

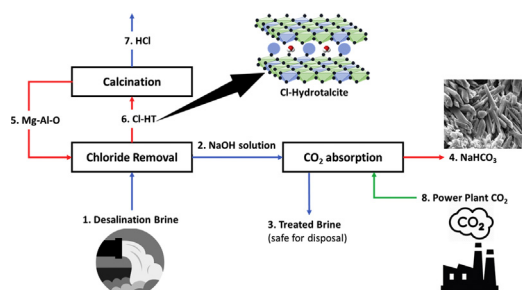
# A process for combined CO<sub>2</sub> utilization and treatment of desalination reject brine



Abdallah Dindi, Dang Viet Quang, Inas AlNashef, Mohammad R.M. Abu-Zahra\*

Department of Chemical Engineering, Khalifa University of Science and Technology, Masdar city Campus, P.O. Box 54224, Abu Dhabi, United Arab Emirates

## GRAPHICAL ABSTRACT



## ARTICLE INFO

### Keywords:

Desalination  
Reject brine  
Hydroxalites  
Mg-Al-O  
CO<sub>2</sub> utilization  
NaHCO<sub>3</sub>

## ABSTRACT

One of the problems facing the desalination industry is the management of concentrated brine wastes which constitute an environmental pollution when disposed into surface water bodies. This problem is huge in regions where desalination is heavily relied upon for potable water. In this work, a process is proposed for the first time, which could make these brines safer for disposal in combination with CO<sub>2</sub> capture and utilization for the production of chemicals like NaHCO<sub>3</sub>. Revenues from the sale of this product could offset some of the high costs which have hampered the growth of Carbon Capture, Utilization and Storage (CCUS) technology. As such, the process tackles two environmental problems while generating a valuable product. Results of the process evaluation are promising on account of the high CO<sub>2</sub> absorption capacity of about, 1.86 mol CO<sub>2</sub>/kg carbonated solution, NaHCO<sub>3</sub> yield of 44 g/kg carbonated solution, and a 70% and 20% overall reduction in the brine chloride and sodium concentration respectively, making it much less saline and safer to dispose into the sea.

## 1. Introduction

In arid regions around the world, desalination is heavily relied upon for the production of potable water, making up over 90% of the total water supply in some regions [1,2]. On account of this, a huge volume of concentrated brines is often disposed into surface water bodies where they pose threat to marine life, due to increased salinity and reduced levels of dissolved oxygen [1,3]. As fresh water resources get depleted and demand for potable water increases globally [4], technologies like

sea water desalination will increasingly be employed to produce water, resulting in the disposal of even larger volumes of concentrated brine into the sea with its attendant environmental impact. As such, the treatment of concentrated brine wastes before disposal into water bodies is a pressing environmental need.

CO<sub>2</sub> utilization can be used to mitigate the impact of disposing concentrated brines into water bodies. By reacting CO<sub>2</sub> from power plants with cations in the brine, it is possible to make carbonates which can be used as building aggregates or industrial feedstock chemicals,

\* Corresponding author.

E-mail address: [mabuzahra@masdar.ac.ae](mailto:mabuzahra@masdar.ac.ae) (M.R.M. Abu-Zahra).

thereby reducing the salinity of the brine in the process. The produced chemicals can then be sold to generate revenue. A major cause of the slow progress of Carbon Capture and Storage (CCS) is the high costs which have deterred investment in the technology. As the window of opportunity to avoid the danger of climate change narrows, it has become more important than ever to accelerate the deployment of technologies which can facilitate rapid reduction of global CO<sub>2</sub> emissions such as CCS. CO<sub>2</sub> utilization has been recognized as a possible means to achieve this. In fact, many CCS projects in the past few years have thrived as a result of CO<sub>2</sub> utilization [5]. Examples of CO<sub>2</sub> utilization options include, but are not limited to, enhanced oil recovery, urea manufacture and soda ash manufacture, and coal seam bed methane recovery [6–8].

While various options already exist for utilizing CO<sub>2</sub>, there is a need to harness new opportunities which may arise to promote the development of CCS technologies. CO<sub>2</sub> utilization with brine is particularly attractive among a number of strategies that have been proposed to address the problem of brine wastes from desalination plants, since it is able to tackle two environmental problems at the same time [9].

Earlier work in this regard often involves the use of the Solvay process, or its modifications, to accomplish CO<sub>2</sub> conversion into NaHCO<sub>3</sub> and the removal of Na<sup>+</sup> cations from the brine solution. El Naas et al. demonstrated the use of the conventional Solvay process for CO<sub>2</sub> capture and brine management [10,11]. Huang and coworkers showed a modified Solvay process which uses a sterically hindered amine, Methyl aminoethanol (MAE), in the place of ammonia with good results [12]. Also, our group reported the enhanced performance of 2-amino, 2-methyl propanol (AMP) used in a modified Solvay process for simultaneous brine treatment and CO<sub>2</sub> conversion [13,14]. A drawback of this approach, however, is the inability to regenerate amine-chloride produced during the carbonation step of the process. While the conventional Solvay process uses CaO to recover NH<sub>3</sub> from NH<sub>4</sub>Cl, the use of lime results in additional CO<sub>2</sub> emissions because for every ton of CaO produced, 2 tons of CO<sub>2</sub> is released [12]. Also, when amines are used, there is potential degradation which may lead to formation of toxic species that could further contaminate the brine. These issues make the modified Solvay process a less desirable option for brine treatment and CO<sub>2</sub> capture.

In this work, a novel approach to this problem is proposed which makes use of mixed metal oxides derived from the calcination of Mg-Al layered double hydroxide (LDH). LDHs are a class of anionic clays whose structures are similar to the those found in the naturally occurring mineral, hydrotalcite, earning them the name hydrotalcite-like materials (HT) [15]. As shown in Fig. 1, they are made of alternating layers of positively charged di or tri-valent cations and negatively charge balancing anions with the formula  $[M_{1-x}^{2+} M_x^{3+} (OH)_2]_x + (A^-)_x /$

$n\text{-mH}_2\text{O}$ , where M<sup>2+</sup> represents divalent cations like Mg, Fe, Co, Cu, Ni, or Zn, M<sup>3+</sup> represents trivalent cations like Al, Cr, Ga, Mn or Fe, and A<sup>-</sup> is the interlayer anion such as CO<sub>3</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>, OH<sup>-</sup>. The value of x is equal to the molar ratio of M<sup>2+</sup>/(M<sup>2+</sup> + M<sup>3+</sup>) and is generally in the range 0.2–0.33.

HTs often bind with CO<sub>3</sub><sup>2-</sup> to give a CO<sub>3</sub><sup>2-</sup>-HT, a poor ion exchanger, due to the strong affinity of the HT to CO<sub>3</sub><sup>2-</sup> ion. However, when calcined, they release CO<sub>2</sub> to form mixed-metal oxides which can reconstruct the original HT structure when exposed to an aqueous solution of the appropriate anion. This forms the basis of the good anion sorption properties observed for mixed-metal oxides [16,17]. Several workers have reported the use of such HT-derived mixed-metal oxides for the removal of chloride ions from wastewater. Kameda and coworkers used Mg-Al-O synthesized from a CO<sub>3</sub>-LDH for the removal of Cl<sup>-</sup> from various salt solutions with excellent results [18,19]. About 97% of the chloride could be removed from an HCl solution while 87% was removed from a NaCl solution [19]. Similarly, high removal rates were observed when they applied the mixed metal oxide for the simultaneous removal of Cl<sup>-</sup> and SO<sub>4</sub><sup>-</sup> from seawater [20]. In another study, Zhao reported a cyclic system for the removal of Cl<sup>-</sup> from waste water in which the Cl<sup>-</sup>-HT was regenerated with Na<sub>2</sub>CO<sub>3</sub> solution to form CO<sub>3</sub><sup>2-</sup>-HT which was then calcined to obtain the original Mg-Al mixed metal oxide [21].

Obviously, HT has high potential to remove Cl<sup>-</sup> from waste water, however, the exploitation of this functionality for treatment of waste brines combined with CO<sub>2</sub> capture and utilization process has never been investigated. Here, a novel process that combines CO<sub>2</sub> capture and utilization and reject-brine treatment simultaneously, using a mixed metal oxide obtained from Layered double hydroxide (LDH) was proposed and evaluated. The process partially treats reject brine and sequesters CO<sub>2</sub> by producing NaHCO<sub>3</sub> and Cl<sub>2</sub> which can be used to produce chlorine based products such as Hydrochloric acid. The proposed process has an advantage over earlier work because it can generate two chemical products and has the potential to treat the brine more extensively through the removal Na<sup>+</sup> and Cl<sup>-</sup> ions, while earlier processes could remove only Na<sup>+</sup>. In subsequent sections of this paper, the evaluation results of this process were reported through performance indices for brine treatment, expressed as overall chloride and sodium removal efficiencies, and for CO<sub>2</sub> capture and utilization, expressed as CO<sub>2</sub> absorption capacity and yield of NaHCO<sub>3</sub>, respectively. The ability of the Mg-Al-O to be used over several cycles of the process was also assessed and reported.

## 2. Process description

A schematic of the proposed process is given in Fig. 2. In the first

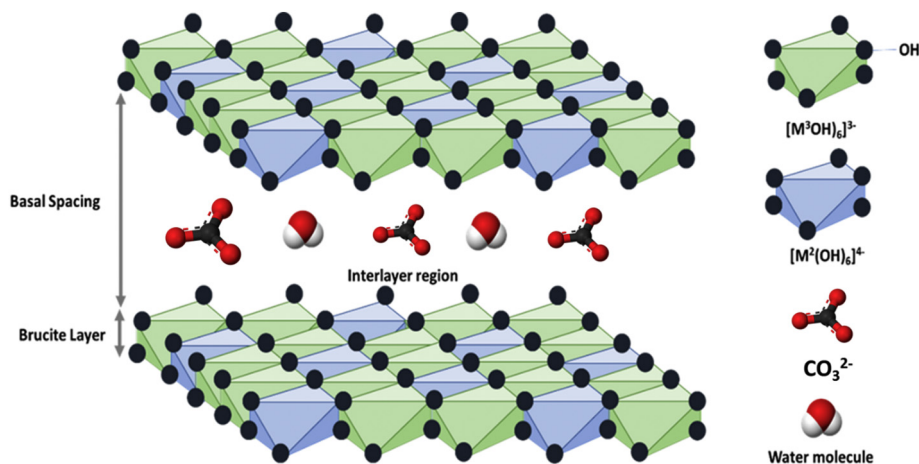


Fig. 1. Hydrotalcite structure.

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