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NEXAFS and Raman spectroscopic investigations of proto-dolomite mineralization directly on carboxylic functional groups derived from naturally occurring exopolymeric substances (EPS) in hypersaline microbial mats

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Despite many studies over the last century, low-temperature (low-T) dolomite remains an enigmatic mineral. Although supersaturated in modern aquatic environments, dolomite is inexplicably scarce even though it is abundant in the rock record, a phenomenon known today as “the dolomite problem”. Although our understanding of “the dolomite problem” has progressed significantly over the past two decades, due to the development of nucleation surface (1,2) and saturation undersaturation cycling models (3), however these mechanisms are still not fully understood nor are they delineated well in natural environments. In terms of the nucleation surface model, the lack of direct observations confirming the preferential dolomite nucleation on carboxylic functional groups gives rise to many unanswered questions. These questions include the exact mechanisms of dolomite formation in the presence of exopolymeric substances (EPS), which functional groups are contributing to dolomite formation, and what are the impact of these formation pathways on the mineralogical properties of primary dolomite. In order to evaluate these questions, controlled laboratory experiments were performed using naturally occurring EPS as a substrate to form proto-dolomite, which was subsequently examined using high-resolution spectroscopic techniques including Raman and Absorption Fine Structure (NEXAFS) analysis. The data from our work showed that carbonates preferentially nucleating on carboxylic functional groups within EPS from naturally occurring mats as opposed to other functional groups commonly found, such as phosphoryl and hydroxyl groups. This data in conjunction with detailed geochemical measurements indicated that not only are the properties of nucleation substrates key factors in low-T dolomite but also evolution of geochemical conditions play a significant role in the dolomite mineralization process.

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