

Trash to treasure: Seawater pretreatment by CO₂ mineral carbonation using brine pretreatment waste of soda ash plant as alkali source



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HIGHLIGHTS

- The application of trash to treasure of the brine pretreatment waste of soda ash plant
- A possible application of the waste for acidic gas absorption was investigated.
- Alkali-metal-based hydroxides can be used to assist in seawater decalcification by CO₂.
- Permit the use of moderate temperature and pressure conditions for mineral carbonation of CO₂

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ABSTRACT

Mineral carbonation is one of the ways to reduce carbon dioxide gas in the atmosphere, which has attracted industrial and academic interest for its permanent storage of CO₂ with no leakage of threat. This paper focuses on development of new energy-saving and economic alkali sources for seawater decalcification by mineral carbonation process, with which not only carbon dioxide could be sequestered, but also the calcium ions in the seawater could be removed. Magnesium hydroxide was firstly tested as the alkali source in a simple Ca²⁺, Mg²⁺//Cl⁻-H₂O system, showing the ability of assisting in the decalcification according to the monitor of calcium ions concentration in solution and the analytical results of the sediments using XRD and SEM. This innovative approach permits the use of moderate temperature and pressure conditions in the process of seawater decalcification with mineral carbonation and the production of pure CaCO₃ with a potential sale value. Then it is inspired that the brine pretreatment waste of soda ash plant, mainly containing Mg(OH)₂ and CaCO₃, may act as the alkali source. Therefore, the seawater decalcification processes using the brine pretreatment waste of soda ash plant as alkali source by CO₂ were carried out.

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1. Introduction

The latest research reports by scientists in Germany, Spain, the United States and other countries of interdisciplinary research teams draw the conclusion that the sea level rise faster than expected through tracking sea level changes over the past 2800 years, and the rising sea level has already triggered floods all over the world [1–2]. The research team observed global warming is one of the leading causes of rising sea levels, and the increase of the concentration of pollutants in the atmosphere result in the melting of the glaciers and ice. As is well known, carbon dioxide is considered as the main source of the greenhouse gas emitted through human activities.

Carbon capture and storage (CCS) is considered as one of the effective methods of carbon emission reduction, which capture, compress and recycle the carbon dioxide released into the atmosphere to the depleted oil and gas fields or other safe underground places, and it has been attracted the attention of the researchers [3–4]. However, this method is of high cost of investment, which is estimated that carbon capture treatment costs about \$30 per tonne of carbon dioxide. Moreover, the transportation of carbon dioxide from power plants to safe place is a huge project. Carbon capture projects are also subject to economic cost and energy loss in the United States, European Union, Japan, Australia and other countries. Mineral carbonation technologies are widely drawn attention in recent years [5–7], as it not only alleviate CO₂ emissions, but also reduce the production cost, and then it is expected to become the most direct and effective means to deal with carbon dioxide crisis.

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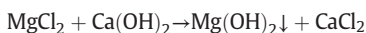
As proposed in previous study [8], CaCO_3 was controlled to precipitate using the carbon dioxide as precipitant by alkalization using NaOH to remove calcium ions and then avoid the fouling during desalination. In such process, carbon dioxide is captured by calcium ion in seawater and reacts with the help of alkali source. Meanwhile, the result showed calcium ions in seawater were effectively moved before desalination whereas magnesium ions were nearly removed, and then the ultimate decalcified sediments are only include the calcium carbonate, which indicate the waste carbon dioxide can be recycled as the valuable chemical—calcium carbonate, and the seawater is pretreated for seawater desalination.

Problems with water are expected to grow worse in the coming decades, with water scarcity occurring globally, even in regions currently considered water-rich [9]. Over the last decades, reverse osmosis (RO) membrane method has become the prevalent technology of water desalination for the increasing demand for freshwater [10]. Nevertheless, high energy consumption, low water recovery and membrane fouling restrain its development [11]. One of the causes of low water recovery is the existence of calcium scaling limiting the water recovery, for seawater is in rich of Ca^{2+} and SO_4^{2-} , and calcium sulfate is precipitated during seawater concentration process. Many researchers focus on the scaling process during seawater desalination and utilization, but seawater pretreatment nearly studied for not only calcium ions but also magnesium ions can lead to the scaling, if both calcium and magnesium ions are removed indiscriminately, the early treatment cost is quite high for the demand of amount of alkali source. Meanwhile, if calcium carbonate and magnesium carbonate formed during the desalination, acidification can help to decrease the scaling. However, remaining scale is very difficult to remove as calcium sulfate is also deposited during the process and accumulated over a long period.

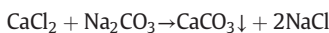
Herein, how to achieve the single decalcification is our focus, as a decalcification pretreatment will prevent the scale formation and increase the desalination performance. However, if sodium hydroxide is used as alkali source, the cost is relatively high, which will restrict the industrial application of seawater decalcification technology using flue gas as precipitant. In the recently published paper [12], the decalcification pretreatment process is proved to be a complex reaction involving first formation of $\text{Mg}(\text{OH})_2$ and $\text{Ca}(\text{OH})_2$ before CO_2 is injected, and then the dissolving of $\text{Mg}(\text{OH})_2$ and $\text{Ca}(\text{OH})_2$ but crystallizing of CaCO_3 with the continuous influx of CO_2 . The material conversion process indicated that the reaction essence probably is $\text{Mg}(\text{OH})_2$ played the role of alkali source.

However, the solubility of magnesium hydroxide is very low [13], and it should be testified whether it is effective to be used as the source of hydroxide for the precipitant of calcium carbonate. Therefore, a series of experiments and analyses have been conducted, including the material conversion of precipitate, Ca^{2+} , Mg^{2+} , CO_3^{2-} , HCO_3^- , and OH^- ion concentrations and pH in solutions during the carbonation of seawater with magnesium hydroxide.

In the further research, it is found that brine pretreatment waste of soda ash plant may be recycled as alkali source. In soda ash production through Solvay process, the first step is brine preparation, as sodium chloride solutions obtained by dissolving salt deposits usually contain a low concentration of impurities such as magnesium and calcium salts, which can cause the scaling of processing equipment and contamination of the product. The 'lime and soda' method is generally used to purify raw brine from calcium and magnesium ions [14–15]. The precipitation of magnesium ions in the form of $\text{Mg}(\text{OH})_2$ proceeds under the influence of lime milk.



The calcium ions present in brine as well as those introduced in the lime milk are precipitated with a sodium carbonate (soda) solution.



The suspension formed during this process creates a problematic waste product ($\text{Mg}(\text{OH})_2/\text{CaCO}_3$), which was carried away into sediment ponds, contaminating the environment [14]. Since the waste stream mainly contains $\text{Mg}(\text{OH})_2$ and CaCO_3 , it is speculated that the suspension may be used as the alkali source and it is named the brine pretreatment waste of soda ash plant (hereinafter referred to as white clay) in this paper.

Consequently, a possible application of $\text{Mg}(\text{OH})_2$ and white clay for acidic gas absorption was investigated in the present study, to explore the new method of carbon sequestration and decalcification of seawater at lower cost, while at the same time minimizing the damage on the environment. If $\text{Mg}(\text{OH})_2$ can be used to replace NaOH as the alkali source in the seawater decalcification by CO_2 , the cost for seawater decalcification would be accepted by the companies of seawater desalination or seawater utilization, meanwhile, this technology can realize the carbon dioxide fixation under moderate conditions. The further application of the brine pretreatment waste of soda ash plant makes trash to treasure, and expand the research scope of the alkali source.

2. Materials and methods

2.1. Materials

The white clay used in this study was obtained from a limited company to produce soda ash in North China, and was used as received. The major constituents were CaCO_3 , CaSO_4 , $\text{Ca}(\text{OH})_2$, $\text{Mg}(\text{OH})_2$, CaCl_2 , NaCl and combined water, while the major impurities were SiO_2 , Al_2O_3 , and Fe_2O_3 . The detailed composition is of 4.98% magnesium, 11.23% calcium and 32.12% water. The other analytical chemicals with purity above 99.0% were purchased from chemical agent companies in Tianjin.

2.2. Experimental unit

The experimental unit of seawater pretreatment by carbon sequestration using magnesium hydroxide as alkali source is the same with the unit represented in previous literature [12], and experimental parameters were simultaneously measured, including pH, calcium concentration [Ca^{2+}], magnesium concentration [Mg^{2+}], and CO_3^{2-} , HCO_3^- , and OH^- ion concentrations. Sediment characterization was performed by X-ray diffraction analysis (XRD, Bruker) and scanning electron microscope (SEM, FEI NanoSEM 450). In this experiment, the studied solution is prepared only using calcium chloride and magnesium chloride and the detailed composition is presented in Table 1.

The experimental unit of seawater decalcification pretreatment by carbon sequestration using white clay as alkali source is drawn in Fig. 1. A blade stirrer (10) was installed at the center of the mixer (9) to agitate the slurry, and the slurry was transferred into a glass column (6) by a magnetic drive pump (8). The seawater decalcification pretreatment was conducted in the glass column with a diameter of 200 mm at room temperature and atmospheric pressure. A pH meter (5) was also installed to monitor the progress of the decalcification reaction in the glass column. CO_2 (1) was input into the surge flask (2) firstly and then injected into the glass column quantitatively through a rotameter (3). Meanwhile, air was generated by air generator and also injected into the glass column quantitatively through a rotameter (4). The volume ratio of CO_2 and air was 1:9, the gas was well dispersed

Table 1

Chemical composition of solution used in decalcification by mineral carbonation using magnesium hydroxide as alkali source.

Ions	Concentration (g/L)
Mg^{2+}	1.24
Ca^{2+}	0.41
Cl^-	4.51

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