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Calcium silica speciation and carbonation in cement systems

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Deep disposal concepts for radioactive waste are usually based on a multiple barriers which may involve cementitious materials and thus alkaline media. In such regard, silica and calcium speciation is of major importance for geochemical calculations. The existence of mixed calcium-silica complexes was inferred in previous works [1, 2]. The present study proposes first an assessment for the influence of such complexes on cement hydrates (C-S-H) solubility. Then, a set of lab experiments and measurements is realised to assess the stability of such complex in alkaline media. Experiments consists in preparing solutions in CO₂ preserved conditions. The solutions are filtered up to 3kD. Several analytical technics are used to characterize the filtered solutions on the filtered solutions (Raman, ²⁹Si NMR, High energy X-ray, acidic titrations, Ca-ISE, ICP). Measurements do not provide unambiguous proof of the presence of mixed calcium-silica complexes in the solutions. However, the results clearly indicate a partial carbonation of the system. Modelling the titration experiments helps in understanding the extent for such carbonation, with strong suspicion for calcite and C-S-H precipitation in some cases. Finally, previous works dedicated to calcium-silica complexes are re-investigated and previous results are re-assessed regarding for the influence of carbonation, which is able to explain modifications of the overall speciation without involving the presence of mixed silica complexes. This contribution evaluates also the main sources of uncertainty affecting to the previous experimental studies [1, 2], and the possible impact in the reported thermodynamic functions.

[1] Santschi P. H. and Schindler P. W. (1974) Journal of the Chemical Society, Dalton Transactions, 181-184.

[2] Nicoleau L. and Schreiner E. (2017) Cement and Concrete Research 98, 36-43.