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Geneviève Robert

Bates College, grobert@bates.edu

Rebecca A. Smith

Alan G. Whittington

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Viscosity of melts in the NaAlSiO_4 - KAlSiO_4 - SiO_2 system: configurational entropy modelling

Geneviève Robert^{a,*}, Rebecca A. Smith^{a,**}, Alan G. Whittington^{b,c}

^a*Department of Geology, Bates College, Lewiston, Maine 04240, USA*

^b*Department of Geological Sciences, University of Missouri, Columbia, Missouri 65211, USA*

^c*Department of Geological Sciences, The University of Texas at San Antonio, San Antonio, Texas 78249, USA*

Abstract

We provide new viscosity data in the system SiO_2 -(Na,K) AlSiO_4 , for the nepheline-kalsilite and jadeite-leucite joins. We present a configurational entropy model for the viscosity of melts in the system as a function of $\text{Na}/(\text{Na}+\text{K})$ and $\text{Al}/(\text{Al}+\text{Si})$ ratios. Our modelling indicates that: i) Viscosity data are reproduced well by a non-ideal, symmetrical form of the parameters $S^{\text{conf}}(T_g)$ and B_e , ii) Na-K mixing is the main source of additional entropy in the system based on the limited dependence of $S^{\text{conf}}(T_g)$ and B_e parameters on $\text{Al}/(\text{Al}+\text{Si})$ ratio, iii) A_e likely varies as a function of $\text{Al}/(\text{Al}+\text{Si})$ ratio. Melt fragility in the system increases with increasing $\text{Al}/(\text{Al}+\text{Si})$ ratio and is greater for Na or K end-member melts than mixed melts. The viscosity of nominally fully-polymerized melts in the SiO_2 -(Na,K) AlSiO_4 system can be modelled through chemical mixing, without explicit consideration of the important changes in structure related to changes in $\text{K}/(\text{Na}+\text{K})$ and $\text{Al}/(\text{Al}+\text{Si})$ ratios.

Keywords: viscosity, aluminosilicate melts, alkali mixing, configurational entropy modelling

*Corresponding author

**Present address: Department of Geosciences, University of Massachusetts – Amherst, Amherst, Massachusetts, 01003, USA

Email address: grobert@bates.edu (Geneviève Robert)

1. Introduction

The nepheline-kalsilite-quartz system (Figure 1) contains a number of important compositions for modelling magmatic systems, and offers an ideal analog system for the study of mixed-alkali melts, Na_2O and K_2O being the most important alkali oxides by weight in magmas. Viscosity studies in the system have largely concentrated on the the NaAlO_2 - SiO_2 join and the haplogranite subsystem (Riebling, 1966; Taylor and Rindone, 1970; Sharma et al., 1979; Urbain et al., 1982; Stein and Spera, 1993; Kim and Lee, 1997; Toplis et al., 1997a,b), with obvious applications to granites and rhyolites. Le Losq and Neuville (2013) investigated Na-K mixing along the silica-rich alkali feldspar and $(\text{Na,K})\text{AlSi}_5\text{O}_{12}$ joins and Le Losq et al. (2017) investigated compositions along the $(\text{Na,K})\text{AlSi}_{1.3}\text{O}_{4.7}$ and $(\text{Na,K})\text{AlSiO}_4$ joins. We expand the systematic quantification of the effects of alkali mixing on viscosity to silica-poor aluminosilicate melts, at two different $\text{Al}/(\text{Al}+\text{Si})$ ratios, specifically along the $(\text{Na,K})\text{AlSi}_2\text{O}_6$ and the $(\text{Na,K})\text{AlSiO}_4$ joins (Figure 1). The metaluminous melts along the $\text{NaAlSi}_x\text{O}_{2x+2}$ - $\text{KAlSi}_x\text{O}_{2x+2}$ joins in the nepheline-kalsilite-quartz system have a nominal ratio of non-bridging oxygen to tetrahedrally-coordinated cations (NBO/T) of 0, as each alkali atom charge-balances one tetrahedrally-coordinated aluminum atom, and are nominally fully polymerized. However, the work of Stebbins and Xu (1997) suggests the presence of small amounts of non-bridging oxygen atoms (NBOs) in metaluminous (and therefore nominally fully polymerized) glasses; NBOs in metaluminous glasses are also suggested by the presence of a peraluminous viscosity maximum in Na_2O - Al_2O_3 - SiO_2 melts instead of the expected metaluminous one (Toplis et al., 1997b). Murdoch et al. (1985) demonstrated that smaller, highly-charged cations favor a wider distribution of silicate and aluminate tetrahedra in aluminosilicate melts, that is, they favor more Al-Si disorder in contrast to melts in which the aluminum avoidance principle is strictly observed (Loewenstein, 1954). It could be expected that with increasing Al/Si ratio, more order is required to satisfy the aluminum avoidance principle (Murdoch et al., 1985; Toplis et al., 1997a), in these sodium-potassium aluminosilicate melts.

Le Losq and Neuville (2013) showed the effects of mixing of Na and K (mixed alkali effect; Isard, 1969; Day, 1976) on the viscosity of silica-rich aluminosilicate melts. Indeed, the potassium and sodium end-members on each join in the system have different viscosities, and the intermediate viscosities are not a simple linear function of Na/K ratio, suggesting changes in structure

Figure 1: *Black circles: glass and melt compositions from this study. Grey circles: glass and melt compositions studied by Le Losq and Neuville (2013) and Le Losq et al. (2017). The Si:O ratio is shown for each join on the right side and the Al/Si ratio is shown on the left side. Not shown are samples studied by Toplis et al. (1997a), Stein and Spera (1993), N'dala et al. (1984), Urbain et al. (1982), Riebling (1966) along the NaAlSiO₄-SiO₂ join, and samples studied by N'dala et al. (1984) and Urbain et al. (1982) along the KAlSiO₄-SiO₂ join.*

38 along each join. Le Losq and Neuville (2013) further explored the structural
 39 changes that occur as K substitutes for Na by Raman spectroscopy and more
 40 recently by molecular dynamics simulations (Le Losq et al., 2017). They
 41 suggest, on the basis of their Raman spectroscopy data on silica-rich glasses,
 42 that: i) an increase in K relative to Na results in an increase of three- and
 43 four- membered rings relative to rings with higher members; ii) an increase
 44 in K favors the formation of Si-O-Si-O-Si three-membered rings relative to
 45 Si-O-Al-O-Si rings; iii) two populations of six-membered rings distinguished
 46 by their intertetrahedral (T-O-T) angles may coexist, one of which is sug-
 47 gested to have an evolving T-O-T angle with increasing K whereas the other
 48 remains unchanged by the alkali substitution. They attribute the viscosity

49 patterns they observed in the silica-rich melts to these structural changes.
50 They explain the small observed viscosity changes at high Na/K ratios by a
51 predominance of a Si-Al-Na-O subnetwork where K is easily accommodated.
52 At low Na/K ratio, they explain the large viscosity increases they observed
53 by a predominance of a more ordered Si-Al-K-O subnetwork. They explain
54 the slight viscosity decrease relative to the end-members observed at inter-
55 mediate Na/K ratios by either a pairing of the Na^+ and K^+ ions or by a
56 random distribution of the two subnetworks at the medium scale.

57 Le Losq et al. (2017) proposed that the clusters of alkalis (K vs. Na
58 subnetworks) associate with Al-rich regions of the network, violating the Al-
59 avoidance principle, and that this effect is stronger in melts with higher Al/Si
60 ratio. Indeed, their molecular dynamics simulations predict Al-Al to be only
61 $\sim 18\%$ of all inter-tetrahedral distances, consistent with $\sim 13\%$ observed
62 with ^{17}O MAS NMR (Lee and Stebbins, 2009), which is less than the $\sim 25\%$
63 expected from a random distribution of the Al in the network. The mixed
64 alkali effect is more important at higher Al/Si ratios, and percolation in
65 alkali channels, as proposed by Greaves (1985) and Greaves and Ngai (1995)
66 with the Modified Random Network and Compensated Continuous Random
67 Network models for the structure of alkali-rich silicate melts, is only attained
68 in melts with high Al/Si ratios and at high X_K whereas only clusters are
69 found at low Al/Si ratios or low X_K . Le Losq et al. (2017) conclude that
70 increasing the proportion of K over Na dilates tetrahedral rings and cages
71 of the network, which shortens and strengthens the Si-O and Al-O bonds,
72 increasing the viscosity of the melt. This strengthening of both Si-O and
73 Al-O bonds allows for more Al-O-Al bonds to be stable in the network.

74 1.1. The mixed alkali effect and configurational entropy theory

75 The mixed alkali effect refers to a non-negligible deviation from the sim-
76 ple additivity of physical properties, such as viscosity, upon mixing of alkalis
77 (Isard, 1969; Day, 1976). This behavior can be explained in the framework
78 of the configurational entropy theory (Richet, 1984) based on the Adam and
79 Gibbs (1965) theory of melt relaxation, where viscosity is expressed as a func-
80 tion of the configurational entropy of melts. The theory assumes that struc-
81 tural relaxation times are determined by the probability of configurational
82 rearrangements in the melt. The structural relaxation time of a melt (τ) is
83 proportional to the relaxed Newtonian shear viscosity of a melt ($\tau = \frac{\eta}{G_\infty}$),
84 where G_∞ is the unrelaxed elastic shear modulus (typically assumed to be a

constant for silicate melts at $\sim 10^{10}$ Pa; Dingwell and Webb, 1990). Combining the structural relaxation equation with the Adam-Gibbs theory of melt relaxation gives (Richet, 1984):

$$\eta(T) = A_e \exp \left(\frac{B_e}{T S^{conf}(T)} \right) \quad (1)$$

where A_e is a pre-exponential term and high-temperature viscosity limit of the melt, and B_e is a constant proportional to the Gibbs free-energy barriers hindering rearrangements in the melt.

The variation of configurational entropy with temperature is defined as:

$$S^{conf}(T) = S^{conf}(T_g) + \int_{T_g}^T \frac{C_P^{conf}}{T} dT \quad (2)$$

where C_P^{conf} is the difference in heat capacity between the melt and the glass and may be obtained by calorimetry (Richet, 1984; Richet et al., 1986; Neuville and Richet, 1991) and $S^{conf}(T_g)$ is the configurational entropy of the melt at the glass transition. Although $S^{conf}(T_g)$ may only be directly obtained by calorimetry for congruently melting compositions, comprehensive calorimetry and viscosity datasets allow $S^{conf}(T_g)$ to be fitted, just like the A_e and B_e parameters, when Equations 1 and 2 are combined. We refer to fits obtained through configurational entropy modelling as “Adam-Gibbs” or “AG” for short. Other successful viscosity models exist (e.g., Tamman-Vogel-Fulcher, Avramov-Milchev, and MYEGA models; Vogel, 1921; Avramov and Milchev, 1988; Mauro et al., 2009), but the AG model is particularly interesting because it allows for the possibility of using independently measured heat capacity to predict viscosity and, as we use it in this paper, to predict thermodynamic properties of a system using viscosity data.

For viscosity, deviations from linearity upon mixing are most important at temperatures near the glass transition (T_g) and are smaller at superliquidus temperatures. At low temperatures near the glass transition, there is a greater relative contribution of configurational entropy, which includes chemical (mixing) and topological (distribution of bond angles and bond lengths) components, whereas the heat capacity of the liquid is the most important term at higher temperatures (Richet, 1984; Le Losq and Neuville, 2013, Equation 2). Although some studies investigating Ca-Mg cation mixing in silicate melts have found the assumption of ideal mixing to be appropriate

(e.g., Neuville and Richet, 1991), this does not appear to be the case for Na-K aluminosilicate or Mg-K (alumino)silicate melts (e.g., Allwardt and Stebbins, 2004; Le Losq and Neuville, 2013; Le Losq et al., 2017). We propose a configurational entropy model for mixed-alkali aluminosilicate melts of the (Na,K)AlSiO₄-SiO₂ system spanning Al/Si ratios between 0.2 and 1.

2. Experimental methods

2.1. Sample synthesis

We weighed reagent grade oxide (SiO₂, Al₂O₃) and carbonate (Na₂CO₃, K₂CO₃) powders using a Sartorius Cubis analytical balance. We mixed the powders under acetone for ~20 minutes. We transferred the slurry in roughly equal amounts into two separate PtRh crucibles of 120 mL capacity and known weight. Each slurry was ignited and the acetone allowed to burn off. We let the dry mixtures cool in a desiccator. After weighing, we heated the dry mixtures very slowly overnight to 1100 °C in a Nabertherm box furnace to drive off CO₂.

We fused the decarbonated powder mixtures to temperatures between 1600 and 1745 °C and immediately quenched the glass upon reaching temperature. The glass was then completely tapped out of each crucible, combined and mechanically ground with a high density alumina dish and puck in a Spex[®] ShatterBox[®] swing mill. We carefully scraped out the powdered glass out of the dish and put it into a single crucible for melting. For each melting step, we kept the melt at maximum temperature for one to two hours, and quenched it to a glass by pouring it onto a copper tray. We crushed and remelted the glass slabs two or three more times, until we obtained a homogeneous, bubble-free, and crystal-free glass. Melting temperatures varied between 1600 and 1750 °C. For some very viscous melts, we were not able to obtain entirely bubble-free glasses. The estimated bubble fraction for those glasses is in Table 1.

We drilled the glass slabs to produce 5 mm (quarter inch) diameter cylinders roughly one centimeter long. We cut parallel ends for each cylinder using a low-speed wafer saw, and polished the cylinder ends to obtain smooth parallel surfaces.

Table 1: Chemical and physical sample characterization

Sample no. of analyses	Ne ₁₀₀	Ne ₇₅	Ne _{62.5}	Ne ₅₀	Ne _{37.5}	Ne ₂₅	Jd ₁₀₀	Jd ₇₅	Jd _{62.5}	Jd ₅₀	Jd _{37.5}	Jd ₂₅	Jd ₀	Or	K-petalite
(wt.%)	18	17	20	16	30	17	12	11	18	10	25	5	3	6	6
SiO ₂	40.51	39.41	40.33	37.87	39.34	37.28	57.32	55.98	57.12	54.85	55.86	53.12	54.51	63.38	71.56
Al ₂ O ₃	37.12	35.87	35.01	34.84	33.80	34.17	26.14	25.49	24.66	24.79	23.77	24.76	25.04	18.93	17.63
FeO	0.04	0.04	0.04	0.05	0.00	0.06	0.04	0.06	0.01	0.05	0.03	0.05	0.00	0.04	0.15
Na ₂ O	21.61	15.74	13.29	10.11	7.57	4.89	15.24	11.34	9.24	7.27	5.57	3.52	0.07	0.04	0.06
K ₂ O	0.02	8.29	12.26	16.07	19.52	22.62	0.03	5.92	8.68	11.54	14.22	17.12	21.57	15.97	15.37
Total	99.29	99.36	100.92	98.94	100.23	99.02	98.76	98.79	99.72	98.50	99.49	98.57	101.19	98.36	104.77
std (wt.%)															
SiO ₂	0.60	0.55	0.16	0.45	0.29	0.60	0.88	1.19	0.41	0.79	0.59	1.54	0.29	0.41	1.04
Al ₂ O ₃	0.28	0.24	0.35	0.21	0.24	0.22	0.44	0.69	0.28	0.38	0.40	1.23	0.10	0.10	0.06
FeO	0.01	0.02	0.02	0.02	0.00	0.02	0.02	0.02	0.01	0.03	0.02	0.02	0.01	0.02	0.02
Na ₂ O	0.15	0.13	0.09	0.13	0.07	0.11	0.22	0.28	0.09	0.15	0.14	0.14	0.01	0.01	0.01
K ₂ O	0.01	0.09	0.09	0.10	0.11	0.16	0.00	0.07	0.10	0.15	0.12	0.17	0.05	0.16	0.30
mol.%															
SiO ₂	48.61	48.60	49.39	48.27	49.77	48.68	65.50	65.26	66.28	65.41	66.24	64.75	65.60	74.78	77.95
Al ₂ O ₃	26.24	26.06	25.26	26.17	25.20	26.29	17.60	17.51	16.86	17.42	16.61	17.79	17.76	13.16	11.31
Na ₂ O	25.14	18.81	15.77	12.50	9.28	6.19	16.88	12.82	10.39	8.40	6.41	4.16	0.08	0.04	0.06
K ₂ O	0.02	6.52	9.57	13.06	15.75	18.84	0.02	4.40	6.43	8.77	10.75	13.31	16.56	12.02	10.68
Total	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
NBO/T	-0.02	-0.01	0.00	-0.01	0.00	-0.02	-0.01	-0.01	0.00	0.00	0.01	-0.01	-0.02	-0.02	-0.01
Al/(Al+Si) ^a	0.52	0.52	0.51	0.52	0.50	0.52	0.35	0.35	0.34	0.35	0.33	0.35	0.35	0.26	0.22
Na/(Na+K) ^a	1.00	0.74	0.62	0.49	0.37	0.25	1.00	0.74	0.62	0.49	0.37	0.24	0.00	0.00	0.01
gfw ^b	71.55	73.58	74.22	75.73	76.19	77.64	67.78	69.16	69.51	70.53	70.83	72.15	73.17	69.69	68.46
no. of atoms	3.52	3.52	3.51	3.52	3.50	3.53	3.35	3.35	3.34	3.35	3.33	3.36	3.36	3.26	3.23
(kg/m ³)															
ρ_{pre}^c	2487(2)	2489(2)	2492(2)	2487(1)	2488(1)	2481(2)	2411(2)	2412(3)	2411(2)	2409(2)	2408(2)	2396(2)	2333	2288	2333
ρ_{post}^d	2500(2)	2497(2)	2496(2)	2494(2)	2492(1)	2487(2)	2417(1)	2422(3)	2416(1)	2417(2)	2412(1)	2407(2)	2334	n.a.	2337
Bubble fraction	<0.01	<0.01	~0.18	<0.01	<0.05	<0.01	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.01	n.d.	n.d.

All samples are nominally anhydrous and expected to contain less than 0.1 wt.% dissolved water (Ryan et al., 2015). This corresponds to an effect on the glass transition temperature of at most 20 K (Davis, 1999).

Measurements of compositions post parallel-plate and concentric cylinder viscosity experiments are provided as supplementary material. Post-experiment analyses overlap with pre-experiment analyses, within the analytical uncertainty of the order of 0.2-0.3 wt.% for Na₂O or K₂O, except in the case where samples crystallized during an experiment and crystals affected the composition of the glass.

^a Cation units.

^b Gram formula weight for one mole of oxides, equivalent to a two oxygen basis in this system.

^c Density of glass before viscometry measurement. Standard deviation on 5 measurements in parentheses.

^d Density of glass after parallel-plate viscometry measurement. Standard deviation on 5 measurements in parentheses.

147 2.2. Sample characterization

148 We measured the composition of the glasses pre- and post-viscometry
 149 experiments with a Cameca SX-50 electron microprobe at the University of
 150 Massachusetts – Amherst (Table 1). We tried to mitigate Na migration by
 151 using a low beam current density, that is, by using a beam diameter of 20 μm
 152 and a beam current of 10 nA at an accelerating voltage of 15 kV. Counting
 153 for Na was done on a very high count rate monochromator (Cameca’s PC0
 154 detector, otherwise known as the ovonyx W-Si multilayer with 2d-spacing of
 155 60Å), which allows short count times with good precision. This allowed us
 156 to measure Na with 6 second counting on peak to minimize any inaccura-
 157 cies arising from both diffusion (migration of Na toward space charge) and
 158 surface carbon buildup. Matrix corrections are done via Cameca’s PAP pro-
 159 gram which is integrated into the quantitative analysis package within the
 160 SXRayN50 automation.

161 We measured the density of the glass cores before and after parallel-plate
 162 viscometry experiments (Table 1). We used the Archimedeian method, with
 163 anhydrous ethanol as the immersion liquid. Each density reported is an
 164 average of five measurements. We estimated bubble or crystal fraction (in
 165 the case where samples crystallized during experiments) by transmitted light
 166 microscopy. We did not observe crystals in the starting glasses.

167 2.3. Viscometry

168 We measured the viscosity of melts at low temperatures near the glass
 169 transition by parallel-plate viscometry ($\eta = 10^{8.5}-10^{13}$ Pa s) at Bates College.
 170 The instrument recovers viscosity data within 0.1 log units as tested by mea-
 171 suring National Institute of Standards and Technology SRM 717a glass over
 172 the full range of viscosities testable. Samples generally underwent less than
 173 10% shortening during the viscosity measurements and the cores maintained
 174 a cylindrical shape. We therefore calculated viscosity from the shortening
 175 rate of the samples at constant load using the perfect slip condition (Ding-
 176 well et al., 1993), using the following equation:

$$\eta_s = \frac{mgh^2}{3V \frac{dh}{dt}} \quad (3)$$

177 where m is the mass applied to the sample (kg), g gravity (ms^{-2}), h the
 178 sample height (m), V the volume of the sample (m^3), and t is time (s).

179 We report the viscometric glass transition as the temperature at which the
 180 viscosity is 10^{12} Pa s (T_{12}).

181 We measured the viscosity of melts at high temperature ($\eta = 1$ to 10^5
 182 Pa s) in the concentric-cylinder viscometer at the University of Missouri
 183 – Columbia using a Theta Industries Rheotronic II Rotating Viscometer,
 184 equipped with a Brookfield HBDV-III Ultra measuring head and full range
 185 spring torque of $57496 \cdot 10^{-4}$ Nm). The crucible holding the melt and the
 186 cylindrical spindle rotating inside of the melt are both PtRh alloy. The in-
 187 strument measures the torque exerted by the melt on the spindle rotating at
 188 a constant angular velocity, at a given temperature. For each temperature,
 189 we record the torque for three different angular velocities to test for any non-
 190 Newtonian behavior. The viscosity we report for a given temperature is an
 191 average of the viscosity calculated for each angular velocity. The instrument
 192 is calibrated using NIST standards. The liquidus temperature of the melt
 193 investigated was generally the lower temperature limit for a given experi-
 194 ment, except for melts that exceeded the viscosity limit of the instrument
 195 or which were surprisingly stable in the supercooled state. Ne_{50} is the most
 196 potassium-rich composition we were able to measure using the instrument
 197 at the University of Missouri – Columbia. Melts with more potassium than
 198 Ne_{50} have liquidus temperatures higher than the upper limit of the instru-
 199 ment. Similarly, we were only able to obtain 2 measurements on sample
 200 Jd_{25} because of crystallization at temperatures near the upper limit of the
 201 instrument. We sent samples of $Ne_{37.5}$ and Ne_{25} to Anton-Paar in Ostfildern,
 202 Germany for measurements at higher temperature. The samples were mea-
 203 sured in a Anton-Paar FRS 1800, DSR 502 rheometer. Sample $Ne_{37.5}$ was
 204 measured in an Ar atmosphere using a graphite crucible and spindle, and
 205 Ne_{25} was measured in air using an alumina crucible and spindle. Measure-
 206 ments reported for $Ne_{37.5}$ and Ne_{25} were performed at 1750 °C. We estimate
 207 the uncertainty on viscosity to be less than 0.1 log units, based on compari-
 208 son with measurements on standard melts NIST 710a and 717a (Getson and
 209 Whittington, 2007).

210 3. Experimental results

211 3.1. Density

212 Overall, density is relatively constant across a join up to X_K ratios of 0.5,
 213 then decreases slightly at higher X_K (Figure 2d). Density variations across
 214 a join range from 1 to 3.5%. Density decreases linearly from highest Al/Si

ratio to lowest Al/Si ratio (Figure 2b). For any given Al/Si ratio, the increase in molar volume caused by the substitution of K for Na is roughly a linear function of the K/(Na+K) ratio; this increase is more important at higher Al/Si ratio (Figure 2a, c). These changes reflect changes in density across a join ranging from ~ 4 kg/m³ for high SiO₂ glasses to ~ 30 kg/m³ for low SiO₂ glasses. There is a larger spread in molar volume for glass compositions with higher Al/Si (3.5 cm³/mol) ratio than for glass compositions with small Al/Si ratios (1.3 cm³/mol). The potassium end-members range from 28.7 to 32.0 and sodium end-members range from 27.6 to 28.8 cm³/mol.

All samples have a higher density post parallel-plate viscometry than pre-viscometry (Table 1). All synthesized samples had fewer than 5% bubbles, except for *Ne*_{62.5}, which had between 15-20%. We expect bubbles to have little effect on viscosity measurements. Density measurements for samples in each of the two joins are in good agreement with those of Le Losq and Neuville (2013) and Le Losq et al. (2017), except for our leucite and orthoclase samples, whose density are $\sim 4\%$ lower (Figure 2). We used the density model of Lange (1997) to recalculate density and molar volume at T = 1098 K for compositions along each join. When the density data are normalized to the same fictive temperature, we see a slight increase (1-3 kg/m³) in density with increasing K along the Si₅O₁₂ and *AbOr* joins. For all other joins, a decrease in density is observed with increasing K, and this decrease becomes more important at higher Al/Si ratios (e.g., 3 kg/m³ for *JdLct* and 19 kg/m³ for *NeKls*). Normalizing the data to the same fictive temperature for all samples results in very good agreement between our data and those of Le Losq and Neuville (2013) and Le Losq et al. (2017). The trends in molar volume remain relatively unchanged with the normalization to a common fictive temperature, except for the molar volume for leucite decreasing from 31.36 to 30.60 cm³/mol, in better agreement with the data of Le Losq et al. (2017). Molar volume increases linearly with increasing K/(Na+K) along each join.

3.2. Viscosity

Viscosity data are reported in Tables 2-4. At high temperatures (T > 1300 °C), jadeite-leucite (*JdLct*) melts have viscosities ~ 1.25 log units higher than nepheline-kalsilite (*NeKls*) melts (Figure 3). For both joins, melt viscosity increases progressively with increasing potassium. Melts in both joins have approximately the same slope in the high temperature range.

Figure 2: *Black symbols: this study. Open triangles and diamonds: Le Losq and Neuville (2013). Grey circles, grey triangles, and squares: Le Losq et al. (2017). Shaded areas highlight trends in the data.*

At low temperatures above the glass transition ($T < 1000^\circ\text{C}$), *JdLct* melts have slightly higher viscosities than *NeKls* melts overall, and the rate of change in viscosity with temperature near the glass transition is higher for *NeKls* than for *JdLct* melts (Figure 3). Orthoclase and K-petalite have shallower slopes than jadeite-leucite melts. Our viscosity measurements are in good agreement with those of Le Losq et al. (2017) for nepheline-kalsilite melts, except for the sodium end-member. Our viscosity data for nepheline are also in good agreement with the measurements by Toplis et al. (1997a,b), but our jadeite data are ~ 0.3 log units higher, with a parallel slope.

The differences in viscosity between the melts we measured in this study

are on the order of ~ 2.5 log units across the whole temperature range. For the jadeite-leucite join, there is 3.1 log units of viscosity difference between the Na- and K-end members at 1300 K; at 1823 K, the difference is 1.6 log units (Figure 4b). Increasing K only has a small effect on T_{12} at low and intermediate K/(Na+K) ratios up to 0.4, but results in a rapid increase at higher K content (Figures 4a and 5). Although we did not measure the viscosity of kalsilite melt, extrapolation from available data suggests that the effects on T_{12} of substituting K for Na are similar for both joins.

Minima in isothermal viscosity occur at the sodium end-member for each join (Figure 4b). In the low-temperature range ($T = 1300$ K), viscosity varies non-linearly across a join and the mixed-alkali effect results in a depression in viscosity at intermediate Na/K ratios of as much as one log unit relative to a linear variation. In the high-temperature range ($T = 1823$ K), viscosity increases with increasing K across a join is nearly linear (± 0.25 log units) for most joins with the exception of the *JdLct* join for which a viscosity depression of ~ 0.4 log units is observed at intermediate Na/K ratios. There is a difference of ~ 50 K in the temperature at which the viscosity is 10^{12} Pa.s (T_{12}) between the *Si₅O₁₂* and the *NeKls* joins (Figure 4a), with melts of higher Al/(Al+Si) ratio having the lowest T_{12} on average. At $T = 1300$ K, Na-rich melts vary in viscosity from $10^{6.4}$ - $10^{8.9}$ Pa.s across Al/Si ratios from 0.2 to 1. K-rich melts span viscosities of $10^{9.4}$ - $10^{10.9}$ Pa.s for the same Al/Si ratios.

The data show that, overall, melts with high Al/(Al+Si) ratio are more fragile than melts with lower ratios (Figure 4d). This is also seen in the slopes of the viscosity data near the glass transition in Figure 3. T_{12} varies in a different way as a function of Al/(Al+Si) ratio for Na and K end-member melts (Figure 5a). The largest difference of temperature at the glass transition (T_{12}) between the end-member melts happens at an Al/(Al+Si) ratio of ~ 0.35 - 0.43 , that is, for the *JdLct* and *Si_{1.3}O_{4.7}* joins. This is both because of the higher viscosities of the K end-member and because these joins have lower viscosities for their Na end-members relative to the melts on other Al/(Al+Si) ratio joins. The largest ΔT_{12} occurs at the *Si_{1.3}O_{4.7}* join (Figure 5b).

Figure 3: All symbols as in legend shown in panel **a**. Some symbols are different shades of grey for clarity. All TVF fits use a constant A parameter equal to -4.3 (Russell et al., 2003). Numbers next to TVF fits refer to sodium end-member content (e.g., "25" on a dashed line means Ne_{25}). **a**. Viscosity data for all melts measured in this study, over the full temperature range. **b**. Near-glass transition viscosity data for nepheline-kalsilite melts. **c**. Near-glass transition viscosity data for jadeite-leucite melts. **d**. High-temperature range.

Table 2: Viscosity data – *JdLct*

Jd_{100}				$Jd_{62.5}$				$Jd_{37.5}$				Jd_0			
T (K)	$\log \eta$ (log Pa.s)	order	T (K)	$\log \eta$ (log Pa.s)	order	T (K)	$\log \eta$ (log Pa.s)	order	T (K)	$\log \eta$ (log Pa.s)	order	T (K)	$\log \eta$ (log Pa.s)	order	order
1103	11.25	6	PP	1082	11.76	8a	PP	1123	12.08	10a	PP	1195	13.02	10	PP
1113	10.90	4	PP	1142	10.51	7a	PP	1148	11.36	6a	PP	1204	12.73	9	PP
1122	10.71	2	PP	1161	10.16	4a	PP	1162	11.13	8a	PP	1214	12.45	8	PP
1127	10.49	7	PP	1179	9.84	5a	PP	1173	10.85	9a	PP	1219	12.30	15	PP
1136	10.39	3	PP	1191	9.60	3a	PP	1191	10.43	4a	PP	1224	12.28	7	PP
1146	10.04	5	PP	1217	9.16	11a	PP	1198	10.30	7a	PP	1229	12.07	14	PP
1170	9.58	1	PP	1774	3.59	3b	CC	1222	9.84	3a	PP	1234	12.02	6	PP
1177	9.50	9	PP	1799	3.42	4b	CC	1230	9.72	5a	PP	1238	11.86	13	PP
1710	3.61	8b	CC	1824	3.25	2b	CC	1244	9.51	11a	PP	1244	11.79	5	PP
1742	3.39	7b	CC	1849	3.09	5b	CC	1819	3.61	4b	CC	1249	11.60	11	PP
1763	3.25	6b	CC	1866	2.98	6b	CC	1840	3.47	3b	CC	1254	11.47	4	PP
1783	3.12	5b	CC	1866	2.99	1b	CC	1854	3.37	5b	CC	1257	11.54	12	PP
1803	2.99	4b	CC					1860	3.34	2b	CC	1263	11.29	10a	PP
1824	2.87	3b	CC					1864	3.30	6b	CC	1266	11.24	12	PP
1841	2.77	2b	CC					1869	3.28	1b	CC	1269	11.28	11	PP
1854	2.68	9b	CC					1870	3.26	7b	CC	1270	11.17	2	PP
1864	2.64	1b	CC												
Jd_{75}				Jd_{50}				Jd_{25}							
T (K)	$\log \eta$ (log Pa.s)	order	T (K)	$\log \eta$ (log Pa.s)	order	T (K)	$\log \eta$ (log Pa.s)	order	T (K)	$\log \eta$ (log Pa.s)	order	T (K)	$\log \eta$ (log Pa.s)	order	order
1057	12.07	10b	PP	1153	10.74	5a	PP	1163	11.66	6	PP				
1072	11.80	8b	PP	1166	10.62	7a	PP	1186	11.00	7	PP				
1088	11.44	9b	PP	1166	10.30	1a	PP	1195	10.75	3	PP				
1103	11.13	5a	PP	1175	10.21	6a	PP	1205	10.58	5	PP				
1107	10.96	6b	PP	1192	9.92	3a	PP	1227	10.04	1	PP				
1113	10.78	7b	PP	1200	9.74	4a	PP	1252	9.73	9	PP				
1122	10.70	2a	PP	1202	9.65	5b	PP	1256	9.74	8	PP				
1134	10.48	4a	PP	1213	9.44	3b	PP	1852	3.62	2b	CC				
1136	10.42	3a	PP	1216	9.43	2a	PP	1865	3.49	1b	CC				
1146	10.18	6a	PP	1225	9.30	9a	PP	1867	3.49	3b	CC				
1151	10.01	4b	PP	1240	9.02	11b	PP								
1159	9.85	5b	PP	1711	3.95	10c	CC								
1163	9.79	8a	PP	1735	3.76	9c	CC								
1186	9.33	3b	PP	1758	3.60	8c	CC								
1198	9.12	11b	PP	1782	3.44	7c	CC								
1745	3.55	4c	CC	1807	3.26	6c	CC								
1771	3.38	3c	CC	1826	3.16	11c	CC								
1796	3.21	5c	CC	1833	3.10	5c	CC								
1822	3.05	2c	CC	1833	3.09	12c	CC								
1847	2.89	6c	CC	1850	2.99	4c	CC								
1864	2.80	1c	CC												
1864	2.78	7c	CC												

PP: Parallel-plate viscometry.

CC: Concentric-cylinder viscometry.

Order: Order in which viscosity measurements were performed. Order resets for each viscometry method and is indicated by a letter following the number. If multiple viscosity experiments were performed using one method on a given sample, this is also indicated by a letter following the number.

Table 3: Viscosity data – *NeKls*

Ne ₁₀₀				Ne _{62.5}				Ne _{37.5}			
T (K)	log η (log Pa.s)	order		T (K)	log η (log Pa.s)	order		T (K)	log η (log Pa.s)	order	
1073	12.07	9a	PP	1087	11.80	10a	PP	1147	11.49	9	PP
1101	11.04	7b	PP	1092	11.60	8a	PP	1158	11.17	10	PP
1115	10.79	8c	PP	1107	11.22	6a	PP	1172	10.90	7	PP
1129	10.24	5	PP	1110	11.03	9a	PP	1178	10.55	8	PP
1143	9.95	6	PP	1118	10.82	7a	PP	1192	10.33	5	PP
1158	9.50	4	PP	1136	10.37	4a	PP	1211	9.82	4	PP
1811	1.70	3b	CC	1150	10.02	5a	PP	1219	9.70	6	PP
1821	1.65	5b	CC	1178	9.32	3a	PP	1238	9.29	12	PP
1830	1.60	2b	CC	1188	9.14	11a	PP	2023	1.04	1a	CC
1846	1.53	1b	CC	1775	2.48	3b	CC	Ne ₂₅			
1861	1.45	4b	CC	1800	2.32	4b	CC	T	log η	order	
Ne ₇₅				1825	2.16	2b	CC	(K)	(log Pa.s)		
T	log η	order		1851	2.01	5b	CC	1183	11.49	8	PP
(K)	(log Pa.s)			1865	1.94	1b	CC	1197	10.90	6	PP
1072	11.82	10c	PP	1866	1.91	6b	CC	1220	10.50	9	PP
1078	11.89	11a	PP	Ne ₅₀				1234	10.13	7	PP
1086	11.48	8c	PP	T	log η	order		1245	9.83	1	PP
1093	11.40	9a	PP	(K)	(log Pa.s)			1248	9.72	4	PP
1100	11.24	11c	PP	1113	11.72	11a	PP	1251	9.68	3	PP
1110	10.84	7a	PP	1122	11.44	9a	PP	1252	9.74	10	PP
1114	10.66	6c	PP	1133	11.08	10a	PP	1264	9.43	2	PP
1120	10.58	10a	PP	1143	10.88	7a	PP	1264	9.37	5	PP
1127	10.45	7c	PP	1153	10.58	8a	PP	2023	0.93	1a	CC
1131	10.23	9c	PP	1163	10.32	5a	PP				
1135	10.20	8a	PP	1170	10.05	1a	PP				
1141	10.07	12c	PP	1182	9.86	3a	PP				
1149	9.74	5a	PP	1189	9.66	2a	PP				
1152	9.66	1a	PP	1198	9.51	4a	PP				
1154	9.65	5c	PP	1205	9.32	6a	PP				
1160	9.50	6a	PP	1218	9.04	12a	PP				
1166	9.35	3a	PP	1806	2.36	3b	CC				
1171	9.25	2a	PP	1830	2.21	2b	CC				
1175	9.25	12a	PP	1840	2.15	1b	CC				
1179	9.03	4a	PP	1850	2.06	4b	CC				
1682	2.75	7b	CC								
1707	2.58	6b	CC								
1730	2.43	5b	CC								
1757	2.27	4b	CC								
1780	2.13	3b	CC								
1805	1.99	2b	CC								
1841	1.80	1b	CC								

PP: Parallel-plate viscometry.

CC: Concentric-cylinder viscometry.

Order: Order in which viscosity measurements were performed. Order resets for each viscometry method and is indicated by a letter following the number. If multiple viscosity experiments were performed using one method on a given sample, this is also indicated by a letter following the number.

Table 4: Viscosity data for *Or* and K-petalite

K-petalite				Or			
T	log η	order		T	log η	order	
(K)	(log Pa.s)			(K)	(log Pa.s)		
1189	12.45	5	PP	1201	12.31	8	PP
1199	12.31	8	PP	1205	12.26	23	PP
1214	12.02	9	PP	1211	12.11	6	PP
1223	11.93	8b	PP	1214	12.07	22	PP
1229	11.73	7	PP	1215	12.02	15	PP
1244	11.47	10	PP	1225	11.82	16	PP
1252	11.33	6	PP	1226	11.77	5a	PP
1254	11.25	4	PP	1228	11.77	5b	PP
1262	11.13	4b	PP	1231	11.65	4	PP
1265	11.02	2	PP	1233	11.67	6b	PP
1268	11.07	7b	PP	1234	11.62	6a	PP
1269	11.04	11	PP	1235	11.63	21	PP
1274	11.03	9b	PP	1235	11.62	17	PP
1274	10.92	6b	PP	1236	11.59	5	PP
1279	10.86	5b	PP	1243	11.45	4	PP
1287	10.67	3	PP	1244	11.38	7	PP
1289	10.67	3b	PP	1245	11.44	18	PP
				1252	11.25	7	PP
				1252	11.24	9b	PP
				1252	11.26	9a	PP
				1255	11.26	19	PP

PP: Parallel-plate viscometry.

CC: Concentric-cylinder viscometry.

Order: Order in which viscosity measurements were performed. Order resets for each viscometry method and is indicated by a letter following the number. If multiple viscosity experiments were performed using one method on a given sample, this is also indicated by a letter following the number.

Figure 4: **a.** T_{12} vs. X_K . **b.** Isotherms for $T = 1300$ K (high viscosity) and $T = 1823$ K (low viscosity). Lines are dashed for extrapolated data. **c.** ΔT_{12} (relative to sodium end-member) vs. X_K . **d.** Fragility (m) as calculated by Equation 4 vs. X_K . **e** and **f**: TVF fit parameters. All data reported using a constant A parameter equal to -4.3 (Russell et al., 2003). All symbols as in legend shown in panel a.

Figure 5: **a.** T_{12} vs. X_{Al} , and **b.** ΔT_{12} (relative to sodium end-member) vs. X_{Al} . All symbols as in legend shown in panel **a**. Vertical bars at $Al/(Al+Si) = 0.52$ represent extrapolations of our viscosity data ($T_{12} \sim 1300$ K and $\Delta T_{12} \sim 200$ K) to the K end-member for the NeKls join, which predict much higher values than those of Le Losq et al. (2017). Values for the Na end-members, K end-members, and for $K/(Na+K) = 0.5$ are shown.

294 4. Modelling

295 4.1. Tamman-Vogel-Fulcher empirical fits

296 We report empirical fits to the viscosity data for each of the samples
 297 studied in Table 5. We fitted the viscosity data using three approaches:
 298 i) fit each sample individually without imposing constraints on any of the
 299 parameters, ii) fit each sample individually constraining $A_{TVF} = -4.3$ as per
 300 Russell et al. (2003), and iii) impose a common, but unconstrained A_{TVF}
 301 to all samples along the same join. In Figures 4e and f, we compare the fit
 302 parameters obtained with the constraint of $A = -4.3$ for our samples and
 303 those of Le Losq and Neuville (2013) and Le Losq et al. (2017). B_{TVF} shows
 304 an overall increase with increasing X_K , except for our *NeKls* measurements
 305 which increase until $X_K = 0.4$ and then decrease at higher X_K . C_{TVF} shows
 306 a concave up trend across the joins, with lowest values at intermediate X_K .
 307 Fragility (Equation 4) decreases slightly overall with increasing X_K , except
 308 for our *NeKls* measurements and the $Si_{1.3}O_{4.7}$ join which show a minimum
 309 at intermediate X_K (Figure 4d).

Table 5: TVF parameters for fits to individual compositions following equation: $\log(\eta) = A + \frac{B}{T-C}$

Sample	n	A_{ind}^{TVF}	B_{ind}^{TVF}	C_{ind}^{TVF}	RMSD	T_{12} (K)	m
K-petalite	17	-4.30	13878	363.3	0.04	1215	23.3
Or	21	-4.30	12446	452.7	0.02	1216	26.0
Jd ₁₀₀	17	-4.30	9687	475.0	0.05	1069	29.3
Jd ₇₅	22	-4.30	10157	442.1	0.05	1065	27.9
Jd _{62.5}	12	-4.30	10523	431.9	0.05	1077	27.2
Jd ₅₀	24	-4.30	10141	474.3	0.07	1096	28.7
Jd _{37.5}	16	-4.30	10584	474.5	0.03	1124	28.2
Jd ₂₅	10	-4.30	10816	479.8	0.07	1143	28.1
Jd ₀	16	-4.30	11241	543.8	0.04	1233	29.2
JdLct^a	117				0.06		
Ne ₁₀₀	11	-4.30	7016	646.9	0.07	1077	40.8
Ne ₇₅	27	-4.30	7578	609.6	0.08	1075	37.7
Ne _{62.5}	15	-4.30	7978	591.7	0.04	1081	36.0
Ne ₅₀	16	-4.30	7905	621.6	0.06	1107	37.2
Ne _{37.5}	9	-4.30	7501	677.2	0.15	1137	40.3
Ne ₂₅	11	-4.30	7150	736.3	0.17	1175	43.7
NeKls^a	89				0.10		
Sample	n	A_{join}^{TVF}	B_{join}^{TVF}	C_{join}^{TVF}	RMSD	T_{12} (K)	m
K-petalite ^b	17	-11.37	28412	0.0	0.04	1216	23.4
Or ^b	21	-5.80	14882	380.2	0.02	1216	25.9
Jd ₁₀₀		-5.01	11136	411.6	0.06	1066	27.7
Jd ₇₅		-5.01	11613	380.3	0.05	1063	26.5
Jd _{62.5}		-5.01	12042	366.5	0.03	1074	25.8
Jd ₅₀		-5.01	11563	414.1	0.05	1094	27.4
Jd _{37.5}		-5.01	12087	411.1	0.04	1122	26.9
Jd ₂₅		-5.01	12334	415.7	0.08	1141	26.8
Jd ₀		-5.01	12244	513.7	0.04	1233	29.2
JdLct^a	117				0.05		
Ne ₁₀₀		-5.63	9290	546.2	0.04	1069	32.1
Ne ₇₅		-5.63	9795	514.7	0.06	1066	30.9
Ne _{62.5}		-5.63	10413	486.4	0.05	1073	29.1
Ne ₅₀		-5.63	10246	520.7	0.03	1097	30.3
Ne _{37.5}		-5.63	9747	580.1	0.08	1141	31.3
Ne ₂₅		-5.63	9228	646.8	0.10	1105	42.1
NeKls^a	89				0.06		

Values either fit with the constraint $A = -4.3$ (top) or with common A for each join (bottom).

^a Overall RMSD values for the whole join.

^b Values for individual composition with $A =$ unconstrained.

$$m = \frac{B_{TVF}}{T_{12} \left(1 - \frac{C_{TVF}}{T_{12}}\right)^2} \quad (4)$$

Root-mean-square deviation (RMSD) values obtained by TVF fitting are slightly better when A^{TVF} is unconstrained for an individual composition or is a common, but unconstrained value for a given join (Table 5). The greatest improvement of fit is for the *NeKls* join. When A is unconstrained, it varies between -5.8 and -5.0 for *NeKls*, *JdLct*, and *Or*, and returns a value of -11.4 for K-petalite.

4.2. Configurational entropy modelling

4.2.1. Individual compositions

We used configurational entropy modelling to obtain values of A_e , B_e , and $S^{conf}(T_g)$ (Equations 1 and 2). We used the model of Richet (1987) for the heat capacity of multicomponent oxide glass. This model predicts a smooth, and very small, decrease for the heat capacity of the glass at the glass transition temperature as X_K increases. We used the model of Stebbins et al. (1984) for the heat capacity of the liquid. This model predicts trends parallel to those of the glass, when calculated at the glass transition temperature, but we note that the model does not account for temperature-dependent liquid heat capacity (e.g., Richet and Bottinga, 1984; Bouhifd et al., 1999). The patterns are the same for both the *NeKls* and *JdLct* joins. We used the viscometric glass transition (T_{12}) as calculated with TVF fits with constraint $A = -4.3$ as T_g in the modelling. We show results obtained by fitting each sample individually without any constraints on the Adam-Gibbs parameters, and we provide data from the literature for comparison in Table 6 and Figure 6. The individual configurational entropy model fits all return equivalent RMSD values as for individual, unconstrained TVF fits, or better RMSD values than TVF fits when A^{TVF} is constrained to -4.3 (Table 5). The only exception to this is $KAlSi_3O_8$, which is better fit by TVF. Overall, the values we obtain for parameters A_e , B_e , and $S^{conf}(T_g)$ are reasonable, but there is significant noise. The *JdLct* join shows an obvious peak in B_e and $S^{conf}(T_g)$ at intermediate K/(Na+K) ratios (Figure 6). Our fits show a greater range of values across the *NeKls* and *AbOr* joins than those presented by Le Losq et al. (2017) who constrained the A_e parameter for each join. Our unconstrained results suggest a dependence of the fit parameters on Al/Si ratio, whereas there is no systematic variation with respect to Na/K ratio.

Figure 6: *Individual Adam-Gibbs fits for melts along joins in the $(\text{Na},\text{K})\text{AlSiO}_4\text{-SiO}_2$ system using model heat capacity for glass and liquid. Values for NeKls LN and AbOr LN are from Le Losq et al. (2017) and have common, but unconstrained A_e value for each join instead of individual unconstrained A_e values for each composition along the join. Panels a, b, c and d show A_e , $10^{-3}B_e$, $S^{\text{conf}}(T_g)$, and $10^{-3}B_e/S^{\text{conf}}(T_g)$ as a function of X_K , respectively.*

4.2.2. Parameterization of the $(\text{Na},\text{K})\text{AlSiO}_4\text{-SiO}_2$ system

We also used the models for the heat capacity of the glass (Richet, 1987) and that of the liquids (Stebbins et al., 1984) to investigate all viscosity datasets for which multiple compositions across a join of constant Al/Si ratio were available within the $(\text{Na},\text{K})\text{AlSiO}_4\text{-SiO}_2$ system. The A_e parameter was either treated as having a common, but unconstrained value for all compositions along each join of constant Al/Si ratio, or as being -3.51 (Russell and Giordano, 2017). We used forms allowing for non-ideal mixing for both

351 the B_e and the $S^{conf}(T_g)$ parameters.

352 B_e is the enthalpy term:

$$B_e = X_{Na}B_{Na} + (1 - X_{Na})B_K + w_B X_{Na}(1 - X_{Na}) \quad (5)$$

353 where B_{Na} is the value of B_e for the sodium end-member, B_K is the value
 354 of B_e for the potassium end-member, X_{Na} is the mole fraction of sodium
 355 across a join, and w_B is the Margules parameter for B_e . The first two terms
 356 of Equation 5 represent linear (ideal) symmetrical mixing and the third term
 357 is a non-ideal mixing term that reduces to ideal mixing when $w_B = 0$.

358 $S^{conf}(T_g)$ is the entropy term:

$$\begin{aligned} S^{conf}(T_g) = & X_{Na}S^{conf}(T_g)_{Na} + (1 - X_{Na})S^{conf}(T_g)_K \\ & - R[X_{Na}\ln X_{Na} + (1 - X_{Na})\ln(1 - X_{Na})] \\ & + w_S X_{Na}(1 - X_{Na}) \end{aligned} \quad (6)$$

359 where $S^{conf}(T_g)_{Na}$ is the value of $S^{conf}(T_g)$ for the sodium end-member,
 360 $S^{conf}(T_g)_K$ is the value of $S^{conf}(T_g)$ for the potassium end-member, X_{Na} is
 361 the mole fraction of sodium across a join, R is the gas constant, and w_S is
 362 the Margules parameter for $S^{conf}(T_g)$. The first three terms of Equation 6
 363 represent ideal mixing and the third term is a non-ideal mixing term with a
 364 symmetrical form across the join. We tested an asymmetrical form but did
 365 not obtain better fits. All fit parameters obtained for $A_e = -3.51$ and B_e and
 366 the $S^{conf}(T_g)$ as in Equations 5 and 6 above are shown in Figure 7.

367 RMSD values for the joins range from 0.11 to 0.23. Parameters B_{Na} , B_K ,
 368 w_b , $S^{conf}(T_g)_{Na}$, and $S^{conf}(T_g)_K$ show a negative linear relationship with
 369 Al/Si ratio. Overall, B_K is smaller than B_{Na} , and $S^{conf}(T_g)_K$ is smaller than
 370 $S^{conf}(T_g)_{Na}$. We see a larger difference between the Na and K end members
 371 of the B_e and $S^{conf}(T_g)$ parameters at lower Al/Si ratio. Parameter w_s has
 372 a much weaker relationship with Al/Si ratio.

Table 6: Configurational entropy modelling unconstrained fit results to individual compositions

Suite	$\frac{Al}{Al+Si}$	$\frac{K}{Na+K}$	A_e^{ind} (log Pa s)	$10^{-3}B_e^{ind}$ (KJ mol ⁻¹)	$S^{conf}(T_g)^{ind}$ (J mol ⁻¹ K ⁻¹)	$\frac{10^{-3}B_e}{S^{conf}(T_g)}^{ind}$ (10 ³ K)	RMSD Individual	RMSD Join
Si ₅ O ₁₂	0.17	0.00	-0.63	154.3	10.9	14.1	0.03	
Si ₅ O ₁₂	0.17	0.25	3.51	44.5	4.5	9.8	0.03	
Si ₅ O ₁₂	0.17	0.50	4.91	32.2	4.0	8.1	0.03	
Si ₅ O ₁₂	0.17	0.73	3.21	59.7	5.7	10.5	0.02	
Si ₅ O ₁₂	0.17	0.99	5.86	27.5	3.6	7.6	0.02	0.03
Na-petalite T97	0.18	0.00	-4.43	4372.0	249.7	17.5	0.06	
K-petalite	0.22	0.99	-0.34	175.3	11.7	15.0	0.04	
Ab T97	0.25	0.00	-5.72	5451.0	289.1	18.9	0.01	
Ab SS93	0.27	0.00	1.34	207.2	6.9	29.9	0.09	
AbOr	0.25	0.00	-0.08	111.6	8.5	13.1	0.01	
AbOr	0.25	0.20	-1.27	165.5	11.5	14.4	0.02	
AbOr	0.25	0.39	-0.39	140.8	10.1	13.9	0.02	
AbOr	0.25	0.50	0.63	117.1	9.3	12.6	0.03	
AbOr	0.25	0.59	-0.75	163.5	11.2	14.6	0.04	
AbOr	0.25	0.79	0.78	111.9	8.5	13.2	0.02	
AbOr	0.25	1.00	1.56	96.8	7.6	12.8	0.03	0.02
Or	0.26	1.00	-4.51	245.5	12.2	20.1	0.08	
Jd SS93	0.32	0.00	-3.19	1486.9	91.9	16.2	0.08	
Jd SS93	0.34	0.00	-6.17	4241.2	225.8	18.8	0.05	
Jd T97	0.33	0.00	-4.38	2519.6	148.3	17.0	0.04	
JdLct	0.35	0.00	-2.64	178.9	11.4	15.7	0.05	
JdLct	0.35	0.26	-3.39	273.8	16.7	16.3	0.05	
JdLct	0.34	0.38	-4.28	417.2	24.0	17.4	0.02	
JdLct	0.35	0.51	-4.37	378.3	21.2	17.8	0.04	
JdLct	0.33	0.63	-2.87	227.2	13.6	16.7	0.03	
JdLct	0.35	0.76	-2.59	216.2	12.9	16.7	0.07	
JdLct	0.35	1.00	-2.98	237.5	12.9	18.5	0.04	0.05
60:50 T97	0.40	0.00	-4.27	2099.9	124.9	16.8	0.03	
Si _{1.4} O _{4.7}	0.43	0.00	-0.29	88.0	6.7	13.2	0.01	
Si _{1.4} O _{4.7}	0.42	0.24	-0.10	101.2	7.7	13.2	0.02	
Si _{1.4} O _{4.7}	0.42	0.47	-0.82	124.4	8.8	14.2	0.02	
Si _{1.4} O _{4.7}	0.42	0.73	1.33	88.5	7.2	12.2	0.02	
Si _{1.4} O _{4.7}	0.42	0.99	4.24	35.9	3.7	9.7	0.03	0.02
Ne T97	0.51	0.00	-3.57	1156.4	68.8	16.8	0.04	
NeKls	0.52	0.00	-3.84	170.9	10.1	16.9	0.04	
NeKls	0.52	0.26	-3.68	189.8	11.3	16.7	0.06	
NeKls	0.51	0.38	-3.34	185.8	11.2	16.5	0.03	
NeKls	0.52	0.51	-3.78	210.6	12.1	17.3	0.03	
NeKls	0.50	0.63	-5.01	275.3	14.4	19.1	0.05	
NeKls	0.52	0.75	-5.82	314.2	15.3	20.5	0.06	0.05
NeKls LN	0.51	0.00	1.43	55.1	4.8	11.4	0.02	
NeKls LN	0.52	0.26	-0.99	110.1	7.9	13.9	0.01	
NeKls LN	0.50	0.50	0.27	93.2	7.2	13.0	0.01	
NeKls LN	0.50	0.78	-0.24	110.1	8.0	13.8	0.03	
NeKls LN	0.50	0.99	3.03	51.4	4.8	10.7	0.02	0.02
RMSD ALL								0.04

We refit all previously published viscosity data, *Si₅O₁₂*, *AbOr*, *Si_{1.4}O_{4.7}*, *NeKls* LN, SS93: Ab and Jd, T97: Na-petalite, Ab, Jd, 60:50, and Ne (Stein and Spera, 1993; Toplis et al., 1997a,b; Le Losq and Neuville, 2013; Le Losq et al., 2017).

Figure 7: *Parameterization of the $(\text{Na},\text{K})\text{AlSiO}_4$ system. All data were obtained using model heat capacity data for glasses and liquids and fitting each join separately with Equations 5 and 6, and modelling A_e as a constant equal to -3.51 (Russell and Giordano, 2017). In panels **c** and **d**, the larger symbols are for the sodium end-member, and the smaller symbols are for the potassium end-member. There are multiple data points for a given join due to slight Al/Si ratio variations.*

373 4.2.3. Global fit

374 We expanded our modelling of the viscosity of melts in the $(\text{Na},\text{K})\text{AlSiO}_4$ -
 375 SiO_2 system to also include compositions along the end-member joins SiO_2 -
 376 NaAlSiO_4 and SiO_2 - KAlSiO_4 (Toplis et al., 1997a,b; Stein and Spera, 1993;
 377 N'dala et al., 1984; Urbain et al., 1982; Riebling, 1966) in addition to our
 378 data and those of Le Losq and Neuville (2013) and Le Losq et al. (2017).

379 A total of 49 datasets were included, representing 598 viscosity data points.
 380 None of these studies reported measurements of dissolved water contents for
 381 their samples and we consider them nominally anhydrous and all similarly
 382 affected by any dissolved water (<0.1 wt.%, Davis, 1999; Ryan et al., 2015).
 383 Our high-temperature viscosity data for nepheline melt agree with those of
 384 Toplis et al. (1997b) and Riebling (1966). The nepheline data of N'dala et al.
 385 (1984) are roughly one log unit higher viscosity for the same temperatures.
 386 At temperatures near the glass transition, there is good agreement between
 387 our data and Toplis et al. (1997b) and Le Losq et al. (2017). Our high-
 388 temperature viscosity data for jadeite are about 0.3 log units higher than
 389 previous data (Riebling, 1966; Toplis et al., 1997b; Stein and Spera, 1993).
 390 This is also the case at temperatures near the glass transition, when compared
 391 to the data of Toplis et al. (1997b).

392 Based on our parameterization results shown in Figure 7, we chose to use
 393 the non-ideal symmetrical mixing forms for parameters B_e and $S^{conf}(T_g)$ and
 394 parameterize the end-member values and Margules parameters as a function
 395 of X_{Al} , as follows:

$$B_{Na} = b_{Na}^1 + b_{Na}^2 X_{Al} \quad (7)$$

$$B_K = b_K^1 + b_K^2 X_{Al} \quad (8)$$

$$w_B = w_B^1 + w_B^2 X_{Al} \quad (9)$$

$$S^{conf}(T_g)_{Na} = c_{Na}^1 + c_{Na}^2 X_{Al} \quad (10)$$

$$S^{conf}(T_g)_K = c_K^1 + c_K^2 X_{Al} \quad (11)$$

$$w_S = w_S \quad (12)$$

401 We present two different parameterizations for A_e :

$$A_e^{-3.51} = -3.51 \quad (13)$$

$$A_e^{AlSi} = a_1 + a_2 X_{Al} \quad (14)$$

403 The second one (Equation 14) allows A_e to vary as a function of Al/Si
 404 ratio as suggested by the results shown in Figure 6a. Equations 5-14 break
 405 down for pure SiO_2 , and we therefore exclude it from the model. Indeed, the
 406 addition of trace amounts of Al, Na, or K to SiO_2 melt dramatically changes
 407 its properties, in a non-linear way (e.g., Knoche et al., 1994; Wu et al., 2015,
 408 and references therein), whereas our model forces a linear relationship with

409 respect to X_{Al} . Figure 8 shows the comparison of measured vs. calculated
 410 viscosity for all of the datasets included in the model, as well as viscosity
 411 vs. reciprocal temperature fits using the model for *NeKls*, *JdLct*, *Or*, and
 412 K-Petalite compositions. Figures 9 and 10 show the two different forms of
 413 the model. We obtain a RMSD of 0.32 for $A_e^{-3.51}$ and a RMSD of 0.26 for
 414 A_e^{AlSi} (Table 7). When A_e is constrained to a single value, we obtain a range
 415 of values between joins for the B_e and $S^{conf}(T_g)$ parameters (Figure 10b,c).
 416 Both parameters converge at $X_K = 1$ whereas they show a greater range in
 417 values at the sodium end-member. When A_e is allowed to vary linearly as a
 418 function of Al/Si ratio, the $S^{conf}(T_g)$ parameter does not vary across joins
 419 (Figure 9c). B_e^{AlSi} converges at the sodium end-member but shows a range of
 420 values at $X_K = 1$ (Figure 9b). A_e^{AlSi} varies between -2.5 and -4 (Figure 9a).
 421 The $B_e/S^{conf}(T_g)$ ratios show similar patterns for both models, but opposite
 422 relationships with respect to Al/Si ratio (Figures 9d and 10d).

Table 7: Fit parameters for the global configurational entropy model

	$A_e=-3.51$	$A_e=AlSi$
a_1	-3.51	-1.56
a_2	—	-4.93
w_b^1	199715	280675
w_b^2	32187	18470
B_{Na}^1	155049	164971
B_{Na}^2	116661	27060
B_K^1	177611	152918
B_K^2	3968	76119
w_s	-10.2	-5.1
c_{Na}^1	8.0	10.6
c_{Na}^2	10.5	-0.3
c_K^1	8.9	9.0
c_K^2	1.0	1.0
n	598	598
RMSD	0.32	0.26
Se	0.33	0.27

Figure 8: Comparison of measured and calculated viscosity for the two parameterizations of the configurational entropy models: Panels **a** and **b**: $A_e = -3.51$ (Russell and Giordano, 2017); Panels **c** and **d**: $A_e = \text{linear function of Al/Si ratio}$. Dashed lines in panels **b** and **d** indicate uncertainties on viscosity of ± 0.3 log units. Symbol size has been reduced in the insets for a better visualization of the fits. Thick grey dashed line and grey symbols in panels **a** and **c** represent fits for haplogranite HPG8 (Holtz et al., 1992), which was not used in the calibration of the models.

Figure 9: Global viscosity model for system $(Na,K)AlSiO_4$, for A_e a linear function of Al/Si ratio. Panels **a**, **b**, **c**, and **d** show A_e , $10^{-3}B_e$, $S^{conf}(T_g)$, and $10^{-3}B_e/S^{conf}(T_g)$ as a function of X_K , respectively. Black dashed line shows model for $X_{Al} = 0.17$, grey dashed line for $X_{Al} = 0.5$. Comparison with $S^{conf}(T_g)$ values calculated for ideal mixing is shown in panel **c** (thin solid lines). See Figure 10 for legend.

Figure 10: Global viscosity model for system $(Na,K)AlSiO_4$, for $A_e = -3.51$ (Russell and Giordano, 2017). **a**. Legend for this figure and Figure 9. Panels **b**, **c**, and **d** show $10^{-3}B_e$, $S^{conf}(T_g)$, and $10^{-3}B_e/S^{conf}(T_g)$ as a function of X_K , respectively. Black dashed line shows model for $X_{Al} = 0.17$, grey dashed line for $X_{Al} = 0.5$. Comparison with $S^{conf}(T_g)$ values calculated for ideal mixing is shown in panel **c** (thin solid lines).

5. Discussion

Our density data are consistent with the structural data and interpretations of Le Losq and Neuville (2013) and Le Losq et al. (2017) that the dilation of rings and cages in the network and the formation of percolation

channels at high K in high Al/Si melts causes a significant increase of molar volume.

We note that the Kls_{99} melt of Le Losq et al. (2017) has a much lower viscosity at the glass transition than what our viscosity data would predict by extrapolating our $NeKls$ T_{12} trend to the end-member Kls . Considering that the glass structure data obtained by Raman spectroscopy and molecular dynamics simulations clearly demonstrate that the addition of K has greater effects at higher Al/(Al+Si) ratios, producing percolation channels and other changes in the melt which Le Losq and Neuville (2013) and Le Losq et al. (2017) have linked to the observed increase in viscosity as K/(Na+K) increases, we would not expect end-member Kls to have the lowest viscosity of all K end-member melts. If we used the extrapolation of our $NeKls$ viscosity data instead (vertical bars in Figures 5a,b) the magnitude of change in T_{12} relative to the Na end-member melt across a join would increase with increasing Al/(Al+Si) ratio. The viscosity data by Le Losq and Neuville (2013) and Le Losq et al. (2017) instead suggest that the greatest effect on viscosity due to K/(Na+K) increase happens at intermediate Al/(Al+Si) ratios.

5.1. Configurational entropy modelling

5.1.1. Unconstrained fits to individual compositions

The goodness of fit obtained with configurational entropy modelling (AG) for individual compositions is equivalent to that of TVF (Tables 5 and 6). The overall RMSD obtained by fitting all compositions along the $JdLct$ and $NeKls$ joins using individual, unconstrained TVF or AG fits is ~ 0.05 , an uncertainty equivalent or better than viscosity measurement uncertainty.

5.1.2. Global model

Fit parameters for both parameterizations of the global model are provided in Table 7, and the corresponding values of A_e , B_e , and $S^{conf}(T_g)$ are provided in Appendix Table A1. We used the same 598 data in each parameterization of the global model. Only 20 compositions have both high and low-temperature viscosity data available: 12 compositions from this study, 5 from Toplis et al. (1997a), and 3 from Stein and Spera (1993). Much more data are available at low temperatures near the glass transition, without any high-temperature constraints (25 datasets). Four compositions were only measured at high viscosity.

Several factors affect the goodness of fit of the global models. For example, Toplis et al. (1997a,b) clearly showed the effects on viscosity of small

463 variations in composition in nominally fully-polymerized melts. Some of the
 464 older datasets do not include chemical characterization of the samples (Ur-
 465 bain et al., 1982; N’dala et al., 1984), and we included them assuming the
 466 nominal compositions provided by the authors. Of the 45 analyzed compo-
 467 sitions, 21 fall between $0.98 < (\text{Na}+\text{K})/\text{Al} < 1.02$ ($\pm 2\%$), and 37 between
 468 $0.95\text{--}1.05$ ($\pm 5\%$). All compositions that are outside of $(\text{Na}+\text{K})/\text{Al} = 1 \pm 5\%$
 469 are peraluminous. The largest difference is 16% relative for composition
 470 83.8.0 from Le Losq and Neuville (2013) (Si_5O_{12} join), which corresponds
 471 to 0.1 log units or less variation in viscosity at 1596 °C, but to ~ 1 log unit
 472 near the glass transition (Toplis et al., 1997a,b). Overall, 7 compositions are
 473 peralkaline, all within $\pm 5\%$ of the metaluminous join, corresponding to a
 474 variation in viscosity of ~ 0.02 log units at 1596 °C and less than 1 log unit
 475 near the glass transition (Toplis et al., 1997a,b). These include all composi-
 476 tions from Stein and Spera (1993), T97 60:50 (Toplis et al., 1997a), $Jd_{37.5}$,
 477 $Ne_{62.5}$, and LN Ne_{75} (Le Losq et al., 2017). We chose to include each dataset
 478 without any corrections, even in cases of disagreement between datasets (see
 479 Section 4.2.3) since we do not have any evidence of systematic errors in the
 480 datasets, nor can we accurately correct all compositions that are not exactly
 481 metaluminous. All of these factors, of course, introduce uncertainty in the
 482 model and it is difficult to evaluate its magnitude. However, our primary goal
 483 was to use the power of a large dataset in order to extract thermodynamic
 484 information concealed in the noise of the smaller datasets.

485 Our model with A_e parameterized as a function of Al/Si ratio is visibly
 486 more accurate overall than when A_e is modelled as a constant (Figure 8).
 487 Russell and Giordano (2017) obtained a A_e value of -3.51 ± 0.25 by fitting
 488 mostly complex multicomponent natural melts covering a wide range of silica
 489 contents. For binary and ternary silicate and aluminosilicate melts including
 490 nominally fully polymerized albite, jadeite, and nepheline, Toplis (1998) es-
 491 timated a value of -2.6 ± 1 that varied linearly as a function of $B_e/(\text{Si}+\text{Al})$.
 492 We obtain A_e that varies between -2.4 (Al/Si = 0.17) and -4.0 (Al/Si = 1)
 493 when we parameterize it as a function of Al/(Al+Si) ratio.

494 5.2. Configurational entropy at the glass transition; $S^{\text{conf}}(T_g)$

495 $S^{\text{conf}}(T_g)$ varies in a concave down pattern as a function of Na/K ratio.
 496 This pattern is consistent with a positive contribution of mixing to the con-
 497 figurational entropy. The entropy of mixing is lower than predicted by ideal
 498 mixing (see thin solid lines in Figures 9c and 10c), which represents a random
 499 distribution of Na and K cations in the network. When it is allowed to vary

500 as a function of Al/Si ratio, A_e accommodates most of the variability, and
 501 $S^{conf}(T_g)$ values do not vary substantially between joins (Figure 9c) and are
 502 closer to ideal mixing. Configurational entropy at the glass transition at X_K
 503 = 1 is smaller than at $X_K = 0$ for all joins and for both parameterizations of
 504 A_e (Table A1). When we hold A_e constant at -3.51, most of the variability
 505 in the model is accommodated in $S^{conf}(T_g)$ and melts of higher Al/Si ratio
 506 have the highest values. We see a spread in $S^{conf}(T_g)$ values at $X_K = 0$, and
 507 convergence for all joins at $X_K = 1$.

Figure 11: Configurational entropy at the glass transition in $J\ g\text{-atom}^{-1}K^{-1}$. Left panel: $A_e = -3.51$; Right panel: $A_e = \text{linear function of Al/Si ratio}$. Black dashed line shows model for $X_{Al} = 0.17$, grey dashed line for $X_{Al} = 0.5$.

508 We recalculated $S^{conf}(T_g)$ on the basis of a gram-atom mole (Figure 11)
 509 for a direct comparison between the different joins. For A_e^{AlSi} , patterns re-
 510 main unchanged. For $A_e^{-3.51}$, there is a significant change in the pattern where
 511 $S^{conf}(T_g)$ is highest for *NeKls* and lowest for *Si₅O₁₂* from $X_K = 0$ -0.75, then
 512 the trend reverses. In both parameterizations, the values of $S^{conf}(T_g)$ are still
 513 the lowest at the potassium end-member. Patterns for isothermal configura-
 514 tional entropy are the same as at the glass transition for both parameteriza-
 515 tions of A_e (Figure 12). As expected, $S^{conf}(T = 1800K)$ is higher and shows
 516 a slightly greater spread in values across all joins than $S^{conf}(T = 1300K)$. All
 517 isotherms are concave down, consistent with a positive entropy of mixing and

518 $S^{conf}(T)$ of the potassium end-member is lower than all other compositions
 519 along each join.

Figure 12: Isothermal configurational entropy in $\text{J g-atom}^{-1} \text{K}^{-1}$ calculated at $T = 1300 \text{ K}$ (left panels) or $T = 1800 \text{ K}$ (right panels). Top panels: $A_e = -3.51$; Bottom panels: $A_e = \text{linear function of Al/Si ratio}$.

520 The similar variation in $S^{conf}(T_g)$ across a join (Figure 11) and the small
 521 differences in $S^{conf}(T_g)$ between the different joins imply that Si-Al mixing
 522 contributes little to $S^{conf}(T_g)$, and that most of the configurational entropy
 523 at the glass transition is a result of Na-K mixing. For A_e^{AlSi} , the *NeKls* join

has the smallest entropy of mixing for all K/(Na+K) ratios. This could be explained by a highly ordered melt caused by the higher ratio of alkalis to network-formers, especially with the formation of percolation channels with increasing potassium as described by Le Losq et al. (2017). For $A_e^{-3.51}$, the *NeKls* join has the greatest entropy of mixing up to K/(Na+K) ratios of 0.7. This could be explained by greater Na-K mixing as this join has a greater ratio of alkalis to network-formers, assuming a negligible contribution of Si-Al mixing. $S^{conf}(T_g)$ of the potassium end-member melts seems unaffected by Al/Si ratio and both parameterizations predict very similar values. Si-Al mixing therefore does not seem to provide an additional source of available configurations for the melt at high K content. Both parameterizations of A_e reproduce the viscosity data well, suggesting that chemical mixing is more important than structural changes in viscosity modelling.

The A_e^{AlSi} parameterization reproduces relationships between end-member values of $S^{conf}(T_g)$ when compared to values of residual entropy (S_0) measured independently by calorimetry (Richet, 1984). Indeed, these measurements show that S_0 (reported in J g-atom⁻¹K⁻¹) for nepheline (1.38 ± 0.30) is lower than S_0 values for both albite (2.82 ± 0.46) and orthoclase (2.18 ± 0.46).

Le Losq et al. (2017) obtained a concave down $S^{conf}(T_g)$ pattern for *AbOr* in which configurational entropy decreases with increasing X_K and a concave up pattern for *NeKls*, in which configurational entropy increases with increasing X_K . For each fit, they constrained the value of the sodium end-member to values obtained by calorimetric measurements (Richet, 1984). The concave up pattern they obtain for the *NeKls* join is reflective of a negative configurational entropy of mixing, which would imply mixed Na-K melts have fewer available configurations relative to the end-member melts. This is not reasonable considering the mixing of Na-K results in a viscosity reduction for all joins in the (Na,K)AlSiO₄-SiO₂ systems. In addition, the increasing configurational entropy at the glass transition from the Na to the K end-member melts in *NeKls* calculated by Le Losq et al. (2017) is inconsistent with an increase in viscosity across the join. In both parameterizations of our model, the smallest $S^{conf}(T_g)$ occurs at the potassium end-member, consistent with the higher measured viscosities.

5.3. B_e , $B_e/S^{conf}(T_g)$, and fragility

The B_e parameter follows the same patterns as $S^{conf}(T_g)$ (Figures 9b and 10b). Values for B_e are very similar for both parameterizations of A_e ,

561 with the main difference being that $B_e^{-3.51}$ shows no spread at the potassium
 562 end-member whereas B_e^{AlSi} shows an increase with increasing Al/Si ratio.

563 The ratio $B_e/S^{conf}(T_g)$ is proportional to the height of the average poten-
 564 tial energy barrier to viscous flow (Toplis, 1998). In both parameterizations,
 565 the average potential energy barrier to viscous flow increases with increasing
 566 potassium. However, because $S_{-3.51}^{conf}(T_g)$ shows a greater spread across Al/Si
 567 ratios, the trends as a function of Al/Si ratio for $B_e/S^{conf}(T_g)$ go in opposite
 568 directions: $B_e^{-3.51}/S_{-3.51}^{conf}(T_g)$ decreases with increasing Al/Si ratio (Figure
 569 10d), whereas $B_e^{AlSi}/S_{AlSi}^{conf}(T_g)$ increases (Figure 9d). It follows that, if A_e is
 570 assumed to be constant, the *NeKls* join has the lowest potential energy bar-
 571 rier to viscous flow, whereas the same join has the greatest potential energy
 572 barrier to viscous flow if A_e is a linear function of Al/Si ratio. The patterns
 573 we obtain are similar in shape to those obtained by Neuville and Richet
 574 (1991) for Ca-Mg mixing in pyroxene and garnet melts (shown in Figure 5
 575 of Toplis, 1998). Values for Ab, Jd, and Ne and are within 2-3 KJ mol⁻¹ of
 576 the values obtained by Toplis et al. (1997a).

577 We calculated the fragility (m) of the melts modelled in the (Na,K)AlSiO₄
 578 system following Toplis et al. (1997a):

$$m = \frac{B_e}{T_{12}S_{conf}(T_g)} \left[1 + \frac{C_P^{conf}(T_g)}{S_{conf}(T_g)} \right] \quad (15)$$

579 At constant Al/Si ratio, melt fragility decreases with increasing X_K to
 580 be lowest at $X_K = [0.25-0.75]$, then increases again to its highest at $X_K = 1$.
 581 The most silicic joins show a “bump” in fragility at K/(Na+K) ratio = 0.5
 582 relative to ratios immediately above or below, also seen as a different slope
 583 from the other compositions on the join near the glass transition (Le Losq
 584 and Neuville, 2013). The dependence of m on K/(Na+K) ratio parallels that
 585 of the $B_e/S^{conf}(T_g)$ ratio. As expected, melts with a higher Al/Si ratio are
 586 more fragile (Figure 13).

Figure 13: *Fragility as calculated using Equation 15. Left panel: $A_e = -3.51$; Right panel: $A_e = \text{linear function of Al/Si ratio}$.*

5.4. Implications for viscosity models of mixed aluminosilicate melts

We have presented two different families of models based on two different assumptions about the high-temperature viscosity limit of aluminosilicate melts at very high temperatures. The two different parameterizations show that melt viscosity data in the $SiO_2-(Na,K)AlSiO_4$ system can be fitted well, either by keeping A_e constant and allowing $S^{conf}(T_g)$ to vary as a function of Al/Si (in addition to Na/K) ratio, or by allowing A_e to vary as a linear function of Al/Si ratio, in which case $S^{conf}(T_g)$ has only a small dependence on Al/Si ratio.

These global models have a standard error, defined as:

$$Se = \frac{\sum(\eta_{obs} - \eta_{calculated})^2}{(n - f)} = \frac{n(RMSD)^2}{(n - f)}, \quad (16)$$

where n is the number of data and f the number of fit parameters, of 0.33 for the $A_e^{-3.51}$ parameterization (12 adjustable parameters) and of 0.27 for the A_e^{AlSi} (13 adjustable parameters). Allowing A_e to vary linearly as a function of Al/Si ratio therefore clearly provides a better fit than using a constant $A_e = -3.51$ in the $SiO_2-(Na,K)AlSiO_4$ system, with only one additional adjustable parameter.

Figure 14: Model parameters as a function of $Al/(Al+Si)$ ratio. Left panels: $A_e = -3.51$; Right panels: $A_e = \text{linear function of } Al/Si \text{ ratio}$. Top: B_e , middle: $S^{conf}(T_g)$, and bottom: $10^{-3}B_e/S^{conf}(T_g)$.

When A_e varies as a linear function of Al/(Al+Si) ratio, there is a spread in B_e as a function of Al/(Al+Si) ratio and only a very small dependence of $S^{conf}(T_g)$ on Al/(Al+Si) ratio (~ 0.4 J g-atom $^{-1}$ K $^{-1}$ across all Al/(Al+Si) ratios, for all K/(Na+K) ratios; Figures 9, 11, 14). B_e increases linearly with increasing Al/(Al+Si) ratio; B_e increases at a faster rate for high potassium melts than for high sodium melts (Figures 9b, 14b). We observe that $S^{conf}(T_g)$ decreases with increasing Al/(Al+Si) ratio for all K/(Na+K) ratios, with a shallower slope for compositions with Al/(Al+Si) ratio above 0.5 (Figure 14d). The ratio of $B_e/S^{conf}(T_g)$ increases quasi-linearly with increasing Al/(Al+Si) ratio for all K/(Na+K) ratios; melts with the highest K/(Na+K) ratios see the most dramatic increase. This may be a result of the larger size of the K $^{+}$ cation compared to the Na $^{+}$ cation. Indeed, for alkali tetrasilicates (M $_2$ Si $_4$ O $_9$), $B_e/S^{conf}(T_g)$ increases linearly with increasing cation radius (Toplis, 1998).

An increase in the height of the energy barrier to allow configurational changes with increasing Al/(Al+Si) ratio should make configurational changes more difficult. Based solely on this, viscosity may be expected to increase with increasing Al/(Al+Si) ratio. However, our observations are that with increasing Al/(Al+Si) ratio, viscosity decreases, and that with increasing K/(Na+K) ratio, viscosity increases. Le Losq et al. (2017) show that as K replaces Na in a melt at constant Al/(Al+Si) ratio, it causes the size of tetrahedral cages and rings to increase. This shortens and strengthens the Al-O and Si-O bonds and causes an increase of Al-Al and Al-Si distances. They also show that alkali (Na or K) cluster size increases with increasing Al/(Al+Si) ratio, causing an increase in Al-O-Al linkages. Their molecular dynamics simulations predict Al-O-Al (and therefore Si-O-Si) linkages on the order of 18% for the *NeKls* join (consistent with NMR studies, e.g., Dubinsky and Stebbins, 2006), and on the order of 3% for the *AbOr* join. Percolation channels in the melt are observed only at high Al/(Al+Si) ratio for high K/(Na+K). Le Losq et al. (2017) argue that the segregation of alkali into clusters or channels reduces the mobility of Si, Al, and O, increasing the viscosity of the melts. The model trends in $S^{conf}(T_g)$ and $B_e/S^{conf}(T_g)$ as a function of Al/(Al+Si) ratio may therefore be consistent with an increasing size of the smallest rearranging unit in the melt, z^* , caused by alkali clustering at high Al/(Al+Si) ratio, and an associated increase in the height of the energy barrier to configurational changes, $\Delta\mu$. In this parameterization of our global model, the lower viscosity of the *NeKls* join compared to all other joins in the system is explained by A_e being smaller (more negative) for melts

641 with higher Al/(Al+Si) ratio, reflecting the lower overall bond strength of
642 high Al/(Al+Si) melts because of their greater proportion of Al-O bonds.

643 6. Summary

644 The global configurational entropy model for viscosity where A_e is mod-
645 elled to vary as a linear function of Al/(Al+Si) ratio has a lower RMSD
646 and lower standard error than when A_e is modelled as a constant (Table 7).
647 The better fit obtained by allowing A_e to vary as a function of Al/(Al+Si)
648 ratio is clearly demonstrated in Figure 8. The variable A_e model is in bet-
649 ter agreement with measurements of residual entropy obtained by calorime-
650 try on congruently melting nepheline, albite, and orthoclase (Richet, 1984),
651 with nepheline having a lower residual entropy than SiO₂. We argue that
652 the parameters obtained with the variable A_e model are consistent with re-
653 cent structural data obtained on nominally fully polymerized, mixed Na-K
654 aluminosilicate melts (Le Losq et al., 2017), with the implication that the
655 varying A_e accounts for the overall bond strength of the network in the SiO₂-
656 (Na,K)AlSiO₄ system.

657 We conclude that the viscosity of nominally fully-polymerized melts in
658 the SiO₂-(Na,K)AlSiO₄ system can be modelled through chemical mixing,
659 without explicit consideration of the important changes in structure related
660 to changes in K/(Na+K) and Al/(Al+Si) ratios. Most of the configurational
661 entropy at the glass transition is a result of Na-K mixing; Si-Al mixing pro-
662 vides little additional contribution.

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7. Appendix

Table A1: Fit results for the global configurational entropy model

Suite	$\frac{\Delta H}{RT}$	$\frac{K}{Na+K}$	$A_e = -3.51$										$A_e = \text{AlSi}$									
			$A_e^{-3.51}$ (log Pa s)	$10^{-3}B_e^{-3.51}$ (KJ mol ⁻¹)	$S^{conf}(T_g)^{-3.51}$ (J g atom ⁻¹ K ⁻¹)	$S^{conf}(T_g)^{-3.51}$ (J g atom ⁻¹ K ⁻¹)	$\frac{10^{-3}B_e^{-3.51}}{S^{conf}(T_g)^{-3.51}}$ (K)	RMSD Individual	RMSD Join	A_e^{AlSi} (log Pa s)	$10^{-3}B_e^{\text{AlSi}}$ (KJ mol ⁻¹)	$S^{conf}(T_g)^{\text{AlSi}}$ (J g atom ⁻¹ K ⁻¹)	$S^{conf}(T_g)^{\text{AlSi}}$ (J g atom ⁻¹ K ⁻¹)	$\frac{10^{-3}B_e^{\text{AlSi}}}{S^{conf}(T_g)^{\text{AlSi}}}$ (K)	RMSD Individual	RMSD Join						
Si ₅ O ₁₂	0.17	0.00	-3.51	175.2	9.8	3.1	17.8	0.23	0.39	-2.41	169.6	10.6	3.3	16.0	0.27	0.38						
Si ₅ O ₁₂	0.17	0.25	-3.51	213.8	12.4	3.9	17.3	0.57		-2.38	221.6	13.9	4.4	15.9	0.55							
Si ₅ O ₁₂	0.17	0.50	-3.51	227.7	12.6	4.0	18.0	0.22		-2.39	238.6	14.4	4.5	16.6	0.14							
Si ₅ O ₁₂	0.17	0.73	-3.51	217.4	12.1	3.8	18.0	0.56		-2.39	222.1	13.4	4.2	16.6	0.50							
Si ₅ O ₁₂	0.17	0.99	-3.51	181.2	9.6	3.0	19.0	0.28		-2.41	170.1	9.8	3.1	17.4	0.40							
Na-petalite T97	0.18	0.00	-3.51	176.1	9.9	3.1	17.8	0.75		-2.45	169.9	10.6	3.3	16.1	0.29							
K-petalite	0.22	0.99	-3.51	179.7	9.4	2.9	19.2	0.23		-2.67	171.6	9.5	3.0	18.0	0.18							
Ab T97	0.25	0.00	-3.51	183.9	10.6	3.3	17.3	0.40		-2.78	171.7	10.5	3.2	16.3	0.16							
Ab SS93	0.27	0.00	-3.51	186.5	10.9	3.3	17.2	0.10		-2.89	172.3	10.5	3.2	16.3	0.15							
AbOr	0.25	0.00	-3.51	184.3	10.7	3.3	17.3	0.28	0.31	-2.79	171.8	10.5	3.2	16.3	0.15	0.25						
AbOr	0.25	0.20	-3.51	215.7	12.9	4.0	16.8	0.29		-2.79	216.6	13.6	4.2	15.9	0.21							
AbOr	0.25	0.39	-3.51	231.4	13.2	4.1	17.5	0.14		-2.78	239.8	14.4	4.4	16.6	0.05							
AbOr	0.25	0.50	-3.51	233.3	13.1	4.0	17.8	0.36		-2.79	243.1	14.4	4.4	16.9	0.35							
AbOr	0.25	0.59	-3.51	221.0	12.9	4.0	17.9	0.14		-2.77	240.7	14.2	4.4	16.9	0.06							
AbOr	0.25	0.79	-3.51	214.8	12.1	3.7	17.8	0.55		-2.79	219.9	13.0	4.0	16.9	0.51							
AbOr	0.25	1.00	-3.51	178.6	9.1	2.8	19.5	0.34		-2.81	172.2	9.3	2.9	18.5	0.26							
Or	0.26	1.00	-3.51	179.4	9.3	2.9	19.3	0.25		-2.84	173.7	9.5	2.9	18.3	0.19							
Ab Urban	0.25	0.00	-3.51	184.2	10.6	3.3	17.3	0.12		-2.79	171.7	10.5	3.2	16.3	0.19							
Or Urban	0.25	1.00	-3.51	178.6	9.1	2.8	19.5	0.32		-2.79	171.9	9.3	2.9	18.5	0.69							
Jd SS93	0.32	0.00	-3.51	192.7	11.4	3.4	16.9	0.18		-3.15	173.7	10.5	3.2	16.5	0.17							
Jd SS93	0.34	0.00	-3.51	195.0	11.6	3.5	16.8	0.16		-3.25	174.2	10.5	3.1	16.6	0.12							
Jd T97	0.33	0.00	-3.51	193.9	11.5	3.5	16.8	0.31		-3.20	174.0	10.5	3.2	16.5	0.31							
JdLet	0.35	0.00	-3.51	196.1	11.8	3.5	16.7	0.34	0.29	-3.28	174.8	10.6	3.2	16.5	0.37							
JdLet	0.35	0.26	-3.51	231.6	13.9	4.1	16.7	0.22		-3.28	230.3	14.0	4.2	16.5	0.16							
JdLet	0.34	0.38	-3.51	238.2	13.8	4.1	17.3	0.32		-3.22	243.6	14.4	4.3	16.9	0.25							
JdLet	0.35	0.51	-3.51	239.8	13.7	4.1	17.6	0.32		-3.27	248.7	14.4	4.3	17.2	0.33							
JdLet	0.33	0.63	-3.51	233.8	13.2	4.0	17.7	0.25		-3.21	243.8	14.1	4.2	17.3	0.12							
JdLet	0.35	0.76	-3.51	221.5	12.6	3.7	17.6	0.41		-3.31	230.7	13.3	4.0	17.3	0.30							
JdLet	0.35	1.00	-3.51	180.1	9.5	2.8	19.0	0.16		-3.29	181.0	9.6	2.9	18.8	0.12							
60:50 T97	0.40	0.00	-3.51	202.1	12.3	3.6	16.5	0.23		-3.55	175.9	10.5	3.1	16.8	0.28							
Si ₁₄ O ₁₇	0.43	0.00	-3.51	204.7	12.5	3.6	16.4	0.16	0.31	-3.66	176.5	10.5	3.1	16.8	0.03							
Si ₁₄ O ₁₇	0.42	0.24	-3.51	236.8	14.4	4.2	16.4	0.31		-3.64	230.4	13.9	4.0	16.6	0.29							
Si ₁₄ O ₁₇	0.42	0.47	-3.51	245.6	14.2	4.1	17.3	0.19		-3.64	252.4	14.5	4.2	17.4	0.17							
Si ₁₄ O ₁₇	0.42	0.73	-3.51	227.6	13.0	3.8	17.5	0.31		-3.64	239.1	13.6	4.0	17.6	0.32							
Si ₁₄ O ₁₇	0.42	0.99	-3.51	181.1	9.6	2.8	18.8	0.50		-3.62	187.0	9.8	2.9	19.1	0.39							
Ne T97	0.51	0.00	-3.51	214.3	13.4	3.8	16.0	0.59		-4.06	178.7	10.5	3.0	17.1	0.10							
NeKls	0.52	0.00	-3.51	215.8	13.5	3.8	16.0	0.50	0.37	-4.12	179.3	10.5	3.0	17.1	0.15	0.19						
NeKls	0.52	0.26	-3.51	247.6	15.2	4.3	16.3	0.34		-4.11	237.9	14.0	4.0	17.0	0.12							
NeKls	0.51	0.38	-3.51	251.8	15.0	4.3	16.8	0.39		-4.05	251.6	14.4	4.1	17.4	0.19							
NeKls	0.52	0.51	-3.51	251.4	14.6	4.2	17.2	0.69		-4.12	258.5	14.5	4.1	17.8	0.20							
NeKls	0.50	0.63	-3.51	242.6	14.0	4.0	17.4	0.32		-4.04	254.2	14.2	4.0	17.9	0.20							
NeKls	0.52	0.75	-3.51	228.9	13.2	3.7	17.4	0.38		-4.12	243.2	13.5	3.8	18.0	0.37							
NeKls LN	0.51	0.00	-3.51	214.1	13.3	3.8	16.0	0.41	0.25	-4.05	178.7	10.5	3.0	17.1	0.14	0.18						
NeKls LN	0.52	0.26	-3.51	247.6	15.2	4.3	16.3	0.22		-4.11	237.9	14.0	4.0	17.0	0.19							
NeKls LN	0.50	0.50	-3.51	250.4	14.6	4.2	17.2	0.15		-4.02	257.1	14.5	4.1	17.7	0.08							
NeKls LN	0.50	0.78	-3.51	224.5	12.9	3.7	17.4	0.07		-4.04	238.5	13.3	3.8	18.0	0.10							
NeKls LN	0.50	0.99	-3.51	180.9	9.6	2.8	18.8	0.17		-4.01	192.2	9.8	2.8	19.6	0.37							
Ne Ndala	0.50	0.00	-3.51	213.4	13.3	3.8	16.1	0.15		-4.02	178.5	10.5	3.0	17.1	0.51							
Kls Ndala	0.50	1.00	-3.51	179.6	9.4	2.7	19.1	0.04		-4.02	191.0	9.5	2.7	20.0	0.19							
HPGS	0.16	0.38	-3.51	223.3	12.6	4.0	17.8	0.32		-2.34	234.1	14.4	4.6	16.3	0.42							