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In-situ identification of the biomacromolecules relevant to diagenetic isotope exchange in biocalcites

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The oxygen isotope compositions of fossil biocalcites such as foraminifera, bivalves, brachiopods and belemnites are a comprehensive proxy for ocean temperatures stretching back into the Phanerozoic. Biocalcites differ from abiotic calcites by their hierarchical structures and abundant organo-mineral interfaces, which provide huge surface areas for diagenetic fluids. As a result of these differences, biocalcites are particularly susceptible to fluid-mediated diagenesis that can potentially bias paleoclimate reconstructions [1], without any evidence of textural changes. In our previous work [2] we experimentally simulated the effects of fluid-mediated isotopic exchange on biocalcites by incubating multiple species of modern foraminifera in ^{18}O-labeled artificial seawaters. Evidence of the resulting preferential fluid penetration and ^{18}O-exchange was obtained using NanoSIMS imaging. Consistent differences in bulk ^{18}O-exchange between the studied foraminifera revealed a species-specific susceptibility to diagenesis, which is related to specific organo-mineral ultrastructures, the proportions of which vary with each species of foraminifera.

We recently extended this work to include two species of modern bivalves, which similarly showed substantial differences in bulk ^{18}O-exchange due to differences in their ultrastructures. This has motivated an investigation into the composition of the biomacromolecules that compose these diverse biocalcite ultrastructures. We used photo-induced force microscopy [3] (PiFM), a new cutting-edge technique that can simultaneously acquire 3D topographic data at high spatial resolution (~ 5 nm) — ideally suited to locate ultrastructures of interest — and high spectral resolution (~1 cm^{-1}) molecular chemical information, to identify the organic compounds most relevant to diagenesis in each of the investigated species. These *in-situ* PiFM observations permit to chemically fingerprint the organo-mineral ultrastructures relevant to diagenesis and hence further advance our understanding of the role biomacromolecules play during diagenesis of biocalcites.

[1] Bernard, S. *et al.* (2017) Burial-induced oxygen-isotope re-equilibration of fossil foraminifera explains ocean paleotemperature paradoxes. *Nature Communications* 8, 1-10.

[2] Cisneros-Lazaro, D. *et al.* (2022) Fast and pervasive diagenetic isotope exchange in foraminifera tests is species-dependent. *Nature Communications* 13, 1-11.

[3] Otter, L. M. *et al.* (2021) Chemical Imaging by Photo-Induced Force Microscopy : Technical Aspects and Application to the Geosciences. *Geostandards* 45, 5-27.