Why pH buffering may lead to a more homogeneous rock dissolution

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An important effect of CO_2 and some preexisting organic compounds in deep saline aquifers, e.g., acetate, is chemical buffering, i.e. making system pH less sensitive to the release of cations from the dissolving rocks [1]. This buffering effect changes the apparent reaction order of water-rock interactions and affects the microstructural evolution [2]. Evaluating the residence time distribution and earliness of mixing of a reactive fluid [3] in porous media based on high resolution tomography allows us to predict the dissolution-driven structural changes of rocks from nanoscale. Here we show that the chemical buffering effects of aqueous species may lead to a more homogeneous pattern of rock dissolution because the first and the second derivatives of the apparent rate law regarding the reactant concentration in a buffered system favors both macro- and micro-mixing. In contrast, an unbuffered system with an identical initial geometry tends to form channelized structures in which solid removal is occuring primarily at the fluid-rock interface of the channels. Our results concorded with *in situ* synchrotron X-ray microtomographic measurements and laid a foundation for a computationally affordable simulation approach for emergent processes in geologic carbon storage systems.

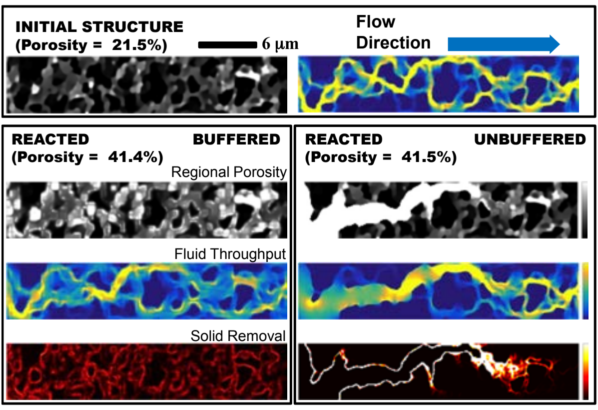

Figure 1: Comparisons of simulated microstructure development (gray), flow field (blue) and chemical reaction (red) between a buffered and an unbuffered system with an identical initial geometry. Colorbars are normalised and shared among the same type of images. A buffered system showed homogeneous removal of materials while the unbuffered system showed channelised morphology with surface controlled dissolution.

[1] Yang *et al*. (2011) *Energy Environ. Sci* **4**, 4596-4606. [2] Noiriel (2015) *Rev. Mineral. Geochem* **80**, 247-285. [3] Levenspiel (1999) *Chemical reaction engineering*, Wiley & Sons, Inc.