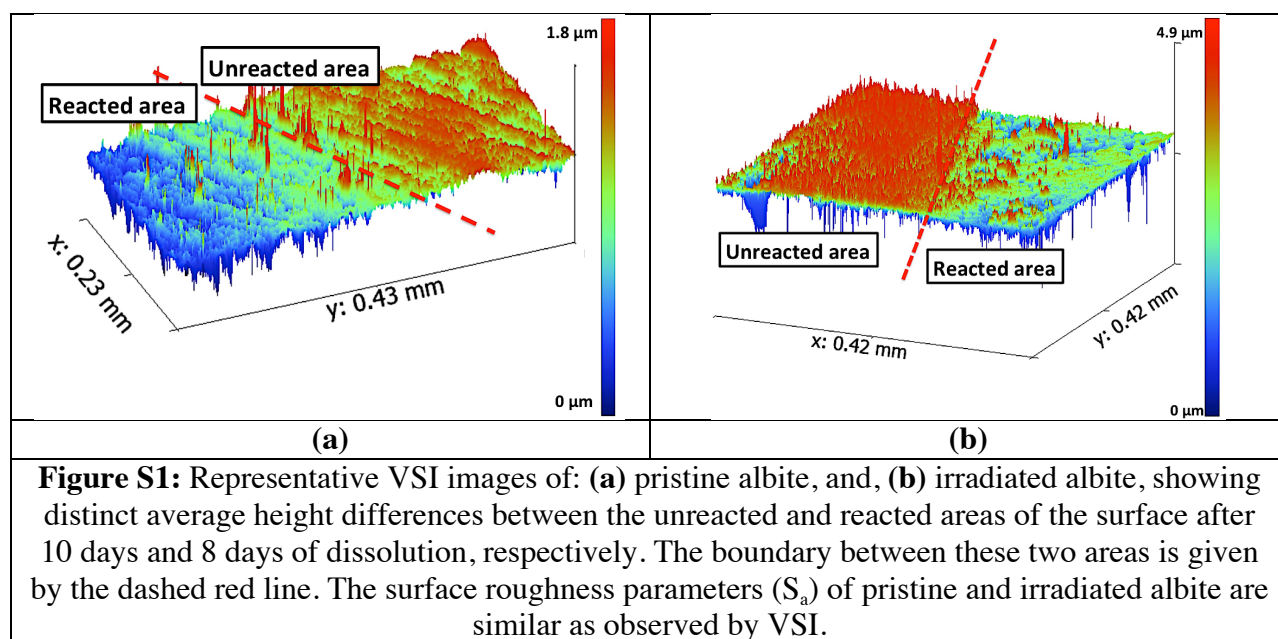


## Effects of Irradiation on Albite's Chemical Durability

Yi-Hsuan Hsiao <sup>(1)</sup>, Erika Callagon La Plante <sup>(1)</sup>, N. M. Anoop Krishnan <sup>(2)</sup>, Yann Le Pape <sup>(3)</sup>, Narayanan Neithalath <sup>(4)</sup>, Mathieu Bauchy <sup>(2)</sup>, Gaurav Sant <sup>(1, 5, 6)</sup>

- Surface topography change assessing by vertical scanning interferometry (VSI):** The dissolution rates of pristine (non-implanted) and irradiated (implanted) albite surfaces were measured using vertical scanning interferometry (VSI), from which quantifications of sub-nanometer alterations of surface height can be obtained. Surface topography parameters (i.e., surface roughness ( $S_a$ ), surface average height ( $\Delta h$ )) were determined for the unreacted and reacted areas on the surface. The dissolution rates were obtained by dividing the difference in average heights between the reference area (unreacted) and sample area (reacted) by the reaction time ( $\Delta t$ ). The analysis of the VSI data was carried out using Gwyddion (version 2.47).<sup>1</sup>



<sup>1</sup> Laboratory for the Chemistry of Construction Materials (LC<sup>2</sup>), Department of Civil and Environmental Engineering, University of California, Los Angeles, CA, USA

<sup>2</sup> Laboratory for the Physics of Amorphous and Inorganic Solids (PARISlab), Department of Civil and Environmental Engineering, University of California, Los Angeles, CA, USA

<sup>3</sup> Oak Ridge National Laboratory, Oak Ridge, TN, USA

<sup>4</sup> School of Sustainable Engineering and the Built Environment, Arizona State University, Tempe, AZ, USA

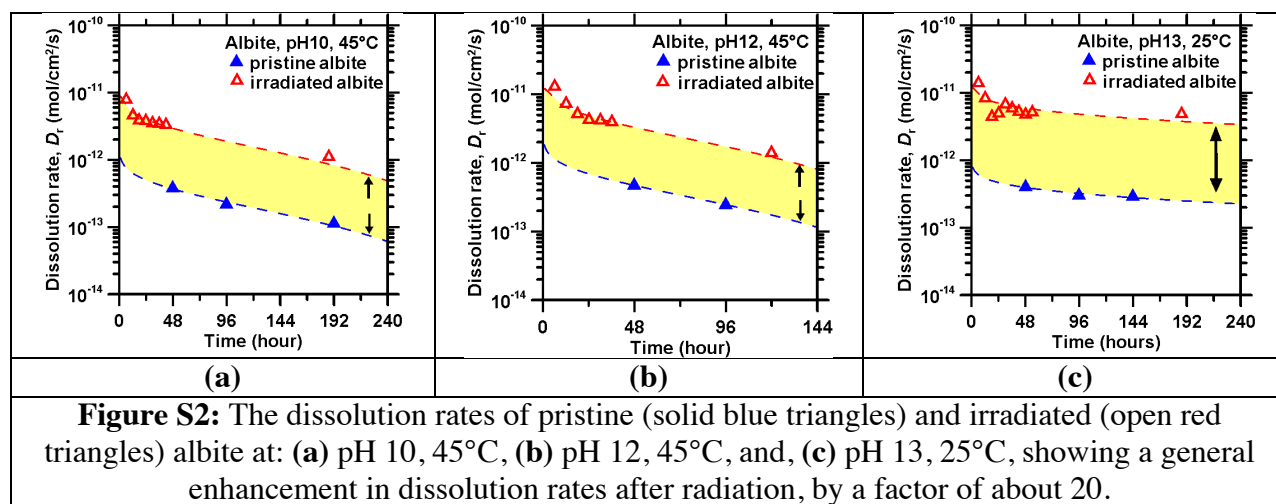
<sup>5</sup> Department of Materials Science and Engineering, University of California, Los Angeles, CA, USA

<sup>6</sup> California Nanosystems Institute, University of California, Los Angeles, CA, USA

\* Corresponding author: G. Sant, Phone: (310) 206-3084, Email: [gsant@ucla.edu](mailto:gsant@ucla.edu)

A section on the albite surface was covered by an inert silicone mask (Silicone Solutions SS-38) to preserve an unreacted part of the surface, which serves as a reference location from which sample height change is measured against. The mask was peeled off after every measurement and then reapplied at the same position before resuming reaction of the sample with the solution. The mask was found to adhere well to the surface, and the solution did not penetrate under the silicone mask as would be apparent from peeling of the mask (or a vertical swelling in the region of the mask), which was not observed. As shown in Figure S1 (a,b), the boundary between the reacted and unreacted areas remained sharp after extended times (up to 10 days) of exposure to the solution.

- The dissolution rates of irradiated and pristine albite across a range of pH and temperature:** Dissolution rates were measured for 2 temperatures (25 and 45 °C) and 3 solution pH levels (pH 10, pH 12 and pH 13). Specifically, 0.1 mM NaOH (pH 10), 10 mM NaOH (pH 12), and 100 mM NaOH (pH 13) were selected to simulate the alkaline conditions that persist in the pore solution in concrete. In general, the dissolution rates of irradiated albite are about 20 times higher compared to pristine albite, over the entire range of temperature and pH studied herein, as shown in Figure S2 and Table S1 below.



**Table S1:** The steady state dissolution rates of pristine and irradiated albite at pH 10, 12, 13, and at 25 and 45 °C, measured using VSI. These values are used to calculate the dissolution enhancement ratios (DER) for each experimental condition.

Pristine albite ( $\times 10^{-13}$ mol/cm <sup>2</sup> /s)	pH 10	pH 12	pH 13
25 °C	$1.10 \pm 0.86$	$1.74 \pm 0.44$	$3.32 \pm 0.61$
45 °C	$2.36 \pm 1.33$	$2.84 \pm 1.70$	$2.66 \pm 1.66$
Irradiated albite ( $\times 10^{-13}$ mol/cm <sup>2</sup> /s)	pH 10	pH 12	pH 13
25 °C	$24.70 \pm 1.70$	$34.40 \pm 4.40$	$48.10 \pm 2.01$
45 °C	$33.40 \pm 1.06$	$39.70 \pm 0.63$	$42.03 \pm 1.51$

- **Topological constraints calculation:** In fully connected glasses, the number of constraints ( $n_c$ ) created by each atomic element  $i$  is determined by its coordination number ( $r_i$ ). Namely, each atomic species features  $r_i/2$  bond-stretching (BS) constraints (since each radial constraint is shared by two atoms) and  $2r_i - 3$  bond-bending (BB) constraints (the number of independent angles required to define a polyhedron). The total number of constraints per atom ( $n_c$ ) is then obtained by summing the BS and BB constraints. However, more complex ionic systems can feature some thermally-broken constraints (e.g., non-directional Si–O–Na bonds do not feature any angular BB constraint). To overcome these difficulties, the number of BS and BB constraints is directly computed by molecular dynamics simulations following a well-established methodology.<sup>2,3</sup> In brief, we compute the radial and angular excursions of the neighbors of each atom. For instance, low (high) radial excursions indicate the presence (absence) of an underlying constraint, since each constraint prevents the relative motion between a pair of atoms. Note that this methodology clearly discriminates intact from thermally-broken constraints.

## References

- (1) Nečas, D.; Klapetek, P. Gwyddion: an open-source software for SPM data analysis. *Cent. Eur. J. Phys.* **2012**, 10, 181-188.
- (2) Bauchy, M.; Micoulaut, M. Atomic scale foundation of temperature-dependent bonding constraints in network glasses and liquids. *J. Non-Cryst. Solids* **2011**, 357, 2530-2537.
- (3) Bauchy, M.; Qomi, M.J.A.; Bichara, C.; Ulm, F.-J.; Pellenq, R. J.-M. Nanoscale Structure of Cement: Viewpoint of Rigidity Theory. *J. Phys. Chem. C* **2014**, 118, 12485–12493.