Simulating the Formation of ASR Gels

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PROBLEM

Alkali-silica-reaction (ASR) is a significant cause of premature concrete deterioration. Although it has received much research attention, an essential question still remains—how does a soft gel, as is formed by this reaction, induce the critical level of stress to exceed the strength of, and crack the concrete matrix?

One possibility is that the ASR gel increases in viscosity as it imbibes calcium with age and a concomitant expansion causes the concrete failure**.** Another is that the gel's rate of flow into the porous microstructure is slower than the rate at which it forms, which builds up stresses that cannot be relieved.

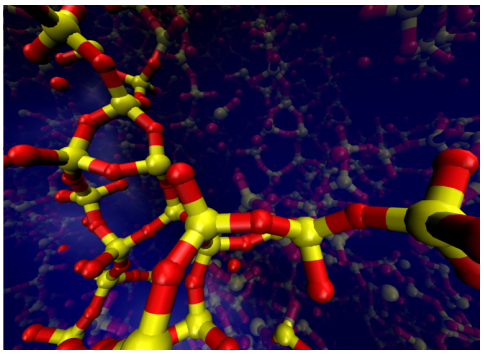

Figure 1. Configuration of a pure silicate gel after the gelation in a wet environment. The structure of these chains is a complex 3D skeleton that includes branching and silicate rings.

To better understand ASR mechanisms and to eventually test these hypotheses, we sought a basic understanding at the atomic scale.

APPROACH

We approached this issue by simulating the formation of gels at the atomic scale. While this allowed us to determine their structure and estimate their mechanical properties, it also presents a challenge; gels form over longer periods that cannot be readily modeled by classical molecular dynamics. We, thereby, used state-of-the-art atomistic methods to overcome the time-scale issue.

Specifically, we coupled parallel tempering to accelerate the dynamics with grand canonical Monte Carlo method to account for the wet environment inside the pores during gel formation. As silicate chains are formed, our model is able to add or remove water molecules. This same method can also be used to change the alkali concentration, to study alkali-Ca exchange during calcification, or to look at the effect of pH.

WHY DOES THIS RESEARCH MATTER?

- To better control ASR, it is necessary to know the molecular structure of the ASR gels.
- A simulation approach offers a more efficient way to examine the structure of gels. Understanding the formation and evolution of gels will also have implications for research on geopolymers and other gel-based materials.
- Gels are not homogeneous, and their structure is not in a state of equilibrium. This makes them challenging to characterize. Progress in these simulation techniques can be used to study other phases such as solutions and liquids.

FINDINGS

This new method allows us to efficiently simulate the formation of gels. This enables us to evaluate the impact of the water to silicon ratio. The modeled gels respond to chemical changes, such as water content, and relative composition of alkali and calcium. More importantly, it demonstrates that drying the gel strongly affects its structure. This drying effect has not been considered in past research and provides an opportunity to reconsider proposed mechanisms from past research. Additionally, we observed that there is a reduction in the volume of gels as it calcifies., which offers further insight into reasons for concrete failure. Currently, we are simulating the formation of the gels that are being investigated in the laboratory. We will compare the mechanical properties (elasticity) with experimental data. These results will give us the basis to test the hypotheses posed in the problem section above.

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