

Structure changes of hydromagnesite $\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$ with temperature and pressure

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The global warming has become one of the most important environment problems in recent years. Geological storage of CO_2 has great potential to eliminate atmospheric CO_2 emitted by fossil fuel combustion (XUE and Nakao 2008). Regarding mineral CO_2 sequestration, CO_2 is chemically stored through mineral carbonation in solid carbonates. Magnesium and/or calcium carbonate minerals such as magnesite MgCO_3 , dolomite $\text{CaMg}(\text{CO}_3)_2$, calcite CaCO_3 , and aragonite CaCO_3 have long been recognized as the best CO_2 storage. Because hydrous magnesium carbonates can be directly precipitated from aqueous solution in a range between 0 and 200 °C (Cheng et al. 2009; Yamamoto et al. 2017), we consider that hydrous magnesium carbonates would be the best candidate for the CO_2 storage process. The major hydrous magnesium carbonates formed from aqueous solution are nesquehonite $\text{MgCO}_3 \cdot 3\text{H}_2\text{O}$ and hydromagnesite $\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$. The physical properties of these minerals are critically important factors to assess environmental safety and phase stability over geological time scale, but they have not been fully investigated yet, especially of hydromagnesite. In this study, we performed investigated the structural behaviors of hydromagnesite under high temperature and high pressure using HT-XRD, TG-DTA, HP-XRD to reveal the physical properties and structural stability of hydromagnesite. The results of *in-situ* high-temperature XRD showed that, no significant change was observed up to 170 °C. With increasing temperature, however, the intensities started to decrease at 200 °C, and all peaks disappeared at 290 °C. Above the decomposition temperature a few peaks corresponding to periclase appeared. The results of TG-DTA clearly showed two weight loss steps in the temperature range from 200 to 340 °C and from 340 to 500 °C, which correspond to the dehydration and decarbonation of hydromagnesite, respectively. Therefore, hydromagnesite decomposes into periclase, carbon dioxide, and water as following reaction: $\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O} \rightarrow 5\text{MgO} + 4\text{CO}_2 + 5\text{H}_2\text{O}$. No magnesite is formed from hydromagnesite with temperature. The results of *in-situ* high-pressure XRD showed that the intensities gradually decreased with compression, and then most of peaks disappeared above 6.4 GPa. After decompressing to 4.2 GPa, however, the hydromagnsite reappeared. It suggests that no dehydration occurs below at least 6.4 GPa. The amorphousization of hydromagnesite with compression would be caused by disordered arrangements of MgO_6 octahedra and CO_3 triangles in the structure.

Keywords: CO_2 geological storage, hydromagnesite $\text{Mg}_5(\text{CO}_3)_4(\text{OH})_2 \cdot 4\text{H}_2\text{O}$, structure change, high-temperature XRD, high-pressure XRD