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## **Advances in the rheology of natural multiphase silicate melts: Import for magma transport and lava flow emplacement**

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**Abstract**

A review of recent advances in the field of rheology of multicomponent silicate melts and multiphase silicate melt suspensions is presented here. The advances include the development of new experimental devices and field and remote sensing methods for measuring the rheological properties of natural melts and magmas. These promising approaches combine laboratory experiments, theoretical models, numerical simulations and remote sensing data derived from ground, airborne and satellite-based tools. Each of these sub-disciplines has evolved rapidly in recent years and the growing range of complementary data appears now to provide an opportunity for the development of multi-disciplinary research. Ultimately, these multidisciplinary initiatives seek to provide near-real-time forecasting of hazardous volcanic processes such as lava flow field evolution. The results and approaches described here focus on multiphase (i.e. melts, bubbles, crystals) rheology of natural systems and are pertinent to the effusive emplacement of lavas, dykes and sills, as well as, to the eruption dynamics attending explosive eruptions.

Keywords: Multicomponent and Multiphase silicate melts, Rheology,

## 1. Introduction

The transport of magmas and volcanic materials is characterized by very dynamic, interdependent and complex, physical and chemical processes that all are affected by and affect the materials physical properties. Understanding the dynamic processes operating during magma ascent and eruption and the timescales and mechanisms of emplacement, welding and remobilization of fragmental or massive volcanic deposits, constitutes one of the main challenges in the Earth sciences (Dingwell, 1996; Papale, 1999; Sparks, 2004; Russell and Quane, 2005; Giordano et al., 2005). Accurate description of these processes requires the characterization of a wide range of transport and thermodynamic properties for the melt or magma (e.g. viscosity, density, enthalpy, entropy, heat capacity, thermal conductivity, solubility of volatile phases). These properties play crucial roles at micro- to macroscopic scale and many are correlated, in a non-linear manner (e.g. Richet et al., 1984; Giordano et al., 2008a; Russell and Giordano, 2017).

Lava flow dynamics are strongly governed by subsurface buoyancy forces, resulting from the density contrast with the host rock, which push the magma toward the surface (e.g. Wilson & Head, 2016a; Wieczorek et al. 2001), and by the evolving internal and external frictional forces (e.g. with dyke, conduit wall and topography) that oppose to the movement of magmas and lavas (e.g. Nemeth, 2010; Cañón-Tapia, 2016, Dragoni, 1993; Dragoni et al. 2005; Giordano et al., 2007; Cashman et al., 2013; Kolzenburg et al., 2016a,b; 2018a,b; Hulme, 1974; Hiesinger et al., 2007, Chevrel et al., 2013, 2015, Castruccio et al. 2014).

The rheological properties of magmas undergo tremendous changes from transport in the subsurface to eruption or emplacement at the surface and to final deposition and cooling. These changes are caused dominantly by the evolving of thermo-chemical and deformational conditions, imposing phase transitions and therewith heterogeneous textural and morphological variations of the magmatic and volcanic suspensions which evolve in space and time. The complex rheological evolution of lava flows can tentatively be constrained by carrying out laboratory measurements

60 under controlled conditions, simulating natural systems, and by monitoring flow emplacement at the  
61 field-scale and via satellite-based platforms. In parallel with this, the sophistication of physical  
62 models of lava flows and domes have improved significantly and are capable of providing fast  
63 simulations (see, amongst the others, Costa and Macedonio, 2003, 2005; Del Negro et al, 2008,  
64 2013, 2016; Melnik and Sparks, 1999, 2005; Melnik et al., 2009; Kilburn 2015 for reviews on this  
65 topic). These models are increasingly informed by, or validated by, satellite-derived parameters  
66 such as lava flow discharge rate or periodic updates on flow advance/geometry. Together these  
67 capabilities represent an emergent strategy that may provide timely reliable projections of lava flow  
68 field evolution and derive information for hazard assessment and mitigation measures. Yet, to date  
69 they do not always provide coherent results reproduced in nature.

70 This highlights the necessity to estimate the rheological properties of magmas and volcanic  
71 materials at conditions pertinent to nature and to investigate the effect of each variable over the  
72 range of relevant environmental conditions (e.g. pressure, temperature, volatile contents) during  
73 varying thermodynamic (equilibrium and non-equilibrium) conditions, and deformation regimes.

74 Our understanding of the single- and multi-phase (liquid+crystals+bubbles) rheology of  
75 magmas and volcanic products has greatly improved in the last two decades. This can largely be  
76 attributed to the growing availability of empirical data from the following sources (each of which  
77 will be reviewed in detail below):

78 1) laboratory experimentation on natural and simplified silicate melts. These data support  
79 the creation of robust models for predicting the Newtonian viscosity of pure liquid natural melts as  
80 a function of temperature (T), pressure (P) composition (X), volatile content ( $X_v$ ) and structural  
81 features (see Chapter 2).

82           2) the rheological experimentation and modelling of non-reactive multiphase suspensions  
83 (liquid+bubbles; liquid+crystal and liquids+bubbles+crystals) constituted by analogue materials or  
84 simplified or natural silicate melts mixtures (Chapter 4);

85           3) dynamic cooling rheological measurements on natural multiphase suspensions at non-  
86 isothermal and non-equilibrium conditions to explore the interdependent effects of composition,  
87 cooling-rate, shear-rate and oxygen fugacity acting during magma and lava transport in nature  
88 (Chapter 5);

89           4) rheological measurements of actively flowing lava. These represent snapshots of actual  
90 lava flow rheology at specific conditions and provide data that helps to constrain the conditions  
91 required to be reproduced in systematic laboratory studies (Chapter 6).

92           5) studies on the 3D and 4D evolution of lava flows at increasing spatial and temporal  
93 resolution and contemporary estimates of effusion rate and flow development from satellite data.  
94 These provide data for cross correlation and benchmarking of laboratory measurements (Appendix  
95 A1) and to re-visit long standing methods for deriving rheological parameters from morphologic  
96 data (Chapter 6).

97           These studies document that the effective viscosity of natural silicate melts and magmas can  
98 span more than 15 orders of magnitude ( $10^{-1} - 10^{14}$  Pa s), primarily in response to variations in melt  
99 composition (X), dissolved volatile content ( $X_v$ ), temperature (T), pressure (P), as well as the  
100 proportions, size, and shape distributions of suspended solid and/or exsolved fluid phases (i.e.  
101 crystals and bubbles). The deformation rate, which in nature would depend on the discharge rate  
102 will determine whether flow behavior will be Newtonian (i.e. one for which there is a linear  
103 relationship between stress and strain rate; or spatial variation of velocity) or non-Newtonian (e.g.  
104 Caricchi et al., 2007, 2008; Costa et al., 2007a, 2009; Vona et al. 2011, Hess et al., 2009).  
105 Deformation rate also exerts an influence on the crystallization kinetics (Vona et al., 2013, Kouchi,

106 1986, Kolzenburg 2018). It may further determine whether the melt will deform viscously or  
107 elastically and, therewith, whether or not it will eventually fracture giving origin to effusive rather  
108 than explosive eruptive styles (Dingwell, 1996). Combined the above experimental data and  
109 computational models form a basis from which to understand the flow behavior of natural magmatic  
110 and volcanic suspensions.

111 In the following I present a review of the research advances in the rheological  
112 characterization of pure silicate melts and multiphase silicate mixtures (i.e. lavas and magmas)  
113 achieved in the past decades. I follow the structure of points 1-5 outlined above to group the  
114 individual fields. In the Appendices (A1-A3) I summarize the most commonly employed  
115 experimental devices and technological advances to measure the single and multiphase silicate  
116 melts also reporting the most common equations used to describe the viscosity variation as a  
117 function of P, T, X (Appendix A1) as well as suspended solids phase and/or porosity (Appendices  
118 A2 and A3). I conclude with a discussion of how new laboratory developments and of the growth in  
119 complementary datasets (e.g. remote-sensing; drone technology; high-speed calculation facility) is  
120 providing greater understanding of magma and lava transport on Earth.

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## 122 **2. Pure liquid melt Newtonian viscosity experiments and models**

### 123 ***2.1. T – dependent models for predicting melt viscosity***

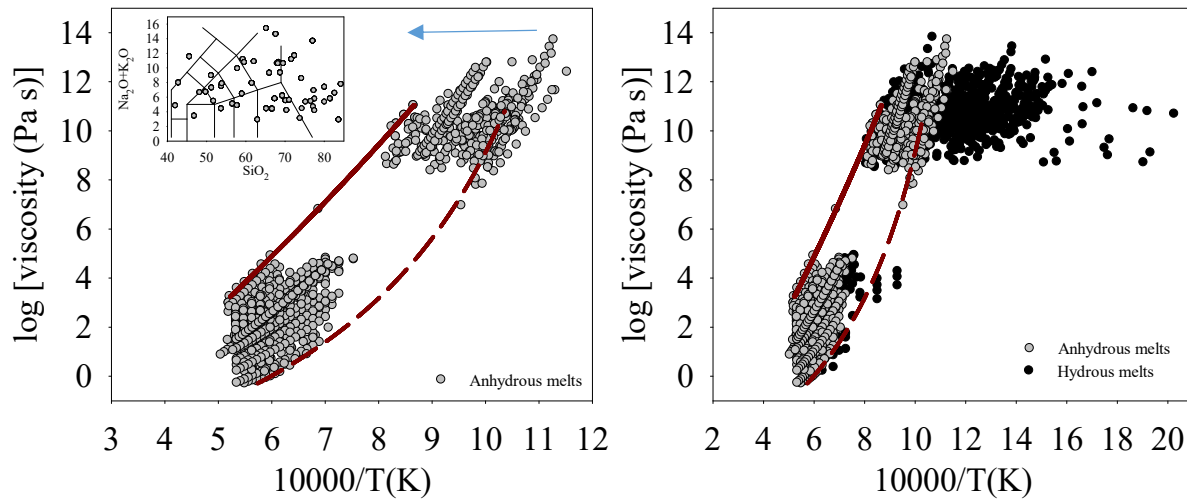
124 The first step toward characterizing multiphase rheology of natural silicate melts mixture is  
125 the knowledge of multicomponent viscosity of pure liquids as a function of their composition  
126 (including dissolved volatile species such as H<sub>2</sub>O, C and S –species, F, Cl) temperature (T) and  
127 pressure (P). Early models for predicting the viscosity of silicate melts were developed using data  
128 that spanned relatively small ranges of temperature (T) and viscosity ( $\eta$ ). These experimental data,  
129 restricted to superliquidus temperatures and a narrow compositional range, showed a nearly linear  
130 trend of viscosity in reciprocal temperature space. Thus, early models adopted an Arrhenian

131 formulation of the temperature-viscosity relationship (Shaw, 1972; Bottinga and Weill, 1972).  
132 Expansion of the melt viscometry database over a wider range of compositions and temperatures  
133 exposed the limitations of Arrhenian models. With the emergence of viscometry data closer to the  
134 glass transition temperature ( $T_g$ ) (i.e. the temperature of transition between a liquid-like and a solid-  
135 like behavior) (e.g. Angell, 1991, Giordano et al., 2005), the Arrhenian models proved unsuitable to  
136 describe the temperature dependence of silicate melt viscosity. These measurements were enabled  
137 by experimental devices that allow very small displacements to be monitored (e.g. Linear Voltage  
138 Displacement Transducers) and, the production of quenched glasses, freezing in the crystal free  
139 melt structure. In these experiments, supercooled glasses are reheated above  $T_g$ , where the “relaxed  
140 melt” viscosity (e.g. Angell, 1991; Scherer, 1984) could then be measured. These experiments are  
141 performed at timescales shorter than phase transitions timescale, therewith allowing anhydrous and  
142 hydrous pure liquid viscosity measurements (Angell, 1991; Scherer, 1984; Giordano et al., 2008b).

143         Based on the large number of experimental studies (e.g. Richet et al., 1995; Richet et al.,  
144 1996; Hess and Dingwell, 1996; Whittington et al., 2000, 2001; Giordano et al., 2009 amongst the  
145 others), models of melt viscosity were developed (e.g. Avramov, 1998; Angell, 1991; Russell et al.,  
146 2003; Giordano and Dingwell, 2003a, b; Russell and Giordano, 2005; Giordano and Russell, 2007;  
147 Hui and Zhang, 2007; Giordano et al., 2006, 2008a,b; Ardia et al., 2008; Mauro et al., 2009), also  
148 accounting for the non-Arrhenian viscosity behaviour (e.g. Vogel, 1921, Fulcher, 1925; Tammann  
149 and Hess, 1926; Adam and Gibbs, 1965). These models describe the P-T-X dependence of the  
150 viscosity of silicate melts. Some of the most relevant empirical and theoretical formulations  
151 describing the T-dependence of silicate melts and the relationships between constitutive parameters  
152 are reported in Appendix A2.

153         The growing database and the new models show that silicate melts display various degrees  
154 of non-Arrhenian behavior, from *strong* to *fragile* (Angell, 1991; Russell et al., 2002, 2003), which  
155 depend on composition and dissolved volatile content (Fig. 1).





157 *Fig. 1. The figure shows the variation of viscosity as a function of the reverse of temperature for the anhydrous melts*  
 158 *(a) and, per comparison, the anhydrous and hydrous melts (b) as reported by Giordano et al. (2008). The curves in a)*  
 159 *represent the most Arrhenian (continuous line in a))(strong) and the least Arrhenian (dashed curve in a) (fragile) melts*  
 160 *amongst those reported in panel of Fig 1a. The effect of water is that of significantly reducing viscosity and the fragility*  
 161 *(deviation from Arrhenian behavior) of the melts (details in Giordano et al., 2008).*

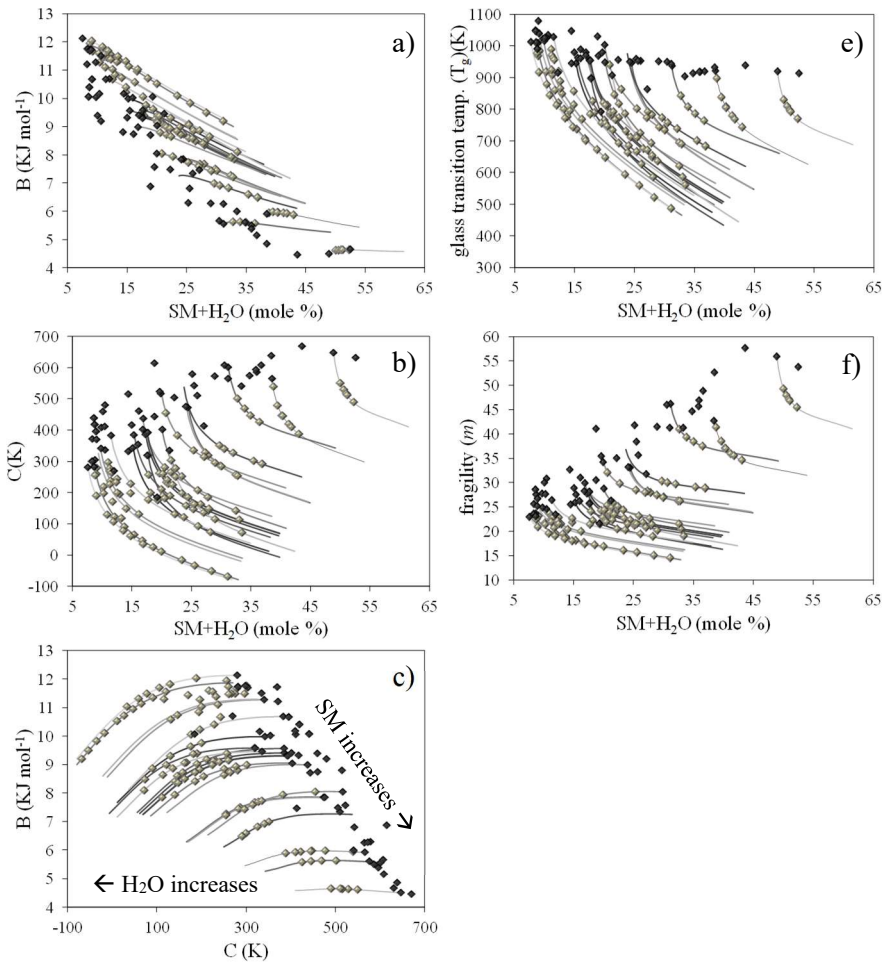
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163 All these models provide viscosity predictions based on composition commonly expressed  
 164 in terms of oxide abundances or combination of oxides and a range of adjustable parameters. Of the  
 165 various models only the HZ model (Hui and Zhang, 2007) and the GRD model (Giordano et al.,  
 166 2008a) accounts for the effects of dissolved volatile species (H<sub>2</sub>O, F). The GRD model is based on  
 167 the well-known VFT (Vogel-Fulcher-Tammann) equation, such that:

$$168 \quad \log [\eta \text{ (Pa s)}] = A_{\text{VFT}} + B_{\text{VFT}} / (T - C_{\text{VFT}}) \quad (\text{Eq. 1})$$

169 where  $A_{\text{VFT}}$  is the pre-exponential factor,  $B_{\text{VFT}}$  is the pseudo-activation energy and  $C_{\text{VFT}}$  is the VFT-  
 170 temperature. In contrast the HZ model uses a purely empirical T-dependent viscosity formulation of  
 171 non straightforward correlation with thermo-physical amounts. The GRD model has gained support  
 172 due to its simplicity and direct correlation of constitutive parameters (i.e. Appendix A2) to other  
 173 important physical and structural properties such as the glass transition ( $T_g$ ), the fragility ( $m$ ) (i.e.  
 174 the rate at which viscosity varies with temperature, that is an indication of melts capacity to store  
 175 energy), calorimetric properties (configurational entropy,  $S_{\text{conf}}$  and the configurational heat  
 176 capacity  $C_p^{\text{conf}}$ ; see Eq A2.4)(e.g. Giordano & Dingwell, 2003a; Giordano et al., 2008b; Chevrel et

177 al., 2013; Giordano and Russell, 2017; Russell and Giordano, 2017) and structural properties (e.g.  
178 Qn-species and Raman Ratio)(i.e. Le Losq and Neuville., 2017; Giordano and Russell, 2018;  
179 Giordano et al., 2019). These models show that, to a first approximation, the viscosity of silicate  
180 melts can be correlated at constant temperature to empirical, composition-based pseudo-structural  
181 parameter (i.e. the SM - structural modifiers - and the NBO/T - i.e. the Non Bridging Oxygen over  
182 Tetrahedra - parameters). The NBO/T and SM parameters are commonly assumed as proxies for the  
183 degree of polymerization of silicate melts and glasses (e.g Giordano and Dingwell, 2003a, b;  
184 Giordano and Russell, 2018; Giordano et al., 2019)(Figs. 2, 3). Compositions with low values of the  
185 SM-parameter (or low NBO/T values) are associated to strong (Arrhenian-like) rheological  
186 behavior, i.e. a linear behavior in the  $\log\eta$ - $1/T$  space, and more polymerized melts. On the other  
187 hand high values of SM (or high NBO/Ts) are related to more depolymerized melts which show  
188 fragile rheological behavior (i.e. the  $\log\eta$  vs  $1/T$  paths are significantly non-linear)(e.g. Angell,  
189 1991; Giordano and Dingwell, 2003)(Figs. 1-2). Russell et al. (2003), in agreement with early  
190 theoretical studies (e.g. Angell, 1991 amongst others), showed that the pre-exponential factor of the  
191 VFT and AG formulations, i.e. the viscosity at infinite temperature (Appendix A2), is a constant  
192 independent of compositions (Russell et al., 2003; Giordano et al., 2008a). The current models are  
193 applicable within the compositional space that they are based upon, but some compositional regions  
194 (e.g. peralkaline compositions) still remain unmapped and the models struggle to reproduce  
195 measured viscosity values (Giordano et al., 2006, 2008a, Di Genova et al., 2017). Those  
196 formulations also put in evidence that the role of water ( $H_2O$ ) dissolved in the melt is  
197 counterintuitive being opposite to that of network modifier cations. In fact, although dissolved  $H_2O$   
198 strongly decreases the viscosity of silicate melts (Fig 1b), the parameters describing the T-  
199 dependence of viscosity (e.g.  $B_{VFT}$  and  $C_{VFT}$  in Eq. 1) are differently affected by  $H_2O$  and by the  
200 most common structure modifiers (Fig. 2).



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Fig. 2. Relationships between constitutive parameters of the GRD model (Giordano et al., 2008), based on the VFT formulation (Eq. 1), as a function of the modified SM (Structure Modifiers) parameter (Giordano and Dingwell, 2003). The role of increasing SM on the constitutive parameters of anhydrous melts (black symbols) is that of decreasing  $B_{VFT}$  and increasing  $C_{VFT}$  (Fig. 2a, b) while increasing the fragility ( $m$ ) (Fig. 2f). On the other hand adding  $H_2O$  to the melt structure (gray symbols) results in decreasing  $B_{VFT}$  while decreasing  $C_{VFT}$ , the glass transition temperature  $T_g$  (as taken at a viscosity of  $10^{12}$  Pas) (Fig. 2e) and the fragility ( $m$ ). This observation put in evidence that the structural role of  $H_2O$  is different from that of those cations which simply modify silicate melts structure (Giordano et al., 2008, 2009).

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## 2.2. P – dependent models for predicting melt viscosity.

211

Measuring the effect of pressure (P) on the viscosity of melts is a complex experimental task

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and, as a result, has not been investigated extensively. A short summary of applied techniques and

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technological advances is reported in Appendix A1, together with some of the main results.

214

Largely, the available data imply that the viscosity of silica-rich melts decreases with increasing P,

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whereas the viscosity of silica-poor melts increases as pressure increases (Liebske et al., 2005;

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Ardia et al., 2008 and references therein). However, the available data suggest that the effect of P is

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negligible at near surface conditions pertinent to explosive and effusive volcanism. As a

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consequence this effect will not be discussed any further in this contribution. Fig. A2.1 shows for

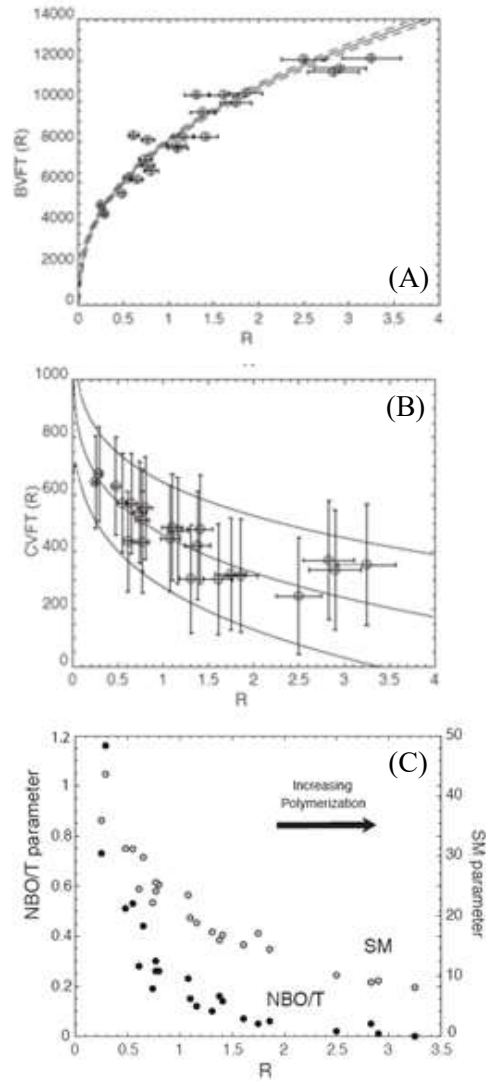
219 the Ab-Di system what is the effect of P which changing composition in the binary system, by using  
220 fitting procedure as adopted by Ardia et al. (2008). This system is considered to show what is the  
221 effect of P on polymerized (Ab) to depolymerized (Di) synthetic compositions from low to high P.  
222 Similar behaviours is expected for natural compositions, but, as shown by previous authors (e.g.  
223 Giordano et al., 2008b; Chevrel et al., 2013; Whittington et al., 2009), simplified systems (e.g. An,  
224 Di, Ab) should not be considered as proxies for natural compositions.

225

### 226 ***2.3. Toward a structural model for geological melts.***

227 More recently, Le Losq and Neuville (2017), Giordano and Russell (2018) and Giordano et  
228 al. (2019), following different approaches, showed that the viscosity of simple and multicomponent  
229 anhydrous silicate melts over a temperature interval of  $\sim 700$  to  $1600^\circ\text{C}$ , can be predicted from the  
230 Raman spectra obtained from the corresponding glasses (i.e. fast quenched melts). These methods  
231 prove to be very promising methods for in situ rheological investigations and may have great  
232 importance for planetary sciences studies (Angel et al., 2012; Giordano and Russell, 2018). Le Losq  
233 and Neuville (2017) developed a 13 - parameters model for melt viscosity in the simple system  
234  $\text{SiO}_2\text{-Na}_2\text{O-K}_2\text{O}$  which connects the transport and thermodynamic properties of these simple melts  
235 explicitly to the structural state of the melt expressed via the abundances of  $Q^n$  -species recovered  
236 from Raman spectral analysis of the glasses. Giordano and Russell (2018) first and Giordano et al.  
237 (2019), later, presented a first order model predicting the viscosity of multicomponent natural  
238 melts by the employment of the so-called Raman  
239 ratio (R) and normalized Raman ratio ( $R_n$ ) derived by Raman spectra measured on the  
240 corresponding glasses as defined by Mercier et al. (2009, 2010). As shown in Fig. 2 a strong  
241 relationship exists between  $B_{VFT}$  and  $C_{VFT}$  parameters and R which allows the viscosity of  
242 anhydrous multicomponent natural melts to be predicted with a great accuracy. Although, the model  
243 requires expansion to use of the structural information of volatile-bearing melts, it allows accurate

244 description of the viscosity of anhydrous melts by the employment of a simple equation with 6  
 245 adjustable parameters and the measured R. Also the SM and NBO/T parameters, calculated from  
 246 compositions, are shown to be strongly correlated with R.



247

248 *Fig. 2. Model VFT parameters  $B_{VFT}(R)$ (A) and  $C_{VFT}(R)$ (B) as defined by Giordano and Russell (2018) (panels A, B)*  
 249 *and relationships between pseudo-structural parameters (SM, NBO/T)(C), as a function of the Raman ratio (R).*  
 250 *According to the above mentioned authors:  $B_{VFT}(R) = b_1 R^{b_2}$  and  $C_{VFT}(R) = c_1 R^{c_2} + c_3$  where  $b_1, b_2, c_1, c_2, c_3$  are*  
 251 *adjustable parameters.*  
 252

### 253 **3. From pure liquids to multiphase analogues and magmas: advantages and** 254 **disadvantages of the different experimental approaches.**

255 Being magmas and volcanic materials very complex mixtures of crystals and vesicles  
 256 suspended in a silicate melt phase which evolve as a function of the evolving P, T and  
 257 compositional variations and dynamic regimes, the description of effect of suspended phases on the

258 viscosity of these natural suspensions has followed different approaches. The early models devoted  
259 to describe the multiphase rheology were historically based on the investigation of analogue  
260 materials (e.g. Einstein, 1906; Einstein and Roscoe, 1952). More recently, the basis for the  
261 description of natural multiphase suspensions has been largely developed using natural and  
262 simplified silicate melts mixtures at experimental conditions at around thermodynamic equilibrium  
263 (e.g. Campagnola et al. 2016; Chevrel et al. 2015; Robert et al. 2014; Sehlke et al. 2014; Soldati et  
264 al. 2016; Vona and Romano 2013; Vona et al. 2011, 2013). As such, their application to natural  
265 environments requires extrapolation into the thermal and deformational disequilibrium state at  
266 which magmatic and volcanic processes commonly operate. This is only possible to a limited  
267 extent, as natural magmatic and volcanic processes often operate quite far from equilibrium. Recent  
268 studies on the disequilibrium rheology of crystallizing natural silicate melts have documented that  
269 deformation-rate and cooling-rate may significantly affect the phase transitions of magmatic  
270 mixtures so to forcing the material toward a thermal and mechanical disequilibrium state (e.g.  
271 Giordano et al., 2007; Kolzenburg et al., 2016, 2017a, b; 2018a,b; Arzilli and Carroll, 2013).

272         Following, the experimental efforts aimed at retrieving information of the multiphase  
273 rheology of natural silicate mixtures have been broadly summarized by subdividing it into,  
274 experiments on non reactive materials (chapter 4) or reactive silicate melts mixtures undergoing  
275 variable thermal or deformational variation (chapter 5). These kind of experiments can be further  
276 subdivided into three main categories: a) experimentation on analogue materials; b) experiments on  
277 simplified silicate mixtures and c) experiments on natural volcanic products. Each of these  
278 experimental approaches has different advantages/disadvantages which are listed below.

279

### 280 ***3.1. Analogue materials***

281         Multiphase analogue materials are commonly constituted by non-reactive mixtures of mono-  
282 or poly-disperse particles and/or bubbles, with varying content and shape and size distributions,

283 immersed in some Newtonian synthetic fluid (e.g. silicon oil, syrup, liquid paraffin), which can be  
284 investigated at room temperature. These kinds of multiphase mixtures can normally be investigated  
285 at room temperature conditions and therefore their rheological characterization is simplified as it  
286 does not involve the need for high temperature or pressure equipment and the sample texture can  
287 readily be controlled (e.g. the solid/bubble proportion or variation). These kinds of experiments are  
288 commonly performed on transparent multiphase mixtures and therefore allow observing and  
289 characterizing strain partitioning processes occurring amongst the phases during the deformation.  
290 The main disadvantage of this kind of experimentation is that they cannot reproduce neither the  
291 transient disequilibrium processes occurring in natural mixtures (e.g. crystallization or degassing  
292 stages) nor the natural dynamic physical properties of silicate melts (e.g. viscous and cohesive  
293 forces between the natural residual melt and suspended particles and bubbles). The largest part of  
294 these studies investigate two phase suspensions of either liquid and solid particles (simulating  
295 crystal bearing magma) or liquid and bubbles or vesicles (simulating the exsolution of volatile  
296 gases).

297

### 298 ***3.2. Multiphase silicate melt suspensions***

299 The experiments of categories b) and c) require significantly more complex experimental  
300 infrastructure and are substantially more complex to be characterized in terms of textural  
301 parameters (crystal and bubble content; crystals and bubbles size and shape distributions) but they  
302 offer the opportunity to perform measurements on materials with direct application to the Earth  
303 Sciences. The inherent inhomogeneity of geo-materials and the large variations of the size- and  
304 shape-distributions found in natural products can, to date, not be captured in a satisfactory manner  
305 by the available theoretical or empirical models. Experiments on natural materials at controlled  
306 conditions have the advantage of being representative of natural scenarios and, in most cases, they  
307 allow retrieving, at least, the final stage of textural evolution as a function of the imposed

308 environmental conditions (i.e. isothermal; non-isothermal; isobaric; non-isobaric) as well as varying  
309 deformation regimes (i.e. constant or varying stress and/or strain rate). This allows the  
310 reconstruction of the rheological parameters in a tightly constrained parameter space, however it  
311 requires unique experimental characterization for each studied scenario. When volatile free samples  
312 are investigated, this kind of experiments can be performed, using a variety of experimental  
313 techniques (e.g. rotational concentric cylinder/Patterson deformation rig and uniaxial compression  
314 and/or micropenetration and parallel plates techniques; see Appendix A1 for details), over the entire  
315 temperature-viscosity interval from super- to sub-liquidus conditions that are characteristic of  
316 natural environments. For volatile-bearing natural melts and suspensions, this becomes more  
317 complex as limited experimental infrastructures exist to date to measure at the elevated pressures  
318 required to maintain volatiles in solution. There have been some recent advances which take  
319 advantage of the metastable liquid state close to  $T_g$  or by using devices which allow the sample to  
320 be pressurized (Paterson, 1978; Paterson and Olgaard, 2000; Caricchi et al., 2007; 2008; Ardia et  
321 al., 2008; Robert et al., 2008a, b; Piermarini et al., 1978). A further advancement is the 4D  
322 characterization of the sub liquidus evolution of natural melts is represented by experiments within  
323 synchrotron facilities which allow real time monitoring of the textural evolution of samples of  
324 volcanological interest during crystallization and/or degassing (e.g. Ohtani et al. 2005; Pistone et al.  
325 2015; Pleše et al. 2018; Polacci et al. 2018; Polacci et al. 2010; Song et al. 2001). These techniques  
326 are starting to be coupled with devices for rheometry, which may in the future allow for in situ  
327 measurements of both the crystallization kinetics and the rheological response of evolving natural  
328 systems (Coats et al. 2017; Dobson et al. 2015; Dobson et al. 2016; Raterron and Merkel 2009). The  
329 results on experimental campaigns and modelling of the multiphase rheology of natural magmatic  
330 suspensions performed on natural or analogue silicate melts at high temperatures will be presented  
331 in § 4.2.

332



## 333 4. Experiments and models of non-reactive multiphase mixtures

334 Following the results obtained on isothermal bubble-bearing or particle-bearing suspensions  
335 rheology of analogue materials, simplified silicate melt mixtures and natural melts and magmas are  
336 here first introduced. Finally I summarize what is known on the effect of the presence of  
337 bubbles+crystals on suspension rheology measurements performed at constant temperature.

338

### 339 4.1. Models of bubble suspension rheology

340 Early studies estimating the effect of void spaces within natural and simplified silicate melts  
341 (e.g. Bagdassarov and Dingwell 1992, 1993; Lejeune et al. 1999; Vona et al., 2016; Ryan et al.,  
342 2019) or synthetic analogues (e.g. Manga et al., 1998; Llewellyn et al. 2002a, b; Llewellyn and  
343 Manga, 2005) has been carried out by several authors. Those investigations showed, largely, that,  
344 two end member cases can be considered: 1) bubbles behave as rigid objects (capillary number  
345  $Ca < 1$ ); 2) bubbles are deformed ( $Ca > 1$ ) (Llewellyn et al., 2002a,b). For the different regimes various  
346 empirical equations were proposed (Bagdassarov and Dingwell., 1992; Lejeune et al., 1999;  
347 Llewellyn et al., 2002; Llewellyn and Manga, 2005) (details in Appendix A3) which suggested, that  
348 during steady flow: a) an increase in relative viscosity in the case of the first end-member ( $Ca < 1$ )  
349 and b) a decrease of the relative viscosity in the case of second end member condition ( $Ca > 1$ ) can  
350 be observed. Additional complexities are introduced, as discussed in Appendix A3, for non-steady  
351 flow for which the definition of a dynamic capillary number ( $Cd$ ) is required. The same authors  
352 (e.g. Lejeune and Richet, 1996; Bagdassarov and Pinkerton, 2004, Llewellyn et al., 2002, Llewellyn  
353 and Manga, 2005) also provided important attainments concerning the understanding of the effect  
354 of closed and opened voids on liquid viscosity (e.g. Lejeune et al., 1999; Bagdassarov and  
355 Dingwell, 1992; Quane and Russell, 2004; Llewellyn et al., 2002a, b; Llewellyn and Manga, 2005;  
356 Mader et al., 2013; Vona et al., 2016; Ryan et al., 2019). A summary of recent formulations and

357 works related to both crystal bearing and bubble bearing rheological studies is reported in Appendix  
358 A3.

359

#### 360 ***4.2. From shear-rate independent to shear-rate dependent particle suspension rheology*** 361 ***models of analogue materials***

362 Early studies on the rheological behavior of multiphase suspensions (e.g. Einstein, 1906;  
363 Roscoe, 1952; Krieger and Dougherty, 1959; Gay et al., 1969; Pinkerton and Stevenson, 1992)  
364 suggested a threshold in solid fraction, the so-called crystal maximum packing fraction ( $\phi_c$ ), that  
365 separates a liquid dominated rheology from a solid-dominated rheology. For dilute suspensions of  
366 solid mono-disperse spherical particles ( $\phi < 3$  vol%) Einstein (1906) proposed that the relative  
367 viscosity  $\eta_r$  (i.e. the ratio between the viscosity of the particle-bearing suspension and that of the  
368 particule-free melts) could be calculated as:  $\eta_r = (1 + B\phi)$ , where  $B$  is a constant depending on object  
369 geometrical features ( $B=2.5$  for spheres). Roscoe (1952) extended Einstein's expression to higher  
370 concentration of spheres, by first defining the maximum crystal packing fraction ( $\phi_m$ ) and providing  
371 for the relative viscosity the following expression:  $\eta_r = (1 + \phi/\phi_m)^{-2.5}$ . Different  $\phi_m$  values were  
372 proposed by different authors depending on crystal geometry (see appendix A3 for more details).  
373 Later, Krieger and Daugherty (1959) generalized the previous expressions as it follows:  $\eta_r =$   
374  $(1 + \phi/\phi_m)^{-B_e\phi_m}$  where  $B_e$  is a constant called the Einstein coefficient (KD model). Others similar  
375 expressions were formulated for which different value of the  $B_e$  coefficients were determined (see  
376 appendix A3). Although widely applied, a limitation of those empirical or semi-empirical laws is  
377 that they do not account for neither the strain-rate dependence nor the existence of, although still  
378 debated, yield strength (Moitra and Gonnermann, 2015) of multiphase mixtures typical of non-  
379 Newtonian fluid (see appendix A3 for more details). For a review on the two phase rheology of  
380 particle bearing analogue suspensions the reader can refer to Mader et al. (2013), who presented a  
381 comprehensive review on this topic.

382

383 ***4.3. Non-Newtonian models for particle suspension rheology of simplified silicate mixtures***

384 Concerning magma-equivalent suspensions, more recently Caricchi et al. (2007), Costa et al.  
385 (2007a, 2009), based on the available experimental data obtained at constant temperature, presented  
386 models describing the non-Newtonian strain-rate-dependent rheological effects of crystals in the  
387 range of solid fractions from 0 to 0.8 and over. These models cover the transition from the regime  
388 where the deformation behavior is controlled by melt viscosity up to the beginning of the regime  
389 where the deformation behavior is controlled by a solid framework of interlocking particles. The  
390 most detailed and comprehensive model to date proposed by Costa et al. (2009) model (CM) (Eqs.  
391 A3.3-A3.4.), describes the relative viscosity  $\eta_r$  (i.e. the viscosity of a crystal melt mixture ( $\eta_{\text{mix}}$ )  
392 divided by the viscosity of the melt phase ( $\eta_l$ )). The CM model is the result of the combined  
393 mathematical and experimental efforts condensed in the works of Costa (2005), Costa et al. (2007a)  
394 that was used by Caricchi et al (2007) to describe their experimental data. Compared to previous  
395 models (e.g., Einstein-Roscoe, 1952; Costa, 2005; Caricchi et al., 2007a), the CM model accounts  
396 for the strain-rate dependent changes in the rheology of liquid+crystal mixtures. The model in  
397 particular shows that the strain rate dependence of the relative viscosity at varying crystal volume  
398 fractions follows a sigmoid curve with exponential increase above a critical solid fraction ( $\phi_c \sim 0.3$ -  
399 0.4). This model is consistent with the early Einstein-Roscoe equation (Einstein, 1906; Roscoe,  
400 1952) for crystal fractions in the range of 0 to 0.1-0.3 depending on crystal shape and size (e.g.  
401 Cimarelli et al., 2011). A summary of the main results obtained by the employment of the CM and a  
402 summary of its original formulation are reported in Appendix A3. Extension of CM devoted to  
403 characterize the effect of crystal size and shape distribution and suspended particle ratio and particle  
404 roughness are discussed in Appendix A3.

405

406 ***4.4. Non-Newtonian strain-rate dependent models for particle suspension rheology of***  
407 ***natural mixtures.***

408 The fermenting production of studies (Shaw et al., 1968; Lejeune and Richet, 1995;  
409 Giordano et al., 2007; Caricchi et al., 2007, 2008; Ishibashi, 2009; Vetere et al., 2010, 2017; Vona  
410 et al., 2011; Pistone et al., 2012, 2016; Chevrel et al., 2015, 2017; Campagnola et al., 2016) devoted  
411 to the characterization of the isothermal viscosity evolution of silicate melts at subliquidus  
412 temperature as a function of presence and size and shape distributions of crystals and bubbles and  
413 deformation regimes of the last twenty years has permitted extraordinary advances that are  
414 condensed in empirical and theoretical models of suspension rheology (Saar et al., 2001; Caricchi et  
415 al., 2007; Costa et al., 2009; Mueller et al. 2011; Vona et al., 2011; Moitra and Gonnermann, 2015).  
416 According to the comprehensive model of Costa et al (2009)(CM)(see § 3.1), inspired by the  
417 previous work of Costa (2005), Costa et al. (2007a) and Caricchi et al (2007), the relative viscosity  
418 of two-phase mixture increases following a sigmoid curve with exponential increase above a critical  
419 solid fraction ( $\phi_c$ ) corresponding to the first ( $\phi \sim 0.3-0.4$ ) inflection point. A second inflection point  
420 ( $\phi_m$ ) at  $\phi \sim 0.6-0.7$  is determined by the beginning of crystal dominated rheology (Fig. A3.3).

421 Since the seminal contributions of Caricchi et al. (2007) and Costa et al. (2009), numerous  
422 scientists provided new and more complete formulation of the critical crystal fraction ( $\phi_c$ ) for the  
423 natural variability in of crystal size and shape distribution which would also account for new  
424 variables (e.g. crystal surface roughness)(e.g. Mueller et al. 2011; Klein et al., 2018). The  
425 employment of these critical contributions have allowed interpreting, based on model calculations,  
426 the effect of rheological constraints on eruptive behavior.

427

428 ***4.5. Models for particles and bubbles suspension rheology***

429           Complex three-phase suspensions (i.e. liquid+bubbles+crystals) have been investigated in  
430 only a few studies (Cordonnier et al., 2009; Robert et al., 2008a, b; Lavallée et al., 2007, 2008;  
431 Vona et al., 2013, 2017; Campagnola et al., 2016; Pistone et al., 2012, 2015, 2016). Given their  
432 complexity only a few studies have provided preliminary models describing the complex  
433 rheology of three-phase mixtures (Pistone et al., 2012, 2013, 2015, 2016). The viscosity data  
434 presented in those studies are the same as those presented in Pistone et al (2012), but the authors  
435 apply their results to different geological context by showing that size- and shape-distributions of  
436 crystals and bubbles may significantly vary while undergoing certain stress-strain regimes. The  
437 experiments by Pistone et al. (2012) were performed at pressurized and isothermal temperature  
438 conditions in a Paterson device (Appendix A1) on samples for which the liquid+crystal rheology  
439 was characterized by Caricchi et al. (2007). They show that bubbles strongly affect the rheological  
440 properties of crystal-rich mushes. By presenting a comprehensive review of existing literature and  
441 performing new measurements, they estimated that a decrease of up to 4 orders of magnitude is  
442 observed by the addition of only 9 vol% of bubbles to a liquid+crystals suspension containing 70  
443 vol% of crystals. They also established that two non-Newtonian deformation regimes originate as a  
444 consequence of the bubble and crystal interaction: i) a shear thinning behavior result of the crystal  
445 size reduction and shear banding due to strain localization (also observed by Caricchi et al. 2008)  
446 which is typical of magmas which are transported and emplaced in Earth's crust and may feed  
447 eruptions; ii) a shear thickening behavior which is the consequence of crystal interlock and flow  
448 blockage which they argue locks plutonic rocks in the lower and upper crust, inhibiting eruptions.  
449 More details of the results obtained by the works of Pistone and coauthors are provided in Appendix  
450 A3.3.

451

452           **5. Non-Isothermal cooling-rate and strain-rate dependent rheology of volcanic**  
453 **materials**

454           Efforts to systematically describe and predict magma migration and lava flow behavior rely  
455 heavily on these experimental measurements to derive empirical models. However, during  
456 migration and transport of silicate melts in the Earth's crust and at its surface magma/lava can  
457 experience varying cooling and deformation conditions which may drastically influence its  
458 thermorheological evolution; see for example Rhéty et al. (2017) and Robert et al. (2014). As a  
459 consequence, data intended for application to the natural environment will have to account for the  
460 disequilibrium behavior of natural magmatic suspensions. Cooling rates of basaltic lavas, measured  
461 at the surface and within active lava channels during emplacement range from ~0.01 to 15 °C/min  
462 (Cashman et al., 1999; Flynn and Mougini-Mark, 1992; Hon et al., 1994; Witter and Harris, 2007;  
463 Kolzenburg et al., 2017). These values are largely representative for the exterior part of lava flows  
464 or for the initial cooling of newly emplaced dikes. They can, therefore, be taken as maximum  
465 cooling rates that are expected to be lower in the interior of the lava flow or a cooling dike. The  
466 importance of varying thermal conditions on the crystallization kinetics and textural development of  
467 silicate melts has been recognized for decades and inspired disequilibrium experimentation in  
468 petrology and volcanology (e.g. Walker et al., 1976; Arzilli and Carroll, 2013; Coish and Taylor,  
469 1979; Gamble and Taylor, 1980; Hammer, 2006; Lofgren, 1980; Long and Wood, 1986; Pinkerton  
470 and Sparks, 1978; Giordano et al., 2007; Vetere et al., 2013). These studies highlight that significant  
471 differences in textures and paragenesis emerge when moving from equilibrium to disequilibrium  
472 conditions that can, in turn, affect the flow behavior. Albeit a growing experimental disequilibrium  
473 database is becoming available no models for the disequilibrium phase dynamics of natural silicate  
474 melts have been developed to date.

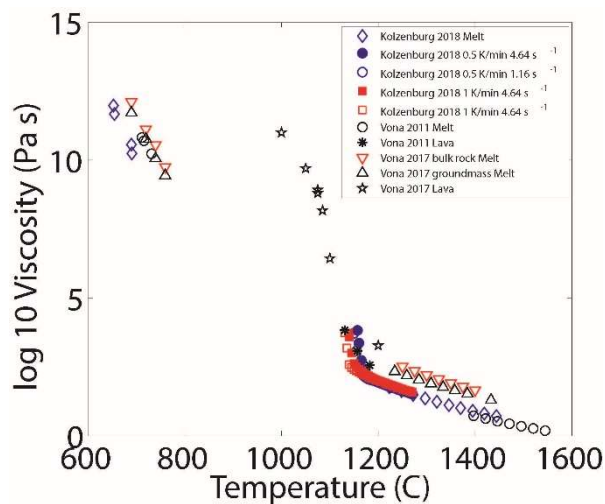
475           Understanding the rheological evolution of crystallizing melts, magmas and volcanic  
476 products requires direct measurement of the flow properties of investigated materials at such  
477 disequilibrium conditions in the field or in the laboratory. In such environments, the studied  
478 materials are degassed and undergoes transient increases in viscosity as it is increasingly

479 undercooling until a “rheological cut-off temperature” (Giordano et al., 2007, Kolzenburg et al.,  
480 2016, 2017, 2018a, b, c; 2019) is reached and the lava rheologically solidifies. This transient  
481 rheological gradient, which occurs in all natural, non-isothermal environments, governs the lavas  
482 emplacement style. In recent years, the first sets of measurements were presented that constrain the  
483 rheological evolution of natural silicate melts under temperature- and deformation-conditions  
484 pertinent to the transport of silicate melts on the earth’s surface and in shallow magma plumbing  
485 systems. The recovered data show a strong dependence of composition (Kolzenburg et al., 2017,  
486 2018a), cooling-rate (Giordano et al., 2007, Kolzenburg et al., 2016, 2017), oxygen fugacity  
487 (Kolzenburg et al., 2018a) and shear-rate (Kolzenburg et al., 2018) on the thermorheological  
488 evolution of natural silicate melts. They represent the first contributions to a growing database of  
489 lava rheology under natural conditions. However, significant experimental effort in this field is  
490 required to expand the range of available data to cover the most relevant compositions and to  
491 experimentally map the range of parameters pertinent to flow of natural silicate melts under  
492 disequilibrium. Such a database would then allow deducing the underlying processes and  
493 expanding these into a theoretical description of the flow behavior of magma and lava. So far, the  
494 main limitation of these kind of studies is the difficulty to monitor, and therefore extend, the results  
495 to non-degassed materials and therefore the application to intra-crustal magmatic or explosive  
496 volcanic processes. According to previous authors (e.g. Melnik and Sparks, 1999, 2005; Costa and  
497 Macedonio, 2003, 2005; Costa et al., 2007b; Hess et al., 2008; Cordonnier et al., 2012) an  
498 additional complexity could be due to the effects of nonlinear thermal effects, potentially generated  
499 by viscous dissipation and loss by conduction at the contact between the molten material and the  
500 hosting rock, in conduits, and channels or tunnels after eruption to the surface. The nonlinear  
501 behaviour of thermal effect are mainly governed by specific non-dimensional numbers (Graetz;  
502 Nahme; Prandtl; Reynolds; regime), which according to Costa et al. (2007b), amongst the others  
503 above mentioned, may determine the necessity to distinguish between three main regimes - a  
504 conductive-heat-loss-dominated regime, an intermediate regime and a viscous-heating-dominated -

505 may have significant effects for the definition of the rheological behaviour and emplacement  
506 dynamics of lava flows and lava domes.

507 Figure 3 shows a summary of rheological data recovered using a variety of experimental  
508 methods on Etna melts. The melt compositions, albeit stemming from different eruptions, are  
509 similar for most major oxides with the exception of the sample from Vona et al. (2017), that is more  
510 rich in silica and poor in iron and, as a result, more viscous than the samples in Vona et al. (2011)  
511 and Kolzenburg (2018).

512 For the investigated degassed materials these data summary highlights a number of effects  
513 acting during the transport of magma and lava at sub liquidus conditions. Comparison of the pure  
514 liquid viscosity of the remelted bulk rock and the separated groundmass (Vona et al., 2017); open  
515 triangles documents that, for basaltic melts, crystallization induced changes in melt composition  
516 result in relatively small changes in the viscosity of the liquid phase of the evolving suspension.



517  
518  
519 *Fig. 3. Summary of available melt and crystal-suspension viscosity data on remelted Etnean lava as a function*  
520 *of temperature. Melt viscosity measurements (open blue diamonds, black circles and black and red triangles) were*  
521 *performed via concentric cylinder viscometry, micro penetration and differential scanning calorimetry; Sub liquidus*  
522 *viscosity measurements were preformed using 1) concentric cylinder viscometry at constant temperature (black plus*  
523 *symbols), 2) concentric cylinder viscometry at varying cooling- and shear-rates (open and filled red squared and blue*  
524 *circles) and 3) parallel plate viscometry via unconfined uniaxial deformation (open black stars).*

525  
526 Therefore, the variations of the flow behavior of crystallizing basalts are controlled by  
527 variations in the volumetric fractions of crystals and bubbles. These data also reflect the



528 measurement limits of the respective methods that are described in more detail in Kolzenburg et al.  
529 (2016a). Concentric cylinder suspension viscometry for these Etnean lavas is confined to  $<10^4$  Pa s  
530 and shows that the measured viscosities at constant temperature (i.e. at or near thermodynamic and  
531 textural equilibrium) are commonly higher than non-isothermal measurements at the same  
532 temperature. This is due to the fact that under dynamic thermal conditions, the crystal nucleation  
533 and growth kinetics lag behind the equilibrium state and commonly produce lower crystal contents.  
534 The non-isothermal viscosity data from Kolzenburg et al. (2018c) document that both cooling-rate  
535 (blue circles vs. red squares) and shear rate (open vs. filled symbols) exert a modulating effect on  
536 the disequilibrium rheology of the Etna melt. Measurements beyond the mechanical limit of  
537 concentric cylinder (CC) viscometry were presented in Vona et al. (2017) who employed parallel  
538 plate (PP) viscometry via unconfined uniaxial deformation (open black stars) to measure the  
539 viscosity of three phase magmatic suspensions. The data form an apparent continuing trend with  
540 respect to the concentric cylinder viscometry measurements but document lower lava viscosities  
541 than extrapolation from the two phase measurements would suggest. This is likely a result of the  
542 differences in sample texture, where all CC data are restricted to bubble free two phase suspensions  
543 of crystals and melt, whereas the PP data are measured on three phase (i.e. crystal and bubble  
544 bearing) suspensions.

545 In summary, the rheological evolution of lava at sub-liquidus conditions can be  
546 reconstructed neatly by combination of datasets from differing sources. This is also shown in Figure  
547 3 in Kolzenburg et al., 2019 (this issue), where laboratory and field estimates of lava rheology at  
548 emplacement conditions are compared and the respective data fall within a range of similar values.  
549 This highlights the potential of cross correlation of data from different experimental and field  
550 sources and the need to expand the available experimental database in order to generate a holistic  
551 view of the dynamics of magma and lava transport.

552

553           **6. Alternative ways of retrieving rheological information from remote sensing ground-**  
554 **or satellite-based techniques.**

555           Besides laboratory viscometry (i.e. the direct measurement of melt / suspension viscosity  
556 under controlled conditions) there are several other sources of rheological information that are  
557 useful to place the laboratory measurements in context of the natural environment. This kind of  
558 information is important as it allows accounting for the multiphase nature of lava bodies and can  
559 serve to place the laboratory measurements within the framework of conditions relevant in natural  
560 scenarios. However, to date such data only represents a very limited source of information of the  
561 rheological evolution of lava flows, in space and time. This is largely due to large logistical and  
562 financial efforts required for some of these measurements and the uncertainties associated to others.  
563 Broadly these approaches can be separated into:

- 564 1. Direct measurement of viscosity on active lava flows via penetration- or rotational-viscometry  
565 (Einarsson, 1949; Gauthier 1973; Panov et al. 1988; Pinkerton and Sparks, 1978; Belousov et  
566 al. 2015; Belousov and Belousova, 2018; Shaw et al., 1968; Pinkerton and Norton, 1995;  
567 Pinkerton and Wilson, 1994; Chevrel et al., 2018). These represent snapshots of actual lava  
568 flow rheology at specific conditions and provide data that helps to constrain the conditions  
569 required to be reproduced in systematic laboratory studies. However, such measurements are  
570 quite difficult and require significant logistical effort and manpower. Further, the available  
571 devices (e.g. Belousov and Belousova, 2018; Chevrel et al., 2018) for such measurements are  
572 only slowly advancing to be able to measure all relevant parameters sufficiently well to  
573 recover high quality viscosity data (Appendix, A1.5).
- 574 2. Calculation of the apparent viscosity based on Jeffreys' equations (e.g. Jeffrey, 1925; Hulme,  
575 1974)(Appendix 4, SMO) using flow rate measurements of active lavas in channelized flows  
576 (Naboko 1938; Nichols 1939; Minakami 1951; Einarsson 1966; Walker 1967; Gautier 1973;  
577 Moore 1978; Andreev 1978; Fink and Zimbelman 1986; Vande-Kirkov 1987; Panov 1988;

578 Soldati 2016; Belusov and Belousova 2018). Such data are still few due to the difficulty of  
579 accessing active lava flows. However, the development of affordable unmanned aerial  
580 vehicles (UAV's) in recent years appears to be promising making this method widely  
581 applicable with the opportune considerations. In fact, the above mentioned approach has  
582 strong limitations as it is based on the assumption of parabolic velocity profile that is not  
583 generally valid because of thermal effects (e.g. Costa and Macedonio, 2003, 2005; Costa et  
584 al., 2007b; Filippucci et al., 2013 and Filippucci et al., 2019, this issue)(details at § 3.1.2).  
585 Such aspect is still never considered to describe the nonlinear dynamic of lava flows and lava  
586 domes rheology (Melnik and Sparks, 1999, 2005; Melnik et al., 2009). To an adequate  
587 analysis of this contribution for specific cases, it is recommended to refer, for instance, to the  
588 above mentioned works and e.g. Filippucci et al (2019)(this issue).

589 3. Ties between lava flow geometry and viscosity. Morphological-derived rheological  
590 parameters (i.e. viscosity and yield strength) are commonly obtained in planetary sciences  
591 (Heisinger et al., 2007; Castruccio et al., 2010 and Chevrel et al., 2015 provide excellent  
592 reviews of the employed equations and results). Rheological information is obtained by  
593 retrieving, in the field or remotely also from satellites, length, width, thickness and slope of  
594 emplacement of lava flows. This methodology has also been applied based remote sensing  
595 data collected during active flow emplacement (James et al., 2015, Farquarson et al., 2015;  
596 Kolzenburg et al., 2018a). Also in this case, the emplacement of lava flows is commonly  
597 modelled using a single rheological parameter (apparent viscosity or apparent yield strength)  
598 calculated from morphological dimensions using Jeffreys' and Hulme's (Jeffrey and Acrivos,  
599 1976; Hulme, 1974) equations. The rheological parameters are then typically further  
600 interpreted in terms of the nature and chemical composition of the lava (e.g., mafic or felsic).  
601 Chevrel et al. (2013, 2015) employing this methodology has shown that providing an unique  
602 factor to describe rheology of lava flows is definitely far from being representative of the real  
603 emplacement dynamics of lava flows. As above mentioned (point 2), given the nonlinear

604 dynamics of lava flows and domes, which may determine significant thermal effects,  
605 significant limitations may be observed and should be carefully considered before applying to  
606 any natural context (e.g. Costa and Macedonio, 2003, 2005; Costa et al., 2007b; Filippucci et  
607 al., 2013 and Filippucci et al., 2019, this issue).

608 4. Ties between the intensity of thermal anomalies generated by actively flowing lava and its  
609 silica content and therewith discharge rate of lavas (e.g. Coppola et al., 2013, 2017). This  
610 approach takes advantage of the fact that low viscosity lavas are readily able to spread into  
611 thin sheets during flow, whereas high viscosity lavas usually retain lower aspect ratios. Since  
612 the heat loss of a lava is largely governed by its surface to volume ratio, its spreading ability  
613 (i.e. viscosity) can, empirically, be correlated to the measured heat loss. Over the last decades  
614 such satellite-based remote sensing and data processing techniques have proved well suited to  
615 complement field observations and to allow timely eruption detection, as well as for flow  
616 tracking.

617

## 618 **7. Concluding remarks and outlook**

619 The present review shows the extraordinary improved knowledge of rheological properties  
620 of multicomponent and multiphase silicate melts occurring in the last twenty years. Such knowledge  
621 advancement has been due to the necessity of constraining natural processes and parallel the  
622 development of new technological advances, frequently obtained to face specific problems. It has  
623 been observed that the continuously evolving rheology of magmas and eruptive products during  
624 their ascent, eruption and emplacement can be described with increasing accuracy and specifically  
625 applied to geological issues with improved confidence. The observed transition between Newtonian  
626 to strongly non-Newtonian rheological behaviour is typical of both simple liquids and/or multiphase  
627 natural mixtures. These transitions govern the observed eruption dynamics and the eruption  
628 dynamic transitions, potentially determining also whether an eruption will be effusive or explosive.

629           The employment of the rheological flow laws for multicomponent and multiphase silicate  
630 melts find a very promising application to constraining the advancement and halting of lava flows.  
631 For these superficial phenomena, the opportunity of monitoring important variables such as the  
632 discharge rate and the topography of emplacement provide fundamental advantage for the  
633 employment of numerical simulations tools. These have allowed showing that more accurate  
634 estimates of the effects of crystals and bubbles during lava flow emplacement can be obtained only  
635 by real-time monitoring of lava flows through field and remote sensing methods paralleled by a  
636 proper experimental campaign, which in particular would account for the non-equilibrium, non-  
637 isothermal rheology of multiphase mixtures.

638           This progress in understanding the mechanisms of advancement and emplacement of lava  
639 flows and domes has also been made possible recently thanks to the recent emplacement of large  
640 long-lasting silicic to basaltic effusive eruptions. Prior to 2008, for instance, no rhyolite lava flow-  
641 forming eruptive event was observed or documented. Hence, the real-time observations of active  
642 rhyolitic flow and dome emplacement at the Chilean volcanoes of Chaitén (Carn et al. 2009; Lara  
643 2009; Bernstein et al. 2013; Pallister et al. 2013) and Puyehue-Cordón Caulle significantly  
644 developed our knowledge of rhyolitic lava emplacement (Castro et al. 2013; Schipper et al. 2013;  
645 Tuffen et al. 2013; Bertin et al. 2015; Farquharson et al. 2015; Magnall et al. 2017). Analogously  
646 the long lasting 2014-2015 basaltic eruption at Holuhraun, Bardarbunga system, Iceland (e.g.  
647 Pedersen et al., 2017), offered the opportunity to establish/calibrate, through the contemporaneous  
648 employment of field work, remote sensing techniques (Kolzenburg et al., 2018a) and laboratory  
649 experimentation (Kolzenburg et al., 2017), which allow retrieving thermal properties, estimates of  
650 effusion rate (Coppola et al., 2013, 2017) and evaluate the effect of bubbles by comparison with  
651 experimental campaign on liquid+ crystals material as collected during eruption (Kolzenburg et al.,  
652 2017, 2018a,b). Worth mentioning is also the integrated field, remote sensing, physical properties  
653 and physical modelling and numerical simulations studies performed in the recent years for

654 intermediate compositions producing effusive activities (Chevrel et al., 2013a,b; 2015). For  
655 andesitic domes huge progresses in understanding the non-linear thermal effects which determine  
656 non-linear eruption dynamics has been made by previous authors (e.g. Costa et al., 2007b; Melnik  
657 and Sparks, 1999, 2005; Melnik et al., 2009). Although the purposes of this paper is to mostly  
658 describe the development of rheological properties in relationship to the emplacement of lavas,  
659 most of the general results obtained here, and in particular those related to the effect of crystals and  
660 vesicle on multiphase rheology, can be extended to eruption dynamics of explosive phases.

661         Some of the main results deduced by application of the existing rheological models and  
662 experimental studies, supported by petrological analysis and field work, allowed to unequivocally  
663 show that lava flow emplacement may be a long lasting process also for silicic magmas and that  
664 flow may continued also unrooted from the vent for long times (e.g. Farquarson et al., 2015) and  
665 that extremely voluminous silicic lava flows may be emplaced in relatively short time without  
666 giving origin to significant explosive stages (Tuffen et al., 2013; Farquarson et al., 2015; Giordano  
667 et al., 2017; Polo et al., 2018a, b). In addition, Kolzenburg et al (2016, 2017, 2018) showed that  
668 disequilibrium, cooling- and shear-rate controlled rheological properties may have fundamental  
669 influence in determining the effective length of basaltic lava flows.

670         Although the results evidenced by performing non-equilibrium, non-isothermal, transient  
671 rheology of basaltic lava flows, are promising and provided a first understanding of lava flow  
672 rheology under natural conditions, it is possible to anticipate that future studies will require  
673 performing this kind of experiments also to a wider range of effusive products.

674

## 675         **Acknowledgements**

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680

681 **APPENDICES**

682 **Appendix A1. Short review of some of the most common methods to measure viscosity.**

683 The basic nature of magmas and volcanic products changes from liquids, through foams,  
684 emulsions, crystal suspensions, and partially molten aggregates. The rheology of such widely  
685 varying materials, each complex in its own right, and the fact that the viscosity of these materials  
686 can span over, at least, 16 orders of magnitude, defies the use of one single method to fully  
687 characterize their behavior. A large variety of devices for the measurement of the deformation and  
688 transport behavior of magmas, lavas and other volcanic materials have been presented in literature  
689 to date and this continuously evolving field produces novel techniques on a regular basis. These  
690 range from new devices for laboratory measurements to novel methods for ground- and satellite-  
691 based viscosity estimates. This field is growing in order to accomplish the complex task of defining  
692 the rheology of natural systems or analogues.

693 In principle, viscosity information of a material can be obtained from any data where time  
694 evolution of strain ( $\gamma = (dx/dz)$ ) and strain-rate ( $\dot{\gamma} = d\gamma/dt = 1/dt (dx/dz)$ ) in response to an  
695 applied stress ( $\sigma$ ) can be recorded.

696 Viscometry can therefore be performed in devices where either strain-rate is maintained  
697 constant while the resulting stress is measured (e.g., concentric cylinder) or when the imposed stress  
698 is kept constant and the resulting strain-rate is measured (e.g., micropenetration and fiber  
699 elongation; Dingwell et al., 1993). For the sake of brevity, we will not report the details and  
700 analytical procedure of each experimental device here but rather we will provide the most relevant  
701 reference papers describing the devices and the experimental procedures in detail.

702 Experimental efforts are to date largely subdivided into two categories of two phase  
703 suspension rheometry, namely i) those measuring volatile-bearing suspensions and, ii) those  
704 measuring particle bearing suspensions where volatiles cannot be retained dissolved in the liquid  
705 phase at experimental conditions. At room temperature conditions this distinction fundamentally



706 identifies two temperature intervals: i) low-temperature experiments, at or close to the glass  
707 transition temperature, where volatile dissolution kinetics and, in most cases, crystals formation  
708 processes are slow enough not to allow gas exsolution and crystal formation during the experiment  
709 (these span viscosities in the range between  $\sim 10^8$  to  $10^{12}$ ) and, ii) the high-temperature  
710 experiments, where pure liquid and liquid+crystals measurements can be performed. A small  
711 number of laboratory studies of viscous deformation of volatile and crystal-bearing mixtures at high  
712 pressure and temperature also exist. The main advantage of these apparatus is that they attain P-  
713 T conditions more realistic for geologically relevant multiphase assemblages and they allow  
714 measurements on volatile bearing liquids at superliquidus conditions.

715

#### 716 *41.1. Rotational (Couette) rheometry*

717 The advantage of the rotational concentric cylinder viscometry, compared to other  
718 viscometers, is the possibility to operate continuously at given conditions (e.g. shear rate or shear  
719 stress), so that steady-state measurements can be performed while systematically varying the  
720 experimental conditions. This allows detecting and quantifying any time dependency of the  
721 viscosity of the studied material via measurement at different shear rates, temperature, etc.  
722 Therefore, rotational viscometers are among the most widely used devices for measurements on  
723 magma rheology. The most common type are Searle-type viscometers, where the inner cylinder  
724 rotates (at constant strain-rate or stress). The sample is deformed in an annulus of liquid filling the  
725 gap between a rotating inner spindle (commonly a cylinder, or a cone-plate geometry) and an outer  
726 cylindrical cup. Rheometry on natural silicate melts is commonly performed in a wide gap  
727 geometry, where the velocity profile across the deforming liquid or suspension is non-linear. As  
728 such, determination of the sample viscosity relies on calibration of the measured torque against well  
729 characterized standard materials for which the temperature viscosity relationship is accurately  
730 known.

731 Viscosities are calculated from the equation for Newtonian liquids:

$$732 \quad \eta = \frac{M}{4\pi\Omega(L)} \left( \frac{1}{r^2} - \frac{1}{R^2} \right) \quad (\text{Eq. A1.1})$$

733 where  $M$  is the torque,  $\Omega$  is the angular velocity of the outer cylinder,  $r$  and  $R$  are the respective  
734 radii of the inner and outer cylinders, and  $(L)$  is the effective length of the inner cylinder.

735 As above mentioned, the common approach for natural silicate melt viscometry at high  
736 temperature is to use a wide gap geometry and calibrating the torque exerted on a spindle to the  
737 melt viscosity using a standard. As it can be retrieved by Eq. A1.1 wide gap geometry has the  
738 disadvantage of being slightly less accurate than small gap geometry, but the advantage of  
739 introducing small viscosity reading errors in the case of crystallization on the adopted rotating  
740 spindles or in the gap space.

741 Early devices of this type that produced a plethora of experimental data are described in a  
742 series of papers measuring the viscosity of synthetic silicate melts (Dingwell 1986; Dingwell 1989;  
743 Dingwell and Virgo 1988). For these high temperature rheological experiments a solid precious  
744 metal spindle is hung from the measurement head and immersed into the sample while being rotated  
745 at a constant rate. In this setup, the torque needed to maintain a constant rotation rate is proportional  
746 to the melt/suspension viscosity and is recorded at a frequency of  $\sim 1$  Hz. The spindles used in these  
747 experiments vary in diameter and are chosen depending on the expected melt viscosity, to suit the  
748 torque and deformation rate range of interest. They are commonly machined to have a  $45^\circ$  conical  
749 top and bottom to reduce edge effects. Calibration of these devices is performed for shear-rates and  
750 temperatures exceeding those used in the experiments to account for mechanical effects in the  
751 measurement setup. The precision of the viscosity determination is ca.  $\pm 3\%$  as described in  
752 Dingwell (1986). Since direct temperature measurement during viscometry was not possible up  
753 until recently, the thermal evolution of the sample at the imposed temperatures is commonly  
754 calibrated over the entire experimental temperature range using a platinum sheathed type-S

755 thermocouple immersed in an inert standard glass that does not crystallize over the entire calibration  
756 range.

757 The concentric cylinder is used to investigate both volatile-free liquids (at above liquidus  
758 conditions) and liquid+crystal suspensions (at subliquidus conditions) over the viscosity range  
759 between  $10^{-1}$  and  $10^4$  Pa s (Dingwell and Virgo, 1988; Chevrel et al. 2015; Chevrel et al. 2013a;  
760 Dingwell 1989; Dingwell and Virgo 1987; Giordano et al. 2005, 2006; Kolzenburg et al. 2016b;  
761 2017; Sato 2005; Vetere et al. 2017; Vona et al. 2011, 2013). It has also been employed to study the  
762 effect of changing oxygen fugacity on melt and suspension viscosity via combination with gas  
763 mixing furnaces (see for example Dingwell and Virgo,1987, Chevrel et al., 2013a, Kolzenburg et  
764 al., 2018b).

765 There are two dominant mechanical constraints of these experimental apparatuses that  
766 inhibit rheometry at high viscosity values presented here 1) the torque limit of the rheometer head  
767 and 2) the fact that the crucible containing the experimental sample may start to slip and rotate in its  
768 holder at high torque, rendering the measured torque data useless. This issue was addressed recently  
769 in the construction of a new rheometer by Morgavi et al. (2015), who designed a new crucible  
770 coupling, allowing both viscometry at higher torques, expanding the measurement range to  $10^4$  Pa s,  
771 and allowing also for chaotic mixing experiments.

772 A further advancement in concentric cylinder viscometry at sub liquidus conditions was the  
773 development of an experimental device and method for in-situ differential thermal analysis during  
774 concentric cylinder rheometry as presented in Kolzenburg et al. (2016a). This device allows  
775 tracking the crystallization relates change in the effective viscosity of the suspension due to  
776 crystallization as well as the latent heat released during crystallization. Such a device, in turn  
777 allowed studying the shear rate dependence of sub liquidus lava rheology at conditions pertinent to  
778 lava flow emplacement (Kolzenburg et al., 2018c,d).

779

## 780 *A1.2. Dilatometric methods*

781 These methods can be used to measure, at close to glass transition temperature (e.g.  
782 viscosity between  $10^8$  and  $10^{14}$  Pa s), pure liquid as well as two-phase and three-phase suspensions  
783 (Robert et al., 2008a,b; Vona et al., 2013). The most widely such a devices are the dilatometric  
784 method of the micropenetration (Hess and Dingwell, 1986) and the uniaxial parallel plate (e.g.  
785 Robert et al., 2008a, b) the details of which are provided below.

### 786 *Micropenetration*

787 Micropenetration technique involves determining the rate at which an hemispherical Ir-  
788 indenter moves into the melt surface under a fixed load. The sample is placed in a silica rod sample  
789 holder under an Argon gas flow. The indenter is attached to one end of an alumina rod. The other  
790 end of the alumina rod is attached to a mass. The metal connection between the alumina rod and the  
791 weight pan acts as the core of a calibrated linear voltage displacement transducer (LVDT)(e.g. Hess  
792 and Dingwell, 1996). The movement of this metal core as the indenter is pushed into the melt yields  
793 the displacement. The absolute shear viscosity is determined via the following equation  
794 (Pocklington, 1940; Tobolsky and Taylor, 1963):

$$795 \quad \eta \text{ (Pa} \cdot \text{s)} = \frac{0.1875 \cdot P \cdot t}{r^{0.5} \cdot \alpha^{1.5}} \quad \text{(Eq. A1.2)}$$

796 where  $P$  is the applied force,  $r$  is the radius of the hemisphere,  $t$  is the penetration time and  $\alpha$   
797 is the indentation distance. This provides an accurate viscosity value if the indentation distance is  
798 lower than 150 – 200 microns. The technique allows viscosity to be determined at T up to 1100°C  
799 in the range  $10^{8.5}$  to  $10^{12}$  Pa·s without any problems with vesiculation. One advantage of the  
800 micropenetration technique is that it only requires a small amounts of sample (other techniques used  
801 for high viscosity measurements, such as parallel plates and fiber elongation methods instead  
802 necessitate larger amount of material).

803 *Parallel plate*

804 Using the parallel plate technique, the shear viscosity of the cores is computed for a given  
805 applied load ( $F$ ;  $N$ ), sample volume ( $V$ ;  $m^3$ ), sample length at time  $t$  ( $L$ ;  $m$ ), and rate of shortening  
806 ( $\partial L/\partial t$ ;  $m/s$ ) using the “no-slip” (Eq. A1.2), and “perfect-slip” (Eq. A1.3) models of Gent (1960) (cf.  
807 Dingwell et al. 1993):

$$808 \quad \eta_S (Pa \cdot s) = \frac{2\pi \cdot L^5 F}{3V \frac{dL}{dt} (2\pi L^3 + V)} \quad (\text{Eq. A1.3})$$

809 and

$$810 \quad \eta_S (Pa \cdot s) = \frac{L^2 F}{3V \frac{dL}{dt}} \quad (\text{Eq. A1.4})$$

811 respectively. The “no slip” equation is used for the case in which the surface area of contact  
812 between the melt and the parallel plates remains constant and the cylinder bulges with increasing  
813 deformation. The “perfect slip” equation is used for the case in which the surface area between the  
814 cylinder and the plate increases with deformation and the cylinder does not bulge.

815

### 816 ***A1.3. Falling Sphere method (Simple and centrifuge)***

817 The falling sphere method relies on determining the speed of a sphere (typically made of  
818 metal) falling through molten material. Viscosity is calculated according to the Stokes law as it  
819 follows:

$$820 \quad \eta (Pa \cdot s) = \frac{2}{9} \cdot \frac{(\rho_S - \rho_M) \cdot a \cdot r^2}{v} \cdot C_F \quad (\text{Eq. A1.5})$$

821 where  $\eta$  is the viscosity ( $Pa \cdot s$ ),  $\rho_S - \rho_M$  the density contrast between the sinking sphere and  
822 the melt ( $kg/m^3$ ),  $a$  the acceleration of typically  $1g$  ( $m/s^2$ ),  $r$  the radius of the sphere ( $m$ ), and  $v$  the  
823 velocity of the sinking sphere ( $m/s$ ).  $C_F$  is the Faxè correction term (Faxè, 1922), which accounts  
824 for interactions between the sinking sphere and the wall of the capsule expressed by

825 
$$C_F = 1 - 2.104 \cdot \left(\frac{r}{R}\right) + 2.09 \cdot \left(\frac{r}{R}\right)^3 - 0.95 \cdot \left(\frac{r}{R}\right)^5 \quad (\text{Eq. A1.6})$$

826 where R is the radius of the container, in which the sphere is positioned in the center. The  
827 Fax n correction is considered to result in a slight underestimation of the calculated viscosity, but  
828 constitutes the best expression to account for the wall-effect on viscosity (Kahle et al., 2003).

829 Maximizing the density contrast ( $\rho_S - \rho_M$ ) between the sphere and the melt reduces the  
830 uncertainty associated with the estimation of melt density. As a consequence Pt ( $\rho_S=21450 \text{ kg/m}^3$ )  
831 or Mo spheres ( $\rho_S=10220 \text{ kg/m}^3$ ) are commonly employed.

832 Experiments that allow to investigate the variation of viscosity at the P-T-X(H<sub>2</sub>O)  
833 conditions relevant to magmatic and volcanological environments are incredibly time consuming  
834 and require significant experimental effort. The range of viscosities investigated with the falling  
835 sphere technique varies from  $10^{-4} \text{ Pa s}$  (Brearley et al., 1986; Behrens and Schulze, 2003; Audetat  
836 and Keppler, 2004; Kanzaki et al., 1987; Taniguchi, 1995; Suzuki et al., 2005) to  $10^{4.5} \text{ Pa s}$   
837 (Kushiro et al., 1976; Kushiro, 1976, 1977, 1978; Baker and Vaillancourt, 1995; Schulze et al.,  
838 1999; Liebske et al., 2005; Vetere et al., 2006; Del Gaudio et al., 2007). At the lower viscosity end  
839 ( $<1 \text{ Pa s}$ ), represented by ultramafic melts, real time movies of the falling sphere are necessary and  
840 are obtained through the use of X-rays from a synchrotron source. At the upper viscosity end ( $10^{4.5}$   
841  $\text{Pa s}$ ), represented by granitic melts, the sinking distance of the sphere becomes smaller than its  
842 diameter and the sum of the experimental errors in determining the sphere location. Falling sphere  
843 viscometry has been employed at ambient (Dorfman et al., 1997; Riebling, 1966) and high-P  
844 conditions (e.g. Kushiro, 1978; Ryan and Blevins, 1987; Persikov et al., 1990; Scarfe et al., 1986;  
845 Dingwell, 1987; Brearley and Montana, 1989; and White and Montana, 1990; Baker and  
846 Vaillancourt, 1995; Schulze et al., 1999; Liebske et al., 2005; Vetere et al., 2006; Del Gaudio et al.,  
847 2007) within piston cylinders and multi anvils (up to 130 kbar, Liebske et al., 2005) for  
848 simultaneous determination of density and viscosity. Maximizing the density contrast between the

849 falling sphere and the surrounding melt reduces errors associated with the estimation of melt density  
850 and viscosity. Errors in density contrast can easily be reduced below the uncertainties inherent in  
851 the other variables affecting viscosity determination. Total errors for falling sphere viscosity  
852 determinations using the piston cylinder apparatus are probably nearer 20%. Burnham (1963)  
853 describes a variant on the falling sphere method in which the fall of a sphere, connected to a wire  
854 isolated electrically from the metal capsule, was electrically detected by the contact of the sphere  
855 against the capsule wall. Persikov et al. (1990) used a radioactive-tracer doped falling sphere in an  
856 internally-heated pressure vessel. The descent of the sphere is recorded radiographically as the  
857 sphere transits two "windows" in a lead shield. Very high pressure measurements of viscosity have  
858 been made by Kanzaki et al. (1987) who imaged the falling sphere in real time using a synchrotron  
859 radiation source. This method extends the lower limit of measurable viscosity using the falling  
860 sphere method at high pressure to  $10^{-3}$  Pa s.

861         The highest viscosity that can be investigated by the falling sphere method may be expanded  
862 by the use of a centrifuge apparatus (Dorfman et al., 1996; Dorfman et al., 1997; Bagdassarov and  
863 Dorfman, 1998; Schmidt et al., 2006; Ardia et al., 2008). With viscosity and acceleration scaling  
864 linearly, a centrifugal force of 1000g expands the viscosity range to  $10^7$  Pa s. The elevated  
865 acceleration (up to 1000g) also allows investigating viscosities between the solidus and the glass  
866 transition temperature ( $T_g$ ). This kind of measurements are commonly performed on pure liquids.  
867 We are aware of only one published paper (Vetere et al. 2010) which investigated multiphase  
868 rheology of andesitic lavas under pressure conditions. In a few cases these experiments were run at  
869 acceleration of 1000g by means of centrifuge falling-sphere methods (e.g. Ardia et al., 2008).

870

#### 871 *A1.4. Paterson type apparatus*

872         The gold standard for quantitative high pressure deformation studies is the gas apparatus  
873 developed by Paterson (1970) and Paterson and Olgaard (2000). The apparatus can either compress

874 or extend the sample. Some gas apparatus, for example, the Paterson gas apparatus (Paterson,  
875 1970), can also deform the sample in torsion. Torsion allows much higher strains to be reached.  
876 Because the amount of strain in the sample varies radically from the centre to the edge in torsion  
877 experiments, torsion samples are constructed in the shape of a thin ring with jacket materials filling  
878 the centre. What gives the gas apparatus its edge is that the gas confining medium (usually Ar)  
879 provides a perfect uniaxial stress field. Because the gas supports no shear tractions along the sides  
880 of the sample or pistons, an external load cell can accurately measure the load supported by the  
881 sample. Unfortunately, the gas apparatus can only achieve a confining pressure of 500 MPa (5  
882 kbar). This pressure is equivalent to the pressure at 15 km depth in the earth. The limited pressure  
883 range is a severe limitation for students of the deep earth. Online resources: an Introduction to High  
884 Pressure Rock Deformation Techniques can be found at:

885 [https://serc.carleton.edu/NAGTWorkshops/mineralogy/mineral\\_physics/deformation\\_mechanisms.html](https://serc.carleton.edu/NAGTWorkshops/mineralogy/mineral_physics/deformation_mechanisms.html).

886

### 887 ***A1.5. Direct viscosity measurements on active flows in the field***

888 Few direct measurements of lava rheology under natural conditions have been reported  
889 (Pinkerton and Sparks, 1978; Shaw et al., 1968; Belousov and Belousova, 2018; Chevrel et al.  
890 2018). Nonetheless, some of these methods are based on the assumption of a parabolic velocity  
891 profile (Jeffrey's eq) and have the limitations highlighted at paragraph 3.2.1. These measurements  
892 are crucial for benchmarking of experimental data, but insufficient to develop a systematic  
893 understanding of the evolution of lava-flow properties in response to varying external and internal  
894 parameters (composition, cooling-and shear-rate, oxygen fugacity etc.), as they represent snapshots  
895 of the system at one specific condition (Belousov and Belousova, 2018). Belousov and Belousova,  
896 (2018) and Chevrel et al (2018) present the most recent technological advances of this unique  
897 technique and new field measurement results. Such direct measurements in the field require  
898 inserting a custom-built penetrometer, constituted by a steel bar with a half-spherical penetration



899 head (e.g. Belousov and Belousova, 2018), or a viscometer (Chevrel et al., 2018), into the molten  
 900 lava while it is flowing. Such in-situ viscometry is challenging due to the difficulty of accessing an  
 901 active lava flow, and the lack of appropriate instrumentation. Measurements under such conditions  
 902 are therefore very restricted and have mostly been performed on slow advancing lava flows and far  
 903 from the solidification point of the lava.

904 1) During penetration viscometry (similar to that used to perform micropenetration experiment in  
 905 the lab), where a rod is pushed into the liquid lava, the force required to penetrate the lava is kept  
 906 constant and penetration is monitored with time until penetration ceases. Viscosity of lava is then  
 907 calculated based on Stoke's law for half a sphere (i.e. the half spherical penetrating tip of the  
 908 penetrating rod). Belousov and Belousova (2018) employed the relationship proposed by Panov  
 909 (1988), according to which:

$$910 \quad \eta_S (Pa \cdot s) = \frac{F}{3\pi v R} \quad (A1.7)$$

911 where  $F$  is the force (N),  $v$  is the speed of penetration (m/s) and  $R$  is the radius of the penetration  
 912 head (m).

913 2) The rotational viscometry method, requires a shear vane to be inserted into the molten  
 914 lava and the shear stress values corresponding to various applied rotation rates is measured.

915 Viscosity is estimated as the shear stress over the strain-rate ratio ( $\eta = \frac{\tau}{\dot{\gamma}}$ ). The shear stress ( $\tau$ ) is

916 calculated by:  $\tau = \frac{M}{2\pi h R_i^2}$  where  $M$  is the torque recorded by the torque sensor,  $h$  is the length of the

917 vane and  $R_i$  is the equivalent radius of the rotating vane assuming a wide gap concentric cylinder

918 geometry. The strain-rate ( $\dot{\gamma}$ ) is calculated according to the following formula:  $\dot{\gamma} = \frac{2\omega}{n \left[ 1 - \left( \frac{R_i}{R_o} \right)^{\frac{2}{n}} \right]}$

919 where  $\omega$  is the rotational velocity (rad/s) and  $R_i$  and  $R_o$  are the inner and the outer radius of the

920 spindle.  $n$  is the flow index which is determined by performing measurements at various rotational

921 speed. The advantage of this approach is that it allows for evaluation of the lavas effective viscosity

922 at a range of shear rates. Viscosity estimates obtained by employing this procedure are reported in  
923 Chevrel et al. (2018).

924 **Appendix A2. Empirical and theoretical based formulations to express the T- and P-**  
925 **dependence of the viscosity of silicate liquids.**

926 Among the known theoretical relations for coefficient of viscous flow of liquids, the Eyring  
927 arrhenian equation (Glasstone et al., 1941) is the most popular:

928 
$$\eta = \eta_0 \exp(\Delta G/RT)$$
 (Eq. A2.1);

929 where R is the gas constant, T the absolute temperature and  $\Delta G$  is the free activation energy  
930 of yield of the flow, that is, a function of the internal energy, entropy, P, and V (i.e. the volume of  
931 particles). The pre-exponential factor  $\eta_0$ , is related to the jump frequency of an atom from one site to  
932 another and represent the microscopic view of the “viscosity”. It can be expressed as  $\eta_0 = hN_A / V_m$   
933 where  $h$  is the Planck's constant, ( $6.63 \cdot 10^{-34}$  J s) and  $N_A$  the Avogadro's number ( $6.02 \cdot 10^{27}$ ) and  $V_m$   
934 is the molar volume of the investigated liquid at very high temperature. This equation can be  
935 rewritten in logarithmic scale as it follows:

936 
$$\log \eta = A + B/T$$
 (Eq. A2.2);

937 where  $A=2.303 \cdot \log \eta_0$  and  $B=\Delta G/R$ . This Arrhenian form for viscosity has been used by  
938 Shaw (1972) and Bottinga and Weill (1972) to describe the first description of the viscosity of  
939 natural silicate melts.

940 Expansion of the melt viscometry database over a wider range of melt compositions and  
941 temperatures (closer to the so-called calorimetric glass transition temperature,  $T_g^{cal}$ , Angell, 1991;  
942 Scherer, 1984; Giordano et al., 2008a) exposed the limitations of Arrhenian models and new  
943 empirical and theoretical-based models have been provided to describe the temperature dependence  
944 of the viscosity of silicate liquids. In particular, the T-dependence of viscosity is accounted for by  
945 the three parameters in each of the most commonly employed formulation: (i) Vogel–Fulcher–  
946 Tamman (VFT)(Vogel, 1921; Fulcher, 1925; Tamman and Hesse, 1926); (ii) Adam–Gibbs (AG)  
947 (Adam and Gibbs, 1965) and (iii) Avramov (AV) (Avramov, 1998). These formulations  
948 accommodating the non-Arrhenian T -dependence of silicate melts, can be written as following:

949 Vogel–Fulcher–Tammann (VFT) :  $\log \eta = A_{VFT} + B_{VFT}/(T - C_{VFT});$  (Eq. A2.3);  
 950 Adam and Gibbs (AG):  $\log \eta = A_{AG} + B_{AG}/[T S^{\text{conf}}(T,x)];$  (Eq. A2.4);  
 951 Avramov (AV):  $\log \eta = A_{AV} + (B_{AV}/T)^{CAV};$  (Eq. A2.5);

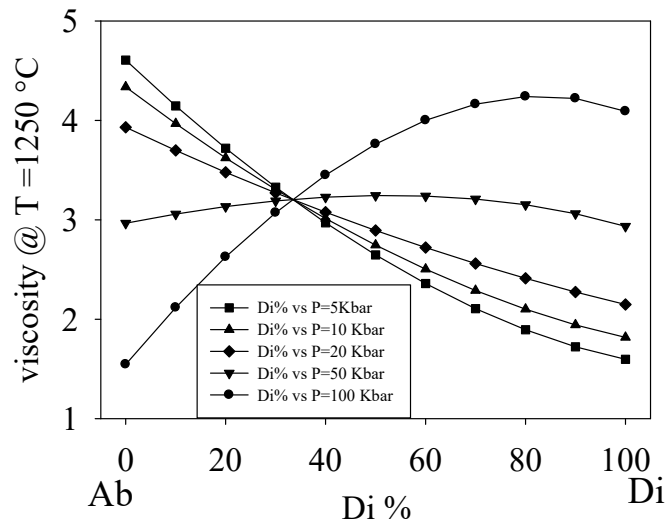
952

953 where  $\eta$  is the viscosity in Pa s,  $T$  is the absolute temperature, and  $A$ ,  $B$ ,  $C$ ,  $D$  and  $S^{\text{conf}}(T,$   
 954  $x)$ (the so-called configurational entropy; representing the number of configurations accessible to the  
 955 liquids at the glass transition ( $T_g$ ), Richet, 1984) are adjustable parameters (e.g. Giordano and  
 956 Russell, 2007; Giordano et al., 2008a, b; Russell and Giordano, 2017). The literature shows that, in  
 957 these systems, viscosity converges to a common value of the pre-exponential factors ( $A$ ) that can be  
 958 assumed, at fixed pressure, to be independent of composition (e.g. Russell et al., 2003; Russell and  
 959 Giordano, 2005;; Giordano and Russell, 2007; Giordano et al., 2008a). The other adjustable  
 960 parameters in each equation are expanded to capture the effect of composition (Hui and Zhang,  
 961 2007; Giordano et al., 2008a). Several other formulations exist that can be reconciled with the three  
 962 model equation above mentioned. For instance, the most recent formulation of Mauro et al. (2009),  
 963 which is based on the AG theory, assumes that  $S^{\text{conf}}(T)$  is closely connected with the topological  
 964 degrees of freedom of atoms which provide an increased number of adjustable parameters. The  
 965 most recent GRD and HZ models used to describe the viscosity of anhydrous and volatile bearing  
 966 melts where  $B$ ,  $C$  and  $S^{\text{conf}}$  parameters accommodate the effect of composition.

967

968 Empirical formulations used to constrain the effect of pressure on the viscosity of liquids  
 969 and characteristic parameters of Equations (Eqs. A2.1-A2.5) are provided by various authors (e.g.  
 970 Liebske et al., 2003; Ardia et al., 2008; Hui et al., 2009). In particular, Liebske et al (2003) by  
 971 modelling the viscosity of andesitic magmas and Ardia et al. (2008) modelling the viscosity of more  
 972 silicic magmas (rhyolites), used VFT expression (Eq. A2.3) where  $B_{VFT}$  parameter was function of  
 973  $H_2O$  and cubic and a linear function of  $P$ , respectively, and  $C_{VFT}$  parameter was only function of  
 974  $H_2O$  content and independent of  $P$ . Fig. A2.1 reports, as an example, the variation of viscosity at

975 constant T (1250 °C) as a function of P for melts of the Ab - Di system as fitted by using a linear  
 976 dependence from P of the  $B_{VFT}$  parameter.



977  
 978 *Fig. A2.1. Isothermal viscosity variation as a function P (up to 100 kbar) for melts of the Ab-Di systems. The*  
 979 *viscosity was calculated here using VFT equation (Eq. A2.3) where the  $B_{VFT} = B_0, VFT + b_1 * P(\text{Kbar})$ , where  $B_0$ ,  $VFT$*   
 980 *is the pseudo-activation energy at 1 bar pressure and  $b_1$  is an adjustable parameter accounting for a linear P-*  
 981 *dependence of  $B_{VFT}$ . The effect of composition is shown in the figure. At  $T=1250$  °C the pure liquid viscosity of the*  
 982 *albitic term (Ab100, left), proxy for a polymerized melt, is decreasing as P increases. On the other hand the pure liquid*  
 983 *viscosity for the diopsidic melt (Di100, right), proxy for more depolymerized compositions, increases as P increases.*  
 984

985 Online resources: a detailed summary of the philosophical approach employed to the construction  
 986 of the GRD model, predictable variables and downloadable versions of the viscosity calculator can  
 987 be found online at: <https://www.eoas.ubc.ca/~krussell/VISCOSITY/grdViscosity.html>

988 **Appendix A3. Empirical formulations to express the effect of varying crystal/vesicle**  
989 **content, size and shape distributions and strain-rate on the viscosity of multiphase**  
990 **mixtures.**

991 Viscosity is defined as the resistance to flow under specific applied stress ( $\sigma$ ) conditions and  
992 it is expressed by complex functions of applied stress and resulting strain ( $\gamma$ ) and strain-rates (e.g.,  
993 Herschel and Bulkley, 1926). For a Newtonian liquid,  $\sigma = \eta\dot{\gamma}$  where  $\eta$  is the Newtonian viscosity. It  
994 is well-known from both laboratory studies on analogue materials (e.g. Mueller et al., 2011;  
995 Cimarelli et al, 2011; Truby et al., 2015) and natural remelted volcanic rock (e.g. Caricchi et al.,  
996 2008; Vona et al., 2011; Campagnola et al., 2016; Kolzenburg et al., 2016; Soldati et al., 2016,  
997 2017) that suspended solids and natural crystals lead to the increase of the suspension's bulk  
998 viscosity of up to several orders of magnitude compared to the solid particles free counterpart. On  
999 the other hand the role of an addition of bubbles on a suspension may lead to both a viscosity  
1000 increase (spherical bubbles) or decrease (oblate deformed bubbles)(e.g. Llewellyn et al., 2002). The  
1001 presence of a solid phase or a gaseous phase to form a solid or bubble suspension commonly yield  
1002 non-Newtonian behavior, expressed in the more general equation:

$$1003 \quad \sigma = \sigma_0 + K\dot{\gamma}^n \quad (\text{Eq. A3.1})$$

1004 where  $\sigma_0$  is a stress threshold (or yield stress) to be overcome in order to start flow;  $K$  is the  
1005 flow consistency (which corresponds to shear viscosity at  $\dot{\gamma} = 1 \text{ s}^{-1}$ ) and  $n$  is the flow index which  
1006 describes the degree of non-Newtonian behavior, being equal to 1 for Newtonian fluids,  $n > 1$  for  
1007 shear-thickening and  $n < 1$  for shear-thinning fluids. For non-Newtonian fluid ( $\sigma_0 = 0$ ), the term  $\sigma /$   
1008  $\dot{\gamma}$  is equal to  $K\dot{\gamma}^{n-1}$  and an apparent viscosity will be defined as  $\eta_{app} = \sigma / \dot{\gamma}$ , while the relative  
1009 viscosity ( $\eta_r$ )(ratio between stress and strain rate divided by the viscosity of the suspending liquid,  
1010  $\eta_l$ ) will be expressed by:

$$1011 \quad \eta_r = \left(\frac{K}{\eta_l}\right) \dot{\gamma}^{n-1} \quad (\text{Eq. A3.2})$$

1012 where  $K_r = K/\eta_l$  represents the relative consistency.

### 1013 ***A3.1. Bubble-melt suspensions***

1014 When a bubble suspension flows bubble deformation is promoted, through the viscous  
1015 forces, by shear (tending to deform bubbles) and opposed by surface tension which tends to  
1016 restore/maintain bubble sphericity. The adimensional capillary number ( $Ca$ ), i.e. the measure of the  
1017 relative importance (i.e. the ratio) of shear and interfacial stresses, is:  $Ca = \frac{\eta_{melt} \cdot r \cdot \dot{\gamma}}{\Gamma}$ , where  $\eta_{melt}$ ,  $r$ ,  
1018  $\dot{\gamma}$  and  $\Gamma$  are the viscosity of the suspension, the radius of the undeformed bubble, the shear  
1019 rate of the flow and the liquid-vapour surface tension, respectively. The presence of bubbles  
1020 can either increase or decrease the viscosity of a suspension depending on the dynamic regime  
1021 (Manga et al., 1998; Lejeunne et al., 1999; Llewellyn et al., 2002a,b; Rust and Manga, 2002; Stein  
1022 and Spera, 2002; Llewellyn and Manga, 2005). Similar to solid particles, bubbles deform flow lines  
1023 within the suspending medium, which tends to increase the viscosity. However, at the same time,  
1024 they provide free-slip surfaces which favour flow. For  $Ca < 1$ , interfacial tension forces dominate  
1025 and bubbles are approximately spherical (e.g., Taylor, 1932). In this case flow-line distortion is  
1026 great and free-slip surface area is small, hence the overall effect is to increase the suspension  
1027 viscosity. In contrast, at high deformation regimes, the bubbles will undergo significant elongation  
1028 ( $Ca > 1$ ), thus favouring small flow-line distortion, greater free-slip surface area and a decrease in  
1029 the suspension viscosity (Hinch and Acrivos, 1980, Llewellyn and Manga, 2005; Vona et al., 2013,  
1030 2017).

1031 However, the capillary number  $Ca$  implies an equilibrium between viscous and interfacial  
1032 forces, and it can be applied only for steady flows, in which the conditions of shear have remained  
1033 constant for a long enough time. As a consequence, if the shear strain rate is changing, the flow is  
1034 unsteady and to describe unsteadiness Llewellyn et al. (2002a, b) introduced the dynamic capillary  
1035 number  $Cd = \frac{\eta^0 \cdot r \cdot \ddot{\gamma}}{\tau \dot{\gamma}}$ , where the double derivative of strain-rate is the speed of strain-rate variation.  
1036 For  $Cd \ll 1$ , the changes in shear environment are slow enough to allow the bubbles to reach their  
1037 equilibrium shape, hence flow is steady and the dynamic regime is controlled by the capillary

1038 number  $Ca$ . On the other hand, if  $Cd \gg 1$ , flow is unsteady, the bubbles are not able to reach their  
 1039 equilibrium shape in response to fast strain rate and they are, therefore, unrelaxed (they are actively  
 1040 deforming). In other words, in these conditions the rate of bubble deformation is large compared  
 1041 with the bulk strain rate and, therefore, most of the strain is accommodated by deformation of the  
 1042 gas in the bubbles. Since the gaseous phase has a negligible viscosity, this leads to a decrease of  
 1043 viscosity as the bubble content increases. Llewellyn and Manga (2005) parameterized the effect of  
 1044 bubbles on the relative viscosity of a bubbly suspension, considering a single equation for the  
 1045 positive dependence of  $\eta_r$  on  $\phi_b$  and a single equation for the negative dependence of  $\eta_r$  on  $\phi_b$ ,  
 1046 regardless of whether the decrease in viscosity in the latter case is related to steady ( $Ca > 1$ ) or  
 1047 unsteady ( $Cd > 1$ ) flow. Based on existing literature models (Bagdassarov and Dingwell, 1992, 1993;  
 1048 Pal 2003, Llewellyn et al., 2002a,b), the authors suggested two different parameterizations for each  
 1049 viscous regime (increasing and decreasing  $\eta_r$ ), considering two limiting cases corresponding to a  
 1050 minimum (MIN) and a maximum (MAX) effect of the bubbles on the viscosity of the suspensions.

1051 For  $Ca < 1$  (increasing  $\eta_r$ ):

$$1052 \quad \eta_r = (1 - \phi_b)^{-1} \text{ (MIN: Pal 2003); } \eta_r = (1 + 9\phi_b) \text{ (MAX: Llewellyn, 2002a,b)} \quad (\text{Eq. A3.13})$$

1053 For  $Ca > 1$  or  $Cd > 1$  (decreasing  $\eta_r$ ):

$$1054 \quad \eta_r = (1 - \phi_b)^{5/3} \text{ (MIN: Pal 2003); } \eta_r = [1 / (1 + 22.4\phi_b)] \text{ (MAX: Bagdassarov \& Dingwell, 1992) (Eq.}$$

1055 A3.14)

1056 Online resources: a synthesis of the results obtained by Llewellyn research can be found at  
 1057 the following webpage: <http://community.dur.ac.uk/ed.llewellyn/rheology.htm>

1058 Quane and Russell (2005), Russell and Quane (2005) and Robert et al (2008a, b) by  
 1059 performing uniaxial compression experiments on porous samples modelled the effective evolving  
 1060 viscosity ( $\eta_e$ ) and porosity ( $\phi$ ) as from Russell and Quane (2005):



1061  $\log \eta_e = \log \eta_0 - [\alpha\phi/(1-\phi)]^\beta$  (Eq. A3.15)

1062 where  $\alpha$ ,  $\beta$  are unknown adjustable coefficients and  $\eta_0$  is the unknown effective viscosity of the  
1063 melt plus crystal cargo at zero porosity. The parameter  $\beta$  was added to the original model of  
1064 Ducamp and Raj (1989) to capture the full range of data and preserve a concave down equation  
1065 form in the variables space ( $\eta$ ,  $\phi$ ). The employed model was used to estimate the timescale for the  
1066 welding of the block and ash flow deposits at Mount Meager. The results of the specific  
1067 measurements of Quane and Russell (2005), Russell and Quane (2005), Robert et al (2008a, b) and  
1068 new data on natural samples were compared by Vona et al. (2016) with the modelling proposed by  
1069 Mader et al (2013), based on the analysis of analogue materials. Those results are in better  
1070 agreement with the model proposed by Bagdassarov and Dingwell (1992). Finally, Vona et al.  
1071 (2016) proposed a model to evaluate the strength of vesiculated magmas based on Eq. A3.15.

1072

### 1073 ***A3.2. Crystal-melt suspensions***

1074 In the last decades a significant number of experimental studies have investigated the  
1075 rheological properties of different solid-bearing suspensions constituted by synthetic analogues  
1076 (Mueller et al., 2011; Cimarelli et al., 2011; Moitra and Gonnermann, 2015; Truby et al., 2015;  
1077 Klein et al., 2017, 2018; Dobson et al., 2015, 2016); synthetic silicate melts (Lejeune and Richet,  
1078 1995; Caricchi et al., 2007; Champallier et al., 2008 and Costa et al., 2007a, 2009) and crystal-  
1079 bearing natural magmas at subliquidus temperatures (e.g., Gay et al., 1969; Shaw, 1969; Marsh,  
1080 1981; Ryerson et al., 1988; Pinkerton and Stevenson, 1992; Pinkerton and Norton, 1995; Sato,  
1081 2005; Ishibashi and Sato, 2007; Caricchi et al., , 2008; Ishibashi, 2009; Vona et al., 2011, 2017;  
1082 Campagnola et al., 2016; Kolzenburg et al., 2016, 2017, 2018).

1083 In crystal-melt suspensions, the dispersed phase acts as a ‘hard’ (non-deformable) inclusion  
1084 which increases the viscosity of the suspension through both hydrodynamic and mechanical  
1085 interaction among crystals. For low solid fractions, the viscosity increases slowly with the particle

1086 volume fraction ( $\phi$ ), and the suspension maintains a Newtonian rheological behavior (strain-rate  
1087 independent). When  $\phi$  exceeds a critical value ( $\phi_c$ ), particles start to interact with each other and a  
1088 solid network of particles begins to form, causing a strong increase in viscosity and the onset of  
1089 non-Newtonian flow, characterized by Bingham-like rheology and/or shear thinning effects (Eq.  
1090 (1)). As the solid fraction is further increased, the system reaches another rheological threshold,  
1091 corresponding to the maximum packing density of solid particles ( $\phi_m$ ) Fig. 2), which causes the  
1092 transition from melt and melt+crystal to solid-state creep rheology (e.g., Kohlstedt and Zimmerman,  
1093 1996; Lavallée et al., 2007, 2008, 2012). The value of  $\phi_c$  is defined by both the crystal content as  
1094 well as by the crystal size and crystal shape distributiosn (e.g. Costa et al., 2009; Cimarrelli et al.,  
1095 2011; Vona et al., 2011). The increase in viscosity and the non-Newtonian flow depend on textural  
1096 features (crystal and bubble distribution) and deformation regimes (e.g., Costa et al., 2009; Petford,  
1097 2009; Mueller et al., 2011).

1098 A number of models have been proposed describing the effect, on the suspensions rheology,  
1099 of the crystal volume fraction and their,shape and size distributions as well as particles roughness  
1100 and deformability (e.g., Einstein, 1906; Roscoe (1952); Maron and Pierce, 1956; Krieger and  
1101 Dougherty, 1959; Frankel and Acrivos, 1970; Jeffrey and Acrivos, 1976; Marsh, 1981; McBirney  
1102 and Murase, 1984; Costa, 2005; Hsueh and Becher, 2005; Stickel and Powell, 2005; Caricchi et al.,  
1103 2007; Champallier et al., 2008; Costa et al., 2009; Ishibashi, 2009; Mueller et al., 2011; Cimarrelli et  
1104 al., 2011; Vona et al., 2011, Mader et al., 2013; Moitra and Gonnermann; 2015; Klein et al., 2017,  
1105 2018). The various models can be applied to represent the viscosity variation from a dilute ( $\phi < 0.03$ ,  
1106 where viscosity increase is linear with crystal content and it is Newtonian) to a semi-dilute ( $0.25 < \phi < 0.40 < \phi_c$ ,  
1107 where viscosity shows an increasing dependence of crystal fraction, but it is still  
1108 Newtonian) until a concentrated regimes ( $\phi_c < \phi < \phi_m$ , for which viscosity shows a rapid increase and  
1109 the onset of non-Newtonian rheology). For  $\phi > \phi_m$  a solid creep-dominated rheology is observed  
1110 until the occurrence of brittle failure. The crystal fraction  $\phi$  at the transition between one and the

1111 other regime depends strongly from the shape and size distributions (e.g. Saar et al., 2001; Costa et  
1112 al., 2009).

1113 So far, one of the most comprehensive models describing the relative viscosity variation  
1114 from dilute to highly concentrated regimes is that proposed by Costa et al. (2009)(CM). According  
1115 to that model:

$$1116 \quad \eta r(\phi) = \frac{1+\phi^\delta}{[1-F(\phi,\xi,\gamma)]^B \phi^*} \quad (\text{Eq. A3.3})$$

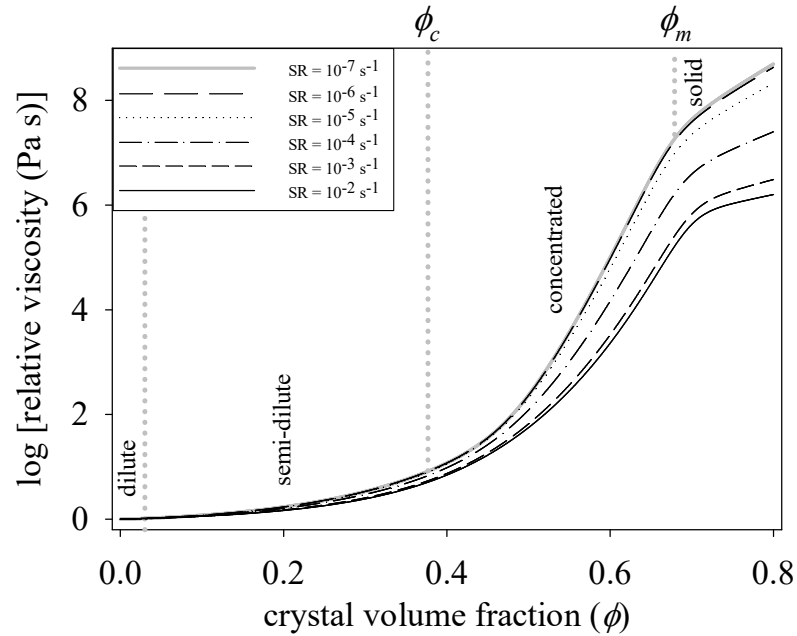
1117 where

$$1118 \quad F = (1 - \xi) \operatorname{erf} \left[ \frac{\sqrt{\pi}}{2(1-\xi)} \phi(1 + \phi^\gamma) \right] \quad (\text{Eq. A3.4})$$

1119 with  $\phi = \phi/\phi^*$  where  $\phi^*$ ,  $\xi$ ,  $\gamma$  and  $\delta$  are adjustable parameters that depend on the  
1120 deformation rate (Costa et al., 2009) and  $B$  is the Einstein coefficient (Einstein, 1906). The  
1121 parameter  $\phi^*$  represents the critical solid fraction at which the rheological transition from a  
1122 dominant liquid phase regime switches to a regime where the effect of crystals is predominant and  
1123 the viscosity values are much higher [Costa, 2005; Costa et al., 2009]. The CM is calibrated using  
1124 suspensions containing crystal fractions, isotropically distributed, in the interval  $\phi = 0.1 - 0.8$ .

1125 Figure A2.1 shows, based on the Costa et al. (2009) model (CM), the strain-rate dependence  
1126 of the relative viscosity at varying crystal volume fractions. The relative viscosity of two-phase  
1127 mixture increases following a sigmoid curve with exponential increase above a critical solid  
1128 fraction ( $\phi_c$ ), corresponding to the first inflection point in Fig. 2. This documents how variations of  
1129 crystal content may cause orders of magnitude changes in suspension viscosity. A further important  
1130 point in models of suspension rheology is that the maximum critical crystal volume fraction ( $\phi_m$ )  
1131 depends strongly on crystal shape, size distribution, crystal surface roughness and crystal  
1132 orientation (e.g. Mueller et al., 2011; Mader et al., 2013; Klein et al., 2017; 2018). In general,  $\phi_c$  and  
1133  $\phi_m$  decrease with increasing crystal alignment and particle shape anisotropy (i.e., equant vs

1134 elongated) and increases with dispersion in object size and surface roughness (e.g. Chong, 1971;  
 1135 Lejeune and Richet, 1995; Saar et al., 2001; Caricchi et al., 2007., 2008; Costa et al., 2009; Vona et  
 1136 al., 2011; Mueller, 2011; Mader et al., 2013; Klein 2018).



1137

1138 *Fig. A.2.1. (a) Relative viscosity as a function of crystal volume fraction of spheres at different strain-rates as*  
 1139 *calculated with the Costa et al. (2009) model. The figure shows, at first approximation, the inflection points at  $\phi_c$  and*  
 1140  *$\phi_m$ , separating the semi-dilute, concentrated and solid-like deformation regimes and the dilute regime as described in*  
 1141 *the text. The inflection points are a function of the applied shear rates and shape and size distributions.*  
 1142

1143 Mueller et al. (2011) by studying the rheological response of monodisperse suspensions with  
 1144 different aspect ratios, and adopting the Maron-Pierce equation, where

1145 
$$\eta_r = \left(1 - \frac{\phi}{\phi_m}\right)^{-2} \quad (\text{Eq. A3.5})$$

1146 concluded that both the flow index  $n$  and (Eq A3.1) the critical crystal fraction depend on  
 1147 the average particle aspect ratio ( $R$ ), such that:

1148 
$$n = 1 - 0.2R \left(\frac{\phi}{\phi_m}\right)^4 \quad (\text{Eq. A3.6})$$

1149 where the dependence of the  $\phi_c$  from  $R$ -parameter is given, as in Mueller et al. (2011) by:

1150 
$$\phi_m = \frac{2}{0.321R+3.02} \quad (\text{Eq. A3.7})$$

1151 It is worth mentioning that the parameterization of Mueller et al (2011) and Mader et al. (2013)  
 1152 cannot be extended above the critical solid fraction  $\phi_m$ . Other authors (e.g. Ishibashi, 2009; Mueller et  
 1153 al., 2011; Vona et al., 2011) proposed parameterizations obtained by modifications of the strain rate  
 1154 independent KD equation (Krieger and Dougherty, 1959)

1155 
$$\eta_r = \left(1 - \frac{\phi}{\phi_m}\right)^{-B\phi_m} \quad (\text{Eq. A3.8})$$

1156 to take into account the shear thinning effect on the rheology of suspensions. These authors  
 1157 provided modified strain rate dependent KD equations based on rheological measurements on low  
 1158 SiO<sub>2</sub> melts. Ishibashi (2009) provided the following expression for the relative viscosity:

1159 
$$\eta_r = \left(1 - \frac{\phi}{\phi_m}\right)^{-B_1\phi_m[1+\lambda \ln(1-\phi/\phi_c)\ln\dot{\gamma}]} \quad (\text{Eq. A3.9})$$

1160 where  $\phi_m = 0.6$  (as in the ER equation),  $B_1 = 5.46$  represents the intrinsic (melt) viscosity at  
 1161  $\dot{\gamma} = 1 \text{ s}^{-1}$  and  $\lambda = 0.118$  is an empirical constant which takes the shear thinning effect into account. In  
 1162 the Ishibashi (2009) parameterization, the fitting parameters were not related to the textural features  
 1163 of the suspensions (e.g., crystal shape, crystal shape dispersion, crystal size dispersion and  
 1164 orientation dispersion), such that the parameterization cannot be applied to other suspensions.

1165 Later, Cimarelli et al. (2011) by using a KD expression extended the application of the  
 1166 parameterization provided by Mueller et al (2011) to account for bimodal shape polydispersion of  
 1167 particles.

1168 Finally Vona et al. (2011) on the basis of rheological measurements on crystallizing  
 1169 polydispersed crystal-rich basalts provided a similar KD-derived parameterization

1170 
$$\eta_r = \left(1 - \frac{\phi}{\phi_m}\right)^{-2[1-\alpha \log(\dot{\gamma})]} \quad (\text{Eq. A3.10})$$

1171 in which  $\alpha$  is an empirical parameter equal to 0.06 and the effect of crystal shape can be  
1172 evaluated using the equation proposed by Mueller et al. (2011) (Eq. (9)) where, in order to account  
1173 for the disperse particle size distribution of the natural samples investigated, the average particles  
1174 ratio ( $\bar{R}$ ) is calculated by averaging the contribution of each group of particles having a certain  
1175 aspect ratio and is called as it follows:

$$1176 \quad \bar{R} = \frac{\sum \phi_i \bar{R}_i}{\phi} \quad (\text{Eq. A3.11})$$

1177 where  $\phi_i$  and  $\bar{R}_i$  are the crystal fraction and the mean aspect ratio of phase  $i$ . The model by  
1178 Vona et al. (2011) is able to describe the rheological behavior of suspensions as a function of strain  
1179 rate and textural features (i.e., crystal fraction, aspect ratios and shape polydispersion) and can be  
1180 therefore applied to the complexities of natural magmas. An additional step ahead is provided by  
1181 the work of Klein et al (2017, 2018) which express the critical crystal aspect ratio ( $\phi_c$ ) as a function  
1182 of a polydispersity parameter ( $\chi$ )( $\gamma$  in Klein et al., 2018):

$$1183 \quad \phi_c = 1 - (1 - \phi_{c,0}) \chi^\alpha \quad (\text{Eq. A3.12})$$

1184 where  $\phi_{c,0}$  represents the  $\phi_c$  for a unimodal distribution of particles and  $\alpha$  is an adjustable  
1185 parameter ( $\alpha = 0.173$ ). The polydispersity employed is the ratio of the specific surface of a  
1186 polydispersed system and that of a monodisperse system (Torquato, 2013; Wadsworth et al., 2017).  
1187 Klein and coauthors employing Maron-Pierce kind of equation (Eq. A3.5) to express the relative  
1188 viscosity and the parameterization of Mueller et al (2011) to account for the crystal ratio obtain and  
1189 interesting expression for the relative viscosity of a polydisperse suspension and provide as  
1190 supplementary material online an user-friendly spreadsheet .to calculate such a value  
1191 (<https://doi.org/10.1016/j.jvolgeores.2018.04.018>).

1192

### 1193 *A3.3. Three phase mixtures*

1194           Only few studies have explored the rheology of crystal and bubble-bearing magmas (e.g.  
1195 Cordonnier et al., 2009; Pistone et al. 2012, 2013; 2015, 2016; Campagnola et al., 2016; Plese et al.,  
1196 2018). Lavallée et al. (2007), Avard and Whittington (2011) have investigated natural lavas from  
1197 domes by uniaxial deformation experiments. The authors have observed pseudo-plastic behavior  
1198 with a strong shear thinning component for all the investigated magmas and provided equations  
1199 describing the apparent viscosity as a function of temperature and strain rate for the multiphase  
1200 magmas. Pistone et al. (2012) showed that three crystal and vesicle contents domains where non-  
1201 Newtonian rheological behaviour varying from shear thinning (e.g. viscosity decreases as strain-rate  
1202 increases and the flow index  $n < 1$ , Eq. A3.1) to shear thickening (e.g. viscosity increases as strain-  
1203 rate increases and the flow index  $n > 1$ , Eq. A3.1) through intermediate between shear thickening  
1204 and shear thinning were observed. The authors concluded that shear thinning occurred in crystal-  
1205 rich suspensions (55-65 vol%) and bubble content of 9-10 vol% as due to crystal size reduction and  
1206 shear localization. Shear tickening was instead observed in dilute suspensions (24 vol% crystal  
1207 content, 12 vol% bubbles) as due to bubbles coalescence and boudinage which favoured water loss  
1208 from the melts and sample degassing. Intermediate behaviour was observed for samples with 44  
1209 vol% crystals and 12 vol% bubbles probably due to the different temperatures and the time of  
1210 experiments which did not give always time to the bubbles and crystal to orient along the flow lines,  
1211 passing from prolate to oblate. The authors also showed that, caused by crystals and bubbles  
1212 interactions, the interactions between flow-lines and crystals plus bubbles cannot be compared to  
1213 that occurring for crystals and bubbles in only two-phase suspensions. Results from Pistone et al  
1214 (2012) were applied to understand the process of viscous death and rejuvenation of magmatic  
1215 bodies stored at depth (e.g. Bachmann and Bergantz, 2006).

1216           The individual effect of crystal and bubbles was theoretically parameterized by Phan-Thien  
1217 and Pham (1997) and later applied by Harris and Allen (2008) for the study of basaltic magmas

1218 from Mauna Loa and Mount Etna. Depending on the relative size of crystals ( $\Phi_{xtl, tot}$ ) and vesicles  
1219 ( $\Phi_b$ ), Phan-Thien and Pham (1997) present three equations:

1220 1. For crystals smaller than vesicles:

$$1221 \quad \eta = \eta_l \left(1 - \frac{\Phi_{xtl, tot}}{1 - \Phi_b}\right)^{-\frac{5}{2}} (1 - \Phi_b)^{-1} \quad (\text{Eq. A3.16})$$

1222 2. For crystals and vesicles of equal size:

$$1223 \quad \eta = \eta_l \left[1 - \Phi_{xtl, tot} - \Phi_b\right]^{\frac{(-5\Phi_{xtl, tot} + 2\Phi_b)}{(2(\Phi_{xtl, tot} + \Phi_b))}} \quad (\text{Eq. A3.17})$$

1224 3. For crystals larger than vesicles:

$$1225 \quad \eta = \eta_l \left(1 - \frac{\Phi_b}{1 - \Phi_{xtl, tot}}\right)^{-1} (1 - \Phi_{xtl, tot})^{-\frac{5}{2}} \quad (\text{Eq. A3.18})$$

1226 This treatment does not take into account the effect of textural variability, being applicable  
1227 to spherical particles only, and strain rate dependency on the rheology.

1228

1229



1230 **Appendix A4. Equations to estimate rheological properties or constrain characteristic**  
1231 **flow laws parameters starting from field observation of the geometrical features of lava**  
1232 **flows.**

1233  
1234 Where direct measurement of the viscosity of lava flows (see A1.5) is not accessible the  
1235 most accurate method to estimate the viscosity of lava flows is the so-called flow rate method (e.g.  
1236 Kolzenburg et al., 2017, 2018a; Belousov and Belousova, 2018). Measuring accurately the flow rate  
1237 of active lava flows allows calculating the apparent viscosities of the lava flows using Jeffreys'  
1238 equations:

1239 
$$\eta = \rho g h^2 * \frac{\sin(\alpha)}{3v} \text{ (Eq. A4.1); } \eta = \rho g h^2 * \frac{\sin(\alpha)}{2v} \text{ (A4.2)}$$

1240 where  $\eta$  is the viscosity of lava (Pa s),  $\rho$  is the bulk density (kg/ m3),  $g$  is the gravitational  
1241 acceleration (9.8 m/s2),  $h$  is the thickness of the lava flow (m),  $\alpha$  is the surface slope (degrees), and  
1242  $v$  is the velocity of lava flow (m/s). Eq. A4.1 is commonly used for data observing the flow front  
1243 velocity, and the Eq. A4.2 is used for velocity observations of the flow surface behind the flow front  
1244 (e.g. Nichols 1939; Gauthier 1973; Panov et al. 1988; Belousov nd Belousova, 2018).  
1245 Measurements using this procedure are reported in several studies (e.g. Nichols 1939; Minakami  
1246 1951; Macdonald 1963; Einarsson 1966; Walker 1967; Gautier 1973; Andreev 1978; Fink and  
1247 Zimbelman 1986; Vande-Kirkov 1987; Moore 1987; Panov 1988; Soldati 2016). Limitations of  
1248 Jeffrey's equations have been discussed at 3.1.2.

1249 Lava flow morphologies are thought, under particular assumptions, to reflect the rheological  
1250 characteristic of the lavas (e.g., Wilson and Head, 1983; Hiesing et al., 2007). Assuming that: i) the  
1251 rheological properties can be estimated from remote sensing data; ii) the geometry of the flow  
1252 depends on lava rheology and that lava behaves as a Bingham fluid (i.e. a fluid whose rheological  
1253 law is expressed by  $\tau = \tau_0 + \eta * \dot{\gamma}$ ; where  $\tau$  is the shear stress and  $\tau_0$  is yield strength, that is, the shear  
1254 stress necessary to first determine the beginning of the flow); iii) no inflation of the lava has  
1255 occurred; then the yield strength of the lava flow (Pa) and the lava flow viscosity can be related to

1256 its geometry according to the equations proposed by Moore et al. (1978). These equations,  
1257 commonly employed in planetary science, have been recently adopted in numerous scientific  
1258 contributions which also provide exhaustive reviews of the present state of art for the application of  
1259 this method (e.g. Hiesinger et al., 2007; Chevrel et al., 2015; Castruccio et al., 2010, 2014;  
1260 Kolzenburg et al., 2018a amongst the others). These contributions together with the new advances,  
1261 due to comprehend the effect of crystalline and bubbles phases on the rheology of multiphase lavas,  
1262 pointed out that this method, given the strong constitutive assumptions (in particular, single value of  
1263 viscosity and yield strength during lava field evolution and emplacement), are not accurate enough  
1264 to be employed, for instance, for real-time monitoring of lava flows (Chevrel et al., 2015).

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