condition was achieved for 80 mL of the nanofluid including 0.02 wt.% of NPs. Increasing NPs concentration from 0.001 to 0.02 favors CO_2 absorption while the higher increase caused the absorption rate to decrease. Besides, CO_2 molecules showed a better absorption rate in lower volumes of nanofluids. Indeed, it was found that although both the grazing effect and the Brownian motion of NPs served a crucial role in increasing CO_2 absorption, the Brownian motion of NPs was the predominant mechanism. Moreover, mass transfer parameters affecting CO_2 diffusivity into the Co/SiO_2 nanofluid like Sherwood (*Sh*) and Schmidt (*Sc*) numbers were calculated. Finally, a new correlation was

introduced to predict the *Sh* number over the *Sc* number in a gas-nanofluid column (Re ~ 1300) with a high accuracy of R^2=0.99.

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Conflicts of Interest

The authors declare no conflict of interest.

Competing Interests

This research received no external funding.

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