Supplementary Materials

We examined the drop cast $CsPbI_2Br$ sample on the TEM copper grid without any annealing process (**Figures S1a-c**). Also **Figure S1d** displays the selected area electron diffraction (SAED) pattern of $CsPbI_2Br$ displaying the phase impurity, which is different from the XRD pattern in Figures 7a and 8 (see Results and Discussion in *Article*). This indicates that without annealing, the sample has high phase-impurity, e.g., non-perovskite yellow phase (see **Table S1** containing estimated 2θ date for comparison purpose). Hence, the pseudo-cubic α-phase $CsPbI_2Br$ with 'iodine atoms' might be kinetically stable (not thermodynamically) due to polymorphism [1,2]. Therefore, 'iodine' has double sides, increasing light absorption but causing stability problems under ambient conditions.

Table S1. Analysis of selected area electron diffraction (SAED) image for CsPbI2Br without annealing at room temperature.

Perovskite	Diameter (1/nm)	Radius (1/nm)	*d*-Spacing (nm)	2θ (°)
$CsPbI_2Br$	3.91	1.96	0.51	17.3
	5.23	2.63	0.38	23.2
	6.32	3.16	0.32	28.2
	8.05	4.03	0.25	36.1
	10.08	5.04	0.20	45.7
	12.10	6.05	0.17	55.5

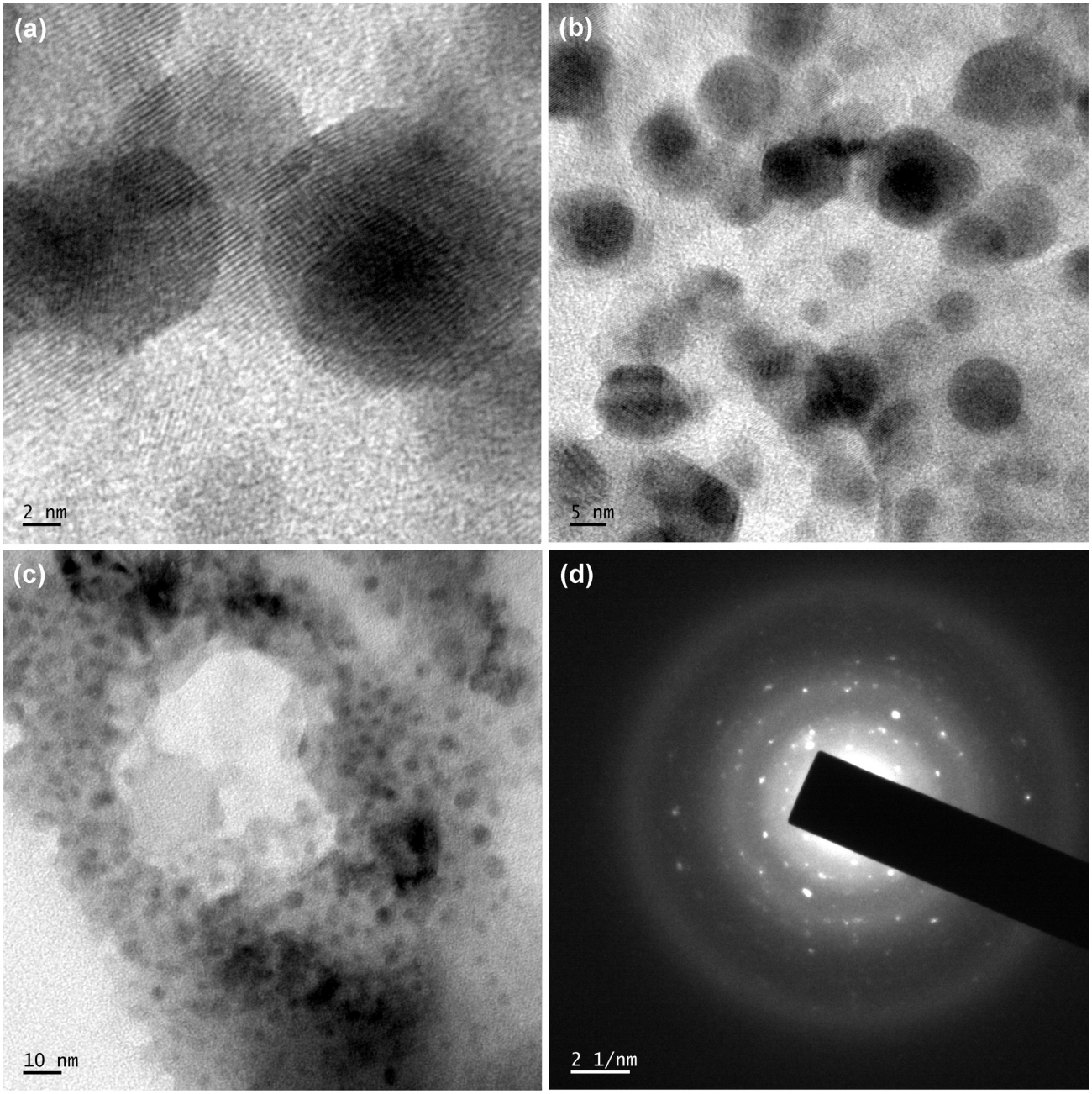

Figure S1. HR-TEM image of $CsPbI_2Br$ with different scale bar: (**a**) 2 nm, (**b**) 5 nm, and (**c**) 10 nm. (**d**) SAED pattern of $CsPbI_2Br$ displaying the phase impurity at room temperature.

References

1. Alaei, A.; Circelli, A.; Yuan, Y.; Yang, Y.; Lee, S.S. Polymorphism in metal halide perovskites. *Mater. Adv*. **2021**, *2*, 47-63.
2. Wang, B.; Novendra, N.; Navrotsky, A. Energetics, Structures, and Phase Transitions of Cubic and Orthorhombic Cesium Lead Iodide (CsPbI3) Polymorphs. *J. Am. Chem. Soc*. **2019**, *141*, 14501–14504.