

Temperature-dependent thermal expansivities of multicomponent natural melts between 993 and 1803 K

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Abstract

The thermal expansivities of three multicomponent glasses and liquids have been obtained over a large temperature interval (298–1803 K) which combine the results of low and high temperature measurements. The sample compositions investigated were derived from three natural lavas; Vesuvius 1631 eruption, Etna 1992 eruption and an Oligocene–Miocene lava flow from Slapany in the Bohemian massif. The original rocks are tephriphonolite, trachybasalt and basanite, respectively. The density values of the glassy samples were derived from dilatometric measurements of each sample after cooling at 5 K min^{-1} at 298 K, followed by measurements of the glass thermal expansion coefficient from 298 K to the samples' respective glass transition interval. Supercooled liquid volumes and thermal molar expansivities were determined by combining scanning calorimetric and dilatometric measurements, assuming that the kinetics of enthalpy and shear relaxation are equivalent [Webb, S.L., 1992. Shear, volume, enthalpy and structural relaxation in silicate melts. *Chem. Geol.* 96, 449–457.]. Thermal molar expansivity at supercooled liquid temperature varies from $16.86 \pm 0.48 \times 10^{-4} \text{ cm}^3 \text{ mol}^{-1} \text{ K}^{-1}$ for basalt/basanite, to $18.99 \pm 0.48 \times 10^{-4} \text{ cm}^3 \text{ mol}^{-1} \text{ K}^{-1}$ for trachybasalt, and $20.98 \pm 0.62 \times 10^{-4} \text{ cm}^3 \text{ mol}^{-1} \text{ K}^{-1}$ for tephriphonolite. High temperature densities were measured using Pt double bob Archimedean densitometry. Across the super-liquidus temperature interval investigated, the densities range from 2.655 ± 0.002 to $2.708 \pm 0.012 \text{ g cm}^{-3}$ for basalt–basanite, from 2.578 ± 0.003 to $2.601 \pm 0.002 \text{ g cm}^{-3}$ for trachybasalt and 2.458 ± 0.006 from $2.467 \pm 0.002 \text{ g cm}^{-3}$ to tephriphonolite. In addition, the oxidation state of iron was analyzed using a wet chemistry method. The measured high temperature densities have been compared with predicted densities across the same temperature interval calculated using the multicomponent density models of Lange and Carmichael [Lange, R.A., Carmichael, I.S.E., 1987. Densities of Na_2O – K_2O – CaO – MgO – FeO – Fe_2O_3 – Al_2O_3 – TiO_2 – SiO_2 liquids: new measurements and derived partial molar properties. *Geochim. Cosmochim. Acta* 51, 2931–2946.] and Lange [Lange, R.A., 1997. A revised model for the density and thermal expansivity of K_2O – Na_2O – CaO – MgO – Al_2O_3 – SiO_2 liquids from 700 to 1900 K: extension to crustal magmatic temperatures. *Contrib. Mineral. Petrol.* 130, 1–11].

The resulting data for volumes near glass transition temperature (993–1010 K) and at super-liquidus temperature (1512–1803 K) are combined to yield temperature-dependent thermal expansivities over the entire supercooled and stable liquid range. These results confirm the observation of Knoche et al. [Knoche, R., Dingwell, D.B., Webb, S.L., 1992a. Non-linear temperature dependence of liquid volumes in the system albite–anorthite–diopside. *Contrib. Mineral. Petrol.* 111, 61–73], Knoche et al. [Knoche, R., Dingwell, D.B., Wegg, S.L., 1992b. Temperature-dependent thermal expansivities of silicate melts: the system anorthite–diopside. *Geochim. Cosmochim. Acta* 56, 689–699], Toplis and Richet [Toplis, M.J., Richet, P., 2000. Equilibrium density and expansivity of silicate melts in the glass transition range. *Contrib. Mineral. Petrol.* 139, 672–683], Gottsmann and Dingwell [Gottsmann, J., Dingwell, D.B., 2002. Thermal expansivities of supercooled haplobasaltic liquids. *Geochim.*

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Cosmochim. Acta 66 (12) 2231–2238] of the temperature dependence of thermal expansivity. In the systems investigated here, the temperature dependence of thermal expansivity increases from basalt/basanite to tephriphonolite compositions.

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1. Introduction

Information on the density of silicate liquids is a fundamental prerequisite in order to derive the buoyancy forces associated with melt transport in magmatic processes. The thermal expansivity of silicate liquids is a crucial parameter in the calculation of melt densities over the wide range of temperatures relevant for magmatic processes (Bottinga et al., 1983). Thus, accurate data on the thermal expansivity of magmatic melts should play an important role in the construction of PVT equations of state. Expansivity data are also required as thermodynamic input for the calculation of physical properties, such as melt compressibilities from fusion curves of minerals (Bottinga, 1985; Herzberg, 1987). Such data are also necessary for the reduction of adiabatic wave velocity data to isothermal conditions (Rivers and Carmichael, 1987). The uncertainties associated with the thermal expansion of silicate liquids have been emphasized in numerous studies (e.g., Bottinga, 1985; Herzberg, 1987; Lange and Carmichael, 1987; Knoche et al., 1992a; Webb, 1992; Knoche et al., 1994; Lange, 1996; Lange, 1997; Gottsmann et al., 1999; Gottsmann and Dingwell, 2000, 2002; Toplis and Richet, 2000; Bouhifd et al., 2001; Liu and Lange, 2001; Tangeman and Lange, 2001).

Simple dilatometric measurements of glassy expansivity can be performed directly on a free standing cylindrical glass sample. However, the viscous deformation obstructs the direct measurement of relaxed supercooled liquid thermal expansivity. In order to use dilatometric data to predict the supercooled liquid expansivities of silicate melts, a method for removing this deformation from the dilatometric trace, is required.

The first method, which excludes the gravitational viscous deformation effect, was introduced by Webb et al. (1992). In this method, the comparison of the behaviour of heat capacity and thermal molar expansivity in the glass transition region permits the dilatometric data to be reconstructed, which provides the required supercooled liquid volume and expansivity data. These supercooled temperature data can then be used in combination with high-temperature data to provide a more precise determination of thermal molar expansivity in silicate melts. The advantage of this

method lies in the fact that the temperature difference between the accessible temperature regions of the double-bob Archimedean buoyancy and dilatometric methods is commonly more than 1000 K, a range far wider than that accessible by methods currently used to determine liquid density. The extraction of low-temperature liquid molar expansivities from dilatometry/calorimetry is based on an assumed equivalence of the relaxation of volume and enthalpy at the glass transition.

However, the validity of the extraction procedure has not received general acceptance, given that volume and enthalpy relaxation are not necessarily equivalent (Moynihan et al., 1976). The existence of this problem led Lange (1996, 1997) to introduce an alternative method. In this method the volume of the sample has been determined at the limiting fictive temperature and combined with measurements made on the same material at superliquidus temperature. The calculation of thermal molar expansivity and molar volume in this way is critically dependent on precise determination of the limiting fictive temperature of the glass. This problem has been discussed in detail by Gottsmann and Dingwell (2000).

Gottsmann et al. (1999) developed and tested another method, where they directly observe the thermal expansivity of a silicate melt in the relaxed liquid state. The principle of this technique is that the sample is placed within a metal container composed of a hollow cylinder and two sliding cylindrical end pieces. Expansion of the volume of liquid, the hollow cylinder and the end pieces contribute to the overall change in length of the assembly. Two correction factors can eliminate the effect of the last two, allowing the liquid expansion to be calculated. Reported precision of this method is about 3.5%.

More recently Toplis and Richet (2000) used a dilatometry technique, which was described in details by Sipp and Richet (2002). In their study, three glasses in the $\text{CaAl}_2\text{Si}_2\text{O}_8\text{--CaMgSi}_2\text{O}_6$ (anorthite–diopside) system were not initially heated through the glass transition but the samples were annealed isothermally until relaxation occurs with time at constant temperature. The annealing temperatures were chosen with respect to the viscosity where the samples are rigid

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