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RETRACTED: Polymer-Coated Nickel Nanoparticles for CO₂ Capture in Seawater

Abhishek, Abhishek Ratanpara , Adib Mahmoodi Nasrabadi  and Myeongsub Kim * 

Department of Ocean and Mechanical Engineering, Florida Atlantic University, Boca Raton, FL 33431, USA; abhisheklhu2023@fau.edu (A.); aratanpara2020@fau.edu (A.R.); amahmoodinas2023@fau.edu (A.M.N.)

* Correspondence: kimm@fau.edu

Abstract: Carbon capture and storage (CCS) technologies are employed to mitigate global warming by removing carbon from the atmosphere. To enhance carbon capture efficiency, nanoparticles have gained considerable attention as catalysts due to their large surface area, tunable properties, regeneration, and enhanced reactivity. However, it poses some challenges, such as nanoparticle aggregation and reduced effectiveness in sustainable solvents like seawater. To address these limitations and promote an environmentally sustainable method for carbon capture, this study evaluates the CO₂ capture efficiency of seawater using nickel nanoparticles (NiNPs) coated with polyvinylpyrrolidone (PVP) as a catalyst. We examined the time-dependent size variations of CO₂ bubbles in a flow-focusing microchannel using high-speed bubble-based microfluidics, directly associated with transitory CO₂ dissolution into the surrounding solution. We hypothesized that smaller polymer-coated NiNPs, due to their higher surface-to-volume ratio, can enhance CO₂ solubility and capture rates under identical environmental conditions. To verify this, polymer-coated NiNPs of three different sizes—5 nm, 10 nm, and 20 nm—were synthesized and tested. The experiments revealed that 5 nm NiNPs achieved a CO₂ dissolution rate of 77%, in contrast to 71% for 10 nm and 43% for 20 nm particles. These findings validate the hypothesis, demonstrating that smaller nanoparticles facilitate more effective CO₂ capture using equivalent material quantities, thereby potentially improving the overall efficiency of CO₂ reduction. This innovative approach contributes to advancing NiNP-based catalysts for saltwater-based CO₂ capture.

Keywords: carbon capture; nickel nanoparticles; polyvinylpyrrolidone; zeta potential; dispersion; dissolution



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1. Introduction

The increase in greenhouse gas emissions associated with human activities has been significant over the past century and continues accelerating, with global carbon dioxide (CO₂) emissions more than doubling in the past 50 years, rising from approximately 17 billion metric tons to over 37 billion metric tons [1]. The observed increase leads to considerable impacts on global warming, exacerbating climate-related challenges like the unprecedented elevation of sea levels, as well as an uptick in the frequency and severity of wildfires, floods, droughts, and tropical storms. These negative impacts not only disturb natural ecosystems but also pose considerable risks to human populations [1]. For example, the combustion of fossil fuels and various industrial activities has led to CO₂ accounting for 78% of total greenhouse gas emissions [2]. Natural systems like the ocean and lakes can absorb CO₂ as part of the carbon cycle; however, the processes involved in this uptake are

inadequate to offset the rising levels of anthropogenic CO₂ emissions in the atmosphere [3]. The decrease of CO₂ emissions is a pressing issue that demands prompt focus through both policy initiatives, such as carbon pricing and renewable energy subsidies, and technological progress, including carbon capture and storage. As a result, various approaches have been proposed to lower global atmospheric CO₂ concentrations [4–6]. Among these strategies, carbon capture, utilization, and storage (CCUS) technologies have emerged as a pivotal approach for mitigating CO₂ emissions and decelerating climate change [7]. These technologies focus on capturing CO₂ emissions at their source, preventing their release into the atmosphere, and either repurposing them for industrial applications or storing them safely underground [8,9]. The implementation of CCUS technologies is particularly critical in major CO₂-emitting industrial sectors such as coal-based thermal power plants, cement production, and steel manufacturing, where emissions are substantial and difficult to reduce through alternative means alone [6].

Currently, post-combustion CO₂ capture is recognized as a prominent technology in commercial fossil fuel power plants focused on reducing CO₂ emissions [10]. This procedure entails the extraction of CO₂ from flue gas post-combustion, with the captured CO₂ generally subjected to sequestration and storage via complex chemical processes [11]. The modular nature of post-combustion CO₂ capture presents a significant advantage, allowing for greater adaptability in retrofitting when compared to pre-combustion and oxy-fuel combustion methods, which tend to be more complex and costly [12,13]. Among the various techniques for post-combustion capture, amine scrubbing stands out as the most widely adopted method, owing to its demonstrated effectiveness in industrial settings. During this procedure, CO₂-laden flue gas is introduced to an amine solution within an absorber unit, facilitating the binding of CO₂ to the amine molecules. The flue gas, depleted of CO₂, is subsequently discharged into the atmosphere, whereas the amine solution, rich in CO₂, is directed to a regeneration unit for further processing. Heat is applied to separate CO₂ from the amine, enabling the CO₂-free amine to be reused in the absorption process [14].

While amine scrubbing is commonly employed, it has notable drawbacks, such as significant energy demands during the regeneration process, the deterioration of amine solvents over time, and possible environmental risks associated with amine emissions [14]. To tackle these challenges, current research investigations are focused on sustainable alternative solvents, with seawater standing out as a promising option. Seawater is a most abundant natural resource, and its inherent alkalinity presents a compelling opportunity for CO₂ capture, especially in coastal power plants, where it can lessen reliance on freshwater resources. Nonetheless, seawater exhibits comparatively slow reaction kinetics and restricted absorption capacity due to salinity, highlighting the need for improvements via advanced materials. To address these inherent limitations of seawater, investigations have been conducted into the use of nanoparticles (NPs) as a catalyst for CO₂ capture in seawater. Nanoparticles provide an extensive surface area, exhibit catalytic properties, and possess adjustable functionalities, all of which can greatly enhance the efficiency of CO₂ absorption. Metal oxide nanoparticles, including MgO and CaO, have demonstrated potential by offering nucleation sites for CO₂ absorption [15]. Nonetheless, these materials exhibit limitations, such as structural and chemical alterations over time that diminish their reactivity and stability in ongoing capture systems [16]. This has initiated the exploration of more environmentally friendly alternatives. Among other nanoparticle materials, nickel nanoparticles (NiNPs) have attracted interest because of their potential in CO₂ capture and regenerative functions [17]. NiNPs demonstrate distinctive catalytic characteristics that improve CO₂ dissolution, especially in intricate aqueous settings, positioning them as a promising option for enhancing CO₂ capture in seawater systems [18–21]. NiNPs offer superior corrosion resistance in saline environments and exhibit regenerative properties due to their inherent metallic characteristics, enabling

them to maintain their functionality after multiple cycles of CO₂ capture. Moreover, NiNPs exhibit more stable, pH-independent CO₂ capture performance, which is advantageous as the solution pH tends to drop during continuous CO₂ dissolution [20,21]. Nonetheless, a significant obstacle linked to NiNPs is their propensity to cluster in seawater environments due to van der Waals forces. This aggregation diminishes the available surface area, and, as a result, their catalytic efficiency is reduced.

To mitigate this challenge, the steric stabilization of NPs plays a crucial role in enhancing their dispersion and performance in aqueous solutions, leading to improved catalytic efficiency. Small molecule ligands in polymers regulate nanoparticle stability while maintaining their material properties [22–24]. Therefore, additional investigation on efficient steric stabilization is necessary to fully utilize polymer-NP catalysts for enhanced CO₂ capture.

This study investigates the capabilities of polyvinylpyrrolidone (PVP)-coated NiNPs to improve CO₂ capture efficiency in natural seawater. Compared to the conventional study [22], where the polymer dispersants were directed to NPs after synthesis through the spontaneous process [22], the PVP coating occurs through chemical bonding during NP synthesis, which could effectively stabilize the NiNPs, preventing aggregation and ensuring optimal dispersion. We hypothesize that the individual dispersed NPs preserve their catalytic activity, so the catalytic performance of NPs per volume would be maximized by this approach [22]. To explore the influence of nanoparticle size on CO₂ dissolution, we synthesize PVP-coated NiNPs of different sizes, enabling us to evaluate the size dependency on dissolution rates (i.e., the greater the surface area, the higher the reactivity). We utilize bubble-based microfluidic techniques, providing the quantification of CO₂ dissolution at the gas–liquid interface through the observation of bubble size and morphology [25–30]. The degree of NP dispersion at different polymer concentrations in various sizes and the resultant CO₂ dissolution performance are quantitatively compared.

2. Materials and Methods

A microfluidic channel with a flow-focusing geometry was used to generate consistent CO₂ microbubbles in all test solutions. The microbubble-based quantification of CO₂ absorption in microfluidic platforms has been well established in the literature and provides an excellent experimental framework where variables (e.g., pressure, flow rate) can be easily and precisely controlled [31,32]. The high surface area-to-volume ratio of microbubbles enables rapid mass transfer, leading to swift alterations in bubble size over short timescales [30,33–38]. Examining the behavior of CO₂ microbubbles offers a dependable method for measuring dissolution rates and assessing the impact of NiNP-polymer stabilization on improving CO₂ sequestration.

2.1. Materials

Natural seawater was obtained from the Atlantic Ocean coastal areas of Boca Raton, Florida, with a measured salinity of 34 ± 1 ppt. Nickel (II) nitrate hexahydrate, 98%; sodium borohydride, 98%; and polyvinylpyrrolidone (M.W. = 50,000) were purchased from Thermo Fisher Scientific, Inc. PHD ULTRA 4400, Harvard Apparatus, Holliston, MA, USA; the syringe pump was used to maintain the flow rate of the test solution. The Olympus Corp., Japan IX71 microscope is used to observe the experiment, and an attached camera (Fastc) is used to record videos. The microfluidic chip was made from polydimethylsiloxane (PDMS) elastomer kits (Sylgard 184, Dow Corning Corp., Midland, MI, USA) through two well-known subsequent processes, including photolithography and soft lithography. The zeta potential measurements were performed using a Malvern Zetasizer Nano, USA series to assess the electrostatic stability of the nanoparticles.

2.2. Nanoparticle Synthesis

Nickel nanoparticles (NiNPs) were synthesized via the polyol method, a widely adopted approach known for its cost-effectiveness and precise control over particle morphology and size [39]. In this process, 60 mg of nickel(II) nitrate hexahydrate was dissolved in 20 mL of ethylene glycol, which served both as a solvent and a mild reducing agent. To control nanoparticle growth and prevent agglomeration, 600 mg of polyvinylpyrrolidone (PVP) was added as a stabilizer. Subsequently, 180 mg of sodium borohydride (NaBH_4), a strong reducing agent, was slowly introduced into the mixture under continuous stirring at 200 rpm. The gradual addition of NaBH_4 (20 mg/min) facilitated the reduction of Ni^{2+} ions to metallic Ni^0 , initiating the nucleation of NiNPs.

Once reduction commenced, the reaction mixture was heated to temperatures ranging between 100 °C and 130 °C. Varying the temperature enabled controlled nucleation and growth, allowing the formation of nanoparticles in distinct sizes. The presence of PVP played a critical role in regulating particle formation by capping the surface, thereby preventing excessive growth and maintaining uniformity.

After the reaction concluded, the nanoparticles were collected by centrifugation at 11,000 rpm at room temperature and subsequently washed multiple times with deionized water to remove residual reagents and polyol, yielding high-purity, PVP-coated NiNPs. This method's tunability—achieved by adjusting factors such as reaction temperature, precursor concentration, and stabilizer content—makes it highly suitable for applications where particle characteristics are crucial. Using this approach, we successfully fabricated NiNPs of approximately 5 nm, 10 nm, and 20 nm in diameter for subsequent experiments aimed at enhancing CO_2 dissolution efficiency.

2.3. Experimental Procedure

Gaseous CO_2 was introduced into a microfluidic chip featuring a flow-focusing geometry and Y-shaped channel configuration concurrently with the injection of an aqueous test solution (Figure 1). The CO_2 inlet is 50 μm in width, whereas both the solvent inlet and the Y-shaped channel are 100 μm wide. The total length from the junction to the outlet was 20 mm. The CO_2 microbubbles were generated at the junction, where the aqueous solution flows at a 60° angle in relation to the CO_2 gas stream. The interplay of various forces, such as inertia, shear, interfacial tension, and pressure forces, results in the formation of CO_2 microbubbles in the aqueous solution [40]. The frequency of bubble generation can be modified by changing the flow rate of the aqueous solution, the relative angle of the aqueous solution to the CO_2 gas inlet, and the pressure of the injected CO_2 gas. An in-depth exploration of the mechanisms of bubble generation in liquid flow has been conducted in various microfluidic studies [40,41]. Along the channel, the shrinkage of CO_2 microbubbles represents its dissolution into the surrounding solvent. The reduction in CO_2 microbubble size was observed using high-speed microscopic imaging, which consisted of an Olympus IX71 microscope equipped with a 4× objective lens and a recording speed of 1000 frames per second. The solvent flow rate and CO_2 gas pressure were maintained at 35 $\mu\text{L}/\text{min}$ and 4.5 psig, respectively, to ensure stable and consistent CO_2 microbubble generation. Three different test solutions were prepared using natural seawater with NiNPs of varying diameters at 5 nm, 10 nm, and 20 nm. The test solutions were prepared using a sonicator (Fisher Scientific, Hampton, USA) operating at a frequency of 20 kHz for 30 min. To prevent overheating, a rest interval of 30 s per minute was applied. This process ensured the homogeneous dispersion of NiNPs in seawater.

In all test solutions, the NiNP concentration was kept constant at 28 mg/L to isolate the effect of nanoparticle size on CO_2 dissolution. The concentration of 28 mg/L for NiNPs was selected based on preliminary optimization experiments conducted in our laboratory.

Each experiment was repeated three times to ensure the repeatability and reliability of the results, with a maximum standard deviation of 2.88%.

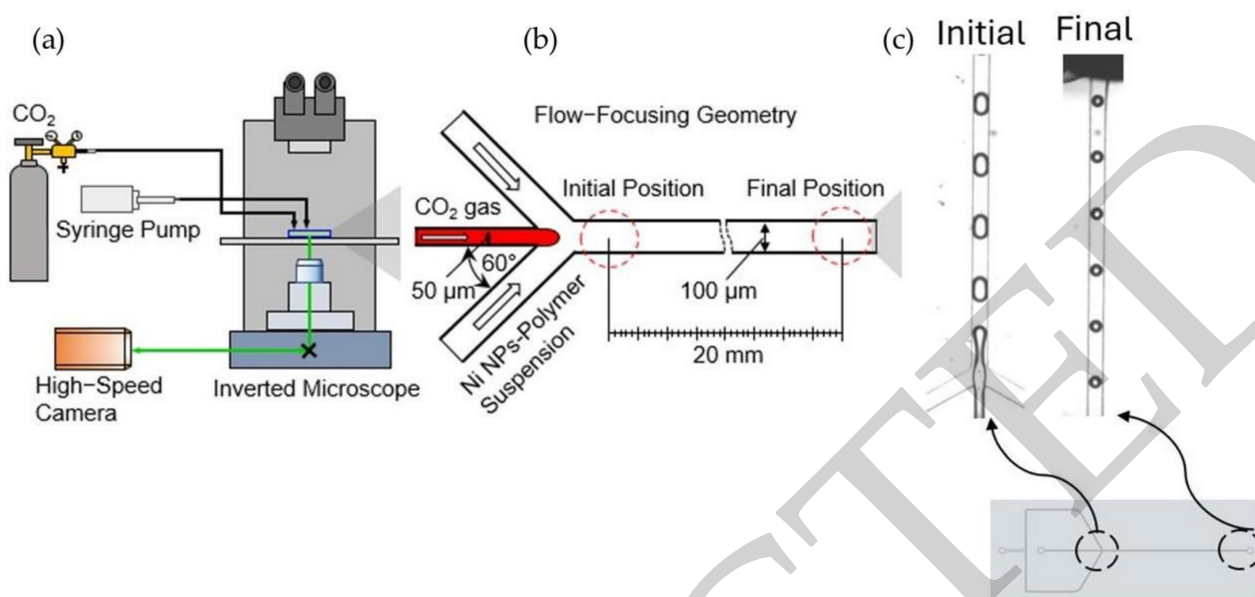


Figure 1. (a) The schematic experimental setup includes a CO₂ gas source connected to a syringe pump, which regulates gas flow into a microfluidic chip observed using a microscope and a high-speed camera. (b) Flow-focusing microchannel geometry, where a NiNP-polymer suspension interacts with CO₂. (c) Microscopic images of microbubble formation show the transition from the initial state, where bubbles are generated, to the final stabilized state downstream in the microchannel.

As shown in Figure 1c, recordings were taken at two locations because a single field of view could not encompass the entire straight channel. The first field of view was set immediately after the junction to capture the generation of CO₂ microbubbles, while the second was positioned at the outlet to record the final size of the shrunk CO₂ microbubbles. The recorded videos were analyzed using MATLAB software (2024). Figure 2 illustrates the image processing steps used to estimate the size reduction in the CO₂ microbubbles. As shown in Figure 2a, a reference frame was manually created by removing a microbubble. This reference frame was then subtracted from all recorded frames to determine the initial bubble size by calculating the pixel area of the CO₂ microbubbles, as shown in Figure 2b. All recorded bubbles exhibited similar sizes for their respective test solutions, with a maximum standard deviation of 0.96%. A similar process was applied to the recordings taken in the second field of view. Finally, the average initial and final bubble sizes were compared to analyze the total CO₂ dissolution in each test solution.

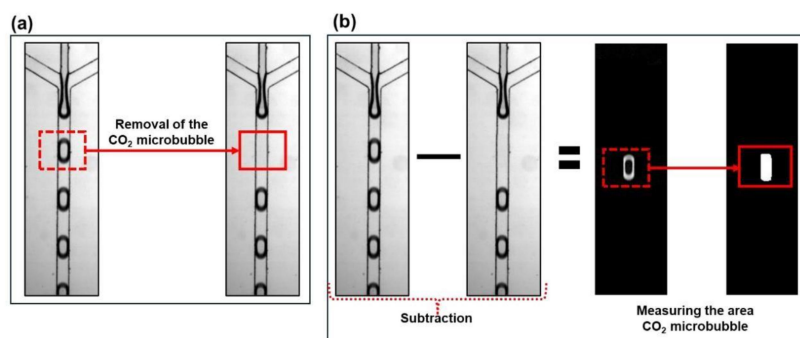


Figure 2. (a) Image sequence illustrating the reference frame and removal of a CO₂ microbubble within the microchannel. The red box highlights the region where the CO₂ bubble is detected and analyzed. (b) Image processing approach used for measuring the CO₂ microbubble area.

3. Results and Discussion

3.1. Temperature and Time Effects on Nanoparticle Size

This experimental investigation examines the effect of PVP coating and NiNP size on CO₂ dissolution performance. Parametric modification was performed during the synthesis process to obtain NiNPs of varying sizes. While this process is well established, this subsection outlines the observed trends under certain experimental conditions. The variation in nanoparticle size was obtained by interplaying nucleation and growth dynamics during NiNP synthesis. At higher temperatures, the nucleation rate of nickel atoms increases, leading to the rapid formation of a larger number of nickel nuclei, which limits the growth of individual nanoparticles. Simultaneously, a shorter reaction time restricts the growth phase, preventing excessive particle expansion. On the contrary, lower temperature and longer reaction time would allow the extended growth of nickel nuclei, resulting in large nanoparticle size. As shown in Table 1 and Figure 3, these effects are reflected during the synthesis process. When the temperature and reaction time were set to 130 °C and 90 min, respectively, the average NiNP diameter size was found to be 20 nm. Upon temperature increment from 130 °C to 160 °C with a reduced reaction time of 45 min, the synthesized NiNP's average diameter was found to be 10 nm. Further increment of temperature to 180 °C and reduction of reaction time to 30 min lead to 5 nm diameter-sized NiNP production. The size observation of NiNPs was performed using a transmission electron microscope (TEM).

Table 1. Nanoparticle size variations according to temperature and time during synthesis.

Nanoparticle Size	Time (Minutes)	Temperature
5 nm	30	180 °C
10 nm	45	160 °C
20 nm	90	130 °C

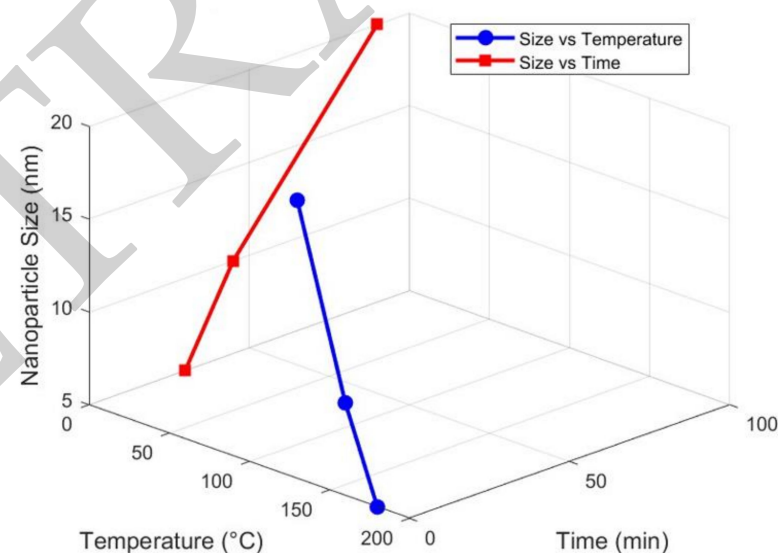


Figure 3. The 3D plot of changes in nanoparticle size according to temperature and time during synthesis.

3.2. Characterization of PVP-Coated NiNPs

Figure 4 presents TEM images of NiNPs with average sizes of 5 nm, 10 nm, and 20 nm, highlighting their morphology and size distribution. In Figure 4a, the TEM image of 5 nm PVP-coated NiNPs shows that the particle sizes largely range from 4.36 nm to 5.92 nm, with a nominal diameter of 4.87 nm based on 12 samples. The PVP polymer (appearing as a dark

gray coating) effectively disperses the nanoparticles, preventing aggregation. The irregular shapes and high surface roughness of these nanoparticles arise from dominant surface energy effects, which prevent the formation of smooth structures. This rough morphology increases the actual exposed surface area, thereby enhancing the surface-area-to-volume ratio. As a result, more active sites are available for catalytic reactions, improving their overall catalytic efficiency.

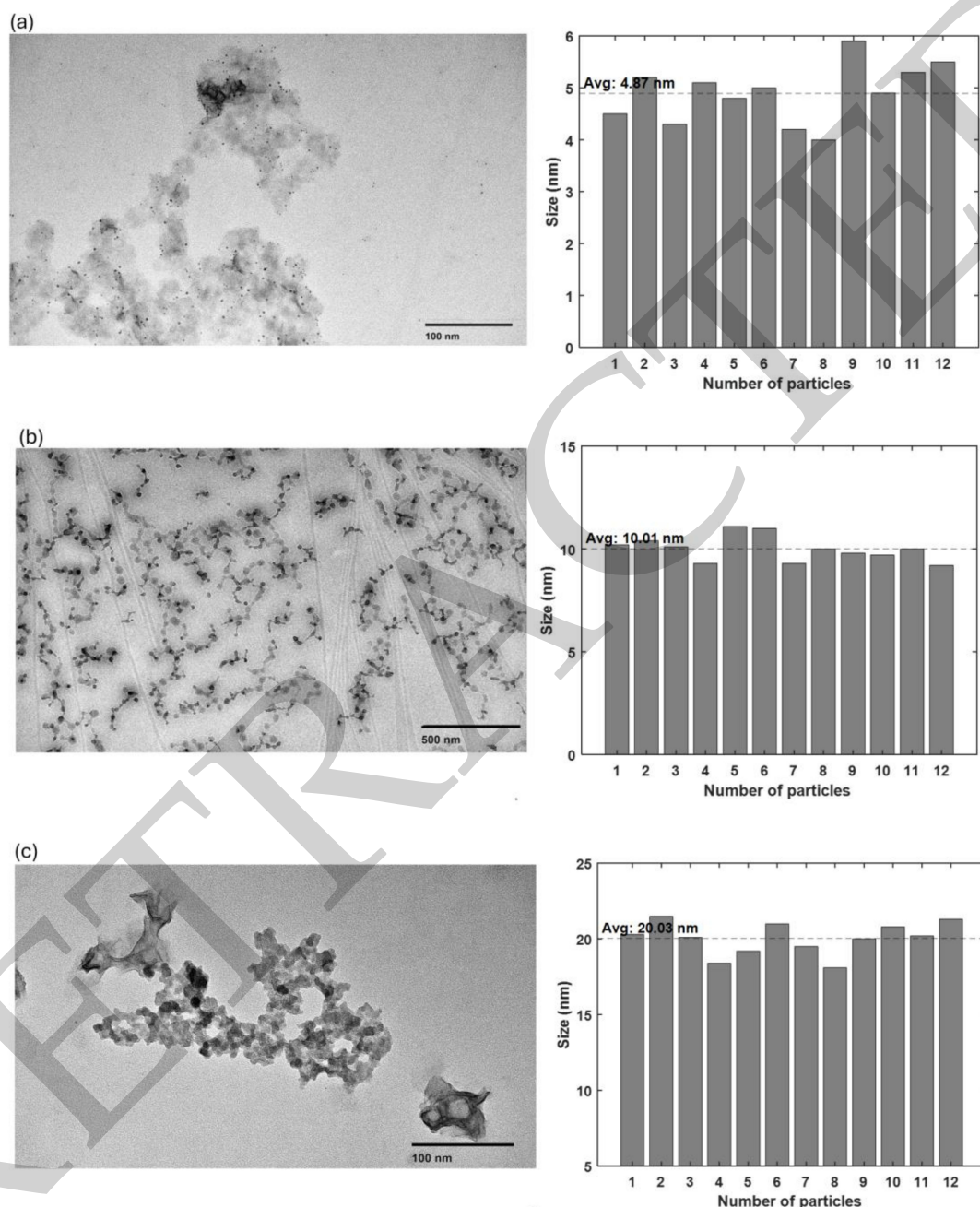


Figure 4. Transmission electron microscopy (TEM) of PVP-coated NiNPs in (a) 5 nm; (b) 10 nm; (c) 20 nm. The dotted line in each figure represents the average size of NPs.

In Figure 4b, the TEM image of NiNPs with an average size of 10.01 nm (ranging from 8.44 nm to 12.4 nm) reveals asymmetrical particle morphologies with some clustering, particularly in darker regions. These particles show more faceted shapes with clearer lattice fringes, indicating improved crystallinity and stability. Compared to the 5 nm NiNPs, they are more prone to agglomeration and beam-induced deformation, making

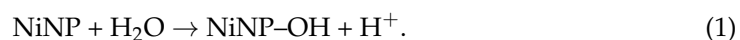
them less suitable for catalytic applications due to reduced surface area-to-volume ratio. In Figure 4c, the TEM image of NiNPs with an average size of 20.3 nm (ranging from 20.1 nm to 22.0 nm). However, their lower surface area-to-volume ratio slightly reduces their reactivity compared to smaller nanoparticles.

The observed structural variations across different NiNP sizes are expected to impact CO₂ capture efficiency. Smaller nanoparticles provide more active sites due to their high surface-area-to-volume ratio, which can enhance the CO₂ dissolution rate in seawater. In contrast, larger nanoparticles have a lower surface area-to-volume ratio, aligning with the findings of this research.

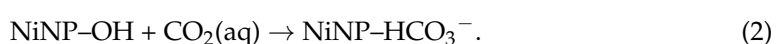
3.3. CO₂ Capture Mechanism by NiNPs

The process of CO₂ dissolution in water is broadly understood and follows a series of well-defined chemical equilibrium steps. When CO₂ gas comes into contact with water at the gas–liquid interface, a portion of the CO₂ molecules dissolves into the liquid phase and form carbonic acid (H₂CO₃). Carbonic acid then rapidly dissociates into bicarbonate ions (HCO₃[−]) and hydrogen ions (H⁺). The extent to which bicarbonate further dissociates into carbonate ions (CO₃^{2−}) depends largely on the alkalinity (pH) of the water. In freshwater, most of the dissolved CO₂ remains in the form of bicarbonate ions with minimal further dissociation into carbonate ions due to lower alkalinity. In contrast, seawater is more alkaline, which allows a higher conversion of bicarbonate into carbonate ions compared to fresh water. However, despite its higher alkalinity, seawater has a lower overall CO₂ dissolution rate compared to freshwater due to its high salinity. The presence of dissolved salts increases the ionic strength of the solution, which reduces CO₂ solubility through salting-out effects. The salting-out effect occurs because dissolved ions in seawater compete with dissolved salts for water molecules, reducing the availability of free water to interact with CO₂ and thereby lowering its solubility. As a result, while seawater enhances CO₂ dissociation into carbonate, it simultaneously hinders the initial dissolution of CO₂ into the liquid phase. To overcome the salting-out effect and make seawater more practical for carbon capture use, NiNPs can be used to catalyze the CO₂ dissolution process.

In this section, we have described a possible mechanism of NiNPs as a catalyst for CO₂ capture. The exact mechanism by which NiNPs enhance CO₂ dissolution is not yet fully understood. Figure 5 illustrates a CO₂ microbubble in a microchannel surrounded by seawater containing colloidally suspended NiNPs. NiNPs interact with water molecules, leading to the formation of hydroxyl groups on their surface. This process can be represented as follows:

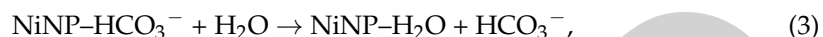


These NiNPs-OH are observed to migrate toward the gas–water interface, likely due to the higher interfacial energy at this boundary. Nanoparticles tend to adsorb at such interfaces to lower the system's overall free energy, stabilizing the gas–liquid boundary. Once at the interface, as shown in Figure 5, CO₂ molecules adsorb onto the ligands present on the NiNP-OH surface [42]. The hydroxyl groups provide active sites where CO₂ molecules adsorb, potentially forming a bicarbonate intermediate. This interaction can be expressed as follows:



After adsorption, NiNP-HCO₃[−] moves away from the interface through two primary mechanisms. The shuttle mechanism suggests that NiNPs carrying HCO₃[−] migrate into the bulk liquid due to a diffusion driven by the concentration gradient of CO₂ in the bulk liquid and the CO₂ gas bubble. Another documented possible mechanism is based on fluid

dynamics, including Brownian motion and micro-convection, which create continuous nanoparticle movement, reduce mass transfer resistance, and ensure a steady exchange of NiNPs at the gas–water boundary. When NiNP-HCO_3^- reaches the bulk liquid, it undergoes further transformation or desorption, releasing bicarbonate ions into the solution while regenerating NiNP-OH for further CO_2 capture,



where CO_2 desorbs, dissolving into the surrounding water. Once CO_2 is released, NiNPs reposition within the system, allowing fresh nanoparticles to take their place at the interface. Additionally, fluid dynamics, including Brownian motion and micro-convection, create continuous nanoparticle movement, reducing mass transfer resistance and ensuring a steady exchange of NiNPs at the gas–water boundary. When NiNP-HCO_3^- reaches the bulk liquid, it undergoes further transformation, releasing bicarbonate ions into the solution while regenerating NiNP-OH for further CO_2 capture.

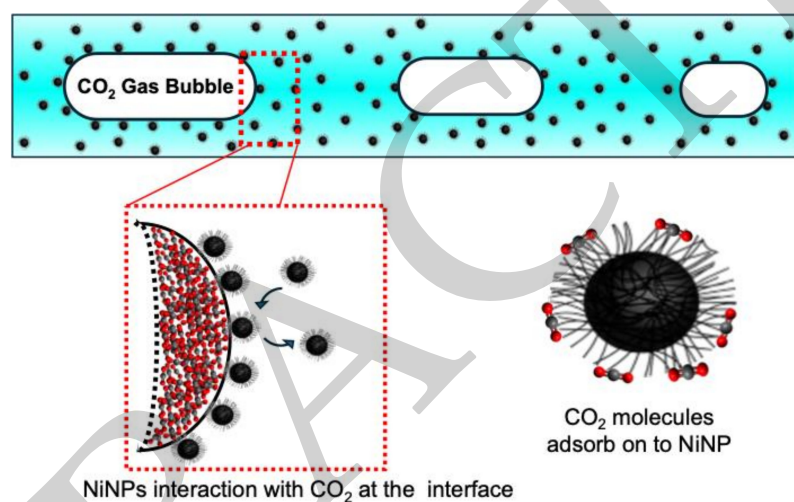


Figure 5. Illustrative mechanism of the catalytic activity of NiNPs in CO_2 dissolution. The shrinkage of the CO_2 gas bubble along the microchannel represents CO_2 dissolution into the surrounding liquid. The red box highlights the NiNP adsorption and desorption of CO_2 at the interface (exaggerated). The bottom right image illustrates a NiNP with multiple active sites with adsorbed CO_2 molecules.

This adsorption–desorption cycle suggests that NiNPs act as carriers, facilitating the continuous transport of CO_2 from the gas phase into the liquid phase, effectively catalyzing the dissolution process. While the term “catalytic” is commonly used in the literature to describe this process [42,43], it should be noted that it does not fully align with the traditional definition of catalysis, which involves the breaking of chemical bonds. Instead, this mechanism primarily promotes CO_2 dissolution through physical interactions. As mentioned earlier, the exact mechanism is still not fully understood, but it clearly differs from processes like CO_2 hydrogenation and reduction [19,44].

Based on this hypothesis, we believe that smaller NiNPs with a higher surface-to-volume ratio would be more effective in enhancing CO_2 dissolution. The increased surface area provides more active sites for CO_2 adsorption with less mass of nickel, thereby improving transport efficiency and accelerating the dissolution process [45,46].

3.4. CO_2 Capture Performance of PVP-Coated NiNPs

In this study, PVP was selected due to its superior durability and enhanced performance in solvent-saline environments. Seawater’s high ionic strength often compromises the functionality of nanoparticles by reducing their solubility or altering the structural

behavior of polymer coatings. PVP, however, is widely recognized for its outstanding ability to stabilize nanoparticles—minimizing aggregation and enhancing dispersion efficiency. This enhanced dispersion plays a critical role in facilitating efficient CO₂ capture, as it directly influences the dissolution behavior of CO₂ bubbles in the microfluidic system. The dissolution rate refers to the total number of CO₂ molecules dissolved into the surrounding liquid per time. This concept can be translated to the percentage reduction in the projected area of CO₂ bubbles over time (%/s) as measured via high-speed imaging within the microfluidic channel. This serves as one of the key indicators to evaluate the nanoparticle-assisted CO₂ absorption performance.

Figure 6 illustrates the percentage change in CO₂ dissolution relative to the size of PVP-coated nickel nanoparticles (NiNPs), tested at 5 nm, 10 nm, and 20 nm. The error bars represent variability in the experimental data and highlight the dissolution efficiency associated with each nanoparticle size. A clear inverse relationship was observed: as nanoparticle size increased, CO₂ dissolution decreased. The 5 nm PVP-coated NiNPs achieved the highest dissolution rate, nearly 77%, while the 10 nm and 20 nm variants demonstrated lower rates of 71% and 43%, respectively. This trend supports our hypothesis that smaller PVP-coated NiNPs yield superior CO₂ capture performance. To explore the underlying cause of this variation, we performed zeta potential analysis to examine the nanoparticles' surface properties. Zeta potential quantifies surface charge and serves as a critical indicator of colloidal stability [47,48]. Nanoparticles with high positive or negative zeta potentials exhibit strong electrostatic repulsion, which reduces aggregation and supports better dispersion in the solution. The stabilization plays a vital role in CO₂ capture applications, as well-dispersed nanoparticles offer an increased reactive surface area for gas–liquid interactions. In theory, smaller nanoparticles are expected to improve capture performance because of their greater surface-to-volume ratio, leading to an increase in the number of available reaction sites. Nevertheless, the existing literature indicates that smaller nanoparticles show a greater propensity to agglomerate, which is at odds with the anticipated improvement in performance. In order to explore this discrepancy, we examined the interaction forces that influence nanoparticle stability through the application of the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory.

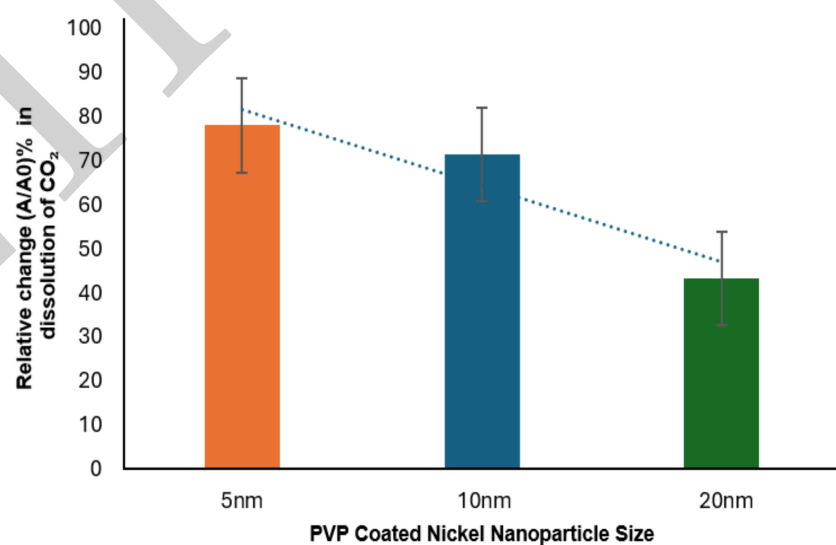


Figure 6. The relative dissolution rate of CO₂ in seawater according to the size of PVP-coated nickel nanoparticles. A is the initial area, and A₀ is the final area of the CO₂ bubble. The dotted line represents the trend of CO₂ dissolution in different NP diameters.

The DLVO theory posits that the overall interaction energy among particles is influenced by the interplay of coulombic (electrostatic) repulsion and van der Waals attraction. The electrostatic repulsion originates from surface charges and is predominant at short separation distances, establishing an energy barrier that inhibits aggregation. The undesirable energy diminishes with increasing separation distance, whereas van der Waals attraction—an intrinsically attractive force—continues to be relevant at very short distances, which could result in aggregation if the repulsive energy barrier is surpassed. At intermediate distances, a consistent energy barrier upholds nanoparticle dispersion, thereby guaranteeing system stability. Higher zeta potential and strong electrostatic repulsion counteract van der Waals forces, preventing nanoparticle aggregation and maintaining colloidal stability. This suggests that nanoparticle stability, rather than just surface area, plays a crucial role in enhancing CO₂ dissolution rates. Conversely, at low zeta potential, van der Waals attraction dominates, leading to aggregation and reduced stability. The balance between these forces directly influences the effectiveness of nanoparticles in gas–liquid interactions, highlighting the importance of electrostatic repulsion in optimizing CO₂ capture efficiency.

Figure 7 illustrates the correlation between the size of PVP-coated nickel nanoparticles (NiNPs) and their corresponding zeta potential values (in millivolts, mV), thereby emphasizing the electrostatic stability across different nanoparticle sizes of 5 nm, 10 nm, and 20 nm. The left *x*-axis denotes the particle sizes, while the right *y*-axis displays the zeta potential measurements. From the data, it is evident that smaller nanoparticles tend to possess higher zeta potential values, which is indicative of enhanced electrostatic stability resulting from increased surface charge. Specifically, the 5 nm NiNPs exhibit the highest zeta potential at 23 mV, suggesting strong charge repulsion and improved dispersion behavior in solution. In contrast, the 10 nm particles demonstrate moderate stability with a zeta potential of 19 mV, whereas the 20 nm particles show a lower value of 13 mV, implying weaker repulsive forces and a greater tendency to aggregate. This observed trend—where zeta potential decreases with increasing particle size—can be attributed to the diminishing surface area-to-volume ratio in larger nanoparticles. Such a reduction may limit the ability of the PVP coating to establish a sufficiently strong electrostatic field around the particles, thereby weakening their colloidal stability.

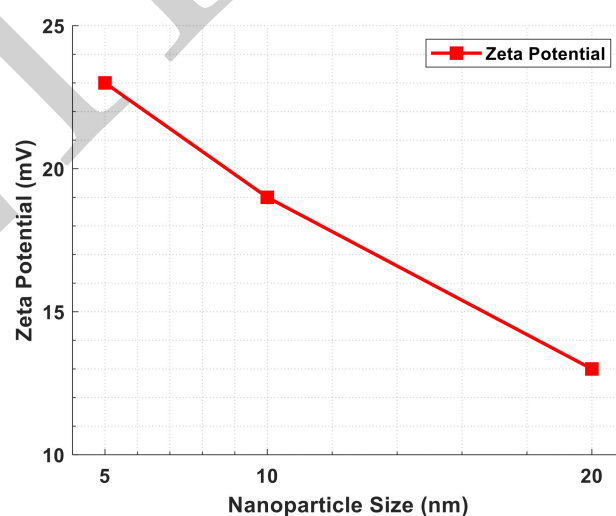


Figure 7. The relationship between zeta potential and PVP-coated nickel nanoparticle size.

Overall, the results highlight the significance of nanoparticle size optimization for maintaining favorable zeta potential values, which is particularly important in applications like CO₂ dissolution and other industrial processes that demand stable colloidal systems.

3.5. Sedimentation of PVP-Coated NiNPs

In seawater, PVP-coated NiNPs experience a force balance between electrostatic repulsion, van der Waals attraction, steric stabilization, aggregation, and sedimentation. Initially, all three sizes (5 nm, 10 nm, and 20 nm) carry a negative charge due to interactions with water molecules and hydroxyl (OH[−]) ion adsorption. This creates an electric double layer (EDL) that prevents aggregation by repelling particles. However, seawater's high salt content, including sodium (Na⁺), calcium (Ca²⁺), and magnesium (Mg²⁺), weakens this repulsion by compressing the EDL. As a result, attractive forces become stronger, especially affecting the smaller 5 nm NiNPs, which depend more on electrostatic stability [49].

PVP coating plays a critical role in mitigating these effects by providing steric hindrance. PVP molecules form a hydrated polymer shell around each nanoparticle, physically preventing them from coming close enough for van der Waals attraction to dominate. The effectiveness of PVP stabilization is highest for the 5 nm NiNPs, as their higher surface area allows for better polymer coverage, creating a strong steric barrier. The 10 nm NiNPs experience moderate stabilization, while the 20 nm NiNPs, due to their lower surface-to-volume ratio, have a weaker steric effect, making them more susceptible to aggregation despite the polymer coating. As aggregation progresses, sedimentation becomes increasingly significant. Individually dispersed NiNPs settle slowly due to Brownian motion counteracting gravitational forces, particularly in the case of the 5 nm NiNPs. However, as aggregation leads to the formation of larger clusters, these clusters behave as larger particles, accelerating their sedimentation rate. The 20 nm NiNPs, which aggregate quickly without sufficient steric stabilization, form the largest clusters and settle the fastest. The 10 nm NiNPs sediment at a moderate rate, while the 5 nm NiNPs, if well stabilized by PVP, remain suspended for the longest duration.

Figure 8 depicts the sedimentation process of PVP-coated NiNPs over time within a seawater environment. At the outset, the three sizes of NiNPs (20 nm, 10 nm, and 5 nm) exhibit a well-dispersed nature, showing minimal visible sedimentation, which can be attributed to the electrostatic repulsion and steric stabilization offered by the PVP coating. As time advances, sedimentation initiates, especially for the larger 20 nm NiNPs (Figure 8), since the elevated ionic strength of seawater compresses the electric double layer, diminishing electrostatic repulsion and enabling van der Waals forces to facilitate aggregation. During the intermediate phase, the 20 nm NiNPs begin to exhibit noticeable sedimentation, whereas the 10 nm and 5 nm NiNPs maintain a relatively stable suspension. In the final stage (Figure 8), notable sedimentation occurs, characterized by the formation of a substantial sediment layer at the bottom due to the 20 nm NiNPs, while the 10 nm NiNPs exhibit moderate aggregation and settling, and the 5 nm NiNPs predominantly remain suspended. The findings indicate that smaller NiNPs (5 nm) demonstrate enhanced steric stabilization attributed to superior polymer coverage, whereas larger particles (20 nm) undergo swift aggregation and sedimentation due to diminished steric effects and prevailing attractive forces. In summary, PVP-coated NiNPs in 5 nm remain stable for an extended period due to strong steric effects, as compared to 10 nm and 20 nm NiNPs, which experience increasing aggregation and sedimentation as steric hindrance becomes less effective and attractive forces dominate.

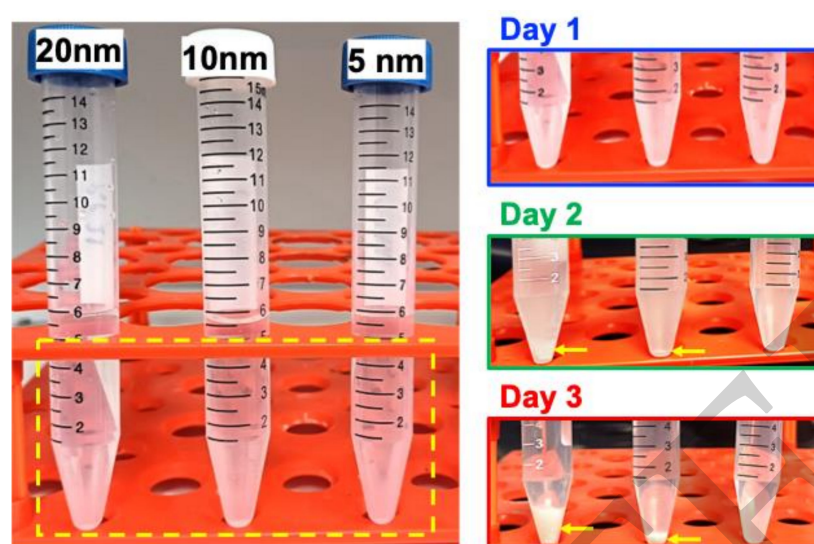


Figure 8. The sedimentation process of PVP-coated NiNPs over three days. The yellow arrows in each inset figure show precipitation.

4. Discussion

Previous investigations have thoroughly examined the capabilities of NiNPs as catalysts for CO₂ capture in saline conditions. Previous studies showed that the stabilization of polymers, especially using PVP and carboxymethyl cellulose (CMC), notably improved CO₂ dissolution by inhibiting nanoparticle aggregation, which poses a significant challenge in high-salinity environments [22]. Although their investigation validated the advantages of polymer coatings, it failed to determine the ideal nanoparticle size for maximizing catalytic efficiency. Furthermore, the stabilization mechanisms remain inadequately investigated, resulting in ambiguities about the optimal polymer type and coating technique necessary for maintaining long-term nanoparticle dispersion. Additional studies examined seawater as a potential solvent for amine-based CO₂ capture, revealing that it absorbed merely 1.79% less CO₂ compared to deionized (DI) water. The inclusion of NiNPs further reduced this difference to 1.15%, underscoring its potential as a sustainable capture medium. Nonetheless, these investigations were confined to amine-based solutions, which limited their wider relevance for carbon sequestration in natural seawater [48]. Moreover, although the investigation into NiNPs in conjunction with CMC polymers pinpointed an ideal concentration for CO₂ dissolution (90 mg/L NiNPs in 300 mg/L CMC, resulting in a 67.2% dissolution rate), it lacked a thorough examination of the impacts of nanoparticle size, polymer stabilization, and catalytic efficiency [50]. The study investigated NiNP-CMC interactions, but it did not employ a systematic methodology that included variations in nanoparticle size and the optimization of polymer coatings.

Expanding on these gaps, our investigation concentrated on enhancing nanoparticle dimensions, polymer stabilization, and catalytic efficiency for CO₂ capture in seawater, tackling the size-dependent catalytic performance of PVP-coated NiNPs, as shown in Table 2. The results indicated a distinct size-dependent trend, with 5 nm NiNPs demonstrating the highest dissolution rate at 77%, followed by 10 nm at 71% and 20 nm at 43%. This confirms that smaller NiNPs possess enhanced catalytic efficiency, which is attributed to their greater surface-to-volume ratio. Furthermore, our investigation reinforces the role of PVP as a potential stabilizing agent, facilitating nanoparticle dispersion and reducing efficiency losses associated with aggregation. All experimental conditions in our study were carefully controlled to isolate the effect of nanoparticle size, ensuring consistent flow and pressure conditions. Our findings demonstrate that under these conditions, smaller

nanoparticles (e.g., 5 nm) exhibit superior dissolution performance due to their higher surface area-to-volume ratios and better dispersion stability. The findings not only demonstrate a more efficient, amine-free approach to CO₂ capture in seawater but also offer a structured framework for refining nanoparticle size and polymer coatings, setting the stage for scalable and eco-friendly carbon sequestration technologies.

Table 2. The comparison of carbon dissolution with different nanoparticle sizes and concentrations associated with flow conditions.

	Channel Type	Pressure (psig)	Flow Rate (μL/min)	Concentration (mg/L)	Nanoparticle Size (nm)	Relative Change (A/A_0) in CO ₂ Dissolution
[22]	Y channel	1	300	30	100	40.85
[48]	Serpentine	5.5	27	35	100	78.30
[50]	Y channel	4	35	30	100	53
Present work	Y channel	4.5	35	28	5	77
					10	71
					20	43

5. Conclusions

This study investigates CO₂ dissolution using a microfluidic approach, integrating advanced image processing and nanoparticle synthesis techniques to enhance measurement accuracy and nanoparticle control. The primary objective is to demonstrate the effectiveness of polymer-coated NiNPs for CO₂ capture compared to a NiNP-polymer mixture, with the influence of NiNP size on dissolution efficiency. A microfluidic chip was designed to enable controlled bubble generation and dissolution, while automated image processing in MATLAB ensured precise bubble size measurements. Additionally, PVP polymer-coated NiNPs were synthesized via the polyol method, where temperature (130–180 °C) and reaction time (30–90 min) were adjusted to achieve precise nanoparticle sizes of 5, 10, and 20 nm. The PVP coating played a crucial role in stabilizing the nanoparticles, particularly for smaller particles, effectively preventing aggregation and enhancing dispersion. Experimental results demonstrated that smaller nanoparticles (5 nm) significantly improved CO₂ dissolution, reducing bubble diameter from 375 μm near the junction to 84 μm near the outlet, achieving a dissolution rate of 77%. This effect was attributed to the increased surface area-to-volume ratio and higher stability, as indicated by a higher zeta potential (~23 mV). In contrast, larger nanoparticles (10 and 20 nm) exhibited lower dissolution rates due to decreased zeta potential (19 and 13 mV, respectively), greater aggregation, and reduced surface interaction. These findings highlight the significant influence of nanoparticle size, stability, and surface modifications in optimizing CO₂ dissolution efficiency. This study demonstrates the potential of nanoparticle-based solutions for carbon capture and storage (CCS) applications. Future research should explore the impact of different nanoparticle compositions, surface functionalization strategies, and varying salinity and pH to further enhance dissolution efficiency. Additionally, scaling up this approach for industrial applications and investigating its long-term environmental impact would be valuable directions for continued study.

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