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Triple Oxygen Isotope Fractionation of Calcite

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Researchers have debated whether calcite and water precipitate in equilibrium ever since the technique was developed 70 years ago. Unlike conventional δ^{18}O analysis where the formation water's isotopic value is assumed, paired δ^{17}O-δ^{18}O measurements allow for the water's isotopic composition to be calculated because there is only one unique solution for equilibrium fractionation using Δ^{17}O-δ^{18}O values (where Δ^{17}O= δ^{17}O-0.528δ^{18}O). To a first approximation, the calcite-water equilibrium fractionation factor, θ (where θ=ln$^{17}\alpha$/ln$^{18}\alpha$), varies with temperature by 0.00001/°C. This study proposes an equilibrium Δ^{17}O-δ^{18}O fractionation line using modern brachiopod shells formed at 0 and 30 °C in ocean water with a δ^{18}O value of 0 and calcite precipitated in New Mexico tap water at 20 and 40 °C catalyzed with the addition of bovine carbonic anydrase precipitated in New Mexico tap water.

The triple oxygen isotope calcite-water equilibrium fractionation line was applied to well preserved brachiopod shells ranging from the Mid Maastrichtian to Ordovician from a variety of locales and Early Triassic ammonite shells from the Western United States. Based on paired δ^{17}O-δ^{18}O measurements, all but three samples formed in equilibrium with an ice-free ocean with an oxygen isotopic value of -1 ‰ or the modern ocean value of 0. The Ordovician brachiopod shell and the Early Triassic ammonite shells suggest diagenetic alteration affected the oxygen isotopic composition. However, we can 'see through' this diagenesis by using a simple fluid-rock interaction model because the δ^{18}O values change appreciably before the Δ^{17}O values change when plotted on a Δ^{17}O-δ^{18}O graph. Seeing through the diagensis suggests that samples from the Smithian thermal maximum formed in water ~5 °C warmer than samples from after the thermal maximum, where the Smithian sample formed in ~20 °C water and the Spathian samples formed in ~15 °C water. The Ordovician brachiopod sample formed in ~18 °C water. By using triple oxygen isotope analyses, we are able to suggest both past ocean temperatures and oxygen isotopic composition. Paired δ^{17}O-δ^{18}O measurements of ancient carbonates provides a better understanding of past ocean conditions during climate change events.