



In situ time-resolved X-ray diffraction of tobermorite formation in autoclaved aerated concrete: Influence of silica source reactivity and Al addition

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ABSTRACT

The hydrothermal formation of tobermorite during the processing of autoclaved aerated concrete was investigated by *in situ* X-ray diffraction (XRD) analysis. High-energy X-rays from a synchrotron radiation source in combination with a newly developed autoclave cell and a photon-counting pixel array detector were used.

To investigate the effects of the silica source, reactive quartz from chert and less-reactive quartz from quartz sand were used as starting materials. The effect of Al addition on tobermorite formation was also studied. In all cases, C-S-H, hydroxyllestadite and katoite were clearly observed as intermediates.

Acceleration of tobermorite formation by Al addition was clearly observed. However, Al addition did not affect the dissolution rate of quartz. Two pathways, via C-S-H and katoite, were also observed in the Al-containing system. These results suggest that the structure of initially formed C-S-H is important for the subsequent tobermorite formation reactions.

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1. Introduction

Recently, the demand for improved heat insulation of buildings has been increasing; accordingly, autoclaved aerated concrete (AAC) has attracted attention as a construction material because of its excellent heat insulation properties [1,2]. A typical process for the production of AAC includes hydrothermal treatment of a mixture of quartz sand, lime, cement, gypsum and other additives at high temperature (typically, 180–200 °C) under saturated steam pressure, which results in the formation of crystalline calcium silicate hydrates, namely, tobermorite ($\text{Ca}_5\text{Si}_6\text{O}_{16}(\text{OH})_2 \cdot 4\text{H}_2\text{O}$) [1,3–7]. Both the quantity and crystallinity of tobermorite formed in AAC have been reported to affect the mechanical properties of AAC significantly [4–7]. Therefore, understanding the mechanism of tobermorite formation during hydrothermal treatment (i.e., the autoclave process) is important in AAC production.

The phase evolution of calcium silicate hydrates under hydrothermal conditions, especially the formation of tobermorite, has been extensively studied [8–42]. Several kinetic approaches have also been attempted to clarify the mechanism of tobermorite formation [14,15,35–42]. It is well known that the reactivity of the silica source [8–15], the addition of Al compounds [11,16–29,31,42], the addition

of alkali [11,16,28–30] and the addition of sulfate compounds [19,31–34] strongly affect tobermorite formation. As starting material, a highly reactive silica source such as amorphous silica enhances the polymerization of C-S-H, which precedes tobermorite formation, consequently inhibiting the rearrangement of C-S-H to tobermorite [8,9,15,20]. On the other hand, the presence of Al slows the initial formation of C-S-H by reducing the solubility of quartz, which then accelerates tobermorite formation from C-S-H [16,18,20,22,42]. In addition, this inhibits the conversion to xonotlite [9,29], extending the temperature range over which tobermorite exists. These processes are more complex in industrial AAC production, which includes cement as starting material, and the formation mechanism of tobermorite is not thoroughly understood for such processes.

For investigating the formation process, X-ray diffraction (XRD) is a useful technique, but it is difficult to quantify non-crystalline phases, for example, calcium silicate hydrate (C-S-H), which is an important intermediate in the tobermorite formation process. XRD has been used in many *ex situ* studies, in which specimens were taken out from the autoclave at various stages of the reaction. However, such *ex situ* approaches may include ambiguity caused by undesired reactions during the cooling process.

Therefore, an *in situ* approach is more desirable for understanding the reaction in detail. Several *in situ* approaches have been reported for monitoring hydrothermal reactions, cement hydration and pozzolanic reactions by means of transmission XRD [42–48], transmission energy-dispersive XRD [42,49–52], and neutron

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