

Supporting Information

Why does B_2O_3 suppress nepheline ($NaAlSiO_4$) crystallization in
aluminosilicate glasses?

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Table S1. Viscosities of melts of BL, SB-10, SB-20, BA-10 and BA-20 compositions as a function of temperature.

BL		SB-10		SB-20		BA-10		BA-20	
T (K)	$\log_{10}\eta$ (Pa.s)	T (K)	$\log_{10}\eta$ (Pa.s)	T (K)	$\log_{10}\eta$ (Pa.s)	T (K)	$\log_{10}\eta$ (Pa.s)	T (K)	$\log_{10}\eta$ (Pa.s)
1927.1	1.078	1863.1	0.561	1617.4	0.866	1670.4	1.1157	1573.0	0.139
1927.1	1.078	1769.1	0.937	1580.2	1.049	1671.5	1.135	1534.7	0.266
1907.7	1.161	1769.7	0.939	1580.3	1.0492	1632.9	1.2353	1495.1	0.341
1907.7	1.161	1721.4	1.147	1541.9	1.204	1632.9	1.2396	1455.6	0.469
1888.3	1.249	1721.6	1.146	1541.6	1.209	1593.8	1.3712	1416.0	0.584
1888.3	1.248	1673.0	1.382	1503.0	1.394	1593.8	1.3822	1336.4	0.868
1863.2	1.362	1673.1	1.386	1503.0	1.396	1551.3	1.5089	1296.7	1.049
1863.2	1.359	1624.	1.601	1463.8	1.568	1551.4	1.51	1296.7	1.043
1843.3	1.455	1624.7	1.604	1463.7	1.566	1511.1	1.6554	1256.7	1.143
1843.3	1.452	1576.7	1.864	1424.3	1.770	1510.9	1.6568	1256.7	1.144
1823.5	1.551	1576.7	1.863	1424.2	1.767	1470.6	1.8077	1216.7	1.486
1823.5	1.551	1537.3	2.109	1385.0	1.997	1470.7	1.8081	1216.7	1.478
1803.5	1.655	1537.3	2.107	1384.9	1.999	1430.8	1.9707	1176.6	1.767
1803.5	1.657	1476.1	2.552	1345.4	2.239	1430.7	1.9701	1176.5	1.762
1783.9	2.150 [†]	1478.1	2.548	1345.4	2.240	1391.4	2.1389	808.1	11.300
1101.3	11.296	927.5	11.121	1305.9	2.497	1391.3	2.1359	805.5	11.493
1095.2	11.500	922.8	11.294	1305.7	2.504	1350.1	2.3016	799.6	11.910
1089.2	11.705	917.1	11.498	1275.6	2.701	1350.1	2.2986	798.1	11.999
1084.0	11.909	911.4	11.699	1278.5	2.713	822.4	11.195	807.7	11.319
1080.6	12.006	905.4	11.907	834.5	11.199	818.9	11.395	805.1	11.508
1103.9	11.198	903.1	11.998	829.9	11.395	815.0	11.617	802.4	11.699
1098.0	11.396	925.6	11.196	824.8	11.605	811.6	11.808	799.5	11.909
1091.7	11.607	920.0	11.394	820.2	11.807	808.2	12.002		
1086.6	11.807	914.4	11.594	832.0	11.302	822.1	11.198		
		908.5	11.802	827.9	11.492	818.9	11.394		
		903.0	12.01	822.9	11.703	814.7	11.616		
				818.5	11.912	811.3	11.806		
				816.2	11.999	807.4	12.015		

[†] In case of BL, the viscosity at 1783.9 K was not found to gradually increase with time instead of stabilizing, indicating crystallization

Table S2. Viscosity of melts with NaAlSiO₄ or near-NaAlSiO₄ compositions obtained from literature

NaAlSiO ₄ from Le Losq et al. ¹		Toplis et al. NAS50:49 ²		Toplis et al. NAS50:51 ²	
T, K	$\log_{10}\eta$ (Pa.s)	T, K	$\log_{10}\eta$ (Pa.s)	T, K	$\log_{10}\eta$ (Pa.s)
1058.9	12.57	1164.4	9.46	1162.9	8.99
1072.5	12.04	1147.4	9.98	1138.2	9.57
1085.4	11.59	1125.0	10.72	1120.4	10.05
1093.2	11.33	1117.9	10.78	1102.5	10.73
1102.1	11.09	1100.9	11.4	1919.1	1.117
1110.7	10.79	1919.1	1.118	1894.1	1.226
1121.1	10.5	1894.1	1.229	1869.1	1.341
1134.3	10.12	1869.1	1.345	1845.1	1.456
1150.7	9.72	1845.1	1.46	1820.1	1.579
1172.5	9.26	1820.1	1.586		

Table S3. ^{23}Na and ^{27}Al chemical shift (δ_{CS}), quadrupolar coupling product (P_{Q}) and quadrupolar coupling constant (C_{Q}) parameters

	^{23}Na MAS NMR		^{27}Al MAS NMR	
	δ_{CS} (ppm)	P_{Q} (MHz)	δ_{CS} (ppm)	C_{Q} (MHz)
BL	-4.1	1.75	65.8	5.4
SB-10	-4.4	1.67	65.8	5.4
SB-20	-4.2	1.53	64.7	5.2
BA-10	-3.8	1.75	63.7	4.7
BA-20	-3.5	1.90	63.1	4.8

Table S4. ^{23}Na chemical shifts (δ_{CS}) and quadrupolar coupling product (P_{Q}) parameters from MQMAS NMR data

	Crystalline sites			Residual glass	
		δ_{CS} (ppm)	P_{Q} (MHz)	δ_{CS} (ppm)	P_{Q} (MHz)
BL 950C 24h	Site 1	11.0	1.9	--	--
	Site 2	6.3	2.3		
	Site 3	5.4	2.1		
	Site 4	-1.8	1.5		
SB-10 950C 24h	Site 1	6.3	1.8	--	--
	Site 2	5.3	1.6		
	Site 3	4.3	1.6		
	Site 4	-1.4	1.5		
	Site 5	-3.4	1.5		
	Site 6	-11.6	0.7		
	Site 7	-13.3	0.8		
SB-20 950C 24h	Site 1	5.9	1.6	--	--
	Site 2	0.1	1.5		
	Site 3	-1.7	1.5		
	Site 4	-9.9	0.7		
	Site 5	-11.6	0.8		
BL 850C 24h	Site 1	5.7	2.06	-3.7	1.70
	Site 2	-2.2	1.77		
BA-10 850C 24h	Site 1	-5.4	2.01	-3.3	1.12
	Site 2	-19.2	1.53		
BA-20 850C 24h		--	--	-3.5	1.99

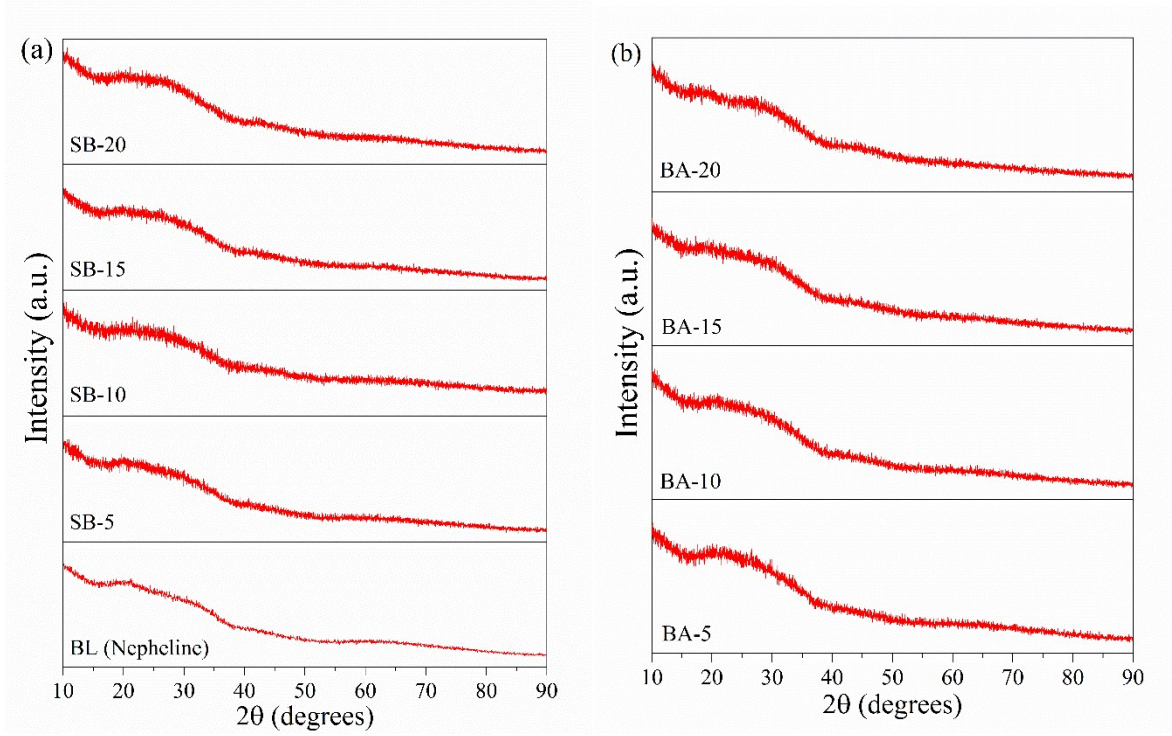


Figure S1. X-ray diffractograms of glasses of (a) SB-series and (b) BA-series

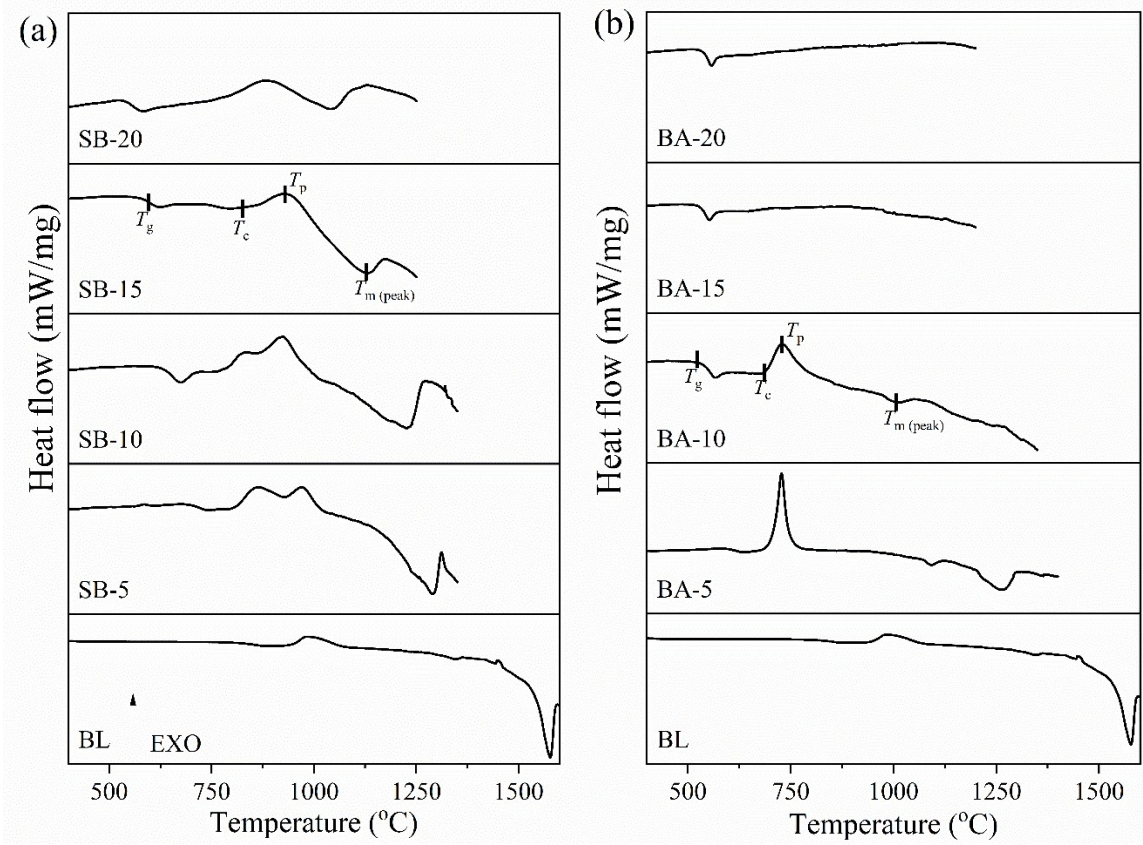


Figure S2. DSC scans of glasses of (a) SB-series and (b) BA-series

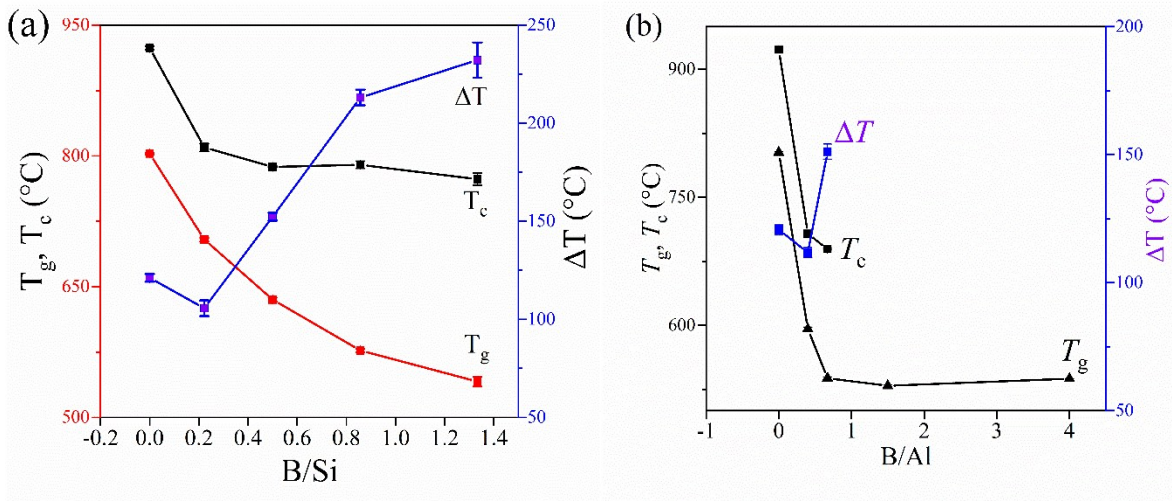


Figure S3. T_g , T_c and ΔT values vs. mol.% B_2O_3 for (a) SB-series and (b) BA-series glasses obtained from thermal analysis at 10 K/min.

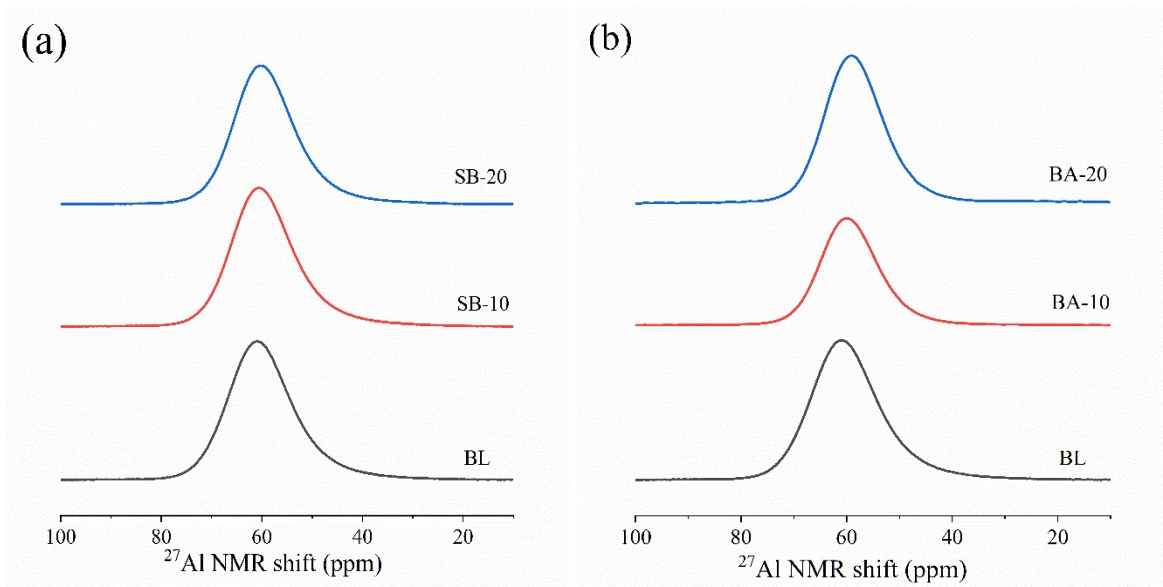


Figure S4. ^{27}Al MAS NMR spectra (a) SB-series and (b) BA-series glasses

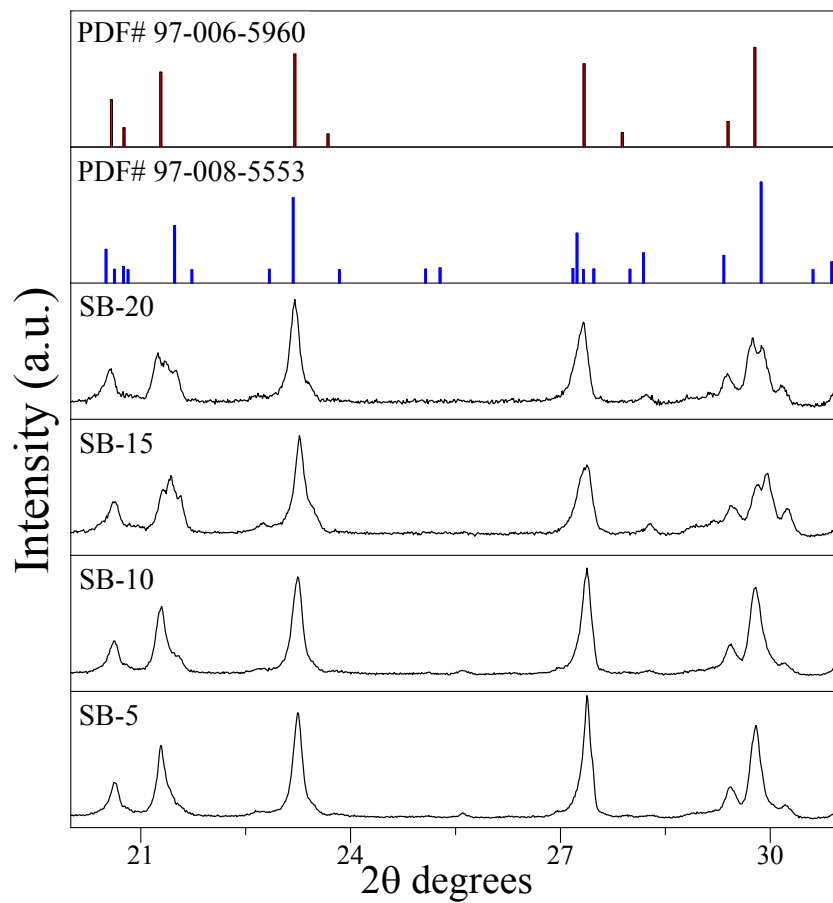


Figure S5. XRD diffractograms of isothermally heat-treated SB-series glass-ceramics and comparison with different crystal phases of nepheline - PDF#97-008-5553 and PDF# 97-006-5960 within the 2θ range of 19° to 31°

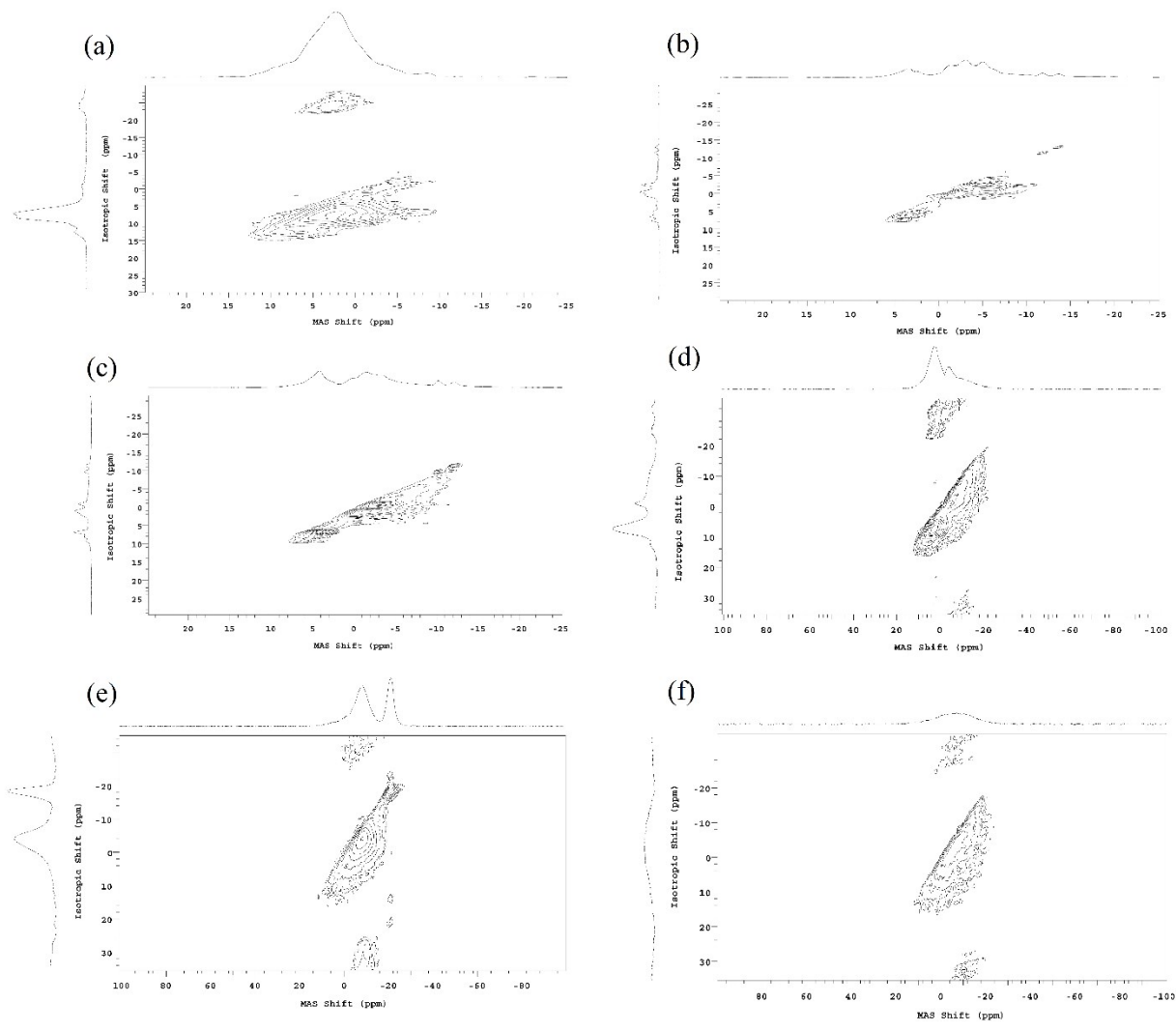


Figure S6. 2D ^{23}Na MQMAS NMR spectra of isothermally heat-treated glass-ceramics- (a)BL 950 °C 24h, (b) SB-10 950 °C 24h, (c) SB-20 950 °C 24h, (d) BL 850 °C 24h (e) BA-10 850 °C 24h and (f) BA-20 850 °C 24h

References

1. C. Le Losq, D. R. Neuville, W. Chen, P. Florian, D. Massiot, Z. Zhou, and G. N. Greaves, *Sci. Rep.*, 2017, **7**, 16490.
2. M. J. Toplis, D. B. Dingwell, K.-U. Hess, and T. Lenci, *Am. Mineral.*, 1997, **82**, 979-90.