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Reunion, Indian Ocean, France**

***Stephan Kolzenburg, Daniele Giordano, Andrea Di Muro, D. B.  
Dingwell***

# Equilibrium Viscosity and Disequilibrium Rheology of a high Magnesium Basalt

2 from Piton De La Fournaise volcano, La Reunion, Indian Ocean, France

S. Kolzenburg<sup>1,2,3\*</sup>, D. Giordano<sup>3,4,5</sup>, A. Di Muro<sup>6</sup>, D.B. Dingwell<sup>1</sup>

4 1) Ludwig-Maximilians-University Munich, Department of Earth and  
Environmental Sciences, Theresienstr. 41, 80333 Muenchen, Germany

6

8 2) McGill University, Department of Earth and Planetary Sciences, 3450 University  
Street, H3A 0E8 Montreal, Quebec, Canada

10 3) Universita degli Studi di Torino, Dipartimento di Scienze della Terra, Via  
Valperga Caluso 35, 10125 Torino, Italy

12

14 4) Istituto Nazionale di Geofisica e Vulcanologia-Sezione di Pisa, Via della  
Faggiola, 32, 56126 Pisa, Italy.

16 5) Institute of Geoscience and Earth Resources (IGG-CNR), Italian National  
Research Council (CNR), Pisa, Italy

18

20 6) Institut de Physique du Globe de Paris, Observatoire Volcanologique du Piton de  
la Fournaise, 1 Rue Jussieu, 75005 Paris, France

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\* Corresponding author: Stephan Kolzenburg; skolzenburg@gmail.com

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28

## Abstract

Lava flows are a common hazard at basaltic to intermediate volcanoes and have posed a significant threat to La Reunion Island over the past centuries. In sustained flow units, the efficiency of lava transport away from the vent is dominated by cooling. For basaltic to intermediate lavas, it is the ability of the lava to solidify during cooling which exerts a first-order control on spatial extent and flow distance. As a consequence, understanding the *sub-liquidus* rheology of lavas has become a key focus in lava flow research in the past decade. To date, the development of a systematic understanding of lava rheology during emplacement conditions has been significantly hampered by a lack of experimental data. Here we present new data on the rheological evolution of crystallizing high-Mg basalt from Piton de la Fournaise. Sub-liquidus experiments were performed at constant cooling rates ranging from 0.5 to 5 K/min. Those rates mimic thermal conditions experienced 1) by lava during flow on the surface and 2) by magma during dike and sill emplacement. Our data show that the effective viscosity of the crystallizing suspension increases until reaching a specific sub-liquidus temperature, the so-called “rheological cut-off temperature” ( $T_{\text{cutoff}}$ ), at which the lava becomes rheologically immobile and flow ceases. This departure from the pure liquid viscosity curve to higher viscosity is a consequence of rapid crystallization and its variability for a given lava is found to be primarily controlled by the imposed cooling rate. Based on these experimental data, we adapt the failure forecasting method (FFM) - commonly used to describe the self-accelerating nature of seismic signals to forecast material failure - to predict the rheological cut-off temperature ( $T_{\text{cutoff}}$ ). The presented data substantially expand the modest experimental database on non-equilibrium rheology of lavas and represent a step towards understanding the underlying process dynamics.

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## 1. Introduction

### 1.1. Motivation and scope of this study

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The flow of lava is dominantly governed by viscosity, effusion-rate and the slope and morphological features of the underlying topography (Cashman et al., 2013; Dragoni et al., 1986; Hallworth et al., 1987; Harris and Rowland, 2001; Pinkerton, 1987).

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Mitigating the hazards posed by lava flows that threaten infrastructure has only shown

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moderate success to date (Barberi et al., 2003). This is largely due to an incomplete understanding of how lava flows, how and when flows stop and is further hampered by a

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lack of methods to provide up-to-date topographic surface models (Cashman et al., 2013; Farquharson et al., 2015; Favalli et al., 2010; James and Robson, 2014; Kolzenburg et al.,

62

2016a; Kolzenburg et al., 2018c). These shortfalls drastically reduce our ability to predict lava flow emplacement.

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One of the most challenging aspects of predicting the flow dynamics of lavas is that the rheological properties of lava flows evolve during eruption and emplacement since they

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occur under conditions that impose a state of dynamic disequilibrium. The transient nature of the rheological properties is caused by changes in the temperature, composition and

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phase state of the lava or magma resulting from gas loss, cooling, crystallization and vesiculation. The importance of *non-isothermal* effects on the crystallization kinetics and

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textural development of silicate melts have in principle been recognized for decades (Arzilli and Carroll, 2013; Coish and Taylor, 1979; Gamble and Taylor, 1980; Hammer,

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2006; Lofgren, 1980; Long and Wood, 1986; Vetere et al., 2013; Walker et al., 1976). Yet experimental parameterisation of these effects on magma flow behaviour has lagged

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behind badly. Such a detailed understanding of the rheological evolution of crystallizing

76 magmas or lavas requires direct measurement of the flow properties at emplacement  
conditions.

To date, direct measurements of the rheology of lavas or magmas under natural  
78 conditions are few (Belousov and Belousova, 2018; Chevrel et al., 2018; Einarsson, 1949;  
Gauthier, 1973; Panov et al., 1988; Pinkerton, 1994; Pinkerton and Norton, 1995;  
80 Pinkerton and Sparks, 1978; Shaw et al., 1968). They represent crucial measurements for  
benchmarking of experimental data. In conjunction with systematic laboratory studies on  
82 the evolution of lava flow properties as a function of the physico-chemical environmental  
conditions such data enable the production of new input parameters for predictive models  
84 of lava rheology and emplacement. In an effort to generate a systematic empirical database  
that will help to elucidate the systematics of the underlying processes, a range of  
86 concentric cylinder viscometry data has been presented in the past decade. Concentric  
cylinder viscometry typically utilises measurements of the torque exerted by the liquid on a  
88 spindle inserted into a sample melt and rotated at a constant rate. The available datasets can  
broadly be separated into measurements at constant, isothermal conditions (i.e. constant  
90 undercooling and at fixed shear rates and oxygen fugacities) e.g. (Chevrel et al., 2015;  
Ishibashi and Sato, 2007; Robert et al., 2014; Sato, 2005; Sehlke et al., 2014; Soldati et al.,  
92 2016; Vona et al., 2011; Vona et al., 2013) and measurements at constant cooling rates that  
map suspension rheology at varying experimental shear-rates and/or changing oxygen  
94 fugacity (Giordano et al., 2007; Kolzenburg et al., 2018a; Kolzenburg et al., 2018b;  
Kolzenburg et al., 2016b; Kolzenburg et al., 2017; Kouchi et al., 1986; Shaw et al., 1968).  
96 The former dataset aims at attaining mechanical and textural equilibrium and permit the  
sampling of the equilibrated material. These data are used to correlate the viscosity  
98 measurements to the sample textures. The latter dataset aims at reproducing natural flow

conditions and allows the quantification of the “rheological cut-off temperature” ( $T_{\text{cutoff}}$ ), at  
100 which the lava becomes rheologically immobile and flow ceases. However, sampling of  
the texture or modelling of the change in crystal content throughout the experiment for  
102 correlation to the rheological data is not possible during these dynamic, disequilibrium  
experiments, since crystallization under these conditions occurs far from equilibrium and  
104 very rapidly.

Here we present new experimental data on a high-Mg Basalt from Piton de la  
106 Fournaise that investigate the sub liquidus evolution of melt rheology as a function of  
cooling rate at atmospheric conditions. While these data can also facilitate the  
108 understanding of kinetic flow evolution at thermal conditions (i.e. absolute temperature  
and cooling rates) of partly degassed magma during transport toward the surface in dike-  
110 swarms, the decrease in oxygen fugacity occurring at sub-surface conditions will likely  
require a shift of the presented data to lower temperatures, as shown in Kolzenburg et al.  
112 (2018a). Nevertheless, the presented dataset serves to expand a growing database on non-  
isothermal rheology, which is necessary for the development of empirical models of the  
114 effective lava viscosity at emplacement conditions. Such data are essential for robust  
prediction of lava flow behaviour and flow geometry.

## 116 ***1.2. Compositional characteristics of the 2007 eruption of Piton de la Fournaise.***

Piton de la Fournaise volcano is located on La Reunion Island ~800 km east of  
118 Madagascar. In 2007, it produced one of its largest volume eruptions of the past centuries.  
A detailed account of the eruption can be found in Staudacher et al. (2009). The 2007  
120 eruption represents an interesting end-member with respect to lava rheology since it  
erupted a wide range of basalt compositions and crystal contents, spanning almost the

122 entire range of magma compositions typically erupted at Piton de la Fournaise (Villemant  
et al., 2009). Based on regular sampling of the lava and compositional analyses,  
124 Staudacher et al. (2009) and Di Muro et al. (2014) report that the lava was, initially,  
evolved (MgO ca. 7wt%) and poorly phyric with less than 5 vol% of small  
126 olivine/clinopyroxene phenocrysts and plagioclase microlites. Throughout the eruption, the  
volume fraction (and size) and of olivine crystals increased, reaching up to 40 vol% of  
128 olivine phenocrysts at the end of the eruption (Di Muro et al., 2014; Staudacher et al.,  
2009; Villemant et al., 2009). Villemant et al. (2009) report that the April 2007 eruption  
130 produced some of the most primitive and most Mg-rich melts erupted at Piton de la  
Fournaise (MgO~9wt%) and that they correspond to the first step of the differentiation  
132 trend for the parental magma at Piton de la Fournaise (Albarède et al., 1997; Boivin and  
Bachèlery, 2009; Villemant et al., 2009). Villemant et al. (2009) and Di Muro et al. (2014)  
134 further note that early erupted lavas and the matrix glass of the 2007 lava flows (bulk MgO  
content ~25–30%) are less Mg-rich (MgO~6%) than matrix glass of scoria (glass MgO  
136 content ~7% and bulk rock MgO content ~18–20%) or Pélé's hair (glass MgO  
content=7.5–9%) emitted at the same vent; see (Tables 1a–b) in Villemant et al. (2009).  
138 Melt inclusions trapped in the most Mg-rich olivines (Fo86) record mafic melt  
compositions (MgO 10-11 wt%) representative of the primary melts feeding the central  
140 and shallow plumbing system of Piton de la Fournaise (Di Muro et al., 2014). Average lava  
composition at Piton de la Fournaise record extensive early olivine+/-pyroxene  
142 fractionation at depth, while primitive magnesian melts (MgO > 14wt%) are never erupted  
on the La Réunion island (e.g. See Fretzdorff and Haase (2002), Di Muro et al. (2016) and  
144 references therein). In summary, the available data demonstrate that the magma is  
significantly more Mg-rich during subsurface transport and eruption than is preserved in

146 the lava flow matrix glass and thus that the formation of olivine  $\pm$  clinopyroxene  $\pm$   
plagioclase plays a central role in the solidification of magma and lavas and, thereby their  
148 rheological evolution at Piton de la Fournaise volcano.

## 2. Experimental Materials and Methodology

### 150 *2.1. Experimental Material and Melt Preparation*

The samples we selected from the 2007 eruption display relatively high crystal  
152 contents (~25-40 vol%). For rheological experimentation we created a partial melt of the  
crystallized matrix and some amount of the crystal cargo. The rock sample was crushed  
154 using a jaw crusher and fragments of ~ 1cm diameter were hand-picked for matrix melt  
separation. These fragments were then partially melted in large thin-walled Pt crucibles to  
156 separate the olivine phenocrysts from the matrix. Melting was performed in a  
Nabertherm® MoSi<sub>2</sub> box furnace at 1250°C in multiple, small, batches for ~20 minutes per  
158 batch. During re-melting, the dense olivine phenocrysts sank to the base and the interstitial  
melt percolated through the crystal framework, forming a lens of melt on top of the crystal  
160 cumulate. This melt lens was carefully decanted to collect the interstitial partial melt.  
Fifteen batches were required to recover sufficient melt volume to perform viscometry.  
162 The quenched melt batches were crushed and ground using an agate mortar and thoroughly  
mixed for homogenization. These powders were then re melted at 1350°C for 12h to  
164 ensure chemical homogeneity. After homogenization the sample was quenched by pouring  
it onto a steel plate in order to ensure a cooling rate fast enough to suppress crystal growth.  
166 Visual assessment and Raman spectroscopy following the approach of Di Genova et al.  
(2018) affirmed that the glasses remained crystal-free. The glass was crushed and re-  
168 melted into Pt<sub>80</sub>Rh<sub>20</sub> cylindrical crucibles of 51 mm height and 26.6 mm diameter. The  
crucible was then transferred into a Deltech bottom-loading furnace adapted for



170 rheological experimentation, the device is described in successive configurations in  
(Dingwell, 1991; Dingwell, 1986; Dingwell, 1990; Dingwell, 1992).

## 172 **2.2. Glass chemistry analysis**

The bulk chemical composition of the experimental material was determined for  
174 the quenched glass using an electron probe micro analyser (EPMA). The sample was  
impregnated in a low viscosity epoxy resin, polished and carbon coated. The major  
176 element composition was determined at the Department of Earth and Environmental  
Sciences at the University of Munich, using a Cameca SX100 EPMA with 15 kV  
178 acceleration voltage and 5 nA beam current. In order to reduce the alkali volatilization the  
beam diameter was defocused down to 10  $\mu\text{m}$ . Synthetic wollastonite (Ca, Si), periclase  
180 (Mg), hematite (Fe), corundum (Al), natural orthoclase (K), and albite (Na) were used as  
standards, and matrix correction was performed by PAP procedure (Pouchou and Pichoir,  
182 1991). The accuracy was better than 2.5% for all analysed elements. Normalized results of  
the average of 10 EPMA analyses are presented in Table 1; analytical totals average 99.76  
184 %.

## 186 **2.3. Concentric Cylinder (CC) viscometry and Differential scanning calorimetry (DSC).**

Glass transition temperatures at onset ( $T_{\text{g}_{\text{onset}}}$ ) and peak ( $T_{\text{g}_{\text{peak}}}$ ) were obtained  
188 from heat capacity curves derived from differential scanning calorimetry on aliquots of the  
re-melted glass undergoing cooling/heating cycles at rates of 5, 10, 15 and 20 K/min.  
190 Reproducibility of both  $T_{\text{g}_{\text{onset}}}$  and  $T_{\text{g}_{\text{peak}}}$  are better than 0.5 % (i.e.  $\sim 3$  K); see also  
Giordano et al. (2015) and Giordano and Russell (2017). These data are used to recover the  
192 low temperature viscosity data using the method presented by Giordano et al. (2008a), that  
correlates  $T_{\text{g}_{\text{onset}}}$  and  $T_{\text{g}_{\text{peak}}}$  to fixed values of viscosity; these data are reported in Table 2.

194 High temperature rheological experiments were performed using a Brookfield DV-  
III+ viscometer head (full range of torque = 0 - 0.7187 mNm). A Pt<sub>80</sub>Rh<sub>20</sub> spindle is  
196 suspended from the viscometer head, immersed into the sample and rotated at a constant  
rate. The torque exerted by the sample at constant rotation rate is proportional to the melt  
198 or suspension viscosity. The spindle head used in these experiments is 33.2 mm long with  
14.4 mm diameter and a 45° conical top and bottom to reduce edge effects, this bob is hung  
200 on a 2.4 mm diameter stem; see also: (Dingwell, 1986; Dingwell and Virgo, 1988). The  
torque reading for this spindle and crucible were calibrated against DGG1 standard glass,  
202 with a known viscosity-temperature relationship (Meerlender, 1975). Calibration was  
performed for shear rates and temperatures exceeding those used in this study. The  
204 precision of the viscosity determination is ±3% as described in Dingwell (1986). The  
thermal evolution of the sample at the imposed cooling rates was calibrated with an  
206 accuracy of ±1 °C over the entire experimental temperature range using a platinum sheathed  
Type-S thermocouple immersed in DGG1 standard glass. The super liquidus melt viscosity  
208 was measured at between 1444 and 1321 °C in temperature steps of ~25 K; see Table 2  
(corresponding to furnace set points of 1450 - 1325 °C). Each temperature step was  
210 maintained for 120 min and a stable torque reading was commonly achieved after ~70 min,  
indicating thermal and chemical (i.e. redox) homogeneity.

212 The high temperature viscosity measurements in combination with the low  
temperature melt viscosity data recovered via application of a shift factor to the DSC data,  
214 are used to fit a VFT-model (Tammann and Hesse, 1926) to the experimental data. This  
model describes the theoretical crystal-free liquid viscosity at sub-liquidus temperatures  
216 (Giordano et al., 2008a; Gottsmann et al., 2002; Hess and Dingwell, 1996; Hess et al.,  
1995) and is used to calculate the relative viscosity of the suspension (see section 2.4 for

218 details). The model parameters are reported together with the liquid viscometry results in  
Table 2.

#### 220 ***2.4. Non-Isothermal Concentric Cylinder (CC) viscometry.***

Non-isothermal experimentation was performed following methods similar to the  
222 ones described in previous work (Giordano et al., 2007; Kolzenburg et al., 2018a;  
Kolzenburg et al., 2017). A series of experiments were run at constant cooling rates of 0.5,  
224 1, 3 and 5 K/min and an initial shear rate of  $0.766 \text{ sec}^{-1}$  (corresponding to 2.5 rpm).  
Experiments at lower initial shear rates were not possible for the given experimental  
226 temperatures because at low rotation rates, the surface tension of the melt in the meniscus  
between spindle and crucible is sufficiently high to pull the spindle to the sidewalls of the  
228 crucible. During experimentation, the measured torque increased as a function of the  
apparent viscosity of the suspension. Upon reaching the maximum torque limit of 0.7187  
230 mNm, the shear rate was automatically dropped to 0.368, 0.184, 0.092, 0.061, 0.031  $\text{s}^{-1}$   
(1.2, 0.6, 0.3, 0.2, 0.1 rpm, respectively), each successive time the limit was reached.

232 Between consecutive dynamic cooling experiments the sample was re-heated to  
1396 °C (corresponding to a furnace set point of 1400 °C) for a minimum of two hours and  
234 homogenized by stirring at a shear rate of  $3.06 \text{ s}^{-1}$  (10 rpm) in order to ensure complete  
melting of all crystalline phases formed during previous experimentation. After the end of  
236 the last experimental cycle, the sample was re-melted and quenched to a glass. Based on  
the results of Mysen and Virgo (1978) and Dingwell and Virgo (1987) we assume that  
238 redox equilibrium was achieved during the super-liquidus melt viscosity determination,  
which lasted more than 24 hours at atmospheric conditions. Since the crystallization  
240 experiments lasted between 0.5 and less than 5.8 hrs (at lower temperatures; i.e. higher

viscosities and slower redox equilibration) and we assume that the melt redox state was re-  
242 equilibrated to the initial, pre-experimental state within the time spent at high temperature  
under constant stirring during remelting. The assumption of thermal and redox  
244 equilibration and re-melting of previous crystal phases was confirmed by the recovery of a  
steady torque measurement at  $\pm 1\%$  of the value of the pure melt over more than an hour.

246 In order to resolve the crystallization induced rheological deviation from the pure  
melt viscosity at greater detail than in absolute viscosity space we calculate the relative  
248 viscosity  $\eta_r$ , that is the ratio between the measured shear viscosity of the mixture  $\eta_s$  of the  
crystallizing suspension and the liquid viscosity  $\eta_l$ , (Eq. 3):

250 
$$\eta_r = \frac{\eta_s}{\eta_l} \text{ (eq. 3)}$$

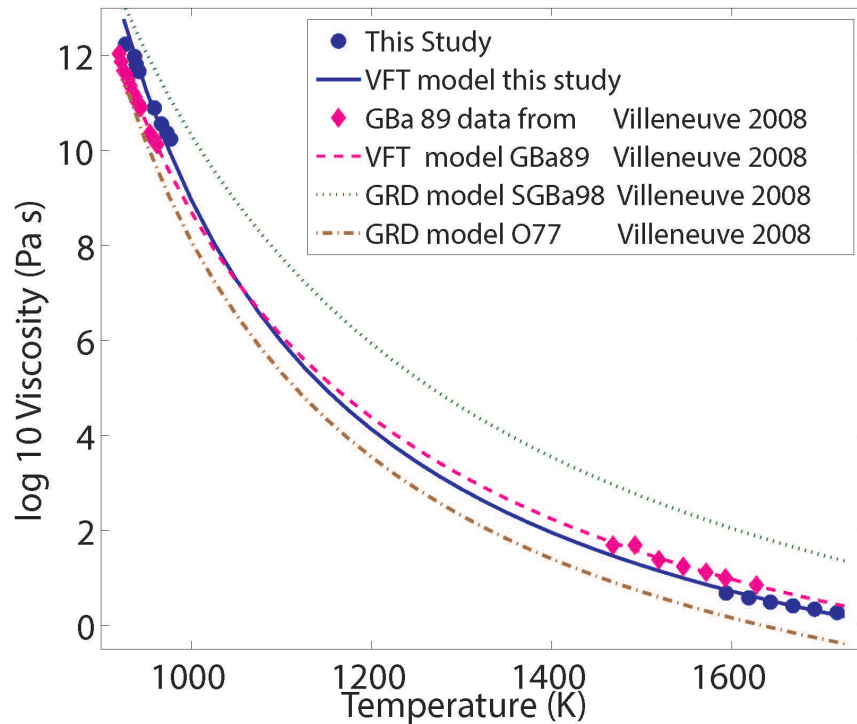
where  $\eta_l$  was calculated by employing a VFT-fitting of the low temperature  
252 viscosity data reported in Table 2 and the super liquidus viscometry data of each  
experiment. In the following sections, the results are presented as the logarithms of both  $\eta_s$   
254 and  $\eta_r$ .

### 3. Results

256 **3.1. Liquid viscosities of magmas at Piton de la Fournaise.**

The melt composition studied here is reported in Table 1. It is representative of the  
258 parental melt feeding the central activity of Piton de la Fournaise (Di Muro et al., 2016; Di  
Muro et al., 2014; Famin et al., 2009; Villemant et al., 2009). The viscosity measurements  
260 of the crystal free liquid are summarized, together with the VFT model fitted to the low  
and high temperature viscometry data in Figure 1. For comparison we also report viscosity  
262 measurements as well as GRD model estimates (Giordano et al., 2008b) of three other

compositions presented in Villeneuve et al. (2008). These span the compositional variety  
 264 of the Piton de la Fournaise volcano, where SGBa98 is representative of the more evolved  
 and O77 representative of the more primitive melt compositions erupted at Piton de la  
 266 Fournaise (Albarède et al., 1997; Boivin and Bachèlery, 2009; Famin et al., 2009;  
 Villemant et al., 2009).



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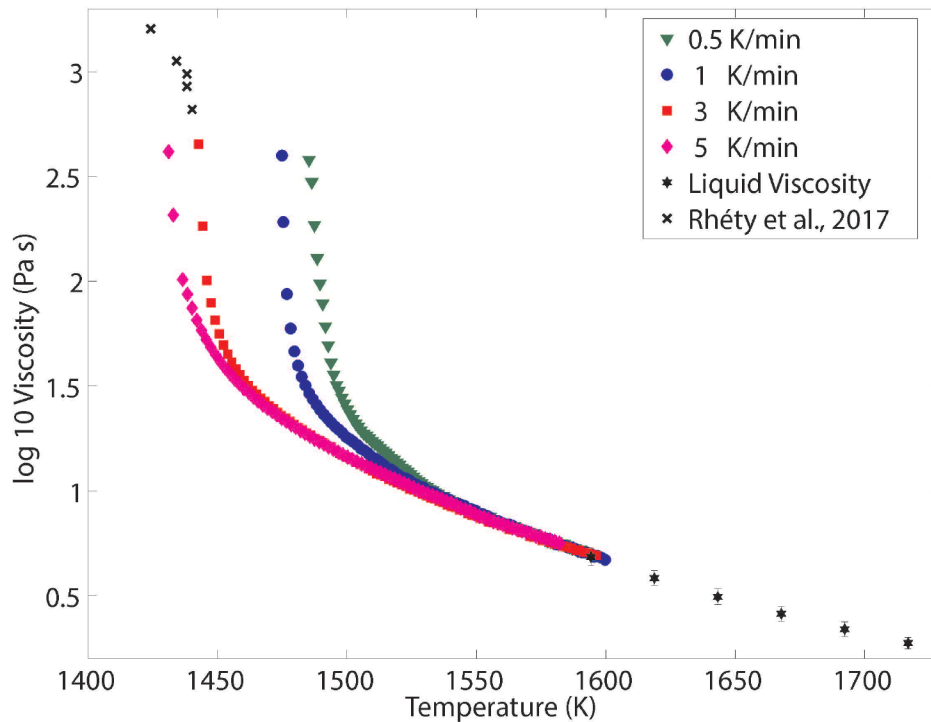
**Figure 1:** Summary of crystal free liquid viscometry data on Piton lavas. Blue dots  
 270 represent high temperature CC data and low temperature DSC measurements converted to  
 viscosity, measured for this study. The blue solid line represents the VFT-model fitted to  
 272 these data. Magenta diamonds and dashed line represent viscometry data from Villeneuve  
 et al. (2008). Green dotted and brown dash-dotted lines represent GRD model curved for  
 274 compositions SGBa90 and O77 reported in Villeneuve et al. (2008), respectively. Errors  
 are smaller than symbol size.

276

### 3.2. Viscosity evolution of the crystallizing suspension

278 The temperature-dependent viscosity of the Piton de la Fournaise lava, when subjected to  
 varying cooling rates is plotted as absolute and relative viscosity vs. temperature in Figures

280 2 and 3, respectively; the data are reported as absolute and relative viscosity in Tables 3  
and 4, respectively.



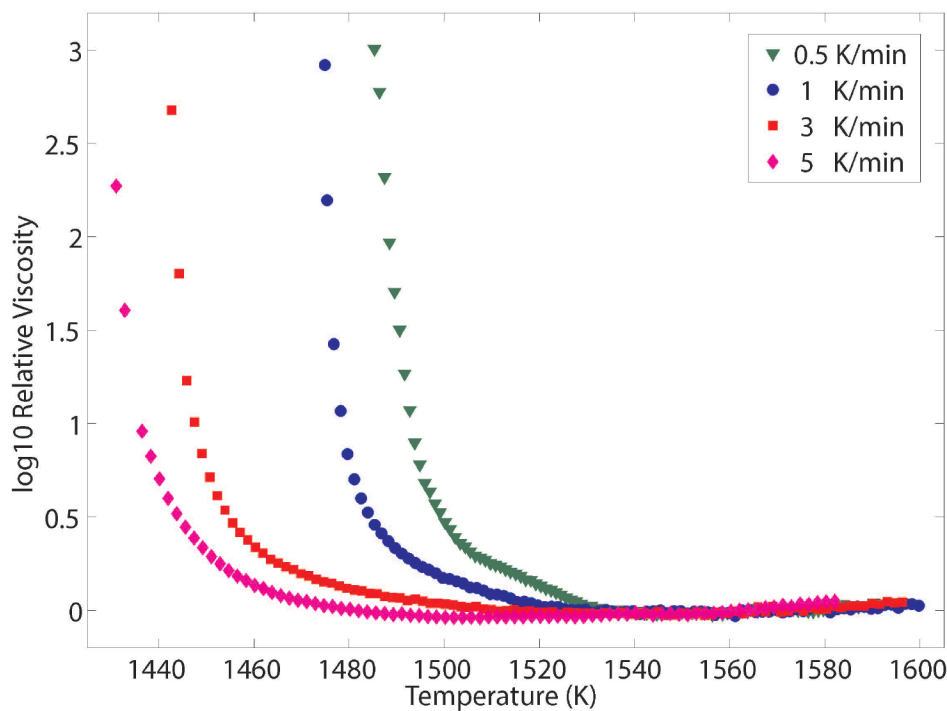
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**Figure 2:** Summary of non-isothermal viscometry data on Piton lavas presented as absolute viscosity. Black stars represent high temperature CC data. Green triangles, blue dots, red squares and magenta diamonds represent constant cooling viscometry data on the evolving suspension at cooling rates of 0.5, 1, 3 and 5 K/min, respectively. Black crosses represent viscosity estimates based on sample textures from (Rhéty et al., 2017). The precision of the viscosity determination is  $\pm 3\%$  of the measured value.

The rheological evolution of the lava follows the trend of the pure liquid data ( $\log \eta_r = 0$ ) up until the point where the influence of crystalline phases (via nucleation and growth of crystals) starts to measurably increase the sample's apparent viscosity. At this deviation, the apparent viscosity of the suspension increases with respect to the theoretical liquid ( $\log \eta_r > 0$ ). This increase in relative viscosity accelerates rapidly as temperature decreases. At higher cooling rates this departure commences at progressively lower temperatures (i.e. higher degrees of undercooling) since crystallisation of the melt is delayed. Further, the nature of this departure is correlated with the imposed cooling rate,

where slower imposed cooling rates result in a steeper rheological departure. This is in  
298 agreement with earlier non-isothermal rheological experiments at atmospheric conditions  
(Giordano et al., 2007; Kolzenburg et al., 2018b; Kolzenburg et al., 2016b).

300 The crystallization induced rheological departure accelerates continuously until the  
samples reach the torque limit of the experimental device. This point is close to a specific  
302 sub-liquidus temperature, the so-called “rheological cut-off temperature” ( $T_{\text{cutoff}}$ ) at which  
the sample becomes rheologically immobile and flow ceases. Albeit direct measurements  
304 at high viscosities, approaching solidification, are not possible due to mechanical  
constraints of the experimental setup, the data show that  $T_{\text{cutoff}}$  shifts systematically to  
306 lower temperatures with increasing cooling rate.



308 **Figure 3:** Summary of non-isothermal viscometry data on Piton lavas presented as relative  
310 viscosity. Green triangles, blue dots, red squares and magenta diamonds represent constant  
cooling viscometry data on the evolving suspension at cooling rates of 0.5, 1, 3 and 5  
312 K/min, respectively. The precision of the viscosity determination is  $\pm 3\%$  of the measured  
value.

## 4. Discussion

### 314 *4.1. Implications for magma transport and lava emplacement at Piton de la Fornaise*

316 The data reported in Figure 2 show that the viscosity measurements presented here  
are in good agreement with the down-flow viscosities calculated by Rhéty et al. (2017).  
318 The latter study estimated the bulk lava viscosity of the 2007 lava flow using the texture-  
based approach for multiphase rheology presented in Phan-Thien and Pham (1997) in  
320 combination with the temperature-dependent melt viscosity model reported in Villeneuve  
et al. (2008). Direct comparison with our measurements is consistent with down-flow  
322 cooling rates of 3 to 5 K/min. Rhéty et al. (2017), reconstruct initial cooling rates of 7  
K/km that increase down-flow to 42 K/km. Accounting for the average flow channel  
324 geometries (i.e. widths between 4 and 30 m and heights between 0.8 and 2 m) reported for  
the flow zones defined in Rhéty et al. (2017) and the average effusion rate of  $54 \text{ m}^3\text{s}^{-1}$   
326 reported in Staudacher et al. (2009), the distance-dependent cooling rates convert to time-  
dependent cooling rates of 0.1 to 2 K/min and 0.7 to 12 K/min for the reported values of 7  
328 K/km and 42 K/km, respectively.

Lava cooling rates increase sharply during late stages of flow once the lava leaves  
330 the well-insulated transport system (Cashman et al., 1999; Harris et al., 2005; Harris and  
Rowland, 2009), therefore the lower cooling rates reported in the former studies are likely  
332 most representative of the dominant flow phase. Values as low as 0.1 K/min, however, are  
lower than the cooling rate imposed in the experiments (see viscometry data plotted in  
334 Figure 1). We attribute this mismatch to the changes in oxygen fugacity between the  
experiments, carried out in atmospheric conditions, and the natural lava, being erupted in a  
336 more reduced state; see Vlastélic et al. (2016), Pichavant et al. (2016) and references



therein. The pre-eruptive  $fO_2$  for PdF magmas is estimated at  $\sim$ NNO-0.5 and shallow level  
338 degassing of  $SO_2$  and  $H_2O$  might further decrease the oxygen fugacity; this is suggested by  
late stabilization of sulphides at shallow level; see (Vlastélic et al., 2016). According to the  
340 data presented in Kolzenburg et al. (2018a) such a shift in  $fO_2$  may result in a decrease of  
the rheologic departure and  $T_{cutoff}$  of  $\sim$  40 to 80 K. When accounting for such a  $\sim$ 40-80 K  
342 decrease in the rheologic departure due to varying  $fO_2$  conditions, the reconstructed  
cooling rates of 0.1 to 2 K/min match with the experimental measurements, indicating that  
344 the erupted lava did not undergo efficient redox equilibration during subaerial flow.

#### ***4.2. Towards forecasting the rheological cut-off temperature ( $T_{cutoff}$ ).***

346 The decrease in  $T_{cutoff}$  with increasing cooling rate is a result of the balance  
between crystal nucleation and growth rate and the rate of undercooling experienced by the  
348 sample (Kolzenburg et al., 2018a; Kolzenburg et al., 2016b). There are two dominant  
mechanical constraints of the experimental apparatus that do not allow to continue the  
350 rheological measurement beyond the maximum viscosity values presented here: 1) the  
torque limit of the rheometer head and 2) the fact that the crucible containing the  
352 experimental sample may start to slip and rotate in its holder at high torque, rendering the  
measured torque data invalid. Here we present a new, numerical, approach that uses the  
354 self-accelerating nature of the rheological departure to forecast the  $T_{cutoff}$  value (i.e. the  
temperature at which the sample becomes rheologically immobile and infinite viscosity is  
356 reached) of the lavas under the imposed experimental conditions. This approach follows  
the failure forecast method (FFM) adopted in volcano seismology and material science to  
358 predict failure of a material based on the rate of increase in seismic energy released prior to  
failure (Voight, 1988; Voight, 1989).

360 It is based on the following equation:

$$\frac{d^2\Omega}{dt^2} = A \left(\frac{d\Omega}{dt}\right)^\alpha \quad \text{Equation (1)}$$

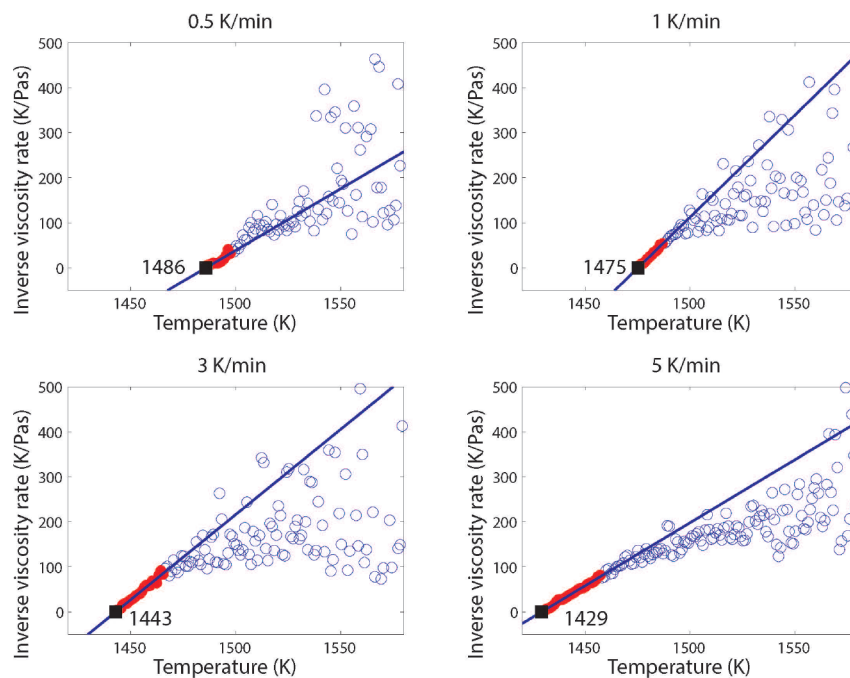
362 where  $d^2\Omega/dt^2$  and  $d\Omega/dt$  are the acceleration and rate of the phenomenon being  
monitored, and  $A$  and  $\alpha$  are empirical parameters. For predicting rock failure, this equation  
364 is commonly simplified to describe only peaks in signal acceleration rate using a constant  
 $\alpha=2$ , as shown in Kilburn (2003) and Lavallée et al. (2008) resulting in:

$$366 \left(\frac{d\Omega}{dt}\right)^{-1} = \left(\frac{d\Omega}{dt}\right)_0^{-1} - A(t - t_0) \quad \text{Equation (2)}$$

where  $t$  is time, and  $(d\Omega/dt)_0$  is the value of  $(d\Omega/dt)$  when  $t=t_0$ . The FFM utilizes  
368 the production rate of a precursory phenomena and correlates its acceleration to the point at  
which the process reaches an uncontrolled (i.e. runaway) state. This state is reached at an  
370 infinite  $d\Omega/dt$ , that implies an uncontrolled rate of change and, thereby, the point at which  
the monitored process reaches infinite velocity. The forecasting potential of Eq. 1 lies in its  
372 potential for describing the rate at which  $d\Omega/dt$  approaches uncontrolled conditions.  
Plotting inverse rate against time returns a negative linear trend, where the time at which  
374 the inverse rate equals zero corresponds to the uncontrolled condition when  $d\Omega/dt$  tends to  
infinity (Kilburn, 2003; Lavallée et al., 2008). This point can then be obtained by simple  
376 linear extrapolation of the measured trend to the time axis. In volcano seismology or  
laboratory experimentation on the brittle ductile transition of magma, the precursory signal  
378 of the fracturing and failure phenomena are for example, seismicity rate, acoustic emission  
rate or seismic energy release. The forecasting potential for volcanic eruptions has been  
380 demonstrated on seismic data from active volcanoes (Kilburn, 2003; Kilburn and Voight,  
1998) as well as for experiments on natural lavas at the brittle ductile transition (Lavallée

382 et al., 2008). For further details see also (Cornelius and Voight, 1995; De la Cruz-Reyna  
and Reyes-Dávila, 2001; Tokarev, 1963).

384 In the case of crystallizing lava that experiences ever increasing undercooling, the  
precursory signal to  $T_{\text{cutoff}}$  (i.e. the lava becomes rheologically immobile resulting from an  
386 accelerating crystallization rate) is the rate of increase in suspension viscosity. This is  
measured as a function of temperature instead of time. The formulation for forecasting then  
388 reads  $d\eta/dT$  (instead of  $d\Omega/dt$ ), where  $\eta$  and  $T$  are the effective suspension viscosity and  
temperature, respectively. The temperature at which the inverse rate reaches zero  
390 corresponds to the uncontrolled condition when  $d\Omega/dT$  tends to infinity. In Figure 4 we  
present plots of the inverse rate of viscosity increase against temperature.

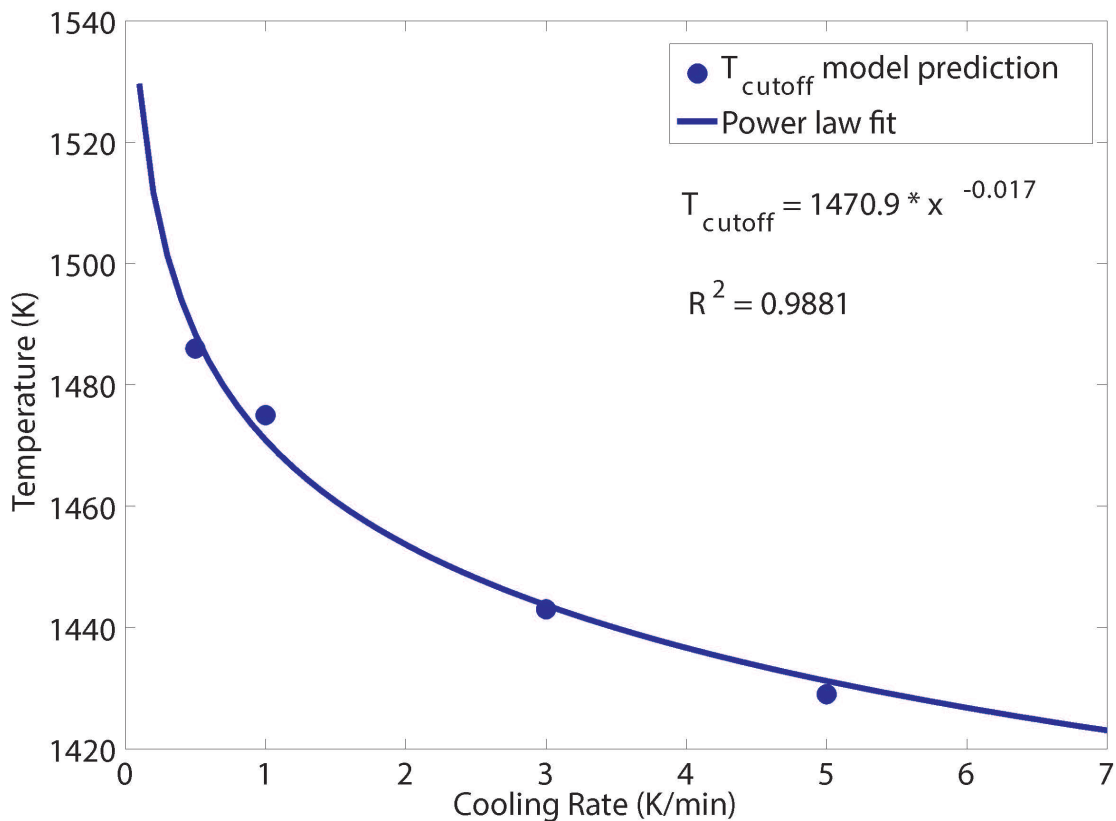


392

**Figure 4:** Plot of viscosity acceleration rate vs Temperature. Data for cooling rates of 0.5,  
394 1, 3 and 5 K/min are plotted in subplots a, b, c and d, respectively. Blue open circles  
represent the experimental data interpolated at 1 degree intervals. Red filled circles  
396 represent the linear part of the data used to fit the linear model of viscosity acceleration for  
extrapolation to  $T_{\text{cutoff}}$ . Black squares represent the predicted  $T_{\text{cutoff}}$  (i.e.  $d\Omega/dT = \infty$ ). The  
398 precision of the viscosity determination is  $\pm 3\%$  of the measured value.

The data plot randomly at high temperatures, where the acceleration is small (i.e. large numbers of inverse acceleration). Once crystallization sets in, the acceleration of the measured suspension viscosity increases, forcing the data to converge on a negative linear trend. Thus,  $T_{\text{cutoff}}$  can be obtained by simple linear extrapolation of the measured trend to the temperature axis.

Figure 5 reports the values forecasted using this method as a function of cooling rate. We fit a power law model to the predicted  $T_{\text{cutoff}}$  values that describes the data with an R-squared value of 0.9881.



**Figure 5:** Plot of the predicted  $T_{\text{cutoff}}$  temperatures as a function of cooling rate. Blue circles represent the forecasted values from the data in figure 4 for cooling rates of 0.5, 1, 3 and 5 K/min. the blue solid line represents a power law model fitted to these data describing the  $T_{\text{cutoff}}$  lava immobilization threshold as a function of cooling rate. Errors are smaller than symbol size.

Extrapolation of the data using this model suggests that further increasing cooling  
414 rate would result in a smaller effect of cooling rate on this rheological threshold, whereas  
decreasing cooling rates would further increase the temperature at which this threshold is  
416 reached. This is due to the longer time spent at higher temperatures, allowing for crystal  
growth approaching near equilibrium conditions. However, since the model describes a  
418 crystallization induced rheological threshold, it is important to note that the model is only  
valid at sub-liquidus temperatures and non-equilibrium conditions (i.e. at constant cooling  
420 rates).

Incorporating  $T_{\text{cutoff}}$  measurements in lava-flow models will allow for the concept  
422 of yield strength to be replaced by a melt specific rheological  $T_{\text{cutoff}}$  that is dependent on  
composition, shear- and cooling-rate (Kolzenburg et al., 2018a; Kolzenburg et al., 2018b;  
424 Kolzenburg et al., 2016b). The concepts of yield strength or other arbitrary or empirically  
chosen parameters such as fixed crystal contents, temperature or degrees of undercooling  
426 have been introduced in numerical simulations of lava flows (especially in cellular  
automata type models; e.g. (Miyamoto and Sasaki, 1997)) in order to ascribe a halting  
428 criterion to a modelled lava parcel. It has also been introduced as a rheologic criterion in  
planetary sciences for the derivation of rheological parameters from flow morphology,  
430 derived from experiments using analogue materials that possess yield strength e.g. (Fink  
and Griffiths, 1990; Hulme, 1974). In these analogue experiments these materials were  
432 chosen in order to mimic the development of a crust during cooling and they do not to  
represent actual flow rheology as demonstrated in Kolzenburg et al. (2018c).  $T_{\text{cutoff}}$  data  
434 would therefore represent a more realistic description of the lavas rheological evolution  
and therewith better ability to forecast their emplacement and flow cessation. The new  
436 approach to predict flow cessation may eventually be combined with satellite remote

sensing data of the thermal evolution of lava flows such as presented in Coppola et al.  
438 (2013) to provide near real time assessment and forecasting of flow evolution.

## 5. Conclusions

440 In conclusion, we find that:

1. Cooling rate exerts a first order control on the sub liquidus rheological evolution of  
442 the high Mg Basalt from Piton de la Fournaise through its influence on the melt  
crystallization-kinetics.
- 444 2. The failure forecasting method (FFM) can be adopted to extrapolate disequilibrium  
viscometry data beyond the mechanical constraints of the experimental apparatus  
446 for prediction of the lavas  $T_{\text{cutoff}}$ .
3. Modelling of magmatic flow behaviour requires melt-composition specific flow-  
448 and crystallization-models describing  $T_{\text{cutoff}}$ .
4. Developing a systematic process understanding of the non-equilibrium rheology of  
450 natural silicate melt suspensions requires an expanded database of dynamic  
measurements such as those presented here.
- 452 5. Implementation of data on the dynamic rheological evolution of magma during  
ascent in dykes and during the emplacement of lavas in computational models  
454 would allow to more accurately constrain the results of physical property based  
magma and lava transport models.

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## Tables

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**Table 1:**

Oxide	<i>Wt % Oxide</i>
SiO <sub>2</sub>	48.56
TiO <sub>2</sub>	2.40
Al <sub>2</sub> O <sub>3</sub>	12.15
FeO	12.19
MnO	0.20
MgO	11.61
CaO	9.76
Na <sub>2</sub> O	2.33
K <sub>2</sub> O	0.56
P <sub>2</sub> O <sub>5</sub>	0.25
analytical total	99.76

EPMA analysis of major element components as oxides; Data is normalized to 100 wt%

**Table 2**

Temperature (K)	Temperature (C)	log $\eta$ (Pa s)	Standard deviation
1717	1444	0.27	0.03
1692	1419	0.34	0.03
1668	1395	0.41	0.04
1643	1370	0.49	0.04
1619	1346	0.58	0.04
1594	1321	0.68	0.04
977	704	10.24	0.17
973	700	10.37	0.17
967	694	10.56	0.15
959	686	10.90	0.14
942	669	11.67	0.26
939	666	11.81	0.25
937	664	11.98	0.24
927	654	12.24	0.24
	A	-3.77	
	B	4163.79	
	C	673.18	

704

Table 3: Summary of absolute viscosity measurements for all experiments. For simplicity we report interpolated values of the experimental data at equal temperature steps with exception of the final datapoint for some experiments; highlighted in grey. Temperatures related to those points are reported in the last two rows in Kelvin and Celsius, respectively.

Cooling Rate (K/min)		0.5	1	3	5
Shear Rate (sec <sup>-1</sup> )		0.77	0.77	0.77	0.77
T (K)	T (C)	log $\eta$ (Pa s)			
1573	1300	0.79	0.79	0.78	0.78
1568	1295	0.81	0.81	0.80	0.81
1563	1290	0.83	0.84	0.81	0.83
1558	1285	0.85	0.86	0.84	0.85
1553	1280	0.89	0.89	0.87	0.87
1548	1275	0.91	0.91	0.89	0.90
1543	1270	0.94	0.94	0.91	0.92
1538	1265	0.96	0.96	0.94	0.95
1533	1260	1.00	0.99	0.97	0.98
1528	1255	1.04	1.02	0.99	1.00
1523	1250	1.10	1.06	1.02	1.03
1518	1245	1.15	1.09	1.05	1.06
1513	1240	1.20	1.14	1.08	1.09
1508	1235	1.26	1.18	1.11	1.11
1503	1230	1.32	1.23	1.14	1.14
1498	1225	1.44	1.28	1.17	1.17
1493	1220	1.66	1.34	1.21	1.21
1488	1215	2.16	1.41	1.25	1.24
1483	1210	2.58	1.53	1.29	1.28
1478	1205		1.78	1.33	1.32
1473	1200		2.60	1.37	1.36
1468	1195			1.43	1.40
1463	1190			1.48	1.46
1458	1185			1.56	1.51
1453	1180			1.67	1.58
1448	1175			1.86	1.67
1443	1170			2.54	1.78
1438	1165			2.65	1.94
1433	1160				2.28
1428	1155				2.62
T Final	K	1485	1475	1443	1431
Datapoint	C	1212	1202	1170	1158

Table 4: Summary of the calculated relative viscosity for all experiments. For simplicity we report interpolated values of the experimental data at equal temperature steps with exception of the final datapoint for some experiments; highlighted in grey. Temperatures related to those points are reported in the last two rows in Kelvin and Celsius, respectively.

Cooling Rate (K/min)		0.5	1	3	5
Shear Rate (sec <sup>-1</sup> )		0.77	0.77	0.77	0.77
T (K)	T (C)	log $\eta$ (Pa s)			
1573	1300	0.01	0.00	0.01	0.02
1568	1295	0.00	-0.01	0.00	0.01
1563	1290	-0.01	0.00	-0.02	0.00
1558	1285	-0.02	-0.01	-0.01	-0.01
1553	1280	-0.01	-0.01	-0.01	-0.02
1548	1275	-0.01	-0.01	-0.03	-0.02
1543	1270	-0.01	-0.01	-0.02	-0.02
1538	1265	-0.01	-0.01	-0.02	-0.02
1533	1260	0.01	0.00	-0.02	-0.03
1528	1255	0.04	0.00	-0.02	-0.04
1523	1250	0.10	0.02	-0.01	-0.03
1518	1245	0.16	0.04	-0.01	-0.03
1513	1240	0.22	0.07	0.00	-0.04
1508	1235	0.28	0.11	0.01	-0.04
1503	1230	0.36	0.15	0.02	-0.04
1498	1225	0.57	0.20	0.04	-0.04
1493	1220	1.00	0.27	0.06	-0.03
1488	1215	2.07	0.37	0.07	-0.02
1483	1210	3.01	0.56	0.10	-0.01
1478	1205		1.09	0.13	0.01
1473	1200		2.92	0.16	0.03
1468	1195			0.22	0.06
1463	1190			0.28	0.10
1458	1185			0.39	0.16
1453	1180			0.57	0.24
1448	1175			0.94	0.37
1443	1170			2.41	0.54
1438	1165			2.68	0.84
1433	1160				1.53
1428	1155				2.27
T Final	K	1485	1475	1443	1431
Datapoint	C	1212	1202	1170	1158