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Viscosity measurements of subliquidus magmas: Alkali olivine basalt from the Higashi-Matsuura district, Southwest Japan

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Abstract

We carried out viscosity measurements and sampling of a crystal suspension derived from alkali olivine basalt from the Matsuura district, SW Japan, at subliquidus temperatures from 1230 °C to 1140 °C under 1 atm with NNO oxygen buffered conditions. Viscosity increased from 31 to 1235 Pa s with a decrease in temperature from 1230 to 1140 °C. On cooling, olivine first appeared at 1210 °C, followed by plagioclase at 1170 °C. The crystal content of the sample attained 31 vol.% at 1140 °C (plagioclase 22%, olivine 9%). Non-Newtonian behaviors, including thixotropy and shear thinning, were pronounced in the presence of tabular plagioclase crystals. The cause of such behavior is discussed in relation to shear-induced changes in meltcrystal textures. Relative viscosities, η_r (= η_s/η_m , where η_s and η_m are the viscosities of the suspension and the melt, respectively), were obtained by calculating melt viscosities from the melt composition and temperature at 1 atm using the equation proposed by Giordano and Dingwell [Giordano, D., Dingwell, D.B., 2003. Non-Arrhenian multicomponent melt viscosity: a model. Earth and Planetary Science Letters, 208, 337-349.]. The obtained relative viscosities are generally consistent with the Einstein-Roscoe relation, which represents η_r for suspensions that contain equant and equigranular crystals, even though the crystal suspension analyzed in the present experiments contained tabular plagioclase and granular olivine of various grain sizes. This consistency is attributed to the fact that the effect of crystal shape was counterbalanced by the effect of the dispersion of crystal size. The applicability of the Einstein-Roscoe equation with respect to crystal shape is discussed on the basis of the present experimental results. Our experiments and those of Sato [Sato, H., 2005. Viscosity measurement of subliquidus magmas: 1707 basalt of Fuji volcano. Journal of Mineralogical and Petrological Sciences, 100, 133–142.] show that the relationship between relative viscosity and crystal fraction is consistent with the Einstein-Roscoe relationship for axial ratios that are smaller than the critical value of 4-6.5, but discrepancies occur for higher ratios.

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1. Introduction

Viscosity is a critical parameter in terms of controlling the flow properties of magma. Most natural magmas exist at subliquidus temperatures and are crystal suspensions. The effects of crystals on the rheological

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properties of magma should therefore be properly evaluated when modeling magmatic processes such as convection and crystal settling within a magma chamber, magma ascent within a volcanic vent, volcanic eruption, and the flow of lava upon the Earth's surface. The rheological properties of magma are affected not only by crystal content but also by crystal shape, mean crystal size, and size distribution (e.g. Pinkerton and Stevenson, 1992; Pinkerton and Norton, 1995). Although a number of laboratory experiments have examined the rheological properties of different types of natural magmas at subliquidus temperatures (e.g. Shaw, 1969; Murase and McBirney, 1973; Murase et al., 1985; Ryerson et al., 1988; Spera et al., 1988; Pinkerton and Norton, 1995; Sato, 2005) and synthetic silicate suspensions (e.g. Lejeune and Richet, 1995; Deubener and Brückner, 1997; Yue et al., 1999; Müller et al., 2003), a generalized method for predicting the flow properties of magma has yet to be proposed, largely because of a lack of textural and compositional analyses of experimental products.

The rheological properties of magmas that contain equant crystals are relatively well understood (e.g. Lejeune and Richet, 1995). At relatively low crystal contents, magma behaves as a Newtonian flow, and its relative viscosity, $\eta_r (=\eta_s/\eta_m$, where η_s and η_m are the viscosities of the crystal suspension and the melt, respectively), is well described by the Einstein–Roscoe relationship (Roscoe, 1952):

$$\eta_{\rm r} = (1 - \Phi/\Phi_{\rm m})^{-2.5},\tag{1}$$

where Φ is the volume fraction of crystals and Φ_m is the maximum packing fraction of crystals. Φ_m is commonly assumed to be 0.6, as determined empirically by Marsh (1981). Lejeune and Richet (1995) analyzed the rheological properties of Mg₃Al₂Si₃O₁₂ melt that contained varying amounts of spherulites of aluminous enstatite. In their experiment, the measured relative viscosity was in good agreement with Eq. (1), indicating the validity of the equation.

At high crystal contents, magma behaves as a non-Newtonian fluid. The equation proposed by Gay et al. (1969) provides a good description of the flow properties of suspensions that contain high particle contents. This equation has been used in predicting the flow behavior of dacite lava at Mt. Saint Helens and basaltic lava at Mt. Etna (Pinkerton and Stevenson, 1992; Pinkerton and Norton, 1995). The volume fraction of equant crystals that marks the transition from Newtonian to non-Newtonian behavior is approximately 0.3 [0.25 for picrite magma that contains olivine crystals (Ryerson et al., 1988), 0.4 for Mg₃Al₂Si₃O₁₂ melt that contains spherulite (Lejeune and Richet, 1995), and 0.29 determined from numerical calculations (Saar et al., 2001)]. This critical value may also be dependent on factors such as crystal shape, crystal size distribution, and shear strain rate, although the nature of such a dependence has yet to be investigated.

In contrast to magmas that contain equant crystals, the rheological properties of magmas that contain tabular crystals are poorly understood. Sato (2005) provided the first experimental analysis of the rheological properties of a tabular-plagioclase suspension produced from Fuji 1707 basaltic lava. Sato's experiments demonstrated that the behavior of the suspension is non-Newtonian, even at a low crystal fraction of 0.11, and that the relative viscosities deviate markedly from those predicted by the Einstein–Roscoe relationship. These experiments show that the rheological properties of magma are strongly influenced by crystal shape in addition to the influence of the crystal volume fraction.

In the present study, we used a concentric cylinder viscometer to measure the viscosity of a crystal suspension, derived from alkali olivine basalt, that contains both equant olivine and tabular plagioclase crystals. The experimental products were polished and thin-sectioned, and their phase compositions and textures were analyzed to evaluate the effects of crystals on the rheological properties of the suspension. We discuss the effects of crystal shape and crystal size distribution on values of relative viscosity and describe a constraint on the applicability of the Einstein–Roscoe equation. We also discuss texture-related causes of non-Newtonian behavior in crystal suspensions.

2. Experimental methods

2.1. Starting material

A block of alkali olivine basaltic lava was collected from the Higashi-Matsuura district, Southwest Japan, and processed as a starting material. In this district, a relatively massive succession of basaltic lava flows ($\sim 6.2 \text{ km}^3$; Kurasawa, 1967) covers basement that comprises the Middle-Late Miocene Sasebo Group and Cretaceous granitic plutons. The basaltic succession consists of nine flow units, each with a thickness of 10-30 m (Kobayashi et al., 1955, 1956). K-Ar ages of the flows are 3.00 ± 0.03 Ma (Nakamura et al., 1986). The lava collected for the starting material was sampled from the B2 unit defined by Kobayashi et al. (1955, 1956), which is the most widespread flow unit close to basement. The major element composition of the starting material is listed in Table 1, which shows the most undifferentiated composition within the B2 unit.

Table 1 Major element compositions of the starting material, melts, and mineral phases within the samples

<i>T</i> (°C)	Phase	SiO_2	TiO ₂	Al_2O_3	Cr_2O_3	FeO*	MnO	MgO	NiO	CaO	Na ₂ O	K ₂ O	P_2O_5	Total	Mg#	An#
Starting material		49.45	1.75	16.21		9.58	0.17	8.38		10.03	3.02	1.30	0.23	100.12		
1230	Melt	49.50	1.81	16.41		9.37	0.21	8.17		10.08	3.07	1.30	0.26	100.18		
	Spinel	0.72	1.86	26.03	28.44	25.17	0.24	14.44	0.06	0.27				97.23	62.9	
1210	Melt	49.63	1.75	16.41		9.45	0.18	8.29		10.06	3.05	1.30	0.23	100.35		
	Olivine	40.57				12.04	0.19	45.75	0.16	0.30				99.01	87.2	
	Spinel	0.59	1.50	23.87	28.17	28.26	0.15	13.62	0.12	0.26				96.54	61.1	
1190	Melt	49.27	1.75	16.22		9.55	0.16	8.09		10.11	3.06	1.32	0.21	99.74		
	Olivine	40.29				13.01	0.22	45.34	0.17	0.29				99.32	86.2	
	Spinel	0.52	1.29	24.52	27.57	28.76	0.29	13.65	0.17	0.31				97.08	61.6	
1170	Melt	49.44	1.79	16.22		9.71	0.18	7.54		10.15	3.12	1.28	0.23	99.66		
	Olivine	40.34				13.60	0.25	45.15	0.11	0.31				99.76	85.6	
	Plagioclase	50.71		30.87		0.99				14.64	3.35	0.24		100.80	69.8	69.8
	Spinel	0.45	1.30	23.67	26.97	30.34	0.23	13.47	0.12	0.30				96.85	61.1	
1160	Melt	50.13	2.17	15.28		10.55	0.18	6.48		10.16	3.17	1.57	0.33	100.02		
	Olivine	40.03				15.79	0.28	43.49	0.09	0.32				100.00	83.2	
	Plagioclase	50.26		30.94		1.04				14.82	3.21	0.24		100.51	70.9	70.9
	Spinel	0.47	2.67	17.94	22.60	41.12	0.39	10.66	0.10	0.35				96.30	49.1	
1150	Melt	50.38	2.37	14.61		10.90	0.20	6.08		10.15	3.15	1.70	0.36	99.90		
	Olivine	39.52				16.40	0.27	42.75	0.06	0.32				99.32	82.4	
	Plagioclase	50.08		30.92		1.01				14.67	3.27	0.23		100.18	70.3	70.3
	Spinel	0.39	3.51	14.96	21.88	45.52	0.32	9.21	0.07	0.35				96.21	42.5	
1140	Melt	50.42	2.46	14.25		11.00	0.21	5.68		10.12	3.05	1.94	0.35	99.48		
	Olivine	39.44				18.55	0.33	41.03	0.06	0.40				99.81	79.9	
	Plagioclase	50.34		30.92		1.04				14.80	3.15	0.24		100.49	71.2	71.2
	Spinel	0.68	5.43	11.97	19.42	48.24	0.29	8.23	0.11	0.37				94.74	37.2	

Mg# and An# are calculated by Mg/(Mg+Fe) and Ca/(Ca+Na+K) in mol fraction, respectively.

FeO* represents total iron as FeO.

2.2. Experimental equipment and method

We used an atmosphere-controlled furnace consisting of a vertical double-spiral heating element of SiC that is detached from the sample crucible by an alumina tube (60 mm internal diameter). The temperature of the sample crucible was monitored and controlled by a thermocouple located outside of the heating element. The temperature in the crucible was calibrated against the melting point of gold. Variations in temperature around the sample crucible were within 5 °C. The redox condition within the furnace was controlled using mixed H₂ and CO₂ gas so as to realize oxygen fugacities similar to those of Ni-NiO buffer at experimental temperatures. The flow rates of the H₂ and CO₂ gases were 10 ml/min and 500 ml/min, respectively (Huebner, 1987). The mixed gas was introduced from the bottom of the alumina tube; from there, it flowed upward within the tube at a rate of 2-3 mm/s.

We measured viscosity using an electromagnetic rotational viscometer (TV-10U, Toki Sangyo Co. Ltd., Japan). The fixed outer cylinder comprised a 60 mm high Pt crucible (1 mm wall thickness) with a 30 mm internal diameter. An alumina rod with an outer diameter of 6 mm

was installed as the rotating inner cylinder. At a constant rotation rate, viscosity is obtained from the following equation:

$$M = 4\pi\eta \Omega R_{\rm b}^2 R_{\rm c}^2 / \left(R_{\rm c}^2 - R_{\rm b}^2 \right) \tag{2}$$

where *M* is the moment, η is viscosity in Pa s, Ω is the angular velocity, and R_b and R_c are the radii of the rod and crucible, respectively (Shaw, 1969; Landau and Lifschitz, 1970). To correct for the end-effect of the rod, the system was calibrated with a standard-viscosity oil (JS160000) by varying the distances between the rod end and the bottom of the crucible and by varying the depth of the oil. The JS160000 standard oil has a viscosity of ~148 Pa s at 20 °C and shows Newtonian behavior.

Fig. 1 shows the temperature-time schedule of the experiment. Viscosity measurements and sampling were performed at 1230, 1210, 1190, 1170, 1160, 1150, and 1140 °C. The lava sample was kept at 1230 °C for 48 h to ensure that the starting material was perfectly molten. The viscosity was then measured and a sample was collected. Subsequently, the temperature of the melt was lowered to that of the next measurement step. This



Fig. 1. Temperature–time schedule of viscosity measurements. The arrows indicate the timing of viscosity measurements and sampling. Viscosity measurements and sampling were carried out after maintaining the sample at a constant temperature for 1-2 days.

sequence of viscosity measurement, sampling, and stepwise cooling was repeated every ~ 24 h. We believe that after ~ 20 h, a near-equilibrium state is attained prior to viscosity measurement and sampling. This view is based on the observation that similar results were obtained for both 24 h/step and 48 h/step experiments (Sato, unpublished data), as well as the fact that experiments using a wire-loop method indicated near-equilibration conditions after 10 h (Sato, 1995). The cooling rate during the transition between steps was 1 °C/min at temperatures above 1170 °C and 0.5 °C/min at temperatures below 1170 °C. At temperatures below 1140 °C, we were unable to perform viscosity measurements because the large number of crystals and resulting high viscosity of the magma made it impossible to insert the rod.

The procedures used in the viscosity measurements and sampling were as follows. The top of the alumina tube of the furnace was kept closed with a ceramic plate cover, except for times of viscosity measurement. When measuring viscosity, the plate cover was removed and the rotation rod connected to the viscometer was inserted into the molten basalt within the fixed Pt crucible. Each viscosity measurement involved several sessions with variable rotation rates to record the dependence of viscosity on time and shear strain rate. The rotation rate was first increased in a stepwise mode from the lowest to the highest rate before being reduced again in a stepwise mode. During measurement, the top of the tube was kept as closed as possible with the ceramic plates so as to minimize temperature fluctuations and minimize the movement of air into the tube (Sato, 2005). Upon completion of the measurements, the top of the tube was opened to extract the rod and then closed again. The

basalt sample attached to the tip of the rod was quenched, processed to make polished thin sections, and analyzed with an electron probe analyzer for phase chemistry and texture. The depth of molten basalt within the crucible was ~ 49 mm at the start of the experiment. This amount decreased to 28 mm after the run due to repeated sampling during the experiment. The depth of molten basalt within the crucible for each measurement was obtained by linearly interpolating the depths of the sample before and after the experiment. Minor dissolution of the alumina rod was detected close ($\sim 50 \ \mu m$) to the rod at the highest temperature, but this did not affect the bulk melt compositions listed in Table 1. Dissolution of the alumina rod was negligible below the plagioclase liquidus temperature, as a cover of very fine ($<5 \mu m$) plagioclase crystals crystallized upon the surface of the alumina rod, thereby preventing dissolution. In contrast



Fig. 2. Variations in the volume fractions of melt (a), plagioclase (b), and olivine (c) with changing temperature. As temperature decreases, the volume fraction of melt decreases and the fraction of plagioclase and olivine increases.

to Sato (2005), who added basalt powder to his experiments, no seed crystals were used in the present study.

Yield strength is petrologically a very interesting aspect of magma rheology (e.g. McBirney and Noyes, 1979; Ryerson et al., 1988). Conventionally, the yield strength is estimated experimentally by extrapolating the relationship between stress and strain rate at a finite strain rate to a strain rate of zero (e.g. Ryerson et al., 1988; Lejeune and Richet, 1995); however, this method is highly model-dependent (e.g. Ryerson et al., 1988; Kerr and Lister, 1991). Bagdassarov and Pinkerton (2004) used a rotational viscometer to obtain precise estimates of the yield strength of viscoelastic foam, as an analogue of bubble-bearing melt, by measuring residual stress when the strain rate was reduced from a finite value to zero. In our experiment, yield strength was not estimated because of difficulties encountered in using our viscometer when measuring residual stress at a strain rate of zero.

3. Results

3.1. Phase chemistry and modal abundances

Quenched basalt samples were analyzed using a scanning electron microprobe (JEOL JSM5800LV) equipped with an EDS analytical system (Oxford, Link ISIS) at Kyushu University under analytical conditions of 500 pA beam current, 20 kV accelerating voltage, and 100 s counting time.

The phase assemblages in the samples were as follows: melt+spinel at 1230 °C, melt+olivine+spinel at 1210 °C and 1190 °C, and melt+plagioclase+olivine+spinel below 1170 °C. The phase compositions are listed in



Fig. 3. Relations between volume fraction and the concentrations of major elements within the melt.

Table 1. The volume fractions of major phases were calculated using the mass-balance calculation method of Le Maitre (1979) from starting and phase compositions (Table 1); the results are listed in Table 2. The volume fractions of spinel were obtained from a backscattered electron (BSE) image (Table 2).

The volume fractions of olivine and plagioclase increased steadily to 0.088 and 0.218 with decreasing temperature, respectively (Fig. 2). The mean Fo number decreased from 87.2 to 79.9 with increasing volume fraction of olivine. The composition of plagioclase was largely constant throughout the experiment runs, with An numbers [=100Ca/(Ca+Na+K) in mol fraction] from 69.8 to 71.2 (Table 1). The volume fraction of melt decreased continuously to 0.694 with decreasing temperature, with a rapid decrease from 0.954 to 0.694 over the temperature range from 1170 to 1140 °C (Fig. 2). SiO_2 , TiO_2 , $FeO*(=FeO+0.9Fe_2O_3)$, and K_2O contents increased with decreasing volume fraction of melt, whereas Al₂O₃ and MgO contents decreased and CaO and Na₂O contents were largely constant (Fig. 3). The volume fraction of spinel increased from 0.0002 at 1230 °C to 0.0027 at 1170 °C before decreasing again.

The chromite component decreased with decreasing temperature, whereas the magnetite component increased (Tables 1 and 2).

3.2. Texture

Fig. 4 shows BSE images of samples oriented normal to the rotation axis. Crystals are evenly distributed regardless of the distance from the inner rod. Olivine crystals are mostly euhedral, with some being skeletal. The olivine crystals are smaller than 100 μ m and have length/width ratios of less than 3.

Plagioclase crystals are generally tabular (thin parallelepipeds with an edge angle of 116°), with some skeletal crystals. Crystal size distributions (CSD), length/ width ratios, and orientations of plagioclase crystals were determined from digitized BSE images of samples using SCION Image analytical software. The means and standard deviations of plagioclase crystal sizes for each temperature are listed in Table 2, and the CSDs for plagioclase crystals are shown in Fig. 5. The 2D intersection data were converted to 3D using the conversion method proposed by Higgins (2000). For temperatures



Fig. 4. Back scattered electron images of samples within the plane oriented normal to the rotation axis at 1140 °C (a), 1150 °C (b), 1160 °C (c) and 1170 °C (d). The width of the images is 660 μ m. The dark gray and elongate rectangles are plagioclase; the equant mid-gray crystals are olivine; the white equant crystals are spinel; and the light gray area is melt. The dark area with darker pits is the alumina rod used for the viscosity measurements and sampling.

Table 2 Results of viscosity measurements, phase proportions, and textural analyses

2							
<i>T</i> (°C)	1230	1210	1190	1170	1160	1150	1140
$\eta_{\rm s}$ (Pa s)	31	38	52	94	233	674	1235
$\eta_{\rm m}$ (Pa s)	30	42	58	94	141	183	241
$\eta_{ m r}$	1.03	0.91	0.88	1.00	1.65	3.67	5.13
$\eta_{r(1)}$	1.00	1.01	1.02	1.22	2.69	4.07	5.95
$\Phi_{ m melt}$	100	99.7	99.5	95.4	80.4	74.2	69.4
(vol.%)							
$\Phi_{ m olv}$ (vol.%)	_	0.3	0.5	2.1	5.9	7.3	8.8
$\Phi_{\rm plg}$ (vol.%)	_	_	_	2.5	13.7	18.5	21.8
$*\Phi_{\rm spl}$	0.02	0.04	0.09	0.27	0.13	0.08	0.04
(vol.%)							
L_{plg}	_	_	_	98.5	42.0	55.9	52.4
$SD_{L_{n/n}}$	_	_	_	106.2	68.5	58.1	53.2
ϵ_{Total}	_	_	_	350	850	475	330

 $\eta_{\rm m}$ is estimated by the method of Giordano and Dingwell (2003).

 $\eta_{r(1)}$ is the relative viscosity calculated by Eq. (1).

 $\Phi_{\rm spl}(vol.\%)$ is obtained from the BSE image.

 L_{plg} and $SD_{L_{plg}}$ are mean and standard deviation of long axis of plagioclase crystals, respectively.

 ϵ_{Total} is total strain required to attain η_s to steady state.

below 1160 °C, the population densities of plagioclase crystals decreased exponentially with increasing crystal length. These CSD patterns may imply that nucleation and growth continued until immediately prior to measurement and that significant crystal fractionation

did not occur (e.g. Marsh, 1998; Cashman and Marsh, 1988), although size-dependent growth can also contribute to the formation of linear CSDs (Eberl et al., 2002). The length of plagioclase crystals shows only a moderate negative correlation with the log population densities at 1170 °C; this is attributed to the fact that 1170 °C is close to the liquidus temperature of plagioclase and the fact that only a small number of plagioclase crystals were sampled.

Plagioclase crystals have a tendency to become more elongate via an increase in the length of the major axis. The frequency distributions of the apparent axial ratios of plagioclase crystals are shown in Fig. 6. Plagioclase crystals with length/width ratios of around 4 were dominant at all run temperatures. The frequency of these crystals decreased as the length of the major axis increased. Fig. 7 shows that the highest frequency of plagioclase crystals occurs for crystals with their major axes oriented parallel to the direction that is tangential to the rotation. The direction that is tangential to the rotation is normal to the flow velocity gradient within the cylinder. At temperatures below 1160 °C, plagioclase crystals have a tendency to align parallel to the direction that is tangential to the rotation. As the orientation of plagioclase crystals was random before applying shear to the suspension, the alignment of plagioclase crystals almost certainly formed in response to the shear.



Fig. 5. Crystal size distributions of plagioclase crystals at 1140 °C (a), 1150 °C (b), 1160 °C (c), and 1170 °C (d). The 2D intersection data were converted to 3D using the conversion method proposed by Higgins (2000).



Fig. 6. Frequencies of length/width ratios of plagioclase crystals at 1140 °C (a), 1150 °C (b), 1160 °C (c), and 1170 °C (d).

3.3. Viscosity

Fig. 8 shows variations in viscosity with run time, with results for 1150 °C and 1190 °C presented as

examples. As mentioned above, plagioclase is present at 1150 °C but is absent at 1190 °C. The observed variation in viscosity at 1150 °C comprises two distinct parts (Fig. 8a). The rotation rate was increased in a stepwise



Fig. 7. Frequencies of the orientations of the long axes of plagioclase crystals relative to the tangential direction of rotation at 1140 °C (a), 1150 °C (b), 1160 °C (c), and 1170 °C (d). Note that plagioclase crystals are aligned parallel to the tangential direction of rotation at temperatures below 1160 °C.



Fig. 8. Time-dependent variations in viscosity at 1150 °C (a) and 1190 °C (b). Thin black lines, thick gray lines, thick black lines, and thin gray lines represent rotation rates of 1, 2.5, 5, and 10 rpm in (a), and 5, 10, 20, and 50 rpm in (b). At 1150 °C, the measured viscosity decreases markedly over time to a minimum value at 10 rpm before increasing slightly with decreasing rotation rate. The former behavior is thixotropy and the latter is shear thinning. At 1190 °C, the measured viscosity is constant regardless of lapse time and rotation rate.

mode during the first half of the run (< 3000 s) and decreased during the second half ($> \sim 3000$ s). In the first half of the run, the viscosity decreased over time, even at a constant rotation rate, before achieving a steady state. The total strain required to achieve a steady state at temperatures below the plagioclase liquidus is listed in Table 2. The viscosity at 1150 °C decreased from \sim 5000 to 670 Pa s during the first 3000 s of the run. In the second half, the viscosity was largely constant at each rotation rate, and increased slightly from 670 to 830 Pa s as the rotation rate decreased from 10 to 1 rpm. The variation in viscosity recorded in the first half of the run is known as thixotropy, a time-dependent decrease in viscosity, whereas that in the second half of the run is the so-called shear thinning, in which viscosity decreases with an increasing rate of shear strain. In contrast, the viscosity at 1190 °C was almost constant, regardless of both the lapse time and the rotation rate (Fig. 8b).

Fig. 9 shows the rotation-rate dependence of the viscosity. At temperatures above 1190 °C, plagioclase crystals were absent, and the viscosity was largely constant regardless of the rotation rate. In contrast, at temperatures below 1170 °C, plagioclase crystals were present and the viscosity decreased slightly with increasing rotation rate.

To evaluate the effect of crystals on the rheology of magma, it is necessary to determine the viscosity of a pure silicate melt phase. According to Webb and Dingwell (1990), pure silicate melt shows Newtonian behavior when the reciprocal of the shear strain rate is more than three orders of magnitude greater than the Maxwell relaxation time, τ (=viscosity/shear modulus), even at temperatures close to the glass transition. The shear modulus of basaltic melt is generally considered to be in the order of 10^8-10^{10} Pa (Dingwell and Webb, 1990; James et al., 2004). In the case of our experiments, pure basaltic melt with a viscosity in the order of 10^2 Pa s showed Newtonian behavior because the highest shear



Fig. 9. Relationship between measured viscosity and rotation rate at different temperatures. At temperatures above 1190 °C, where plagioclase crystals are absent, the measured viscosity is constant regardless of the rotation rate. At temperatures below 1170 °C, where plagioclase crystals occur, we observe shear-thinning behavior, with a minor increase in measured viscosity with decreasing rotation rate.

strain rate in our experiment is 2-4 orders of magnitude less than that required to induce visco-elastic behavior. Several empirical methods have been proposed to calculate the Newtonian viscosity of multicomponent silicate melts as a function of temperature and melt composition (e.g. Bottinga and Weill, 1972; Shaw, 1972; Giordano and Dingwell, 2003); however, it is difficult to accurately estimate the viscosity of silicate melt at subliquidus temperatures. The methods of Bottinga and Weill (1972) and Shaw (1972) are both based on the Arrhenius equation $(\ln(\eta) = A + B/T)$, where η is viscosity, T is absolute temperature, and A and B are constants); however, the temperature-viscosity relationship of silicate melt is Arrhenian only at high superliquidus temperatures and is generally non-Arrhenian at lower temperatures (e.g. Spera, 2000; Taniguchi, 2000). These methods generate serious errors in viscosity estimates at lower temperatures because their calibrations are based solely on experiments conducted at high superliquidus temperatures (Taniguchi, 2000; Giordano and Dingwell, 2003). In contrast, the method described by Giordano and Dingwell (2003) is based on the Tammann-Vogel-Fulcher (TVF) equation $(\ln \eta = A + B/(T - T_0))$, where A, B, and T_0 are constants), which takes non-Arrhenian effects into account. To assess the reliability of this method, we compared the calculated viscosity with observed data for an alkali basaltic melt (Taniguchi, 1993) and the viscosity measured at 1230 °C in the present study, which included only a negligible amount of crystals. The calculated viscosities are about 0.2 log units higher than observed viscosities at temperatures from 1250 to 1150 °C.

In this study, the viscosity of melt was estimated by adding 0.2 log units to the viscosity calculated using the method described by Giordano and Dingwell (2003). This approach was taken to correct the discrepancy that exists between observed and calculated viscosity values. The estimated melt viscosities for each run temperature are listed in Table 2. The calculated melt viscosity increased consistently from 30 to 241 Pa s with a decrease in temperature from 1230 to 1140 °C.

The steady-state viscosities at 5 rpm for all run temperatures are listed in Table 2. A rotation rate of 5 rpm corresponds to a shear strain rate of 0.57 s^{-1} at the surface of the rotation rod, which is of the same order as that of a natural basaltic magma ascending within a dyke or flowing upon the land surface $(0.1-1 \text{ s}^{-1}; \text{Spera et al.}, 1988)$. Fig. 10 shows a comparison of measured melt viscosities and viscosities calculated using the method of Giordano and Dingwell (2003), with these values plotted against the reciprocal of absolute temperature. For reference, we also plotted the viscosities of melts with the composition of the starting material at temperatures



Fig. 10. Relationship between the viscosity coefficient and the inverse of temperature (K^{-1}). Black squares are the measured viscosity at a steady state with a rotation rate of 5 rpm, and gray diamonds are the viscosity of melt calculated using the method of Giordano and Dingwell (2003) with the analyzed melt composition at each temperature (see details in text). The gray line is the relationship between temperature and viscosity, which is also estimated using the method of Giordano and Dingwell (2003) from the starting melt composition. Across the range of experiment temperatures, the calculated viscosity for the starting melt composition is largely linearly proportional to the reciprocal of absolute temperature, and the relationship is successfully regressed by following equation: $\log \eta = 1.615 - 9.293/T (R^2 = 0.9998)$.

from 1230 to 1140 °C, as calculated using the method of Giordano and Dingwell (2003). Across the range of experiment temperatures, the estimated viscosity for the starting melt composition increased approximately linearly with reciprocal absolute temperature, and is expressed as $\log(\eta) = 1.615 - 9.293 / T (R^2 = 0.9998)$. The measured viscosities vary from 31 to 1235 Pa s with a decrease in temperature from 1230 to 1140 °C. At temperatures above 1170 °C, the measured viscosities are similar to both the calculated viscosities of melts and the calculated viscosity of melt with the starting material composition, indicating that the effects of crystallization are not discernible. At temperatures below 1170 °C, both the measured and calculated viscosities of the analyzed melts differ from the calculated viscosity of melt with the composition of the starting material. The measured viscosity increased by a factor of ~ 13 over the 30 °C interval from 1170 to 1140 °C, and the temperature that marked the start of the rapid increase in viscosity coincided with the first appearance of plagioclase (Fig. 2). At temperatures below 1170 °C, the calculated viscosities of the analyzed melts are higher than the extension of the calculated viscosity of the melt with the composition of the starting material. However, the viscosities of the



Fig. 11. Comparison of measured relative viscosities and those estimated using the Einstein–Roscoe equation. Black squares and gray triangles are measured values of viscosity for the present study and that of Sato (2005), respectively. The gray line shows the Einstein–Roscoe relationship.

analyzed melts were all significantly lower than the measured viscosities of the crystal suspension at all run temperatures below 1170 °C. This finding indicates that the increase in the measured viscosity of the crystal suspension is mainly due to the effect of crystals; the effect of melt composition is relatively small in comparison.

The effect of crystals on the measured viscosities was evaluated from the relative viscosity, $\eta_{\rm r}$ In Fig. 11, the relative viscosity is plotted against the volume fraction of the crystals, Φ . The relative viscosity increased consistently up to 5.1 for a volume fraction of crystals up to Φ =0.306. The Einstein–Roscoe relationship shown in Eq. (1) with $\Phi_{\rm m}$ =0.6 (Marsh, 1981; Pinkerton and Stevenson, 1992) is also plotted in Fig. 11. The data obtained in the present study are largely in agreement with the Einstein–Roscoe relationship, except for those measurements taken at 1160 °C.

4. Discussion

4.1. Effect of crystals on the viscosity of the suspension

The relative viscosity of a crystal suspension is generally expressed using the Krieger–Dougherty equation (Krieger and Dougherty, 1959):

$$\eta_{\rm r} = \left(1 - \Phi/\Phi_m\right)^{-[\eta]\Phi_m} \tag{3}$$

where $[\eta]$ is the intrinsic viscosity. In geological applications, the exponential term $[\eta]\Phi_{\rm m}$ is commonly assumed to be 2.5, which equates to the Einstein–Roscoe relationship under the assumption that $\Phi_{\rm m}=0.6$

(Marsh, 1981; Pinkerton and Stevenson, 1992). The relationship successfully represents the relative viscosity of magmas that contain equant crystals (e.g. Pinkerton and Stevenson, 1992; Lejeune and Richet, 1995; Pinkerton and Norton, 1995), but fails for magmas that contain tabular plagioclase (Sato, 2005). This failure of the relationship reflects the fact that both the intrinsic viscosity and Φ_m are not constant and are dependent on crystal characteristics such as crystal shape, crystal orientation, and crystal size distribution; these factors are not evaluated in the Einstein–Roscoe equation. Accordingly, we examined the nature of the dependence of intrinsic viscosity and Φ_m on these crystal characteristics.

The intrinsic viscosity is theoretically formulated for suspensions that contain ellipsoidal particles as a function of the particle axial ratio and orientation under an assumption that inter-grain interaction is negligible (Jeffery, 1922). We calculated the intrinsic viscosity using Jeffery's equation for a suspension with oblate rotating ellipsoids whose orientation distribution achieves a steady state. The steady-state distribution of particle orientations was determined as follows. For a simple shear flow with a flow direction parallel to the *x*-axis and a shear-gradient direction parallel to the *y*-axis in an x-y-z coordinate system, the orientation of the rotation axis of an ellipsoid rotating within the flow is described by the following equations (Jeffery, 1922):

$$d\theta/dt = G(1-\alpha^2)\sin\theta\cos\theta\sin\psi\cos\psi/(1+\alpha^2), \qquad (4)$$

$$d\psi/dt = G(\cos^2\psi + \alpha^2 \sin^2\psi)/(1+\alpha^2), \qquad (5)$$

$$C = \tan\theta \left(\cos^2\psi + \alpha^2 \sin^2\psi\right)^{1/2},\tag{6}$$

where θ is the angle between the *z*-axis and the axis of rotation, ψ is the angle between the *y*-*z* plane and the plane that contains both the axis of rotation and the *z*axis, *G* is the shear strain rate, α is the axial ratio (defined as long axis/short axis), and *C* is the orbit constant that characterizes the trajectory of a particle's rotation axis. The steady-state fraction of particles with orientations in the interval d ψ at ψ at time *t*, $p(\psi,t)$, is obtained by solving the following equation (Anczurowski and Mason, 1967):

$$\partial p(\psi, t) / \partial t = \partial [p(\psi, t) \partial \psi / \partial t] = 0$$
(7)

To determine the distribution of θ , it is necessary to determine the initial orientation and orbit constant of the analyzed particles. We assumed that the initial orientation and orbit constant of the particles were random and

that the orbit constant was independent of time. The steady-state distribution of θ was obtained numerically by integrating Eqs. (4) and (5) for 5000 random initial orientations and orbit constants. The calculated intrinsic viscosity obtained from the equation of Jeffery (1922) increased consistently with increasing particle axial ratio, and was approximated by the following equation:

$$[\eta] = 1.642 + 0.5124\alpha (R^2 = 0.9993, 3 \le \alpha \le 20), \qquad (8)$$

where α is the axial ratio as defined above. The intrinsic viscosity of a suspension that contains spherical particles should be 2.5 (Einstein, 1906, 1911) if intergrain interaction is negligible. However, the value derived from the Einstein-Roscoe equation is 4.167, on the assumption of $\Phi_m = 0.6$. This value is significantly higher than that determined theoretically for spherical particles, and corresponds to $\alpha = 4.9$ in Eq. (8). The high obtained value indicates that inter-grain interaction cannot be ignored. The effect of inter-grain interaction is difficult to determine quantitatively; however, such interaction is expected to increase with an increasing axial ratio of particles because the excluded volume of an oblate rotating ellipsoid increases with increasing axial ratio (Garboczi et al., 1995). The excluded volume is defined as the volume surrounding and including a given particle that excludes other particles (Onsager, 1949; Santalo, 1976). As the excluded volume increases, the probability of collision and hydrodynamic interaction among particles also increases, resulting in an increase in intrinsic viscosity. Consequently, the intrinsic viscosity increases with increasing axial ratios of particles, and Eq. (8) provides the minimum value of $[\eta]$ for an arbitrary axial ratio.

The maximum 3D disordered packing density, $\Phi_{\rm m}$, of uniform oblate rotating ellipsoids was calculated by Donev et al. (2004) for axial ratios up to 3.3. They showed that $\Phi_{\rm m}$ increases from 0.64 to 0.71 with an increase in the axial ratio from 1 to 1.5, and then decreases to 0.65 with a further increase in the axial ratio to 3.3. Although $\Phi_{\rm m}$ was not calculated for $\alpha > 3.3$, it is possible that $\Phi_{\rm m}$ is approximately 0.6 for a value as high as $\alpha = 10$; this inference is based on an extrapolation from the relationship between $\Phi_{\rm m}$ and $1/\alpha$ in Donev et al. (2004), and is consistent with the finding of Gay et al. (1969) that $\Phi_{\rm m}$ is ~0.55 for alumina suspensions with a shape factor (which is defined as the ratio of the surface area of a sphere with a volume that is equivalent to the surface area of the particle itself) of alumina particles of ~ 0.164 . The shape factor value of ~ 0.164 corresponds to an axial ratio of ~ 15 for an oblate ellipsoid. These results indicate that $\Phi_{\rm m}$ varies only within a narrow

range of 0.63 ± 0.08 for axial ratios of 1 to 15, and that such minor variations in Φ_m have an insignificant effect on relative viscosity. Φ_m increases with a decrease in the dispersion of particle orientations and increasing dispersion of particle size. The former trend is due to the fact that the packing structure becomes close to the maximum ordered packing structure, while the latter trend reflects the fact that the space between larger particles is filled by smaller particles. The effect of the former trend on relative viscosity is insignificant because Φ_m varies within a relatively narrow range from 0.64 to 0.74 (Donev et al., 2004); however, the effect of the latter trend is significant because Φ_m increases from 0.64 to 1 with increasing dispersion of particle size.

4.2. Effect of crystals on the relative viscosity at a steady state

The viscosity values measured in the present study for the crystal suspension at steady state plot close to the Einstein-Roscoe relationship (Fig. 11), even though the suspensions contained tabular plagioclase crystals (Fig. 6). This observation can be explained by the counterbalancing effects of the distributions of crystal shape and crystal size. With increasing crystal axial ratios, the intrinsic viscosity increases but $arPsi_{\mathrm{m}}$ remains largely constant. This results in an increase in the exponential term of Eq. (3). In contrast, $\Phi_{\rm m}$ increases with increasing dispersion of crystal size; this results in only a slight increase in the exponential term of Eq. (3) and a significant reduction in the effective volume fraction, $\Phi/\Phi_{\rm m}$. The crystal size distributions of plagioclase obtained in the present study were all dispersed (Fig. 5), resulting in values of $\Phi_{\rm m}$ that were greater than 0.6 and small values of $\Phi/\Phi_{\rm m}$. The relative viscosity increased because of the increase in the exponential term of Eq. (3), but decreased because of the reduction in $\varPhi/\varPhi_{\mathrm{m}}.$ The effect of particle shape on relative viscosity was counterbalanced by the effect of crystal size distribution, resulting in an approximately Einstein-Roscoe relationship between relative viscosity and the crystal volume fraction.

The relative viscosities of suspensions produced from Fuji 1707 high-alumina basalt and that contain thin tabular plagioclase crystals are provided by Sato (2005); these values are also plotted in Fig. 11. The suspensions contain only plagioclase crystals, except for the most crystalline sample obtained at 1130 °C. Plagioclase crystals within the suspension, which were thinner than those analyzed in the present study, showed a dispersed size distribution and a dominant axial ratio of approximately 8. The suspensions had relative viscosities that were significantly higher than those predicted by the Einstein-Roscoe relationship for an equivalent crystal volume fraction. This discrepancy may be related to the effect of crystal shape. The intrinsic viscosity has no upper limit and increases with the crystal axial ratio, whereas $\Phi_{\rm m}$ has an upper limit of 1. For crystal volume fractions of less than 0.3, Eq. (3) with $\Phi_{\rm m}=1$ and $[\eta]=5$ yields a similar relationship (within $\pm 5\%$) to that of the Einstein–Roscoe relationship with $\Phi_{\rm m}$ =0.6. This observation indicates that for intrinsic viscosities greater than 5, the relative viscosity deviates from the Einstein-Roscoe relationship because the intrinsic viscosity is too large to be counterbalanced by the effect of increasing $\Phi_{\rm m}$. The largest axial ratio that produces an intrinsic viscosity of less than 5 is estimated to be 6.5, as estimated from Eq. (8) for negligible inter-grain interaction. This value falls below 6.5 for non-negligible inter-grain interaction. The dominant length/width ratio of plagioclase in this study was less than 6.5, and the relative viscosity was consistent with the Einstein-Roscoe equation. The dominant length/width ratio reported by Sato (2005), however, was greater than 6.5, indicating that the intrinsic viscosity would have been too high to be counterbalanced by $\Phi_{\rm m}$, thereby resulting in a significant deviation from the Einstein-Roscoe relationship. The presence of olivine in all of the products of subliquidus runs conducted in the present study (in contrast to the absence of olivine in most of the run products of Fuji basalt) may also have contributed to the lower relative viscosity of the present experiment due to a decrease in the average aspect ratio of the crystals.

The results of the present study and those of Sato (2005) reveal a constraint on the applicability of the Einstein-Roscoe equation. This study demonstrated that the Einstein-Roscoe equation is applicable to magma with moderately thin crystals ($\alpha < 4$) when the crystal size distribution is dispersed; however, for thin crystals $(\alpha > 6.5)$, the relative viscosity deviates from the Einstein-Roscoe equation, even for a dispersed crystal size distribution. The axial ratio threshold that determines whether the Einstein-Roscoe equation is applicable or not lies between 4 and 6.5. As plagioclase crystals within natural magma generally show a dispersed size distribution (e.g. Cashman and Marsh, 1988; Mangan, 1990), this axial ratio threshold is applicable to most natural magmas. For example, Fig. 12 shows the CSD and length/width ratios of plagioclase crystals in the groundmass of the starting basaltic rock, as measured from X-ray maps. The mean length of the crystals is 108 μ m, with a standard deviation (1 σ) of 120 μ m and a mean length/width ratio of 4.9. As the CSD pattern was altered by post-emplacement annealing, the obtained standard deviation of crystal length is a minimum

plagioclase crystals in the groundmass of the starting basaltic rock. The mean length is 108 μ m (standard deviation $1\sigma = 120 \mu$ m) and the

value. Both the ratio of the standard deviation of the length to the mean length and the mean length/width ratio of the rock are similar to those of the present experiments. This correspondence indicates that the lava can be modeled by the Einstein-Roscoe equation.

4.3. Cause of thixotropic behavior

The suspensions containing plagioclase crystals showed thixotropic behavior (Fig. 8a), which we explained in terms of the time-dependent alignment of tabular plagioclase crystals. The time-dependent alignment of non-spherical particles such as tabular plagioclase crystals acts to reduce the viscosity of the suspension (e.g. Pinkerton and Norton, 1995; Deubener and Brückner, 1997; Yue et al., 1999). This occurs because the alignment of non-spherical particles reduces inter-grain interaction and increases the maximum packing fraction. Both of these factors contribute to reducing the relative viscosity, as discussed in Section

Fig. 12. CSD (a) and frequencies of length/width ratio (b) of mean length/width ratio is 4.9.



4.1, leading to thixotropic behavior. As shown in Fig. 7, plagioclase crystals analyzed in the present study tend to become aligned parallel to the direction of rotation. This alignment is considered to have evolved during the measurement as the plagioclase crystals were randomly oriented prior to shearing. This indicates that the thixotropic behavior resulted from the time-dependent alignment of plagioclase crystals during measurement. Such a view is consistent with the fact that the thixotropic behavior was observed for the suspension containing plagioclase crystals but not for the suspension that contained only olivine crystals (Fig. 8b).

Other causes of thixotropic behavior have been proposed in previous studies, including shear heating, the Weissenberg effect (i.e., climbing of the melt to the rotating rod), shear-induced crystallization, the migration of crystals along the rotation radius (e.g. Pinkerton and Norton, 1995), and the visco-elastic behavior of the pure melt phase. However, these mechanisms are unable to explain the thixotropic behavior observed in the present study. Shear heating acts to increase the temperature of magma, resulting in the thixotropic behavior of melt viscosity. The degree of increase in temperature induced by shear heating is, however, less than 1 °C, even for rhyolitic melt (Spera et al., 1988). Such an increase would be negligible for lower-viscosity basaltic melt because the degree of temperature increase is proportional to viscosity (Spera et al., 1988).

The Weissenberg effect induces a heterogeneous distribution of mass along the rotation radius; however, this phenomenon has only been observed at higher rotation rates (Ω >50 rpm) for the concentric cylinder system used in this study (see Sato, 2005). As the rotation rate applied in the present study was less than 50 rpm, the Weissenberg effect could not have occurred, and hence this mechanism was not the cause of the observed thixotropy. Crystallization is enhanced by shear deformation (e.g. MacDonald, 1953; Kouchi et al., 1986; Pinkerton and Norton, 1995), which affects the viscosity of the suspension via compositional changes to the melt, thereby leading to an increase in the volume fraction of crystals. Under the conditions of the present study, changes in the composition of the melt led to an increase in melt viscosity (Fig. 10). Both the change in composition and increase in the crystal volume fraction acted to increase the viscosity of the suspension with increasing lapse time. This phenomenon is known as rheopexy, which is the opposite of thixotropic behavior. Although it is possible that shear-enhanced crystallization occurred during the measurement, it had an insignificant effect on viscosity, and was therefore not the cause of the observed thixotropy.

The radial migration of crystals away from the inner rod, which resulted from secondary induced radial convection and/or the grain dispersive force (Bagnold, 1954), tends to result in a decrease in viscosity. However, crystals observed in the present study were evenly distributed along the rotation radius, as mentioned above (Fig. 4), and the Reynolds number of the flow in the concentric rotational cylinder of our experiment was four orders of magnitude less than the critical Reynolds number for radial convection, as calculated using the method of Van Wazer et al. (1963). This indicates that the radial migration of crystals did not occur in the present study and that this mechanism was not the cause of the observed thixotropy. Although the time-dependent development of viscosity is expected if pure silicate melt behaves as a visco-elastic material, melt can also behave as a Newtonian fluid under our experimental conditions, as described in Section 3.3. This is consistent with the fact that liquidus basaltic melt in the present experiments did not show time-dependent viscosity.

4.4. Cause of shear-thinning behavior

The basaltic magma that contained plagioclase crystals showed shear-thinning behavior at temperatures below 1170 °C (Fig. 9). There are two possible causes of such behavior, both of which are related to changes in melt-crystal texture that are dependent on the shear strain rate. The first possible cause is an increase in the dispersion of the orientation of tabular plagioclase crystals. As the shear strain rate increases, the long axes of tabular particles in a suspension tend to align parallel to the flow direction; consequently, the dispersion of particle orientations tends to decrease, resulting in a decrease in viscosity via both a decrease in the intensity of inter-grain interaction and an increase in the maximum packing fraction. Although this has not been examined experimentally in terms of magmatic suspensions, an analogous feature was observed for suspensions containing plate-like kaolin particles (e.g. Brown et al., 2000). A similar phenomenon probably occurs in magma that contains tabular plagioclase crystals, thereby leading to shear-thinning behavior.

The second possible cause of shear-thinning behavior is the shear strain rate-dependent decrease in intergrain interaction induced by the grain dispersive force (Ryerson et al., 1988). Ryerson et al. (1988) used this mechanism to explain the shear-thinning behavior of picritic magma that contained only equant olivine crystals. Ryerson et al. (1988) introduced contiguity as a measure of the intensity of inter-grain interaction, which was defined as the fraction of the internal surface area of a phase that is shared with grains of the same phase in a two-phase microstructure (Underwood, 1970; German, 1984). As the grain dispersive force is positively proportional to shear strain rate (Bagnold, 1954), the intergrain repulsive force increases with increasing shear strain rate, resulting in a reduction in contiguity. This decrease in contiguity results in a decrease in inter-grain interaction, leading in turn to shear-thinning behavior.

Other causes of shear-thinning behavior, including shear heating, the Weissenberg effect, and the viscoelastic behavior of a pure silicate melt phase (e.g. Pinkerton and Norton, 1995), are also rejected in terms of explaining the results of the present study. Both shear heating and the Weissenberg effect are rejected because their effects were negligible in the present experiment, as discussed in the previous section. The visco-elastic behavior of a pure silicate melt phase is also rejected because it behaves as a Newtonian fluid under our experimental condition, as mentioned in Section 3.3.

5. Concluding remarks

In this study, viscosity measurements were performed for a crystal suspension produced from alkali olivine basalt lava from the Higashi-Matsuura district, Southwest Japan. The relationship between relative viscosity, $\eta_{\rm r}$, and crystal volume fraction, Φ , measured in this study is close to the Einstein-Roscoe relation, even though the suspension contained tabular plagioclase crystals with a dispersed crystal size distribution. This observation is interpreted to indicate that the effect on relative viscosity of crystal shape was counterbalanced by the opposing effect of crystal size distribution. This indicates that the Einstein-Roscoe relationship represents the relative viscosity even for magma containing thin crystals ($\alpha < 4$) with a dispersed crystal size distribution. However, for suspensions that contain extremely thin crystals ($\alpha > 6.5$), the relative viscosity deviates from the Einstein-Roscoe relationship because the effect of crystal shape is too large to be counterbalanced by the effect of crystal size dispersion. These results indicate that the Einstein-Roscoe equation is applicable to magma that contains crystals with axial ratios of less than 5.3 (± 1.3) .

The magma that contained plagioclase crystals showed non-Newtonian behaviors such as thixotropy and shear thinning. The thixotropic behavior is explained by the time-dependent alignment of plagioclase crystals. The crystals gradually became aligned parallel to the flow direction during measurement, resulting in the observed thixotropic behavior. The shear-thinning behavior is explained by a decrease in the intensity of intergrain interaction resulting from a shear rate-dependent decrease in the dispersion of plagioclase orientations and/or the shear rate-dependent grain dispersive force.

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