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Experimental investigation of capillary effects on solid-liquid interactions in porous media at the decimetric column scale

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The capillary state of water in porous media is of paramount importance at the caprock-aquifer interface to prevent any CO2 leakage. But it also significantly influences gas dissolution and solid mineralization reactions, crucial for CO2 trapping and earth science engineering. Capillarity-based geochemistry has been experimentally documented and quantified at the one-pore sale. Yet, capillary water exerts super-solubilizing effects on solids and gases [1], induces cold boiling, and modifies the stress field within the host matrix [2]. These effects must now be established experimentally at the upper core scale. At this thousands of pores scale, the effect(s) of capillarity competes with the dry conditions intrinsic to unsaturated media that limits the water availability, and with the evolution of the reactive surface area which depends on the liquid/air distribution and the passivation of solid surfaces through secondary phase precipitation. We are discussing the experimental design and first results to help unravel this interwoven sketch, which is much needed for accurate predictions in any long-term scenarios of aquifer-air 'tandem'technique (CO2 storage, nuclear wastes tunnels, energy storage, ⋯).

In this study, we examine the brucite (Mg(OH)2) carbonation in unsaturated columns flushed with a CO2 gas flow. The design varied from one experiment to the others by the initial water content, the gas flushing composition, the gas flow rate, and the granulometry of the column materials. These experiments provide simplified models for comprehending carbon mineralization reactions in partially water-saturated porous media, resembling conditions in unsaturated zones of natural systems or disturbed and industrial settings. The main goal of our investigation is to evaluate how capillary water impacts the precipitation of hydrous Mg-carbonates.

Experiments are performed in decimetric unsaturated columns filled with brucite grains and quartz sand (Figure 1). Capillary conditions are established by flushing the column with N2 air at low relative humidity (RH) illustrated below for one dataset (Stage 0, RH 50%, Figure 2). After stabilization, the gas composition is turned to CO2-H2O, initiating carbonation (hydromagnesite formation) by pore-scale interaction between CO2, liquid water, and brucite. The column mass variation shows a first decreasing stage linked to the evaporation of 15% of the initial water, before reaching a plateau, and then displaying an increasing stage related to the carbonation reaction. The CO2 breakthrough curve reveals three distinct stages (Figure 2). Stage 1 exhibits minimal CO2 at the column outlet, primarily due to bulk brucite carbonation. In Stage 2, a decline in reactive brucite abundance slowdowns the carbonation rate, leading to an increase in CO2 content in the effluent gas. Finally, Stage 3 demonstrates a CO2 concentration aligning with the supplied gas composition, signifying the cessation of the carbonation reaction. This is also evident by the stability of the column weight at this stage.

In summary, the evaporation process and the carbonation reaction occurring in an extended pore network at the column scale both appear sensitive to the capillary conditions in porous media which can grow and express oneself depending on various factors that will be discussed in this contribution.

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