Viscoelastic behaviour of basaltic lavas

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Abstract

The rheological properties of basaltic lavas from Etna, Hawai'i and Vesuvius have been	
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investigated at temperatures between ~500 and 1150 °C using a small strain oscillatory shear.	
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The viscoelastic response of the lavas to small, forced, sinusoidal torques (<10 ⁻⁵ Nm) at	Deleted: oscillatory
frequencies between 0.002 and 20 Hz was measured. A purely viscous regime was only	
approached during experiments with Hawai'i samples. These experiments indicated that at	
temperatures between ~1070 and 1130 °C, strain rate independent viscosities (>10 ⁹ Pa s)	
could be measured at strain rates less than $\sim 10^{-2}$ to 10^{-1} s ⁻¹ . At 800 °C, temporal variations in	
complex shear modulus and internal friction suggest that, over durations of up to 120 hours,	
structural adjustments were occurring within some of the samples. This time-varying	
behaviour of lava samples <u>may be</u> attributed to the slow closing (healing) of microcracks <u>and</u>	Deleted: is
small pore spaces resulting in the apparent stiffening of lava samples under annealing. Thus,	
those parts of lava flows that undergo slow cooling <u>will</u> have more elastic properties. Regions	Deleted: ing
which cool faster possess smaller shear moduli and higher internal friction due to thermal	
microcracking and less cohesion between crystals and the bulk glassy matrix.	Deleted: grains

Key words: basalt lava, Etna, Vesuvius, Hawai'i, shear modulus, shear viscosity, oscillatory rheology

1. Introduction

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their eruption temperature, lavas display viscoelastic behaviour and, at temperatures below the glass transition temperature, T_{g} , of their glass matrix, they are anelastic (Sakuma, 1953; Bagdassarov, 2000). Field measurements carried out at high stresses (~10³ Pa) and typical eruption temperatures indicate that basaltic lavas have a rather moderate viscosity (>10² Pa s)

and an appreciable static yield-strength ($\sim 10^2$ to 10^3 Pa) (Shaw et al., 1968; Pinkerton and Sparks, 1978; Pinkerton and Norton, 1995; Norton and Pinkerton, 1997).

Experiments designed to characterise the high temperature anelastic and viscoelastic behaviours of glassy, crystalline and partially molten rocks are based on measurements of elastic modulus (C^*) and internal friction (C^{-1}) . Traditionally, these measurements are performed using an inverted torsional pendulum (e.g. Day and Rindone, 1961; Gueguen et al., 1981; Weiner et al., 1987; Versteeg and Kohlstedt, 1994) or by forced torsional oscillation (Beckhemer et al., 1982; Jackson and Paterson, 1987; Bagdassarov and Dingwell, 1993; Gribb and Cooper, 1998). The forced torsional oscillation method used in this work has previously been used to study the behaviour of both rocks (Berckhemer et al., 1982; Jackson and Paterson 1987; Gribb and Cooper, 1998; Bagdassarov and Dorfman, 1998) and glasses (Bagdassarov and Dingwell, 1993; Bagdassarov et al., 1993,1994). This technique allows the magnitude of the complex shear modulus and angle of internal friction to be measured over a range of temperatures and frequencies.

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2. Apparatus

The experimental method consists of exerting small strain oscillatory torsion deformation on cylindrical samples. The equipment (described in detail previously (Berckhemer et al., 1982; Kampfmann and Berckhemer, 1985, Bagdassarov and Dingwell, 1993)) exerts a small sinusoidal torque (of amplitude $\sim 10^{-3}$ N m) to the end of a cylindrical sample (8 mm in diameter, ~ 20 to 30 mm in length). A simple schematic of the device is shown in Fig. 1a. The <u>sinusoidal torque applied to the sample is generated using a pair of</u> electromagnets (two microphone-type coils) connected to a synthesiser via a power amplifier. The sample is fixed between two aligned alumina rods, onto which two sets of light aluminium wings are also attached. The angular deformation across the sample is measured by pairs of capacitive pick-ups which respond to the movement of pure iron plates located at the ends of the aluminium wings. The capacitive signal is detected and amplified using a 5 kHz-frequency bridge which is sampled using a PC. Calibration of the equipment has been described previously (Bagdassarov, 2000), with shear modulus measurements being accurate to 3 to 5 % (due to thermal drift of the calibration at high temperatures).

Although the mechanical design of the equipment has not changed from that used during previous work, the data acquisition hardware and processing software have been significantly improved. For each measurement, data are collected over two periods of the torsional oscillation (Fig. 1b). Data are sampled at up to 10 kHz, allowing 1000 samples per channel to be acquired at the highest frequency used during experiments (20 Hz). At torsional oscillation frequencies of 2 Hz or lower the number of samples per channel is limited to 10,000. Sinusoids are automatically fitted to the collected data using a Levenberg-Marquardt algorithm, and the shear modulus and phase difference between the applied torque and the angular displacement across the sample are calculated from the phase and amplitude parameters of the fitted curves.

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Experiments were carried out over the frequency range 0.002 to 20 Hz (at approximately 0.3 log intervals) and at temperatures between ~500 and 1150 °C. At high temperatures, low frequency measurements were prevented by the onset of non-linear sample response. This was revealed by Fourier analysis of the data indicating the presence of harmonics of the torsional driving frequency. Automation of sampling and processing allows repeated measurements and at high frequencies an average of twenty measurements was used for most temperature-frequency points. At low frequencies, individual measurements take up to 17 minutes, so fewer experiments were averaged.

During experiments the furnace was purged with a flow of Ar gas (5 cm³ s⁻¹). Inspection of the samples after experiments showed that oxidation (as indicated by growth of Fe-(Ti)-oxides) had taken place only on the surfaces of the samples. Temperatures were recorded using Pt-PtRd (S-type) or Ni-NiCr (K-type) thermocouples. Direct measurements of the temperature field inside the furnace indicated that temperatures were reduced by up to 15 °C at distances of 10 mm from the hottest point. Although this spatial sensitivity implies that recorded values were only accurate to ~15 °C as indicators of the sample temperature, relative temperature changes within any one experiment are much better constrained (\pm 3 °C at 1000 °C).

The data collected at each temperature-frequency point allowed calculation of the magnitude of the complex shear modulus $G^*(\omega, T)$, and the phase shift $\varphi(\omega, T)$ between the applied torque and the resultant angular strain of the sample, where ω is the angular velocity (equal to 2π multiplied by the applied frequency). From these results, the real, G', and the imaginary, G'', parts of the complex shear modulus, the internal friction, Q^{-1} , and the complex shear viscosity, \underline{n} can be calculated from

 $G^* = G' + iG'' = G^* \cos(\varphi) + iG^* \sin(\varphi) \tag{1},$

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$$Q^{-1} = \tan(\varphi) = \frac{G''}{G'}$$
(2),

$$\eta = \eta' - i\eta'' = \frac{G''}{\omega} + i\frac{G'}{\omega}$$
(3),

(e.g. Nowick and Berry, 1972). The zero-rate shear viscosity (macroscopic viscosity), \underline{n}_{0} , may be obtained from the frequency dependence of G'' by

$$\eta_0 = \lim_{\omega \to 0} \frac{G''(\omega)}{\omega} \tag{4}$$

(Marin, 1998),

2.1 Sample bonding

During experiments it is essential that each end of the sample is securely bonded to the alumina rods. In order to do this efficiently small conical grips (angle ~1°, length 4 mm) were machined at both flat ends of the sample with a diamond tool. Complementary mating grips were produced in the alumina rods and samples were cemented between the rods with a high temperature cement (Polytec[®]). The assembly was placed in the torsion apparatus and the sample was then sintered to the rods for 2 hours at 150 °C and then for 24 hours at 500 °C, under an axial load of ~8 N (e.g. Berckhemer et al., 1982). Measurements carried out using a dummy sample of Al_2O_3 have demonstrated that the effect of the cement on phase delay measurements was less than 10^{-3} rad. (Bagdassarov and Dorfman, 1998).

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2.2 Size and shape factors

When temperatures increased during experiments, thermal expansion of the sample and the alumina ceramic rods was accommodated by a spring located at one end of the apparatus. At temperatures sufficiently high for the sample to deform, some of the **Deleted:** If these parameters are frequency dependent, then the material is viscoelastic and a more complex relation must be found for stress and strain (e.g. Bagdassarov and Dingwell, 1993).

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accumulated stress dissipates by flow shortening of the sample. Changes in sample length were calculated from micrometer readings taking at the spring (to a precision of ~0.02 mm) and <u>the</u> correspondingly changed sample diameter (calculated by assuming conservation of _ sample volume) were then used to calculate the material properties. However, samples recovered after experiments have shown that flow deformation was not continuously distributed through the samples but was concentrated in the centre, producing distorted, barrel-shaped, cylinders. This is a consequence of temperature gradients across the sample and due to it being supported at both ends. Thus, despite efforts to account for changes in the sample shape, the deviation from a cylindrical form introduced errors (<2 %) in the assumed diameter of the sample, once sample shortening has started.

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2.3 Sample description

The experiments were carried out on samples of basaltic lavas collected from three different volcanoes, Etna, Hawai'i and Vesuvius.

a. Etna. Two samples from Etna were collected from lava erupted in 1992 which had ponded after overflowing from a skylight in the Valle del Bove. One of these samples was from the top surface and one was taken from the base (~10 cm down from the surface) and thus represent samples with different cooling regimes. <u>Digital images of polished sections</u> were obtained through an optical microscope and computer analysed, omitting corrections for <u>3D effects</u>. The surface sample has smaller vesicles (~0.2 to 0.5 mm, 15 to 20 vol.%) and smaller crystal content (~20 to 30 vol.%) than the basal sample (~20 vol.% of 1 to 2 mm vesicles and ~30 vol.% phenocrysts. <u>Images of polished thin sections</u> are shown in Fig. 2. A further sample was collected from near the south east cone in 1999. This sample was taken from the least vesicular area found in a recently emplaced flow near hornito H3 (Calvari and Pinkerton, 2002) at the top of the active flow field.

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b. Hawai'i. The Hawai'ian basalt was sampled in the east rift eruption zone of Kilauea, Hawai'i from a lava flow from a pahoehoe toe in September, 1984 (eruption temperature ~1147 °C). This pahoehoe lava flow corresponds to the episode 25 of the eruption Pu'u 'O'o of Kilauea Volcano. Its bulk chemical composition has been presented elsewhere (Garcia et al., 1992). The chemical composition of the groundmass glass obtained by microprobe analysis differs from the bulk rock composition in SiO₂ (52.3 wt%) and CaO (9.2%) content (Bagdassarov, 2000). The vesicularity estimated from 2-D image analysis varies from 46.8 to 57.6 vol.%, or ~50 vol.% when calculated on the "dry" density rock basis. The sample has about ~10 vol.% of olivine quenched from magma during sampling and a few percent of other phenocrysts.

c. Vesuvius. The Vesuvius samples were collected from the 1834 flow at Cava Ranieri in the national protected area of Terzigno approximately 6.3 km ESE of the central cone of Vesuvius by the group from University College of London. Chemical analysis of the samples is given in Belkin et al. (1993). The same sample was also used by Rocchi et al. (2002) in experiments to determine Young's modulus and tensile strength.

3. Results

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for basaltic glass (Ryan and Sammis, 1981), For Pu'u 'O'o paheohoe lavas, the glass transition and melting temperatures are 655°C and 1149°C respectively (Burkhard, 2001). In contrast, at

temperatures below 1000 °C, the internal friction of the basal sample (Fig. 3d) is a strong power law of frequency, Q⁻¹ ∝ ω^{-0.35}. The response of a purely elastic material would be
independent of frequency, and this frequency dependence indicates a partially viscous
response even at ~600 °C.

Similar experiments carried out on cores cut from the 1999 Etna sample not only reproduced the internal friction peak at low temperatures (~700 °C) but demonstrated considerable temporal variations in the results (Fig 4). Over periods of up to ~120 hours, the measured shear modulus increased and internal friction decreased, After annealing, the 1999 samples produced similar shear modulus and internal friction results (Fig. 4c, d) to those of the 1992 Etna crustal material. At temperatures >1100 °C, Etna samples demonstrated an increase of fluidity of several orders of magnitude and it was not possible to maintain them in the torsion apparatus.

The experiments carried out on the Hawai'i sample were extended to higher temperatures because of the <u>higher temperature of the</u> sample's <u>softening point</u>. Similar temporal variations to those observed with the Etna lavas were found and are shown in Fig. 5 at a temperature of 1102 °C. The results are broadly similar to those of the Etna samples with shear moduli of ~18 GPa decreasing to ~0.2 GPa as <u>sample fluidity starts to rapidly increase</u>.

The results from the experiments carried out on the Vesuvius sample are given in Fig. 6. During sample annealing the shear modulus increased but to a lesser extent than in Etna samples. An increase in tensile strength and Young's modulus after annealing was also reported during bending experiments (Rocchi et al., 2002). At the highest temperature attained during experiments (1132 °C) the shear modulus was less than 0.5 GPa and practically frequency independent. With a further increase of temperature the sample became too fluid for it to be held within the torsion apparatus.

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Deleted: Similar changes have been previously observed in thermally cycled quartzite, granite and sandstones (Johnson and Toksöz, 1980; Lu and Jackson, 1998) and are thought to be due to the presence and healing of microcracks. For example, rapid thermal cycling of granite and diabase samples from room temperature to ~800 °C results in order of magnitude increases in crack porosity and a factor of 2 increase in Q^{-1} due to the thermal production of cracks (Johnson and Toksöz, 1980). Thus we interpret our data, which indicate an increasingly elastic response with time, as demonstrating the effects of healing microcracks in the samples. It is likely that the microcracks were originally created due to thermal stresses during cooling. Progressive volatile loss could also be responsible for strengthening the samples, however this could not be used in order to explain the power law dependence of internal friction in the 1992 Etna samples at low temperatures.

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4. Discussion

Measurements of the complex shear modulus (G_{\star}^{*}) and internal friction (Q^{-1}) of lava, samples from Etna, Vesuvius and Hawai'i show that they possess an appreciable shear modulus (>0.5 GPa) and an internal friction generally less than 1, at temperatures where field measurements indicate viscosities of ~10³ Pa s. For comparison, polyerstalline rocks at room temperature have typical values of unrelaxed shear modulus, G_{∞} of about 50 GPa (Jackson, 1993) and 25 GPa is representative for silicate glasses (Bagdassarov et al., 1993). Between room and high temperatures prior to the onset of melting, values of Q^{-1} vary between ~10⁻³ and 10⁻¹ (Manghnani et al., 1981; Weiner et al., 1987; Jackson, 1993; Bagdassarov, 2002).

Natural silicate melts and glasses show a linear viscoelastic behaviour in the glass transition temperature range (e.g. Bagdassarov and Dingwell, 1993; Bagdassarov et al., 1993). With increasing temperature and decreasing strain-rate, one expects progressively decreasing shear modulus and increasing internal friction. However, the viscoelastic behaviour of lavas is often controlled by the presence of deformable (vesicles) and non-deformable (crystals) heterogeneities. Viscosity increases with increasing crystal content but decreases with increasing vesicle content (Bagdassarov and Dingwell, 1992) and the presence of these macroscale obstacles extend the shear stress relaxation spectrum towards longer relaxation times. Shear modulus relaxation can be extended over several hundred °C above the T_{e} of a basaltic groundmass glass (~655 °C) up to ~1100 to 1150 °C. Over the same range, the frequency dependence of shear modulus and internal friction becomes weaker, for example, in lavas $Q^{-1} \propto a^{0.35}$, compared to $a^{0.5}$ for silicate melts.

<u>The shear modulus and internal friction of our lava samples varied significantly with</u> <u>annealing time with, at temperatures between ~500 and 1000 °C, some samples becoming</u> <u>increasingly stiff and elastic (Fig. 4). Similar temporal variation of elastic modulus and</u> <u>internal friction</u> have been previously observed in thermally cycled quartzite, granite, <u>dunite</u>

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and sandstones (Johnson and Toksöz, 1980; Jackson, 1993; Lu and Jackson, 1998) and is thought to be due to the presence and healing of microcracks and pores. For example, rapid thermal cycling of granite and diabase samples from room temperature to ~800 °C results in order of magnitude increases in crack porosity and a factor of 2 increase in  $Q^{-1}$  due to the thermal production of cracks (Johnson and Toksöz, 1980). Jackson (1993) interpreted the observed increase in shear modulus and decrease in internal friction with the increasing annealing time to be a result of the reduction of porosity and increase of the grain boundary cohesion by a sintering processes. Progressive decreases in  $O^{-1}$  have been associated with a decrease of centres of stress concentration. Thus, we interpret our data, which indicate an increasingly elastic response with time, as demonstrating the effects of healing microcracks and thermally produced pore space (not vesicles) in the samples. It is likely that the microcracks were originally created due to thermal stresses during rapid cooling of the lava flows, however, we cannot exclude the possibility that they could have been produced during sample preparation or the subsequent heating. In thin sections, microcracks within crystals and between crystals and the groundmass can be observed to have undergone some healing

after annealing (Fig. 7),

The production of microcracks suggests that rocks which have undergone slow cooling will possess higher shear moduli and have higher strengths than rocks which cooled more rapidly. Thus, before annealing, samples taken from the centre of a cooled flow would be expected to be more elastic than samples taken from the top of a flow. Under annealing at high temperature, the crack healing process consists of cracks pinching off and healing to a pore-like shape before subsequent decreasing of the pore diameter (Atkinson, 1984). During this period, the shear modulus may significantly increase and the internal friction decrease. The time dependence of changes in crack lengths can be described as an Arrhenius function of temperature and, as a first approximation, a linear dependence between shear modulus and a crack density parameter may be assumed (O'Connel and Budiansky, 1974). In Fig. 8a the

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results of annealing on the complex shear modulus of the 1999 Etna lava sample are presented as a function of time, *t*. By fitting these data with an exponential time-dependence.

$$|G^*| = (G_{\infty} - G(0)) \left\{ 1 - e^{-\left(\frac{t}{\tau}\right)} \right\} + G(0)$$
 (5)

where  $G_{\infty}$  is the stable shear modulus at any temperature, G(0) is the shear modulus at t=0, the characteristic time,  $\tau$ , of the crack healing process may be estimated. The results of the fitting are presented in Fig. 8b in the form of an Arrhenian dependence of  $\tau$ , yielding an activation energy of  $150 \pm 20 \text{ kJ mol}^{-1}$ . According to Atkinson (1984) this value should relate to the activation energy of the effective diffusion coefficient on grain boundaries of species participating in the pinching process at microcrack tips. For example, the activation energy of crack healing in dry quartz at T < 600 °C is about 80 to 85 kJ mol⁻¹ (Atkinson and Meredith, 1987), corresponding to the activation energy of water diffusion in quartz (~60 kJ mol⁻¹, Dersch et al. (1997) or ~100 kJ mol⁻¹, Brady (1995)). At high temperatures, crystal lattice diffusion is the rate controlling mechanism of grain boundary diffusion. Thus, the observed activation energy for the time dependence of shear modulus may be associated with an inter-diffusion of Ca, Mg and alkaline species between olivine, clinopyroxene and plagioclase crystals and basalt glass, with activation energies between 130 and 180 kJ mol⁻¹ (Brady, 1995). Alternatively, our results are also consistent with a self-diffusion of Si and O, network formers in basaltic glass, with an activation energy of 170 kJ mol⁻¹ (Lesher et al., 1996).

Alternative mechanisms which could contribute to the increase of shear modulus during annealing of samples are redox and re-crystallisation processes in the basalt groundmass glass. Progressive volatile loss could also be responsible for strengthening the samples, however this cannot explain the power law frequency dependence of internal friction in the 1992 Etna samples at low temperatures. A contribution to shear modulus increase from sample oxidation would be due to the production of interface-controlled intergrowths of pyroxene dendrites and Fe-(Ti)-oxides between 850 and 940 °C (Burkhard, 2001). However,

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the activation energy of this process is ~100 kJmol⁻¹, which is less than that observed from the time variation curves of shear modulus. The oxidation kinetics of basaltic glass are determined primarily by the diffusion of divalent cations  $Ca^{2+}$  and  $Mg^{2+}$  to the surface of the sample, which is charged compensated by the inward flux of electron holes (Cooper et al., 1996). The activation energy of this process at temperatures below  $T_g$  is 210 kJmol⁻¹. The calculated maximum depth (~1 µm at 600 °C and ~20 µm at 800 °C) of surface oxidation by using the average divalent cation diffusion coefficient (Cooper et al., 1996) is small compared with the size of lava samples used in torsion experiments and is in agreement with observations made from thin sections of the samples. It should also be noted that these rates relate to initially non-oxidised basalt glass samples and the Java samples should be considered as slow cooled, partially crystallised glasses, with Jong oxidation histories.

At T> 920 °C, the bulk crystallisation of basaltic glass and growth of plagioclase crystals may also contribute to the increase of stiffness of annealed lava samples, but the timescales required are generally much longer than those of the observed changes in shear modulus. For example, a noticeable growth of plagioclase and Fe-Ti-oxides has been observed at 850-934 °C after >200 h of annealing basalt glass (Burkhard, 2001) and this process is likely to have continued for an order of magnitude longer time. In contrast, shear modulus changes occurred much more rapidly in our experiments and generally ceased on a timescale of <100 hours. Thus, neither redox or re-crystallisation processes are believed to significantly contribute to the measured changes in the rheological properties of the samples

Converting the shear modulus and internal friction data into viscosity (Eq. 3) shows that both the Vesuvius and Etna samples maintained a frequency dependent real component of their viscosity ( $\underline{n}_{i}$ ) up to the highest temperatures attainable during the experiments (Fig. 9). Therefore, no "Newtonian" viscosity can be given for these temperatures at low strain rates. In contrast, the Hawaiian sample demonstrated marked decreases in the frequency dependence of  $\underline{n}_{i}$  at high temperatures and low frequencies (Fig. 10a). The main difference

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	samples after experiments revealed the textural changes only in thin surface layer (few µm) and not in
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between the low strain rate behaviour of, on the one side, Etna and Vesuvius basalts, and on the other, that from Hawai'i, lies in the relative proportion of crystals and vesicles. The small strain-rate and small stress viscosity,  $\eta_0$ , measured for the Hawaiian sample represents a creep-type mechanism underlying the Bingham yield stress which is expressed under conditions of slow, small scale deformation, where the internal structure of the lava sample remains intact. The presence of non-deformable crystals results in increases of this effective viscosity, but also increases the Bingham yield stress (Bagdassarov et al., 1994). Higher ratios of crystals to vesicles in Etna and Vesuvius lavas increase the Bingham creep-viscosity,  $\eta_0$ , and shift it toward smaller strain-rates which were unattainable in our torsion experiments. In contrast, the Hawai'ian lava sample, with a smaller crystals to vesicles ratio, possessed a smaller Bingham creep-viscosity which was observable at the lower strain rates of the torsion apparatus (10⁻³ s⁻¹).

If a material has a strain rate dependent <u>rheology</u>, then two different viscosities, can be assigned,  $\eta_0$  and  $\eta_{\infty}$ , which relate to the low and high strain rate limits respectively. The Cross model then gives the viscosity as

$$\eta = \eta_{\infty} + \frac{\eta_0 - \eta_{\infty}}{1 + \left(K \cdot \gamma\right)^m}$$

where *m* and *K* are empirical constants and  $\gamma$  is strain rate (e.g. Barnes, 1999), with *m* characterising a stretching parameter for shear stress relaxation during the viscoelastic transition. For a Maxwell body rheology, *m*=2. In the operational window of the oscillatory shear apparatus, only  $\eta_0$  can be measured. The high strain rate viscosity,  $\eta_{\infty_3}$  is interpreted as the effective viscosity of a fluid at high strain-rates or stresses. In the case of lavas, it corresponds to the results of field or laboratory experiments in which high strains induced by the measurement disrupt the internal structure of the lava.

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The high temperature data in Fig. <u>10a</u> have been fitted with Cross curves (assuming
that $\underline{\eta}_0 >> \underline{\eta}_{p_0}$ and using $m=1$ ) and the values of $\underline{\eta}_0$ obtained are plotted in Fig. <u>10b</u> along with
other viscosity measurements made with a parallel plate viscometer (a $B\ddot{a}hr^{\mbox{\tiny $^{(n)}$}}$ high
temperature dilatometer, University Bayreuth (Bagdassarov, 2000)) and rotational
viscometers (Shaw, 1968, 1969; Ryan and Blevins, 1987; Ryerson et al., 1988; Pinkerton et
al., 1995). The parallel plate viscometer uses shear rates of 10 ⁻⁵ to 10 ⁻⁷ s ⁻¹ , compared with
equivalents of $\sim 10^{-2}$ to $10^{2}$ s ⁻¹ for the torsion apparatus. The activation energy for viscous flow /
obtained by the dilatometer and torsion experiments (given by the gradient of the dashed lines
in Fig. <u>10b</u> ) is $\sim 950 \pm 5 \text{ kJ mol}^{-1}$ . The small offset (0.67 log units) between the two lines is
due to the effect of the volume (or bulk) viscosity of a porous sample, $\eta_{\nu}$ , which affects the
parallel plate deformation results of the dilatometer experiments. The pure shear deformation
used in the dilatometer gives a compressional viscosity, $\eta_c$ , which is usually related to the
shear viscosity by $\eta_c/3$ . However, this is valid only for incompressible samples (which have
an infinite volume viscosity), and is therefore not applicable to material with ~50 % porosity,
which will have a finite volume viscosity. For high porosity materials, the relationship
$\underline{\qquad \qquad } \eta_c = \eta_v + \frac{4}{3}\eta_s \underline{\qquad \qquad } (7)$
is appropriate (e.g. Bagdassarov and Dingwell, 1992). Thus measured values should be
corrected for a finite volume viscosity, the value of which is unknown. From Fig. 10b the
compressional viscosity data from the dilatometer experiments are related to the simple shear

viscosity results of the torsion experiments,  $\eta_s$ , by  $\eta_c \approx 3 \times 10^{0.67} \eta_s$ , or  $10^{1.15} \eta_s$ . Applying Eq. 7

viscosity as function of porosity has been considered elsewhere (Prud'homme and Bird, 1978;

thus gives  $\underline{\eta_v} \approx 12.7 \eta_s$  (at ~50 % porosity). The theoretical dependence of the volume

Aksel, 1995), with the classical formula

 $\underline{\qquad} \eta_v = \frac{4}{3} \cdot \frac{\rho^2}{\rho_0(\rho_0 - \rho)} \cdot \eta_s^0 \underline{\qquad}$ 

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being derived for small concentrations of vesicles in a fluid, where  $\rho_0$  is the density of fluid,  $\rho_0$  is the density of the suspension and  $\eta_s^0$  is the viscosity of the fluid without vesicles. <u>Converting</u>  $\eta_s^0$  to  $\eta_s$  by  $\eta_s^0 \approx 0.2 \eta_s$  (for ~50 % porosity, Fig. 5, Rust and Manga 2002), gives the theoretical relation of  $\eta_v \approx 3.3 \eta_s$ . The discrepancy between our result and this theoretical estimation may be easily attributed to experimental errors in viscositiy as well as the nonspherical shape of some vesicles and their non-uniform size distribution in the lava samples.

Alternatives to the Cross model were reviewed by Yue and Brückner (1994) who suggested a strain rate dependent viscosity model depending on three phenomenological constants. Their model is appropriate for parallel plate viscometry in which significant strain increases during experiments can produce thermal effects and violations of non-slip boundary conditions. However, in the torsion experiments the maximum torque was  $10^{-3}$  N m, and maximum angle deformation  $10^{-5}$  rad, resulting in a negligible heat production (< $10^{-7}$  J s⁻¹) even at the highest strain rates used.

Field measurements, corrected to unit strain rate, have reported significantly smaller shear viscosities for Hawaiian basalts. Shaw et al. (1968) measured plastic viscosities of 650 to 750 Pa s at 1130 °C and Pinkerton et al. (1995) measured viscosities of 234 to 548 Pa s at 1146 °C (Fig. 8b). Unit shear rate results from the torsion experiments (as given by the fitted Cross models) are five orders of magnitude greater than these field measurements. For Mount St. Helens dacite, Pinkerton and Stevenson (1992) demonstrated that a combination of factors was responsible for a 10 order of magnitude variation within measured and calculated apparent viscosities at sub-liquidus temperatures. For Hawaiian basalt, although differences in volatile content, crystallinity and vesicularity exist between the experimental and the fieldwork samples, the main difference in the results (Fig. <u>10b) is due to the magnitude of the</u> strain used, with rotational viscometry producing high strains and torsion and dilatometer experiments producing small strains

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<u>The</u> small stress and strain rate experiments <u>carried out in torsion and dilatometer</u> <u>equipment</u> demonstrate a second, relatively high viscosity <u>(creep-type rheology)</u>, plateau in the viscosity-strain rate dependence (Barnes, 1999) which cannot be observed with high stress or strain rate measurements. However, at temperatures above ~1145 to 1150 °C the groundmass of lava becomes so fluid that the viscosity decreases a few orders of magnitude (Fig. <u>10b</u>) and the high viscosity plateau is no longer present. <u>Fig. 10b</u> demonstrates the difference between the  $y_0$  (creep-type) and  $y_0$  (viscoplastic) viscosities. In the small strain and small stress torsion equipment, the internal structure of lava sample is not destroyed, and the measured viscosity is of a creep-type. This type of viscosity may be expressed in lava dome growth or relaxation processes. In experiments with rotational viscometers where strains and stresses are considerable, the internal structure of the lava is destroyed and the apparent viscosity is several magnitudes smaller. This type of viscosity will control lava flows over steep topography. Thus, at the same temperature, lavas may possess two viscosities depending on a stress-strain scale and numerical modelling of lava flows should consider the high viscosity and viscoelasticity of lavas when strain rates are below unity.

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## 5. Conclusions

- The oscillatory torsional apparatus allows low strain investigations of shear modulus and internal friction. Experiments carried out on lavas over a range of temperatures (~500 to 1150 °C) and frequencies (20 to 0.002 Hz) demonstrate that in general the complex shear modulus of lavas decreases with decreasing frequency and increasing temperature. Internal friction is usually <1.</li>
- 2. At temperatures close to their eruption temperature (~1080 to 1100 °C) no purely viscous regime was detected for the Etna and Vesuvius samples and their measured <u>creep-type</u> viscosity remained frequency dependent over the range 20 to 0.002 Hz. However, results for the Hawai'i sample indicate a shear rate independent regime for low shear rates at temperatures between ~1070 and 1130 °C, with viscosities >10⁹ Pa s.
- 3. The samples from Etna and Vesuvius exhibited <u>extended</u> anelastic behaviour at temperatures <u>within</u> the "glass transition" temperature of the groundmass, and that may be attributable to the <u>bad</u> cohesion between crystal grains and the groundmass glass. Annealed lava at ~900 to 950°C possesses a shear modulus about 15 to 20 GPa. Below ~800 °C, <u>but still within basalt glass transition temperature range</u>, when the <u>dilatometric</u> <u>effect of between groundmass glass and crystals are significant</u>, intensive microcracking may be expected and can decrease the shear modulus to 7 to 10 GPa. <u>By reheating</u> <u>hetween 700 and 950°C the characteristic time-constant of the lava hardening process</u> may be on a scale of several to <u>a</u>, hundred hours.

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Acknowledgements

We thank J. Deubener and one anonymous referee for helpful comments which improved the manuscript.

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**Fig. 1.** (a) Schematic of the torsion oscillation apparatus (redrawn from Berckhemer et al., 1982). Capacitive pickups detect the motion of the iron plates at the ends of the aluminium wings providing two data channels which can be calibrated to provide angular deflection and the applied torque. (b) An example of two periods of data collected in order to measure internal friction (from the phase shift) and magnitude of the complex shear modulus (from the relative amplitudes of the curves). Further details are given in Bagdassarov (2000).

Fig. 2. (a) Thin section of the crustal sample from Etna, 1992. The lava contains ~15 to 20 vol.% of vesicles (mean diameter between ~0.2 to 0.5 mm) and ~20 to 25 vol.% of plagioclase, olivine and magnetite phenocrysts. (b) Thin section of the 1992 Etna sample which was collected from the base of an overflow. This sample contains ~20 vol.% of vesicles with a mean diameter of 1 to 2 mm and about 30 vol.% of crystal (from analysis of polished thin sections without corrections for 3D effects).

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**Fig. 3.** Shear modulus (**a**, **b**) and internal friction (**c**, **d**) results from the 1992 Etna lavas. The crustal sample (**a**, **c**) shows a higher shear modulus than the basal sample, and neither sample has an internal friction approaching 1 over the conditions investigated.

**Fig. 4.** In (**a**) and (**b**), temporal variations in shear modulus and internal friction are given for the 1999 Etna sample. After annealing for 118 hours at 800 °C the sample had attained a greater shear modulus than it originally possessed at 700 °C. The increasing shear modulus and decreasing internal friction with time indicate the sample was becoming increasingly elastic and less viscous. The shear modulus and internal friction results collected after annealing are given in (**c**) and (**d**).

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**Fig. 5.** Time dependent shear modulus (**a**) and internal friction (**b**) during annealing experiments on the Hawai'i sample. Numbers in the legend indicate annealing time in hours.

Fig. 6. Shear modulus (a) and internal friction (b) results from the Vesuvius lava.

**Fig. 7.** Thin sections of Etna (**a** and **b**) and Vesuvius (**c** and **d**) lava samples (each image represents 1250 × 950 μm). (**a**) and (**b**) are starting material and show microcracks between crystals and groundmass glass as well as in the interior of phenocrysts. (**b**) shows the Etna sample after being annealed at 812 °C for 118 hours and (**d**) shows the Vesuvius sample after being annealed at 1102 °C for 96 hours. The reduction in the number of microcracks demonstrates that healing has occurred both within the groundmass and the phenocrysts.

**Fig. 8.** (a) The complex shear modulus of 1999 Etna lava measured at 20 Hz and different temperatures. The temporal change at each temperature is modelled using a curve of characteristic time-constant which is believed to represent the characteristic time for crack healing (see text). In (b), these characteristic times are plotted against absolute reciprocal temperature. The straight line fit represents an Arrhenian dependence with an activation energy of  $150 \pm 20 \text{ kJ mol}^{-1}$ .

**Fig. 2.** Dynamic viscosity as given by  $G''(\omega)/\omega$  (see Eq. 4) for the Vesuvius (**a**) and Etna (**b**) samples. At the highest temperatures and lowest frequencies used these samples maintained a frequency dependent rheology.

**Fig. 10.** Dynamic viscosity of the Hawai'i sample. In (a), the high temperature values of  $G''(\omega)/\omega$  are given, demonstrating the decreasing dependence on frequency at low frequencies. The curves show the results of using a Cross model in order to extract the zero-shear viscosity (see text). Fitting parameter of Eq 5 m=1 at all temperatures,  $K=1.6 \log(n_0 Pa)$ 

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# <u>s)= 9.23 at 1126°c, 3.7 and 9.7 at 1100°, 30 and 10.65 at 1072°.</u> In (b), the zero-shear

viscosity values <u>no</u> obtained <u>for the Hawai'j lava sample</u> are compared with those given by dilatometer (<u>Bagdassarov</u>, 2000) and rotational viscometer (Shaw, 1968, 1969; Ryerson et al. 1988; Ryan and Blevins, 1974; Pinkerton et al. 1995) experiments <u>on Hawai'jan basalt lavas</u>.

There is good agreement between the dilatometer and the torsional results, with the slightly greater values from the dilatometer being expected due to the effect of compression viscosity

on extracting shear viscosity values from the pure shear experiments (Bagdassarov and Dingwell, 1992). The gradient of the dashed best fit lines represents an activation energy for viscous flow of ~950 kJ mol⁻¹. The plot demonstrates the ~3 order of magnitude change in

viscosity which occurs around  $7.0 \times 10^{-4} \text{ K}^{-1}$  (~1155 °C) and separates the high temperature,

Newtonian region from the lower temperature, viscoplastic region. The 1 s⁻¹ line indicates the torsion results at unit shear rates which are more applicable for comparison with the rotational viscometer measurements. High values of  $p_0$  obtained in torsion and dilatometer, small strain-, stress experiments relate to the creep-type rheology displayed by lava when its internal structure is not destroyed by the measurements. Field experiments carried out at high stresses and strains show  $p_{\infty}$  or a viscoplastic viscosity, expressed when the internal structure (the organisation of vesicles and crystals) of the lava is altered by the viscometer or the flow itself.

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these characteristic times are plotted against absolute reciprocal temperature. The straight line fit represents an Arrhenian dependence with an activation energy of  $150 \pm 20 \text{ kJ mol}^{-1}$ .¶ **Fig. 10.** Thin sections of Etna (A and B) and Vesuvius (C and D) lava samples. Photos 1250 x 950 µm. A and C is starting material. The contacts between crystals and

ava samples. Findos 1250 × 950 μm. A and C is starting material. The contacts between crystals and groundmass glass as well as the interior of phenocrystals are marked by microcracks. B is Etna sample annealed over 118 h at 812°C. D is Vesuv sample annealed over 96 h at 1102°C. The contacts between groundmass glass and crystals as well as the interior of phenocrysts are healed.¶

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according to the princ	cipie of the thermorheological	simplicity
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The main difference in	n	
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In this case,		
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viscoity		
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(effective $T_a$ of a sample)		
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increase or		
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in the case of th		
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e		
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This effect is clearly demons	strated by an extended	ed temperature dependence of
<b></b>		
shear modulus and internal friction		
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on frequency and temperature	e	
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The relaxation of the s	WINC	21/03/2002 13.37.00
The relaxation of the 5		
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in basalt lava		
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At the same time		
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rather		
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than		
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Dama 12, [27] Dalatad		22/02/2002 11/12/02
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At $1 > 920$ C, the t	fulk crystallisation of basaltic gr	
nla sis slags amostala mary sl	as sontributs to the increase of	tiffe and of sum caled laws
plagioclase crystals may al	so contribute to the increase of s	summess of annealed lava
samples. Analysis of thin s	ections of samples after experin	ients revealed the textural

changes only in thin surface layer (few  $\mu$ m) and not in the bulk of samples. A

noticeable growth of plagioclase and Fe-Ti-oxides was observed at 850-934°C after

>200 h of annealing (Burkhard, 2001) that is longer that the duration of torsion

annealing experiments. The end of these processes must take 10 times longer time,

despite the constant steady state values of shear modulus were observed after <100 h.

Page 13: [38] DeletedMike James22/08/2002 14:12:00the additional annealing of lava samples in laboratory is not believed to

Page 13: [39] DeletedMike James22/08/2002 14:12:00significantly contribute to further oxidation

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in the bulk of the samples.

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Bingham yield stress and		
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under conditions of slow s	small scale deformation where the	
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	Nilke James	23/08/2002 13:05:00
internal structure of lava sa	mples remains intact	
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of lava samples is not destr	royed	
Ĩ	5	
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Parameter <i>m</i> characterises a	stratching character of shear strass	relevation during
ratalieter m characterises a	a stretching character of shear stress	relaxation during
viscoelastic transition. For a	a Maxwell body rheology <i>m</i> =2 in E	q 5.
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Parameter $m=1$ in E	q 5 and indicates on a distribution of	of a shear stress
	-	
relevation encotror		
relaxation spectrum.		
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$\mu$		
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in nure shear deformation	WIKE James	22/06/2002 14.34.00
, in pure shear derormation,	1	
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Parallel plate		
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samples. Thus, the viscosity calcul	ated from a simple	shear deformation
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in a dilatometer and shown in Fig.	8b is ¹ / ₃ "	
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shear viscosity from parallel plate	viscometry. The co	prrection of the measured
	2 - 4	
viscosity by dividing to the factor.	5 stems from the ass	sumption that the material is
incompressible and possesses an ir	finite volume visco	sity. It is not true for a sample
incompressione and possesses an in		sity. It is not true for a sample
with porosity of c 50 yol %		
with porosity of c. 50 vol.70.		
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must	Wike James	22/00/2002 14.37.00
must		
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Thus, assuming that the measured		
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dilatometer viscosity is a	Wilke Suffics	2270072002 14.07.00
and the set of the set		
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compressional viscosity, $\mu_c$ , and, f		
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, is r		
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from the torsion experiments		
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provides the viscosity for simple s	shear, $\eta$	
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and they are related as follows		
s,		
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(6).

Applying the relationship  $\mu_c = \mu_v + 4/3 \cdot \mu_s$ 

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appropriate for materials with hi	gh porosity	
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(e.g. Bagdassarov and Dingwell	, 1992), thisthis	
Page 15: [62] Deleted	Mike James	22/08/2002 17:37:00
the volume viscosity as estimate	ed from the observed dif	ference between the two
types of experiments (pure and s	simple shear) as $\sim$	
	1 /	
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3.35		
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4/3 (Prud'homme and Bird 197	8)	
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difference between our result s	and	23/06/2002 13:11:00
difference between our result a	lind	
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Eq		
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which	MIKe	21/08/2002 18:34:00
which		
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the	WIKE	21/06/2002 10.34.00
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where		
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significantly	Mike	21/00/2002 10:07:00
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the		
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High strains		
8		
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of deformation,		
Page 16: [68] Deleted	Mike	21/08/2002 16:35:00
some		
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, transient flow after ap	plying a new load etc.	
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which		
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 This heat production cannot result in a significant temperature effect.

 Transient flow in oscillatory shear does not play any significant role, amplitude and

 phase of the angle deformation were calculated from sinusoidal signals after a many

 repeated oscillations.

Page 17: [71] DeletedMike James22/08/2002 15:59:00In torsion experiments the viscosity results from the Hawai'i sampledemonstrate the effect of a large volume percentage of deformable vesicles. Only withthe Hawai'i sample (~50 vol.% of vesicles) were our experiments close to a shear rateindependent viscosity. For the Etna and Vesuvius samples (<10 vol.% of vesicles) a</td>frequency independent viscosity was not detected, even at temperatures well abovethe glass transition temperature of basalt glass (~820 to 850 °C). This is in agreementwith previous measurements (on partially molten rocks and melt-crystal suspensions)which indicate that a frequency independent shear viscosity is unobtainable at lowstrains and stresses for samples with a melt phase <40 vol.% (Bagdassarov et al.,</td>1994; Bagdassarov and Dorfman, 1998).

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At temperatures about their glass transition point, lavas exhibit viscoelastic		

behaviour which can be of importance in problems such as dome growth and collapse and the slow development of lava flow fronts. Numerical modelling of lava flows should consider the high viscosity and viscoelasticity of lavas when the strain rates

are below unity. The main difference

Page 17: [72] Deleted Mike James 22/08/2002 16:04:00 between the low strain rate behaviour of, on the one side, Etna and Vesuvius basalts Page 17: [73] Deleted 22/08/2002 16:04:00 Mike James , and on the other, that from Hawai' Page 17: [74] Deleted Mike James 22/08/2002 16:04:00 lies in the relative proportion of crystals and vesicles. Page 17: [75] Deleted Mike James 22/08/2002 16:04:00 presence of deformable inclusions leads to 22/08/2002 16:04:00 Page 17: [76] Deleted Mike James decreases at small strain-rates and small stresses (Bagdassarov and Dingwell, 1992). The presence of non-deformable crystals results in increases Page 17: [77] Deleted Mike James 22/08/2002 16:04:00 the Bingham yield stress (Bagdassarov et al., 1994). Higher ratios of crystals to Page 17: [78] Deleted Mike James 22/08/2002 16:04:00 vesicles in Etna and Vesuvius Page 17: [79] Deleted Mike James 22/08/2002 16:04:00 lavas increase the Bingham creep-viscosity,  $\eta_0$ , Page 17: [80] Formatted Mike 21/08/2002 13:34:00 Formatted Page 17: [81] Deleted Mike James 22/08/2002 16:04:00 which was observable at the strain rates of the torsion apparatus. Page 17: [81] Deleted 23/08/2002 09:01:00 Mike James 8 Page 17: [81] Deleted Mike James 26/08/2002 11:01:00

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strain-rates which were	e unattainable in	
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the operational window	v of	
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our torsion experiment	$s (10^{\circ} s^{\circ})$ . In contr	
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Page 17: [86] Formatted	Strobl	14/08/2002 11:53:00
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was much smaller and	because of this fact	21/00/2002 10:01:00
was much smaller and	because of this fact	
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Bingham creep-viscos	ity	
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