LETTERS

Thermal expansion of silicate liquids: Direct determination using container-based dilatometry

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ABSTRACT

The expansivity of a silicate melt in the relaxed liquid state was obtained by direct dilatometric measurement for the first time. During the measurement, the liquid sample resides within a metal container composed of a hollow cylinder and two solid cylindrical end pieces. The sample is loaded at room temperature as a precisely machined glass cylinder. Heating the assembly beyond the glass transition results in an intimate space-filling seal of the liquid to the container surfaces. At temperatures above the glass transition, the linear expansion of the combined assembly is measured using a push rod resting on the upper end piece. The change in length of the assembly contains contributions due to the liquid volume expansion, as well as two correction terms, one each for the expansion of the enclosing hollow cylinder and the end pieces. The precision of expansivity data so obtained is ±3.5%.

The method was tested, using NIST SRM 710 standard soda-lime glass against the previously used method of combined dilatometry and calorimetry. Comparison of these two methods reveals a good agreement for the liquid expansivity. The agreement of the new and old methods supports the assumption built into the latter that volume and enthalpy relaxation follow identical kinetics in these silicate melts. The new method makes no assumptions regarding the relaxation processes of enthalpy and volume relaxation and opens up many further experimental possibilities including applications at higher pressures.

INTRODUCTION

The density of silicate liquids is a fundamental input into the derivation of buoyancy forces associated with melt transport in magmatic processes (Lange and Carmichael 1990). As such, the derivation of an adequate equation of state for the PVT properties of liquid silicates is an important experimental goal of petrology, geophysics, and geochemistry (Bottinga and Weill 1970; Bottinga et al. 1982). Considerable effort has therefore been invested in determining melt density as a function of composition, temperature, and pressure in the past decade. Several of these efforts have relied on traditional and widely accepted methods such as high-temperature immersion methods (e.g., Lange and Carmichael 1987; Dingwell and Brearley, 1988; Dingwell et al. 1988; Dingwell 1990) and both high-temperature and high-pressure Stokesian falling sphere techniques (Scarfe et al. 1987; Taniguchi 1989). Those methods are well suited to low viscosity melts (<10¹ Pa s and <10⁵ Pa s, respectively). This, however, restricts their range of applicability to relatively basic melt compositions at relatively high temperatures. Recent development of dilatometric methods for the determination of density and thermal expansion of relaxed silicate liquids (Webb et al. 1992) is highly complementary to the above traditional methods and was used, for example to first obtain the temperature dependence of the expansivity of geologically relevant silicate melt compositions (Knoche et al. 1992a, 1992b, 1992c, 1994, 1995; Dingwell et al. 1993a, 1993b). The derivation of liquid expansivity from dilatometry is based on the testable assumption that the kinetic parameters controlling the relaxation of enthalpy and volume in the glass transition are equivalent and transferable. Although evidence was presented to support this assumption, its validity has been questioned. Several dilatometric investigations of silicate melts report relaxed density at or near the glass transition but did not include direct estimates of the liquid expansivity (Lange 1997a, 1997b; Ochs and Lange 1997). As a result, the methods used to obtain expansivity for silicate melts just above the glass transition and the data themselves are in dispute. Direct determination of liquid expansivity data just above the glass transition could potentially resolve this controversy. Unfortunately, push rod dilatometric systems operating on unconstrained cylinders of glass cannot yield such data because uncontrolled longitudinal sample deformation sets in at the glass transition. Sample containment however provides a practical solution to the exact measurement of total volume expansion above the glass transition, and in fact a “thermometer” style geometry of a semi-contained sample was employed by Fiske and Stebbins (1994) recently for liquid expansivity. For the first time ideas from Gottsmann et al. (1998) are demonstrated by direct determinations of the expansivity of a silicate liquid above the glass transition using container-based linear dilatometric techniques. For the standard glass investigated, comparison with our earlier dilatometric/calorimetric techniques, reveals good agreement for the derived values of liquid expansivity.

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