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Magnesium facilitates sulfate incorporation into carbonate

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Carbonate associated sulfate (CAS) refers to trace quantities of sulfate occurring in bulk carbonate rocks as well as in biogenic carbonates. Its sulfur isotope value reflects that of the ambient seawater, hence, CAS is used as an indicator for redox conditions and microbial activity in former oceans. Although its sulfur isotope value is widely applied as a seawater proxy, the factors controlling CAS concentration in carbonates are still not fully constrained.

In order to explore CAS incorporation in carbonates, hydrothermal alteration experiments with shells of the modern marine bivalve *Arctica islandica* were performed. The obtained results suggest preferential incorporation of CAS into Mg-rich calcite compared to pure calcite. Pieces of *Arctica islandica* shells were altered at 175°C in sulfate-depleted seawater. During alteration from one to twelve weeks, the primarily aragonitic shell successively recrystallized to calcite. After twelve weeks, a magnesium-bearing carbonate seam (10 µm thick) developed along the inner shell rim. NanoSIMS images of Mg and Ca reveal regions with different magnesium concentrations. Moreover, concentration differences in O and S exist at the same spatial resolution. The images show regions with different magnesium concentration and indicate a 1-to-1 exchange between magnesium and calcium. A contemporary increase of oxygen and sulfur not only indicates sulfate incorporation in those regions where magnesium is present but also exhibits a maximum sulfate incorporation into the carbonate with a Mg/Ca ratio of 1:5. This result explains why the CAS concentration of Mg-rich biogenic carbonates is usually higher than that of Mg-poor carbonates. Knowing the preferred CAS incorporation into carbonates with a Mg proportion of 20% helps to reconstruct the sulfate concentrations of former seawater from CAS in bulk rocks as well as from biogenic carbonates.