

Crystal structure of the high-pressure phase of calcium hydroxide, portlandite: In situ powder and single-crystal X-ray diffraction study

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ABSTRACT

The crystal structure of a high-pressure phase of calcium hydroxide, Ca(OH)₂ (portlandite), was clarified for the first time using the combination of in situ single-crystal and powder X-ray diffraction measurements at high pressure and room temperature. A diamond-anvil cell with a wide opening angle and cell-assembly was improved for single-crystal X-ray diffraction experiments, which allowed us to successfully observe Bragg reflections in a wide range of reciprocal space. The transition occurred at 6 GPa and the crystal structure of the high-pressure phase was determined to be monoclinic at 8.9 GPa and room temperature [*I*121; *a* = 5.8882(10), *b* = 6.8408(9), *c* = 8.9334(15) Å, β = 104.798(15)°]. The transition involved a decrease in molar volume by approximately 5.8%. A comparison of the structures of the low- and high-pressure phases indicates that the transition occurs by a shift of CaO₆ octahedral layers in the **a-b** plane along the **a**-axis, accompanied by up-and-down displacements of Ca atoms from the **a-b** plane. The crystal structure of this high-pressure phase is considered to be an intermediate state between the low-pressure phase and the high-pressure–high-temperature phase. The complicated diffraction patterns of the high-pressure phase suggest that the phase transition occurred toward three directions around the **c**-axis of the low-pressure phase. This explains the difficulties encountered in previous structural analyses. The present results will provide key information for discussing the behavior of hydrogen bonds in these hydrous minerals under pressure.

Keywords: Portlandite, phase transition, crystal structure, high pressure, X-ray diffraction, hydrogen bond