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Experimental investigation of magma rheology at 300 MPa: From pure hydrous melt to 76 vol.% of crystals

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Abstract

The rheological behaviour of synthetic crystal-bearing magmas containing up to 76 vol.% of crystals ($0 \le \phi_S \le 0.76$) has been investigated experimentally at a confining pressure of 300 MPa and temperatures between 475 and 1000 °C at shear rates between 10^{-4} and $2 \times 10^{-3} \text{s}^{-1}$. Starting hydrated crystal-bearing glasses were synthesized from a dry haplogranitic glass ($Qz_{36}Ab_{39}Or_{29}$) and 2.5 wt.% water mixed with 0 (pure hydrous melt), 16, 34, 54, 65 or 76 vol.% of Al_2O_3 sieved ($45 < \emptyset < 90 \ \mu m$) crystals. Shear viscosity measurements were performed in torsion (simple shear) in a Paterson gas-medium apparatus.

For pure hydrated melt and for 16 vol.% of crystals, the rheology is found to be Newtonian. At higher crystal contents, the magmas exhibit shear thinning behaviour (pseudoplastic). The Einstein–Roscoe equation adequately estimates viscosities of the crystal-bearing magmas at low crystal contents ($\phi_S \le \sim 0.25$), but progressively deviates from the measured viscosities with increasing crystal content as the rheological behaviour becomes non-Newtonian. On the basis of a power–law formulation, we propose the following expression to calculate the viscosity as a function of temperature, crystal content and applied stress (or shear rate):

$$\label{eq:delta_def} \dot{\gamma} = A_0 (1 - \varPhi/\varPhi_m)^K \tau^{\left(1 + K_1 \varPhi^{K_2}\right)} \exp{\left(\frac{-Q}{RT}\right)},$$

where $\dot{\gamma}$ is shear rate (s⁻¹), τ is shear stress (MPa), Φ is the crystal volume fraction, T is temperature (K), $\Phi_{\rm m}$ is the relative maximum packing density, R is the gas constant, Q=231 kJ mol⁻¹ is the activation energy of the viscous flow and A_0 , K, K₁ and K₂ are empirical parameters. © 2007 Elsevier B.V. All rights reserved.

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1. Introduction

Viscosity of magmas is recognized as a critical factor controlling magma ascent and emplacement in the Earth's crust and eruptive styles of volcanoes. An important goal during the last

40 years has been to measure and model the viscosity of silicate liquids (Bottinga and Weill, 1972; Shaw, 1972; Neuville and Richet, 1991; Neuville et al., 1993; Hess et al., 1995, 2001; Dingwell et al., 1996, 2000; Hess and Dingwell, 1996; Dingwell and Hess, 1998; Scaillet et al., 2000; Whittington et al., 2000, 2001; Giordano and Dingwell, 2003; Toplis and Dingwell, 2004). However, although most natural magmas are crystal suspensions, studies on the effect of crystals on the rheological properties of magmas have long been scarce (Sabatier, 1959; Shaw, 1969; Murase and McBirney, 1973; Ryerson et al., 1988) until the late

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nineties (Bagdassarov et al., 1994; Lejeune and Richet, 1995; Pinkerton and Norton, 1995; Deubener and Bruckner, 1997; Lemke et al., 1999; Yue et al., 1999; Muller et al., 2003; Sato, 2005; Sonder et al., 2006; Ishibashi and Sato, 2007). Despite these new data, the existence, significance and importance of some rheological thresholds are still under debate (Renner et al., 2000; Burg and Vigneresse, 2002; Rosenberg and Handy, 2005) and a unified model for predicting the viscosity of magmas over their whole range of crystallisation is still to be proposed.

At low crystal contents, it is generally assumed that magmas can be approximated as Newtonian fluids (e.g. Pinkerton and Stevenson, 1992) and that their viscosity can be estimated according to the Einstein–Roscoe equation (Einstein, 1906, 1911; Roscoe, 1952, 1953):

$$\eta = \eta_0 (1 - \Phi_{\rm s}/\Phi_{\rm m})^{-k} \tag{1}$$

where η_0 is the viscosity of the melt, Φ_s the crystal fraction, Φ_m the maximum packing fraction of crystals and k an adjustable parameter. However, the value of Φ_m depends on the size, shape and distribution of crystals (Chong et al., 1971, Jefferey and Acrivos, 1976, Lejeune and Richet, 1995) and has been extensively debated in the literature. For suspensions of particles in a fluid, values of Φ_m in the range 0.32 to 0.71 have been measured (Gay et al., 1969), while values between 0.5 and 0.7 are considered realistic for magmas (Marsh, 1981). Moreover, Lejeune and Richet (1995) highlight that Φ_m and k are interdependent when adjusting (Eq. (1)) to experimental data.

At higher crystal contents, the rheological behaviour of magmas is no longer Newtonian and two phenomena are classically described and discussed. First, many studies report that the development of a 3-D framework of crystals inhibits flow movement at low stresses, causing a yield strength to appear (e.g. Gay et al., 1969; Pinkerton and Norton, 1995). Studies based on the percolation theory show that the value of the critical (crystal) volume fraction Φ_c for the onset of crystal or particle networks, which depends on particle shape and size distributions and on the particles' overall orientation distribution (Pike and Seager, 1974; Garboczi et al., 1995; Lorenz and Ziff, 2001; Saar et al., 2001), can be as low as Φ_c =0.08 for highly anisotropic particles (Saar et al., 2001; Saar and Manga, 2002). Second, apparent viscosities depend on shear rate for highconcentration suspensions (Krieger, 1972; Jefferey and Acrivos, 1976; Lejeune and Richet, 1995; Sato, 2005; Sonder et al., 2006) and the nature of this dependence is still not well understood.

In the present study, we investigate the rheological behaviour of synthetic magmatic suspensions containing up to 76 vol.% of crystals. Viscosity measurements are conducted under a confining pressure (300 MPa) in simple shear (torsion) in a servo-controlled internally-heated high-pressure deformation vessel (Paterson apparatus). The effect of temperature (475 °C \leq $T \leq$ 600 °C) and of shear rate ($10^{-4}~{\rm s}^{-1} \leq \dot{\gamma} \leq 2 \times 10^{-3}~{\rm s}^{-1}$) on viscosity are characterized as a function of crystal content. Our data are compared with previous studies and are used to test the model proposed by Gay et al. (1969) for the viscosity of crystal-bearing magmas. The evolution of viscosity with increasing crystal content is discussed. Moreover, the development of microstruc-

tures studied in a companion paper (Arbaret et al., 2007) is presented and correlated with the rheological behaviour.

2. Experimental methods

2.1. Starting material and analytical techniques

Starting crystal-bearing hydrated glasses with a crystal fraction of up to 76 vol.% $(0 \le \phi_S \le 0.76)$ were prepared following the procedure of Martel et al. (2001). A finely ground powder of dry haplogranitic glass (SiO₂: 78.8, Al₂O₃: 12.4, Na₂O: 4.6, K₂O: 4.2 wt.%, microprobe analyses) prepared by Schott AG (Germany) was mixed with 0, 16, 34, 54, 65 or 76 vol.% of Al₂O₃ sieved (45< ϕ <90 µm) crystals (insulating powder from Friatec AG, Germany—Corundum). The glass+crystal mixture was loaded in large gold capsules (20 mm in diameter and 35 mm in length) with demineralised water. The amount of added water was calculated to obtain 2.5 wt.% in melt in the final product independently of solid fraction. Capsules were arc-welded while surrounded by liquid nitrogen in order to prevent water loss. Melt hydrations were performed at IST Orléans in a large bore furnace of an internally-heated pressure vessel (IHPV) at 1000 °C and 100 MPa during 1 month, then isobarically quenched.

Observation of polished sections of the starting products shows that insignificant chemical reaction occurred between the melt and the crystals during hydration due to the subaluminous composition of the melt (molar $Al_2O_3/(Na_2O+K_2O)=1.025$) (Martel et al., 2001). As expected due to the limited solubility of aluminium in subaluminous melts (Holtz et al., 1992), microprobe analyses show a small enrichment in Al₂O₃ of the melt (less than 1.5 wt.%) in samples containing corundum crystals compared to the crystal-free hydrous melt. After hydration, all samples contain some residual bubbles (Fig. 1). For the crystalfree starting product (HPG-10), the volume fraction of bubbles (Φ_b) was determined by image analysis of polished sections (Fig. 1a) to be less than 0.01. For crystal-bearing starting products, determination of ϕ_b is not very accurate because the polishing procedure causes plucking of alumina grains, and it is often impossible to distinguish plucked grains from bubbles (Fig. 1c).

The $\rm H_2O$ content of the starting products was controlled by Karl–Fisher titration (KFT) and by Fourier-transform infrared (FTIR) on some samples (see Table 1). The precision of KFT analyses is known to be ± 0.15 wt.% $\rm H_2O$ and depends mainly on the amount of sample available and on measurement duration (Behrens, 1995). For crystal-bearing samples, it also requires a correction for crystal content.

FTIR analyses were performed on doubly polished glass plates using a Nicolet 760 Magna FTIR spectrometer. Spectra were collected with 128 scans, with a resolution of 4 cm $^{-1}$, a gain of 4, a CaF $_2$ beamsplitter, visible light, and a liquid N $_2$ cooled MCT/A detector. The concentrations of OH and H $_2$ O species were determined by the peak heights of the absorption bands at 4500 and 5200 cm $^{-1}$, respectively, using molar absorptivities from Behrens and Nowak (2003) for haplogranitic compositions. Glass densities were determined using the model of Richet et al. (2000).

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