

A long-duration experiment on hydrous species geospeedometer and hydrous melt viscosity

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Abstract

A 2.4-year controlled-cooling-rate experiment was carried out to investigate the dependence of hydrous species concentrations in rhyolitic melt on cooling rate. The experiment allows us to obtain speciation for a cooling rate of 1.68×10^{-6} K/s, extending previous experimental data by two orders of magnitude. Furthermore, a viscosity as high as $10^{17.2}$ Pa s is inferred for this hydrous rhyolitic melt with 0.85 wt% total H₂O at 671 K. The results are applied to examine whether a geospeedometry model and four viscosity models may be extrapolated to slower cooling rates or lower temperatures. Two of the viscosity models and the geospeedometry model can be extrapolated by two orders of magnitude upwards in terms of viscosity or downwards in terms of cooling rate.

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1. INTRODUCTION

Cooling rate plays a critical role in the degree of degassing, welding, crystallinity, and other textures of volcanic rocks. Hence, accurate inference of cooling rate is important to the understanding of properties and reconstruction of history of volcanic rocks. Furthermore, cooling rate of pyroclasts may be used to infer the eruption column temperature (Xu and Zhang, 2002). For these purposes, a hydrous species geospeedometer has been developed and calibrated experimentally over a cooling rate range of 10^{-4} –100 K/s (Zhang et al., 1997b, 2000). The speedometer is based on the kinetics of the interconversion reaction between two hydrous species (H₂O molecules and OH groups). However, estimated cooling rates of natural volcanic glasses using the geospeedometer range from 10^{-8} to 100 K/s (Zhang et al., 2000; Xu and Zhang, 2002; Wallace et al., 2003), much larger than the experimental range. The extrapolation down to 10^{-8} K/s is uncertain because the geospeedometer is calibrated empirically and the formulation is complex. One motivation of this work is to carry

out slow-cooling experiments to extend the calibration of the hydrous species geospeedometer.

In addition to the above motivation, another goal is to test whether available viscosity models may be extrapolated to extreme viscosities. Magma viscosity plays an important role in volcanic eruption styles and dynamics, magma fragmentation and magma chamber processes. Numerous authors have conducted experiments to determine the viscosity of silicate melts and constructed viscosity models. Viscosity data usually cover two viscosity ranges: one from 0.1 to 10^5 Pa s, and the other from 10^8 to 10^{14} Pa s. In the low viscosity range, rotation viscometry (e.g., Dingwell, 1989; Whittington et al., 2001) and the falling sphere method (e.g., Kushiro et al., 1976; Schulze et al., 1996) are often applied, which cannot be easily extended to viscosities above 10^5 Pa s because rotation rate or falling speed would be too slow to measure. In the high viscosity range, parallel plate viscometry (e.g., Neuville and Richet, 1991; Schulze et al., 1999) and the micro-penetration method (Hess et al., 1995) are often applied, which cannot be easily extended to viscosity below 10^8 Pa s because the melt would flow too easily, nor viscosity above 10^{14} Pa s because deformation time scale would be too long. Furthermore, a liquid with viscosity between 10^5 and 10^8 Pa s often crystallizes easily, making it difficult to determine the viscosity.

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An Arrhenian relation (straight line in terms of $\ln \eta$ versus $1/T$ where η is viscosity and T is temperature) usually describes the viscosity data well in either the low-viscosity or the high-viscosity range. However, to simultaneously fit viscosity data in both ranges, it is necessary to use non-Arrhenian models. Four non-Arrhenian viscosity models are applicable to hydrous rhyolitic melts, of which three are specifically tailored for rhyolitic compositions (Hess and Dingwell, 1996; Zhang et al., 2003; Giordano et al., 2004). One significant difference between the three models occurs at H_2O content below 0.2 wt%: the model of Hess and Dingwell (1996) is constructed for hydrous rhyolitic melt and does not apply to completely dry melt. For example, as H_2O content decreases from 0.2 wt% to zero, calculated viscosity would first increase, reach a maximum, and then decrease to zero. Some might argue that the difference is not important because natural melts usually contain more than 0.1 wt% H_2O . The other two models (Zhang et al., 2003; Giordano et al., 2004) do not have this difficulty. Another difference between the three models is a small discrepancy in terms of calculated viscosity values for H_2O content between 0.2 and 8.0 wt%: usually about a factor of 2–3 (or 0.3–0.5 log units). In addition to viscosity models specifically constructed for rhyolites, Hui and Zhang (2007) developed a non-Arrhenian viscosity model for all anhydrous and hydrous natural silicate melts, which includes rhyolitic melt. Because non-Arrhenian models are curved in $\ln \eta$ versus $1/T$, it is of interest to examine whether such models can be extrapolated to even greater viscosities (lower temperatures). (Other viscosity models are either for dry melts only or for non-rhyolitic melts.) Because viscosity and cooling rate indicators (geospeedometry) are related (e.g., Dingwell and Webb, 1990; Zhang et al., 2003), controlled cooling rate experiments can also be used to examine the accuracy of viscosity models.

Here, we report a long-duration controlled-cooling-rate experiment on hydrous rhyolitic melt to determine hydrous species concentrations after very slow cooling (1 K/week = 1.7×10^{-6} K/s). The results are applied to test the hydrous species geospeedometry model and to assess whether current viscosity models can be extrapolated to extremely high viscosity at low temperature. Because inference of viscosity from controlled cooling rate experiments is not commonly used, the principles are introduced below.

2. PRINCIPLES FOR VISCOSITY INFERENCE

Viscosity inference in this work is not based on direct physical measurement of the flow rate of the melt, but based on the reaction kinetics of hydrous species interconversion in rhyolitic melt:



where H_2O_m is molecular H_2O (H_2O_t denotes total H_2O content), O is anhydrous bridging oxygen, and OH is a hydroxyl group, all dissolved in silicate melt. The equilibrium constant K for reaction (1) is expressed as:

$$K = \frac{[\text{OH}]^2}{[\text{H}_2\text{O}_m][\text{O}]}, \quad (2)$$

where brackets mean activity (approximated by mole fractions) on a single oxygen basis (Stolper, 1982; Zhang, 1999). The above equilibrium constant has been determined experimentally and is a function of temperature (Dingwell and Webb, 1990; Zhang et al., 1997a; Ihinger et al., 1999; Nowak and Behrens, 2001) with the approximate expression of $K = \exp(1.876 - 3110/T)$ (Zhang et al., 1997a). The experimentally determined $[\text{OH}]/[\text{H}_2\text{O}_m]$ ratio at equilibrium depends not only on temperature, but also on H_2O_t because the ratio is not the equilibrium constant. Concerning the experimental determination of the equilibrium constant K , some readers may remember two debates about possible quench effects on measured speciation. These have been resolved (see Dingwell and Webb, 1990; Zhang et al., 1995; Withers et al., 1999; Zhang, 1999; Nowak and Behrens, 2001) and the conclusion is that species concentrations would be altered during quench if the experimental temperature is high (such as >1100 K), but would not be altered during quench if the experimental temperature is intermediate (such as <900 K depending on quench rate and H_2O_t). In other words, for a given H_2O_t and temperature, species concentrations would be altered if cooling is slow but would be maintained if cooling is rapid. The freezing of OH and H_2O contents (i.e., no more interconversion) is attributed to the crossing from the liquid to the glass state (Dingwell and Webb, 1990; Romano et al., 1994). Our experiment exploits the effect of slow controlled cooling on speciation.

The species concentrations are obtained from infrared band intensities of the 452 mm^{-1} peak for OH and the 523 mm^{-1} peak for H_2O_m . The band intensities (linear absorbances) per unit sample thickness are denoted as A_{452} and A_{523} . For applications that do not require the calculation of species concentrations (such as calculation of the equilibrium temperature), infrared band intensities are used directly (e.g., Ihinger et al., 1999) to avoid the extra uncertainty in converting band intensities to species concentrations. In such applications, A_{523} is used as a proxy for H_2O_m concentration, A_{452} for OH concentration, $A_{523} + A_{452}$ for H_2O_t , and A_{452}^2/A_{523} for K in Eq. (2) because the relative variation in [O] is small.

The principles for viscosity inference were explained in Zhang et al. (2003) and are briefly outlined below. The viscosity of a melt at the glass transition temperature (T_g) is inversely proportional to the cooling rate (q) (Dingwell and Webb, 1990). The final concentrations of H_2O_m and OH depend on the cooling rate, meaning that the apparent equilibrium temperature (T_{ae}) of Reaction (1) calculated from the final species concentrations also depends on the cooling rate (Dingwell and Webb, 1990; Zhang, 1994). Because both T_g and T_{ae} are controlled by melt relaxation, it is assumed that the apparent equilibrium temperature of a cooled melt is equivalent to the glass transition temperature (Dingwell and Webb, 1990). Therefore, for a given cooling rate, the viscosity of the melt at T_{ae} is a constant divided by the cooling rate. Based on the above assumption and calibration using experimental data, Zhang et al. (2003) obtained the following:

$$\eta|_{atT_{ae}} = \eta|_{atT_g} = \frac{10^{11.45}}{q}, \quad (3)$$

where η is viscosity, q is cooling rate, and the unit of the constant $10^{11.45}$ is Pa K. The uncertainty of the constant $10^{11.45}$ was estimated to be about 0.20 log units (Zhang et al., 2003), $10^{11.45 \pm 0.20}$. The value of $10^{11.45}$ is similar to that of $10^{11.5}$ obtained by Toplis et al. (2001) from comparison of viscosity and heat capacity data in the glass transition range. The constant is not expected to depend on cooling rate q , and the compositional dependence of the constant has been found to be small, about 0.22 log units from 0 to 80 wt% SiO₂ (Sipp and Richet, 2002).

Based on the above, viscosity may be inferred as follows. First, a controlled cooling rate experiment is carried out. Because the cooling rate q is specified, the viscosity at T_{ae} is also specified by Eq. (3). After the experiment, hydrous species concentrations are measured by infrared spectroscopy, and T_{ae} is calculated as shown in detail below. Therefore, viscosity at this temperature is inferred. That is, in this method, the real measurement is to determine T_{ae} , whereas the viscosity value at this temperature is calculated from q using Eq. (3).

The calculation of T_{ae} of the quenched glass is based on previous experimental calibrations. A couple of calibrations have been carried out (e.g., one calibration in Zhang et al., 1997a; and three in Ihinger et al., 1999), and all are consistent within errors. For consistency with the geospeedometer of Zhang et al. (2000) and the viscosity models of Zhang et al. (2003) and Hui and Zhang (2007), the calibration of Ihinger et al. (1999) has been used to calculate T_{ae} of the quenched glass:

$$\ln \frac{A_{452}^2}{A_{523}} = 2.482 - \frac{2660 + 89.6A_{523} + 1082A_{452}}{T_{ae}}, \quad (4)$$

Table 1
Chemical composition of the starting material (on the anhydrous basis)

Oxide	SiO ₂	TiO ₂	Al ₂ O ₃	FeO	MgO	CaO	Na ₂ O	K ₂ O	Total
wt%	76.59	0.08	12.67	1.00	0.03	0.52	3.98	4.88	99.75

From Zhang et al. (1997a).

Table 2
FTIR data

Pt#	A_{523} (mm ⁻¹)	A_{452} (mm ⁻¹)	$\ln Q'$	H ₂ O _m (wt%)	OH (wt%)	H ₂ O _t (wt%)	$\ln K$	T_{ae} (K)
Natural KS sample (no heating)								
1	0.0408	0.1440	-0.68	0.173	0.682	0.855	-1.62	893
2	0.0410	0.1426	-0.70	0.174	0.674	0.848	-1.65	885
KS sample after heating and controlled cooling experiment								
1	0.0740	0.1172	-1.68	0.315	0.530	0.845	-2.72	671
2	0.0738	0.1176	-1.67	0.314	0.532	0.846	-2.71	672
3	0.0749	0.1179	-1.68	0.319	0.534	0.852	-2.72	671
4	0.0766	0.1187	-1.69	0.326	0.538	0.864	-2.73	670

Notes. A_{523} means the IR peak height at 523 mm⁻¹ per mm sample thickness; A_{452} = peak height at 452 mm⁻¹ per mm sample thickness; $Q' = A_{452}^2/A_{523}$; and $K = [\text{OH}]^2/([\text{H}_2\text{O}_m][\text{O}])$. Because the large KS piece before the experiment was too large to be directly analyzed by FTIR, "natural KS sample (no heating)" is a different and untreated piece of KS.

where A_{452} and A_{523} are in mm⁻¹ (per-mm thickness), and T_{ae} is in K. The above equation is based on the regular solution model for mixing of hydrous species. It has the advantage that IR band intensities are used directly so as to avoid extra uncertainties in converting band intensities to H₂O_m and OH concentrations.

3. EXPERIMENTS AND ANALYSES

A 2.4-year controlled-cooling-rate experiment was conducted in a 1-atm tube furnace following the procedure of Zhang et al. (1997b, 2000). Two natural samples (KS and bb5b) of hydrous rhyolitic glasses with different initial H₂O contents were cooled in this experiment but only one succeeded (KS). Due to very high reproducibility of kinetic data based on previous experiments (Zhang et al., 1997b, 2000), duplicates were not planned. The KS sample is from a big chunk of rhyolitic glass from Mono Craters, which was used before in studies of H₂O diffusion and reaction in rhyolitic melt (Zhang et al., 1991, 1995, 1997b, 2000). The crystallinity in the clear parts of the glass is low (of the order of a few percent) (Zhang et al., 1991). The composition is listed in Table 1. The H₂O content is about 0.85 wt% (Table 2). The sample is roughly cubic.

The furnace was first heated to and stabilized at 725 K. The KS sample (initial mass 2.7932 g; roughly 10 mm in each dimension) was placed into the tube furnace. The sample was cooled at 1 K/week by decreasing the set temperature by 1 K at 14:30 every Monday (or at 15:30 with daylight saving time). The temperature record is shown in Fig. 1. In any heating experiment, there is also diffusive loss of water from the surface (dehydration), which is not the purpose of our study. Hence, the initial experimental temperature must be low enough so that diffusion would not affect the central region of the sample. On the other hand, the initial experimental temperature must be high enough that

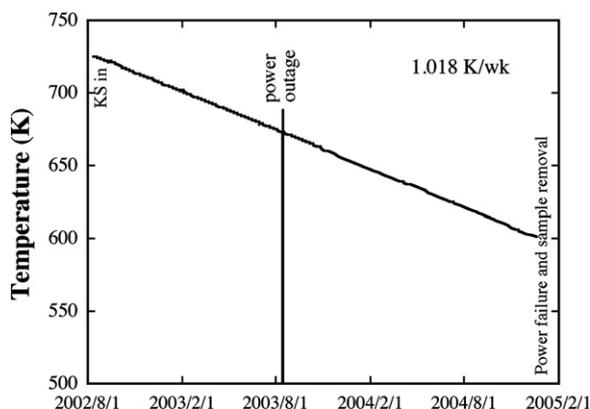


Fig. 1. Temperature history of the experiment. The horizontal axis gives time in year/month/day format. The time at which sample KS was placed into the furnace is marked. The times of two power outages are also marked.

hydrous species concentrations reach equilibrium during the initial phase of the experiment. Hence, we chose the initial temperature of each sample based on an estimation of diffusion distance (Zhang and Behrens, 2000) and the reaction time scale (Zhang et al., 1995, 1997b, 2000).

Unfortunately, two power outages occurred in the course of the long-duration experiment. One happened at roughly 1 year into the experiment ($t = 1.005$ years). After the power outage, the sample cooled all the way to room temperature. Based on kinetic considerations (Zhang et al., 1995, 1997b, 2000), the cooling rate was high enough that species concentrations were quenched without alteration during this cooling event. For example, rough calculation using the program of Zhang (1994) shows that the variation in T_{ae} is of the order 10^{-4} K during quench due to power outage. On the other hand, when the power was restored, overheating occurred (Fig. 1). Fortunately, we were present and took out the samples after the overheating was noticed. The samples were placed back into the furnace after the furnace temperature stabilized. The total overheating duration was about 3 min and the maximum overheating was by 14 K (from 674 to 688 K). Based on the kinetic data of Zhang et al. (1995, 1997b, 2000), at 688 K, the mean (or re-folding) reaction time for the hydrous species reaction to reach equilibrium in a hydrous rhyolitic glass containing 0.85 wt% H_2O_t is of the order 8000 min. In 3 min, $\ln K$ of the reaction is estimated to change by about 0.0004, or a change of T_{ae} by 0.06 K. Hence, overheating by 14 K for 3 min has an insignificant effect on the hydrous species concentrations. The second power outage also happened during daytime, after 2.384 years from the beginning of the experiment. Learning from the experience of the first power outage, the samples were removed right after the outage. Because the temperature was low enough (601 K) at the second power outage based on our estimation of the reaction kinetics, we decided to terminate the experiment (Our original plan included the addition of a third sample with much higher H_2O content when the temperature became low enough. Because of the frequent power outages, we decided to stop the experiment and acquire

the data, rather than risking another power outage that could happen during night and lead to large overheating when the power is restored.)

After the experiment, the KS sample was found to be in excellent shape. The after-experiment mass of the KS sample was 2.7879 g, meaning 0.19% mass loss. That is, on average, 0.19 wt% H_2O_t (or about 22% of the initial H_2O_t content) was lost during the experiment. Furthermore, the calculated mean diffusion distance ($\int D_{out} dt$)^{1/2} is 0.33 mm using H_2O diffusivities of Zhang and Behrens (2000), where D_{out} is diffusion-out diffusivity (Zhang et al., 1991; Wang et al., 1996). Because the mean diffusion distance is much smaller than the half-thickness (5 mm) of the sample, the central part of the sample is not affected by dehydration. We analyzed four different points by Fourier transform infrared spectroscopy (FTIR) on the central part of the sample.

The KS sample was cut near the center to produce a thin wafer of 2.5 mm thickness and 10.4 mm by 10.9 mm in area. The wafer was doubly polished. There is no noticeable increase in crystallinity. To avoid the dehydrated layer, four infrared spectra were taken from near-central points (within a circle of 6 mm diameter) in the wafer, at least 2 mm away from the dehydration surface. H_2O_m and OH species concentrations were obtained using the calibration of Zhang et al. (1997a). The data are used to check the validity of the geospeedometer of Zhang et al. (2000), and to estimate viscosity at this H_2O content. We did not look for dehydration gradients because resolving the dehydration profile would require the section to be thinned to less than 0.5 mm thickness.

Compared to sample KS, the second sample (bb5b) contained higher initial H_2O_t (1.2 wt%) and was placed into the furnace at a later time (at 696 K) because the reaction is expected to be more rapid. However, it failed because it cracked heavily, resulting in significant H_2O loss, even from the center of the sample.

4. RESULTS

The cooling rate obtained from the recorded temperature history is 1.68×10^{-6} K/s. Using Eq. (3), the viscosity is $10^{17.22}$ Pa s at the apparent equilibrium temperature. The FTIR data for the KS sample (four different points) after the experiment are listed in Table 2. H_2O_t content of the four points ranges from 0.845 to 0.864 wt%, a 2.2% relative variation. Because relative precision in FTIR is better than 1% relative, the variation in H_2O_t is real and reflects the initial heterogeneity, which was not homogenized during the experiment because the diffusive distance during the experiment is small (only 0.33 mm) compared to the size of the sample. The apparent equilibrium temperature ranges from 670 to 672 K. The precision in T_{ae} is high (with 2σ error of 2 K), but there is additional error in terms of the calibration accuracy (Ihinger et al., 1999). The T_{ae} after the experiment is about 220 K below the initial T_{ae} (890 K) of the natural sample, meaning that the extent of reaction during the cooling experiment was large. Coincidentally, the apparent equilibrium temperature (671 K) is very close to the temperature at the first power outage (674 K).

5. DISCUSSION

5.1. Comparison with previous geospeedometry model

The new experiment provides data on the relation between the final species concentrations and the cooling rate. Zhang et al. (1995, 1997b, 2000) investigated the reaction kinetics, and empirically quantified the experimental data relating species concentrations to cooling rates as a geospeedometer. Experimental data will be compared with the model of Zhang et al. (2000) because it supersedes that of Zhang et al. (1997b). The cooling rate range covered by previous experimental data is from 0.00017 to 100 K/s. Zhang et al. (2000) presented two methods for the calculation of cooling rates. Their preferred method, employed here, uses the following iteration:

$$\ln q = 8.7905 + 7.8096\xi - 3.4937\xi^2, \quad (5)$$

where $\xi = \ln(A_{452}/A_{523}) - f(x, y) + f(-1.7, y)$, where $f(x, y) = -5.4276 - 1.196x - 0.044536y - 0.023054xy + 3.7339 \times \exp(0.21361x + 0.030617y) - 0.37119 \times \exp(1.6299x)$ with $x = \ln(A_{452} + A_{523})$, and $y = \ln q$. Given A_{452} and A_{523} , the only unknown in the above iteration is q .

The geospeedometer has found applications in inferring cooling rates and temperature in volcanic eruption columns (Xu and Zhang, 2002), as well as cooling rates and thermal history of Plinian-fall and pyroclastic flow deposits in Bishop Tuff (Wallace et al., 2003). In these applications, sometimes the geospeedometer has to be extrapolated to cooling rate as low as 10^{-8} K/s (Wallace et al., 2003), four orders of magnitude below the experimental data of the calibration by Zhang et al. (2000). The uncertainty of the extrapolation was difficult to gauge but a rough and generous estimate of uncertainty by Wallace et al. (2003) was a factor of 50 at cooling rate of 10^{-8} K/s.

The new experiment with a cooling rate of 1.68×10^{-6} K/s (two orders of magnitude smaller than the previous minimum cooling rate) is applied to examine the reliability of the extrapolation of the geospeedometer of Zhang et al. (2000) to such low cooling rate. (To extend the cooling rate by another two orders of magnitude, down to 10^{-8} K/s using this approach would require a duration of hundreds of years and hence is impractical.) Note that in the discussion below and in Fig. 2, the IR peak heights are directly related to cooling rate, and that there are no intermediate parameters that may introduce additional uncertainties. The curve for 1.68×10^{-6} K/s (1 K/week) cooling rate is calculated from Zhang et al. (2000) and plotted on a $\ln(A_{452}/A_{523})$ versus $\ln(A_{452} + A_{523})$ diagram (Fig. 2), along with experimental data. The experimental points (squares) from this study lie on the calculated curve, meaning good agreement with the geospeedometry model of Zhang et al. (2000). The numerical comparison of calculated versus experimental cooling rates is shown in the last three columns of Table 3. The difference is ≤ 0.21 log units (similar to the uncertainty of prediction of experimental data in Zhang et al., 2000), and ≤ 0.38 log units if the alternative method proposed by the same authors is employed. Hence, extrapolating the geospeedometry model of Zhang et al. (2000) to a cooling rate of 10^{-6} K/s would have an er-

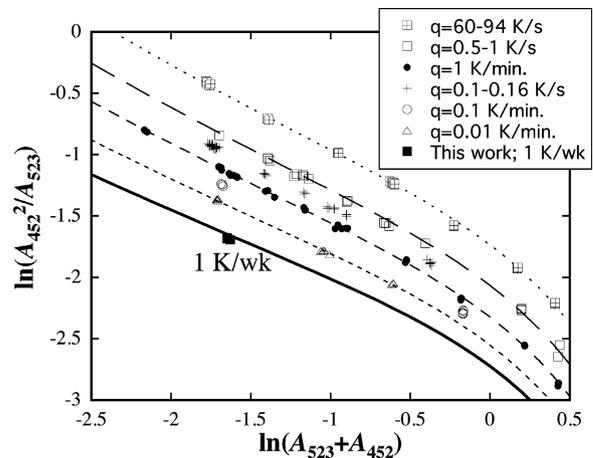


Fig. 2. The relation between species concentrations and cooling rate. The curves in the figure, from top down, are calculated using the model of Zhang et al. (2000) for cooling rates of 70, 1, 0.0167 K/s (1 K/min), 1.67×10^{-4} K/s (0.01 K/min), and 1.65×10^{-6} K/s (1 K/week). Points are data from Zhang et al. (1997b, 2000) and this work.

ror of at most 0.4 log units (a factor of 2.5). Based on these results, the preferred method in Zhang et al. (2000) (Eq. (5)) is recommended over the alternative, for future use.

Because extrapolation from 10^{-4} to 10^{-6} K/s does not result in additional error compared to prediction of cooling rate in the range covered by experiments of Zhang et al. (1997b, 2000), uncertainty of extrapolation to 10^{-8} K/s using Eq. (5) above is now estimated to be less than a factor of 5 (or 0.7 log unit), much smaller than the generous estimate of a factor of 50 in Wallace et al. (2003).

5.2. Comparison with previous viscosity models

The new, extremely high viscosity, data point may also be applied to examine whether previous viscosity models can be extrapolated to such high viscosities. We first compare three viscosity models of hydrous rhyolitic melts (Hess and Dingwell, 1996; Zhang et al., 2003; Giordano et al., 2004) with the new experimental data (Table 3). The difference between the calculated and experimental viscosity values is $\leq 0.2 \log \eta$ units for the models of Hess and Dingwell (1996) and Zhang et al. (2003), smaller than the stated 2σ uncertainty of either model (0.92 for Hess and Dingwell, 1996, and 0.36 log units for Zhang et al., 2003). Hence, these two non-Arrhenian models for rhyolitic melts can be extrapolated to viscosities at least as high as $10^{17.2}$ Pa s. However, the calculated viscosity of the more recent model of Giordano et al. (2004) is 1.7 log units smaller than the experimental data. Hence, extrapolation of the model of Giordano et al. (2004) to viscosity beyond their data range of 10^2 to 10^{12} Pa s is not recommended. For example, even though Giordano et al. (2004) stated that their model can be used from 648 to 1673 K, our data at 671 K cannot be predicted by their model.

The new data are also employed to examine whether the model of Hui and Zhang (2007) for all natural silicate melts may be extrapolated to lower temperatures. The compari-

Table 3
Comparison of experimental and calculated viscosity and cooling rate

Pt#	H ₂ O _t (wt%)	T _{ac} (K)	log η _{exp}	log η _{calc} HD1996	log η _{calc} Z2003	log η _{calc} G2004	log η _{calc} HZ2007	log q _{exp}	log q _{calc} Z2000 (1)	log q _{calc} Z2000 (2)
1	0.845	671	17.22	17.13	17.42	15.51	16.26	-5.77	-5.98	-6.16
2	0.846	672	17.22	17.06	17.34	15.46	16.20	-5.77	-5.90	-6.06
3	0.852	671	17.22	17.10	17.38	15.47	16.24	-5.77	-5.95	-6.11
4	0.864	670	17.22	17.12	17.39	15.45	16.26	-5.77	-5.96	-6.13

Notes. Unit of viscosity is Pa s; unit of cooling rate is K/s. References: HD1996, Hess and Dingwell (1996); Z2000 (1), the preferred method in Zhang et al. (2000); Z2000 (2), the alternative method in Zhang et al. (2000); Z2003, Zhang et al. (2003); G2004, Giordano et al. (2004); HZ2007, Hui and Zhang (2007).

son is also shown in Table 3. The calculated viscosity values are about one log unit smaller than the experimental data, implying that the model of Hui and Zhang (2007) cannot be reliably extrapolated to high viscosities. Hence, as pointed out by Hui and Zhang (2007), the model should only be applied for interpolation (data used in their model covered viscosity 10⁻¹ to 10¹⁵ Pa s) but not for extrapolation. There is clearly room to further improve the general viscosity model for all natural silicate melts.

In summary, results from this long-duration experiment provide critical data to evaluate previous models, and to demonstrate that the geospeedometry model of Zhang et al. (2000) can be extrapolated to cooling rates as low as 10⁻⁶ K/s. It is also found that the viscosity models of Hess and Dingwell (1996) and Zhang et al. (2003) can be extrapolated to viscosities up to 10^{17.2} Pa s. However, the viscosity models of Giordano et al. (2004) and Hui and Zhang (2007) cannot be extrapolated.

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