Viscosities of granitic (sensu lato) melts: Influence of the anorthite component

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ABSTRACT

The viscosities of a series of granitic (*sensu lato*) melts have been determined in the range of 10^3 to 10^{12} Pa·s. The anhydrous melt compositions are based on the addition of 10, 20, 50, and 75 wt% of the anorthite component (CaAl₂Si₂O₈) to a haplogranitic melt (HPG8) whose composition lies near the 2 kbar water-saturated minimum melt composition in the system NaAlSi₃O₈-KAlSi₃O₈-SiO₂. Melts with 10 and 20 wt% normative anorthite were subjected to high-pressure hydration syntheses using a piston-cylinder apparatus to generate water contents up to 2 wt%. Viscosities were determined for the anhydrous melts using the concentric cylinder method in the viscosity range of 10^2 to 10^5 Pa·s, and for both anhydrous and hydrated melts in the range of 10^9 to 10^{12} Pa·s.

The results for the temperature dependence of viscosity in the anhydrous system indicate that the influence on melt viscosity, caused by the addition of normative anorthite to the haplogranitic melt composition, is strongly temperature-dependent. Viscosity-temperature relationships of the melts become much more non-Arrhenian with addition of normative anorthite. The addition of water to melts with 10 and 20 wt% normative anorthite results in strong nonlinear decreases in viscosity. In the high viscosity range, the results for hydrous melts with 10% normative anorthite are adequately reproduced using the calcalkaline melt viscosity model of Hess and Dingwell (1996), whereas those for hydrous melts with 20 wt% normative anorthite are higher than the model predictions by amounts that depend on the water content. It appears that, at the higher temperatures anticipated for intermediate granitic magmatism, the calcalkaline model can adequately deal with up to 15 wt% normative anorthite in the melt composition in the range of temperatures relevant for intermediate magmas in nature.

INTRODUCTION

The voluminous quantities of granite (*sensu lato*) associated with orogenic terrains on the Earth's continents are dominated by crystalline rocks of intermediate composition that, in the modal classification of Streckeisen (1976), fall within the fields of granodiorite to diorite and their more monzonitic equivalents. The granitic series, broadly defined, can be approximated chemically by the fractionation of plagioclase component together with mafic phases to generate melt compositions that progressively approach petrogeny's residua system. We have chosen, in the present study, to use the simple addition of the anorthite component, as 10, 20, 50, and 75 wt% to the base melt compositions responsible for these extensive plutonic terrains.

Since the pioneering works of Saucier (1952) and Sabatier (1956) in the high viscosity range and Shaw (1963) and Burnham (1963) in the low viscosity range, experimental work on the viscosities of granitic melts have concentrated almost exclusively on natural multicomponent melts or on synthetic

composition melts that were more or less (>95%) confined compositionally to the haplogranitic system (NaAlSi₃O₈-KAlSi₃O₈-SiO₂). The results of the now numerous studies have been recently summarized and subjected to numerical techniques to derive a model for the viscosity of calcalkaline granitic melts as a function of temperature and water content (Hess and Dingwell 1996). The results of that parameterization indicate that variations of a few percent in normative albite, orthoclase, quartz, and anorthite, will have negligible effect on the viscosity of the melt (Hess and Dingwell 1996). This has been recently confirmed experimentally for the haplogranitic system (Hess et al. 2000). However, subsequent systematic extensions of the compositional data base for hydrous granitic melt viscosities have indicated that the model holds well for compositions that are much more peraluminous (up to 5% normative corumdum) than the initial compositional calibration range (Dingwell et al. 1998a). However, the model fails badly for melt compositions that are even moderately peralkaline (Dingwell et al. 1998b) and for the addition of other components such as F, P, and B (Dingwell et al. 1996a). In general, then, we remain confronted with the task of further testing and/ or calibration of the existing model with successive sets of data for the important compositional variables in intermediate melt compositions. We contribute to this task here with the investigation of the influence of the anorthite component. The viscosity-temperature relationship for pure anorthite melt has been

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determined previously (Urbain et al. 1982; Scarfe et al. 1983; Hummel and Arndt 1985), and indicates that at high temperatures significantly lower melt viscosities might be expected for anorthite-rich granitic melts whereas at lower temperature, the variation of melt viscosities with anorthite content is minimal.

METHODS

For our investigations, we have chosen as a base composition a haplogranitic melt composition (designated HPG8 by Holtz et al. 1992), which lies near the 2 kbar ($P_{\rm H_20}$) ternary minimum in the system SiO₂-NaAlSi₃O₈-KAlSi₃O₈. This melt composition was chosen to enable a direct comparison of the present results for the influence of water on melt viscosity with the data provided by Hess et al. (1995, 1996) for the effects of alkali and alkaline earth oxides and to serve as a model for calcalkaline rhyolites.

The base melt used in the present study was generated by direct fusion of powder mixes of oxides and carbonates at 1 atm and 1400–1650 °C. The fusions were performed in thin-walled platinum crucibles with a $MoSi_2$ box furnace. The partially fused products of this initial fusion were transferred in the crucibles to the viscometer furnace and a stirring spindle was entered into the samples from above. The samples were fused for several tens of hours until inspection of the stirring spindle, removed periodically from the melt, indicated that the melts were crystal- and bubble-free. The melts were removed from the furnace and allowed to cool in air in the Pt crucibles. The glass compositions were analyzed by solution-based ICP-AES methods. The results of bulk anhydrous analyses are presented in Table 1.

The low range viscosities were obtained using the concentric cylinder system and methods described by Dingwell (1989a, 1989b, 1991). Viscosity is determined in steps of decreasing temperature with a final reoccupation of the highest temperature data point as a check against instrumental or sample drift. The system is calibrated against DGG-1, a viscosity standard glass. The viscometer head employed was the Brookfield DVIII+, which has a full-scale torque of 7.2×10^{-2} N-m. The spindle used was the Pt₈₀Rh₂₀ low viscosity spindle described by Dingwell and Virgo (1988). After concentric cylinder viscometry, the samples were permitted to cool in the crucibles. From the glassy products, cores were drilled with a diamond coring tool and the cores were cut into disks and polished in preparation for micropenetration viscometry.

For hydrations, glass cylinders (diameter = 5 mm; length = 1 cm) were drilled by water-cooled diamond coring tools, cleaned ultrasonically in acetone, and then dried at 110 °C to remove any residue of acetone. Glass samples (~100 mg) were

then loaded together with known amounts of doubly distilled H₂O in capsules formed from platinum tubing (outer diameter = 5 mm; length = 10 mm; wall thickness = 0.1 mm) and sealed with an arc-welder. The capsules were checked for possible leakage by testing for weight loss after drying in an oven at 110 °C for at least one hour. After an hour at 110 °C, the added water was considered to be distributed randomly in the platinum capsule, and thus around the glass sample. Synthesis of hydrous glasses was performed in a piston cylinder apparatus. The piston-cylinder runs were performed at 1550 to 1600 °C and 10 kbar pressure for durations of several hours. The runs were quenched isobarically by pumping during the quench. The quenched run products consisted of crystal-free limpid glasses. Doubly polished sections of 1 mm thickness were prepared for micropenetration viscometry by grinding with alumina abrasive and then polishing with diamond paste using water as a lubricant. Polished samples were cleaned with acetone to remove epoxy. The homogeneity of water in the glasses was tested by analysis with an infrared spectrometer (see below).

MICROPENETRATION VISCOMETRY

The high range viscosities were measured using a micropenetration technique. This involves determining the rate at which an Ir-indenter under a fixed load moves into the melt surface. These measurements were performed in a BÄHR DIL 802 V vertical push-rod dilatometer. The sample is placed in a silica rod sample holder under an Ar gas flow. The indenter is attached to one end of an alumina rod, which is attached at the other end to a weight pan. The metal connection between the alumina rod and the weight pan acts as the core of a calibrated linear voltage displacement transducer (LVDT). The movement of this metal core as the indenter is pushed into the melt yields the displacement. The present system uses hemispherical Ir-indenters with diameters of 2 mm, and a force of 1.2 N. The absolute shear viscosity is determined from

$$\eta = \frac{0.1875Pt}{r^{0.5}\alpha^{1.5}} \tag{1}$$

(Pocklington 1940; Tobolsky and Taylor 1963) for the radius of the half-sphere *r*, the applied force *P*, indent distance a, and time *t* (t = 0, a = 0 upon application of the force). The measurements are performed over indentation distances of less than 100 µm. The technique yields an absolute determination of viscosity up to 1100 °C in the range of 10⁸ to 10^{11.5} Pa·s. Viscosities determined on the DGG-1 standard glass using 3 mm samples have been reproduced within an error of \pm 0.06 log units.

All samples were heated up to the measuring temperature,

TABLE 1. Melt compositions (wt%) determined by ICP-AES

SiO ₂	AI_2O_3	Na ₂ O	K₂O	CaO	Total	Excess mole fraction	Sample	
78.6 ± 0.4	12.5 ± 0.2	4.6 ± 0.3	4.2 ± 0.2		100.0	-0.003	HPG8	
73.6 ± 0.5	15.6 ± 0.6	4.4 ± 0.4	3.8 ± 1.8	2.1 ± 0.6	99.5	-0.003	HPG8An10	
71.5 ± 0.4	17.3 ± 0.1	4.2 ± 0.6	3.5 ± 0.2	4.0 ± 0.6	100.5	0.004	HPG8An20	
64.0 ± 0.3	23.1 ± 0.2	2.6 ± 1.1	1.9 ± 2.0	8.7 ± 0.7	100.3	-0.006	HPG8An50	
56.2 ± 0.4	27.2 ± 0.5	1.6 ± 0.8	1.5 ± 3.4	13.3 ± 0.6	99.8	0.008	HPG8An75	

Note: Numbers quoted as uncertainties are in percent relative to the analysed amount. Excess mole fraction is the moles of Ca, Na, and K in excess of those needed to charge balance Al.

held at this temperature for at least 15 min to allow thermal equilibration and complete structural relaxation of the sample and then the viscosity measurement was performed. After the measurement, each sample was cooled at a rate of approximately 20 K/min.

A primary advantage of the micropenetration technique is that small amounts of sample and simple sample geometry constraints are required compared to other high-viscosity methods such as parallel plate or fiber elongation (Dingwell et al. 1993; Dingwell 1995). Using the formulation of Pocklington (1940), the penetration distance should be small compared to the sample height. As this criterion cannot always be maintained in our present experiments, we have evaluated the effect of sample size on a DGG-1 standard glass with a thickness of 1 mm. Between 10^8 to $10^{10.5}$ Pa·s, there appears to be a slight but significant deviation of the determined viscosities to values higher than the calculated ones reported in the Deutsche Glastechnische Gesellschaft data sheet. The data listed in Table 3b, all performed on samples of 1 mm thickness, are corrected for this sample size effect.

Control of water content

Water contents of the samples, both as total water dissolved in the melt and as the fractions of water dissolved as molecular water and as hydroxyl groups, were determined by IR spectroscopy. Because the present samples were investigated under water-oversaturated conditions at 1 bar, samples were analyzed before and after each viscosity measurement to check for water loss during viscometry. Samples were then positioned over an aperture in a brass disc in order to aim the beam at areas of interest in the glasses. A Bruker IFS 120 HR fourier transform spectrophotometer was used to obtain transmission infrared spectra in the NIR region (2500-8000 cm⁻¹), using a W source, CaF₂ beamsplitter, and a narrow band MCT detector. The spectrophotometer operated at a resolution of 4 cm⁻¹ with a scanning speed of 20.0 KHz and the objective used was a Cassegrainian 15X. Typically, 200-1000 scans were collected for each spectrum. Background was recorded and subtracted from every spectrum. The two bands of interest of this regions are at 4500 cm⁻¹ and 5200 cm⁻¹.

The H₂O content of the samples was determined by measuring the heights at the maximum of the absorption bands near 4500 and 5200 cm⁻¹ attributed respectively to the combination stretching + bending mode of X-OH groups, and a combination of stretching + bending modes of molecular water groups (Stolper 1982). The precision of the measurements presented here is based on the reproducibility of the measurements of glass fragments repeated over a long period of time as well as on the uncertainty assigned to the background subtraction procedure. We estimate that the typical uncertainty in our measurements for the 4500 cm⁻¹ and the 5200 cm⁻¹ band is approximately 0.003 absorbance units. The concentration of dissolved "water" in a glass contributing to a given band can be determined as follows:

$$c = (18.02)(absorbance)/(\delta)(\rho)(\epsilon)$$
(2)

where c is the H₂O concentration in weight fraction, 18.02 is

the molecular weight of water, absorbance is the height of the absorption peak, δ is the thickness of the specimen in cm, ϵ is the molar absorptivity (or extinction coefficient) in liter/molcm, and ρ is the density of the sample in g/liter.

In addition, the absolute water contents of the hydrous samples was determined using the Karl-Fischer Titration (KFT) method. The analytical method is described by Westrich (1987) and Behrens (1995). The water of the freshly crushed sample is released by rapidly (200 °C/min) heating it up to 1300 °C in a platinum sample holder using a high-frequency generator. The water is than transported by an Ar stream into the moisture cell, where the amount of water is measured quantitatively by the reaction with coulometrically generated iodine. A critical point for the determination of water contents by KFT is how much water remained dissolved in the glass during the analyses. This has been determined to be 0.10 wt% for a wide variety of compositions (Holtz et al. 1994) and thus this amount was added to the determined values here.

Density measurements were performed using a Mettler Toledo AG 204 balance (with density determination kit) using ethanol as a reference liquid (Table 2). Precision, for samples whose masses range from 0.02 to 0.06 g, is estimated to be ± 0.05 g/cm³ based on reproducibility of measurements on different samples. The thickness of each glass plate was measured with a digital Mitutoyo micrometer (precision $\pm 3.10^{-6}$ cm) (Table 2).

In order to determine quantitatively the wt % of the water species in the samples investigated, the molar absorptivities are required. We considered for our study molar absorptivity values presented by Nowak and Behrens (1995) for an haplogranitic composition very similar to that investigated here ($e_{OH} = 1.56 \pm 0.04$; $e_{H_2O} = 1.79 \pm 0.04$). Infrared determination of the water content (molecular water and water dissolved as hydroxyl groups) was performed after each viscosity measurement in order to test the homogeneity of the sample and to rule out significant water loss.

The water contents of all samples remained constant during the high-temperature dwells employed to assure fully relaxed viscosity determinations in the viscometry measurements. The viscosity data obtained thus pertain to structurally equilibrated, fully relaxed, water-bearing melts at 1 bar pressure and the temperatures of measurement.

RESULTS AND DISCUSSION

The results of the concentric cylinder and micropenetration viscometric determinations for anhydrous melts are presented in Table 3 and illustrated in Figures 1a and 1b as a function of the reciprocal absolute temperature. The high-viscosity data set (Fig. 1a) illustrates that the anorthite component initially (at HPG8An10) contributes to a decrease of melt viscosity at approximately constant activation energy, and then to a diminished viscosity decrease (at HPG8An20) accompanied by an increase in the activation energy. At HPG8An50 the viscosity is lower than the extrapolation of the trend from HPG8 to HPG8An20 would predict but the activation energy is similar to that of HPG8An20. For compositions beyond 50 wt% anorthite component, the viscosity shows a crossover to higher values at higher activation energies indicative of more fragile

Composition	BV/AV	Sample	A H ₂ O (5200 cm ⁻¹)	A OH (4500 cm ⁻¹)	Thickness (mm)	Density (g/l)	FTIR H ₂ O (wt%)	Rem	KFT H ₂ O (wt%)	
HPG8An10	BV	717-4	0.037	0.144	1.018	2386	0.84		0.89	
HPG8An10	AV	717-4	0.034	0.131	1.028	2386	0.76	*	0.89	
HPG8An10	BV	716-4	0.162	0.243	1.024	2381	1.82		2.10	
HPG8An10	AV	716-4	0.179	0.216	1.031	2381	1.75		2.10	
HPG8An10	BV	716-3	0.167	0.243	1.024	2381	1.84		2.10	
HPG8An10	AV	716-3	0.175	0.214	1.031	2381	1.72		2.10	
HPG8An10	BV	716-2	0.171	0.243	1.024	2381	1.86		2.10	
HPG8An10	BV	716-1	0.151	0,238	1.024	2381	1.75		2.10	
HPG8An20	AV	715-3	0.031	0.124	1.024	2437	0.70	*	1.07	
HPG8An20	AV	715-2	0.037	0.134	1.024	2437	0.77		1.07	
HPG8An20	AV	715-1	0.033	0.128	1.024	2437	0.73	*	1.07	
HPG8An20	BV	714-4	0.138	0.230	1.026	2432	1.62		2.03	
HPG8An20	AV	714-4	0.153	0.211	1,031	2432	1.59		2.03	
HPG8An20	BV	714-3	0.141	0.223	1.026	2432	1.60		2.03	
HPG8An20	AV	714-3	0.155	0.209	1.031	2432	1.58		2.03	
HPG8An20	BV	714-2	0.136	0.266	1.026	2432	1.78		2.03	
HPG8An20	AV	714-2	0.149	0.207	1.032	2432	1.55		2.03	
HPG8An20	BV	714-1	0.130	0.244	1.026	2432	1.65		2.03	
HPG8An20	AV	714-1	0.142	0.207	1.032	2432	1.52		2.03	

Notes: BV/AV: refer to FTIR measurements before or after the viscosity determination.

A H₂O, A OH: Absorbance, error = 0.003 units; values reported refer to multiple spectra.

KFT H₂O: refers not to the specific sample, but rather to a bulk analysis of the sample material.

Note that the FTIR measurements were done only to check for possible water loss before and after the viscosity measurements and for homogeneity. An absolute determination of water is not yet possible using FTIR on the melts, because the molar absorption coefficients change with increasing anorthite content.

12

10

HPG8

HPG8An10

HPG8An20

0

 ∇

* Slightly foamed after viscosity measurement.



Viscosity (Pars) HPG8An50 HPG8An75 0 An100 0 9 ğ n 6 7 8 10000/T (K)

FIGURE 1A. The low range viscosity-temperature relationships for haplogranitic melts with variable amounts of normative anorthite. Addition of 10% anorthite component to the haplogranite system leads to a small but systematic decrease of the viscosities. Further increase of the anorthite component significantly changes the apparent activation energy of the viscous flow process, resulting in a crossover of individual viscosity-temperature curves.

behavior and leading to relatively high activation energies for HPG8An75 as for An100 (Hummel and Arndt 1985). These trends are consistent with the independently obtained low-viscosity-range data from concentric cylinder viscometry. The two data sets-high-viscosity, micropenetration measurements and low-viscosity, concentric-cylinder measurements-are combined in Figure 1b. The total range of viscosity variation is much greater at high temperatures in the HPG8-anorthite system than at lower temperatures. This feature of viscosity variation in granitic (sensu lato) melts contrasts with the general tendency of viscosity-temperature relationships of silicate melts

FIGURE 1B. The complete viscosity regime for the same melts. The addition of the anorthite component leads to a strong reduction in viscosity in the low-viscosity range. The situation is more complex in the high-viscosity range, where the addition of anorthite leads to a slight initial decrease and then an increase in melt viscosity. Increasing normative anorthite content yields therefore an increasingly non-Arrhenian temperature dependence of viscosity.

to converge with increasing temperature. It presents a clear signal to multicomponent modeling of silicate melt viscosity-temperature relationships that sufficient detail must be available in the viscosity data base before sensible attempts at modeling the results can be obtained. In contrast to the high-viscosity range, where the total variation in isothermal viscosity is approximately 2 log units, the range at low viscosities is over 3 log units. Activation energies decrease monotonously with added anorthite component, demonstrating clearly, when taken together with the high-viscosity data, that the anorthite component leads to more-pronounced non-Arrhenian viscosity-tem-

TABLE 3A. Concentric cylinder and micropenetration viscometry data for dry samples

 TABLE 3C. Comparison of viscosity data with Hess and Dingwell (1996) model

	, ,			
T(°C)	HPG8An10	HPG8An20	HPG8An50	HPG8An75
±0.5 °C	log₁₀ η	log₁₀η	log₁₀ η	log₁₀η
	(Pa·s)	(Pa·s)	(Pa·s)	(Pa·s)
1649	2.97	2.60		
1599	3.24	2.85		1.11
1549	3.50	3.12	2.01	1.35
1499	3.81	3.40	2.27	1.61
1449	4.13	3.71	2.56	1.89
1399		4.04	2.88	2.27
1349		4.40	3.22	2.61
1299			3.59	2.99
1249			4.02	3.40
1199			4.49	3.90
1149				4.46
927.4	9.75			
892.6	10.25			
876.9	10.51			
933.7		9.25		
893.2		10.03		
871.1		10.50		
928.3			8.70	
916.0			9.10	
894.2			9.52	
852.9			10.60	
931.8				8.60
928.2				8.69
910.7				9.12
8/2.5				10.22

 TABLE 3B. Micropenetration viscometry data for wet samples

Composition	Sample	T (°C) + −0.5 °C	log ₁₀ η (Pa·s)
		± 0.0 0	(1 4 3)
HPG8+10An-0.89	717-4	643.6	9.81
HPG8+10An-0.89	717-3	665.2	10.30
HPG8+10An-0.89	717-1	678.2	9.62
HPG8+10An-0.89	717-2	703.0	9.15
HPG8+10An-2.10	716-4	521.8	10.99
HPG8+10An-2.10	716-3	522.8	10.85
HPG8+10An-2.10	716-2	562.8	9.79
HPG8+10An-2.10	716-1	607.3	8.97
HPG8+20An-0.87	715-1	615.5	11.32
HPG8+20An-0.87	715-2	641.0	10.40
HPG8+20An-0.87	715-3	670.5	9.84
HPG8+20An-0.87	715-4	689.2	9.40
HPG8+20An-2.03	714-3	532.4	11.41
HPG8+20An-2.03	714-1	539.8	10.87
HPG8+20An-2.03	714-2	543.6	10.80
HPG8+20An-2.03	714-4	547.4	10.28
HPG8+20An-2.03	714-6	561.0	10.08
HPG8+20An-2.03	714-5	582.9	9.75

Sample	H_2O		log ₁₀ η (Parc)	log ₁₀ η (Parc)	Residuals
	(₩1/0)	(0)	measured	calculated	4
HPG8	0.02	16/2 00	3.24	3 /1	0 17
HPG8	0.02	1593 99	3 58	3.69	0.17
HPG8	0.02	1543 99	3.81	3 99	0.18
HPG8	0.02	1494 99	4 15	4 30	0.15
HPG8	0.02	1445.99	4 53	4 63	0.10
HPG8	0.02	1396.99	4 90	4 98	0.08
HPG8	0.02	1179.99	6 79	6.83	0.00
HPG8	0.02	938.79	10.16	9.71	-0.45
HPG8	0.02	925.69	10.28	9.90	-0.38
HPG8	0.02	904.99	10.63	10.21	-0.42
HPG8	0.02	881.69	11.02	10.58	-0.44
HPG8An10	0.02	1649.00	2.97	3.38	0.41
HPG8An10	0.02	1599.00	3.24	3.66	0.43
HPG8An10	0.02	1549.00	3.50	3.96	0.46
HPG8An10	0.02	1499.00	3.81	4.28	0.47
HPG8An10	0.02	1449.00	4.13	4.61	0.48
HPG8An10	0.02	927.40	9.75	9.88	0.13
HPG8An10	0.02	892.60	10.25	10.41	0.16
HPG8An10	0.02	876.90	10.51	10.66	0.15
HPG8An20	0.02	1649.00	2.60	3.38	0.77
HPG8An20	0.02	1599.00	2.85	3.66	0.81
HPG8An20	0.02	1549.00	3.12	3.96	0.84
HPG8An20	0.02	1499.00	3.40	4.28	0.88
HPG8An20	0.02	1449.00	3.71	4.61	0.91
HPG8An20	0.02	1399.00	4.04	4.97	0.93
HPG8An20	0.02	1349.00	4.40	5.35	0.95
HPG8An20	0.02	933.70	9.25	9.78	0.53
HPG8An20	0.02	893.20	10.03	10.40	0.37
HPG8An20	0.02	871.10	10.50	10.75	0.25
	0.89	643.60	9.01	9.90	0.17
	0.09	679.20	10.30	9.09	-0.71
HPG910An-0.09	0.09	702.00	9.02	9.30	-0.20
HPG810An-2 10	2 10	521.80	10.99	10 71	-0.20
HPG810An-2 10	2 10	522.80	10.85	10.68	-0.17
HPG810An-2 10	2 10	562.80	9 79	9.80	0.01
HPG810An-2.10	2.10	607.30	8.97	8.94	-0.03
HPG820An-0.87	0.87	615.50	11.32	10.58	-0.74
HPG820An-0.87	0.87	641.00	10.40	10.08	-0.32
HPG820An-0.87	0.87	670.50	9.84	9.54	-0.30
HPG820An-0.87	0.87	689.20	9.40	9.22	-0.18
HPG820An-2.03	2.03	532.40	11.41	10.54	-0.87
HPG820An-2.03	2.03	539.80	10.87	10.38	-0.49
HPG820An-2.03	2.03	543.60	10.80	10.29	-0.51
HPG820An-2.03	2.03	547.40	10.28	10.21	-0.07
HPG820An-2.03	2.03	561.00	10.08	9.92	-0.16
HPG820An-2.03	2.03	582.90	9.75	9.47	-0.28
Note: Viscosity da	ata for HF	G8 from Di	ngwell et al.	(1996b).	
* Trace water con	tents of d	Iry melts are	e estimated	to be 200	ppm.

perature relationships for these melts. All of the viscosity-temperature relationships of the combined high- and low-viscosity data ranges have been fitted to the VFT relationship and the coefficients as well as the standard errors of the fit are included in Table 4.

The variation of viscosity (expressed as isokom temperatures) with the addition of anorthite component to HPG8 is illustrated in Figure 2 for the 10^{12} , 10^8 , and 10^2 Pa·s isokoms. The 10^2 Pa·s isokom illustrates perhaps most clearly that the decrease in viscosity with added anorthite component is not a simple monotonous decrease in viscosity at this isokom. Rather, there is a discernable step drop in viscosity between 20 and 50 wt% anorthite component. The effect can be seen better in the variation of the VFT-parameters of Table 4. These are plotted in Figure 3 where the discontinuity between 20 and 50 wt% anorthite is apparent. It is tempting to speculate on what structural causes exist for this behavior. Perhaps the structural role of Al in these melts goes through a subtle transition in the composition range between 20 and 50 wt% anorthite component. There are many topological possibilities for such a structural transition, including the potential of Al-bearing tricluster stabilization (Toplis et al. 1997). The reasons must be investigated in the future using direct probes of the melt structures.

The viscosity data for the hydrous melts are presented in Figure 4 together with Arrhenian fits to those data. It is clear from Figure 4 that Arrhenian fits suffice to describe the temperature dependence of the viscosities of these melts over these relatively restricted temperature ranges.



FIGURE 2. Temperature as a function of anorthite component for dry melts at three different viscosity isokoms. At low viscosities (superliquidus regime), the temperature dependence scales linearly with anorthite component in the melt. At high viscosities (in the glass transition regime), the temperature for equal structural relaxation times is nearly independent of composition.

Comparison with the Hess and Dingwell model

A comparison of the viscosities of anorthite-bearing haplogranitic melts with the predictions of the Hess and Dingwell (1996) model of metaluminous melt viscosities is presented for the 10^{12} Pa·s isokom in Figure 5. There it can be seen that the model functions quite well for anhydrous anorthite-bearing melts up to anorthite contents of 20 wt%. Beyond that anorthite content, the model underestimates the viscosities significantly.

The viscosities of all melts measured in this study, anhydrous and hydrous, are compared in Table 3c with the predictions of the Hess and Dingwell (1996) metaluminous (anorthite-free) model. Again it can be seen from the residuals of Table 3c that the model works reasonably well for anorthite contents up to 20 wt% in anhydrous and in hydrous melts near the glass transition. Beyond 20 wt% anorthite, the model does not work well and should not be applied.

CONCLUDING REMARKS

The influence of the anorthite component on the viscosity of haplogranitic melts results in a strongly temperature-dependent variation in the viscosity. The Hess and Dingwell (1996) calcalkaline model is capable of reproducing viscosities for melts of up to 15 wt% anorthite component within reasonable error estimates. The high-temperature viscosities of anorthiterich melts display the greatest deviation from haplogranitic melt viscosities. Further investigation of calcalkaline melt viscosities must include the influences of components such as FeO and MgO before intermediate melanocratic granite (*sensu lato*) melts can be modeled adequately.



FIGURE 3. The VFT parameters (a,b,c) exhibit a behavior appearing to indicate a change in flow behavior between 10 and 20% anorthite component in melts at high and low temperatures.



FIGURE 4. The high range viscosities of wet tectosilicate melts as a function of reciprocal absolute temperature. Water significantly decreases the viscosities of the dry melt compositions. However, the influence of water on the apparent activation energy remains minor.

TABLE 4. Fit Parameters for the Tamann-Vogel-Fulcher equation and calculated isokom temperatures

VFT-FIT	а	b	C	Fit Std Err*	T (K) at	T (K) at	T(K) at		
	(log ₁₀ Pa·s)	(K)	(K)		$\eta = 10^{12}$	η =10 ⁸	$\eta = 10^{2}$		
					Pa·s	Pa·s	Pa·s		
HPG8	-6.2191	15950	232.24	0.050	1107.7	1354.0	2172.8		
HPG8An10	-6.2527	15678	216.72	0.041	1075.7	1316.7	2116.5		
HPG8An20	-4.2905	10061	463.88	0.004	1081.5	1282.5	2063.3		
HPG8An50	-4.1134	7223.6	637.5	0.053	1085.8	1233.8	1819.1		
HPG8An75	-4.2026	6067.6	727.91	0.058	1102.4	1225.1	1706.1		
An100	-4.2808	4862.1	826.18	0.076	1124.8	1222.1	1600.3		
Notes: Fit St	Votes: Fit Std Err* refers to (sum of residuals squared / degree of freedom) × 100.5.								

Viscosity data for anorthite from Hummel and Arndt (1985) and Scarfe et al. (1983)



FIGURE 5. Temperature as a function of H_2O component for melts at the 10^{12} Pa·s isokom, corresponding to the structural relaxation time near the conventional glass transition. The wet HPG8An10 melt compositions are in fair agreement with the HD-model. For higher amounts of the anorthite component, the HD-model predicts significantly lower temperatures, resulting in an overestimation of the effect of water on reducing viscosities of anorthite-rich melts.

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REFERENCES CITED

- Behrens, H. (1995) Determinations of water solubilities in high viscosity melts. European Journal of Mineralogy, 4, 905–920.
- Burnham, N.L. (1963) Viscosity of a H₂O rich pegmatite melt at high pressure. Geological Society of America Special Paper, 76, 26.
- Dingwell, D.B. (1989a) The effect of fluorine on the viscosity of diopside melt. American Mineralogist, 74, 333–338.
 - ——(1989b) Shear viscosities of ferrosilicate liquids. American Mineralogist, 74, 1038–1044.
 - ——(1991) Redox viscometry of some iron-bearing silicate liquids. American Mineralogist, 76, 1560–1562.
- (1995) Viscosity and anelasticity of melts and glasses. In T. Ahrens, Ed., Mineral Physics and Crystallography. A Handbook of Physical Constants, AGU Reference Shelf 2, p. 209–217. American Geophysical Union, Washington, D.C.
- Dingwell, D.B. and Virgo, D. (1988) Melt viscosities in the Na₂O-Fe₂O₃-SiO₂ system and factors controlling the relative viscosities of fully polymerized silicate melts. Geochimica et Cosmochimica Acta, 52, 395–403.
- Dingwell, D.B., Bagdassarov, N.S., Bussod, G.Y., and Webb, S.L. (1993) Magma Rheology. Handbook on Experiments at High Pressure and Applications to the Earth's Mantle, 21, 1–42.
- Dingwell, D.B., Hess, K.-U., and Knoche, R. (1996a) Granite and granitic pegmatite melts: volumes and viscosities. Transactions of the Royal Society of Edinburgh: Earth Sciences, 87, 65–72.

- Dingwell, D.B., Romano, C., and Hess, K.-U. (1996b) The effect of water on the viscosity of a haplogranitic melt under P-T-X-conditions relevant to silicic volcanism. Contributions to Mineralogy and Petrology, 124, 19–28.
- Dingwell D.B., Hess K.-U., and Romano C. (1998a) Viscosity data for hydrous peraluminous granitic melts: incorporation in a metaluminous model. American Mineralogist, 83, 236–239.
- ——(1998b) Extremely fluid behavior of hydrous peralkaline rhyolites. Earth and Planetary Science Letters, 158, 31–38.
- Hess, K-U. and Dingwell, D.B. (1996) Viscosities of hydrous leucogranitic melts: a non-Arrhenian model. American Mineralogist, 81, 1297–1300.
- Hess, K.-U., Dingwell, D.B., Gennaro, C. and Mincione, V. (2000) Viscosity-temperature behaviour of dry melts in the Qz-Ab-Or system. Chemical Geology, in press.
- Hess, K.-U., Dingwell, D.B., and Webb, S.L. (1995) The influence of excess alkalies on the viscosity of a haplogranitic melt. American Mineralogist, 80, 297– 304.
- ——(1996) The influence of alkaline earth oxides on the viscosity of granitic melts: systematics of non-Arrhenian behavior. European Journal of Mineralogy, 8, 371–381.
- Holtz F., Behrens H., Dingwell D.B., and Taylor R. (1992) Water solubility in aluminosilicate melts of haplogranitic composition at 2 Kbar. Chemical Geology, 96, 289–302
- Holtz, F., Behrens, H., and Dingwell, D.B. (1994) Water solubility in haplogranitic melts: compositional, pressure and temperature dependence. American Mineralogist, 80, 94–108.
- Hummel, W. and Arndt, J. (1985) Variation of viscosity with temperature and composition in the plagioclase system. Contributions to Mineralogy and Petrology, 90, 83–92.
- Nowak, M. and Behrens, H. (1995) The speciation of water in haplogranitic glasses and melts determined by in situ near-infrared spectroscopy. Geochimica et Cosmochimica Acta, 59, 3445–3450.
- Pocklington, H.C. (1940) Rough measurement of high viscosities. Proceedings of the Cambridge Philosophical Society, 36, 507–508.
- Sabatier, G. (1956). Minéralogie—Influence de la teneur en eau sur la viscosité d'und rétinite, verre ayant la compsition chimique d'un granite. Academie des Sciences, 1340–1342.
- Saucier, H. (1952). Quelques expérimences sur la viscosité a haute température de verres avant la composition d'un granite. Compt. Rendu–Academie des Sciences.
- Scarfe, C.M., Cronin, D.J., Wenzel, J.T., and Kauffman, D.A. (1983) Viscositytemperature relationships at 1 atm in the system diopside-anorthite. American Mineralogist, 68, 1083–1088.
- Shaw, H.R. (1963) Obsidian-H₂O viscosities at 1000 and 2000 bars in the temperature range 700 to 900°C. Journal of Geophysical Research, 68, 6337–6343.
- Stolper, E. (1982) The speciation of water in silicate melts. Geochimica et Cosmochimica Acta, 46, 2609–2620.
- Streckeisen, S.T. (1976) To each plutonic rock its proper name. Earth Science Reviews, 12, 1–33.
- Tobolsky, A.V. and Taylor, R.B. (1963) Viscoelastic properties of a simple organic glass. Journal of Physical Chemistry, 67, 2439–2442.
- Toplis, M.J., Dingwell, D.B., and Lenci, T. (1997) Peraluminous viscosity maxima in Na₂O-Al₂O₃-SiO₂ liquids; the role of triclusters in tectosilicate melts. Geochimica et Cosmochimica Acta, 61, 2605–2612.
- Urbain, G., Bottinga, Y., and Richet, P. (1982) Viscosity of liquid silica, silicates and alumino-silicates. Geochimica et Cosmochimica Acta, 46, 1061–1072.
- Westrich, H.R. (1987) Determination of water in volcanic glasses by Karl-Fischer titration. Chemical Geology, 63, 335–340.

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