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The influence of Cl, F and Mg ions on the experimental replacement of Carrara marble by apatite

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Fluid-solid interactions span a very wide range of possible reactions, and are responsible for most of the mineral assemblages in the Earth's crust. Reequilibration processes in a wide range of materials are dominated by dissolution and reprecipitation mechanisms. The interaction of aqueous fluids with these materials results in a remarkable consistency of behaviour, especially in the way that the dissolution and precipitation processes are coupled, and how the relative solubility and molar volume combine to create porosity which allows fluid to continuously migrate to the reaction interface. The recent discovery that the coupling can be manipulated by changing the fluid composition not only emphasizes the role of the fluid in the coupling but opens new avenues for materials syntheses (Putnis, 2009).
Recently, a study by Jonas et al., (2013) used as a model the pseudomorphic replacement of Carrara marble (calcite – $CaCO_3$) by calcium phosphates (Ca5P(O4)3(OH, Cl,F) to show that the grain boundaries present in the rock and the transient porosity structures developing throughout the replacement reaction, enabled the reaction front to progress further into the rock, as well as to the center of each single grain until complete transformation.
In the present study, the same system was used to explore the rates and evolution of such replacement in the presence of different ions in the fluid used for reaction. Small Carrara marble cubes of around 2 mm3 were treated in hydrothermal reaction vessels at 200°C using phosphate bearing solutions with and without the presence of the different ions: chlorine, fluorine and magnesium. Mounted and polished cross sections of the reacted samples were examined by scanning electron microscopy and electron microprobe analysis to determine the extent of replacement, the textural evolution and the composition of the product phases. Results show that changes in the chemistry of the fluid seem to influence the evolution of the reaction front: whether the reaction front is more effective through the grain boundaries towards the centre of the grains, or through the developing porous structures towards the centre of the rock. Moreover, the micro-porosity of the replaced product also shows significant dependence on the solution composition, varying from being very heterogeneous as reported by Jonas et al., (2013), to a more uniform micro-porosity distribution as observed in this study.

References: Jonas, L., John, T., King, H. E., Geisler, T., & Putnis, A. (2014). The role of grain boundaries and transient porosity in rocks as fluid pathways for reaction front propagation. Earth and Planetary Science Letters, 386, 64–74. doi:10.1016/j.epsl.2013.10.050

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