Role of alkali cation in early stage of oligomerization in silicate fluids: a molecular dynamics study

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Abstract (300 word limit)

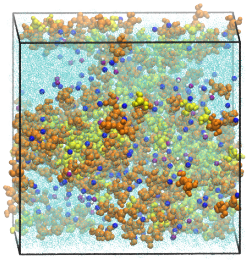
Alkali activation solutions of alumino-silicates are widely used in building and construction industry as "sealing layers". They are considered as green chemistry. They lead to solid material by a sol-gel transition that strongly depends on the composition.

It has been proved that the nature and the concentration of the alkali cation drive the final properties of the gel[1]. We developed theoretical tools to characterize concentrated alkali solutions of silicates to understand the structural and dynamical properties of such fluids. The influence of the composition of the alkali aqueous solutions on the physico-chemical properties of silicate oligomers identified in aqueous solutions by NMR[2] and mass spectrometry[3] has been observed. Here, the influence of the MOH concentration (M+ being an alkali cation), as well as the nature of the alkali cation has been investigated by means of classical molecular dynamics using explicit polarization. To this end, polarizable force fields for describing the solvation properties of the hydroxide anion and the silicate oligomers have been developed from non-polarizable force field[4,5].

Simulation of hydroxide alkali solution from 0.5 up to 9.66 mol L-1 was carrying and structural properties were studied. The density, WAXS spectra were calculated and directly compared to experiments measurements. A good agreement between the theoretical results and the experiments was observed[6]. Furthermore, we investigated the effect of the addition of silicate oligomers on structural properties of the solutions. Changes in the oligomerization process are observed as a function of the nature and the concentration of the oligomers i.e., monomers ($Si(OH)_4$, $SiO(OH)_3^-$), dimers ($Si_2O_2(OH)_5^-$, $Si_2O_3(OH)_4^{2-}$).

Recent Publications (minimum 5)

1. A. Poulesquen, F. Frizon and D. Lambertin, Rheological behavior of alkali-activated metakaolin during geopolymerization, Journal of Non-Crystalline Solids, 357 (21): 3565-3571
2. C. T. G. Knight, R. J. Balec, and S. D. Kinrade. (2007), The Structure of Silicate Anions in Aqueous Alkaline Solutions. Angewandte Chemie International Edition, 46: 8148–8152
3. P. Bussian, F. Sobott, B. Brutschy, W. Schrader, and F. Schüth, (2000). Speciation in Solution: Silicate Oligomers in Aqueous Solutions Detected by Mass Spectrometry.Angewandte Chemie International Edition, 39: 3901-3905.
4. J. R. Pliego, Jr. and J. M. Riveros (2000), On the Calculation of the Absolute Solvation Free Energy of Ionic Species: Application of the Extrapolation Method to the Hydroxide Ion in Aqueous Solution. The Journal of Physical Chemistry B, 104 (21): 5155-5160
5. M. Jorge, J. R. B. Gomes, M. N. D. S. Cordeiro, and N. A. Seaton, Molecular Dynamics Simulation of the Early Stages of the Synthesis of Periodic Mesoporous Silica. The Journal of Physical Chemistry B, 113: 708-718.
6. A. Coste, J.-F. Dufrêche, O. Diat, A. Poulesquen and M. Duvail, Investigation of the structure of concentrated NaOH solutions: development of a polarizable model for molecular dynamics, in preparation



Image

Figure 1: simulation box of monomers $Si(OH)_4$ (yellow) and dimers $Si2O2(OH)_5^-$ (orange) at 2.0 mol kg^{-1} in aqueous solutions of 5 mol L^{-1} of NaOH. For clarity, only Na^+ (blue) and OH^- (purple) close to silicate oligomers are represented (distance ions-Si atoms inferior to 6 Å).



Biography (150 word limit)

Amaury Coste is a second year PhD student of the University of Montpellier. He is working at the Marcoule Institute of Marcoule (ICSM) in the Mesoscopic Modelling and Theoretical Chemistry Laboratory (LMCT). He is supervised by Magali Duvail, researcher at the CEA and Jean-François Dufrêche, Professor at the University of Montpellier.

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Session name/ number: Theoretical and Computational Chemistry 1-5
Category: (Oral Presentation/Poster Presentation)

Any Speaker Suggestions:

Notes/Comments: