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The effect of microstructure in the dissolution and alteration kinetics of biogenic Carbonates

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Carbonate archives, being geochemical proxy carriers, form the basis of a considerable amount of environmental information. These archives are subject to variable degrees of post-depositional alteration, diagenesis, that takes place during mineral interaction with meteoric water, interstitial solutions, connate water or subsurface brines. Complex diagenetic-reaction pathways are induced and cause recrystallization and transformation to new polymorphs, morphologies and macrostructures. Hence, an improved quantitative understanding of precipitation, the alteration processes and its parameters is of key importance. Of special significance is the ability to assess the degree to which primary environmental and/or metabolic signals are affected, ranging from well preserved to entirely overprinted stages.

In order to assess the effect of biomineral microstructures on the extent, mode of diagenetic alteration of pristine skeletons we subjected modern hard tissues of the coral Porites sp., the gastropod Haliotis ovina and the bivalves Arctica islandica and Aequipecten operculatis to laboratory-based hydrothermal alteration. Experiments were conducted at 175°C, lasted for up to 28 days and were carried out in the presence of three different alteration solutions simulating meteoric, burial and marine diagenetic realms. We examined for each microstructure: (a) the kinetics of biogenic to non-biogenic carbonate transition, (b) new carbonate mineral formation on the micro- and the nanoscale, (c) micro- and nanostructural changes at different stages of alteration and (d) differences in grain areas in pristine and altered shells. With our statistical evaluation of grain area we are able to determine the degree of diagenetic overprint, especially low degrees of hydrothermal overprint. Carbonate phase analysis, micro- and nanostructure characterization and biomineral grain size evolution was determined with XRD and Rietveld analysis, AFM imaging, electron backscatter diffraction (EBSD) analysis and statistical grain size eveluation.

We demostrate the significant influence of the biogenic microstructure at alteration. In the shell of modern Arctica islandica aragonite prisms are surrounded by a thin network of organic fibrils. These get easily destroyed at alteration, render space for fluid percolation and facilitate the prevaisive transformation from biogenic aragonite to abiotic calcite. Calcite formation starts after a dormant period of about 4 days. Once started, transformation progresses quickly such that with 28 days of alteration almost all aragonite is transformed to calcite. Acicular, Porites sp. aragonite displays a different behaviour at alteration. When altered for 28 days only a minor part of the coral skeleton is transformed to calcite. The alteration fluid enters the coral skeleton at the centers of calcification, new calcite formation starts mainly at these sites. The highly porous prismatic shell portion of modern Haliotis ovina is easily transformed to calcite. Aragonite prisms encased by a network of biopolymer fibrils get destroyed when altered and space becomes available for fluid infiltration that causes an extensive overprint. The nacreous shell portion of Haliotis ovina is little affected.

Our study shows, that laboratory-based alteration of bioaragonite to calcite occurs via interface-coupled dissolution-reprecipitation. Alteration strength is temperature but also fluid dependent. The fluid simulating meteoric water has little effect, fluids simmulating burial and marine diagenesis have a major influence.