

Viscosities of hydrous leucogranitic melts: A non-Arrhenian model

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ABSTRACT

An equation is presented for the calculation of the shear viscosity of hydrous (0–12.5 wt% H₂O) leucogranitic melts from 10² to 10¹³ Pa·s. The equation is a multiple nonlinear least-squares regression of a data set of 111 viscosity determinations in the literature. It is based on the Vogel-Fulcher-Tammann (VFT) form and thus accounts for the very important non-Arrhenian temperature dependence of the viscosity. This is possible because of the inclusion of data obtained recently in the high-viscosity region.

The equation includes a logarithmic dependence of the three adjustable parameters of the VFT equation on the H₂O content of the melt. The root-mean-standard deviation for the 111 data points included is 0.46 log₁₀ units. In comparison with earlier methods based on the Arrhenian approximation of the temperature dependence of viscosity, the present method provides significant improvement in the accurate prediction of viscosity. We recommend its use in all petrologic calculations involving hydrous, metaluminous, leucogranitic melts. It is an especially powerful method for calculating the high viscosities thought to be relevant to erupting silicic volcanic systems and magmatic hydrothermal granitic and pegmatitic systems at the brittle-ductile transition.

INTRODUCTION

The viscosity of silicate melts is a powerful constraint on the nature and efficiency of magmatic processes. As early as the 1930s Bowen (1934) noted that one of the most important physical distinctions between basaltic and rhyolitic volcanism lies in the extraordinary difference in viscosities between the two systems at their eruptive temperatures.

Silicic magmas are often inferred to have been H₂O-rich during their petrogenesis, and the influence of H₂O on the viscosity of silicic melts has a long history of investigation (Saucier 1952; Sabatier 1956; Friedman et al. 1963; Shaw 1963; Dingwell 1987; Scaillet et al. 1996; Baker 1996), which in recent years has concentrated on the effects of relatively high H₂O contents (several weight percent H₂O) on the viscosity of rhyolitic and granitic melts (Dingwell 1987; Scaillet et al. 1996; Schulze et al. 1996). Very recently, however, attention has turned again to the region of relatively low H₂O contents and relatively high viscosities (Dingwell et al. 1996; Richet et al. 1996). This is due in part to increasing interest in the physics of eruptive processes and the potentially dominant role of viscosity in controlling the processes involved in melt degassing and fragmentation (e.g., Thomas et al. 1994).

Despite the fact that models for the calculation of melt viscosity have been present for 25 years (Bottinga and Weill 1972; Shaw 1972), prediction of the viscosity of hydrous granitic melts is complicated by the fact that the temperature dependence of the viscosity is significantly non-Arrhenian (e.g., Richet et al. 1996; Dingwell 1997). Here we present an empirical model to account for the non-Arrhenian temperature dependence.

METHOD

For the purposes of our fit we extracted all the available viscosity data for hydrous, metaluminous, leucogranitic melts, a total of 111 data points, from the literature. The data, along with their sources, are listed in Table 1. These data were used as input in the fitting. Two aspects of this input should be noted. First, although these data have been obtained for a range of pressures, from 1 bar to several kilobars, there is clear and adequate evidence from previous studies that the pressure dependence of the viscosity of hydrous granitic melts is insignificant in comparison with the dependence of viscosity on temperature and H₂O concentration over this pressure range (Dingwell 1987; Schulze et al. 1996). Thus, no explicit pressure dependence is included in the present analysis. Second, speciation of the dissolved H₂O was ignored. A direct link between H₂O speciation, as inferred from spectroscopic studies of melts and glasses, and melt viscosity has yet to be established.

In our approach, the VFT equation

$$\log \eta = A + B/(T - C) \quad (1)$$

is rewritten so that the VFT parameters are replaced by expressions *a*, *b*, and *c*, introducing a composition dependence (H₂O concentration in weight percent) for each VFT parameter:

$$\log \eta = a(\text{H}_2\text{O}) + b(\text{H}_2\text{O})/[T - c(\text{H}_2\text{O})] \quad (2)$$

We fitted the data of Table 1 to different functional forms using standard nonlinear regression techniques with a Levenberg-Marquard algorithm (Press et al. 1986). This algorithm seeks the values of the parameters that mini-

TABLE 1. Viscosities of hydrous, metaluminous melts used to construct the model

H ₂ O (wt%)	T (K)	log η (Pa·s)	Ref.	Resid.	H ₂ O (wt%)	T (K)	log η (Pa·s)	Ref.	Resid.	H ₂ O (wt%)	T (K)	log η (Pa·s)	Ref.	Resid.
0.02	1673	4.91	3	-0.05	1.00	867	10.98	7	0.22	3.98	1180	4.45	9	0.11
0.02	1453	6.79	5	-0.04	1.00	885	10.45	7	0.05	4.14	646	12.38	2	-0.68
0.02	1212	10.16	6	0.45	1.00	940	9.42	7	0.06	4.30	1073	5.51	11	0.44
0.02	1199	10.28	6	0.38	1.05	1473	4.21	3	0.28	4.30	1123	5.26	11	0.61
0.02	1155	11.02	6	0.44	1.05	1573	3.77	3	0.38	4.30	1173	4.88	11	0.60
0.02	1178	10.63	6	0.42	1.27	771	12.38	2	-0.19	4.30	1073	5.45	11	0.38
0.02	1916	3.24	6	-0.17	1.33	802	11.39	7	-0.26	4.30	1123	5.17	11	0.52
0.02	1867	3.58	6	-0.11	1.33	841	10.58	7	-0.15	5.00	1423	2.75	3	0.03
0.02	1817	3.81	6	-0.18	1.33	823	10.96	7	-0.18	5.00	1273	3.37	3	-0.07
0.02	1768	4.15	6	-0.15	1.33	856	10.38	7	-0.02	5.00	1173	3.95	3	-0.10
0.02	1719	4.53	6	-0.10	1.33	876	9.88	7	-0.12	5.15	630	12.38	2	-0.44
0.02	1670	4.90	6	-0.08	1.33	895	9.60	7	-0.04	5.20	1073	3.97	8	-0.77
0.02	1108	9.60	10	-1.76	1.33	886	9.83	7	0.02	5.20	1473	1.67	8	-0.81
0.05	1058	11.50	10	0.13	1.55	1473	3.85	3	0.25	5.90	1073	4.26	3	-0.26
0.10	1058	10.30	10	-0.31	1.55	1573	3.45	3	0.34	5.90	1173	3.69	3	-0.11
0.10	1008	12.00	10	0.49	1.55	1673	2.98	3	0.30	5.90	1273	3.18	3	-0.04
0.10	1008	11.50	10	-0.01	1.80	1073	4.41	1	-2.10	5.94	642	12.38	2	0.60
0.15	1008	10.30	10	-0.71	1.85	820	10.10	7	-0.35	6.20	973	5.52	11	0.20
0.15	1008	10.60	10	-0.41	1.85	781	11.30	7	-0.07	6.20	1023	5.05	11	0.20
0.20	1058	10.10	10	0.32	1.85	850	9.69	7	-0.12	6.20	1073	4.57	11	0.14
0.30	1008	10.10	10	0.02	1.90	756	12.38	2	0.41	6.20	1073	4.63	11	0.20
0.30	1008	10.10	10	0.02	2.09	1473	3.61	3	0.28	6.20	1123	4.25	11	0.20
0.42	922	11.80	7	0.61	2.09	1573	3.20	3	0.33	6.66	1133	3.58	9	-0.29
0.42	977	10.76	7	0.62	2.10	1073	5.10	8	-1.16	6.66	1183	3.22	9	-0.33
0.42	960	11.40	7	0.97	2.10	1373	3.25	8	-0.62	6.66	1073	3.93	9	-0.37
0.50	1058	9.30	10	0.72	2.30	818	9.62	7	-0.33	6.66	1228	2.99	9	-0.31
0.50	1008	9.30	10	-0.04	2.30	775	10.49	7	-0.47	6.66	1086	3.86	9	-0.34
0.50	1008	9.60	10	0.26	2.58	1573	3.07	3	0.36	7.03	1173	3.46	3	-0.07
0.60	1058	8.90	10	0.57	3.00	788	9.93	7	0.00	7.03	1073	4.03	3	-0.17
0.60	908	11.30	10	0.44	3.00	822	9.12	7	-0.10	8.10	610	12.38	2	0.78
0.60	908	10.90	10	0.04	3.00	1073	4.80	8	-0.88	8.21	1173	3.12	3	-0.16
0.65	1008	9.10	10	0.15	3.22	1473	3.27	3	0.33	8.21	1073	3.68	3	-0.23
0.65	1008	9.20	10	0.25	3.22	1573	2.88	3	0.35	8.80	973	5.11	4	0.55
0.85	908	10.30	10	0.07	3.30	1473	2.25	8	-0.67	8.80	1073	4.14	4	0.36
0.85	908	9.80	10	-0.43	3.75	1573	2.54	3	0.14	8.80	1173	3.45	4	0.28
1.00	923	9.68	7	0.02	3.98	1373	3.10	9	-0.09	12.30	1123	3.20	8	0.33
1.00	884	10.48	7	0.08	3.98	1274	3.77	9	0.04	12.30	1223	2.40	8	-0.01

Note: References (Ref.) are as follows: 1 = Baker 1996, 2 = Dingwell 1996, 3 = Schulze et al. 1996, 4 = Burnham 1964, 5 = Dorfman et al. 1996, 6 = Hess et al. 1995, 7 = Dingwell et al. 1996, 8 = Persikov et al. 1990, 9 = Scaillet et al. 1996, 10 = Friedman et al. 1963, 11 = Shaw 1963.

mize the sum of the squared differences between the observed and predicted values of the dependent variable (viscosity). We tested the following simple functional forms:

model 1:

$$\log \eta = (a_1 + a_2 w) + (b_1 + b_2 w) / [T - (c_1 + c_2 w)] \quad (3)$$

model 2:

$$\log \eta = (a_1 + a_2 e^{-w}) + (b_1 + b_2 e^{-w}) / [T - (c_1 + c_2 e^{-w})] \quad (4)$$

TABLE 2. The fit parameters and statistics

Param.	Model 1	Model 2	Std. err.	Model 3	Std. err.	Model 4	Std. err.
a1	—	-2.23	0.76	-1.59	0.72	-3.54	0.66
a2	—	-3.89	0.58	-1.90	0.09	0.83	0.07
b1	—	5502	1007	2665	769	9601	1219
b2	—	14197	2023	7390	612	-2368	247
c1	—	259	51	337	46	196	55
c2	—	-352	85	-197	17	32	94
Std. dev.		0.580		0.560		0.46	

Note: Standard error (std. err.) = (std. dev.)/ $\sqrt{n}$; standard deviation (std. dev.) = $\sqrt{\sum(x_i - \bar{x})^2/(n-1)}$.

model 3:

$$\log \eta = (a_1 + a_2/w^{0.5}) + (b_1 + b_2/w^{0.5}) / [T - (c_1 + c_2/w^{0.5})] \quad (5)$$

model 4:

$$\log \eta = [a_1 + a_2 \ln(w)] + [b_1 + b_2 \ln(w)] / [T - [c_1 + c_2 \ln(w)]] \quad (6)$$

where a_1 , a_2 , b_1 , b_2 , c_1 , and c_2 are the fit parameters, η is the viscosity in Pascal seconds, w is the concentration of H₂O in weight percent, and T is the temperature in kelvins. The results of the fits are listed in Table 2 together with the standard deviation.

No convergence of the fit parameters was achieved using model 1. Model 4 yielded the best fit, generating the following equation for the calculation of the melt viscosities of metaluminous leucogranitic melts under crustal pressures with a standard deviation of 0.46 log₁₀ units:

$$\log \eta = [-3.545 + 0.833 \ln(w)] + [9601 - 2368 \ln(w)] / [T - [195.7 + 32.25 \ln(w)]] \quad (7)$$

where η is in Pascal seconds, w is H₂O concentration in weight percent, and T is in kelvins.

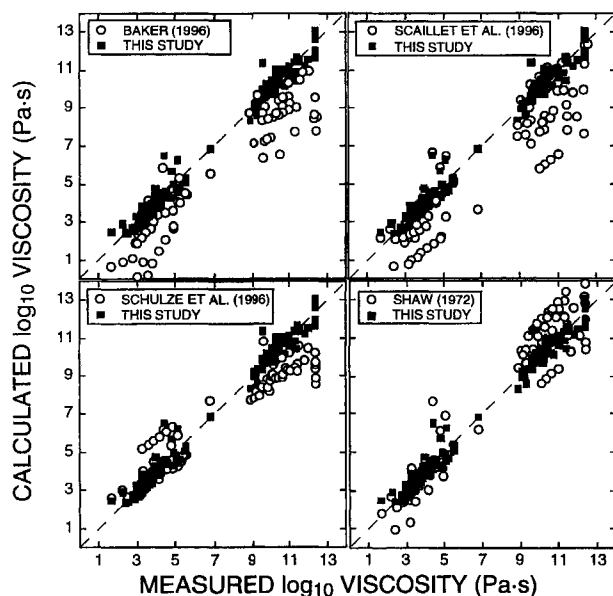


FIGURE 1. Comparison of previous viscosity models with the new model.

DISCUSSION OF THE FIT RESULTS

The non-Arrhenian temperature dependence incorporated into Equation 7 contrasts with all preceding methods for the calculation of hydrous granitic melt viscosities that have been based exclusively on Arrhenian relationships (Shaw 1972; Baker 1996; Scaillet et al. 1996; Schulze et al. 1996). The comparison of those methods with the present one (Fig. 1) clearly reveals the significant improvement in the quality of fit obtained by incorporating the very important non-Arrhenian temperature dependence of the hydrous melt viscosities.

All essential features of the temperature dependence and composition dependence of the viscosity of hydrous metaluminous granitic melts appear to be well reproduced by the new model. Figure 2 illustrates the predicted isothermal variations of viscosity with H_2O content (Fig. 2a) and the variation of viscosity with temperature at constant H_2O content (Fig. 2b). Several aspects of these calculations are noteworthy. First, the model is capable of predicting viscosities over the entire H_2O content range from 0 to 12.5 wt% within accuracy of 0.46 $\log_{10}$ units. The particularly sensitive region of very low H_2O contents is well described, and no apparent cross over of viscosity curves at high temperatures and low H_2O contents exists, at least up to 2500 K. Second, the degree of nonlinearity of the decrease in viscosity with H_2O content is a strong function of the temperature. At lower temperatures, very extreme decreases in viscosity are caused by the addition of a few tenths of 1 wt% H_2O . Third, slight variations in the compositions of the granitic, haplogranitic, and rhyolitic melts used in the various studies from which the input data were obtained [$SiO_2 = 73.2 - 78.6$ wt%; $K/Na = 0.56 - 0.80$; $CaO + MgO + FeO = 0 -$

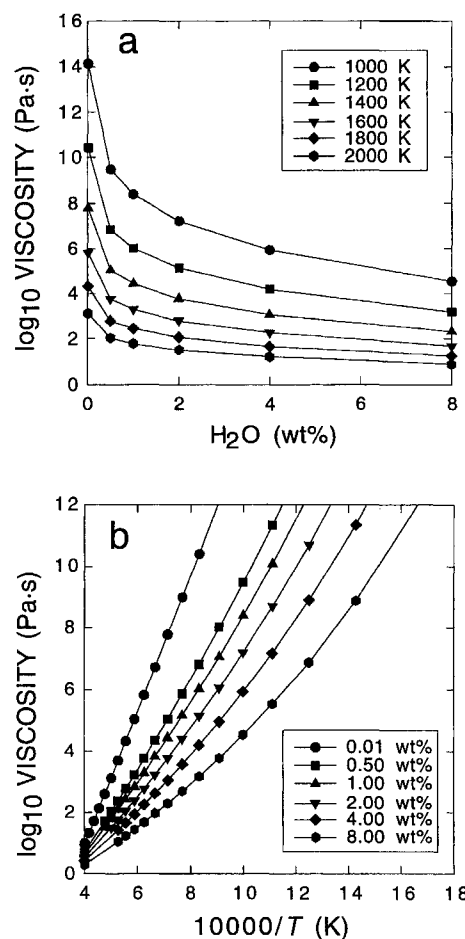


FIGURE 2. (a) Predicted isothermal variation of viscosity with H_2O concentration. (b) Variation of viscosity with temperature at constant H_2O concentration.

2.34 wt%; $(2Na + 2K + Mg + Ca + Fe^{2+})/(2Al + 2Fe^{3+}) = 0.93 - 1.02$] can be neglected for the purpose of hydrous melt-viscosity calculation. On the basis of the success of this parameterization, the neglect of pressure and speciation effects would appear to be justified. We recommend its use in cases that require estimation of the viscosity of hydrous, metaluminous leucogranite or calcalkaline rhyolitic melts. It should not be used for viscosity estimation if the anhydrous base composition differs significantly from those considered here.

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