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Mineral carbonation of carbon dioxide using exchangeable cations of vermiculite

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Increasing carbon dioxide concentration due to anthropogenic emission is emerging as a serious problem leading to global warming. One way to reduce CO_2 is to solidify it into more stable carbonate minerals by reacting CO_2 gas with silicate minerals containing alkaline earth metals (e.g., feldspar, olivine, pyroxene, serpentine, etc.). However, such a mineral carbonation method is still a challenge in practical application owing to: 1) slow reaction kinetics, 2) high temperature reaction conditions, and 3) continuous mineral consumption. As an alternative metal source for advancing the efficiency of mineral carbonation, exchangeable cations of clay minerals can be selected. The exchangeable cations are easily extracted even in ambient conditions without spending extra-energy and time for the dissolving or destroying of mineral structures. We attempted here to mineralize CO_2 by supplying Ca^{2+} from vermiculite using a cationic exchange method. The 100 ml of 1M NaCl solution was injected into a columnar vessel containing 20 g vermiculite powders using a peristaltic pump at a flow rate of 10 ml/min. The reacted solution was continuously dropped into 100 ml 1M NaOH solution, and, simultaneously, CO₂ gas was purged at a flow rate of 2 l/min. Reaction temperature was regulated at 293 K and 333 K. White precipitates were filtrated immediately without further aging processes for crystal growth, then characterized using an X-ray diffractometer and a scanning electron microscopy. As a result, rhombohedral calcite grains with multiple step growth facets and polycrystalline vaterite grains with a barrel-like shape were homogeneously precipitated at 293 K with the grain size of 5 - 10 μ m. In contrast, spindle-shaped vaterite grains (10 - 30 μ m in length and 2 - 5 μ m in width) are predominantly produced at 333 K (\sim 98 wt.%). The calcite grains showed more perfect rhombohedral morphology even in low abundance. These morphological characters of vaterite and calcite are seemingly subjected to the difference in crystal growth mechanisms (step growth for calcite and coalescence for vaterite). For validating a possibility of vermiculite recycling, we recharged the used vermiculite powders with 0.5 M CaCl₂ solution, subsequently conducting the mineral carbonation sequence. As the number of recycling times increased, the vermiculite powders became gradually finer because of particle dissociation by hydration and osmotic swelling due to Na⁺. However, the recycled vermiculite was still useful even after five-time repetitions of the mineral carbonation sequence. Average 0.85 g calcium carbonates were collected per each sequence.