

## ARTICLES

# **In-situ synchrotron study of the kinetics, thermodynamics, and reaction mechanisms of the hydrothermal crystallization of gyrolite, $\text{Ca}_{16}\text{Si}_{24}\text{O}_{60}(\text{OH})_8 \cdot 14\text{H}_2\text{O}$**

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## ABSTRACT

The hydrothermal crystallization of gyrolite was studied dynamically at 190–240 °C using synchrotron-based in situ Energy Dispersive Powder Diffraction (EDPD). The reaction mechanism involves the initial crystallization of a calcium silicate hydrate (C-S-H) gel, which has a sheet structure with well ordered Ca(O,OH) layers and disordered silicate layers. This is followed by the intermediate formation of Z-phase which finally transforms to gyrolite. This process involves ordering of the silicate layers and an increase in the order along *c*.

Kinetics data for all stages of the crystallization process were determined by analyzing the growth and decline of various diffraction peaks with time. The activation energy ( $E_a$ ) (nucleation) for Z-phase is ~39 kJ/mol while that for gyrolite is ~56 kJ/mol.  $E_a$  (crystallization) of gyrolite is higher at ~80 kJ/mol. The reaction occurs via a two-dimensional, diffusion-controlled mechanism and is a continuous process that suggests that Z-phase is an unstable, transient phase.