

Water Redistribution and Carbonation in Drying Unsaturated Porous Media

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Evaporation is a key process in porous hydrosystems affected by human activities or climate-driven underground changes. Basically, it is influenced by complex interactions between the pore structure and external boundary conditions such as relative humidity [1]. Extensive drying not only reduces available pore water but also establishes capillary conditions that modify geochemical dynamics by influencing the driving forces of solid-gas-liquid interactions. [2]; yet these impacts remain largely unexplored at column-scale. In this study, we investigated how evaporation influences pore water redistribution and carbonation reactions in an unsaturated sand-brucite column (Fig. 1). A non-reactive gas with 50% relative humidity was injected from the bottom for over five weeks, and water evaporation was monitored using Time Domain Reflectometry (TDR) sensors and weight measurements. Gas injection initially caused evaporation from bottom and subsequent increasing saturation at top, while continued injection led to evaporation throughout the column, reducing total saturation from 30% to 5% (Fig. 2). At this stage, CO₂ was introduced into the injected gas, triggering brucite carbonation. The CO₂ breakthrough analysis showed less carbonation at the bottom due to limited water availability, while a stepwise trend in the breakthrough curve suggests a possible role of capillarity in CO₂ transport and reaction progression. A separate experiment at the same final saturation (5%) revealed that evaporation history significantly affects water redistribution and carbonation reaction. These findings highlight that water saturation during evaporation does not change in a linear or continuous manner, and the limited water availability induces capillary effects that influence the carbonation reaction. Understanding these effects is essential for predicting long-term processes in diverse subsurface applications such as arid soils management, or geothermal, CO₂ and energy storage systems.

[1] M. A. B. Zanoni, J. Wang, J. L. Torero, and J. I. Gerhard, "Multiphase modelling of water evaporation and condensation in an air-heated porous medium," *Appl Therm Eng*, vol. 212, p. 118516, Jul. 2022, doi: 10.1016/J.APPLTHERMALENG.2022.118516.

[2] C. Hulin and L. Mercury, "Capillarity-driven supersolubility in dual-porosity systems," *Geochim Cosmochim Acta*, vol. 252, pp. 144–158, May 2019, doi: 10.1016/J.GCA.2019.02.026.

