Supporting information for

Phase Formation and Ionic Conduction in Na-Doped $Sr_2MgSi_2O_7$ Melilite-type Silicate

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Table S1. Final refined structural parameters of $Sr_{1.6}Na_{0.4}MgSi_2O_{6.8}$.*

Atom	Site	*x*	*y*	*z*	Occupancy	B_{iso}(Å^2)
Sr	4e	0.33454(4)	0.16546(4)	0.50837(3)	0.984(2)	0.63(2)
Na	4e	0.33454(4)	0.16546(4)	0.50837(3)	0.016(2)	0.63(2)
Mg	2a	0	0	0	1	0.65(6)
Si	4e	0.1387(1)	0.3613(1)	0.9437(2)	1	0.53(5)
O1	2c	0.5	0	0.16001(3)	1	0.49(7)
O2	4e	0.1413(2)	0.3587(2)	0.2544(5)	1	0.49(7)
O3	8f	0.0809(2)	0.1893(2)	0.7996(4)	1	0.49(7)

*$a = b = 8.00087(3)$ Å, $c = 5.169(1)$ Å, $\alpha = \beta = \gamma = 90°$, Z = 2, $V = 330.889(3)$ Å^3, space group: $P\bar{4}2_1m$; Reliability factors are $R_{wp} \sim 2.79\%$, $R_p \sim 2.04\%$, $R_B \sim 0.83\%$.

Table S2. Selected interatomic bond lengths of $Sr_{1.6}Na_{0.4}MgSi_2O_{6.8}$.

Bond	Length (Å)	Bond	Length (Å)
Sr1/Na1-O3 (×2)	2.534(2)	Mg1-O3 (×4)	1.962(2)
Sr1/Na1-O2 (×1)	2.550(3)	Si1-O2 (×1)	1.606(3)
Sr1/Na1-O1 (×1)	2.598(1)	Si1-O3 (×2)	1.632(2)
Sr1/Na1-O2 (×2)	2.750(2)	Si1-O1 (×1)	1.659(2)
Sr1/Na1-O3 (×2)	2.788(2)		

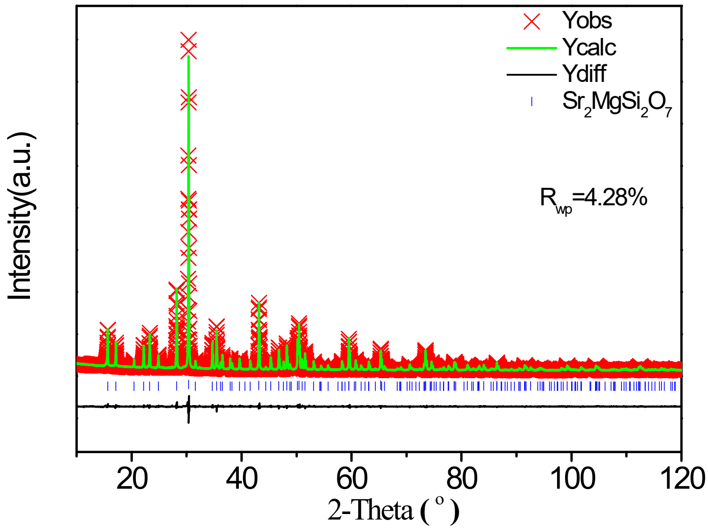


Figure S1. Rietveld profile of XRD data for $Sr_{1.6}Na_{0.4}MgSi_2O_{6.8}$.

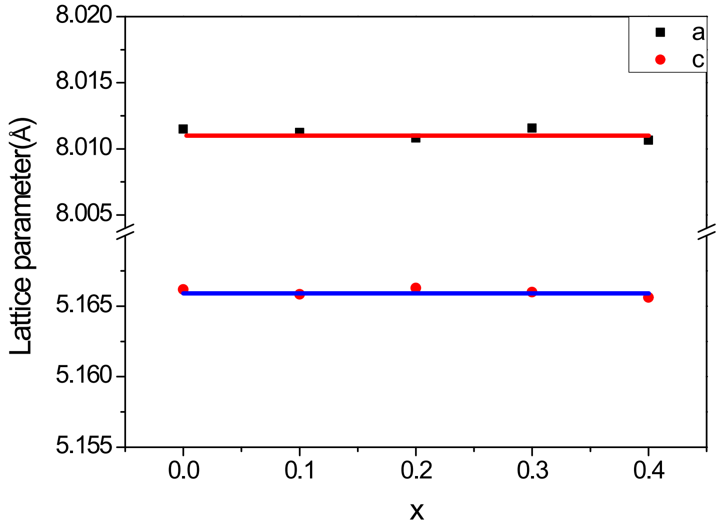


Figure S2. The refined cell parameters of $Sr_{2\text{-}x}Na_xMgSi_2O_{7\text{-}0.5x}$ (x = 0-0.4).

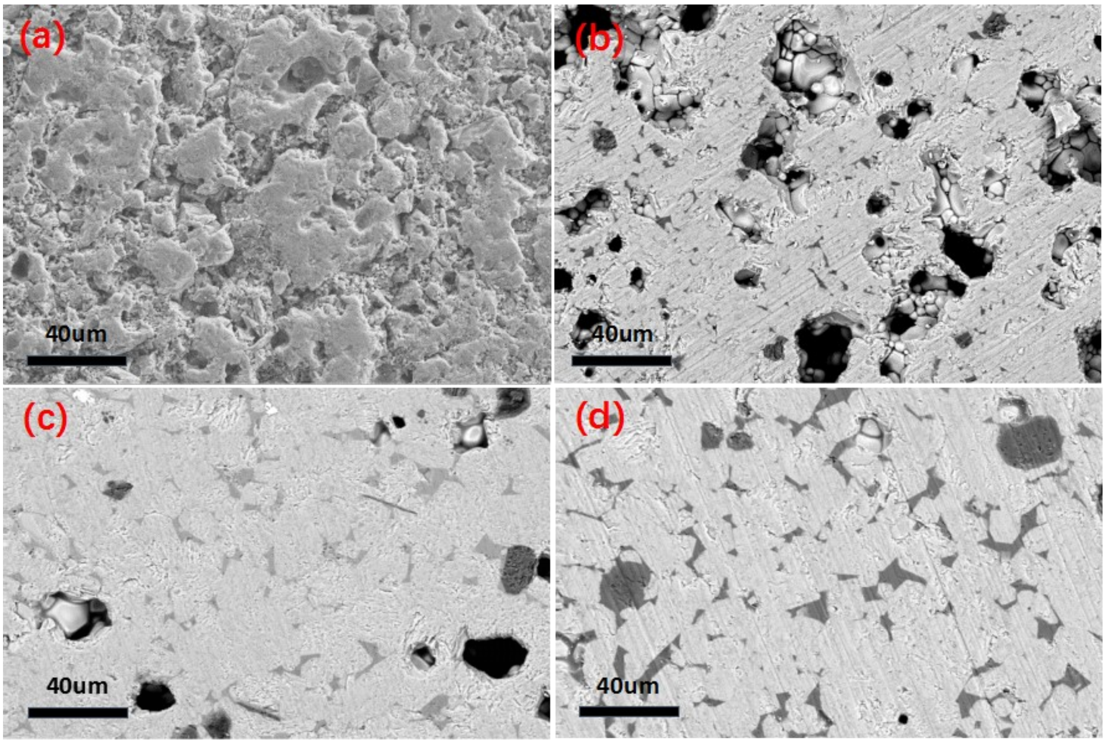
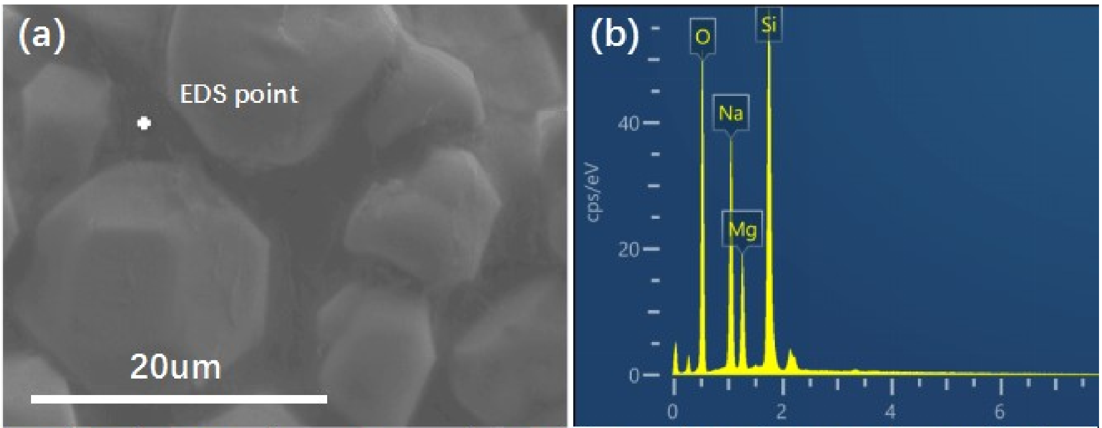


Figure S3. The SEM BSE images of polished surface for the (a) $Sr_2MgSi_2O_7$, (b) $Sr_{1.9}Na_{0.1}MgSi_2O_{6.95}$, (c) $Sr_{1.8}Na_{0.2}MgSi_2O_{6.9}$ and (d) $Sr_{1.7}Na_{0.3}MgSi_2O_{6.85}$ pellets.



Element	Ledge	Wt%	Wt% Sigma	Atomic percent	Normalization
O	K	41.35	0.23	53.15	4.62
Na	K	16.60	0.15	14.85	1.29
Mg	K	10.70	0.13	9.05	0.79
Si	K	31.34	0.20	22.95	2
Totality :		100.00		100.00	

Figure S4. Typical SEM-EDS elemental quantitative analysis of the glassy phase in the $Sr_{1.6}Na_{0.4}MgSi_2O_{6.8}$ pellet: (a) SEM image, (b) EDS spectrum and the table for atomic contents.

Table S3. The TEM-EDS element analysis results for the $Sr_{1.6}Na_{0.4}MgSi_2O_{6.8}$ sample on the crystal shown in Figure 3c.

Element	Edge	Wt% Sigma	Atomic percent	Normalization
O	K	31.9	58.0	6.2
Na	K	0	0	0
Mg	K	7.7	9.3	1
Si	K	17.9	18.6	2
Sr	K	42.4	14.1	1.5
Totality:		100.0	100.0	

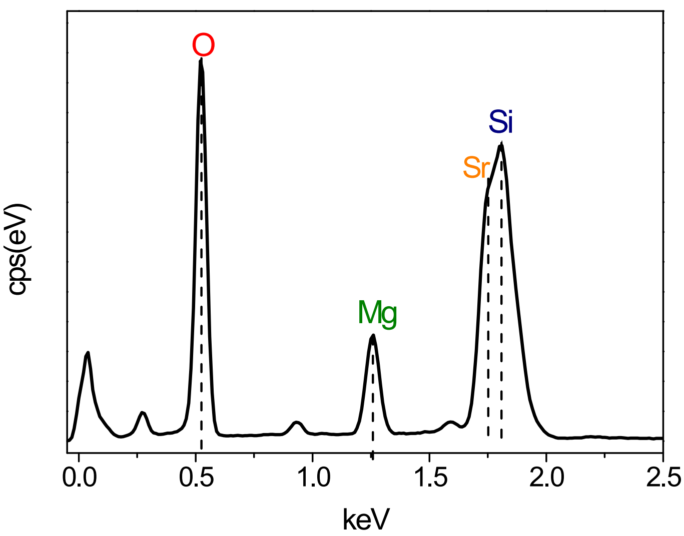
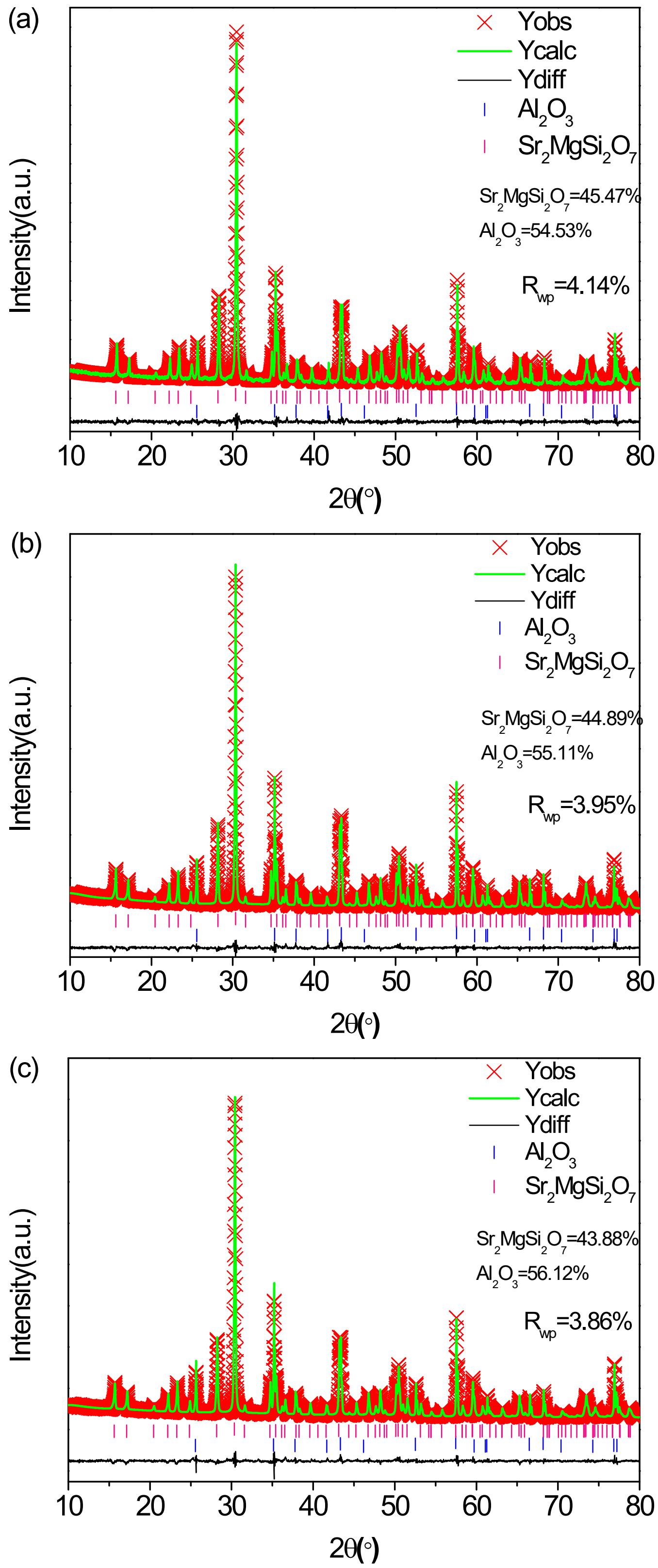
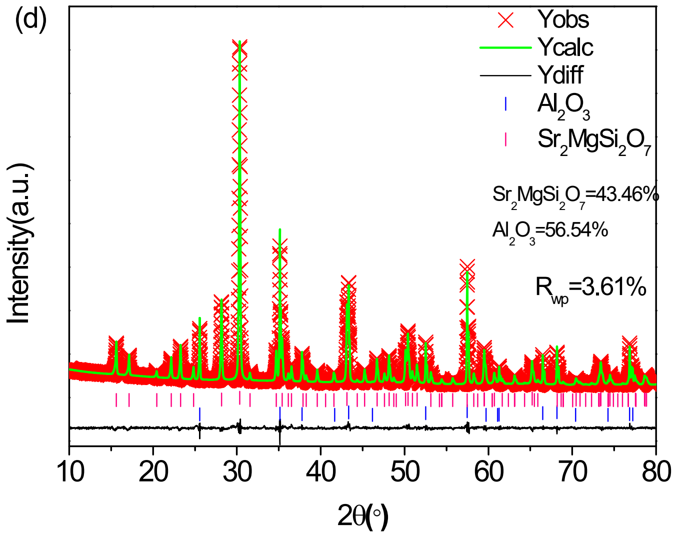


Figure S5. The TEM-EDS spectrum for the $Sr_{1.6}Na_{0.4}MgSi_2O_{6.8}$ sample on the crystal shown in Figure 3c.


(a)
Yobs
Ycalc
Ydiff
Al_2O_3
$Sr_2MgSi_2O_7$
$Sr_2MgSi_2O_7$=45.47%
Al_2O_3=54.53%
R_{wp}=4.14%
Intensity(a.u.)
10 20 30 40 50 60 70 80
2θ(°)
(b)
Yobs
Ycalc
Ydiff
Al_2O_3
$Sr_2MgSi_2O_7$
$Sr_2MgSi_2O_7$=44.89%
Al_2O_3=55.11%
R_{wp}=3.95%
Intensity(a.u.)
10 20 30 40 50 60 70 80
2θ(°)
(c)
Yobs
Ycalc
Ydiff
Al_2O_3
$Sr_2MgSi_2O_7$
$Sr_2MgSi_2O_7$=43.88%
Al_2O_3=56.12%
R_{wp}=3.86%
Intensity(a.u.)
10 20 30 40 50 60 70 80
2θ(°)



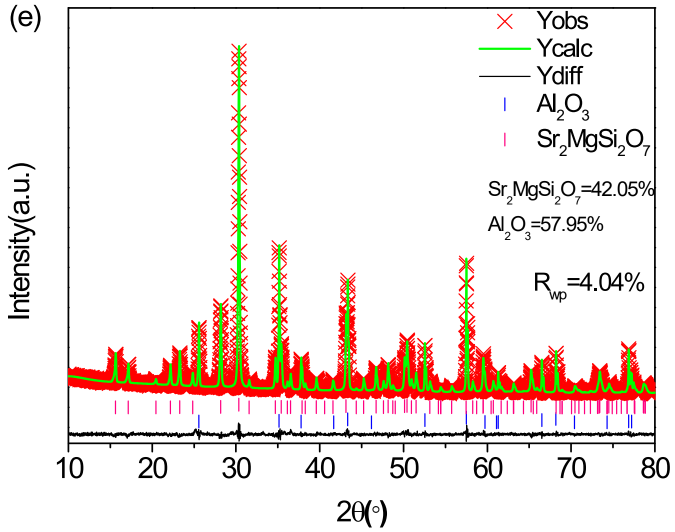


Figure S6. Two-phase Rietveld refinement of the mixture containing $Sr_{2-x}Na_xMgSi_2O_{7-x}$ and Al_2O_3 phases as internal standard with mass percentages of 50%: (a) $x = 0$, (b) $x = 0.1$, (c) $x = 0.2$, (d) $x = 0.3$, (e) $x = 0.4$.

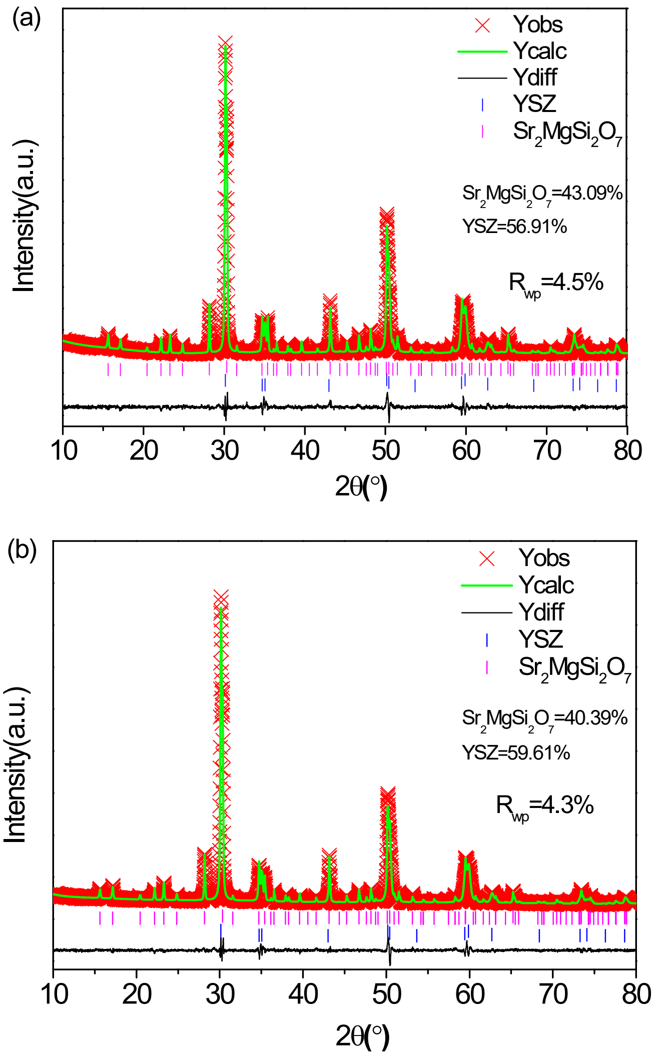


Figure S7. Rietveld plots of (a) $Sr_2MgSi_2O_7$ and (b) $Sr_{1.7}Na_{0.3}MgSi_2O_{6.85}$ with YSZ as internal standard. The glassy phase contents from the two-phase Rietveld analysis are 24(1) wt% and 32(1) wt% in $Sr_2MgSi_2O_7$ and $Sr_{1.7}Na_{0.3}MgSi_2O_{6.85}$, respectively. Although the difference between their glassy phase content 7.9 % is close to that (6.9%) from the analysis using the Al_2O_3 standard, the systematic error (24.3 wt%) is even higher than that (16.6 wt%) from the analysis using Al_2O_3 as standard.

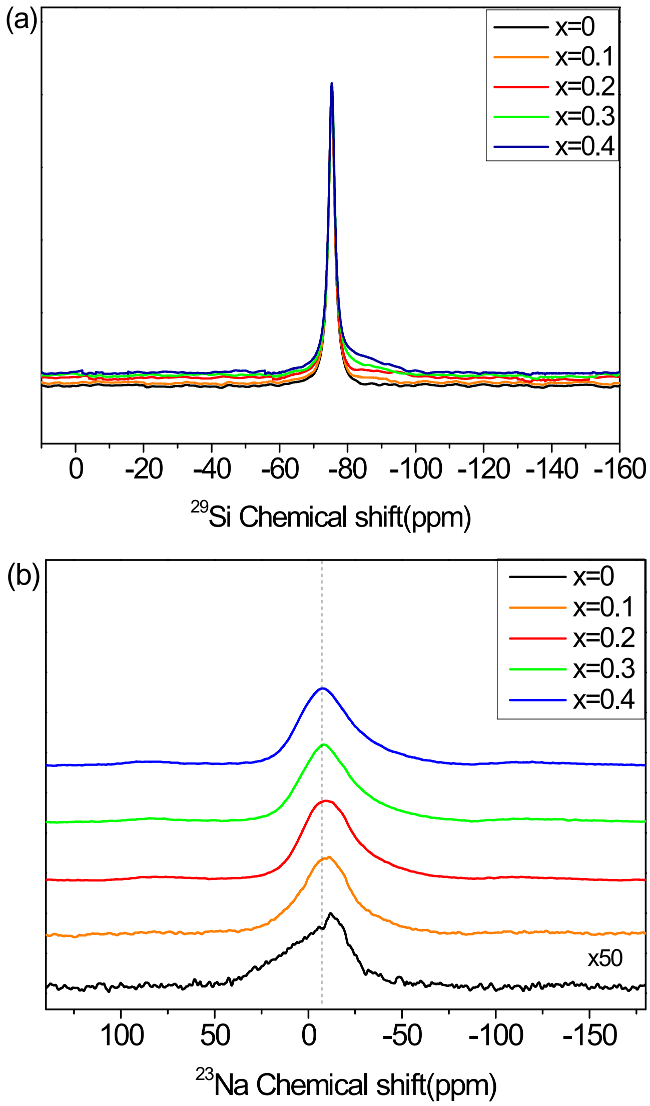


Figure S8. (a) ^{29}Si and (b) ^{23}Na solid state NMR spectra for the x = 0-0.4 compositions of $Sr_{2-x}Na_xMgSi_2O_{7-0.5x}$.

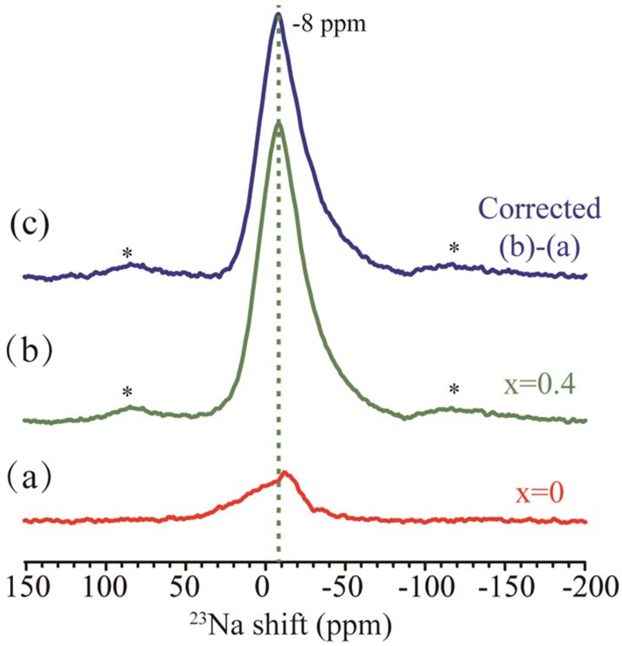


Figure S9. The correction of ^{23}Na NMR spectrum by subtracting (a) the signal of undoped pristine composition from (b) the raw NMR spectra for $Sr_{1.6}Na_{0.4}MgSi_2O_{6.8}$. (c) is the corrected spectrum.

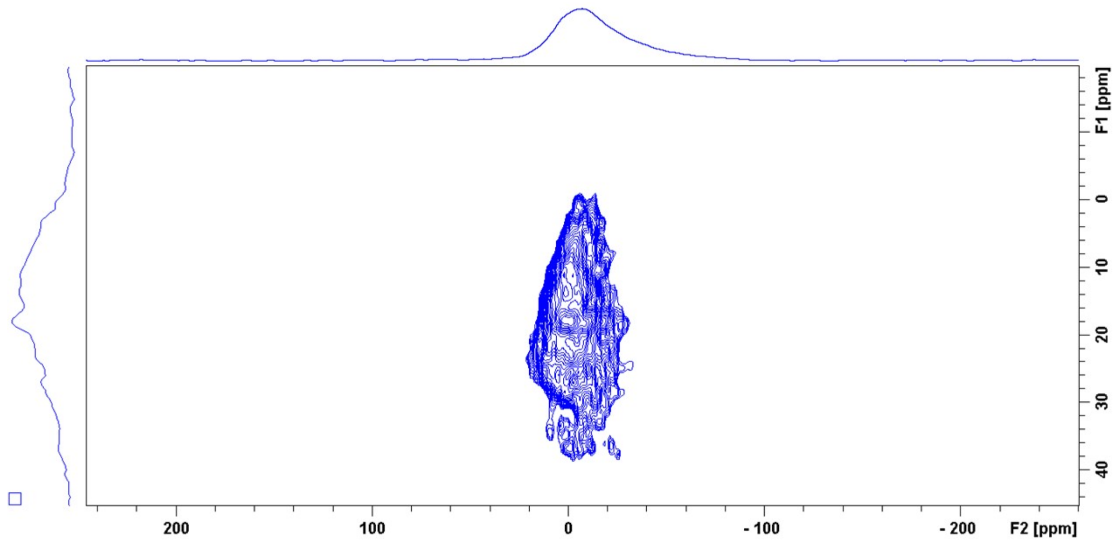


Figure S10. Two dimensional ^{23}Na MQMAS NMR spectrum of $Sr_{1.6}Na_{0.4}MgSi_2O_{6.8}$.

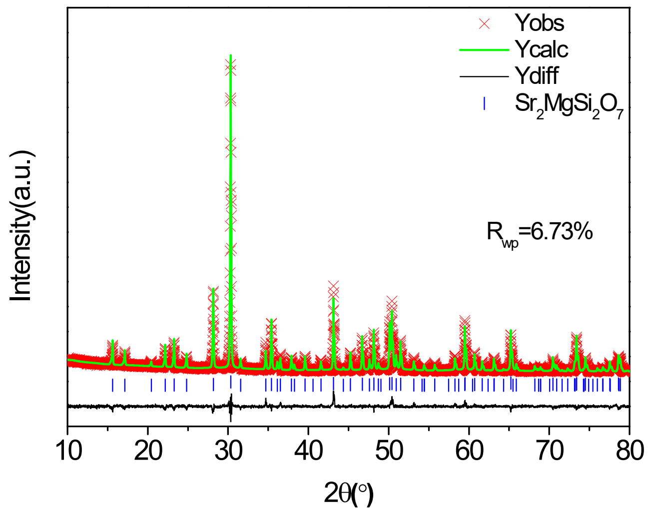


Figure S11. Rietveld refinement of the XRD data for the $Sr_{1.6}Na_{0.4}MgSi_2O_{6.8}$ sample at 1000 °C.