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Biomimetic Control of Carbonate Biomineralisation via Protein Templating

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Biomineralisation in marine calcifiers is regulated within specialised microenvironments where organic matrices, ion-transporters, enzymes cooperatively control pH, nucleation pathways and carbonate polymorph selection [1]. Unravelling the respective roles of organic templating and catalytic carbon chemistry remains challenging in vitro conditions, limiting mechanistic understanding of mineral–organic interactions that underpin biogenic carbonate formation and long-term carbon storage. Here, we present a recombinant, biomimetic platform that reconstructs key elements of coral calcification to probe protein–enzyme regulation of carbonate biomineralisation under environmentally relevant aqueous conditions. We combine coral acid-rich proteins (CARPs) with a halotolerant, thermostable carbonic anhydrase (CA) to dissect how organic templating and enzymatic CO_2 hydration jointly influence nucleation, polymorph selection and mineral composition across supersaturated solutions, artificial seawater. CARP1 and CARP3 exhibit distinct phase-directing behaviour, preferentially stabilising vaterite and calcite, respectively, while CARP3 promotes Mg incorporation and Mg–calcite formation under seawater-like ionic conditions. CA accelerates inorganic carbon supply and promotes rapid formation of amorphous precursors in the absence of structural templating. When combined, CARP3–CA displays cooperative behaviour, yielding densely aggregated crystalline carbonates and enhanced Mg incorporation, revealing complementary roles of catalytic carbon hydration and organic templating at the mineral–solution interface. Morphological and mineralogical analyses (SEM, FTIR) demonstrate that carbonate phase outcomes are governed by the balance between inorganic carbon availability and template-mediated interfacial control. This modular, protein-based system provides a flexible platform to interrogate mineral–organic interactions driving carbonate formation and preservation in complex aqueous environments. Beyond fundamental implications for biomineralisation and early diagenetic processes, these findings inform bio-inspired strategies for low-energy carbonate formation relevant to carbon cycling and mineral-based CO_2 sequestration.

[1] Sanyal, S. K., Scott, C., Nagaraj, V., Speight, R., & Ahmed, F. H. (2025). Harnessing the biomolecular mechanisms of marine biomineralisation for carbon sequestration. *Biotechnology Advances*, 108644.