Fundamental study of CO_2 gas exsolution and flow in the shallow subsurface during leakage from a geologic sequestration site

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Abstract. Geologic sequestration of CO_2 can potentially reduce the effects of global warming by reducing the amount of greenhouse gases that are emitted into the atmosphere. However, leakage of sequestered CO_2 can cause significant impacts to groundwater resources and ecosystems, so the physiochemical processes that CO_2 undergoes in shallow subsurface environments are important to understand. When supercritical CO_2 leaks from a deep storage formation, it dissolves into the groundwater or brine into which it leaks. Conditions can exist for this dissolved CO_2 to migrate upward into the shallow subsurface, where it has the potential to come back out of solution (exsolve). Once enough exsolution has occurred, discrete gas phase migration pathways may develop through the system, at which time, two-phase flow of water and CO_2 gas ensues. An experimental study is underway which investigates the fundamental mechanisms of CO_2 gas exsolution and flow. Degassed, deionized water is saturated with CO_2 gas under a specified pressure (the saturation pressure) and then injected at the bottom of a 4.5 meter tall vertical column of highly characterized test sand that is initially saturated with water. The column is equipped with 12 soil moisture, electrical conductivity, and room temperature sensors, as well as 2 soil temperature sensors that all continuously monitor the physiochemical behavior of the system. Water pressure is monitored at 13 locations along the column using a physical multiplexer connected to a pressure transducer. At the top of the column, gas and liquid phase effluents are separated, the pH and CO_2 concentration of the liquid phase are measured continuously, and the mass outflow rate of each phase is monitored. To evalate the mechansms that contribute to dynamics of CO_2 gas exsolution and flow, a variety of homogeneous and heterogeneous packing configurations, saturation pressures, and injection rates were simulated in the test column. For all cases, both homogeneous and heterogeneous, 1) the rate of water outflow, which is initially equal to the inflow rate, increases when gas starts to exsolve, 2) after some time, discrete gas migration pathways develop, which cause the saturation values to stabilize, the gas phase outflow rate to increase, and the water outflow rate to decrease back to the inflow rate, 3) the water pressure distribution in the column is initially hydrostatic, then the pressure increases when injection begins, and increases further when gas phase develops, 4) the final vertical extent of gas phase corresponds to the saturation pressure (for example, when the saturation pressure is equal to 2 meters of water, gas will only exsolve in the top 2 meters of the column), and 5) the final, steady-state gas saturation always approaches 30-40%, regardless of packing, injection rate, or saturation pressure. These results indicate that, once gas exists in the column, there are two subsequent physiochemical processes. The first consists of kinetic mass transfer of CO_2 from the aqueous phase to the gaseous phase, and the second consists of steady two-phase flow of water and CO_2 gas. Results from experiments with homogeneous packings indicate that, 1) when the saturation pressure is greater than the hydrostatic water pressure at the injection port, gas exsolves first at the bottom of the column and the exsolution front coincides with the front of the migrating CO_2-satruated water, and that 2) when the saturation pressure is lower than the hydrostatic water pressure at the injection port, the CO_2-saturated water reaches the top of the column before any gas exsolves, then gas exsolves first at or near the top of the column and the exsolution front migrates downward. When a layer of fine sand exists between two coarse layers, the initial exsolution occured in the fine layer. However, this seemed to depend on where the fine layer was (i.e., the behavior depends on water pressure). These results will be used to validate numerical tools that are being developed in conjunction with researchers at Los Alamos National Laboratory.

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