

## Supporting Information

# Roll-to-Roll Friendly Solution-Processing of Ultrathin, Sintered CdTe Nanocrystal Photovoltaics

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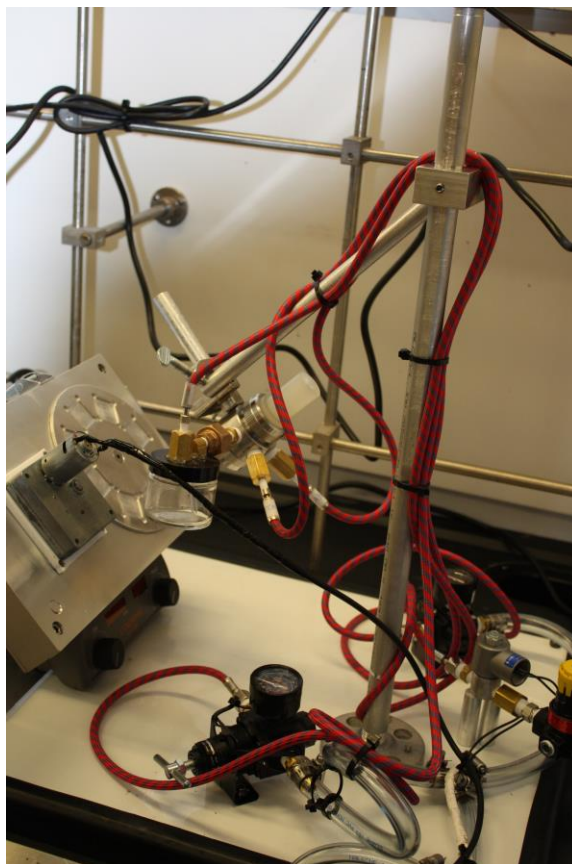
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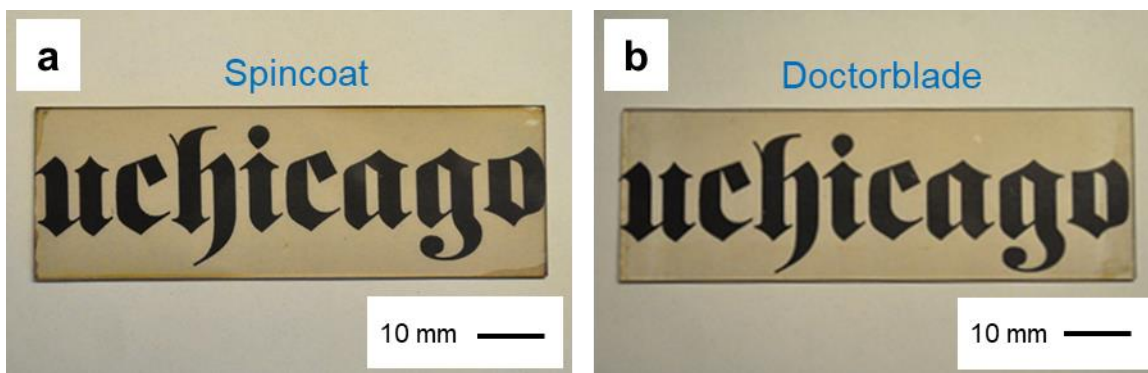
**1. Chemicals.** Cadmium oxide (CdO, 99.99+%), tellurium shot (Te, 99.999%), cadmium chloride (CdCl<sub>2</sub>, 99.99%), pyridinium hydrochloride (pyr·HCl, 98%), zinc acetate dihydrate (Zn(OAc)<sub>2</sub>·2H<sub>2</sub>O, 99.999%), indium chloride (InCl<sub>3</sub>, 99.999%), oleic acid (OA, technical grade, 90%), 1-octadecene (ODE, technical grade, 90%), tributylphosphine (TBP, 97% with isomers), toluene (≥99.8%, anhydrous), ethanol (≥99.5%, anhydrous), pyridine (99.8%, anhydrous), hexane (95%, anhydrous), *N*-methylformamide (NMF, 99%), hexamethylphosphoramide (HMPA, 99%), 1-propanol (1-PA, 99.7%, anhydrous), 2-methoxyethanol (99.9%, anhydrous), and ethanolamine (99.5%, redistilled) were purchased from Aldrich. Molecular sieves grade 564 (Type 3A, 8-12 mesh beads), acetone (certified ACS), methanol (certified ACS), and 2-propanol (IPA, certified ACS) were purchased from Fisher Scientific. Aluminum (Al, 99.99%) and silver (Ag, 99.99%) pellets were purchased from Kurt J. Lesker Company. PELCO colloidal silver paste (Ag paste) was purchased from Ted Pella, Inc. 10 wt% TBP:Te was prepared by dissolving 10 g of Te shot in 90 g of TBP overnight in a N<sub>2</sub>-filled glove box. ODE was recrystallized by cooling the bottle in a chiller overnight at 12 °C and decanted to remove impurities. OA was cooled to 16 °C overnight and vacuum filtered to remove high melting point impurities. Pyridine, NMF, and 1-PA were distilled to remove low and high boiling point impurities. NMF and HMPA were dried over molecular sieves and filtered with 0.2 μm PTFE filter before use.



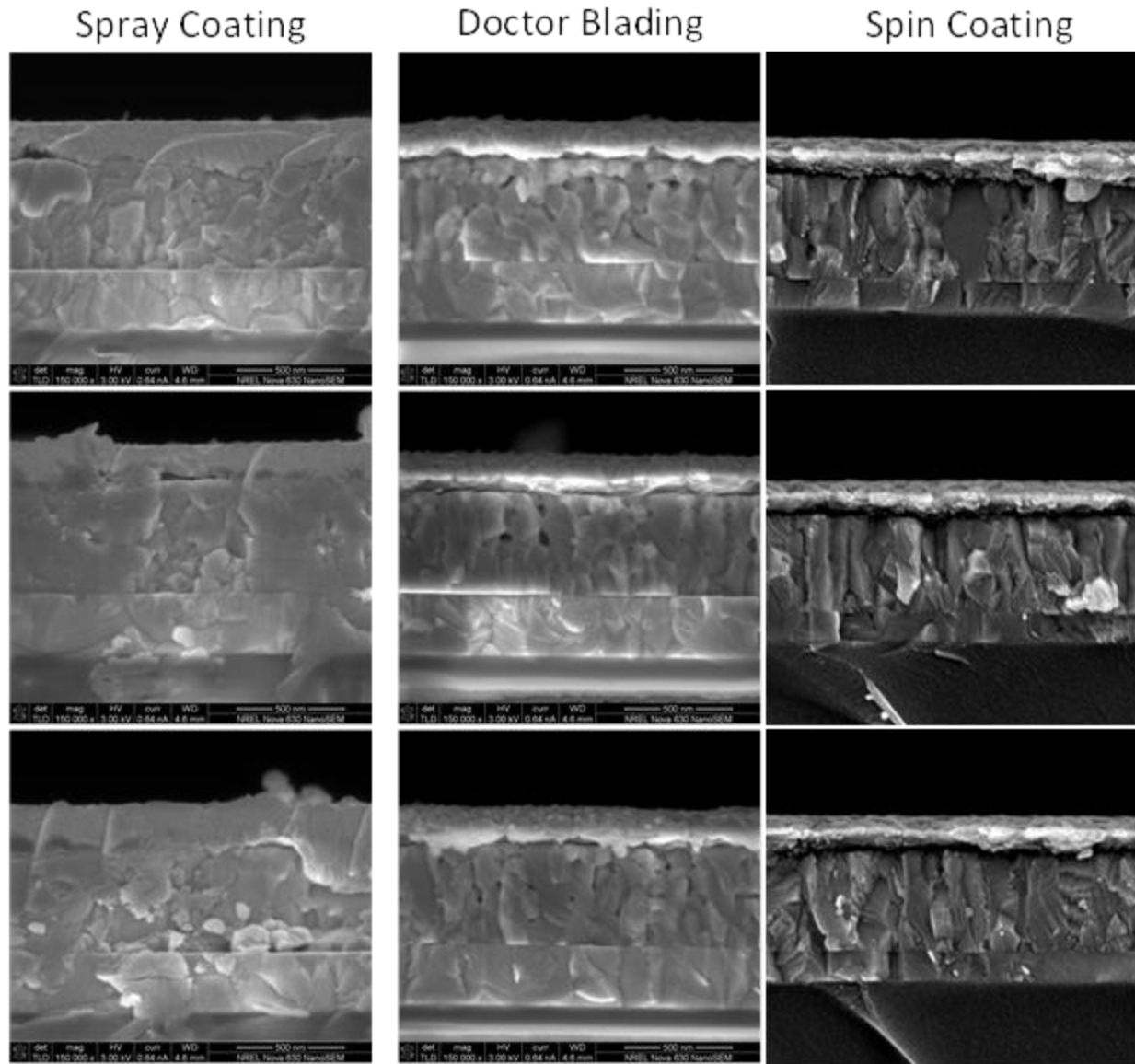
**Figure S1.** Picture of the home-built spray-coating system.



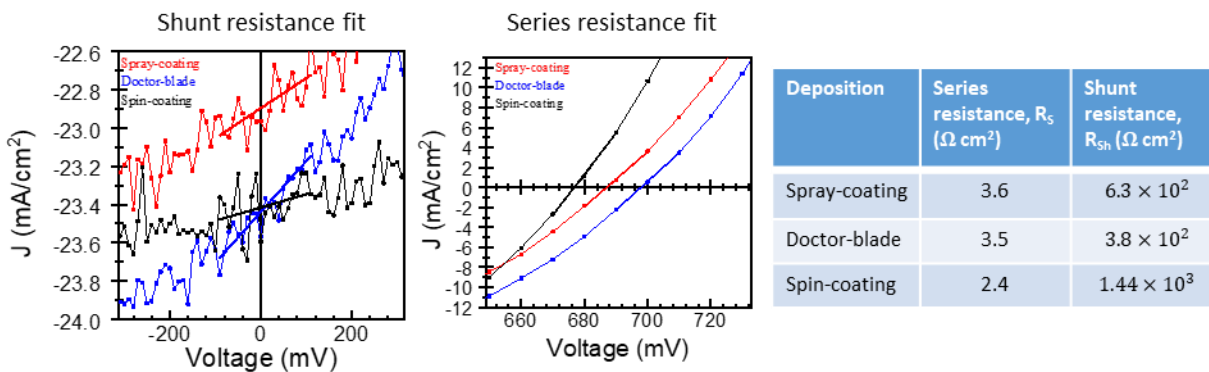
**Figure S2.** Substrates spray-coated using 60 mg/mL pyridine-capped CdTe NCs in a 50/50 mixture of pyridine and 1-propanol as a base. The solution was diluted to 6 mg/mL in chloroform (left), 50/50 pyridine/1-propanol (middle), and methanol (right).



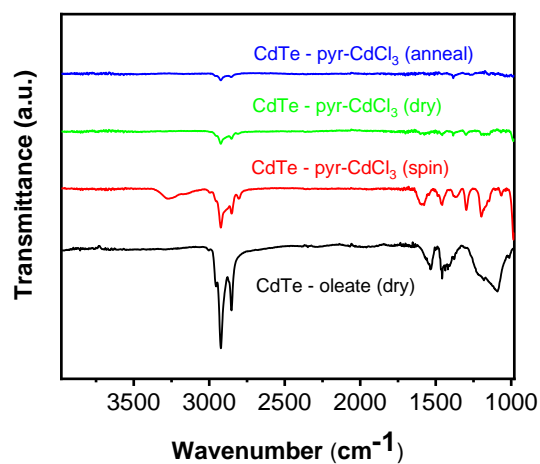
**Figure S3.** A single layer of CdTe NCs deposited on a glass slide via spin-coating (a) or doctor-blading (b). The good uniformity of the film is evident from the clarity of the printed “uchicago” characters on a paper placed behind



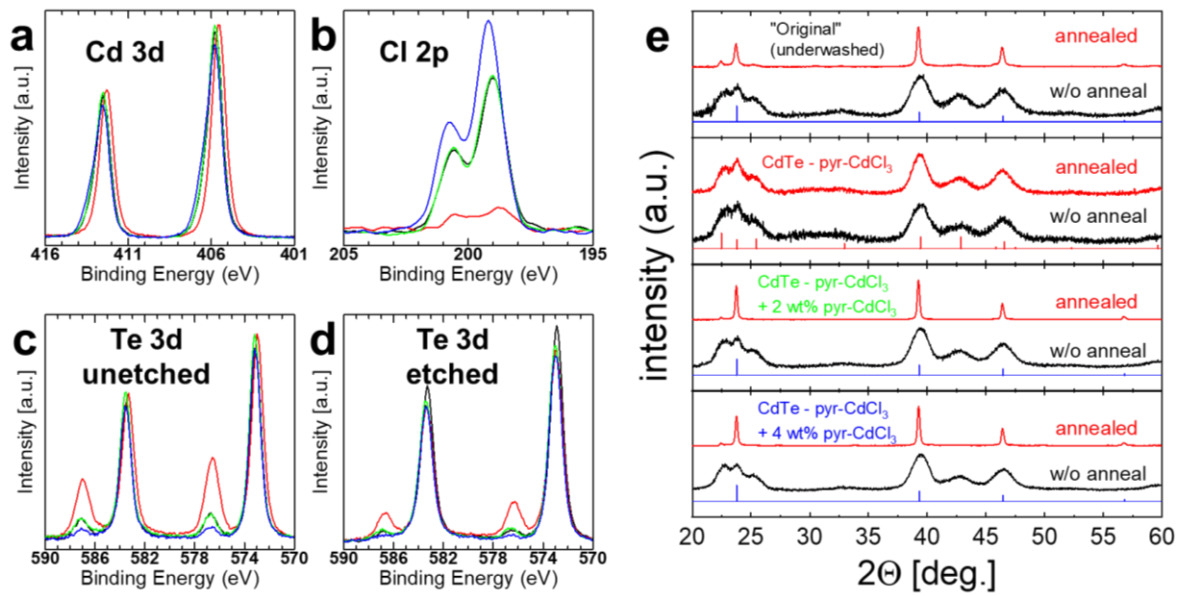
**Figure S4.** Additional cross-sectional SEM images of spin-coated, spray-coated and doctor-bladed devices.



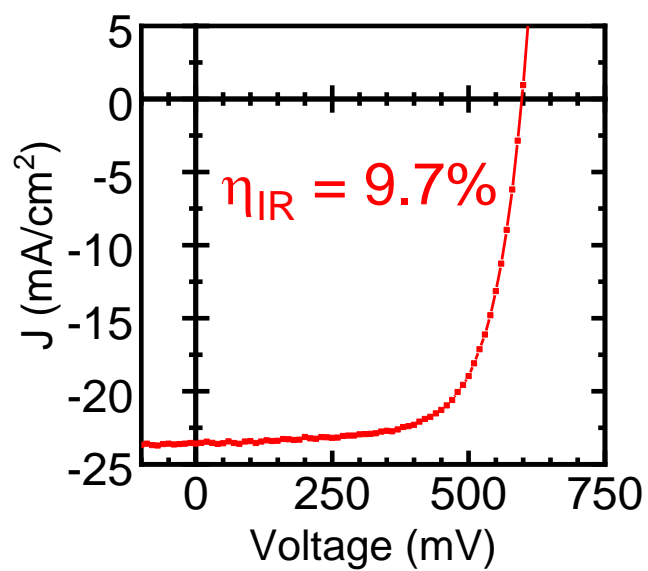
**Figure S5.** Fittings to obtain shunt and series resistances from J-V curves for devices made with different deposition techniques.



**Figure S6.** FTIR analysis of (properly-washed) pyr- $\text{CdCl}_3$ -capped CdTe NCs directly after spin-coating, upon drying at 150 °C and after annealing at 350 °C. The spectrum of oleate-capped CdTe NCs is also shown for reference.

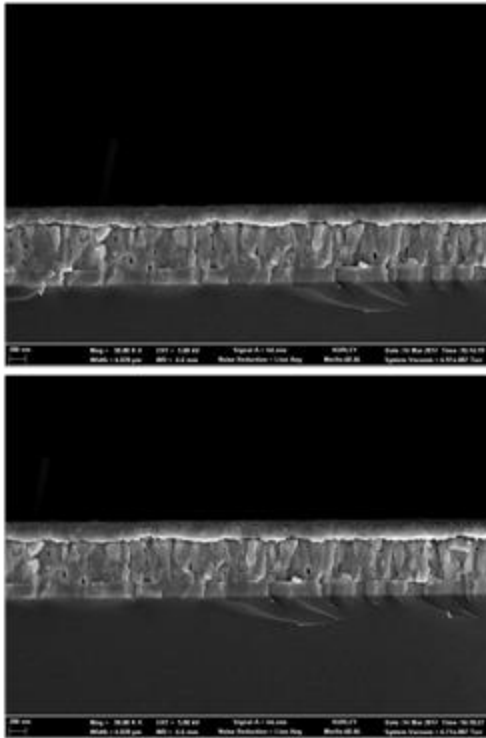


**Figure S7.** (a-d) XPS spectra comparing sintered single-layer CdTe thin films made from “original”, under-washed pyr-CdCl<sub>3</sub>-capped NC ink (black), properly washed pyr-CdCl<sub>3</sub>-capped NC ink with 0 wt% (red), 2 wt% (green), and 4 wt% (blue) pyr-CdCl<sub>3</sub> added in (a) Cd 3d, (b) Cl 2p, (c) Te 3d (unetched), and (d) Te 3d (etched) regions. (e) XRD patterns of single-layer CdTe with (red) and without (black) annealing comparing grain growth of the “original”, under-washed ink and the properly washed ink with 0, 2, and 4 wt% pyr-CdCl<sub>3</sub> added. The vertical lines show the corresponding X-ray diffractions of bulk CdTe in wurtzite (red) and zinc-blende (blue) phase.

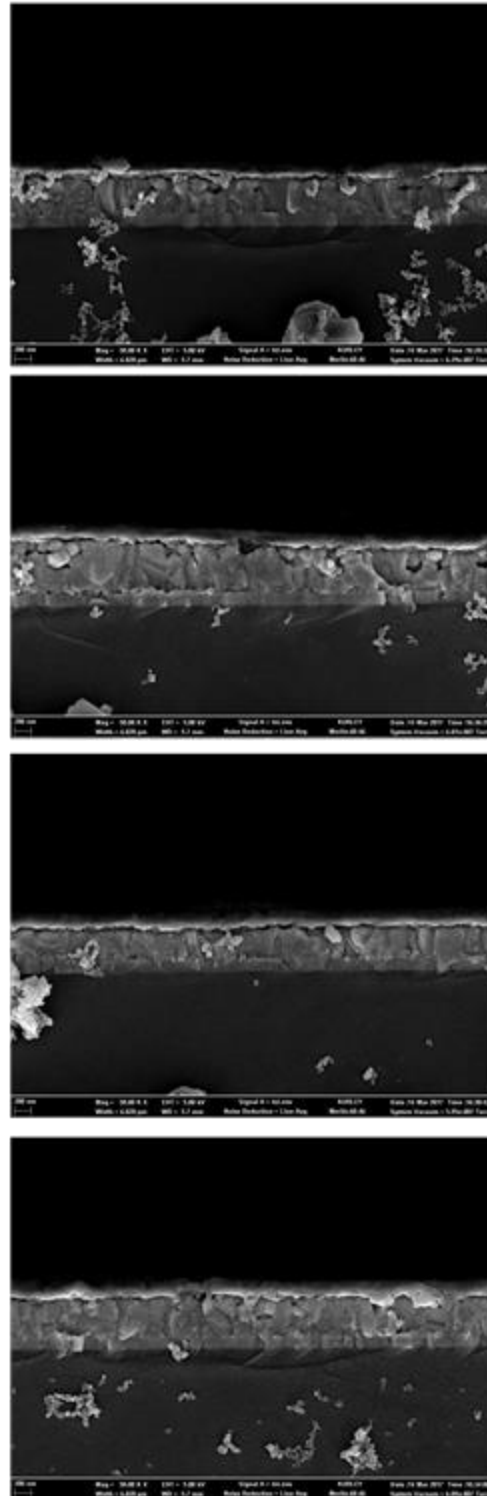


**Figure S8.** JV curves under AM1.5G illumination for solar cells made with IR lamp used for sintering.

Control

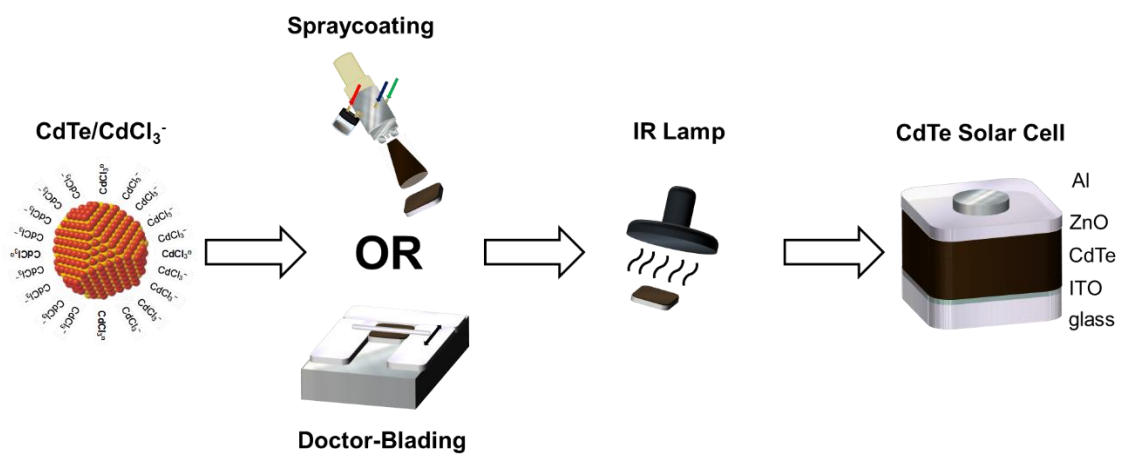


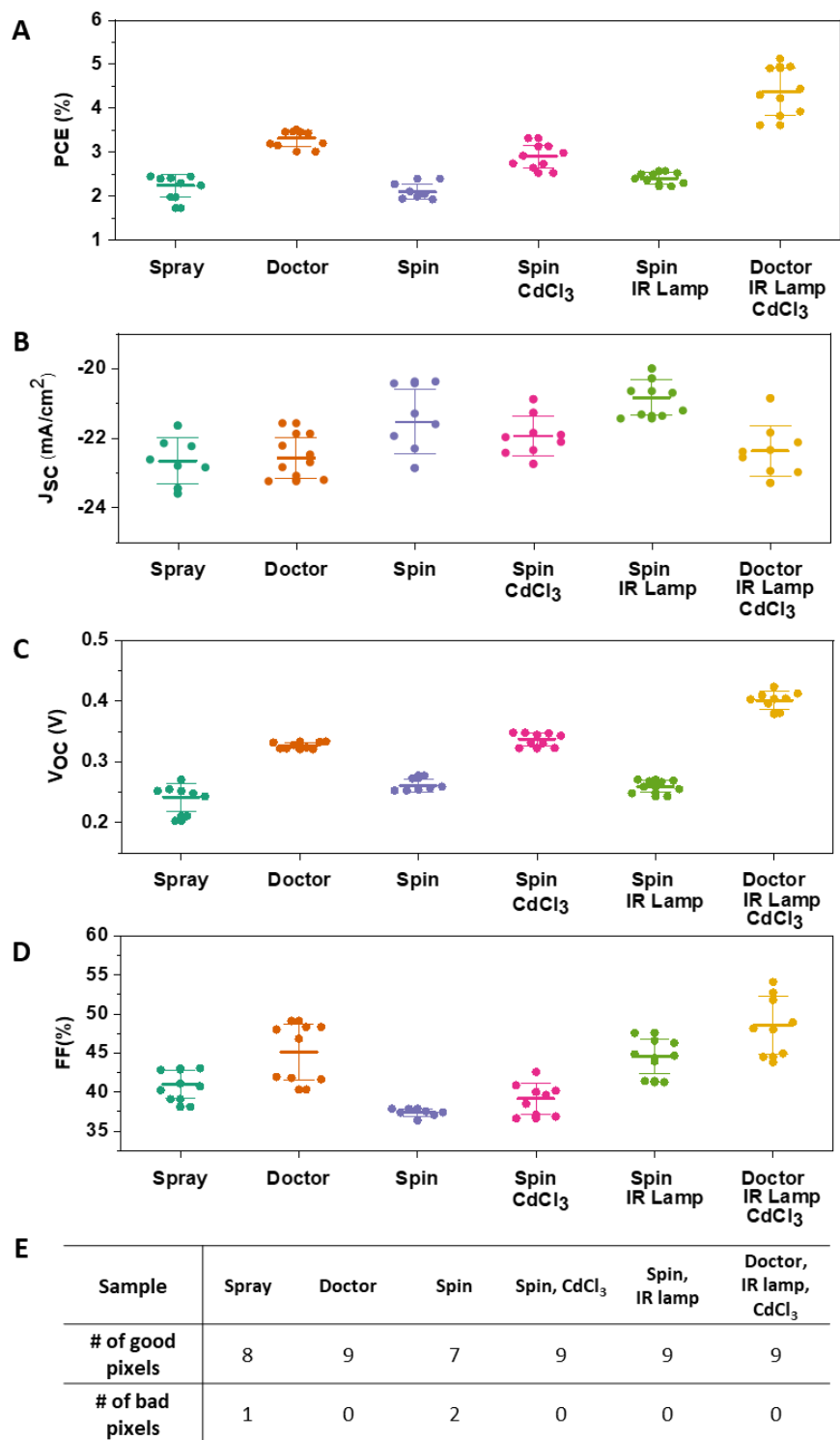
Integrated Device



**Figure S9.** Additional cross-sectional SEM images of devices made or with standard, non-R2R-friendly techniques (left column) or with R2R-friendly techniques (right column).

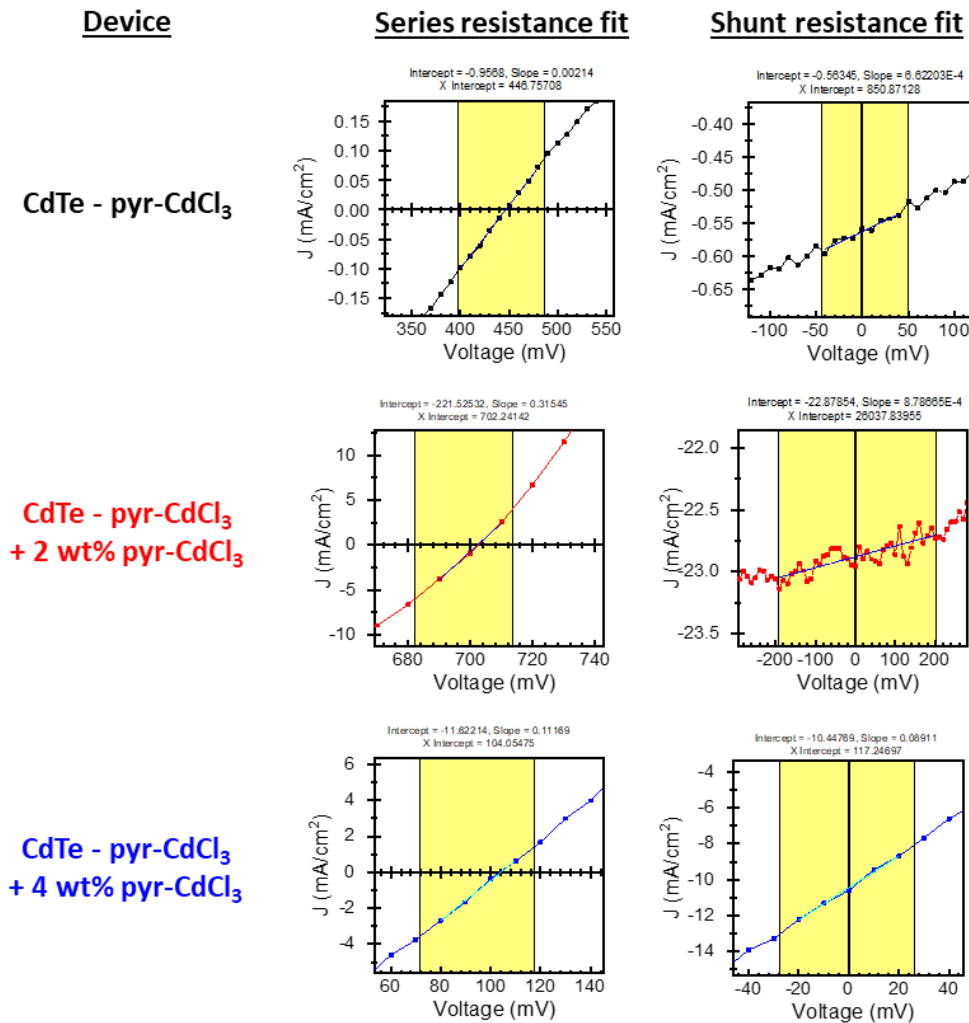
**Scheme S1.** Integrated R2R-friendly deposition of sintered CdTe NCs.





**Figure S10.** Statistics of device performance before current/light soaking. Unless otherwise noted, devices were made using a CdCl<sub>2</sub> bath and annealing with a hot plate. (A–D) Power conversion efficiency (A), short-circuit current (B), open-circuit voltage (C) and fill-factor (D) statistics. (E) Table showing the number of good and bad pixels in a 9-pixel device. The bad pixels were not included in the analysis.

| Deposition  | Series resistance, $R_s$<br>( $\Omega \text{ cm}^2$ ) | Shunt resistance, $R_s$<br>( $\Omega \text{ cm}^2$ ) |
|---|---|--|
| CdTe - pyr-CdCl <sub>3</sub>                                  | 467   | $1.51 \times 10^2$                                   |
| CdTe - pyr-CdCl <sub>3</sub><br>+ 2 wt% pyr-CdCl <sub>3</sub> | 3.17  | $1.14 \times 10^2$                                   |
| CdTe - pyr-CdCl <sub>3</sub><br>+ 4 wt% pyr-CdCl <sub>3</sub> | 8.95  | $1.12 \times 10^1$                                   |



**Figure S11.** Fittings to obtain shunt and series resistances from J-V curves for devices made with various amounts of CdCl<sub>2</sub> added.

## Supplementary References

1. MacDonald, B. I.; Gaspera, E. D.; Watkins, S. E.; Mulvaney, P.; Jasieniak, J. J., Enhanced Photovoltaic Performance of Nanocrystalline CdTe/ZnO Solar Cells Using Sol-Gel ZnO and Positive Bias Treatment. *J. Appl. Phys.* **2014**, *115* (18), 184501.
2. Jasieniak, J.; MacDonald, B. I.; Watkins, S. E.; Mulvaney, P., Solution-Processed Sintered Nanocrystal Solar Cells via Layer-by-Layer Assembly. *Nano Lett.* **2011**, *11* (7), 2856-2864.
3. Zhang, H.; Kurley, J. M.; Russell, J. C.; Jang, J.; Talapin, D. V., Solution-Processed, Ultrathin Solar Cells From  $\text{CdCl}_3^-$  - Capped CdTe Nanocrystals: The Multiple Roles Of  $\text{CdCl}_3^-$  Ligands. *J. Am. Chem. Soc.* **2016**, *138* (24), 7464-7467.