

SUPPLEMENTARY INFORMATION

High-performing Wide Bandgap Perovskite Solar Cells Fabricated in Ambient High-Humidity Conditions

Ugur Deneb Menda, Guilherme Ribeiro, Daniela Nunes, Tomás Calmeiro, Hugo Águas, Elvira Fortunato, Rodrigo Martins, Manuel J. Mendes

i3N/CENIMAT, Department of Materials Science, Faculty of Science and Technology, Universidade NOVA de Lisboa and CEMOP/UNINOVA, Campus de Caparica, 2829- 516 Caparica, Portugal

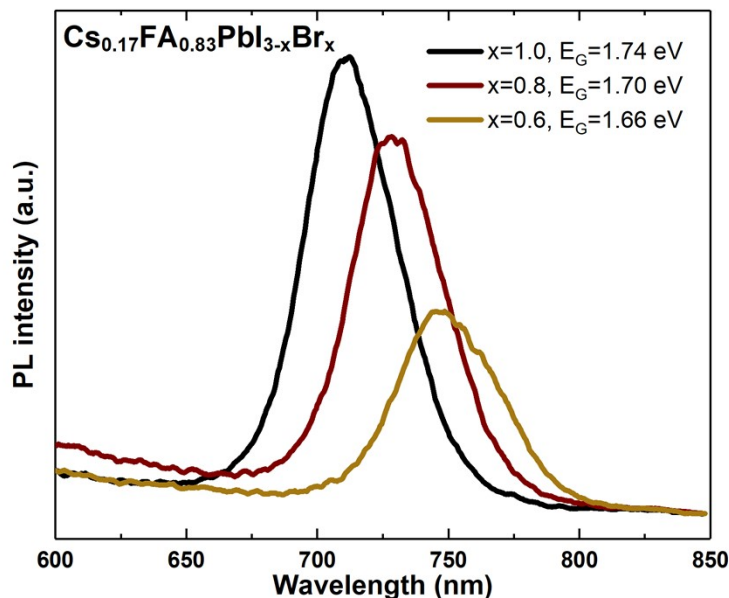


Figure S1. Photoluminescence (PL) spectra of the WBG perovskite ($\text{Cs}_{0.17}\text{FA}_{0.83}\text{PbI}_{3-x}\text{Br}_x$) layers with unnormalized intensity.

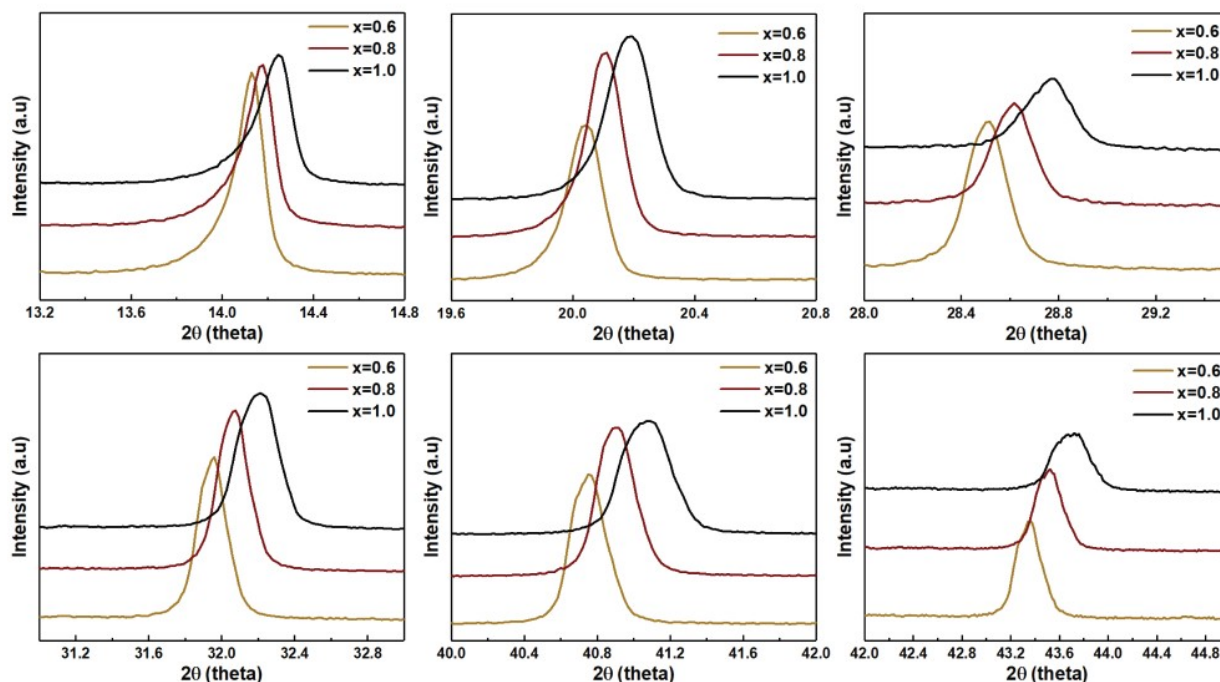


Figure S2. Details of the XRD peaks in Fig. 2d of the main article, of the $\text{Cs}_{0.17}\text{FA}_{0.83}\text{PbI}_{3-x}\text{Br}_x$ layers with different x values, showing more clearly the shift of the characteristic peaks to larger scattering angles with the increment of the Br:I ratio. This occurs due to the contraction of the crystal lattice with higher bromide content.

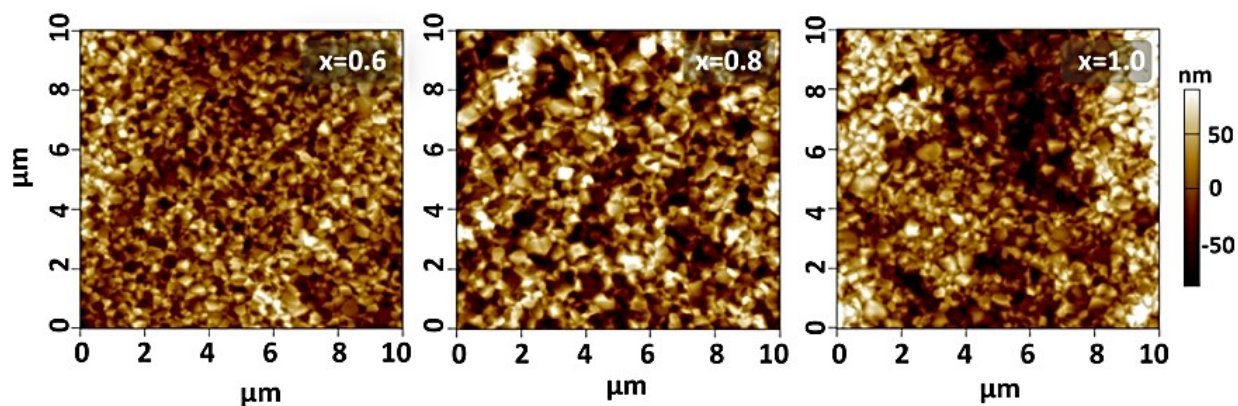


Figure S3. AFM images of $\text{Cs}_{0.17}\text{FA}_{0.83}\text{PbI}_{3-x}\text{Br}_x$ WBG perovskite layers with different Br content.

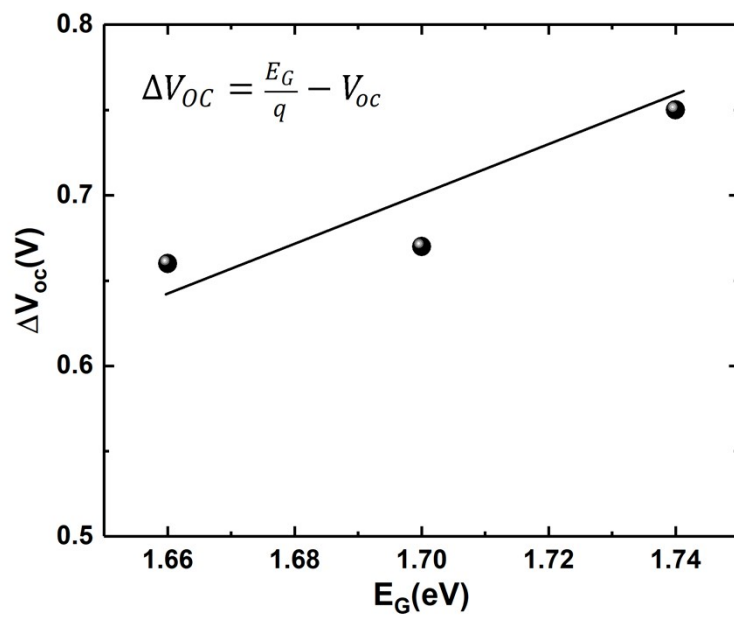


Figure S4. Open-circuit voltage, V_{oc} , deficit values of the WBG PSCs with the 3 different bandgaps, E_G .

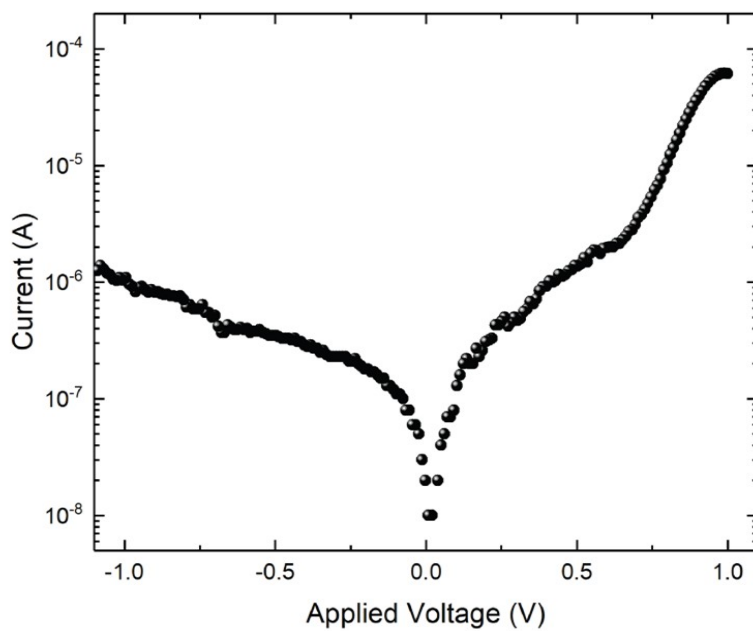


Figure S5. Dark I-V curve of the best performing solar cell with excess reverse leakage current.

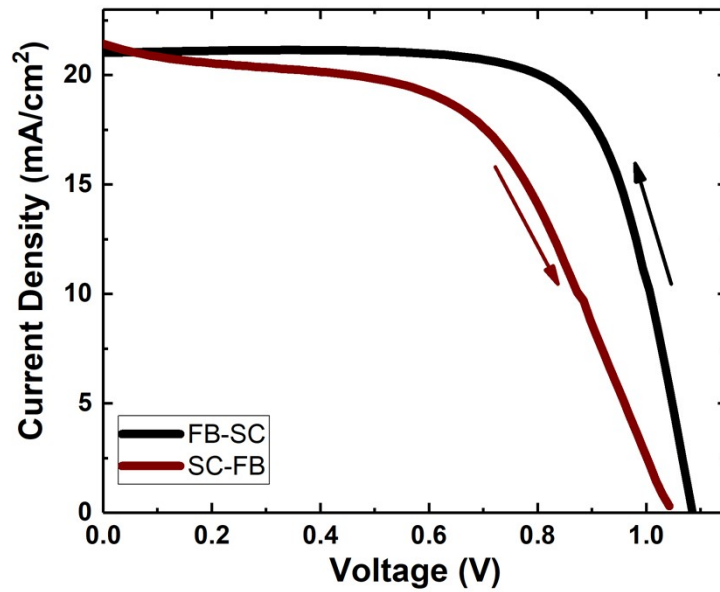


Figure S6. J-V curves in reverse scan (from Forward bias, FB, to Short Circuit, SC) and forward scan (from SC to FB). The plot shows a distinct hysteresis effect which is attributed to the mobile ions/ion-vacancies in the perovskite material [1,2]. This phenomenon is still being actively researched both on theoretical [3–5] and experimental [1,2] grounds, but the general consensus in the PSCs field is that the results comparison should focus in the reverse scan JVs of the best performing devices [5].

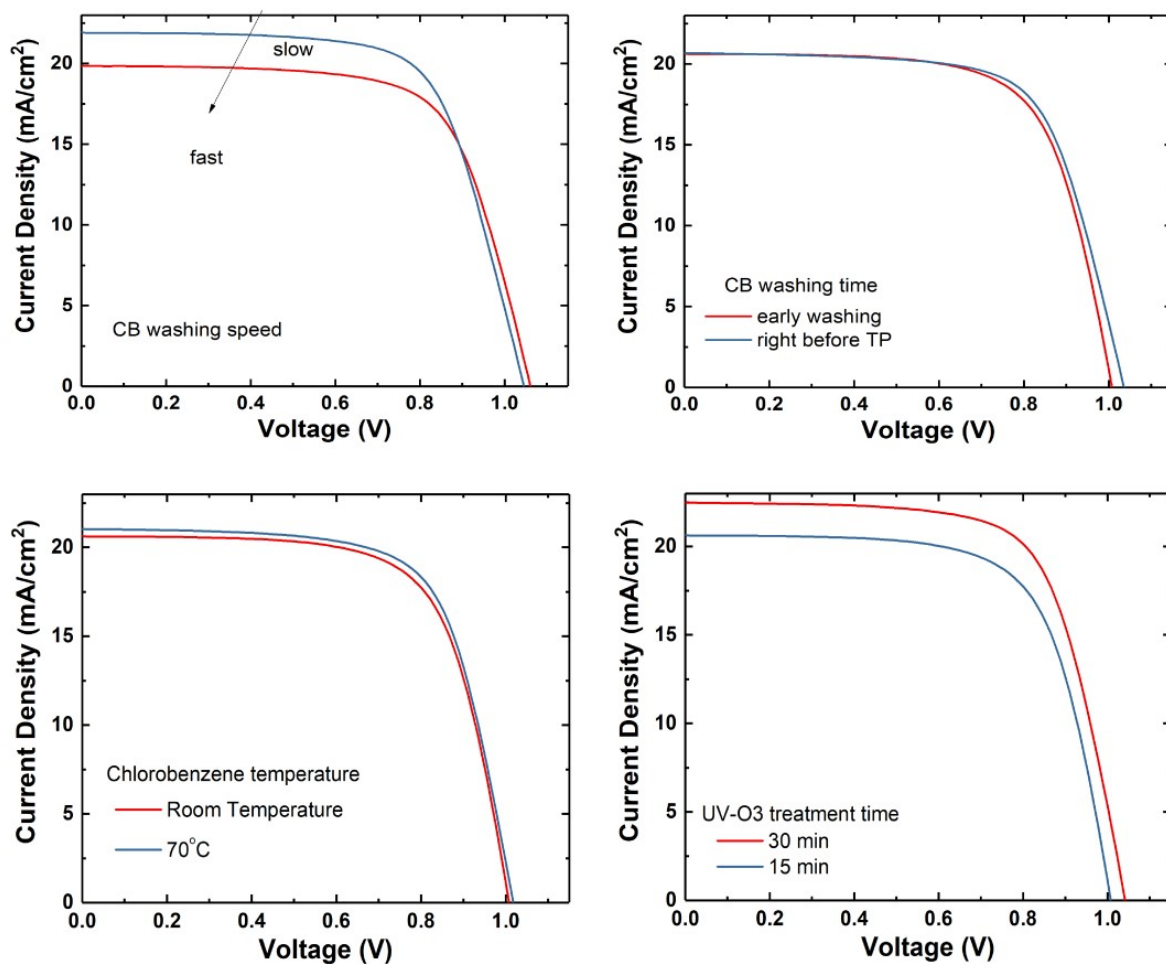


Figure S7. The effects of key deposition conditions, affecting the defect density of the fabricated $(\text{CsI})_{0.17}(\text{FAI})_{0.83}(\text{PbI}_2)_{0.6}(\text{PbBr}_2)_{0.4}$ perovskite layers, in the resulting JV characteristics of the PSCs. In view of the optimization studies performed in this work, it was ascertained that chlorobenzene washing should be performed in a slow process, close to the end of the spinning (right before the turbid point) and a slight improvement has been observed when the antisolvent was applied after heating to 50-70°C. Additionally, increased UV-O₃ treatment time assists the perovskite coverage on the TiO₂ layer in high humid ambient which causes an increase in the solar cell performance.

Table S1. Specific fabrication parameters applied for the optimized WBG (1.70 eV) PSCs.

	Antisolvent (CB) drip	UV-O ₃ treatment on mesoporous TiO ₂	Substrate pretreatments
Time	1-2 s before TP	30 minutes	10 min
Temperature	50-70 °C	Room temperature	70 °C
Distance from substrate	5 mm	2 cm	-
Volume	80-120 μl	-	-

References:

- [1] B. Chen, M. Yang, S. Priya, K. Zhu, Origin of J-V Hysteresis in Perovskite Solar Cells, *J. Phys. Chem. Lett.* 7 (2016) 905–917. <https://doi.org/10.1021/acs.jpcllett.6b00215>.
- [2] C. Li, S. Tscheuschner, F. Paulus, P.E. Hopkinson, J. Kießling, A. Köhler, Y. Vaynzof, S. Huettner, Iodine Migration and its Effect on Hysteresis in Perovskite Solar Cells, *Adv. Mater.* 28 (2016) 2446–2454. <https://doi.org/10.1002/adma.201503832>.
- [3] J. Bisquert, A. Guerrero, C. Gonzales, Theory of Hysteresis in Halide Perovskites by Integration of the Equivalent Circuit, *ACS Phys. Chem. Au.* (2021). <https://doi.org/10.1021/acspchemau.1c00009>.
- [4] S. Van Reenen, M. Kemerink, H.J. Snaith, Modeling Anomalous Hysteresis in Perovskite Solar Cells, *J. Phys. Chem. Lett.* 6 (2015) 3808–3814. <https://doi.org/10.1021/acs.jpcllett.5b01645>.
- [5] S.N. Habisreutinger, N.K. Noel, H.J. Snaith, Hysteresis Index: A Figure without Merit for Quantifying Hysteresis in Perovskite Solar Cells, *ACS Energy Lett.* 3 (2018) 2472–2476. <https://doi.org/10.1021/acsenergylett.8b01627>.