Supporting Information

for

Annealing Induced Structural Evolution in Feldspar Dental Glass-Ceramics Investigated by Solid-State NMR Spectroscopy

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Table S1. Cell parameters for commercial and lab-grade dental ceramics.[a]

Sample	Annealing temp (°C)	a (Å)	b (Å)	c (Å)	α (°)	β (°)	γ (°)	V (Å^3)
Commercial	800	8.412(1)	12.760(2)	7.091(1)	89.90(1)	116.21(1)	89.18(8)	682.8(2)
Commercial	850	8.417(1)	12.773(2)	7.102(2)	89.90(1)	116.17(1)	89.21(1)	685.2(2)
Commercial	900	8.457(1)	12.817(2)	7.130(1)	89.94(1)	116.2(1)	89.15(1)	693.3(2)
Commercial	950	8.508(2)	12.975(2)	7.138(1)	90.16(2)	115.94(1)	89.84(2)	708.6(3)
Lab-grade	900	8.334(1)	12.828(1)	7.095(1)	89.94(1)	116.14(1)	89.69(1)	680.9(2)
Ga-doped	900	8.413(1)	12.896(1)	7.083(1)	89.96(1)	115.95(1)	89.74(1)	690.9(1)
B-doped	900	8.435(2)	12.893(2)	7.093(1)	90.29(2)	115.99(1)	89.80(2)	693.4(2)

[a] Refined in triclinic (pseudo-monoclinic) lattices. The standard uncertainties are shown in parentheses.

Table S2. Amorphous and crystalline phase fractions obtained by fitting ^{23}Na NMR spectra of commercial dental ceramics annealed at various temperatures.[a]

Annealing temperature (°C)	Amorphous (%)	Crystalline (%)
800	82	18
850	71	29
900	87	13
950	98	2

[a] Relative errors are 3%.

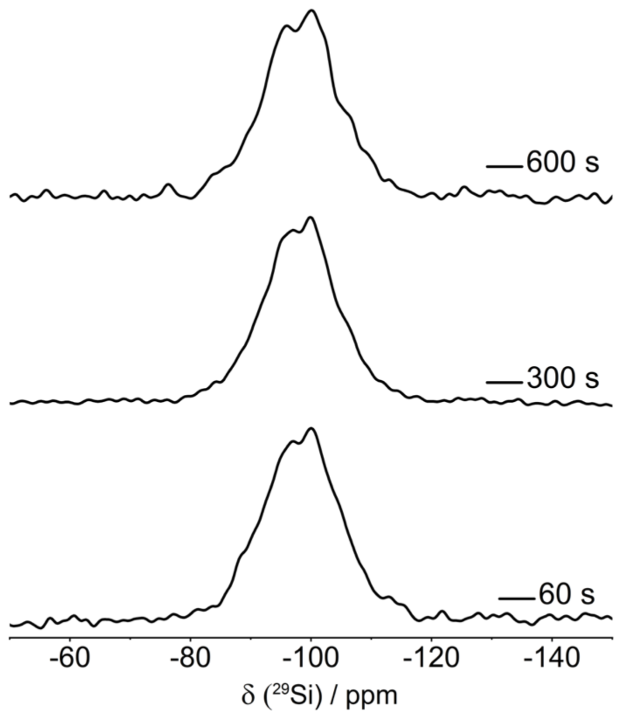

Figure S1. Silicon-29 MAS NMR spectra (B_0 = 7.05 T; $\omega_r/2\pi$ = 5 kHz; short tip angle of ~10°) of VITA VM9 dental ceramic (annealed at 900 °C) with varying recycle delays.

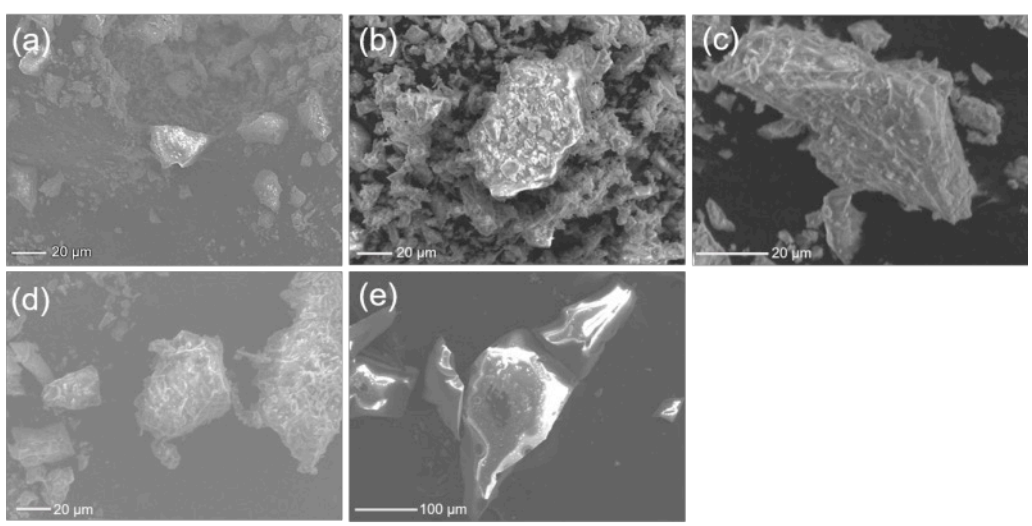

Figure S2. SEM images of the commercial dental ceramics annealed at different temperatures (a) RT, (b) 600 °C, (c) 800 °C, (d) 900 °C and (e) 1100 °C.

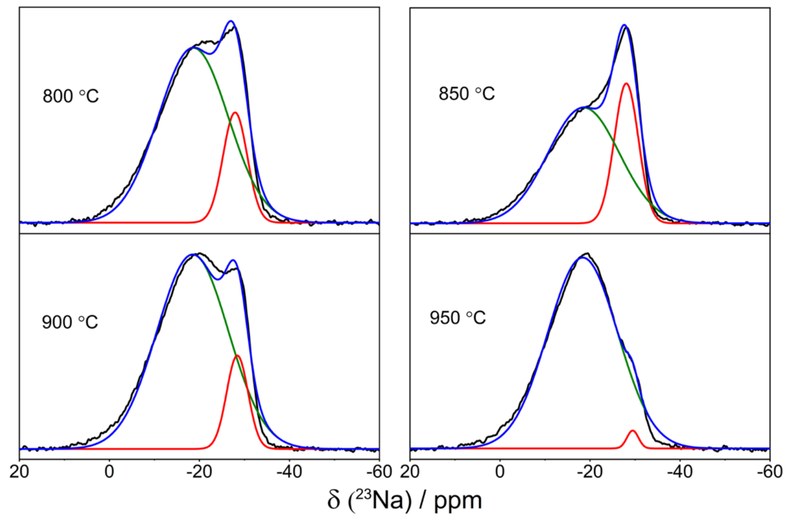

Figure S3. Sodium-23 MAS NMR spectra (B_0 = 21.1 T; $\omega_r/2\pi$ = 31.25 kHz) of the VITA VM9 dental ceramic with annealing temperatures between 800 and 950 °C. The central transition was fit using a Gaussian lineshape considering amorphous (Green) and crystalline (Red) contributions. The cumulative fit is shown in blue.

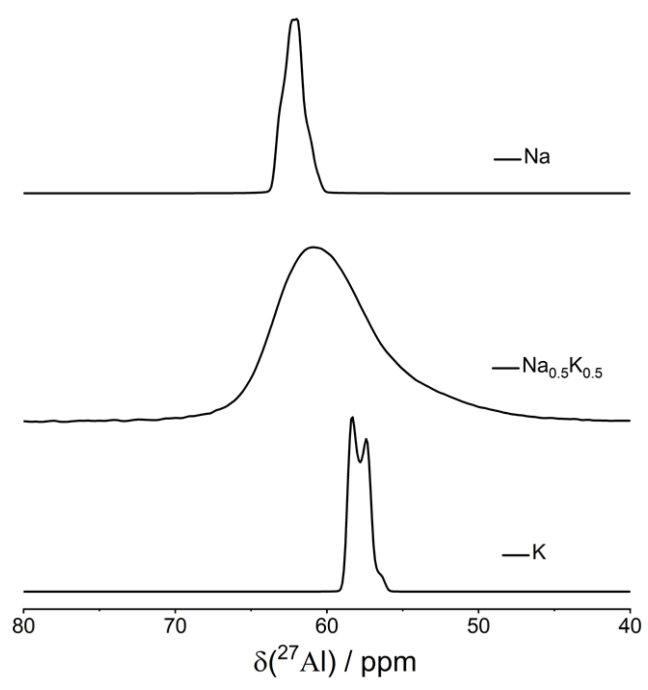

Figure S4. Comparison of the experimental ^{27}Al NMR spectrum for $Na_{0.5}K_{0.5}AlSi_3O_8$ (middle) with simulated spectra for $NaAlSi_3O_8$ (top) and $KAlSi_3O_8$ (bottom) at 21.1 T. Simulations were performed using parameters given in reference 15 of the manuscript.

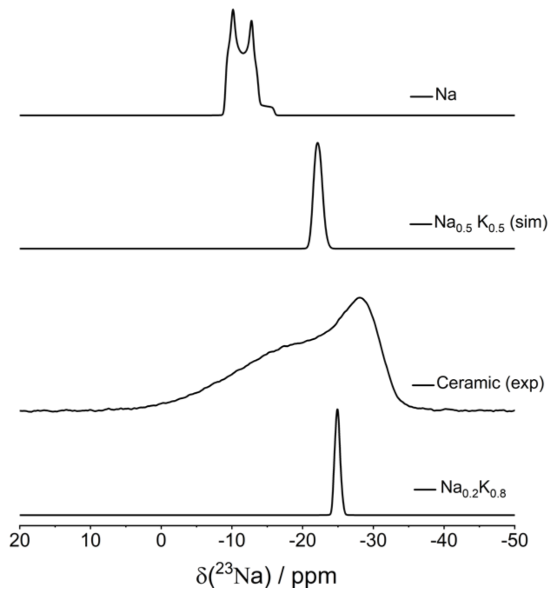

Figure S5. Comparison of experimental ^{23}Na MAS NMR spectrum for $Na_{0.5}K_{0.5}AlSi_3O_8$ glass-ceramic with simulated spectra for crystalline $NaAlSi_3O_8$, $Na_{0.5}K_{0.5}AlSi_3O_8$, and $Na_{0.2}K_{0.8}AlSi_3O_8$ at 21.1 T. Simulations were performed using parameters given in reference 15 of the manuscript.

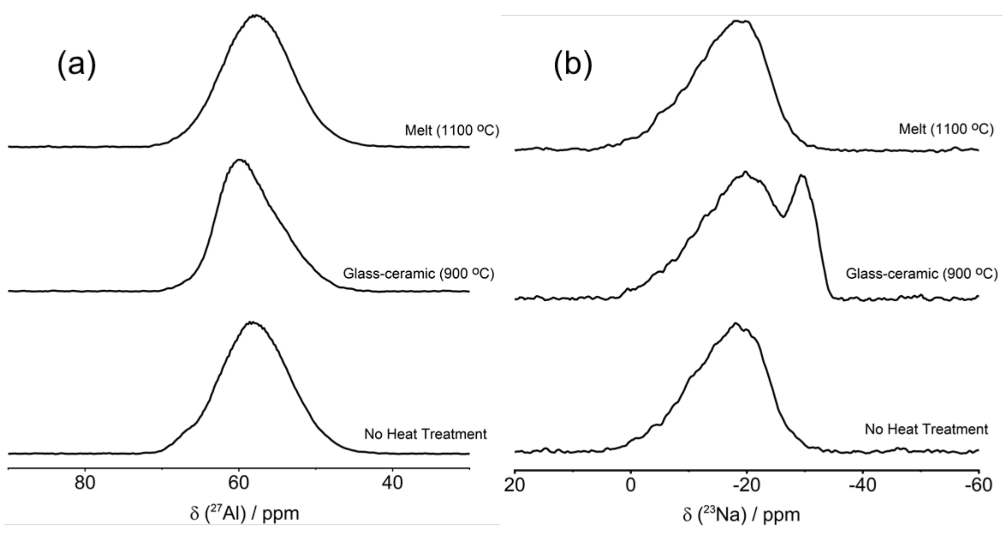

Figure S6. Projections along the isotropic dimension of triple quantum (3Q) (a) ^{27}Al and (b) ^{23}Na MAS NMR spectra (B_0 = 21.1 T; $\omega_r/2\pi$ = 31.25 kHz) of VITA VM9 dental ceramic at 25 °C (bulk powder, no heat treatment), 900 °C (glass-ceramic), and 1100 °C (melt/quench).

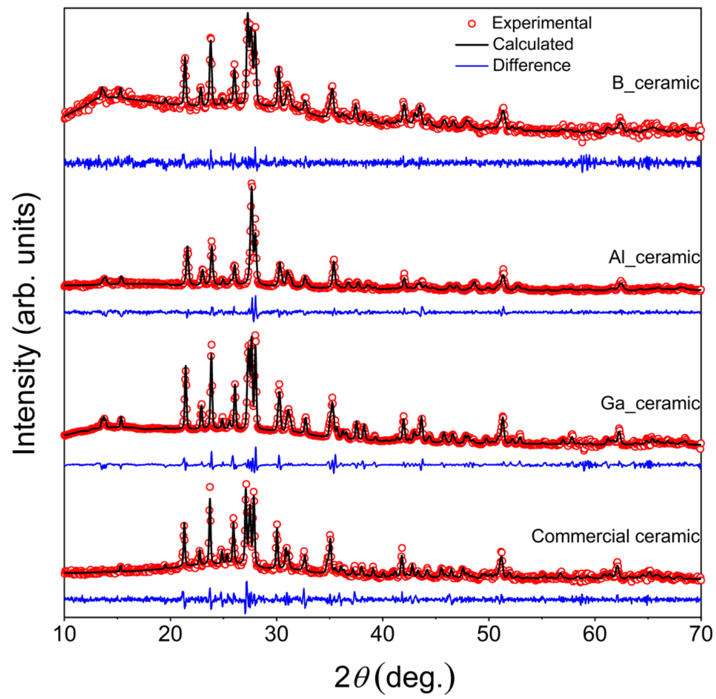

Figure S7. Powder XRD patterns of the commercial dental ceramic in comparison with lab-grade ($Na_{0.5}K_{0.5}AlSi_3O_8$), Ga-doped ($Na_{0.5}K_{0.5}Al_{0.9}Ga_{0.1}Si_3O_8$), and B-doped ceramics ($Na_{0.5}K_{0.5}Al_{0.9}B_{0.1}Si_3O_8$).

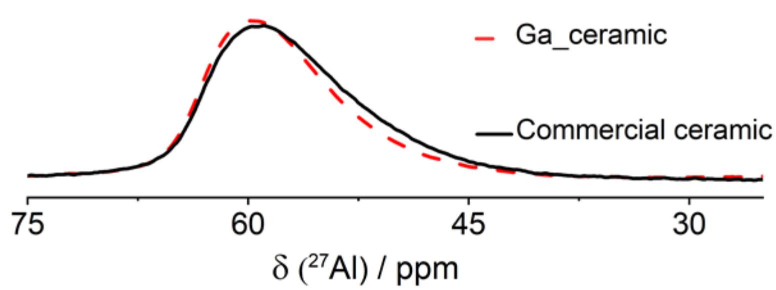

Figure S8. Overlay of the ^{27}Al MAS NMR spectra for the commercial VITA VM9 (black) and Ga-doped (red, $Na_{0.5}K_{0.5}Al_{0.9}Ga_{0.1}Si_3O_8$) glass ceramics.

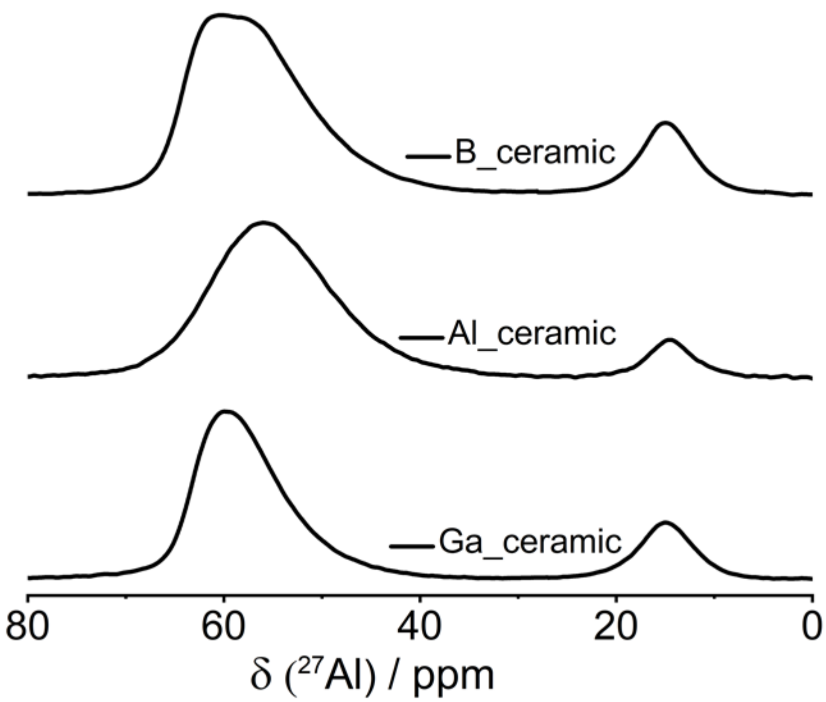

Figure S9. Aluminum-27 MAS NMR spectra of the group 13 synthetic glass-ceramic feldspar compounds ($Na_{0.5}K_{0.5}Al_{0.9}B_{0.1}Si_3O_8$, $Na_{0.5}K_{0.5}AlSi_3O_8$, and $Na_{0.5}K_{0.5}Al_{0.9}Ga_{0.1}Si_3O_8$). The spectra reveal the presence of both $^{[4]}Al$ (~40 to 70 ppm) and $^{[6]}Al$ (~10 to 20 ppm).

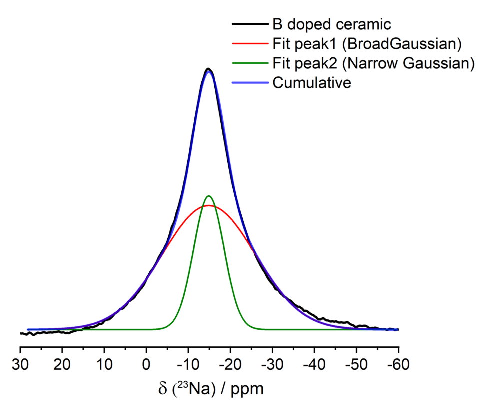

Figure S10. Sodium-23 MAS NMR spectra (B_0 = 14.1 T) of $Na_{0.5}K_{0.5}Al_{0.9}B_{0.1}Si_3O_8$ fit using a two-site Gaussian model.