

Supporting Information

## New Insights into the Synthesis of Sillén–Aurivillius Oxyhalides: Molten Salt induces Interlayer Halogen Competing Reaction

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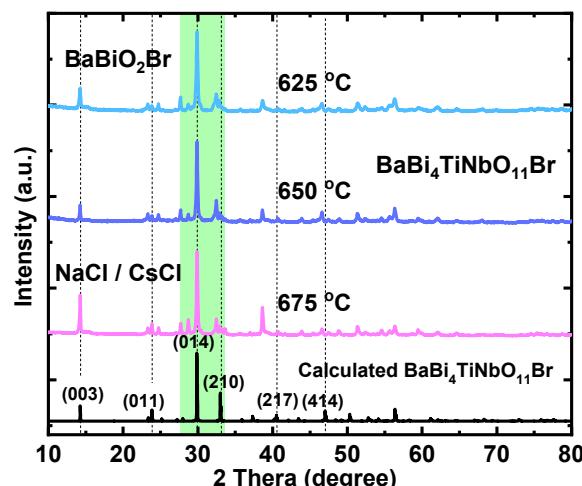
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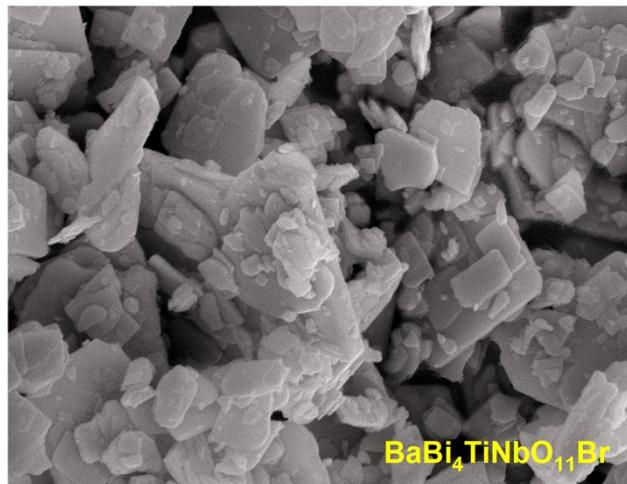
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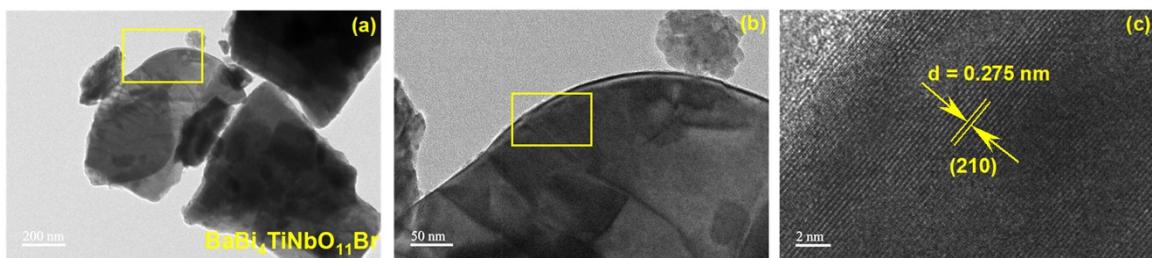
### Figure Captions



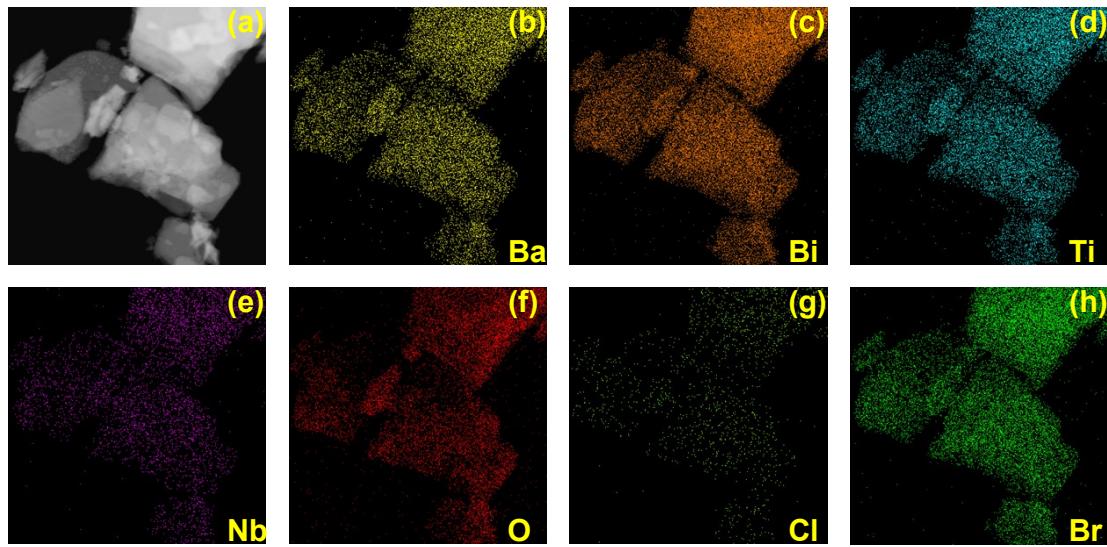
**Figure S1.** XRD patterns of  $\text{BaBi}_4\text{TiNbO}_{11}\text{Br}$  sample calcined from MSS at the temperature range of 625 - 675  $^\circ\text{C}$  for 12 h



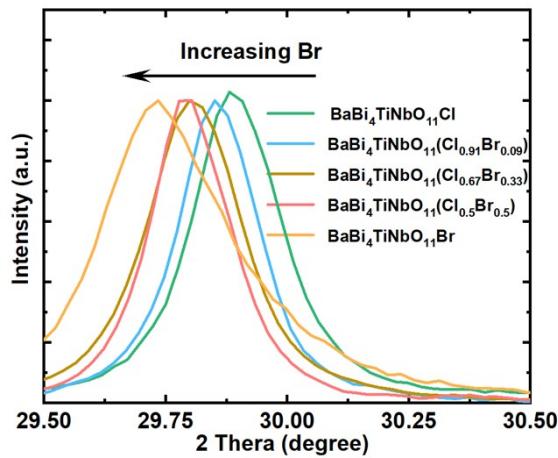
**Figure S2.** SEM image of  $\text{BaBi}_4\text{TiNbO}_{11}\text{Br}$  at the calcination temperature of 650 °C.



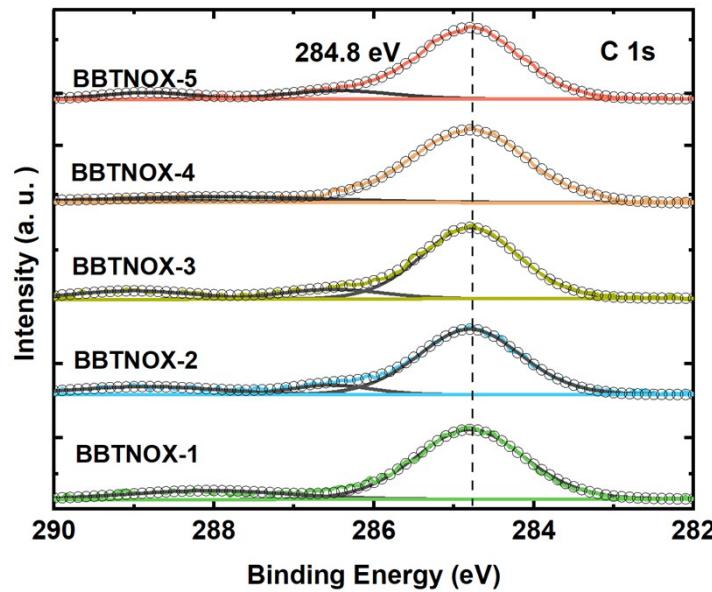
**Figure S3.** (a) TEM and (b, c) HRTEM images of  $\text{BaBi}_4\text{TiNbO}_{11}\text{Br}$ . in (c).



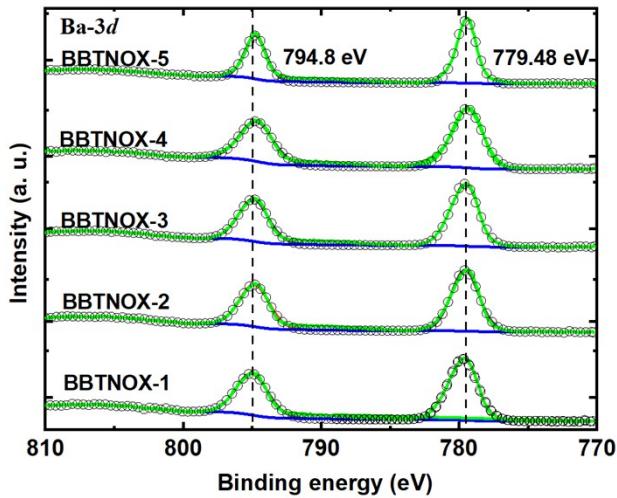
**Figure S4.** (a) TEM images of  $\text{BaBi}_4\text{TiNbO}_{11}\text{Br}$ . The corresponding TEM-EDS elemental mappings of (b) Ba, (c) Bi, (d) Ti, (e) Nb, (f) O, (g) Br, and (h) Cl.



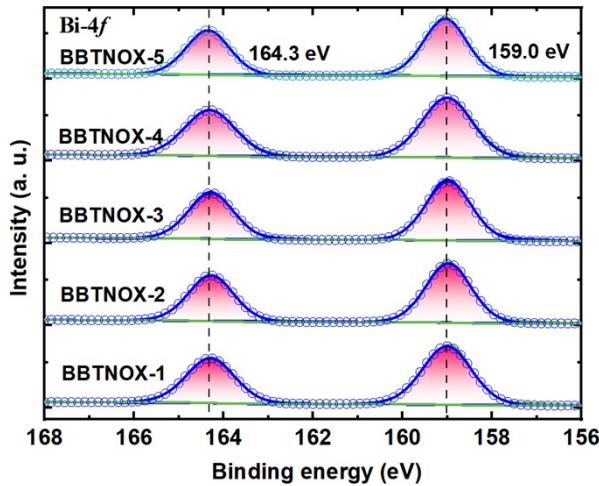
**Figure S5.** XRD patterns of  $\text{BaBi}_4\text{TiNbO}_{11}(\text{Cl}_{1-x}\text{Br}_x)$  with different Br content.



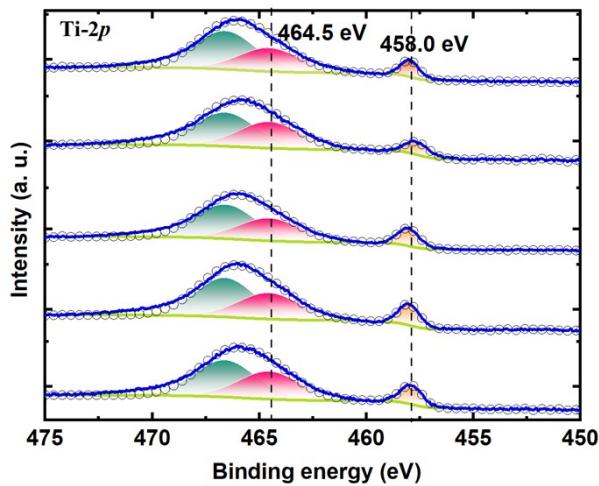
**Figure S6.** XPS peaks of the C-1s core levels.



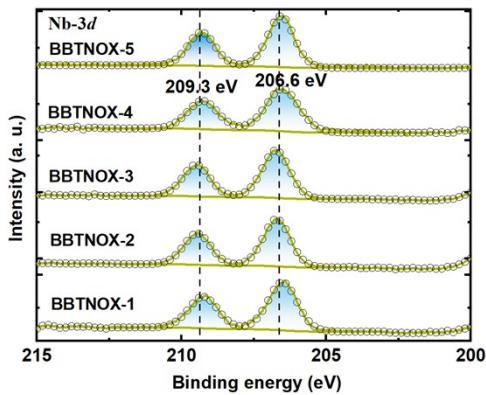
**Figure S7.** XPS peaks of the Ba-3d core levels.



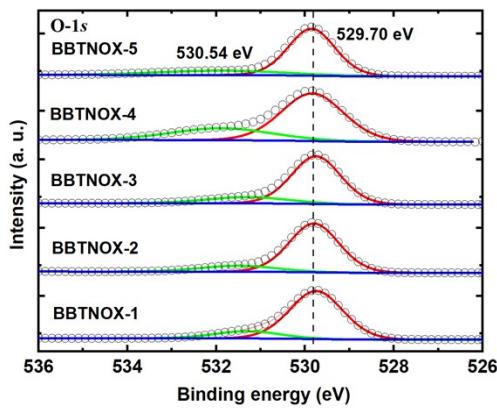
**Figure S8.** XPS peaks of the Bi-4f core levels.



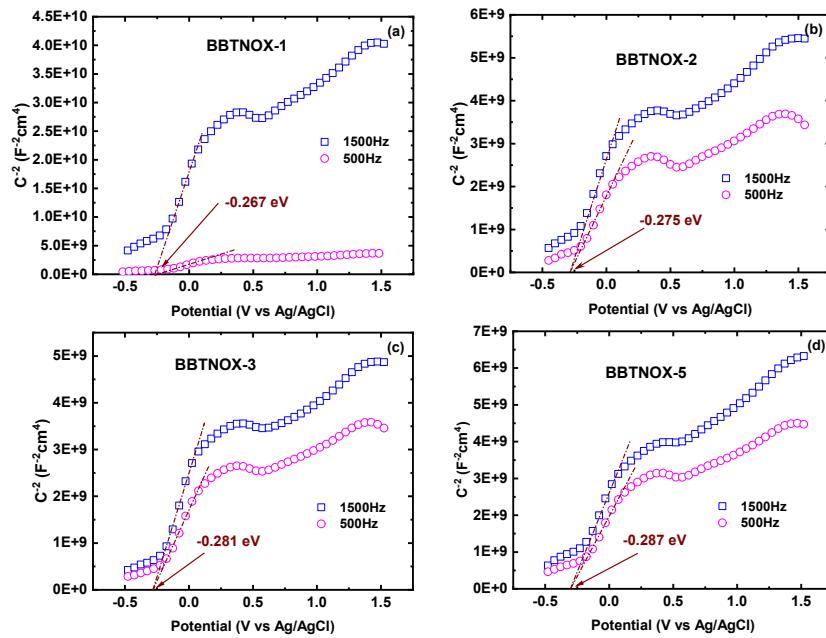
**Figure S9.** XPS peaks of the Ti-2p core levels.



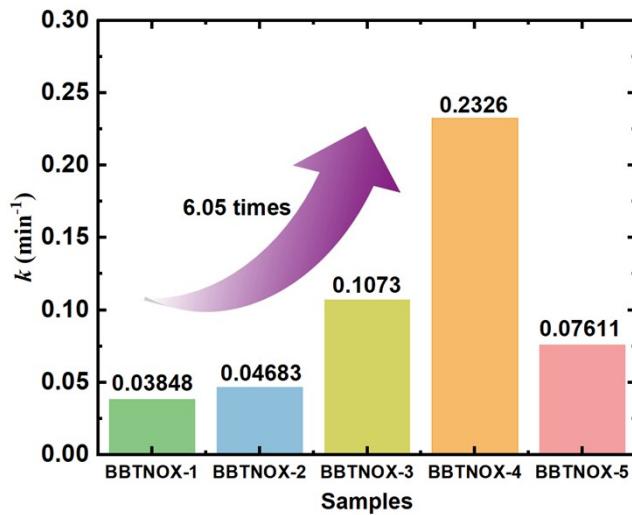
**Figure S10.** XPS peaks of the Nb-3d core levels.



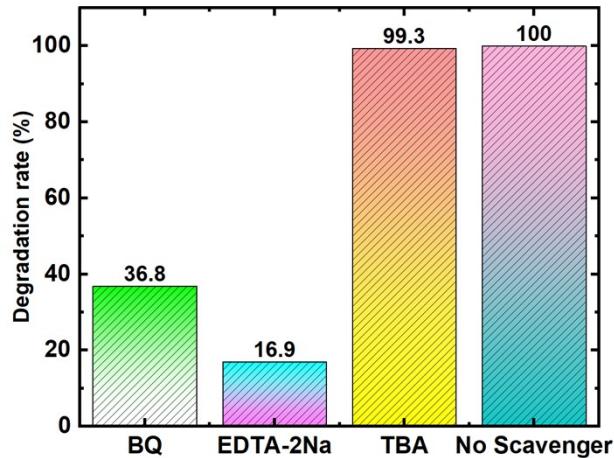
**Figure S11.** XPS peaks of the O-1s core levels.



**Figure S12.** Mott-Schottky curves of (a)  $\text{BaBi}_4\text{TiNbO}_{11}\text{Cl}$ , (b)  $\text{BaBi}_4\text{TiNbO}_{11}(\text{Cl}_{0.91}\text{Br}_{0.09})$ , (c)  $\text{BaBi}_4\text{TiNbO}_{11}(\text{Cl}_{0.67}\text{Br}_{0.33})$ , and (d)  $\text{BaBi}_4\text{TiNbO}_{11}\text{Br}$ .



**Figure S13.** The corresponding  $k$  values of samples.



**Figure S14.** The degradation rate of RhB over  $\text{BaBi}_4\text{TiNbO}_{11}(\text{Cl}_{0.5}\text{Br}_{0.5})$  with the addition of BQ, EDTA-2Na, and TBA.

### Table Captions

**Table S1.** Components of halogen oxide materials from TEM-EDS in Figure S4.

Element	Line Type	k Factor	k Factor type	Absorption Correction	Wt%	Wt% Sigma	Atomic %
O	K series	2.020		1.00	8.04	0.18	44.07
Cl	K series	1.026		1.00	2.78	0.13	6.87
Ti	K series	1.090		1.00	3.53	0.11	6.46
Br	K series	1.801		1.00	0.41	0.07	0.45

Nb	K series	3.857		1.00	6.85	0.21	6.47
Ba	L series	2.056		1.00	12.67	0.27	8.09
Bi	L series	2.469		1.00	65.73	0.34	27.59
Total:				100.00			100.00