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Fluorine and the viscosity of jadeite-leucite and nepheline-kalsilite melts at atmospheric pressure

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MR43A-0463: Fluorine and the Viscosity of Jadeite-Leucite and Nepheline-Kalsilite Melts at Atmospheric Pressure

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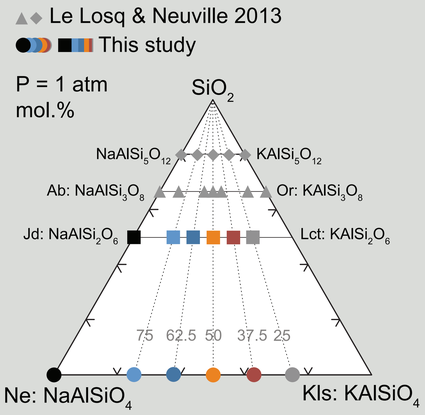
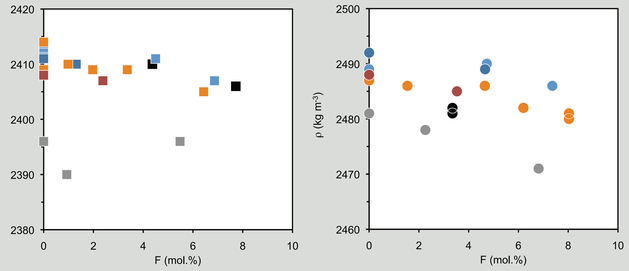
Samples, Goals, and Methods



Sample	F wt.%	F mol%	F_2O_{-1} mol%	NBO/T	Al/(Al+Si)	Na/(Na+K)	n
Jd25+0F	0.00	0.00	0.00	-0.01	0.35	0.24	5
Jd25+2F	0.25	0.93	0.46	0.00	0.34	0.25	20
Jd25+4F	1.52	5.48	2.74	0.00	0.34	0.25	22
Jd37.5+0F	0.00	0.00	0.00	0.01	0.33	0.37	25
Jd37.5+2F	0.65	2.38	1.19	0.01	0.33	0.38	19
Jd50+0F	0.00	0.00	0.00	0.00	0.35	0.49	10
Jd50+1F	0.27	0.98	0.49	-0.01	0.34	0.51	20
Jd50+2F	0.54	1.97	0.99	0.00	0.34	0.50	20
Jd50+2F post	0.52	1.91	0.95	0.00	0.34	0.50	25
Jd50+3F	0.93	3.36	1.68	-0.01	0.34	0.51	20
Jd50+4F	1.83	6.43	3.21	0.00	0.34	0.50	20
Jd62.5+0F	0.01	0.05	0.02	0.00	0.34	0.62	18
Jd62.5+2F	0.36	1.31	0.66	0.00	0.34	0.60	20
Jd75+0F	0.00	0.00	0.00	-0.01	0.35	0.74	11
Jd75+2F	1.28	4.50	2.25	0.00	0.34	0.75	16
Jd75+4F	1.99	6.87	3.43	-0.01	0.34	0.75	22
Jd100+0F	0.00	0.00	0.00	-0.01	0.35	1.00	12
Jd100+2F	1.27	4.37	2.18	-0.01	0.34	1.00	20
Jd100+4F	2.31	7.72	3.86	-0.01	0.33	1.00	22
Ne25+0F	0.00	0.00	0.00	-0.02	0.52	0.25	17
Ne25+2F	0.57	2.26	1.13	-0.02	0.51	0.25	20
Ne25+4F	1.75	6.81	3.41	-0.01	0.50	0.26	25
Ne37.5+0F	0.00	0.00	0.00	0.00	0.50	0.37	30
Ne37.5+2F	0.90	3.53	1.76	-0.02	0.50	0.38	30
Ne50+0F	0.00	0.00	0.00	-0.01	0.52	0.49	16
Ne50+1F	0.39	1.54	0.77	0.00	0.50	0.50	21
Ne50+2F	1.22	4.65	2.32	-0.01	0.51	0.51	17
Ne50+3F	1.64	6.20	3.10	-0.02	0.51	0.51	21
Ne62.5+0F	0.00	0.01	0.00	0.00	0.51	0.62	20
Ne2.5+2F	1.24	4.66	2.33	-0.01	0.50	0.62	30
Ne75+0F	0.00	0.00	0.00	-0.01	0.52	0.74	17
Ne75+2F	1.27	4.73	2.36	-0.01	0.51	0.75	18
Ne75+4F	2.03	7.36	3.68	0.01	0.50	0.76	25
Ne100+0F	0.00	0.00	0.00	-0.02	0.52	1.00	18
Ne100+2F	0.91	3.35	1.67	-0.03	0.51	1.00	16



RESEARCH QUESTIONS:

1. What is the effect of Na-K mixing, and
2. What is the effect of dissolved fluorine

on the viscosity of melts with compositions along the $NaAlSi_2O_6$-$KAlSi_2O_6$ (jadeite-leucite) and $NaAlSiO_4$-$KAlSiO_4$ (nepheline-kalsilite) joins of the quartz-nepheline-kalsilite system?

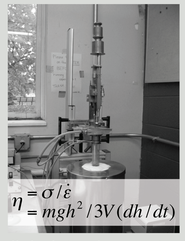
WHY THIS SYSTEM?

All melts nominally fully polymerized (NBO/T=0), yet:

a. Have different Al/Si ratios
b. Have different Na/K ratios

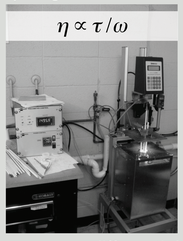
F bonds with Al (Zeng & Stebbins, 2000; Mysen et al., 2004; Schaller et al., 1992) and depolymerizes the melts. Is this effect Al/Si or Na/K dependent?

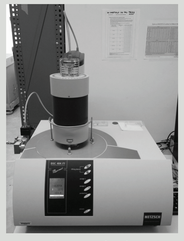
synthesis of glasses
(T = 1600-1745°C)



parallel-plate viscometer
max T = 1100°C
η range 10^9-10^{12} Pa s

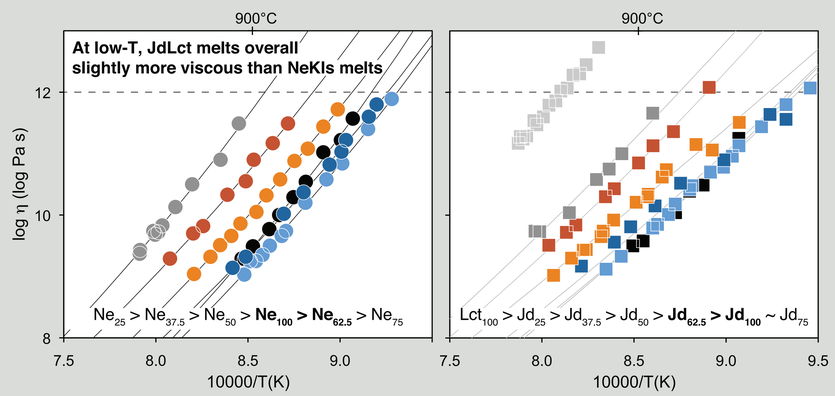
concentric-cylinder viscometer
max T = 1600°C
η range ~ 10^{-1}-10^5 Pa s

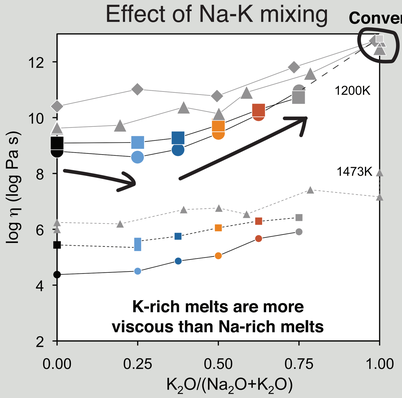


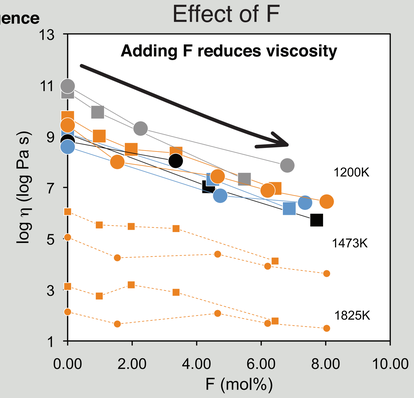


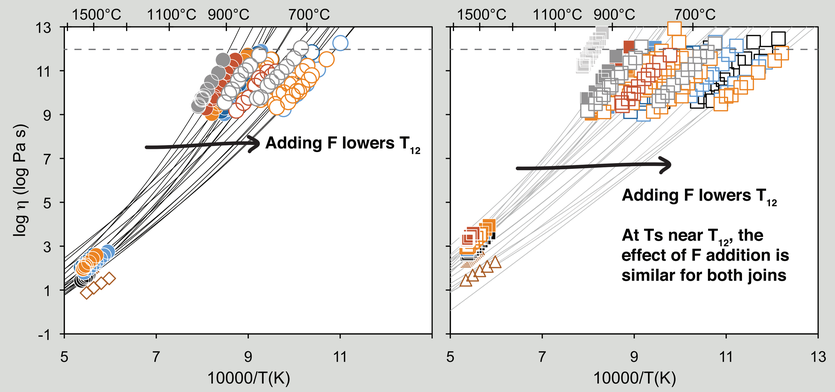
differential scanning calorimeter
max T = 1500°C
heating rate = 20K/min
(future work)

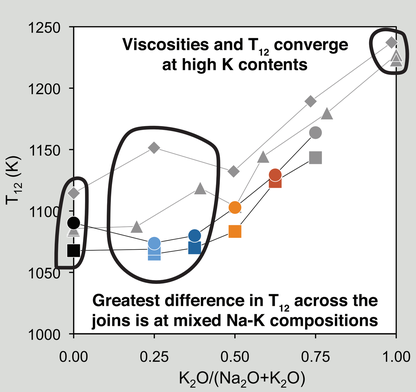
Results and Discussion

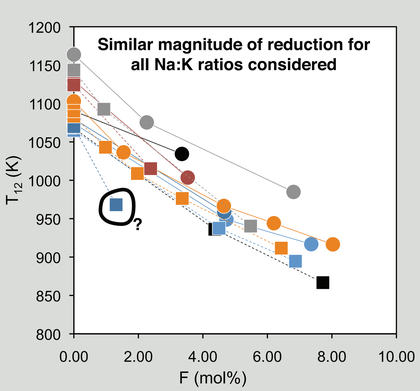


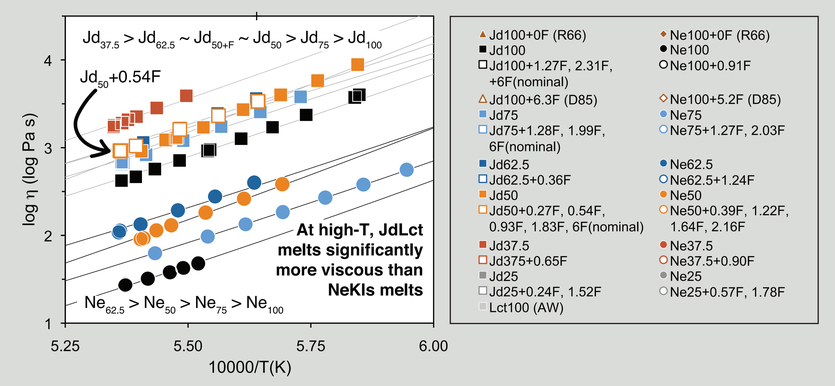


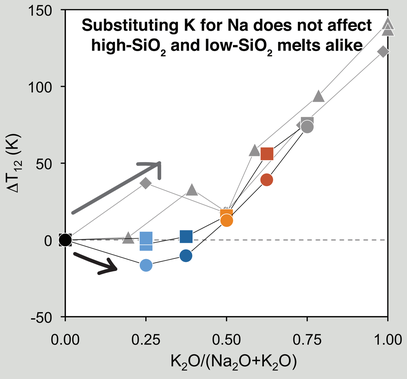


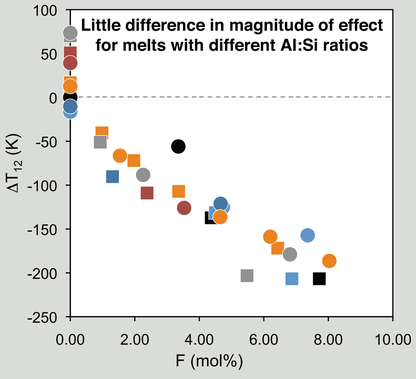












High-T results are consistent with those of Riebling (1966; R66 in legend). NeKls results in perfect agreement with those of Le Losq et al. 2017 (not shown for clarity). Data for F-free leucite from Whittington (Lct100 AW). Preliminary viscosity data on F-bearing melts at high temperature suggest F does not affect viscosity or increases viscosity marginally (Jd 50+0F vs. Jd50+0.54F in bottom left figure; closed and open orange squares; 0.02 wt.% F loss during high-T measurement).

The lack of marked difference in the magnitude of viscosity/T_{12} reduction due to the addition of F as a function of either Al/Si or Na/K ratio suggests that the mechanisms of F dissolution are independent of the alkali present or of the Al/Si ratio in melts with nominal NBO/T of 0. This is interesting in light of recently published results by Le Losq et al. (2017), who show via MD simulation and Raman spectroscopy that important structural changes occur in melts with high Al/Si ratio as K^+ replaces Na^+.

Works Cited

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