

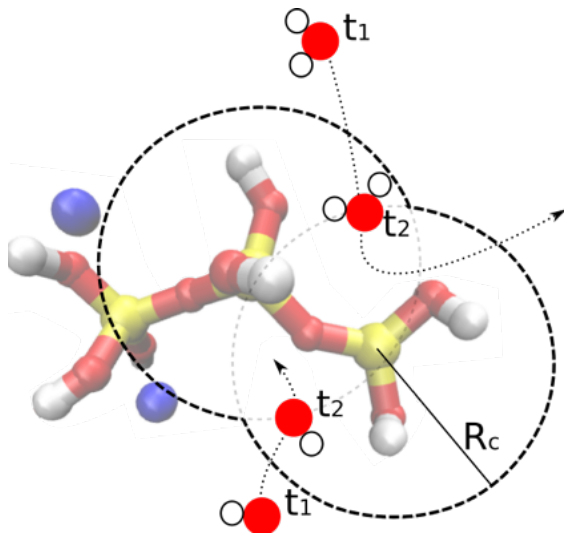
## Questions at the Atomic Scale

With 20% of highway pavements in the U.S. in poor condition, mitigating pavement failure has become increasingly important. However, the mechanisms of the alkali-silica-reaction (ASR), a leading cause of damage in concrete pavements, remain unclear.

Although it is well known that ASR can lead to cracking over many decades, a need still exists to understand the reaction at the atomic scale—in particular, the dissociation and formation of silicates in water. For this, it is necessary to have an atomistic approach.

## Simulating ASR Formation

In this work, we first studied the dissociation of silicates in potentially reactive aggregate when exposed to sodium hydroxide in the cement paste. We then investigated the subsequent formation of silicates in pure water



**Figure 1.** A schematic representation of the algorithm developed to analyze the local environment around silicates during the dissociation by sodium hydroxide. The silicon atoms are represented in yellow, oxygen atoms in red, hydrogen atoms in white, and sodium atoms in blue.

or the presence of alkali to form ASR gel. Since it remains challenging to study ASR over time, we have used state-of-the-art atomistic methods to simulate the different mechanisms that occur over several decades. As shown in **Figure 1**, we have developed a new algorithm to do a statistical analysis of the chemical environment around the silicon atoms.

## Toward Limiting the Reaction

We have found that the dissociation and formation of silicate chains are strongly dependent on the presence of water molecules. Specifically, we have found that in about 20% of the cases, not only one but two water molecules are acting together to dissociate silicates in the presence of sodium hydroxide. This is important for finding the correct reaction pathways and evaluating the energy necessary to dissolve or form the silicates that lead to the formation of ASR gel: energy barriers associated with these reactions offer insight into the speed of the reactions and, therefore, into which step in the mechanism can limit the reaction. With a better understanding of the behavior of these silicates, engineers and scientists could eventually develop new strategies that mitigate ASR in concrete mixtures.

### Key Findings:

- Because the structure of ASR gel is difficult to assess with precision, simulations are an important tool to understand the basic mechanisms of its formation.
- We have developed new algorithms to detail the reaction pathways. This enables the calculation of energy barriers and, therefore, the time-scale of these reactions.
- With this new method, we can study the effect of additives that mitigate the formation of ASR gel or associated expansive cracking.