

# Viscosity of crystal-free silicate melts from the active submarine volcanic chain of Mayotte

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2	Mayotte
3	
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16

## 17 Abstract

18 Following an unprecedented seismic activity that started in May 2018, a new volcanic edifice, 19 now called Mont Fani Maoré, was constructed on the ocean floor 50 km east of the island of 20 Mayotte (Indian Ocean). This volcano is the latest addition to a volcanic chain characterized 21 by an alkaline basanite-to-phonolite magmatic differentiation trend. Here, we performed 22 viscosity measurements on five silicate melts representative of the East-Mayotte Volcanic 23 Chain compositional trend: two basanites from Mont Fani Maoré, one tephri-phonolite and two 24 phonolites from different parts of the volcanic chain. A concentric cylinder viscometer was employed at super-liquidus conditions between 1500 K and 1855 K and a creep apparatus was 25 26 used for measuring the viscosity of the undercooled melts close to the glass transition 27 temperature. At super-liquidus temperatures, basanites have the lowest viscosity (0.11 - 0.99  $\log_{10}$  Pa·s), phonolites the highest (0.91 - 3.89  $\log_{10}$  Pa·s), while the viscosity of the tephri-28

29 phonolite falls in between (0.89 - 1.97  $\log_{10}$  Pa·s). Near the glass transition, viscosity 30 measurements have only been performed for one phonolitic melt because obtaining pure glass 31 samples for the basanitic and tephri-phonolitic compositions was unsuccessful due to the 32 formation of nanolites as evidenced by Raman spectroscopy. The phonolite has a viscosity of 33 10.19 to 12.30 log<sub>10</sub> Pa s at 1058 to 986 K. Comparison with existing empirical models revealed 34 an underestimation of 1.2 to 2.0 log units at super-liquidus and undercooled temperatures, 35 respectively, for the phonolite. This emphasizes (i) the lack of data falling along the alkaline basanite-to-phonolite magmatic differentiation trend to calibrate empirical models, and (ii) the 36 37 complexity of modeling the variations in viscosity as a function of temperature and chemical 38 composition for alkaline composition. The presented new measurements indicate that, at 39 eruptive temperatures between 1050 °C and 1150 °C (1323-1423 K), the oxidized, anhydrous, 40 crystal-free and bubble-free basanite melt is very fluid with a viscosity around 1  $\log_{10}$  Pa·s. In 41 contrast, the anhydrous phonolitic crystal- and bubble-free melt at eruptive temperatures 42 ranging from 800 to 1000 °C (1073-1273 K) would have a viscosity around 6 - 10  $\log_{10}$  Pa·s. 43 Considering that both basanitic and phonolitic melts from the Mayotte submarine volcanic chain contain less than 6 % crystals and a significant amount of water, such viscosity values 44 45 are probably upper limits. These new viscosity measurements are essential to define eruptive models and to better understand the storage and transport dynamics of Comoros Archipelago 46 47 magmas, and of alkaline magmas in general, from the source to the surface.

48

#### 49 keywords

50 viscosity, alkali magmas, submarine volcano, volcanic eruption, Raman spectroscopy

#### 51 1. Introduction

52 Following an unprecedented intense seismic crisis that started in May 2018 (Lemoine et al. 53 2020; Feuillet et al. 2021), surveys revealed a new submarine volcanic edifice, recently named 54 Mont Fani Maoré, 936 m tall with a base at around 3,500 m depth, 50 km east of Mayotte (Indian Ocean). This large eruption has extruded around 6.55 km<sup>3</sup> of basanite magma, with 55 56 first estimates of magma transfer rates from source to surface of a minimum of 30 days (Cesca et al. 2020; Berthod et al. 2021a). The source of this magma is evaluated to be located at 30 -57 58 50 km depth into the underlying mantle, with the potential involvement of an intermediate 59 magma chamber located at ~17 km depth (Berthod et al. 2021b). This new volcano is an 60 addition to the East-Mayotte Volcanic Chain, characterized by the emission of magmas falling 61 along an alkaline basanite-to-phonolite magmatic differentiation trend. This volcanic chain is 62 quite complex and characterized by large effusive lava flow fields, and by the presence of more explosive volcanoes (Figure 1) (Puzenat et al. 2022; Gurioli et al. 2023; Komorowski et al. 63 64 2023). To better understand and constrain the storage processes and transfer rates of the magmas that feed the East-Mayotte Volcanic Chain and the dynamics of their emplacement at 65 66 surface, it is necessary to constrain the viscosity of the basanite-to-phonolite melts, which 67 determines the mobility of magmas toward the surface as well as their fragmentation behavior in the conduit and flow at the surface (Dingwell et al. 1996; Papale 1999; Pistolesi et al. 2011). 68 69 To constrain the viscosity of magmas and lavas, one can potentially rely on the various 70 experimental and modeling efforts that led to important improvements in our knowledge of 71 how it is controlled by temperature (Vogel 1921; Tamman and Hesse 1926; Fulcher 1925; 72 Adam and Gibbs 1965; Nascimento and Aparicio 2007), chemical composition (Bottinga and 73 Weill 1972; Shaw 1972; Giordano et al. 2008), volatile elements (e.g., Whittington et al. 2000, 2001), bubbles (Lejeune et al. 1999; Llewelin and Manga et al. 2005) and crystal contents 74 75 (Lejeune and Richet 1995; Kolzenburg et al. 2022). Yet, there is not a general and very accurate

76 magma viscosity model to date, due to the complexity in estimating the influence of the 77 different parameters listed above. Focusing only on the silicate melt phase of magmas and lavas, many effects exist at the atomic level, such as aluminum coordination changes or metal 78 79 cation mixing, that result in strong and non-linear variations in melt viscosity depending on 80 composition. In particular, in alkali-rich melts, the way Na and K mix may strongly affect 81 viscosity (Poole, 1948; Richet, 1984; Le Losq and Neuville, 2013, 2017). In Al-poor melts, the 82 ideal mixing between Na and K results in an important decrease in viscosity (Richet 1984), whereas in Al-rich melts, Na and K occupy different environments and do not mix ideally (Le 83 84 Losq and Neuville 2013, 2017; Le Losq et al. 2021). At a given temperature, this leads to increasing the melt viscosity by several orders of magnitude upon increasing the melt 85 86 K/(K+Na) ratio. Existing parametric viscosity models (e.g., Hui and Zhang 2007; Giordano et 87 al. 2008) failed to properly reproduce such an effect (Le Losq and Neuville 2013; Robert et al. 88 2019). Therefore, their accuracy for the prediction of the viscosity of melts along alkaline 89 magmatic series is to be improved. Only models integrating thermodynamic and physico-90 chemical knowledge allow to properly model the way Na and K non-ideal mixing alter the 91 viscosity of Al-rich alkali melts. However, this has been applied only on quaternary alkali 92 aluminosilicate melts far from natural compositions (Robert et al. 2019; Starodub et al. 2019; Le Losq et al. 2021, Neuville and Le Losq 2022). Therefore, to properly constrain the viscosity 93 94 of alkali melts, one needs to use ad hoc models for increased precision, as it has been performed 95 for the phonolite of Erebus volcano (Antarctica) for instance (Le Losq et al. 2015a). 96 In this contribution, we present new viscosity measurements performed on samples

97 representative of the lava compositions that can be found along the East-Mayotte submarine
98 volcanic chain. Those samples fall along the moderately silica-undersaturated trend ("Karthala99 trend") of the Comoros Archipelago (Pelleter et al. 2014; Bachèlery et al. 2016; Bachèlery and
100 Hémond 2016). The studied compositions include two basanite lavas from the recent eruption

101 (Mont Fani Maoré volcano), and two phonolite and a tephri-phonolite samples from other 102 emission sites in the submarine volcanic chain (Berthod et al. 2021a; Feuillet et al. 2021; 103 Puzenat et al. 2022; Figure 1). Viscosity is measured with a concentric cylinder viscometer at super-liquidus temperatures, in the  $10^{\circ}$  -  $10^{\circ}$  Pa·s range, and with a creep apparatus close to the 104 glass transition temperature between  $10^8$  and  $10^{13}$  Pa·s. We also examine the effect of the water 105 106 content on the viscosity of the different magma compositions present at Mayotte. The results (i) provide the foundation necessary to investigate the eruptive behavior of the Mayotte 107 108 volcanic system, (ii) strengthen the current knowledge on alkaline series and (iii) could 109 contribute to refine the pre-existing empirical viscosity models based on chemical composition.

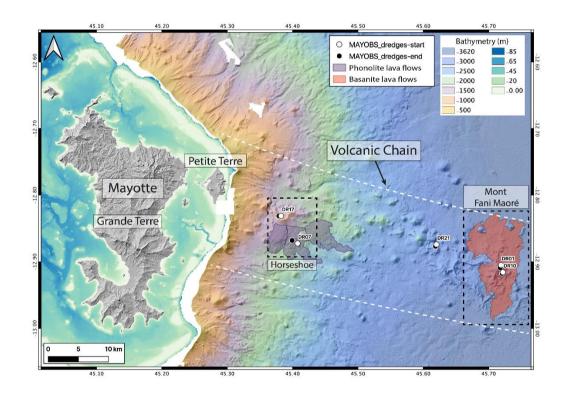


Figure 1. Geological map of the submarine volcanic chain of Mayotte showing the location of
the dredged samples (DR labels). Mont Fani Maoré lava flows are modified from Feuillet et
al. (2021). Modified from Berthod et al. (2021a). Background is the bathymetry from the
Homonim project (SHOM 2015), DEM Litto3D IGN-SHOM (SHOM 2016) and MAYOBS1
(doi:10.17600/18001217).

## 116 2. Materials and Methods

#### 117 2.1. Starting material

118 In this study, the investigated samples were collected by dredging operations that collected

- between 100 1000 kg of rocks, from water depths ranging from 1,370 to 3,455 m (Figure 1).
- 120 These dredges were operated by the R/V Marion Dufresne II and R/V Pourquoi Pas? during
- 121 the following oceanographic cruises (Rinnert et al., 2019): MAYOBS 1 (Feuillet, 2019),
- 122 MAYOBS 2 (Jorry 2019), MAYOBS 4 (Fouquet and Feuillet, 2019), MAYOBS 15 (Rinnert
- 123 et al., 2020), and GEOFLAMME (Rinnert et al., 2021) (Table 1).
- **Table 1.** Location of the dredges performed during the oceanographic cruises. Latitudes and
- 125 Longitudes are given in decimal degrees (DD).

	Oceanographic cruises	DOI of the oceanographic cruises	S	Start dredging			End dredging		
Dredges		<u>10.18142/291</u>	Latitude	Longitude	Depth	Latitude	Longitude	Depth	
DR01	MAYOBS 1	10.17600/18001217	-12.905	45.719	3050 m	-12.909	45.718	2820 m	
DR07	MAYOBS 2	10.17600/18001222	-12.872	45.407	1590 m	-12.868	45.399	1585 m	
DR10	MAYOBS 4	10.17600/18001238	-12.916	45.722	3120 m	-12.918	45.721	2950 m	
DR17	MAYOBS 15	10.17600/18001745	-12.831	45.382	1370 m	-12.831	45.379	1340 m	
DR21	GEOFLAMME	10.17600/18001297	-12.874	45.620	2719 m	-12.876	45.618	2629 m	

126

We selected five samples that represent the diverse composition of the volcanic deposits along
the Mayotte submarine volcanic chain (Fig. 1 and Table 2). Two samples (MAY01-DR010101
and MAY04-DR100504, hereafter named DR01 and DR10, respectively) are from Mont Fani
Maoré, the new volcano that has been in activity from 2018 to 2021 (Fig. 1, Lemoine et al.

131 2020; Berthod et al. 2021b; Feuillet et al. 2021). These two samples are fragments of basanite 132 lava flows collected on the central edifice and were emitted during the first year of the eruption 133 (before May 2019, Berthod et al., 2021b). Two other samples (MAY02-DR070201 and 134 MAY15-DR170404, hereafter named DR07 and DR17, respectively) were recovered at 10 -135 15 km east of Petite Terre Island of Mayotte on the "Horseshoe" volcanic structure (Fig. 1, 136 Berthod et al. 2021a; Feuillet et al. 2021; Puzenat et al. 2022; Gurioli et al. 2023). DR07 is a 137 fragment of a Holocene phonolitic lava flow located on the southeastern part of the Horseshoe 138 site. DR17 is a phonolitic pyroclast bomb collected at the bottom of the internal western side 139 slope of the Horseshoe structure. Finally, the fifth sample (GFL-DR2110, hereafter named 140 DR21) was collected on a seamount located in the middle of the submarine volcanic chain, 10 141 km west of Mont Fani Maoré (Rinnert et al., 2021). This sample is a fragment of lava and has 142 a tephri-phonolitic composition.

About 200 g of each sample were used to produce the starting glasses necessary for the experiments. The rock samples were first crushed and powdered. A portion of the powder was used for bulk rock chemical analyses and the rest for viscosity measurements. Chemical analyses have also been performed on the glass chips produced after high- and lowtemperatures viscometry.

148

#### 149 2.2. Major element analyses

Bulk rock major element composition of the starting materials was analyzed at Laboratoire Magmas et Volcans (LMV, Clermont-Ferrand, France). Samples were crushed into millimetersize chips using home-made thermally hardened steel jaws, and powdered in a motorized agate mortar. Major elements were analyzed by Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES). Powdered samples were melted with LiBO<sub>2</sub> in a magnetic induction oven at 1100 °C for 5 min using graphite crucibles. The glass beads were then dissolved in a solution of deionized water and nitric acid (1 M) and diluted by a factor of 2000 to produce the solution analyzed by an Agilent 5800 VDV ICP AES in radial mode. Analytical uncertainties ( $\pm 2\sigma$ ) vary between 1 and 3 % except for K<sub>2</sub>O, MnO (4 %) and P<sub>2</sub>O<sub>5</sub> (7 %) for the DR-N standard (diorite). For the analysis, the plasma flow was 12 L/min, the nebulizer flow was 0.7 L/min and the radio frequency power was 1.2 kW.

161 Glass chips resulting from spindle quench after the super-liquidus viscometry were mounted 162 as polished sections for chemical analysis carried out with the CAMECA SX Five Tactis 163 electron microprobe at LMV. We used an accelerating potential of 15 kV at current of 8 nA, 164 with a defocused beam of 20 µm diameter to avoid Na migration under the electron beam. 165 Natural and synthetic mineral standards, including orthoclase (K, Al), albite (Na), wollastonite 166 (Si, Ca), fayalite (Fe), forsterite (Mg), TiMnO<sub>3</sub> (Ti, Mn), NiO (Ni), Cr<sub>2</sub>O<sub>3</sub> (Cr), and fluorapatite 167 (P) were used for routine calibration.

We also calculated the NBO/T parameter (number of non-bridging oxygens per tetrahedral),
representing the degree of polymerization of the melt following Mysen et al. (1982):

170 
$$\frac{NBO}{T} = \frac{(2O) - (4T)}{T},$$
 (1)

171 with O the atomic proportion of oxygen atoms and T that of cations entering as network formers in tetrahedral coordination, namely SiO<sub>2</sub>, TiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and Fe<sub>2</sub>O<sub>3</sub> (see for a review of such 172 173 concepts Le Losq et al. 2019; Neuville and Le Losq 2022). Here, the total iron is expressed as 174  $Fe^{3+}$  as the viscosity experiments were performed in an oxidizing environment. This 175 assumption is a valid simplification: estimations with the Moretti (2005) model yields 90 % or 176 more of Fe residing in the melt as Fe<sub>2</sub>O<sub>3</sub> at the conditions of our experiments. The NBO/T of 177 the natural melt may thus be higher, considering the occurrence of reduced iron and its 178 influence on the melt structure (e.g., Le Losq et al. 2021).

179

### 2.3. <u>High temperature viscometry</u>

180 High temperature viscosity measurements are performed at super-liquidus conditions, in air, using a concentric cylinder viscometer (Dingwell 1986; Spera et al. 1988) at the Institut de 181 physique du globe de Paris (IPGP, Paris, France). Powdered glass samples are first melted at 182 183 1800 K in a Pt<sub>95</sub>Au<sub>5</sub> cylindrical crucible (50 mm height, 27 mm inner diameter, 1 mm wall 184 thickness) in a muffle furnace. Once the crucible is full, it is inserted in the hot zone of the 185 viscometer vertical tube furnace, heated by Super Kanthal 33 elements in air. The length of the 186 hot zone was determined by temperature measurement inside the crucible: the vertical and 187 radial temperature gradients inside the crucible are respectively less than 2 K for 5 cm (vertical 188 gradient) and 1 K for 2.7 cm (radial gradient) at 1500 K and a little less at 1800 K. The inner 189 rotating spindle size is 14 mm in diameter, 21 mm in height and has 23° conical extremities to 190 reduce termination effects and a 5 mm diameter stem. Viscosity is measured using a Rheomat 191 115 rheometer head, which allows rotating the cylinder at angular velocities ranging between 192 0.05 and 780 rpm (Neuville 2006). The torque exerted on the cylinder by the sample is recorded 193 digitally. The correspondence between the exerted torque and the viscosity is calibrated against 194 the reference sample NBS SRM 710a, for which the viscosity-temperature relationship is 195 accurately known (Neuville 2006). The accuracy is of the order of 0.02 log Pa·s (e.g., Neuville 2006). 196

197

#### 198 2.4. Low temperature viscometry

Low temperature viscosity measurements are performed near the glass transition temperature  $(T_g)$  using a creep apparatus at IPGP (Neuville and Richet 1991; Neuville 2006). Glass samples used for measurements are (i) a parallelepiped of 7.9 mm length, and (ii) a small cylinder of 2.2 mm diameter x 8.1 mm length. A silver cylinder is placed around the sample, creating a small chamber in which temperature is homogeneous. Lateral and vertical temperature

gradients are controlled using two Pt-PtRh<sub>10</sub> thermocouples (ITS90 type S thermocouples): 204 205 before and during each measurement, lateral and vertical temperature gradients were always 206 lower than 0.2 K. To measure sample viscosity at a given temperature, we performed 20 to 30 measurements at different stresses (between 6.4 and 8.2 log Nm<sup>-2</sup>) to check for the occurrence 207 208 of a non-Newtonian behavior, which could be a sign of crystallization of the sample. Each 209 reported viscosity value at a given temperature is the statistical mean of these measurements. 210 Measurements carried out on the NBS 717 glass show that errors on viscosity measurements 211 are lower than 0.03 log Pa·s with this apparatus (e.g., Neuville 2006).

212

#### 213 2.5. <u>Raman spectroscopy</u>

214 The Raman spectra of the glasses (pre- and post-experiments) were recorded with the Labram 215 HR Evolution spectrometer available at IPGP, equipped with a Peltier-cooled CCD and a 1800 lines mm<sup>-1</sup> grating. The samples were excited with a Coherent MX 488 nm solid-state laser 216 217 focused through a  $\times 50$  Olympus objective on the sample surface. The confocal aperture of the spectrometer was set to 50. With this setup, spectral resolution is  $\sim 3 \text{ cm}^{-1}$  and spatial resolution 218 is <1 cm<sup>-1</sup>. All spectra were recorded with the laser focused at 3-5 µm below the sample's 219 220 surface to avoid any surface effects (Behrens et al. 2006, Schiavi et al. 2018). The laser power 221 on the sample has been adjusted to lower than 10 mW to avoid any potential damage on the 222 sample (iron oxidation effects or melting). Potential damage was checked by recording several 223 spectra on the same spot and varying laser power, without the identification of any effect. 224 Raman data treatment was performed using the Python programming software, with the *rampy* 225 open-source software library (Le Losq 2018).

### 226 **3.** <u>Results</u>

#### 227 3.1.<u>Major element composition</u>

The bulk rock chemical composition of the pre-experiment samples and the post-experiments glass compositions, determined by ICP-AES and by electron microprobe analyses respectively, are reported in Table 2 and presented in Figure 2.

231 DR01 and DR10 basanites have a very similar composition between 47.1 and 47.7 wt% SiO<sub>2</sub> 232 and 7.2 to 7.8 alkali (Na<sub>2</sub>O + K<sub>2</sub>O) wt% (Fig. 2). These samples have low MgO contents, 233 between ~4.8 and ~5.4 wt%, and high FeO<sub>tot</sub> contents, between ~12.5 and ~12.7 wt% (Table 234 2), making them the most iron-rich among all the studied samples from the East-Mayotte 235 submarine volcanic chain. DR07 and DR17 phonolite samples have similar silica content of 236 57.7 - 58.9 wt% SiO<sub>2</sub> but DR07 has an alkali content of 13.0 wt% while DR17 is more enriched 237 in alkali with 14.2 wt% (Fig. 2 and Table 2). Compared to basanite samples, phonolite samples 238 have lower FeO<sub>tot</sub> contents, ranging from 6.1 to 7.2 wt%. Finally, sample DR21 has a tepri-239 phonolitic composition, falling in between the basanite and phonolite fields of the TAS diagram 240 (Fig. 2) with SiO<sub>2</sub> and alkali contents of 53.2 wt% and 10.8 wt%, respectively (Table 2) and 241 an intermediate FeO<sub>tot</sub> content of 10 wt%.

242 For most samples, the major element contents show no significant differences before and after

viscosity experiments (Table 2). We however note that the composition of DR17 after the high-

temperature experiment has a slightly higher SiO<sub>2</sub> content of 1.5 wt%. This is accompanied by

245 iron loss of 0.5 wt% (Table 2).

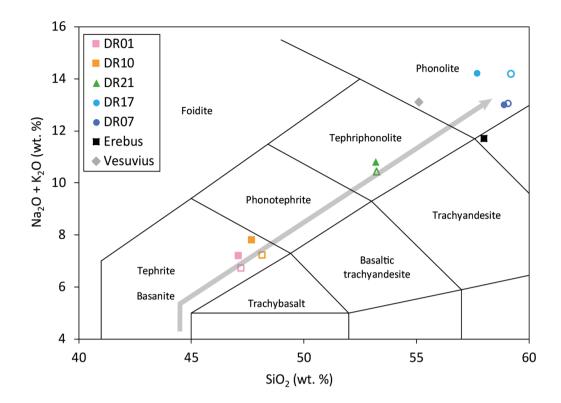




Figure 2. TAS diagram showing normalized compositions of the studied samples before (filled symbols, ICP-AES) and after (empty symbols, Electron Microprobe) viscosity measurements. Compositions of phonolite lavas from Erebus and Vesuvius (GP79, Grey Pumice of the 79 eruption) are also shown for comparison (data from Le Losq et al. 2015a). The grey arrow shows the moderately silica-undersaturated trend ("Karthala-trend") identified by Pelleter et al. (2014) and Bachèlery and Hémond (2016). Error bars are smaller than symbol size. 

261 *Table 2.* Normalized major element composition in wt% pre- (first line) and post- (second line)
262 experiments of the studied samples. The post-experiment composition are an average of at least

ten measurements made on glass. The composition of Erebus and Vesuvius GP79 from Le Losq

264 *et al.* (2015a) are also reported for comparison.

Sample	$SiO_2$	$TiO_2$	$Al_2O_3$	FeO(t)	MnO	MgO	CaO	Na <sub>2</sub> O	$K_2O$	$P_2O_5$	NBO/T
Before and after	Before and after experiments										
DR01	47.1	3.3	15.3	12.7	0.2	5.4	7.3	4.7	2.5	1.6	0.3
DR01	47.2	3.4	14.8	13.2	0.3	5.4	7.3	4.4	2.4	1.6	-
DR10	47.7	3.1	15.2	12.5	0.2	4.8	6.8	5.2	2.6	1.9	0.3
DKIU	48.1	3.2	15.3	12.6	0.3	4.9	6.8	4.6	2.6	1.7	-
DR21	53.2	1.6	16.5	9.8	0.3	1.9	4.2	6.8	4.0	1.0	0.2
DR21	53.2	1.7	16.9	10.0	0.3	2.0	4.5	6.5	4.0	1.0	-
DB07	58.9	0.4	18.2	7.2	0.3	0.4	1.6	7.6	5.4	0.3	0.1
DR07	59.1	0.4	18.6	6.4	0.3	0.3	1.6	7.6	5.7	0.3	-
DD17	57.7	0.1	18.7	6.1	0.3	0.1	1.3	8.6	5.6	0.1	0.1
DR17	59.2	0.1	19.1	5.6	0.2	0.1	1.3	8.4	5.8	0.1	-
Erebus melt	58.0	1.0	20.0	5.4	0.2	0.9	2.6	7.1	4.6	0.0	0.1
Vesuvius GP79	55.1	0.5	19.5	4.1	0.1	1.8	5.3	4.4	8.7	0.0	0.3

265

The calculated NBO/T values of the oxidized melts range from 0.1 to 0.3 (Table 2). DR01 and
DR10 basanite samples have the highest NBO/T (0.32 and 0.33). On the other hand, DR07 and
DR17 phonolite samples have the lowest NBO/T (0.10 and 0.11) related to their higher SiO<sub>2</sub>
content. The tephri-phonolite has an intermediate NBO/T of 0.20.

270

#### 271 3.2. <u>Viscosity data</u>

The viscosity in air for anhydrous and crystal- and bubble-free basanitic, tephri-phonolitic and phonolitic melts of the Mayotte submarine volcanic chain are presented as a function of temperature in Figures 3 and 4 and given in Table 4. High-temperatures viscosity was obtained for all samples while low-temperature viscosity was only obtained for DR07 (Fig. 3 and Table 4). As discussed below, this is because it was not possible to prepare the crystal-free glasssamples needed for the low-temperature measurements for the other samples.

High-temperature viscosity measurements were performed between 1565 K and 1860 K. The
lower bond of this range is motivated by the fact that, according to the MELTS model (Ghiorso
and Sack 1995), magnetite crystallization is expected to occur below 1450 K for the studied
samples. In this restricted high-temperature range, the relationship between viscosity and
temperature is mostly linear (Fig. 3), in agreement with Bottinga et al. (1982). The data are
therefore interpolated using the Arrhenius relation:

284 
$$\log_{10} \eta = A_{Arr} + \frac{Ea}{RT},$$
 (2)

where  $\eta$  is the melt viscosity (Pa·s), A<sub>Arr</sub> is a constant, T is temperature (K), R is the gas constant (J mol<sup>-1</sup> K<sup>-1</sup>), and E<sub>a</sub> is the viscous flow activation energy (kJ mol<sup>-1</sup>).

For DR07, as we obtained both high- and low-temperatures data (**Fig. 3** and **Table 4**), we use the Vogel-Fulcher-Tamman (VFT; Vogel 1921; Tamman and Hesse 1926; Fulcher 1925) equation to interpolate the viscosity data:

290 
$$\log_{10} \eta = A + \frac{B}{T-C},$$
 (3)

with A, B and C the pre-exponential factor, the pseudo-activation energy and the VFT
temperature, respectively. The viscosity-temperature variation of the phonolite melts from
Vesuvius and Erebus are also shown as comparison (Fig. 3).

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- 296
- 297
- 298
- 299

	Basanit	tic melt		Tephri-	phonolitic melt	Phonolitic melt				
	DR01 DR				DR21		DR07	DR17		
Т	η	Т	η	Т	η	Т	η	Т	η	
(K)	(log10 Pa·s)	(K)	(log10 Pa·s)	(K)	(log10 Pa·s)	( <i>K</i> )	(log10 Pa·s)	(K)	$(log_{10} Pa \cdot s)$	
						986	12.30			
						1006	11.61			
						1016	11.30			
						1025	11.00			
						1027	10.87			
						1037	10.67			
						1048	10.44			
						1058	10.19			
								1499	3.89	
								1525	3.71	
								1551	3.54	
								1576	3.37	
1595	0.99	1596	1.16	1596	1.97	1597	3.10	1596	3.27	
1646	0.79	1648	0.94	1647	1.73	1648	2.80	1646	2.95	
1698	0.61	1699	0.74	1696	1.50	1700	2.52	1698	2.67	
1749	0.43	1753	0.56			1751	2.25	1750	2.40	
1761	0.40			1762	1.23					
				1766	1.20			1766	2.32	
1801	0.27	1809	0.38	1809	1.04	1804	2.01	1807	2.12	
		1810	0.34							
1855	0.11			1858	0.89	1860	1.75	1855	1.91	

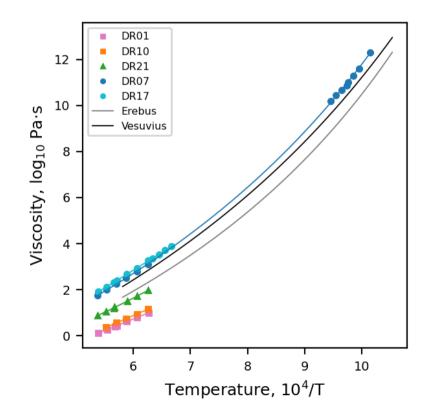
300 Table 3. Viscosity measurements of the basanitic-to-phonolitic melts along the Mayotte

301 submarine volcanic chain. The uncertainty is equal to or lower than 0.03 log (Pa·s).

302

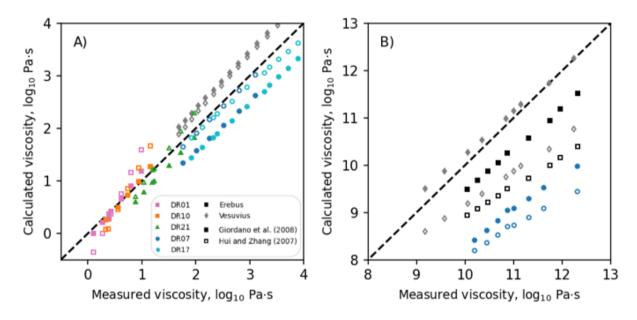
303 At super-liquidus temperatures, the basanitic melts have the lowest viscosity  $(0.99 \text{ to } 0.11 \log_{10} 10)$ 304 Pa·s at 1595 to 1855 K) followed by the tephri-phonolitic (1.97 to 0.89 log<sub>10</sub> Pa·s at 1596 to 305 1858 K) and the phonolitic melts (3.89 to 0.91  $\log_{10}$  Pa s at 1499 to 1855 K). Figure 3 shows 306 that, at a given temperature (between 1500 to 1855 K), the viscosity of the basanitic melts differ 307 from those of the phonolitic ones by about two orders of magnitude. In particular, at 1750 K, 308 the viscosity is of 0.43, 1.23 and 2.40  $\log_{10}$  Pa·s, for the basanitic (DR01), tephri-phonolitic 309 (DR21) and phonolitic (DR17) melts, respectively (Table 3). At undercooled temperatures, the 310 viscosity of DR07 ranges from 12.3 log<sub>10</sub> Pa·s at 986 K to 10.2 log<sub>10</sub> Pa·s at 1058 K (Fig 3.).

311 We compare our measurements to the Giordano et al. (2008) and Hui and Zhang (2007) 312 empirical models (hereafter abbreviated as the GRD and HZ models, respectively). Based on 313 Figure 4, the GRD model tends to fit well the viscosity of basanite while a slight 314 underestimation is observable for the tephri-phonolitic melts at high-temperatures. This 315 becomes critical for the phonolite melts for which the GRD model significantly underestimates 316 the viscosity at both high- and low-temperatures (Fig. 4). Conversely the HZ model tends to 317 slightly overestimate the viscosity of the basanites and tephri-phonolites at high-temperatures. 318 It is in good agreement with the measured viscosity of phonolites at high-temperatures but 319 significantly underestimates the values at low-temperatures (Fig. 4). As also shown by Le Losq 320 et al. (2015a) the GRD model underestimates by ~0.5 log units the viscosity of Erebus (Fig. 321 4). In contrast, for the Vesuvius melt, at super-liquidus temperatures there is a difference of 322 less than almost ~0.2 log units between the predicted and the measured viscosity (Fig. 4A), 323 while at low-temperatures the GRD model fits well the viscosity data (Fig. 4B).



**Figure 3.** Viscosity  $(log_{10} Pa \cdot s)$  as a function of inverse temperature  $(K^{-1})$  for the Mayotte basanite-to-phonolite magmatic differentiation trend. Squares, diamonds and circles represent measurements made on basanites (DR01 and DR10), tephri-phonolite (DR21) and phonolites (DR07 and DR17), respectively. Erebus (grey curve) and Vesuvius (black curve) are also represented (data from Giordano et al. 2009; Le Losq et al. 2015a). Solid lines are interpolations of the data with the VFT equation (Eq. 3, Table 4). Error bars are smaller than the symbol size.

332



**Figure 4.** Comparison between the measured viscosity  $(log_{10} Pa \cdot s)$  and the calculated ones with the models of Giordano et al. (2008) (filled symbols) and of Hui and Zhang (2007) (open symbols) in the super-liquidus (A) and undercooled (B) temperature ranges. Error bars are smaller than the symbol size.

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333

#### 339 3.3. <u>Raman spectroscopy</u>

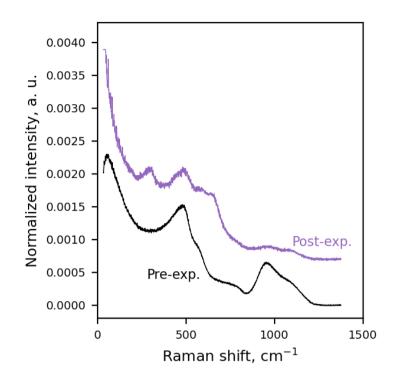
To check for crystallization during the low-temperature viscosity experiments, we compare the Raman spectra of DR07 before and after the low-temperature viscosity experiments (**Fig. 5**).

- 342 Before the experiment, the DR07 glass Raman spectrum exhibits three main broad bands near

343 80, 490 and 1000 cm<sup>-1</sup>. The first is the Boson peak assigned to transverse acoustic vibrational 344 modes promoted in silicate glasses by cooperative inter-tetrahedral vibrations (Buchenau et al. 345 1986; Malinovsky and Sokolov 1986; Hehlen et al. 2002). This is a universal signature of the 346 glassy state (Malinovsky and Sokolov 1986), that tends to quickly disappear in the presence of 347 crystals (e.g., Takahashi et al. 2009). The second is the inter-tetrahedral T-O-T (T = Si, Al) 348 vibrations in the aluminosilicate network. The third is assigned to intra-tetrahedral T-O 349 stretching vibration (Mysen et al. 1982; McMillan 1984; Le Losq et al. 2014).

After the low temperature viscometry, the recovered DR07 sample does not show the Boson peak anymore (**Fig. 5**). This indicates that some degree of crystallization must have occurred during the viscosity measurements near the glass transition.

353 We also acquired Raman spectra on all the other samples quenched during the initial glass 354 preparation (see supplementary Fig. A1). Although microscopic inspection of the samples appeared crystal-free, and although the Raman spectra above 200 cm<sup>-1</sup> may not present sharp 355 356 peaks typical of crystals, no Boson peak was visible. This indicates that initial glass samples 357 other than DR07 contained crystals at a sub-nanometric to nanometric scale (see Discussion). 358 Interestingly, we did not succeed to obtain a pure glass for the DR17 phonolite despite a very 359 similar chemical composition with DR07. This may be explained by the slightly higher Na<sub>2</sub>O and Al<sub>2</sub>O<sub>3</sub> contents that could be responsible for a different behavior. This testifies that 360 361 basanitic, tephri-phonolitic and phonolitic crystal-free glass samples of the required size for 362 low temperature viscosmetry are extremely difficult to obtain.



363

364 *Figure 5.* Uncorrected Raman spectra of DR07 phonolite products pre (black curve)- and post
365 (purple curve)-experiments at low-temperature.

366

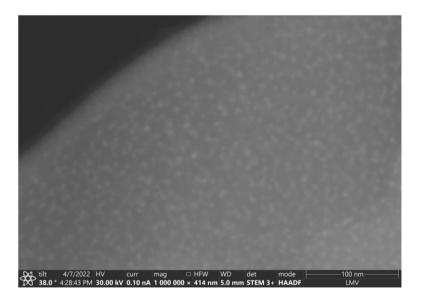
#### 367 4. Discussion

#### 368

## 4.1. Nanolite crystallization during low-temperature measurements

369 DR07 is the only sample for which we obtained a glassy piece large enough to perform near- $T_g$  viscometry. However, during the experiment, the sample seems to have undergone some 370 371 degree of crystallization as evidenced by the disappearance of the Boson peak (Fig. 5). To 372 confirm that crystallization occurred we acquired a scanning transmission electron microscope 373 (STEM) image with the Helios 5 (ThermoFisher Scientific) scanning electron microscope 374 coupled with a focused ion beam (Xe plasma FIB-SEM) at LMV. Images show the presence 375 of homogeneously distributed brighter particles at the nanoscale (Fig. 6). Similar particles with 376 a size ranging from 5 to 30 nm have been observed and characterized as nanolites (Di Genova 377 et al. 2017, 2018, 2020).

378 The effect of crystallization on viscosity may reach several orders of magnitudes (e.g., Lejeune 379 et Richet, 1995; Costa et al. 2009; Mader et al. 2013), particularly in presence of microlites 380 (Del Gaudio et al. 2013) or nanolites (Di Genova et al. 2020; Le Losq et al. 2021). In the present 381 case, we observe neither a deviation of the viscosity measurements as a function of time nor 382 the apparition of a non-Newtonian behavior. Therefore, it seems that the presence of nanolites 383 only has limited effects on our measurements. However, this finding suggests that nanolite 384 crystallization happens very quickly in melts of iron-bearing alkaline compositions and even 385 in silica-rich melts such as phonolitic. To check for this, we placed a DR07 glassy sample in 386 an annealing furnace and setup the temperature at the glass transition, with a particular care in 387 avoiding overshooting while checking the sample temperature with a Pt-PtRh10% 388 thermocouple placed in contact with the sample. The Raman spectra of the retrieved sample 389 also showed the presence of crystals in the glass after annealing and is identical to the post-390 experiment spectra (Fig. 5). These observations indicate that viscosity measurements for such 391 compositions in the undercooled temperature domain that were presented in previous studies 392 (Giordano et al. 2000, 2005; Giordano and Dingwell 2003; Le Losq et al. 2015a; Whittington 393 et al., 2000, 2001) may probably have been also affected by nanolite formation. We add that 394 because diffusion increases with decreasing Si content (Zhang and Ni 2010), this effect is expected to be even stronger for tephri-phonolitic and basanitic composition, preventing pure 395 396 glass samples from being obtained. Great care should therefore be taken during low 397 temperature viscometry even for phonolitic melt, and Raman spectra down to the Boson peak 398 wavelength range should be systematically acquired.



400 *Figure 6.* Scanning transmission electron microscope image of the DR07 sample post low401 temperatures viscosity measurements.

402

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#### 403 4.2. <u>Melt viscosity and comparison with models</u>

404 A discrepancy between experimental measurements and parametric viscosity models, as 405 observed here (Fig. 4), has also been previously reported for alkaline composition by Le Losq 406 et al. (2015a). There are two likely causes for the significant difference between the GRD 407 model and the measured viscosity at low temperature. First, unlike at high temperatures, in 408 undercooled melts, important and nonlinear variations in melt configurational entropy result 409 from changes in melt composition and drive important changes in viscosity (Richet 1984; 410 Neuville and Richet 1991; Le Losq et al. 2021). It is challenging for parametric models to 411 reproduce such strongly nonlinear behavior of viscosity variations and so, this could explain 412 the discrepancy between the GRD model and our low-temperature viscosity measurements. 413 Secondly, the GRD model has been calibrated with a restricted alkaline basanite-to-phonolite compositional data subset (Giordano et al. 2008). This could reduce the accuracy of the 414 415 viscosity prediction of this model for such compositions. This also explains why the Vesuvius 416 melt is better reproduced as this composition was part of the database used to parametrize the417 GRD model (Giordano et al. 2008).

418 Additional comparison has been performed between our viscosity measurements and the 419 predictions from the Hui and Zhang (2007) model. The HZ model is in relatively good 420 agreement with our viscosity measurements at high-temperatures (Fig. 4A) whereas it 421 significantly underestimates by ~1.3  $log_{10}$  units the viscosity of phonolitic melts at low-422 temperatures (Fig. 4B). Similar differences are observed between the HZ model and the 423 measurements made on the Erebus (Le Losq et al. 2015a) and Vesuvius GP79 (Giordano et al. 424 2009; Le Losq et al. 2015a) samples. According to Hui and Zhang (2007), the model could be 425 improved in part by adding new viscosity measurements at low-temperatures and by 426 considering the effect of ferric and ferrous iron. Also, as for the GRD model, the HZ model is 427 a general model leading to an increase in uncertainty for specific compositions.

In order to refine such pre-existing models and thus improve their accuracy in the viscosity prediction, it is important to keep measuring natural samples. The present viscometry data could then be used to improve such models.

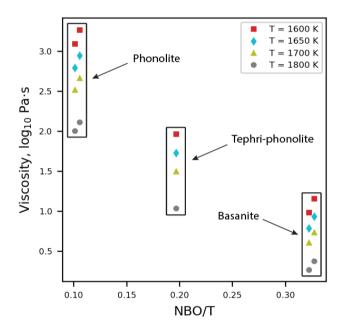
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## 2 4.3. <u>Polymerization and viscosity</u>

433 The relationships between the super-liquidus viscosity and melt polymerization, in terms of T-434 O-T bridging as quantified by the chemically-derived, oxidized NBO/T (Eq. 1), are illustrated 435 in Figure 7. At given temperature, basanite melts show the lowest viscosity ( $< 1.2 \log Pa \cdot s$ ) 436 and the highest NBO/T (0.32 to 0.33) whereas phonolite melts have the highest viscosity (> 2 log Pa $\cdot$ s) and lowest NBO/T (0.10 and 0.11) (Fig. 7). Accordingly, the tephri-phonolite sample, 437 438 having an intermediate chemical composition (Fig. 2 and Table 2), falls in between the basanite and phonolite compositions (Fig. 7). The fact that the highest viscosity data are obtained for 439 440 the samples with the lowest NBO/T, and inversely, is an expected pattern (Mysen et al. 1982,

441 1985; Scarfe et al. 1987; Mysen and Richet 2019). A higher degree of polymerization (lower 442 NBO/T) will lead to lesser solutions for performing the cooperative rearrangements of the 443 molecular sub-units necessary for melt viscous flow. This results, according to the Adam-Gibbs 444 theory of viscous flow, in a lower melt configurational entropy and, hence, in a higher melt 445 viscosity (Adam and Gibbs, 1965; Richet, 1984). Such variations in melt polymerization with 446 composition, albeit expected, may have an important impact on the eruptive style (see 447 Implications).



448

**449** *Figure 7.* Variations of the viscosity  $(log_{10} Pa \cdot s)$  at a given temperature relative to composition 450 as represented by the ratio of non-bridging oxygens to tetrahedrally-coordinated cations 451 (*NBO/T*). See also *Table 2* and *3* for chemical compositions and viscosity results, respectively. 452

453

#### 3 *4.4. <u>Influence of iron redox state and volatile concentration on melt viscosity</u>*

454 All the presently reported experiments were performed in air. In addition, we measured the 455 viscosity of volatile-free melts, as they degassed upon preparation. Therefore, to provide 456 accurate viscosity estimates of the basanite, tephri-phonolite and phonolite melts in a natural 457 context, we should take into account the influence of both water and iron oxidation state on the viscosity of the silicate melts. The reduction to Fe<sup>2+</sup> can lead to a decrease in melt viscosity of
0.2 - 0.5 log units at super-liquidus conditions, which can reach up to 1.5 log units at
undercooled temperature conditions (Dingwell and Virgo 1987; Dingwell 1991; Liebske et al.
2003; Drucwrr 2007; Chevrel et al. 2013). Water also strongly participates in lowering the
viscosity of alkaline magmas (e.g., Whittington et al., 2000, 2001).

To quantify the effect of water, we here follow the methodology of Le Losq et al. (2015a) who calculated the relative effect of water on the B and C terms of the VFT equation (**Eq. 3**) for phonolite melts, based on previously published data from Whittington et al. (2001). In that way we use the following equation:

467

468 
$$log_{10} \eta = A + (B^{anh} + K_1 * C_{H2O} + K_2 * C_{H2O}^2) / (T - (C^{anh} + K_3 * CH_2O + K_4 * C_{H2O}^2)), (4)$$

469

470 where K<sub>1</sub>, K<sub>2</sub>, K<sub>3</sub> and K<sub>4</sub> are the parameters of the polynomial functions that describe the effect of the water concentration in wt%, C<sub>H2O</sub>, on the value of the parameters B and C. B<sup>anh</sup> and C<sup>anh</sup> are the VFT B 471 472 and C parameters of the anhydrous melt. Regarding the basanite composition no viscosity 473 measurements at undercooled temperatures were obtained for this study. Hence, we calculated the VFT 474 parameters of the anhydrous melt by combining our high-temperature data with the low- temperatures 475 data from Whittington et al. (2000) obtained for an iron-free basanite melt (Table 4). To calculate the 476 K<sub>1</sub>, K<sub>2</sub>, K<sub>3</sub> and K<sub>4</sub> parameters, we then used then used the VFT parameters published by Whittington et 477 al. (2000) for a hydrous iron-free basanite. Table 5 reports the  $K_1$ ,  $K_2$ ,  $K_3$  and  $K_4$  parameters from Le 478 Losq et al. (2015a).

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- 481
- 482
- 483

- *Table 4.* Parameters obtained for Arrhenius relation at high-temperature (*Eq. 2*) and VFT
- *parameter over the entire temperature range* (*Eq. 3*). *The VFT parameters calculated for the*
- 486 basanite DR10 result from a combination of the high temperatures data from this study and

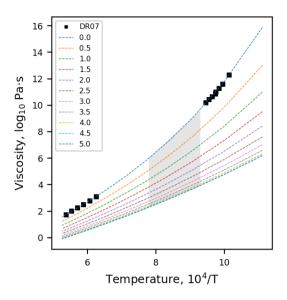
Temperature range	Viscosity range	A <sub>Arr</sub>	EaArr	RMSEArr	AVFT	$B_{\rm VFT}$	$C_{VFT}$	RMSEVFT
(K)	(log10 Pa·s)		(kJ mol <sup>-1</sup> )					
1600 - 1855	0.99 - 0.11	-5.34	-44.37	0.003	-	-	-	-
1600 - 1810	1.16 - 0.34	-5.6	-46.57	0.015	-3.5	4448	645	0.05
1600 - 1860	1.97 - 0.89	-5.8	-48.23	0.011	-	-	-	-
1500 - 1855	3.89 - 1.91	-6.46	-53.72	0.008	-	-	-	-
990 - 1860	12.30 - 1.75	-	-	-	-3.9	7572	517	0.04
	(K) 1600 - 1855 1600 - 1810 1600 - 1860 1500 - 1855	(K)         (log <sub>10</sub> Pa·s)           1600 - 1855         0.99 - 0.11           1600 - 1810         1.16 - 0.34           1600 - 1860         1.97 - 0.89           1500 - 1855         3.89 - 1.91	(K)         (log10 Pars)           1600 - 1855         0.99 - 0.11         -5.34           1600 - 1810         1.16 - 0.34         -5.6           1600 - 1860         1.97 - 0.89         -5.8           1500 - 1855         3.89 - 1.91         -6.46	(K)         (log10 Pars)         (kJ mol <sup>-1</sup> )           1600 - 1855         0.99 - 0.11         -5.34         -44.37           1600 - 1810         1.16 - 0.34         -5.6         -46.57           1600 - 1860         1.97 - 0.89         -5.8         -48.23           1500 - 1855         3.89 - 1.91         -6.46         -53.72	(K)         (log <sub>10</sub> Pa·s)         (kJ mol <sup>-1</sup> )           1600 - 1855         0.99 - 0.11         -5.34         -44.37         0.003           1600 - 1810         1.16 - 0.34         -5.6         -46.57         0.015           1600 - 1860         1.97 - 0.89         -5.8         -48.23         0.011           1500 - 1855         3.89 - 1.91         -6.46         -53.72         0.008	(K)         (log <sub>10</sub> Pa·s)         (kJ mol <sup>-1</sup> )           1600 - 1855         0.99 - 0.11         -5.34         -44.37         0.003         -           1600 - 1810         1.16 - 0.34         -5.6         -46.57         0.015         -3.5           1600 - 1860         1.97 - 0.89         -5.8         -48.23         0.011         -           1500 - 1855         3.89 - 1.91         -6.46         -53.72         0.008         -	(K)         (log10 Pa·s)         (kJ mol <sup>-1</sup> )           1600 - 1855         0.99 - 0.11         -5.34         -44.37         0.003         -         -           1600 - 1810         1.16 - 0.34         -5.6         -46.57         0.015         -3.5         4448           1600 - 1860         1.97 - 0.89         -5.8         -48.23         0.011         -         -           1500 - 1855         3.89 - 1.91         -6.46         -53.72         0.008         -         -	(K)         (log <sub>10</sub> Pa·s)         (kJ mol <sup>-1</sup> )           1600 - 1855         0.99 - 0.11         -5.34         -44.37         0.003         -         -         -           1600 - 1855         0.99 - 0.11         -5.64         -46.57         0.015         -3.5         4448         645           1600 - 1860         1.97 - 0.89         -5.8         -48.23         0.011         -         -         -           1500 - 1855         3.89 - 1.91         -6.46         -53.72         0.008         -         -         -

*low-temperatures data from Whittington et al. (2000) (see Discussion).* 

*Table 5. K* parameters for estimating the effect of water on the parameters B and C of the VFT

491 equation (*Eq. 4*) for the phonolite (from *Le Losq et al. 2015a*) and the basanite melts.

Parameter	Values for phonolite DR07	Values for basanite melts
K1	-455.52	403.15
$\mathbf{K}_2$	32.626	-133.87
$K_3$	-110.61	-140.77
$K_4$	13.241	32.043



#### 494

495 Figure 8. Viscosity (log10 Pa·s) as a function of inverse temperature (K<sup>-1</sup>) for the DR07
496 phonolite. Measured viscosity values (squares) are shown together with values predicted
497 (curves) by Eq. 4 for different water concentrations (wt %). Numbers in the label refer to water
498 content in wt% and the expected magmatic conditions are indicated by the grey box. The 4.5
499 and 5.0 wt% water curves are superposed. Error bars are smaller than the symbol size.

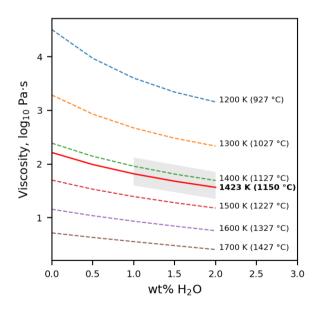


Figure 9. Isothermal viscosity as a function of water concentration (wt%) for the DR10
basanite. The predicted magmatic conditions proposed by Berthod et al., (2021b) are indicated
by the red line (1150 °C) and its uncertainty by the grey box (± 50 °C).

We present the effect of water on the phonolite melt (DR07) in Figure 8 and on the basanite 504 505 melt (DR10) in Figure 9. As expected, the water strongly lowers the viscosity for both melts. 506 The addition of 1 wt% of water reduces the viscosity from 2.50 to 1.68 log<sub>10</sub> Pa·s at 1700 K 507 for the DR07 phonolite. The decreasing effect of water on viscosity is higher at lower 508 temperatures, where the difference between anhydrous and hydrous melts may reach up to 4 509 orders of magnitude at 900 K (Fig. 8). In contrast, the addition of 1 wt% of water decreases the 510 viscosity of the basanite melt by almost 0.4 log<sub>10</sub> units at 1700 K (Fig. 9). These results in 511 agreement to those reported by Whittington at al. (2000, 2001). Hence, our results confirm that 512 (i) further addition of water results in a progressively less important decrease in viscosity and 513 (ii) the effect of water is more effective at reducing the viscosity of polymerized melts 514 (phonolites) than for depolymerized melts (basanites). As it has been previously demonstrated, 515 the chemical composition of the melt affects water solution mechanisms, resulting in different 516 effects on melt structure (e.g., Whittington et al. 2000, 2001; Xue and Kanzaki 2004, 2006; 517 Cody et al. 2005; Mysen and Cody 2005; Mysen and Richet 2005; Giordano et al. 2009; Le 518 Losq et al. 2015b). For polymerized melts, a common and simple water solution mechanism is 519 the reaction of water molecules with the silicate network, breaking T-O-T bonds (with T = Si, 520 Al) (e.g., see Mysen and Richet 2019; Le Losg et al. 2015b). In contrast, for depolymerized 521 melts, silicate network and network modifiers (e.g., Ca, Mg) may be implied into the 522 mechanism leading to slight change of the melt polymerization (Xue and Kanzaki 2004; 523 Moretti et al. 2014; Le Losq et al. 2015b). For the present basanite and phonolite melts that 524 present relatively low NBO/T, the dissolution of water is expected to result in melt 525 depolymerization, but a stronger effect is expected for phonolites as the mechanism implying 526 reaction between T-O-T bonds and water should be more active. This can explain observations 527 made on viscosity (Figs. 8, 9).

528 Thus, the reported values for the viscosity of the silicate melts represent upper limits for529 aluminosilicate melts due to the effect of water combined with the iron oxidation state.

- 530
- 531

#### 4.5. <u>Comparison of phonolites viscosities from different areas</u>

We can compare the viscosity of the phonolites from Mayotte (DR07 and DR17) to data from 532 533 previous studies on similar Al- and alkali-rich compositions. For this, the viscosity 534 measurements of the phonolitic Erebus (Le Losg et al. 2015a) and Vesuvius GP79 (Giordano 535 et al. 2009; Le Losq et al. 2015a) magmatic melts have been selected (Fig. 3). The viscosity of 536 the Vesuvius GP79 sample is lower by an order of magnitude compared to the phonolite from 537 the East-Mayotte submarine volcanic chain. This may be explained by a lower SiO<sub>2</sub> content 538 (54 wt% against 59 wt%). Despite a similar composition in SiO<sub>2</sub> and alkali elements (Fig. 2 539 and Table 2), the viscosity of DR07 differs from the Erebus one by about 0.5 orders of 540 magnitude. The phonolites of Mayotte (DR07 and DR17) are richer by 1 to 2 wt% of iron 541 compared to the Erebus sample (Table 2). In contrast to Chevrel et al. (2013), a decrease in 542 melt viscosity with increasing iron concentration is not observed. The observed viscosity 543 difference more probably finds its origin in variations in the concentrations of a few elements 544 such as K, Mg and Ca between the phonolites of Mayotte and Erebus. Indeed, the DR07 and 545 DR17 samples are slightly more enriched in K<sub>2</sub>O (Table 2). In aluminosilicate compositions, 546 addition of  $K_2O$  can lead to decreasing the melt configurational entropy and so, to increasing 547 its viscosity at constant temperature (Le Losq et al. 2013, 2017, 2021; Robert et al. 2019). 548 Therefore, the difference in K<sub>2</sub>O between the Erebus and Mayotte phonolites could explain the 549 observed viscosity distinctions. In addition, the Erebus sample contains a bit more alkaline-550 earth elements (Mg and Ca), that may act as network modifiers and favor the creation of nonbridging oxygen (NBO) atoms and, hence, a decrease in viscosity (Mysen et al. 1980; Mysen 551 552 1995; Richet et al. 1984; Stebbins et al. 1992; Stebbins and Xu 1997). Hence, it seems

553 consistent that the DR07 phonolite from Mayotte has a higher viscosity than the Erebus sample. 554 As explained previously, no viscosity measurements at undercooled temperatures could be 555 obtained for the DR17 phonolite. However, according to the viscosity at the super-liquidus 556 temperatures and the enrichment in alkali (Figs. 2 and 3) we may assume that the viscosity at 557 undercooled temperatures will be higher than the DR07, Erebus and Vesuvius melts.

558

## 559 5. Implication on eruption dynamics

560 Upon magma ascent in a conduit, the style of a volcanic eruption depends on whether the 561 magma fragmentation threshold is crossed (explosive) or not (effusive). This threshold will 562 depend on the magma viscosity but also on the ascending rate and bubble overpressure (e.g., 563 Dingwell et al. 1996; Papale 1999; Ma et al. 1999; Pistolesi et al. 2011; Gonnermann et al. 564 2011; Gonnermann 2015). Generally, in magmas with high viscosity (silica-rich composition), 565 the enhanced resistance to bubble expansion favors bubble overpressure and reduces the ability 566 of volatiles to stream out of the melt. This in turn promotes more efficient magma 567 fragmentation and a greater explosivity of eruptions. Of course, many exceptions break such 568 general rules, such as basaltic Plinian eruptions (e.g., Moitra et al. 2018) and Si-rich lava flows (e.g., Fink 1983; Farquharson et al. 2015; Prival 2021). Additionally, eruption style can switch 569 570 between effusive and explosive activities during an eruption due to variations in magma 571 composition, degassing, and crystallization that all drive magma viscosity (e.g., Vesuvius 572 Giordano et al. 2009; Myers et al. 2021, Andújar and Scaillet 2012; Popa et al. 2021). Attempts 573 have been done to define the viscosity threshold to discriminate explosive to effusive eruptions, 574 but no consensus has yet been achieved (Papale 1999; Di Genova et al. 2017; Wadsworth et al. 575 2018).

576 Our results show that basanite melts have the lowest viscosity of the series followed by the 577 tephri-phonolite and the phonolite melts and that the presence of water reduces the viscosity. 578 Assuming that the eruptive temperature (T<sub>e</sub>) of the Mont Fani Maoré basanite lavas is around 579 1150 °C (Berthod et al., 2021b), and that water content could range from 1.0 to 2.3 wt% 580 (Berthod et al. 2021b), we thus expect lava viscosity upon eruption to range from 1.5 to 2.5 581  $\log_{10}$  Pa s (Fig. 8). Such a viscosity range is close to, for example, the viscosity of subaerial 582 pahoehoe basaltic lavas from Hawaii (Chevrel et al. 2018) or submarine basaltic lava flows 583 (McClinton et al. 2014). This low viscosity range is therefore consistent with the effusive 584 eruption style and morphologies (pillow lavas and pahoehoe lavas) of the lava flow field as 585 observed at Mont Fani Maoré (Feuillet et al., 2021; Berthod et al. 2021b). However, several 586 questions remain. Why did the basanite melt come out from the Mont Fani Maoré rather than 587 ascend upwards to erupt in the Horseshoe site, where its hypothesized magmatic storage is 588 located (Lemoine et al. 2020; Feuillet et al. 2021)? Does the low viscosity of the basanite melt favored a lateral pathway? Could the low viscosity favors high flux rates ( $150 - 200 \text{ m}^3 \text{ s}^{-1}$ 589 590 averaged over the first year, Berthod et al. 2021b) and the long duration eruption if the volume 591 was there?

592 Around the Horseshoe volcanic site of the Mayotte volcanic chain, a large diversity of edifice 593 morphologies and deposits textures corresponding to both phonolitic effusive and explosive 594 eruptions have been observed (REVOSIMA, 2022; Puzenat et al. 2022; Gurioli et al. 2023; 595 Komorowski et al. 2023). Such diversity can be explained by the fact that, depending on magma 596 ascent rates, eruptive temperatures and initial water contents, phonolite magmas can be 597 involved in a broad range of magmatic effusive and explosive activity (e.g., see Andújar and 598 Scallet 2012). Indeed, the Mayotte phonolite magmas evolved from basanite magma by  $\sim 80\%$ 599 of fractional crystallization (Berthod et al. 2021a). They are expected to present a high viscosity 600 at expected eruption temperatures (typically in the 800-1000 °C range, e.g. see Andújar and 601 Scaillet 2012). However, phonolite magmas are also expected to contain a large amount of dissolved water. If assuming a phonolite melt stored in a deep magma chamber at  $T_e = 800$  -602

603 1000 °C and containing ~5 wt% water (Andújar and Scaillet 2012), this magma could have a 604 viscosity as low as  $2.0 - 3.2 \log_{10} \text{Pa} \cdot \text{s}$  (Fig. 8), not that far from that previously estimated for 605 the basanite melt. However, upon migration toward the surface, vigourous water exsolution is 606 expected to occur. According to the composition of the residual glass of the submarine 607 phonolites from Mayotte (encountered at 1,300 to 1,600 m b.s.l.), the residual water content in 608 erupted phonolite glass ranges between 0.8 and 1.2 wt% (Thivet, personal communication). 609 Considering such values, the viscosity of the phonolite melts may increase of up to 4 to 7 orders 610 of magnitudes upon ascent (Fig. 8). In parallel, the significant water exsolution will lead to the 611 formation of a magmatic foam of low density in the conduit, reinforcing the buoyant force that 612 pushes the magma out toward surface. This effect is particularly promoted in silica-rich melts 613 like phonolites, because their high viscosity favors entrapment of the bubbles (Thomas et al. 614 1994; Gardner et al. 1996). Therefore, exsolution of water from such magmatic liquids often 615 results in foaming, largely enhancing the probability of brittle fragmentation of the foam (Jones 616 et al. 2019; Scheu and Dingwell 2022) in the conduit (e.g., Dingwell, 1996; Papale 1999; 617 Gonnerman, 2015). Therefore, the amount of water initially stored in the chambers will 618 determine the eruptive styles. The occurrence of both effusive and explosive phonolite 619 eruptions at Mayotte, testified by the observed deposits and products, indicate that a broad 620 range of storage conditions and degassing history exist along this volcanic chain, leading to 621 open questions regarding the future events that could imply phonolite melts.

622

## 623 6. Conclusion

This study provides precise quantification of the temperature-viscosity relationship for basanite to phonolite melts through a large temperature range and discusses the effect of dissolved water on melt viscosity. We show that at eruption temperatures basanite lavas are more propitious to erupt in an effusive manner while phonolite may erupt effusively or explosively. However, we

628 do not account for the effect of bubbles and crystals on magma viscosity. Future studies should 629 accounted for their effect to provide a holistic view of magma rheology which directly impacts 630 the dynamics and eruption style as well as lava flow emplacement. For this, detailed sample 631 texture analyses should be undertaken on collected samples to quantify the bubble and crystal 632 content, shape, size and distribution that are necessary to constrain magma suspension rheology 633 (see Harris and Allen 2008; Mader et al. 2013; Kolzenburg et al. 2022). Measuring the 634 viscosity of magmatic liquids along the Mayotte alkali magmatic differentiation trend 635 combined with known volatile content, and crystal and bubbles characteristics, is essential to 636 quantify the evolution of magma viscosity during ascent rate, outgassing and crystallization, 637 which directly influence the prevailing conditions determining the eruptive style (whether it is 638 effusive or explosive).

639

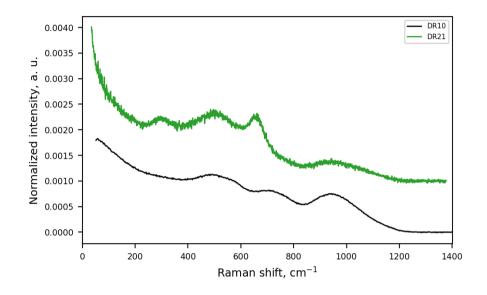
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## 678 Appendix

- 679 *Figure A1.* Uncorrected Raman spectra of (a.) DR01 basanite (black curve) and (b.) DR21
- 680 tephri-phonolite (green curve) products. No Raman Boson peaks are observed on these

681 samples between 50 and 80 cm<sup>-1</sup>.



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