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Probing the effect of composition on structural disorder of basaltic and slab-driven melts using solid state NMR

Sun Young Park[1*], Sung Keun Lee[1]

[1]Seoul National University

Whereas the structure of multi-component silicate melts has strong implication for the properties of natural silicate melts and relevant magmatic processes in mantle and crust of the Earth, little is known about their atomic structures due to lack of suitable experimental probes of multi-component amorphous oxides. Whereas most of the progress in melt structure has been made for relatively simple binary and ternary silicate glasses, recent advances in high-resolution solid-state NMR unveil previously unknown structural details of multi-component silicate melts. Here, we report the experimental results of the effect of composition on the atomic structure and disorder in quaternary [CaO-MgO-Al_2O_3-SiO_2 (CMAS)] using multi-nuclear high-resolution 1D and 2D solid-state NMR. We also report the first NMR results for the diverse glasses with compositions of natural silicate melts. The Al-27 NMR results for diopside- Ca-Tschermakite pseudobinary join, suggest a increases in topological and configurational disorder with increasing diopside content. While the glasses with basaltic compositions show that $^{[4]}$Al is dominant, non-negligible fraction of $^{[5]}$Al were observed for basaltic composition melts while negligible fraction of $^{[5]}$Al was observed for the slab-driven melts . The high-resolution O-17 3QMAS NMR spectra of diopside-Ca-Tschermakite pseudobinary join show that three types of bridging oxygens (BO; Si-O-Si, Al-O-Al, and Si-O-Al) and two types of NBO (Ca-NBO, and mixed?NBO) are partially resolved. Previously unknown structural details in Ca-Mg aluminosilicate glasses include nonrandom distributions of Ca^{2+} and Mg^{2+} around NBO and BO and significant fraction of Al-O-Al in natural basaltic magmas. The preferential partitioning of Ca^{2+} and Mg^{2+} between NBO and BO may results in a variation of activity coefficient of CaO and MgO, thus controlling composition of melts generated at the mid-ocean ridge and subduction zone.

Keywords: basaltic melt, multi-component glass, NMR, atomic structure