Supporting Information

High Luminescent Antimony-based Organic-inorganic Hybrid Halides for X-ray Imaging and Detection

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Experimental Section

Materials: Antimony trichloride ($SbCl_3$, Macklin, 99%), Antimony bromide ($SbBr_3$, Macklin, 99%), Benzyltriethylammoniumchloride ([TEBA]Cl, Bide, 98%), Benzyltriethylammonium bromide ([TEBA]Br, Bide, 98%), N, N-Dimethylformamide (DMF, Macklin, AR), Acetonitrile (ACN, Aladdin, 99%). Anhydrous ethyl ether was purchased from Sinopharm Chemical Reagent Co., Ltd. (China).

Synthesis of TEBA-0: The compound was synthesized by the classical antisolvent method at room temperature. $SbCl_3$ (1 mmol, 0.228 g) and [TEBA]Cl (2 mmol, 0.228 g) were dissolved in 5 mL DMF solution and kept stirring for 8h at room temperature. Subsequently, the transparent block crystals of $(TEBA)_2SbCl_5$ were collected by slowly diffusing anhydrous ethyl ether into the DMF for 3 days and lastly dried through vacuum for 24 h. Yield: 92.5%.

Synthesis of TEBA-1: The synthesis procedure of TEBA-1 is similar to that of TEBA-0, except $SbCl_3$ (1 mmol, 0.228 g), [TEBA]Cl (1 mmol, 0.228 g) and [TEBA]Br (1 mmol, 0.272 g) were used as raw materials. Yield: 91.2%.

Synthesis of TEBA-2: The synthesis procedure of TEBA-2 is similar to that of TEBA-0, except $SbCl_3$ (1 mmol, 0.228 g) and [TEBA]Br (2 mmol, 0.544 g) were used as raw materials. Yield: 94.1%.

Synthesis of TEBA-3: The compound was prepared by cooling hot saturated solution crystallization. $SbBr_3$ (1 mmol, 0.361 g) and [TEBA]Cl (2 mmol, 0.456 g) were dissolved in 5 mL ACN solution at 60 ℃ and then kept stand for 3 days at room temperature. After that, the transparent block crystals of TEBA-3 were obtained by anhydrous ethyl ether washing and lastly dried through a vacuum for 24 h. Yield: 89.7%.

Synthesis of TEBA-4: The synthesis procedure of TEBA-4 is similar to that of TEBA-3, except $SbBr_3$ (1 mmol, 0.361 g), [TEBA]Cl (1 mmol, 0.228 g) and [TEBA]Br (1 mmol, 0.272 g) were used as raw materials. Yield: 86.4%.

Synthesis of TEBA-5: The synthesis procedure of TEBA-5 is similar to that of TEBA-3, except $SbBr_3$ (1 mmol, 0.361 g) and [TEBA]Br (2 mmol, 0.544 g) were used as raw materials. Yield: 88.5%.

Fabrication of TEBA-2 film: Firstly, TEBA-2 crystals were ground into fine powders for 30 min in the agate mortar. Then, the powders (0.3 g) were dissolved and continuously stirred in 5 mL of 0.1 g/mL toluene/PMMA mixing solution for 5 h. Lastly, the homogeneous and viscous liquid was slowly poured into the polytetrafluoroethylene mold and evaporated for approximately 12 h to obtain the thin film of TEBA-2 at room temperature.

Characterizations

Structural Characterization: The single crystal structures of $(TEBA)_2SbCl_{5-x}Br_x$ were collected on a Bruker Smart Apex CCD diffractometer at room temperature and the graphite monochrome Mo-K radiation (λ = 0.71073 Å) was used as the radiation source. The structures were solved by direct methods using SHELXS-2014[1] and refined by full-matrix least squares on F^2 using SHELXL-2014[2]. Powder X-ray diffraction (PXRD) tests of $(TEBA)_2SbCl_{5-x}Br_x$ were performed on an X-ray diffractometer D8 Advance A25. Thermogravimetric analysis (TGA) was conducted at a heating rate of 10 °C/min in an air atmosphere (30 ~ 500 °C) on the NETZSCH STA-2500 synchronous thermal analyzer. X-ray photoelectron spectroscopy (XPS) was tested by a Shimazu Axis Supra spectrometer. The C 1s peak at 284.8 eV was used as the internal standard for XPS measurements.

Photoluminescence Measurements: The photoluminescence excitation (PLE) spectra, photoluminescence (PL) emission spectra, emission lifetimes and photoluminescence quantum yields (PLQYs) of $(TEBA)_2SbCl_{5-x}Br_x$ crystals were obtained on an Edinburgh Instrument FLS980 Spectrophotometer. UV-visible absorption spectra were measured on a Shimadzu UV-2600 spectrophotometer.

Scintillation Characterization: The radioluminescence (RL) spectra and linear response to X-ray dose rate were measured with a spectrofluorometer (FLS980, Edinburgh Instruments) under irradiation of X-ray source (Mini-X2, Amptek; Au target; tube voltage, 50 kV). The X-ray dose rates were altered by adjusting the current of the X-ray tube (5-70 μA). In advance, the dose rates were calibrated by an X-ray ion chamber dose meter (Radcal Accu-DOSE$^+$, 10X6-60).

Determination of X-ray excited relative light yields: The X-ray relative light yields of $(TEBA)_2SbCl_{5-x}Br_x$ single crystals were measured with the help of commercial LuAG: Ce as a reference. $(TEBA)_2SbCl_{5-x}Br_x$ with a thickness of 100 μm and LuAG: Ce were placed in the same position. The corresponding photon counting results ($PC_{measured}$) were then obtained by integrating the steady-state XEL spectra. Compared with reference LuAG: Ce, the corresponding relative light yields of $(TEBA)_2SbCl_{5-x}Br_x$ were calculated according to equation (1) and (2). The equations were given as follows:

$$PC_{normalized} = \frac{PC_{measured}}{AE(d)} \quad (1)$$

$$LY_s = LY_{LUAG:Ce}\frac{PC_{normalized}(S)}{PC_{normalized}(LUAG:Ce)} \quad (2)$$

where $LY_{LuAG:\,Ce}$ is the light yield of LuAG: Ce (22000 photons MeV^{-1}), $PC_{normalized}$ (S) and $PC_{normalized}$ (LuAG: Ce) are the photon counts of our scintillators and LuAG: Ce normalized to respective X-ray attenuation efficiencies.

Table S1. Detailed Single crystal X-ray data of TEBA-0, TEBA-1, and TEBA-2.

Complex	**TEBA-0**	**TEBA-1**	**TEBA-2**
Chemical Formula	$C_{26}H_{44}Cl_5N_2Sb$	$C_{26}H_{44}Br_{0.8}Cl_{4.2}N_2Sb$	$C_{26}H_{44}Br_{1.6}Cl_{3.4}N_2Sb$
Formula Weight	683.68	719.46	753.9
Crystal System	$P2_12_12_1$	$P2_12_12_1$	$P2_12_12_1$
Space Group	orthorhombic	orthorhombic	orthorhombic
a (Å)	9.3198(7)	9.3120(15)	9.3572(9)
b (Å)	13.7258(10)	13.829(3)	13.8687(13)
c (Å)	24.4550(17)	24.661(4)	24.805(2)

α (°)	90	90	90
Complex	**TEBA-0**	**TEBA-1**	**TEBA-2**
β (°)	90	90	90
γ (°)	90	90	90
V (Å^3)	3128.3(4)	3175.8(10)	3218.9(5)
Z value	4	4	4
D(calcd) (g cm^{-3})	1.452	1.505	1.556
Temperature (K)	297.0 K	295.0 K	295.0 K
F(000)	1400.0	1457.9	1510.3
μ (mm^{-1})	1.327	2.252	3.145
R_1/wR_2 [I>2σ(I)]	R_1=0.0372 wR_2=0.0605	R_1=0.0252 wR_2=0.0578	R_1=0.0525 wR_2=0.0953
R_1/wR_2 [all data]	R_1=0.0555 wR_2=0.0671	R_1=0.0288 wR_2=0.0594	R_1=0.0729 wR_2=0.1012
GOF	0.997	1.045	1.048

Table S2. Detailed Single crystal X-ray data of TEBA-3, TEBA-4 and TEBA-5.

Complex	**TEBA-3**	**TEBA-4**	**TEBA-5**
Chemical Formula	$C_{26}H_{44}Br_{3.04}Cl_{1.96}N_2Sb$	$C_{26}H_{44}Br_{3.97}Cl_{1.03}N_2Sb$	$C_{26}H_{44}Br_5N_2Sb$
Formula Weight	817.03	859.81	905.93

Crystal System	$P2_12_12_1$	$P2_12_12_1$	$P2_12_12_1$
Complex	**TEBA-3**	**TEBA-4**	**TEBA-5**
Space Group	orthorhombic	orthorhombic	orthorhombic
a (Å)	9.4310(3)	9.3820(3)	9.747(10)
b (Å)	14.0220(4)	25.1850(8)	14.399(14)
c (Å)	25.166(7)	13.979(5)	25.67(2)
α (°)	90	90	90
β (°)	90	90	90
γ (°)	90	90	90
V (Å^3)	3328.0(16)	3302.7(19)	3603.0(6)
Z value	4	4	4
D(calcd) (g cm^{-3})	1.631	1.729	1.670
Temperature (*K*)	296.0 K	297.0 K	296.0 K
F(000)	1613.0	1681.3	1754.3
μ (mm^{-1})	4.608	5.730	6.326
R_1/wR_2 [I>2σ(I)]	R_1=0.0552 wR_2=0.110	R_1=0.0771 wR_2=0.1436	R_1=0.0497 wR_2=0.0849
R_1/wR_2 [all data]	R_1=0.0753 wR_2=0.1180	R_1=0.1214 wR_2=0.1585	R_1=0.0882 wR_2=0.0957
GOF	1.063	1.023	1.000

Table S3. The cell parameters of $(TEBA)_2SbCl_{5-x}Br_x$ single crystals.

	TEBA-0	TEBA-1	TEBA-2	TEBA-3	TEBA-4	TEBA-5
Cell volume	3128.32	3175.74	3219	3327.99	3303.04	3602.71
$\delta d = 1/5\sum [(d_i-d)/d]^2$	1.67442*10³	2.62247*10³	3.23355*10³	3.00529*10³	2.28386*10³	1.64349*10³

Table S4. Fractional Atomic Coordinates (×10⁴) for TEBA-0.

Atom	X	Y	Z
Sb1	3215.35(16)	705.58(11)	3391.69(6)
Cl1	2698.0(7)	2387.1(4)	3499.7(3)
Cl5	5076.0(7)	722.0(6)	4210.4(3)
Cl4	1009.9(8)	415.7(5)	4067.3(3)
Cl3	1596.7(8)	610.6(5)	2531.1(3)
Cl2	5278.2(8)	1236.6(6)	2767.9(3)
N1	1903(2)	3422.1(13)	1719.4(8)
N2	7570(2)	3410.0(15)	4051.6(8)
C12	664(3)	2775.9(18)	1888.3(11)
C19	5142(2)	4269(2)	3968.4(9)
C7	1908(3)	3586.4(17)	1099.4(10)
C8	3250(3)	2893.0(17)	1907.1(11)
C6	1881(3)	2691.8(17)	752.9(10)
C20	6041(3)	3397.1(19)	3833.8(10)
C25	8625(3)	2406(2)	3262.4(12)
C3	1800(4)	1107(2)	38.6(11)
C24	8315(3)	2476.4(19)	3869.0(11)
C13	-824(3)	3187(2)	1801.2(13)
C26	7578(3)	3447(2)	4674.4(10)

Atom	X	Y	Z
C5	3114(3)	2208(2)	603.8(12)
C10	1790(3)	4433.0(16)	1971.4(10)
C21	8318(3)	4311(2)	3836.9(10)
C1	588(3)	2358(2)	536.1(12)
C4	3077(4)	1421(2)	251.2(13)
C9	4644(3)	3444(2)	1825.5(14)
C17	4373(3)	4326(2)	4453.0(12)
C23	9913(3)	4381(2)	3977.7(13)
C11	1836(4)	4447(2)	2590.5(11)
C18	4965(3)	5005(2)	3589.2(12)
C22	6914(4)	2584(2)	4959.7(12)
C14	4061(3)	5778(2)	3691.5(13)
C16	3469(3)	5104(3)	4555.5(14)
C15	3326(3)	5831(2)	4175.7(14)
C2	554(4)	1570(2)	182.7(13)

Table S5. Fractional Atomic Coordinates ($\times 10^4$) for TEBA-1.

Atom	X	Y	Z
Cl1	4813.9(7)	687.1(6)	5774.8(2)
Br1	9031.1(7)	419.1(5)	5919.6(3)
Cl4	7283.4(9)	2390.2(7)	6502.0(3)
Br4	8414.1(9)	603.9(7)	7472.5(3)
Br3	4699.7(9)	1241.4(8)	7242.4(3)
N2	2404(3)	3425(2)	5949.9(11)
N1	8079(3)	3416.3(19)	8279.6(11)
C8	9313(4)	2770(3)	8111.1(15)
C20	3947(4)	3404(3)	6161.4(14)
C25	1677(4)	4323(3)	6168.7(14)

Atom	X	Y	Z
C21	2387(4)	3466(3)	5330.6(13)
C7	8081(4)	3579(3)	8894.0(13)
C23	1656(4)	2499(3)	6131.2(15)
C18	5026(2)	4996.2(19)	6412.7(7)
C19	4862(2)	4268.8(16)	6028.7(9)
C14	5610(3)	4315.3(18)	5541.7(8)
C15	6523(3)	5089(2)	5438.7(9)
C16	6687(3)	5816.4(18)	5822.8(12)
C17	5939(3)	5769.9(17)	6309.7(11)
C12	6746(4)	2885(3)	8093.0(15)
C26	82(4)	4406(4)	6034.5(18)
C9	10812(4)	3178(3)	8196.3(17)
C22	3023(5)	2608(3)	5044.1(16)
C10	8190(4)	4417(3)	8027.8(13)
C1	9415(2)	2367.7(19)	9455.6(10)
C6	8112(3)	2681.7(15)	9244.6(9)
C5	6846(2)	2214.4(18)	9387.8(10)
C4	6884(3)	1433.0(18)	9742.0(11)
C3	8187(3)	1118.9(16)	9953.0(9)
C2	9453(2)	1586(2)	9809.8(11)
C24	1364(4)	2426(3)	6740.1(16)
C11	8152(5)	4430(3)	7415.0(15)
C13	5335(4)	3439(3)	8174.6(19)
Br2	4813.9(7)	687.1(6)	5774.8(2)
Cl2a	8414.1(9)	603.9(7)	7472.5(3)
Cl5a	9031.1(7)	419.1(5)	5919.6(3)
Cl3a	4699.7(9)	1241.4(8)	7242.4(3)
Br5	7283.4(9)	2390.2(7)	6502.0(3)

Table S6. Fractional Atomic Coordinates ($\times 10^4$) for TEBA-2.

Atom	X	Y	Z
Sb1	3235.1(4)	9279.8(2)	3385.97(13)
Br4	5230.7(8)	9323.9(6)	4231.2(3)
Br1	953.3(9)	9579.3(6)	4087.1(3)
Cl4	1577.7(13)	9407.0(7)	2523.6(4)
Cl1	5328.7(13)	8751.5(9)	2750.3(5)
Cl5	2729.9(14)	7602.8(8)	3501.5(5)
N1	7611(4)	6563(3)	4049.9(17)
N2	1935(4)	6592(3)	1719.7(17)
C7	6075(5)	6597(4)	3844(2)
C25	701(5)	7235(4)	1891(2)
C12	8322(5)	5666(4)	3830(2)
C8	7645(6)	6514(4)	4659(2)
C26	-808(5)	6820(4)	1803(3)
C10	8370(7)	7484(4)	3867(2)
C5	4979(4)	5012(3)	3591.3(12)
C6	5152(3)	5736(2)	3973.3(14)
C1	4411(4)	5692(3)	4458.3(13)
C2	3496(4)	4924(3)	4561.3(14)
C3	3323(4)	4200(2)	4179.3(18)
C4	4065(4)	4244(2)	3694.3(16)
C9	7023(8)	7383(4)	4952(3)
C13	9923(6)	5571(5)	3958(3)
C23	1830(6)	5596(3)	1968.0(19)
C21	3266(6)	7115(4)	1907(2)
C20	1929(6)	6424(3)	1111(2)
C24	1875(7)	5594(4)	2579(2)
C11	8639(7)	7552(5)	3260(2)

Atom	X	Y	Z
C18	597(3)	7630(3)	548.2(16)
C19	1895(4)	7320(2)	758.8(13)
C14	3154(3)	7786(3)	614.9(16)
C15	3115(4)	8562(3)	260.6(16)
C16	1817(5)	8872(2)	50.0(14)
C17	559(4)	8406(3)	193.8(16)
C22	4673(6)	6565(4)	1823(3)
Cl3	953.3(9)	9579.3(6)	4087.1(3)
Cl2	5230.7(8)	9323.9(6)	4231.2(3)
Br2	1577.7(13)	9407.0(7)	2523.6(4)
Br3	5328.7(13)	8751.5(9)	2750.3(5)
Br5a	2729.9(14)	7602.8(8)	3501.5(5)

Table S7. Fractional Atomic Coordinates ($\times 10^4$) for TEBA-3.

Atom	X	Y	Z
Sb1	6736.6(6)	9263.2(4)	3391.3(2)
Br1	4730.2(11)	9322.1(9)	4235.4(4)
Br4	9026.3(12)	9588.8(8)	4092.3(4)
Br2	8426.4(16)	9422.6(10)	2520.4(5)
Cl1	4607.3(16)	8732.1(12)	2741.6(5)
Cl2	7249.9(19)	7570.3(13)	3506.9(7)
N1	2347(7)	6548(5)	4051(2)
N2	8058(7)	6594(5)	1715(3)
C1	4810(6)	5732(4)	3983(2)
C26	5534(7)	5693(5)	4463(2)
C25	6455(7)	4941(5)	4567(2)
C3	6653(7)	4230(4)	4189(3)
C13	5929(7)	4270(4)	3709(3)
C8	5008(7)	5021(5)	3605.3(18)

Atom	X	Y	Z
C4	1646(10)	5651(7)	3832(3)
C6	1313(11)	7512(8)	3267(4)
C10	2294(10)	6516(7)	4652(3)
C12	10803(10)	6845(8)	1802(4)
C14	6710(11)	7118(7)	1895(4)
C16	3878(9)	6592(7)	3849(3)
C18	1577(12)	7453(7)	3865(4)
C20	9272(10)	7227(7)	1882(4)
C22	8118(12)	5610(7)	2565(3)
C24	8154(10)	5618(6)	1964(3)
C7	8072(10)	6442(6)	1106(3)
C2	50(11)	5554(8)	3945(4)
C17	6870(6)	7781(5)	620(2)
C5	8119(7)	7317(3)	759(2)
C11	9404(6)	7618(5)	547(3)
C23	9440(7)	8383(5)	196(2)
C15	8191(9)	8846(4)	56(2)
C21	6906(7)	8545(4)	269(3)
C9	2883(14)	7350(7)	4949(4)
C19	5302(12)	6584(8)	1806(4)
Cl3a	9026.3(12)	9588.8(8)	4092.3(4)
Cl2a	8426.4(16)	9422.6(10)	2520.4(5)
Br1a	4607.3(16)	8732.1(12)	2741.6(5)
Br3	7249.9(19)	7570.3(13)	3506.9(7)
Cl5	4730.2(11)	9322.1(9)	4235.4(4)

Table S8. Fractional Atomic Coordinates ($\times 10^4$) for TEBA-4.

Atom	X	Y	Z
Sb1	5786.9(8)	4102.2(3)	6753.9(5)
Br1	7785.3(14)	3263.2(5)	6814.8(9)
Br3	7928.4(16)	4760.7(6)	6216.9(11)
Cl2	5255.2(19)	3986.2(7)	5035.3(11)
N1	4443(9)	5801(4)	4104(5)
N2	10178(9)	3452(3)	4028(5)
C1	4334(14)	5545(4)	3111(7)
C4	5786(13)	5629(5)	4620(8)
C6	1741(13)	5691(5)	4350(9)
C8	7715(7)	3510(3)	3231(5)
C10	7000(9)	3027(3)	3199(5)
C12	6073(9)	2917(3)	2450(7)
C14	5860(8)	3290(4)	1732(5)
C16	6574(9)	3774(3)	1763(5)
C18	7501(8)	3884(2)	2513(6)
C20	4409(13)	6407(4)	3942(7)
C22	8651(12)	3654(4)	4078(8)
C24	11232(15)	4248(6)	4959(10)
C26	10873(12)	3678(5)	3126(8)
C13	3229(13)	5623(5)	4734(8)
C7	4421(15)	4955(5)	3125(8)
C15	10931(14)	3645(5)	4911(8)
C2	10251(13)	2849(5)	4001(8)
C17	12475(12)	3556(6)	3012(9)
C9	7169(14)	5711(6)	4066(9)
C19	3063(7)	6956(3)	5100(6)
C5	3006(9)	7307(3)	5867(6)

Atom	X	Y	Z
C21	4251(11)	7448(3)	6344(4)
C11	5551(9)	7239(4)	6053(5)
C23	5608(7)	6888(3)	5287(5)
C3	4363(9)	6747(3)	4810(4)
C25	9659(17)	2558(6)	4847(9)
Br1A	5255.2(19)	3986.2(7)	5035.3(11)
Cl3	7928.4(16)	4760.7(6)	6216.9(11)
Br4A	3501.7(15)	3405.0(5)	7094.9(9)
Br2C	4075.6(17)	4975.8(6)	6925.7(9)
Cl4	4075.6(17)	4975.8(6)	6925.7(9)
Cl5	3501.7(15)	3405.0(5)	7094.9(9)
Cl1	7785.3(14)	3263.2(5)	6814.8(9)

Table S9. Fractional Atomic Coordinates ($\times 10^4$) for TEBA-5.

Atom	X	Y	Z
Sb1	6698.0(5)	756.4(3)	3401.82(15)
Br5	7244.6(8)	2502.7(5)	3522.4(3)
Br1	4706.7(8)	692.1(6)	4237.0(2)
Br2	8967.7(8)	397.3(6)	4096.0(3)
Br4	4555.8(8)	1291.6(6)	2736.7(3)
Br3	8419.2(9)	576.8(5)	2529.1(3)
N1	2303(5)	3498(4)	4043.7(19)
N2	1949(6)	8405(4)	3303(2)
C6	4739(6)	4273(5)	3989(2)
C23	1811(8)	9388(5)	3057(3)
C10	1628(7)	4394(5)	3828(3)
C5	5486(7)	4313(6)	4480(3)
C7	3813(7)	3436(5)	3857(3)

Atom	X	Y	Z
C1	5008(7)	4987(6)	3628(3)
C25	3286(8)	7898(5)	3136(3)
C9	2217(8)	3519(5)	4646(2)
C24	1902(9)	9390(5)	2461(3)
C20	1922(8)	8563(5)	3909(2)
C12	1283(9)	2573(7)	3249(3)
C13	1536(9)	2617(6)	3847(3)
C16	3088(8)	7205(5)	4393(3)
C22	-761(8)	8151(6)	3203(3)
C15	3026(10)	6452(6)	4733(3)
C3	6628(8)	5759(6)	4208(3)
C21	744(8)	7755(5)	3133(3)
C2	5909(9)	5728(7)	3737(3)
C26	4660(8)	8446(6)	3227(3)
C17	1864(8)	7695(5)	4245(2)
C8	2799(10)	2661(6)	4931(3)
C4	6396(8)	5046(7)	4577(3)
C14	1756(10)	6163(6)	4943(3)
C11	44(8)	4511(6)	3941(3)
C19	545(9)	6639(6)	4803(3)
C18	609(8)	7388(6)	4463(3)

Table S10. Atomic Occupancy for TEBA-1, TEBA-2, TEBA-3 and TEBA-4.

Atom (TEBA -1)	Occupancy	Atom (TEBA -2)	Occupancy	Atom (TEBA -3)	Occupancy	Atom (TEBA -4)	Occupancy
Cl1	0.658(3)	Br4	0.602(4)	Br1	0.872(7)	Br1	0.939(9)

Atom (TEBA -1)	Occupancy	Atom (TEBA -2)	Occupancy	Atom (TEBA -3)	Occupancy	Atom (TEBA -4)	Occupancy
Br1	0.262(3)	Br1	0.514(5)	Br4	0.828(8)	Br3	0.792(9)
Cl4	0.991(3)	Cl4	0.760(4)	Br2	0.602(7)	Cl2	0.524(9)
Br4	0.097(3)	Cl1	0.790(4)	Cl1	0.471(7)	Br1A	0.476(9)
Br3	0.078(3)	Cl5	0.964(4)	Cl2	0.793(7)	Cl3	0.208(9)
Br2	0.342(3)	Cl3	0.486(5)	Cl3a	0.172(8)	Br4A	0.922(9)
Cl2a	0.903(3)	Cl2	0.398(4)	Cl2a	0.398(7)	Br2C	0.837(8)
Cl5a	0.738(3)	Br2	0.240(4)	Br1a	0.529(7)	Cl4	0.163(8)
Cl3a	0.922(3)	Br3	0.210(4)	Br3	0.207(7)	Cl5	0.078(9)
Br5	0.009(3)	Br5a	0.036(4)	Cl5	0.128(7)	Cl1	0.061(9)

Table·S11. Comparison of Cl:Br ratios of measured by SCXRD·and EDS.

	Cl:Br	Cl:Br	Cl:Br	Cl:Br	Cl:Br	Cl:Br
Raw materials ratio	5:0	4:1	3:2	2:3	1:4	0:5
SCXRD	5:0	4.2:0.8	3.4:1.6	1.96:3.04	1.03:3.97	0:5
EDS	5:0	92.5:19.9	50.3:23.5	22.9:35.5	15.6:64.7	0:5

Table S12. The optical parameters of $(TEBA)_2SbCl_{5-x}Br_x$ powders.

	PLE (nm)	PL (nm)	FWHM (nm)	Stokes shift (nm)	Lifetime (μs)	PLQY (%)	Eg (eV)
TEBA-0	371	586	117	215	3.83	99.25	3.16
TEBA-1	382	598	125	216	2.72	98.10	2.99
TEBA-2	385	603	128	218	2.04	73.83	2.95

	PLE (nm)	PL (nm)	FWHM (nm)	Stokes shift (nm)	Lifetime (μs)	PLQY (%)	Eg (eV)
TEBA-3	395	621	129	226	1.26	37.90	2.93
TEBA-4	401	627	132	226	0.84	17.86	2.88
TEBA-5	404	635	132	231	0.29	2.8	2.84

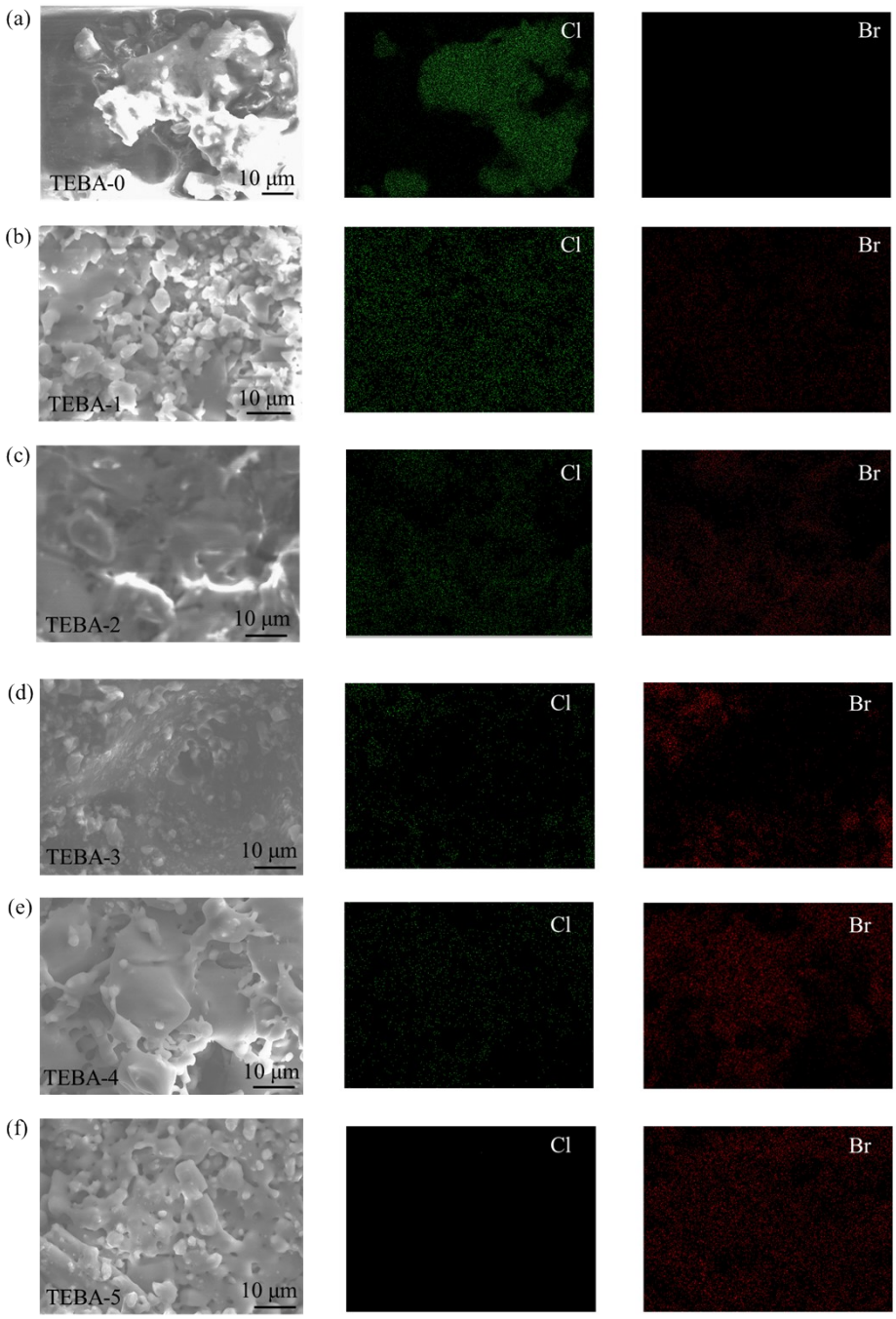

Figure S1 EDS elemental mapping of $(TEBA)_2SbCl_{5-x}Br_x$. (a) TEBA-0, (b) TEBA-1, (c) TEBA-2,

(d) TEBA-3, (e) TEBA-4 and (f) TEBA-5.

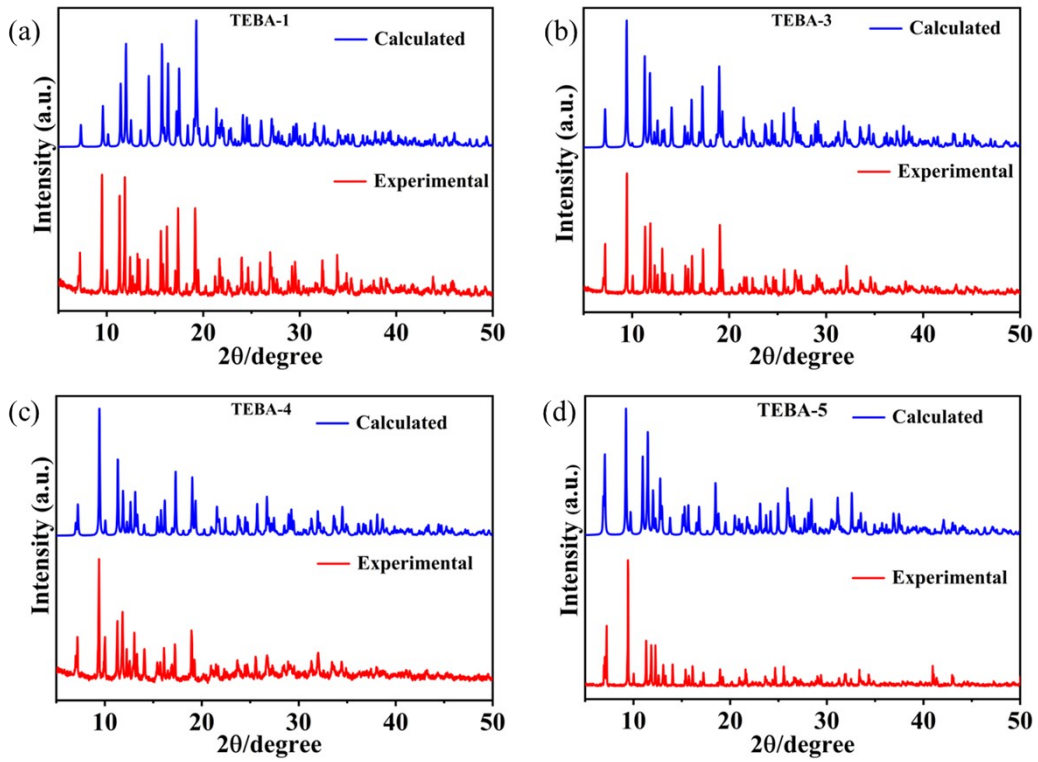

Figure S2. PXRD patterns and simulated patterns of SCXRD of TEBA-1, TEBA-3, TEBA-4, and TEBA-5.

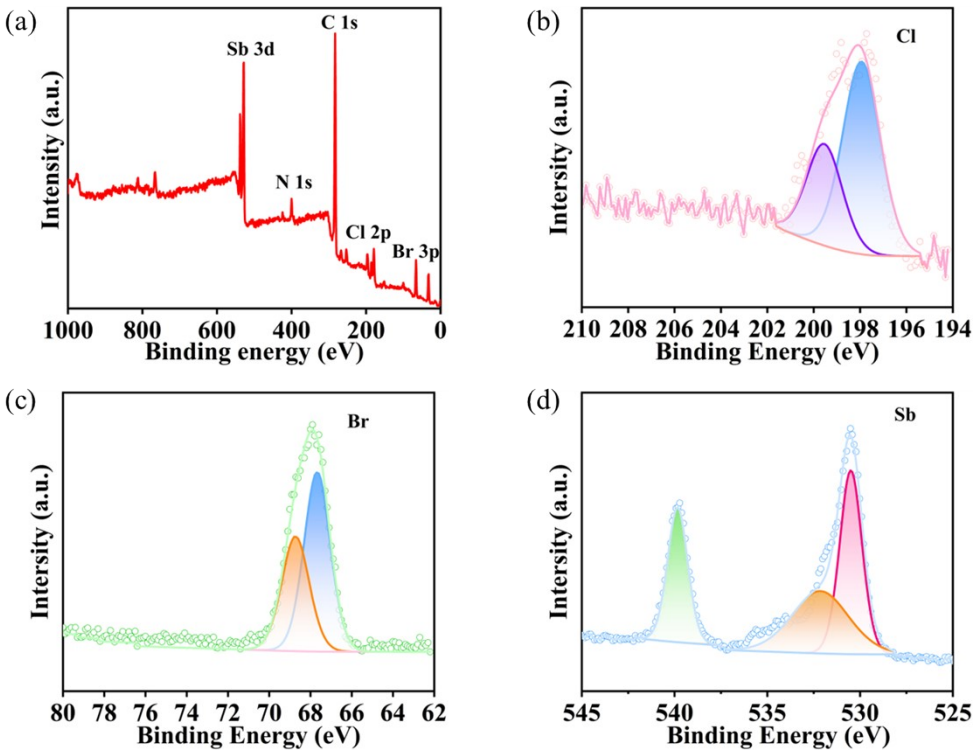

Figure S3. (a) X-ray photoelectron spectroscopy (XPS) of TEBA-2. High-resolution spectra of (b)

Cl 2p, (c) Br 3p, and (d) Sb 3d.

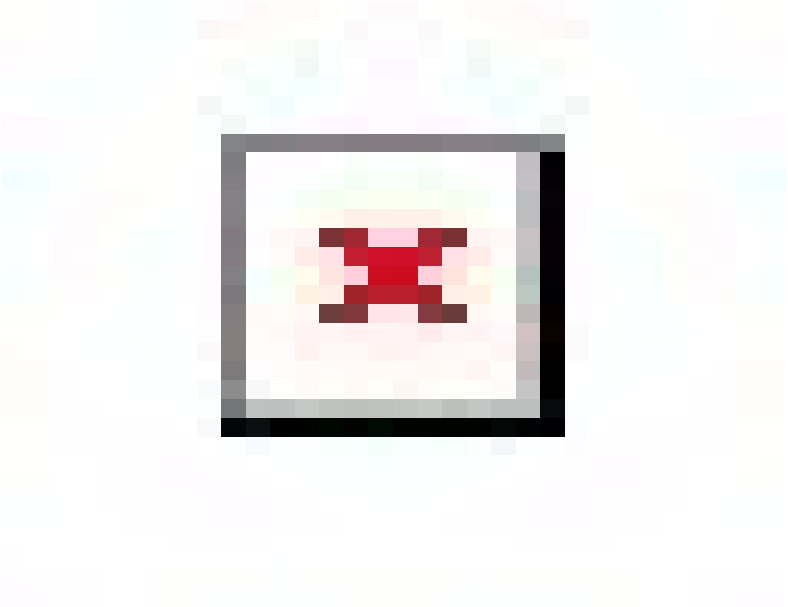

Figure S4. TGA curves of $(TEBA)_2SbCl_{5-x}Br_x$.

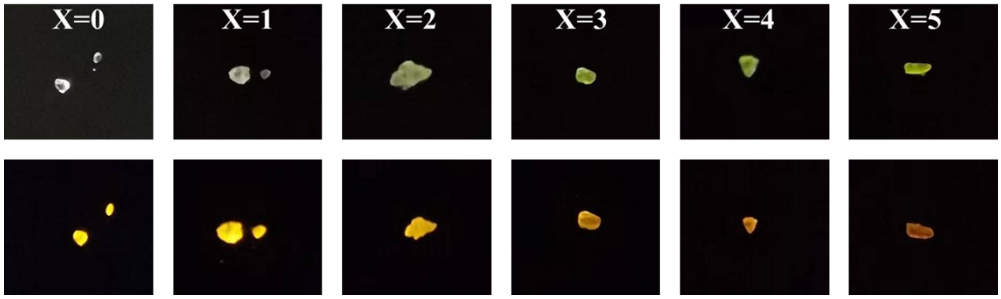

Figure S5. The photographs of $(TEBA)_2SbCl_{5-x}Br_x$ single crystals under ambient light and ultraviolet light of 365 nm.

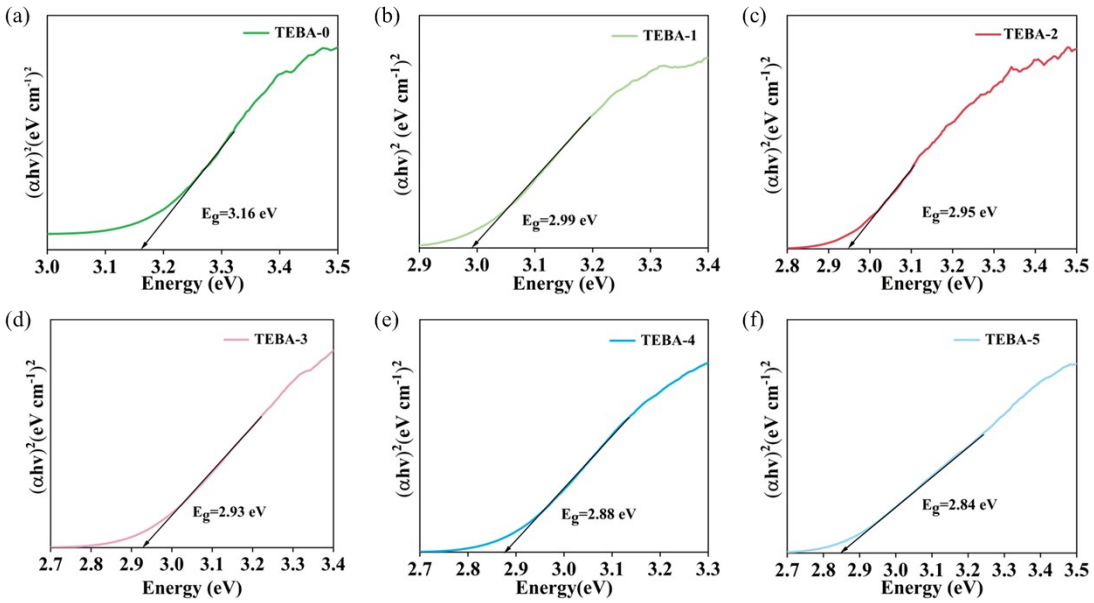

Figure S6. The energy gaps of $(TEBA)_2SbCl_{5-x}Br_x$ single crystals. (a) TEBA-0. (b) TEBA-1. (c) TEBA-2. (d) TEBA-3. (e) TEBA-4. (f) TEBA-5.

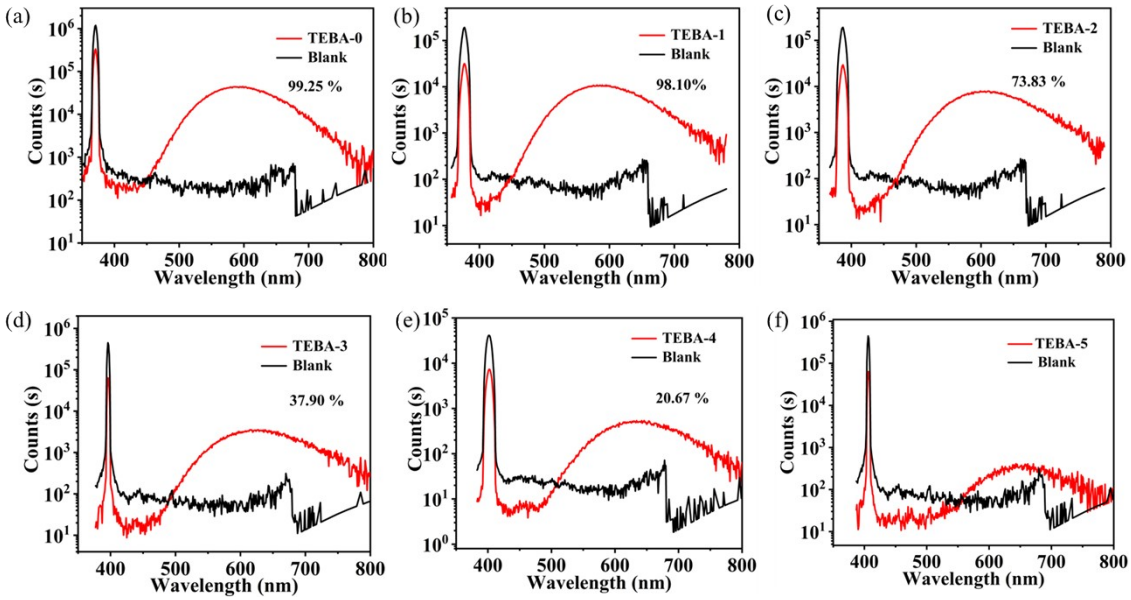

Figure S7. The PLQYs of $(TEBA)_2SbCl_{5-x}Br_x$ single crystals. (a) TEBA-0. (a) TEBA-1. (a) TEBA-2. (a) TEBA-3. (a) TEBA-4. (a) TEBA-5.

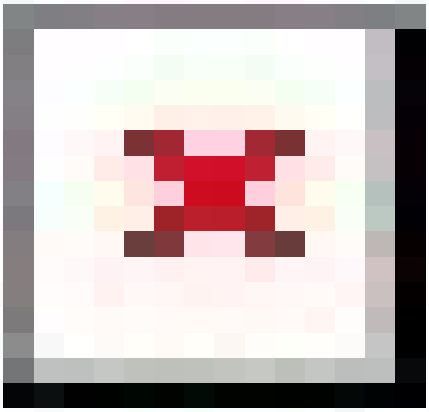

Figure S8. The 1931 Commission Internationale de L'Eclairage (CIE) chromaticity diagram of $(TEBA)_2SbCl_{5-x}Br_x$.

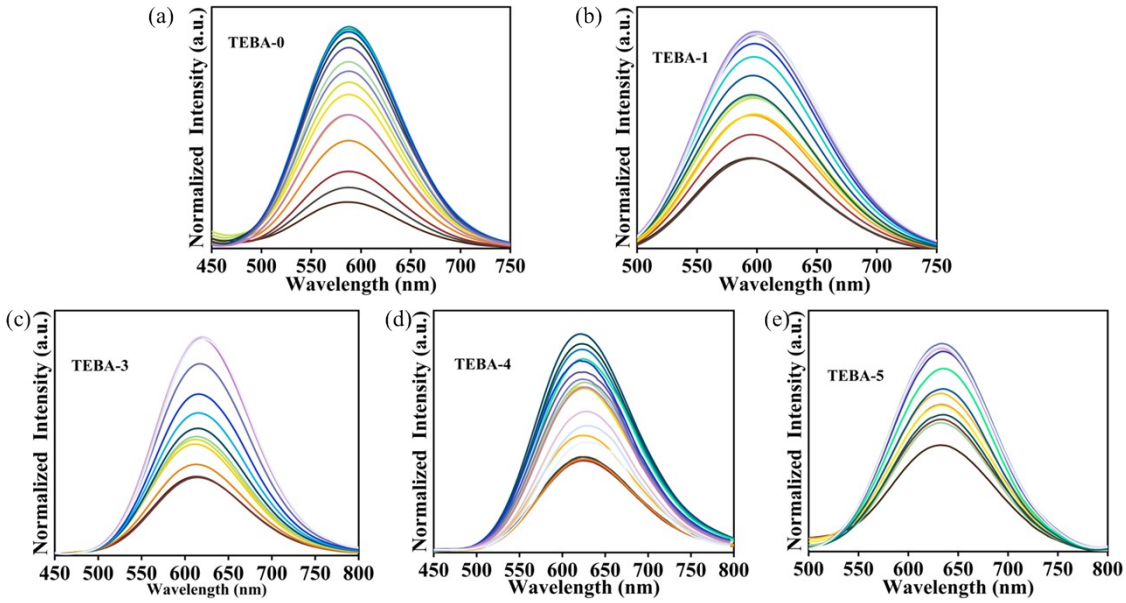

Figure S9. Excitation-wavelength dependent PL spectra of (a) TEBA-0. (b) TEBA-1. (c) TEBA-3. (d) TEBA-4. and (e) TEBA-5.

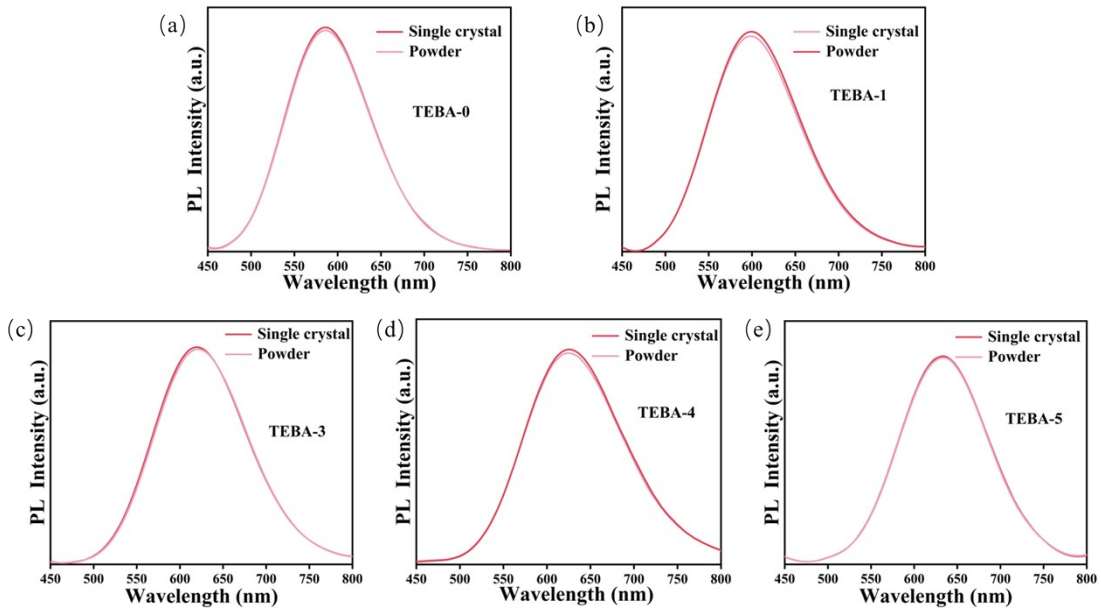

Figure S10. PL spectra of powders and crystals of (a) TEBA-0. (b) TEBA-1. (c) TEBA-3. (d) TEBA-4. and (e) TEBA-5.

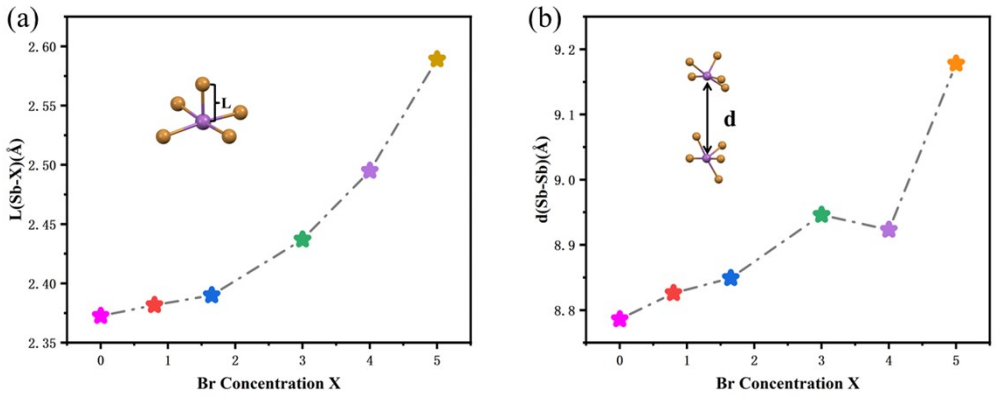

Figure S11. (a) The vertical Sb-Cl bond length (b) The nearest polyhedral distance versus the Br concentration in $(TEBA)_2SbCl_{5-x}Br_x$.

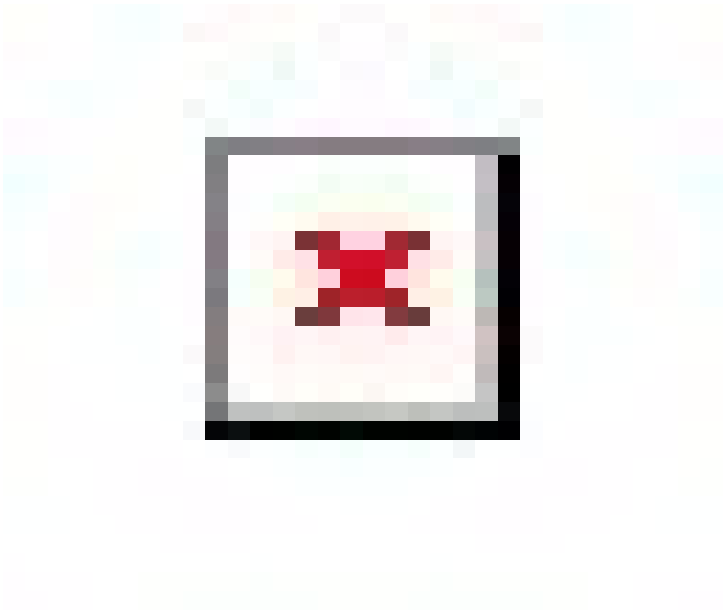

Figure S12. RL spectra with a dose rate range from 0.578 to 5.01 mGy_{air} s^{-1}.

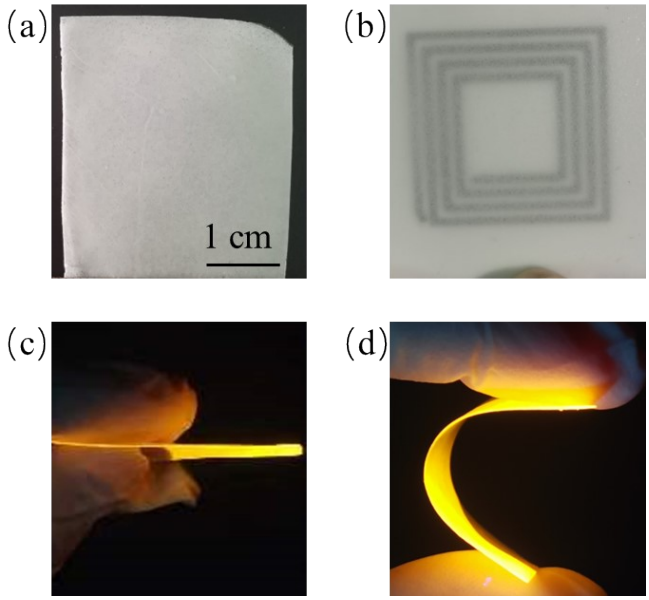

Figure S13. The relevant physical properties of the scintillator film (a) Size. (b) Transparency. (c) and (d) flexibility.

References

[1] G. M. Sheldrick, SHELXS-2014, Program for Crystal Structure Solution, University of Gottingen, Germany, 2014.

[2] G. M. Sheldrick, SHELXL-2014, Program for the Refinement of Crystal Structure, University of Gottingen, Germany, 2014.