

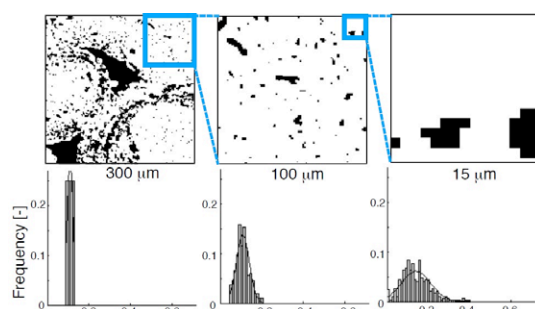
## Pore scale heterogeneity in the reactive surface area of rocks

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There are major knowledge gaps in the ability to characterize reactive transport in porous media at scales larger than individual pores. This precludes prediction of the field-scale impact of geochemical processes on fluid flow [1]. This is a source of uncertainty for CO<sub>2</sub> injection, which results in a reactive fluid-rock system, particularly in carbonate rock



**Figure 1:** Ratio of surface area to pore volume [ $\text{m}^2/\text{m}^3$ ] in an Indiana limestone at various length scales.

reservoirs. A potential cause is the inability of the continuum approach to incorporate the impact of heterogeneity in pore-scale reaction rates. This results in part from pore-scale heterogeneities in surface area of reactive minerals [2,3].

In this study we have created  $\mu\text{m}$  resolution 3D images of 3 sandstone and 4 carbonate rocks using x-ray microtomography. Using in-house image processing techniques we quantified the surface area from the images. This quantification was validated against N<sub>2</sub> BET surface area and He porosity measurements of the imaged samples. Distributions in reactive surface area were constructed by calculating surface areas in thousands of randomly selected subvolume images of the total sample (Fig. 1), each normalized to the pore volume in that image. Berea sandstone was far less heterogeneous and has a characteristic pore size at which a surface area distribution may be used to quantify heterogeneity. In carbonates, heterogeneity is more complex and surface area must be characterized at multiple length scales for an accurate description of reactive transport.

[1] Maher *et al* (2006) *Geochimica et Cosmochimica Acta*, **70**, 337-363. [2] Landrot *et al* (2012) *Chemical Geology* **318-319**, 113-125. [3] Li *et al* (2007) *American Journal of Science* **307**, 1146-1166.

## Termination and hydration of Forsterite (010) and Diopside (010) surfaces

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The hydrous alteration of ultramafic rocks has been paid much attention for its attribution to the abiogenic hydrocarbon generation from crystalline rocks. The molecular processes at the interface of water and those mineral surfaces, however, have not been described. We use in-situ high-resolution X-ray reflectivity to examine the forsterite (010) and diopside (010) surfaces in water. By modelling the electron density profile in the surface normal direction and fitting the measured data with least-square method, the atomic structures of hydrated single crystal surfaces could be depicted. We found, for the forsterite surface polished with alumina colloidal suspension under acidic environment (pH 3.5), a homogeneous termination with about half of the surface magnesium depleted and replaced with possibly a water species. In contrast, the forsterite surface polished with silica colloids under basic condition (pH 9-10) does not show such homogenous surface, but substantial morphological difference from the alumina-polished surface is observed by Atomic Force Microscopy measurement. The difference indicates the importance of solution chemistry to constrain the surface reactivity and dissolution mechanism. On the other hand, the diopside (010) surface is a naturally grown surface, which shows qualitatively identical features in its termination plane and the first adsorbed water electron density profile compared to those of the forsterite (010) surface. This similarity indicates common features in the hydration structure of these mineral surfaces, although the silicate compositions and structures are quite different.