

Editorial

Editorial for Special Issue “Geological and Mineralogical Sequestration of CO₂”

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Received: 16 June 2020; Accepted: 29 June 2020; Published: 2 July 2020



Carbon Capture Utilization and Storage (CCUS) has been substantiated by the International Panel on Climate Change (IPCC) [1] as a necessary measure to reduce greenhouse gas emissions in the short-to-medium term. Considered as a “climate change technology”, CCUS encompasses an integrated number of different technologies aimed at preventing large amounts of CO₂ from being further released into the atmosphere through the use of fossil fuels. Along with fuel switch, energy efficiency, and use of renewables, CCUS is thus currently considered a key option within the portfolio of approaches required to reduce greenhouse gas emissions.

CCUS basically involves (i) capturing CO₂ from stationary sources of C-gases, (ii) compressing and transporting it at the injection point, and (iii) injecting it in deep geological repositories (Geological Carbon Storage, GCS). Storage options include geological storage, ocean storage, and mineral carbonation [2]. Deep saline formations and depleted oil and gas reservoirs are currently envisaged as the most appropriate targets of geological storage.

Promising, alternative options to GCS, that guarantee a permanent, although on a smaller scale, capture of CO₂, are the in situ and ex-situ fixation of CO₂ in the form of inorganic carbonates by carbonation of mafic and ultramafic rocks and of Mg/Ca-rich fly ash, iron and steel slags, cement waste, and mine tailings [3–5]. Moreover, the industrial utilization of CO₂ as technical fluid for diverse applications in the field of material and chemical engineering may contribute to reduce CO₂ emission.

Since the late 1990s (e.g., Weyburn-Midale, 1996 [6], Sleipner, 2000 [7], projects in Canada and Norway, respectively), a number of large-scale CCUS facilities exist, mostly in North America, Europe, China, Australia and the Arabian Peninsula. Pilot projects are now growing in number around the world that are expected to evolve to an operational stage by the end of the 2020s, and further development is expected over the next years to help in accomplishing the ambitious task of keeping the increase in global average temperature below 1.5 °C [8]. From this perspective, key success factors will be the availability of financial incentives for deployment of CCUS technology, and the demonstrated technical and operational capability to effectively manage the risks of storage.

According to this general framework, this Special Issue assimilates contributions covering various aspects of recent scientific advances in CCUS, GCS in particular, that include the assessment of the storage potential of candidate injection sites, numerical modelling of geochemical–mineralogical reactions and CO₂ flow, studies of natural analogues, and experimental investigations of carbonation processes.

Wide-scale deployment of large storage projects over time requires a preliminary screening and ranking of the geological reservoirs for their suitability for storage. Following the recommendations of the international Carbon Sequestration Leadership Forum [9], an integrated assessment of the geological storage capacity of a prospect area requires integrating geological considerations with engineering, legal, regulatory, infrastructure, and general economic constraints.

The studies by Saftić et al. [10], Koukoulzas et al. [11] and Sundal and Hellevang [12] tackle this issue by providing geological information on the storage potential of selected areas in Croatia, Greece and Norway. In particular, Saftić et al. [10] performed a potential assessment of three prospect areas in the southern part of the Pannonian basin, in the Northern and Central Adriatic Sea, by taking advantage of a detailed stratigraphic knowledge of the Croatian territory derived from previous oil prospection activity. A ranking of the three prospect sites was finally proposed based on the integration of geological and infrastructure constraints.

Koukoulzas et al. [11] present a preliminary assessment of the trapping potential of two promising geological formations in the Volos area, Central Greece. The investigated lithologies are volcanic rocks (basalts and trachyandesitic lava flows) formed during the Pleistocene back-arc extension of the Aegean Sea. Based on their high porosity, low alteration grade, silica under-saturated alkaline composition, and the presence of Ca-bearing minerals, the authors estimated that the basalts of the Volos area have the appropriate physical-chemical characteristics to act as a storage reservoir, with a maximum capacity of about 110,000 tonnes of CO₂.

By combining reservoir geology, petrographic observation and geochemical modelling techniques, Sundal and Hellevang [12] assessed the storage capacity of a specific reservoir in the Northern Sea, Norway. In this study, the specific reactive areas of minerals used in the numerical simulations were proposed as an additional parameter to be considered for the geological characterization of the reservoir. The target of the study was the Johansen formation, a new CCUS prospect in Norway, licenced for the storage of CO₂ as of 2019.

A complex interplay of multiphase flow, diffusion, and chemical reactions is expected in the storage sites after injection of CO₂ deep underground. A wide range of homogeneous and heterogeneous reactions have the potential to significantly impact on both injection performance and storage security. In this framework, numerical modelling techniques emerge as an efficient tool to integrate fundamental research into the study of real-world complex processes. By applying reactive transport modelling techniques, Wasch and Koenen [13] set up a field-scale wellbore model aimed at predicting CO₂ leakage along possible fractures at the cement–rock interface. Contrasting evolutionary scenarios were predicted, primarily based on variable initial leakage rates considered in the model. Hypotheses were advanced about the most relevant parameters controlling the process of potential leakage, and to design leakage mitigation measures.

Understanding carbonation in natural systems provides constraints to develop efficient engineering strategies for CO₂ sequestration with both in situ and ex situ methodologies [2,4,14]. Following this approach, the studies of Boschi et al. [15] and Picazo et al. [16] furnish information on the processes and the physical-chemical conditions characterizing serpentinite replacement by carbonates in two different environments.

The research of Boschi et al. [15] is focussed on the spontaneous CO₂ mineral sequestration on serpentinite walls of the Montecastelli copper mine located in Southern Tuscany, Italy. On the basis of the analytical data of solid and liquid phases present in the mine and on geochemical modelling, the authors explain the process which triggered the formation of hydromagnesite and kerolite from the interaction of condensed mine waters and a layer of serpentinite powder accumulated on mine walls during the excavation of the mine adits.

Picazo et al. [16] studied the process of serpentinite replacement by carbonates in brecciated serpentinitized peridotites, recovered in the frame of the International Ocean Discovery Program (IODP) (site 1277), from the Newfoundland margin. The authors presented micro-textural, micro-chemical and O and C isotopic data. The analytical results coupled with a thermodynamic model of fluid/rock interaction during seawater transport in serpentine were utilized to constrain the most probable temperature condition of carbonation process and to discuss the role of temperature and seawater flows (i.e., influx vs. discharge) for the efficiency of CO₂ mineral sequestration.

Experimental studies are also fundamental to develop reliable methods for the mineralogical sequestration of CO₂ and to better understand the effectiveness and mechanisms of CO₂ geological

storage. The papers of De Brueil et al. [17], Martin et al. [18], Kim and Cho [19], and Park et al. [20] deal with this topic.

De Brueil et al. [17] examined the efficiency of the thermal activation of serpentine for mineral carbonation in the presence of the aqueous-phase at ambient temperature and moderate pressure in flue gas conditions. In particular, the study emphasizes the importance of amorphous phases, quantified by means a new original approach based on XRD analyses and Rietveld refinements, which formed during the dihydroxylation processes, and their role on the magnesium leaching during carbonation reaction.

The paper of Martin et al. [18] investigated the possibility to use ceramic construction waste (brick, concrete, tiles) for carbonation reactions. The proposed methodology includes two steps: a sample pre-selection based on in situ carbonation and the mineralogical and chemical characterization of the samples, and laboratory carbonation tests at room temperature and at relatively low-pressure on a brick selected according to the previous analysis. The study highlights the potential use of Ca-silicate-rich bricks as raw material for direct mineral carbonation under surface condition.

The study of Kim and Cho [19] deals with the possibility of storing CO₂ in marine unconsolidated sediments. In this case, CO₂ hydrates-bearing sediments, formed during the CO₂ liquid injection process, would act as cap rocks preventing leakage from the CO₂-stored layer. The feasibility of such a CO₂-storage method was experimentally examined and temperature, pressure, P-wave velocity, and electrical resistance were measured during the experiments. Minimum breakthrough pressure and maximum absolute permeability of CO₂ hydrate-bearing sediment were also estimated.

Park et al. [20] investigated the potential supercritical CO₂ storage capacity of conglomerate and sandstone and the sealing performance of the cap rocks (i.e., dacitic tuff and mudstone) in the Janggi Basin (Korea). To these aims, the authors presented the results of laboratory measurements of the amount of supercritical CO₂ replacing the pore water in each reservoir rock core and of the initial supercritical capillary entry pressure for the cap rocks. Moreover, they also examined the mineralogical changes of the cap rocks related to supercritical CO₂-water-rock reaction.

Acknowledgments: The authors thank the Editorial Board for their suggestions which improved the quality of this editorial.

Conflicts of Interest: The authors declare no conflict of interest.

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