

Reverse chemical garden reaction of cementitious materials

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Recent advances in the nano-scale mineralogy extend from the extraterrestrial materials known as in cosmic dusts and asteroids to ordinary industrial materials like the cementitious materials. The distinct property of nano materials can be characterized from the points of view of (1) nucleation, (2) self-assembly behavior and (3) flexibility in the form.

A very common industrial material, cement is a typical one consisted of nano particles of calcium silicate hydrates (C-S-H).

Crystal growth experiment of cementitious materials can be recently performed using interferometric and nanoscopic techniques. Although the cement reaction producing C-S-H from silicates with Ca(OH)₂ (portlandite: CH) or more alkaline solution is extensively occurring at buildings under and after construction, similar reaction is expected at the interface of natural rock and concrete-building such like tunnel, dam and underground repository for radioactive wastes.

Hyper alkaline alteration experiment using vertical scanning interferometer revealed the precipitation behavior of C-S-H by reverse chemical garden reaction on natural rock (Satoh et al., in press). Very slow growth rate of the C-S-H on rock was observed to be $\sim 2.4E-3$ nm/s. The ionic selection of the solutes through the C-S-H wall having nanopores like membrane was also confirmed.

Most recently, we applied newly developed ultramicroscopic technique with fluid reaction TEM (FR-TEM: Poseidon) for study of reverse chemical garden reaction between silica fume (Elkem Microsilica 940-U, ~ 150 nm) and CH-saturated solution. It revealed that the reaction caused silica hydration (volume expands) and subsequently form string and veil of C-S-H. The growth rate of string C-S-H was calculated to be $\sim 4.5E-2$ nm/s, which is fast enough to form frame network preparing veil-formation. It was chemically confirmed by FESEM-EDS that this C-S-H veil evolved toward Ca-rich over time. Our observed result could be a fundamental process of reverse chemical garden reaction, i.e., cement-solidification.

Ultramicroscopic investigation of C-S-H growths may improve the simulation of groundwater conditions in the future.

Keywords: reverse chemical garden reaction, cementitious material, C-S-H, fluid reaction TEM