Pressure-induced phase transitions of vaterite, a metastable phase of $CaCO_3$

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Calcium carbonate plays an important role in global carbon cycle. Calcium carbonate has three polymorphs, calcite, aragonite and vaterite. Vaterite is a meta stable phase and no systematic experiments on the behavior under high pressure have been condcuted so far. In this study, pressure-induced phase transitions of vaterite, a metastable phase of $CaCO_3$, were observed from Raman spectra and X-ray diffraction patterns at high pressure using diamond anvil cells at room temperature. Pressure dependence of Raman peaks assignable to the symmetric stretching vibration modes of carbonate ion indicated the phase transitions of vaterite. At 4.3 GPa, vaterite (vaterite I) transformed to a high-pressure form of vaterite (vaterite II). With increasing pressure, a part of vaterite II gradually transformed to calcite III, and the remainder of vaterite II transformed to another new phase (vaterite III). The phase transition from vaterite II to calcite III was irreversible, and calcite III back-transformed to calcite I during decompression. Moreover, at the pressures higher than 13.1 GPa, several diffraction spots were observed on an imaging plate, indicating another phase transition from vaterite III to a phase having coarse grains (vaterite IV). Our results indicate that vaterite undergoes more complex phase transitions at high pressure than other polymorphs of calcium carbonate, calcite and aragonite. These phase transitions of vaterite will open a window to understanding phase transitions of metastable minerals at high pressure.

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