Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.
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SUPPORTING INFORMATION

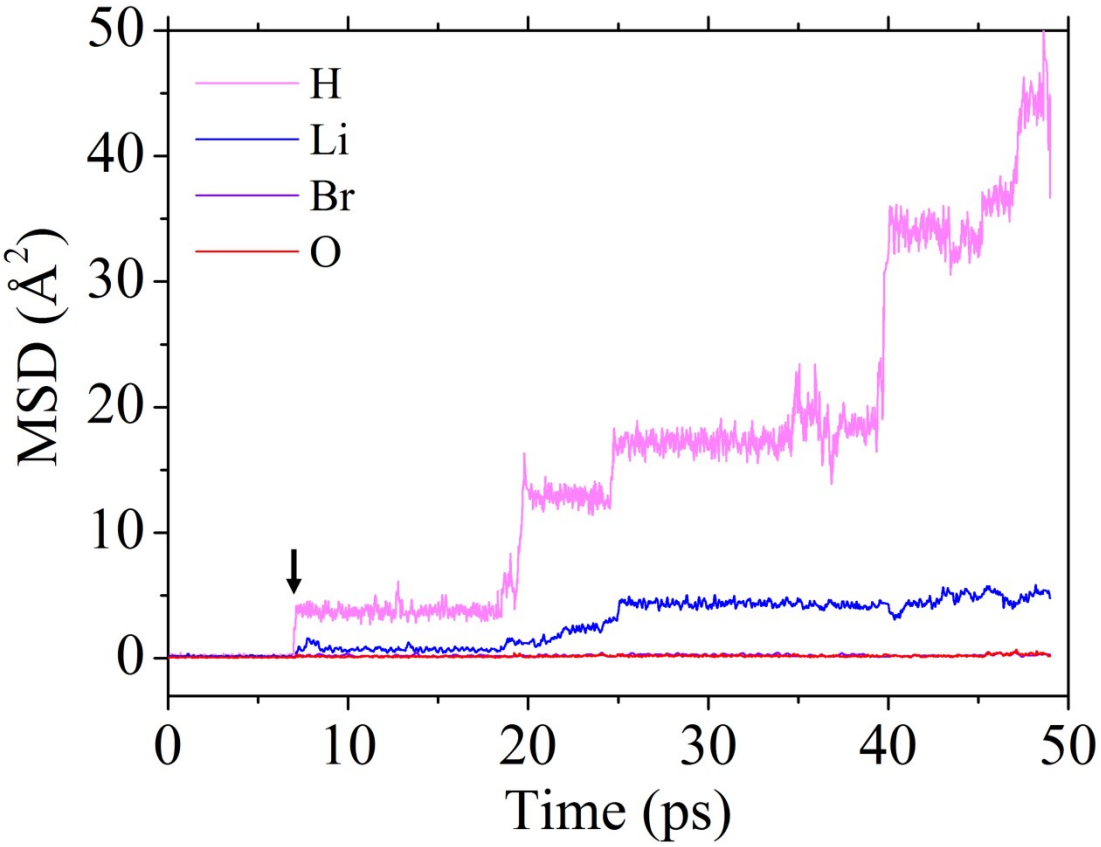

Figure S1. Calculated mean squared displacements of all the atomic species in the model structure of Li_2OHBr using data from molecular dynamics simulations at a simulating temperature of 300 K (see Method). After about 7 ps into the simulation (indicated by the black arrow), hydroxide ions start decomposition, leading to proton fast-ion conduction.

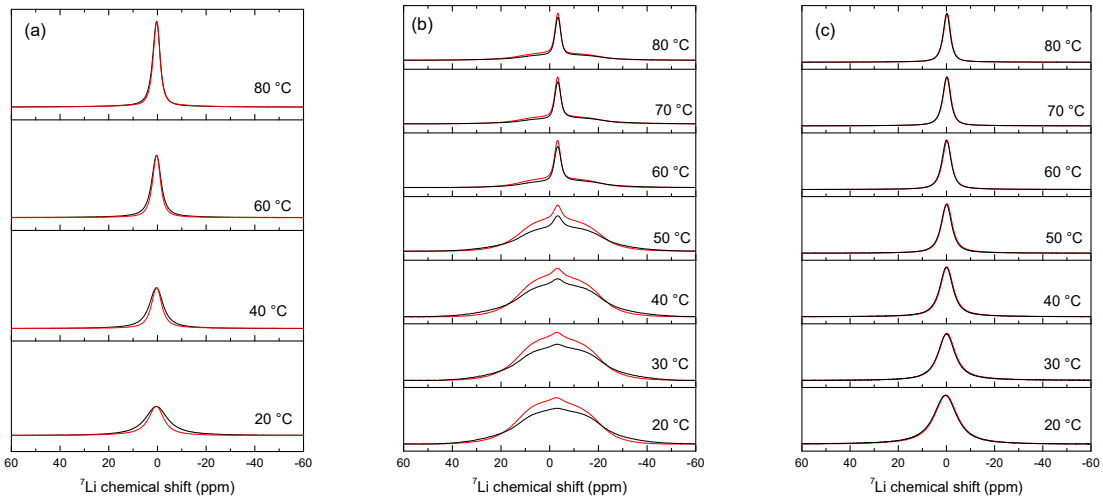

Figure S2. Temperature-dependent ^{7}Li NMR spectra with (S_{Li-H}, red) and without (S_{Li}, black) proton decoupling for (a) $Li_2OHCl_{0.37}Br_{0.63}$, (b) Li_2OHCl, and (d) Li_2OHBr.

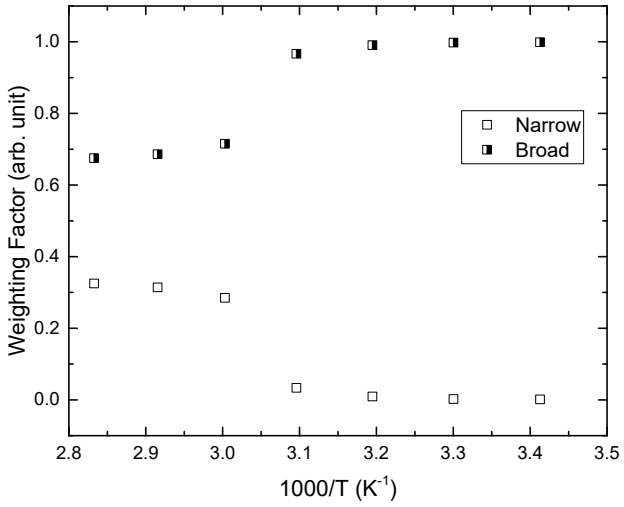

Figure S3. Temperature-dependent weighing factor obtained from S_{Li} of Li_2OHCl plotted in Figure S2(b).

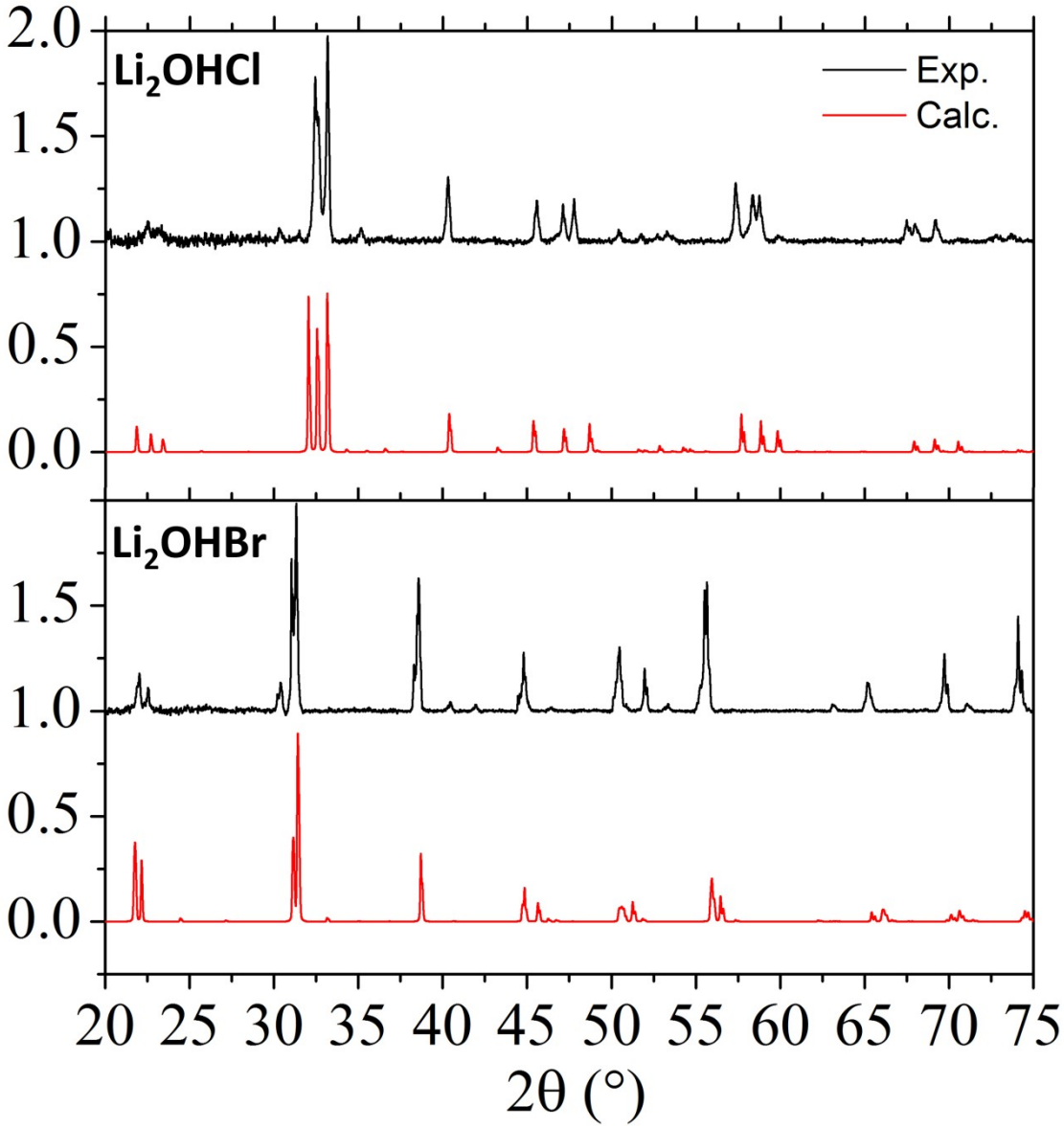

Figure S4. Simulated XRDs from the optimized structural model of Li_2OHX (X=Cl, Br) agree well with the measured ones.

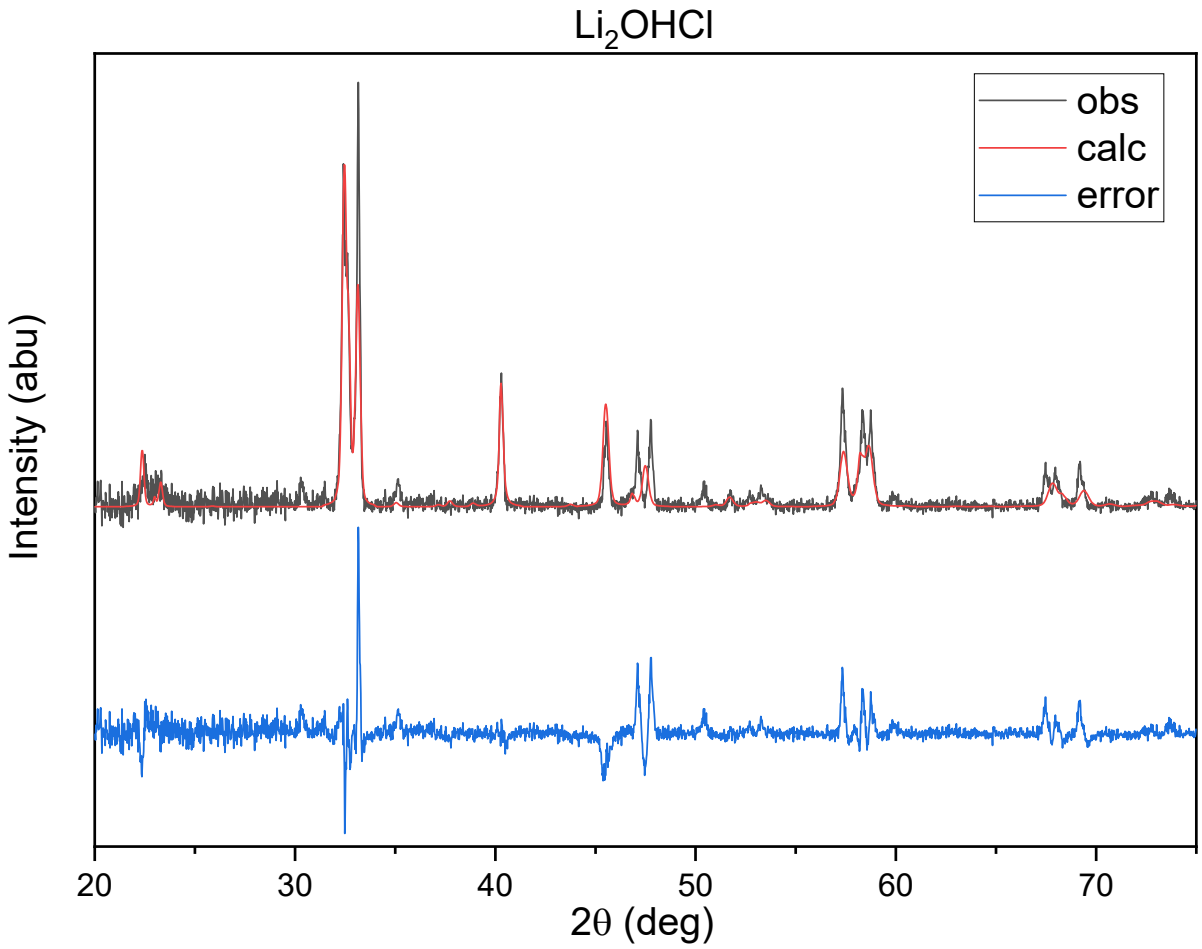

Figure S5. XRD of Li_2OHCl is primarily orthorhombic

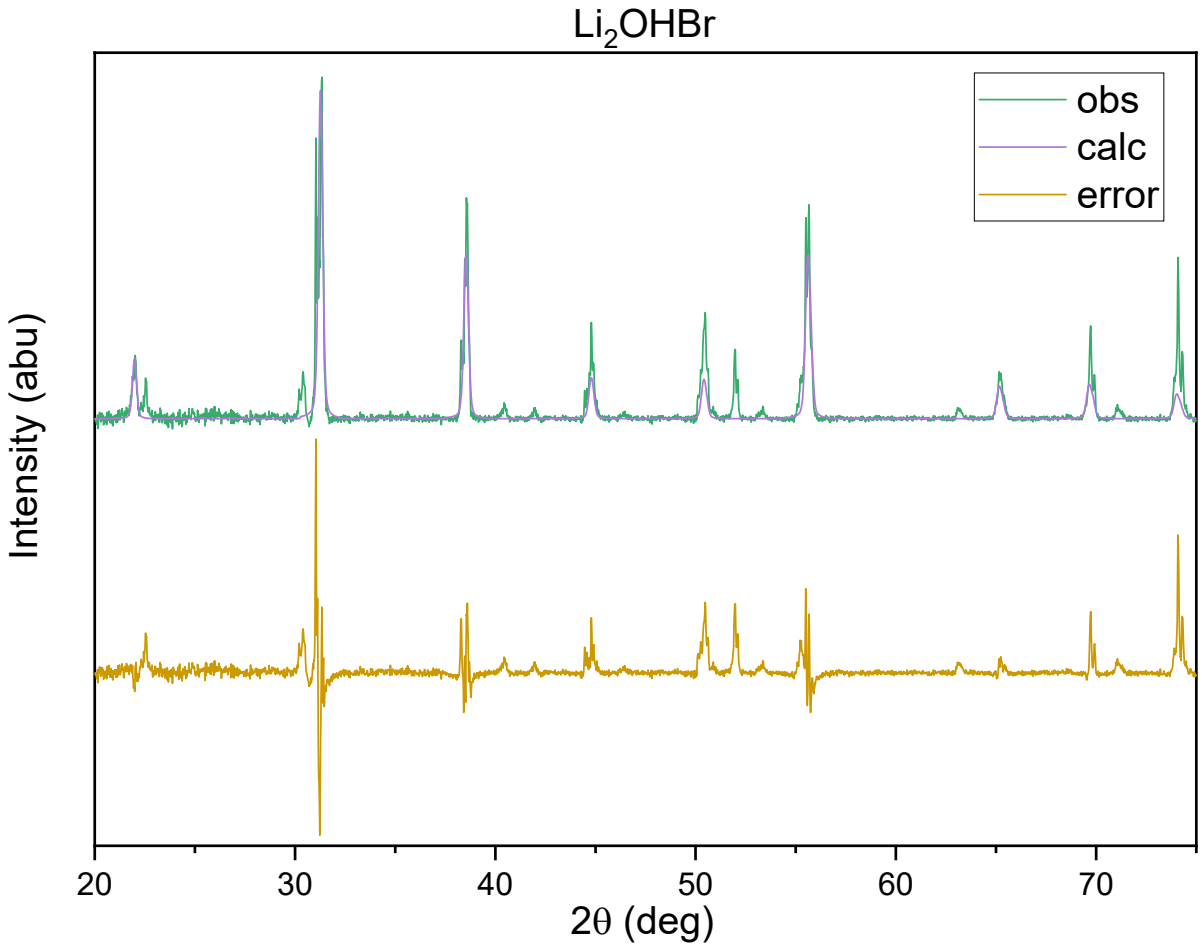

Figure S6. XRD of Li_2OHBr in cubic phase

Table S1. Lattice parameters (LP) and Li-Halide bond lengths (BL) of the DFT optimization compared to those of experiment. For the cubic phase, the DFT optimization is carried out with atomic position relaxation and the experimental lattice parameters.

Antiperovskites	LP (Å)		BL (Å)	
	Exp.	DFT	Exp.	DFT
Li_2OHCl	3.828/3.877/7.999 (Ortho)	3.697/3.890/7.922	2.664-2.785	2.386-2.747
Li_2OHBr	4.044 (Cubic)	4.044	2.862	2.499-3.237
$Li_2OHCl_{0.37}Br_{0.63}$	3.997 (Cubic)	3.997	2.831	2.389-2.511(Cl) 2.557-2.947(Br)