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Experimental studies on the evolution of mineralogy and microstructure during fluid-mediated mineral replacement of calcium carbonate by calcium phosphates

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Rock and mineral porosity is important to geological processes such as hydrothermal fluid migration and ore deposition, as well as industrial processes such as in situ leaching of ore minerals, oil and gas recovery, and geothermal energy extraction. While recent research has demonstrated that mineral porosity can be created by fluid-mediated mineral replacement reactions and can evolve with time, factors influencing such reaction-induced porosity have not been fully studied.

Carbonate minerals are often replaced by other minerals during fluid-mineral interactions and consequently porosity is generated in the product phases. Here, our experimental study shows that in phosphate rich hydrothermal solutions, calcium carbonate ($CaCO_3$) dissolves and is replaced by hydroxyapatite ($Ca_{10}(PO_4)_6(OH)_2$), whitlockite ($Ca_9Mg(PO_4)_6(PO_3OH)$) and other calcium phosphate minerals. The results suggest that the reaction pathway as well as the generation and evolution of porosity are dependent on reaction parameters such as temperature (60-220 °C), solution pH (pH 5 and 8), time (20-730 hours) and the initial composition and texture of calcium carbonate precursors, i.e., porous limestone and pore-free calcite single crystal Icelandic spar.

Porous limestone was replaced by equally porous assemblage of calcium phosphate minerals while pore-free single crystal Icelandic spar was replaced by more porous calcium phosphate product phases. The shape and uniformity of the pores within the product minerals differ when factoring in solution pH. Product phases formed at pH 5 contained elongated pores that follow the phase orientation towards the centre of the mineral grains, while phases produced at pH 8 showed nano-micrometres scale pores with a wider size distribution. At 60 °C, fractures were induced in the parent phase by the reaction, while at higher temperatures (e.g., 220 °C) faster reaction as well as porosity coarsening were observed. These preliminary data provide some basis for more detailed and quantitative studies into reaction-induced porosity in carbonate and phosphate minerals.