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The role of grain boundaries and transient porosity increase as fluid pathways for reaction front propagation

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The pseudomorphic replacement of Carrara marble by calcium phosphates was studied as a model system to examine the influence of different fluid pathways for reaction front propagation induced by fluid-rock interaction. In this model system, the grain boundaries present in the rock and the transient porosity structures developing throughout the replacement reaction enable the reaction front to progress further into the rock as well as to the center of each single grain until complete transformation. Hydrothermal treatment of the marble using phosphate bearing solutions at temperature levels of 150˚C and 200˚C for different durations lead to the formation of two product phases which were identified as hydroxyapatite [$Ca_5(PO_4)_3OH$] as well as β-tricalcium phosphate [β-$Ca_3(PO_4)_2$] (β-TCP). The formation of β-TCP was probably favored by the presence of $\sim$0.6wt.% of Mg in the parent phase. Completely transformed single grains show a distinctive zoning, both in composition and texture. Whereas areas next to the grain boundary consist of nearly pure hydroxyapatite and show a coarse porosity, areas close to the center of the single grains show a high amount of β-TCP and a very fine porous microstructure. If F was added as an additional solution component, the formation of β-TCP was avoided and up to 3wt.% of F were incorporated into the product apatite. The use of the isotope ^{18}O as a chronometer for the replacement reaction makes it possible to reconstruct the chronological development of the calcium phosphate reaction front. Raman analysis revealed that the incorporation of ^{18}O in the PO_4 tetrahedron of hydroxyapatite results in the development of distinct profiles in the calcium phosphate reaction front perpendicular to the grain boundaries of the marble. Through the use of the ^{18}O chronometer, it is possible to estimate and compare the time effectiveness of the different fluid pathways in this model system. The results show that the grain boundaries serve as a very effective pathway that enable the fluid to penetrate the rock more than one order of magnitude faster compared to the newly developing channel-like porosity structures which act as pathways towards the center of single mineral grains. Thus, it may be possible for the fluid to progress relatively large distances along the grain boundaries after only short reaction durations without producing broad reaction fronts along the path.