Viscosity measurement of subliquidus magmas: 1707 basalt of Fuji volcano

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We have installed a new furnace equipped with a rotational viscometer for use in viscometry at high temperature (one atmosphere) under controlled oxygen fugacities. This furnace allows sampling at the time of the viscosity measurement. Utilizing this furnace, we examined the relationship between textural parameters (crystallinity and the apparent aspect ratio of crystals) and viscosity during cooling of a basaltic magma. The 1707 basalt from Fuji volcano was used for the experiment and the result shows a drastic increase in viscosity from the liquidus temperature (1230°C; $\eta = 52$ Pa s), through 1170°C (crystallinity = 11%; $\eta = 350$ Pa s), to 1130°C (crystallinity = 23%; η = 1950 Pa s). The increase in viscosity during cooling is affected by both the increase of the crystallinity and the change of melt composition as a result of crystallization. For the basalt from Fuji volcano, crystallization of thin tabular plagioclase enhanced the strong effect of crystal entanglement, whereas the changes of melt compositions did not affect the viscosity because of the increase of ferromagnesian components and the decrease of feldspathic component in the residual liquid during crystallization. The effect of the presence of crystals on the viscosity is much larger than that expected from the Einstein-Roscoe equation with Marsh's constant ($\eta = \eta_0 (1 - \varphi/0.6)^{-2.5}$), where φ denotes the crystal fraction. The additional correction of Simha on the solid shape parameter slightly overestimates the increase in relative viscosity in the presence of thin platy crystals. A shear rate dependence of the viscosity was observed at lower temperature crystal-rich conditions.

Keywords: Basalt, Crystallinity, Einstein-Roscoe equation, Plagioclase, Viscosity

INTRODUCTION

The viscosity of magma is one of the most critical parameters of the various magmatic processes, such as magma segregation, magma uprise, crystallization differentiation, magma degassing and the mode of volcanic eruptions (Dingwell, 1998; Spera, 2000). Natural magmas almost always contain some crystals, where the bulk viscosity is often estimated through simple equations such as the Einstein-Roscoe equation (Marsh, 1981). Pinkerton and Stevenson (1992), among others, reviewed the methods of correction to obtain the bulk viscosity for melt + crystal systems and indicated that crystal size distribution and the aspect ratio of crystals, in addition to the proportion of crystalline matter, greatly affect the bulk rheological properties of magmas. Previous experimental studies of subliquidus viscometry on basaltic magmas mostly pay little attention to the composition and textures of the crystals and melts (Shaw, 1969; Ryerson et al., 1988; Pinkerton

and Norton, 1995). In the present study, we installed a new rotational viscometer to operate at high temperature and one atmosphere under controlled oxygen fugacity, which allows temporal sampling of the melt + crystal at the time of the viscosity measurement. Utilizing the viscometer, we examined the rheological behavior of highalumina basalt from Fuji volcano to obtain the relationship between textural parameters (crystallinity and the apparent aspect ratio of crystals) and viscosity during cooling of the system. The result shows a drastic increase of viscosity from liquidus temperature to subliquidus temperature, along with an increase of the percentage of crystals to 23% by volume. The phase compositions were obtained and viscosities were calculated utilizing the equations of Shaw (1972) and Einstein-Roscoe with a parameter of Marsh (1981). The calculated viscosities were compared with the observed ones to reveal discrepancies due to the application of the Einstein-Roscoe equation for systems with tabular plagioclase (in the present experiments). The effect of crystal shape is also examined with an equation of Simha (1940).

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EXPERIMENTAL METHODS

We used a newly installed atmosphere-controlled furnace for viscometry. The furnace consists of vertical double spiral heating element of SiC, which is separated from the sample crucible by an alumina tube of 60 mm inner diameter. The temperature of the furnace was monitored and controlled by a thermocouple outside the heating element, that is calibrated against the temperature within the sample crucible. The temperature variation along the length of the sample location is within 5°C. The furnace atmosphere was controlled by a mixed gas of H₂ (10 ml/min) and CO₂ (400 ml/min), yielding a redox condition near the Ni-NiO buffer conditions at the temperature of the experiment (Huebner, 1982). The mixed gas was introduced at the bottom of the alumina tube and flew upward at a rate of ca. 2-3 mm/s in the tube. The top of the alumina tube was closed by ceramic plates, which are removed at the time when the rotating rod used in viscometry is being inserted, are roughly closed during the viscometry and opened again after the viscometry to pull out the rod for sampling of the basalt. The basalt sample at the tip of the rod was quenched in air and made into polished thin sections for electron probe microanalyses and textural analyses.

The viscometry was carried out using an electromagnetic rotational viscometer (Model TV-10U, Toki Sangyo Co. Ltd., Japan). The sample was placed in a Pt crucible, made especially for viscometry, with a 30 mm inner diam-



Figure 1. Velocity and strain-rate profiles in the crucible for the rotational viscometer at 10 rpm, calculated after the equation of Landau and Lifschitz (1970). Note the large gradient of the strain rate in the rotational viscometry of the present experiment.

eter, height of 60 mm, and a wall thickness of 1 mm. The depth of the basaltic sample was ca. 44 mm at the start of the experiment, decreasing to 38.5 mm after the run because of repeated viscosity measurement and sampling during the experiment. The depth of the sample at the time of each measurement was obtained by linear interpolation between the depths of the sample before and after the experiment. For rotational viscometry we used an alumina rod with a diameter of 6 mm. As will be described later, the alumina rod did not melt or react with the basaltic melt during measurement and sampling. One of the features of rotational viscometry is that the shear rate is not uniform within the sample. Figure 1 shows the velocity and strain rate for conditions of 10 rpm (rotations per minute), calculated after the equation of Landau and Lifschitz (1970). The strain rate varies from 0.09 s^{-1} to 1.13 s⁻¹, a more than 1 order of magnitude variation of the strain rate in the rotational viscometry. Because of the non-uniform shear rate in these experiments, the viscosity obtained may be dependent on the size of rod and crucible for the non-Newtonian melt + crystal. The viscometer runs at constant rotation rate, and viscosity is obtained from the following equation, $M = 4\pi\eta\Omega R_1^2R_2^2/(R_2^2 - R_1^2)$, where M is the moment, η is viscosity in Pa s, Ω is angular velocity, and R1 and R2 are the radii of rod and crucible, respectively. To account for the end effects of the rod and the depth of the basalt, we calibrated the system with standard viscosity oils (JS160000 and JS52000) against variations of the distance between the rod end and the bottom of the crucible, and of the depth of the sample. The standard oils of JS160000 and JS52000 have viscosi-

Run schedule: R-3



Time (not to scale)

Figure 2. Temperature-time schedule of the viscosity measurement R-3 of the present experiment. Viscometry and sampling were carried out after keeping the sample for ca. 1-3 days at constant temperature. The viscosity measurements at each temperature are designated R-3-1 at 1230°C, R-3-2 at 1210°C, etc. After the R-3-1 measurement and sampling, a small amount of the basalt powder was inserted into the crucible to facilitate crystal nucleation.



Figure 3. Viscosity variation during R-3-3 measurement at 1190°C. (A) shows all the session results, and (B) is enlarged to show fine details. In session 1, we recorded a marked decrease of viscosity in the first several minutes at 5 rpm. From session 1 to 4, the rotation rate is increased from 5 rpm to 50 rpm, each recording the temporal change of viscosity. From session 4 to 7, the rotation rate is decreased from 50 rpm to 5 rpm, where viscosity is almost constant in each session. These data are used for discussion and Figures 4, 5 and 8. A break of ca. 15-30 s occurred while the rate of rotation was changed between sessions.

ties of ca. 148 and 42 Pa s at 20°C. respectively, and show Newtonian behavior (no dependence on the shear rate).

Figure 2 shows the temperature-time schedule of the experiment. The sample was first melted at 1230°C for ca. 70 hours and a viscosity measurement and sampling were carried out. The sample was then cooled by 20°C within 5 minutes, and kept for another ca. 20 hours. The sequence of stepwise cooling, holding at constant temperature for ca. 20 hours, viscosity measurement, and sampling was repeated five times down to 1130°C. At each temperature, viscosity measurements were carried out several times. Figure 3 illustrates the record of measurement at 1190°C, which consists of seven sequential sessions. Each session records the variation of viscosity at a constant rotation rate of the rod. Usually, the first session records a marked decrease of viscosity, which may be due to shear heating, the tendency for parallel alignment of plagioclase plates, or the tendency for melt to climb the rod (Shaw, 1969; Spera et al., 1988; Ryerson et al., 1988). The significance of the temporal variations of viscosity in each session will be discussed later. After each measurement, the alumina rod was pulled out of the sample, quenched in air, and the basalt made into polished thin sections for electron probe microanalyses. This allowed determination of the melt and crystal compositions and textural features. Specifically after the viscosity measurement and sampling at 1230°C, we added a small amount of the basalt powder to the crucible to facilitate nucleation of crystals in the following measurements. The basaltic sample is composed of scoria fragments with phenocrysts of olivine (0.9 vol%) and plagioclase (0.4%) in the matrix of fine microlites of plagioclase, pyroxene, titanomagnetite and tachylyte glass. Without the seeding of nuclei to the melt, delays in nucleation may occur for several tens of centidegrees below the liquidus temperature, as experimentally demonstrated by Gibb (1974).

EXPERIMENTAL RESULTS

Viscosity measurements

Viscosity measurements and sampling were carried out at the end of each temperature step. Figure 2 illustrates the temperature-time schedule of the experiment R-3. Viscosity measurements at 1230°C, 1210°C, 1190°C, 1170°C, 1150°C and 1130°C are called R-3-1, R-3-2, R-3-3, R-3-4, R-3-5 and R-3-6, respectively. Each of the measurement consists of a sequence of six to eight sessions, which record the change of viscosity at constant rotational rate. The rotational rate is firstly increased from session to session and then decreased to obtain stable viscosity data. Figure 3 illustrates the seven sessions of the R-3-3 measurement. The first session (5 rpm) records a marked decrease of viscosity for the first several minutes.



Figure 4. Shear rate dependence of the viscosity of the basaltic magma of Fuji volcano. At higher temperature with little crystals (1210°C), the viscosity is almost constant against variation of the shear rate (Newtonian behavior) whereas at lower temperature, the shear rate dependence becomes pronounced because of a higher crystal concentration.

The second and the third sessions (10 rpm and 20 rpm, respectively) also show a decrease of viscosity within the session, and the fourth session (at 50 rpm, the highest rotation rate) records the lowest viscosity. The rotation rate is decreased from the fourth session (50 rpm) to the seventh session (5 rpm) and the viscosity is almost constant within each of the sessions. Viscosity values increased from the fourth to the seventh sessions, which are employed for the following data presentation.

Figure 4 demonstrates the shear rate dependence of the viscosity. At 1210°C, where there is only 1.3 vol% plagioclase, the viscosity is almost constant against the variation of the rotation rate from 5 to 50 rpm, whereas at lower temperatures, where there is a higher content of crystals, the viscosity apparently decreases as the rotation rate increases. At 1170°C, a factor of 1.23 increase of the viscosity was observed from 10 rpm to 1 rpm, and at 1130°C, a factor of 1.62 increase of the viscosity was observed from 6 rpm to 0.6 rpm of the rotation rate.

Figure 5 shows the viscosity versus reciprocal temperature (K) of the Fuji basalt in the present experiment (solid circle). We used the viscosity values at the lowest shear rate for each measurement. The viscosity varies from 52 (= $10^{1.72}$) Pa s at 1230°C (crystal-free) through 350 (= $10^{2.54}$) Pa s at 1170°C (11 vol% crystal), to 1950 (= $10^{3.29}$) Pa s at 1130°C (23 vol% crystal). An increase in viscosity by a factor of 37.5 was observed within 100°C of the liquidus temperature of the Fuji basalt. Fitting of



Figure 5. Viscosity coefficient versus inverse temperature (K⁻¹) relations. The measured values (solid circles) are approximated by an equation: = $4.89*10^{-23} \exp(82700/T)$ with R = 0.99. Open squares represents the viscosity of melt calculated for the melt composition at each temperature after the equation of Shaw (1972). Open circles illustrate the temperature effect on the viscosity of melt, calculated for the bulk composition at each temperature after the equation of Shaw (1972). Subliquidus viscosity of Hawaiian picritic tholeiite (crosses) is after the experiments of Ryerson et al. (1988).

the data with an exponential equation gave the following, with a correlation coefficient of 0.99; $\eta = 4.89 \times 10^{-23} \exp(82700/\text{T})$, where T denotes absolute temperature, and η in Pa s.

Phase composition and mode

Figure 6 shows the back-scattered electron images of the products of the R-3 experiment. The pictures show only the glass of ca. 1 mm thick attached to the alumina rod. Plagioclase appears as a liquidus at 1227°C (Hara, 1991), followed by olivine at 1130°C. The increase of the amount of plagioclase is slow initially, from 0 vol% at 1230°C to 3.6% at 1190°C, but becomes more rapid from 11.0 vol% at 1170°C to 23.2% at 1130°C. The maximum size of plagioclase crystals is apparently similar in all the runs, and number density of crystals increases at lower temperatures. Plagioclase crystals have a thin tabular form with a an apparent length/width ratio of 12.9 ± 1.8 . They often show skeletal texture. Thin tabular plagioclase is generally aligned parallel to the tangential direction of the rotation of the rod. The plagioclase section parallel to the rotation direction shows a rhombic form with nearly equal width ratio and an intersecting angle of 128-130°. The angle is somewhat larger than the usually reported β angle of ca. 116° for plagioclase (Deer et al., 1992).

R-3-1; 1230°C



R-3-3; 1190°C



R-3-5; 1150°C



R-3-2; 1210°C



R-3-4; 1170°C



R-3-6; 1130°C



Figure 6. Back-scattered electron images of the charges of R-3 experiment. The scale bars represent 200 μ m. The bright area represents basaltic glass, the dark elongated (sometimes skeletal) rectangles are plagioclase and the slightly dark equant crystals are olivine (only in R-3-6). Dark areas with dots of darker pits are the alumina rods used for the viscometry and sampling.

Table 1. Phase chemistry of the run products and

Phase	Temp.	SiO ₂	TiO ₂	Al_2O_3	FeO	MnO	MgO	CaO	Na ₂ O	K ₂ O	P_2O_5	Total
n	°C	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%	wt%
		s.d.	s.d.	s.d.	s.d.	s.d.	s.d.	s.d.	s.d.	s.d.	s.d.	s.d.
R3-1-gl	1230	52.51	1.39	16.78	10.10	0.18	5.20	9.69	2.60	1.03	0.13	99.61
5		0.34	0.03	0.10	0.19	0.03	0.16	0.20	0.07	0.23	0.02	0.27
R3-2-gl	1210	52.38	1.38	16.72	10.27	0.18	5.26	9.60	2.58	1.06	0.12	99.55
5		0.20	0.04	0.09	0.19	0.02	0.14	0.13	0.07	0.18	0.01	0.36
R3-2-pl		51.49	0.08	29.99	0.93	0.01	0.20	14.67	2.97	0.15	0.00	100.49
6		0.53	0.03	0.29	0.08	0.01	0.02	0.32	0.19	0.02	0.00	0.35
R3-3-gl	1190	52.21	1.40	16.05	10.33	0.20	5.33	9.47	2.66	0.96	0.14	98.74
5		0.13	0.03	0.14	0.31	0.04	0.22	0.11	0.09	0.09	0.03	0.57
R3-3-pl		51.36	0.06	29.94	0.96	0.00	0.21	14.53	2.96	0.14	0.00	100.16
7		0.22	0.02	0.21	0.05	0.00	0.02	0.19	0.12	0.01	0.00	0.32
R3-4-gl	1170	52.03	1.51	14.99	11.09	0.19	5.85	9.10	2.55	0.99	0.13	98.44
6		0.15	0.02	0.13	0.60	0.01	0.07	0.08	0.08	0.05	0.03	0.73
R3-4-pl		51.57	0.07	29.59	0.95	0.01	0.22	14.29	3.04	0.16	0.00	99.90
6		0.30	0.02	0.40	0.13	0.01	0.02	0.30	0.14	0.01	0.00	0.28
R3-5-gl	1150	52.36	1.67	14.19	12.25	0.23	6.32	8.83	2.49	1.09	0.19	99.62
6		0.17	0.08	0.05	0.09	0.02	0.08	0.06	0.06	0.03	0.02	0.10
R3-5-pl		51.26	0.07	29.88	0.91	0.02	0.21	14.63	2.94	0.14	0.00	100.05
6		0.45	0.02	0.51	0.09	0.01	0.01	0.31	0.14	0.02	0.00	0.24
R3-6-gl	1130	52.59	1.75	13.50	12.46	0.24	5.76	8.73	2.41	1.24	0.19	98.86
6		0.21	0.04	0.11	0.60	0.03	0.09	0.06	0.10	0.03	0.03	0.84
R3-6-pl		51.81	0.07	29.35	1.18	0.01	0.24	14.09	3.17	0.19	0.00	100.11
9		0.62	0.02	0.56	0.15	0.01	0.04	0.45	0.22	0.03	0.00	0.53
R3-6-ol		39.84	0.05	0.27	20.44	0.35	40.27	0.34	0.08	0.03	0.05	101.72
8		0.48	0.04	0.33	0.33	0.03	0.89	0.11	0.09	0.04	0.06	0.55

^{\$} Least square calculation from the phase compositions.

^{\$\$} Calculated from the equation of Shaw (1972) for each melt at the experimental temperatures.

Analytical methods: The electron microprobe analyses were made at 15 kV and 12 nA. Full ZAF corrections were applied to the background-corrected count ratios. The counting time was 20 s at peak position with 10 s for background on both sides of the peak, except for Na, for which 4 and 2 s are counted at peak and background wavelength. The crystals were analyzed using a focused beam but for glass analyses a beam diameter of ca. $10 \,\mu$ m was used. Standards used for quantitative analyses were pure synthetic oxides for Si, Ti, Al, Fe, Mn, Mg; CaSiO₃ for Ca; NaCl for Na; and natural adularia for K.

Table 1 shows the phase chemistry of the run products, together with the modal contents calculated by the least squares method and the results of viscometry. Figure 7 illustrates the compositional variations of the glass from the run products. The glass compositions in any one charge are mostly uniform, even very near to the alumina rod, indicating that there is a negligible effect of contamination from the rod. As the temperature decreases, the SiO₂ content of the glass is almost constant, varying from 52.5 wt% at 1230°C to 52.6% at 1130°C, whereas Al₂O₃ decreases from 16.8% at 1230°C to 13.5% at 1130°C. The CaO content also decreases from 9.7 to 8.7 wt% while FeO* increases from 10.1 to 12.5 wt% from 1230°C to 1130°C. MgO firstly increases from 5.2 to 6.3 wt%, from 1230°C to 1150°C, then decreases to 5.8% at 1130°C due to the crystallization of a small amount of olivine in the 1130°C charge. The average plagioclase composition shows only limited change: $\#Ca (= Ca/(Ca + Na) mol) = 0.73 \text{ to } 0.71 \text{ from } 1210^{\circ}\text{C} \text{ to } 1130^{\circ}\text{C}$. The value of #Mg (= Mg/(Mg + Fe) mol) in olivine at 1130°C is 0.78.

DISCUSSION

Basic features of the rotation viscometry at subliquidus temperatures

As illustrated in Figures 3 and 4, the subliquidus basalt showed viscosity variations dependent on the elapsed time and on the rotation rate. These are discussed in this section and the next section. The time-dependent viscosity variations shown in Figure 3 may be called thixotropy. This phenomenon may be relevant to the four problems pertinent to the present rotational viscometry: (1) shear heating, (2) concentration of crystals toward the outer part

the result of viscometry

	#Ca or	Mode	Mode	Viscosity	Viscosity	
	#Mg*	wt% ^{\$}	vol fraction	bulk	melt ^{\$\$}	
_				Pa s	Pa s	
	0.48*	100.00	1.00	52	62	
				(20 rpm)		
	0.48*	98.70	0.99	73	74	
	0 72	1 20	0.01	(10 rpm)		
	0.73	1.30	0.01			
	0.40*	0(20	0.07	117	02	
	0.48*	96.30	0.96	(5 rnm)	92	
	0.73	3.70	0.04	(5 (piii)		
	0.48*	88.80	0.89	317	93	
				(2.5 rpm)		
	0.72	11.20	0.11			
	0.48*	82.40	0.83	909	91	
	0.73	17.60	0.17	(0.6 rpm)		
	0.75	17.00	0.17			
	0.45*	76.20	0.77	1050	138	
	0.43*	70.20	0.77	(0.3 rbm)	130	
	0.71	22.00	0.22	(
	0.78*	1.80	0.01			

of the container due to flow differentiation, (3) normal stress causes the basalt to rise up the rotating rod and (4) parallel alignment of plagioclase lath. Shear heating may increase the temperature, which in turn decreases the viscosity during rotational viscometry. The amount of shear heating has been evaluated by an equation discussed by Spera et al. (1988). The temperature increase was estimated to be less than 1°C increase and may be ignored in the present experiments.

Because of the variation of shear rate in the radial direction of rotational viscometry, crystals tend to move toward areas of low gradient of shear rate; i.e., the outer part of the crucible. Komar (1972) discussed the flow differentiation in dikes, and suggested that the grain dispersive force active on the solids in a gradient of shear rate tends to bring the larger grains toward locations with a lower gradient of shear rate. R-3-6 in Figure 6 shows that



Figure 7. Melt composition-temperature relations. From 1230°C to 1150°C, Al₂O₃ and CaO show a monotonous decrease and FeO and MgO show a monotonous increase in concentration because of the crystallization of plagioclase alone. At 1130°C, crystallization of additional olivine caused a decrease of MgO in the melt.

plagioclase tends to be sparse near the rotating rod, in accord with the prediction that the solid tends to move away from the rotating rod. Although we have no quantitative means to evaluate the effect of flow differentiation on the temporal variation of viscosity of the subliquidus magmas, the apparent similarity of the modal content of crystals in Figure 6, representing basalt very near to the rod, with the calculated mode (Table 1) suggests that the effect of flow differentiation may not be large enough to fully explain marked decrease of viscosity in the first session of the measurement.

Shaw (1969) and Spera et al. (1988) discussed the normal stress on the melt surrounding the rotating rod. This was actually observed when we performed calibration of the crucible/rod system with standard oils, with the result that oils tends to climb up the rod at higher rotation rate (> 50 rpm). Most of the measurements on basalt were carried out at a lower rate so that this effect was minimized.

As noted in Figure 6, plagioclase crystals have a thin tabular form and tend to show parallel alignment to the rotational direction. This is caused by the gradient of the shear rate around the rotating rod, like imbrication of tabular fragments in sediments. This effect may weaken the interaction of the plagioclase and decrease the apparent viscosity of the magma (Manga, 1999).

These characteristics pertinent to the rotational viscometry of the present study may affect the apparent time-dependent viscosity of the magma. As illustrated in Figure 3, the first session of viscosity measurements recorded a marked decrease of viscosity during the first several minutes. Viscous heating, flow differentiation, melt climbing the rod and parallel alignment of plagioclase plates may each cause a decrease of viscosity during rotational viscometry. Crystal movement toward the outer part of the crucible and melt climbing the rod may decrease the apparent viscosity during measurement but their effects are not large and cannot account for the observed decrease of the viscosity. The most likely explanation of the temporal decrease of the viscosity is the change of texture of crystals due to alignment of plagioclase plates. Another possible cause of the temporal decrease of the viscosity in the first session of R-3-3 is the thixotropic behavior of the liquid phase. However, the nearly Newtonian behavior of near liquidus melt (Fig. 4) and the similarity of the composition of the melt for liquidus and subliquidus conditions (Table 1) suggest that this is not the case, and textural change during rotational viscometry may be the most likely cause of the marked decrease of viscosity during the first session of the viscosity measurements.

Non-Newtonian behavior of the basaltic melt

As illustrated in Figure 4, near liquidus basalt at 1210°C shows Newtonian behavior, whereas basalts at lower temperatures (1170°C and 1130°C) with higher crystal contents (11-23 vol%) show non-Newtonian behavior. A lower shear rate causes an increase of the viscosity, and a one order of magnitude decrease of the rotation rate caused a factor of 1.62 increase of viscosity at 1130°C. This is called shear thinning. The shear rate dependence of subliquidus magmas has been noted by previous experimental studies. Webb and Dingwell (1990) conducted fiber elongation experiments on silicate glasses and showed that the strain rate dependence of shear viscosity only occurs at high strain rate. This observation, however, is related to the structural relaxation of silicate melt and is not relevant to the non-Newtonian feature observed in the present work, in which interaction of suspended solids is the main factor causing the non-Newtonian behavior. Lejeune and Richet (1995) showed by experiment that interaction of spherical garnet crystal caused slight shear thinning behavior at high crystal contents (> 40 vol%). Pinkerton and Norton (1995) conducted subliquidus viscometry on Etna basalt, in which plagioclase was the liquidus phase. The basalt showed Newtonian behavior at temperatures above 1120°C, but became non-Newtonian with some yield strength at lower temperatures. The present experimental data are slightly different from the previous experimental observations, i.e., no yield strength was observed and a shear rate dependence of viscosity was observed at crystal contents of 10-23 vol%. The present experimental results suggest that in utilizing the rheological data to simulate basaltic lava flows, a shear rate dependence of the viscosity should be taken into account.

The non-Newtonian behavior is intimately related to the Bingham behavior of magma, in which yield strength creates a threshold for the start of fluidal movement of magmas. Hoover et al. (2001) demonstrate that the yield strength occurs at much lower crystal fractions (ca. 0.1-0.2) for thin prismatic kaolin suspensions compared with spherical poppy seed suspensions (ca. 0.5). This points to the importance of the crystal shape on the rheological properties of crystal suspensions. Ryerson et al. (1988) noted the presence of yield strength in Hawaiian picrites, in which near spherical olivine was crystallized in abundance. The present work utilized a rotational viscometer with constant rotation rate, so that it did not afford data on the yield strength of magma with plagioclase plates. It is likely that the presence of thin tabular plagioclase caused the non-Newtonian behavior in the subliquidus conditions, as shown in Figure 4. The non-Newtonian behavior of plagioclase-bearing magma may be relevant to the flow behavior of basaltic lava flows. Cashman et al. (1999) noted that the Pahoehoe to Aa transition of Hawaiian basalt lava flow corresponds to the increase of plagioclase lath from 15 (interior) to 40 (surface) vol%, which caused disruption of the surface of the lava because of the high yield strength required for flow. Present experiments are limited to lower crystallinity conditions, but the increase of the shear thinning behavior at higher crystal content is consistent with this idea of Cashman et al. (1999), and may facilitate understanding of the behavior of basaltic lava flows.

Temperature dependence of the viscosity of subliquidus basalt

Figure 5 illustrates an approximately 40-fold increase of viscosity over a 100°C decrease of temperature (from 1230°C to 1130°C). The increase of the viscosity may be caused by three factors; i.e., a decrease of temperature, a change of melt composition as a result of crystallization or an increase of crystallinity. We now evaluate these factors with respect to the change of viscosity of the melt + crystal system. Although there are models of non-Arrhenian equations for melt viscosity over wide temperature ranges (Taniguchi, 1995; Giordano et al., 2003), melts of near liquidus conditions may follow Arrhenian relations; i.e., a linear relation of log η and 1/T (K) (Bottinga and Weill, 1992; Shaw, 1972; Goto et al., 1997). We calculated the temperature effect on the melt viscosity by the equation of Shaw (1972), assuming a constant melt

composition of the bulk, shown as open circles in Figure 5. The viscosity increases by a factor of only 4 from 1230°C to 1130°C, and accounts for only 10% of the observed increase of the viscosity of the subliquidus basalt.

Utilizing the glass compositions in Table 1, we calculated the viscosity of the melt at the temperature of the measurement. Open squares in Figure 5 show the values of the viscosity of the melt that were obtained. These show an increase of the viscosity by a factor of only ~2 from 1230°C to 1130°C. This is mostly because the crystallization of plagioclase increases the ferromagnesian component and decreases the feldspathic component in the melt (Fig. 7). Therefore, a factor of 37.5 increase of the subliquidus magma from 1230°C to 1130°C is mostly accounted by the presence of plagioclase plates. The data are obtained in the later sessions of the measurements (Fig. 3) when the plagioclase plates are probably aligned parallel to the rotational directions.

Comparison with Einstein-Roscoe equation

In crystal + melt systems, the effects of crystals are generally expressed by the Einstein-Roscoe equation; i.e., $\eta_r =$ $\eta/\eta_{\rm m} = (1 - \varphi/\sigma)^{-2.5}$, where $\eta_{\rm r}$ is the relative viscosity, $\eta_{\rm m}$ is melt viscosity, φ is crystallinity, and σ is a constant (Roscoe, 1952). Marsh (1981) pointed out that σ may reach 0.6, suggesting that a crystal fraction of 0.6 corresponds to the rigid state of the crystal + melt system. The resulting equation, $\eta/\eta_{\rm m} = (1 - \varphi/0.6)^{-2.5}$, is often used to estimate the effect of crystals on the relative viscosity in the simulation of natural magmas. Lejeune and Richet (1995) noted that their experimental data on spherical garnet + melt system is neatly explained by this equation. Figure 8, which compares the present experimental results with the Einstein-Roscoe equation incorporating Marsh's constant, illustrates a marked departure of the data from the equation. When we fit the equation of Einstein-Roscoe to the present experimental data, we obtain a value of the constant σ of 0.3, indicating that entanglement of thin plates of plagioclase may become rigid at a crystallinity of ca. 30 vol%. This is similar to the critical rigidity threshold of crystal mesh in basaltic samples examined by Philpotts and Carroll (1996) and Hoover et al. (2001), who showed that heating of a basalt cube caused collapse of the cube after melting only 65-80% of the basalt; i.e., a crystallinity of 0.20-0.35.

The departure of the experimental data from the Einstein-Roscoe equation may be related to the morphology of the crystals or their size distribution (Pinkerton and Stevenson, 1992). Simha (1940) derived a theoretical equation of the effects of non-sphericity on the relative viscosity of crystal suspensions and gave approximate



Figure 8. Comparison of the measured and calculated relative viscosities. Solid squares represent the observed relative viscosity, obtained by dividing the bulk viscosity by the melt viscosity, calculated after the equation of Shaw (1972). The dotted bold line shows the Einstein-Roscoe equation with a constant of 0.6 (Marsh, 1981). It is noted that the observed relative viscosity is markedly larger than the calculated viscosity, which assumes the presence of spherical solids. See the text for further discussion.

equations for large axial ratios. For thin disks (axial ratio: f = a_2/a_1), the relative viscosity is expressed as η_r = 16f/15tan⁻¹f. For the axial ratio of 12.9, which is the measured average axial ratio of the experimental plagioclase, the relative viscosity becomes 9, which is slightly larger than the observed relative viscosity (4-5 times relative to the values of Einstein-Roscoe equation with a parameter of Marsh, Fig. 8). The slightly smaller relative viscosity in the present experiment may be caused either by dispersion of the crystal size or by the parallel alignment of plagioclase plates in the viscosity measurements. The original equation of Simha (1940) assumed random orientation of anisotropic ellipsoidal solids, whereas parallel alignment of the solids may decrease the apparent viscosity of the system (Manga, 1999). Although the present experiment showed an example of the effect of plagioclase plates on the viscosity of subliquidus magma, a general formulation of the effects of textural features on the viscosity of subliquidus magmas requires further experimental work under various conditions and starting materials with different crystal textures.

CONCLUSIONS

 Viscosities were measured on a high-Al basalt from Fuji volcano over the temperature range from 1230°C to 1130°C. The viscosity increased from ca. 52 Pa s at 1230°C (above liquidus) to ca. 1950 Pa s at 1130°C where the crystal content was 23 vol.%. The calculated viscosities of melts from 1230 °C to 1130°C increased only by a factor of ca. 2, indicating that the 37.5-fold increase of the viscosity of bulk basalt is mostly due to the presence of thin tabular plagioclase.

- 2. The relative viscosity-crystallinity relation obtained in the experimental results is larger by a factor of 4-5 than the calculated values using the Einstein-Roscoe equation with a parameter value from Marsh $(\eta/\eta_m = (1 - \varphi/0.6)^{-2.5})$. The discrepancy may largely be due to the effect of entanglement of thin tabular crystal shape of plagioclase.
- A tendency to non-Newtonian shear-thinning increases as the crystal content increases in the Fuji basalt.

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REFERENCES

- Bottinga, Y. and Weill, D.F. (1972) The viscosity of magmatic silicate liquids: a model for calculation. American Journal of Science, 272, 438-475.
- Cashman, KV, Thornber, C. and Kauahikaua, J. (1999) Cooling and crystallization of lava in open channels, and the transition of pahoehoe lava to 'a'a. Bulletin of Volcanology, 61, 306-323.
- Deer, W.A., Howie, R.A. and Zussman, J. (1992) An introduction to the rock-forming minerals, 2nd edition. pp. 696, Longman, London.
- Dingwell, D.B. (1998) Recent experimental progress in the physical description of silicic magma relevant to explosive volcanism. In The Physics of Explosive Volcanic Eruptions (Gilbert, J.S. and Sparks, R.S.J. Eds.). Geological Society, London, Special Publication, 145, 9-26.
- Gibb, F.G.F. (1974) Supercooling and crystallization of plagioclase from a basaltic magma. Mineralogical Magazine, 39, 641-653.
- Giordano, D. and Dingwell, D.B. (2003) Non-Arrhenian multicomponent melt viscosity: a model. Earth and Planetary Science Letters, 208, 337-349.
- Goto, A., Ohshima, H. and Nishida, Y. (1997) Empirical method of calculating the viscosity of peraluminous silicate melts at high temperatures. Journal of Volcanology and Geothermal Research, 76, 319-327.
- Hara, I. (1991) 1-atmosphere experiment on the 1707 basalt of Fuji volcano. pp.43, MS Thesis, Hiroshima University, Hiro-

shima, Japan.

- Hoover, S.R., Cashman, K.V. and Manga, M. (2001) The yield strength of subliquidus basalts - experimental results. Journal of Volcanology and Geothermal Research, 107, 1-18.
- Huebner, J.S. (1982) Use of gas mixtures at low pressure to specify oxygen and other fugacities of furnace atmospheres. In Hydrothermal Experimental Techniques (Ulmer, G.C. and Barnes, H.L. Eds.). John Wiley & Sons, NewYork, 20-60.
- Komar, P.D. (1972) Mechanical interactions of phenocrysts and flow differentiation of igneous dikes and sills. Geological Society of America Bulletin, 83, 973-981.
- Landau, L.D. and Lifschitz, E.M. (1970) Fluid Mechanics, Japanese edition translated by Takeuchi, H., Tokyo-Gakujyutsu Tosho Co. Ltd, 64-65.
- Lejeune, A.M. and Richet, P. (1995) Rheology of crystal-bearing silicate melts: an experimental study at high viscosities. Journal of Geophysical Research, 100, 4215-4229.
- Manga, M. (1999) Orientation distribution of microlites in obsidian. Journal of Volcanology and Geothermal Research, 86, 107-115.
- Marsh, B.D. (1981) On the crystallinity, probability of occurrence, and rheology of lava and magma. Contribution to Mineralogy and Petrology, 78, 85-98.
- Philpotts, A.R. and Carroll, M (1996) Physical properties of partly melted tholeiitic basalt. Geology, 24, 1029-1032.
- Pinkerton, H. and Stevenson, R.J. (1992) Methods of determining the rheological properties of lavas from their physico-chemical properties. Journal of Volcanology and Geothermal Research, 53, 47-66.
- Pinkerton, H. and Norton, G. (1995) Rheological properties of basaltic lavas at sub-liquidus temperatures: laboratory and field measurements on lavas from Mount Etna. Journal of Volcanology and Geothermal Research, 68, 307–323.
- Roscoe, R. (1952) The viscosity of suspensions of rigid spheres. British Journal of Applied Physics, 3, 267-269.
- Ryerson, F.J., Weed, H.C. and Piwinskii, A.J. (1988) Rheology of subliquidus magmas, 1. picritic compositions. Journal of Geophysical Research, 93, 3421-3436.
- Shaw, H.R. (1969) Viscosity of basalt in the melting range. Journal of Petrology, 10, 510–535.
- Shaw, H.R. (1972) Viscosities of magmatic silicate liquids: an empirical method of prediction. American Journal of Science, 272, 870-893.
- Simha, R. (1940) The influence of Brownian movement on the viscosity of solutions. Journal of Physical Chemistry, 44, 25-34.
- Spera, F.J. (2000) Physical properties of magma. In Encyclopedia on Volcanoes(Sigurdsson, H. Ed.). Academic Press, San Diego, California, 171-190.
- Spera, F.J., Borgia, A. and Strimple, J. (1988) Rheology of melts and magmatic suspensions, Design and calibration of concentric cylinder viscometer with application to rhyolitic magma. Journal of Geophysical Research, 93, 10273-10294.
- Taniguchi, H. (1995) Universal viscosity-equation for silicate melts over wide temperature and pressure ranges. Journal of Volcanology and Geothermal Research, 66, 1-8.
- Web, S.L. and Dingwell, D.B. (1990) Non-Newtonian rheology of igneous melts at high stresses and strain rates: experimental results for rhyolite, andesite, basalt, and nephelinite. Journal of Geophysical Research, 95, 15695-15701.

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