AMORPHOUS MATERIALS: PROPERTIES, STRUCTURE, AND DURABILITY

Effects of chemical composition and temperature on transport properties of silica-rich glasses and melts

ANNE M. HOFMEISTER¹,*, ALAN G. WHITTINGTON², JONAS GOLDSAND¹ AND REINHARDT G. CRISS³†

¹Department of Earth and Planetary Sciences, Washington University, St. Louis, Missouri 63130, U.S.A.
²Department of Geological Sciences, University of Missouri, Columbia, Missouri 65201, U.S.A.
³Horton-Watkins High School, Ladue, Missouri 63124, U.S.A.

ABSTRACT

Combining new measurements of thermal diffusivity (D) and viscosity (η) of 13 silica-rich glasses and their melts with previous data reveals specific effects of Al, Ca, and Fe cations on heat and mass transport for diverse glasses and melts. We investigated rhyolites, tektites, leucogranite, haplogranite, and chemically complex commercial glasses. Highly polymerized samples, with high-Al but low-Ca contents, yield high values for η, D, and glass transition temperatures (T_g,12), whereas less polymerized samples with high-Ca but low-Al contents, have low η, D, and T_g,12. Upon crossing the glass transition, D decreases substantially, to ~0.35 mm²/s for Ca-rich melts, but D decreases only weakly, to ~0.52 mm²/s for Al-rich melts. The magnitude of the decrease in D at T_g,12 correlates with the melt fragility, and also to the configurational heat capacity. High-Ca contents result in low D for glasses and melts, whether or not Al is present. At high T, ∂D/∂T is positive for glasses and melts containing Fe²⁺, which we attribute to diffusive radiative transfer involving electronic-vibronic coupling. Thermal conductivity of all glasses increases with T, flattening out as the transition is approached. For melts with >1 wt% FeOtotal, ∂k/∂T is positive. We predict that upon melting, 1-type are granite liquids should have lower thermal diffusivity than calcium-poor A- or S-types, and calc-alkaline basalts will have lower D than tholeitic basalts, such that D of granitic melts is ~0.2 mm²/s higher than basaltic. Ferrous iron enhancing heat transport could alter the predicted order at higher temperatures.

Keywords: Laser-flash analysis, high-temperature, thermal diffusivity, viscosity, hydration, impurities, glass, melt

INTRODUCTION

Transport properties of rocks and magmas strongly influence igneous processes (e.g., Nabelek et al. 2012). Mass transport of a melt is described by viscosity (η), for which several predictive models exist as functions of temperature and composition (e.g., Hui and Zhang 2007; Giordano et al. 2008). Heat transport is described by thermal conductivity (k). Direct measurements of k of melt using contact methods are not reliable, because at the high temperatures (T) required, ballistic radiative transfer gains exceed the lattice contribution (e.g., Hofmeister et al. 2007, 2009). Ballistic (direct or boundary-to-boundary) radiative transfer, which goes roughly as T³, is not a material property and is due to light passing essentially unattenuated through the sample. This ballistic radiative transfer does not occur in geological settings, but over the small length-scales encountered in the laboratory it occurs at all temperatures, even cryogenic (e.g., Hofmeister 2010; Hofmeister and Whittington 2012).

Thermal diffusivity (D) can be measured a few hundred degrees Kelvin above the glass transition using laser-flash analysis (LFA), thereby providing information on the liquid state. The contact-free LFA technique (Parker et al. 1961) lacks systematic errors associated with conventional methods, such as thermal losses at interfaces of ~10% per contact. Furthermore, ballistic radiative transfer gains are removed after Degiovanni et al. (1994) and Mehling et al. (1998), making the LFA technique essential for measuring thermally insulating, but highly transparent, glasses and melts with a high degree of accuracy (~±2%). Combining our data with heat capacity (C_p) and density (ρ) data, which are generally available or can be estimated with reasonable accuracy (e.g., Richet 1987; Lange 1997), therefore constrains thermal conductivity (k) associated with vibrational modes, needed for thermal models of igneous processes,

\[ k_{lat} = \rho C_p D. \]  

(1)

Previously, we focused on simple glass compositions corresponding to the stoichiometry of crustal minerals, quartz (SiO₂), alkali feldspar (XAlSiO₄, where X = Li, K, Na), anorthite (CaAl₂Si₂O₈), and clinopyroxene (XYSi₂O₆, where XY = CaMg, LiAl, NaAl) (Pertermann et al. 2008; Hofmeister et al. 2009; Hofmeister and Whittington 2012). Studying natural rhyolites...