Supplementary Information (SI) for Journal of Materials Chemistry A.

Supporting Information:

CsCl Seed Layer Engineering for Buried-Interface Modification Enabling >20% Efficiency in Single-Source Vapor-Deposited Perovskite Solar Cells

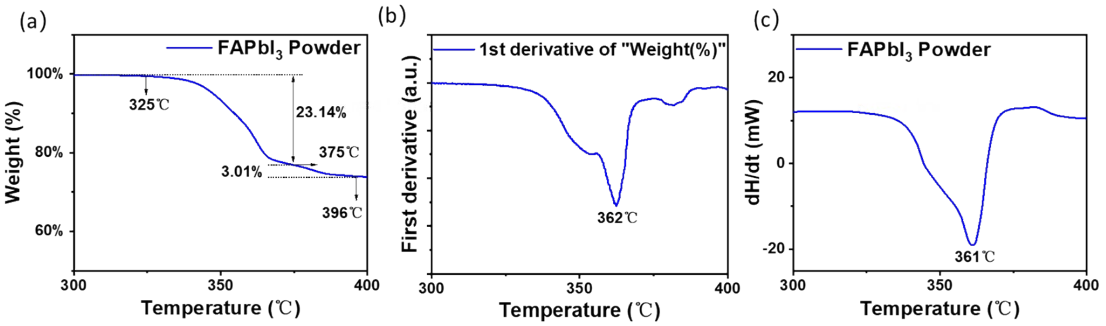

Fig. S1: (a) TGA curve, (b) 1st derivative of TGA curve and (c) DSC curve of $FAPbI_3$ powder.

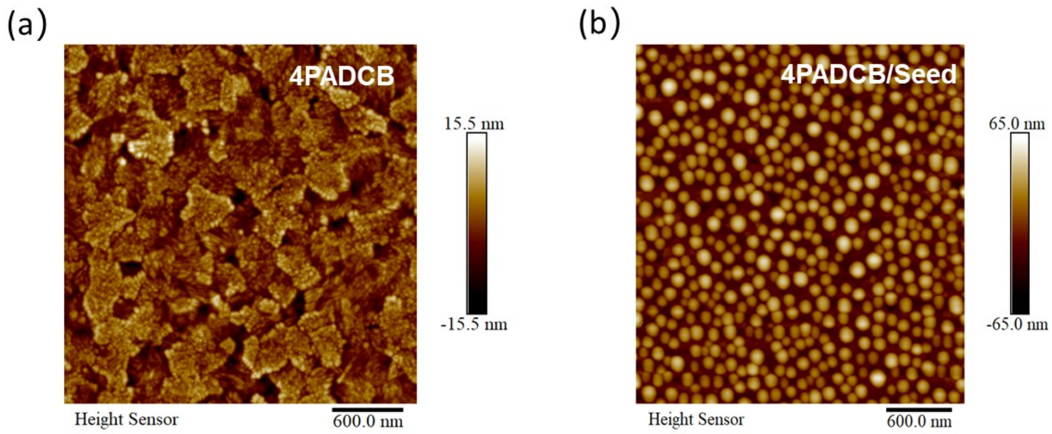

Fig. S2: The AFM images of (d) 4PADCB film and (e) 4PADCB/Seed film.

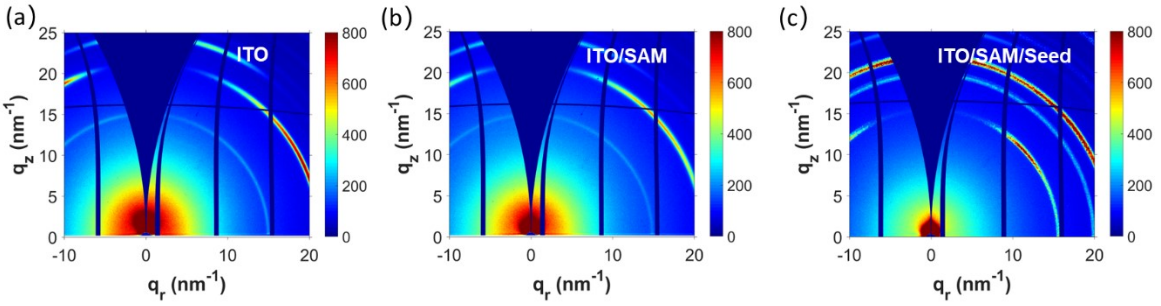

Fig S3: Two-dimensional GIWAXS (2D-GIWAXS) patterns of (a) ITO, (b) ITO/4PADCB (c) ITO/4PADCB/Seed.

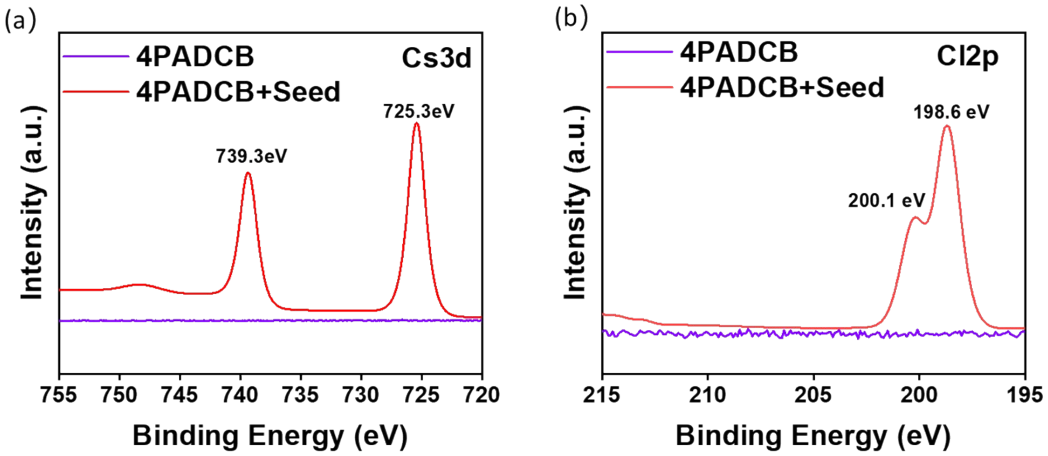

Fig. S4: The XPS patterns of (a) Cs3d and (b) Cl2p of 4PADCB and 4PADCB+Seed.

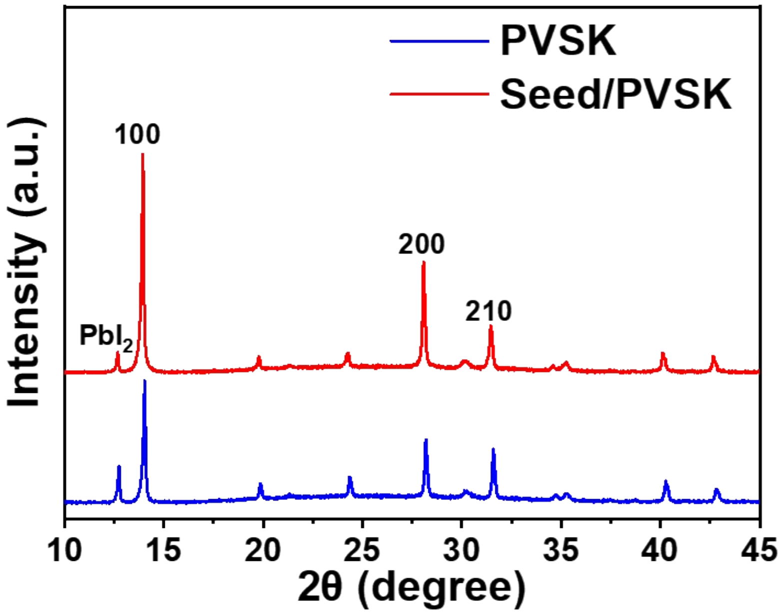

Fig. S5 : The XRD patterns of PVSK and Seed/PVSK films.

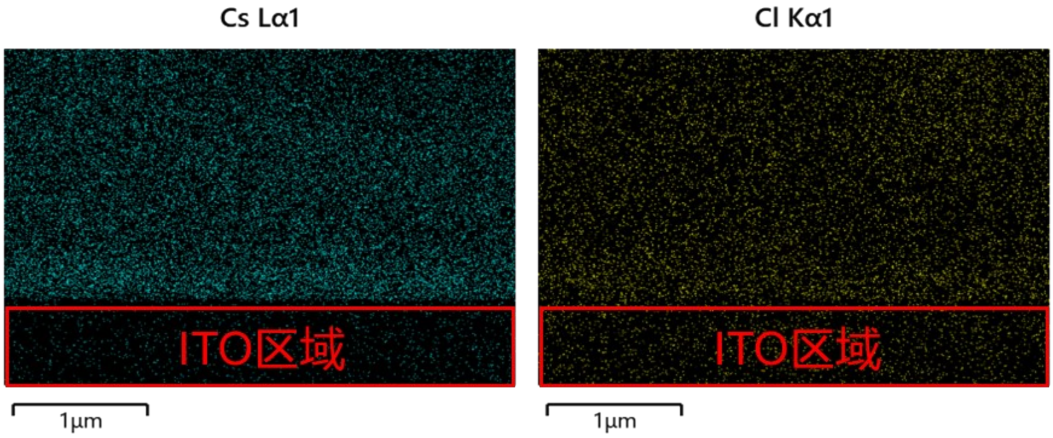

Fig. S6: EDS data of the ITO/4PADCB/Seed/perovskite structure, including Cs, Cl.

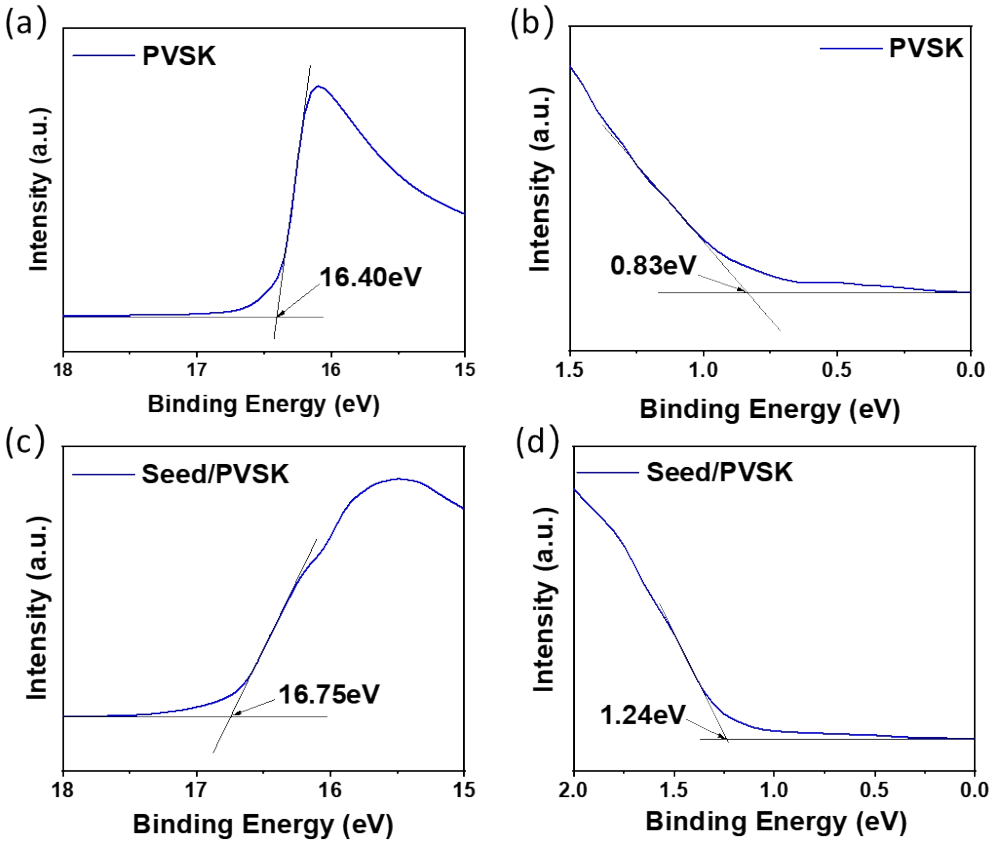

Fig. S7: UPS spectra of PVSK sample in the (a) onset and (b) cutoff energy region. UPS spectra of Seed/PVSK samples in the (c) onset and (d) cutoff energy region.

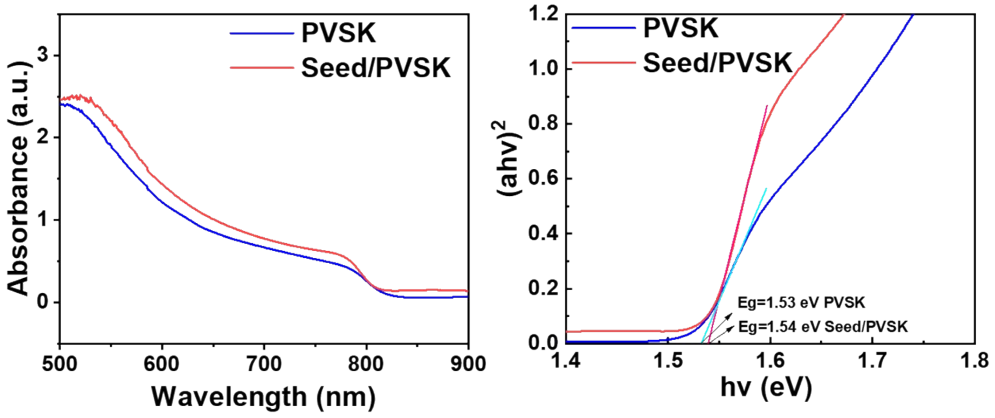

Fig. S8: The Absorbance (a) and UPS spectra (b) of perovskite films on PVSK and Seed/PVSK.

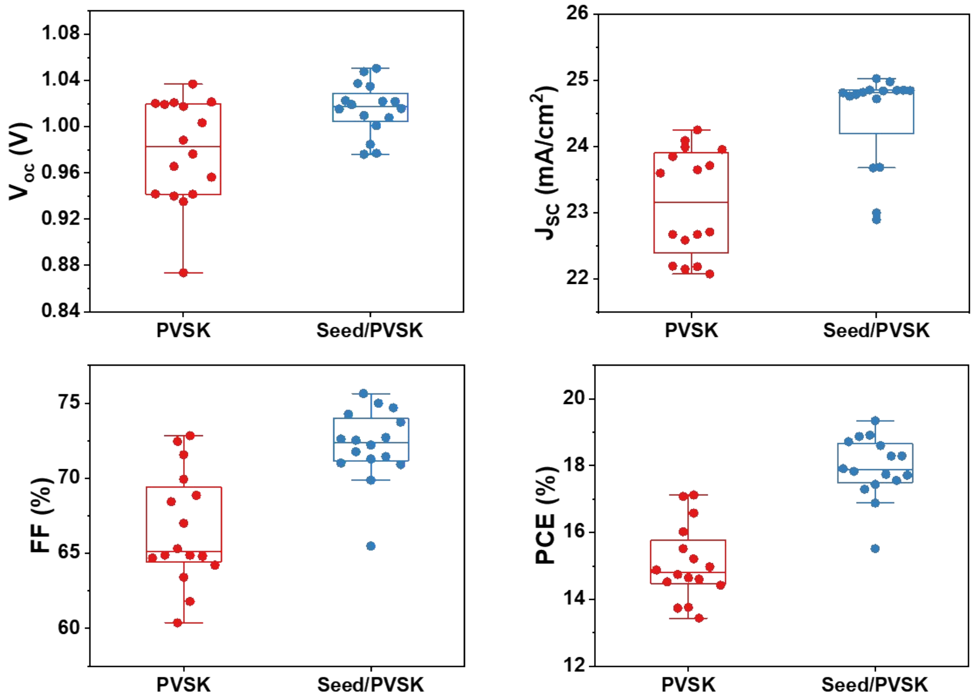

Fig. S9: Statistical photovoltaic parameters of (a) V_{oc}, (b) J_{sc}, (c) FF, (d) PCE of PSCs using PVSK and Seed/PVSK.

Table S1. Fitted TRPL parameters of PVSK with and without CsCl seed layer.

	τave(ns)	A1	τ_1(ns)	A2	τ_2(ns)
PVSK	54.80	0.77	3.91	0.14	70.07
Seed/PVSK	177.33	0.46	25.7	0.48	196.2

Table S2. PV parameters extracted from the most efficient PSCs under different structures.

Structure	Voc(V)	Jsc(mA/cm^2)	FF(%)	PCE(%)
ITO/2PACz/FAPbI$_3$/C60/SnO$_X$/Ag	1.001	20.546	62.003	12.747
ITO/Meo-2PACz/FAPbI$_3$/C60/SnO$_X$/Ag	0.991	23.550	72.203	16.851
ITO/4PADCB/FAPbI$_3$/C60/SnO$_X$/Ag	1.018	23.709	72.268	17.458
ITO/4PADCB:Meo-2PACz=1:1/FAPbI$_3$/C60/SnO$_X$/Ag	0.947	24.318	68.858	15.862
ITO/NiOx/Me-4PACz/FAPbI$_3$/C60/SnOX/Ag	1.083	20.158	63.572	13.879
ITO/4PADCB/5nmCsCl/FAPbI$_3$/C60/SnO$_X$/Ag	1.008	24.877	73.950	18.536
ITO/4PADCB/15nmCsCl/FAPbI$_3$/C60/SnO$_X$/Ag	1.042	25.097	77.435	20.258

ITO/4PADCB/25nmCsCl/$FAPbI_3$/C60/SnO_X/Ag	1.012	25.450	73.821	19.009
ITO/4PADCB/35nmCsCl/$FAPbI_3$/C60/SnO_X/Ag	0.985	24.841	72.529	17.741

Table S3. The report presents the device efficiency and publication dates corresponding to the single-source vapor deposition methods for solar cell devices with different compositions.

Ref.	Method	Device stack (Composition)	PCE (%)	Year
1	Thermal evaporation	ITO/PEDOT:PSS/ $MAPbI_3$/PCBM/Ag	10.9	2016
2		ITO/PEDOT:PSS/$MAPbI_3$/$PC_{61}BM$/Ag	7.7	2018
3		ITO/PEDOT:PSS/ $MAPbI_3$/PCBM/Ag	9.9	2019
4		FTO/TiO_2/$Cs_2AgBiBr_6$/Spiro-OMeTAD/Ag	0.7	2019
5		FTO/PCBM/$CsSn_{0.5}Ge_{0.5}I_3$/Spiro-OMeTAD/Au	7.1	2019
6		FTO/c-TiO_2/$CsPbBr_3$/Spiro-OMeTAD/Au	8.7	2020
7		FTO/TiO_2/$CsSn_{1-y}Pb_yBr_3$/carbon	9	2021
8		FTO/TiO_2/$CsPbBr_3$/Spiro-OMeTAD/Ag	1.37	2021
9		FTO/TiO_2/$Cs_2AgBiBr_6$/SpiroOMeTAD/Ag	1.38	2022
10		ITO/PEDOT:PSS/$MASnI_3$/C_{60}/Au	1.7	2022
11		FTO/TiO_2/$CsPbBr_3$/Carbon	7.5	2023
12		ITO/MeO-2PACz/$Cs_xPbI_yBr_z$/C_{60}/BCP/Ag	15.0	2023
13		FTO/TiO_2/$CsPbBr_3$ QDs/$CsPbBr_3$/Spiro-OMeTAD/Au	7.0	2023
14		ITO/TiO_2:SnO_2NRs/$(BA)_2(MA)_3Pb_4I_{13}$/Spiro-OMeTAD/Au	13.6	2024
This work		ITO/4PADCB/PVSK/C_{60}/SnO_2/Ag	17.8,	2025

		ITO/4PADCB/CsCl/PVSK/C$_{60}$/SnO$_2$/Ag	20.26	
15	Flash evaporation	ITO/PEDOT:PSS/PolyTPD/MAPbI$_3$/PCBM/Ba/Ag	12.2	2015
16		FTO/PCBM/MAPЬI$_3$/Spiro-OMeTAD/Au	10.0	2016
17		FTO/TiO$_2$/PCBM/MAPbI$_3$/Spiro-OMeTAD/Au	16.8	2018
18		FTO/PCBM/MAPЬI$_3$/Spiro-OMeTAD/Au	12.2	2019
19		FTO/PCBM/FAPbI$_3$/Spiro-OMeTAD/Au	12.5	2019
20		ITO/SnO$_2$/CsPb(I$_{0.67}$Br$_{0.33}$)$_3$/Spiro-MeOTAD/Au	15.0	2024
21	PLD	ITO/ZnO/MAPb(I$_y$Cl$_{1-y}$)$_3$/Spiro-OMeTAD/Au	7.7	2015
22		FTO/TiO2/MAPbI$_3$/Spiro-OMeTAD/Au	10.9	2017
23		FTO/c-TiO$_2$/m-TiO$_2$/CsPbBr$_3$/Spiro-OMeTAD/Ag	6.3	2019
24		ITO/SnO2/C60/MAPbI$_3$/TaTm/MoO$_3$/Au	6.7	2021
25		ITO/SnO$_2$(ALD)/C60/MA$_x$FA$_{1-x}$PbI$_3$/TaTm/MoO$_3$/Au ITO/SnO$_2$/PCBM/MA$_x$FA$_{1-x}$PbI$_3$/Spiro-OMeTAD/Au	9.9	2023
26		ITO/2PACz/MA$_{1-x}$FA$_x$PbI$_3$/C$_{60}$/BCP/Ag	19.7	2024
27	Electron beam evaporation	FTO/c-TiO$_2$/CsPbBr$_3$/carbon	7.8	2022
28	sputtering	MAPbI$_3$, MAxPbI$_3$Cly	4.7 15.2	2021

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