

Experimental approaches to marine and meteoric dissolution-to-precipitation cycles of fine-grained marine carbonate sediments

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Fine-grained carbonate (carbonate ooze), or microcrystalline carbonate (micrite), its lithified counterpart, forms a main constituent of limestones throughout much of Earth's history. Fine-grained carbonates are deposited below the permanent fair-weather wave base in neritic lagoonal environments or below the storm-wave base in basinal settings. The origin of components forming these fine-grained carbonates often remains poorly understood and represents a major challenge in carbonate sedimentology, particularly when these materials are used as carbonate archives (bulk micrite geochemistry). Here we present a novel experimental approach exposing natural, fine-grained carbonate sediments to dissolution-precipitation cycles under non-sterile conditions that mimic earth-surface conditions. In a first stage, the experiment simulated subaerial exposure of an ooid (aragonite) shoal and leaching and carbonate dissolution under meteoric phreatic conditions. In a second stage, CO₂ was added to the experimental fluid (natural rainwater) representing soil-zone activity. In a third stage, partly dissolved (microkarstified) sediments were exposed to marine phreatic conditions simulating renewed flooding of the shoal carbonates. During the third stage, precipitation was induced by degassing the CO₂ in the fluid with N₂. Degassing induced nucleation and growth of a diagenetic inorganic aragonite (and subordinate calcite) phase upon the surface of carbonate particles. The outcome of these first experiments is promising. The CO₂ concentration of the fluid and the air are low under atmospheric conditions and increase as expected due to adding CO₂ to the experiment resulting in a lower pH. Carbonate dissolution increases conductivity, alkalinity, and calcium concentration reaching a plateau at the end of the first experimental phase. Small surficial damages to ooids represent zones of weakness and form the preferred sites of dissolution leading to a deepening and widening of these features and a general smoothing of ooidal surfaces. Following N₂ addition, CO₂ concentrations of water and air, conductivity, alkalinity, and calcium concentrations decrease, whilst the pH increased. Scanning electron microscope images reveal the re-precipitation of dissolved calcium carbonate in the form of newly formed diagenetic needle-like aragonite crystal on the surface of carbonate particles. Oxygen isotope analyses of altered sediments document a ca 0.3 permil shift to lower bulk carbonate ratios whereas carbon isotope data are lower by up to 1 permil in the leached and cemented samples.