Supporting Information

Effect of Halide-mixing on Tolerance Factor and Charge-carrier Dynamics in ($CH_3NH_3PbBr_{3-x}Cl_x$) Perovskites Powders

Zumaira Siddique[a,b,c], Julia L. Payne[c], John T.S. Irvine[c], Lethy K. Jagadamma[d], Zareen Akhter[a], Ifor D.W. Samuel[d], Azhar Iqbal[*a]

Table S1. The calculated miller indices for perovskite $CH_3NH_3PbBr_{3-x}Cl_x$ (x = 0.5, 1, 1.25, 1.75) powders.

Angle (2θ) / (°)	**Miller indices**
15.1	001
21.3	110
30.2	002
34.0	210
37.3	211
43.3	220
46.1	003
48.7	310
53.6	222
56.0	320
58.3	321
62.7	004

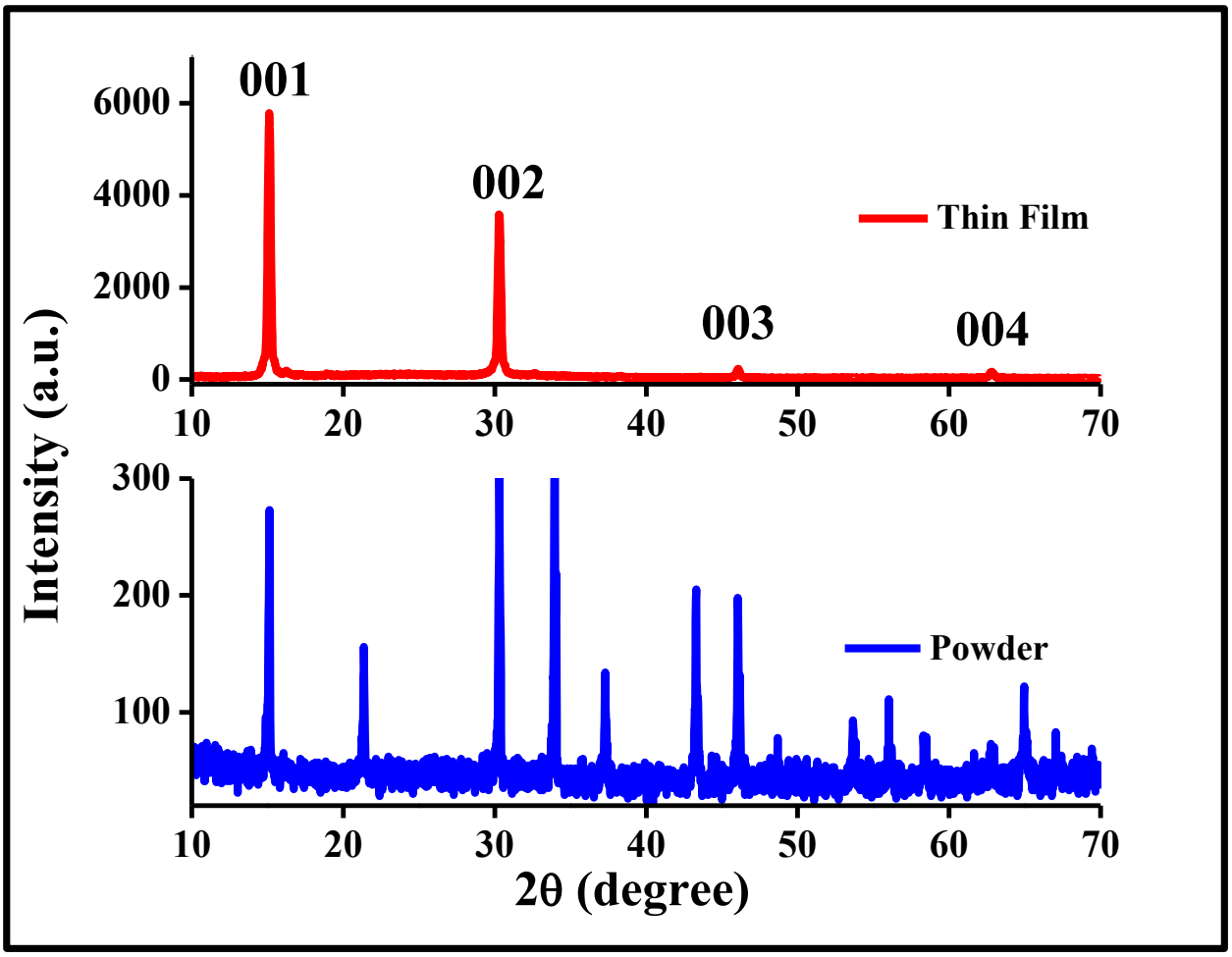

Figure S1. Comparison of XRD diffraction pattern of $CH_3NH_3PbBr_3$ powder and thin film used in solar cell device.

Table S2. The intensity and FWHM of 002 peak in XRD pattern of $CH_3NH_3PbBr_3$ (x=0), $CH_3NH_3PbBr_{2.5}Cl_{0.5}$ (x=0.5), $CH_3NH_3PbBr_2Cl_1$ (x=1), $CH_3NH_3PbBr_{1.75}Cl_{1.25}$ (x=1.25) and $CH_3NH_3PbBr_{1.25}Cl_{1.75}$ (x=1.75) perovskites.

Compound	**Intensity of peak 002 (counts)**	**FWHM of 002 peak (°)**
$CH_3NH_3PbBr_3$ (x=0)	251.7	0.147
$CH_3NH_3PbBr_{2.5}Cl_{0.5}$ (x=0.5)	224.2	0.151
$CH_3NH_3PbBr_2Cl_1$ (x=1)	91.8	0.158
$CH_3NH_3PbBr_{1.75}Cl_{1.25}$ (x=1.25)	69.9	0.228
$CH_3NH_3PbBr_{1.25}Cl_{1.75}$ (x=1.75)	74.3	0.226

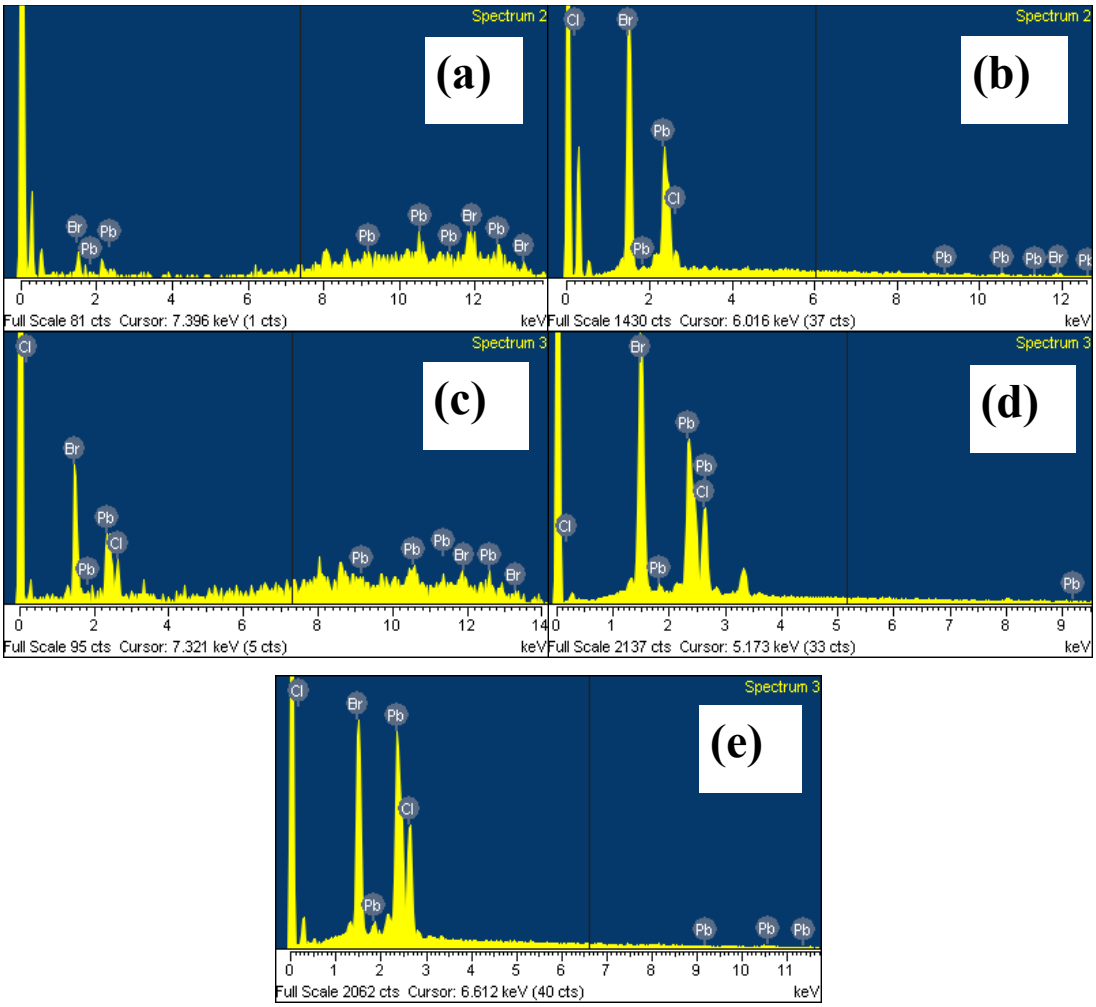

Figure S2. EDX spectra of (a) $CH_3NH_3PbBr_3$ powder (x=0), (b) $CH_3NH_3PbBr_{2.5}Cl_{0.5}$ (x=0.5), (c) $CH_3NH_3PbBr_2Cl_1$ (x=1), (d) $CH_3NH_3PbBr_{1.75}Cl_{1.25}$ (x=1.25) and (e) $CH_3NH_3PbBr_{1.25}Cl_{1.75}$ (x=1.75).

Table S3. PL lifetimes of $CH_3NH_3PbBr_3$ (x=0), $CH_3NH_3PbBr_{2.5}Cl_{0.5}$ (x=0.5), $CH_3NH_3PbBr_2Cl_1$ (x=1), $CH_3NH_3PbBr_{1.75}Cl_{1.25}$ (x=1.25) and $CH_3NH_3PbBr_{1.25}Cl_{1.75}$ (x=1.75) perovskites.

Compound	**τ_1 (ns)**	**A_1 (%)**	**τ_2 (ns)**	**A_2 (%)**	**$\tau_{average}$ (ns) (amplitude weighted)**
$CH_3NH_3PbBr_3$ (x=0)	3.3±0.22	69	15.4±0.8	31	7.07
$CH_3NH_3PbBr_{2.5}Cl_{0.5}$ (x=0.5)	2.0±0.09	66	10.0±0.21	34	4.80
$CH_3NH_3PbBr_2Cl_1$ (x=1)	1.9±0.13	63	11.3±0.32	37	5.42
$CH_3NH_3PbBr_{1.75}Cl_{1.25}$ (x=1.25)	2.4±0.17	60	10.9±0.36	40	5.82
$CH_3NH_3PbBr_{1.25}Cl_{1.75}$ (x=1.75)	2.2±0.32	71	15.2±1.72	29	5.98

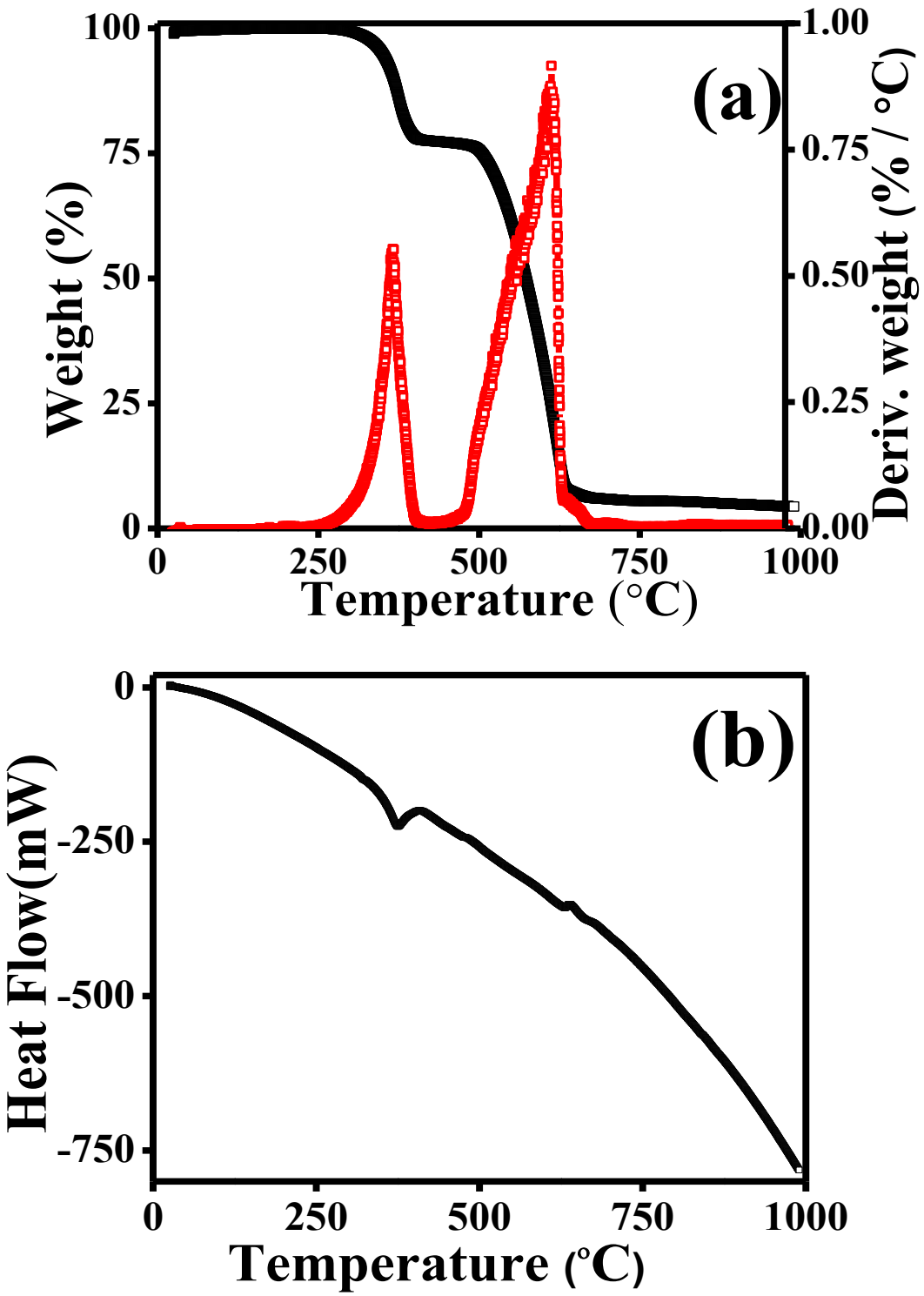

Figure S3. (a) TGA (black line) and DTG (red line) thermograms and (b) DSC thermogram in nitrogen at 10 °C/minute of $CH_3NH_3PbBr_3$ (x=0) powder.

Table S4. Thermal decomposition data for $CH_3NH_3PbBr_3$ perovskite powder.

Temperature (°C)		Peak Temperature T_p (°C)	Weight (%) at peak Temperature	Weight (%) at final temperature	Weight loss at final temperature
Initial	Final				
254	430	375	86	77.5	22.5
450	642	622	17	7.5	70
643	688	645	7	6	1.5

Table S5. Comparison of the power conversion efficiencies of $CH_3NH_3PbBr_3$ based solar cells from this work and previously reported, without different treatments.

Compound	PCE (%)	V_{OC} (V)	Reference
$CH_3NH_3PbBr_3$	4.85	0.84	[1]
$CH_3NH_3PbBr_3$	2.00	0.79	[2]
$CH_3NH_3PbCl_3$ (best)	0.24	1.11	[3]
$CH_3NH_3PbCl_{2.4}Br_{0.6}$	0.42	1.13	[3]
$CH_3NH_3PbBr_3$	2.79	1.1	[4]
$CH_3NH_3PbBr_3$	2.10	0.72	[5]
$CH_3NH_3PbI_3$	1.35	0.65	[6]
$CH_3NH_3PbBr_3$ (best) FW	1.17	1.17	Our work
$CH_3NH_3PbBr_3$ (best) RW	1.41	1.18	Our work

References

1. H. Mehdi, A. Mhamdi, R. Hannachi, A. Bouazizi, $MAPbBr_3$ perovskite solar cells via a two- step deposition process. RSC Adv. **9**, 12906-12912 (2019)
2. T.D. McFarlane, C.S.D. Castro, P.J. Holliman, M.L. Davies, Improving the light harvesting and colour range of methyl ammonium lead tri-bromide ($MAPbBr_3$) perovskite solar cells through co-sensitisation with organic dyes, Chem. Commun. **55**, 35-38 (2019)
3. D. Liu, C. Yang, Lunt, R. R. Halide perovskites for selective ultraviolet-harvesting transparent photovoltaics. Joule **2**, 1827-1837 (2018)
4. X. Hu, X.F. Jiang, X. Xing, L. Nian, X. Liu, R. Huang, K. Wang, H.L. Yip, G. Zhou, Wide bandgap perovskite solar cells with large open-circuit voltage of 1653 mv through interfacial engineering. Solar RRL **2**, 1800083/1-10 (2018)
5. E. Belarbi, M.V. Pelarda, B.C. Hames, R.S. Sanchez, E.M. Barea, H.M. Meherzi, I.M. Sero, Transformation of PbI_2, $PbBr_2$ and $PbCl_2$ salts into $MAPbBr_3$ perovskite by halide exchange as an effective method for recombination reduction. Phys. Chem. Chem. Phys. **19**, 10913-10921 (2017)
6. S-H. Huang, K-Y. Tian, H-C. Huang, C-F. Li, W-C. Chu, K-M. Lee, Y-C. Huang, W-F. Su, Controlling morphology and interface of perovskite layer for scalable high-efficiency solar cell fabricated by using green solvents and blade coating in ambient environment. ACS Appl. Mater. Interfaces **12**, 26041-26049 (2020)