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Alteration of nanocrystalline Calcium Silicate Hydrate (C-S-H) at pH 9.2 and room temperature

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In the French design of deep underground radioactive waste disposal, cementitious materials will be used to build access structures, galleries, vaults and waste packages for Intermediate Level Long-lived Waste (ILLW). Part of these cementitious materials will be in physical contact with the Callovian-Oxfordian clay host rock formation (COx). The pH of the COx pore water is around 7 and its composition is very different from the cementitious pore water. Due to this chemical contrast, a chemical gradient will develop at the cement/clay interfaces and will induce mineralogical transformations.

Nanocrystalline calcium silicate hydrates (C-S-H) are the main constituents of concretes and are essential for cohesion of cement paste. It is therefore fundamentally important that we be able to reliably estimate C-S-H weathering/dissolution rates if we wish to estimate the durability of various cementitious materials. In order to mimic a clay/concrete interface, C-S-H alteration was studied with flow-through experiments at 25 °C and pH 9.2. Three materials with apparent Ca/Si ratios of 1.47, 1.38 and 0.86 were investigated. Physical (TGA/DTA), mineralogical (XRD) and chemical (EPMA, TEM/EDX) analyses were performed to characterize the reacting minerals. Initial stoichiometric Ca/Si ratios (C/S) were 1.22, 1.22 and 0.85, respectively. The excess of calcium is attributed mainly to the presence of CH intimately mixed in with C-S-H particles. Under our experimental conditions the stoichiometric C/S ratios decreased continuously with time. C-S-H compositions were monitored during flow-through experiments in order to know the mineral stoichiometry. The kinetics were found to be a function of the C/S. A decrease in layer-to-layer distance in the early stage of the alteration process is interpreted as interlayer Ca/Na exchange. A second dissolution step, marked by the stoichiometric release of calcium and silicate, certainly results from layer dissolution.