Supplementary Information

Two-dimension additive diethylammonium iodide promoting crystal growth for

efficient and stable perovskite solar cells

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Figure S1. Top view of SEM images of perovskite films (a) with 10% PEAI content, (b) with 10% BAI content. This figure shows that the films doped with PEAI or BAI present small grains less than 100 nm, and even smaller than the MAPbI₃ film. The results indicate PEAI or DAI could not induce the crystal grain growth of 3D film.



Figure S2. XRD patterns of the perovskite films without and with 7%, 10% and 15% DA contents. The inset is the XRD pattern of DA₂PbI₄. Compared with the XRD patterns of 2D perovskite DA₂PbI₄ films, the peaks around 7.5° in the XRD patterns of DA mixed films mainly belong to DA₂PbI₄.



Figure S3. *J*-V characteristics of the perovskite devices without and with 7%, 10% and 15% DAI contents. The measurements were conducted during reverse scanning under one sun illumination at 100 mW cm⁻². The devices based on the perovskite film with 10% DAI content exhibits the best performance.



Table S1. Performance parameters obtained from the J-V characteristics of the perovskite devices without and with 7%, 10% and 15% DAI contents.

| DA content (%) | J _{SC} (mAcm ⁻²) | Voc (V) | FF (%) | PCE (%) |
|----------------|--|------------|-----------|------------|
| 0 | 22.47 | 1.02 | 68.60 | 15.73 |
| 7 | 22.83 | 1.04 | 72.09 | 17.12 |
| 10 | 22.95 | 1.05 | 79.04 | 19.05 |
| 15 | 22.33 | 1.02 | 77.91 | 17.72 |

Figure S4. The performance of best devices without and with 10% DAI content films under reverse scan (1.2V--0.2 V) and forward scan (-0.2V-1.2 V). The hysteresis of the (DA₂PbI₄)_{0.05}MAPbI₃ perovskite solar cell is slightly reduced.



Table S2. Performance parameters of the device without and with 10% DA contents under reverse scan (1.2V-0.2 V) and forward scan (-0.2V-1.2 V).

| DA content | Scan direction | $J_{\rm SC}$ (mA cm ⁻²) | Voc (V) | FF (%) | PCE (%) |
|------------|-------------------|-------------------------------------|------------|-----------|------------|
| Without | Reverse | 22.47 | 1.02 | 68.60 | 15.73 |
| | Forward | 22.44 | 0.95 | 54.56 | 11.63 |
| 10% DAI | Reverse | 22.95 | 1.05 | 79.04 | 19.05 |
| | Forward | 22.92 | 0.97 | 77.73 | 17.26 |

Figure S5. The long-term stability of normalized PCE of the fabricated devices without and with 10% DA content. The PCE of the (DA₂PbI₄)_{0.05}MAPbI₃ device shows better stability than the MAPbI₃ device.



Table S3. Fitted parameters of the TRPL curves of ITO/perovskite films.

| DA content | B1 (%) | τ_1 (ns) | B2 (%) | τ ₂ (ns) | $	au_{average}$ (ns) |
|------------|-----------|---------------|-----------|------------------------|----------------------|
| Without | 41.76 | 8.59 | 58.24 | 66.24 | 61.34 |
| 7% | 51.90 | 38.02 | 48.10 | 88.66 | 72.65 |
| 10% | 42.40 | 34.59 | 57.60 | 103.04 | 89.48 |
| 15% | 47.76 | 4.88 | 52.24 | 39.16 | 35.65 |