

Figure S.1. XRD data for $A = \text{Na}^+$ as prepared and after treatment with concentrated nitric acid. **a)** Data from $10 - 60^\circ$, inset shows the (110) reflection, **b)** the (100) reflection, **c)** the (111) reflection, **d)** the (200) reflection, and **e)** the (211) reflection.

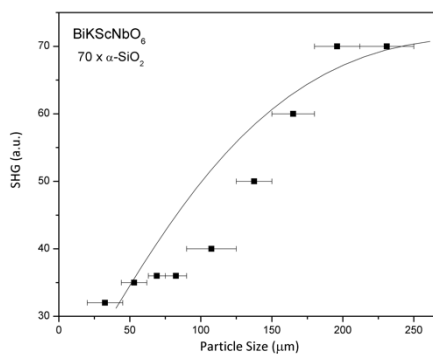


Figure S.2. SHG efficiency as a function of particle size for $A = \text{K}^+$.

Table S.1. Space group, R_{wp} , χ^2 , and A -site B_{eq} for Rietveld refinements on $A = \text{K}^+$. Results shown for $P4mm$, $Amm2$, and $R3m$ have been refined without constraints on the atomic positions of A -site cations.

Space Group	R_{wp}	χ^2	B_{eq}^\dagger
$Pm\bar{3}m$	7.44	3.14	10.51(9)
$P4mm$	6.86	2.72	10.73(3)
$Amm2$	6.81	2.63	10.70(2)
$R3m$	6.96	2.74	10.70(4)
$Pm\bar{3}m[100]$	7.05	2.82	4.58(5)

† isotropic displacement parameter of the A -site Cation

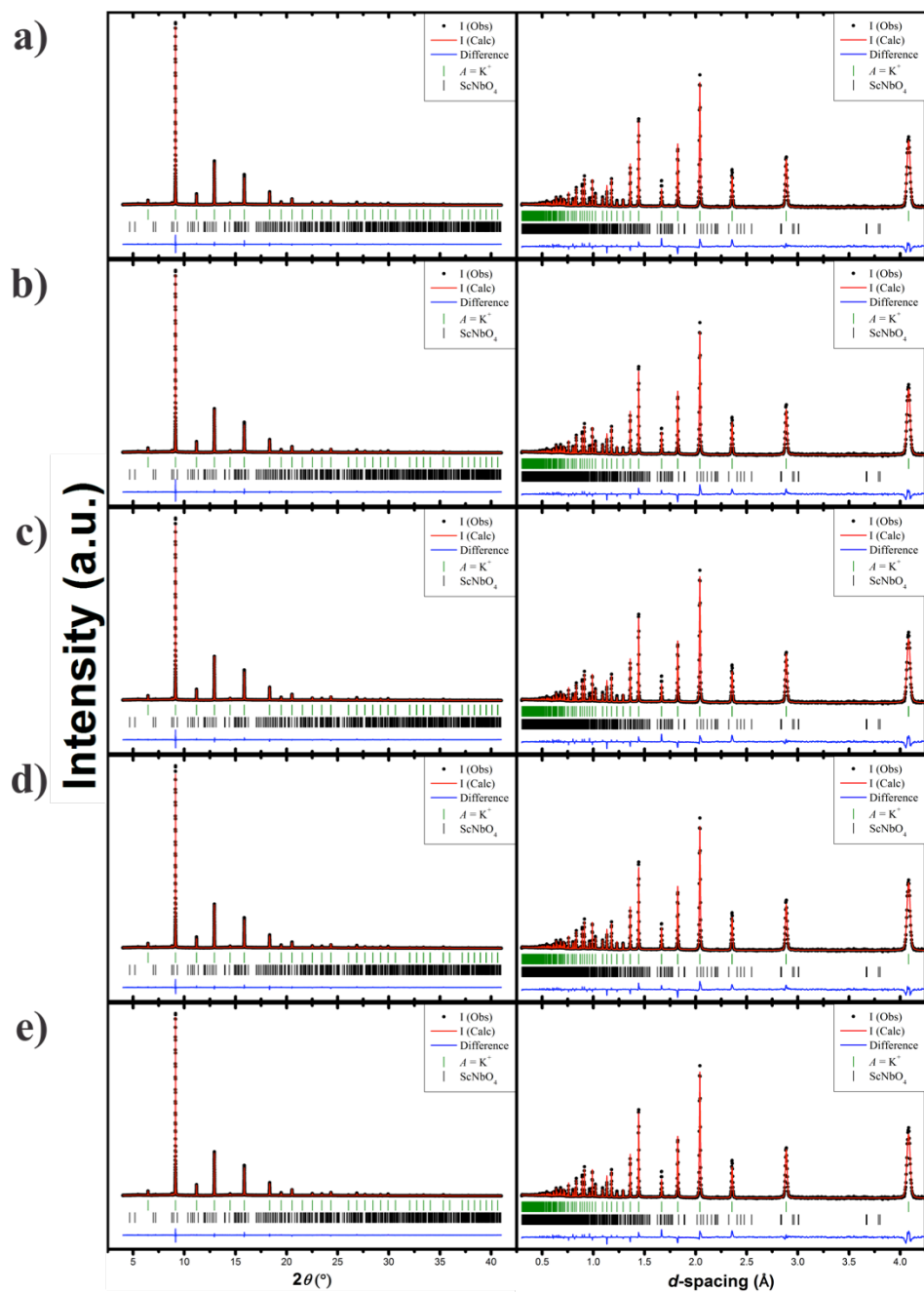


Figure S.3. Results of combined Rietveld refinement on $A = K^+$ using synchrotron XRD and NPD data for a) $Pm\bar{3}m$, b) $P4mm$, c) $Amm2$, d) $R3m$, and e) $[100]$ distorted $Pm\bar{3}m$.

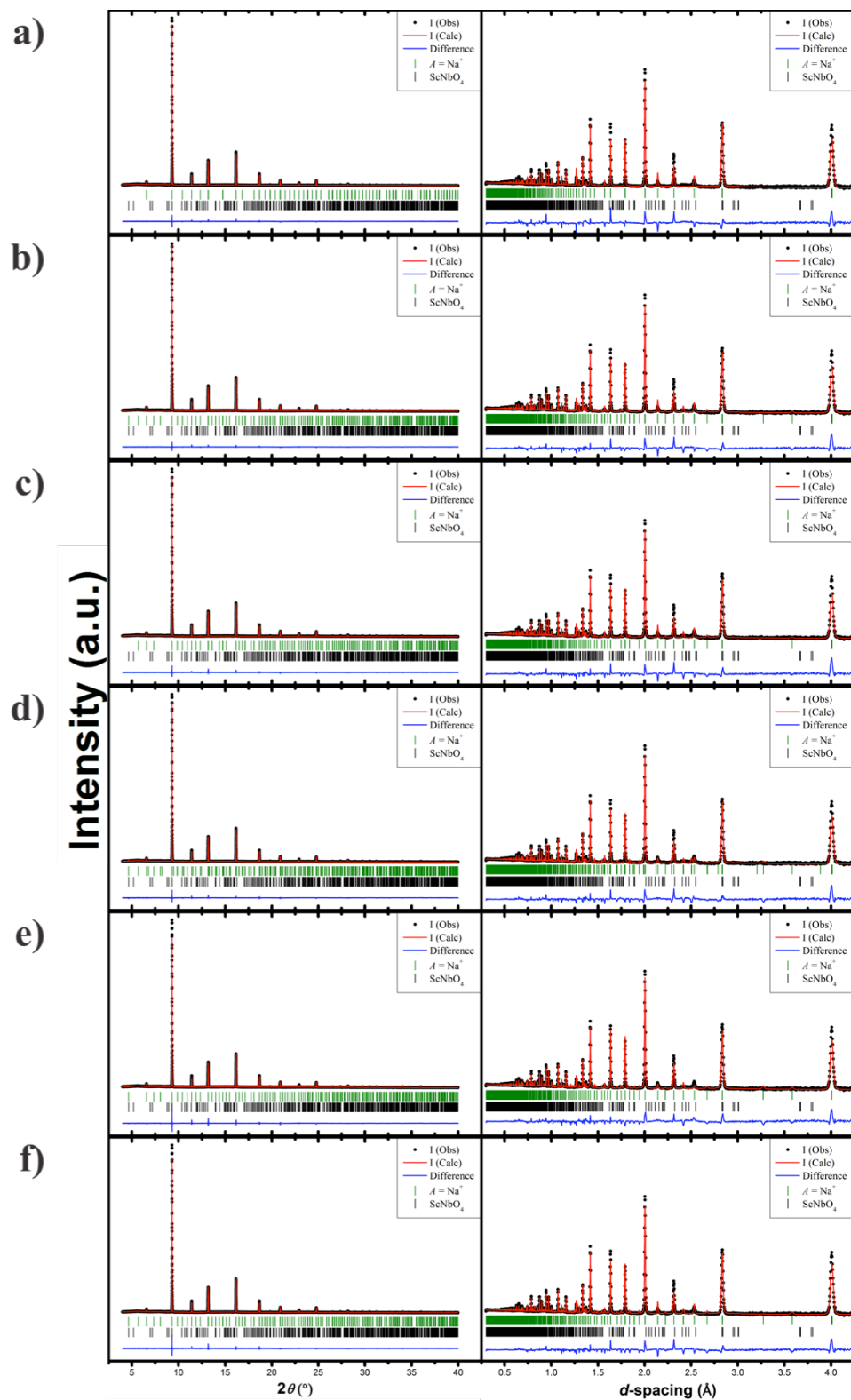


Figure S.4. Results of combined Rietveld refinement on $A = \text{Na}^+$ using synchrotron XRD and NPD data for a) $P4/mbm$, b) $Cmcm$, c) $Pbnm$, d) $Pbcm$, and e) $R3c$, and f) $P2_1/m$.

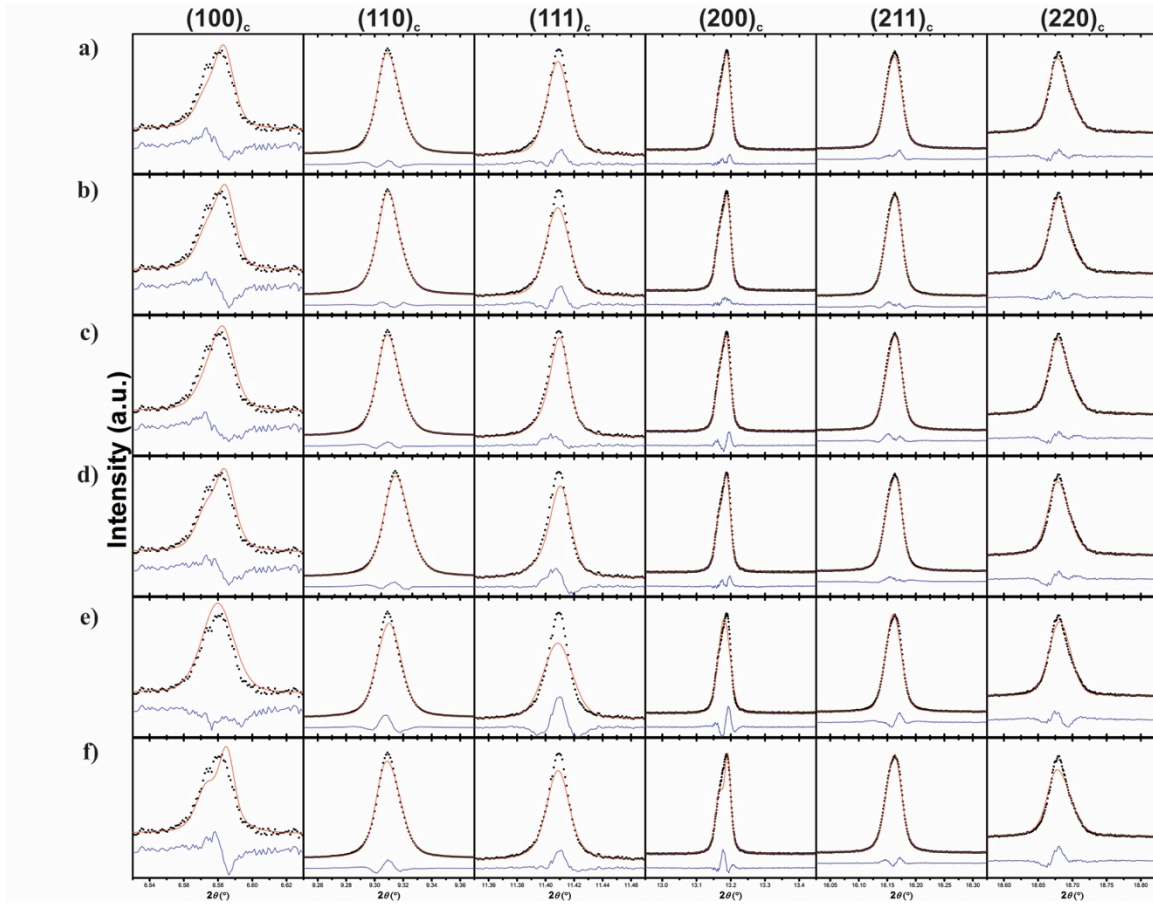


Figure S.5. The fits of selected peaks for Rietveld refinement of XRD data for $A = \text{Na}^+$ in **a)** $P4/mbm$, **b)** $Cmcm$, **c)** $Pbnm$, **d)** $Pbcm$, and **e)** $R3c$, and **f)** $P2_1/m$ symmetry.

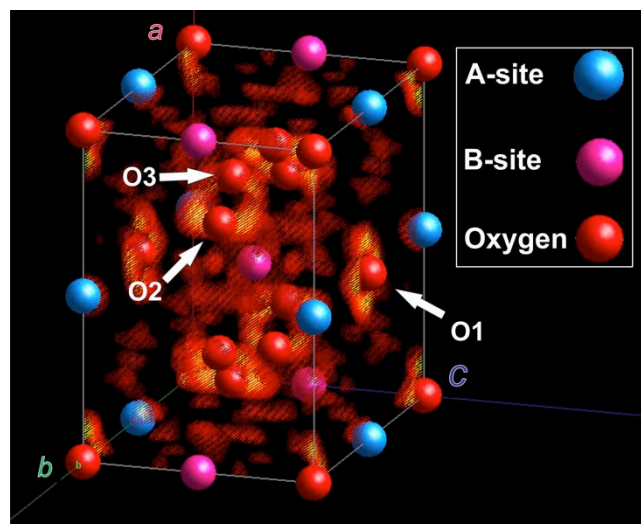


Figure S.6. Fourier difference map calculated from the Rietveld refinement of $A = \text{Na}^+$ in $P4/mbm$, with a split oxygen site, on NPD data