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

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

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## Abstract

The present work explores the effects of melt chemistry on diffusion controlled creep of partially molten labradorite plagioclase (An<sub>50</sub>) at anhydrous conditions. Using sol-gel and hot pressing techniques we produced: 1) nominally melt-free samples, with < 1 vol. % residual glass confined solely to multiple-grain junctions; 2) two types of partially molten samples, containing respectively ~ 1 and ~ 5 vol. % silica-rich partial melts, wetting numerous grain boundaries by thin (< 10 nm) amorphous films. Energy dispersive X-ray analysis showed that the amorphous phases of the latter materials contained ~ 85 and 95 wt. % SiO<sub>2</sub>, thus representing different polymerization degrees. Infrared spectroscopy showed that the initial traces of water (~ 0.05 wt. %) were dried out by annealing in air above 1100°C. Uniaxial creep tests performed at temperatures and flow stresses ranging respectively between 1100 – 1250°C and 3 – 60 MPa showed dominantly linear viscous flow, with a strong grain size dependence indicating grain boundary diffusion control. Counter-intuitively strength and activation energy increased with the content of melts. However, for the sample suite silica content co-varies with melt proportion, and thus our results suggest that the kinetics of grain boundary diffusion controlled creep strongly depends on melt chemistry. Instead of acting as short-cut for diffusion, thin films of highly viscous amorphous phases may in turn considerably hinder grain boundary transport properties.

**Key words:** “creep and deformation”, “plasticity, diffusion, and creep”, “microstructure”, “rheology: crust and lithosphere”

## 1. Introduction

The dynamics of various tectonic settings depend on the rheological properties of partially molten silicate rocks. The stability of the high plateau resulting from the collision context of the Himalayan-Tibetan orogen is proposed to be ensured by channel flow localized within the lower crust, and

1 43 characterized by exceptionally low viscosity owing to partial melting (Harris, 2007). The accretion of  
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4 44 oceanic crust from magma chambers beneath mid-ocean ridges involves lateral flow of gabbroic magmatic  
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6 45 suspension, as demonstrated by bedding fabrics presenting shape preferred textures, without crystal  
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8 46 preferred orientations (Nicolas and Ildefonse, 1996; Ildefonse and Nicolas, 1997). Lamoureux et al. (1999)  
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11 47 and Nicolas and Ildefonse (1996) proposed that magmatic flow could operate at melt fractions as low as  
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13 48 10 vol. %, provided grain sliding and dissolution – precipitation creep mechanisms.

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15 49 Partially molten silicates rocks are usually substantially weaker than their fully crystalline  
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17 50 counterparts (Vigneresse et al., 1996). The extent of softening depends on several different (and  
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20 51 sometimes interdependent) physical aspects, such as: 1) the volume fraction of the molten phase and its  
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22 52 grain scale topology, 2) the dominant creep mechanisms, 3) the mineralogy and chemistry of the  
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24 53 considered system. Extreme softening of partially molten silicate rocks is observed in two cases. On the  
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27 54 one hand, if the melt fraction exceeds the so called Rheologically Critical Melt Percentage (RCMP,  
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29 55 typically > 20 vol.%), the rock experiences loss of cohesion and a switch from ductile mechanisms to  
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31 56 enhanced granular and cataclastic type of flow (van der Molen and Paterson, 1979). On the other hand, at  
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34 57 low melt fractions (< 10 vol.%), grain size sensitive creep may be promoted by dissolution-precipitation  
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36 58 and diffusional mass transfer through the molten phase (Cooper and Kohlstedt, 1984; 1986; Dell'Angelo  
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38 59 et al., 1987). The latter mechanisms are especially efficient when large proportions of grain boundaries are  
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40 60 lost to the melt, and thus the extent of weakening strongly depends on the grain-scale melt distribution  
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43 61 (Cooper et al., 1989; Kohlstedt, 1992; Hirth and Kohlstedt, 1995; Dimanov et al., 1998; 2000).

44  
45 62 Melt topology depends essentially on the wetting angle  $\theta$ , which is defined as the dihedral angle  
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48 63 between the solid – liquid interfaces of two neighboring grains, at a melt-bearing triple junction. The  
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50 64 dihedral angle  $\theta$  depends on the local equilibrium of interfacial tension forces (Beere, 1975; Bulau et al.,  
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52 65 1979; Jurewitz and Watson, 1985; Fujii et al., 1986). The limit case  $\theta = 60^\circ$  corresponds to non-wetting  
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55 66 melt, forming isolated spherical pockets. When  $60^\circ > \theta > 0^\circ$ , the melt forms an interconnected network  
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1 67 of tubules along three- and higher order grain junctions. The smaller is  $\theta$ , the larger is the proportion of  
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4 68 wetted grain boundaries and the more efficient are the diffusive mass transfer processes. The limit case  
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6 69  $\theta = 0^\circ$  corresponds to wetting melt, spreading as melt films along all grain boundaries. For the usual mafic  
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8 70 and felsic silicates the dihedral angles range between  $10 - 40^\circ$ , and therefore, silicate melts form  
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11 71 interconnected networks along multiple grain junctions (Kohlstedt, 1992; Laporte et al., 1997). In more  
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13 72 details however, melt topology is sensitive to mineralogy and chemistry, to grain size distribution, but  
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15 73 also to the mechanical stress state (Fujii et al., 1986; Dell'Angelo et al., 1987; Dell'Angelo and Tullis,  
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18 74 1988; Bussod and Christie, 1991; Jin et al., 1994; Hirth and Kohlstedt, 1995; Laporte and Watson, 1995;  
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20 75 Drury and FitzGerald, 1996; Wirth, 1996; Franz and Wirth, 1997; Daines and Kohlstedt, 1997; Laporte  
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22 76 et al., 1997; Jung and Waff, 1998; Cmíral et al, 1998; Schäfer and Foley, 2002, Walte et al., 2003;  
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25 77 Dimanov et al., 2000; De Kloe et al, 2000; Wark et al., 2003; Mecklenburgh and Rutter, 2003; Hier-  
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27 78 Majumder et al., 2004; Katz et al., 2006; Holtzman and Kohlstedt, 2007; Spiess et al., 2012; Carapic et  
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29 79 al., 2013).

31 80 At low melt fraction, the extent of weakening in the grain-size sensitive regime is strongly  
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34 81 conditioned by the melt topology, because the diffusion coefficients in silicate melts are usually much  
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36 82 faster than in the solid or along grain boundaries. Partial melts confined solely to multiple grain junctions  
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38 83 cause only little to moderate weakening (Cooper and Kohlstedt 1984; 1986; Kohlstedt, 1992; Hirth and  
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41 84 Kohlstedt, 1995; Dimanov et al., 1998). Conversely, melt films wetting numerous grain boundaries induce  
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43 85 substantial creep rate enhancement (Raj, 1982; Pharr and Ashby, 1983; Cooper et al., 1989; Paterson,  
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45 86 2001; Dimanov et al., 2000). To date, the influence of melt chemistry on creep rates has only been  
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48 87 considered from the view point of the dihedral angles (Wanamaker and Kohlstedt, 1991; Kohlstedt, 1992;  
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50 88 Wolfenstine and Kohlstedt, 1994). Yet, because diffusivities differ by orders of magnitude depending on  
51  
52 89 the considered species and melt composition (Zhang et al., 2010), it may be expected that the chemical  
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54 90 composition of the molten phase may also influence the mass transfer kinetics, and hence the creep rates.  
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1 91 In the present work we focus on the rheological effects of melt chemistry at high silica content (> 80 wt.  
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4 92 % SiO<sub>2</sub>) for partially molten plagioclase aggregates presenting wetted grain boundaries.  
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## 8 94 **2. Experimental procedures**

### 10 95 **2.1. Specimen preparation**

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13 96 We used fine grained (< 16 µm) synthetic glass powder (Corning Inc.), with An<sub>50</sub>Ab<sub>50</sub> (labradorite)  
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15 97 chemical composition. X – ray fluorescence (XRF, SRS 303AS) showed that the impurity content was  
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17 98 less than 1 wt. % (Table 1). After being stored at 120°C in oven for 24 hours, the powder was first  
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19  
20 99 uniaxially cold – pressed at 300 MPa in steel jackets of 20 mm in length, 10 mm in diameter and 0.5 mm  
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22 100 thick. Then, the green bodies were subjected to hot – isostatic pressing (HIP) for 3 hours at 1150°C and  
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24 101 300 MPa in a gas-medium Paterson apparatus, resulting in nearly glass – free polycrystalline material,  
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26 102 with about 1 vol. % residual glass (Dimanov et al., 1998; 1999; 2000; 2003; Rybacki and Dresen, 2000).  
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29 103 However, this procedure results sometimes in spherulites due to fast crystallization kinetics (Rybacki and  
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31 104 Dresen, 2000). Hence, the HIPed samples were re-crushed and milled in alcohol in an agate mortar to  
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33 105 obtain homogeneously grained crystalline powders. Laser – particle analysis (Fritsch analysette 22)  
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36 106 showed that about 80% of the grains were < 10 µm. The larger particles constituted agglomerates of  
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38 107 smaller grains. The chemical composition of the powder was checked by XRF (Table 1). To remove  
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40 108 adsorbed water, the crystalline powders were annealed at 800°C for 24 hours in air and subsequently  
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43 109 stored in an oven at 120°C prior to further processing.  
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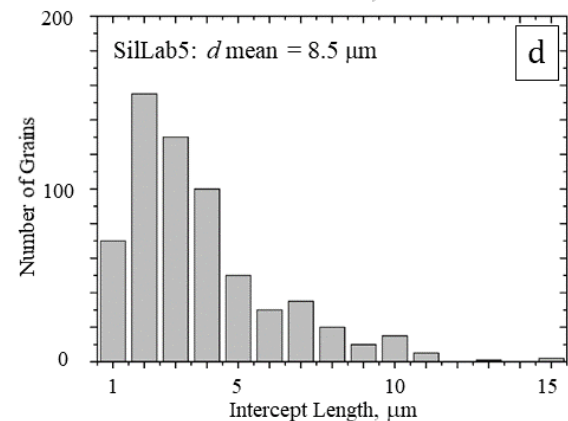
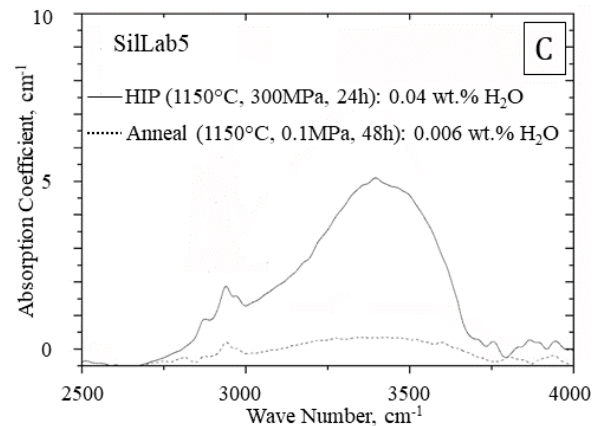
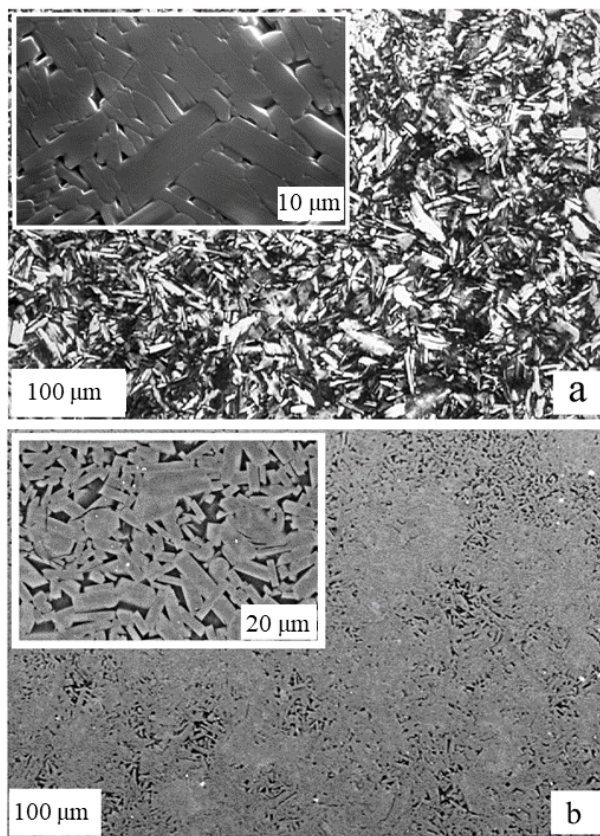
### 47 111 **Melt-free samples**

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50 112 Nominally melt-free samples were first produced, in order to have a rheological reference. The  
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52 113 prepared crystalline powders were subjected to a second round of cold- and hot-pressing. HIP at 1150°C  
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54 114 and 300 MPa for 24 hours. The resulting materials, further referred to as “Lab” samples, are dense and  
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115 polycrystalline. They are considered as nominally melt-free, although small amounts (< 1 vol. %) of  
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 4116 residual amorphous phase persisted at multiple grain junctions.

6117 Scanning electron microscopy (SEM, Zeiss DSM 962) and the line intercept method were applied  
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 8118 on polished and thermally etched specimens (Dimanov et al., 1998, 1999, 2000; 2003). The obtained  
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 10119 microstructures are characterized by lath shaped grains with aspect ratios of 2 to 3, and log – normal grain  
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 12 size distribution (Fig. 1a). A mean arithmetic grain size of  $9.8 \pm 3.2 \mu\text{m}$  was obtained from five SEM  
 1320 micrographs, based on the linear intercept method and a correction factor accounting for the aspect ratio  
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 1521 (for further details, see Dimanov et al., 1998).

2023 Transmission electron microscopy (TEM, Philips CM 200 Twin) showed that nearly half of the  
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 2224 grains present numerous and dense growth twins, but average dislocation densities are low ( $< 10^7 \text{ cm}^{-2}$ )  
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 2425 (Fig. 2a). Residual amorphous phase with plagioclase-like composition may remain at some multiple grain  
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 2626 junctions (Fig. 2b).



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Fig. 1: Characterization of microstructures at optical and SEM scales. a) Optical micrograph of large representative area from thin section and SEM micrograph (insert, SE mode) of polished and thermally etched melt - free specimen (Lab). b) SEM micrographs (BSE mode) of polished melt-bearing specimen (SilLab5). At sub-mm scale melt distribution shows local heterogeneities, with local melt fractions reaching up to 10 vol. % (insert). c) Water content after initial HIP and after drying anneal from FTIR measurements on SilLab5 samples d) Grain size distribution for SilLab5 samples obtained by the intercept method performed on SEM micrographs.

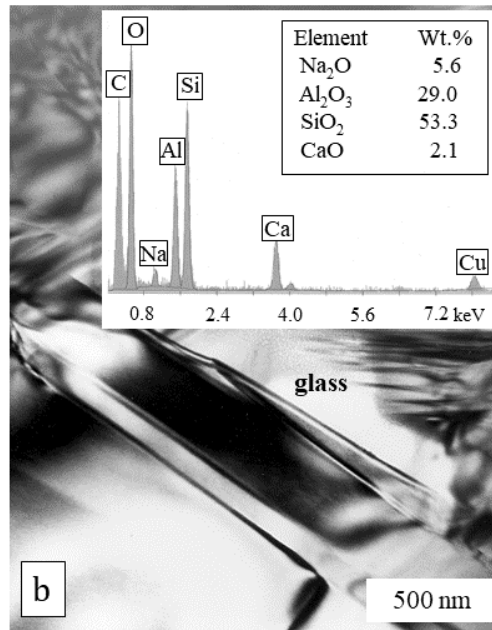
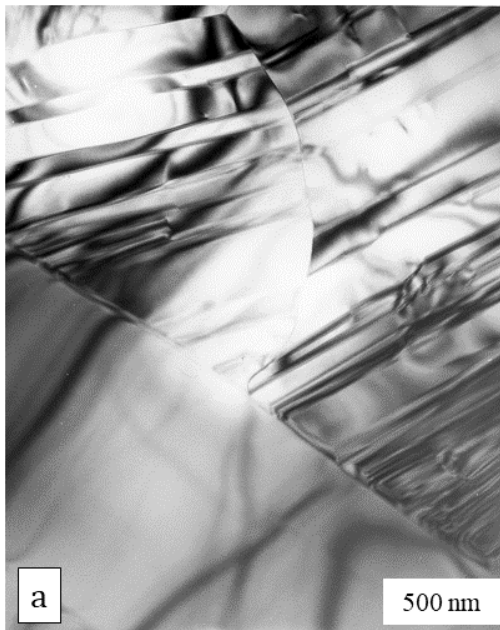
Fluid inclusions were also observed within crystals, at grain boundaries and in the glassy-phase. Fourier transform infra-red spectroscopy (FTIR, Brüker IFS 66v) was applied on 5x5 mm<sup>2</sup> doubly polished sections of 150 µm in thickness. We operated in the range 2000 - 4000 cm<sup>-1</sup> and we applied a third – order polynomial fit for background correction. We observed a broad absorption band centered at about 3350 cm<sup>-1</sup> (Fig. 1c), indicating free molecular water present in intra – and inter – crystalline fluid inclusions, and/or dissolved within the residual glass and at grain boundaries (Dimanov et al., 1999). A few sharp peaks indicated that hydrous species were also incorporated within the crystalline structure (Hofmeister and Rossman, 1985; Beran, 1986; 1987). The samples contain 0.05 ± 0.025 wt. % H<sub>2</sub>O (≈ 6000 ± 3000 ppm H/Si), based on the calculation approach of Paterson (1982) and the linear molar absorptivity coefficient for plagioclase (Beran, 1987). For further details, see Dimanov et al. (1998; 1999) and Rybacki and Dresen (2000). The incorporated water is due to adsorption onto the starting glass and crystalline particles, during temporary storage and handling at room condition for cold-pressing. Annealing of the samples for 24 – 48 hours in air at temperatures above 1100°C at 0.1 MPa resulted in drying out most of the water traces down to < 0.002 wt. % H<sub>2</sub>O (Fig.1, Dimanov et al., 1999).

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 <sub>2</sub> O(wt.%)	5.68	5.16	0.04	5.5	1.1	2.7
Al <sub>2</sub> O <sub>3</sub> (wt.%)	28.43	29.07	0.23	29.0	3.3	8.7
SiO <sub>2</sub> (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 <sub>2</sub> O <sub>3</sub> (wt.ppm)	0.0	0.0	0.0	0.0	0.0	0.3
ZrO <sub>2</sub> (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

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



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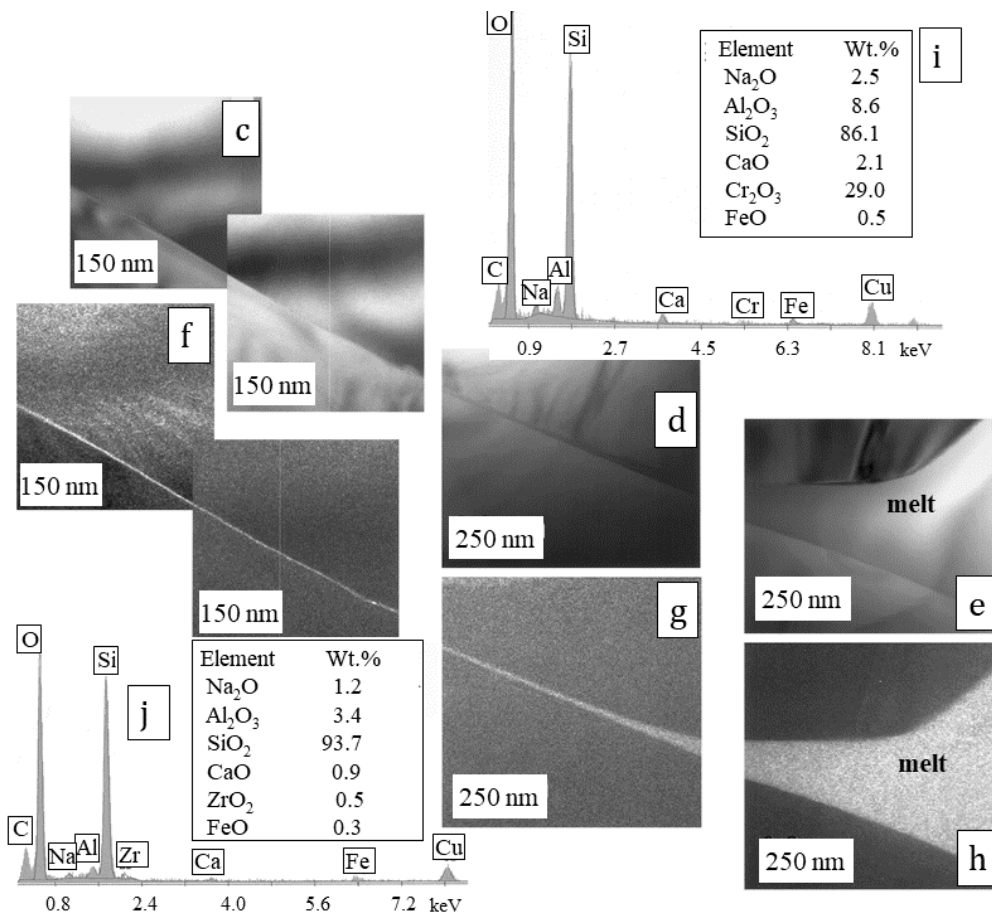


Fig. 2: Characterization of microstructures at TEM scale and EDX analysis of the chemical composition of amorphous phases.

a) Grains exhibit low dislocation densities, but growth twins are numerous. b) Residual glass pockets (< 1 vol.%) are commonly found at multiple grain junctions in Lab samples. EDX analyses (insert) show that their composition is comparable to plagioclase. c-h) SilLab1 and SilLab5 materials present amorphous pockets at multiple junctions, but most interfaces also show thin amorphous films (< 10 nm) extending from triple junction channels, as evidenced by bright versus dark field imaging. i-j) EDX analysis of amorphous phases performed at melt pockets located at multiple grain junctions in SilLab1 and SilLab5 samples. Note that carbon coating of the non-conductive samples results in the presence of carbon.

### Melt – bearing samples

Dimanov et al. (2000) studied partially molten labradorite materials with a few percent of melt wetting numerous grain boundaries and containing up to 80 wt. % SiO<sub>2</sub>. These samples showed to be substantially weaker than their fully crystalline counterparts. In the present study we aimed to investigate the rheological effects of wetting melts with variable and higher silica contents. For this purpose, we

1 applied the sol–gel method (Hamilton and Henderson, 1968), which was previously used to precipitate  
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3 synthetic basalt onto olivine crystalline powders (Cooper and Kohlstedt, 1984; Beeman and Kohlstedt,  
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5 1993; Hirth and Kohlstedt, 1995). We used pure grade tetraethyl orthosilicate ( $\text{Si}(\text{C}_2\text{H}_5\text{O})_4$ , TEOS from  
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7 Aldrich) diluted with ethanol in proportions of 10 to 1. Ammonium hydroxide solution (1 mol/l, Prolabo)  
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9 was used as catalyst. In the solution, water hydrolyzes the TEOS and produces silanol groups (Si-OH),  
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11 which interact through hydrogen bonding and form hydrolyzed silica. After drying out all volatiles, we  
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13 performed XRF analysis on the precipitation product that consisted of nearly pure silica (Table 1). We  
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15 prepared two aqueous solutions, where labradorite powders and diluted TEOS were mixed at the  
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17 appropriate proportions in order to obtain 1 and 5 vol. % excess silica. The precipitation took place within  
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19 a few minutes after the addition of the ammonium hydroxide, but the suspensions were continuously  
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21 stirred for an hour. The precipitation products were dried at 100°C for 24 hours and ground in agate mortar.  
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23 The resulting powders were fired into platinum crucibles at 700°C (in air) for 24 hours in order to release  
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25 remaining volatiles. Finally, the silica – coated powders were stored in an oven at 120°C. The coated  
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27 crystalline powders were cold pressed and HIPed for 24 hours at 1150°C and 300 MPa in order to obtain  
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29 dense and equilibrated melt – bearing polycrystalline samples for deformation. Samples containing 1 and  
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31 5 vol. % excess silica glass are called SilLab1 and SilLab5, respectively. SEM investigations were applied  
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33 to carefully polished and thermally etched specimens (Fig. 1). The microstructure is comparable to those  
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35 of melt – free samples, with prismatic and lath shaped grains, and with log – normal grain size distributions  
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37 (Fig. 1d). The intercept method determined mean arithmetic grain sizes of  $8.6 \pm 3.4$  and  $9.3 \pm 3.2$   $\mu\text{m}$  for  
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39 SilLab5 and SilLab1 samples, respectively. The amorphous phases are more or less homogeneously  
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41 distributed at multiple grain junctions, as evidenced by SEM in back scattering mode (Fig. 1b). TEM  
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43 consistently showed that the amorphous phases are present at most multiple grain junctions, with variable  
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45 apparent dihedral angles. Additionally, most two grain boundaries are wetted by thin amorphous layers <  
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47 10 nm in thickness. These features are shown in Figures 2c to 2h. In spite of apparently large dihedral  
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191 angles melt films extend from triple junctions within grain boundaries. In addition, the melt film  
192 thicknesses gradually increase in the vicinity of the triple junctions, hence suggesting that there are not  
193 finite wetting angles. These observations are in part similar to those of Dimanov et al. (2000) for partially  
194 molten plagioclase and of Hiraga et al. (2002) for olivine – basalt system, but we did observe some  
195 differences too. In the present study we observed that 1) the melt thickness stabilizes at around 10 nm at  
196 distances of less than a micrometer from the triple junction, 2) the melt films are a general feature and not  
197 exclusively related to low index crystallographic planes as in the latter study. Energy dispersive X ray  
198 analysis (EDX) was performed at triple junctions and evidenced that during HIPing the precipitated silica  
199 equilibrated chemically with the residual labradorite glass ( $\approx 1$  vol. %), resulting in liquid phases with  $\approx$   
200  $95 \pm 2$  wt. % and  $\approx 85 \pm 2$  wt. %  $\text{SiO}_2$ , respectively for SilLab5 and SilLab1 samples (Fig. 2i and 2j, Tab.  
201 1). TEM also evidences the presence of nm scale fluid inclusions in the amorphous phases. FTIR  
202 spectroscopy showed similar broad absorption bands and corresponding hydroxyl concentrations ( $\approx 0.05$   
203  $\pm 0.025$  wt. %  $\text{H}_2\text{O}$ ) for both melt – bearing and melt – free samples (Fig. 1c). The traces of water could  
204 be substantially dried out by annealing in air above  $1100^\circ\text{C}$  for 48 hours (Fig. 1c).

## 2.2. Experimental deformation

207 Parallelepiped specimens with dimensions of 2.5 mm aside and 5 mm in length were cut from hot  
208 pressed samples with low speed diamond saw. Compression and observation surfaces were polished to a  
209 finish of  $1 \mu\text{m}$  and  $0.3 \mu\text{m}$ , respectively. Uniaxial creep tests were performed stepwise at constant  
210 temperatures and constant loads in a dead – load apparatus (Dimanov et al., 1998; 1999; 2000, 2003).  
211 Finite strains for individual steps ranged from 3 to 0.5 %. The details of deformation assemblies and  
212 procedures are same as reported in previous works (Dimanov et al., 1998; 1999; 2000). Axial stresses and  
213 temperatures were varied stepwise between 3 – 60 MPa and  $1100 - 1225^\circ\text{C}$ , respectively. Total sample  
214 strain was less than 15 %. Samples were cooled under load. Fast cooling within 10 minutes was only

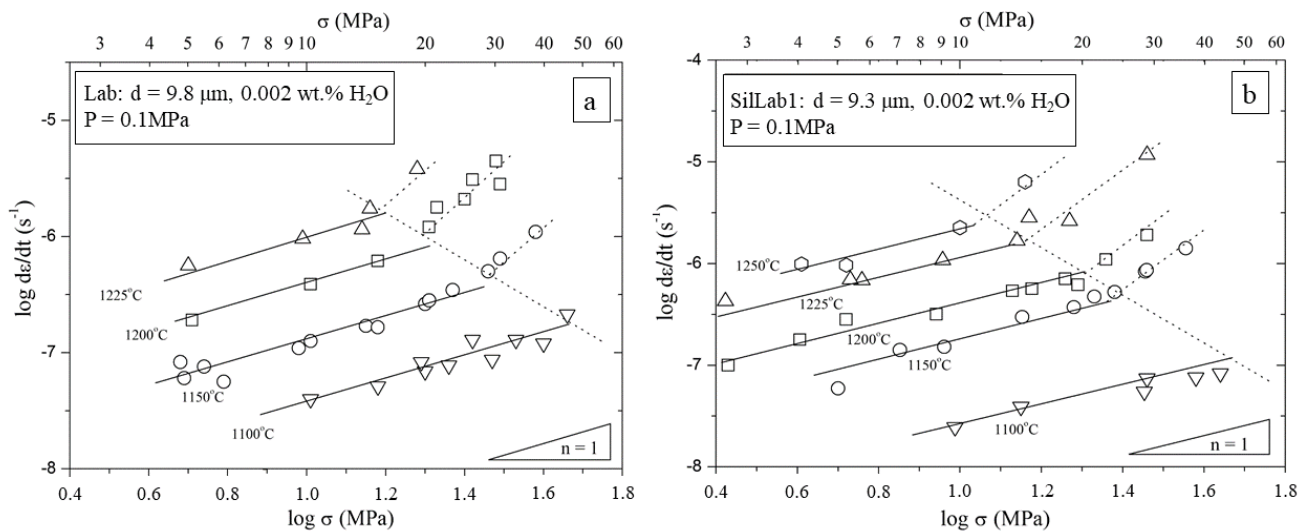
possible down to 900°C, due to furnace inertial. Further cooling down to room temperature lasted for about two hours. For comparison between static and dynamic conditions, one specimen (lab-04) was only statically annealed at 1200°C, which is an intermediate temperature with respect to the experimental temperature range for the crept samples (Tab. 2, electronic supplement).

### 3. Results

#### 3.1. Mechanical Data

##### Stress sensitivity

The mechanical data are provided as electronic supplement (Tab. 2, electronic supplement). All materials showed an extended range of stress conditions characterized by a linear relation between strain rates and flow stresses (stress exponent  $n \approx 1$ , Fig. 3a,b,c), evidencing linear viscous (Newtonian) flow. A transition to power-law creep ( $n \approx 3$ ) could be marginally observed at the highest stresses.



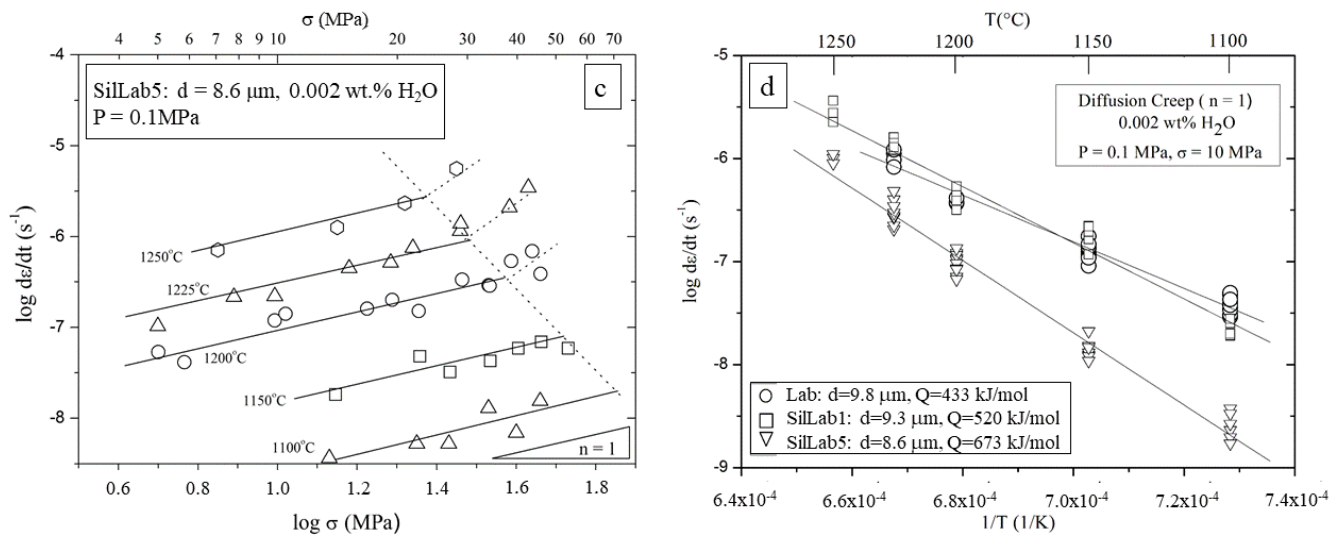


Fig. 3: Thermo-mechanical data for all tested materials. a, b and c: log Stress - log Strain rate plots for Lab, SilLab1 and SilLab5 samples, with respective mean grain sizes of 9.8, 9.3 and 8.6 μm). All materials show dominantly linear viscous (Newtonian) flow (stress exponent  $n = 1$ ) and a transition (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). d: Arrhenius diagram of the data for the linear viscous regime for Lab, SilLab1 and SilLab5 samples, recalculated to 10 MPa flow stress showing the Newtonian flow laws. Noteworthy, Lab and SilLab materials present comparable strain rates, whilst SilLab5 material is considerably more resistant. Activation energy increases with melt content.

In this study we are essentially interested in the linear viscous Newtonian flow. Such regimes either relate to Harper-Dorn dislocation-mediated mechanisms, or to grain sliding and/or diffusional mass transfer. In the former and latter cases, the strain rates are respectively grain size sensitive and insensitive. The previous works of Dimanov et al. (1998; 1999; 2000; 2003; 2011) have evidenced for similar materials grain size sensitive flow, which may be described by the constitutive equation:

$$\dot{\epsilon} = A \sigma d^m e^{-\frac{Q}{RT}}, \quad (1)$$

where  $A$  (s<sup>-1</sup>MPa<sup>-n</sup>μm<sup>m</sup>) is a material related constant.

## Grain size sensitivity

1247 On the one hand, we did not investigate samples with different grain sizes. On the other hand,  
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4248 grain growth was not observed during annealing and creep (Table 2, electronic supplement), possibly  
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6249 because of slow grain boundary migration in the presence of interfacial molten phases. Therefore, we  
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8250 cannot directly measure the dependence of strain rate on grain size. However, a first order estimation may  
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11251 be obtained if we compare the present data for Lab samples (mean grain size  $d \approx 10 \mu\text{m}$ ) with data from  
12  
13252 Dimanov et al. (1998, 2000). These former data correspond to similar but coarser grained labradorite  
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15253 samples, with mean grain sizes  $d \approx 14.5 \mu\text{m}$  (Dimanov et al., 2000) and  $d \approx 16.5 \mu\text{m}$  (Dimanov et al.,  
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17  
18254 1998). We considered exclusively the data corresponding to the Newtonian regime, for temperatures  
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20255 ranging between 1200°C and 1225°C. The data from Dimanov et al. (1998) and from Dimanov et al (2000)  
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22256 correspond respectively to 1213°C and 1210°C (their samples HIP1-01, -02 and -03). The data from the  
23  
24  
25257 present work correspond to 1200°C and 1225°C (samples lab03 and lab05, Table 2, supplementary data).  
26  
27258 The activation energies of the flow laws reported by Dimanov et al (2000) and in the present work (396  
28  
29259 kJ/mol and 433 kJ/mol, respectively) are comparable within uncertainties. The data from the three studies  
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31  
32260 were reduced to the same flow stress  $\sigma = 10 \text{ MPa}$  using  $n = 1$ , and to an intermediate temperature  $T =$   
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34261 1213°C using their respective activation energies. A linear regression of the reduced data sets for the three  
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36262 distinct grain sizes yields  $m = -2.7 \pm 0.2$  (Fig. 4a), in agreement with previous studies for anorthite  
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39263 plagioclase (Wang et al., 1996; Dimanov et al., 1999) and suggests grain boundary diffusion controlled  
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41264 mechanisms (Coble, 1963).  
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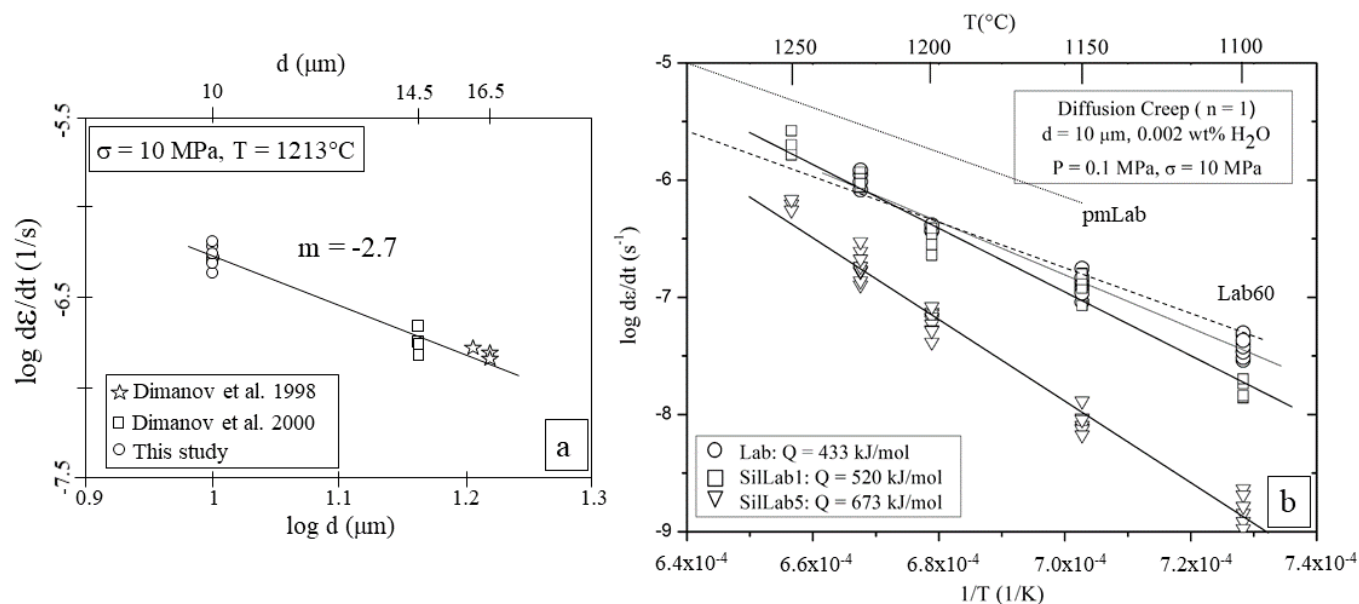


Fig. 4: Thermo-mechanical data for present and previously tested materials. a: log Grain Size - log Strain rate diagram. The data sets are from the present work and from previously published data for similar materials with different grain sizes and reduced to the same flow stress and temperature (see text, Dimanov et al., 1998; 2000). b: Arrhenius diagram, comparing the flow laws for Lab, SilLab1 and SilLab5 materials, for data reduced to the same grain size  $d = 10 \mu\text{m}$ . Are reported for comparison previously published flow laws for 1) nominally melt-free labradorite Lab60 containing ca 2 vol. % non-wetting melt from residual glass (comparable to Lab material), 2) partially molten labradorite pmLab containing ca 2 vol. % of residual non-wetting melt, plus ca 1 vol.% of wetting incongruent partial melt with ca 80 wt.% silica (Dimanov et al., 2000).

Considering the former experimental studies, the evidences for grain sliding mechanisms documented by Dimanov et al. (2007) for fine grains anorthite – diopside materials, and the theoretical approach of Ashby and Verall (1973), the present experimental data corresponding to the linear – viscous regime are compatible with the mechanisms of grain sliding accommodated by grain boundary diffusion. In the following, the raw mechanical data will be normalized with respect to grain size with the theoretical value for Coble type of creep:  $m = -3$ . It is noteworthy that the mean grain sizes for the three materials are not very different, so that the normalization procedure introduces correction factors of respectively  $\approx 0.7$  and  $\approx 0.6$  for SilLab5 and SilLab1, which is actually within the experimental scatter. Also, a hypothetical

normalization of mean grain sizes with  $m = -2$  provides corresponding correction factors of 0.9 and 0.7, which do not modify the ground analysis of the experimental results provided in the following.

### Activation energy

The parameters of the flow law equation (1) for the different materials Lab, SilLab1 and SilLab5 were obtained by least square regression fits of the respective mechanical data, corresponding to the Newtonian regime (Fig. 3a, 4b, 4c). The results are reported in an Arrhenius diagram (Figure 4b), where the grain sizes for three materials are normalized to a grain size  $d = 10 \mu\text{m}$  and to a flow stress  $\sigma = 10 \text{ MPa}$  using a grain size exponent  $m = -3$  and a stress exponent  $n = 1$ . The corresponding flow laws are listed respectively below:

$$\text{for Lab : } \log A_0 (\text{s}^{-1}\text{MPa}^{-n}) = 9.03 \pm 0.52, n = 1, \quad Q = 433 \pm 14 \text{ kJ/mol};$$

$$\text{for SilLab1 : } \log A_0 (\text{s}^{-1}\text{MPa}^{-n}) = 12.08 \pm 0.76, n = 1, \quad Q = 520 \pm 21 \text{ kJ/mol};$$

$$\text{for SilLab5 : } \log A_0 (\text{s}^{-1}\text{MPa}^{-n}) = 16.73 \pm 0.64, n = 1, \quad Q = 673 \pm 18 \text{ kJ/mol}.$$

Note the pre-exponential factor  $A$  is written as  $A_0 (\text{s}^{-1}\text{MPa}^{-n})$ , incorporating the microstructural state variable  $d (\mu\text{m})$ . The activation energy and the strain rates for Lab samples are consistent with previously published data for diffusion controlled creep of similar labradorite (Lab60) aggregates (Fig. 4), containing very small amounts of residual non-wetting plagioclase-like melt (Dimanov et al., 1998, 2000). The flow laws for Lab and SilLab1 are nearly identical (Fig. 4). Conversely, SilLab5 samples exhibit substantially lower strain rates and higher activation energy, despite the fact they contain the highest proportion of melt.

### Rheological effects of melts

1 305 The materials from the present study (Lab) and a previous work (Lab60, Dimanov et al., 2000),  
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3  
4 306 containing of 1 – 2 vol. % non-wetting melts (residual starting glass: plagioclase-like composition, with  
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6 307 ca 50 wt.% silica) exhibit similar flow laws and are considered as nominally melt-free reference  
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8 308 labradorite (Fig. 4). The pmLab material from Dimanov et al. (2000) contains < 2 vol. % of non-wetting  
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10 309 residual plagioclase melt plus ca 1 vol. % wetting incongruent partial melt (with 80 wt.% silica) resulting  
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13 310 from localized grain boundary pre-melting at hyper solidus conditions. It is substantially weaker than the  
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15 311 reference labradorite. Conversely, the SilLab1 and SilLab5 materials, containing wetting partial melts  
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17 312 with varying compositions (ca 85 and 95 wt. % silica, respectively) exhibit unexpected behaviours. Whilst  
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20 313 SilLab1's strength compares to that of Lab material, SilLab5 is substantially stronger, although it contains  
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22 314 the highest melt proportion. It is noteworthy that the latter material also exhibits the highest activation  
23  
24 315 energy.

26 316 The presence of small amounts of non-wetting partial melt do not strongly influence the diffusion  
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28  
29 317 controlled creep laws of labradorite aggregates. In contrast, the presence of small amounts of wetting  
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31 318 partial melt have strong but contrasting rheological impacts, which may be either strengthening or  
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33 319 weakening. The differences in the rheological effects of the different wetting melts relates to their  
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36 320 compositions. Increasing silica content translates in progressive strengthening and increase of activation  
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38 321 energy. These observations are discussed in the following at the light of the effects of silica content on  
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40 322 polymerization, viscosity and diffusion coefficients of silicate melts.

### 43 323

## 45 324 3.2. Microstructures

47 325 Observations of *post mortem* microstructures by SEM did not show noticeable grain growth for  
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49 326 both crept and statically annealed specimens (Tab.2, electronic supplement). Deformation did not result  
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52 327 in shape preferred orientation. SEM and TEM observations and EDX analysis did not show obvious  
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54 328 evolution of melt proportion (within 1 vol. %), topology, or chemistry (within 2 wt. %) during creep. For

1 329 Lab samples the small amount of residual glass was never found along grain boundaries. There was no  
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4 330 indication for dynamic wetting of grain boundaries as observed sometimes in olivine-basalt assemblages  
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6 331 (Jin et al., 1994; Hier-Majumder et al., 2004). For SilLab1 and SilLab5 samples, the initial melt  
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8 332 distribution was preserved during deformation: melt still resided at multiple grain junctions and the thin  
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10 333 melt films were still present at most grain boundaries. Yet, some evidences for local redistribution of melt  
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12 334 along wetted interfaces could be found, in the form of concentric strain contrasts located at wetted grain  
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14 335 boundaries (Fig. 5b,c) Studying creep of glass-ceramic systems, Burger et al. (1997) reported the same  
15  
16 336 kind of strain contrast features, that they called “strain whirls”. According to Burger et al. (1997), the  
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18 337 latter result from residual elastic strain, corresponding to local stress enhancement associated with the  
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20 338 establishment of local dry contacts along previously wetted interfaces. The presence of such localized  
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22 339 solid – solid contacts along wetted grain boundaries may be compared with the theoretical island – channel  
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24 340 structure proposed by Raj (1982). However, we did not observe massive redistribution or complete melt  
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26 341 expulsion out of normally stressed grain boundaries as described for olivine – basalt systems (Jin et al.,  
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28 342 1994). Our observations rather support previous authors promoting the stability of thin amorphous  
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30 343 interfacial phases, in both static and dynamic conditions (Clarke, 1987; Clarke et al., 1993; Hess, 1994;  
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32 344 Drury and FitzGerald, 1996; Wilkinson, 1998).

33 345 Curved free dislocations and dislocation arrays could be occasionally observed. Though,  
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35 346 consistently with Newtonian viscous flow accommodated by diffusional mass transfer mechanisms,  
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37 347 overall dislocation densities,  $\rho$ , remained low ( $\rho \approx 10^7 \text{ cm}^{-2}$ ). Growth twins are very common in  
38  
39 348 undeformed specimens; hence it is difficult to estimate if mechanical twinning could contribute to  
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41 349 deformation. In contrast to polished but undeformed samples which simply experienced static thermal  
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43 350 etching (see Fig. 1), deformed samples present a strong development of surface topography (Fig. 5).  
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45 351 Several previous studies on high temperature uniaxial creep of fine grained materials, such zirconia based  
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47 352 ceramic materials (Clarisse et al., 2000; Duclos et al., 2002) and superplastic alloys (Huang and Langdon,  
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2002), interpreted the development of grain scale topography onto the free surfaces as evidence for grain sliding mechanisms.

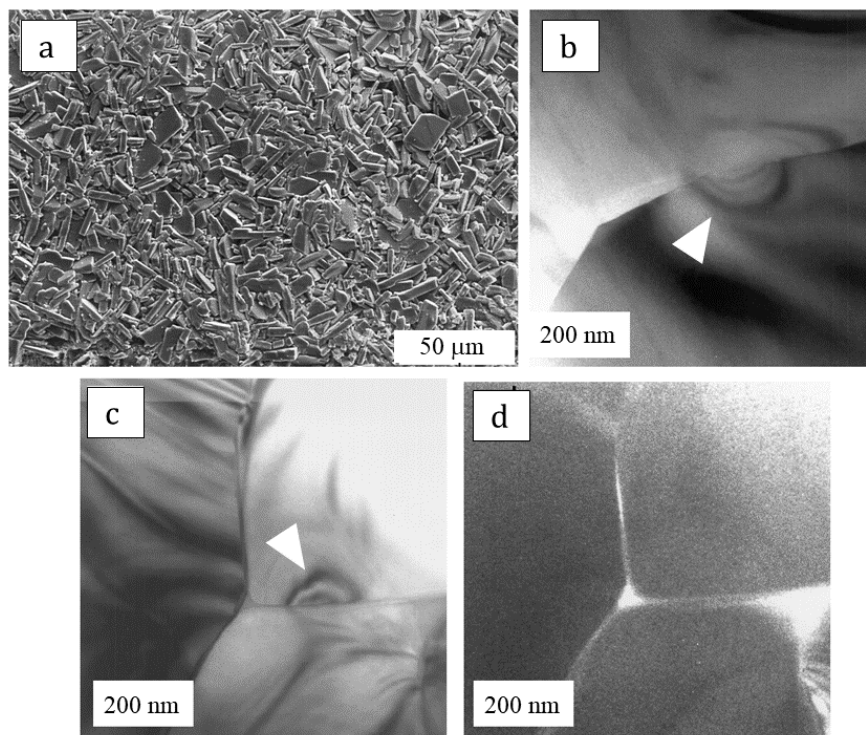


Fig.5: SEM and TEM micrographs of deformed SiLab1 material. a) SEM shows topography development on the sample surface, indicating grain boundary sliding processes. b-c) Bright field images showing dislocation-free grains and whirl shaped strain contrasts (white arrows) next to melt filled pockets (b) or along interfaces (c), which indicate local residual stress related to “dry” contact area (see text). d) Dark field image corresponding to c) showing the presence of amorphous material at interfaces.

#### 4. Discussion

From a general point of view, the mechanical data indicate two dominant creep mechanisms (Figure 4), and thus show show phenomenological similarities with previously reported data for fine grained plagioclase, clinopyroxene and plagioclase-clinopyroxene aggregates (Dimanov et al., 1998; 1999; 2003; Rybacki and Dresen, 2000; Bystricky and Mackwell, 2001; Dimanov and Dresen, 2005; Hier-

1369 Majumder et al., 2005), and olivine aggregates (Hirth and Kohlstedt, 1995; Mei and Kohlstedt, 2000; Mei  
 2 et al., 2002). From a point of view focussing on the rheological effect of the molten phases, our results  
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 4370 present however some striking differences with previous works. To date, all rheological studies of  
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 6371 partially molten silicates invariably report melt related weakening, which magnitude depends on melt  
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 8372 proportion, grain scale topology and dominant creep mechanisms. Contrastingly, we show that in the case  
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 10373 of grain boundary diffusion controlled creep, the presence of extremely silica-rich (95 wt. % SiO<sub>2</sub>) wetting  
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 12374 melt results in strengthening. Interestingly, zirconia (Kajihara et al., 1995), silicon nitride and silicon  
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 14375 carbide ceramics, containing thin amorphous silica films along grain boundaries, systematically exhibit  
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 16376 high activation energies and exceptional creep strengths (Meléndez-Martinez and Dominguez-Rodriguez,  
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 18377 2004).  
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 20378

21  
 22379 In the specific case of grain boundary diffusional mass transfer, whether wetting partial melts  
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 24380 result in weakening or strengthening would be determined by the diffusion properties of the molten  
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 26381 phases, which are actually conditioned by their chemical compositions (Zhang et al., 2010). We  
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 28382 investigate the effects of melts containing between 80 and 95 wt. % silica, which compositions are close  
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 30383 to the end member amorphous silica. At comparable melt content, we observe that the higher is the silica  
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 32384 content of the melt and the lower is the strain rates, but the higher is the activation energy.  
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 36386 Diffusivities in silicate melts, and more specifically diffusion rates of network formers as Si, relate  
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 38387 to the polymerization degree of the melt. Si diffusion in polymerized Jadeite melt is slow with high  
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 40388 activation energy, whilst it is much faster with considerably lower activation energy in depolymerized  
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 42389 diopside melt (Shimizu, 1984). The polymerization degree conditions the melt viscosity, and the latter  
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 44390 relates to the effective molecular diffusion through the Eyring equation (Ayring, 1936; Glasstone et al.,  
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 46391 1941; Mungal, 2002; Nascimento and Zanotto, 2006; Huaiwei et al., 2015):  
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$$D_{eff} = \frac{k_B T}{\lambda \eta} \quad , \quad (2)$$

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1 393 where  $\eta$  is the melt viscosity,  $D_{eff}$  is the effective (molecular) diffusivity and  $\lambda$  is the molecular radius.  
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 4 394 The latter is taken as an approximation for the jump distance of the mobile species or the activated  
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 6 395 transition state molecular complexes. For complex compositions the effective diffusion coefficient  
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 8 396 represents the multicomponent (coupled) diffusion of all the constitutive species. The slowest diffusing  
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 10 397 species, which is rate limiting the coupled diffusion process, remains formally unknown.  $D_{eff}$  is however  
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 12 398 usually considered as a proxy for the self-diffusion coefficients of the basic network formers, namely Si  
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 14 399 and O (Huaiwei et al. 2015). Experimental investigations show that Si and O have the lowest diffusion  
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 16 400 rates in dry silicate melts, Si being always the slowest (Zhang et al., 2010). Si is also the slowest diffusing  
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 18 401 species in silicate minerals (Béjina and Jaoul, 1996; 1997). The Eyring equation (2) is especially  
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 20 402 applicable to anhydrous and polymerized melts (Shimizu and Kushiro, 1984; Tinker et al., 2004). For O  
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 22 403 it is convenient at silica contents < 65 wt.%, while for Si it is suited to any composition (Zhang et al.,  
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 24 404 2010).  
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29 405 Amorphous silica presents a complete tri-dimensional network of  $\text{SiO}_4$  tetrahedrons. The major  
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 31 406 difference with quartz is that its structure is very distorted. Flow of amorphous silica involves transition  
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 33 407 state molecular complexes within the distorted network of tetrahedrons, which activation involves braking  
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 35 408 of Si-O bonds. Ojovan and Lee (2004) reported that the formation and the migration enthalpies for such  
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 37 409 defects to be of 197 and 515 kJ/mol, respectively. The activation energy for silica flow is therefore the  
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 39 410 summation of the latter values, which is in astonishing agreement with the value we report for creep in  
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 41 411 SilLab5 samples (673 kJ/mol). Silicon is the primary network former species in silicate melts. Si is  
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 43 412 therefore most likely the rate limiting species for creep of SilLab5 samples, controlled by diffusion through  
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 45 413 the silica-rich melt films present at the grain boundaries. However, the investigated melts show varying  
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 47 414 compositions, with silica contents ranging between 80 and 95 wt.%  $\text{SiO}_2$ . It is therefore necessary to  
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 49 415 evaluate their respective diffusion properties, in order to provide quantitative kinetic analysis of the  
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 51 416 corresponding creep rates. In the following, based on models we will evaluate the viscosities of the silicate  
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1417 melts present in our samples, from which we will derive the diffusivities of the slowest (rate controlling)  
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4418 species.

#### 8420 **4.1. Viscosities of the silicate melts**

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11421 The viscosity  $\eta$  of a silicate melt depends on the degree of polymerisation of its structural units  
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13422 (Mysen, 1983), which correspond to the network formers species in tetrahedral coordination (in particular  
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15423  $\text{Si}^{4+}$  and  $\text{Al}^{3+}$ , but also  $\text{Ti}^{4+}$ ,  $\text{Fe}^{3+}$ ...). The higher is the polymerization degree and the higher is the  
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17424 concentration of network formers. In opposition, bivalent metals, alkaline and earth alkaline cations  
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20425 represent network modifiers, which decrease the polymerization state, because they relate to non-bridging  
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22426 oxygen within the network of structural tetrahedrons.  
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25427 Based on a large experimental data sets Giordano et al. (2006, 2008) developed a  
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27428 phenomenological model, which allows to calculate the viscosity of anhydrous silicate melts for  
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29429 temperatures between 700°C and 2000°C, on the basis of their content of network modifiers. The silica  
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31430 richest rhyolitic melt used to derive the phenomenological fitting parameters contained 80 wt. % silica.  
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33431 However, the authors suggested that their model might be applicable for melts containing up to 90 wt. %  
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36432 silica, hence covering the range of compositions for the partial melts of pmLab samples ( $\approx 80$  wt. %  $\text{SiO}_2$ )  
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38433 and SilLab1 samples ( $\approx 85$  wt. %  $\text{SiO}_2$ ). Lacking any alternative model, we extrapolated the model  
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40434 towards more siliceous compositions to estimate the viscosity for the melt present in our SilLab5 ( $\approx 95$   
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43435 wt. %  $\text{SiO}_2$ ) samples. Consistently, the calculated viscosities are higher, intermediate and lower for melts  
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45436 present respectively in SilLab5, SilLab1 and pmLab samples, whilst the concerned viscosity range lies  
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47437 between these of pure amorphous silica and albite melt (see appendix A, Fig. A1).  
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50438 The consistency of the viscosities of the melts present in our samples, calculated with the model  
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52439 of Giordano et al. (2008), may be tested by making a comparison with experimental data as a function of  
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54440 their number of non-bridging oxygen per structural tetrahedron, NBO/T. The latter represents the relative  
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1441 amounts of network formers and network modifiers, and thus the polymerization degree of a silicate melt.  
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4442 The impurities were neglected and only the major cations (Si, Al, Na, Ca) were taken in account. Al  
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6443 content does not exceed the sum of the charge weighted contents of Na and Ca ( $Al < Na + 2Ca$ ), and  
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8444 hence Al was entirely assigned to tetrahedral coordination. Following Mysen (1983, 1988; 2003) we  
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10445 calculated  $NBO/T \approx 0.01, 0.02$  and  $0.06$  for the melts present in SilLab5, SilLab1 and pmLab materials  
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12  
13446 respectively. The calculated viscosities line up consistently with the experimental ones (see appendix, A,  
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15447 Fig. A1), which indicates that the extrapolation of the model of Giordano et al. (2008) necessary to  
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17448 SilLab5 is reasonable.  
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## 21 22450 **4.2. Diffusion coefficients in melts and at grain boundaries**

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24451 Ion diffusion properties in aluminosilicate melts correlate with their NBO/T and hence correlate  
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26452 inversely with their viscosities (Hofmann, 1980; Shimizu and Kushiro, 1991; Mysen, 1988, Liang et al.,  
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28  
29453 1996). In the temperature range of interest the diffusion rates of network formers Si and Al in silicate  
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31454 melts decrease over many orders of magnitude with increasing silica. This is shown in Figure 6 (and  
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33455 appendix B, Fig. B1), which is a compilation of self-diffusion and inter-diffusion coefficients in silicates  
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36456 materials, considering: i) volume diffusion in single crystals; ii) grain boundary diffusion in polycrystals;  
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38457 iii) diffusion in melts. We restricted to dry systems and to the slowest diffusing network formers (Al, Si  
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40458 and O), which are susceptible to control the kinetics of diffusion controlled creep. In general, diffusion  
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42459 rates of Al, Si and O are way faster at grain boundaries of silicate aggregates than in their single crystal  
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4560 counterparts (Fig. 6 and appendix B, Fig. B1).  
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47461 Newtonian flow of polycrystalline materials involves bulk diffusion, which operates both in  
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50462 volume and at grain- or interphase boundaries, owing to their specific crystallographic structural defects.  
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52463 The slowest volume diffusion rates relate to point defects (vacancies, interstitials, substitutional impurities  
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54464 and their associations). Alternatively, faster volume diffusion may operate through shortcuts provided by  
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1 465 extended linear defects (dislocation and dislocation arrays) and planar defects (low angle– or sub–grain  
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4 466 boundaries), Grain boundary diffusion rates along high angle grain boundaries are usually much faster  
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6 467 than volume diffusion, because grain boundaries are themselves two dimensional crystallographic defects,  
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8 468 presenting the largest crystallographic deviations from bulk crystal structure. Volume diffusion  
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10 469 coefficients in plagioclases are several orders of magnitude slower than grain boundary diffusion  
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12 470 coefficients in similar materials. As far as concerned with diffusion controlled mass transfer and creep,  
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15 471 strain rates are controlled by the slowest diffusing species diffusing through the fastest pathway.  
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17 472 Accordingly, the creep rates of nominally crystalline plagioclase would be controlled by grain boundary  
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19 473 diffusion rates. Consistently, the grain size exponent we derived from creep of nominally melt-free  
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21 474 labradorite indicate grain boundary diffusion control. The creep rates of partially molten labradorite are  
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24 475 also likely governed by diffusion along grain boundaries wetted by melt. The presence of melt films  
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26 476 wetting grain boundaries would in principle modify the diffusional mass transfer mechanisms, switching  
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29 477 from grain boundary diffusion to diffusion through the melt. The net result on the mass transfer kinetics  
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31 478 would depend on the ratio of the diffusion coefficient in the melt relative to the grain boundary diffusion  
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33 479 coefficient. This is to say the effective diffusivities would be strongly conditioned by the melt  
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35  
36 480 composition. Indeed, diffusion rates in melts are very fast in basalts, intermediate in rhyolitic melts and  
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38 481 substantially slow in plagioclase melts (Fig. 6). Noteworthy, the extremely low self-diffusion coefficient  
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40 482 of Si in pure amorphous silica is nearly comparable with that in pure single crystal quartz, which reflects  
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42 483 the similarity of the polymerized tri-dimensional network of SiO<sub>4</sub> tetrahedrons in both materials.  
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45 484 The calculated viscosities of the melts wetting grain boundaries in pmLab, SilLab1 and SilLab5  
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47 485 samples (see appendix A, Fig. A1) can be interpreted in terms of effective diffusivities,  $D_{eff}$ , based on  
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49 486 equation (2) and taking  $\lambda = 10^{-10}$  m as a first order approximation. The results are convincingly consistent  
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52 487 when compared with appropriate experimental data (Fig. 6 and appendix B, Fig.B1): the calculated  
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54 488 effective diffusion coefficients are intermediate between volume diffusion and grain boundary diffusion  
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in silicates. Alternatively, if we use for equation (2)  $\lambda \approx 0.3$  nm, which is the effective radius for oxygen (Shannon, 1976), the effective diffusion coefficients would be lower with about half an order of magnitude. It is noteworthy that grain boundary diffusion rates in plagioclase were estimated from the kinetics of reaction rim growth in wet conditions (Liu et al., 1997; Yund, 1997; Yund and Farver, 1999; Milke et al., 2001; Abart et al., 2004). However, direct comparison cannot be done with our data, corresponding to dry conditions. Alternatively, the existing grain boundary diffusion data for other dry silicates, oxides and aluminosilicates (dotted lines) match reasonably with the slowest diffusivities in the most polymerized aluminosilicate melts (rhyolite, albite).

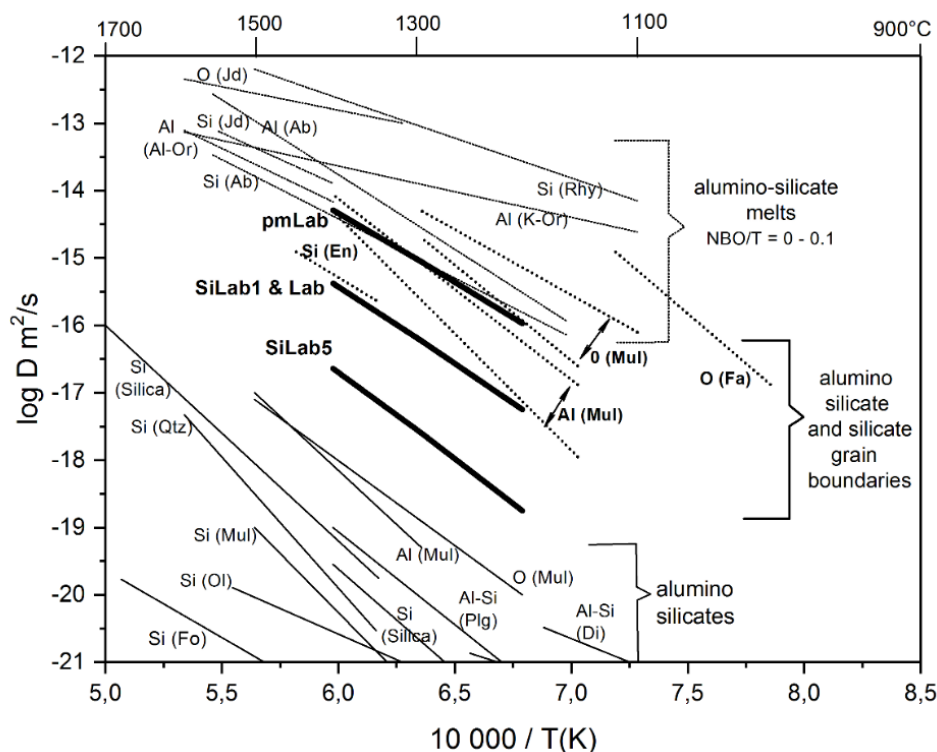


Fig. 6: Arrhenius diagram of volume and grain boundary diffusion of network formers (Si, Al and O) in silicates and oxides and in silicate melts (in dry or unsaturated conditions). The data are recalculated for a pressure of 0.1 MPa (when activation volumes were available). Bas is basalt, Rhy is rhyolitic melt, Di and Jd refer to diopside and jadeite melts, Ab and KAS are albite and potassium-aluminosilicate melts. Diffusivity of Si in silica falls within the lower field for volume diffusion. Diffusivities of O in silica fall in-between the two fields. Grain boundary diffusivities (recalculated for a grain boundary width of  $\sim 3$  nm) are represented by heavy dot lines. The larger vertical brackets indicate the type of diffusivity. For diffusion in

1504 melt the smaller brackets indicate the compositional ranges and the corresponding NBOT numbers. The diffusivities calculated  
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3505 for our melt compositions (see text) are reported as heavy lines and compared to existing grain boundary diffusivities (light  
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5506 dotted lines) in aluminosilicates and silicates (Mulite: Mul, Enstatite: En, Fayalite: Fa) and calculated diffusivities for the  
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7507 present highly polymerized aluminosilicate melts (pmLab, SilLab1 and SilLab5). It appears that grain boundary diffusivities  
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9508 are in general comparable to the lowest diffusivities in alumino-silicate melts, as do the diffusivities for pmLab and SilLab1  
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1509 melts. The diffusivity for SilLab5 melt shows however substantially lower, in-between the lowest diffusivities in alumina-  
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1510 silicate melts and the highest diffusivities in alumina-silicate single crystals. The references for the diffusion data are reported  
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1511 in appendix B, Figure B1.

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1913 The calculated  $D_{eff}$  for the albitic-like melt in pmLab samples (containing ca 80 wt. % silica)  
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2514 matches nearly perfectly the measured Si diffusivity in albite melt (ca 70 wt. % silica), which supports  
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2315 our approach to determine diffusivity from viscosity estimates and the Eyring equation. Consistent with  
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2616 the dependence of Si self-diffusion on silica content in silicate melts (Zhang et al., 2010), the calculated  
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2817 effective diffusivities decrease monotonously from pmLab-melt to SilLab1-melt, and to SilLab5-melt.  
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3018 The faster  $D_{eff}$  for pmLab melt exceeds by about an order of magnitude grain boundary diffusivities in  
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3319 silicates and oxides (enstatite, alumina and mulite), which is fully consistent with the observation that the  
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3520 strain rates for pmLab samples are about an order of magnitude faster than those for nominally melt-free  
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3721 Lab and Lab60 materials. Intermediate  $D_{eff}$  for SilLab1 melt mimics grain boundary diffusion rates for  
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4022 dry silicates, oxides and aluminosilicates, consistent with strain rates for SilLab1 samples comparable to  
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4223 those for nominally melt-free Lab and Lab60 materials. Conversely,  $D_{eff}$  for SilLab5 type of melt (95 wt.  
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4424 % silica) is much lower than the self-diffusion coefficients of Si, Al and O in any of the usually  
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4725 investigated silicate melts (with < 80 wt.% silica).  $D_{eff}$  decreases by about two and half orders of  
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4926 magnitude between pmLab melt and SilLab5 melt. Figure 6 also shows that  $D_{eff}$  for SilLab5 melt is  
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5127 substantially lower than any of the reported grain boundary diffusion coefficients for Si, Al and O. From  
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5428 the perspective of grain boundary diffusion creep, these observations are consistent with the finding that  
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5629 strain rates for Sillab5 are considerably lower than those for nominally melt-free (Lab and Lab60) and  
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1 530 for SilLab1 materials. In summary, on the one hand our findings suggest that thin melt films of  
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4 531 polymerized wetting melt (ca. 85 wt.% silica) do not noticeably affect grain boundary diffusion  
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6 532 coefficients with respect to crystalline grain boundaries. On the other hand, albitic-like wetting melt (ca  
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8 533 80 wt.% silica) considerably enhance the effective grain boundary diffusion coefficients. Conversely, the  
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10 534 latter are strongly decreased by the presence of extremely polymerized wetting melt (ca 95 wt.% silica).  
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### 14 15 536 **4.3 Implications**

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17 537 Extremely silica-rich melts are not very common, but may be found in a few geodynamic contexts.  
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19 538 Hydrothermal weathering and preferential leaching processes of silicate minerals may result in the  
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22 539 formation of small amounts of siliceous amorphous phases (Eggleton and Smith, 1983; Ruiz-Agudo et  
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24 540 al., 2012; Hellmann et al, 2013; Daval et al., 2018; Wild et al., 2019). Some unusually quartz-rich granites  
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26 541 originate from very silica-enriched melts (Chen et al., 2021), whilst some crustal xenoliths exhibit silica-  
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29 542 rich melt inclusions (Frezzotti et al. 2004). Low degree of melting of some mantle xenoliths may result in  
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32 543 the generation of very silica-rich amorphous phases, which are either preserved as volume inclusions  
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34 544 (Schiano et al., 1995), or as glassy grain boundary films (Wirth, 1996; Franz and Wirth, 1997).  
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36 545 Conversely, experiment approaches report low degree of localised melting in relation to stress state or  
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38 546 chemical environment. Samae et al. (2021) report on stress related grain boundary amorphization. The  
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40 547 evolution of point defect populations in iron-bearing diopside may result in lattice precipitation of a few  
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43 548 vol. % of nearly pure silica droplets; a process called early partial melting (Raterron and Jaoul, 1991;  
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45 549 Ingrin et al., 1991; Doukhan et al., 1993, Raterron et al., 1995). From a more general perspective on  
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47 550 partially molten silicates our work demonstrates that melt composition may have strong influences on  
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49 551 mass transfer kinetics, and hence on transport properties involved in creep, but also in electrical  
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52 552 conductivity. Depending on the different geodynamic contexts resulting in low degree of melting, the  
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54 553 mechanical behaviour of the resulting partially molten rocks must also be considered from the point of  
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1 554 view of melt chemistry. However, our work focussed exclusively on dry systems. The role of aqueous  
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4 555 species in depolymerisation of silicate melts and enhancement of diffusion coefficients is well-known  
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6 556 (Brady, 1995, Zhang et al., 2010). Therefore, complementary work would be necessary to investigate the  
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8 557 rheological effects of melt chemistry in water bearing silicate systems.  
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## 10 11 558 **5. Conclusions** 12 559

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16 561 We studied the high-temperature rheology of nominally melt-free and partially molten plagioclase  
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18 562 aggregates by uniaxial creep at dry conditions. The synthetic fine grained materials (ca 10  $\mu\text{m}$  grain size)  
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21 563 contain less than 6 vol. % silica-rich melt, of which less than 1 vol. % resides as thin films (< 10 nm) at  
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23 564 numerous grain boundaries. The mechanical data show dominant linear viscous rheology. The  
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25 565 microstructures and the strong grain size sensitivity (grain size exponent  $m = -2.7$ ) indicate grain boundary  
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27 566 sliding controlled by grain boundary diffusional mass transfer. We specifically focussed on the effect of  
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29 567 the composition of the wetting melt on the latter controlling mechanisms. Varying the silica content of  
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31 568 the wetting melts between 80 and 95 wt. % silica, we demonstrated a strong impact of melt chemistry on  
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33 569 the diffusion controlled Newtonian rheology. Samples containing melts with ca 80, 85 and 95 wt. % silica  
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35 570 were respectively weaker, comparable, and substantially stronger than nominally melt-free samples. The  
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37 571 activation energy for the stronger material was also considerably higher than for the other materials. Based  
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39 572 on existing models, we estimated the viscosities of the silica-rich melts and the effective diffusivities for  
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41 573 their slowest constitutive species. The calculated values are consistent with experimental viscosity and  
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43 574 diffusion data. Regarding the progressively decreasing effective diffusivity with increasing silica content  
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45 575 of the silica-rich melts, we interpret the relative weakening or strengthening effects for our partially  
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47 576 molten samples as respectively representative of enhanced or hindered diffusion properties through the  
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49 577 thin melt films wetting grain boundaries.  
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## Data Availability

The experimental data are available.

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## Appendix A

The viscosities of the wetting melts present in samples pmLab from Dimanov et al. (2000) and in samples SilLab1 and SilLab5 (present work) are calculated based on the model of Giordano et al. (2008). The results are shown in Figure A1, where calculated and experimental viscosities are reported as functions of temperature and the number of non-bridging oxygen per tetrahedron, NBO/T (Mysen, 2004).

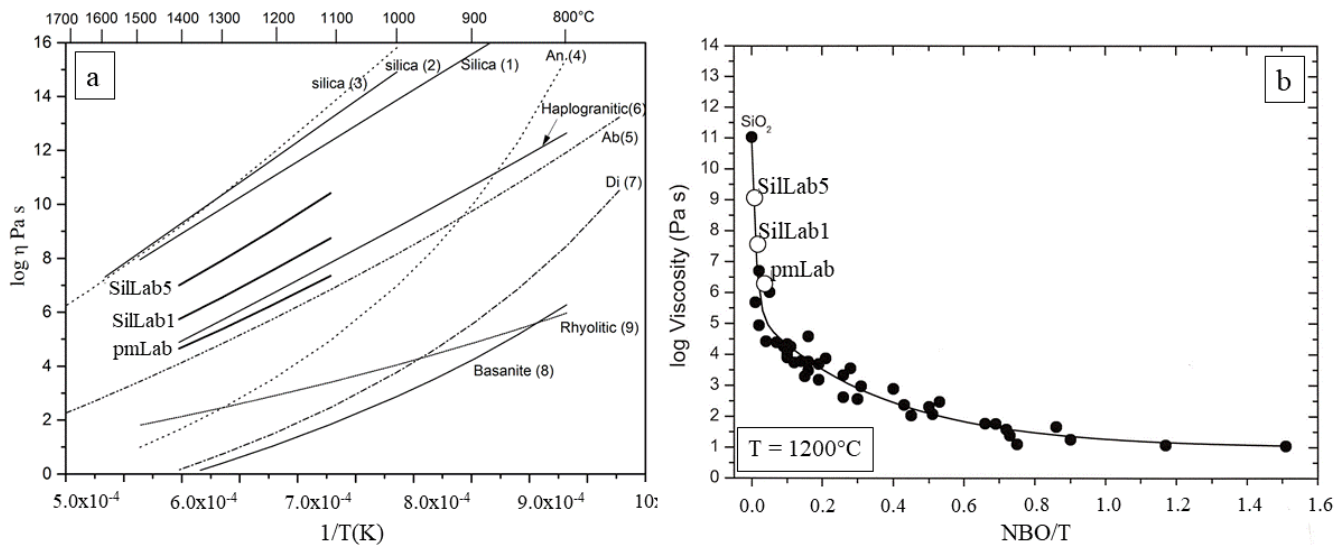


Fig. A1: Comparison between calculated or measured viscosities of typical silicate melts and viscosities calculated for the compositions of the melts present in our materials. a) Arrhenius plot of viscosities of typical silicate melts (not exhaustive, but covering the whole range of viscosities from basaltic composition to vitreous silica) and those of the melts present in our samples (SilLab1 and SilLab5) and in the partially molten samples (pmLab) of Dimanov et al. (2000), computed with the model of Giordano et al. (2008). Data for silica are: (1) from Giordano et al., (2005), (2): from Nascimento and Zanotto, (2006), Silica (3): from Ojovan and Lee (2004) and from experimental data of Urbain et al., 1982 and Hetherington et al (1964). Anorthite (An(4)) and Diopside (Di(7)): from Russel and Giordano (2005), Haplogranitic(6), Albite (Ab(5)) and Rhyolitic(9): from Giordano et al. (2006), Basanite(8): from Giordano et al. (2006). b) Comparison of the experimental viscosities used by Giordano et al. (2008) to set the parameters of their model and the viscosities computed with the latter for compositions of the melts present in our materials as a function of NBO/T (number of non-bridging oxygen per tetrahedron). The solid line is only an eye guiding fit.

**Appendix B**

The effective molecular diffusion coefficients of the wetting melts present in samples pmLab from Dimanov et al. (2000) and in samples SiLab1 and SiLab5 (present work) are calculated based on the Eyring equation (see text, equation (2)) and the corresponding viscosities shown in Figure A1. The results are shown in Figure B1.

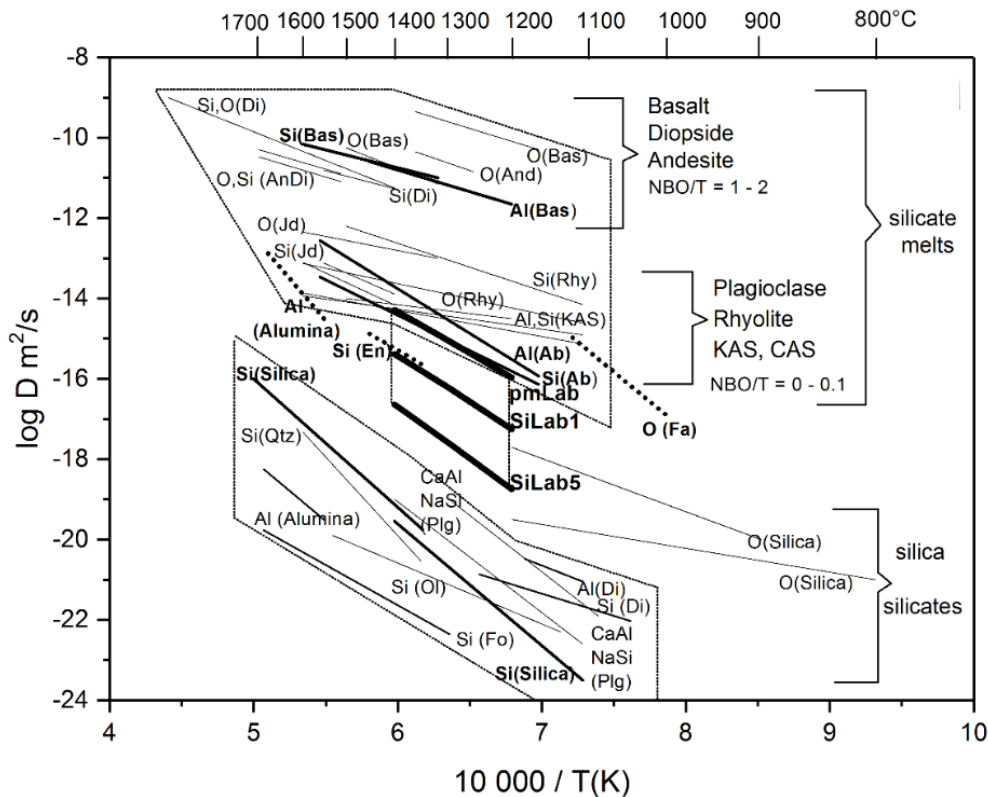


Fig. B1: Arrhenius diagram of volume and grain boundary diffusion of network formers (Si, Al and O) in silicates and oxides and in silicate melts (in dry or unsaturated conditions). The dot line delimited upper and lower fields represent the data for Si, Al and O diffusion in silicate melts and silicate single crystals, respectively. The data are recalculated for a pressure of 0.1 MPa (when activation volumes were available). Bas is basalt, Rhy is rhyolitic melt, Di and Jd refer to diopside and jadeite melts, Ab and KAS are albite and potassium-aluminosilicate melts. Diffusivity of Si in silica falls within the lower field for volume diffusion. Diffusivities of O in silica fall in-between the two fields. Grain boundary diffusivities (recalculated for a grain boundary width of ~ 3 nm) are represented by heavy dot lines. The larger vertical brackets indicate the type of diffusivity. For diffusion in melt the smaller brackets indicate the compositional ranges and the corresponding NBO/T numbers. The

1 35 diffusivities calculated for our melt compositions (see text) are reported as heavy lines. Diffusion data for melts are from Baker  
2  
3 36 (1990, 1992, 1995), Shimizu and Kushiro (1984, 1991), Canil and Muehlenbach (1990), Wendlandt (1991), Chakraborty et  
4  
5 37 al. (1995), Kress and Ghiorso (1995), Lesher et al. (1996), Harmer and Brook (1980), Brebec et al. (1980), Reid et al. (2001).  
6  
7 38 Data for grain boundary diffusion in silicates, aluminosilicates and alumina are from Rubie (1986), Fislser and Mackwell  
8  
9 39 (1994), Fislser et al. (1997), Fielitz et al. (2004, 2007, 2016). Data for volume diffusion in silicates, aluminosilicates and  
10  
11 40 alumina are from Bejina and Jaoul (1996), Houlier et al. (1990), Jaoul et al. (1981, 1991), Grove et al. (1984), Yund (1986),  
12  
13 41 Le Gall et al. (1994), Fielitz et al. (2007).

Table 2. Mechanical data.

Sample	$T$ (°C)	t (h)	$\sigma_{diff}$ (MPa)	$d\varepsilon/dt$ (1/s)	$d$ ( $\mu\text{m}$ )*
lab-01	1150	31	5.5	$7.5 \times 10^{-8}$	9.8
	1150	13	10.3	$1.3 \times 10^{-7}$	-
	1150	8.4	15.3	$1.7 \times 10^{-7}$	-
	1150	2.4	20.5	$2.8 \times 10^{-7}$	-
	1150	1.5	30.5	$6.4 \times 10^{-7}$	-
	1200	0.8	21.3	$1.8 \times 10^{-6}$	-
	1200	0.5	26.2	$3.0 \times 10^{-6}$	-
	1200	0.5	31.0	$2.8 \times 10^{-6}$	-
	1150	25	4.8	$8.4 \times 10^{-8}$	-
	1150	3.5	20.0	$2.7 \times 10^{-7}$	9.9
lab-02	1100	17.2	26.2	$1.3 \times 10^{-7}$	9.8
	1100	23	19.4	$8.3 \times 10^{-8}$	-
	1100	19.4	22.8	$7.8 \times 10^{-8}$	-
	1100	15.2	34.1	$1.3 \times 10^{-7}$	-
	1100	8.4	45.4	$2.2 \times 10^{-7}$	9.6
lab-03	1100	66	10.1	$4.0 \times 10^{-8}$	9.8
	1100	21	15.0	$5.2 \times 10^{-8}$	-
	1100	24.7	19.8	$6.9 \times 10^{-8}$	-
	1100	24	29.7	$8.6 \times 10^{-8}$	-
	1100	8.5	39.6	$1.2 \times 10^{-7}$	-
	1200	4.8	5.2	$1.9 \times 10^{-7}$	-
	1200	1.6	15.0	$6.2 \times 10^{-7}$	-
	1200	2.2	10.3	$3.9 \times 10^{-7}$	-
	1200	1	20.3	$1.2 \times 10^{-7}$	-
	1200	0.5	25.1	$2.1 \times 10^{-7}$	-
	1200	0.2	30.0	$4.4 \times 10^{-7}$	10
lab-04**	1200	72	-	-	9.8



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2						
3	lab-05	1150	20	4.9	$6.0 \times 10^{-8}$	9.8
4						
5		1150	24	9.5	$1.1 \times 10^{-7}$	-
6						
7		1150	6.5	14.1	$1.7 \times 10^{-7}$	-
8						
9		1150	2.0	28.6	$5.0 \times 10^{-7}$	-
10						
11		1150	18	6.2	$5.6 \times 10^{-8}$	-
12						
13		1150	3.0	23.7	$3.5 \times 10^{-7}$	-
14						
15		1150	1.0	38.1	$1.1 \times 10^{-6}$	-
16						
17		1225	2.5	5.0	$5.6 \times 10^{-7}$	-
18						
19		1225	1.5	9.8	$9.5 \times 10^{-7}$	-
20						
21		1225	0.7	14.3	$1.7 \times 10^{-6}$	-
22						
23		1225	0.4	19.2	$3.8 \times 10^{-6}$	-
24						
25		1225	0.5	13.8	$1.1 \times 10^{-6}$	10
26						
27	SiLab1-01	1150	21	7.1	$1.4 \times 10^{-7}$	9.3
28						
29		1150	3.7	14.2	$3.0 \times 10^{-7}$	-
30						
31		1150	3.2	21.3	$4.7 \times 10^{-7}$	-
32						
33		1150	1.5	28.6	$8.6 \times 10^{-7}$	-
34						
35		1150	15	9.2	$1.5 \times 10^{-7}$	-
36						
37		1150	1.8	28.8	$8.3 \times 10^{-7}$	-
38						
39		1150	1.8	36.0	$1.4 \times 10^{-6}$	9.2
40						
41	silab1-02	1200	4	4.0	$1.8 \times 10^{-7}$	9.3
42						
43		1200	16	2.7	$1.0 \times 10^{-7}$	-
44						
45		1200	2.8	8.7	$3.1 \times 10^{-7}$	-
46						
47		1200	1.1	13.5	$5.4 \times 10^{-7}$	-
48						
49		1200	1.2	15.02	$5.6 \times 10^{-7}$	-
50						
51		1200	1.5	18.2	$7.1 \times 10^{-7}$	-
52						
53		1200	1.0	22.9	$1.0 \times 10^{-6}$	9
54						
55	silab1-03	1100	30	9.7	$2.5 \times 10^{-8}$	9.3
56						
57		1100	24.2	14.1	$3.9 \times 10^{-8}$	-
58						
59		1100	14	28.4	$5.5 \times 10^{-8}$	-
60						

	1100	11.4	13.5	$7.6 \times 10^{-8}$	-
	1225	2.6	2.7	$4.3 \times 10^{-7}$	-
	1225	1.3	5.7	$6.9 \times 10^{-7}$	-
	1225	0.8	9.1	$1.1 \times 10^{-6}$	-
	1225	1.5	13.8	$1.7 \times 10^{-6}$	-
	1225	1.0	18.6	$2.6 \times 10^{-6}$	9.4
silab1-04	1200	3.1	5.3	$2.8 \times 10^{-7}$	9.3
	1200	1.9	19.5	$6.2 \times 10^{-7}$	-
	1200	1.1	28.8	$1.9 \times 10^{-6}$	-
	1100	12	28.84	$7.4 \times 10^{-8}$	-
	1100	10	43.7	$8.3 \times 10^{-8}$	-
	1150	14.1	5.01	$5.9 \times 10^{-8}$	-
	1150	2.8	19.0	$3.7 \times 10^{-7}$	-
	1150	1.4	24.0	$5.3 \times 10^{-7}$	-
	1225	1.2	5.4	$6.9 \times 10^{-7}$	-
	1225	0.8	14.8	$2.8 \times 10^{-6}$	-
	1225	0.1	28.8	$1.2 \times 10^{-5}$	-
	1225	2.4	4.0	$9.9 \times 10^{-7}$	-
	1225	2.2	5.3	$9.7 \times 10^{-7}$	-
	1225	1.2	10	$2.3 \times 10^{-6}$	-
	1225	0.2	14.5	$6.4 \times 10^{-6}$	9.2
silab5-01	1100	22.4	22.6	$5.3 \times 10^{-9}$	8.6
	1100	28.3	34.1	$1.3 \times 10^{-8}$	-
	1100	36	46.2	$1.5 \times 10^{-8}$	-
	1150	24.2	23.0	$4.8 \times 10^{-8}$	-
	1150	20.5	34.3	$4.3 \times 10^{-8}$	-
	1150	27	47.0	$7.0 \times 10^{-8}$	-
	1200	30.8	5.8	$4.2 \times 10^{-8}$	-
	1200	8.2	16.8	$1.6 \times 10^{-7}$	-

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3		1200	4.7	22.6	$1.5 \times 10^{-7}$	-
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5		1200	2.4	33.9	$2.9 \times 10^{-7}$	-
6						
7		1200	3.2	45.8	$3.9 \times 10^{-7}$	-
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9		1200	7.2	34.12	$2.9 \times 10^{-7}$	8.7
10						
11	silab5-02	1200	23.2	5.00	$5.4 \times 10^{-8}$	8.6
12						
13		1200	17.0	9.8	$1.2 \times 10^{-7}$	-
14						
15		1200	4.1	19.4	$2.0 \times 10^{-7}$	-
16						
17		1200	2.6	29.0	$3.4 \times 10^{-7}$	-
18						
19		1200	2.7	38.6	$5.3 \times 10^{-7}$	-
20						
21		1200	2.5	43.6	$6.1 \times 10^{-7}$	-
22						
23		1225	10.2	5.0	$1.0 \times 10^{-7}$	-
24						
25		1225	3.0	9.8	$2.2 \times 10^{-7}$	-
26						
27		1225	3.2	19.2	$5.2 \times 10^{-7}$	-
28						
29		1225	1.0	28.8	$1.2 \times 10^{-6}$	-
30						
31		1225	0.7	38.3	$2.1 \times 10^{-6}$	8.6
32						
33	silab5-03	1100	163	13.7	$3.6 \times 10^{-9}$	8.6
34						
35		1100	125	26.7	$5.3 \times 10^{-9}$	-
36						
37		1100	55.8	40.0	$6.9 \times 10^{-9}$	-
38						
39		1150	61.6	14.0	$1.8 \times 10^{-8}$	-
40						
41		1150	27	27.2	$3.3 \times 10^{-8}$	-
42						
43		1150	25	40.3	$5.9 \times 10^{-8}$	-
44						
45		1150	20.6	53.8	$5.8 \times 10^{-8}$	8.6
46						
47	silab5-04	1225	19.2	7.8	$2.2 \times 10^{-7}$	8.6
48						
49		1225	4.3	15.0	$4.5 \times 10^{-7}$	-
50						
51		1225	2.5	21.8	$7.5 \times 10^{-7}$	-
52						
53		1225	1.1	28.8	$1.4 \times 10^{-6}$	-
54						
55		1225	0.3	42.9	$3.5 \times 10^{-6}$	8.5
56						
57	silab5-05	1250	3.0	7.0	$7.1 \times 10^{-7}$	8.6
58						
59		1250	1.0	14.0	$1.6 \times 10^{-6}$	-
60						

	1250	0.5	21.0	$2.3 \times 10^{-6}$	-
	1250	0.2	28.3	$5.6 \times 10^{-6}$	8.6

\* grain size  $d$  was measured only for the starting material and at the end of each mechanical test.

\*\* lab-04 sample was only annealed in static conditions.

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