Quantitative modelling of the degradation processes of cementitious components

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The chemical alteration of cement components is a very relevant process in the performance assessment of radioactive waste repository due the large amounts cementitious materials involved. This alteration process has been repeatedly performed in laboratory, and, in general, the data obtained show a dependence between the pH of porewater and the Ca/Si ratio of the hydrated calcium-silicate (CSH) phases. The Ca/Si ratio decreases as the CSH is progressively replaced by Si-enriched phases since CSH dissolution is clearly incongruent. The short-term data from laboratory experiments are further used in reactive transport numerical calculations to evaluate the long term behaviour. Recently, the application of the aqueous solution-solid solution equilibrium theory has provided a new way to numerically model the geochemical processes occurring during the alteration of cementitious materials. This modelling presents the advantage to easily simulate the incongruent dissolution of the cement, in which Ca is preferentially solubilised with respect to Si.

In this work, the solid solution approaches of Sugiyama and Fujita [1] and Carey and Lichtner [2], which include the same end-members ($Ca(OH)_2$ and SiO_2), are used to reproduce the $[Ca_{aq}]$-$[Si_{aq}]$-pH-Ca/Si_{CSH} relationships from experimental data in literature. In addition, using the Sugiyama and Fujita [1] approach, the porosity changes and the Ca/Si_{CSH} observed in some of the Yamamoto *et al.* [3] experiments are numerically investigated. The results indicate that the initial porosity and the effective diffusion coefficient are key parameters to reproduce the observed alteration pattern.

[1] Sugiyama & Fujita (2006) *Cem. Concr. Res*. **36**, 227-237.
[2] Carey & Lichtner (2007) *Mat. Sci. of Concrete Spec.* Vol.
[3] Yamamoto *et al.* (2007). 3rd Workshop :R&D on low pH cement for a geological repository. Paris.

Zircons from the Apollo 17 Breccias: Implications for the Early History of the Moon

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One of the unique features of the Moon is the overabundance of zircon in many mafic rocks, which appears to be linked to the enrichment of these rocks in the incompatible elements. This enrichment is interpreted to originate from the final stages of crystallization of the Lunar Magma Ocean (LMO), when incompatible elements were concentrated in the residual reservoir referred to as KREEP for its high content in K, REE and P. As this reservoir was enriched in the radioactive elements, it constituted a substantial source of heat and the focus of extensive magmatic activity long after the LMO crystallization. The rocks formed as a result of this activity inherited high concentrations of incompatible elements from the KREEP reservoir, including zirconium, which resulted in the widespread crystallization of zircon in these rocks.

Abundance of zircon in the lunar rocks, their link with the KREEP reservoir and their known stability under extreme P-T conditions create an opportunity to obtain significant chronological and petrological information related to the last stages of LMO crystallization and to the formation of plutonic rocks that followed crystallization of lunar crust. In addition, it is becoming evident that some lunar zircons preserve evidence of impacts that predate the 3.8-3.9 Ga event recorded by the other isotopic systems. Apollo 17 samples are particularly important as the data obtained so far indicate that the zircons from this site reflect the pre-4.2 Ga history of the Moon, with about 75% of the zircon ages older than 4.3 Ga.

Several large and texturally complex zircon grains have been identified in the Apollo 17 breccias. Detailed U/Pb study of these grains, combined with the high resolution imaging to reveal their internal textures, indicate that KREEP reservoir was formed on the Moon by at least 4417±6 Ma and hence the LMO was mainly crystallized at this time. These studies also suggest that major magmatic activity subsided significantly by about 4.3 Ga and completely ceased by 4.2 Ga in the Serenitatis impact area (proposed as a source of Apollo 17 breccias). Finally, zircon data imply that at least two major impacts occurred on the Moon before 3.8-3.9 Ga, one at 4334±8 Ma and the other at 4187±11 Ma.