Supporting Information

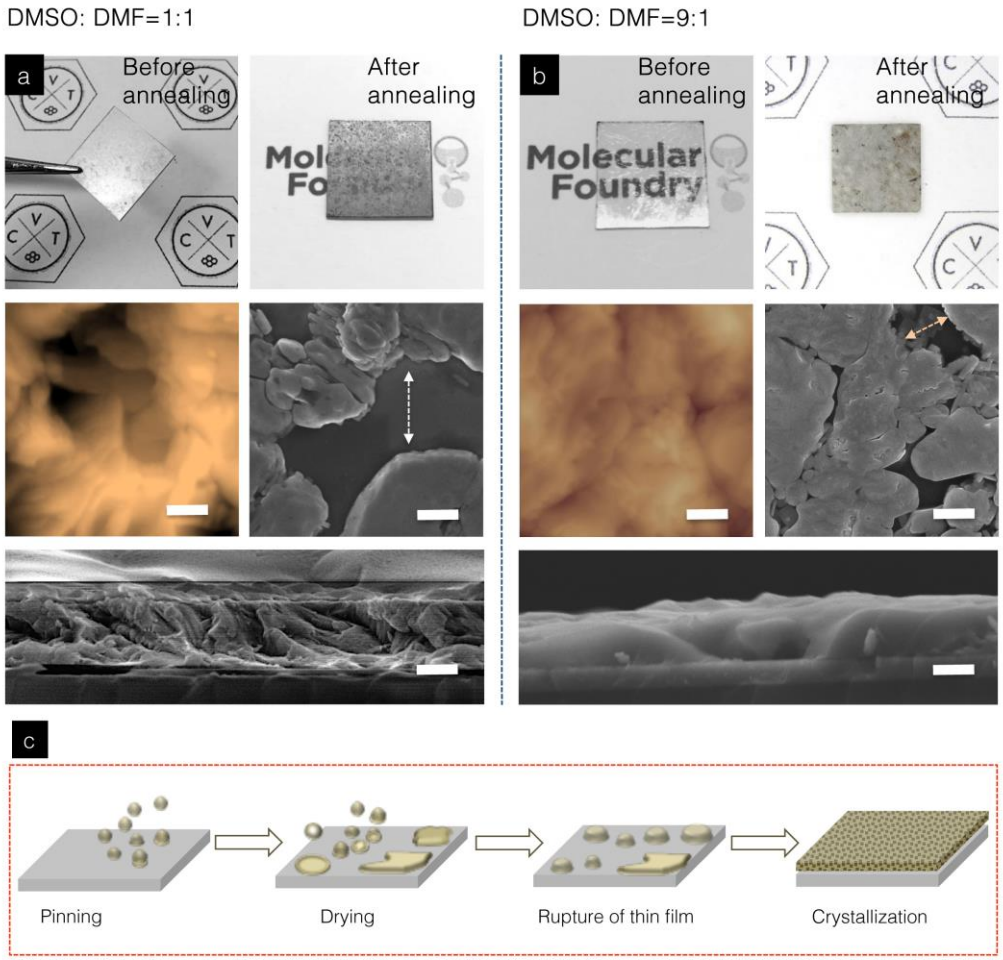

Figure S1. (a) PVSK thin films made of a mixture of DMSO: DMF (1:1, v/v) displayed a poor coverage after deposition and became even worse upon annealing. AFM and SEM images collectively suggest the insufficient coverage stems from the high density of pores and discontinuous crystalline networks. Cross-sectional SEM image further reveals the grainy and rough interfaces. The typical crystalline size is small than 300 nm. (b) Increasing the volume ratio of DMSO decreases both the number and dimension of the pores. However, the lack of dynamic solvent fronts results in unwanted de-wetting process, including (c) pinning, drying and subsequent rupture of wet films. As a result, PVSK thin films mainly comprise discontinuous crystalline domains with peak-to-valley variation up to tenth of μm. Scale bars are 200 nm for AFM and cross-sectional SEM, and 2μm for SEM, respectively.

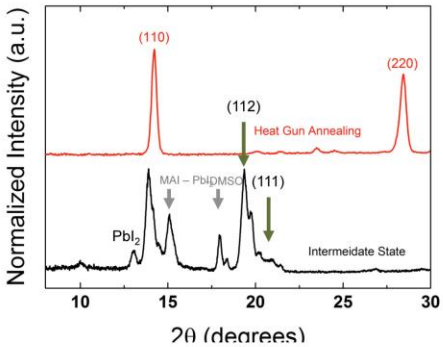

Figure S2. The fully crystalized (red) and intermediate phase (black) PVSKs display distinctively different XRD patterns. Thin films of PVSK precursors collected right before turning into dark brown/matte black in color still show the existence of DMSO that ligates to the $PbCl_2$ and MAI (labeled in gray and olive arrows). Green arrows indicate the XRD patterns that can be assigned to the intermediate phase.

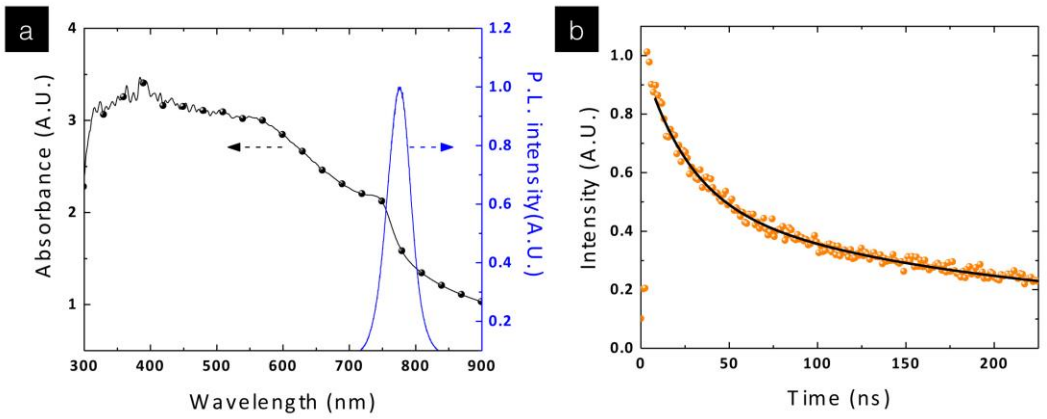

Figure S3. (a) Broad UV/Vis spectrum that spans the entire visible region in conjunction with the narrow distribution of full width at half maximum of PL conclusively indicates the full conversion of PVSK precursors into crystallized thin films. (b) Carrier life time can be deconvoluted into a bimodal mode, an indication of continuous and hybrid morphology.

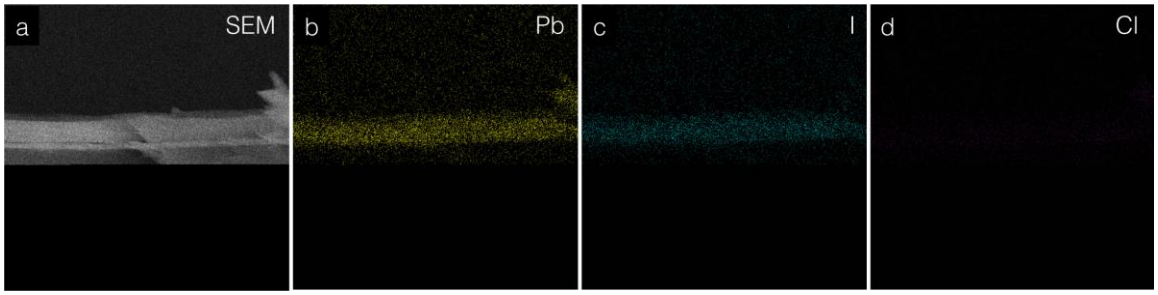

Figure S4. (a) Cross-sectional SEM image of PVSK thin films produced through Marangoni-inspired process. EDX mapping reveals the spatial distribution of relevant elements (b) Pb in yellow, (c) I in cyan and (d) Cl in red, respectively.

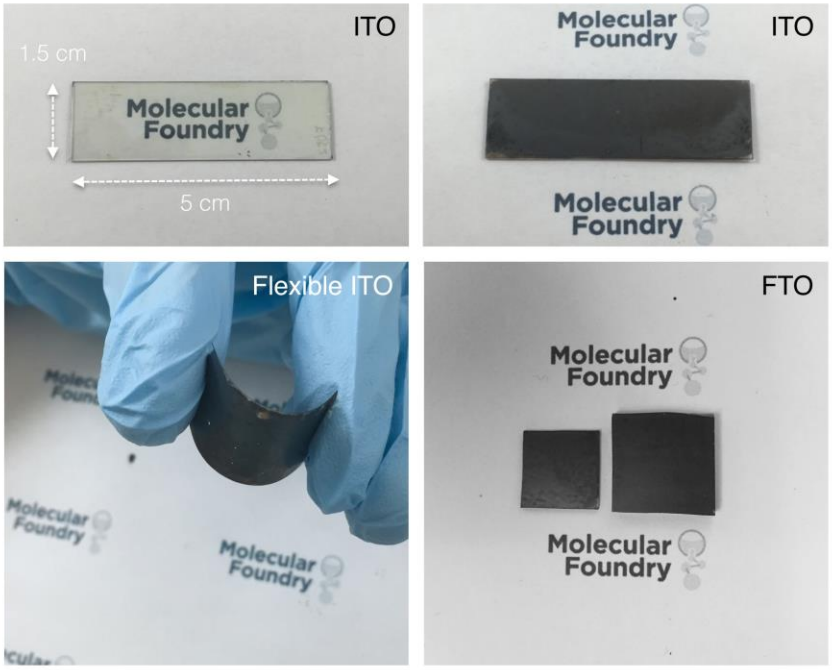

Figure S5. The versatile yet general EHD strategy is manifested in the deposition of large area PVSK thin films as well as on a myriad of different substrates, such as flexible ITO and FTO.

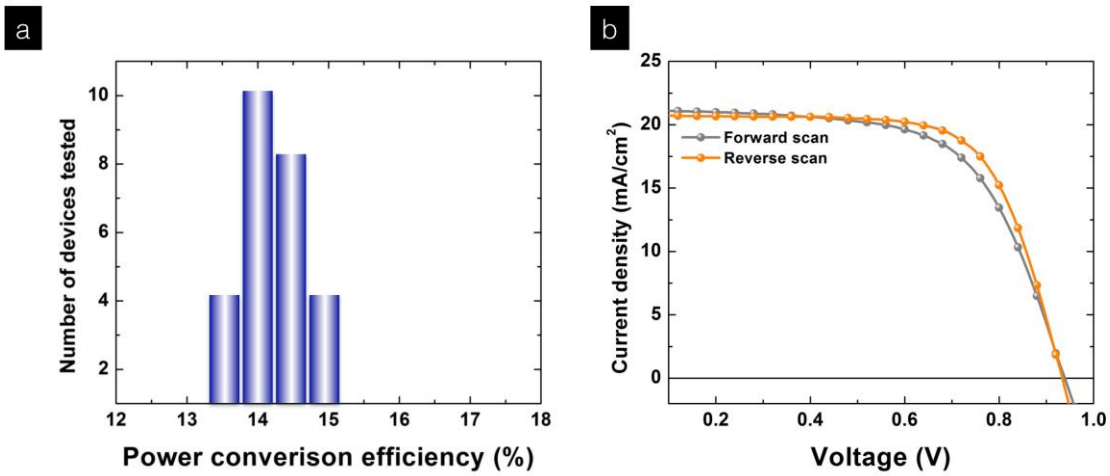

Figure S6. (a) Histogram shows the distribution of power conversion efficiency over 28 devices. (b) In particular, output characteristics only display a minor hysteresis, mainly due to the *FF* when scanned in both directions.

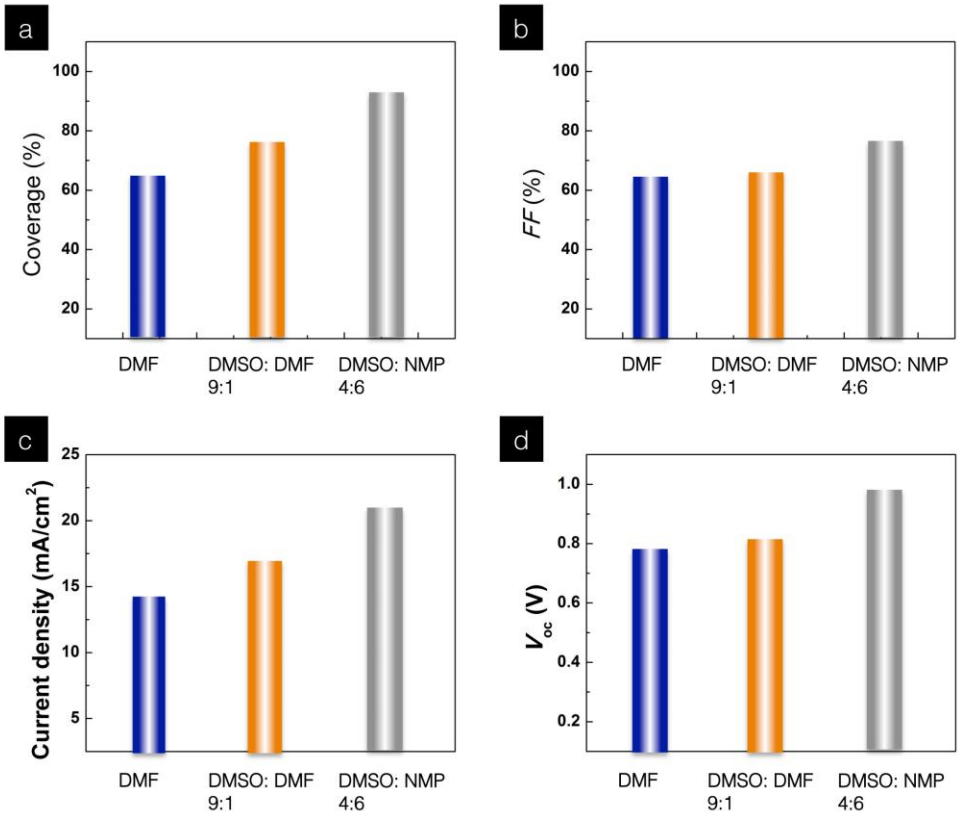

Figure S7. (a) Coverage of PVSK thin films deposited from DMF (blue), DMSO: DMF (orange) and DMSO: NMP (gray), respectively. Solvent ratios that resulted in the highest coverage are selected for comparison. (b) *FF*, (c) J_{sc}, and (d) V_{oc} collectively show the coverage dependence. High coverage delivers improved output characteristics.