

## Viscosity data for hydrous peraluminous granitic melts: Comparison with a metaluminous model

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### ABSTRACT

We performed 27 viscosity determinations on dry and water-bearing peraluminous haplogranitic melts. The dry melt compositions cover the range of normative corundum to be expected in peraluminous granitic melts in nature. The compositions are based on addition of  $\text{Al}_2\text{O}_3$  to a haplogranitic melt (HPG8) whose composition is near that of the projection of the 2 kbar  $\text{H}_2\text{O}$ -saturated minimum melt composition into the system  $\text{NaAlSi}_3\text{O}_8$ - $\text{KAlSi}_3\text{O}_8$ - $\text{SiO}_2$ . The  $\text{H}_2\text{O}$  contents of the hydrous melts were analyzed using Karl Fischer titration ranging from 1 to 3 wt%. The viscosity determinations were performed using a modified micropenetration method in the viscosity range of  $10^{10}$  to  $10^{11}$  Pa-s, at 1 atm pressure, and in the temperature ranges of 880–940 °C and 470–640 °C for the dry and wet melts, respectively. For the dry peraluminous melts in this high viscosity range, addition of the first few percent of normative corundum to a metaluminous granitic melt increases the viscosity, which remains nearly constant despite further addition of  $\text{Al}_2\text{O}_3$ . Thus a viscosity maximum is inferred for dry slightly peraluminous granitic melts. The hydrous melt viscosity data were compared with the recent calculational model of Hess and Dingwell (1996), which was based on and designed for metaluminous melt viscosities. That model is capable of describing the viscosities of hydrous peraluminous granitic melts within the uncertainties stated for its application in metaluminous melts.

### INTRODUCTION

The Newtonian shear viscosity of hydrous granitic melts is the most important material coefficient relating the stresses to which largely molten granitic magmas are subjected to the resulting deformation. Reliable modeling of the mass transfer involved in the physical evolution of granitic magmas and its consequences for chemical evolution of the residual melts of highly fractionated magmas requires a model that can accurately predict melt viscosity. Considerable attention has been focussed on experimental measurement of granitic and analog melt viscosities (e.g., Burnham 1963; Shaw 1963; Friedman et al. 1963; Persikov 1991; Dingwell et al. 1996; Schulze et al. 1996). Because metaluminous compositions dominate granitic terrains, experimental studies (listed above) have largely focussed on a restricted range of melt compositions. Multicomponent models based on the Arrhenian temperature dependence of viscosity well describe the high-temperature viscosities of both dry (Bottinga and Weill 1972) and wet (Shaw 1972; Persikov et al. 1990) silicate melts. However, such models have been recently shown to be inadequate in dealing with the high viscosity region for hydrous melts (Baker and Vaillancourt 1995; Baker 1996). This became quite apparent when new viscosity data were obtained by dilatometric means (Dingwell et al. 1996; Richet et al. 1996). Hess and Dingwell (1996) pro-

duced a non-Arrhenian model for calculation of hydrous granitic melt viscosities and demonstrated that the minor variations in the silica content or alkali/aluminum ratio of hydrous melts produced no noticeable influence on the viscosity at significant  $\text{H}_2\text{O}$  contents. Greater variations in the chemical composition of the dry melt might, however, be expected to produce significant variations in melt viscosity at moderate  $\text{H}_2\text{O}$  contents. In this context, perhaps the most important compositional variable influencing the viscosity of dry granitic melts is the alkali/aluminum ratio. The present study investigates the influence of  $\text{H}_2\text{O}$  on the viscosity of peraluminous granitic melts.

### METHODS

The dry melt compositions are based on the addition of  $\text{Al}_2\text{O}_3$  to a haplogranitic melt (HPG8) whose composition lies near the 2 kbar  $\text{pH}_2\text{O}$  ternary minimum melt composition projected into the system  $\text{NaAlSi}_3\text{O}_8$ - $\text{KAlSi}_3\text{O}_8$ - $\text{SiO}_2$ . The synthesis of the dry base glasses by means of 1 atm direct fusions of oxide-carbonate powder mixes was described by Holtz et al. (1992) and chemical analyses of the glasses by ICP-AES (Table 1) were presented by Knoche et al. (1995).

Hydrothermal syntheses of the  $\text{H}_2\text{O}$ -bearing samples were performed in rapid-quench TZM vessels using methods described in Dingwell et al. (1996). The hydrous samples were analyzed for  $\text{H}_2\text{O}$  content using Karl-Fischer titration (KFT) and methods described in Dingwell

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**TABLE 1.** Composition in weight percent  $\pm$  standard deviation

	HPG8	AI02	AI05
SiO <sub>2</sub>	78.6 $\pm$ 0.4	77.2 $\pm$ 0.4	75.4 $\pm$ 0.3
Al <sub>2</sub> O <sub>3</sub>	12.5 $\pm$ 0.2	13.9 $\pm$ 0.4	16.1 $\pm$ 0.2
Na <sub>2</sub> O	4.6 $\pm$ 0.3	4.5 $\pm$ 0.2	4.7 $\pm$ 0.2
K <sub>2</sub> O	4.2 $\pm$ 0.2	4.3 $\pm$ 0.4	4.3 $\pm$ 0.4
Total	99.9	99.9	100.5

et al. (1996). The homogeneity and the stability of the H<sub>2</sub>O contents of the samples was confirmed by FTIR spectroscopy before and after the viscometry using methods extensively described by Dingwell et al. (1996). The results of both KFT and FTIR spectroscopy are presented in Table 2.

The samples were prepared for micropenetration viscometry by using methods described in Hess et al. (1995) and Dingwell et al. (1996). The small amounts of hydrous samples available in this study necessitated the use of a modified micropenetration method discussed by Dingwell et al. (1996). The indentation vs. time traces were processed using the software described by Hess (1996). The errors associated with these methods have been discussed extensively by Hess et al. (1995) and Dingwell et al. (1996).

## RESULTS AND DISCUSSION

The results of the viscosity determinations for the dry and H<sub>2</sub>O-bearing peraluminous granitic melts are presented in Tables 3 and 4. Viscosities were successfully determined in the interval of 10<sup>10</sup> to 10<sup>11</sup>. The viscosity of the dry granitic starting compositions (Fig. 1) appears to increase significantly with the addition of excess alumina but then to remain constant with further addition of excess alumina. The activation energy of viscous flow remains unchanged. A smooth variation of viscosity as a function of the alkali/aluminum ratio requires a maximum in viscosity at a slightly peraluminous melt composition (between HPG8AI02 and HPG8AI05). Such a variation of viscosity has, in fact, been confirmed in this laboratory along stoichiometrically similar joins in the system Na<sub>2</sub>O-Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub> by Toplis et al. (1997a, 1997b). The variation in viscosity with the alkali/aluminum ratio is significant and cannot be accounted for by the model of Hess and Dingwell (1996), which is based on a metaluminous base composition. Nevertheless the deviations from the pre-

**TABLE 2.** H<sub>2</sub>O content determined by KFT and FTIR

HPG8 Sample no.	H <sub>2</sub> O (FTIR) (wt%)	AI02 Sample no.	H <sub>2</sub> O (KFT) (wt%)	AI05 Sample no.	H <sub>2</sub> O (KFT) (wt%)
528	0.42	632bis	1.07	636	1.17
526	0.99	632	2.18	634	2.03
527	1.33	631	2.54	633	2.77
144	1.85				
280	2.27				
300	3.00				

Note: Uncertainties are  $\pm$ 0.1%.

**TABLE 3.** Viscosity data for dry samples

T (°C)	log $\eta$ (Pa-s)	Sample no.
881.7	11.02	HPG8
905.0	10.63	HPG8
925.6	10.28	HPG8
938.8	10.16	HPG8
902.9	11.30	A102
924.9	11.08	A102
944.0	10.48	A102
961.2	10.14	A102
984.2	10.07	A102
979.7	10.12	A105
960.7	10.35	A105
942.9	10.70	A105
994.7	9.85	A105
939.8	10.58	A105

Note: 3 mm sample thickness. Temperatures accurate to  $\pm$ 0.5 °C. Viscosities are accurate to  $\pm$ 0.06 log units on the basis of DGG-1 standard glass determinations (see Dingwell et al. 1996).

dictions of the Hess and Dingwell (1996) model may be tolerable for some applications. This variation of viscosity among dry granitic melts should be investigated further.

Similarly, the activation energy of the hydrous melts (Fig. 2) does not vary significantly with composition, permitting a straightforward comparison of relative viscosities in this interval. The predictions of the hydrous melt viscosities using the Hess and Dingwell (1996) model are also shown in Figure 2. The results are satisfactory indicating that the model adequately accounts for the viscosities of hydrous peraluminous granitic melts commonly encountered in nature.

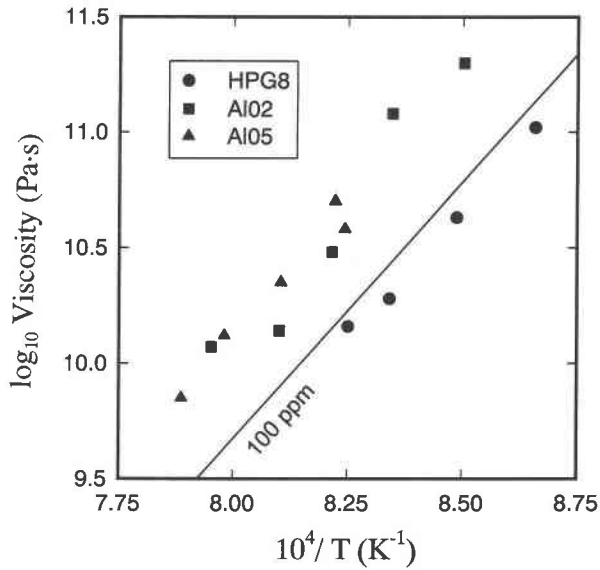
Comparison of the viscosities of the hydrous peraluminous melts with those of hydrous metaluminous melts (data from Dingwell et al. 1996) shows convergence of the melt viscosities with the addition of H<sub>2</sub>O (Fig. 3). This convergence allows the successful application of the Hess

**TABLE 4.** Viscosity data for hydrous samples

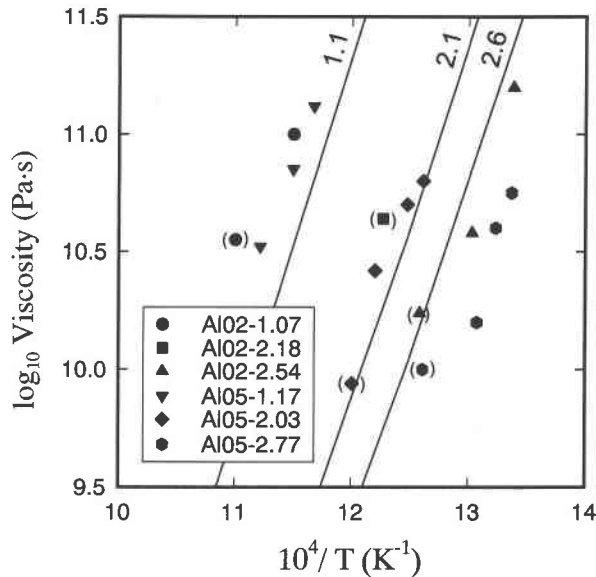
T (°C)	log $\eta$ (Pa-s)	Sample no.
596.2	11.00	632bis
635.6	10.55	632bis*
541.9	10.64	632*
494.1	10.58	631
521.4	10.19	631*
473.9	11.20	631
618.4	10.52	636
596.6	10.85	636
583.4	11.12	636
546.3	10.42	634
559.7	9.84	634*
528.2	10.70	634
519.5	10.80	634
491.6	10.14	633
482.4	10.60	633
519.7	9.91	633*
474.7	10.75	633

Note: 1 mm sample thickness; all viscosity data are corrected for sample size effect. Temperatures accurate to  $\pm$ 0.5 °C. Viscosities are accurate to  $\pm$ 0.06 log units on the basis of DGG-1 standard glass determinations (see Dingwell et al. 1996).

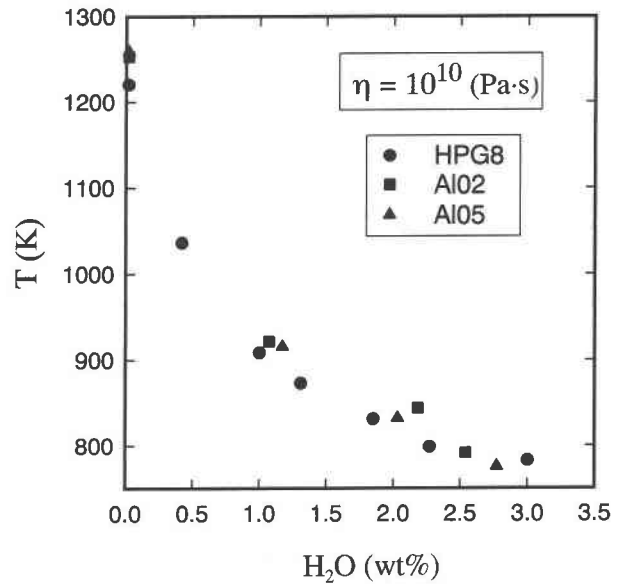
\* Loss of water determined by FTIR.



**FIGURE 1.** The viscosities of dry peraluminous granitic melts presented as a function of the reciprocal absolute temperature. The activation energy of viscous flow is constant and equal for all three compositions. Calculated values of the viscosity using the method of Hess and Dingwell (1996) are presented (solid line) for  $H_2O$  contents of 100 ppm (the approximate  $H_2O$  contents of the nominally dry melts).



**FIGURE 2.** The viscosities of hydrous peraluminous granitic melts presented as a function of the reciprocal absolute temperature. The activation energy of viscous flow is constant and equal for all three compositions. Calculated values of the viscosity using the method of Hess and Dingwell (1996) are presented for the equivalent  $H_2O$  contents. Symbols in parentheses denote viscosity measurements with considerable loss of  $H_2O$ .



**FIGURE 3.** The influence of  $H_2O$  on the viscosities of peraluminous vs. metaluminous granitic melts. The data are presented as the  $10^{10}$  Pa-s isokom (line of constant viscosity) to avoid extrapolative comparison. The isokom is generated from the interpolation of viscosity data using corrected viscosity data in addition to activation energies calculated using the Hess and Dingwell model (1996).

and Dingwell (1996) calculation to the viscosities of peraluminous melts. In conclusion we wish to emphasize that these new data on the influence of  $H_2O$  on the viscosity of moderately peraluminous granitic melts indicate that for  $H_2O$  contents greater than 1–2 wt%, the calculational model presented by Hess and Dingwell (1996) accurately reproduces the melt viscosities in the high viscosity range.

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