



HAL
open science

**Rheology of partially molten plagioclase containing wetting silica-rich anhydrous melt abbreviated title:
Rheology of partially molten plagioclase**

Alexandre Dimanov

► **To cite this version:**

Alexandre Dimanov. Rheology of partially molten plagioclase containing wetting silica-rich anhydrous melt abbreviated title: Rheology of partially molten plagioclase. 2021. hal-03334127

HAL Id: hal-03334127

<https://hal.science/hal-03334127>

Preprint submitted on 3 Sep 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18

Rheology of partially molten plagioclase containing wetting silica-rich anhydrous melt

abbreviated title: **Rheology of partially molten plagioclase**

Alexandre Dimanov

LMS, UMR7649, Ecole Polytechnique

Route de Saclay, 91128 Palaiseau, France.

Corresponding author: Alexandre Dimanov, *e-mail:* dimanov@lms.polytechnique.fr

19 **Abstract**

20 The present work explores the effects of melt chemistry on diffusion controlled creep of partially
21 molten labradorite plagioclase (An₅₀) at anhydrous conditions. Using sol-gel and hot pressing techniques
22 we produced: 1) nominally melt-free samples (Lab), with < 1 vol. % residual glass confined solely to
23 multiple grain junctions; 2) SilLab1 and SilLab5 partially molten samples containing respectively 1 and
24 5 vol. % excess amorphous silica, resulting in partial melts wetting numerous grain boundaries as thin (<
25 10 nm) amorphous films. Energy dispersive X-ray analysis showed that the amorphous phases in Lab,
26 SilLab1 and SilLab5 samples contained about ~ 70, ~ 85 and ~ 95 wt. % SiO₂, respectively. Infrared
27 spectroscopy showed that the initial traces of water (~ 0.05 wt. %) were dried out by annealing in air
28 above 1100°C. Uniaxial creep tests performed at 1100 – 1250°C and 3 – 60 MPa flow stresses showed
29 dominantly linear viscous flow, with a strong grain size dependence indicating grain boundary sliding and
30 diffusion control. Counter-intuitively strength and activation energy increased with the content of melts,
31 but in accord with the silica content of the latter, that is with their polymerization state. Our results show
32 that the kinetics of grain boundary diffusion controlled creep strongly depends on melt chemistry. Instead
33 of acting as short-cut for diffusion, thin films of highly viscous amorphous phases may in turn
34 considerably reduce grain boundary transport properties.

35

36 **Key words:** rheology, plagioclase, partial melt, thin melt films, grain sliding, diffusion creep.

37

38 **1. Introduction**

39

40 The rheological properties of partially molten silicates may substantially deviate from those of
41 their fully crystalline counterparts. The partially molten silicates are usually substantially weaker (Van
42 der Molen and Paterson, 1979; Vigneresse et al., 1996), but the magnitude of weakening may differ

43 depending on the mineralogy and chemistry of the considered system, as well on the proportion of partial
44 melt and its grain scale topology (Cooper and Kohlstedt, 1986; Hirth and Kohlstedt, 1995a; 1995b;
45 Kohlstedt and Zimmerman, 1996; Dimanov et al., 1998; 2000). Understanding the rheological properties
46 of partially molten silicates is therefore of fundamental importance for modelling the dynamics of various
47 tectonic settings. For example, the stability of the high plateau in the collision context of the Himalayan-
48 Tibetan orogen is proposed to be ensured by channel flow localized within the lower crust. The viscosity
49 of the latter is supposedly exceptionally low, in relation with partial melting (see a review by Harris,
50 2007). Another classical example is the accretion of the oceanic crust from the magma chambers beneath
51 the mid-ocean ridges. In this case, plagioclases and clinopyroxene crystallizing from the basaltic liquid
52 settle down from the crystalline mush at the bottom of the magmatic lens. The gabbroic material is further
53 driven apart from the ridge by the convection of the upper mantle. This lateral flow occurs at partially
54 molten state as demonstrated by the magmatic fabrics: bedding, shape preferred textures and lack of
55 crystalline plasticity (Nicolas and Ildefonse, 1996; Ildefonse and Nicolas, 1997). However, magmatic flow
56 is expected to occur for suspensions at relatively high liquid fractions (> 30 vol. %), which disagrees with
57 the estimations from seismic studies (Lamoureaux et al. 1999). Yet, based on microstructural observations
58 Nicolas and Ildefonse (1996) and Lamoureaux et al. (1999), suggested that magmatic flow could still
59 operate at low melt fractions (< 10 vol. %) provided flow is accommodated by grain sliding and
60 dissolution – precipitation processes. Such mechanisms have been experimentally observed by Dimanov
61 et al. (1998, 2000) for partially molten plagioclase (labradorite) aggregates presenting melt fractions
62 between 1 – 10 vol. %. The authors emphasized that the topology of the melt was critical with respect to
63 the flow properties. They reported that Newtonian type of flow could be strongly enhanced even for very
64 low melt fractions (1 – 3 vol. %) in the case where the partial melt was wetting most of grain boundaries.

65 Over more than two decades it was considered that the equilibrium melt distribution in a
66 polycrystalline aggregate is essentially driven by surface tension forces and gradients of chemical

67 potentials in order to minimise the interfacial free energy (Beere, 1975; Waff and Bulau, 1979, 1982;
68 Bulau et al., 1979; Cooper and Kohlstedt, 1984a; 1984b; 1986; Jurewitz and Watson, 1985; Von Bargen
69 and Waff, 1986; Fujii et al., 1986; Bussod and Christie, 1991; Kohlstedt, 1992; Laporte and Watson, 1995;
70 Laporte et al., 1997; Hirth and Kohlstedt, 1995a,b; Jung and Waff, 1998). From this perspective it can be
71 shown that the equilibrium melt topology at hydrostatic conditions in a partially molten aggregate
72 corresponds to a balance between interfacial tensions at triple points, where two neighbouring solid grains
73 are in contact with the melt. Considering single phase solid matrix, single grain size value, isotropic solid
74 – solid and solid – liquid interfacial energies (γ_{ss} and γ_{sl} , respectively), the latter equilibrium determines
75 an unique dihedral angle θ (the angle between the two solid – liquid interfaces of the two neighbouring
76 grains), which satisfies $2\cos(\theta/2) = \gamma_{ss}/\gamma_{sl}$. In addition, because gradients of chemical potentials of the
77 constituent species appear whenever the system presents solid – liquid interfaces with variable curvatures,
78 the chemical and textural equilibrium requires a constant mean curvature of all the solid – liquid interfaces.
79 In turns, this requirement ensures the minimization of solid – liquid interfacial area and implies concretely
80 that the crystalline phase in contact with the liquid does not present sharp corners and flat surfaces. In
81 summary of the previously cited works, we can consider that at low melt fractions for $\theta > 60^\circ$ the molten
82 phase resides in isolated pockets and interconnectivity occurs only above a critical melt fraction that
83 increases with θ . For $0^\circ > \theta > 60^\circ$ a given fraction of the whole amount of melt, called the equilibrium
84 melt fraction (that minimises the free interfacial energy of the system), forms an interconnected network
85 along three – , four – and higher order grain junctions, whilst all exceeding amount of melt distributes in
86 additional high aspect ratio pools. The smaller is θ and the larger is the proportion of grain boundary area
87 lost to the melt. Grain boundaries are expected to be completely wetted by melt only if $\theta = 0^\circ$. However,
88 since most dihedral angles were found to fall between $10 - 40^\circ$ in both mafic (olivine or peridotite and
89 basalt, Kohlstedt, 1992) and felsic (granitic rocks and silicic melts, Laporte et al., 1997) systems, two
90 grain boundaries were supposed to be melt-free. But despite the latter expectation, melt films spreading

91 at two grain boundaries were frequently found (Fujii et al., 1986; Bussod and Christie, 1991; Waff and
92 Faul, 1992; Laporte and Watson, 1995; Hirth and Kohlstedt, 1995a,b; Laporte et al., 1997; Faul, 1997;
93 Jung and Waff, 1998; Wark et al., 2003, Carapic et al., 2013). Moreover, instead of the expected smoothly
94 curved solid – liquid interfaces straight flat interfaces (called F – faces) were often observed, suggesting
95 that minimisation of interfacial free energy does not solely relate to minimisation of interfacial area. It
96 was argued that the deviations from the ideal melt topology could relate a) to the polymineralic nature of
97 some of the studied systems, b) to distributed grain sizes and ongoing grain growth, c) to the fact that
98 interfacial free energies and dihedral angles depend on the relative crystal orientations, because silicates
99 present anisotropic surface energies (Waff and Faul, 1992; Laporte and Watson, 1995; Jung and Waff,
100 1998; Cmíral et al, 1998; Schäfer and Foley, 2002, Walte et al., 2003). Theoretical considerations on bi-
101 crystals have shown that interfacial free energies i) are not necessarily much smaller than surface free
102 energies, ii) strongly depend on interfacial misfits, or structural defects (van der Merwe, 2001), and hence
103 on relative grain twist and tilt disorientation.

104 Conversely, non-hydrostatic conditions have also been considered to explain deviations from the
105 ideal melt topology. Melt – wetted grain boundaries were often reported in experimentally deformed
106 partial melts (Van der Molen and Paterson, 1979; Dell’Angelo et al., 1987; Dell’Angelo and Tullis, 1988;
107 Bussod and Christie, 1991; Jin et al., 1994; Hirth and Kohlstedt, 1995a,b; Drury and FitzGerald, 1996;
108 Daines and Kohlstedt, 1997; Zimmerman and Kohlstedt, 1999; De Kloe et al, 2000; Dimanov et al., 2000;
109 Mecklenburgh and Rutter, 2003; Hier-Majumder et al., 2004). This behaviour was attributed to the non-
110 equilibrium dynamic state and/or possibly to undrained conditions at high melt fractions. At high strains
111 major microstructural changes may also occur: melt initially restricted to multiple grain junctions may
112 segregate into high aspect ratio bands preferentially oriented at a small angle ($\approx 20^\circ$) to the direction of
113 maximum compressive stress (Zimmerman and Kohlstedt, 1999; Zimmerman et al., 1999; Holtzman et
114 al., 2003; 2007; Katz et al., 2006; Spiess et al., 2012).

115 High resolution transmission electron microscopy (HRTEM) studies documented stable thin
116 amorphous films (~ 1 – 10 nm) at grain boundaries in natural xenoliths (Wirth, 1996, Franz and Wirth,
117 1997), in deformed synthetic olivine – orthopyroxene (Drury and FitzGerald, 1996; De Kloe et al, 2000)
118 and plagioclase (Dimanov et al., 2000) aggregates. Many ceramic materials present the same scheme,
119 with stable nm – sized thin amorphous silica-rich films at grain boundaries (see a review by Wilkinson,
120 1998). It has been demonstrated that such films may be stable, because they tend to decrease the interfacial
121 free energy (Clarke, 1987; Clarke et al. 1993). Conversely, based on HRTEM Hiraga et al. (2002) report
122 that wetting of olivine grains by basaltic melt films is not a general feature. The authors indicate that stable
123 basalt films (~ 10 nm) are only observed along grain boundaries where the grains present low index
124 crystallographic planes.

125 In the specific case of diffusion-controlled creep of fine grained partially molten rocks with low
126 melt fractions (< 15 vol. %) the strength depends strongly on melt content, but also on the grain – scale
127 melt distribution. Theoretical (Raj, 1982; Pharr and Ashby, 1983; Cooper et al., 1989; Paterson, 2001)
128 and experimental studies on olivine (Cooper and Kohlstedt 1984; 1986; Kohlstedt, 1992; Hirth and
129 Kohlstedt, 1995a, b) and plagioclase (Dimanov et al., 1998; 2000) have shown that strain rates are
130 substantially enhanced if melt films spread along grain boundaries, whilst partial melts that are confined
131 to multiple grain junctions cause only moderate creep rate enhancement. Because diffusion in a molten
132 phase is often orders of magnitude faster than along grain boundaries (see a review in Dimanov et al.,
133 2000), the larger is the proportion of grain boundary lost to the melt, the larger is the proportion of
134 interfaces providing high-diffusivity paths, and the lower is the strength of the partially molten aggregate
135 (Cooper et al., 1989; Paterson, 2001). To date, the effect of melt chemistry on creep rates has essentially
136 been considered from the view point of the dependence of dihedral angles on melt composition
137 (Wanamaker and Kohlstedt, 1991; Kohlstedt, 1992; Wolfenstine and Kohlstedt, 1994). However, for a
138 given melt topology, melt composition might also influence transport properties. In a previous work we

139 have shown that, at least at the experimental conditions, thin amorphous films were stable in plagioclase
140 aggregates (Dimanov et al., 2000). Most grain boundaries being wetted, the flow strength of the
141 plagioclase aggregates deforming by diffusion controlled grain boundary sliding was substantially
142 depressed, event at very low melt fractions (~ 1 – 2 vol. %). Yet, the activation energy was found to be
143 similar for melt – bearing and nominally melt – free aggregates. Based on extant diffusion data in melts
144 and polycrystalline silicates Dimanov et al. (2000) suggested that in dry conditions the activation energy
145 might be similar for diffusion of network formers along plagioclase grain boundaries and in silica – and
146 aluminium – rich, plagioclase – like melts. In this study we further focus on the effect of the silica content
147 of the wetting intergranular molten phase on grain boundary diffusion controlled creep of partially molten
148 plagioclase aggregates.

149

150 **2. Experimental procedures**

151

152 **2.1. Specimen preparation**

153

154 We used fine grained (< 16 μm) labradorite glass powder (Corning Inc., kindly provided by R.C.
155 Cooper). It has $\text{An}_{50}\text{Ab}_{50}$ chemical composition and X – ray fluorescence (XRF, SRS 303AS) showed that
156 the impurity content was less than 1 wt. % (Tab. 1). After being stored at 120°C in oven for 24 hours the
157 powder was first uniaxially cold – pressed at 300 MPa in steel jackets of 20 mm in length, 10 mm in
158 diameter and 0.5 mm thick. Secondly, the green bodies were hot – isostatically pressed (HIPped) for 3
159 hours at 1150°C and 300 MPa in a gas-medium Paterson apparatus in order to obtain nearly glass – free
160 polycrystalline material, with about 1 vol. % residual glass (Dimanov et al., 1998; 1999; 2000; 2003;
161 Rybacki and Dresen, 2000). However, this procedure results sometimes in spherulites due to fast
162 crystallization kinetics (Rybacki and Dresen, 2000). Hence, the HIPed samples were re-crushed and

163 milled in alcohol in an agate mortar in order to obtain homogeneously grained crystalline powders. Laser
 164 – particle analysis (Fritsch analysette 22) showed that about 80% of the grains were < 10 μm . The larger
 165 particles were mostly due to clustering of smaller grains. The chemical composition of the powder was
 166 checked by XRF (Table 1). To remove adsorbed water the crystalline powders were annealed at 800°C
 167 for 24 hours in air and subsequently stored in an oven at 120°C prior to further processing.

168

169 Table 1. Composition of starting materials and partial melts

Element	An50 glass powder*	An50 crystal powder*	Sol-gel silica*	An50 residual glass**	SilLab5 melt**	SilLab1 melt**
Na ₂ O(wt.%)	5.68	5.16	0.04	5.5	1.1	2.7
Al ₂ O ₃ (wt.%)	28.43	29.07	0.23	29.0	3.3	8.7
SiO ₂ (wt.%)	55.27	54.82	99.53	53.0	94.8	85.4
CaO(wt.%)	10.52	10.81	0.12	12.0	0.7	2.2
MgO(wt.%)	0.25	0.31	0.06	0.3	0.0	0.0
FeO(wt.%)	0.03	0.04	0.0	0.2	0.2	0.5
Cr ₂ O ₃ (wt.ppm)	0.0	0.0	0.0	0.0	0.0	0.3
ZrO ₂ (wt.ppm)	173	166	0.0	0.0	0.2	0.1
Total(wt.%)	100.2	100.22	98.98	100	100.3	99.9

170 * XRF analysis of powders, **EDX analysis in TEM

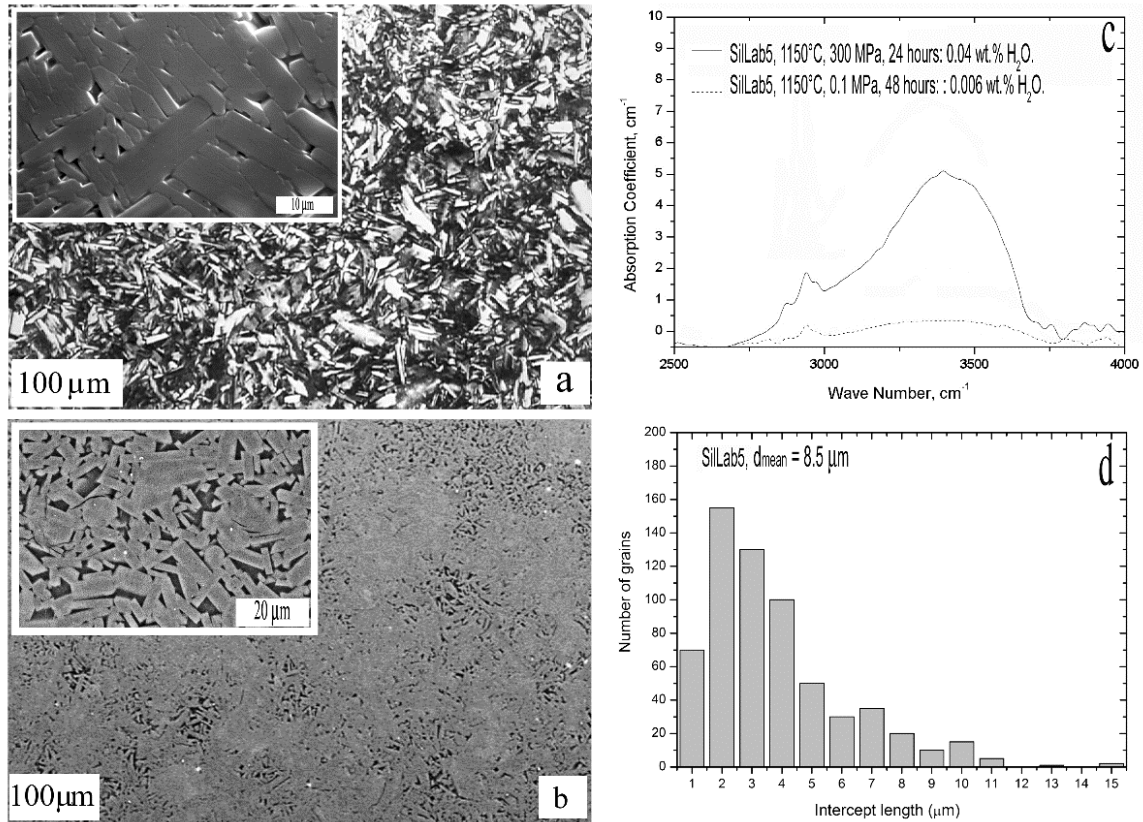
171

172 **2.1. a. Melt – “free” samples**

173

174 Crystalline powders were subjected to a second round of cold pressing, followed by HIPing for 24
 175 hours at 1150°C and 300 MPa in order to obtain dense, melt – free polycrystalline materials, further
 176 referred as Lab samples. Scanning electron microscopy (SEM, Zeiss DSM 962 and Jeol JSM 845) and the
 177 line intercept method were applied on carefully polished and thermally etched specimens (Dimanov et al.,
 178 1998, 1999, 2000; 2003). Figure 1a shows the obtained microstructures, with lath shaped grains
 179 characterized by high aspect ratios and log – normal grain size distribution. We determined a mean
 180 arithmetic grain size of $9.8 \pm 3.2 \mu\text{m}$ from five different SEM micrographs. Transmission electron
 181 microscopy (TEM, Philips CM 200 Twin) showed that grains present low average dislocation densities
 182 and numerous growth twins (Fig. 2a).

183

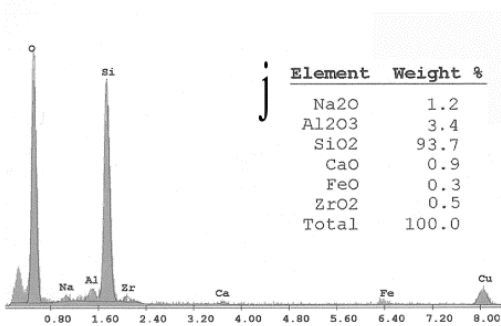
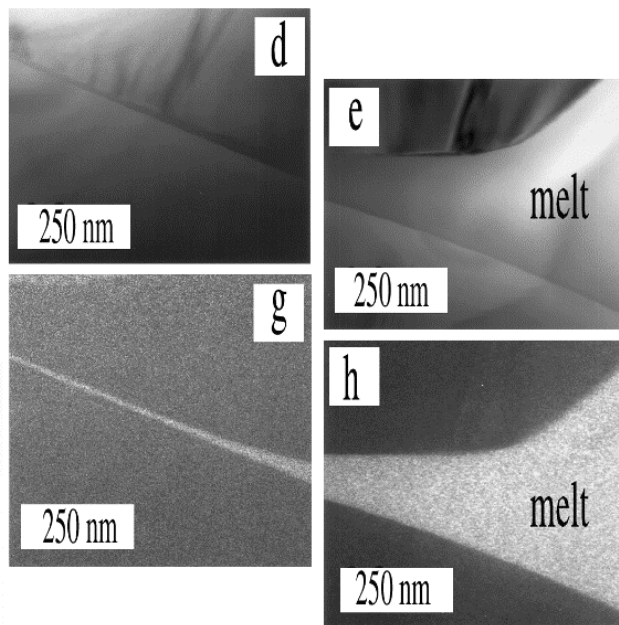
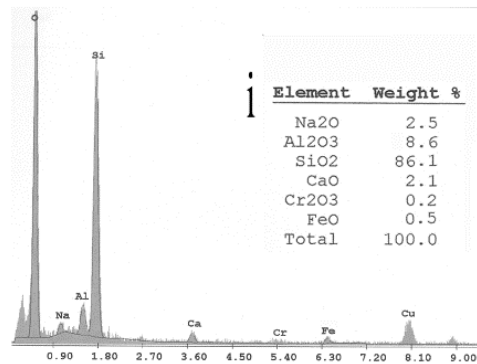
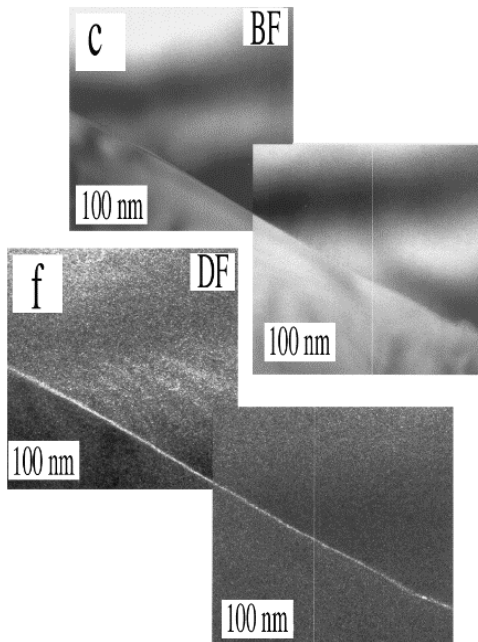
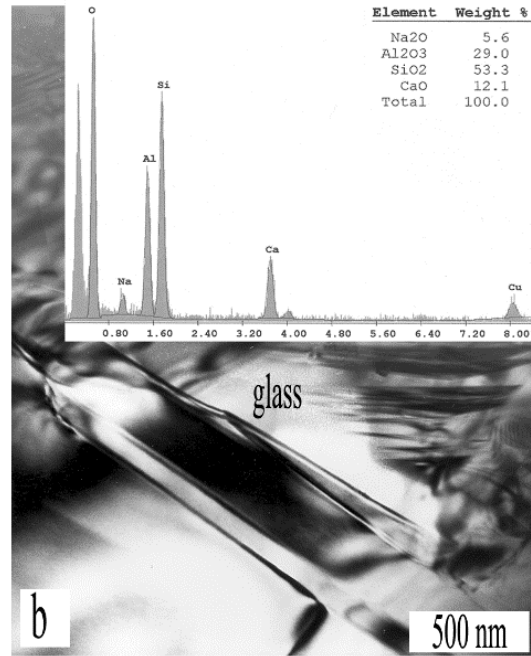


184

185 Fig. 1: Characterization of microstructures at optical and SEM scales. a) Optical micrograph of large representative area from
 186 thin section and SEM micrograph (insert, SE mode) of polished and thermally etched melt - free specimen (Lab). b) SEM
 187 micrographs (BSE mode) of polished melt - bearing specimen (SiLab5). At mm-scale melt distribution is somewhat
 188 heterogeneous, with local melt fractions as high as 10 vol. % (insert). c) Water content after initial HIP and after drying anneal
 189 from FTIR measurements on SiLab5 samples d) Grain size distribution for SiLab5 samples obtained by the intercept method
 190 performed on SEM micrographs.

191

192 As previously observed by Dimanov et al., (1998), small amount (< 1 vol. %) of residual glass persisted,
 193 but it was essentially restricted to multiple grain junctions (Fig. 2b). Fluid inclusions were also observed
 194 within crystals, at grain boundaries and in the glassy-phase. Fourier transform infra-red spectroscopy
 195 (FTIR, Brüker IFS 66v) was applied on 5x5 mm² doubly polished sections of 150 μm in thickness. We
 196 operated in the range 2000 - 4000 cm⁻¹ and we applied a third - order polynomial fit for background
 197 correction.



199 Fig. 2: Characterization of microstructures at TEM scale. a) Grains exhibit low dislocation densities, but growth twins are
200 numerous. b) Residual glass pockets (< 1 vol.%) are commonly found at multiple grain junctions in Lab samples. EDX
201 analyses (insert) show that their composition is comparable to plagioclase. c-h) SilLab1 and SilLab5 materials present
202 amorphous pockets at multiple junctions, but most interfaces also show thin amorphous films (< 10 nm) extending from triple
203 junction channels, as evidenced by bright versus dark field imaging. i-j) EDX analysis of amorphous phases performed at melt
204 pockets located at multiple grain junctions in SilLab1 and SilLab5 samples.

205

206 As in previous studies (Dimanov et al., 1998; 1999; Rybacki and Dresen, 2000) we observed a
207 broad absorption band centered at about 3350 cm^{-1} (Fig. 1c), indicating free molecular water present in
208 intra – and inter – crystalline fluid inclusions, and/or dissolved within the residual glass and at grain
209 boundaries. But, a few sharper peaks indicated that hydrous species were also incorporated within the
210 crystalline structure (Hofmeister and Rossman, 1985; Beran, 1986; 1987). We calculated the water content
211 of the samples (in mol $\text{H}_2\text{O}/\text{L}$ labradorite), based on Paterson (1982), and using the linear molar
212 absorptivity coefficient for plagioclase (32 L/mol/cm , Beran, 1987). For that purposes, the integral of the
213 spectrum is normalised by the half-width of the absorption band. Assuming a Gaussian approximation,
214 this operation gives the height of the band, which is used together with the linear absorbtivity in the Beer-
215 Lambert's law (see also Dimanov et al., 1998, 1999; Rybacki and Dresen, 2000). The samples contained
216 $\approx 0.05 \pm 0.025\text{ wt. \% H}_2\text{O}$ ($\approx 6000 \pm 3000\text{ ppm H/Si}$), which is mostly incorporated by adsorption onto
217 the starting glass particles, and later onto the crystalline particles, during the temporary storage and
218 handling at room condition for cold – pressing. Annealing of the samples for 24 – 48 hours in air at
219 temperatures above 1100°C at 0.1 MPa resulted in drying out most of the water traces down to < 0.002
220 $\text{wt. \% H}_2\text{O}$ (Fig.1, see also Dimanov et al., 1999).

221

222 **2.1.b Melt – bearing samples**

223 Dimanov et al. (2000) annealed plagioclase samples (similar to those used in this study) at
224 hypersolidus conditions. The authors obtained partially molten specimens containing a few percent of
225 silica – enriched melt, with up to 80 wt. % SiO₂. In addition to multiple grain junctions the melt was
226 present as thin films (< 5 nm) wetting many grain boundaries. The samples were about an order of
227 magnitude weaker than melt-free specimens. In this study we aimed to investigate the effects of
228 homogeneously distributed and wetting melts with higher silica contents. For this purpose we applied the
229 sol – gel method (Hamilton and Henderson, 1968), which was previously used to precipitate synthetic
230 basalt onto olivine crystalline powders (Cooper and Kohlstedt, 1984; Beeman and Kohlstedt, 1993; Hirth
231 and Kohlstedt, 1995a). We used pure grade tetraethyl orthosilicate (Si(C₂H₅O)₄, TEOS from Aldrich)
232 diluted with ethanol in proportions of 10 to 1. Ammonium hydroxide solution (1 mol/l, Prolabo) was used
233 as catalyst. In the solution, water hydrolyzes the TEOS and produces silanol groups (Si-OH), which
234 interact through hydrogen bonding and form hydrolyzed silica. After drying out all volatiles, we
235 performed XRF analysis on the precipitation product that consisted of nearly pure silica (Table 1). We
236 prepared two aqueous solutions, where labradorite powders and diluted TEOS were mixed at the
237 appropriate proportions in order to obtain 1 and 5 vol. % excess silica. The precipitation took place within
238 a few minutes after the addition of the ammonium hydroxide, but the suspensions were continuously
239 stirred for an hour. The precipitation products were dried at 100°C for 24 hours and ground in agate mortar.
240 The resulting powders were fired into platinum crucibles at 700°C (in air) for 24 hours in order to release
241 remaining volatiles. Finally, the silica – coated powders were stored in an oven at 120°C. The coated
242 crystalline powders were cold pressed and HIPed for 24 hours at 1150°C and 300 MPa in order to obtain
243 dense and equilibrated melt – bearing polycrystalline samples for deformation. Samples containing 1 and
244 5 vol. % excess silica glass are called SilLab1 and SilLab5, respectively. SEM investigations were applied
245 to carefully polished and thermally etched specimens (Fig. 1). The microstructure is comparable to those
246 of melt – free samples, with prismatic and lath shaped grains, and with log – normal grain size distributions

247 (Fig. 1d). The intercept method determined mean arithmetic grain sizes of 8.6 ± 3.4 and 9.3 ± 3.2 μm for
248 SilLab5 and SilLab1 samples, respectively. The amorphous phases are more or less homogeneously
249 distributed at multiple grain junctions, as evidenced by SEM in back scattering mode (Fig. 1b). TEM
250 consistently showed that the amorphous phases are present at most multiple grain junctions, with variable
251 apparent dihedral angles. Additionally, most two grain boundaries are wetted by thin amorphous layers <
252 10 nm in thickness. These features are shown in Figures 2c to 2h. In spite of apparently large dihedral
253 angles melt films extend from triple junctions within grain boundaries. In addition, the melt film
254 thicknesses gradually increase in the vicinity of the triple junctions, hence suggesting that there are not
255 finite wetting angles. These observations are in part similar to those of Dimanov et al. (2000) for partially
256 molten plagioclase and of Hiraga et al. (2002) for olivine – basalt system, but we did observe some
257 differences too. In the present study we observed that 1) the melt thickness stabilizes at around 10 nm at
258 distances of less than a micrometer from the triple junction, 2) the melt films are a general feature and not
259 exclusively related to low index crystallographic planes as in the latter study. Energy dispersive X ray
260 analysis (EDX) was performed at triple junctions and evidenced that during HIPing the precipitated silica
261 equilibrated chemically with the residual labradorite glass (≈ 1 vol. %), resulting in liquid phases with \approx
262 95 ± 2 wt. % and $\approx 85 \pm 2$ wt. % SiO_2 , respectively for SilLab5 and SilLab1 samples (Fig. 2i and 2j, Tab.
263 1). TEM also evidences the presence of nm scale fluid inclusions in the amorphous phases. FTIR
264 spectroscopy showed similar broad absorption bands and corresponding hydroxyl concentrations (≈ 0.05
265 ± 0.025 wt. % H_2O) for both melt – bearing and melt – free samples (Fig. 1c). The traces of water could
266 be substantially dried out by annealing in air above 1100°C for 48 hours (Fig. 1c).

267

268 **2.2. Experimental deformation**

269

270 Specimens with dimensions of $2.5 \times 2.5 \times 5 \text{ mm}^3$ were cut from hot pressed samples with low speed
271 diamond saw. Compression and observation surfaces were polished to $1 \text{ }\mu\text{m}$ and $0.3 \text{ }\mu\text{m}$ finish,
272 respectively. Uniaxial creep tests were performed stepwise at constant temperatures and constant loads in
273 a dead-load apparatus (Dimanov et al., 1998; 1999; 2000, 2003). Finite strains for individual steps ranged
274 from 3 to 0.5 %. We used alumina pistons and spacers, with thin platinum foils in order to avoid chemical
275 and to limit frictions. The fixed part of the deformation column was isolated from the outer atmosphere
276 by o – ring tightened alumina tubing, whilst the mobile part benefited from a frictionless oil bath sealing.
277 Dry argon flowed continuously in the furnace tubing. Sample shortening was measured using two linear-
278 variable displacement transducers (LVDT), which inner cores were mounted on two alumina rods ending
279 respectively at the top and bottom spacers. Accordingly, the differential measurements were not affected
280 by temperature fluctuations and allowed for strain rates lower than 10^{-8} s^{-1} . Stress and temperature steps
281 were performed between 3 – 60 MPa and 1100 – 1225°C. Steady state creep rate was generally achieved
282 within less than 0.5 % strain. Total sample strain was less than 15 %. Samples were cooled under load at
283 a rate of $50^\circ/\text{min}$ was in order to preserve at best the in-situ melt topology. However, due to the furnace
284 inertial rapid cooling (within less than 10 mn) was only achieved down to about $900 \text{ }^\circ\text{C}$, whilst further
285 cooling down to room temperature lasted for about two hours.

286

287 **3. Results**

288

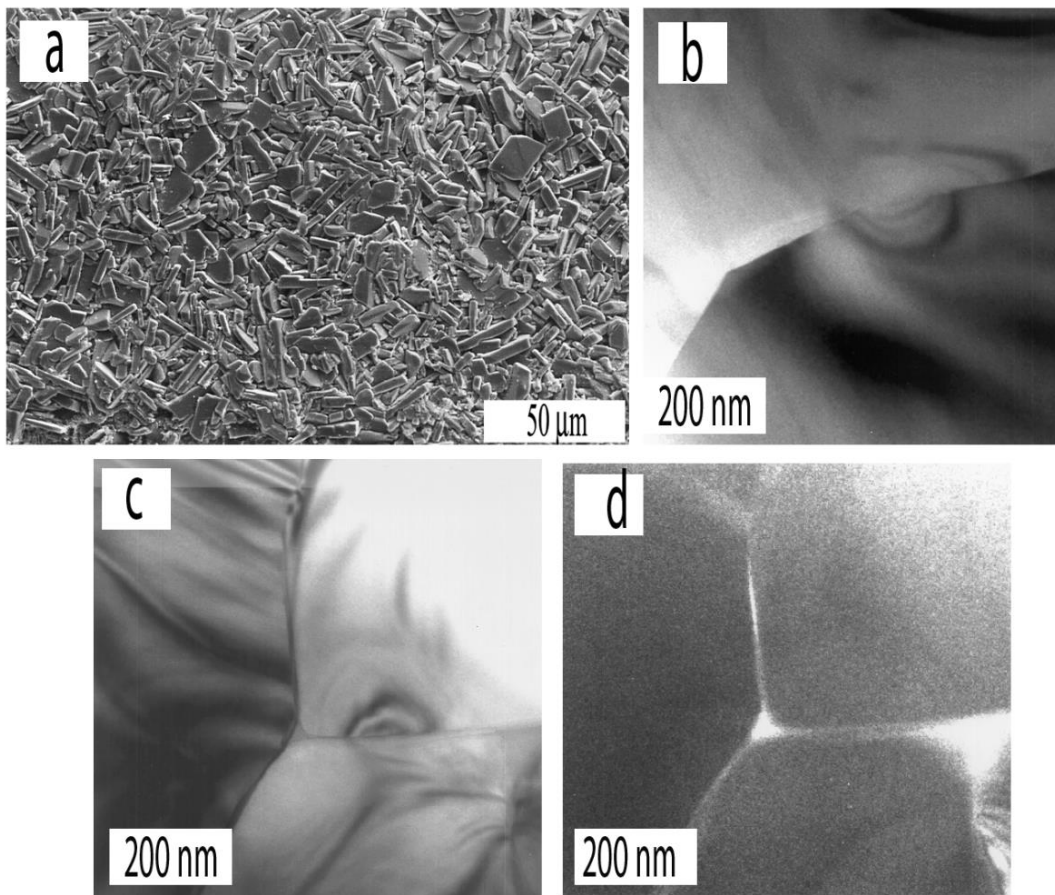
289 **3.1. Microstructures**

290

291 SEM observations did not show any shape preferred orientation of grains in deformed samples.
292 SEM and TEM observations and EDX analysis did not show obvious evolution of melt proportion (within
293 1 vol. %), topology, or chemistry (within 2 wt. %) during creep. For Lab samples the small amount of

294 residual glass was never found along grain boundaries. There was no indication for dynamic wetting of
 295 grain boundaries during deformation as observed sometimes in olivine-basalt assemblages (Jin et al.,
 296 1994; Hier-Majumder et al., 2004). For SilLab1 and SilLab5 samples the initial melt distribution did not
 297 obviously change either: most of the melt still resided at multiple grain junctions and the thin melt films
 298 were still present at most grain boundaries. Yet, we could evidence some local redistribution of melt along
 299 wetted interfaces.

300



301

302 Fig.3: SEM and TEM micrographs of deformed SilLab1 material. a) SEM shows topography development on the sample
 303 surface, indicating grain boundary sliding processes. b-c) Bright field images showing dislocation-free grains and whirl shaped
 304 strain contrasts next to melt filled pockets (b) or along interfaces (c), which indicate local residual stress related to “dry”
 305 contact area (see text). d) Dark field image corresponding to c) showing the presence of amorphous material at interfaces.

306

307 Figure 3 shows that along some wetted interfaces the grains present hemispherical strain contrast
308 features. According to Burger et al. (1997), such “strain whirls” result from residual elastic strain,
309 corresponding to local stress enhancement associated with the establishment of local dry contacts along
310 wetted interfaces during creep of glass-ceramic systems. This local phenomenon may be compared with
311 the theoretical island structure proposed by Raj (1982). However, we did not observe massive
312 redistribution of the melt as observed by Jin et al. (1995) in crept glass-ceramic samples or complete melt
313 expulsion out of normally stressed grain boundaries as described for olivine–basalt systems (Jin et al.,
314 1994). Our observations rather support previous authors arguing about the stability of thin amorphous
315 interfacial phases in both static (Clarke, 1987; Hess, 1994) and dynamic conditions (Drury and FitzGerald,
316 1996).

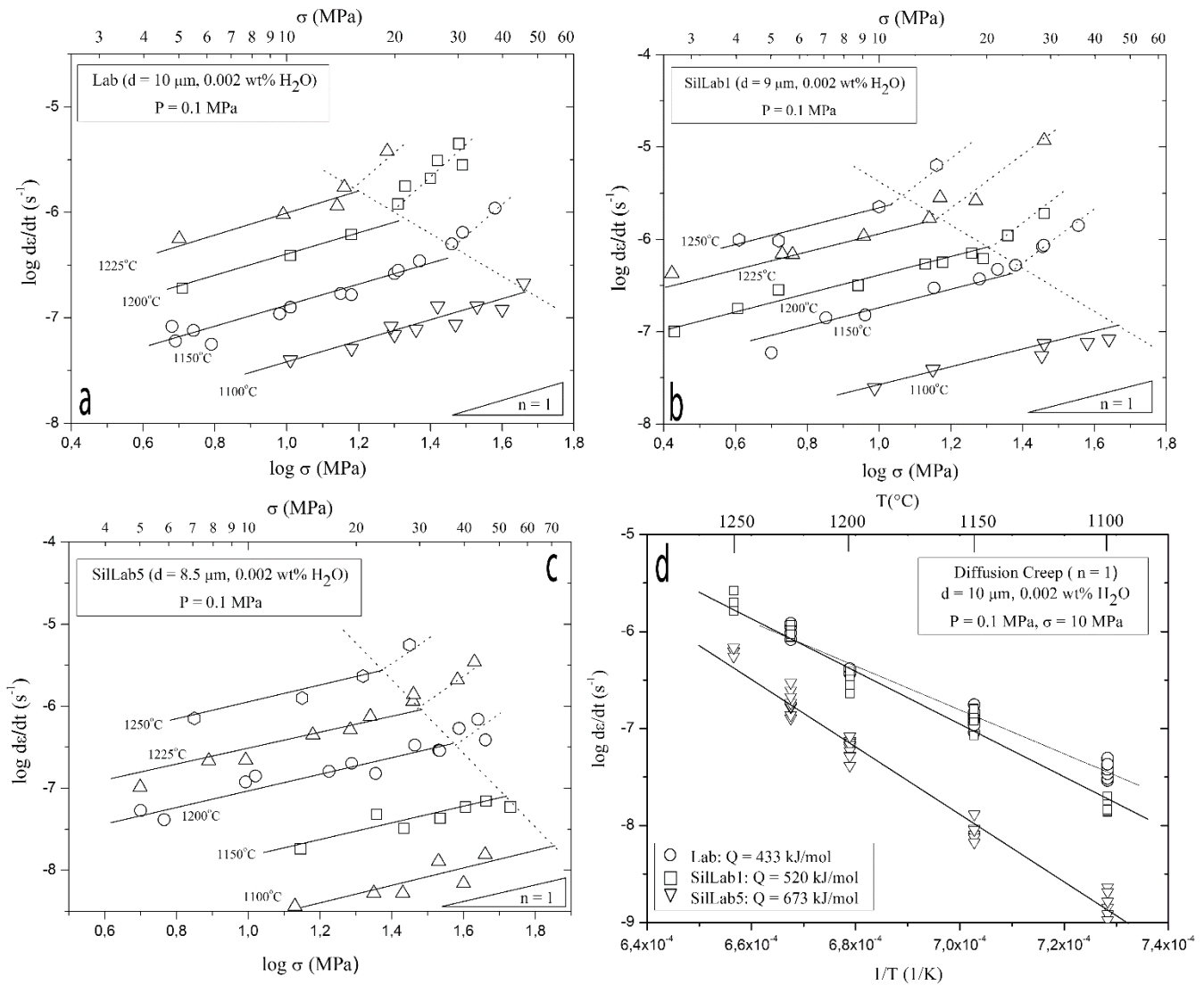
317 Curved free dislocations and dislocation arrays could be occasionally observed, but usually
318 dislocation densities, ρ , remained low ($\rho \approx 10^{10} - 10^{11} \text{ m}^{-2}$), which is consistent with Newtonian viscous
319 flow operating by grain sliding accommodated by diffusional mass transfer mechanisms. Growth twins
320 are very common in undeformed specimens, hence it is difficult to estimate if mechanical twinning could
321 contribute to deformation. In opposition to polished samples which simply experienced static thermal
322 etching (see Fig. 1), deformed samples present a strong development of surface topography (Fig. 3). Some
323 previous studies on high temperature creep of fine grained zirconia based ceramic materials (Clarisse et
324 al., 2000; Duclos et al., 2002) and superplastic alloys (Huang and Langdon, 2002) report the development
325 of grain scale topography onto the sample free surfaces, which characteristic was interpreted as evidence
326 for grain sliding.

327

328 **3.2. Mechanical Data**

329

330 **3.2.1. Stress sensitivity**



331

332

333

334

335

336

337

338

339

340

341

Fig. 4: Thermo-mechanical data for all tested materials presented in log Stress - log Strain rate plots (a, b and c for Lab, SilLab1 and SilLab5 samples respectively) and in Arrhenius diagram (d). All materials show dominantly linear viscous (Newtonian) flow (stress exponent $n = 1$) and a transition (indicated by dot-line) to non-linear behaviour (dislocation creep) at the highest temperatures and stresses, which is in agreement with previous works on similar materials (Dimanov et al., 1998; 1999; 2000). Data corrected for grain size (see text) and recalculated to 10 MPa differential stress are used to establish the Newtonian flow laws (d). Lab and SilLab materials present comparable strain rates, whilst SilLab5 material is considerably more resistant. Activation energy increases with melt content.

342 Table 2. Mechanical data.

Sample	T(°C)	T(h)	σ_{diff} (MPa)	d ϵ /dt (1/s)
lab-01	1150	31	5.48	7.52×10^{-8}
	1150	13	10.34	1.25×10^{-7}
	1150	8.4	15.25	1.66×10^{-7}
	1150	2.4	20.49	2.82×10^{-7}
	1150	1.5	30.54	6.41×10^{-7}
	1200	0.8	21.34	1.79×10^{-6}
	1200	0.5	26.20	3.06×10^{-6}
	1200	0.5	31.06	2.82×10^{-6}
	1150	25	4.76	8.36×10^{-8}
	1150	3.5	20.01	2.65×10^{-7}
lab-02	1100	17.2	26.19	1.30×10^{-7}
	1100	23	19.43	8.33×10^{-8}
	1100	19.4	22.79	7.75×10^{-8}
	1100	15.2	34.13	1.29×10^{-7}
	1100	8.4	45.36	2.15×10^{-7}
lab-03	1100	66	10.14	4.02×10^{-8}
	1100	21	14.98	5.17×10^{-8}
	1100	24.7	19.82	6.89×10^{-8}
	1100	24	29.73	8.62×10^{-8}
	1100	8.5	39.64	1.21×10^{-7}
	1200	4.8	5.16	1.90×10^{-7}

	1200	1.6	14.98	6.20×10^{-7}
	1200	2.22	10.29	3.88×10^{-7}
	1200	0.97	20.28	1.19×10^{-7}
	1200	0.48	25.12	2.07×10^{-7}
	1200	0.23	29.96	4.43×10^{-7}
lab-05	1150	20	4.92	6.01×10^{-8}
	1150	24.4	9.48	1.10×10^{-7}
	1150	6.4	14.08	1.69×10^{-7}
	1150	2.0	28.61	5.03×10^{-6}
	1150	18	6.17	5.62×10^{-8}
	1150	3.12	23.70	3.50×10^{-7}
	1150	1.13	38.09	1.08×10^{-6}
	1225	2.5	5.01	5.60×10^{-7}
	1225	1.5	9.84	9.53×10^{-7}
	1225	0.72	14.31	1.74×10^{-6}
	1225	0.41	19.18	3.79×10^{-6}
	1225	0.48	13.79	1.14×10^{-6}
SiLab1-01	1150	21	7.11	1.41×10^{-7}
	1150	3.7	14.23	2.98×10^{-7}
	1150	3.23	21.34	4.73×10^{-7}
	1150	1.5	28.59	8.63×10^{-7}
	1150	15	9.15	1.51×10^{-7}
	1150	1.78	28.79	8.33×10^{-7}

	1150	1.83	35.91	1.44×10^{-6}
silab1-02	1200	4	4.04	1.78×10^{-7}
	1200	16	2.69	9.97×10^{-8}
	1200	2.77	8.74	3.13×10^{-7}
	1200	1.14	13.45	5.37×10^{-7}
	1200	1.15	15.02	5.60×10^{-7}
	1200	1.45	18.16	7.10×10^{-7}
	1200	1.01	22.87	1.09×10^{-6}
silab1-03	1100	30	9.72	2.45×10^{-8}
	1100	24.2	14.14	3.93×10^{-8}
	1100	14	28.37	5.47×10^{-8}
	1100	11.4	13.45	7.59×10^{-8}
	1225	2.63	2.65	4.27×10^{-7}
	1225	1.33	5.74	6.85×10^{-7}
	1225	0.75	9.06	1.08×10^{-6}
	1225	1.45	13.79	1.68×10^{-6}
	1225	1.01	18.56	2.62×10^{-6}
silab1-04	1200	3.1	5.25	2.82×10^{-7}
	1200	1.9	19.5	6.17×10^{-7}
	1200	1.1	28.84	1.91×10^{-6}
	1100	12	28.84	7.41×10^{-8}
	1100	10	43.65	8.32×10^{-8}
	1150	14.1	5.01	5.89×10^{-8}

	1150	2.8	19.06	3.72×10^{-7}
	1150	1.4	23.99	5.25×10^{-7}
	1225	1.2	5.37	6.92×10^{-7}
	1225	0.8	14.79	2.82×10^{-6}
	1225	0.14	28.84	1.18×10^{-5}
	1225	2.4	4.07	9.87×10^{-7}
	1225	2.2	5.25	9.67×10^{-7}
	1225	1.2	10	2.26×10^{-6}
	1225	0.2	14.45	6.37×10^{-6}
silab5-01	1100	22.4	22.63	5.25×10^{-9}
	1100	28.3	34.12	1.28×10^{-8}
	1100	36	46.17	1.54×10^{-8}
	1150	24.2	22.8	4.81×10^{-8}
	1150	20.5	34.31	4.26×10^{-8}
	1150	27	45.99	6.95×10^{-8}
	1200	30.8	5.84	4.16×10^{-8}
	1200	8.2	16.79	1.61×10^{-7}
	1200	4.7	22.63	1.52×10^{-7}
	1200	2.35	33.94	2.91×10^{-7}
	1200	3.2	45.8	3.90×10^{-7}
	1200	7.15	34.12	2.87×10^{-7}
silab5-02	1200	23.2	5.00	5.37×10^{-8}
	1200	17.0	9.84	1.19×10^{-7}

	1200	4.1	19.39	2.01×10^{-7}
	1200	2.55	29.08	3.36×10^{-7}
	1200	2.65	38.62	5.33×10^{-7}
	1200	2.5	43.62	6.11×10^{-7}
	1225	10.2	5.0	1.03×10^{-7}
	1225	3.00	9.84	2.20×10^{-7}
	1225	3.2	19.23	5.16×10^{-7}
	1225	1.0	28.77	1.15×10^{-6}
	1225	0.65	38.32	2.09×10^{-6}
silab5-03	1100	163	13.64	3.63×10^{-9}
	1100	125	26.65	5.26×10^{-9}
	1100	55.8	40.08	6.88×10^{-9}
	1150	61.6	14.01	1.83×10^{-8}
	1150	27	27.15	3.27×10^{-8}
	1150	25	40.29	5.89×10^{-8}
	1150	20.6	53.8	5.83×10^{-8}
silab5-04	1225	19.2	7.83	2.17×10^{-7}
	1225	4.3	14.99	4.47×10^{-7}
	1225	2.5	21.8	7.50×10^{-7}
	1225	1.1	28.84	1.39×10^{-6}
	1225	0.27	42.92	3.46×10^{-6}
silab5-05	1250	3.0	7.01	7.10×10^{-7}
	1250	1.0	14.01	1.60×10^{-6}

	1250	0.5	21.02	2.33×10^{-6}
	1250	0.2	28.25	5.60×10^{-6}

343

344

345

346

347

348

349

350

351

352

353

354

355

356

357

358

359

360

361

362

Table 2 reports the experimental data set. Figures 4a, 4b and 4c report the diagrams of log strain rates versus log flow stresses obtained at different temperatures for the different types of materials. All materials showed dominantly a linear relation between strain rates and flow stresses (stress exponent $n \approx 1$), evidencing Newtonian linear viscous flow. Thought, a transition to a power law creep ($n \approx 3$) could be marginally observed at the highest stresses. Depending on temperature and material type the transition stress (σ_t) ranges between $\sigma_t \approx 20 - 60$ MPa. These results are phenomenologically similar to those previously reported for fine grained plagioclase, clinopyroxene and plagioclase-clinopyroxene aggregates (Dimanov et al., 1998; 1999; 2003; Rybacki and Dresen, 2000; Bystricky and Mackwell, 2000; Dimanov and Dresen, 2005; Hier-Majumder et al., 2005), and olivine aggregates (Hirth and Kohlstedt, 1995; Mei and Kohlstedt, 2000; Mei et al., 2002). They indicate two dominant creep mechanisms, which are usually considered as being independent and acting in parallel, with overall dominance of the most effective one. The linear viscous flow that dominates at low stresses is grain size sensitive (gss) and often advocated as being diffusion-controlled creep, whilst the highly non-linear viscous flow that dominates at higher stresses is usually found to be nearly grain size insensitive (gsi) and is attributed to dislocation creep. It has been evidenced that for heterogeneous and/or polyphase materials these mechanisms may be locally concomitant and co-operational (Dimanov et al., 2011), but for homogeneous single phase silicates they are consensually considered as mutually independent. The general flow law resulting from the parallel contribution of both mechanisms is therefore of the form:

$$\frac{d\varepsilon}{dt} = A_{gss} \sigma d^{-m} e^{-\frac{Q_{gss}}{RT}} + A_{gsi} \sigma^n e^{-\frac{Q_{gsi}}{RT}} \quad (1)$$

363 where $\frac{d\varepsilon}{dt}$ (s^{-1}) is the strain rate, σ (MPa) is the differential stress, n (usually between 3 and 5) is the stress
 364 exponent for the dislocation creep component, d (μm) is the grain size, m is the grain size exponent, Q_{gss}
 365 and Q_{gsi} (kJ/mol) are the activation energies for the diffusion and the dislocation creep components,
 366 respectively. R is the molar gas constant and T is the absolute temperature (K). A_{gss} ($s^{-1} MPa \mu m^m$) and
 367 A_{gsi} ($s^{-1} MPa^{-n}$) are material related constants corresponding to the diffusion and the dislocation creep
 368 components, respectively. In this study we are essentially interested in the grain size sensitive (Newtonian)
 369 regime. Hence, we will further focus only on the data described by the constitutive equation:

$$370 \quad \frac{d\varepsilon}{dt} = A_0 \sigma e^{-\frac{Q}{RT}} = A \sigma d^{-m} e^{-\frac{Q}{RT}} \quad (2)$$

371
 372 where A ($s^{-1} MPa^{-n} \mu m^m$) is the material related constant, that may be written as A_0 ($s^{-1} MPa^{-n}$) if
 373 incorporating the microstructural parameter d (μm).

375 3.2.2. Grain size sensitivity

376 To estimate the strain rate dependence on grain size for nominally melt – free samples (< 1 vol.
 377 % non-wetting residual glass) we considered the present data for Lab samples with a mean grain size of
 378 $9.8 \mu m$ and data from Dimanov et al. (1998) for similar nominally melt – free labradorite, but coarser
 379 grained with a mean grain size of $16 \mu m$. We obtained a grain size exponent $m = 2.6 \pm 0.3$, which is in
 380 agreement with previous results for anorthite plagioclase (Wang et al., 1996; Dimanov et al., 1999).
 381 Previous studies on olivine and olivine – basalt systems (Hirth and Kohlstedt, 1995) documented similar
 382 grain size sensitivities ($m \approx 2.5 - 3$) indicating grain boundary diffusion controlled creep (Coble, 1963).
 383 Dimanov et al. (2003, 2007) also reported similar grain size exponents for fine grained diopside samples,
 384 and Dimanov et al., (2007) documented microstructural evidences for grain sliding mechanisms
 385 accommodated by diffusional mass transfer in fine grained anorthite and diopside two – phase aggregates.

386 According to Coble (1963) and Ashby and Verall (1973), the present data indicate that grain sliding
387 accommodated by grain boundary diffusion most likely operated in the linear – viscous regime.

388

389 **3.2.3. Activation energy**

390 In Figure 4d we reported the flow laws for the different materials in an Arrhenius type of diagram,
391 where log strain rates (for a fixed grain size and flow stress) are plotted as a function of the inverse absolute
392 temperature. The slopes of the linear regressions provide the activation energies $Q = 433 \pm 14$ kJ/mol, Q
393 $= 520 \pm 21$ kJ/mol and $Q = 673 \pm 18$ kJ/mol for diffusion controlled creep of dry Lab, SilLab1 and SilLab5
394 samples, respectively (Tab. 3). The activation energy and the strain rates for our Lab samples are
395 comparable with those for diffusion controlled creep of labradorite (An60) and anorthite plagioclase
396 aggregates, deformed uniaxially at 0.1 MPa (Dimanov et al., 1998, 1999). Figure 4d also shows that i) the
397 strain rates are nearly identical for Lab and SilLab1, ii) SilLab5 samples (which contain the highest
398 proportion of melt) show substantially lower strain rates, iii) the activation energy increases with the
399 addition of silica – rich melt.

400

401 **3.2.4 Melt effects**

402 Mechanical characterization (stress exponent, grain size exponent) and microstructures indicate
403 that all kind of samples deformed dominantly by grain sliding mechanisms accommodated by grain
404 boundary diffusion. These results agree with previous studies on high temperature creep of partially
405 molten labradorite An60 containing either non – wetting residual An60 glass (Dimanov et al., 1998), or
406 wetting partial melt obtained at sub solidus conditions (Dimanov et al., 2000). For instance, Dimanov et
407 al. (2000) have shown that samples containing $\approx 2 - 3$ vol. % of wetting partial melt (thin melt films
408 present at numerous grain boundaries) were substantially weaker than melt – free samples, or than samples
409 containing similar amounts of non – wetting residual glass (present only at multiple grain junctions).

410 Conversely, in the present study we clearly observed that the presence of thin amorphous films at grain
 411 boundaries did not weaken the samples. SilLab1 samples exhibited comparable strain rates and activation
 412 energy as for melt – free Lab samples, whilst counter intuitively SilLab5 showed to be substantially
 413 stronger and to have much higher activation energy than any other tested samples. The present results
 414 indicate that the mechanical behaviour of the partially molten samples does not only depend on the melt
 415 proportion and distribution, but also on the melt chemistry and more specifically on the silica content of
 416 the amorphous phase: the higher was the silica content and the lower was the creep rate, and hence the
 417 diffusion kinetics controlling mass transfer.

418

419 **3.2.5 Flow laws**

420 The flow law parameters that we obtained by least square regression fits of equation (2) to the
 421 mechanical data of the Newtonian regime for Lab, SilLab1 and SilLab5 materials are listed respectively
 422 below for a grain size $d = 10 \mu\text{m}$ (assuming $m = 3$):

423

424 For Lab : $\log A_0 (\text{s}^{-1}\text{MPa}^{-n}) = 9.026 \pm 0.524, n = 1, Q = 432.889 \pm 14.234 \text{ kJ/mol}$

425 For SilLab1 : $\log A_0 (\text{s}^{-1}\text{MPa}^{-n}) = 12.079 \pm 0.757, n = 1, Q = 520.484 \pm 21.000 \text{ kJ/mol}$

426 For SilLab5 : $\log A_0 (\text{s}^{-1}\text{MPa}^{-n}) = 16.732 \pm 0.642, n = 1, Q = 673.342 \pm 17.864 \text{ kJ/mol}$

427

428 **4. Discussion**

429

430 To our knowledge, the only previous work concerned with high temperature creep of silicate
 431 polycrystalline material containing excess pure silica is from Wolfenstine and Kohlstedt (1994). In the
 432 latter study synthetic Nickel – based olivine aggregates containing 3 % pure silica in excess have been
 433 deformed in uniaxial compression. The presence of the amorphous silica did not affect the creep properties

434 of the samples. Marked weakening was observed only at temperatures promoting kinetic decomposition
435 of the Ni – olivine. However, the material preparation procedure was different. Also, the authors observed
436 that the excess silica was exclusively restricted to multiple grain junctions, whilst two grain boundaries
437 remained melt-free. It is therefore impossible to directly compare the results of Wolfenstine and Kohlstedt
438 (1994) and the present findings. Conversely, our present data may be compared with the results of
439 previous works of Dimanov et al. (1998; 2000). These former studies have clearly documented that for
440 partially molten plagioclases samples containing plagioclase – like partial melts (with silica content < 80
441 wt. %), and deforming by diffusion controlled grain sliding, the creep strength depends mostly on the melt
442 topology. Dimanov et al. (2000) concluded that the samples exhibiting amorphous films along most grain
443 boundaries are substantially weaker than samples presenting an interconnected amorphous network along
444 multiple grain junctions but crystalline grain boundaries. The present study further shows that for diffusion
445 controlled grain sliding at dry conditions (< 0.002 wt.% water), when the physical structure of grain
446 boundaries is amorphous (presence of thin melt films), the chemistry of the amorphous phase (and more
447 specifically the silica content) is the key criterion for the creep strength. On the one hand, materials which
448 are nominally fully crystalline and materials containing ≈ 2 vol. % of wetting partial melt with ≈ 85 wt.
449 % silica have similar creep strengths. In opposition, samples with ≈ 6 vol. % wetting partial melt, but
450 containing ≈ 95 wt. % silica present pronounced strengthening. Though astonishing, this situation is not
451 unique. Numerous studies show that silicon nitride and silicon carbide ceramics almost always present \approx
452 1 – 2 nm amorphous silica films along most grain boundaries. Still, these materials present exceptional
453 creep strengths at high temperatures (see a review of Meléndez-Martinez and Dominguez-Rodriguez,
454 2004 and references therein). Kajihara et al. (1995) have shown that between 1200 – 1380°C tetragonal
455 zirconia polycrystals with amorphous silica present at grain boundaries is characterized by superplasticity
456 involving grain boundary sliding and diffusion controlled viscous flow of the silica phase, with an
457 activation energy of 635 kJ/mol. Ojovan and Lee (2004) reported that enthalpies of formation and

458 migration of defects in pure silica are respectively of 197 and 515 kJ/mol, which summation is in
459 astonishing agreement with both the latter activation energy and the value we report for creep in SilLab5
460 samples (673 kJ/mol), where the controlling mechanisms is supposedly grain boundary diffusion through
461 extremely silica-rich melt films. Considering that we observe diffusion controlled creep and clear effects
462 of the melt chemistry it is useful to estimate the viscosities and the diffusion properties of the melts present
463 in our samples in order to provide a physical – chemical explanation of the creep strength of our materials.

464

465 **4.1. Viscosities of the silicate melts**

466

467 The estimation of the viscosities of natural silicate melts is a crucial issue in earth sciences.
468 Therefore, their dependence on temperature and composition has been extensively studied experimentally
469 and theoretically (see Giordano et al. 2006, 2008 and references therein). From a general view point the
470 viscosity of a melt essentially depends on the degree of polymerisation of its structural units (Mysen,
471 1983). The latter is often expressed in terms of the NBO/T number, which is the number of non-bridging
472 oxygen per tetrahedron. Species in tetrahedral coordination (in particular Si^{4+} and Al^{3+} , but also Ti^{4+} , Fe^{3+}
473 ...) tend to polymerize the melt by establishing a tri-dimensional network of tetrahedrons and are thus
474 called network formers, whilst metal, alkaline and earth alkaline cations have the opposite tendency and
475 are therefore called network destroyers. The NBO/T directly relates to the relative amounts of network
476 formers and network destroyers. The lower is the NBO/T the more viscous is the considered melt. For
477 example, basalts are characterized by NBO/T ranging between 1 – 2 and are many orders of magnitude
478 less viscous than granitic and plagioclase like melts, not even mentioning fully polymerized amorphous
479 silica, which is characterized by NBO/T = 0. Based on numerous experimental data sets Giordano and
480 Dingwell (2003), and more recently Giordano et al. (2006, 2008), proposed semi empirical models for
481 predicting viscosities of anhydrous and hydrous silicate melts over a wide compositional range (from

482 haplogranitic to basaltic compositions). Data sets were first described with the well-known Vogel-
 483 Flucher-Tammann empirical relation:

484

485

$$\log \eta = A + \frac{B}{T - C} \quad (3)$$

486

487 where η is the viscosity (in Pa s), T is the absolute temperature (in K), A , B and C are the fitting
 488 parameters. The analysis is further constrained by the hypothetical assumption that the viscosities of all
 489 silicate melts converge to a common value at extremely high temperatures. This analysis implies that: i)
 490 A is composition independent, ii) B and C are correlated and accommodate all compositional effects. In
 491 particular, B and C correlate with the polymerization degree and the composition. The authors express the
 492 latter by the SM (structure modifier) parameter, which is (for anhydrous melt) the sum of the molar
 493 proportions of alkalis and earth alkaline network modifiers ($SM = \Sigma \text{ mol. \%} = \text{Na}_2\text{O} + \text{K}_2\text{O} + \text{CaO} + \text{MgO}$
 494 $+ \text{MnO} + \text{FeO}_{\text{Tot}}/2$). Based on (3) Giordano et al. (2006) computed the isothermal viscosities for 44
 495 different melt compositions (44 different SM) corresponding to different peraluminous, metaluminous
 496 and peralkaline silicate melts. The obtained isothermal viscosity values were further fitted to empirical
 497 equations of the form:

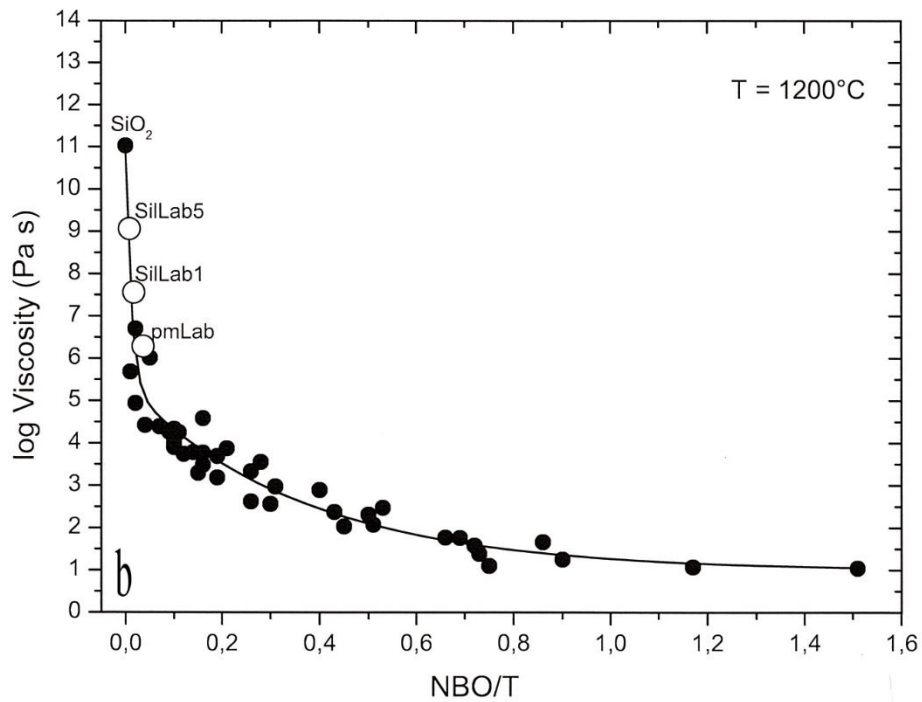
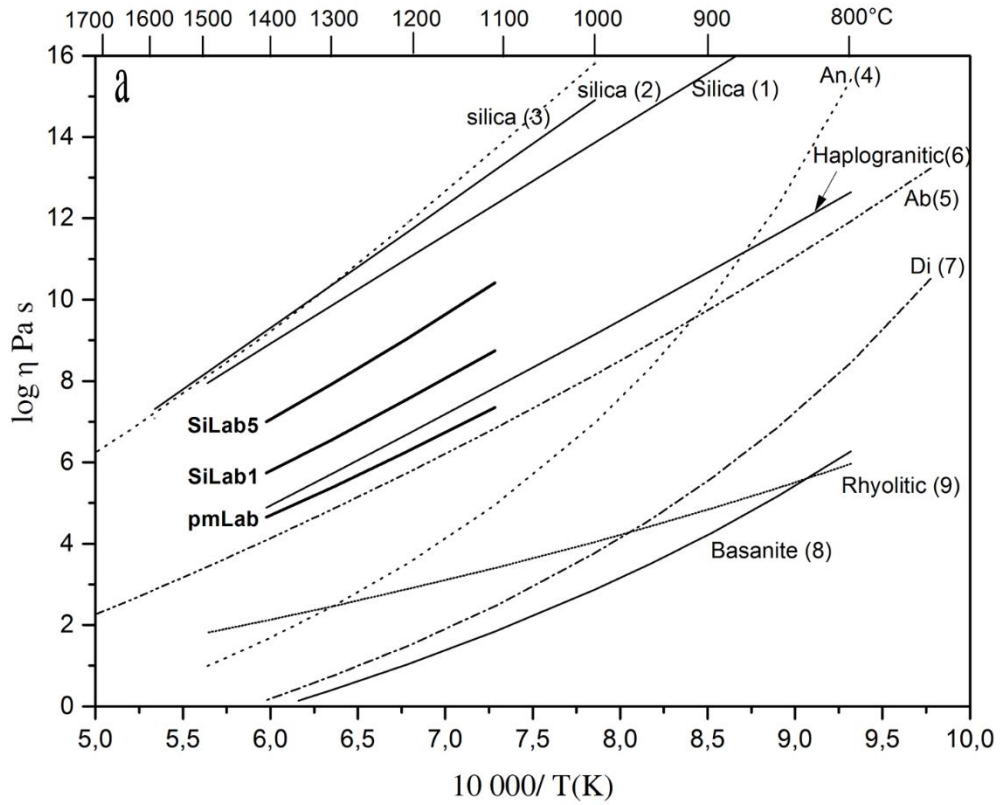
498

499

$$\log \eta = a_1 + \frac{a_2 a_3}{a_3 + SM} \quad (4)$$

500

501 where a_i , ($i = 1, 2, 3$) are empirical (temperature dependent) fitting parameters. According to the
 502 adjustments of the model to the experimental data for 44 different melts the authors derived a_1 , a_2 , a_3 as
 503 functions of temperature between 700°C and 2000°C (Tab. 4, Giordano et al., 2006).



505 Fig. 5: Comparison between calculated or measured viscosities of typical silicate melts and viscosities calculated for the melt
 506 compositions of those present in our materials. a) Arrhenius plot of viscosities of typical silicate melts (not exhaustive, but
 507 covering the whole range of viscosities from basaltic composition to vitreous silica) and those of the melts present in our
 508 samples (SilLab1 and SilLab5) and in the partially molten samples (pmLab) of Dimanov et al. (2000), computed with the
 509 model of Giordano et al. (2008). Data for silica are: (1) from Giordano et al., (2005), (2): from Nascimento and Zanotto,
 510 (2006), Silica (3): from Ojovan and Lee (2004) and from experimental data of Urbain et al., 1982 and Hetherington et al
 511 (1964). Anorthite (An(4)) and Diopside (Di(7)): from Russel and Giordano (2005), Haplogranitic(6), Albite (Ab(5)) and
 512 Rhyolitic(9): from Giordano et al. (2005), Basanite(8): from Giordano et al. (2006). b) Comparison of the experimental
 513 viscosities used by Giordano et al. (2008) to set the parameters of their model and the viscosities computed with the latter for
 514 compositions of melts as those present in our materials as a function of the NBO/T number. The model predicts very well the
 515 viscosities of the melts present in our samples.

516

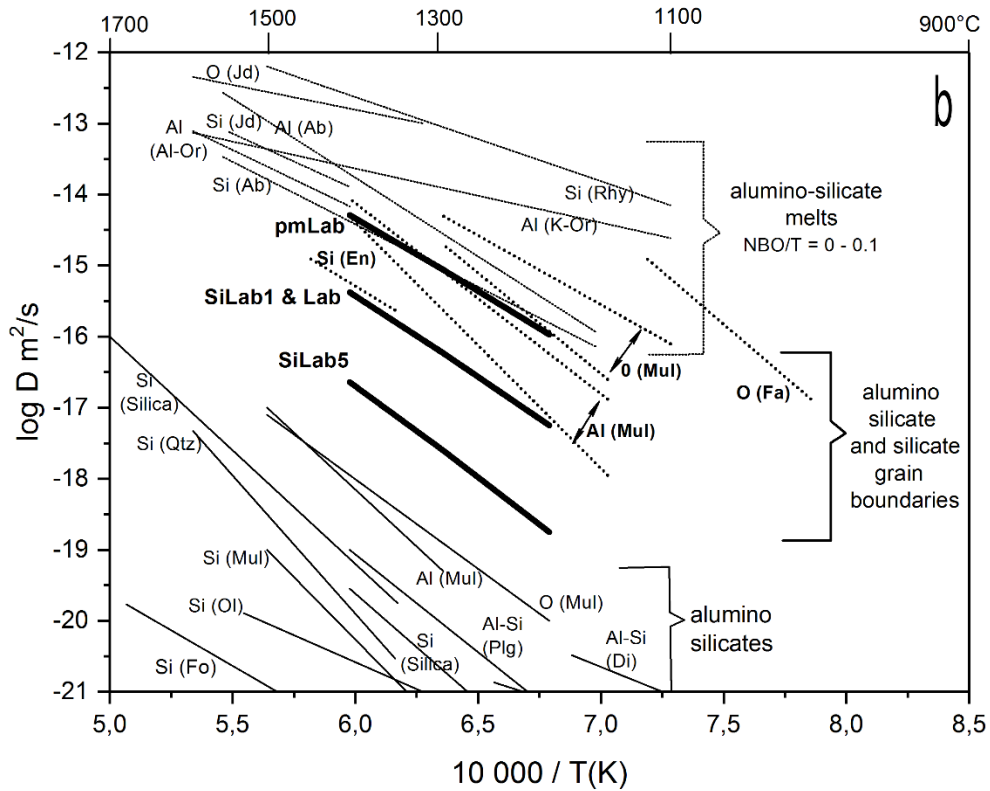
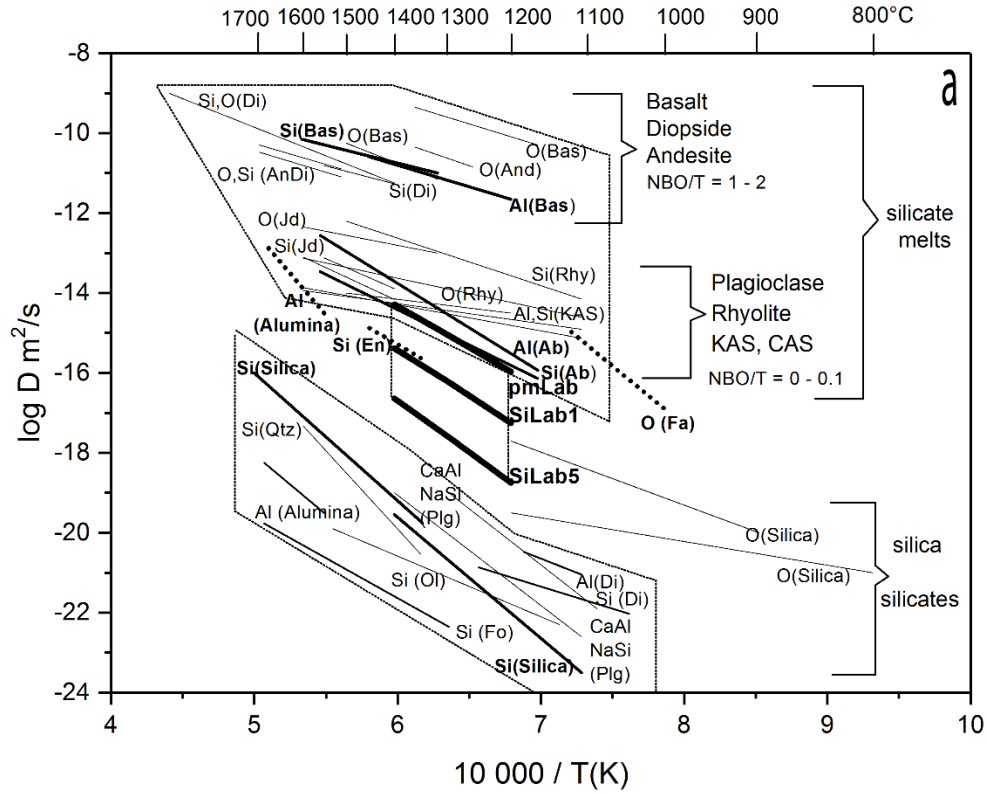
517 The phenomenological model of Giordano et al. (2006) was initially derived on the basis of
 518 experimental results from silicate melts which silica content did not exceed 80 wt. % (rhyolitic melt), but
 519 according to Giordano et al. (2008) the model may be applicable for melts containing up to 90 wt. %
 520 silica, which is the case for the melt present in our SilLab1 materials (≈ 85 wt. % SiO_2 , $\text{SM} = 5.15$). Due
 521 to the lack of any alternative model, and as a first order approximation, we also used the equation (4) and
 522 the a_i parameters given by Giordano et al. (2006) in order to calculate the viscosity for the melt present in
 523 our SilLab5 (≈ 95 wt. % SiO_2 , $\text{SM} = 2.56$) samples. The viscosity results are plotted as functions of the
 524 inverse absolute temperature in Figure 5a. One can see that the melt present in SilLab5 samples (our silica
 525 – richest melt) is about one to two orders of magnitude more viscous than the melt present in SilLab1
 526 samples. We have also reported for comparison (i) the computed viscosities for diverse anhydrous silicate
 527 melts (haplogranitic, plagioclases, rhyolitic, diopside and basanite) calculated by Russel and Giordano
 528 (2005) and Giordano et al. (2006) on the basis of equation (4), (ii) some experimentally derived viscosities
 529 for silica, albite and basalt melt (see references in figure caption). The data in Figure 5a show that silica
 530 is the most possibly viscous melt, whilst mafic and rhyolitic melts are the less viscous ones. Plagioclase

531 and haplogranitic melts present intermediate viscosities. The viscosities we obtain for the melts present
532 in our samples are in between those for pure silica and haplogranitic/albitic melts, which is fully consistent
533 with the fact that their silica contents are extremely high. We also considered for comparison the
534 composition data for the silica – rich (≈ 80 wt. % SiO_2 , $\text{SM} = 8.39$) partial melt present in the partially
535 molten labradorite (called pmLab) from Dimanov et al. (2000). The corresponding viscosity is
536 substantially lower (one to three orders of magnitude) than those of the melts investigated in the present
537 study, and compares well with the viscosities for haplogranitic or albitic melts.

538 We also calculated the NBO/T for the melts in the present study and the study of Dimanov et al.
539 (2000) following the approach of Mysen (1983, 1988; 2003). As a first order approximation we considered
540 only the major cations present (Si, Al, Na, Ca). In particular, all Al present was assigned to tetrahedral
541 coordination (network former), as its content does not exceed the sum of the charge weighted contents of
542 Na and Ca ($\text{Al} < \text{Na} + 2\text{Ca}$). We obtained NBO/T values ≈ 0.01 , 0.02 and 0.06 for SilLab5, SilLab1 and
543 pmLab respectively, which clearly indicate polymerization states in agreement with the calculated
544 viscosities. In addition, we realized a comparison of the viscosities of the melts present in our samples
545 and the material from Dimanov et al. (2000) and the viscosities of all the different melts reported by
546 Giordano et al. (2006) against their NBO/T numbers, at 1200°C (Fig. 5b). The data set is fully consistent,
547 including for the silica-richest melt from our SilLab5 material. The calculated viscosities of the melts
548 from SilLab5, SilLab1 and pmLab materials correlate very well with the viscosity suite for all the melts
549 considered by Giordano et al. (2006), which suggests that the extrapolation we made in using their model
550 for the silica-richest melt from SilLab5 sample is reasonable.

551

552 **4.2. Melt diffusivities versus grain boundary diffusivities**



554 Fig. 6: Arrhenius diagram of volume and grain boundary diffusion of network formers (Si, Al and O) in silicates and oxides
 555 and in silicate melts (in dry or unsaturated conditions). **a:** The dot line delimited upper and lower fields represent the data for
 556 Si, Al and O diffusion in silicate melts and silicate single crystals, respectively. The data are recalculated for a pressure of 0.1
 557 MPa (when activation volumes were available). Bas is basalt, Rhy is rhyolitic melt, Di and Jd refer to diopside and jadeite
 558 melts, Ab and KAS are albite and potassium-aluminosilicate melts. Diffusivity of Si in silica falls within the lower field for
 559 volume diffusion. Diffusivities of O in silica fall in-between the two fields. Grain boundary diffusivities (recalculated for a
 560 grain boundary width of ~ 3 nm) are represented by heavy dot lines. The larger vertical brackets indicate the type of diffusivity.
 561 For diffusion in melt the smaller brackets indicate the compositional ranges and the corresponding NBO/T numbers. The
 562 diffusivities calculated for our melt compositions (see text and Fig. 5) are reported as heavy lines
 563 **b:** Close up from (a) comparing the existing grain boundary diffusivities (light dotted lines) in aluminosilicates and silicates
 564 (Mullite: Mul, Enstatite: En, Fayalite: Fa) and calculated diffusivities for the present highly polymerized aluminosilicate melts
 565 (pmLab, SilLab1 and SilLab5). It appears that grain boundary diffusivities are in general comparable to the lowest diffusivities
 566 in aluminosilicate melts, as do the diffusivities for pmLab and SilLab1 melts. The diffusivity for SilLab5 melt shows however
 567 substantially lower, in-between the lowest diffusivities in alumina-silicate melts and the highest diffusivities in alumina-
 568 silicate single crystals.

569 Diffusion data for melts are from Baker (1990, 1992, 1995), Shimizu and Kushiro (1984, 1991), Canil and Muehlenbach
 570 (1990), Wendlandt (1991), Chakraborty et al. (1995), Kress and Ghiorso (1995), Lesher et al. (1996), Harmer and Brook
 571 (1980), Brebec et al. (1980), Reid et al. (2001).

572 Data for grain boundary diffusion in silicates, aluminosilicates and alumina are from Rubie (1986), Fislser and Mackwell
 573 (1994), Fislser et al. (1997), Fielitz et al. (2004, 2007, 2016). Data for volume diffusion in silicates, aluminosilicates and
 574 alumina are from Bejina and Jaoul (1996), Houlier et al. (1990), Jaoul et al. (1981, 1991), Grove et al. (1984), Yund (1986),
 575 Le Gall et al. (1994), Fielitz et al. (2007).

577 Ion diffusion properties in aluminosilicate melts inversely correlate with their viscosities, that is
 578 to say with their NBO/T (Hofmann, 1980; Shimizu and Kushiro, 1991; Mysen, 1988). The latter explicitly
 579 depends on the relative proportion of network formers and network destroyers, and hence the silica content
 580 is straightforwardly a first order parameter in determining NBO/T, viscosity and diffusivity. The lower is
 581 the NBO/T and the higher is the viscosity (Fig. 5). For example, the NBO/T is between 1 and 2 for

582 andesitic to mafic compositions (basalts), whilst it is < 0.1 for felsic compositions (rhyolites) and strictly
583 zero for dry silica. Consequently, the lower is the NBO/T and the lower is the diffusivity. An illustration
584 is given by Liang et al. (1996), who measured self-diffusivities in calcium aluminosilicate (CAS) melts
585 with low water contents between 0.04 – 0.07 wt.%. They show that D_{Si} , D_{Al} , D_O and D_{Ca} are respectively
586 about 20, 10, 13 and 4 times lower in the melt with 20, 15 and 65 wt.% CaO, Al_2O_3 and SiO_2 (NBO/T =
587 0.305) than with 40, 20, 40 wt. % CaO, Al_2O_3 and SiO_2 (NBO/T = 0.978).

588 In Figure 6 we present a compilation for diffusivities in silicates, considering: i) volume diffusion
589 in single crystals, ii) grain boundary diffusion in polycrystals, and iii) diffusion in melts. The considered
590 data were intentionally restricted to the slowest diffusing network formers (Al, Si and O), which are the
591 species susceptible to control the kinetics of diffusion controlled creep. To be in accord with the present
592 work, we further restricted our consideration to data obtained for dry systems. In the experimental
593 temperature range of interest the diffusion rates of Si and Al in silicate melts decrease over many orders
594 of magnitude with decreasing NBO/T, that is to say with increasing silica content and correspondingly
595 decreasing contents of network destroyers. In general, the diffusion rates are the fastest in basalts,
596 intermediate in rhyolitic melts and substantially lower in plagioclase melts. The extreme lowest diffusion
597 rate of Si in pure amorphous silica is nearly comparable with Si diffusion in single crystal pure quartz.
598 These observations are qualitatively consistent with our findings from creep data, which indicate: i) the
599 highest diffusivity for the partially molten samples of Dimanov et al. (2000), which contain amorphous
600 grain boundaries with ≈ 80 wt.% silica, ii) intermediate diffusivity for our SilLab1 samples, which present
601 amorphous grain boundaries with ≈ 85 wt.% silica, iii) the lowest diffusivity for our SilLab1 samples,
602 which present amorphous grain boundaries with ≈ 95 wt.% silica.

603 Yet, it is most important to address the question whether any silicate melt present along labradorite
604 grain boundaries may enhance the diffusion rates with respect to the crystalline grain boundaries. On the
605 one hand, the data of Dimanov et al. (2000) indicate that thin melt films with ≈ 80 wt. % silica provide

606 substantially faster pathways for grain boundary diffusion as compared with labradorite grain boundaries.
 607 On the other hand, the present data for SilLab1 samples indicate that thin melt films with ≈ 85 wt.% silica
 608 have no noticeable effect, whereas our data for SilLab5 samples indicate that grain boundary diffusion is
 609 substantially slowed down within thin melt films with ≈ 95 wt.% silica. In order to be more quantitative
 610 we need to estimate the effective diffusivities in our different samples and to compare the latter with
 611 diffusivities in silicate melts and grain boundaries. For this purpose we considered the Eyring equation,
 612 relating diffusivity and viscosity of liquids:

$$D_{eff} = \frac{k_B T}{\lambda \eta} \quad (5)$$

613
 614
 615
 616 where D_{eff} and η are respectively the effective diffusivity and the viscosity, and where the molecular radius
 617 λ is taken as an approximation for the jump distance of the mobile species. For instance, the Eyring
 618 equation is especially considered as applicable to the network formers in viscous silicate melts and glasses
 619 (Mungal, 2002; Nascimento and Zanotto, 2006). Based on equation (5) and taking $\lambda = 10^{-10}$ m as a first
 620 order approximation, we calculated the diffusivities of the melts present in SilLab1, SilLab5 and in the
 621 partially molten labradorite (pmLab) of Dimanov et al. (2000). We show in Figure 6a that the latter are
 622 intermediate between volume diffusion and grain boundary diffusion in silicates. Unfortunately, direct
 623 measurements of grain boundary diffusivities for plagioclase materials are not available. Indirect
 624 estimations of grain boundary diffusivities in plagioclase, pyroxene and forsterite from the kinetics of
 625 reaction rim growth do exist (Liu et al., 1997; Yund, 1997; Yund and Farver, 1999; Milke et al., 2001;
 626 Abart et al., 2004), but only for water present conditions, which precludes any direct comparison in the
 627 present study. However, Figure 6b shows that the existing grain boundary diffusion data for dry silicates,
 628 oxides and aluminosilicates (dotted lines) correspond to the slowest diffusivities in the most polymerized
 629 aluminosilicate melts (rhyolite, albite). This observation is in agreement with recent findings based on

630 molecular dynamics, which indicate strong similarities between the cooperative molecular motion at grain
631 boundaries and in glass forming liquids (Zhang et al., 2009). According to the data, the calculated D_{eff} in
632 labradorite partial melt (pmLab) corresponds very well to the measured Si and Al diffusivities in albite
633 melt. On the other hand, we can highlight the fact that within the experimental range (1100 – 1300°C) D_{eff}
634 in labradorite partial melt (pmLab) exceeds by about an order of magnitude the existing grain boundary
635 diffusivities in enstatite, alumina and mulite. This observation is consistent with the findings of Dimanov
636 et al. (2000), who reported that the partially molten labradorite samples (pmLab) were about an order of
637 magnitude weaker than the fully crystalline samples. Conversely, the effective diffusion through melt
638 films with 85 wt.% silica (SilLab1) seems slow enough to mimic grain boundary diffusion rates. This
639 observation provides an explanation why the SilLab1 and the fully crystalline Lab samples have
640 comparable strengths. Most interestingly, diffusion through melts with 95 wt. % silica (SilLab5) appears
641 to be much slower than in any other melt, or at grain boundaries. The corresponding effective diffusivity
642 is only one to two orders of magnitude faster than volume diffusion processes in aluminosilicate single
643 crystals involving network formers (diffusion of Al and O in mullite and CaAl – NaSi interdiffusion in
644 plagioclase). This observation clearly suggests very slow grain boundary diffusion rates for SilLab5
645 samples, which is fully consistent with the fact that the latter are considerably stronger than the SilLab1
646 and Lab samples.

647

648 **4.3. Grain boundary diffusivity versus melt – grain interfacial diffusivity**

649

650 We estimated on the basis of equations (4) and (5) the effective diffusivities for the different
651 amorphous silicate materials present along grain boundaries in our partially molten plagioclase samples.
652 The results are in good agreement with the extant diffusion data in dry conditions for Si, Al and O (network
653 formers and rate limiting species) in both aluminosilicate melts and along grain boundaries (Fig. 6). From

654 this comparison it appears that i) diffusivity in plagioclase – like partial melt (80 wt. % silica) is
655 substantially faster than diffusivities along grain boundaries; ii) diffusivities along grain boundaries
656 correspond to the diffusivity in silica – rich melt (85 wt. % SiO₂); iii) diffusivity in extremely silica – rich
657 melt (95 wt. % silica) is substantially slower than along grain boundaries. The first point indicates that
658 partially molten plagioclase containing plagioclase – like melt forming films along grain boundaries
659 should be weaker than nominally crystalline plagioclase, which is precisely what Dimanov et al. (2000)
660 observed. The second and the third points imply that partially molten plagioclase with silica – rich (85 wt.
661 % SiO₂) and extremely silica – rich (95 wt. % SiO₂) melt films along grain boundaries should be,
662 respectively, as strong- and stronger than nominally crystalline plagioclase, which is what we did observe.
663 Our evaluation of diffusivities from melt viscosities, which were previously evaluated from melt
664 chemistry, is only a first order approximation. Nevertheless, in spite of the roughness of this approach,
665 the order of magnitude and the consistency of the estimated results are convincingly reasonable when
666 compared with i) the experimental data for melts (Figures 5 and 6); ii) our creep data interpreted in terms
667 of efficiency of interfacial diffusion. However, these observations open some additional questions about
668 the structure and the diffusivities at the interfaces between the grains and the intergranular amorphous
669 films, which are developed below.

670 Solid state diffusional mass transfer operates owing to crystallographic structural defects. Volume
671 diffusion relies on point defects, such as vacancies, interstitials, substitution impurities and their
672 associations. Alternatives for faster volume diffusion are provided by extended linear and planar defects,
673 such as dislocation cores and twin boundaries or sub – grain boundaries. The latter are special low – angle
674 tilt – boundaries, which result from the rearrangement of free dislocations (dislocation pile-up) driven by
675 the associated decrease in free energy of the system. Twin boundaries and low – angle tilt – boundaries
676 may be called are coherent semi – coherent, respectively, because they present the most numerous
677 coincident crystallographic sites. Conversely, the common high angle, tilt and twist grain boundaries

678 present the lowest densities of coincident lattice sites. The lower is the density of the coincident lattice
679 sites and inversely higher is the density of the structural defects (local lattice distortion, dangling bonds,
680 impurity segregation...) of the considered interface. Interfacial diffusion is conditioned by the density of
681 interfacial structural defects, and hence, diffusion along common grain boundaries is orders of magnitude
682 faster than diffusion along coherent interfaces and volume diffusion. The classical hypothesis that
683 diffusion creep is controlled by the slowest species diffusing through the fastest path ways implies in the
684 case of nominally crystalline plagioclase creep control by the diffusion of network formers along grain
685 boundaries. Conversely, the fastest path ways would be the interfacial melt films, if providing fastest
686 diffusion kinetics than the crystalline interfaces. It may be, however, more difficult to identify the fastest
687 path ways if the interfaces contain amorphous material where diffusion is supposedly slower than at
688 crystalline high angle grain boundaries. If diffusion occurs through the melt films, the material would
689 strengthen with respect to the nominally crystalline one, which is what we did observe. Still, a question
690 remains whether faster diffusion could not proceed along the grain – melt interfaces, in which case the
691 macroscopic behaviour would differ. In order to clarify this point it is necessary to compare the nature
692 and the density of structural defects at a melt – grain interface with respect to a common grain boundary,
693 which we attempt in the following.

694 Low – angle tilt – boundaries are low – energy (favoured) grain boundaries that present the highest
695 densities of coincident lattice sites. From a geometrical viewpoint their cores have been recognized to
696 form regular arrays of “structural units”, i.e. small atomic groups organized in a characteristic
697 configuration (Sutton and Vitek, 1983). Conceptually, the “structural unit model” is equivalent to describe
698 the interface as closely spaced lattice dislocations (sometimes called primary dislocations) whose elastic
699 stress field concentrates closely to the core interface (Baluffi and Bristowe, 1984). As a consequence, such
700 grain boundaries present little lattice distortion at long-range. More random interfaces (tilt and twist
701 boundaries) were tentatively described by mixing different types of structural units corresponding to

702 different types of favoured boundaries (Sutton and Vitek, 1983), which was shown i) to be equivalent to
703 the classical grain boundary dislocation (or secondary dislocation) structure (Baluffi and Bristowe, 1984),
704 ii) to be of limited success (Sutton, 1989). Furthermore, considering such general interfaces Sutton and
705 Baluffi (1987) concluded that a simple geometric framework cannot provide general rules for optimized
706 (low energy) configurations, because the atomistic and bonding structures must be considered as well.
707 Therefore, random interfaces necessarily exhibit the highest densities of structural misfits and
708 correspondingly the high interfacial energies. The latter are actually partly compensated by local
709 relaxation (rearrangement) of atomic positions at and near the core interface (Merkle, 1997). Relaxation
710 results in various crystallographic distortions of the core boundary. The net result is a strong short range
711 interfacial disordering, which makes the dynamic properties (mobility, co-operational transport and
712 viscosity) of grain boundaries to mimic those of amorphous phases (Zhang et al., 2009). Therefore, the
713 common (high – angle, twist and tilt) grain boundaries are characterized by the highest density of
714 ionic/molecular crystal structural defects (impurities, vacancies, interstitials and their associations), but
715 also extended crystal structural defects (dislocations, sub-grain boundaries, vacancy clusters...), which all
716 contribute to the structural misfits, the so called “packing frustration” and the presence of unsatisfied
717 dangling bonds. As the interfacial free energy relates to the density of the interfacial structural defects
718 (van der Merwe, 2001), it increases gradually from twin – and low angle – to high angle boundaries, which
719 exhibit the highest interfacial energies.

720 For ceramic materials (alumina, zirconia, SiC, Si₃N₄, SrTiO₂...) it has been demonstrated that thin
721 (nm – sized) amorphous films may be thermodynamically stable at grain boundaries, because they may
722 contribute to lower the interfacial free energy of the system (Clarke, 1987; Clarke et al., 1993; Wilkinson,
723 1993; Bobeth et al., 1999). In other words, an amorphous interphase may contribute to partly compensate
724 for the inherent interfacial structural defects. For instance, in order to minimise the excess free energy
725 associated with interfacial misfits the surrounding crystalline lattices can only modestly relax by limited

726 distortion. Conversely, a viscous amorphous phase could more efficiently accommodate to any interfacial
727 structural misfit, providing molecular units to satisfy dangling bonds. The net result would be the
728 reduction of interfacial lattice distortions and interfacial free energy, and thus, of the density of structural
729 defects. If so, the diffusivities at the crystal – melt interface should be lowered with respect to the common
730 grain boundary diffusivities as well. So finally, whether the presence of an interfacial amorphous phase
731 would slow down or enhance the grain boundary diffusion rates would essentially depend on the diffusion
732 properties through the amorphous material itself. In the present case, the extremely silica – rich amorphous
733 phase present along grain boundaries of SilLab5 material tends to slows down the diffusion properties
734 and the samples appear stronger.

735

736 **5. Conclusions**

737

738 We investigated high temperature ($>1000^{\circ}\text{C}$) diffusion controlled creep of plagioclase aggregates
739 containing limited amounts of partial melt (2 – 6 vol. %), with respect to the physical structure (crystalline
740 / amorphous) of the grain boundaries. We specifically focused on the effect of melt chemistry in the
741 situation of thin amorphous films ($< 10\text{ nm}$) wetting grain boundaries. For this purpose we applied sol –
742 gel techniques and isostatic hot – pressing (300 MPa, $T = 1150^{\circ}\text{C}$) in order to synthesize i) nominally
743 crystalline plagioclase material (SilLab), ii) two types of partially molten plagioclase materials (SilLab1
744 and SilLab5), containing limited amounts of silica – rich partial melt, with contrasting chemistries. The
745 SilLab nominally melt – free samples contained $< 1\text{ vol. \%}$ residual glass. The SilLab1 and SilLab5
746 partially molten plagioclase samples contained 2 and 6 vol. % of aluminosilicate melts with c.a. 85 wt. %
747 and 95 wt. % SiO_2 , respectively. The hot-pressed materials were fine grained ($< 15\text{ }\mu\text{m}$) and contained
748 about 0.05 wt. % water species that could be dried out by annealing at 1100°C and at atmospheric pressure.
749 Uniaxial compression creep tests were performed at temperatures and flow stresses between 1100 –

750 1250°C and 3 – 60 MPa, respectively. All samples showed dominantly linear viscous flow with a stress
751 exponent $n \sim 1$ and nominally melt – free samples showed a grain size exponent $m \sim -3$, indicating grain
752 boundary diffusion control. Activation energies were 433 ± 14 kJ/mol, 520 ± 21 kJ/mol and 672 ± 18
753 kJ/mol for nominally melt – free samples, and for samples containing ~ 2 vol. % and 6 vol. % silica – rich
754 melts, respectively. The samples containing ~ 6 vol. % melt presented the highest activation energy and
755 were substantially stronger than all other samples. We explained the fact by the presence of the highly
756 polymerized and viscous melt, with the highest silica content (~ 95 wt. % SiO_2), and thus, with the lowest
757 diffusion rates.

758 Based on viscosity models for aluminosilicate melts (Giordano et al., 2006) we estimated the
759 viscosities and the effective diffusivities of the melts present in our samples and in previously studied
760 partially molten plagioclase samples (≈ 80 wt. % SiO_2 , Dimanov et al., 2000). The data we obtain are in
761 agreement while considering the extant diffusion data of network formers in silicate melts, with respect
762 to their silica contents and polymerization states in dry conditions. Furthermore, we considered volume
763 diffusion data in single crystals and grain boundary diffusivities in silicates and oxides polycrystals.
764 Amorphous silica apart, volume diffusion data in single crystals are the lowest, whilst melt diffusivities
765 are the highest. For the latter, the diffusivities inversely correlate with the polymerization degree. Grain
766 boundary diffusion data are much higher than volume diffusion data, but they overlap with the diffusivities
767 in highly polymerized aluminosilicate melts. Our findings indicate that grain boundary diffusivity in
768 polycrystalline melt-free labradorite is i) comparable to those in highly polymerized silica-rich melts (\sim
769 85 wt. % SiO_2), ii) considerably lower than those in labradorite partial melt (~ 80 wt. % SiO_2), iii)
770 substantially higher than in extremely silica-rich melt (~ 95 wt. % SiO_2).

771 We demonstrated strong physical – chemical couplings between plagioclase rheology based on
772 grain boundary diffusion controlled creep and the presence of aluminosilicate melts. In the particular case
773 where grain boundaries present amorphous structure the rheological properties are strongly related to the

774 melt chemistry, and more specifically to the silica content. The latter determines the polymerization
775 degree and hence the diffusion properties, depending on which samples may either weaken or strengthen.
776 Counter intuitively, our data show that the presence of thin amorphous films at grain boundaries may not
777 always ensure faster diffusion path ways. In turn, in dry conditions grain boundary transport properties
778 may be strongly hindered in the presence of melt films with extreme silica contents ($> 90 - 95$ wt. % SiO_2).

779

780 **Acknowledgements**

781

782 The author is grateful to Jianguo Huang and Michael Naumann who assisted with sample
783 preparation techniques and mechanical testing, to Richard Wirth for his help with TEM observations, to
784 Erik Rybacki for many stimulating discussions and to Georg Dresen, who kindly provided unlimited
785 access to his experimental facilities and unconditional support. The rheological and microstructural
786 investigation data are available.

787

788 **References**

789

790 Abart, R., Kunze, K., Milke, R., Sperb, R. and Heinrich, W., (2004), Silicon and oxygen self-diffusion in
791 enstatite polycrystals: the Milke et al. (2001) rim growth experiments revisited. *Contrib. Mineral. Petrol.*,
792 Vol. 147, 633-646.

793 Ashby, M.F. and Verall, R.A. (1973), Diffusion accomodated flow and superplasticity. *Acta Metallurgica*.
794 Vol. 21, 149-163.

795 Balluffi, R.W. and P.D. Bristowe (1984), On the structural unit/grain boundary dislocation model for grain
796 boundary structure. *Surface Science*, Vol. 144 (1), 28-43.

- 797 Baker, D.R., (1990), Chemical interdiffusion of dacite and rhyolite: anhydrous measurement at 1 atm and
798 at 10 kbar, application of transient state theory and diffusion in zoned magma chambers. *Contrib. Mineral.*
799 *Petrol.*, Vol.104: 407-423.
- 800 Baker, D.R., (1992), Tracer diffusion of network formers and multicomponent diffusion in dacitic and
801 rhyolitic melts. *Geochim. Cosmochim. Acta.*, Vol. 56, 462-473.
- 802 Baker, D.R., (1995), Diffusion of silicon and gallium (as an analogue for aluminum) network-forming
803 cations and their relationship to viscosity in albite melt. *Geochim. Cosmochim. Acta.*, Vol. 59 (17), 3561-
804 3571.
- 805 Beeman M.L. and Kohlstedt, D.L. (1993), Deformation of Fine-Grained Aggregates of Olivine Plus Melt
806 at High Temperatures and Pressures. *J. Geophys. Res.*, Vol. 98, B4, 6443-6452, doi:10.1029/92JB02697
- 807 Beere, W. (1975), The second stage of sintering of powder compacts. *Acta Metal.* 23(1), 139-145.
- 808 Béjina, F. and Jaoul, O., (1996), Silicon self-diffusion in quartz and diopside measured by nuclear micro-
809 analysis methods. *Phys. Earth. Planet. Interiors*, Vol. 97, 145-162.
- 810 Beran, A., (1986), A model of Water Allocation in Alkali Feldspar, Derived from Infrared-Spectroscopic
811 Investigations. *Phys. Chem. Minerals*, Vol. 13, 306-310.
- 812 Beran, A., (1987), O.H. Groups in Nominally Anhydrous Framework Structures: An Infrared
813 Spectroscopic Investigation of Danburite and Labradorite. *Phys. Chem. Minerals*, Vol.14, 441-445.
- 814 Brebec, G., Seguin, R., Sella, C., Benevot, J. and Martin, J.C., (1980), Diffusion du silicium dans la silice
815 amorphe. *Acta Metall.*, Vol. 28, 327-333.
- 816 Bobeth, M., Clarke, D.R. and Pompe, W. (1999), A diffuse interface description of intergranular films in
817 polycrystalline ceramics. *J. Am. Ceram. Soc.*, Vol. 82 (6), 1537-1546.
- 818 Bulau, J.R., Waff, H.S. and Tyburczy, J., (1979), Mechanical and thermodynamic constraints on fluid
819 distribution in partial melts. *J. Geophys. Res.*, vol. 84, B11, 6102-6108.

- 820 Burger, P., Duclos, R. and Crampon, J., (1995), Microstructure characterization in superplastically
821 deformed silicon nitride. *J. Am. Ceram. Soc.*, Vol. 80, 879-885.
- 822 Bussod, G.Y and Christie J.M. (1991), Textural development and melt topology in spinel lherzolite
823 experimentally deformed at hypersolidus conditions. *J. Petrol. Special lherzolite issue*, 17–39.
- 824 Bystricky, M., and S. Mackwell (2001), Creep of dry clinopyroxene aggregates, *J. Geophys. Res.*, Vol.
825 106, 13443-13454.
- 826 Canil, D. and Muehlenbachs, K., (1990), Oxygen diffusion in an Fe-rich basalt melt. *Geochim.*
827 *Cosmochim. Acta.*, Vol. 54: 2947-2951.
- 828 Carapic, G., Faul, U.H. and Brisson, E. (2013), High-resolution imaging of the melt distribution in
829 partially molten upper mantle rocks: evidence for wetted two-grain boundaries. *Geochem. Geophys.*
830 *Geosyst.*, Vol. 14(3), 556-566 doi:10.1029/2012GC004547.
- 831 Chakraborty, S., Dingwell, D.B. and Rubie, D.C., (1995), Multicomponent diffusion in ternary silicate
832 melts in the system $K_2O-Al_2O_3-SiO_2$: II. Mechanisms, systematics, and geological implications.
833 *Geochim. Cosmochim. Acta.*, Vol. 59(2), 265-277.
- 834 Clarisse L., Petit F., Crampon J. and Duclos R., (2000), Characterization of grain boundary sliding in a
835 fine-grained alumina-zirconia ceramic composite by atomic force microscopy. *Ceram. Int.*, Vol. 26(3),
836 295-302.
- 837 Clarke, D.R., (1987), On the equilibrium thickness of intergranular glass phases in ceramic materials. *J.*
838 *Am. Ceram. Soc.*, Vol. 70, 15-22.
- 839 Clarke D.R., Shaw, T.M., Philipse, A.P. and Horn, R.G. (1993), Possible electrical double layer
840 contribution to the equilibrium thickness of intergranular glass films in polycrystalline ceramics. *J. Am.*
841 *Ceram. Soc.*, Vol. 76(5), 1201-1204.
- 842 Cmíral, M., Fitz Gerald, J.D., and Faul, U.H., (1998), A close look at dihedral angles and melt geometry
843 in olivine-basalt aggregates: a ZEM study, *Contrib Mineral Petrol*, Vol.130, 336-345.

- 844 Coble, R.L., (1963), A model for boundary diffusion controlled creep in polycrystalline materials. *J. Appl.*
845 *Phys.*, Vol. 34, 1679-1682.
- 846 Cooper, R.F. and Kohlstedt, D.L., (1984a), Sintering of olivine and olivine – basalt aggregates. *Phys.*
847 *Chem. Mineral.*, Vol. 11, 5 – 6.
- 848 Cooper, R.F. and Kohlstedt, D.L., (1984b), Solution - precipitation enhanced diffusional creep of partially
849 molten olivine-basalt aggregates during hot-pressing. *Tectonophysics*, Vol. 107, 207-233.
- 850 Cooper, R.F. and Kohlstedt, D.L., 1986. Rheology and Structure of Olivine - Basalt Partial Melts. *J.*
851 *Geophys. Res.* Vol. 91, B9, 9315-9323.
- 852 Cooper, R.F., Kohlstedt, D.L. and Chyung. K., (1989), Solution-precipitation enhanced creep in solid-
853 liquid aggregates which display a non-zero dihedral angle. *Acta Metall.*, Vol. 37, 1759-1771.
- 854 Daines, M.J. and Kohlstedt, D.L., (1997), Influence of deformation on melt topology in peridotites. *J.*
855 *Geophys. Res.*, Vol. 102, B5, 10257-10271.
- 856 Dell'Angelo, L.N. and Tullis, J., (1988), Experimental deformation of partially melted granitic aggregates.
857 *J. Metamorph. Geol.*, Vol. 6, 495-515.
- 858 Dell'Angelo, L.N., Tullis, J. and Yund, R.A., (1987), Transition from dislocation creep to melt-enhanced
859 diffusion creep in fine grained granitic aggregates. *Tectonophysics*, Vol. 139, 325-332.
- 860 Dimanov, A., Dresen, G. and Wirth, R., (1998), High-temperature creep of partially molten plagioclase
861 aggregates. *J. Geophys. Res.*, Vol. 103, B5, 9651-9664.
- 862 Dimanov, A., Xiao, X., Dresen G. and Wirth. R., (1999), Grain Boundary Diffusion Creep of Synthetic
863 Anorthite: The Effect of Water. *J. Geophys. Res.*, Vol. 104, 10483-10497.
- 864 Dimanov, A., Wirth, R. and Dresen, G., (2000), The effect of melt distribution on the rheology of feldspar
865 rocks. *Tectonophysics*, Vol. 328, 307-327.
- 866 Dimanov, A., Lavie, M.P., Ingrin, J., Dresen, G. and Jaoul, O. (2003), Creep of polycrystalline anorthite
867 and diopside. *J. Geophys. Res.* Vol. 108, B1, 2061, doi:10.10292002JB001815.

- 868 Dimanov, A., Rybacki, E., Wirth, R. and Dresen G. (2007), Creep and strain-dependent microstructures
869 of synthetic anorthite-diopside aggregates: *J. Struct. Geol.*, Vol. 29, 1049–1069, doi:
870 10.1016/j.jsg.2007.02.010.
- 871 Dimanov, A., Raphanel, J. and Dresen G. (2011), Newtonian flow of heterogeneous synthetic gabbros at
872 high strain: Grain sliding, ductile failure, and contrasting local mechanisms and interactions. *Eur. J. Min.*,
873 Vol. 23(3), 303-322, doi: 10.1127/0935–1221/2011/0023–2110
- 874 Dimanov A. and Dresen, G. (2005), Rheology of synthetic anorthite-diopside aggregates: Implications
875 for ductile shear zones. *J. Geophys. Res.*, Vol. 110, doi:10.1029/2004JB003431.
- 876 De Kloe, R., Drury, M.R., and van Roermund, H.L.M., (2000), Evidence for stable grain boundary melt
877 films in experimentally deformed olivine –orthopyroxene rocks, *Phys Chem Minerals*, Vol. 27, 480-494.
- 878 Drury, M.R. and FitzGerald, J.D., (1996), Grain boundary melt films in an experimentally deformed
879 olivine-orthopyroxene rock: implications for melt distribution in upper mantle rocks. *Geophys. Res. Lett.*,
880 Vol. 23(7), 701-704.
- 881 Duclos, R., Crampon, J. and Carry, C., (2002), Grain-boundary sliding and accommodation mechanism
882 during creep of yttria-partially-stabilized zirconia. *Philos. Mag. Lett.*, Vol. 82(10), 529-533.
- 883 Faul, U.H., (1997) Permeability of partially molten upper mantle rocks from experiment and percolation
884 theory. *J. Geophys. Res.*, Vol. 102, B5, 10299-10311.
- 885 Fielitz, P., G. Borchardt, M. Schmüker, H. Schneider and P. Willich (2004), Oxygen grain-boundary
886 diffusion in polycrystalline Mullite Ceramics. *J. Am. Ceram. Soc.*, Vol. 87(12), 2232-2236. doi:
887 10.1111/j.1151-2916.2004.tb07497.x
- 888 Fielitz, P., G. Borchardt, M. Schmüker and H. Schneider (2007), A diffusion-controlled mullite formation
889 reaction model based on tracer diffusivity data for aluminium, silicon and oxygen. *Phil. Mag.*, Vol. 87(1),
890 111-127.

- 891 Fielitz, P. and G. Borchardt (2016), Self-Diffusion of the Constituent Elements in Alpha-Alumina, Mullite
892 and Aluminosilicate Glasses", *Diffusion Foundations*, Vol. 8, 80-108.
- 893 Nascimento, M.L.F. and E.D. Zannoto, (2006), Mechanisms and dynamics of crystal growth, viscous
894 flow, and self-diffusion in silica glass. *Phys. Rev. B*, Vol. 73, 024209.
- 895 Fislser, D.K. and Mackwell, S.J., (1994), Kinetics of diffusion-controlled growth of fayalite. *Phys. Chem.*
896 *Minerals*, Vol. 21, 156-165.
- 897 Fislser, D.K., Mackwell, S.J. and Petsch, S., (1997), Grain boundary diffusion in enstatite. *Phys. Chem.*
898 *Minerals*, Vol. 24, 264-273.
- 899 Franz, L. and Wirth, R., (1997), Thin intergranular melt films and melt pockets in spinel peridotite
900 xenoliths from the Rhön area (Germany): early stage of melt generation by grain boundary melting.
901 *Contrib. Mineral. Petrol.*, Vol. 129 (4), 268-283.
- 902 Fujii, N., K. Osamura and E. Takahashi (1986), Effect of water saturation on the distribution of partial
903 melt in the olivine-pyroxene-plagioclase system. *J. Geophys. Res.*, Vol. 91, 9253-9259.
- 904 Giordano, D. and Dingwell, D.B. (2003), Non-Arrhenian multicomponent melt viscosity: A model. *Earth*
905 *Planet. Sci. Lett.*, Vol. 208, 337-349.
- 906 Giordano, D., Mangicapra, A., Potuzak, M., Russell, J.K., Romano, C., Dingwell, D.B. and Di Muro, A.
907 (2006), An expanded non-Arrhenian model for silicate melt viscosity: A treatment for metaluminous,
908 peraluminous and peralkaline melts. *Chem. Geol.*, Vol. 229, 42-56.
- 909 Giordano, D., Russell, J.K. and Dingwell, D.B. (2008), Viscosity of magmatic liquids: a model. *Earth and*
910 *Planet. Sci. Letts.*, Vol. 271, 123-134.
- 911 Grove, T.L., Baker, M.B. and Kinzler, R.J., (1984), Coupled CaAl - NaSi diffusion in plagioclase feldspar:
912 experiments and applications to cooling rate speedometry. *Geochim. Cosmochim. Acta.*, Vol. 48, 2113-
913 2121.

- 914 Hamilton, D.L., and Henderson, C.M.B. (1968), The preparation of silicate compositions by a gelling
915 method. *Mineral. Mag.*, Vol. 36, 832-838.
- 916 Harmer M.P. and Brook R.J., (1980), The effect of MgO additions on the kinetics of hot pressing in
917 Al₂O₃. *J. Mater. Sci.*, Vol. 15, 3017-3024.
- 918 Harris, N. (2007), Channel flow and the Himalayan–Tibetan orogen: a critical review, *J. Geol. Soc.*, Vol.
919 164(3), 511-523, doi: 10.1144/0016-76492006-133
- 920 Hess, P.C., (1994), Thermodynamics of thin fluid films. *J. Geophys. Res.*, Vol. 99, B4, 7219-7229.
- 921 Hetherington, G., Jack, K.H. and Kennedy, J.C. (1964), The viscosity of vitreous silica. *Phys. Chem.*
922 *Glass.*, Vol. 5, 130-136.
- 923 Hier-Majumder, S., P.H. Leo and D.L. Kohlstedt (2004), On grain boundary wetting during deformation.
924 *Acta Mater.* Vol. 52, 3425-3433, doi:10.1016/j.actamat.2004.03.040
- 925 Hier-Majumder, S., S. Mei, and D.L. Kohlstedt (2005), Water weakening of clinopyroxene in the diffusion
926 creep regime, *J. Geophys. Res.* Vol. 110, B07406, doi:10.1029/2004JB003414
- 927 Hiraga, T., Anderson, I., Zimmerman, M., Mei, S. and Kohlstedt, D. (2002), Structure and chemistry of
928 grain boundaries in deformed, olivine + basalt and partially molten lherzolite aggregates: evidence of
929 melt-free grain boundaries. *Contrib. Mineral. Petrol.*, Vol. 144, 163-175, doi: 10.1007/s00410-002-0394-
930 1
- 931 Hirth, G. and Kohlstedt, D.L., (1995a), Experimental constraints on the dynamics of the partially molten
932 upper mantle: Deformation in the diffusion creep regime. *J. Geophys. Res.*, Vol. 100, B2, 1981-2001.
- 933 Hirth, G. and Kohlstedt, D.L., (1995b), Experimental constraints on the dynamics of the partially molten
934 upper mantle: 2 Deformation in the dislocation creep regime. *J. Geophys. Res.* Vol. 100, B8, 15441-
935 15449.
- 936 Hofmann, A.W. (1980), Diffusion in silicate melts: a critical review. In *Physics of Magmatic Processes*
937 (ed. R.B. Hargraves), 385-417. Princeton Univ. Press.

- 938 Hofmeister, A. and Rossman, G., (1985), A model for the Irradiative Coloration of Smoky Feldspar and
939 the Inhibiting Influence of Water. *Phys. Chem. Mineral.*, Vol. 12: 324-332.
- 940 Holtzman, B., Groebner, N., Zimmerman, M., Ginsberg, S. and Kohlstedt, D. (2003), Stress-driven melt
941 segregation in partially molten rocks. *Geochem. Geophys. Geosyst.*, Vol. 4, Art. No. 8607.
- 942 Holtzman B.K. and Kohlstedt D.L., (2007), Stress driven melt segregation and strain partitioning in
943 partially molten rocks: Effects of stress and strain. *J. Petrol.*, Vol. 48, 2379-2406.
- 944 Houlier, B., Cheraghmakani, M., Jaoul, O., (1990), Silicon diffusion in San-Carlos olivine. *Phys. Earth
945 Planet. Interiors.*, Vol. 62, 329-340.
- 946 Huang, Y. and Langdon, T.G. (2002), Characterization of deformation processes in a Zn-22% Al alloy
947 using atomic force microscopy. *J. Mat. Sci.*, Vol. 37(23), 4993-4998.
- 948 Ildefonse, B. and Nicolas, B. (1997), Ecoulement magmatique dans les gabbros et problème des chambres
949 magmatiques sous les dorsales océaniques. In "Des grands écoulements naturels à la dynamique du tas de
950 sable : introduction aux suspensions concentrées en géologie et en physique" (B. Ildefonse, C. Allain &
951 P. Coussot, eds.), Cemagref Editions, 37-49.
- 952 Jaoul, O., Poumellec, M., Froidevaux, A. and Havette, A., (1981), Silicon diffusion in forsterite: a new
953 constraint for understanding mantle deformation. In *Anelasticity in the Earth* (Stacey FD, Paterson MS
954 and Nicolas A Eds.), AGU Geodyn. Ser. Vol. 4, 95-100.
- 955 Jaoul, O., Sautter, V. and Abel, F. (1991), Nuclear microanalysis: A powerful tool for measuring low
956 atomic diffusivity with mineralogical applications. In "Advances in Physical Geochemistry, Diffusion,
957 Atomic Ordering, and Mass Transport: Selected Problems in Geochemistry", J. Ganguly, ed., Springer-
958 Verlag, Berlin, Vol. 6, 198-200.
- 959 Jin, Q., Wilkinson, D.S. and Weatherly, G.C. (1995), Redistribution of a grain boundary glass phase
960 during creep of Si₃N₄ ceramics. *J. Am. Ceram. Soc.*, Vol. 72: 211-214.

- 961 Jin, Z.M., Green, H.W. and Zhou, Y. (1994), Melt topology in partially molten mantle peridotite during
962 ductile deformation. *Nature*, Vol. 372, 164-167.
- 963 Jung, H. and Waff, H.S. (1998), Olivine crystallographic control and anisotropic melt distribution in
964 ultramafic partial melts. *Geophys. Res. Lett.*, Vol. 25, 2901-2904.
- 965 Jurewitz, S. R. and E. B. Watson (1985), The distribution of partial melt in a granitic system: the
966 application of liquid phase sintering theory. *geochim. Cosmochim. Acta*, Vol. 49, 1109-1121.
- 967 Kajihara, K., Yoshizawa, Y. and Sakuma, T. (1995), The enhancement of superplastic flow in tetragonal
968 zirconia polycrystals with SiO₂-doping. *Acta Metall. Mater.* Vol. 43, 1235-1242.
- 969 Katz R.F., Spiegelman M. and Holtzman B. (2006), The dynamics of melt and shear localization in
970 partially molten aggregates, *Nature*, Vol. 442, 676-679.
- 971 Kohlstedt, D.L. (1992), Structure, Rheology and permeability of partially Molten Rocks at Low Melt
972 Fractions. *Mantle Flow and Melt Generation at Mid - Ocean Ridges*. AGU Geophys. Monograph Vol. 71,
973 10-121.
- 974 Kohlstedt, D.L. and Zimmermann, M.E. (1996), Rheology of partially molten mantle rocks. *Annu. Rev.*
975 *Earth Planet. Sci.*, Vol. 24, 41-62.
- 976 Kress, V.C. and Ghiorso, M.S. (1995), Multicomponent diffusion in basaltic melts. *Geochim.*
977 *Cosmochim. Acta*. Vol. 59(2) 313-324.
- 978 Lamoureux, G., B. Ildefonse, and D. Mainprice (1999), Modelling the seismic properties of fast-spreading
979 ridge crustal LVZ: insights from Oman gabbro textures. *Tectonophysics*, Vol. 312, 283–301.
- 980 Laporte, D. and Watson, E.B. (1995), Experimental and theoretical constraints on melt distribution in
981 crustal sources: the effect of crystalline anisotropy on melt interconnectivity. *Chem. Geol.*, Vol. 124, 161-
982 184.

- 983 Laporte, D., Rapaille, C. and Provost, A. (1997), Wetting angles, equilibrium melt geometry, and the
984 permeability threshold of partially molten crustal protoliths. In *Granite: from segregation of melt to*
985 *emplacement fabrics.* (ed. J.L. Bouchez), Kluwer Academic Publishers, Netherlands, 31-54.
- 986 Legall, M., Lesage, B. and Bernardini, J. (1994), Self-diffusion in alpha-Al₂O₃. 1. Aluminium diffusion
987 in single-crystals. *Philos. Mag. A*, Vol. 70(5), 761-773.
- 988 Lesher, C.E., Hervig, R.L. and Tinker, D. (1996), Self diffusion of network formers (silicon and oxygen)
989 in naturally occurring basaltic liquid. *Geochim. Cosmochim. Acta.* Vol. 60(3), 405-413.
- 990 Liang, Y., Richter, F.M., Davis, A.M. and Watson, B. (1996), Diffusion in silicate melts: I. Self-diffusion
991 in CaO – Al₂O₃ – SiO₂ at 1500°C and 1 GPa. *Geochim. Cosmochim. Acta.*, Vol. 60, 4353-4367.
- 992 Liu, M., Peterson, J.C. and Yund, R.A. (1997), Diffusion-controlled growth of albite and pyroxene
993 reaction rims. *Contrib. Mineral. Petrol.*, Vol. 126, 217-223.
- 994 Mecklenburgh, J. and Rutter, E.H. (2003), On the rheology of partially molten synthetic granite. *J. Struct.*
995 *Geol.*, Vol. 25, 1575-1585.
- 996 Meléndez-Martinez, J.J. and Dominguez-Rodriguez, A. (2004), Creep of silicon nitride. *Prog. Mat. Sci.*,
997 Vol. 49, 19-107.
- 998 Mei, S. and Kohlstedt, D.L. (2000), Influence of Water on Plastic Deformation of Olivine Aggregates: 1.
999 Diffusion Creep Regime. *J. Geophys. Res.* Vol. 105(21), 457-21,469.
- 1000 Mei, S., Bai, W., Hiraga, T. and Kohlstedt, D.L. (2002), Influence of melt on the creep behaviour of
1001 olivine – basalt aggregates under hydrous conditions. *Earth Planet. Sci. Lett.* Vol. 201, 491 – 507.
- 1002 Merkle, K.L., (1997), Atomic-Scale Grain Boundary Relaxation Modes in Metals and Ceramics.
1003 *Microscopy and Microanalysis*, Vol. 3(4), 339-351.
- 1004 Milke, R., Wiedenbeck, M. & Heinrich, W. (2001), Grain boundary diffusion of Si, Mg, and O in enstatite
1005 reaction rims: a SIMS study with isotopically doped reactands. *Contrib Mineral Petrol*, Vol. 14, 15-26.
- 1006 Mysen B.O. (1983), The structure of silicate melts, *Ann. Rev. Earth Planet Sci.*, Vol. 11, 75-97.

- 1007 Mysen, B.O. (1988), *Structure and Properties of Silicate Melts*. Elsevier, pp 354.
- 1008 Mysen, B.O. (2003), *Physics and chemistry of silicate glasses and melts*. *Eur. J. Mineral.* Vol. 15, 781-
1009 802.
- 1010 Mungal, J.E. (2002), *Empirical models relating viscosity and tracer diffusion in magmatic silicate melts*.
1011 *Geochim. Cosmochim. Acta.*, Vol. 66 (1), 125-143.
- 1012 Nascimento, M.L.F. and Zanotto, E.D. (2006), *Mechanisms and dynamics of crystal growth, viscous flow*
1013 *and self-diffusion in silica glass*. *Phys. Rev. B*, Vol. 73, 024209.
- 1014 Nicolas, A. and Ildefonse, B. (1996), *Flow mechanism and viscosity in basaltic magma chambers*.
1015 *Geophys. Res. Lett.*, Vol. 16, 2013-2016.
- 1016 Ojovan, M.I. and Lee W.E., (2004), *Viscosity of network liquids within Doremus approach*. *J. Appl. Phys.*
1017 Vol. 95, 3803-3810.
- 1018 Paterson, M.S., (1982), *The determination of hydroxyl by infrared absorption in quartz, silicate glass and*
1019 *similar materials*. *Bull. Mineral.*, Vol. 105, 20-29.
- 1020 Paterson, M.S. (2001), *A granular flow theory for the deformation of partially molten rock*.
1021 *Tectonophysics*, Vol. 335, 51-61.
- 1022 Pharr, G.M. and Ashby, M.F. (1983), *On creep enhanced by a liquid phase*. *Acta Metall.*, Vol. 31, 129-
1023 138.
- 1024 Raj, R. (1982), *Creep in polycrystalline aggregates by matter transport through a liquid phase*. *J. Geophys.*
1025 *Res.* Vol. 87, B6, 4731-4739.
- 1026 Rubie D.C. (1986), *The catalysis of mineral reactions by water and restrictions on the presence of aqueous*
1027 *fluid during metamorphism*. *Mineral. Mag.*, Vol. 50, 399-415.
- 1028 Russell, J.K. and Giordano, D. (2005), *A model for silicate melt viscosity in the System CaMgSi₂O₆-*
1029 *CaAl₂Si₂O₈-NaAlSi₃O₈*. *Geochim. Cosmochim. Acta* Vol. 69, 5333-5349.

- 1030 Giordano, D., J. K. Russell and D.B. Dingwell (2008), Viscosity of magmatic liquids: A model, Earth and
1031 Planet Sci Lett, Vol. 271, 123-134.
- 1032 Reid, J.E., B.T. Poe, D.C. Rubie, N. Zotov, M. Wiedenbeck (2001), The self-diffusion of silicon and
1033 oxygen in diopside ($\text{CaMgSi}_2\text{O}_6$) liquid up to 15 GPa. Chem. Geol., Vol. 174, 77–86.
- 1034 Rybacki, E. and Dresen, G. (2000), Dislocation and diffusion creep of synthetic anorthite aggregates. J.
1035 Geophys. Res., Vol. 105 (B11), 26017-26036. DOI:10.1029/2000JB900223
- 1036 Schäfer, F.N. and Foley, S.F. (2002), The effect of crystal orientation on the wetting behaviour of silicate
1037 melts on the surfaces of spinel peridotite minerals. Contrib. Mineral. Petrol., Vol. 143(2), 254-262.
- 1038 Shimizu, N. and Kushiro, I. (1984), Diffusivity of oxygen in jadeite and diopside melts at high pressures.
1039 Geochim. Cosmochim. Acta., Vol. 48, 1295-1303.
- 1040 Shimizu, N. and Kushiro, I. (1991) The mobility of Mg, Ca and Si in diopside-jadeite liquids at high
1041 pressures. In Physical Chemistry of Magmas, Advances in Physical Geochemistry, (eds. L.L. Perchuk and
1042 I. Kushiro), Vol. 9, 192-212, Springer-Verlag.
- 1043 Spiess, R., Dibona, R., Rybacki, E., Wirth, R. and Dresen, G. (2012), Depressurized Cavities within High-
1044 strain Shear Zones: their Role in the Segregation and Flow of SiO_2 -rich Melt in Feldspar-dominated
1045 Rocks. J. Petrol., Vol. 53(9), 1767-1776. doi:10.1093/petrology/egs032.
- 1046 Sutton, A.P. and V. Vitek (1983), On the Structure of Tilt Grain Boundaries in Cubic Metals I.
1047 Symmetrical Tilt Boundaries. Phil. Trans. Roy. Soc. London A, Vol. 309, 1-36;
1048 DOI:10.1098/rsta.1983.0020.
- 1049 Sutton, A.P. and Baluffi R.W. (1987), Overview no. 61 On geometric criteria for low interfacial energy.
1050 Acta Metall. Vol. 35 (9), 2177-2201.
- 1051 Sutton, A.P. (1989), On the structural unit model of grain boundary structure. Phil. Mag. Lett. Vol. 59 (2),
1052 53-59.

- 1053 Urbain, G. Y. Bottinga and P. Richet (1982), Viscosity of liquid silica, silicates and aluminosilicates.
1054 *Geochim. Cosmochim. Acta*, Vol. 46 (6), 1061–1072.
- 1055 Van der Molen, I. and Paterson, M.S. (1979), Experimental deformation of partially molten granite.
1056 *Contrib. Mineral. Petrol.*, Vol. 70, 299-318.
- 1057 van der Merwe (2001), Interfacial energy: bicrystals of semi-infinite crystals. *Prog. Surf. Sci.*, Vol. 67,
1058 356-381.
- 1059 Vigneresse, J.L., Barbey, P. and Cuney, M. (1996), Rheological transitions during partial melting and
1060 crystallization with application to felsic magma segregation and transfer. *J. Petr.*, Vol. 37, 1579-1600.
- 1061 Von Bargen, N. and Waff, H.S. (1986), Permeabilities, interfacial areas and curvatures of partially molten
1062 systems: Results of numerical computations of equilibrium microstructures. *J. Geophys. Res.*, Vol. 91,
1063 B9, 9261-9276.
- 1064 Waff, H.S. and Bulau J.R. (1979), Equilibrium fluid distribution in an ultramafic partial melt under
1065 hydrostatic stress conditions. *J. Geophys. Res.*, Vol. 84, 6109–6114.
- 1066 Waff, H.S. and J.R. Bulau (1982), Experimental studies of near-equilibrium textures in partially molten
1067 silicates at high pressures. In *High Pressure Res. Geophys., Adv. Earth Planet. Sci.*, Vol. 12, 229-236.
- 1068 Waff, H.S. and Faul, U.H. (1992), Effects of crystalline anisotropy on fluid distribution in ultramafic
1069 partial melts. *J. Geophys. Res.*, Vol. 97, 9003-9014.
- 1070 Walte, N.P., P.D. Bons, C.W. Passchier and D. Koehn (2003), Disequilibrium melt distribution during
1071 static recrystallization. *Geology*, Vol. 31, 1009–1012.
- 1072 Wanamaker, B.J. and Kohlstedt, D.L. (1991), The effect of melt composition on the wetting angle between
1073 silicate melts and olivine. *Phys. Chem. Mineral.*, Vol. 18, 26-36.
- 1074 Wolfenstine, J. and Kohlstedt, D.L. (1994), High-temperature creep and kinetic decomposition of
1075 Ni_2SiO_4 . *Phys. Chem. Mineral.*, Vol. 21, 234-243.

- 1076 Wang, Z., Dresen, G. and Wirth, R. (1996), Diffusion creep of fine-grained polycrystalline anorthite at
1077 high temperature, *Geophysical Research Letters*, Vol. 23, 3111-3114.
- 1078 Wark, D.A., Williams, C.A., Watson, E.B., Price, J.D. (2003), Reassessment of pore shapes in
1079 microstructurally equilibrated rocks, with implications for permeability of the upper mantle. *J. Geophys.*
1080 *Res.* Vol. 108 (B1), 2050. doi:10.1029/ 2001JB001575
- 1081 Wendlandt, R.F. (1991), Oxygen diffusion in basalt and andesite melts: experimental results and
1082 discussions of chemical versus tracer diffusion. *Contrib. Mineral. Petrol.* Vol. 108, 463-471.
- 1083 Wilkinson, D.S. (1998), Creep Mechanisms in Multi-phase Ceramic Materials, Feature Article, *J. Amer.*
1084 *Ceram. Soc.*, Vol. 81, 275-299.
- 1085 Wirth, R. (1996), Thin amorphous films (1-2 nm) at olivine grain boundaries in mantle xenoliths from
1086 San Carlos, Arizona. *Contrib. Mineral. Petrol.*, Vol. 124, 44-54.
- 1087 Yund, R.A. (1986), Interdiffusion of NaSi-CaAl in Peristerite. *Phys. Chem. Minerals*, Vol. 13, 11-16.
- 1088 Yund, R.A. (1997), Rates of grain boundary diffusion through enstatite and forsterite reaction rims.
1089 *Contrib. Mineral. Petrol.*, Vol. 126, 224-236.
- 1090 Yund, R.A. and Farver, J.R. (1999), Si grain boundary diffusion rates in feldspar aggregates. *Abstr. EOS*,
1091 *supl. F1077.*
- 1092 Zhang, H., Srolovitz, D.J., Douglas, J. F. and Warren, J.A. (2009), Grain boundaries exhibit the dynamics
1093 of glass-forming liquids, *Proc. Nat. Acc. Sci.*, Vol. 106(19), 7735–774.
- 1094 Zimmerman, M.E. and Kohlstedt, D.L. (1999), Influence of deformation mechanism on melt topology in
1095 partially molten peridotites. *EOS Trans. AGU Suppl.*, 80, S326.
- 1096 Zimmerman, M., Zhang, E., Kohlstedt, D. and Karato, S. (1999), Melt distribution in mantle rocks
1097 deformed in shear. *Geophys. Res. Letts.*, Vol. 26, 1505-1508.