Formation pathways of norsethite controlled by Mg/Ba ratio and implication for dolomite problem

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Dolomite problem is a long-standing puzzle in geology. Due to the common failure in precipitating dolomite in laboratory at low temperatures, our understanding towards the formation pathway of ordered dolomite still remains poor. An investigation on the formation pathways of dolomite-analogues could be a feasible strategy to enhance our comprehension of dolomite formation. Norsethite [$BaMg(CO_3)_2$] is one of the representative dolomite-analogues, which was not only found in wide range of natural settings, but also could be readily precipitated in laboratory. Previous studies indicated that various precursors or seed crystals (e.g., amorphous phases, witherite, magnesite, etc.) were involved in the formation of norsethite, whereas its direct precipitation has not been achieved yet. Herein, to better understand the formation pathways of norsethite, we conducted the mineralization experiments of norsethite by a CO_2 gas diffusion technique, which is an appropriate mineralization method to mimic natural sedimentary environment. The experimental results show that two different pathways, including direct precipitation and dissolution-reprecipitation, are responsible for norsethite formation in mineralizing solutions with different concentrations of Mg^{2+} and Ba^{2+}, respectively. A series of comparative experiments further reveal that the initial Mg/Ba ratio in solution is the primary factor determining the formation pathway of norsethite. At low Mg/Ba ratios (<15), a multistep pathway occurs, i.e., a precursor witherite first forms, followed by norsethite precipitation and transformation from witherite to norsethite. Especially, at Mg/Ba ratio higher than 20, stoichiometric and fully ordered norsethite can be directly precipitated as penetration twins with well-developed {104} faces. To the best of our knowledge, this is the first report of direct precipitation of dolomite analogues under ambient conditions. The findings provide new insights into the formation pathways of dolomite and its analogues.

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