

Structure of CaO-Al₂O₃-SiO₂ melts studied by molecular dynamics and diffraction experiments

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The structural characterization of multicomponent liquids is a challenging problem. From the experimental perspective, measurements need to be performed *in situ* at high temperature (and possibly high pressure). Furthermore, a single method such as x-ray or neutron diffraction, Raman or NMR spectroscopy is usually not sufficient to provide a reliable structural model. On the other hand, molecular modeling approaches using classical interatomic potentials often lack accuracy and computationally expensive first-principles simulations suffer from finite size and time effects. It appears attractive to combine the strengths of different techniques to investigate the structure of chemically complex melts and the relation between their structure and their physical or thermodynamic properties.

Calcium aluminosilicate melts and glasses are important in both geological and technological context. Here, we employ advanced classical interaction potentials that were parametrized using electronic structure calculations [1] to predict the structure of representative liquids of the CaO-Al₂O₃-SiO₂ system at ambient pressure and a temperature of 2500 K as a function of chemical composition. The quality of the simulation model is assessed by comparing the computed total static structure factors to data from x-ray and neutron diffraction experiments (e.g. [2]). In this presentation, we will focus on structural changes across the binary CaO-SiO₂ and CaO-Al₂O₃ as well as the metaluminous (CaAl₂O₄)-SiO₂ joins. Similar to the MgO-Al₂O₃ system [3], the average Al coordination decreases from about 4.4 in pure Al₂O₃ melt to 4.0 in Ca-rich melts. At the same time, the number of OAl₃ triclusters is reduced significantly. The distributions of Ca coordination are rather broad but they are dominated by a sixfold octahedral geometry and show a decreasing average coordination towards the Ca-rich melts. In the glass-forming region of the CaO-Al₂O₃ system, a maximum in the melt viscosity is observed well above the glass transition temperature.

[1] Jahn & Madden (2007) *Phys. Earth Planet. Int.* **162**, 129–139. [2] Drewitt *et al.* (2011) *J. Phys. Condens. Matter* **23**, 155101. [3] Jahn (2008) *Am. Mineral.* **93**, 1486–1492.

Role of microbe in the formation of illite from nontronite: Mesophilic and thermophilic bacterial reaction

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The formation of illite through the smectite-to-illite (S-I) reaction is considered to be one of the most important mineral reactions occurring during diagenesis. In biologically catalyzed systems, however, this transformation has been suggested to be rapid and to bypass the high temperature and long-time requirements. To understand the factors that promote the S-I reaction, the present study focused on the effects of pH, temperature, solution chemistry, and aging on the S-I reaction in microbially mediated systems. Fe (III)-reduction experiments were performed in both growth and non-growth media with two types of bacteria: mesophilic (*Shewanella putrefaciens* CN32) and thermophilic (*Thermus scotoductus* SA-01). Reductive dissolution of NAu-2 was observed and the formation of illite in treatment with thermophilic SA-01 was indicated by X-ray diffraction (XRD) and high-resolution transmission electron microscopy (HRTEM). A basic pH (8.4) and high temperature (65°C) were the most favorable conditions for the formation of illite. A long incubation time was also found to enhance the formation of illite. K-nontronite (non-permanent fixation of K) was also detected and differentiated from the discrete illite in the XRD profiles. These results collectively suggested that the formation of illite associated with the biologically catalyzed smectite-to-illite reaction pathway may bypass the prolonged time and high temperature required for the S-I reaction in the absence of microbial activity.