Dissolution rates and surface chemistry of calcium aluminosilicate glasses in cementitious systems

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Waste calcium aluminosilicate (CAS) glasses are widely used as supplementary cementitious materials in Portland-cement based construction materials for environmental, economical and technical reasons. The amount of cement that can be replaced by waste CAS is largely depending on their reactivity and composition. Understanding the reactivity or rates of dissolution of these wastes in typical Portland cement pore solutions of high ionic strength and alkalinity (pH ≈ 13) is of great importance to optimise the performance, durability and sustainability of the blended Portland cements.

Water quenched synthetic CAS glasses of selected compositions ranging from pure silica framework to CaO-rich depolymerized glasses were subjected to batch dissolution reactions avoiding hydration product precipitation. The obtained dissolution rates scaled linearly with the Ca/(Al+Si) molar ratio, and showed a large difference in reactivity between framework and partially depolymerised glasses (Fig. 1).

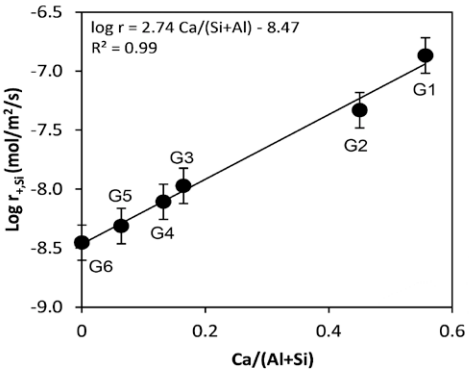

Figure 1: Dependence of glass dissolution rate at pH 13 on the molar Ca(Al+Si) ratio of the glass.

Glass dissolution rates also depended on solution composition. Increasing Al activity decreased dissolution rates of framework glasses, while increasing Ca activity decreased dissolution rates of all glasses. The glass surface chemistry as a function of solution pH and composition was investigated by XPS, zeta potential measurements and surface titrations and lead to a model of CAS glass dissolution for high pH cement type environments.

Isotopic Tomography of Monazite

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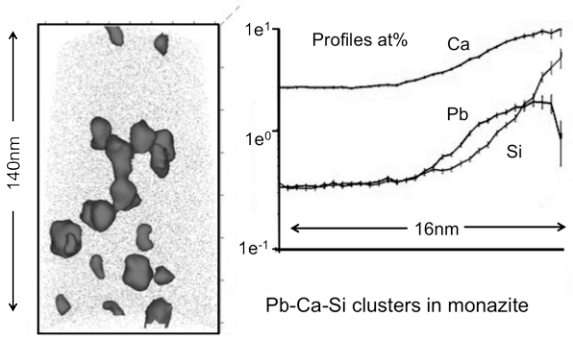
In Atom Probe Tomography (APT) a specimen is evaporated atom by atom, and the mass/charge and original position (X,Y, Z to 0.2nm) of each detected atom is recorded to produce a quantitative 3-D reconstruction at the atomic scale. APT can also provide *in situ* atomic scale isotopic tomography of U, Th and radiogenic Pb, allowing access to otherwise cryptic thermochronologic information.

A polymetamorphic monazite grain from the Churchill Province, Canada was selected for analysis by APT. Prior EPMA analysis indicates monazite crystallization at 2.55Ga, overgrowth at 2.37Ga, and fracture-infill and further overgrowth at 1.84Ga. Datasets of up to 200 million atoms were collected, sampling the various domains and their mutual interfaces.

Distinct clusters of Pb atoms <10nm dia. and spaced at 20–30nm were observed in the older, reheated domain. These clusters contain up to 1.7 at.% Pb, 4 at.% Si and 7 at.% Ca in a radial structure. These clusters closely resemble those found by APT in reheated zircon [1. Valley *et al.*]. The presence of 208Pb, 207Pb and 206Pb in the clusters indicates post-crystallization segregation of radiogenic Pb from both the Th and U decay chains. Because Th and U enter the monazite structure via coupled substitution (e.g. Th+Ca, Th+Si and U+Si), Si and Ca in the clusters may represent the unpaired atoms after radionuclide decay. To measure an isotopic age at this scale, precise 207Pb/206Pb determination requires a minor correction for interfering species such as (ThPO3)3+.



[1] Valley, J. W. *et al.* (2012) Elemental and isotopic tomography at Single-Atom-Scale in 4.0 and 2.4 Ga zircons. Abstract V12A-05 presented at 2012 Fall Meeting, AGU, San Francisco, Calif., 3-7 Dec.

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