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Research paper

Mineral carbonation of ceramic brick at low pressure and room temperature. A simulation study for a superficial CO_2 store using a common clay as sealing material

Domingo Martín*, Patricia Aparicio, Emilio Galán

Departamento de Cristalografía, Mineralogía y Química Agrícola, Universidad de Sevilla, Spain

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ABSTRACT

This research explores the possibilities of CO_2 sequestration on ceramic bricks in a short time and at surface conditions. The experiment was carried out in a specially designed reaction chamber, filled with brick wastes and sealed with common clays. The brick used were composed of quartz, wollastonite, diopside, orthoclase and anhydrite, and the common clay was a marl composed of calcite, quartz, illite, smectite and kaolinite. Experimental condition in the reaction chamber were: reaction time 5 months, pressure of CO_2 0.5 bar, 4:1 solid/water ratio. The experiment was followed by XRD, XRF, BET, physical sorption by N_2 and CO_2, Hg porosity, TG-DTA, SEM and ICP-EOS.

After the CO_2 treatment, wollastonite and anhydrite were practically destroyed and some diopside and orthoclase. Calcite precipitated as new phase (up to 48 wt%), and small amount of illite was the result of orthoclase alteration.

Concerning the sealing clay, the CO_2 produced an increment of calcite content (from 32 to 41 wt%) and a partial destruction of smectite, particularly close to the upper part of the brick layer.

These results are hopeful in relation with the possible mineral carbonation of building ceramic waste in a short time at surface conditions, and open the opportunity to use those wastes for CO_2 trapping in an appropriate system, as a quarry reclamation.

1. Introduction

It is well known the urgent necessity for reducing the emission of anthropogenic greenhouse gases that produce dramatic effects on the Climate Change. Generally, human activities result in emissions of principal greenhouses gases (GHG): CO_2, CH_4, NO_x and halocarbons. All these GHG have increased from the pre-industrial era until nowadays, and particularly CO_2 increased around 40% as a direct consequence of the fossil fuel consumption and the change of soil use (Metz et al., 2005; Stocker et al., 2013). In order to avoid the potentially devastating consequences of global warning and climate change, the anthropogenic CO_2 emissions should be reduced. Main proposed measures lead to reduce the consumption of energy from burning fossil fuels and replacing them with “clean” energy such as renewables, and the use of technologies of capture, transport and storage of CO_2. Such strategy involves the development of innovate, available and cost-effective carbon capture and storage (CCS) technologies.

The storage of CO_2 may be carried out through a number of mechanisms, including mineral carbonation, oceanic storage, underground injection for enhance fossil fuel recovery, and injection into saline aquifers (Giammar et al., 2005; Metz et al., 2005).

Carbon dioxide sequestration by mineral carbonation mimics the naturally occurring rock weathering. The rock weathering has played an important role in the historical reduction of CO_2 concentration in the atmosphere. In this process, CO_2 and water play an important role to dissolve unstable rock-forming minerals releasing alkali and alkaline-earths cations and forming phyllosilicates and carbonates. The mineral carbonated process as an alternative for CO_2 sequestration was originally proposed by Seifritz (Seifritz, 1990). The principal benefit of this process is that the formed carbonates as final products are stable over geological time periods. However, as the weathering is a very slow process, the carbonation of silicates must be accelerated to be considered as a good CO_2 sequestration alternative.

Several investigations on magnesium- or calcium-based rock-forming minerals have been carried out as potential sources for fixing CO_2 by conversion to carbonated, e.g. olivine, serpentine and wollastonite, and, despite those silicates are not very abundant in the Earth, the mineralogical carbon sequestration could contribute significantly to

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* Corresponding author.
E-mail address: dmartin5@us.es (D. Martín).

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Table 1
Chemical composition of original and treated samples (wt%) by XRF; normalized at 100 wt%.

Sample	SiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	TiO_2	P_2O_5	SO_3	LOI	TOTAL
Brick	50.6	11.0	4.4	0.1	2.0	23.8	0.5	2.1	0.6	0.2	1.3	3.6	100.0
Marl	39.8	12.1	5.4	0.1	2.3	16.8	0.6	2.1	0.6	0.1	2.3	17.8	100.0
Marl 0–5 cm	41.2	12.4	5.0	0.1	2.3	16.3	0.7	2.1	0.6	0.2	1.8	17.3	100.0
Marl 5–10 cm	41.1	12.3	5.1	0.1	2.3	16.3	0.6	2.1	0.6	0.2	2.0	17.4	100.0
Marl 10–15 cm	40.8	12.3	4.9	0.1	2.3	16.5	0.6	2.1	0.6	0.2	1.7	17.9	100.0
Marl 15–20 cm	39.9	12.2	5.0	0.1	2.4	17.1	0.7	2.1	0.6	0.2	1.8	18.0	100.0
Marl 20–25 cm	38.7	11.8	4.7	0.1	2.4	18.5	0.6	2.1	0.6	0.2	1.4	18.9	100.0
Marl 25–30 cm	36.4	11.2	4.5	0.1	2.5	20.4	0.6	2.0	0.6	0.2	1.0	20.6	100.0
Brick layer (5 cm)	45.6	9.5	3.9	0.1	1.5	21.6	0.4	1.5	0.5	0.2	0.3	15.0	100.0
Marl at the bottom (5 cm)	40.2	12.0	4.8	0.1	2.2	17.1	0.6	2.0	0.6	0.2	1.6	18.8	100.0
Detection Limit (DL)	0.01	0.01	0.01	0.02	0.01	0.04	0.01	0.02	0.03	0.01	0.22		
Quantification Limit (QL)	0.02	0.02	0.02	0.03	0.02	0.05	0.03	0.03	0.10	0.02	0.23		
Relative Error	0.012	0.020	0.058	0.184	0.007	0.047	0.038	0.028	0.061	0.025	0.063		

LOI: Loss on ignition.

Table 2
Mineralogy composition of original and treated samples (wt%) by XRD.

Sample	Quartz	Wollastonite	Diopside	Orthoclase	Anhydrite	Calcite	Dolomite	Illite	Smectite	Kaolinite
Brick	15	35	25	18	7					
Marl	10					25	tr	17	34	13
Marl 15–20 cm	10					26	tr	18	32	13
Marl 20–25 cm	10					27	tr	19	29	14
Marl 25–30 cm	9					29	tr	19	28	14
Brick layer (5 cm)	18	6	9	7	2	48	tr	10		
Marl at the bottom (5 cm)	9					28	tr	19	29	14

tr: traces.

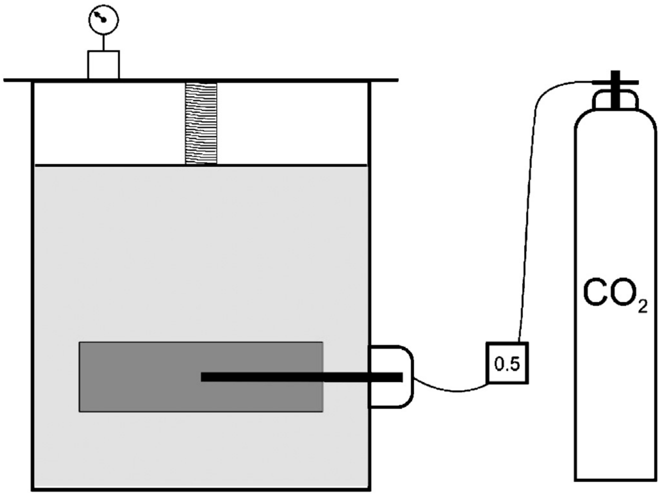

Fig. 1. Scheme of the reaction chamber designed for CO_2 brick reaction. In dark grey is represented the brick layer and in light grey the marl.

the CO_2 sequestration, mainly in the proximity of the emission source. This technology is called ex-situ mineral sequestration of CO_2 as it was proposed by Seifritz and studied in detail by Lackner and co-author (Lackner et al., 1995). The basic reactions, which take places after the Ca and Mg release, are exemplified by the following reactions:

$$CaO + CO_2 \rightarrow CaCO_3 + 179\ \text{kJ/mole} \quad (1)$$

$$MgO + CO_2 \rightarrow MgCO_3 + 118\ \text{kJ/mole} \quad (2)$$

Until now the experiments conducted for the carbonation of magnesium silicates (olivine and serpentine) via magnesium oxide or magnesium hydroxide intermediates, were carried out at high temperature and pressure, 600 °C and 100 bar (allowing for both sub- and supercritical conditions for CO_2) (Fagerlund et al., 2009), or under hydrothermal conditions, in the case of anorthite (Hangx and Spiers, 2009). But these conditions are not very economic, and they are weak points of the accelerated carbonation studies.

In addition, an industrial-scale operation may require the mining and grinding of suitable Mg- and Ca-bearing silicate minerals to accelerate the carbonation process, making the process not economically and environmentally viable.

For these reasons, several authors used other source of Mg and Ca such is the case of Mg- or Ca- containing mine tailings and by-products or waste from industry for carbonation. Such is the case of the carbonation of fly-ash (Montes-Hernandez et al., 2009), steel making slag (Huijgen et al., 2005), asbestos-mining tailings, electric arc furnace (EAF) dust, cement-kiln dust (Huntzinger et al., 2009), waste concrete (Shao et al., 2006), air pollution control (APC) residues (Baciocchi et al., 2006), etc.

This research explores the possibilities of CO_2 sequestration on ceramic bricks by mineral carbonation in a short time and at surface conditions. The results could be used for carbonation of ceramic wastes, at an industrial scale, particularly if such wastes were used as reclamation materials for filling exhausted quarries, in which the CO_2 could be injected. In this way, moreover the role of a possible sealing rock (common clay) was also investigated.

2. Materials and methods

2.1. Raw materials and carbonation experiment

The bricks (AUC1) used for carbonation come from La Puebla de Cazalla (Sevilla, Spain), and were fired at 900 °C. Such bricks were selected because of their high calcium content (Table 1), which can be releases after the CO_2 attack in presence of water. The brick is mainly composed of quartz, diopside, wollastonite and orthoclase, and minor anhydrite (Table 2). This brick was carbonated in an early study (Martín et al., 2016), producing calcite from the partial destruction of silicates and anhydrite. In that previous study, the carbonation was proportional to the reaction time and was independent of the particle-size fraction. The largest quantities of carbonates were obtained for the