

Supplementary material for: Condensation and Growth of Amorphous Aluminosilicate Nanoparticles by Aggregation Process

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I. EXPERIMENTAL DETAILS

Sodium silicate solutions were prepared by dilution of sodium silicate solution: Betol 39T, $[\text{Si}] = 6.26 \text{ mol.L}^{-1}$, $[\text{Na}^+] = 3.64 \text{ mol.L}^{-1}$ and sodium aluminate solution: Stabisil, $[\text{Al}] = 5.88 \text{ mol.L}^{-1}$, $[\text{Na}^+] = 9.44 \text{ mol.L}^{-1}$. Sodium hydroxide pellets were added to adjust the level of sodium and hydroxide anions in both solutions. Deionized water from a Milli-Q Water purification system was used in all dilution processes. Each solution was left at equilibrium for 24 hours before experiments or characterizations. Solutions are mixed steadily at equal volumetric flow rates to avoid high reactivity between the silicate and aluminate entities. The gel was prepared with a final concentrations of: $[\text{Si}] = 1.125 \text{ mol.L}^{-1}$, $[\text{Na}] = 1.31 \text{ mol.L}^{-1}$, $[\text{Al}] = 0.12 \text{ mol.L}^{-1}$. Two SAXS lab benches were used to characterize the gel state and the solutions. The first one was already describe in a first paper [1] and the range of scattering vector is $q = 0.025\text{-}3 \text{ \AA}^{-1}$. The second one uses a Rigaku 002 microfocus X-ray source (Cu-radiation $\lambda = 1.54 \text{ \AA}$) with an Osmic staggered parabolic multilayer optics to focus the beam crossover at the second pinhole. Samples are introduced into a large vacuum chamber that is pumped down to 0.08 mbar. The detector moves from from 100 mm to 1500 mm from the sample. For our experiment, the sample to detector distance was fixed to 1500 mm corresponding to a q-range of $[0.0027\text{-}0.18 \text{ \AA}^{-1}]$. 1.5 mm capillary tubes was used to measure the scattering of the gel and the acquisition time was fixed to 600 s. The scattered intensity is transformed in absolute intensity (in cm^{-1}) after normalization.

II. EVOLUTION OF THE SPECIES WITH THE NUMBER OF ALUMINUM

In the simulation, there is a strong dependancy of the aluminosilicate cluster size with the amount of aluminum included in it. Moreover, aluminosilicates with more aluminum tends to form tighter cages with silicate rings. The figure 1 shows the different structures that can be obtained after the mixing of the two solution. It shows that aluminates are always connected to a silicate in the aluminosilicate structures that have been obtained.

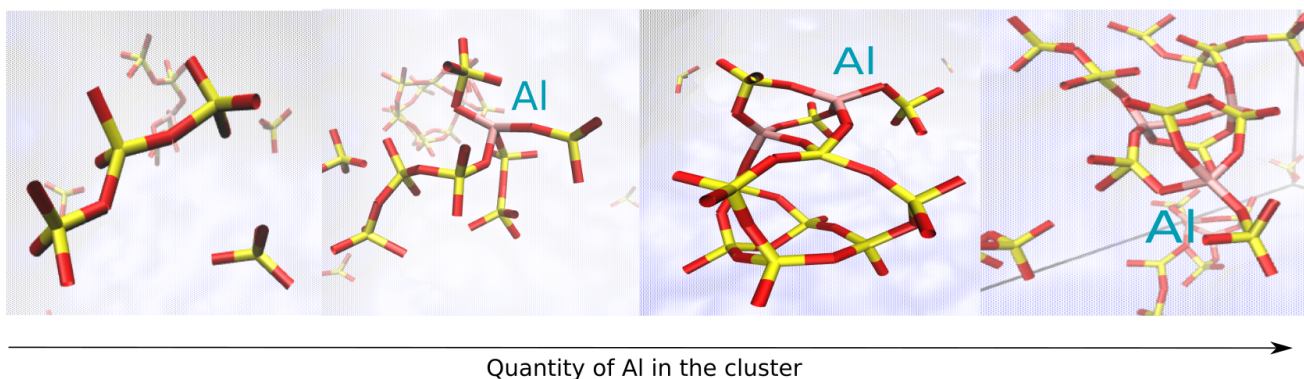


Figure 1. Snapshots of different species with 0,1,2 or 3 aluminates.

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- [1] R. Dupuis, D. G. Rodrigues, J.-B. Champenois, R. Pellenq, and A. Poulesquen, Time resolved alkali silicate decondensation by sodium hydroxide solution, *Journal of Physics: Materials* **3** (2019).