American Mineralogist, Volume 96, pages 744–751, 2011

First-principles study of diffusion and viscosity of anorthite ($CaAl_2Si_2O_8$) liquid at high pressure

BIJAYA B. KARKI,[1,2,]* BIDUR BOHARA,[1] AND LARS STIXRUDE[3]

[1]Department of Computer Science, Louisiana State University, Baton Rouge, Louisiana 70803, U.S.A.
[2]Department of Geology and Geophysics, Louisiana State University, Baton Rouge, Louisiana 70803, U.S.A.
[3]Department of Earth Sciences, University College London, London WC1E 6BT, U.K.

ABSTRACT

We have carried out equilibrium molecular dynamics simulations of $CaAl_2Si_2O_8$ (anorthite) liquid as a function of pressure (up to 160 GPa) and temperature (2500 to 6000 K) within density functional theory. Along the 3000 K isotherm, the Ca self-diffusivity varies most (decreasing by two orders of magnitude between 0 and 50 GPa), whereas the self-diffusion coefficients of Al, Si, and O vary anomalously—they initially increase with pressure, reach the broad maxima (around 5 GPa), and then decrease upon further compression. The calculated melt viscosity also shows a weak anomalous behavior with a local minimum around a similar pressure. Temperature suppresses the dynamical anomalies as well as the overall pressure variations. Therefore, the curvatures of the diffusivity (viscosity) isotherms change from a concave (convex) shape at 3000 K to a convex (concave) shape at 6000 K. We find that anorthite liquid is much more mobile than silica liquid because of its high content of non-bridging oxygen atoms (NBO) and oxygen triclusters (O3). The predicted pressure variations can be associated with structural changes consisting of the pressure-induced maximum in the abundance of pentahedral states (fivefold Al/Si-O coordination) and rapid increase in the O3 abundance. Finally, our predicted first-principles results compare favorably with the available experimental data.

Keywords: Anorthite liquid, diffusion, viscosity, first-principles simulations, high pressure