

# Numerical modeling of self-limiting and self-enhancing caprock alteration induced by CO<sub>2</sub> storage in a depleted gas reservoir

Fabrizio Gherardi <sup>a,\*</sup>, Tianfu Xu <sup>b</sup>, Karsten Pruess <sup>b</sup>

<sup>a</sup> *Istituto di Geoscienze e Georisorse (IGG) – Consiglio Nazionale delle Ricerche (CNR), 56124 Pisa, Italy*

<sup>b</sup> *Earth Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, United States*

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## Abstract

This paper presents numerical simulations of reactive transport which may be induced in the caprock of an on-shore depleted gas reservoir by the geological sequestration of carbon dioxide. The objective is to verify that CO<sub>2</sub> geological disposal activities currently being planned for the study area are safe and do not induce any undesired environmental impact.

In our model, fluid flow, mass transport and mineral alteration are induced in the caprock by penetration of high CO<sub>2</sub> concentrations from the underlying reservoir, where it was assumed that large amounts of CO<sub>2</sub> have already been injected at depth. The main focus is on the potential effect of precipitation and dissolution processes on the sealing efficiency of caprock formations. Concerns that some leakage may occur in the investigated system arise because the seal is made up of potentially highly-reactive rocks, consisting of carbonate-rich shales (calcite+dolomite averaging up to more than 30% of solid volume fraction). Batch simulations and multi-dimensional 1D and 2D modeling have been used to investigate multicomponent geochemical processes. Numerical simulations account for multiphase advection, aqueous diffusion, fracture–matrix interactions (advective and diffusive exchange of species between fractures and matrix rock), gas phase participation in multiphase fluid flow and geochemical reactions, and kinetics of fluid–rock interactions.

The sensitivity of CO<sub>2</sub> concentrations to geochemical processes and parameters is investigated by conceptualizing different mass transport mechanisms (i.e. diffusion and mixed advection+diffusion). The most relevant mineralogical transformations occurring in the caprock are described, and the feedback of these geochemical processes on physical properties such as porosity is examined to evaluate how the sealing capacity of the caprock could evolve in time.

The simulations demonstrate that the occurrence of some gas leakage from the reservoir may have a strong influence on the geochemical evolution of the caprock. In fact, when a free CO<sub>2</sub>-dominated phase migrates into the caprock through pre-existing fractures, or through zones with high initial porosity acting as preferential flow paths for reservoir fluids, low pH values are predicted, accompanied by significant calcite dissolution and porosity enhancement. In contrast, when fluid–rock interactions occur under fully liquid-saturated conditions and a diffusion-controlled regime, pH will be buffered at higher values, and some calcite precipitation is predicted which leads to further sealing of the storage reservoir.

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\* Corresponding author. Tel.: +39 050 315 3409; fax: +39 050 315 2322.  
E-mail address: [f.gherardi@igg.cnr.it](mailto:f.gherardi@igg.cnr.it) (F. Gherardi).

## 1. Introduction

Geologic sequestration is being considered as an effective means for reducing atmospheric emissions of CO<sub>2</sub>. Modeling studies have suggested that under favorable conditions CO<sub>2</sub> can be confined for very long times (thousands of years; see, among others, Bachu et al., 1994; Weir et al., 1996a,b; Hitchon, 1996; White et al., 2005). Despite the great potential benefits of geologic sequestration, primarily because the CO<sub>2</sub> injected in the subsurface can be isolated from the atmosphere for long time periods, risks may nevertheless be present. In particular, CO<sub>2</sub> can leak from storage reservoirs and return to the atmosphere. This is because additional geochemical processes may take place as a consequence of dissolution of supercritical CO<sub>2</sub> into deep brines, which may affect the integrity of the storage reservoir (e.g. Gunter et al., 2000). Leakage may occur through fractures, faults or, more generally, caprock weaknesses which may act as preferential pathways for deep-seated gases (Ennis-King and Paterson, 2000; Pruess and Garcia, 2002; Rutqvist and Tsang, 2002).

Many experimental, modelling and field studies have already been conducted to evaluate the feasibility and integrity of geological carbon storage (see, among others, Lindeberg, 1997; Ennis-King and Paterson, 2000; Pruess and Garcia, 2002; Rutqvist and Tsang, 2002; Kaszuba et al., 2003; Hildebrand et al., 2004; Kaszuba et al., 2005; Gaus et al., 2005; Haszeldine et al., 2005; May, 2005; Marschall et al., 2005; Moore et al., 2005; Li et al., 2006, and references cited therein). From these studies, the following key issues can be identified, which need to be addressed for a quantitative assessment of the sealing efficiency of caprock formations:

- (i) to gain a better understanding of the fluid flow and geochemical processes that would accompany CO<sub>2</sub> migration from a storage reservoir towards shallow depths, it is crucial to explore and evaluate the impacts of dissolution processes of caprock formations by acidic CO<sub>2</sub>-rich fluids;
- (ii) reactions induced by CO<sub>2</sub> in rocks filled with brackish or saline waters likely involve acid hydrolysis of rock-forming minerals. Dissolution reactions are expected in regions with high  $P_{\text{CO}_2}$  in carbonate-rich rocks, with both the type and timing of the mineralogical alteration in caprocks being highly variable and specific to geochemical, physical and hydrologic features of the investigated systems;
- (iii) the sealing capacity of caprock formations above depleted oil and/or gas reservoirs can be significantly reduced after CO<sub>2</sub> injection, due to the lower interfacial tension of the CO<sub>2</sub>–water system in comparison to hydrocarbon–water system originally filling the reservoir before the displacement operated by the injected CO<sub>2</sub> (Li et al., 2006);
- (iv) experimental data on capillary displacement pressure and effective permeability–capillary pressure relationships indicate that dynamic leakage of gases through the caprock depends on ambient pressure conditions; differences in gas breakthrough behavior of gases are governed by interfacial tension and wettability behavior of different gases which are specific to different lithologies (Hildebrand et al., 2004);
- (v) natural analogues provide useful information on geochemical processes occurring in natural environments that laboratory experiments may not be able to reproduce. Such information can assist in the calibration of geochemical models, allowing them to make accurate predictions at the time-scales involved in nature (Haszeldine et al., 2005).

This paper is focused on the study of the reactive mechanisms which may occur as a consequence of CO<sub>2</sub> geological disposal at depth in a potentially highly-reactive caprock, consisting of carbonate-rich shales.

The site investigated here is a depleted on-shore gas reservoir in Northern Italy (Calabrese et al., 2005), in which prefeasibility studies have already identified favorable conditions for CO<sub>2</sub> injection and underground storage. The reservoir consists of a multilayered sandstone–shale formation located at depths generally below 1400 m b.g.l. (below ground level); simulations address the uppermost layer of this reservoir, where CO<sub>2</sub> injection is currently planned.

Available site-specific data include basic physical parameters (temperature, pressure, porosity, permeability, see Section 6), chemical composition of reservoir aqueous solution, and caprock and reservoir mineralogy. The chemical composition of the caprock pore water is unknown, and has been inferred by batch modeling (see below).

Gas–water–rock interactions resulting from CO<sub>2</sub> migration into the caprock have been simulated under dominance of two alternative mass transport conditions: diffusion in the aqueous phase, and gas and/or liquid advection. All simulations concern the geological disposal of an almost pure-CO<sub>2</sub> waste stream. The effects induced by the underground disposal of less pure waste streams, as expected for coal-fired power plants where SO<sub>x</sub>, NO<sub>x</sub>, and H<sub>2</sub>S gas species may be present along with CO<sub>2</sub> (Knauss et al., 2005), are not accounted for.

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