A high-temperature Brillouin scattering study on four compositions of haplogranitic glasses and melts: High-frequency elastic behavior through the glass transition

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ABSTRACT

The sound velocities (νp, vS, vs) and refractive index n of four haplogranitic glasses and melts have been measured as a function of temperature by Brillouin scattering spectroscopy. The measurements were conducted at GHz frequency, through the glass transition temperature (Tg), using both platelet and backscattering geometries. The compositions of the four haplogranites are based on the addition of ~5 wt% of each of the components Li2O, F2O, Na2O, and K2O to a haplogranitic (HPG8) composition. Marked changes in slope and sign are observed in the temperature dependences of sound velocities (νp, vS, vs) as a function of composition. The glass transition temperatures Tg of the haplogranite samples are determined from distinct slope changes of sound velocities (νp, vs) vs. temperature. The lithium-enriched glass has the lowest glass transition temperature (466 °C), while the potassic glass has the highest glass transition temperature (575 °C). The unrelaxed bulk moduli vary markedly with composition below the glass transition, as do their temperature dependencies: the bulk moduli of the F- and Na-rich glasses have positive shifts with temperature. For comparison, the shear moduli have relatively similar temperature dependences below Tg for different alkali contents. At temperatures above the glass transition, the temperature derivatives of the bulk moduli, which for these frequencies reflect the vibrational compressibilities of the liquids, shift to more negative values. However, the compositional range over which the bulk moduli undergo positive or small negative temperature shifts of the vibrational compressibility appears to extend to NBO/T ratios near 0.3–0.4, or spanning most haplogranite compositions.

Keywords: High temperature, elasticity, Brillouin scattering, haplogranites

INTRODUCTION

Properties such as density, compressibility, and viscosity of silicate melts are important in understanding the thermodynamic and fluid dynamic properties of magmatic systems in the Earth’s interior. For example, knowledge of the compressibility of silicate melts at ambient pressures has aided in the construction of accurate pressure–volume–temperature equations of state and modeling of thermodynamic properties of magmatic silicate liquids at high pressure (Lange and Carmichael 1990; Ghiors et al. 2002). Ultrasonic studies have been carried out to determine the sound velocities and compressibility of binary, ternary, and more complex multi-component silicate melts at ambient pressure (Manghnani et al. 1986; Rivers and Carmichael 1987; Kress et al. 1988; Secco et al. 1999b, 1999a; Webb and Dingwell 1994; Webb and Courtial 1996), which show complex temperature and composition dependences of density and compressibility. Moreover, extensive studies have been carried out on the ultrasonic and Brillouin scattering behavior of end-member SiO2-glass and liquid: these studies have yielded major insight into the relaxational and elastic behavior of pure silica glasses and liquids (Polian et al. 2002; LeParc et al. 2006; Ruffle et al. 2010; Yokoyama et al. 2010). In contrast to these studies on a broad suite of silicate melts and glasses, the elastic behavior of haplogranites as a function of temperature, composition, and frequency is not well known. This is despite the fact that these are perhaps the best-investigated silicate melt compositions of geological relevance [e.g., viscosity and viscoelasticity (Dingwell et al. 1992, 1993a, 1993b, 1996, 2000; Hess et al. 1995, 2001), density (Knoche et al. 1992, 1995; Dingwell et al. 1993a), heat capacity (Toplis et al. 2001), surface tension (Bagdassarov et al. 2000), and diffusivities (Chakraborty et al. 1993; Mungall and Dingwell 1997)].

Haplogranites are important model systems for describing the physical state of magmas involved in the petrogenesis of granitic plutons as well as the physics of silicic volcanic eruptions. These melts are also particularly silica-rich. The goal of this study is to characterize the Brillouin spectroscopic properties of a suite of haplogranitic glasses at temperatures extending well into the supercooled liquid regime. Such high-frequency information (if unrelaxed) constrains the vibrational contribution to the compressibility of liquids; static conditions reflect the total compressibility, which is a sum of the configurational and vibrational portions of the compressibility. Thus, the regimes in which we probe these glasses and melts involve both unrelaxed measurements of the glass (a fixed configuration) at ambient and high temperatures, and the relaxed structural configuration of