



Performance optimization of CO₂ mineralization: A comparative study of caustic soda and soda ash as pH-risers under high-pressure reactor

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Abstract

Aqueous mineral carbonation is one of the most important methods of permanent CO₂ sequestration in Carbon Capture, Utilization, and Storage (CCUS). This is carried out using alkaline pH-risers to neutralize the acidity of carbonic acid (H₂CO₃) to cause a change in the chemical balance to carbonate ions (CO₃²⁻). This paper assesses the different sources of alkalinity with special reference to the performance of caustic soda (NaOH) and soda ash (Na₂CO₃) under industrial circumstances. It has been shown in experiments that caustic soda is the most effective reagent in quick mineralization. It causes a sudden early increase in pH (about 2.0 to 2.5 units per gram) and maintains a very alkaline pH (pH ≥ 12), which are critical to the efficiency of CO₂ absorption (95.52 %). Moreover, NaOH has a remarkable stability at high pressure and the rate of change in pH (0.3 ΔpH/gm) does not change at the pressure of 65 bars. Soda ash on the other hand is a moderate buffer that reaches its highest pH at approximately 11.6, with an easily lower absorption efficiency of 72.45%. Soda ash performance improves as the pressure is increased to conserve pressure up to 65 bar, thus making it difficult to use it in high-pressure systems. Optimization of the industry shows that the efficiency is maximized within the temperature regime between 40°C and 60°C, where the accelerated reaction rates resulting decrease in the gas solubility. Furthermore, the best mass transfer rate is 300 rpm that minimizes the CO₂ bubble size. Though the stoichiometric superiority of caustic soda 1.0 kg does the work of 1.3 kg of soda ash, it presents severe challenges to operation because of its expensive nature, energy-consuming nature, and corrosivity. Soda ash is also still a feasible option in large scale sequestration due to its 30-40 % lower cost and the fact that it is dry and can be transported easily. Finally, a tradeoff between chemical reactivity, mechanical stability, and scalability of the economic side should be made even in the choice of a pH riser.

Keywords: CO₂; caustic soda; soda ash; mineralization; pH riser.

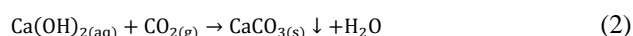
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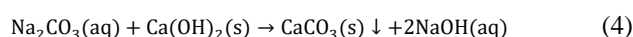
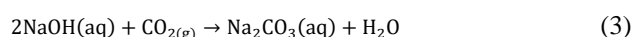
1- Introduction

The basic challenge of aqueous CO₂ mineralization is slowing rate of mineral dissolution and the acidity generated by the formation of H₂CO₃ [1]. To accelerate the precipitation of carbonates like calcite (CaCO₃) or magnesite (MgCO₃) [2, 3], pH-swing or pH-increase agents are employed [4] which substances provide the necessary hydroxyl ions (OH⁻) or neutralize protons (H⁺) to shift the inorganic carbon equilibrium toward carbonate ions (CO₃²⁻).

Oxides are among the most reactive agents for mineralization. Studies such as Sanna et al. [5] and Wehrung et al. [6] highlight that CaO and MgO, often sourced from the calcination of natural rocks, provide a rapid pH boost that facilitates near-instantaneous carbonation as shown in Eq. 1 and Eq. 2 [5]. While highly effective, the "carbon penalty" associated with the energy-intensive production of these oxides remains a primary concern in life cycle assessments (LCA) of the process.



Hydroxide is the standard in Direct Air Capture (DAC) and liquid scrubbing systems. Another study of (Zeman) [7] explored that the use of NaOH in high-volume spray towers, demonstrating that its high solubility allows for a rapid increase in pH to levels above 12, maximizing the mass transfer of CO₂ from gas to liquid as shown in Eq. 3 and Eq. 4 [7].



For large-scale, low-cost sequestration, research has shifted toward magnesium and calcium silicates., through the CarbFix project in Iceland, demonstrated that basaltic rocks and olivine (Mg₂SiO₄) act as natural pH buffers as shown in Eq. 5 and Eq. 6 [8]. As these minerals dissolve,

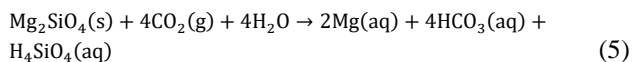


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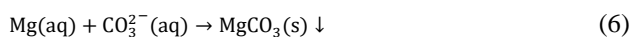
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they consume protons and release divalent cations. While the reaction kinetics are significantly slower than oxides, the abundance of these minerals makes them a cornerstone of Enhanced Rock Weathering (ERW) strategies. The utilization of industrial byproducts offers a dual benefit: waste stabilization and carbon storage.



Followed by the precipitation of Magnesite:



In this case, the Mg_2SiO_4 (Olivine) acts as the "base" that consumes the acidity (H^+) of the CO_2 .

The carbonation of steel converter slag [9], noting that its high content of $\text{Ca}(\text{OH})_2$ and C_2S (dicalcium silicate) provide a ready source of alkalinity as shown in Eq. 7 [10]. Similarly, studies on coal fly ash and cement kiln dust (CKD) show that these materials can raise pH effectively without the environmental cost of fresh mining, though the presence of heavy metals requires careful monitoring of the leachate.

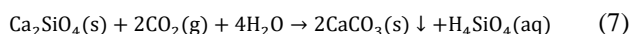


Table 1 summarizes all these pH-Boosting Agents, mechanism, scalability and references.

Table 1. Comparative evaluation of pH-boosting agents

Substance Class	Mechanism	Products and Scalability	Reference
Oxides	Rapidly provides OH^- and $\text{Ca}^{2+}/\text{Mg}^{2+}$	Calcite / magnesite high (industrial)	[5]
Hydroxides	Captures CO_2 into a liquid intermediate Consumes H^+	Sodium carbonate Medium (cost limited) Magnesium carbonate + silica very high (global)	[7]
Silicates	during slow dissolution	Precipitated	[8]
Industrial waste	Leaching of alkali phases	Calcium Carbonate (PCC) high (circular)	[9]

1.1. Factors affecting pH riser and product yield

1.1.1. Temperature

The classical thermodynamics suggests that CO_2 solubility decreases as temperature rises, industrial studies in 2025 confirm that elevated temperatures (up to 50–60°C) enhance the net CO_2 capture efficiency for NaOH solutions. This is because the increase in reaction rate constants and diffusion coefficients outweighs the loss in gas solubility which Optimal Range between 40°C to 60°C for industrial flue gas. At 50°C, NaOH solutions achieve significantly higher conversion to Na_2CO_3 compared to room temperature[11].

1.1.2. Alkali concentration and buffer capacity

The molarity of the pH riser directly impacts the "alkalinity flux" "the speed at which the solution can neutralize carbonic acid. Research shows that 30wt% NaOH provides the highest Na_2CO_3 yield, while lower concentrations (10wt%) lead to a rapid pH drops as stalling the mineralization process [11]. Na_2CO_3 solutions are inherently limited by their maximum pH (~11.6), making them less effective than NaOH (Ph >13) for capturing CO_2 from dilute sources like ambient air.

1.1.3. Stirring speed and mass transfer

Since CO_2 capture is a heterogeneous gas-liquid reaction, the physical dispersion of gas is a dominant factor. Increasing stirring speed from 200 to 300 rpm was found to be the most influential factor in enhancing CO_2 diffusion in pressurized systems [11]. Higher shear forces reduce the size of CO_2 bubbles, dramatically increasing the surface area available for the pH riser to react.

1.2. Caustic soda (NaOH) and soda ash (Na_2CO_3)

The selection of an alkaline pH riser is fundamentally a choice between ionic strength and equilibrium buffering capacity. The mechanism of caustic soda (NaOH) as a pH riser is characterized by its behavior as a strong, monoacidic base that undergoes near-complete dissociation in aqueous solution. Upon dissolution, it provides an immediate and high concentration of hydroxyl ions (OH^-), which rapidly shifts the inorganic carbon equilibrium. This high alkalinity facilitates the conversion of dissolved CO_2 into carbonate ions (CO_3^{2-}) via a two-step neutralization process: first forming bicarbonate (HCO_3^-) and subsequently carbonate as the pH exceeds 10. This mechanism is highly effective for "brute-force" mineralization where rapid cation precipitation is required, although it is susceptible to rapid pH drops as the OH^- ions are consumed during the reaction as Eq. 3.

Using NaOH to raise pH above 12 is necessary to maximize the concentration of CO_3^{2-} ions. If pH riser only reaches 8–9 (like Soda Ash or Bicarbonate), the carbon stays in the "Bicarbonate" form, which is harder to precipitate as a solid mineral. (Park and Fan) [12] showing that the reaction rate of CO_2 with calcium/magnesium is directly proportional to the pH level maintained by the alkali source. NaOH provides a faster reaction, but Na_2CO_3 is more cost-effective for maintaining a steady-state mineralization process over long durations[10]. The reaction between CO_2 and NaOH solution classified as a heterogeneous reaction and is governed by the diffusion-reaction mechanism as in Fig. 1. The rate of this reaction depends on how quickly CO_2 bubbles can diffuse into the NaOH solution[11]. Efficient interaction between CO_2 and NaOH occurs when the diffusion process is fast and unobstructed.

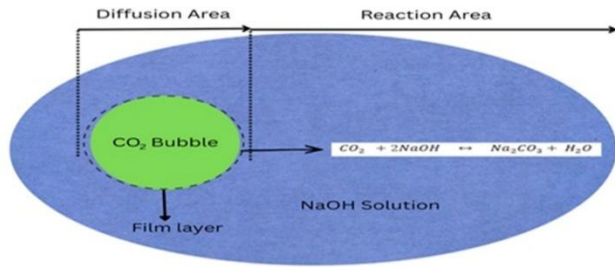


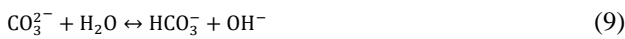
Fig. 1. Reaction mechanism between CO₂ and NaOH

When the reaction system conditions have reached steady state, diffusion rate will be equal to the reaction rate, the Eq. 8 [11] clarified this:

$$D \frac{dc_{CO_2}}{\delta} = k_1 C_{CO_2} C_{NaOH} - k_2 C_{Na_2CO_3} C_{H_2O} \quad (8)$$

where C_{CO_2} : concentration of CO₂ (mol/L); C_{NaOH} : concentration of NaOH (mol/L); $C_{Na_2CO_3}$: concentration of Na₂CO₃(mol/L); C_{H_2O} : concentration of H₂O (mol/L); δ : film layer (dm); D: diffusivity coefficient (dm²/s). $D (dc_{CO_2} / \delta)$: diffusion rate and reaction rate (mol/L. s). k_1 : reaction rate constant in the forward direction(L/(mol. s); k_2 : reverse reaction rate constant (L/(mol. s).

Conversely, the mechanism of soda ash (Na₂CO₃) operates through a hydrolysis-driven equilibrium rather than direct dissociation into hydroxide [13]. As a salt of a strong base and a weak acid, soda ash acts as a buffer. When dissolved, the carbonate ion reacts with water to produce bicarbonate and hydroxyl ions as shown in Eq. 9 [1]:



This results in a lower peak pH (typically 11.4–11.6) compared to caustic soda but provides a more sustained alkaline environment. The mechanism of soda ash in mineralization is particularly advantageous in "alkaline looping" systems; because it already contains the carbonate moiety, it can act as both a pH riser and a direct reagent for mineral precipitation. However, lower OH⁻ flux means that the mass transfer of CO₂ into the liquid phase is slower than in NaOH systems, which requires process intensification such as high-shear stirring or catalysts to achieve comparable sequestration rates as Eq. 4.

Various studies discuss pH risers like Simanjuntak et al. [14] demonstrated that direct contact between CO₂ in coconut shell combustion and a 7 M NaOH solution at room temperature and atmospheric pressure yielded 9.53 g/L of Na₂CO₃, the study of Ruiz et al. [15] utilized membrane separation which effectively isolate Na₂CO₃ from a mixture of NaOH and NaCl at temperatures between 20–40°C. Other studies focused on industrial waste streams, such as Guchhait et al. [16], who confirmed Na₂CO₃ formation from power plant flue gas using a lower NaOH concentration (0.1–1 M), which verified through SEM-EDS and XRD analysis. Kuliyeve et al. [17] investigated the reaction at specific pH levels (12.7–13.7) and flow rates (20–500 ml/min), identifying

NaHCO₃ as a persistent intermediate product. The study of Ghaffari et al. [18] focused on capturing CO₂ directly from the air by 6wt % NaOH concentration at 20°C optimized the mass transfer coefficient for Na₂CO₃ production. Beyond standard sodium hydroxide. Dalia et al. [19] indicated that an optimum CO₂ flow rate of 1.648 L/min and a 10 wt% concentration of Na₂SiO₃ resulted in a Na₂CO₃ yield of 25.63%. Matter et al. [8] found that keeping the pH high (above 9.0) is the optimum for mineralization. If the pH riser (like Soda Ash) allows the pH to dip below 8, the minerals (like calcium carbonate) may begin to redissolve back into the water. Alyousef et al. [20] showed that The addition of 0.6 wt% of sodium hydroxide (NaOH) to smart water elevates the pH, which profoundly alters zeta potential, shifting it from positive to negative in the presence of oil and This shift is critical for enhancing wettability alteration, promoting a water-wet state. Bennet et al. [21]operating at a controlled pH allowed for both complete reaction of available Ca²⁺ (100% carbonation potential or 9.9 g CO₂ .L-1 PW brine) and for reproducible control of the produced calcium carbonate (CaCO₃) product. Yard-scale experiments conducted in a 600 L continuously stirred tank reactor confirmed the selective precipitation of magnesium and calcium hydroxides under controlled pH regimes, consistent with the thermodynamic predictions. Comprehensive analytical characterization verified the near-complete removal of magnesium at pH 8–9, followed by calcium hydroxide formation above pH 10 [22].

The comparison between caustic soda and soda ash in different features, reaction speed and CO₂ adsorption capacity and efficiency summarized in Table 2.

Table 2. Comparison between caustic soda and soda ash

Feature	Caustic Soda (NaOH)	Soda Ash (Na ₂ CO ₃)
pH Level	Very high (~13-14)	Moderate (~11)
Molar mass	39.997 g/mol	105.988 g/mol
Appearance	White, opaque solid (flakes, pearls, or pellets)	White, odorless powder or granular crystals
Solubility	Highly soluble in water (1090 g/L at 20°C).	Moderately soluble (215 g/L at 20°C)
Reactivity	Aggressive / fast	Gentle / moderate
Cost	Expensive	Economical
Safety	High risk (corrosive)	Low Risk (irritant)
Max absorption efficiency	95.52%	72.45%
CO ₂ absorption capacity	1 mol CO ₂ per 2 mol NaOH	1 mol CO ₂ per 1 mol Na ₂ CO ₃

This study aims to evaluate and compare the effectiveness of different alkaline sources in CO₂ mineralization process through examining how varying pressures (from 45 to 65 bar) affect the ability of different alkaline agents to maintain target pH levels (pH ≥ 10) and balance the chemical efficiency of reagents against their cost, energy-intensive production, environmental footprints, and safety requirements.

2- Experimental setup and procedure

The process flow of CO₂ mineralization represents by Fig. 2 when the carbonated water was prepared by mixing 800 ml of produced water using a high-pressure, high-temperature (HPHT) mixing reactor of 1 liter in volume with CO₂ (purity 99%) operating at 500-600 RPM and 30°C for 1 hour using different range of pressures (45,50,55,60 and 65 bar)were tested to determine the optimum pH riser dose , which reaches pH ≥ 10 . Pressures 45–65 bar were evaluated. Selection of 50 bar was based on:

- 1- Achieving target pH equals 10.4 with economically reasonable NaOH consumption (6.1 gm/100 mL vs. >8 gm/100 mL at 65 bar) while in Soda ash didn't reach pH
- 2- Literature precedent from bennet et al. [21] and Zhu et al. [23] at similar pressures.
- 3- Equipment compatibility with standard pilot-scale HPHT systems. This multi-criteria optimization balances mineralization efficiency with practical field deployment considerations.

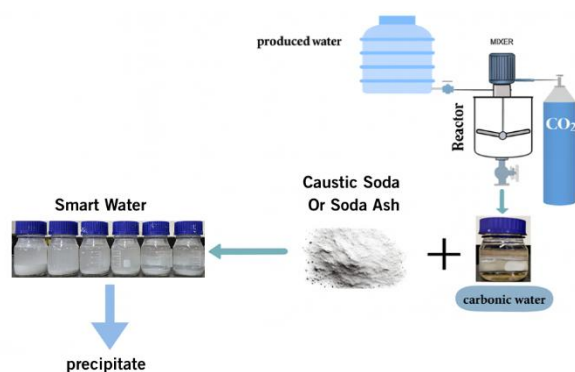


Fig. 2. CO₂ mineralization process flow

3- Results and discussion

The comparative effectiveness of two different alkaline agents soda ash (sodium carbonate) and caustic soda (sodium hydroxide) in raising the pH of a solution across various pressure conditions (45 to 65 bar) which illustrated in Fig. 3 .the circles in the figure which represent caustic soda additions achieve a rapid, steep increase in pH , just 2 grams of addition, the pH jumps from an acidic ~4.5 to a basic range of 8.0–9.0. By 8 grams, it pushes the solution into a highly alkaline state (pH 10.5–11.5). Squares represent soda ash, a weaker base which its curve is much flatter, indicating a high level of buffering. Even at 8 grams of addition, most Soda Ash samples barely reach a neutral pH (7.0) or remain slightly acidic/weakly alkaline (pH = 5.0–8.0).

Caustic soda (indicated by circles) is a significantly stronger base than soda ash (indicated by squares). Even at low additions, Caustic Soda pushes the pH into the alkaline range (pH=8–12) much faster. Caustic Soda: Shows a sharp initial jump in pH between 0 and 2 grams, followed by a more gradual, steady increase. Soda ash

Exhibits a more linear and moderate increase, suggesting it acts as a more controlled buffer compared to the aggressive pH shift of caustic soda. Caustic soda, higher pressures (like 55 and 60 bar) result in higher pH readings for the same amount of additive. Soda ash, the trend is more compressed, though the 45bar line (dark grey squares) surprisingly shows a higher pH response than the higher pressure variants at the same dosage. To calculate the specific rate of pH change, we can look at two distinct phases: the Initial Jump (from 0 to 2 grams) and the steady phase (from 2 to 12 grams). The rate is defined as the change in pH per gram of additive ($\Delta(\text{pH}/\text{gm})$). Caustic soda shows an extremely high rate of change, averaging 2.0 to 2.5 pH units/gm which indicates that lesser amounts of caustic soda immediately neutralize acidity and push the solution toward alkalinity. While soda ash shows a much lower initial rate, averaging 0.1 to 0.2 pH units/gm. It provides a much more gradual start. Steady pH rate (2-12) gm discussed in the Table 3.

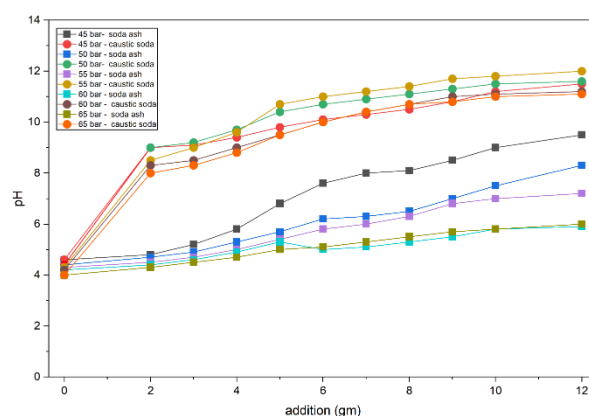


Fig. 3. Comparison between adding caustic soda and soda ash on pH

Table 3. Average rate of change ($\Delta(\text{pH}/\text{gm})$) during the steady phase (2–12 gm addition) highlighting the pressure sensitivity of Soda Ash vs. the stability of Caustic Soda

Pressure (bar)	Additive Type	Avg Rate ($\Delta\text{pH}/\text{gm}$)	Phase Observation
45 bar	Soda ash	0.47	Highest rate for soda ash; shows a jump between 4–6g.
45 bar	Caustic soda	0.3	Steady, linear increase.
55 bar	Soda ash	0.27	Moderate buffering.
55 bar	Caustic soda	0.3	Very consistent increase to pH 12.
65 bar	Soda ash	0.17	Least responsive; pH remains relatively acidic/neutral.
65 bar	Caustic soda	0.31	Maintains a strong alkaline trend despite high pressure.

4- Economic viability, scalability and operational difficulties

Caustic soda is highly efficient. Approximately 1.0 kg can achieve the same neutralization as 1.3 kg of soda ash which reduces the frequency of bulk deliveries and storage volume. However, as of early 2026, the price of caustic Soda remains volatile due to its dependence on the

energy-intensive chlor-alkali process. High electricity costs directly inflate prices, making it a "high-performance but high-risk" financial choice for long-term budgeting. Soda ash requires a higher volume by mass. Soda ash is generally 30-40% cheaper per ton than its caustic counterpart. For systems operating in the 45-bar range, soda ash provides a "safety buffer" economically; its gradual pH response prevents chemical overshoot which reduces the need for expensive secondary acid corrections to fix an over-alkalized solution.

Caustic soda scaling systems are complex due to temperature sensitivity. A 50% caustic solution begins to freeze at roughly 12°C–15°C, requiring large-scale facilities to invest in heat-traced piping and insulated tanks. Furthermore, while it is easier to automate via liquid dosing pumps, the capital expenditure for corrosion-resistant stainless steel. Soda ash is transported as a dry, non-hazardous powder, which simplifies long-distance logistics and storage. However, scalability is hampered by the need for "slaking" systems large tanks used to dissolve the powder into a liquid slurry before dosing. For high-pressure applications (65 bar), the mass of soda ash required to move the pH scale becomes so large that the physical size of the mixing equipment may become a bottleneck for the facility.

Caustic soda is a Class 8 corrosive material. Operational difficulty arises from mandatory high-level PPE, specialized operator training, and liability profile of the facility. Soda ash presents mechanical challenges, such as bridging storage silos and dust inhalation risks. At 65 bar, Soda ash shows a diminished capacity to raise pH compared to lower pressures.

5- Conclusion

The choice of a pH-riser agent is critical to the effectiveness and cost efficiency of CO₂ mineralization. Comparative evaluation of the caustic soda (NaOH) and soda ash (Na₂CO₃) reveals that caustic soda is the preferable agent in terms of mineralization by giving a quick rise in pH of 2.0 to 2.5 units per gram and attaining the necessary critical pH \geq 12. Soda ash, conversely, is a moderate buffer (with a maximum pH of about 11.4 - 11.6).

The highest efficiency of caustic soda absorption is 95.52% and that of soda ash is 72.454. In terms of sensitivity to pressure, caustic soda has a stable and consistent pH change rate regardless of the change in pressure, but soda ash has a lower ability to increase the pH at high pressure (65bar), which requires repeated recalibration.

The optimum range of NaOH industrial efficiency lies between 40 °C and 60 °C and in this range, the rate of reaction is higher and the loss of CO₂ solubility accompanying the reaction is less significant. In addition, stirring speeds should be increased up to 3000 rpm, which significantly enhances mass transfer by decreasing the size of CO₂ bubbles.

Economically and scalability wise, 1.0 kg of caustic soda can do the same as 1.3 kg of soda ash thus it needs a

greater mass volume. However, soda ash costs 30-40% per ton cheaper compared to caustic soda. Caustic soda is also in temperature below \leq 15 °C sensitive substance, but soda ash is a mild irritant. Nonetheless, caustic soda is considered as being very dangerous and corrosive.

In high pressure and efficiently raised industrial applications, the caustic soda is favored due to its stability and high degree of aggressiveness, even though it is more expensive and involves strict safety measures. Using an option to use as an inexpensive low-pressure system (around 45 bars), in which stage-by-stage pH regulation and cost-effective safe margins are more important.

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تحسين أداء عملية تمعدن ثاني أكسيد الكربون: دراسة مقارنة بين الصودا الكاوية و كربونات الصوديوم كمادة رافعة لدرجة الحموضة في مفاعل عالي الضغط

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الخلاصة

تعدّ الكربنة المعدنية المائية إحدى أهم طرق احتجاز ثاني أكسيد الكربون بشكل دائم في تقنية احتجاز الكربون واستخدامه وتخزينه (CCUS) وتُجرى هذه العملية باستخدام مواد قلوية لرفع درجة الحموضة لمعادلة حموضة حمض الكربونيك (H_2CO_3)، مما يحدث تغييراً في التوازن الكيميائي نحو أيونات الكربونات (CO_3^{2-}). تُقيّم هذه الورقة البحثية مصادر القلوية المختلفة، مع التركيز بشكل خاص على أداء الصودا الكاوية (NaOH) وكربونات الصوديوم (Na_2CO_3) في الظروف الصناعية. وقد أظهرت التجارب أن الصودا الكاوية هي الكاشف الأكثر فعالية في التمدن السريع، إذ تُسبب ارتفاعاً مبكراً ومفاجئاً في درجة الحموضة (حوالي ٢,٠ إلى ٢,٥ وحدة لكل غرام) لتحافظ على درجة حموضة قلوية عالية جداً ($pH \geq 12$)، وهو أمر بالغ الأهمية لكفاءة امتصاص ثاني أكسيد الكربون (٩٥,٥٢%) علاوة على ذلك، يتميز هيدروكسيد الصوديوم (NaOH) بثبات ملحوظ عند الضغط العالي، ولا يتغير معدل تغير الرقم الهيدروجيني ($\Delta pH/0.3 \text{ gm}$) عند ضغط ٦٥ بار. أما كربونات الصوديوم، فهو مادة عازلة متوسطة تصل إلى أعلى قيمة للرقم الهيدروجيني عند حوالي ١١,٦، مع كفاءة امتصاص منخفضة نسبياً تبلغ (٧٢,٤٥%) يتحسن أداء كربونات الصوديوم مع زيادة الضغط للحفاظ عليه حتى ٦٥ بار، مما يجعل استخدامه صعباً في أنظمة الضغط العالي. تُظهر الدراسات الصناعية أن الكفاءة تُحقق أقصى قدر من الكفاءة ضمن نطاق درجة الحرارة بين ٤٠ و ٦٠ درجة مئوية، حيث تؤدي معدلات التفاعل المتسارعة إلى انخفاض نوبان الغاز. كما أن أفضل معدل لنقل الكتلة هو ٣٠٠ دورة في الدقيقة، مما يقلل من حجم فقاعات ثاني أكسيد الكربون. على الرغم من التفوق الكمي للصودا الكاوية (١,٠ كجم) مقارنةً بـ ١,٣ كجم من كربونات الصوديوم، إلا أن ذلك يُمثل تحديات كبيرة في التشغيل نظراً لارتفاع تكلفتها واستهلاكها للطاقة وقابليتها للتآكل. مع ذلك، لا يزال كربونات الصوديوم خياراً مُجدياً في عمليات العزل واسعة النطاق نظراً لانخفاض تكلفته بنسبة ٣٠-٤٠%، وجفافه وسهولة نقله. أخيراً، يجب الموازنة بين التفاعل الكيميائي والاستقرار الميكانيكي وقابلية التوسع من الناحية الاقتصادية حتى عند اختيار رافع الرقم الهيدروجيني.

الكلمات الدالة: ثنائي أكسيد الكربون، تمعدن، الصودا الكاوية، كربونات الصوديوم، رافعات الرقم الهيدروجيني.