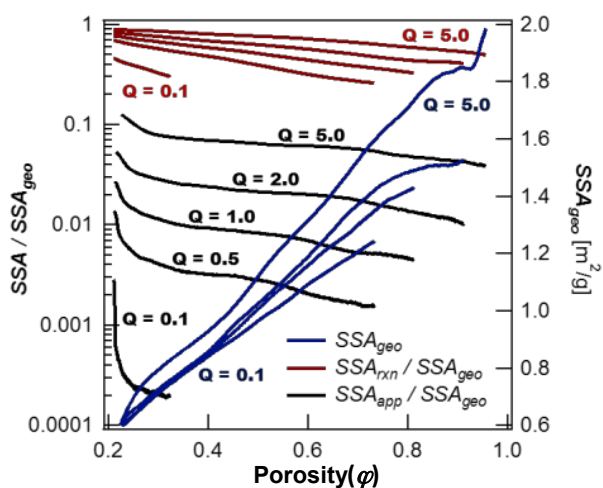


## Infiltration instability amplifies the discrepancy between geometric and reactive surface areas in rocks

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Infiltration instability [1] drives the development of many artificial and natural flow systems, such as are encountered in hydraulic fracturing, geologic carbon storage and watershed evolution. This instability amplifies regional heterogeneities in transport properties, making the microstructural evolution of a porous medium very sensitive to its initial geometry. We used high resolution (100 nm voxel) tomography to characterise the chalk microstructure. Then, using a reactor network model, we used the grey scale tomogram as the initial geometry to predict the evolution of reactive and geometric surface area during dissolution by percolation at a range of normalised macroscopic flowrates ( $Q$ ).



**Figure 1:** Comparison of geometric (*geo*), reactive (*rxn*) and apparent (*app*) specific surface area (*SSA*) and its dependence on the macroscopic flowrate ( $Q$ ) and increased pore volume as reaction proceeds. The subvolume was  $6 \times 6 \times 30 \mu\text{m}^3$  (1,080,000, voxels from a tomogram of a chalk).

The real reactive surface area (*rxn*) is the geometric surface area (*geo*) weighted by the regional fluid reactivity. The apparent surface area (*app*), although often referred to as reactive surface area, is actually a scaled mass conversion, based on an overestimate of the chemical affinity. The discrepancy between *rxn* and *geo* is typically within an order of magnitude, whereas the difference between *geo* and *app* varies more and is much more sensitive to the macroscopic flowrate. This insight sheds light on scaling and reconciling inconsistencies between field and laboratory measurements.

[1] Ortoleva *et al.* (1987) *Am. J. Sci* **287**, 1008-1040.