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Progress in carbon dioxide sequestration via carbonation of aqueous saline wastes

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Abstract

In this article we report the progress made in CO₂ sequestration research *via* formation of synthetic carbonates and explore the engineering aspects of the proposed methodology. The approach to synthesize carbonates involves the reaction of a flux of CO₂ with Mg-chloride solution at room temperature. The kinetics of the carbonation reactions demonstrate that: a) in the experiments performed in solutions having both 7 and 16 g L⁻¹ of Mg the rapid formation of nesquehonite occurred; b) in the experiments with 32 g L⁻¹ of Mg an initial precipitation of chlorartinite was followed by the formation of nesquehonite and a minor amount of lansfordite. Carbonation *via* magnesium chloride aqueous solutions at standard conditions, here reported, represents a simple and permanent method of CO₂ trapping in solid form. It could be applied at point-sources of CO₂ emission and could involve rejected brines from desalination plants and other saline aqueous wastes. Other various aspects such as the influence of the salinity of the solution on the efficiency of carbonation and the kinetics of the reaction are also discussed.

Key words: carbonate synthesis; kinetics; reactor; carbon dioxide sequestration; saline aqueous waste.

Introduction

The 20th century has seen the explosive growth in energy consumption caused by the rapid increase of population and by the growth of industrialized countries, with the prospect of tripling the loading of CO₂ to the atmosphere by

the end of the 21st century. In this view, the scientific community is exploring methods for CO₂ sequestration. There are three options to reduce total CO₂ emissions into the atmosphere, i.e., to reduce energy and carbon intensity, and to enhance the sequestration of CO₂. The first option requires efficient use of energy. The

second option requires switching to using non-fossil fuels such as renewable energy sources. The third option involves the development of technologies to capture and sequester more CO₂ than today.

In this context, we describe here the encouraging results of our experiments on the reaction of CO₂ with Mg-chloride aqueous solutions through a rapid process whose solid products could be applied in several industrial processes (Botha & Strydom, 2001; Freitag & Kleinebudde, 2003; Cheng et al., 2009; Harrison, <http://www.tececo.com>). Moreover, there is little risk of unexpected release of CO₂ back to the atmosphere, because the formation of carbonates assures a safe and long-term CO₂ storage in the conditions that prevail at the Earth's surface (Frost et al., 2008; Ferrini et al., 2009; Ballirano et al., 2010).

In the system MgO–CO₂–H₂O at ambient conditions occur hydrous and hydroxyl metastable carbonates for which an evolution to more stable phases is documented (Langmuir, 1965; Davis and Bubela, 1973; Hopkinson et al., 2008, 2012).

Furthermore, here we report the engineering evolution of the experimental procedure started with a simple laboratory experimental set up, followed by the application of the carbonation process in a pilot plant (at laboratory scale) equipped with an electronic controller of pH conditions and a device to increase the partitioning of the CO₂ bubbles in the solution.

Background on approaches in CO₂ sequestration

In the last decade, efforts to accelerate technology development to provide options for the reduction of the levels of greenhouse gases (GHG) have been explored. Numerous approaches to CO₂ sequestration, including ocean, terrestrial, geological, biological and chemical options are today being studied (Seifritz,

1990; Lackner et al., 1995; Gunter et al., 1997; Holloway, 1997; O'Connor et al., 2002; Lackner, 2003; Xu et al., 2004; Metz et al., 2005; Hansen et al., 2005; Huijgen et al., 2006; Friedmann, 2007; Matter et al., 2009; Wilson et al., 2010) and the retention or sequestration of CO₂ in geological reservoirs is the option currently being applied (e.g., Holloway et al., 2007).

The carbon capture and storage (CCS) has the great advantage that, if applied to a modern conventional power plant, could reduce CO₂ emissions into the atmosphere by approximately 80-90% compared to a plant without CCS (Metz et al., 2005).

However, the common approaches to CCS reveal some drawbacks: 1) the capture and compression of CO₂ require much energy and would increase the fuel needs of a coal-fired plant with CCS by 25-40%, with an estimated increase of energy cost of 21-91% (Metz et al., 2005); 2) the application of CCS technologies to preexisting plants or plants far from a storage location would be very expensive. To reduce the costs the purpose-built plants should be realized close to a storage location. This option could reduce the level of CO₂ in the atmosphere significantly. However, about 20% of injected carbon can be expected to return to the atmosphere in about 300 years (Parson and Keith, 1998). Furthermore, long-term predictions of underground storage safety are very difficult. Studies are under way (e.g., Xu et al., 2004) to understand the behavior of CO₂ in porous and permeable media such as rocks, and the physical and chemical changes that can be expected if CO₂ is injected in these media.

Alternative technologies for CO₂ sequestration, such as neof ormation of minerals *via* reaction of CO₂ with Mg-Ca silicate rocks or of carbonate minerals in aqueous solutions, offer options for the permanent and safe storage of CO₂ in a solid form as hydrated carbonates (Ferrini et al., 2009; Ballirano et al., 2010; De Vito et al., 2012). In nature, the low temperature carbonates of Mg,

Ca, Na-Al occur as aggregates of acicular to fibrous crystals in several sedimentary sequences and in various depositional environments (e.g., associated with magmatic rocks and related to hydrothermal mineralizations; Baker et al., 1995; Ferrini et al., 2003; Sirbescu & Nabelek, 2003). Recently, a possible bio-chemical pathway has been considered in the crystallization of anhydrous carbonate at low-temperature, i.e., magnesite (Deelman, 2012).

The main objectives of CCS technologies *via* mineralization are: 1) to warrant the stable mineral trapping of CO₂; 2) to ensure the formation of materials whose disposal does not involve environmental damage; 3) to be easily applicable also in developing countries; 4) to preserve the biological ecosystem. The methods based on neoformation of carbonates virtually require cations to lock CO₂ (Lackner et al., 1995; O'Connor et al., 2002) in mineral structures and virtually ensure unlimited sequestration capacity. This idea was first proposed by Seifritz in 1990. The carbonation process occurs naturally in ultrabasic and ophiolitic rocks (Hansen et al., 2004; Wilson et al., 2006). Mg and Ca bearing silicates such as olivine, serpentine-group minerals and clinopyroxene could be sources of required alkalis. Actually, the industrial extraction of such cations involves expensive pre-processing with a significant environmental damage and increase of the treatments costs (Lackner, 2002). Furthermore, this option is not at all practical in many countries owing the paucity of exposed basic and ultrabasic rocks.

Conversely, the carbonation process involving the interaction of ions in aqueous solution with gaseous CO₂ could be an attractive solution. In this case, the precipitation of carbonates of Ca and Mg proceeds much more rapidly than if the cations are locked in a silicate structure. The source of such a process could be a brine (De Vito et al., 2011; Mignardi et al., 2011) and saline by-products of several industrial processes. The possibility to involve in the process saline

wastewater as source of Mg makes the method a promising complementary solution in the sequestration of anthropogenic CO₂ at the expense of saline wastewater (Veil et al., 2004; Veil and Puder, 2005). The process, a variant of the Solvay process (Lackner, 2002), is based on carbonating alkaline brines.

Finally, the sequestration method here reported produces a solid material that can be utilized directly in many products such as acoustic panels, non structural panels, insulation, or agglomerated in concrete as a manmade aggregate (Harrison, <http://www.tececo.com>).

Experimental

Syntheses

The experiments have been performed using Mg solutions having concentrations intended to simulate the salinity of the most common industrial wastewater (e.g., oil and gas produced water, reject brines, Veil et al., 2004; Veil and Puder, 2005; Lu et al., 2006; Kanagy et al., 2008).

In eighteen experiments, we synthesized carbonates using MgCl₂·6H₂O as a source of magnesium and bubbling gaseous CO₂ for 15–120 min at a rate of 100–250 mL min⁻¹ through the solution at temperature of 22 ± 2 °C.

The experiments can be grouped in the following two sets (Tables 1 and 2):

1) Static conditions: solution volume 200 and 3600 mL; Mg concentration ranging from 7 to 32 g L⁻¹;

2) Dynamic conditions: solution volume 7200 mL; Mg concentration ranging from 16 to < 28 g L⁻¹.

The time of interaction of the flux of CO₂ with the solution has been prolonged to the point of saturation of the solution in CO₂, as suggested by the stabilization of pH values. The pH, upon stopping the flux of CO₂, ranged between 5.0 and 6.8. Ammonia solution (25%, Merck p.a.) was used to adjust the pH of the solution in the range of carbonate precipitation, i.e., 7.8–8.9.

Table 1. Results of representative experiments and calculated efficiency of the carbonation process in static conditions.

Experiment	Temperature (°C)	Solution volume (mL)	Solid products formed	Residual Mg concentration (g L ⁻¹)	CO ₂ captured (%)
Initial Mg concentration (g L ⁻¹)					
7					
# 1	22 ± 2	200	N	< 0.5	78.3
# 2	22 ± 2	200	N	< 0.5	79.5
# 3	22 ± 2	200	N	~ 0.5	77.4
# 7	22 ± 2	200	N	< 0.35	82.9
# 9	22 ± 2	200	N	< 0.4	81.8
16					
# 11	22 ± 2	3600	N	< 1	73.2
# 14	22 ± 2	3600	N	< 1	74.8
# 15	22 ± 2	3600	N	< 1	76.2
32					
# 18	22 ± 2	3600	C - N	< 1.5	71.4
# 21	22 ± 2	3600	C - N	< 1	72.8
≤ 28					
# 26	22 ± 2	200	N	< 1	65.2
# 27	22 ± 2	200	N	< 1	63.2
# 28	22 ± 2	200	N ± L (~ 7%)	< 1	64.8

Abbreviations: N = nesquehonite, L = lansfordite, C = chlorartinite

Table 2. Results of representative experiments and calculated efficiency of the carbonation process in dynamic conditions.

Experiment	Temperature (°C)	Solution volume (mL)	Solid products formed	Residual Mg concentration (g L ⁻¹)	CO ₂ captured (%)
Initial Mg concentration (g L ⁻¹)					
16					
1d	22 ± 2	7200	N	< 1	71.5
2d	22 ± 2	7200	N	< 1	73.8
3d	22 ± 2	7200	N	< 1	75.7
≤ 28					
7d	22 ± 2	7200	N	< 1.5	66.0
8d	22 ± 2	7200	N	< 1	64.5

Abbreviations: N = nesquehonite

The reactors

The experiments of set 1, carried out in solution volume of 200 mL, have been performed in beakers and bottles, whereas those in solution volume of 3600 mL were executed in a small Plexiglas reactor (Figure 1).

The experiments of set 2 were carried out in a specific reactor with volume of 7200 mL. In the reactor the solution was kept under constant mechanic agitation to increase the partitioning of CO₂ bubbles. Moreover, to reduce the interaction time between gas and solution the rate of CO₂ flux through the solution has been increased.

The reactor consists in a Plexiglass tank having a screw-top where an electric engine drives a propeller which directly acts inside the solution (Figure 2). A small box collects the precipitate at the bottom of the tank. The propeller is inserted in a cylinder having some helixes to help the precipitate to fall down into the box.

The ammonia solution is injected in the Mg-chloride solution by an electric pump.

Analytical methods

The product of the syntheses was investigated morphologically by scanning electron microscopy

(SEM) using a FEI Quanta 400 operating at 30 kV.

X-ray powder-diffraction patterns, obtained using a Seifert diffractometer operating at 40 kV and 30 mA, were recorded from 5° to 60° 2θ at a rate of 0.02° per step and with a counting time of 8 s per step using Cu Kα radiation.

The Mg content in the residual solution was measured by inductively coupled plasma-atomic emission spectrometry (ICP-AES) using a Varian Vista RL CCD simultaneous spectrometer. The detection limit was 0.01 mg L⁻¹, and the analytical error was estimated to be in the order of 3%. The efficiency of the CO₂ mineralization process has been tested by measuring the amount of synthesized carbonates and the concentration of Mg in the residual solutions.

Results and Discussions

Synthesis and characterization of the carbonates

In static conditions (set 1, Table 1) the results of experiments showed that at low Mg concentration the reaction rate was rapid with carbonate deposition within 10 minutes (Figure 3). The XRD patterns of such solid products are

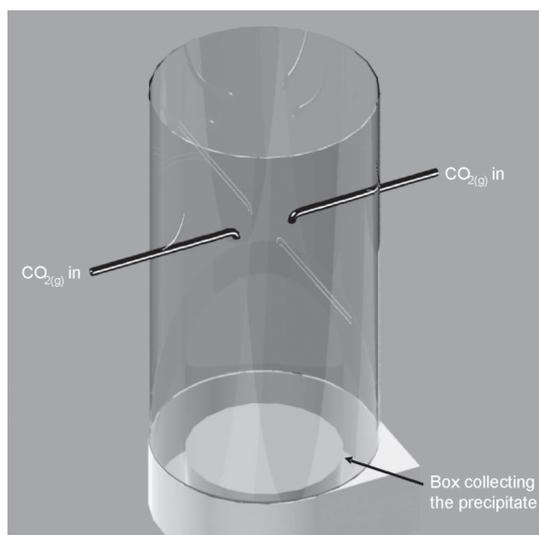


Figure 1. Reactor for carbonate synthesis: volume 3600 mL, static conditions.

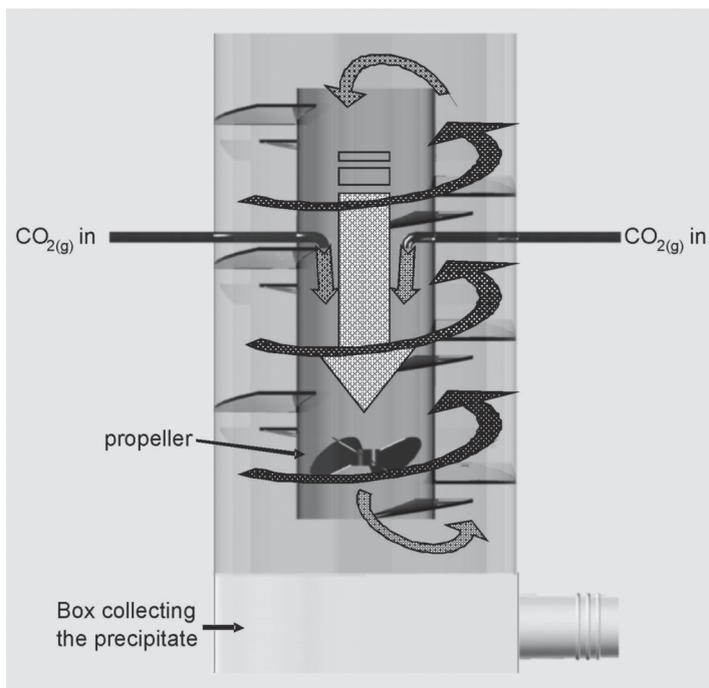


Figure 2. Reactor for carbonate synthesis: volume 7200 mL, dynamic conditions.

in agreement with those reported in JPCS card 20-669 for nesquehonite $\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$ (Figure 4a).

At higher Mg concentration (16 g L^{-1}), nesquehonite formed and the process has been monitored for about 10 days, showing that the Mg concentration in solution after two days was in the range of about 60%. At 32 g L^{-1} of Mg early formed chlorartinite $\text{Mg}_2(\text{CO}_3)\text{Cl}(\text{OH}) \cdot 3\text{H}_2\text{O}$ was followed by abundant precipitation of nesquehonite \pm lansfordite $\text{MgCO}_3 \cdot 5\text{H}_2\text{O}$ (Figure 4b, c).

The kinetics of such experiments highlighted that the reaction rate decreases with increasing salinity and in the residual solution the concentration of Mg was $< 20\%$ of the initial value after 4 and about 10 days for solutions having 16 and 32 g L^{-1} , respectively (Figure 3). On the basis of these results, to avoid the precipitation of a more soluble carbonate such as

chlorartinite, we performed experiments in solutions having Mg concentration $\leq 28 \text{ g L}^{-1}$. In these experiments only nesquehonite \pm lansfordite ($< 10\%$) formed and the reaction rate showed values intermediate with respect to that of the above experiments.

In dynamic conditions only nesquehonite crystallized. In these preliminary experiments the concentration of Mg in the residual solution, sampled at the end of each experiment, was close to that measured in static conditions.

The morphology of all the carbonates (nesquehonite \pm lansfordite) showed a predominant needle-like shape. However, the crystals grown in low concentrated solutions show a typical elongated habit (Figure 5a, b), whereas at higher Mg concentrations the higher nucleation rate determined the formation of more complex aggregates made of thin elongated crystals.

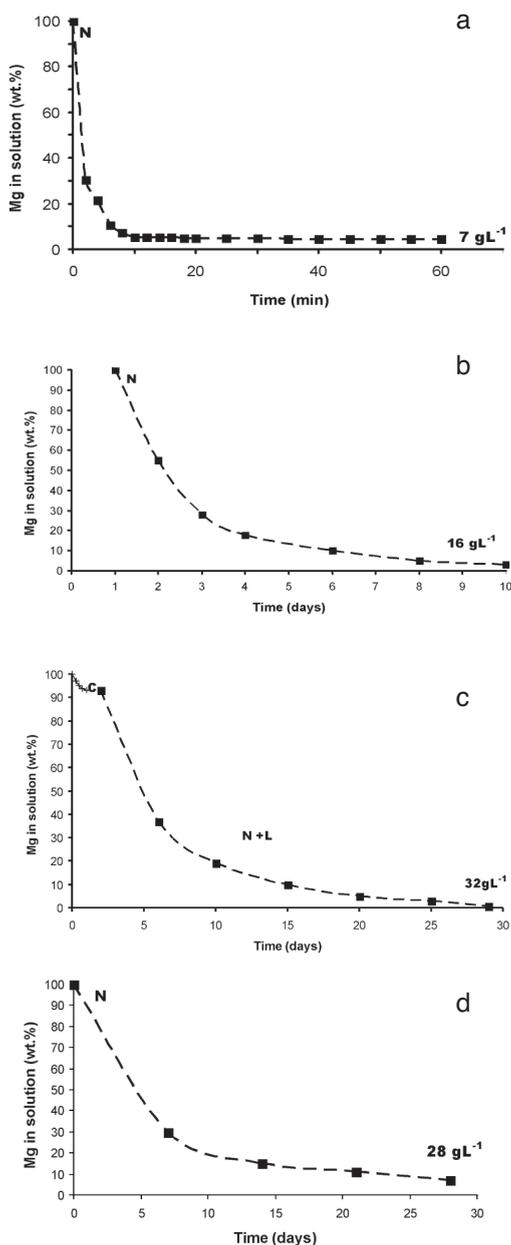


Figure 3. Kinetics of the carbonation reactions. (a) experiments showing the rapid formation of nesquehonite from solutions having 7 g L⁻¹ of Mg; (b) experiments with 16 g L⁻¹ of Mg showing the precipitation of nesquehonite; (c) experiments with 32 g L⁻¹ of Mg showing the initial precipitation of chlorartinite followed by nesquehonite with a minor amount of lansfordite and (d) experiments with ≤ 28 g L⁻¹ of Mg showing the precipitation of nesquehonite.

A large number of methods to synthesize solid magnesium carbonate hydrates have been developed with the aim to generate various morphologies because of their technological importance in different industrial applications (Botha & Strydom, 2001; Freitag & Kleinebudde, 2003; Cheng et al., 2009). In this view, our results can improve the knowledge about the experimental constrains which drive the morphologies of such carbonates.

Estimation of the efficiency of the process

The efficiency of the CO₂ mineralization process as been evaluated on the basis of Mg concentration in the residual solutions and the amount of synthesized carbonates. The results of this estimation are reported in Tables 1 and 2.

In both static and dynamic conditions the experimental results highlighted that the efficiency of the carbonation process decreases with the increasing salinity of the solutions (Figure 6). This result was expected as salinity is a main parameter affecting the solubility of CO₂ in aqueous solutions at ambient conditions (Yasunishi and Yoshida, 1979; Stumm and Morgan, 1996; Al-Anezi et al., 2008). The reduction of CO₂ solubility in concentrated solutions is known as “salting out effect” (Yasunishi and Yoshida, 1979; Al-Anezi et al., 2008). Our results showed that the reduction of the efficiency of CO₂ abatement in more concentrated solutions was lower than expected, probably due to the low degree of CO₂ release from such solutions. This limited release of CO₂ results in a long-term availability of carbonic ions to react with cations in solution to form carbonate (Grace and Piedrahita, 1994; Stumm and Morgan, 1996). A comparison between the

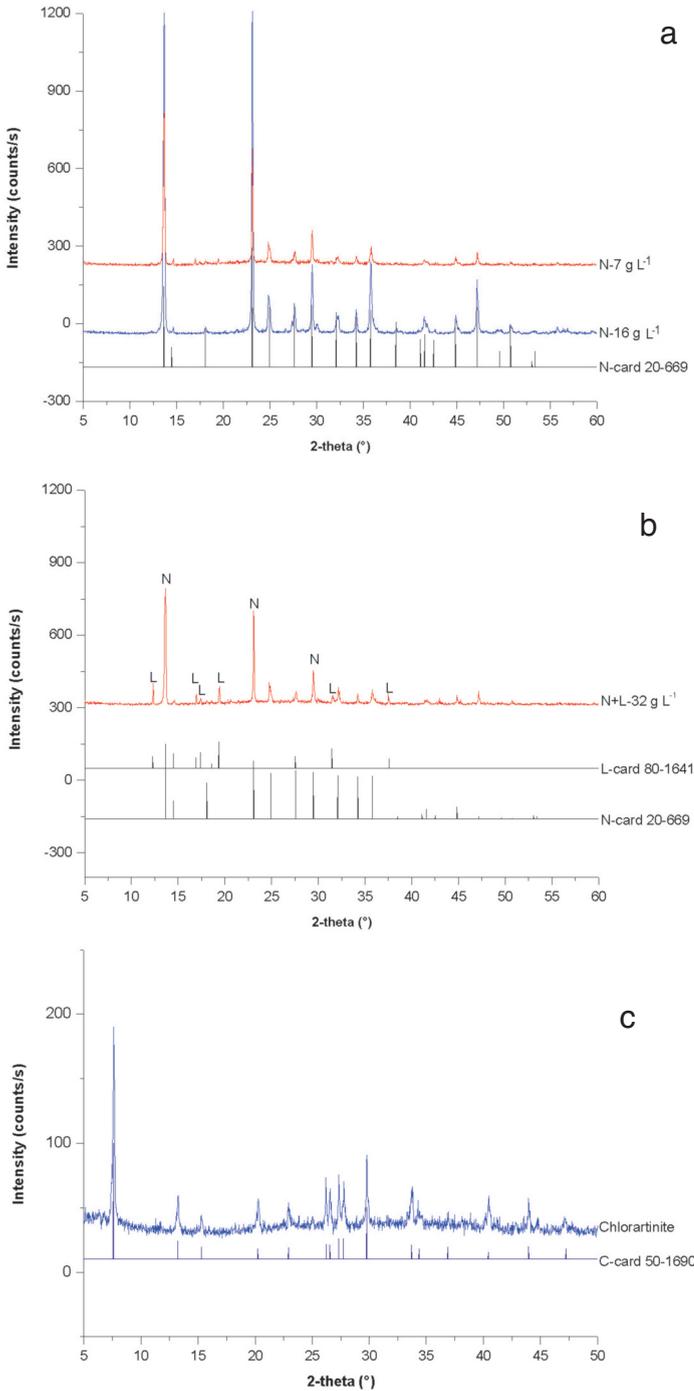


Figure 4. XRD patterns of carbonates synthesized at different Mg concentrations: (a) nesquehonite synthesized in the experiments with 7 and 16 g L⁻¹ of Mg; (b) mixture of nesquehonite and lansfordite (N > L) synthesized in the experiments with 32 g L⁻¹, precipitated after early formation of chlorartinite (c). For comparison the strongest lines recorded in JCPDS card (20-669, 80-1641, and 50-1690) of nesquehonite, lansfordite and chlorartinite have been reported. N=nesquehonite, L=lansfordite.

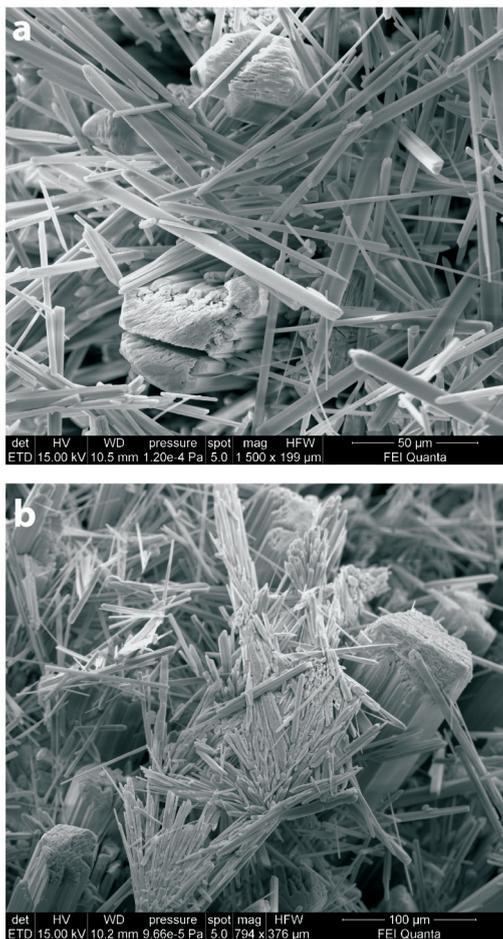


Figure 5. SEM images showing different morphologies depending on the experimental conditions: (a) needle-like morphology of interconnected crystals; (b) more complex aggregates of elongated crystals.

efficiency of CO₂ mineralization process in static and dynamic conditions showed a reduction of the efficiency of the process in dynamic conditions (Figure 6). To this reduction of the efficiency probably contributed also the mechanic agitation of the solution which favored a limited CO₂ release at the free surface of the

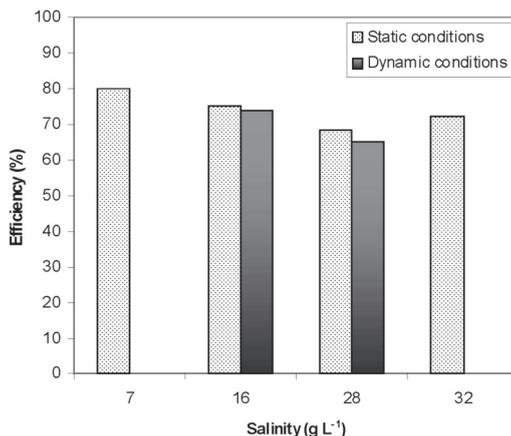


Figure 6. Efficiency of the carbonation process in static and dynamic conditions.

column of solution. Technical solutions are in progress to avoid this drawback. However, the advantages of our method in terms of efficiency of CO₂ removal, kinetics of the process, thermal and structural stability of the solid products (Ballirano et al., 2010) open a new window to the application of our method at industrial scale.

Concluding Remarks

The application of the carbonation method here proposed is desirable for the following advantages:

- the process is kinetically favored (e.g., the overall reaction is exothermic), simple and environmentally friendly;
- the comparison of the efficiency of the process in static and dynamic conditions showed a moderate reduction in these latter experiments;
- the resulting carbonates allow for the long-term storage of CO₂;
- the starting reactants are easy to be found because there exist enormous sources of Mg-enriched aqueous waste;
- the resulting carbonates could be used for

industrial and agricultural purposes, and their near surface or underground disposal involves no environmental damage; however, further investigation is needed for their application in these fields.

Further experimental work is ongoing by means of a pilot plant at laboratory scale in dynamic conditions for a thorough evaluation of the efficiency of the process before its application at industrial scale.

The application in situ of the proposed carbonation process will provide indications on costs, including energy, to produce nesquehonite capturing plant-emitted CO₂.

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