# Understanding the Growth Kinetics of Cementitious Magnesium Silicate Hydrates Using Atomic Force Microscopy

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Alternative cementitious materials, such as magnesium silicate hydrate (M-S-H), address the urgent need for sustainable cement production by reducing embodied CO_2 emissions and increasing durability. Rates and mechanisms of precipitation of the binding phases fundamentally control microstructural and property development in structural materials. Therefore, the kinetics of M-S-H nucleation and growth have become an active area of research, however, growth rates, underlying mechanisms, and transformation pathways remain poorly understood. In this study, M-S-H was grown on freshly cleaved magnesium oxide (MgO) substrates using magnesium nitrate hexahydrate and sodium metasilicate pentahydrate stock solutions to generate growth solutions containing [Mg] = [Si] = 0.4 to 30 mM. An atomic force microscopy (AFM)-based approach was developed and used to quantify growth rates of M-S-H at ambient temperature, including (1) initial (within 15 s) growth rates for varying saturation index (SI) with respect to $M_{0.75}SH$ (–0.76 to 8.05) and (2) time-dependent (over 15 s to 3 weeks) growth rates for a fixed $M_{0.75}SH$ SI of 6.43 ([Mg] = [Si] = 10 mM). From the SI quantified using geochemical modeling and the AFM-derived growth rates, the following rate equation was derived: *ln(rate) = 0.31ln(SI) – 19.87* and interpreted according to geochemical and microstructural models of M-S-H formation. Experimental data yielded a power-law relationship between rate and SI, demonstrating thermodynamic control over growth rates and revealing a plateau indicative of surface-limited growth (**Figure 1**). Furthermore, high-resolution AFM data illustrated surface morphology evolution with increasing Mg/Si ratios, providing insight into heterogeneous nucleation and growth of M-S-H on MgO (**Figure 2**). By inferring specific M-S-H phases from geochemical modeling, these findings provide a foundation for manipulating and refining mechanical, thermal, and chemical properties of M-S-H-based cement. This study highlights the dependence of M-S-H growth kinetics on the evolving composition of the growth solution. Such understanding advances the fundamental knowledge of M-S-H as a durable, sustainable cementitious material while also supporting predictive modeling to improve its application.

Initial Growth Rate of M-S-H on MgO

$ln(Y) = 0.31 ln(X) - 19.87$
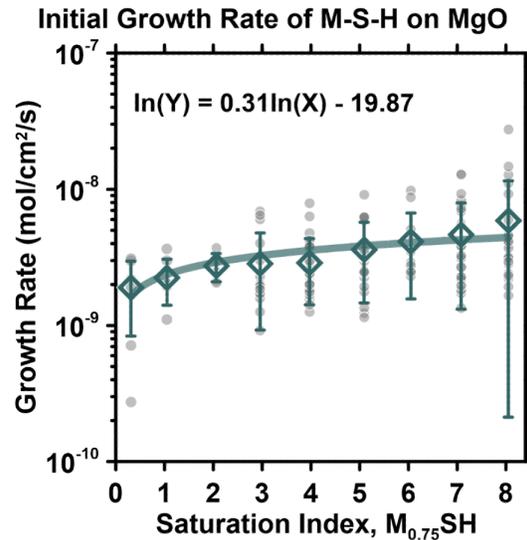
Figure 1. The relationship between growth rate and saturation index (SI). The logarithmic trend indicates a strong dependence on SI at lower Mg/Si concentrations followed by surface-limited growth at higher Mg/Si ratios. Error bars represent the standard deviation ($\pm\sigma$) of the calculated growth rates, applicable only for SIs ranging from –0.76 to 8.05 or [Mg] = [Si] = 0.4 to 30 mM.
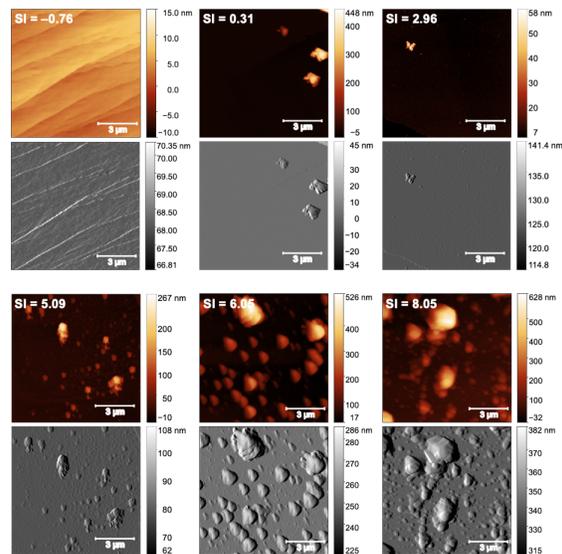
Figure 2. *Ex situ* atomic force microscopy (AFM) height and amplitude images illustrating the evolution of surface morphology of M-S-H growth on MgO across concentrations of [Mg] = [Si] = 0.4 to 30 mM, corresponding to saturation indices (SI) relative to $M_{0.75}SH$ ranging from –0.76 to 8.05. For each SI, the top row displays height images, while the corresponding amplitude images are shown directly below. At [Mg] = [Si] = 0.4 mM, the solution is undersaturated (SI = –0.76), preventing precipitation, which corresponds to the observed absence of nanoparticles. As concentrations increase, nanoparticle formation can be observed, with localized regions of high nanoparticle density and increasing particle size.