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Calcium isotope fractionation during (a)biogenic calcium carbonate precipitation

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Fractionation of calcium isotopes during calcium carbonate precipitation is a long-standing enigma. Different experiments have provided contrasting and even contradictory results. All prior experiments show a different fractionation between calcite and aragonite, with aragonite being isotopically lighter than calcite, with additional control by temperature and/or precipitation rate. Yet, the relationship between precipitation rate and fractionation is significantly different from one experimental setup to the next. Carbonate precipitation by diffusion of CO_2 diffusion [1, 2] and constant addition [3] show that fractionation decreases when precipitation rates increase, while experiments via CO_2 supersaturation using $(NH_4)_2CO_3$ salts [4-6] show the opposite. The hypothesis to explain such variable results are a combination of a surface entrapment model and the existence of Ca^{2+}-NH_3 complexes, combined with a role played by the efficiency of stirring in the solution. With efficient stirring and homogenization of the solution comes a lower role played by aqueous diffusion.

Here, we precipitated synthetic calcium carbonates using a pH-controlled constant addition method. We find that the fractionation depends on the precipitation rate, with no influence of sulfate and magnesium concentrations of the solutions. Our results show a strong similarity to those from Tang et al. [2] for calcite and Gussone et al. [1] for aragonite, despite the different setups used in the various experiments. Our experiments do not rely on $(NH_4)_2CO_3$ salts. As a result, strong interaction with NH3 is avoided, unlike several other studies [4-6], supporting the surface entrapment model to explain calcium isotope fractionation.

We use these new results combined with new calcium isotopic compositions measured from biogenic carbonates to explore biomineralization processes. We confirm that mineralogy is not the only controlling factor as mollusks that precipitate aragonite and/or calcite are characterized by similar calcium isotopic composition. We also find that biomineralization processes induce a relationship between lithium and calcium isotope fractionations and explore the factors controlling the isotopic composition of the shells.

[1] Gussone et al., 2003, GCA
[2] Tang et al., 2008, GCA
[3] Marriott et al., 2004, EPSL
[4] Lemarchand et al., 2004, GCA
[5] Alkhatib et al., 2017, GCA
[6] Alkhatib et al., 2017, GCA