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The partial molar volume, thermal expansivity, and compressibility of H$_2$O in NaAlSi$_3$O$_8$ liquid: new measurements and an internally consistent model

Abstract The molar volumes of 19 hydrous albitic liquids (1.9 to 6.1 wt% H$_2$O^{total}) were determined at one bar and 505–765 K. These volume data were derived from density measurements on hydrous glasses at 298 K, followed by measurements of the thermal expansion of each glass from 298 K to its respective glass transition temperature. The technique exploits the fact that the volume of a glass is equal to that of the corresponding liquid at the limiting fictive temperature (T_f'), and that T_f' can be approximated as the temperature near the onset of the rapid increase in thermal expansion that occurs in the glass transition interval. The volume data of this study were combined with available volume data for anhydrous, Na$_2$O-Al$_2$O$_3$-SiO$_2$ liquids to derive the partial molar volume ($\pm 1\sigma$) of the H$_2$O component ($\overline{V}_{H_2O^{total}} = 15.22 \pm 0.17$ cm^3/mol) in an albitic melt at ~ 565 K and one bar. To extend the determination of $\overline{V}_{H_2O^{total}}$ to higher temperatures and pressures, the molar volumes of the hydrous albitic liquids determined in this study were combined with those measured by previous authors at 1023–1223 K and 480–840 MPa, leading to the following fitted values ($\pm 1\sigma$) at 1673 K and one bar: $\overline{V}_{H_2O^{total}} = 27.75 \, (\pm 0.45)$cm^3/mol, d$\overline{V}_{H_2O^{total}}/dT = 10.86$ (± 0.46) $\times 10^{-3}$ cm^3/mol-K, and d$\overline{V}_{H_2O^{total}}/dP = -3.82$ (± 0.36) $\times 10^{-4}$ cm^3/mol-bar. The measured molar volumes of this study and those of previous authors can be recovered with a standard deviation of 0.5%, which is within the respective experimental errors. There is a significant difference between the values for $\overline{V}_{H_2O^{total}}$ derived in this study as a function of temperature and pressure and those obtained from an existing polynomial, primarily caused by the previous absence of accurate density measurements on anhydrous silicate liquids. The coefficients of thermal expansion ($\alpha = 4.72 \times 10^{-4}$/K) and isothermal compressibility ($\beta_T = 1.66 \times 10^{-5}$/bar) for the H$_2$O component at 1273 K and 100 MPa, indicate that H$_2$O^{total} is the single most expansive and compressible component in silicate liquids. For example, at 1473 K and 70 MPa (conditions of a mid-ocean ridge crustal magma chamber), the presence of just 0.4 wt% H$_2$O will decrease the density of a basaltic liquid by more than one percent. An equivalent decrease in melt density could be achieved by increasing the temperature by 175 degrees or the decreasing pressure by 230 MPa. Therefore, even minor quantities of dissolved water will have a marked effect on the dynamic properties of silicate liquids in the crustal environment.

Introduction

The density of multicomponent silicate liquids is of fundamental importance to the dynamic behavior and chemical differentiation of magmas. The dramatic role that dissolved water may play in modifying the density of silicate melts has long been recognized (Orlova 1962; Burnham and Davis 1971). Measurements of the water concentration in glass inclusions trapped in phenocryst phases have established that substantial amounts of water (up to 6 wt%) commonly occur in both silicic and mafic liquids in the arc environment (e.g., Anderson et al. 1989; Hervig et al. 1989; Geschwind and Rutherford 1992; Sisson and Layne 1993). In addition, the low molecular weight of H$_2$O compared to the other oxide components in a silicate liquid converts even a low weight percent of H$_2$O into a much larger mole percent. Therefore, there is little doubt that density models must include H$_2$O before they can be applied to the entire range of igneous liquids generated in the upper mantle and crust.

Currently, the only direct density measurements available for hydrous silicate liquids are those of Burnham and Davis (1971). Their specific volume data ($\pm 0.4\%$) on hydrous albitic liquids were measured over the temperature and pressure range of 1023 to 1223 K

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and 480 to 840 MPa with a volumometer housed within an internally heated pressure vessel (also used to measure the specific volume of pure H_2O by Burnham et al. 1969). Burnham and Davis (1971) used their volumetric data on hydrous albitic liquids to derive the partial molar volumes of H_2O and $NaAlSi_3O_8$ as a function of temperature and pressure. Their polynomial for $\bar{V}_{H_2O^{total}}$ is the only relation available which attempts to describe the variation of $\bar{V}_{H_2O^{total}}$ with temperature and pressure.

Because the molar volume of natural silicate liquids can be described as a linear function of composition, temperature, and pressure (Lange and Carmichael 1987), this model equation is readily extended to include the volatile components. However, it is inappropriate to use the polynomial for $\bar{V}_{H_2O^{total}}$ of Burnham and Davis (1971) with the general linear model of Lange and Carmichael (1990) since they are based on different calibrations; such a combined model overestimates the measured densities of Burnham and Davis (1971) by 2–7%, an order of magnitude outside the experimental error. The only other estimates of the partial molar volume of water are for $\bar{V}_{H_2O^{molecular}}$, which are based on solubility studies (e.g., Silver and Stolper 1989). However, these estimates are independent of temperature and pressure, and are plagued with problems of quenching the high temperature H_2O speciation.

The goal of this study was to provide volume measurements of hydrous albitic liquids that could be used to derive the partial molar volume, thermal expansivity, and compressibility of the H_2O^{total} component in silicate liquids. A second aim was to evaluate the effect of H_2O speciation (OH^- and $H_2O^{molecular}$) on $\bar{V}_{H_2O^{total}}$. The results of this study indicate that the speciation of water has no effect on derived values of $\bar{V}_{H_2O^{total}}$ within experimental resolution. Moreover, our measurements are completely consistent with the data of Burnham and Davis (1971) and, when combined with recently published measurements of density, thermal expansivity and compressibility of anhydrous silicate liquids, allow derivation of accurate estimates of $d\bar{V}_{H_2O^{total}}$, $d\bar{V}_{H_2O^{total}}/dT$, and $d\bar{V}_{H_2O^{total}}/dP$.

General experimental premise

The measurements of the molar volume of hydrous albitic liquids at one bar (0.1 MPa) were made by taking advantage of the thermodynamic and kinetic properties of glasses. If a liquid is cooled rapidly enough, it may quench to form a glass. The structure of the liquid that is preserved in the glass is the equilibrium structure of the liquid at a temperature called the limiting fictive temperature (T_f'). Unlike the first order, crystal-liquid phase transition, there is no discontinuous change in molar volume across the glass-liquid transition, at T_f'. Therefore, the molar volume of the glass is equal to the molar volume of the liquid at T_f' (Fig. 1). In contrast to the limiting fictive temperature, the glass transition temperature (T_g) is an arbitrarily determined temperature
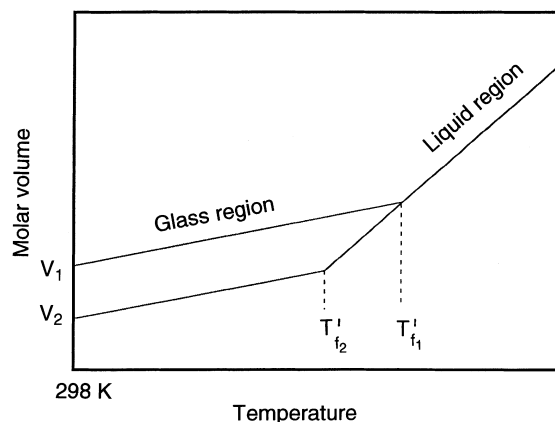

Fig. 1 A schematic diagram of the molar volume of a sample as it is cooled from a liquid to a glass. At the limiting fictive temperature $T_f'(\approx T_g)$, the volume of the glass is equal to that of the corresponding liquid. Two cooling rates are shown; T_{f_1}' is the limiting fictive temperature of the faster cooling rate, and V_1 is the 298 K molar volume of the glass produced from the faster cooling rate. Regardless of cooling rate, if the molar volume of the glass at 298 K and the thermal expansion of the glass up to T_f' are measured, the molar volume determined at T_f' is that of the corresponding liquid

within the glass transition region. In this study, the glass transition temperature (T_g) was chosen to be the temperature near the onset of the rapid rise in thermal expansion (Fig. 2) which has been shown to approximate T_f' within experimental error (Moynihan 1995; Lange 1996). In this study the density and thermal expansion of several albitic glasses with variable amounts of dissolved water were measured up to their respective T_g allowing the density (and thus the molar volume) to be obtained at one bar and $T_g(\approx T_f')$.

Although the above experimental procedure works well for anhydrous glasses (e.g. Lange 1996, 1997), there are difficulties that must be overcome when it is applied

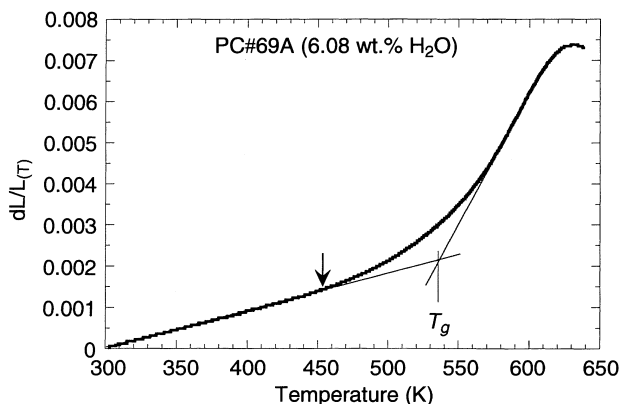**Fig. 2** A typical plot obtained with the Perkin-Elmer TMA-7 dilatometer at a heating rate of 10 K/min. The slope of the curve between 298 K and where the slope diverges from a straight line (at the *arrow*) was used to calculate the coefficient of thermal expansion of the glass. The T_g was determined as the approximate onset of the rapid change in slope of the thermal expansion curve, well before the peak

to hydrous glasses. First, hydrous glasses must be synthesized at pressures greater than one bar in order to dissolve significant quantities of H_2O^{total}. Because glasses synthesized at higher pressure are denser than those synthesized at one bar, a pressure correction is required to obtain the densities of hydrous glasses at one bar. Second, an evaluation of the effect of densification on the thermal expansion of a glass is necessary. Third, a determination must be made of whether or not a densified glass relaxes during the heat treatment of the thermal expansion measurement from 298 K to T_g. Finally, an assessment of any water loss from the hydrous glasses during the thermal expansion measurement is also required.

Experimental technique

Synthesis of starting material

The starting material used in this study was an albitic glass prepared with appropriate proportions of reagent grade Na_2CO_3, Al_2O_3, and silicic acid powders. The powders were mechanically mixed, the mixture decarbonated at 1073 K and fused at 1873 K. The melt was quenched to a glass, then powdered and re-fused twice to ensure homogeneity. A chemical analysis of the glass was provided by I.S.E. Carmichael (Table 1). Once the glass was characterized, it was powdered and kept at 120 °C in a drying oven until needed.

Synthesis and characterization of the hydrous glasses

Each experimental capsule was made of 5 mm O.D. platinum tubing, with a tri-crimp, welded seal at one end so that the capsule would retain a cylindrical shape during compression. The capsule was loaded with half of the required amount of glass powder, followed by the required amount of doubly distilled water measured with a microsyringe, followed by the second half of the powder. The open end of the capsule was then crimped and sealed by arc welding. The charges were run at either 600 or 1000 MPa in a 1.905 cm (3/4″) piston cylinder apparatus, using a NaCl and MgO pressure cell and the cold piston-in technique. No friction correction was used in the pressure determination of each run, as only reproducibility between runs at the same pressure was necessary for this study. Experimental conditions of synthesis are given in Table 2. All samples were held at pressure and temperature for

Table 1 Starting glass composition

Oxide	Analysed composition		Ideal albite (mol%)
	Measured (wt%)[a]	Normalized (mol%)	
SiO_2	68.50	74.99	75.00
Al_2O_3	19.42	12.53	12.50
Na_2O	11.76	12.48	12.50
$H_2O^{(-)}$	0.22	–	–
Total	99.68		

[a] One sigma uncertainties are 0.04 wt% for SiO_2 and Al_2O_3 and 0.02 wt% for Na_2O. $H_2O^{(-)}$ is absorbed water. Uncertainties are independent of composition, and are based on replicate analyses of a basalt standard (I.S.E. Carmichael, 1985, unpublished data)

at least 48 h to obtain a homogeneous water content. The temperature of the runs ranged from 1573 to 1673 K, in all cases well above the liquidus of hydrous albite at the run pressure. During the runs the pressure varied by no more than 40 MPa. An isobaric quench was achieved by turning off the power to the graphite heater while maintaining the run pressure through active pumping. The quenched glass was invariably found to be cracked, although most runs produced glass pieces that were large enough to use for the density and thermal expansion measurements.

Samples were inspected after synthesis for bubbles, cloudiness, and cracks under a microscope. Glass pieces that were clear and free of imperfections had top and bottom faces ground parallel (to within 4 μm). The average thickness of the glass pieces was 1.65 mm. Each glass was measured for H_2O^{total} content with a Fourier-transform infrared (FTIR) spectrometer using a 531 μm spot size centered on the glass piece. The absorption coefficients for the 4485 and 5218 cm^{-1} bands in hydrous albitic glass determined by Silver and Stolper (1989) were used to calculate molecular H_2O and OH^- species concentrations in the glass, which in turn were used to determine the H_2O^{total} concentration. Variations in H_2O^{total} across individual pieces of glass were within 0.01 wt%. The uncertainty in the total water contents is ±5% relative of the measured values, based on all measurements over the lifetime of the machine.

Density and thermal expansion measurements

The room temperature (298 K) density of each sample was measured using the Archimedean method. Each piece of glass was weighed 12 to 24 times on a microbalance, then weighed 24 to 30 times below the microbalance while immersed in toluene. The standard deviation of each set of density measurements on one sample ranged from 0.05 to 0.2%. The masses of the samples ranged from 10 to 90 mg, with a median of 20 mg. The density measurements were replicated on separate occasions and found to be within 0.08 to 0.2% of each other.

The pressure correction used to obtain the values for the density of the hydrous glasses at one bar was determined by comparing the difference in densities of a bubble-free, dry albitic glass synthesized at one bar to that of dry albitic glasses synthesized at 600 MPa (PC#64A) and 1000 MPa (PC#60, PC#75A) (Table 2). The pressure correction to the density of the hydrous glasses is 1.18 (±0.25)% for 600 MPa, and 2.24 (±0.25)% for 1000 MPa.

The thermal expansion of each glass was measured with a Perkin-Elmer TMA-7 dilatometer using a scan rate of 10 K per minute from 298 K up to and beyond the T_g for each glass. A typical dilatometer scan and the method of determining T_g is shown in Fig. 2. The coefficient of linear thermal expansion for each glass was determined by measuring the slope of the dilatometry data from the starting temperature to within ~100 degrees of T_g. The coefficient of volume thermal expansion ($\alpha^{glass} = 1/V_T(\partial V/\partial T)_P$) was calculated by multiplying the coefficient of linear thermal expansion by three. Because of the small sample sizes of this study, the uncertainty in the slope of the dilatometry data is 15%; this uncertainty only weakly (by 0.05%) affects the determination of V^{liq} at T_g.

After each thermal expansion measurement, the density and water content of the glass (if optically clear) were measured again to evaluate how much relaxation (or de-densification) and water loss occurred in the glass during heating. Following this, a second thermal expansion measurement was performed to check for reproducibility. In addition, a sequence of four thermal expansion measurements was performed on a single sample (PC#69C). Between each dilatometer run the density and water content of the glass were measured. The first dilatometer run proceeded up to 450 K, 60 K below T_g. The second and third were measured to 510 K, the initially observed T_g of the glass, and the fourth dilatometer run was stopped at 580 K, 70 K beyond the initially observed T_g. The purpose of the consecutive experiments was to evaluate how much relaxation occurs in the glass prior to T_g and also to determine if the thermal expansion of the glass is significantly affected by

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Table 2 Data for hydrous albitic glass

Sample name	P (MPa)	T (K)	Time (h)	H_2O^{total} (wt%)	H_2O^{molec} (wt%)	OH^- (wt%)	Density (at 298 K) (g/cm^3)	Corrected density (at 298 K) (g/cm^3)	T_g (K)	α^{glass} (10^{-5}, K^{-1})	V^{liq} meas (at T_g) (cm^3/mol)	V^{liq} calc (at T_g) (cm^3/mol)	Residual volume (calc-meas)/meas
Albite	0.1	1673	12.00	0.00 ± 0.00	0.00	0.00	2.376 ± .000	2.376	1073 ± 10	2.035	28.03 ± 0.07	27.87	−0.58%
First runs:													
PC#60	950	1623	62.50	0.15 ± 0.01	0.00	0.15	2.424 ± .001	2.370	883 ± 10	1.768	27.85 ± 0.09	27.62	−0.81%
PC#64A	600	1623	75.50	0.23 ± 0.01	0.00	0.23	2.405 ± .002	2.376	840 ± 15	1.538	27.66 ± 0.08	27.54	−0.41%
PC#75A	970	1673	50.25	0.24 ± 0.01	0.00	0.24	2.437 ± .001	2.382	855 ± 10	1.758	27.61 ± 0.08	27.55	−0.23%
PC#62A	580	1623	46.75	1.87 ± 0.09	0.57	1.30	2.388 ± .007	2.360	670 ± 15	1.932	26.66 ± 0.14	26.69	0.09%
PC#67A	940	1673	48.00	2.12 ± 0.11	0.84	1.29	2.430 ± .005	2.376	607 ± 30	2.040	26.30 ± 0.13	26.53	0.86%
PC#59A	580	1598	69.25	2.28 ± 0.11	1.00	1.28	2.398 ± .001	2.370	692 ± 30	2.366	26.34 ± 0.13	26.54	0.77%
PC#51B	550	1573	49.87	2.44 ± 0.12	0.93	1.51	2.362 ± .004	2.334	676 ± 10	2.095	26.61 ± 0.14	26.46	−0.53%
PC#56	1000	1573	54.50	3.33 ± 0.17	1.61	1.72	2.322 ± .004	2.322	604 ± 10	2.144	26.12 ± 0.17	26.06	−0.25%
PC#46	980	1573	47.50	3.55 ± 0.18	1.73	1.82	2.384 ± .003	2.331	582 ± 10	2.139	25.87 ± 0.17	25.96	0.33%
PC#65A	560	1673	50.00	4.80 ± 0.24	2.81	1.99	2.318 ± .002	2.290	614 ± 10	2.458	25.61 ± 0.21	25.54	−0.28%
PC#61	940	1563	54.25	5.38 ± 0.27	3.40	1.98	2.326 ± .001	2.274	505 ± 15	2.485	25.38 ± 0.23	25.25	−0.52%
PC#69A	950	1573	48.00	6.07 ± 0.30	4.06	2.01	2.327 ± .003	2.275	530 ± 10	2.701	25.00 ± 0.25	25.04	0.14%
PC#69B	950	1573	48.00	6.07 ± 0.30	4.06	2.01	2.323 ± .004	2.271	527 ± 15	2.515	25.03 ± 0.25	25.04	0.03%
Second runs:													
PC#60.02	950	1573	48.00	0.15 ± 0.01	0.00	0.15	2.376 ± .005	2.370	1017 ± 20	1.326	27.82 ± 0.10	27.75	−0.26%
PC#64A.02	950	1573	48.00	0.23 ± 0.01	0.00	0.23	2.364 ± .008	2.376	973 ± 20	1.597	27.73 ± 0.13	27.67	−0.21%
PC#75A.02	950	1573	48.00	0.25 ± 0.01	0.00	0.25	2.378 ± .005	2.382	987 ± 20	1.788	27.68 ± 0.11	27.67	−0.03%
PC#62A.02	950	1573	48.00	1.90 ± 0.01	0.64	1.26	2.364 ± .007	2.360	765 ± 20	1.422	26.63 ± 0.14	26.76	0.50%
PC#51B.02	950	1573	48.00	2.51 ± 0.13	1.06	1.45	2.366 ± .008	2.339	735 ± 20	1.862	26.51 ± 0.16	26.49	−0.09%
PC#56.02	950	1573	48.00	3.38 ± 0.17	1.67	1.71	2.335 ± .004	2.323	725 ± 20	2.245	26.17 ± 0.18	26.15	−0.10%
PC#46.02	950	1573	48.00	3.60 ± 0.18	1.78	1.82	2.357 ± .014	2.331	667 ± 20	1.680	25.84 ± 0.23	26.01	0.66%
PC#61.02	950	1573	48.00	5.29 ± 0.26	3.32	1.97	2.320 ± .005	2.274	577 ± 20	2.305	25.46 ± 0.23	25.33	−0.49%
Consecutive runs with the same sample:													
PC#69C.01	950	1573	48.00	6.08 ± 0.30	4.07	2.01	2.322 ± .003	2.270	510 ± 30	2.085	25.00 ± 0.25	25.02	0.07%
PC#69C.02	950	1573	48.00	6.08 ± 0.30	4.07	2.01	2.320 ± .003	2.270	510 ± 30	2.668	25.03 ± 0.25	25.02	−0.05%
PC#69C.03	950	1573	48.00	6.08 ± 0.30	4.07	2.01	2.314 ± .004	2.270	537 ± 10	2.514	25.04 ± 0.25	25.04	0.00%
PC#69C.04	950	1573	48.00	6.08 ± 0.30	4.07	2.01	2.313 ± .004	2.270	537 ± 10	2.537	25.04 ± 0.25	25.04	0.00%

the degree of relaxation of the glass. The glass densities measured after a dilatometer run were corrected for densification in a similar manner as the glass densities before the dilatometer run; the pressure correction was adjusted to account for any relaxation the glass had undergone during the previous dilatometry measurement.

Results

The density and thermal expansion of hydrous albitic glasses

The densities of all hydrous albitic glasses at 298 K are reported in Table 2. The experiments of PC#69C demonstrated that dilatometry runs that did not heat the glass above T_g did not change the glass density within error. Moreover, FTIR spectroscopic data demonstrate that the glasses did not lose any water during these dilatometry runs (e.g., compare PC#61 to PC#61.02 in Table 2). Therefore, no relaxation (density decrease) or water loss occurred in the hydrous densified glasses during the thermal expansion measurement from 298 K to the T_g of the glass. Some relaxation did occur, however, when the dilatometer runs reached temperatures that exceeded T_g. However, each glass had the same α^{glass} within error whether it was densified or partially relaxed (Table 2, Fig. 3), indicating that the degree of densification of the glass did not affect the observed coefficient of thermal expansion within the resolution of our experiments.

The molar volume of each glass at 298 K was calculated from the density and composition of the glass, using the following equation:

$$V_{298K}^{glass} = \frac{\Sigma X_i (M.W.)_i}{\rho_{298K}^{glass}} \tag{1}$$
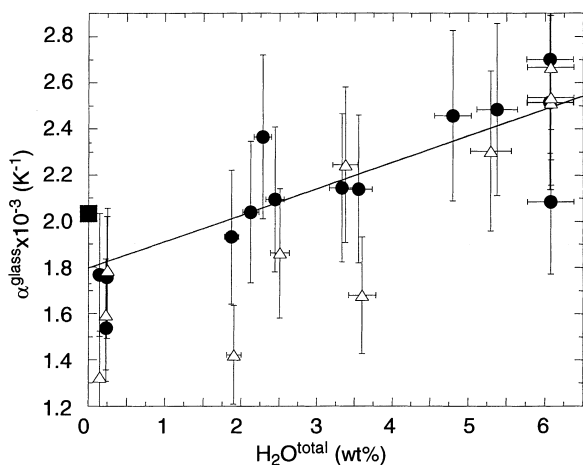

Fig. 3 A plot of α^{glass} for all samples. *Filled circles* first dilatometer runs, *open triangles* replicate runs, *large filled square* coefficient of thermal expansion for an anhydrous albitic glass synthesized at one bar. Error bars only shown for the first runs; uncertainty 15% in the α_{glass} measurement and 5% in the water content measurement. The linear equation of the fit to all the data is: $\alpha^{glass} = [0.135 \times (wt\% H_2O) + 1.82] \times 10^{-3}$ (K^{-1})

where X_i and $(M.W.)_i$ are the mole fraction and molecular weight of each oxide component, respectively.

Determination of liquid molar volumes

The thermal expansivity data were combined with the molar volume measurements of the glasses at 298 K (corrected for densification) to calculate the molar volume of the liquids at their respective glass transition temperatures (Table 2) using the following equation:

$$V_{T_g}^{liq} = V_{298}^{glass} \times \exp[\alpha^{glass} \times (T_g - 298)] \tag{2}$$

where $V_{T_g}^{liq}$ is the molar volume of the glass or liquid at $T_g (\approx T_f')$, and V_{298}^{glass} is the molar volume at 298 K, corrected for densification. The inclusion of replicate density and thermal expansion experiments led to a final data set of 19 molar volumes of hydrous albite liquids at temperatures from 505 to 765 K. The propagated uncertainties (one σ) in the liquid molar volumes range from 0.3 to 1.0%. The two largest sources of error in $V_{T_g}^{liq}$ are derived from the pressure-corrected 298 K density measurements ($\leq 0.3\%$) and the FTIR water analysis (0.1 to 0.6%), whereas the smallest sources of error, which in combination contribute < 0.1% uncertainty to the $V_{T_g}^{liq}$, are caused by the errors in the wet chemical compositional analysis of the albitic glass, the uncertainties in the value of $T_g (\pm 10$–30 K), and the thermal expansion coefficient of the glass ($\pm 15\%$). The relatively large uncertainty in α_{glass} (because of the small sample size) contributes little error to $V_{T_g}^{liq}$ because the projection from 298 K to T_g in the water-rich glasses is only ~ 300 degrees.

Calibration of a linear molar volume equation

The 19 molar volumes of hydrous albitic liquids (between 505 and 765 K) were combined with 56 molar volumes of anhydrous Na_2O-Al_2O_3-SiO_2 liquids (between 701 and 1835 K: Bockris et al. 1956; Stein et al. 1986; Lange 1996) to calibrate the following model equation:

$$V_{T,X_i}^{liq} = \Sigma X_i \left[V_{i,T_{ref}} + \frac{d\bar{V}_i}{dT}(T - T_{ref}) \right] \tag{3}$$

where X_i is the mole fraction of each oxide component i, T is the temperature in Kelvins, and $d\bar{V}_i/dT$ is the partial molar thermal expansivity of each oxide component i. A reference temperature (T_{ref}) of 1023 K was selected for this calibration because it is approximately midway between the temperature range of the hydrous and anhydrous data sets. The partial molar volumes of SiO_2, Al_2O_3, Na_2O, and H_2O^{total}, as well as $d\bar{V}_{Na_2O}/dT$ were determined with a multiple linear regression of Eq. 3, and are given in Table 3.

Fitted $d\bar{V}_i/dT$ values for the SiO_2, Al_2O_3, and H_2O^{total} components were indistinguishable from zero within error and so were dropped from the regression.

Table 3 Derived volume parameters for Na_2O-Al_2O_3-SiO_2-H_2O liquids at 1 bar

Oxide component	$\overline{V}^{liq}_{(T,P,X_i)} = \sum X_i \left[\overline{V}_i + \frac{d\overline{V}_i}{dT}(T - T_{ref}) \right]$	
	$\overline{V}_i$ (cm³/mol)	$\left(\frac{d\overline{V}_i}{dT}\right)$ (10⁻³, cm³/mol-K)
SiO_2	26.87 ± 0.05	–
Al_2O_3	37.48 ± 0.16	–
Na_2O	23.84 ± 0.09	7.69 ± 0.11
H_2O	15.22 ± 0.17	–

Although the results indicate that neither SiO_2 or Al_2O_3 contribute to the thermal expansivity of the liquids over a ∼1300 degree temperature interval (505 to 1835 K), the limited temperature range of the hydrous albitic liquid data set (<260 degrees) prevented resolution of the value for $d\overline{V}_{H_2O^{total}}/dT$. Therefore, the value for $\overline{V}_{H_2O^{total}}$ derived from the linear regression (15.22 ± 0.17 cm³mol) is only valid over the temperature interval 505 to 765 K, and is assumed to represent $\overline{V}_{H_2O^{total}}$ at a median temperature of 567 K (average T_g of glasses with ≥ 2 wt% H_2O^{total}). In order to determine the $\overline{V}_{H_2O^{total}}$ in hydrous albitic liquid at the reference temperature, 1023 K, a separate evaluation of the thermal expansivity of the H_2O^{total} melt component must be made (see discussion).

Application of the model equation (Table 3, Eq. 3) recovers the molar volumes of the hydrous albitic liquids measured in this study within ±0.5% on average. Both the calculated molar volumes and residuals are given in Table 2. There was no correlation between the concentration of molecular H_2O (or hydroxyl groups) and residual volume, which indicated that within the resolution of the experiments of this study, the value of $\overline{V}_{H_2O^{total}}$ is not sensitive to the speciation of H_2O. Nor did the pressure correction to the densities of the glasses introduce a systematic error into the derivation of $\overline{V}_{H_2O^{total}}$ even though these corrections were derived from anhydrous glasses and then applied to hydrous glasses with various amounts of dissolved water. Glasses with different amounts of dissolved water (1.9 to 6.1 wt%) and synthesized at different pressures (600 to 1000 MPa), are all fit by Eq. 3 (Table 3) equally well, with no systematic variation.

Discussion

Thermal expansivity and compressibility of the H_2O^{total} component

The value for $\overline{V}_{H_2O^{total}}$ derived above is for an albitic melt at 500 to 700 K and one bar, and as such is outside the temperature and pressure range of silicate liquids within the Earth's crust. To extend $\overline{V}_{H_2O^{total}}$ into the crustal temperature and pressure regime, the data of Burnham and Davis (1971) were used. The key to employing the specific volume data of Burnham and Davis (1971) on hydrous albitic liquids in the linear volume model is to take advantage of the precise density, thermal expansivity, and compressibility data on anhydrous Na_2O-Al_2O_3-SiO_2 liquids that have been obtained over the past 10 years (Bockris et al. 1956; Stein et al. 1986; Kress et al. 1988; Lange 1996). The combination of the anhydrous density data set with the hydrous density measurements of this study and those of Burnham and Davis (1971) into a multiple linear regression allows the derivation of an internally consistent equation for the molar volume of hydrous sodium aluminosilicate melts.

Three sets of specific volume data for hydrous albitic melts were taken from Burnham and Davis (1971), two with 8.25 wt% H_2O^{total} at 480 MPa and 840 MPa, and one with 10.9 wt% H_2O^{total} at 770 MPa. The temperature range for all three data sets was 1023 to 1208 K. Although additional density measurements on other hydrous albitic melts were reported by Burnham and Davis (1971), much of the actual data are not presented in that paper, which constrains us to use only the three data sets that were directly reported (their Fig. 5). The albite starting material used by Burnham and Davis (1971) was a natural specimen from a granitic pegmatite containing ∼8% each of the orthoclase and anorthite components. In order to derive precise partial molar properties of the H_2O^{total} component from their data, molar volume measurements of silicate liquids with MgO, CaO, and K_2O were also included (from Lange and Carmichael 1987; Lange 1997). Liquids containing TiO_2, FeO, and Fe_2O_3 were not considered since these components occur in negligible quantities in the Burnham and Davis (1971) sample.

To extract compressibility terms for the H_2O^{total} component from Burnham and Davis (1971), it was necessary to incorporate the compressibilities of all the other oxide components into the multiple linear regression. The best compressibility values for the anhydrous components are those derived from sound speed measurements (Rivers and Carmichael 1987; Kress et al. 1988; Kress and Carmichael 1991). The data of Kress and Carmichael (1991) were used to calculate molar volumes at 500 MPa for each liquid in the anhydrous, one bar data set described above. An initial regression using only the molar volumes of the anhydrous liquid data set at one bar and at 500 MPa was performed to verify that the fitted anhydrous compressibility values were comparable to those derived directly from the sound speed data. The compressibility terms are the same within the one sigma fitted error (Table 4).

An internally consistent volume equation for hydrous silicate melts

Both the 500 MPa and the one bar molar volumes from the anhydrous liquid data set, along with the one bar molar volumes of hydrous liquids from this study and the 480 to 840 MPa molar volumes of the hydrous liquid

Table 4 Comparison of the partial molar compressibilities of oxide components

	Oxide component	Dry regression[a]	K&C (1991)[b]
$\left(\dfrac{\mathrm{d}\overline{V}_i}{\mathrm{d}P}\right)_{1673\text{K}}$ $(10^{-4},\ \mathrm{cm}^3/\mathrm{mol\text{-}bar})$	SiO$_2$	-1.87 ± 0.11	-1.89 ± 0.02
	Al$_2$O$_3$	-2.36 ± 0.28	-2.26 ± 0.09
	MgO	0.26 ± 0.21	0.27 ± 0.07
	CaO	0.34 ± 0.16	0.34 ± 0.05
	Na$_2$O	-2.46 ± 0.20	-2.40 ± 0.05
	K$_2$O	-6.79 ± 0.29	-6.75 ± 0.14
	Al$_2$O$_3 \times$ Na$_2$O	11.16 ± 1.31	10.18 ± 0.50
$\dfrac{\dfrac{\mathrm{d}\overline{V}_i}{\mathrm{d}P}}{\mathrm{d}T}$ $(10^{-7},\ \mathrm{cm}^3/\mathrm{mol\text{-}bar\text{-}K})$	SiO$_2$	1.4 ± 0.2	1.3 ± 0.1
	Al$_2$O$_3$	2.7 ± 0.6	2.7 ± 0.5
	MgO	-1.5 ± 0.6	-1.3 ± 0.4
	CaO	-3.1 ± 0.4	-2.9 ± 0.3
	Na$_2$O	-6.9 ± 0.5	-6.6 ± 0.4
	K$_2$O	-14.8 ± 0.8	-14.5 ± 1.5

[a] Fitted values calibrated on volume data at 500 MPa, calculated from the model of Kress and Carmichael (1991)
[b] The values of Kress and Carmichael (1991), derived directly from the sound speed measurements

Table 5 Model equation for hydrous silicate liquids with temperatures from 600 to 1873 K and pressures up to 1000 MPa

$$\overline{V}^{\text{liq}}_{(T,P,X_i)} = \sum X_i \left\{ \overline{V}_{i,T_{\text{ref}}} + \frac{\mathrm{d}\overline{V}_i}{\mathrm{d}T}(T - T_{\text{ref}}) + \left[\left(\frac{\mathrm{d}\overline{V}_i}{\mathrm{d}P}\right)_{T_{\text{ref}}} + \frac{\frac{\mathrm{d}\overline{V}_i}{\mathrm{d}P}}{\mathrm{d}T}(T - T_{\text{ref}}) \right](P - P_{\text{ref}}) \right\}$$

Oxide component	$\overline{V}_{i,1023\text{K}}$ (cm^3/mol)	$\overline{V}_{i,1673\text{K}}$ (cm^3/mol)	$\dfrac{\mathrm{d}\overline{V}_i}{\mathrm{d}T}$ $(10^{-3},$ cm^3/mol-K)	$\left(\dfrac{\mathrm{d}\overline{V}_i}{\mathrm{d}P}\right)_{1673\text{K}}$ $(10^{-4},$ cm^3/mol-bar)	$\dfrac{\left(\frac{\mathrm{d}\overline{V}_i}{\mathrm{d}P}\right)}{\mathrm{d}T}$ $(10^{-7},$ cm^3/mol-bar-K)
SiO$_2$	26.77 ± 0.04	26.77 ± 0.04	–	-1.66 ± 0.13	0.98 ± 0.29
Al$_2$O$_3$	37.42 ± 0.11	37.42 ± 0.11	–	-2.22 ± 0.35	2.58 ± 0.69
MgO	9.70 ± 0.15	11.83 ± 0.09	3.28 ± 0.21	-0.07 ± 0.26	-0.84 ± 0.69
CaO	14.21 ± 0.10	16.64 ± 0.06	3.73 ± 0.15	0.06 ± 0.20	-2.51 ± 0.50
Na$_2$O	23.96 ± 0.09	29.05 ± 0.08	7.83 ± 0.12	-2.81 ± 0.24	-6.36 ± 0.61
K$_2$O	38.40 ± 0.15	46.25 ± 0.11	12.08 ± 0.25	-7.20 ± 0.35	-13.81 ± 0.94
Al$_2$O$_3 \times$ Na$_2$O	–	–	–	10.37 ± 1.64	–
H$_2$O	20.69 ± 0.20	27.75 ± 0.45	10.86 ± 0.46	-3.82 ± 0.36	–

volumes from Burnham and Davis (1971) were used in a multiple linear regression of Eq. 4 to determine the partial molar volume, thermal expansivity, and compressibility of the H$_2$O$^{\text{total}}$ component (Table 5). The values from Table 5 can be used in the following equation:

$$V^{\text{liq}}_{P,T,X_i} = \Sigma X_i \left\{ \overline{V}_{i,T_{\text{ref}}} + \frac{\mathrm{d}\overline{V}_i}{\mathrm{d}T}(T - T_{\text{ref}}) \right.$$
$$\left. + \left[\left(\frac{\mathrm{d}\overline{V}_i}{\mathrm{d}P}\right)_{T_{\text{ref}}} + \frac{\left(\frac{\mathrm{d}\overline{V}_i}{\mathrm{d}P}\right)}{\mathrm{d}T}(T - T_{\text{ref}}) \right](P - P_{\text{ref}}) \right\} \quad (4)$$

where $\frac{\mathrm{d}\overline{V}_i}{\mathrm{d}P}$ is the partial molar compressibility of each oxide component i, $\left(\frac{\mathrm{d}\overline{V}_i}{\mathrm{d}P}\right)/\mathrm{d}T$ is the temperature dependence of the partial molar compressibility of each oxide component i, P is the pressure in bars, P_{ref} is one bar, and all other variables are the same as Eq. 3. Two separate regressions were performed with reference temperatures of 1023 and 1673 K, respectively. The 1023 K reference temperature was chosen since it represents a midpoint for the hydrous density data, whereas the 1673 K reference temperature was chosen so that a direct

comparison could be made to the values given by Lange and Carmichael (1990, their Table 2). The incorporation of the hydrous density data did not change the fitted values derived for the anhydrous components (Tables 4 and 5). The derived values for $\overline{V}_{\text{H}_2\text{O}^{\text{total}}}$ of 27.75 (± 0.45)cm^3/mol, d$\overline{V}_{\text{H}_2\text{O}^{\text{total}}}$/d$T$ of 10.86 (± 0.46)$\times 10^{-3}$/K, and d$\overline{V}_{\text{H}_2\text{O}^{\text{total}}}$/d$P$ of -3.82 (± 0.36) $\times 10^{-4}$/bar at 1673 K are considerably more precise than any previous estimates, and as shown in the following section, are considerably more accurate.

The calibrated model equation (Eq. 4, Table 5) can recover the measured molar volumes of the hydrous albitic liquids of this study and those of Burnham and Davis (1971) within $\pm 0.5\%$ on average (Fig. 4a–c, Table 6). Given the reported uncertainty of $\pm 0.4\%$ in their specific volume measurements, combined with the uncertainty in exact water concentration in their samples (calculated from the amount of H$_2$O weighed into their charge), the recovery of the Burnham and Davis (1971) density data with the calibrated model is exceptionally good. The quality of the fit to the density data further demonstrates that the measurements reported in this study are entirely consistent with the measurements of Burnham and Davis (1971), despite the fact that com-
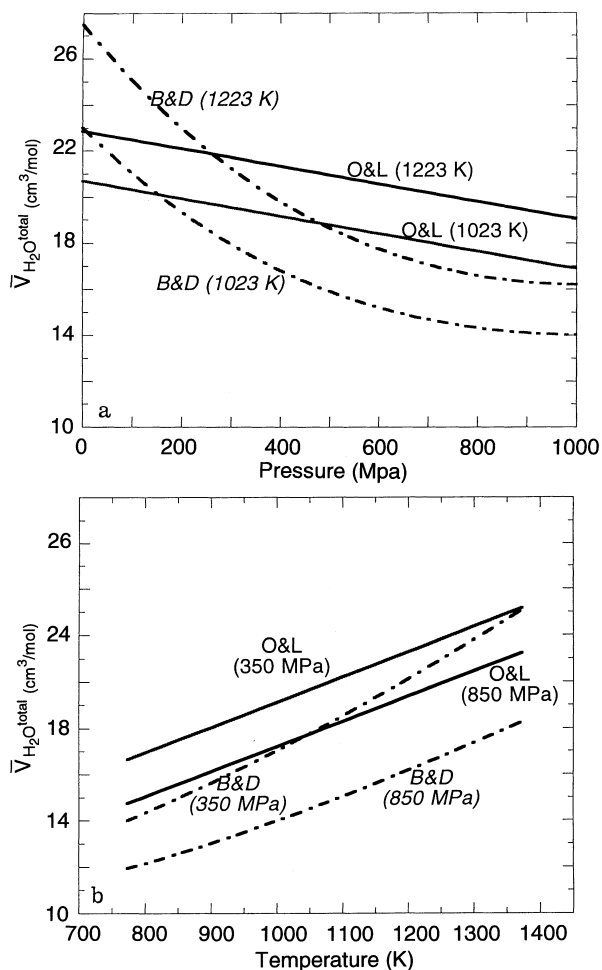
Fig. 4 a A comparison of the $\overline{V}_{H_2O^{total}}$ pressure dependence of this study, shown as a *solid line* (O&L), to that of the polynomial of Burnham and Davis (1971), shown as a *dashed line*. The $\overline{V}_{H_2O^{total}}$ for each model was calculated as a function of pressure at 1023 and 1223 K, the temperature limits of the Burnham and Davis (1971) data. The strong pressure-dependent compressibility of the Burnham and Davis (1971) polynomial is not supported by the combined data sets of hydrous and anhydrous molar volumes. **b** A similar comparison of the temperature dependence of $\overline{V}_{H_2O^{total}}$ between the model of this study and that of Burnham and Davis (1971)

pletely different techniques were employed to measure the density of hydrous silicate melts.

Comparison with the polynomial of Burnham and Davis (1971)

There is a significant discrepancy between the values for $\overline{V}_{H_2O^{total}}$ derived in this study as a function of temperature and pressure and those obtained from the polynomial given by Burnham and Davis (1971). This is illustrated in Fig. 4, where the difference in $\overline{V}_{H_2O^{total}}$ is 22.9 vs 27.5 cm^3/mol at one bar and 1223 K, and 17.4 vs 14.2 cm^3/mol at 840 MPa and 1023 K. The primary cause for these substantial differences in the $\overline{V}_{H_2O^{total}}$ values, derived from nearly the same hydrous data set, is

the absence of accurate density data for anhydrous liquids in the calibration of the Burnham and Davis (1971) polynomial. This study benefits from the fact that the partial molar volumetric properties of all other components in a hydrous albitic melt are relatively well constrained (e.g., Stein et al. 1986; Kress et al. 1988; Lange 1996), leaving only the partial molar volumetric properties of the H_2O^{total} component as the unknown. The quality of the fit in Table 5 is also enhanced by the low temperatures and pressures of the experiments reported in this study. If only the data of Burnham and Davis (1971) are used, the temperature and pressure range is 200 K and 400 MPa; by using the data of this study, the temperature range is tripled and the pressure range is doubled, further constraining the thermal expansivity and compressibility of the H_2O^{total} component.

Prior to this study, the polynomial of Burnham and Davis (1971) provided the best means for estimating $\overline{V}_{H_2O^{total}}$ as a function of temperature and pressure, because it was calibrated using direct measurements. However, when their polynomial for $\overline{V}_{H_2O^{total}}$ is directly combined with the molar volume model for anhydrous liquids of Lange and Carmichael (1990) to calculate the density of hydrous albitic liquids, the errors are significant. The combination of the two equations (Burnham and Davis (1971) polynomial for H$_2$O, Eq. 4 for all other components) overestimates the measured densities of Burnham and Davis (1971) by 2.4 to 6.9%, depending on the pressure and temperature conditions (Fig. 5). However, it has been shown in this study that this deviation is not caused by any problems with the Burnham and Davis (1971) density measurements on hydrous albitic liquid (from which the polynomial was derived), but rather from the lack of an internally consistent model that incorporates precise molar volume data for both hydrous and anhydrous liquids.

Implications of the partial molar volume properties of the H_2O^{total} melt component

The temperature and pressure derivatives of $\overline{V}_{H_2O^{total}}$ determined in this study indicate that H_2O^{total} is one of the most expansive and compressible components in magmatic liquids. Compared to the coefficient of thermal expansion for Na$_2$O $(3.02 \times 10^{-4}$ K$^{-1})$ and K$_2$O $(2.93 \times 10^{-4}$ K$^{-1})$ at 1273 K and 100 MPa, that for H_2O^{total} $(4.72 \times 10^{-4}$ K$^{-1})$ is more than 35% greater (Table 7). Similarly, the coefficient of isothermal compressibility for the H_2O^{total} component $(1.70 \times 10^{-5}$ bar$^{-1})$ at 1473 K and 800 MPa is over 50% greater than the value for K$_2$O $(1.10 \times 10^{-5}$ bar$^{-1})$ and almost three times the value for Na$_2$O $(0.59 \times 10^{-5}$ bar$^{-1})$. The large compressibility of the H_2O^{total} in albitic melts is consistent with the results of an in situ spectroscopic study of hydrous glasses to 34 GPa by Closmann and Williams (1995). In that study, they found that hydrogen-oxygen bond lengths in silicate glasses can change dramatically with increasing pressure, implying that the

Table 6 Data of Burnham and Davis (1971)

Sample	Pressure (MPa)	Temperature (K)	Measured density (g/cm³)	Calculated density (g/cm³)	Residual density (calc-meas)/meas
10.9 wt% H₂O	770	1033	2.174	2.165	0.43%
	770	1050	2.168	2.158	0.46%
	770	1071	2.162	2.150	0.53%
	770	1085	2.161	2.145	0.74%
	770	1085	2.155	2.145	0.48%
	770	1097	2.155	2.140	0.71%
	770	1105	2.148	2.137	0.51%
	770	1118	2.150	2.132	0.84%
	770	1122	2.141	2.131	0.50%
	770	1135	2.143	2.126	0.81%
	770	1146	2.137	2.122	0.69%
	770	1153	2.139	2.119	0.90%
	770	1158	2.134	2.118	0.77%
	770	1166	2.131	2.115	0.78%
8.25 wt% H₂O	480	1061	2.173	2.170	0.15%
	480	1062	2.180	2.170	0.49%
	480	1070	2.169	2.167	0.09%
	480	1074	2.172	2.166	0.30%
	480	1076	2.163	2.165	−0.08%
	480	1084	2.166	2.163	0.15%
	480	1091	2.161	2.161	0.02%
	480	1098	2.156	2.159	−0.11%
	480	1104	2.157	2.157	0.01%
	480	1106	2.155	2.156	−0.05%
	480	1125	2.149	2.150	−0.04%
	480	1132	2.146	2.148	−0.08%
	480	1134	2.144	2.148	−0.17%
	480	1146	2.140	2.144	−0.17%
	480	1152	2.139	2.142	−0.13%
	480	1180	2.125	2.134	−0.43%
	480	1201	2.115	2.127	−0.59%
8.25 wt% H₂O	840	1033	2.246	2.256	−0.43%
	840	1052	2.238	2.249	−0.51%
	840	1055	2.234	2.248	−0.63%
	840	1070	2.232	2.243	−0.51%
	840	1103	2.217	2.232	−0.67%
	840	1120	2.214	2.227	−0.56%
	840	1136	2.209	2.221	−0.57%
	840	1151	2.202	2.216	−0.65%
	840	1155	2.204	2.215	−0.51%
	840	1173	2.197	2.209	−0.57%

Table 7 Selected coefficients of thermal expansion and compressibility[a]

Oxide components	At 1273 K and 100 MPa		At 1473 K and 800 MPa	
	α_T^i (10^{-4}, K^{-1})	β_T^i (10^{-5}, bar^{-1})	α_T^i (10^{-4}, K^{-1})	β_T^i (10^{-5}, bar^{-1})
SiO₂	0	0.77	0	0.73
Al₂O₃	0	0.88	0	0.78
Na₂O	3.02	0.10	2.98	0.59
K₂O	2.93	0.41	3.00	1.10
H₂O	4.72	1.66	4.82	1.70
H₂O (fluid)[b]	10.92	99.15	4.63	5.56

[a] α_T^i coefficient of thermal expansion for component i at temperature T, β_T^i coefficient of isothermal compressibility for component i at temperature T
[b] Values for a pure H₂O fluid phase at the respective temperatures and pressures, calculated from Holloway (1981). For a pure H₂O liquid phase at 100 °C and 1 bar, $\alpha_T = 7.5\,(10^{-4}, \text{K}^{-1})$ and $\beta_T = 4.9\,(10^{-5}, \text{bar}^{-1})$ (Eisenberg and Kauzmann 1969)
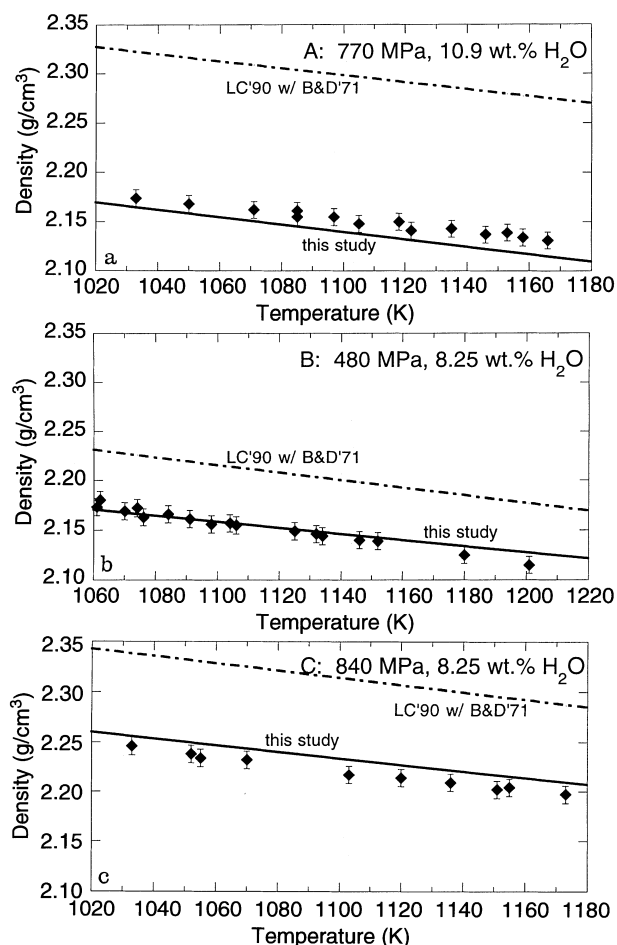
Fig. 5a–c Comparisons of the density measurements of hydrous melts of Burnham and Davis (1971) (*filled diamonds*) with those calculated with the model of this study (*solid line*). Also shown are the densities calculated with a combination of the linear model of anhydrous melts of Lange and Carmichael (1990) and the $\overline{V}_{H_2O^{total}}$ polynomial of Burnham and Davis (1971). The pressure, temperature, and H_2O total content of each melt are shown in the upper right corner of each figure. The error bars on the data of Burnham and Davis (1971) are for the reported uncertainty of 0.4%, not including the uncertainty in the H_2O concentration in their samples

coefficient of isothermal compressibility is large for the hydrous component in silicate liquids.

Interestingly, the coefficients of thermal expansion and isothermal compressibility for the H_2O^{total} component in silicate liquids are similar to but generally lower than those of pure liquid water at low temperatures and pressures. From 20 to 100 °C and 1 bar the coefficient of thermal expansion of pure H_2O liquid varies from ~2.0 to 7.5 (10^{-4}, K^{-1}), whereas the coefficient of isothermal compressibility varies from ~4.4 to 4.9 (10^{-5}, bar^{-1}) (Eisenberg and Kauzmann 1969). When the coefficients of the H_2O^{total} component in a silicate liquid are compared to those calculated for a pure H_2O fluid (using Holloway 1981), the coefficients of the H_2O^{total} component are much lower than those of the pure fluid phase at 100 MPa, but approach those of the pure fluid phase at 800 MPa (Table 7).

Implications of the large thermal expansivity and compressibility of the H_2O^{total} component to the dynamics of melt migration, ascent and differentiation within crustal chambers are numerous. For example, it has been noted in theoretical investigations of magma dynamics (e.g., Sparks et al. 1984) that even small differences in density, on the order of 0.5%, can strongly affect the convective phenomena in magma chambers. For a dry, andesitic melt at 300 MPa, a temperature gradient of 100 degrees across the chamber would provide a 0.5% density contrast; an andesite melt with 4 wt% H_2O^{total}, would require only a 50 degree gradient to achieve the same density contrast.

The effect of even minor amounts of dissolved water has a strong influence on the density of silicate liquids. For example, at 1473 K and 70 MPa (conditions of a mid-ocean ridge crustal magma chamber), the presence of just 0.4 wt% H_2O in a basaltic liquid will change its density by more than one percent. An equivalent change in melt density could be achieved by changing the temperature by 175 K or the pressure by 230 MPa. Therefore, even a few tenths of a percent of H_2O, often considered a negligible quantity, can have a marked effect on the dynamics of melt migration in the crustal environment.

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