A unified relation between viscosity and coordination structure for polymerized liquids

*Yoshio Kono[1], Lei Liu[2], Daijo Ikuta[3], Yanbin Wang[4], Guoyin Shen[5]

1. Geodynamics Research Center, Ehime University, 2. Uppsala University, 3. Tohoku University, 4. University of Chicago, 5. Argonne National Laboratory

Silicate melts are known to have polymerized liquid structures, and the viscosities of silicate melts vary by many orders of magnitude with composition. Structures of silicate melts are traditionally parameterized in terms of the number of non-bridging oxygen per tetrahedral SiO_4 unit (NBO/T). One can roughly estimate the apparent NBO/T of a given substance based on its chemical composition using the relation, NBO=2×O-4×T, where O and T are the numbers of oxygen and Si (also include Al, Ti, etc.), respectively, per chemical formula. Some studies have proposed polynomial relations between viscosity and NBO/T and have shown that the viscosity of silicate melts increases with decreasing NBO/T (i.e., with increasing degree of polymerization).

Viscosity of not only silicate melts but also other polymerized liquids is considered to be primarily controlled by their polymeric architecture. Hence a systematic investigation with a unified polymeric architecture view point may provide important insights into the viscosity-structure relation of silicate melts. Here, as the first step toward this goal, we investigate the structure and viscosity of liquid sulfur and selenium, the simplest elemental “polymers”, under various pressure and temperature conditions. Both liquids, sulfur at high pressures and selenium at ambient pressure, are known to have chained structures. We find a unified relation between viscosity and coordination number for liquid sulfur and selenium. These simple liquids provide fundamental insights into the viscosity-structure relation of polymerized liquids with much more complex compositions and structures such as silicate melts. For silicate melts, we introduce the concept of average oxygen chain length, which is closely related to NBO/T. We find a striking similarity in the correlation between viscosity and structure to that found in simple elemental liquids.

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