Intercalation Route to Complex Perovskites $AM_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$ (*A* = Sr, Ba; *M* = Li, Na): Neutron Diffraction and Nuclear Magnetic Resonance Study

Young-Il Kim,[*,†] Younkee Paik,[‡] Maxim Avdeev[§]

[†] Department of Chemistry, Yeungnam University, Gyeongsan 712-749, Korea

[‡] Korea Basic Science Institute, Daegu 702-701, Korea

[§] Australian Nuclear Science and Technology Organisation, Menai, NSW 2234, Australia

Table S1. Summary of the X-ray Rietveld refinement of $AM_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$ (A = Li, Na; M = Sr, Ba).

	$SrLi_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$	$SrNa_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$	$BaLi_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$	$BaNa_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$
λ (Å)	1.5475	1.5472	1.5472	1.5472
space group	$I4/mcm$	$I4/mcm$	$Pm\bar{3}m$	$Pm\bar{3}m$
Z	4	4	1	1
a (Å)	5.67696(9)	5.7227(1)	4.11049(2)	4.13271(4)
c (Å)	8.0436(2)	8.1165(4)		
V (Å^3)	259.228(5)	265.81(1)	69.451(1)	70.584(2)
2θ range	10–130.5	12–130.5	10–130	12–130
N_{data}	12049	11849	11999	11799
N_{obs}	75	91	24	26
R_{wp} (%)	4.99	6.60	8.03	7.37
R_p (%)	4.24	5.27	7.03	6.10
R_{F2} (%)	1.71	4.14	2.03	5.07
χ^2	1.73	2.74	2.57	2.53

Table S2. Atomic parameters for $SrM_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$ (M = Li, Na) from an X-ray Rietveld refinement.

atom	site	M = Li		M = Na	
		occupancy	U_{iso} (Å^2)	occupancy	U_{iso} (Å^2)
Sr	4b (0, ½, ¼)	$Sr_{1.0}$	0.0147(2)	$Sr_{1.0}$	0.0242(2)
M/Ta	4c (0, 0, 0)	$Li_{0.244(2)}Ta_{0.756}$	0.0072(1)	$Na_{0.19(1)}Ta_{0.81}$	0.0215(1)
O/N1	4a (0, 0, ¼)	$O_{0.933}N_{0.067}$	0.0249(7)	$O_{0.933}N_{0.067}$	0.024(1)
O/N2	8h (x, x + ½, 0)	$O_{0.933}N_{0.067}$	0.0249	$O_{0.933}N_{0.067}$	0.024
		x = 0.7752(7)		x = 0.7820(8)	

Table S3. Atomic parameters for $BaM_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$ (M = Li, Na) from an X-ray Rietveld refinement.

atom	site	M = Li		M = Na	
		occupancy	U_{iso} (Å^2)	occupancy	U_{iso} (Å^2)
Ba	1a (0, 0, 0)	$Ba_{1.0}$	0.0084(2)	$Ba_{1.0}$	0.0123(2)
M/Ta	1b (½, ½, ½)	$Li_{0.206(2)}Ta_{0.794}$	0.0072(2)	$Na_{0.18(1)}Ta_{0.82}$	0.0107(2)
O/N	3c (0, ½, ½)	$O_{0.933}N_{0.067}$	0.0058(8)	$O_{0.933}N_{0.067}$	0.0053(8)

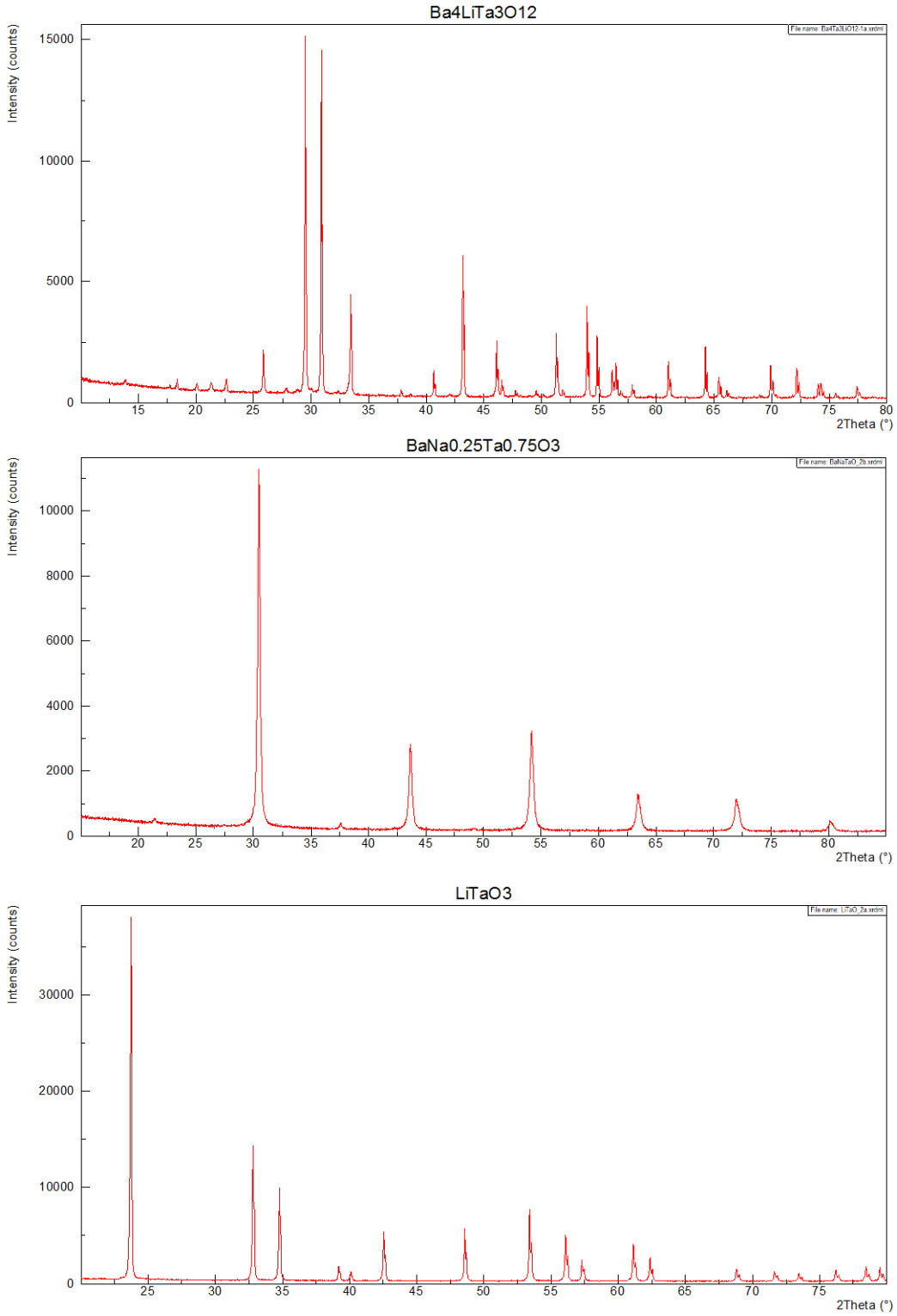


Figure S1. (continued)

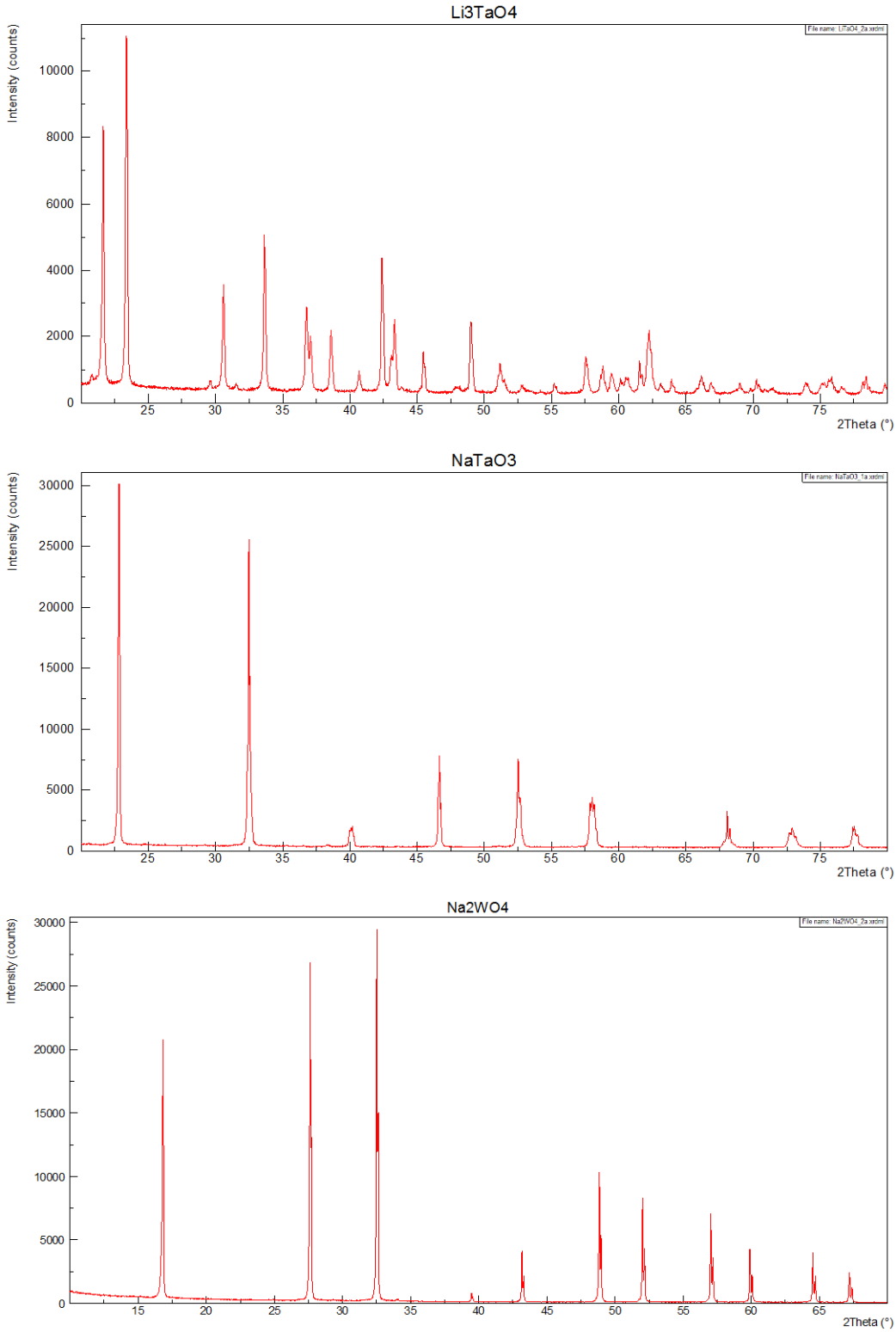


Figure S1. XPD patterns (Cu K_α) of reference compounds, $Ba_4LiTa_3O_{12}$, $BaNa_{0.25}Ta_{0.75}O_3$, $LiTaO_3$, Li_3TaO_4, $NaTaO_3$, and Na_2WO_4.

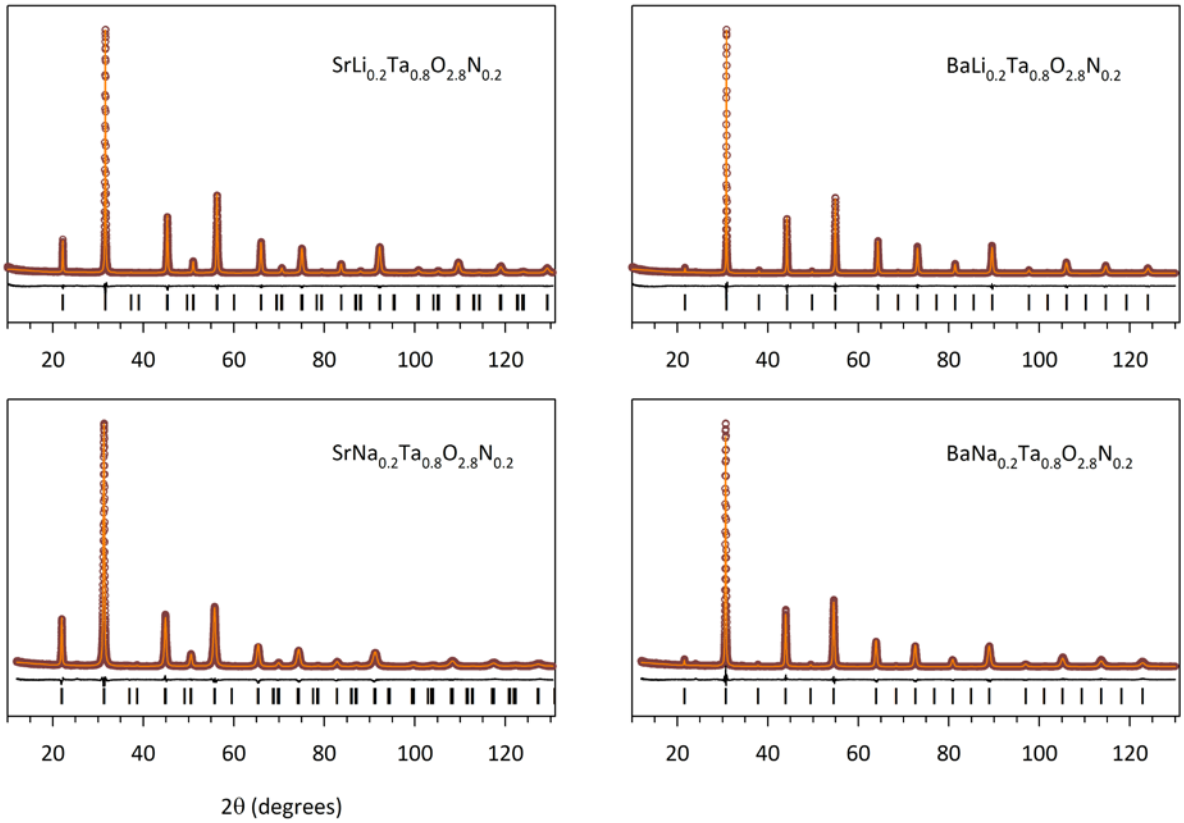


Figure S2. Rietveld refinement of SXPD ($\lambda \approx 1.547$ Å) patterns for $AM_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$ (A = Sr, Ba; M = Li, Na).

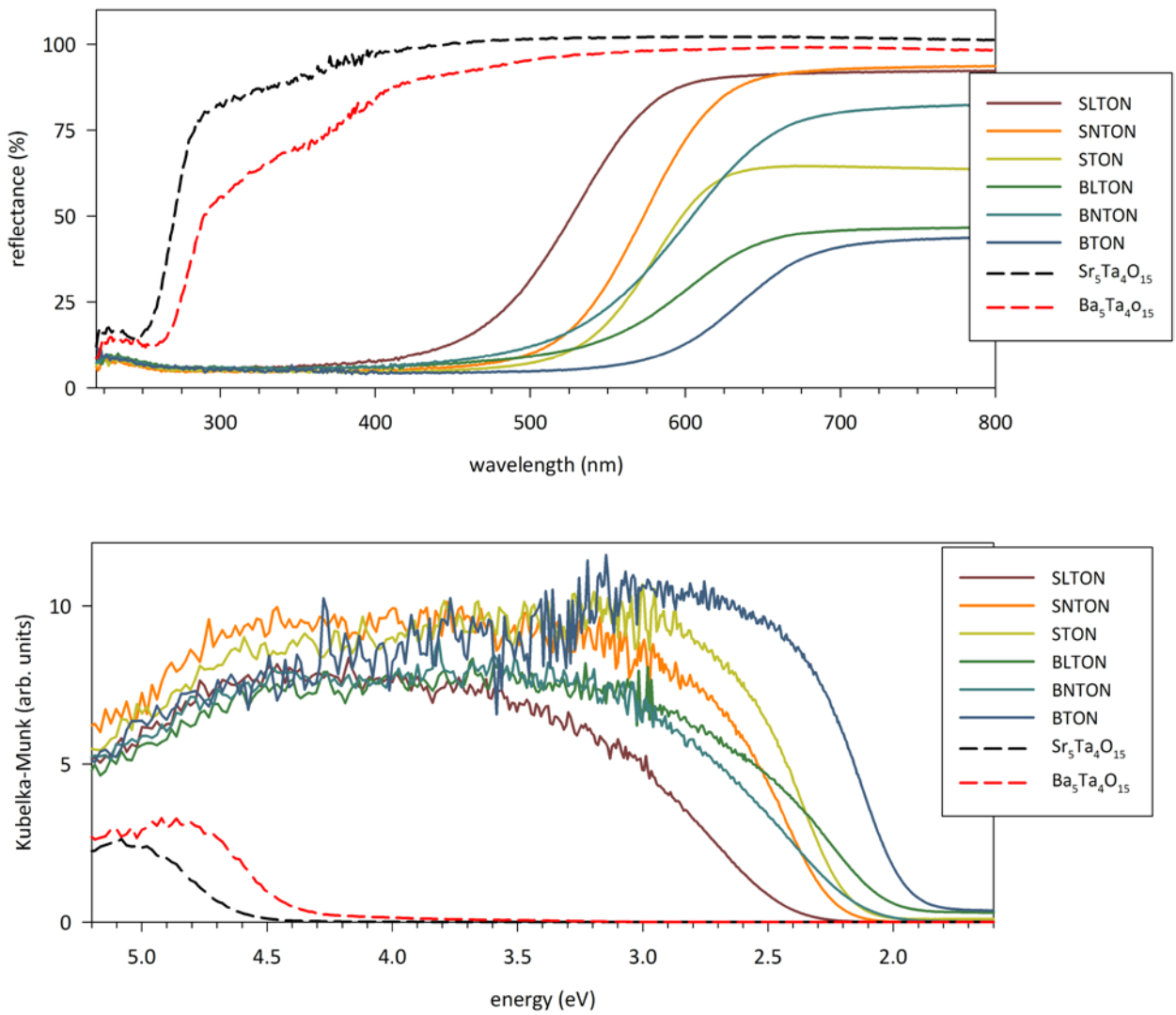
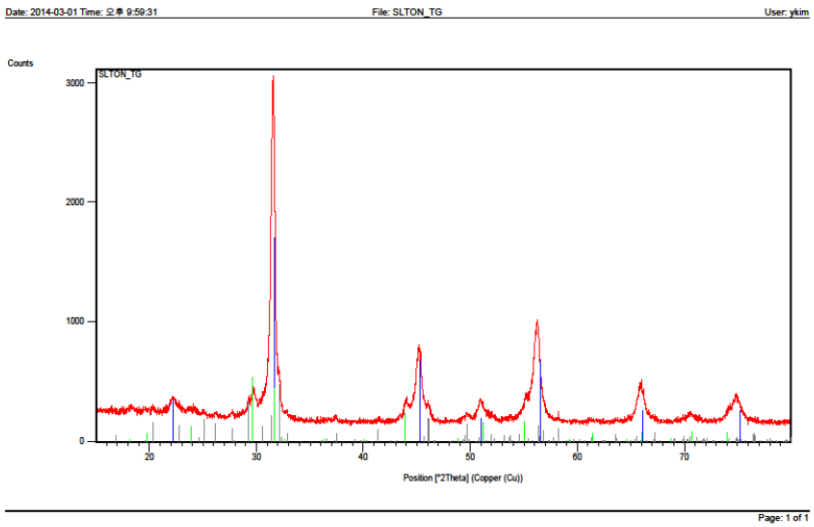
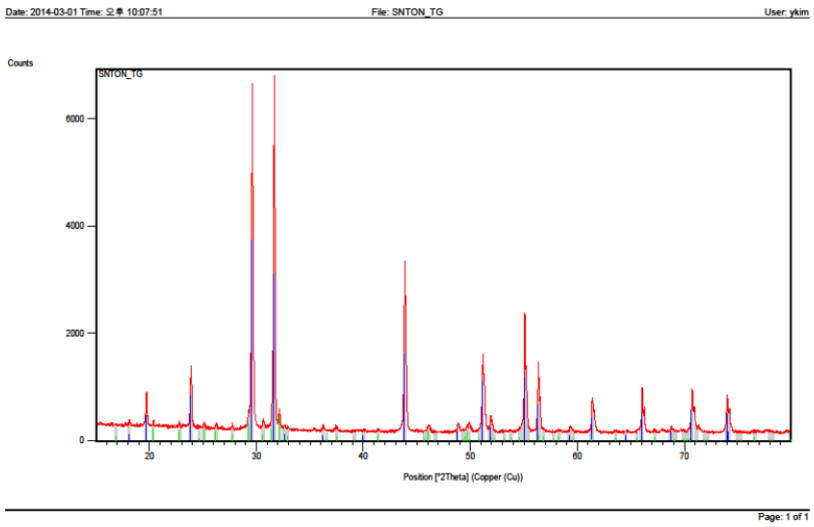


Figure S3. UV-vis spectra for $AM_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$ and $A_5Ta_4O_{15}$ (A = Sr, Ba; M = Li, Na): (top) diffuse-reflectance and (bottom) absorbance.



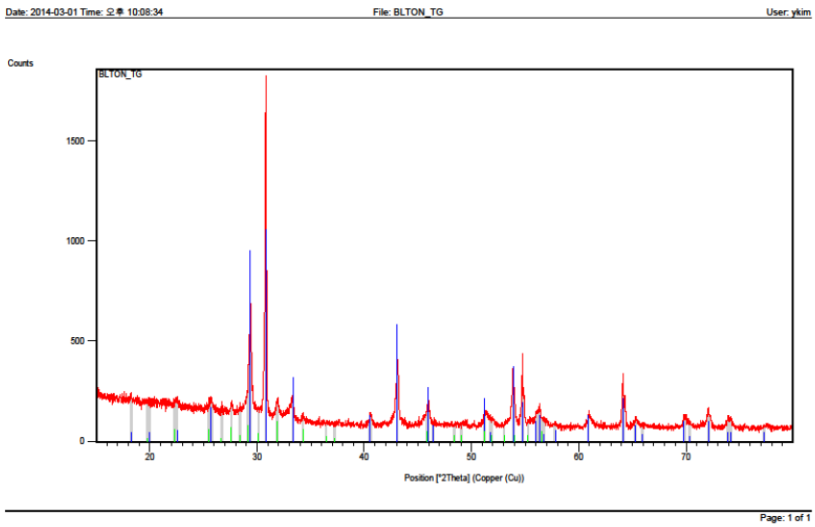
(A)



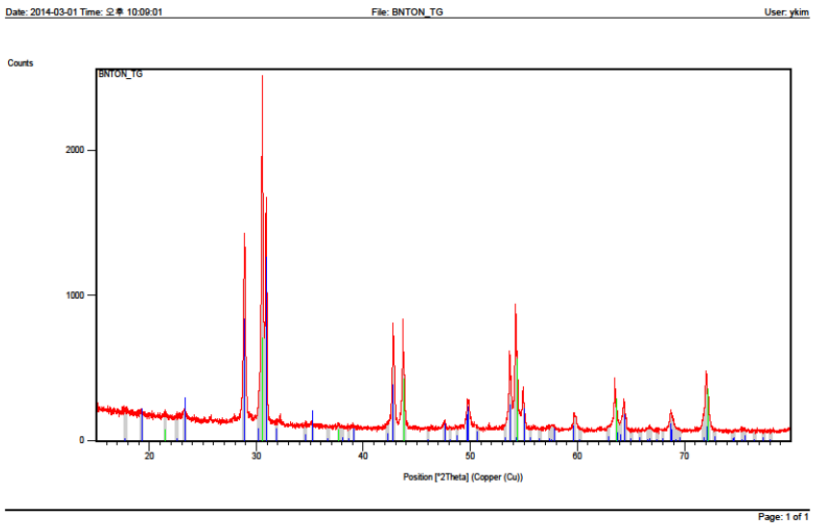
(B)

Before	After (phases and PDF No.)	Diagram
$SrLi_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$	$SrLi_{0.25}Ta_{0.75}O_3$ (#25-1382, blue) $Sr_5Ta_4O_{15}$ (#54-1251, green) $Sr_2Ta_2O_7$ (#30-1304, gray)	A
$SrNa_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$	$Sr_5Ta_4O_{15}$ (#54-1251, blue) $Sr_2Ta_2O_7$ (#30-1304, green)	B

Figure S4. (continued)



(C)



(D)

Before	After	Diagram
$BaLi_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$	$Ba_4LiTa_3O_{12}$ (#27-1216, blue) $Ba_{0.5}TaO_3$ (#17-793, green)	C
$BaNa_{0.2}Ta_{0.8}O_{2.8}N_{0.2}$	$Ba_5Ta_4O_{15}$ (#1-72-631, blue) $BaNa_{0.25}Ta_{0.75}O_3$ (#18-1200, green)	D

Figure S4. XPD patterns (Cu K_α) for the samples after the TGA up to 1670 K in air.

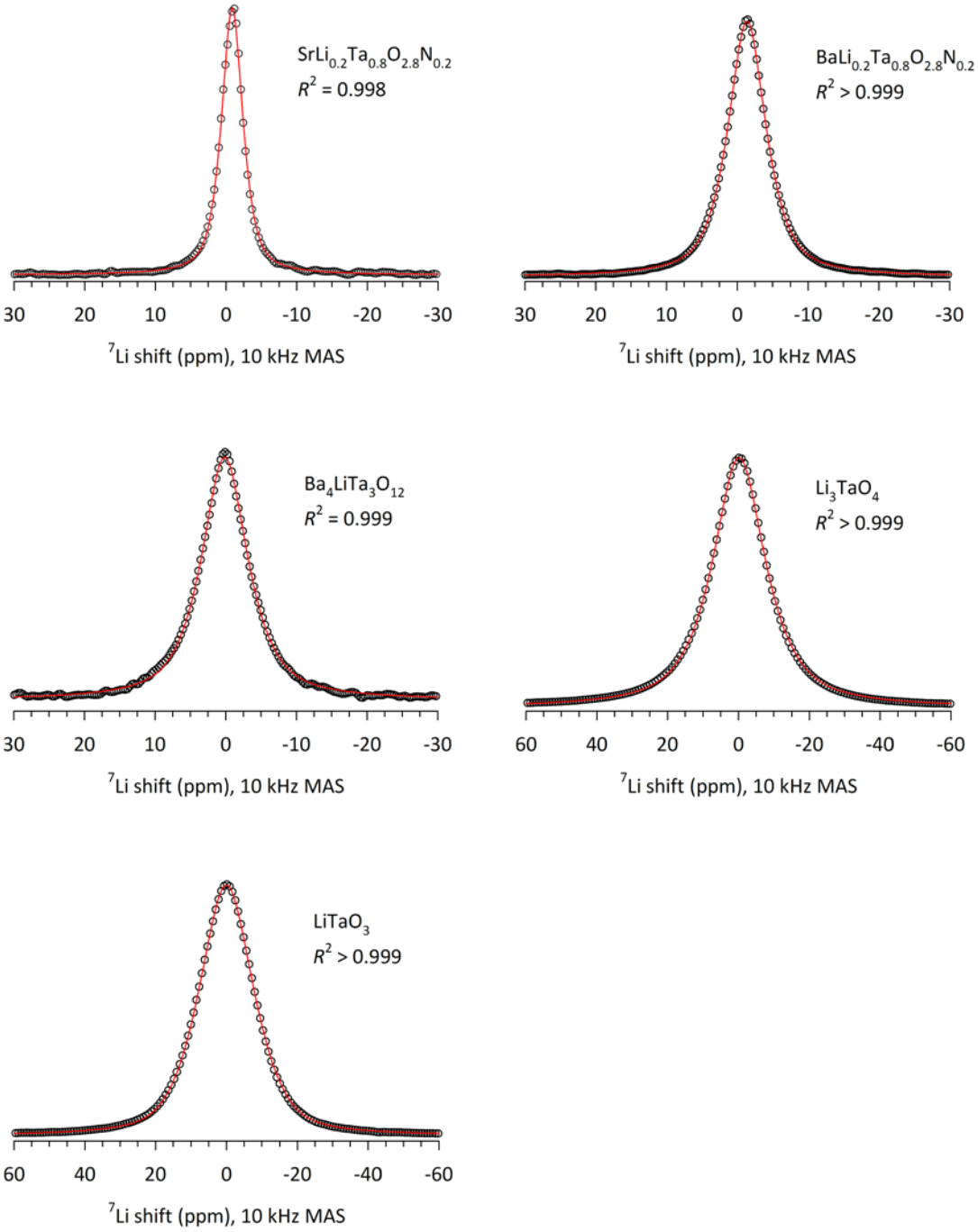


Figure S5. Peak fitting of centerbands from ^{7}Li MAS NMR. (experimental: circles, fit: red line)

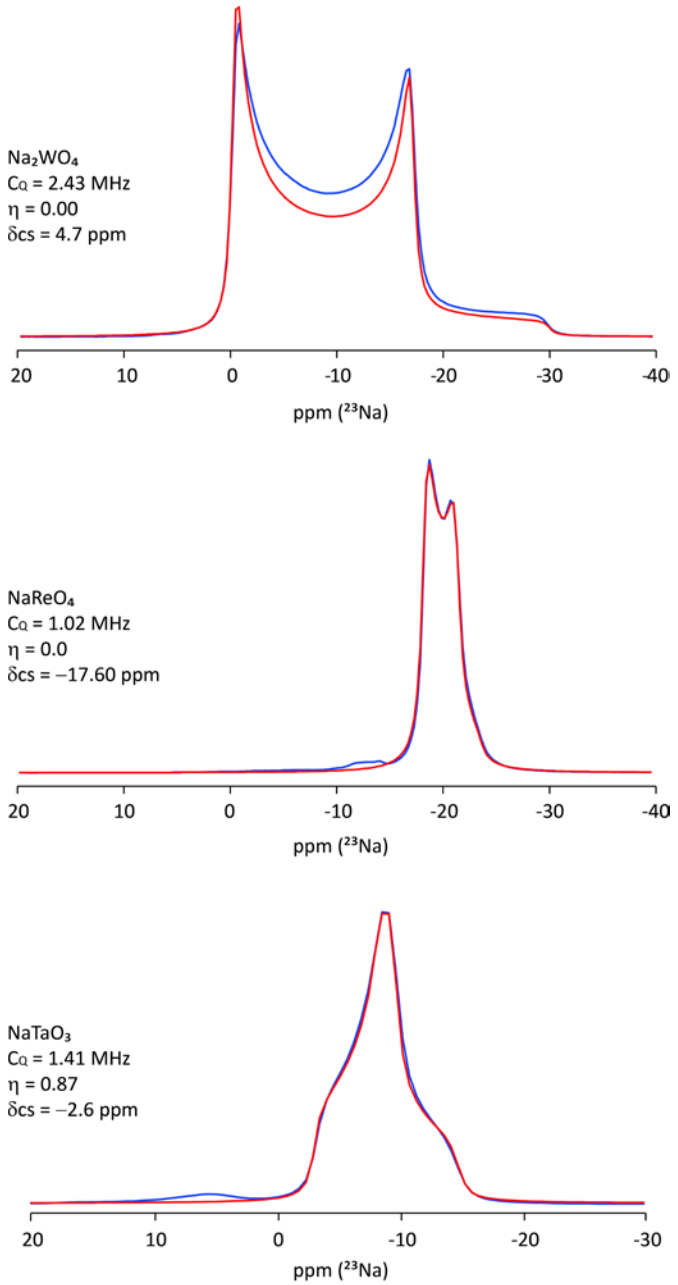


Figure S6. Fitting of ^{23}Na MAS NMR line shapes of Na_2WO_4, $NaReO_4$, and $NaTaO_3$ (experimental: blue, fit: red).