Nanoscale transient porosity controls large-scale reactive fluid flow

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ABSTRACT

The reaction of fluids with rocks is fundamental for Earth's dynamics as they facilitate heat/mass transfer and induce volume changes, weaknesses and instabilities in rock masses that localizes deformation enabling tectonic responses to plate motion. Fluid-rock interactions also play a key role in geothermal energy, hydrocarbon production, CO_2 sequestration and nuclear waste disposal industries. In all of these examples it is the ability of a rock to transmit fluid, its permeability, that dictates geological processes and the industrial use of geological formations. For nuclear waste storage an impermeable wall rock is vital. For anthropogenic CO_2 sequestration, however, impermeable rocks are detrimental. In natural systems some environments (sediments) are open to fluids, but the majority (e.g., oceanic lithosphere) are nearly impermeable. Surprisingly though, even in rocks that are nominally impermeable widespread fluid-rock interactions are observed leading to the question: How can fluids migrate through vast amounts of nominally impermeable rocks? Here we investigate on of the most wide-spread alteration processes in the Earth's crust, the albitization of granitic rocks and compare these to simple X-ray tomography experiments undertaken in the system KBr-KCl. We show that fluid flow and element mobilization during albitization is controlled by an interaction between grain boundary diffusion and reaction front migration through an interface-coupled dissolution-reprecipitation process. Using a combination of focused ion beam scanning electron microscopy (FIB-SEM)-assisted nanotomography combined with transmission electron microscopy (TEM) reveals that the porosity is dictated by pore channels with a pore diameter ranging between 20 to 100 nm. Three-dimensional visualization reveals that the pore channels must have been connected during reaction. Aspect ratio analysis shows a nearly normal distribution indicating that a Rayleigh-Taylor-type instability caused the disconnection of the pore channels. As the transport of fluids in nanometer-sized objects with at least one characteristic dimension below 100 nm enables the occurrence of phenomena that are impossible at bigger length scales we discuss the potential influence of nanofluidic transport phenomena for metamorphic systems and take first numerical approaches using molecular dynamics simulations.