

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30

4841 Revision 2

A general model for calculating the viscosity of natural iron-bearing silicate melts over a wide range of temperature, pressure, oxygen fugacity and compositions

Xianzhe Duan^{1,2*}

¹School of Nuclear Resource Engineering, University of South China, Hengyang, Hunan 421001, China

²State Key Laboratory of Lithospheric Evolution, Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing 100029, China

* Corresponding author (duanxianzhe@126.com , and duanxianzhe@163.com)

31

32

ABSTRACT

33

34

35

36

37

38

39

40

41

42

43

A new general model that takes into account the pressure and redox state effect is presented to calculate melt viscosities of natural Fe-bearing melts. This new model is applicable to melts that span a wide range of temperatures (from 733 K to 1873 K), pressures (0.001-15 kbar), H₂O content (from 0 wt% to 12.3 wt%), and compositions (from ultramafic, mafic to silicic melts). The accuracy of the model is calculated to be ± 0.23 log units of viscosity, which is within or close to experimental uncertainty. The transport properties, including glass transition temperature and melt fragility, can also be calculated from this model. A spreadsheet to calculate the viscosity is provided in an Electronic Supplement.

Keywords: Melt viscosity; Fe-bearing melts; Oxygen fugacity; Iron oxidation state; Glass transition temperature; Melt fragility; Volcanic eruption

44

45

INTRODUCTION

46

47

48

49

50

51

52

53

54

55

56

57

58

59

60

61

62

63

64

65

The viscosity of silicate melts is one of the most important physical properties governing magmatic processes such as the generation, migration, degassing, crystallization and eruption of magmas (e.g., Shaw, 1972; Hui and Zhang, 2007; Giordano et al., 2008). It is well known that the main parameters that control the viscosity of melts are temperature and melt composition (including water) (e.g., Shaw, 1963, 1972; Kushiro, 1976, 1978b, 1986; Persikov, 1991; Baker, 1996; Schulze et al., 1996; Stevenson et al., 1996; Richet et al., 1996; Stevenson et al., 1998; Zhang, 1999; Whittington et al., 2000, 2001; Romano et al., 2003; Zhang et al., 2003; Dingwell et al., 1996, 2000, 2004; Hui and Zhang, 2007; Ardia et al., 2008; Hui et al., 2009; Giordano et al., 2003, 2006, 2008; Vetere et al., 2007, 2008; Misiti et al., 2006, 2011; Bartels et al., 2012; Chevrel et al., 2013). In general, the melt viscosity has non-Arrhenius temperature dependence, and can be significantly reduced by H₂O, which is the most abundant fluid in the upper mantle and crust. In addition to temperature and composition effects, the pressure and redox state (i.e., iron oxidation state or oxygen fugacity) may have important effects (Mysen et al., 1980; Liebske et al., 2003; Mysen and Richet, 2005; Ardia et al., 2008; Hui et al., 2009; Bartels et al., 2012). For instance, Mysen et al. (1980) suggested that it was necessary to consider the pressure effect during the magma ascent from upper mantle to Earth surface, and Liebske et al., (2003) reported that the viscosities of dry andesitic melts could decrease by about 1.6 log units with increasing Fe²⁺/Fe_{tot} from 0.42 to 0.79. In addition, oxygen fugacity controlling the iron oxidation state has been widely recognized to

66 play an important role in the structure of melt, thereby significantly affecting melt viscosity
67 (Cukierman and Uhlmann, 1974; Liebske et al., 2003; Mysen and Richet, 2005; Vetere et al.,
68 2008). Due to the importance of viscosity in geological processes, many experiments on the
69 viscosity of melts have been performed over the past half century (e.g., Shaw, 1963; Kushiro,
70 1986; Persikov, 1991; Toplis et al., 1994; Stevenson et al., 1995; Richet et al., 1996; Giordano
71 et al., 2003b; Liebske et al., 2003, 2005; Vetere et al., 2007, 2008; Chevrel et al., 2013).
72 However, these data are far from sufficient for geological applications over a wide T - P - X
73 range of Earth's deep interior due to the inconsistencies within and between the published
74 data sets, particularly at the high T - P range where the data are scattered due to the difficulty in
75 experimental technique (Avramov, 2007; Ardia et al., 2008; Hui et al., 2009). Therefore, many
76 researchers have devoted extensive efforts to the modeling of viscosity in melts in order to
77 interpolate between data points or extrapolate beyond the data range (e.g., Shaw 1972; Baker
78 et al., 1996; Richet et al., 1996; Schulze et al., 1996; Scaillet et al., 1996; Hess and Dingwell.,
79 1996; Zhang et al., 2003; Misiti et al., 2006, 2011; Vetere et al., 2006, 2007, 2008; Avramov,
80 2007; Hui and Zhang, 2007; Giordano et al., 2003, 2006, 2008; Ardia et al., 2008; Hui et al
81 2009) (see the Electronic Supplement for detail).

82 In this study, I provide a model in which pressure effect and iron oxidation state effect on
83 viscosity have been taken into account. First, the available data are reviewed in Section 2. The
84 framework of the model is presented in Section 3. Then this model is compared with the
85 experimental data with the previous models (Section 4), where evaluation of the model
86 accuracy was performed by the root-mean-square-error (abbreviated as RMSE, hereafter)
87 between the measured and calculated viscosity data, which is defined as follows:

88
$$RMSE = \sqrt{\frac{\sum_{i=1}^n (\log_{10} \eta_{cal}^i - \log_{10} \eta_{exp}^i)^2}{n}} \quad (1)$$

89 where η_{cal}^i and η_{exp}^i are the i^{th} calculated and measured viscosity data in Pa s, respectively;
90 n is the number of the data. As can be seen from Eqn. (1), RMSE can be used to measure the
91 goodness of prediction of certain model of interest. Section 5 contains a discussion of the
92 viscosity behavior of natural melts, with the complicated effects exerted by temperature,
93 pressure, melt composition (including water), and redox state.

94 The new model can predict viscosities of various natural Fe-bearing melts from 733 K to
95 1873 K and from 0.001 kbar to 15 kbar with accuracy within or close to experimental data
96 (e.g., RMSE of 0.23 log units).

97

98 REVIEW OF VISCOSITY DATA

99

100 Over the last half century, experimental scientists have carried out extensive
101 measurements of viscosity of a large variety of natural Fe-bearing silicate melts (from
102 ultra-mafic, mafic and to silicic) over a large temperature-pressure range (Fig. 1).

103 In constructing the literature database for this model, the data used for calibration meet
104 the following minimal criteria: (1) experiments must be on natural silicate melt compositions,
105 (2) complete compositions of melts must have been determined, (3) the redox state (i.e., iron
106 oxidation state or oxygen fugacity) should be known, (4) the melts need not to experience a
107 large variation in the iron oxidation state or the crystallization during the viscosity
108 measurements. Among the experimental measurements for Fe-bearing melts (Fig. 1), some

109 fulfill these requirements (Shaw, 1963; Kushiro, 1986; Persikov, 1991; Toplis et al., 1994;
110 Stevenson et al., 1995; Richet et al., 1996; Giordano et al., 2003b; Liebske et al., 2003, 2005;
111 Vetere et al., 2007, 2008; Chevrel et al., 2013), with the following exceptions: (1) several data
112 points of Giordano et al. (2003b) measured for basaltic melts at temperatures above 1670 K
113 are inconsistent with that of Toplis et al. (1994) with a deviation more than 1 log units; (2)
114 several data points of Liebske et al. (2003) collected during heating are not consistent with
115 their data collected during cooling; (3) the data points of Chevrel et al. (2013) where the melt
116 compositions used were distinctly different from those of natural melts. These small amounts
117 of experimental data are not used for parameterization.

118 The experimental data (Labeled as “Data_redox_state_known”, hereafter) of this
119 database compiled for anhydrous to hydrous Fe-bearing melts covers more than 14.5 orders of
120 magnitude of viscosity ($10^{-1.72}$ - $10^{12.8}$ Pa s), showing near-Arrhenian to non-Arrhenian
121 temperature dependence, includes more than 220 measurements on more than 20 melt
122 compositions spanning the *T-P* range of 733-2523 K and 0.001-130 kbar. In this database,
123 Liebske et al. (2005) provide the only data for pressures above 15 kbar (i.e., 28-130 kbar).
124 This data set was not used for calibration, but instead for a check of the model predictability.
125 Two sets of viscosity on andesitic melts (Liebske et al., 2003) and shoshonitic melts (Vetere et
126 al., 2007) are also not included in the parameterization, but are also chosen to test whether
127 this model can predict data not in the fit (Table 1).

128 Some data (Labeled as “Data_redox_state_unknown”, hereafter) with unknown redox
129 states (Neuville et al., 1993; Stevenson et al., 1996; Alidibirov et al., 1997; Gottsman et al.,
130 2002; Romano et al., 2003; Misiti et al., 2006, 2011) are chosen for checking the

131 predictability of this model in the case where the redox state effect is not under consideration,
132 compared with the H-Z model (Hui and Zhang, 2007) and G-R-D model (Giordano et al.,
133 2008) that are mostly popularly used.

134

135 **MODEL FORMULATION AND PARAMETERIZATION**

136

137 Integrating the strengths of the previous models, the effects of melt composition and iron
138 oxidation on melt viscosity were evaluated in this study by a parameter named VSM, which is
139 a modified version of the SM parameter of Giordano et al. (2003a). SM is a ‘network
140 modifiers’ parameter as the molar percentage oxide sum, as defined as the following equation
141 (Eqn. (2)), and was used as a compositional parameter to calculate anhydrous
142 multi-component melt viscosity.

$$143 \quad SM = \sum(Na_2O + K_2O + CaO + MgO + MnO + FeO_{tot} / 2) \quad (2)$$

144 where FeO_{tot} is the total iron content.

145 However, iron can exist in both divalent and trivalent states in melts. Divalent iron can
146 act as a network modifier, while trivalent iron can act as a network former, like Si^{4+} , Ti^{4+} , Al^{3+} ,
147 P^{5+} (Persikov et al., 1990; Dingwell et al., 1993; Toplis and Dingwell, 1996; Ottonello et al.,
148 2001; Moretti, 2005; Mysen and Richet, 2005). I therefore modified the SM expression by
149 distinguishing between the two different valences and roles of iron in silicate melt, treating all
150 ferric iron as a network former and all ferrous iron as a network modifier. If the iron oxidation
151 state is not given, the method of Moretti (2005) is used to calculate it under the given
152 T - P - X - fO_2 condition. There are some various empirical forms have been proposed to

153 quantitatively calculate the iron oxidation state as a function of oxygen fugacity, pressure,
154 temperature and compositions of the melt (Sack et al., 1980; Kilinc et al., 1983; Kress and
155 Carmichael, 1991). Later work attempted to include the effect of water (Moore et al., 1995),
156 while the most recent attempt to predict iron oxidation state including the effect of water has
157 been presented by Moretti (2005), which is the extension of Ottonello et al. (2001). The
158 method of Moretti (2005) for the determination of the iron oxidation state is mainly based on
159 the following three assumptions: (1) iron oxidation state is dominantly controlled by five
160 variables (oxygen fugacity, pressure, temperature, melt composition, water); (2) FeO can
161 produce Fe^{2+} and O^{2-} (Flood and Forland, 1947) and H_2O can be dissociated to H^+ and O^{2-}
162 (Fraser, 1975, 1977); (3) Fe_2O_3 behaves as an amphoteric oxide in the Lux-Flood acid-base
163 acceptance, i.e., Fe_2O_3 in the melt can be double dissociated into Fe^{3+} (basic dissociation) and
164 FeO^{2-} anions (acidic dissociation) (Fraser, 1975, 1977; Ottonello et al., 2001). With the
165 calculated mole fractions of Fe^{2+} and Fe^{3+} in the silicate melt, I replace the total Fe in the SM
166 expression Eq. (2), by the mole fraction of Fe^{2+} and added the oxides such as SiO_2 , TiO_2 ,
167 Al_2O_3 , Fe^{3+} and P_2O_5 to the denominator to create the VSM parameter.

$$168 \quad VSM = \frac{\sum(Na_2O + K_2O + CaO + MgO + MnO + Fe^{2+})}{\sum(SiO_2 + TiO_2 + Al_2O_3 + Fe^{3+} + P_2O_5)} \quad (3)$$

169 where Fe^{2+} and Fe^{3+} are the relative proportion of ferrous and ferric iron in the total iron,
170 respectively; VSM represents the enrichment of non-bridging oxygen atoms in the melt
171 system. Non-bridging oxygen is related to the depolymerization of melts, which in turn affects
172 the amount of free oxygen in the melts and thus influences the viscosity (Mysen and Richet,
173 2005).

174 The VSM value calculated from the anhydrous melt composition was chosen to

175 characterize the melts. A single value parameter of melt composition, NBO/T, was also used
176 by Giordano et al. (2003a) to model the viscosity of anhydrous melts, while the researchers
177 also used the SM to describe the melt composition in their models (e.g., Giordano et al.,
178 2003a, 2006). The use of individual oxides in models creates additional fitting parameters,
179 typically more than 10; additional model parameters results in better fitting of the model to
180 the calibration data. However, models with many parameters can sometimes produce
181 inaccurate interpolations and extrapolation (Kenny and McCoach, 2003). Because of this
182 possibility, the VSM parameter is used to characterize anhydrous melt composition and
183 produces a model that provides accurate predictions of viscosity for most common,
184 Fe-bearing melts.

185 In spite of the importance of water in affecting the structure and properties of silicate
186 melts, the relative abundance of the species of it dissolved in melts both as hydroxyl groups
187 and molecular species, appears to be a complex function of at least temperature and melt
188 composition that currently defy accurate modeling (e.g., Silver and Stolper, 1989; Silver et al.,
189 1990; Zhang, 1999; Mysen and Richet, 2005). For the purpose of creating a simple-to-use
190 model, a method is needed to reflect the influence of water on the viscosity that does not rely
191 upon experimental measurements of the water species in the melts, but yet adequately
192 quantifies the effect of water on the viscosity. After considering many different methods, I
193 incorporated the effect of water by the product of VSM and the mole fraction of total water in
194 the melts.

195 Based on the Vogel-Fulcher-Tammann equation (abbreviated as VFT equation, hereafter)
196 which expresses the non-Arrhenian temperature dependence of viscosity (Vogel, 1921;

197 Fulcher, 1925; Tammann and Hesse, 1926; Russell et al., 2003), the following equation (Eqn.
198 (4)), which most accurately reproduces the data, is proposed in this study:

$$\log \eta = A + \frac{B}{T - C}$$

$$A = a$$

$$B = b_1 + b_2 \ln(JB)$$

$$C = c_1 + c_2 \ln(JC)$$

$$\begin{aligned} 199 \quad JB &= b_3^2 + VSM(b_4^2 VSM + 2b_3b_4 + b_4X_{H_2O} + b_4b_5P) \\ &\quad + X_{H_2O}(2b_3 + X_{H_2O} + b_3P + b_4VSM) \\ &\quad + P(2b_3b_5 + b_5^2P + b_4b_5VSM + b_5X_{H_2O}) \\ JC &= c_3^2 + VSM(c_4^2 VSM + 2c_3c_4 + c_4X_{H_2O} + c_4c_5P) \\ &\quad + X_{H_2O}(2c_3 + X_{H_2O} + c_3P + c_4VSM) \\ &\quad + P(2c_3c_5 + c_5^2P + c_4c_5VSM + c_5X_{H_2O}) \end{aligned} \quad (4)$$

200 where T is in Kelvin, P is in kbar; η is viscosity in Pa s; The values of parameters a, b₁-b₅
201 and c₁-c₅ are listed in Table 2. X_{H₂O} is molar fraction of the volatile (H₂O) in melts on a single
202 oxygen basis as defined below (Eqn. (5)) (Stolper, 1982; Zhang, 1999).

$$203 \quad X_{H_2O} = (C_{H_2O} / M_{H_2O}) / (C_{H_2O} / M_{H_2O} + (100 - C_{H_2O}) / W) \quad (5)$$

204 where C_{H₂O} and M_{H₂O} are mass percent and mass per mole of H₂O, respectively, W is the mass
205 of dry melt per mole of oxygen.

206 As can be found in Eqn. (4), the viscosity is modeled as the combination of the affects of
207 temperature, pressure, redox state, anhydrous melt composition, and water concentration.

208 The R² value and RMSE for fitting the data used for the calibration of this model (Table
209 1) is 0.996 and 0.23, respectively, indicating that this model reproduces the data within or
210 close to the experimental accuracy. The common value of parameter A indicates that the
211 viscosity at infinite temperature based on the model for various Fe-bearing melts (log
212 $\eta_\infty = -4.75$, negligible compositional dependence) is close to the value of log $\eta_\infty = -4.31 \pm 0.74$

213 obtained by Russell et al. (2003) to be the optimized viscosity limit for silicate melts. With
214 these parameters, the viscosity of natural Fe-bearing silicate melts including anhydrous and
215 hydrous iron-bearing melts can be calculated.

216

217

VALIDATION OF THE MODEL

218

219 More than 10 viscosity models were chosen for comparison in this study (Table 3, which
220 also lists the references denoted by the following model abbreviations).

221 Fig. 2 (a-c) shows the comparisons of the calculated viscosities with this model, H-Z
222 model and the G-R-D model, and the experimental data, including those with known
223 (Data_redox_state_known) and unknown redox states (Data_redox_state_unknown). It should
224 be noted that for proper comparison, the outliers of the H-Z and G-R-D model (Toplis et al.,
225 1994; Richet et al., 1996) were not used in the comparison. In addition, where the information
226 on redox state is absent (Fig. 2a), the contents of FeO and Fe₂O₃ were arbitrarily calculated
227 with the assumption that half of the total iron FeO_{tot} (wt%) is FeO (wt%) and the other half is
228 Fe₂O₃ (wt%); this was also done in cases where the Fe²⁺/Fe³⁺ ratios were known
229 (Data_redox_state_known). Fig. 3 compares the predictions of this model with the
230 experimental data with known redox states (Data_redox_state_known). The experimental
231 measurements of iron oxidation state are taken as inputs for the calculation of viscosities with
232 this model. Only the experimental data within the stated *T-P-X* ranges of models are used in
233 Figs. 2-3.

234 The viscosities calculated from this model are comparable to the H-Z and G-R-D
235 models, and can be applicable in the condition where the redox state effect is unknown (Fig.
236 2). However, the accuracy of previous models for calculating the viscosity of Fe-bearing
237 melts, especially for high iron-bearing melts (e.g., basalt, andesite) (e.g., Toplis et al., 1994;
238 Richet et al., 1996), can be much improved by incorporating redox state effect, as can be seen
239 from Fig. 3.

240 The viscosity model from this study can reproduce the experimental data used in the
241 calibration with a standard deviation of 0.23 log units, and those not used in the calibration
242 within 0.28 log units. These results illustrate the robustness of the model within and beyond
243 its calibration range.

244 Fig. 4 shows the good prediction of this model at high pressures ($P \geq 1$ kbar), with a
245 remarkable accuracy (RMSE=0.27). However, additional experimental data at higher
246 pressures ($P > 15$ kbar) is needed for better constraining the model.

247 Table 3 shows the results of comparisons between the experimental data
248 (Data_redox_state_known) and that calculated by this model, and those of previous general
249 models (e.g., S-B-W model; H-Z model; G-R-D model) and the other specific models in
250 comparison. It can be seen by comparing the different values of RMSE that the model of this
251 study gives better results than the previous models in predicting viscosities of various natural
252 Fe-bearing melts over a wide temperature and pressure range. The S-B-W model produces an
253 RMSE of 0.77 log units when compared to experimentally determined melt viscosities.
254 Moreover, the S-B-W model is only applicable to viscosities below 10^5 Pa s. The comparison
255 between the measurements and the predictions of H-Z model produces the RMSE of 0.7 log

256 units, whereas the comparisons of G-R-D model viscosity predictions with the measurements
257 on various melts yield the RMSE of 0.68 log units. The calculated viscosities of various
258 natural Fe-bearing melts with this model is about 0.5 log units more accurate than previous
259 general models (H-Z model; G-R-D model). In addition, the H-Z and G-R-D models include
260 more parameters (37 and 18, respectively) than this model (11) (Table 3), which may result in
261 less accurate interpolations and extrapolations (e.g., Kenny and McCoach, 2003). The
262 relatively large deviations yielded by both H-Z model and G-R-D model for Fe-bearing melts
263 probably reflects that they did not account for the iron redox state.

264 On the other hand, this model gives comparable results to other models except for
265 shoshonitic melts (Vetere et al., 2007), where the standard deviation of this model is 0.40 log
266 units, which is 0.23 log units higher than the V-B-M model (RMSE of 0.17 log units).
267 However, the V-B-M model used the Vetere et al (2007) data for calibration, whereas in the
268 model presented here the data used as a check.

269 This model is based on the data for Fe-bearing melts, and should not be used for Fe-free
270 melts, where the H-Z or G-R-D models are recommended. In addition, this model is best
271 recommended for Fe-bearing melts with well characterized redox states. If the oxygen
272 fugacity rather than iron oxidation state is given, it is recommended to evaluate the iron
273 oxidation state prior to the determination of melt viscosity according to the method of Moretti
274 (2005), which is the most recent attempt to quantitatively calculate the iron oxidation state as
275 a function of oxygen fugacity, pressure, temperature, melt composition and H₂O. The
276 calculation can be done with the files found in the Electronic Supplement. If neither iron
277 oxidation state nor oxygen fugacity are known, the total FeO content (FeO_{tot}, wt%) could be

278 split equally between FeO (wt%) and Fe₂O₃ (wt%) for the calculation of melt viscosity.

279

280

DISCUSSION

281

The temperature, pressure and compositional effects on the viscosity

283

284 While some workers (e.g., Bottinga and Weill, 1972; Shaw, 1972) in the earlier studies
285 assumed an Arrhenius temperature dependence of melt viscosity, it has been most widely
286 accepted that the viscosities of silicate melts have non-Arrhenian temperature dependence,
287 and that this dependence is more pronounced in the lower temperature range (e.g., Mysen,
288 1988; Richet and Bottinga, 1995; Hess and Dingwell, 1996; Zhang et al., 2003; Mysen and
289 Richet, 2005; Vetere et al., 2006, 2007, 2008; Misiti et al., 2006, 2011; Hui and Zhang, 2007;
290 Giordano et al., 2008).

291

292 The important role that the pressure plays in viscosity was not recognized until 1970s.
293 The pressure dependence of viscosity is complicated, and is still subject of debate (e.g.,
294 Kushiro, 1976, 1978a; Scarfe et al., 1979, 1987; Mysen et al., 1980; Brearley et al., 1986;
295 Gupta, 1987; Bottinga and Richet, 1995; Richet et al., 1996; Schulze et al., 1996, 1999; Mori
296 et al., 2000; Behrens and Schulze, 2003; Reid et al., 2003; Liebske et al., 2003, 2005; Poe et
297 al., 2006; Ardia et al., 2008; Vetere et al., 2006, 2008; Hui et al., 2009; Del Gaudio and
298 Behrens, 2009; Bartels et al., 2012). Kushiro (1976) found that the viscosity of albitic melts
299 could decrease by 1 log units from 5 kbar to 24 kbar in the high temperature range. Kushiro
(1978a) found a similar decrease of viscosity for jadeite with increasing pressure. Gupta

300 (1987) also reported a negative pressure dependence of viscosity in some synthetic melts (e.g.,
301 $\text{Na}_2\text{O}-3\text{SiO}_2$; $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-4\text{SiO}_2$; $\text{K}_2\text{O}-\text{MgO}-5\text{SiO}_2$; $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-6\text{SiO}_2$). Some workers (e.g.,
302 Kushiro, 1976, 1978a; Mori et al., 2000; Ardia et al., 2008) interpreted this remarkable
303 decrease as the consequence of the coordination change of Al in melts, i.e., from four- to
304 six-fold or the decrease in the T-O-T (T=Si, Al) angle weakening the silicate network. Other
305 works (e.g., Scarfe et al., 1979, 1987; Mysen et al., 1980; Brearley et al., 1986; Bottinga and
306 Richet, 1995; Schulze et al., 1999; Behrens and Schulze, 2003; Del Gaudio and Behrens,
307 2009) suggested that the variation in the viscosity was mainly related to the degree of melt
308 polymerization. For instance, Scarfe et al. (1979) noted that the viscosity of highly
309 polymerized melts exhibited a negative pressure dependence, whereas the less highly
310 polymerize melts showed a positive pressure dependence. Scarfe et al. (1987) found that in
311 the low viscosity range, a positive pressure effect existed in some silicate melts with
312 $\text{NBO}/\text{T}>1$, like diopside, whereas most silicate melts with $\text{NBO}/\text{T}<1$, such as andesite,
313 tholeiitic basalt, were characterized by a negative pressure effect. On the other hand, in the
314 high viscosity range, the negative pressure effect on viscosity was also found for polymerized
315 melts such as albitic melts (Schulze et al., 1999; Behrens and Schulze, 2003; Del Gaudio and
316 Behrens, 2009). Del Gaudio and Behrens (2009) observed that at low temperature, the
317 pressure effect appeared to be more important than at high temperature. Nevertheless, Reid et
318 al. (2003) reported an “anomalous” pressure dependence of viscosity for diopside, showing
319 that the viscosity increased from 35 kbar to a maximum at 100 kbar and then decreased at
320 higher pressure. A pressure dependence similar to that reported by Reid et al. (2003) was also
321 observed in Liebske et al. (2005), who reported that the viscosity results for peridotite had a

322 maximum at the pressure (i.e., 85 kbar). This viscosity behavior with pressure was explained
323 to be likely associated with structure changes of melt that occur upon compression (Reid et al.,
324 2003; Liebske et al., 2005; Poe et al., 2006). A minor effect of pressure on viscosity of
325 synthetic rhyolitic to more feldspaitic melts (Schulze et al., 1996; Bartels et al., 2012), but
326 also of iron-free andesite and iron-bearing andesite has been reported (Richet et al., 1996;
327 Liebske et al., 2003; Vetere et al., 2006, 2008). Liebske et al. (2003) found the variation in
328 viscosity of iron-free andesite was less than 0.3 orders of magnitude in the pressure range
329 from 1 kbar to 3 kbar. Vetere et al. (2006) observed no significant dependence of viscosity in
330 hydrous iron-free andesite on pressure in the range of 0.001-5 kbar at high temperature.
331 Vetere et al. (2008) suggested that pressure had a minor influence for hydrous iron-bearing
332 andesite with high water content ($H_2O \cong 2.5$ wt%) in the low viscosity range, although these
333 authors stated that they didn't have experimental pairs where only pressures were changing
334 (identical temperature, melt composition and iron redox state). Fig. 5 shows the prediction of
335 this model on the pressure dependence of viscosity at 1473 K and in the 0.001 kbar-10 kbar
336 pressure range, demonstrating that the pressure has a minor effect on iron-bearing andesitic
337 melts with high H_2O content at high temperature, in good agreement with the pressure trend
338 observed by previous studies (e.g., Liebske et al., 2003; Vetere et al., 2006, 2008). However,
339 when the H_2O decreases, the effect of pressure becomes larger, and cannot be ignored. For
340 instance, the viscosity of iron-bearing andesitic melt with 6 wt% H_2O content, decreases by
341 less than 0.4 orders of magnitude with increasing pressure from 0.001 kbar to 3 kbar, but can
342 decrease by about 0.8 orders of magnitude when H_2O content decreases to about 0.1 wt%. A
343 similar pressure trend can also be found at pressures beyond 3 kbar. Therefore, in spite of

344 smaller influence on viscosity relative to temperature, the pressure effect cannot be neglected
345 to model the viscosity in the geologically relevant conditions. This result is consistent with the
346 point suggested by Mysen et al. (1980) that the pressure effect should be taken into account
347 under the condition during the magma ascent from Earth depth to surface.

348 H₂O is of great importance to viscosity because small variation in H₂O content can cause
349 significant changes in viscosity and thus ultimately influence the mechanisms of
350 transportation and eruption of magma (Shaw, 1963; Kushiro, 1978b; Dingwell et al., 1996,
351 2000; Papale, 1999; Whittington et al., 2000, 2001; Zhang et al., 2003; Morizet et al., 2007;
352 Vetere et al., 2007, 2008; Hui et al., 2009; Misiti et al., 2011; Duan, 2014). It is well
353 recognized that H₂O has a significantly negative effect on viscosities of melts, regardless of
354 the melt types, but the viscosity behavior corresponding to H₂O effect has been hotly debated
355 (e.g., Shaw, 1963; Kushiro, 1978b; Davis and Tomozawa, 1996; Dingwell et al., 1996; Zhang,
356 1999; Whittington et al., 2000, 2001; Zhang et al., 2003; Morizet et al., 2007; Misiti et al.,
357 2011). Shaw (1963) found the addition of 1 wt% water could cause the reduction by two
358 orders of magnitude in the viscosity of initially anhydrous rhyolitic melts. Dingwell et al.
359 (1996) reported that the viscosity of haplogranitic melts dropped drastically with the addition
360 of 0.5 wt% of water and then decrease at water contents of 2 wt%. Richet et al. (1996)
361 suggested a strongest decrease of viscosity for andesitic melts at low water. For example, 1
362 wt% H₂O can decrease viscosity by more than 5 orders of magnitude, but a further viscosity
363 decrease by only 2 orders of magnitude with an additional 2.5 wt% H₂O. A similar effect was
364 observed by Whittington et al. (2000, 2001), who found a decrease of viscosity by 3 orders of
365 magnitude resulted from the first addition of 1 wt% of water, but smaller reductions with

366 further additions of water for both trachyte and phonolite at 900 K. Kushiro (1978b) observed
367 a difference in H₂O dependence of viscosity at high temperature and low temperature. For
368 instance, addition of 3.5 wt% H₂O reduces melt viscosity by 7 orders of magnitude at 1 bar
369 and 1000 K, whereas the viscosity is reduced by only 1.5 orders of magnitude with 4.5 wt%
370 H₂O at 1600 K and 15 kbar. Some works (e.g., Shaw, 1963; Davis and Tomozawa, 1996;
371 Zhang, 1999; Dingwell et al., 2000; Whittington et al., 2000, 2001; Morizet et al., 2007;
372 Misiti et al., 2011) reported that the decreased viscosity for a given amount of H₂O was highly
373 dependent on melt composition, e.g., H₂O effect was more pronounced in more polymerized
374 melts. The decrease of viscosity with H₂O may be attributed to the modification of silicate
375 network and thus depolymerization of melt resulted from the reactions between H₂O and
376 melts (Davis and Tomozawa, 1996; Zhang, 1999)

377

378 **The influence of redox state on the viscosity**

379

380 Although some studies (e.g., Moore et al., 1995; Richet et al., 1996) suggested no effect
381 of redox state on melt viscosity, it has been more widely accepted that the redox state plays an
382 important role on melt viscosity (e.g., Cukierman and Uhlmann, 1974; Klein et al., 1983;
383 Mysen, 1988; Dingwell and Virgo, 1988; Mysen and Virgo, 1989; Dingwell, 1991; Neuville et
384 al., 1993; Toplis et al., 1994; Liebske et al., 2003; Dingwell et al., 2004; Hui and Zhang, 2007;
385 Vetere et al., 2008; Giordano et al., 2008). In general, the increase of Fe²⁺/Fe_{tot} ratio (or
386 decrease of Fe³⁺/Fe_{tot}) leads to the decrease of viscosity (Klein et al., 1983; Dingwell, 1989;
387 Neuville et al., 1993). A decrease in viscosity by 0.81 log units on average when Fe²⁺/Fe_{tot}

388 increases from about 0 to 0.77 was found for the system Na-Si-Fe-O (composition NS4F40),
389 whereas a decrease by 0.34 log units was found for NaFeSi₂O₆ melt when the Fe²⁺/Fe_{tot} ratio
390 increases from 0.08 to 0.82 (Mysen and Virgo, 1989). Liebske et al. (2003) reported that
391 viscosity of dry andesitic melts could decrease by about 1.6 log units when the Fe²⁺/Fe_{tot} ratio
392 increases from 0.42 to 0.79 at 1061 K. Dingwell et al. (2004) found the effect of iron
393 oxidation state on melt viscosity is more pronounced at lower temperature than is at higher
394 temperature. Vetere et al. (2008) indicated that changing Fe²⁺/Fe_{tot} from 0.83 to 0.60 could
395 cause the increase of viscosity of andesitic melts at high temperature ($\cong 1473\text{K}$) with low
396 water (H₂O $\cong 0.1$ wt%) by about 1 log units. The effect of redox state on viscosity of various
397 Fe-bearing melts at 0.001 kbar and 1473 K is shown in Fig. 6a, b, where the oxygen fugacity
398 of each melt corresponds to its iron oxidation state, illustrating that the redox state may exert
399 a significant effect on viscosity. For example, the viscosity of Fe-bearing andesitic melts can
400 increase by about 0.8 log units at 1473 K when increasing the Fe³⁺/Fe_{tot} ratio from 0.2 to 0.6,
401 where for basaltic melts there is 0.86 log units increase in viscosity for the same variation of
402 iron oxidation state (Fig. 6a). In addition, the viscosities of Fe-bearing basaltic melts,
403 andesitic melts and rhyolitic melts can increase by about 0.5 log units, 0.8 log units and 0.6
404 log units, respectively, at 0.001 kbar and 1473 K when increasing the oxidation state from
405 $\Delta\text{NNO}-2$ to $\Delta\text{NNO}+2$ (Fig. 6b) (note here: ΔNNO is the oxygen fugacity relative to NNO
406 buffer, see O'Neill, 1987) . Fig. 6c shows the significant effect of oxygen fugacity on the
407 'network modifiers' parameter (VSM) of various Fe-bearing melts under the same *T-P*
408 conditions as described in Fig. 6(a-b). From Fig. 6, it can be found that the oxygen fugacity
409 may substantially affect iron oxidation state and melt polymerization, and thus exert a

410 significant effect on the viscosity.

411

412 **Application of the model**

413

414 This new model can be used for the calculation of other physical properties of silicate
415 melts, such as the glass transition temperature (T_g) and the melt fragility (m). The glass
416 transition represents the transition from a melt to a solid phase, and plays a vital role in
417 volcanic eruptions because the mechanical response of the magma or lava to applied stress at
418 this brittle/ductile transition governs the eruptive behavior (Sato et al., 1992; Gottsmann et al.,
419 2002). T_g is the temperature division between the relaxed liquids and unrelaxed glasses and is
420 defined as follows (Eqn. (6)) (Dingwell and Webb, 1990; Angell, 1991; Plazek and Ngai,
421 1991; Stevenson et al., 1995; Dingwell, 1995; Moynihan, 1995; Webb and Dingwell, 1995;
422 Gottsmann et al., 2002).

$$423 \log \eta \Big|_{T = T_g} = K - \log |q| \quad (6)$$

424

425 where the K and q are the shift factor and cooling rate, respectively; K may reflect a melt
426 structure influence on the relationship between relaxation time and T_g (Stevenson et al., 1995;
427 Gottsmann et al., 2002). K is treated differently in different works. For example, Gottsmann et
428 al. (2002) and Dingwell et al. (2004) considered it as a function of melt composition, whereas
429 it was treated as a constant in Stevenson et al. (1995). In this study, the method of Gottsmann
430 et al. (2002) is used to calculate T_g as function of cooling rate and silicate melt composition.

431 Melt fragility can be used to measure the degree of deviation of viscosity from an

432 Arrhenian temperature dependence; strong melts tend to have an Arrhenian behavior, whereas
433 fragile melts show a non-Arrhenian behavior (Plazek and Ngai, 1991). It is clearly defined in
434 Toplis et al., (1997) that the fragility (m) of melts is the gradient of the viscosity trace at the
435 glass transition temperature and can be calculated with Eqn. (7).

$$436 \quad m = \text{fragility} = \left. \frac{d(\log_{10} \eta)}{d(T_g / T)} \right|_{T=T_g} = \frac{B}{T_g (1 - C/T_g)^2} \quad (7)$$

437 where B and C are the parameters in this model (Eqn. (4)).

438 Fig. 7a shows the strong effect of H₂O on T_g. The main effect of increased H₂O is to
439 depress values of T_g (Fig. 7a). Model values of T_g decrease sharply with increasing H₂O
440 content (Fig. 7a), which is in agreement with the experimental studies of Giordano et al.
441 (2004a, b). The largest decrease in T_g (145-334 K) is predicted to occur within the first 3 wt%
442 H₂O. Anhydrous melts with T_g values of 893-1125 K have predicted T_g values of 700-725 K
443 at 12 wt% H₂O (Fig. 7a).

444 Numerous discrepancies exist in the literature on the effect of H₂O on melt fragility, and
445 there is currently no consensus. For example, a negative H₂O dependence of melt fragility in
446 various melts was proposed by some works (e.g., Giordano et al., 2003, 2006, Giordano et al.
447 2008; Del Gaudio et al., 2007), whereas positive H₂O dependence of melt fragility in some
448 polymerized melts (e.g., rhyolitic melts) was obtained in others (e.g., Hess and Dingwell 1995;
449 Zhang et al., 2003; Malfait et al., 2013). Though the effect of H₂O on melt fragility is not well
450 understood, some researchers (e.g., Kohn, 2000; Zhang et al., 2003; Del Gaudio et al., 2007;
451 Deubener et al., 2008; Di Genova et al., 2013; Malfait and Sanchez-Valle, 2013) interpreted it
452 as the consequence of the variation in the melt structure mainly associated with the

453 dissolution behavior of water in different silicate melts. A negative correlation between the
454 melt fragility and H₂O is predicted by this model (Fig. 7b). This result is consistent with that
455 obtained by Giordano et al. (2003, 2006, 2008) and Del Gaudio et al. (2007). The implication
456 is that dry melt that shows non-Arrhenius behavior tends to become more Arrhenian with
457 increasing H₂O content. Fig. 7b also shows that anhydrous rhyolitic to basaltic melts have
458 fragility values of 28.5-32.7, but their fragility values virtually overlap at about 22 and with
459 12 wt% H₂O. Although the negative relationship between melt fragility and H₂O in rhyolitic
460 melts contrasts with their positive correlation obtained by some works (e.g., Hess and
461 Dingwell 1995; Zhang et al., 2003; Malfait et al., 2013), there is a common consensus that
462 H₂O indeed exerts a significant effect on fragilities of melts, regardless of the melt types,
463 especially under low water condition (Fig. 7b).

464 As described above, the glass transition temperature (T_g) and fragility (m) of degassing
465 melt would significantly increase as a consequence of the decreasing H₂O water during
466 magma ascent, implying that melt becomes more non-Arrhenian in its temperature
467 dependence and that the potential of melt for explosive fragmentation increases, respectively,
468 ultimately resulting in volcanic eruption (Fig. 7) (e.g., Angell, 1991; Plazek and Ngai, 1991;
469 Toplis et al., 1997; Stevenson et al., 1995; Gottsmann et al., 2002; Giordano et al., 2004a, b;
470 Giordano et al., 2008).

471

472 **Implication for the petrogenesis of magmas**

473

474 The viscosities of Fe-bearing melts calculated by this model are almost 0.5 orders of

475 magnitude higher than those calculated by the previous general models that did not include
476 the Fe oxidation state. The higher viscosities would result in substantially slower ascent
477 velocities of Fe-bearing melts than previously calculated. For example, although the transport
478 rates in dikes are poorly known, a simple model has been proposed for elucidating the
479 rheological relations between the velocity (u) and viscosity of magmas through the volcanic
480 conduits (Eqn. (8)) (Mastin and Ghiorso, 2000; Mastin, 2002; Mastin, 2005):

$$481 \quad u = -(R^2 / 8\eta)(\partial P / \partial z + \rho g) \quad (8)$$

482 where u is the average flow velocity, R is the radius of the conduit; η is the viscosity, P is the
483 pressure; z is vertical position (upwards being positive); ρ is density of melt; g is gravitational
484 acceleration. As the value of $(\partial P / \partial z + \rho g)$ is negative in an upward flowing conduit with z
485 positive in the upward direction (Mastin 2005), the velocity of melt shows inversely
486 dependence upon melt viscosity at the given R and $(\partial P / \partial z + \rho g)$. The rates of separation
487 and ascent through dikes of Fe-bearing melts are slower than previously considered possible
488 because of the higher viscosities calculated by this model than previous general models. The
489 concept that the ascent of melt diapirs through the upward flowing conduit relies on the
490 rheology of the deforming region of wall rock immediately surrounding the ascending diapir
491 was proposed by Mahon et al. (1988), which used S-B-W model for calculating melt
492 viscosities prior to the calculations of the rheology of the deformation region. Although the
493 calculation of diapir ascent viscosities was not conducted in this study, substitution into the
494 Mahon et al. (1988) calculations of partially molten wall rock viscosities estimated by this
495 model would decrease diapiric ascent rate, and thus increase the depth of final diapir
496 emplacement.

497 This new general model presented in this work represents significant improvement over
498 previous general models (e.g., H-Z model; G-R-D model) for the prediction of the viscosities
499 of Fe-bearing melts and is in good agreement with measured viscosities. These higher
500 viscosities result in almost a half orders-of-magnitude decreases in the melt-transport
501 processes occurring in the Fe-bearing magmas.

502

503 ***Acknowledgement*** I greatly appreciate Dr. Fidel Costa and two anonymous reviewers for
504 their valuable and constructive suggestions, which greatly contributed to the improvement of
505 this manuscript. I much thank Dr. Fidel Costa for his significant efforts to kindly polish this
506 manuscript at late stage, Dr. Keith Putirka for his kind assistance in the further improvement
507 of this manuscript, Drs. Keith Putirka, Don R Baker, Bjorn Mysen, Alexander Bartels and
508 Kai-Uwe Hess for their insightful comments on this paper at early stage of this manuscript
509 that have done much help to strengthen this manuscript, and Drs. Zhigang Zhang, Chi Zhang,
510 Ming Geng, and Guangjun Guo for their kind help in this work. This research is supported by
511 the Open Project from State Key Laboratory of Lithospheric Evolution, Institute of Geology
512 and Geophysics, Chinese Academy of Sciences.

513

514

REFERENCES

515 Angell, C.A. (1991) Relaxation in liquids, polymers and plastic crystals -strong/fragile
516 patterns and problems. *Journal of Non-Crystalline Solids*, 131-133, 13-31.

517 Ardia, P., Giordano, D., and Schmidt, M.W. (2008) A model for the viscosity of rhyolite as a
518 function of H₂O-content and pressure: A calibration based on centrifuge piston cylinder

- 519 experiments. *Geochimica et Cosmochimica Acta*, 72, 6103-6123.
- 520 Avramov, I. (2007) Pressure and temperature dependence of viscosity of glassforming and of
521 geoscientifically relevant systems. *Journal of Volcanology and Geothermal Research*,
522 160, 165-174.
- 523 Baker, D. (1996) Granitic melt viscosities: Empirical and configurational entropy models for
524 their calculation. *American Mineralogist*, 81, 126-134.
- 525 Bartels, A., Behrens, H., Holtz, F., Schmidt, B.C., Fechtelkord, M., Knipping, J., Crede, L.,
526 Baasner, A., and Pukallus, N. (2012) The effect of fluorine, boron and phosphorus on the
527 viscosity of pegmatite forming melts. *Chemical Geology* 346, 184-198.
- 528 Behrens, H., and Schulze, F. (2003) Pressure dependence of melt viscosity in the system
529 NaAlSi₃O₈-CaMgSi₂O₆. *American Mineralogist*, 88(8-9), 1351-1363.
- 530 Bottinga, Y., and Weill, D.F. (1972) The viscosity of magmatic silicate liquids; a model
531 calculation. *American Journal of Science*, 272, 438-475.
- 532 Bottinga, Y., and Richet, P. (1995) Silicate melts: The "anomalous" pressure dependence of
533 the viscosity. *Geochimica et Cosmochimica Acta*, 59(13), 2725-2731.
- 534 Brearley, M., and Dickinson, J. (1986) Pressure dependence of melt viscosities on the join
535 diopside-albite. *Geochimica et Cosmochimica Acta*, 50(12), 2563-2570.
- 536 Chevrel, M.O., Baratoux, D., Hess, K.-U., and Dingwell, D.B. (2013) Viscous flow behavior
537 of tholeiitic and alkaline Fe-rich martian basalts. *Geochimica et Cosmochimica Acta*, In
538 Press.
- 539 Cukierman, M., and Uhlmann, D.R. (1974) Effects of iron oxidation state on viscosity, Lunar
540 composition. *Journal of Geophysical Research* 79, 1594-1598.

- 541 Davis, K.M., and Tomozawa, M. (1996) An infrared spectroscopic study of water-related
542 species in the silicate glasses. *Journal of Non-Crystalline Solids*, 201, 177-198.
- 543 Del Gaudio, P., Behrens, H., and Deubener, J. (2007) Viscosity and glass transition
544 temperature of hydrous float glass. *Journal of Non-Crystalline Solids*, 353(3), 223-236.
- 545 Del Gaudio, P., and Behrens, H. (2009) An experimental study on the pressure dependence of
546 viscosity in silicate melts. *Journal of Chemical Physics*, 131, 044504.
- 547 Deubener, J., Hensch, G., Moiseev, A., and Bornhoft, H. (2009) Glasses for solar energy
548 conversion systems. *Journal of the European Ceramic Society*, 29(7), 1203-1210.
- 549 Di Genova, D., Romano, C., Hess, K.-U., Vona, A., Poe, B., Giordano, D., Dingwell, D., and
550 Behrens, H. (2013) The rheology of peralkaline rhyolites from Pantelleria Island. *Journal*
551 *of Volcanology and Geothermal Research*, 249, 201-216.
- 552 Dingwell, D. (1989) Shear viscosities of ferrosilicate liquids. *American Mineralogist*, 74,
553 1038-1044.
- 554 Dingwell, D. (1991) Redox viscometry of some Fe-bearing silicate melts. *American*
555 *Mineralogist*, 76, 1560-1562.
- 556 Dingwell, D., Romano, C., and Hess, K. (1996) The effect of water on the viscosity of a
557 haplogranitic melt under PTX conditions relevant to silicic volcanism. *Contributions to*
558 *Mineralogy and Petrology*, 124, 19-28.
- 559 Dingwell, D.B., and Virgo, D. (1988) Melt viscosities in the Na₂O-FeO-Fe₂O₃-SiO₂ system
560 and factors controlling the relative viscosities of fully polymerized silicate melts.
561 *Geochimica et Cosmochimica Acta*, 52, 395-403.
- 562 Dingwell, D., Courtial, P., Giordano, D., and Nichols, A. (2004) Viscosity of peridotite liquid.

- 563 Earth and Planetary Science Letters, 226, 127-138.
- 564 Dingwell, D., Knoche, R., and Webb, S. (1993) The effect of P₂O₅ on the viscosity of
565 haplogranitic liquid. European Journal of Mineralogy, 5, 133-140.
- 566 Dingwell, D.B. (1995) Relaxation in silicate melts; some applications. Reviews in Mineralogy
567 and Geochemistry, 32, 21-66.
- 568 Dingwell, D. B., Hess, K.-U., and Romano, C. (2000) Viscosities of granitic (sensu lato) melts:
569 Influence of the anorthite component. American Mineralogist, 85, 1342-1348.
- 570 Dingwell, D.B., and Webb, S.L. (1990) Relaxation in silicate melts. European Journal of
571 Mineralogy, 2, 427-449.
- 572 Duan, X. Z. (2014) A general model for predicting the solubility behavior of H₂O-CO₂ fluids
573 in silicate melts over a wide range of pressure, temperature and compositions.
574 Geochimica et Cosmochimica Acta, 125, 582-609.
- 575 Flood, H. and Forland, T. (1947) The acidic and basic properties of oxides. Acta Chemical
576 Scand, 1, 592-604.
- 577 Fraser, D. G. (1975) Activities of trace elements in silicate melt. Geochimica et
578 Cosmochimica Acta, 39, 1525-1530.
- 579 Fraser, D. G. (1977) Thermodynamic properties of silicate melt. Thermodynamics in Geology,
580 301-325.
- 581 Fulcher, G.S. (1925) Analysis of recent measurements of the viscosity of glasses. Journal of
582 the American Ceramic Society, 8, 339-355.
- 583 Giordano, D., and Dingwell, D.B. (2003a) Viscosity of hydrous Etna basalt: implications for
584 Plinian-style basaltic eruptions. Bulletin of Volcanology, 65, 8-14.

- 585 Giordano, D., and Dingwell, D.B. (2003b) Non-Arrhenian multicomponent melt viscosity: a
586 model. *Earth and Planetary Science Letters*, 208, 337-349.
- 587 Giordano, D., Mangiacapra, A., Potuzak, M., Russell, J., Romano, C., Dingwell, D., and Di
588 Muro, A. (2006) An expanded non-Arrhenian model for silicate melt viscosity: A
589 treatment for metaluminous, peraluminous and peralkaline liquids. *Chemical Geology*,
590 229, 42-56.
- 591 Giordano, D., Romano, C., Papale, P., and Dingwell, D.B. (2004a) The viscosity of trachytes,
592 and comparison with basalts, phonolites, and rhyolites. *Chemical Geology*, 213, 49-61.
- 593 Giordano, D., Romano, C., Dingwell, D., Poe, B., and Behrens, H. (2004b) The combined
594 effects of water and fluorine on the viscosity of silicic magmas. *Geochimica et*
595 *Cosmochimica Acta*, 68, 5159-5168.
- 596 Giordano, D., Russell, J. K., and Dingwell, D.B. (2008) Viscosity of magmatic liquids: A
597 model. *Earth and Planetary Science Letters*, 271, 123-134.
- 598 Gottsmann, J., Giordano, D., and Dingwell, D.B. (2002) Predicting shear viscosity during
599 volcanic processes at the glass transition: a calorimetric calibration. *Earth and Planetary*
600 *Science Letters*, 198, 417-427.
- 601 Gupta, P.K. (1987) Negative pressure dependence of viscosity. *Journal of the American*
602 *Ceramic Society*, 70(7), C152-C153.
- 603 Hess, K.U., and Dingwell, D.B. (1996) Viscosities of hydrous leucogranitic melts: A
604 non-Arrhenian model. *American Mineralogist*, 81, 1297-1300.
- 605 Hui, H., and Zhang, Y. (2007) Toward a general viscosity equation for natural anhydrous and
606 hydrous silicate melts. *Geochimica et Cosmochimica Acta*, 71, 403-416.

- 607 Hui, H., Zhang, Y., Xu, Z., Del Gaudio, P., and Behrens, H. (2009) Pressure dependence of
608 viscosity of rhyolitic melts. *Geochimica et Cosmochimica Acta*, 73, 3680-3693.
- 609 Kenny, D.A., and McCoach, D.B. (2003) Effect of the number of variables on measures of fit
610 in structural equation modeling. *Structural. Equation. Modeling*, 10, 333-351.
- 611 Klein, L.C., FA sano, B.V., and Wu, J.M. (1983) Viscous flow behavior of four
612 iron-containing silicates with alumina, effects of composition and oxidation condition.
613 *Journal of Geophysical Research*, 88, A880-A886.
- 614 Kilinc, A., Carmichael, I.S.E., Rivers, M.L., and Sack, R.O. (1983) The ferric-ferrous ratio of
615 natural silicate liquids equilibrated in air. *Contributions to Mineralogy and Petrology*,
616 83(1), 136-140.
- 617 Kushiro, I. (1976) Changes in viscosity and structure of melt of NaAlSi₂O₆ composition at
618 high pressures. *Journal of Geophysical Research*, 81(35), 6347-6350.
- 619 Kushiro, I. (1978a) Viscosity and structural changes of albite (NaAlSi₃O₈) melt at high
620 pressures. *Earth and Planetary Science Letters* **41**, 87-90.
- 621 Kushiro, I. (1978b) Density and viscosity of hydrous calc-alkalic andesite magma at high
622 pressre. *Carnegie Institute of Washington Year Book* **77**, 675-678.
- 623 Kushiro, I. (1986) Viscosity of partial melts in the upper mantle. *Journal of Geophysical*
624 *Research*, 91, 9343-9350.
- 625 Le Bas, M., Le Maitre, R., Streckeisen, A., and Zanettin, B. (1986) A chemical classification
626 of volcanic rocks based on the total alkali-silica diagram. *Journal of Petrology*, 27,
627 745-750.
- 628 Liebske, C., Behrens, H., Holtz, F., and Lange, R. (2003) The influence of pressure and

- 629 composition on the viscosity of andesitic melts. *Geochimica et Cosmochimica Acta*, 67,
630 473-485.
- 631 Liebske, C., Schmickler, B., Terasaki, H., Poe, B., Suzuki, A., Funakoshi, K., Ando, R., and
632 Rubie, D. (2005) Viscosity of peridotite liquid up to 13 GPa: Implications for magma
633 ocean viscosities. *Earth and Planetary Science Letters*, 240, 589-604.
- 634 Mahon, K. I., Harrison, T. M., and Drew, D. A., 1988. Ascent of a granitoid diapir in a
635 temperature varying medium. *Journal of Geophysical Research*. 94, 1174-1188.
- 636 Malfait, W.J., and Sanchez-Valle, C. (2013) Effect of water and network connectivity on
637 glass elasticity and melt fragility. *Chemical Geology*, 346, 72-80.
- 638 Mastin, L.G. (2002) Insights into volcanic conduit flow from an open-source numerical model.
639 *Geochemistry, Geophysics, Geosystems*, 3, 1029-1046.
- 640 Mastin, L.G. (2005) The controlling effect of viscous dissipation on magma flow in silicic
641 conduits. *Journal of Volcanology and Geothermal Research*, 143, 17-28.
- 642 Mastin, L.G., and Ghiorso, M.S. (2000) A numerical program for steady-state flow of
643 magma-gas mixtures through vertical eruptive conduits, USGS Open-File Report 00-209.
644 *U.S. Geological Survey, Vancouver, WA*, 56 pp.
- 645 Morizet, Y., Nichols, A. R. L., Kohn, S. C., Brooker, R. A., and Dingwell, D. B. (2007) The
646 influence of H₂O and CO₂ on the glass transition temperature: insights into the effects of
647 volatiles on magma viscosity. *European Journal of Mineralogy*, 19, 657-669.
- 648 Moynihan, C.T. (1995) Structural relaxation and the glass transition. *Reviews in Mineralogy*
649 *and Geochemistry*, 32, 1-19.
- 650 Misiti, V., Freda, C., Taddeucci, J., Romano, C., Scarlato, P., Longo, A., Papale, P., and Poe, B.

- 651 T. (2006) The effect of H₂O on the viscosity of K-trachytic melts at magmatic
652 temperatures. *Chemical Geology*, 235, 124-137.
- 653 Misiti, V., Vetere, F., Freda, C., Scarlato, P., Behrens, H., Mangiacapra, A., and Dingwell, D.
654 B. (2011) A general viscosity model of Campi Flegrei (Italy) melts. *Chemical Geology*,
655 290, 50-59.
- 656 Mori, S., Ohtani, E., and Suzuki, A. (2000) Viscosity of the albite melt to 7 GPa at 2000 K.
657 *Earth and Planetary Science Letters*, 175, 87-92.
- 658 Moore, G., Richter, K., and Carmichael, I.S.E. (1995) The effect of dissolved water on the
659 oxidation state of iron in natural silicate liquids. *Contributions to Mineralogy and*
660 *Petrology*, 120, 170-179.
- 661 Moretti, R. (2005) Polymerisation, basicity, oxidation state and their role in ionic modelling of
662 silicate melts. *Annals of Geophysics*, 48, 583-608
- 663 Mysen, B., Virgo, D., and Scarfe, C. (1980) Relations between the anionic structure and
664 viscosity of silicate melts—a Raman spectroscopic study. *American Mineralogist*, 65,
665 690–710.
- 666 Mysen, B., Virgo, D., Scarfe, C., and Cronin, D. (1985) Viscosity and structure of iron-and
667 aluminum-bearing calcium silicate melts at 1 atm. *American Mineralogist*, 70, 487-498.
- 668 Mysen, B.O. (1988) *Structure and Properties of Silicate melts*. Elsevier, Amsterdam, 1988,
669 354 pp.
- 670 Mysen, B.O., and Richet, P. (2005) *Developments in Geochemistry 10, Silicate Glasses and*
671 *Melts Properties and Structure*. Elsevier, Amsterdam.
- 672 Mysen, B.O., and Virgo, D. (1989) Redox equilibria, structure and properties of Fe-bearing

- 673 aluminosilicate melts: relationship among temperature, composition and oxygen fugacity
674 in the system $\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2-\text{Fe}-\text{O}$. American Mineralogist, 74, 58-76.
- 675 Neuville, D., Courtial, P., Dingwell, D., and Richet, P. (1993) Thermodynamic and rheological
676 properties of rhyolite and andesite melts. Contributions to Mineralogy and Petrology, 113,
677 572-581.
- 678 O'Neill, H.S.C. (1987) Free energies of formation of NiO, CoO, Ni_2SiO_4 , and Co_2SiO_4 .
679 American Mineralogist, 72(3), 280-291.
- 680 Ottonello, G., Moretti, R., Marini, L., and Vetuschi Zuccolini, M. (2001) Oxidation state of
681 iron in silicate glasses and melts: a thermochemical model. Chemical Geology, 174(1-3),
682 157-179.
- 683 Papale, P. (1999) Modeling of the solubility of a two-component $\text{H}_2\text{O}+\text{CO}_2$ fluid in silicate
684 liquids. American Mineralogist, 84, 477-492.
- 685 Persikov, Y.S., Zharikov, V.A., Bukhtiyarov, P.G., and Pol'skoy, S.F. (1990) The effect of
686 volatiles on the properties of magmatic melts. European Journal of Mineralogy, 2(5),
687 621-642.
- 688 Persikov, E. (1991) The viscosity of magmatic liquids: experiment, generalized patterns. A
689 model for calculation and prediction. Applications. Advance in Physical Geochemistry, 9,
690 1-40.
- 691 Plazek, D.J., and Ngai, K.L. (1991) Correlation of polymer segmental chain dynamics with
692 temperature-dependent time-scale shifts. Macromolecules, 24, 1222-1224.
- 693 Press, W.M., Teukolski, S.A., Vetterling, W.T., and Flannery, B.P. (1992) Numerical recipes in
694 FORTRAN-The art of scientific computing, second edition, 963 p. Cambridge

695 University Press Cambridge, U.K.

696 Richet, P., Lejeune, A., Holtz, F., and Roux, J. (1996) Water and the viscosity of andesite
697 melts. *Chemical Geology*, 128, 185-197.

698 Romano, C., Giordano, D., Papale, P., Mincione, V., Dingwell, D.B., and Rosi, M. (2003) The
699 dry and hydrous viscosities of alkaline melts from Vesuvius and Phlegrean Fields.
700 *Chemical Geology*, 202, 23-38.

701 Russell, J., Giordano, D., and Dingwell, D. (2003) High-temperature limits on viscosity of
702 non-Arrhenian silicate melts. *American Mineralogist*, 88, 1390-1394.

703 Sato, H., Fugii, T., and Nakada, S. (1992) Crumbling of dacite dome lava and generation of
704 pyroclastic flows at Unzen volcano. *Nature*, 360, 664-666.

705 Scarfe, C., Mysen, B., and Virgo, D. (1979) Changes in viscosity and density of melts of
706 sodium disilicate, sodium metasilicate, and diopside composition with pressure. *Year
707 Book Carnegie Institute Washington*, 78, 547-551.

708 Scarfe, C.M., Mysen, B.O., and Virgo, D. (1987) Pressure dependence of the viscosity of
709 silicate melts. *Magmatic processes: Physicochemical principles*, 1, 59-67.

710 Scaillet, B., Holtz, F., Pichavant, M., and Schmidt, M. (1996) Viscosity of Himalayan
711 leucogranites: Implications for mechanisms of granitic magma ascent. *Journal of
712 Geophysical Research*, 101, 27691-27699.

713 Schulze, F., Behrens, H., Holtz, F., Rous, J., and Johannes, W. (1996) The influence of H₂O on
714 the viscosity of a haplogranitic melt. *American Mineralogist*, 81, 1155-1165.

715 Schulze, F., Behrens, H., and Hurkuck, W. (1999) Determination of the influence of pressure
716 and dissolved water on the viscosity of highly viscous melts; application of a new

- 717 parallel-plate viscometer. *American Mineralogist*, 84, 1512-1520.
- 718 Shaw, H. (1963) Obsidian-H₂O viscosities at 1000 and 2000 bars in the temperature range 700
719 to 900 °C. *Journal of Geophysical Research*, 68, 6337-6343.
- 720 Shaw, H.R. (1972) Viscosities of magmatic silicate liquids; an empirical method of prediction.
721 *American Journal of Science*, 272, 870-893.
- 722 Silver, L. and Stolper, E. (1989) Water in albitic glasses. *Journal of Petrology*, 30, 667.
- 723 Silver, L. A., Ihinger, P. D., and Stolper, E. (1990) The influence of bulk composition on the
724 speciation of water in silicate glasses. *Contributions to Mineralogy and Petrology*, 104,
725 142-162.
- 726 Stevenson, R., Bagdassarov, N., Dingwell, D., and Romano, C. (1998) The influence of trace
727 amounts of water on the viscosity of rhyolites. *Bulletin of Volcanology*. 60, 89-97.
- 728 Stevenson, R.J., Dingwell, D.B., Webb, S.L., and Bagdassarov, N. S. (1995) The equivalence
729 of enthalpy and shear stress relaxation in rhyolitic obsidians and quantification of the
730 liquid-glass transition in volcanic processes. *Journal of Volcanology and Geothermal
731 Research*, 68, 297-306.
- 732 Stevenson, R.J., Dingwell, D.B., Webb, S.L., and Sharp, T.G. (1996) Viscosity of
733 microlite-bearing rhyolitic obsidians: an experimental study. *Bulletin of Volcanology*,
734 58, 298-309.
- 735 Stolper, E. (1982) The speciation of water in silicate melts. *Geochimica et Cosmochimica
736 Acta*, 46, 2609-2620.
- 737 Tammann, G., and Hesse, W. (1926) Die abhängigkeit der viskosität von der temperature bei
738 unterkühlten flüssigkeiten. *Z Anorg Allg Chem*, 156, 245-257.

- 739 Toplis, M., Dingwell, D., Hess, K., and Lenci, T. (1997) Viscosity, fragility, and
740 configurational entropy of melts along the join $\text{SiO}_2\text{-NaAlSiO}_4$. American Mineralogist,
741 82, 979-990.
- 742 Toplis, M.J., and Dingwell, D.B. (1996) The variable influence of P_2O_5 on the viscosity of
743 melts of differing alkali/aluminium ratio: Implications for the structural role of
744 phosphorus in silicate melts. *Geochimica et Cosmochimica Acta*, 60(21), 4107-4121.
- 745 Toplis, M.J., Dingwell, D.B., and Libourel, G. (1994) The effect of phosphorus on the iron
746 redox ratio, viscosity, and density of an evolved ferro-basalt. *Contributions to*
747 *Mineralogy and Petrology*, 117, 293-304.
- 748 Vetere, F., Behrens, H., Holtz, F., and Neuville, D. (2006) Viscosity of andesitic melts-new
749 experimental data and a revised calculation model. *Chemical Geology*, 228, 233-245.
- 750 Vetere, F., Behrens, H., Misiti, V., Ventura, G., Holtz, F., De Rosa, R., and Deubener, J. (2007)
751 The viscosity of shoshonitic melts (Vulcanello Peninsula, Aeolian Islands, Italy): Insight
752 on the magma ascent in dikes. *Chemical Geology*, 245, 89-102.
- 753 Vetere, F., Behrens, H., Schuessler, J., Holtz, F., Misiti, V., and Borchers, L. (2008) Viscosity
754 of andesite melts and its implication for magma mixing prior to Unzen 1991-1995
755 eruption. *Journal of Volcanology and Geothermal Research*, 175, 208-217.
- 756 Vogel, D.H. (1921) Das Temperaturabhängigkeitsgesetz der Viskosität von Flüssigkeiten.
757 *Physical.Z.*, 645-646.
- 758 Webb, S.L., and Dingwell, D.B. (1995) Viscoelasticity. *Reviews in Mineralogy and*
759 *Geochemistry*, 32, 95-119.
- 760 Whittington, A., Richet, P., and Holtz, F. (2000) Water and the viscosity of depolymerized

- 761 aluminosilicate melts. *Geochimica et Cosmochimica Acta*, 64, 3725-3736.
- 762 Whittington, A., Richet, P., Linard, Y., and Holtz, F. (2001) The viscosity of hydrous
763 phonolites and trachytes. *Chemical Geology*, 174, 209-223.
- 764 Zhang, Y. (1999) H₂O in rhyolitic glasses and melts: measurement, speciation, solubility, and
765 diffusion. *Reviews of Geophysics*, 37, 493-516.
- 766 Zhang, Y., Xu, Z., and Liu, Y. (2003) Viscosity of hydrous rhyolitic melts inferred from
767 kinetic experiments, and a new viscosity model. *American Mineralogist*, 88, 1741-1752.
- 768

769

Tables

770 Table 1 Experimental data of natural Fe-bearing melt viscosity used in this model (including
 771 data for calibration and for test)

772

Reference	Melt	T (K)	P (kbar)	H ₂ O (wt%)	N [#]
Giordano et al. (2003b)	Basalt	798-1004	0.00	0.02-1.5	22
Liebske et al. (2003)*	Andesite	1006-1098	0.00	0.00	22
Richet et al. (1996)	Andesite	996-1123	0.00	0.00	41
Toplis et al. (1994)	Basalt (MTV1)	1670-1673	0.00	0.00	4
	Basalt (MTV2)	1572-1867	0.00	0.00	4
	Basalt (MTV3)	1621-1769	0.00	0.00	4
	Basalt (MTV4)	1719-1867	0.00	0.00	4
	Basalt (MTV6)	1673-1873	0.00	0.00	4
	Basalt (MTV7)	1673-1873	0.00	0.00	3
	Basalt (MTV8)	1673-1873	0.00	0.00	3
	Basalt (MTV9)	1673-1873	0.00	0.00	3
	Shaw (1963)	Rhyolite	973-1173	1.0-2	4.3-6.2
Persikov (1991)	Rhyolite	973-1173	0.5-7	2.1-12.3	12
Liebske et al. (2005)*	Peridotite	2043-2523	28-130	0.00	21
Vetere et al. (2007)*	Shoshonite	733-1573	0.001-5	0.019-4.75	21
Vetere et al. (2008)	Andesite	747-1573	3.0-10	0.02-6.31	27
Stevenson et al. (1995)	Rhyolite	897-965	0.00	0.16-0.21	6
Kushiro (1986)	Olivine Tholeiite I	1648-1748	7-12.5	0.00	6
	Olivine Tholeiite II	1623-1673	7-12.5	0.00	3
	Low-alkali Tholeiite	1623-1673	10.0-15	0.00	5
	Alkali Olivine Basalt	1623-1698	7.0-15	0-1.08	7
	K-rich Alkali Basalt	1673-1698	7-12.5	0.00	3

773 # Number of measurements.

774 * The data are not used for the calibration of the model, but for the check.

775

776

777 **Table 2 Parameters for this model**

Parameters	Value
a	-4.753468E+00 (1.116472E-01)
b ₁	7.317415E+03 (3.356892E-01)
b ₂	-1.572090E+02 (3.719342E-02)
b ₃	-3.068000E-02 (3.740712E-03)
b ₄	1.816191E-01 (6.108900E-02)
b ₅	2.185230E-01 (4.804092E-02)
c ₁	1.672632E+02 (3.729234E-01)
c ₂	-1.765680E+01 (1.779570E+00)
c ₃	-6.101450E-05 (3.555688E-05)
c ₄	3.044336E-04 (1.376224E-04)
c ₅	6.653222E-04(3.630155E-05)

778

779 The parameters and standard deviations (in parentheses) are estimated by

780 Levenberg-Marquardt method of nonlinear least squares according to Press et al. (1992)

781

782 **Table 3 Comparison between the predictions of models for melt viscosity and the**
 783 **experimental data with known redox states**

Model		N ⁱ	Various ^a	Basalt ^b	Shoshonite ^c	Andesite ^d	Rhyolite ^e	Ref ^j
This model	RMSE ^f	11	0.23	0.26	0.40	0.16	0.32	This study
S-B-W model	RMSE	8	0.77	0.99		0.15		1, 2
H-Z model	RMSE	37	0.70 ^g	0.35 ^g	1.55	0.79 ^g	0.32	3
G-R-D model	RMSE	18	0.68 ^h	0.56	0.62	0.77 ^h	0.99	4
Baker model	RMSE	7					0.54	5
S-B-H model	RMSE	5					1.20	6
H-D model	RMSE	6					0.72	7
Z-X-L model	RMSE	7					2.03	8
A-G-S model	RMSE	6					2.53	9
H-Z-X model	RMSE	11					2.03	10
R-L-H model	RMSE	10				0.89		11
V-B-S model	RMSE	7				0.16		12, 13
V-B-M model	RMSE	6			0.17			14
M-V-F model	RMSE	6			0.82			15, 16

784 Note: Blank range indicates the melt ranges where the specific models are not intended to be
 785 applied; the *T-P-X* range of each model is shown in the text in detail

786 ^a The viscosity calculated for various melts in the stated *T-P-X* range of relevant models

787 ^b The viscosity calculated for basaltic melts in the stated *T-P-X* range of relevant models

788 ^c The viscosity calculated for shoshonitic melts in the stated *T-P-X* range of relevant models

789 ^d The viscosity calculated for andesitic melts in the stated *T-P-X* range of relevant models

790 ^e The viscosity calculated for rhyolitic melts in the stated *T-P-X* range of relevant models

791 ^f The root-mean-square-error (log units) between the measured and calculated data defined in
 792 Eqn. (1) in the Text.

793 ^g The experimental data belong to the outliers of H-Z model are not in comparison here (see
 794 the Text for detail)

795 ^h The experimental data belong to the outliers of G-R-D model are not compared here (see the
 796 Text for detail)

797 ⁱ The number of parameters used in each model

798 ^j The reference number: 1, Shaw (1972); 2, Bottinga and Weill (1972); 3, Hui and Zhang
 799 (2007); 4, Giordano et al. (2008); 5, Baker (1996); 6, Schulze et al. (1996); 7, Hess and
 800 Dingwell (1996); 8, Zhang et al. (2003); 9, Ardia et al. (2008); 10, Hui et al. (2009); 11,
 801 Richet et al. (1996); 12, Vetere et al. (2006); 13, Vetere et al. (2008); 14, Vetere et al.
 802 (2007); 15, Misiti et al. (2006); 16, Misiti et al. (2011)

803
 804
 805
 806

Figure Captions

807

808 Fig. 1. The melt compositions of the database in this study. (a) Total alkalis ($\text{Na}_2\text{O}+\text{K}_2\text{O}$, wt%)
809 versus SiO_2 (wt%) diagram (Le Bas et al., 1986). F, foidite; Ph, phonolite; Pc, microbasalt; B,
810 basalt; R, rhyolite ; T, trachyte or trachydacite; H1, basanite or tephrite; H2, phonotephrite;
811 H3, tephriphonolite; J1, trachybasalt; J2, basaltic trachyandesite; J3, trachyandesite; L1,
812 basaltic andesite; L2, andesite ; L3, dacite. (b) Total iron content (FeO_{tot} , wt%) versus SiO_2
813 (wt%) diagram.

814 Fig. 2. Comparisons of the calculated data by general models (e.g., this model; H-Z model;
815 G-R-D model) with experimental viscosity data, including those with known
816 (`Data_redox_state_known`) and unknown redox states (`Data_redox_state_unknown`). Note:
817 only the experimental data within the stated *T-P-X* ranges of models are used. Since the H-Z
818 and G-R-D models intended to predict the viscosity at the atmospheric pressure, the data
819 beyond their pressure range are not used in Fig 2 (b-c). The circles denote the experimental
820 data with known redox states, which include those used for calibration of the model (filled
821 circles), and the ones used only for testing the predictability of the model (open circles). In
822 addition, the filled squares are the experimental data with unknown redox states. The dashed
823 lines indicate 95% confidence level of the calculated viscosities. RMSE is the abbreviation of
824 root-mean-square-error (log units) between the measured and calculated data. (a) Comparison
825 between the calculated data with this model and the experimental data. Note: in this figure,
826 the FeO and Fe_2O_3 contents were arbitrarily calculated, assuming that half of the total iron
827 FeO_{tot} (wt%) is FeO (wt%) and the other half is Fe_2O_3 (wt%). (b) Comparison between the
828 calculated data with H-Z model and the experimental data. Note: the outliers of H-Z model

829 (Toplis et al., 1994; Richet et al., 1996) are not plotted. (c) Comparison between the
830 calculated data with G-R-D model and the experimental data. Note: the outliers of G-R-D
831 model (Richet et al., 1996) are not plotted. The sources of experimental data with known
832 redox states (Data_redox_state_known): Shaw (1963); Kushiro (1986); Persikov
833 (1991); Toplis et al. (1994); Stevenson et al. (1995); Richet et al. (1996); Giordano et al.
834 (2003b); Liebske et al. (2003, 2005); Vetere et al. (2007, 2008). Note: most of these data are
835 used in the model calibration, except those data (Liebske et al., 2003, 2005; Vetere et al., 2007)
836 that are not used for parameterization, but for checking the predictability of this model. The
837 sources of experimental data with unknown redox states (Data_redox_state_unknown):
838 Neuville et al. (1993); Stevenson et al. (1996); Alidibirov et al.(1997); Gottsman et al. (2002);
839 Roman et al. (2003); Misiti et al. (2006); Misiti et al. (2011).

840 Fig. 3. Comparison between the calculated data with this model and the experimental data
841 with known redox states (Data_redox_state_known). The circles denote the experimental data
842 with known redox states, which include the ones used for the calibration of the model (filled
843 circles), and the ones used for testing the predictability of the model (open circles). The
844 dashed lines indicate 95% confidence level of the calculated viscosities. RMSE is the
845 abbreviation of root-mean-square-error (log units) between the measured and calculated data.
846 In this case, the experimental measurements of iron oxidation state are taken as inputs for
847 calculation of melt viscosity. The experimental data sources are the same as those defined in
848 Fig. 2.

849 Fig. 4. The prediction of this model in the high pressure range ($P \geq 1$ kbar). The symbols are
850 the experimental measurements of melt viscosity. The dashed lines indicate 95% confidence

851 level of the calculated viscosities.

852 Fig. 5. The pressure dependence of viscosity for Fe-bearing andesitic melts with different H₂O
853 contents. The source of the melts: Vetere et al. (2008).

854 Fig. 6. The effect of redox state on viscosity of various melts at 0.001 kbar and 1473 K. (a)
855 The viscosity of various melts varying with Fe³⁺/Fe_{tot}; (b) The viscosity of various melts
856 varying with oxygen fugacity; (c) The 'network modifiers' parameter (VSM) varying with
857 oxygen fugacity. See the Text for detail. The melt source: basalt, Giordano et al. (2003b);
858 andesite, Liebske et al. (2003); rhyolite, Persikov (1991).

859 Fig. 7. The properties of silicate melts predicted by this model. (a) Computed values of T_g (K)
860 vs. H₂O content (wt%) for diverse melt compositions including: alkali basalt, theoleiite,
861 shoshonite, andesite, rhyolite, assuming q (cooling rate) as 0.0833 K/s. (b) Model curves for
862 the values of fragility (m) vs. H₂O content (wt%) for the same melts as shown in Fig.7a ,
863 assuming q (cooling rate) as 0.0833 K/s. Explosive volcanic behavior is favored by melt with
864 low fragility (e.g., strong) at high volatile contents.

865

866

867

Figures

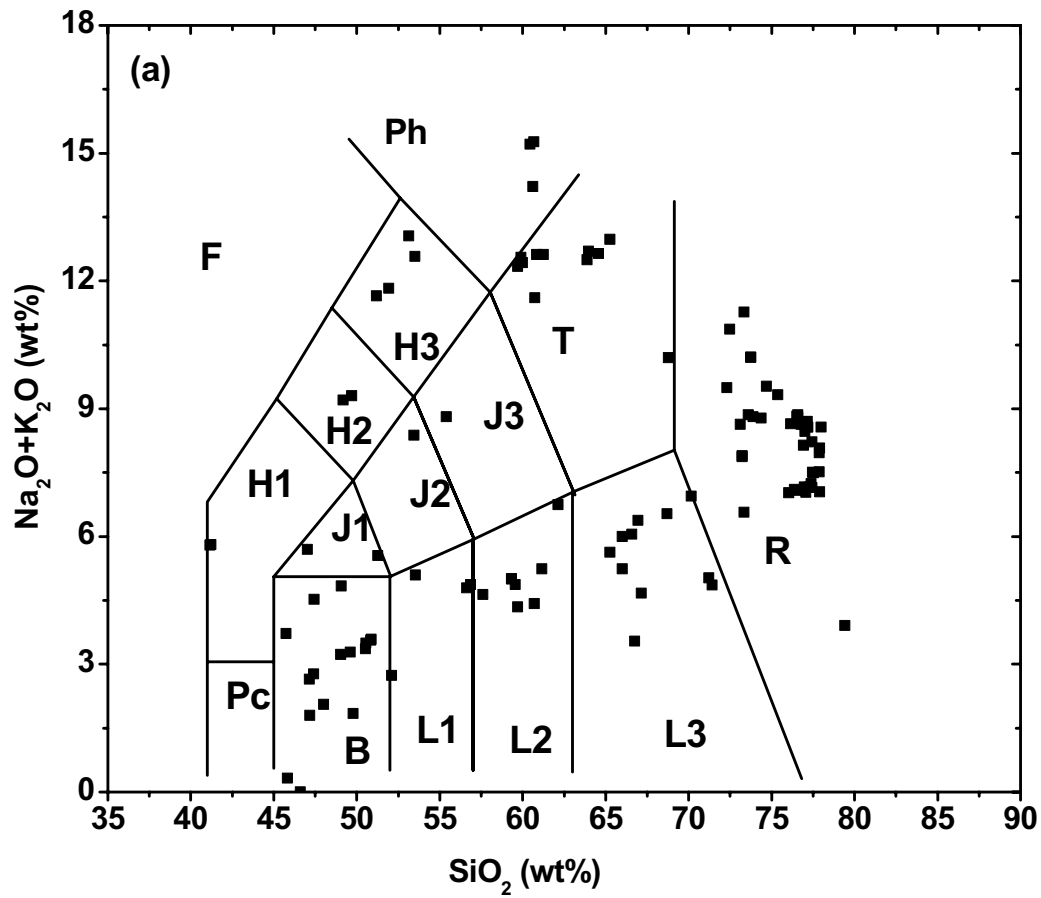


Fig. 1a

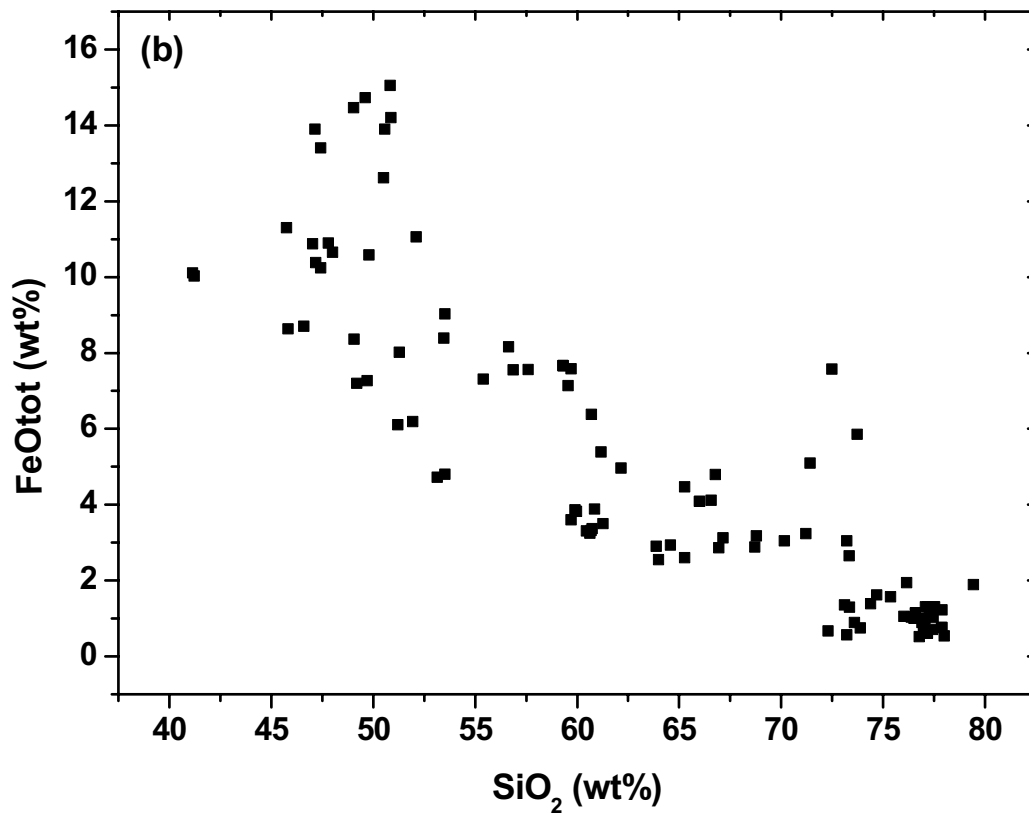


Fig. 1b

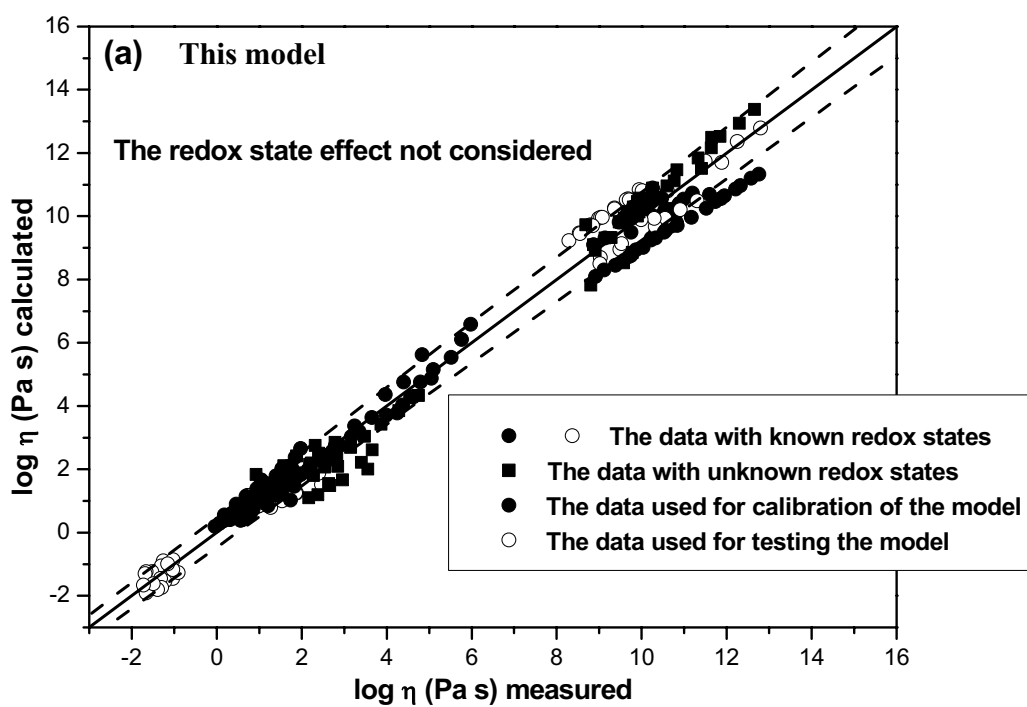


Fig. 2a

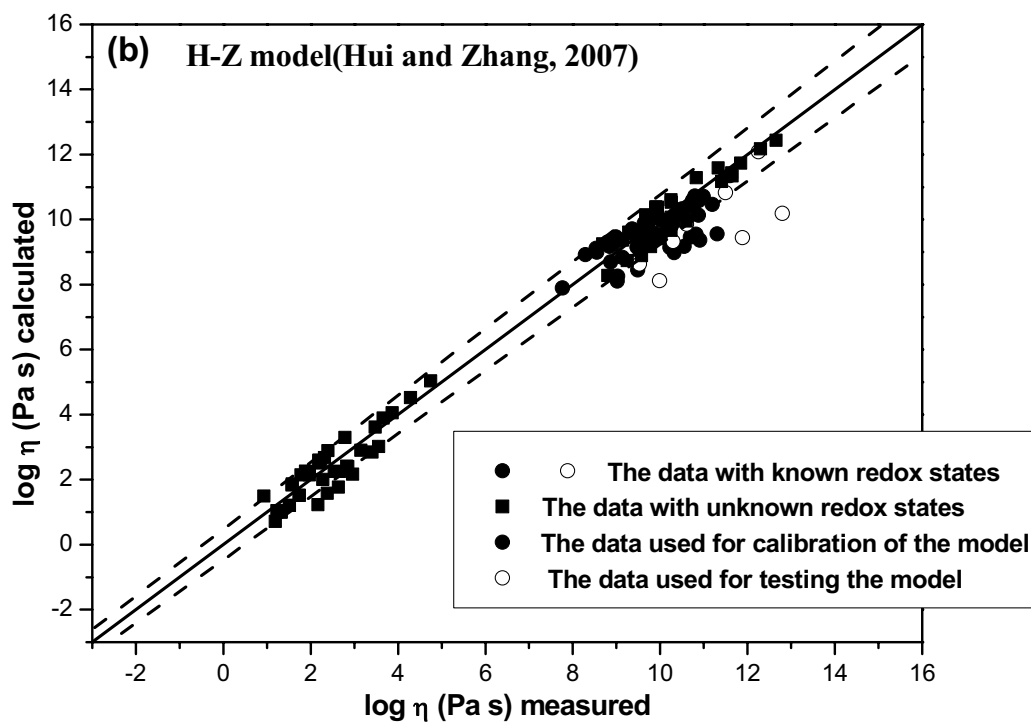


Fig. 2b

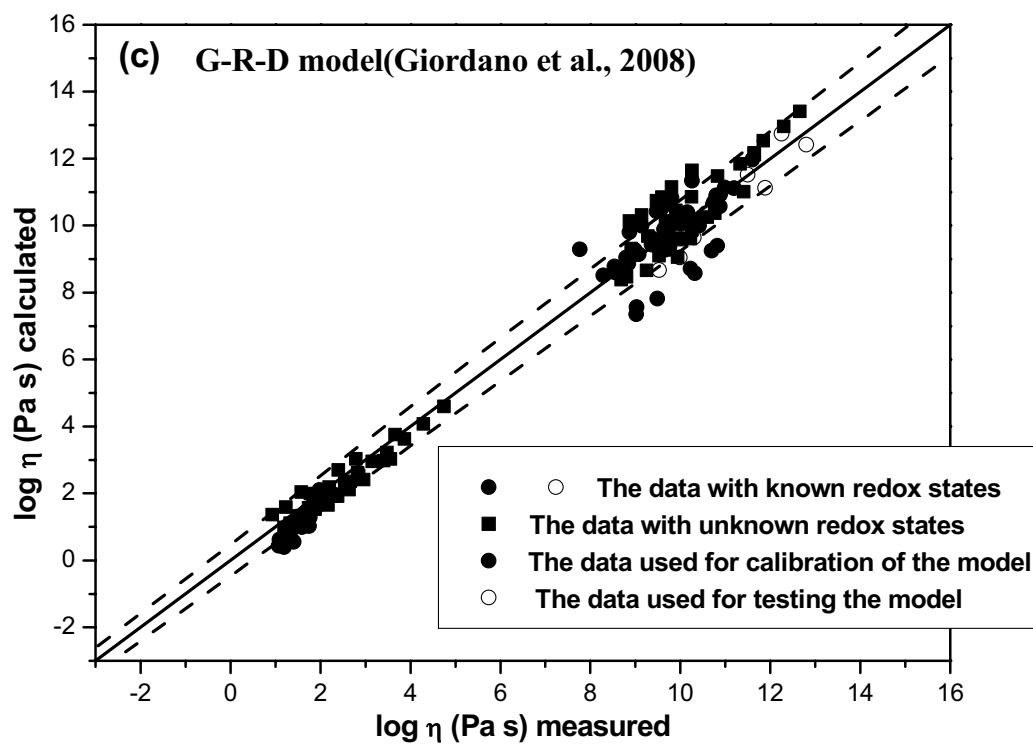


Fig. 2c

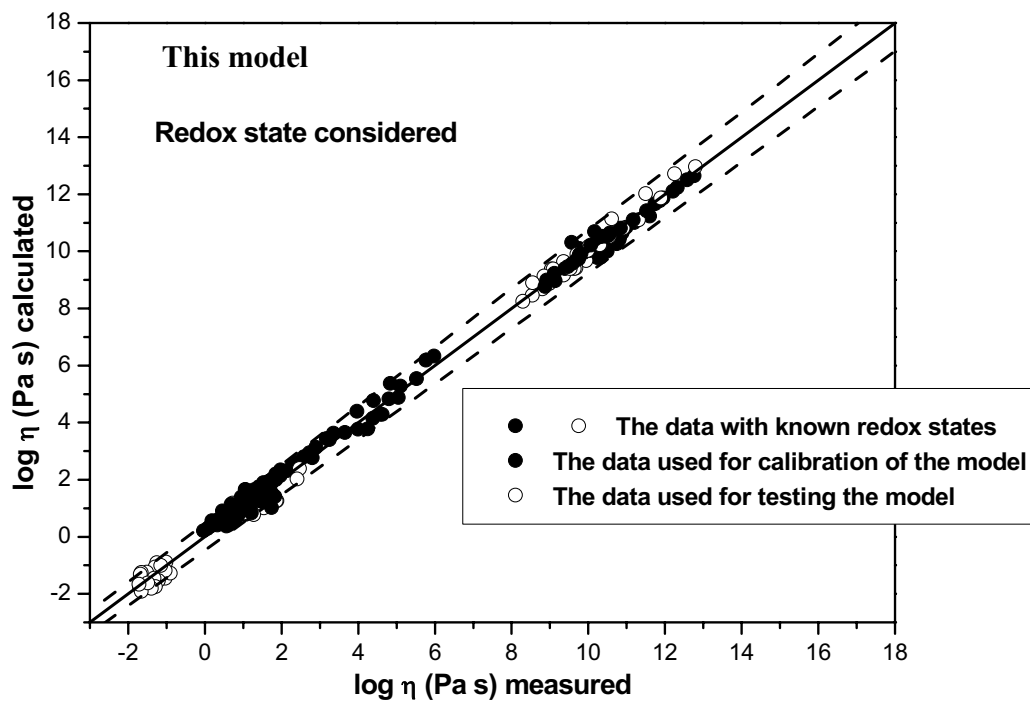


Fig. 3

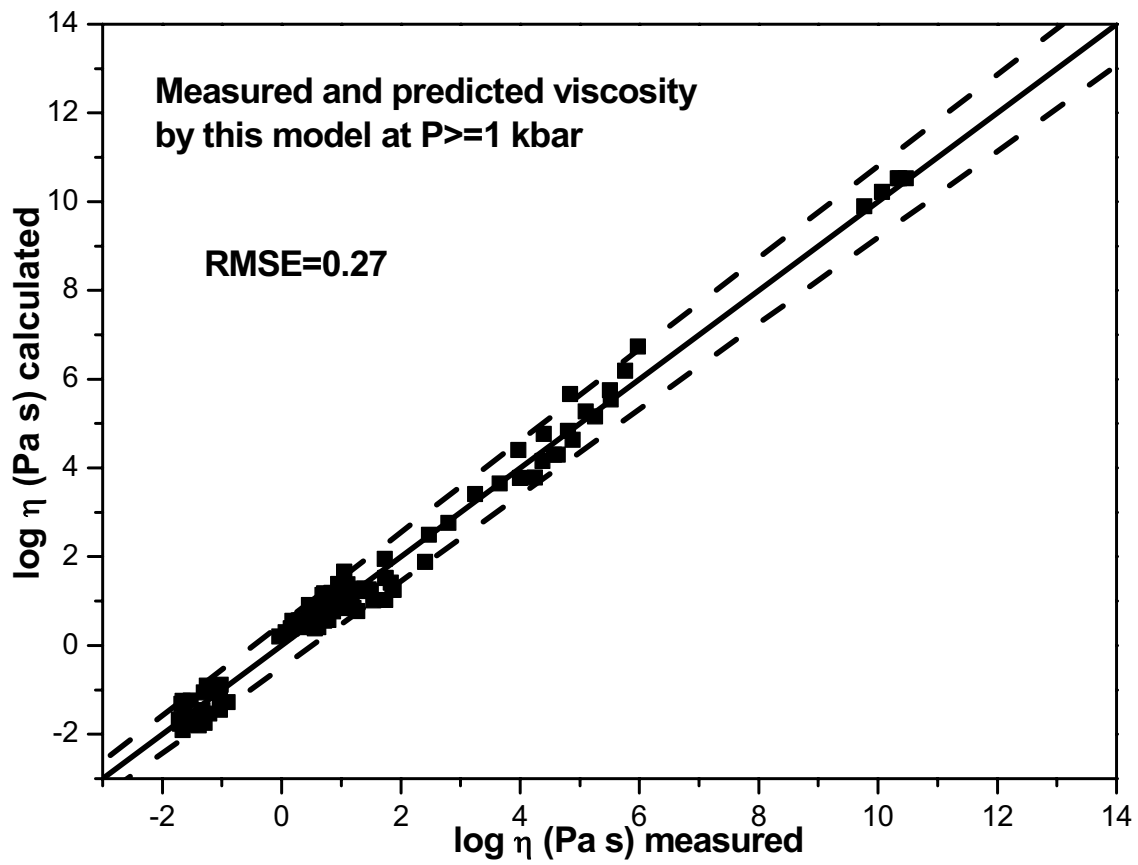


Fig.4

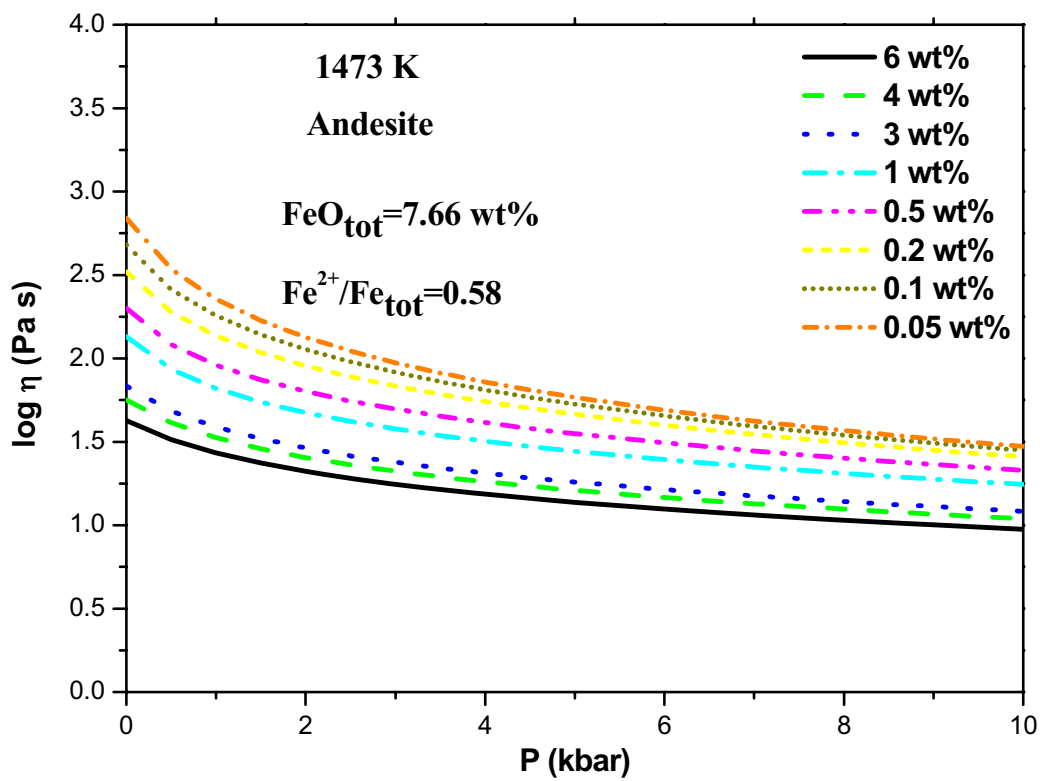


Fig.5

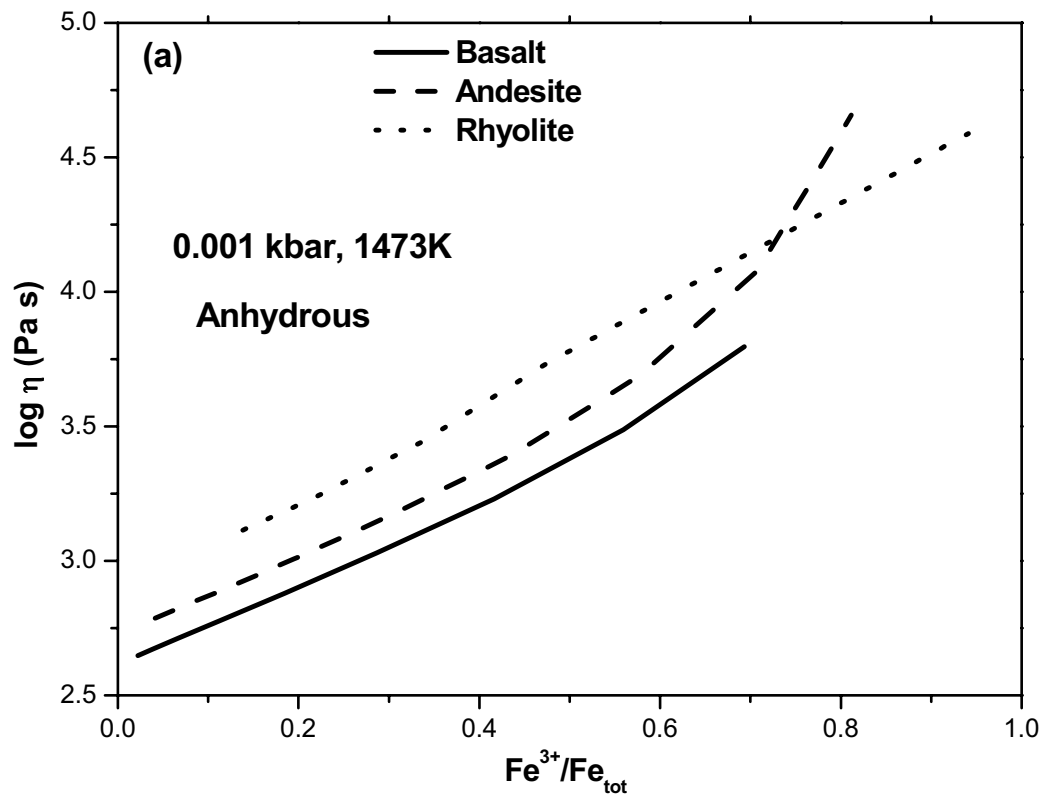


Fig.6a

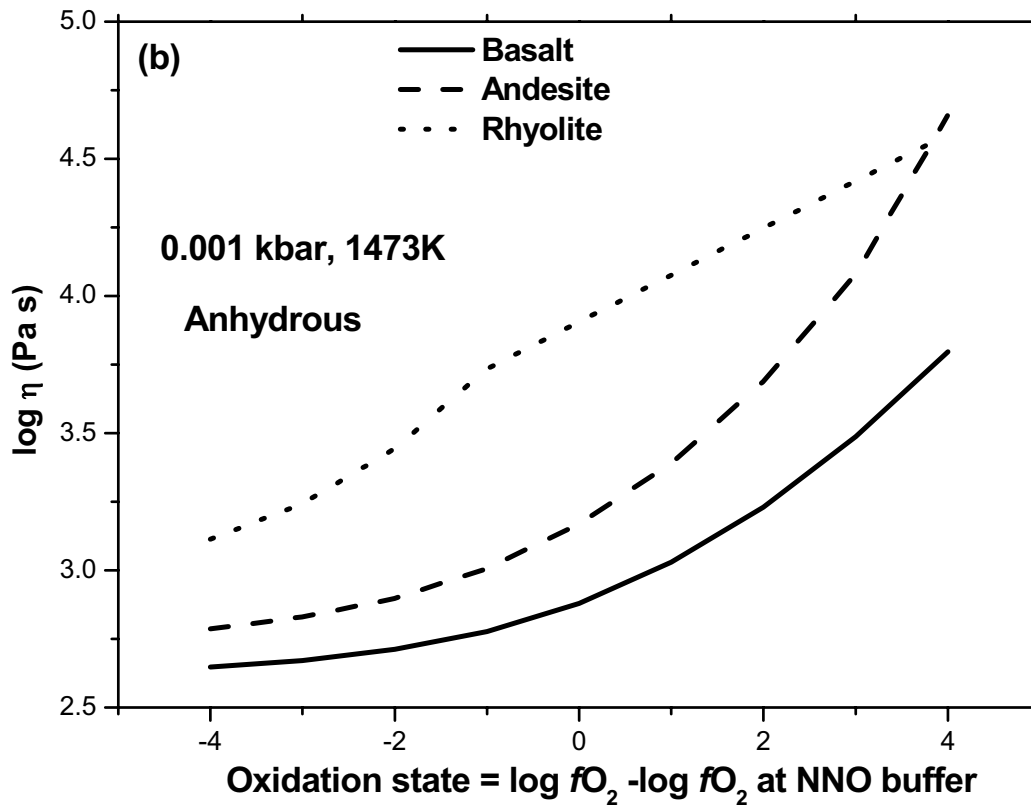


Fig. 6b

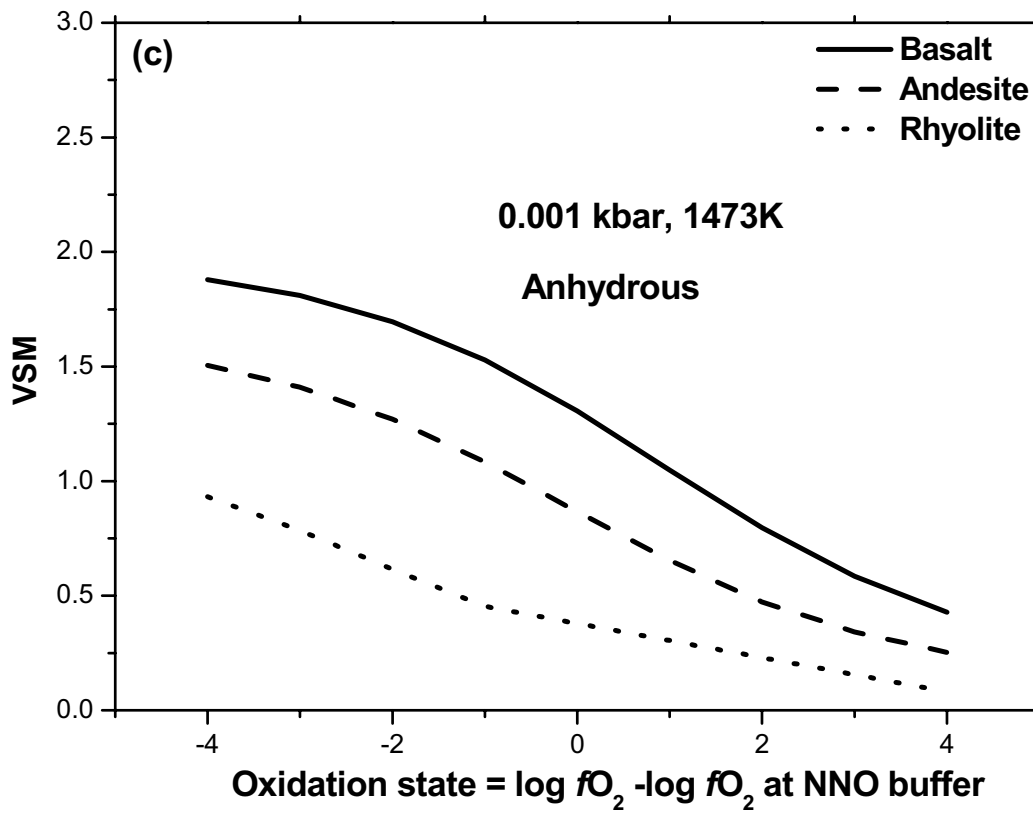


Fig. 6c

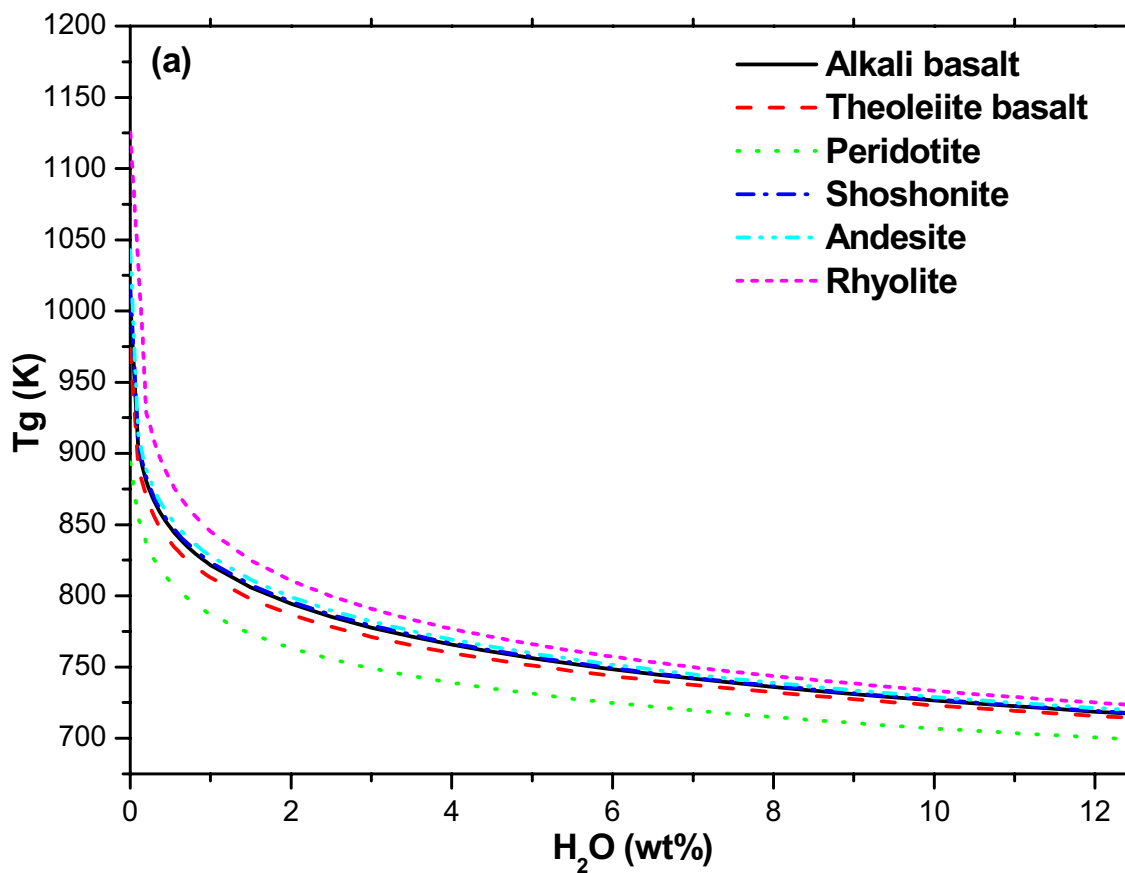


Fig.7a

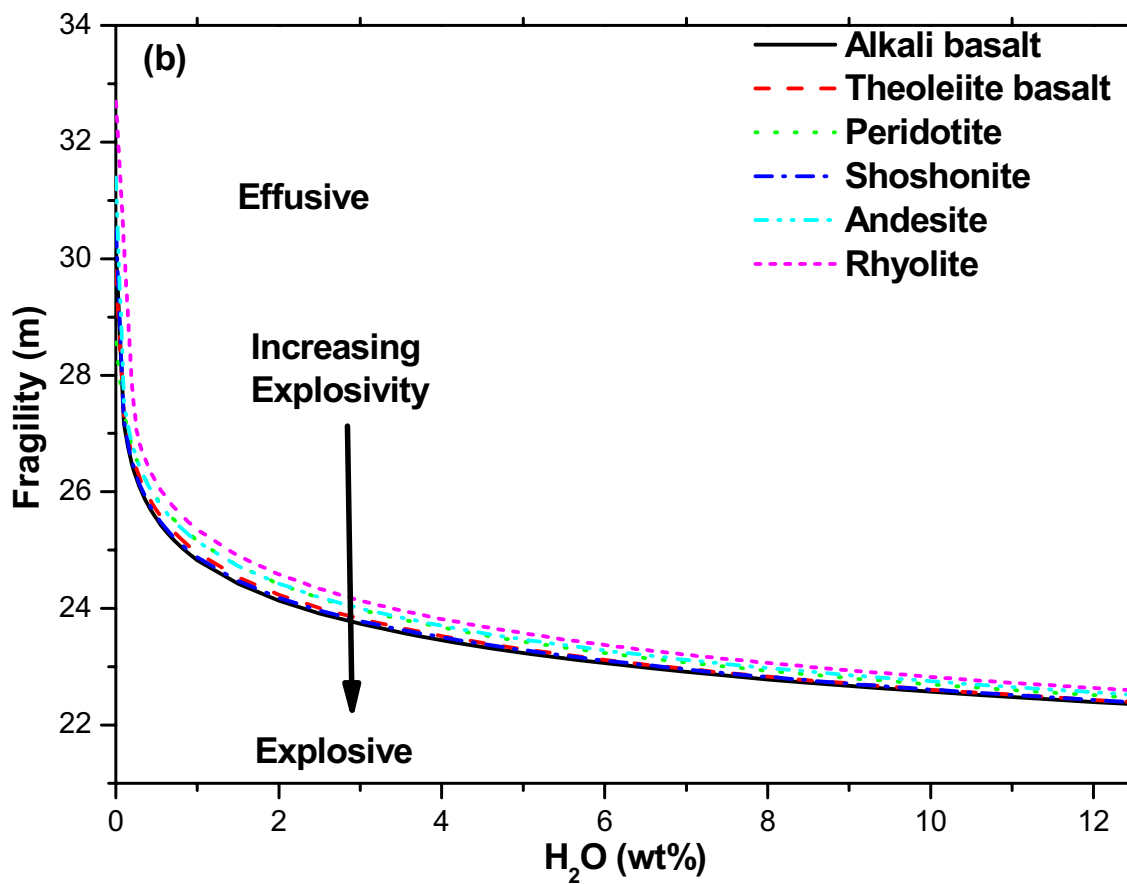


Fig. 7b