Supporting information

Eco-friendly Antisolvent Enabled Inverted MAPbI₃ Perovskite Solar Cells with Fill Factors over 84%

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Fig. S1. Photo-CELIV transients recorded at various light intensity for the control (a) and PA-treated single carrier devices.



Fig. S2. *J-V* curve of the devices with different antisolvents (DE: PA = 1:0, 1:0.1, 1:0.2, 1:0.3 and 1:0.4, respectively).



Fig. S3. *J-V* curves of PSCs with different antisolvents.



Fig. S4. Stability tests of unencapsulated devices with DE and 20% PA-treated PSCs (stored in ambient condition at 25°C, 30% relatively humidity).



Fig. S5. Contact angles of DE and 20% PA-treated perovskite films.



Fig. S6. XRD stability tests of unencapsulated devices with DE and PA-treated PSCs (stored at 50-60% relative humidity and 100 °C).



Fig. S7. UV stability tests of unencapsulated devices with DE and PA-treated PSCs (stored at 50-60% relative humidity and 100 °C).

| 0 | | | | |
|-------------------|-----------------------------|----------------------------|--|--|
| Antisolvent | Environmental impact | ital impact Healthy impact | | |
| DE | 5 | 5 | | |
| PA | 7 | 8 | | |
| TOL | 3 | 4 | | |
| Anisole | 6 | 7 | | |
| EA | 8 | 8 | | |
| isopropyl acetate | 7 | 7 | | |
| IPA | 9 | 8 | | |
| PE | 2 | 2 | | |

Table S1. The environmental and healthy impact of different antisolvents¹. (The higher the score, the greener it is.)

Table S2. The detailed parameters of photo-CELIV current transients for devices with different antisolvents.

| Antisolvent | A (100 V ms ⁻¹) | t _{max} (μs) | Δ <i>j</i> (mA cm ⁻²) | <i>j</i> ₀ (mA cm ⁻²) | d (nm) |
|-------------|-----------------------------|-----------------------|-----------------------------------|--|--------|
| Ctr. | 100 | 2.703 | 2.541 | 1.046 | 420 |
| PA | 100 | 2.186 | 3.276 | 1.049 | 420 |

Table S3. Photovoltaic parameters for the champion PSCs with different antisolvents ratios.

| Antisolvent | Voc (V) | Jsc (mA cm ⁻²) | FF (%) | PCE (%) |
|-------------|---------|----------------------------|--------|---------|
| Ctr. | 1.02 | 22.64 | 79.13 | 18.28 |
| 10% PA | 1.03 | 22.86 | 81.89 | 19.23 |
| 20% PA | 1.05 | 23.20 | 84.49 | 20.61 |
| 30% PA | 1.04 | 23.30 | 81.37 | 19.63 |
| 40% PA | 1.02 | 22.88 | 80.19 | 18.65 |

Table S4. The detailed performance data of PSCs with different antisolvents.

| Antisolvent | $V_{\rm oc}$ (V) | $J_{\rm sc}$ (mA cm ⁻²) | FF (%) | PCE (%) |
|-------------------------|------------------|-------------------------------------|--------|---------|
| СВ | 1.02 | 21.17 | 77.25 | 16.65 |
| DE | 1.02 | 22.64 | 79.13 | 18.28 |
| IPA | 0.97 | 18.42 | 76.75 | 13.75 |
| PA | 1.00 | 21.06 | 78.26 | 16.52 |
| Toluene | 0.97 | 17.62 | 74.49 | 12.73 |
| <i>n</i> -butyl alcohol | 1.02 | 17.85 | 78.82 | 14.35 |

| Method | Antisolvent | $V_{ m oc}$ (V) | $J_{\rm sc}$ | FF (%) | PCE (%) | Refs |
|------------------|-------------|-----------------|------------------------|--------|---------|-----------|
| | | | (mA cm ⁻²) | | | |
| Antisovent | EA | 1.08 | 22.00 | 79.90 | 18.98 | 2 |
| Single crystal | - | 1.14 | 21.93 | 81.00 | 21.93 | 3 |
| Antisovent | Toluene | 1.13 | 22.19 | 81.29 | 20.46 | 4 |
| Antisovent | СВ | 1.10 | 23.38 | 81.87 | 21.11 | 5 |
| Thin monocrystal | - | 1.08 | 22.60 | 82.50 | 20.10 | 6 |
| Antisovent | СВ | 1.13 | 23.10 | 83.81 | 21.88 | 7 |
| Blade coating | - | 1.10 | 22.60 | 86.30 | 21.50 | 8 |
| Antisovent | PA | 1.05 | 23.20 | 84.49 | 20.61 | This work |

Table S5. Representative inverted MAPbI₃ PSCs with high fill factors.

References

- R. K. Henderson, C. Jiménez-González, D. J. C. Constable, S. R. Alston, G. G. A. Inglis, G. Fisher, J. Sherwood, S. P. Binks and A. D. Curzons, *Green Chem.*, 2011, 13, 854-862.
- 2. J. Liu, N. Li, J. Jia, J. Dong, Z. Qiu, S. Iqbal and B. Cao, Sol. Energy, 2019, 181, 285-292.
- A. Y. Alsalloum, B. Turedi, X. Zheng, S. Mitra, A. A. Zhumekenov, K. J. Lee, P. Maity, I. Gereige, A. AlSaggaf, I. S. Roqan, O. F. Mohammed and O. M. Bakr, *ACS Energy Lett.*, 2020, 5, 657-662.
- 4. Y. Kang, S. Kwon, S. Cho, Y. Seo, M. Choi, S. Kim and S. Na, *ACS Energy Lett.*, 2020, 5, 2535-2545.
- 5. S. Wang, Z. He, J. Yang, T. Li, X. Pu, J. Han, Q. Cao, B. Gao and X. Li, *J. Energy Chem.*, 2021, **60**, 169-177.
- W. Kong, S. Wang, F. Li, C. Zhao, J. Xing, Y. Zou, Z. Yu, C. Lin, Y. Shan, Y. H. Lai, Q. Dong, T. Wu, W. Yu and C. Guo, *Adv. Energy Mater.*, 2020, 10, 2000453.
- S. Xiong, Z. Hou, S. Zou, X. Lu, J. Yang, T. Hao, Z. Zhou, J. Xu, Y. Zeng, W. Xiao, W. Dong, D. Li, X. Wang, Z. Hu, L. Sun, Y. Wu, X. Liu, L. Ding, Z. Sun, M. Fahlman and Q. Bao, *Joule*, 2021, DOI: <u>https://doi.org/10.1016/j.joule.2020.12.009</u>.
- W. Wu, J. Zhong, J. Liao, C. Zhang, Y. Zhou, W. Feng, L. Ding, L. Wang and D. Kuang, *Nano Energy*, 2020, 75, 104929.