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The influence of solution composition and grain boundaries on the replacement of calcite by dolomite

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Dolomite formation is a mineral replacement reaction that affects extensive rock volumes and comprises a large fraction of oil and gas reservoirs [1,2]. The most accepted hypothesis is the 'dolomitization' of limestone by Mg-rich fluids [3]. The objective of this research is to study the replacement mechanism of calcite by dolomite, the role of grain boundaries, highlighted by Etschmann et al. (2014), and the possible influence of solutions in dolomite formation under the presence of ions that are normally in crustal aqueous fluids.

To accomplish this purpose, we performed hydrothermal experiments using Carrara marble cubes of ∼1.5 mm size and 7-9 mg weight as starting material, reacted with 1M (Mg,Ca)Cl2 aqueous solutions, with Mg/Ca ratios of 3 and 5 at 200°C, for different reaction times. Additional experiments were performed adding 1mM of Na2SO4, NaCl or NaF to the previous solutions. After the reaction, the product phases were identified using Raman spectroscopy, X-Ray powder diffraction (XRD), electron microprobe analysis (EMPA), and the textural evolution was studied by scanning electron microscopy (SEM).

Samples reacted with aqueous solutions resulted in the replacements of the calcite rock into magnesite and dolomite. The amount and type of reaction strongly depends on the Mg/Ca ratio. Samples reacted with a Mg/Ca ratio of 5 resulted in an almost complete replacement reaction and more favorable for magnesite formation than for dolomite. When the Mg/Ca ratio was 3 dolomite formed but the replacement was located in the core of the sample. We show that grain boundaries are very important for the infiltration of solution and the progress of a replacement reaction, acting as fluid pathways. Solution composition controls the nature of the replacement product.

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