

Observations of chaotic mixing in porous rocks using X-ray microtomography

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Chemical reactions in porous rocks are typically modelled assuming well-mixed conditions, i.e., homogeneous reactant concentrations, within the pores of the rock. Recent findings using optical imagery in transparent bead packs challenge this assumption, showing that laminar flow through a bead assembly can lead to chaotic mixing of solutes, like those formed under turbulent flow. Yet, proving the existence of chaotic mixing in other porous media such as natural rocks remains unresolved due to limitations in directly observing pore-scale processes. In this study, we present direct evidence of chaotic fluid trajectories in porous rock samples using high-resolution X-ray tomography, which takes about 3 to 9 minutes and achieves a resolution of around 1 micron at the European Synchrotron Radiation Facility (ESRF). Using a custom-designed core holder and highly permeable sandstone and sand packs, we achieve high Peclet numbers (up to 10^4) during the co-injection of two miscible, high-viscosity fluids (glycerin and glycerin mixed with brine). These high Peclet numbers enable us to visualize the stretching and folding of the mixing interface between the two fluids within the pores of the samples before molecular diffusion blurs fluid front deformations. The existence of such chaotic fluid deformation leads to the persistence of microscale concentration gradients. The latter could potentially impact chemical reaction rates, causing deviations from predictions made by conventional models that assume homogeneous, well-mixed conditions. These findings highlight the critical role of high-resolution imaging in uncovering pore-scale processes and emphasize the need to revise kinematic models for more accurate predictions of transport and reaction processes in subsurface applications such as soil remediation, wastewater treatment, and geological CO₂ storage.