Perovskite Solar Cells

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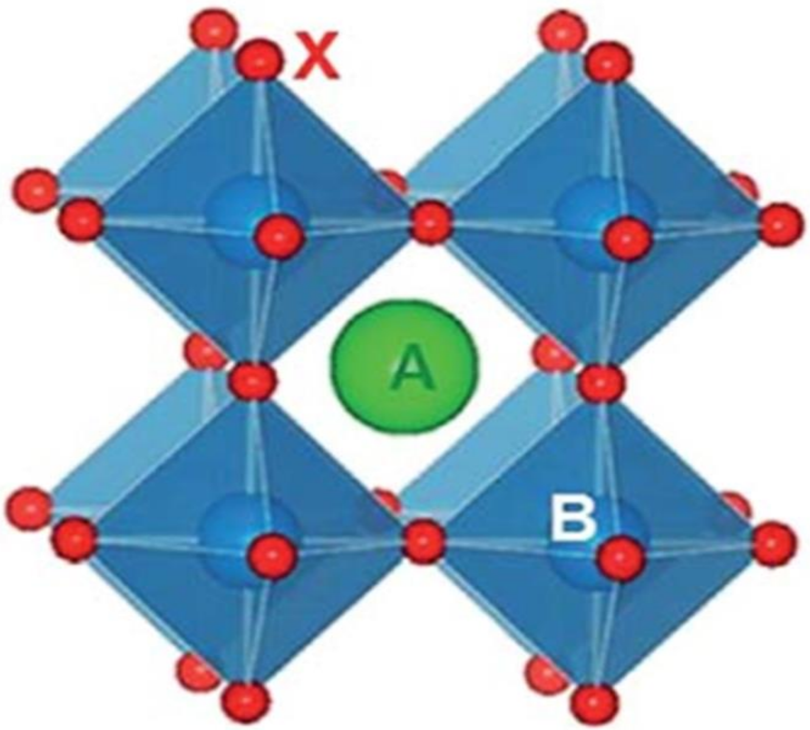
Department of Applied Chemistry
Institute of Molecular Science
National Yang-Min Chiao-Tung University (NYCU)
Hsinchu, Taiwan

Crystal Structure of Organic-Inorganic Perovskites

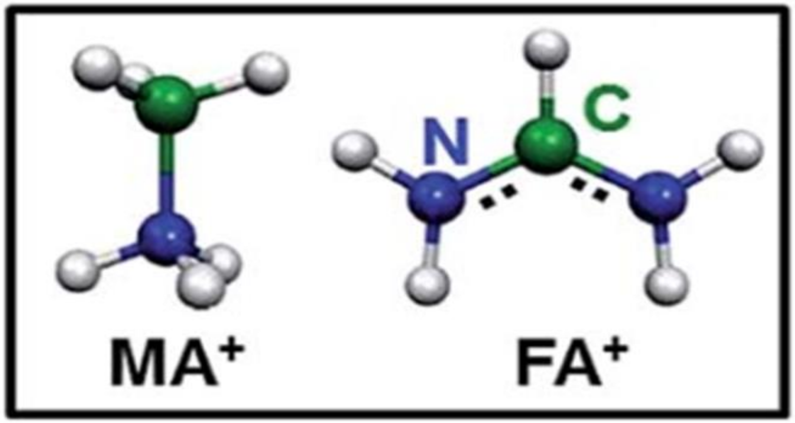


Methyl Ammonium Lead Iodide

A = Cs^+, MA^+ or FA^+

X = I^-, Br^- or Cl^-

B = Pb^{2+} or Sn^{2+}



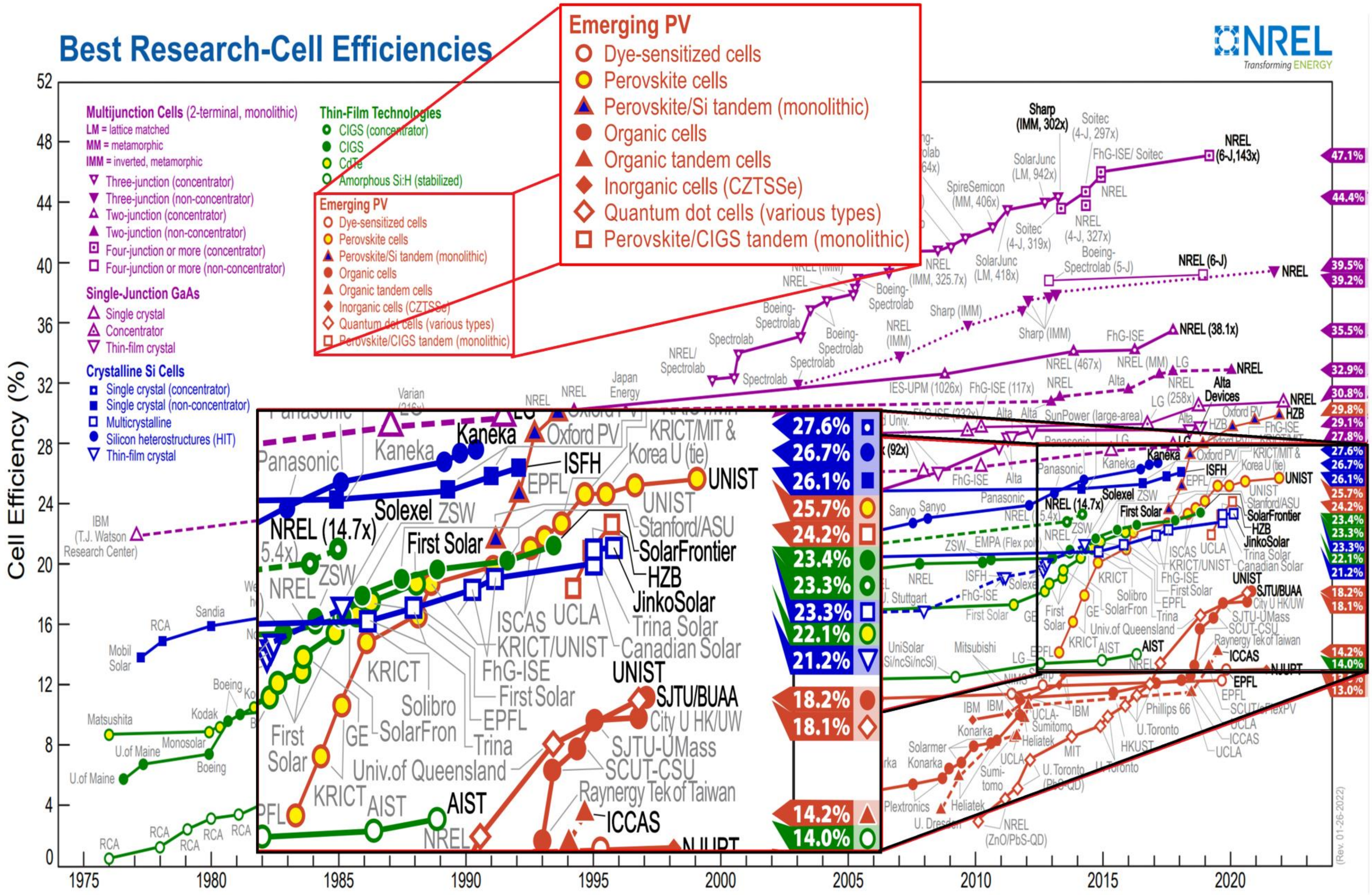
$\underline{CH_3NH_3}\ \underline{Pb}\ \underline{I_3}$

A B X_3

ORGANIC

INORGANIC

Rapid Progress of PSC on 2014-2022



The First Perovskite-sensitized Solar Cells

Tom Miyasaka
(Toin U of Yokohama)

J|A|C|S COMMUNICATIONS

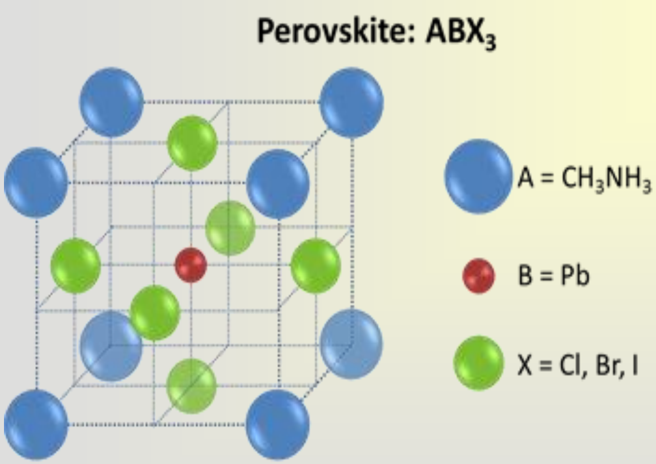
Published on Web 04/14/2009

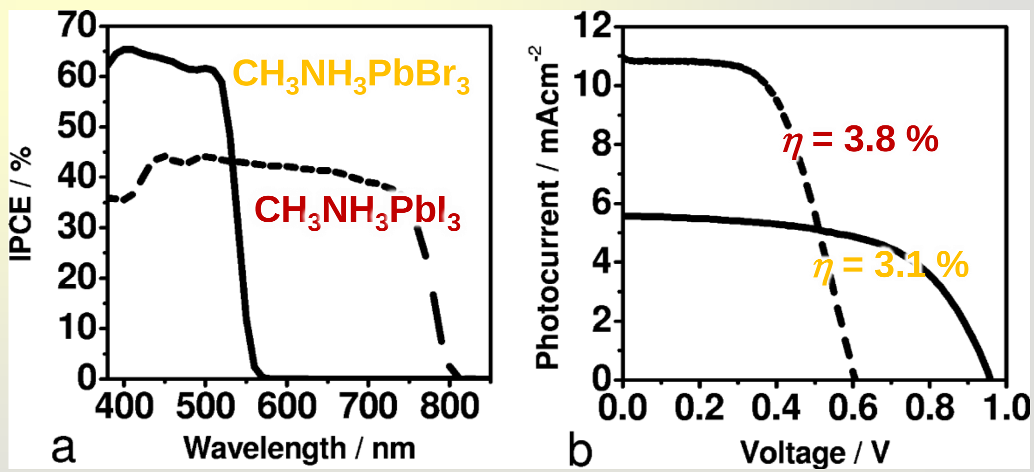
Organometal Halide Perovskites as Visible-Light Sensitizers for Photovoltaic Cells

Akihiro Kojima,[†] Kenjiro Teshima,[‡] Yasuo Shirai,[§] and Tsutomu Miyasaka*,[†,‡,∥]

Graduate School of Arts and Sciences, The University of Tokyo, 3-8-1 Komaba, Meguro-ku, Tokyo 153-8902, Japan, Graduate School of Engineering, Toin University of Yokohama, and Peccell Technologies, Inc., 1614 Kurogane-cho, Aoba, Yokohama, Kanagawa 225-8502, Japan, and Graduate School of Engineering, Tokyo Polytechnic University, 1583 Iiyama, Atsugi, Kanagawa 243-0297, Japan

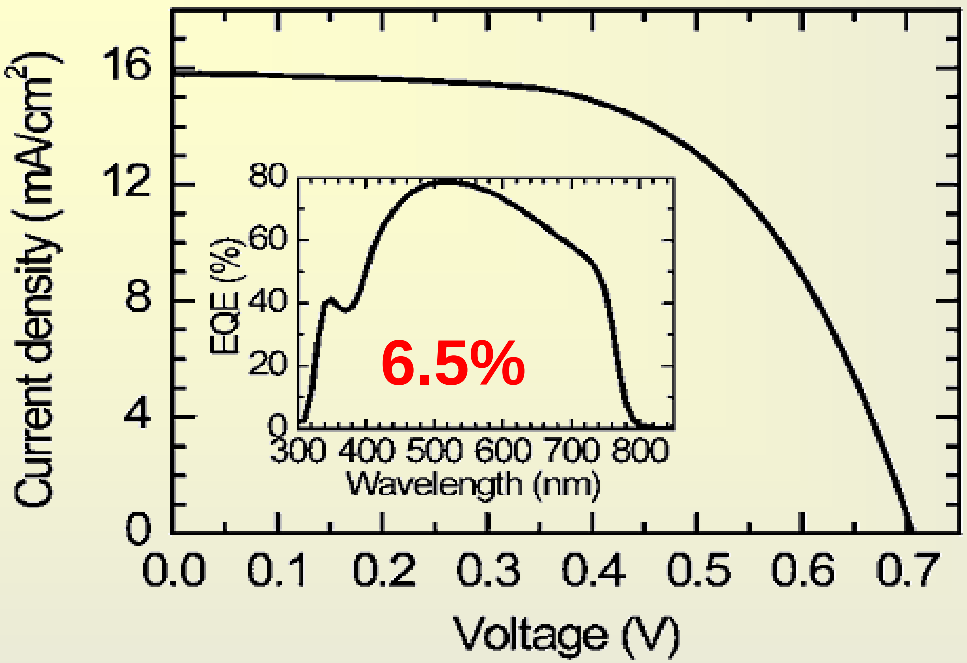
Received December 9, 2008; Revised Manuscript Received April 1, 2009; E-mail: miyasaka@cc.toin.ac.jp





J. Am. Chem. Soc. 131(17), 2009, 6050–6051.

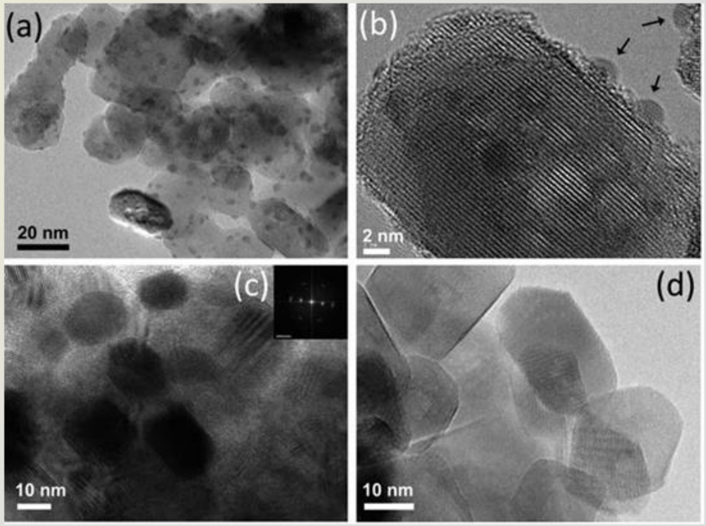
Liquid-type Perovskite-sensitized Solar Cells

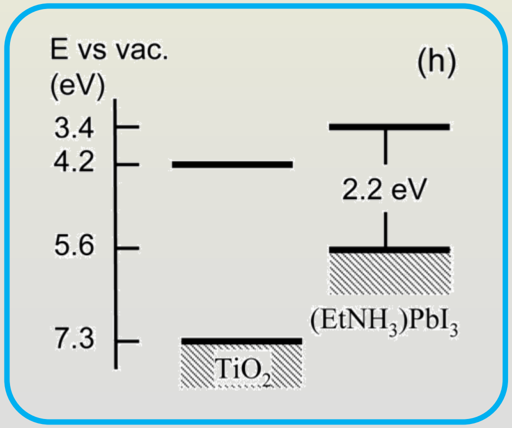


Electrolyte : Iodide/triiodide

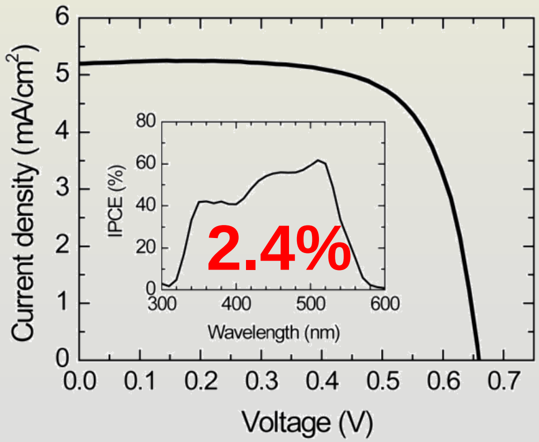
Sensitizer : $CH_3NH_3PbI_3$

Nam Gyu Park (SKKU)





$C_2H_5NH_3PbI_3$

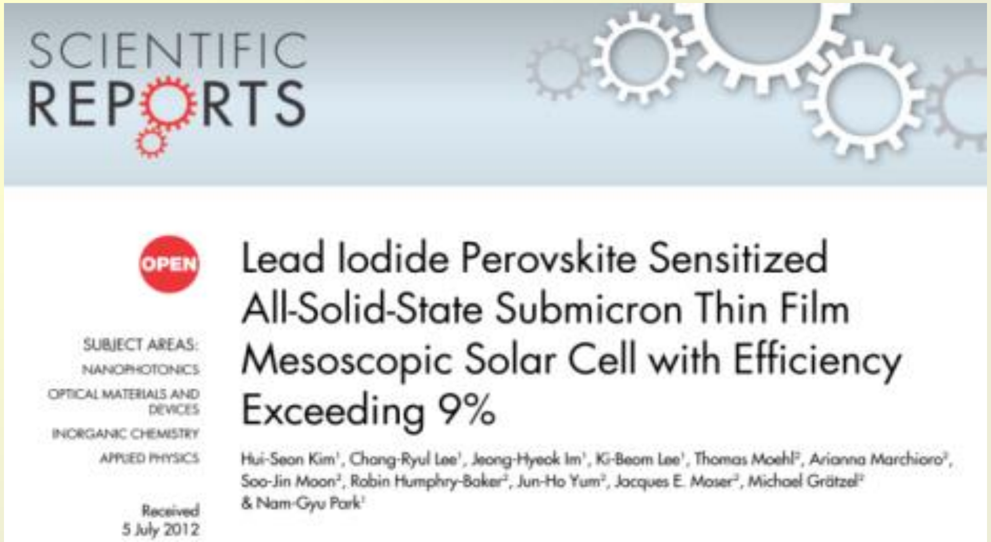


Nanoscale **2011**, *3*, 4088–4093

Nanoscale Research Letters **2012**, *7*, 353

All-Solid-State Perovskite Solar Cells

TiO_2-based Perovskite-sensitized SC


SCIENTIFIC REPORTS

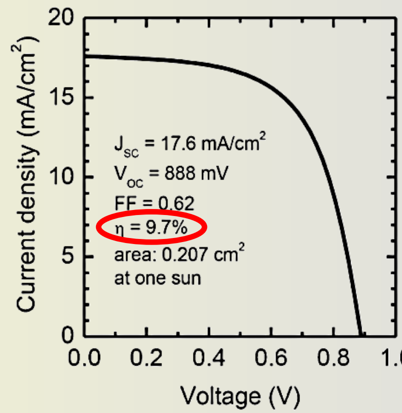
OPEN

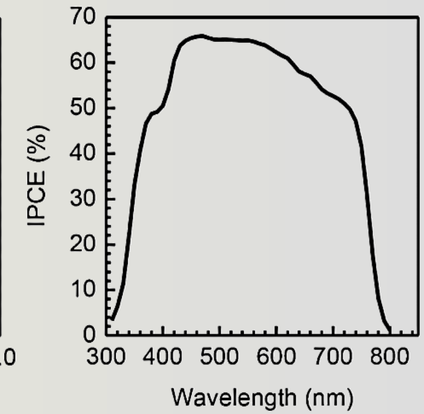
SUBJECT AREAS:
NANOPHOTONICS
OPTICAL MATERIALS AND DEVICES
INORGANIC CHEMISTRY
APPLIED PHYSICS

Received
5 July 2012

Lead Iodide Perovskite Sensitized All-Solid-State Submicron Thin Film Mesoscopic Solar Cell with Efficiency Exceeding 9%

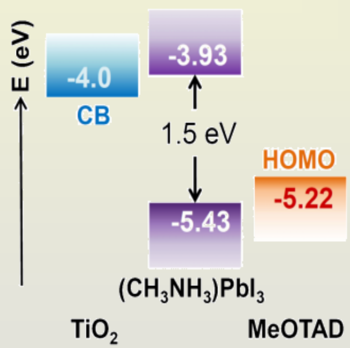
Hui-Seon Kim[1], Chang-Ryul Lee[1], Jeong-Hyeok Im[1], Ki-Beom Lee[1], Thomas Moehl[2], Arianna Marchioro[2], Soo-Jin Moon[2], Robin Humphry-Baker[2], Jun-Ho Yum[2], Jacques E. Moser[2], Michael Grätzel[2] & Nam-Gyu Park[1]

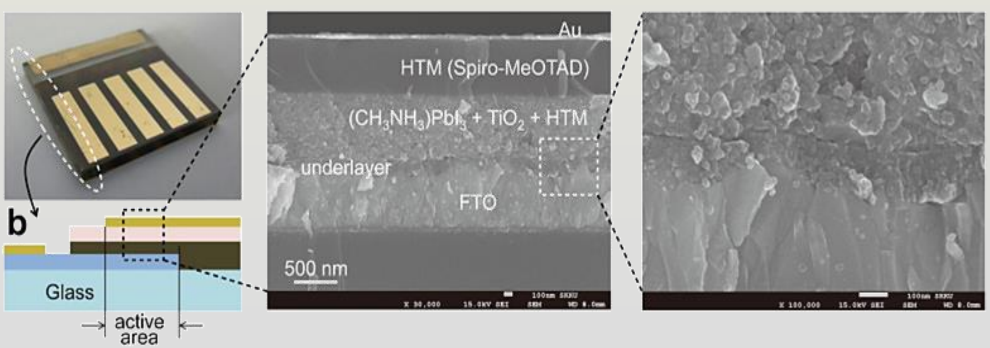


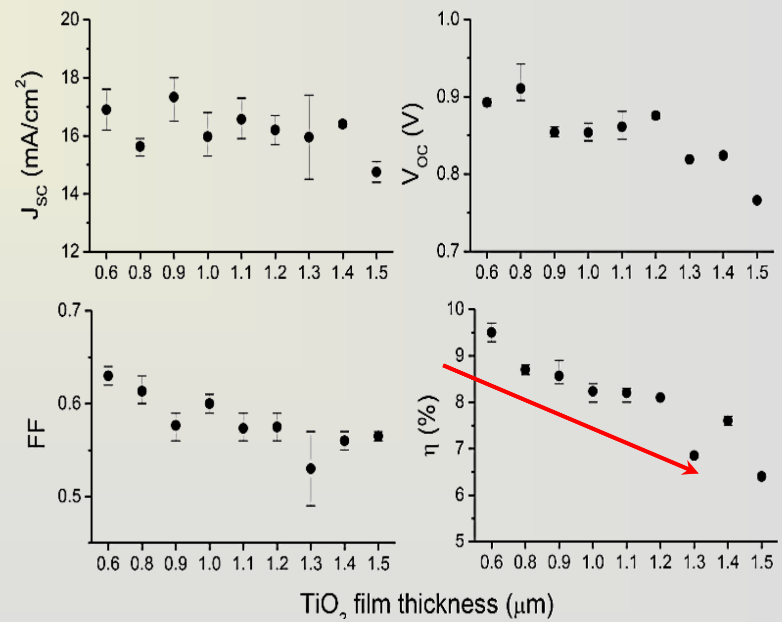



Nam Gyu Park


Michael Grätzel







Scientific Reports 2(1), 2012, 591

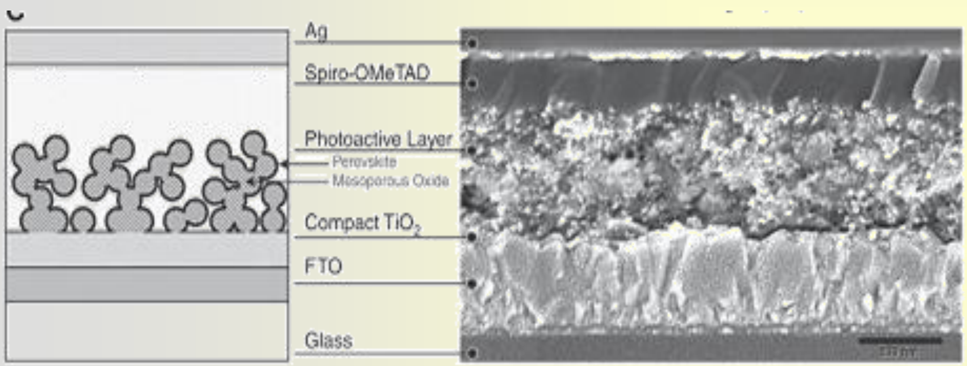
Al_2O_3-based Perovskite Solar Cells

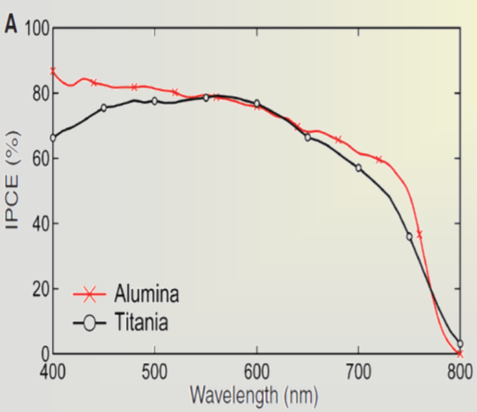
Efficient Hybrid Solar Cells Based on Meso-Superstructured Organometal Halide Perovskites

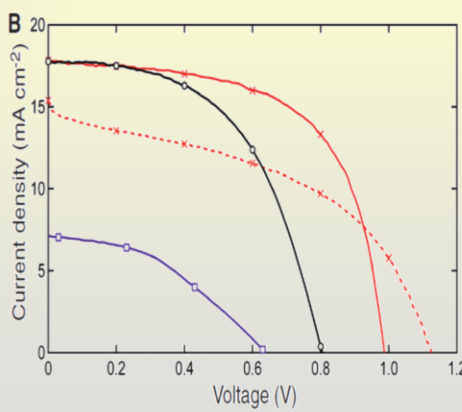
Michael M. Lee,[1] Joël Teuscher,[1] Tsutomu Miyasaka,[2] Takurou N. Murakami,[2,3] Henry J. Snaith[1]*

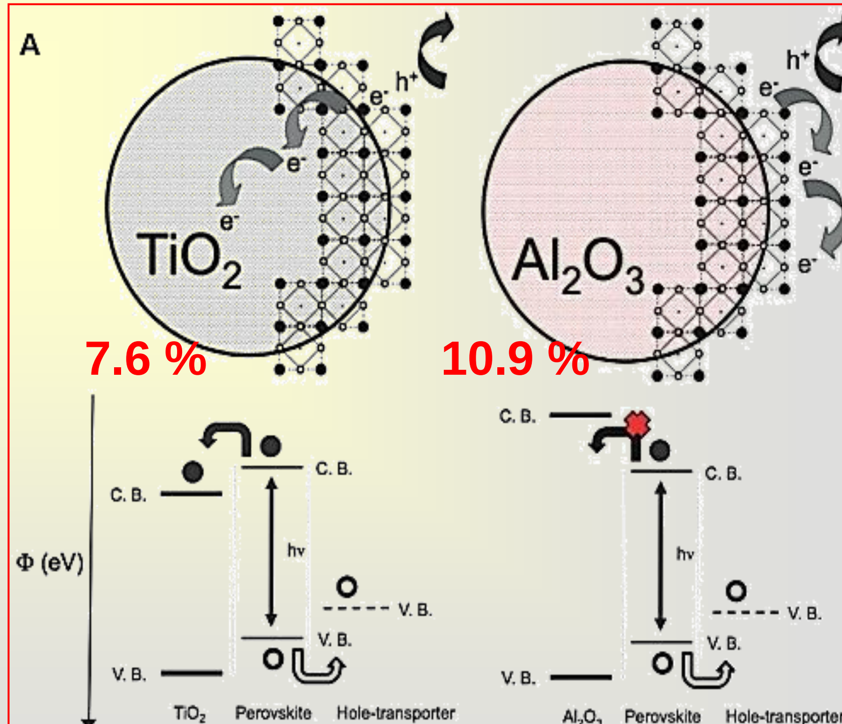
Tom Miyasaka
(Toin University)

Henry Snaith
(Oxford U)









Science, 338, 2012, 643−647.

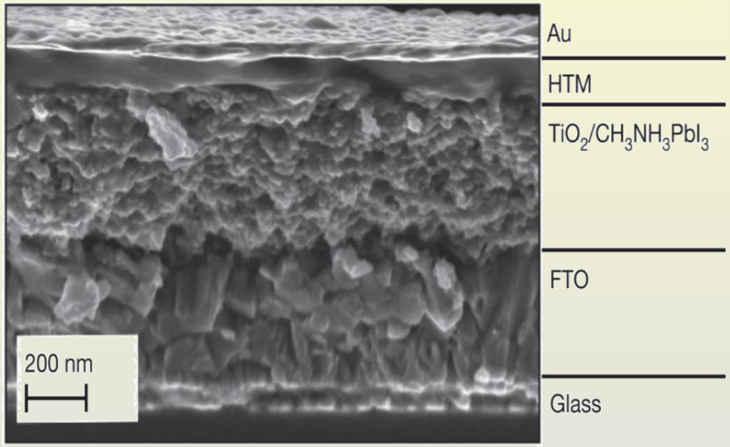
A Two-step Approach for PCE 15 %

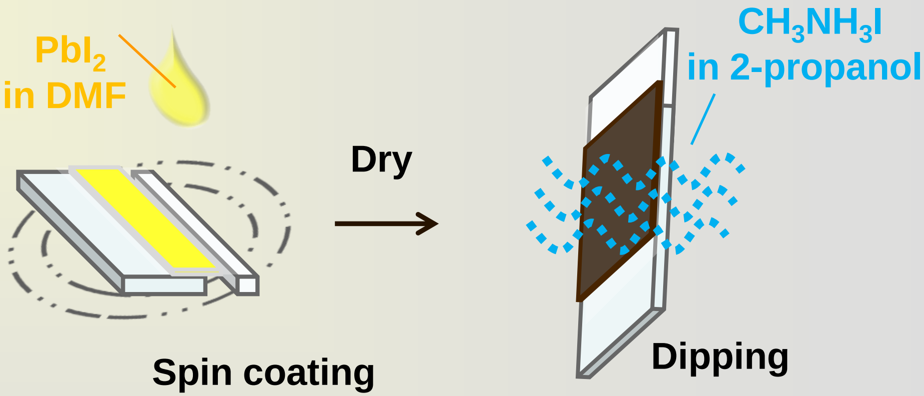
LETTER

doi:10.1038/nature12340

Sequential deposition as a route to high-performance perovskite-sensitized solar cells

Julian Burschka[1]*, Norman Pellet[1,2]*, Soo-Jin Moon[1], Robin Humphry-Baker[1], Peng Gao[1], Mohammad K. Nazeeruddin[1] & Michael Grätzel[1]





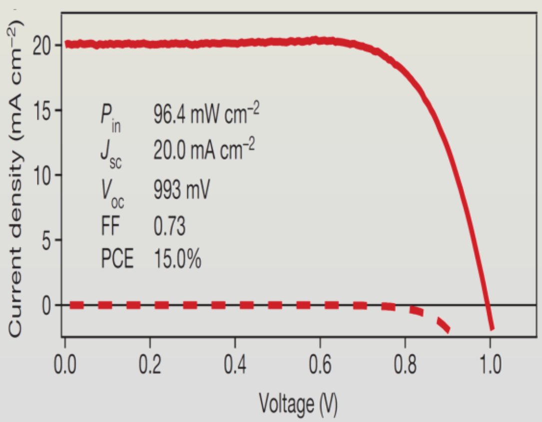


Table 1 | Photovoltaics performance at different light intensities

Intensity (mW cm^{-2})	J_{sc} (mA cm^{-2})	V_{oc} (mV)	Fill factor	PCE (%)
9.3	1.7	901	0.77	12.6
49.8	8.9	973	0.75	13.0
95.6	17.1	992	0.73	12.9

Nature **2013**, *499*, 316-320

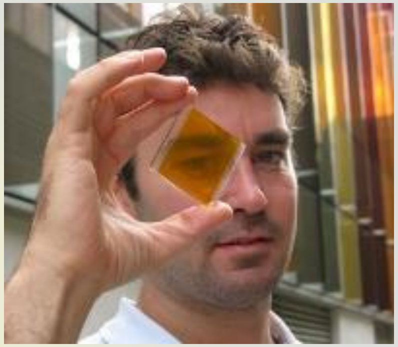
A Thin-film Approach for PCE Exceeding 15 %

LETTER

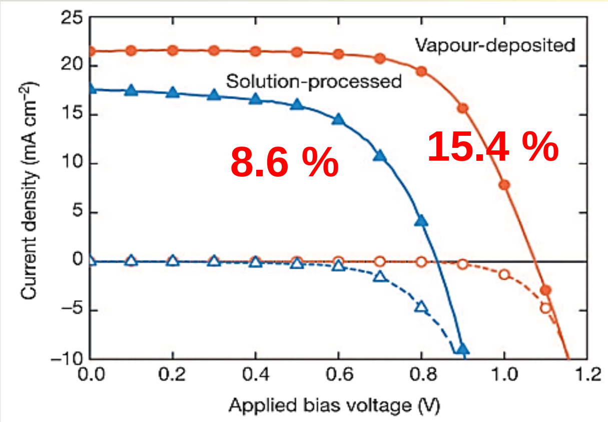
doi:10.1038/nature12509

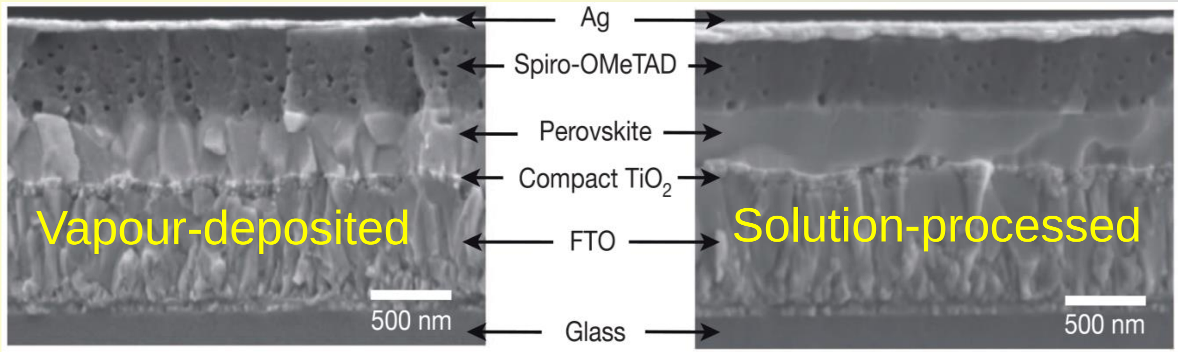
Efficient planar heterojunction perovskite solar cells by vapour deposition

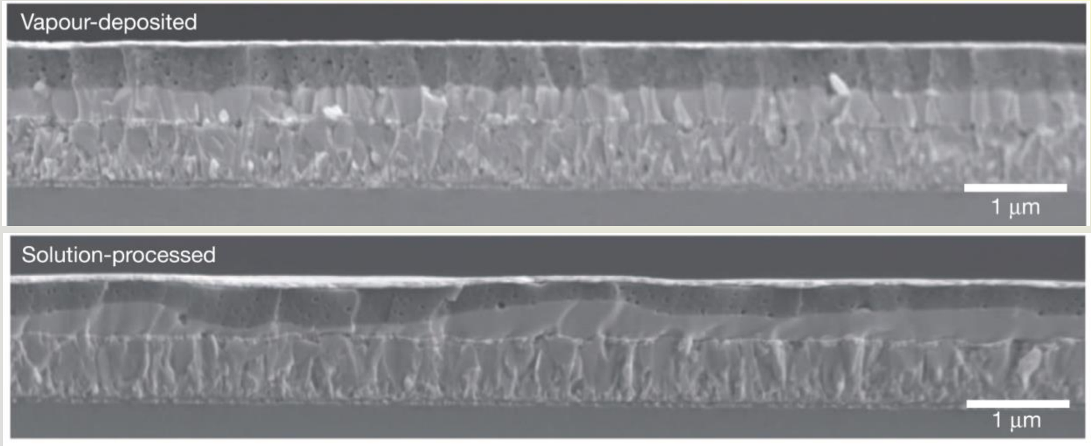
Mingzhen Liu[1], Michael B. Johnston[1] & Henry J. Snaith[1]

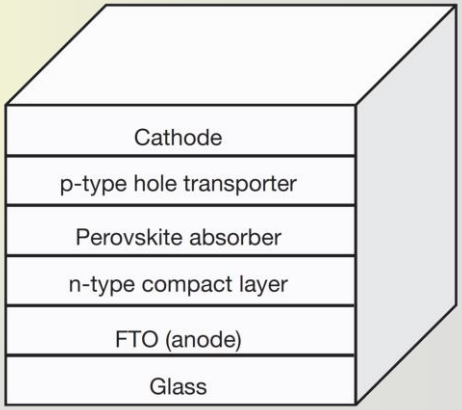


Henry Snaith









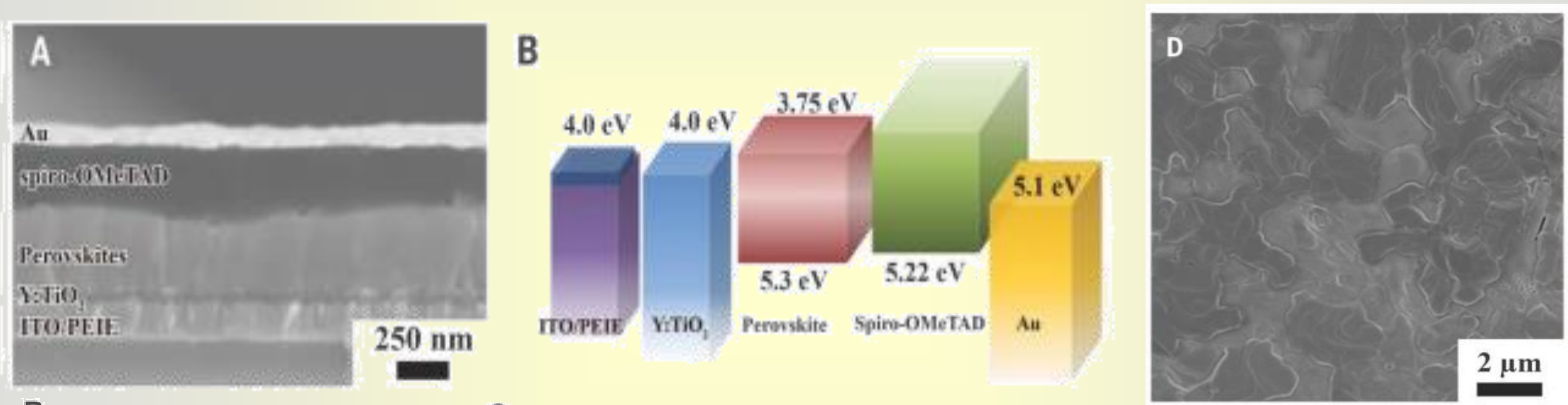
Nature **2013**, *501*, 395-398.

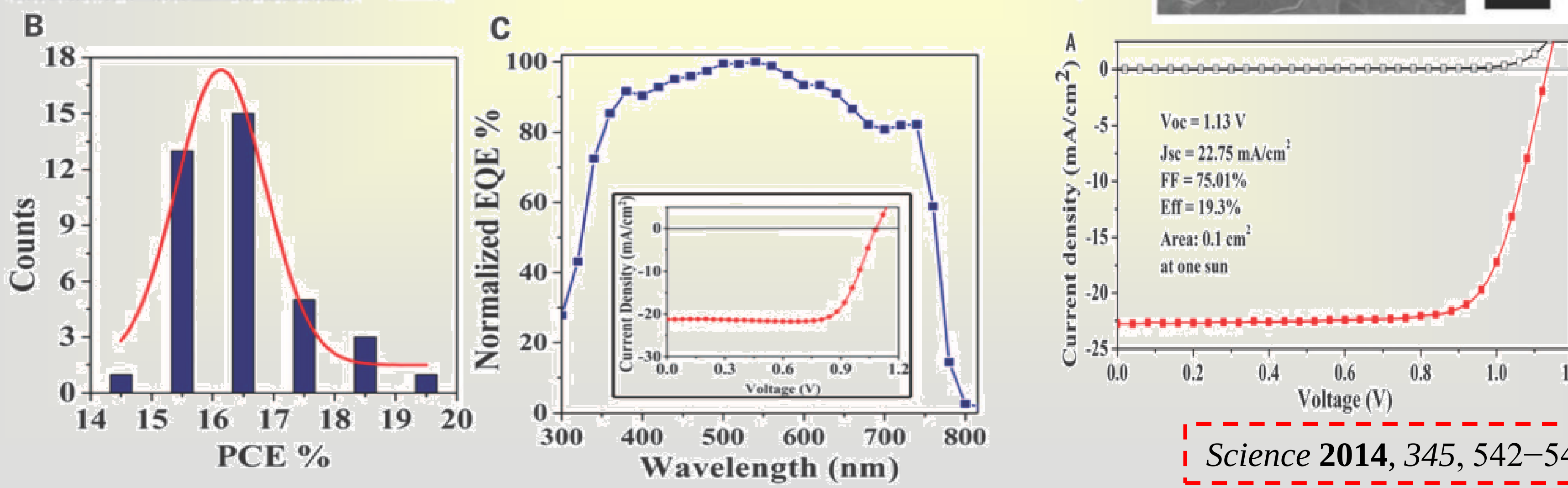
PHJ PSC Reaching PCE 19.3 % in 2014

Interface engineering of highly efficient perovskite solar cells

Huanping Zhou,[1,2*] Qi Chen,[1,2*] Gang Li,[1] Song Luo,[1,2] Tze-bing Song,[1,2] Hsin-Sheng Duan,[1,2] Ziruo Hong,[1] Jingbi You,[1] Yongsheng Liu,[1,2] Yang Yang[1,2]†

Yang Yang
UCLA





Science **2014**, *345*, 542−546.

World Record Attaining PCE 20.2 % in 2015

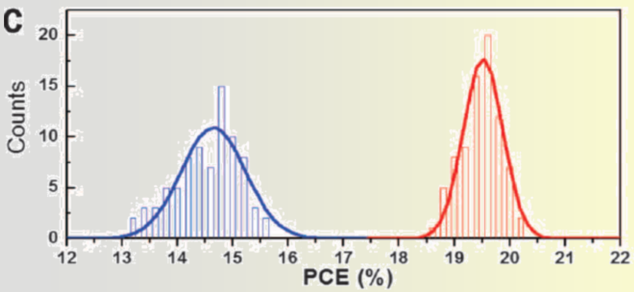
SOLAR CELLS

High-performance photovoltaic perovskite layers fabricated through intramolecular exchange

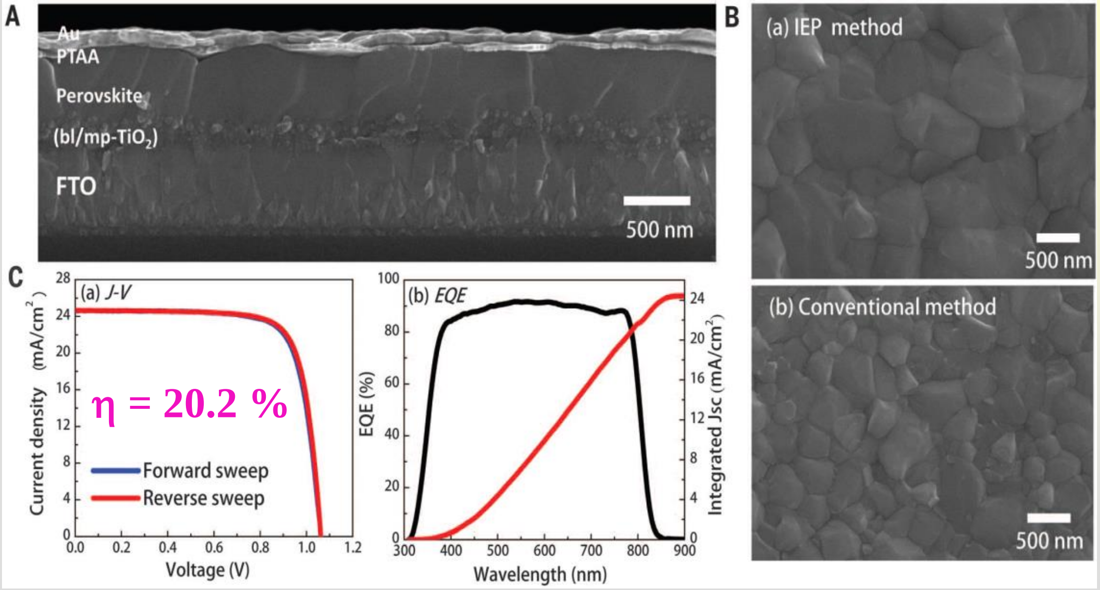
Woon Seok Yang,[1*] Jun Hong Noh,[1*] Nam Joong Jeon,[1] Young Chan Kim,[1] Seungchan Ryu,[1] Jangwon Seo,[1] Sang Il Seok[1,2†]

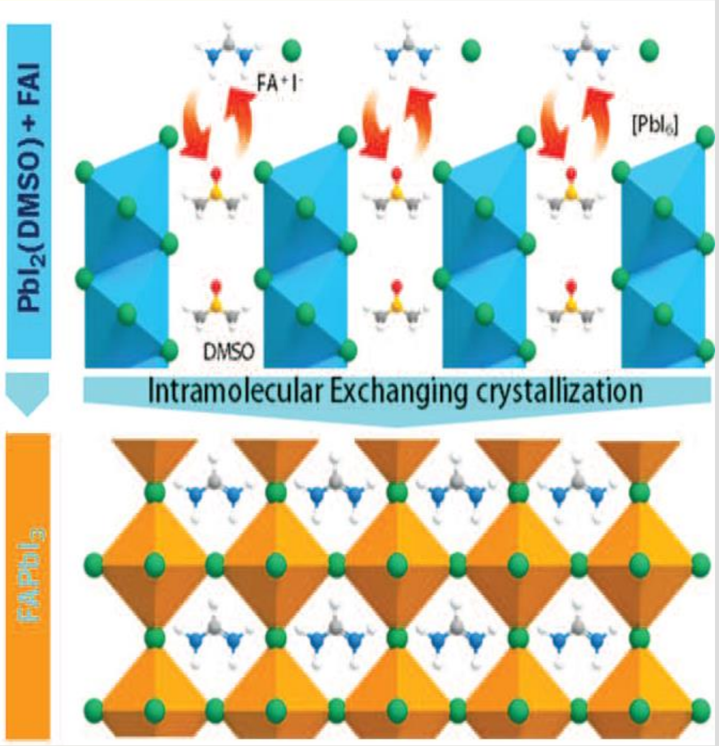
Sang Il Seok
UNIST, Korea

World record
PCE = 25.8%



$$PbI_2\text{-DMSO} + FAI \rightarrow PbI_2\text{-FAI} + DMSO \uparrow$$



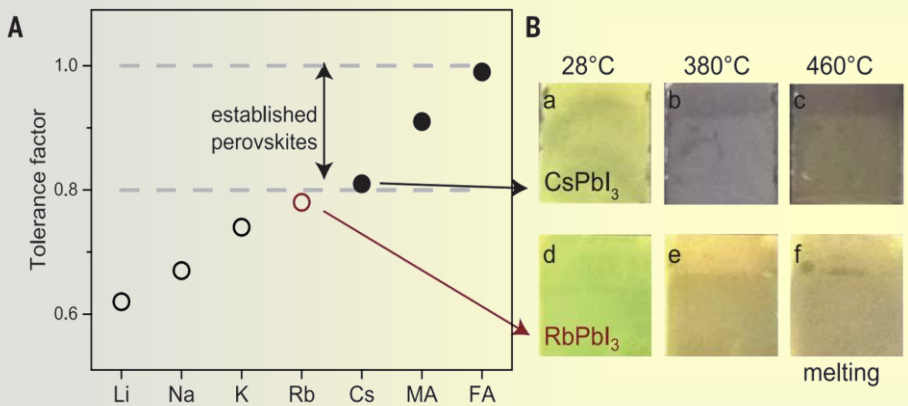


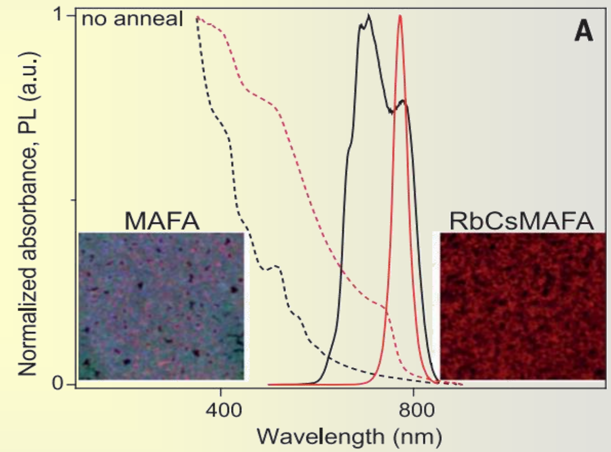
Science **2015**, *348*, 1234-1237.

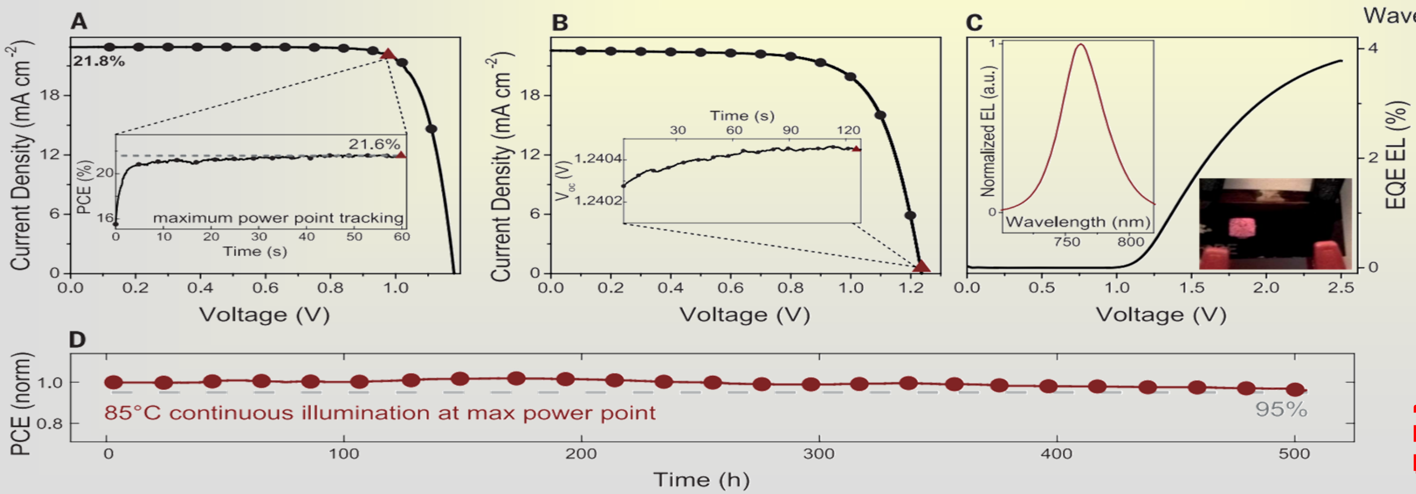
Multiple Cations for PCE 21.6 % in 2016

Incorporation of rubidium cations into perovskite solar cells improves photovoltaic performance

Michael Saliba,[1*†] Taisuke Matsui,[1,2†] Konrad Domanski,[1†] Ji-Youn Seo,[1] Amita Ummadisingu,[1] Shaik M. Zakeeruddin,[1] Juan-Pablo Correa-Baena,[3] Wolfgang R. Tress,[1] Antonio Abate,[1] Anders Hagfeldt,[3] Michael Grätzel[1*]







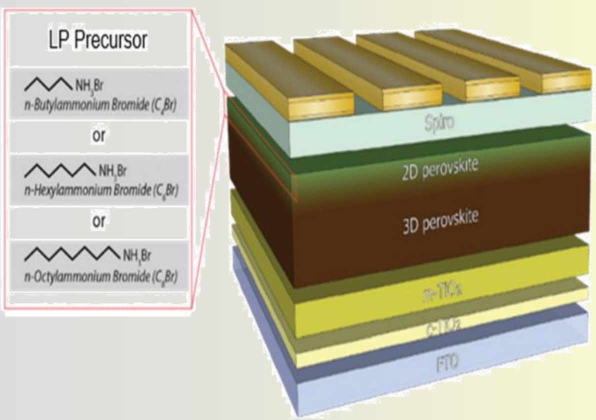
Michael Grätzel

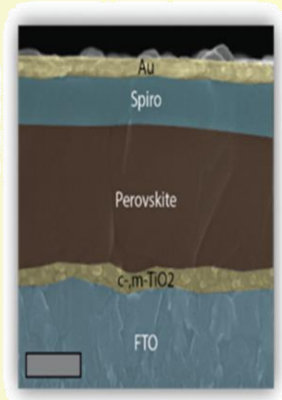
Science **2016**, *354*, 206-209.

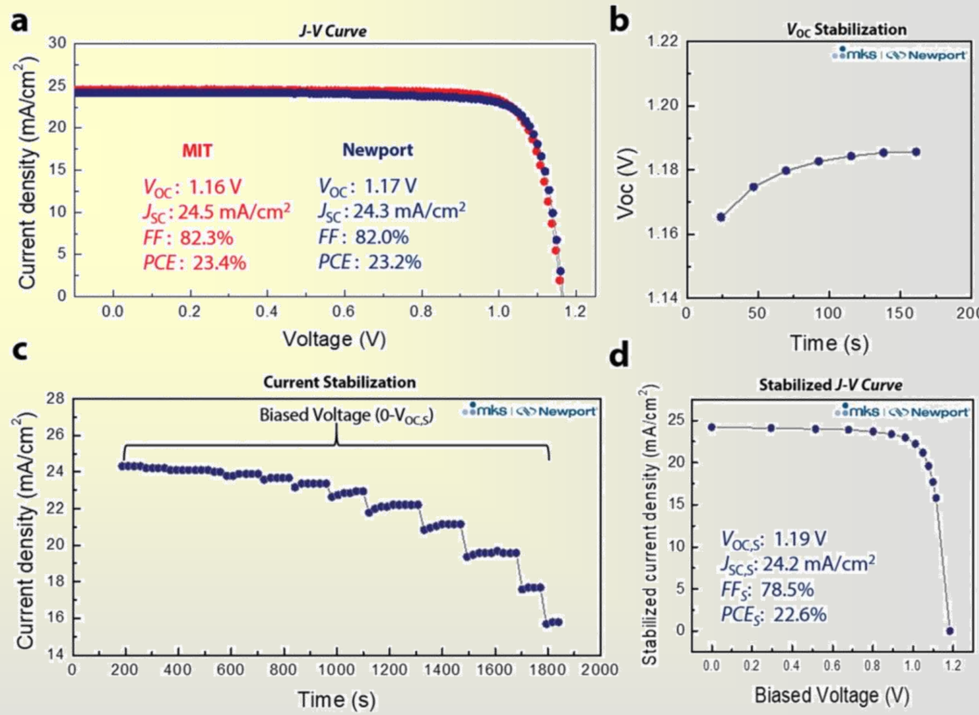
2D/3D Structure for PCE 23.4 % in 2019

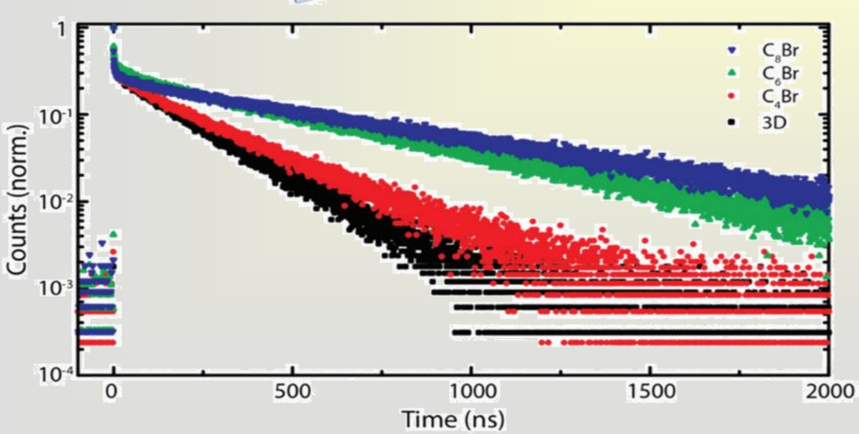
An interface stabilized perovskite solar cell with high stabilized efficiency and low voltage loss†

Jason J. Yoo,[a] Sarah Wieghold,[b] Melany C. Sponseller,[c] Matthew R. Chua,[c] Sophie N. Bertram,[a] Noor Titan Putri Hartono,[b] Jason S. Tresback,[d] Eric C. Hansen,[a] Juan-Pablo Correa-Baena,[b] Vladimir Bulović,[c] Tonio Buonassisi,[b] Seong Sik Shin*[be] and Moungi G. Bawendi*[a]









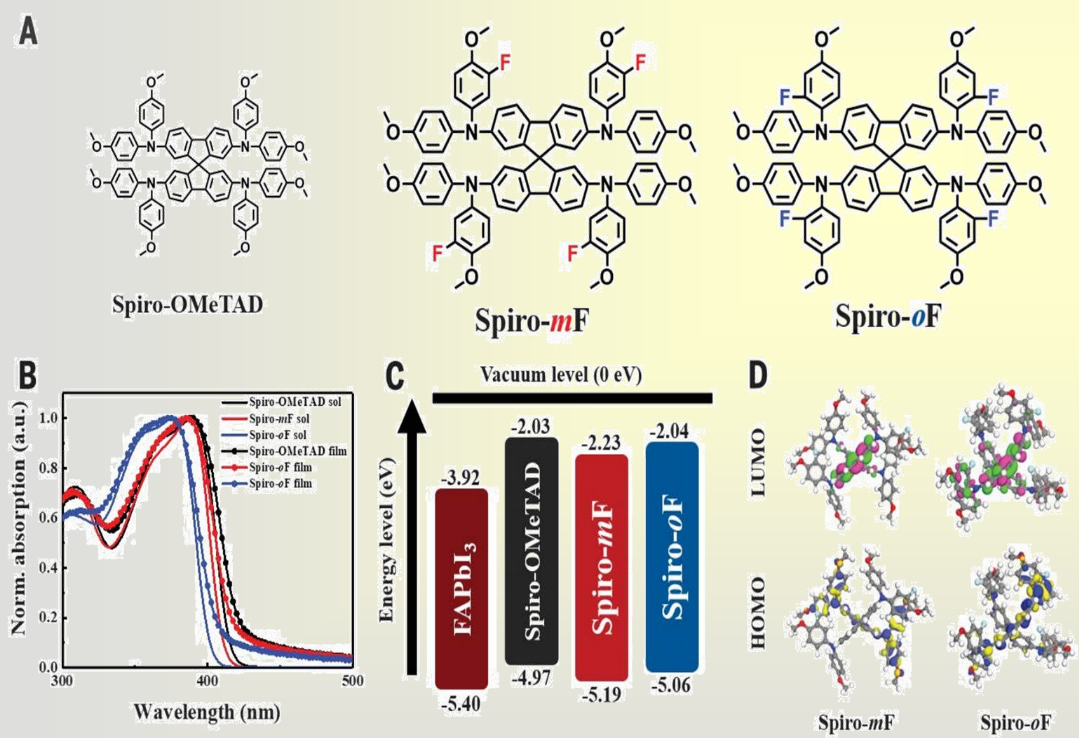
Energy Environ. Sci., **2019**, *12*, 2192-2199.

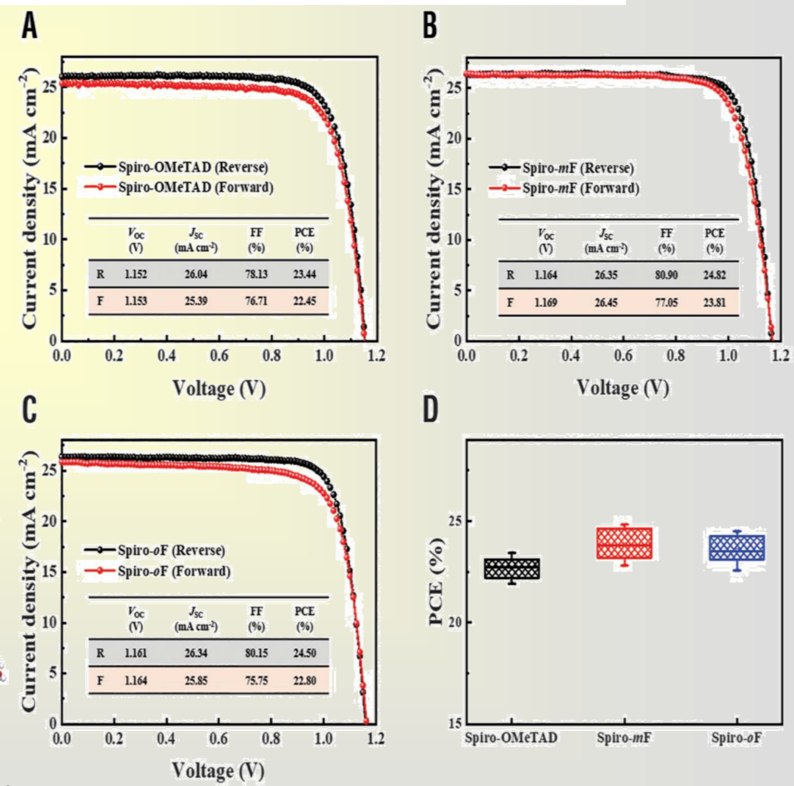
Flouro-Spiro HTM for PCE 24.8 % in 2020

SOLAR CELLS

Stable perovskite solar cells with efficiency exceeding 24.8% and 0.3-V voltage loss

Mingyu Jeong[1]*, In Woo Choi[2,3]*, Eun Min Go[4]*, Yongjoon Cho[1], Minjin Kim[2], Byongkyu Lee[1], Seonghun Jeong[1], Yimhyun Jo[2], Hye Won Choi[2], Jiyun Lee[4], Jin-Hyuk Bae[3], Sang Kyu Kwak[4]†, Dong Suk Kim[2]†, Changduk Yang[1]†





Chemical structures of Spiro-OMeTAD and its fluorinated analogs Spiro-mF and Spiro-oF. (B) UV-Vis absorption spectra of Spiro-OMeTAD, Spiro-mF, and Spiro-oF in dilute chlorobenzene solution (sol) and as thin films. (C) Molecular energy level alignments. (D) Electron density distributions of HOMO and LUMO for Spiro-mF and Spiro-oF.

Science **2020**, *369*, 1615–1620.

Anion-engineering for $FAPbI_3$ PSC with PCE 25.6%

Pseudo-halide anion engineering for α-$FAPbI_3$ perovskite solar cells

Nature **2021**, *592*, 381–385.

https://doi.org/10.1038/s41586-021-03406-5

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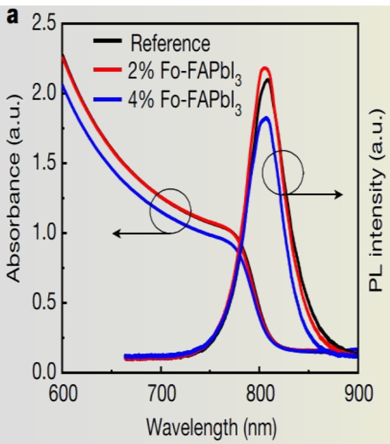
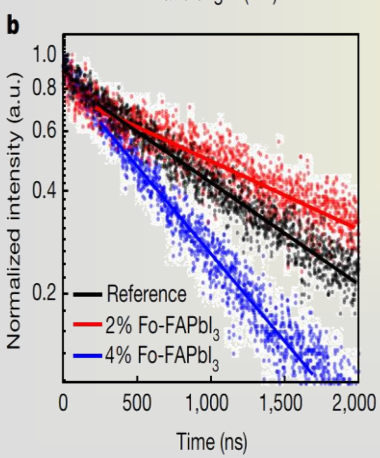
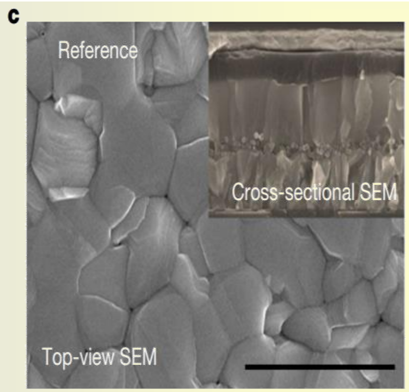
Published online: 5 April 2021

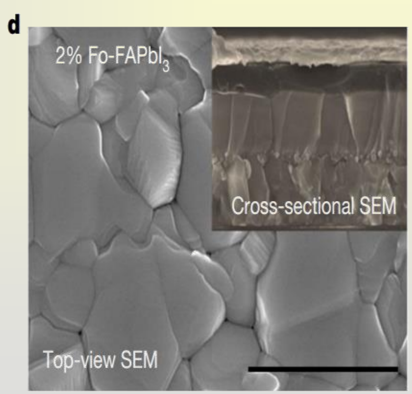
Jaeki Jeong[1,2,3,11], Minjin Kim[4,11], Jongdeuk Seo[1,11], Haizhou Lu[2,3,11], Paramvir Ahlawat[5], Aditya Mishra[6], Yingguo Yang[7], Michael A. Hope[6], Felix T. Eickemeyer[2], Maengsuk Kim[1], Yung Jin Yoon[1], In Woo Choi[4], Barbara Primera Darwich[8], Seung Ju Choi[4], Yimhyun Jo[4], Jun Hee Lee[1], Bright Walker[9], Shaik M. Zakeeruddin[2], Lyndon Emsley[6], Ursula Rothlisberger[5], Anders Hagfeldt[3,10]✉, Dong Suk Kim[4]✉, Michael Grätzel[2]✉ & Jin Young Kim[1]✉

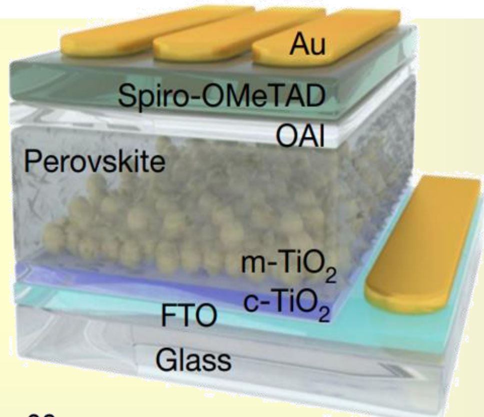
2% Formamidine Formate (FAHCOO) was added to the $FAPbI_3$ precursor solution to enhance the PCE to 25.6 %.



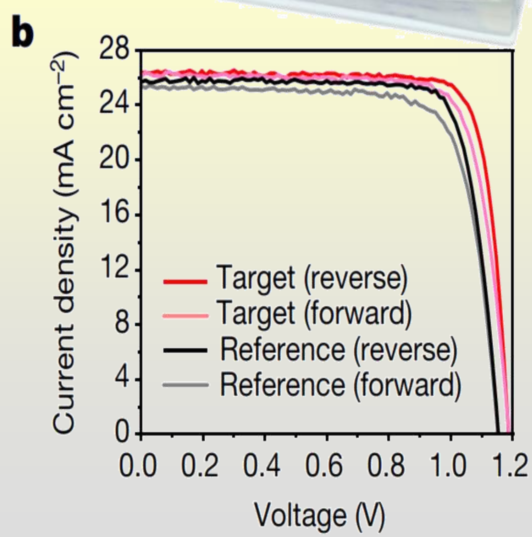


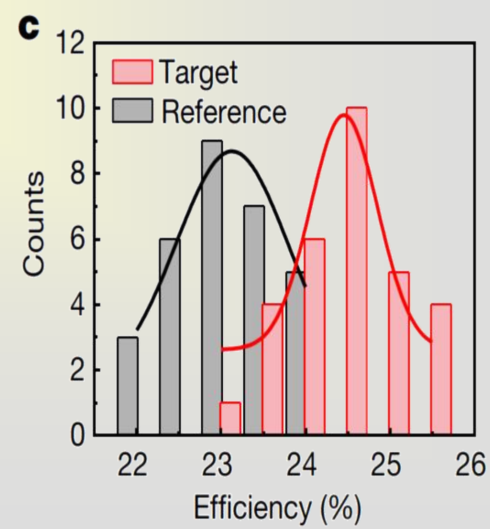




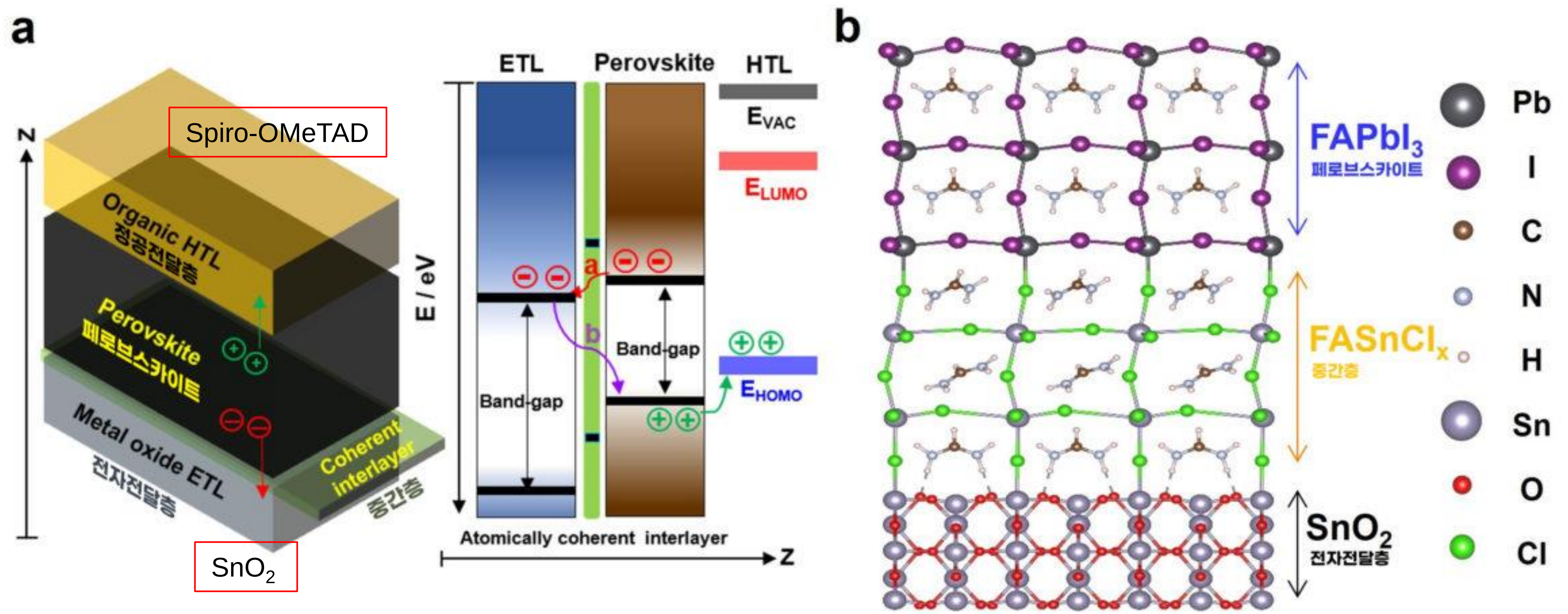


Michael Grätzel





Coherent Interlayer Engineering with PCE 25.8%



New world record of PCE 25.8% of $FAPbI_3$ PSC was achieved by forming a coherent interlayer $FASnCl_3$ between electron-transporting (SnO_2) and perovskite layers by coupling chlorine (Cl)-bonded SnO_2 with a Cl-containing perovskite precursor to reduce interfacial defects for retarded interfacial charge recombination.

NREL certified PCE = 25.7%

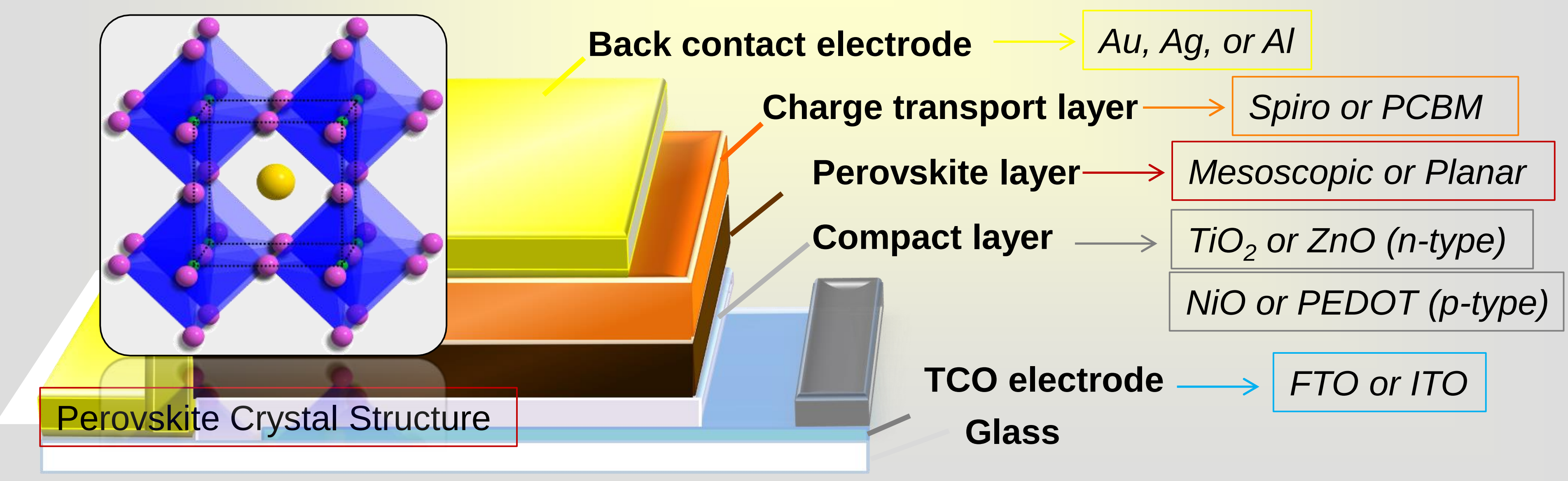
Sang Il Seok
UNIST, Korea

Nature **2021**, *598*, 444–450.

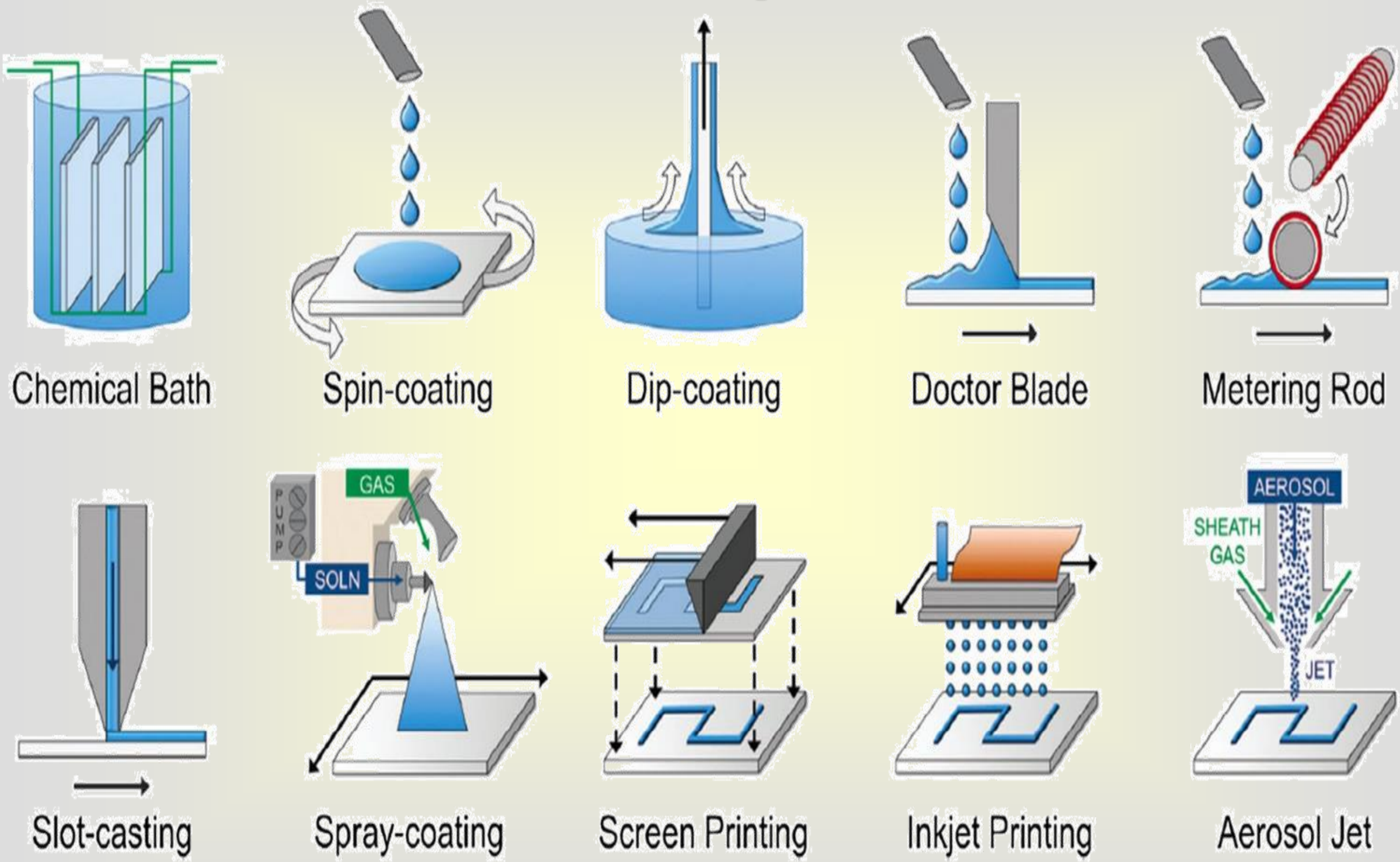
Device Configuration and Fabrication of PSC

Perovskite Solar Cells

1. Device Configurations: n-type vs. p-type and mesoscopic heterojunction (MHJ) vs. planar heterojunction (PHJ).
2. Electrode Configurations: carbon-based and HTM-free.
3. Morphology Control: anti-solvent, delayed annealing, solvent annealing and effect of additives.
4. Device and Film Characterizations: IV/IPCE, EIS, EA/EPL Spectra, PIA/TAS, Femtosecond Spectroscopy.

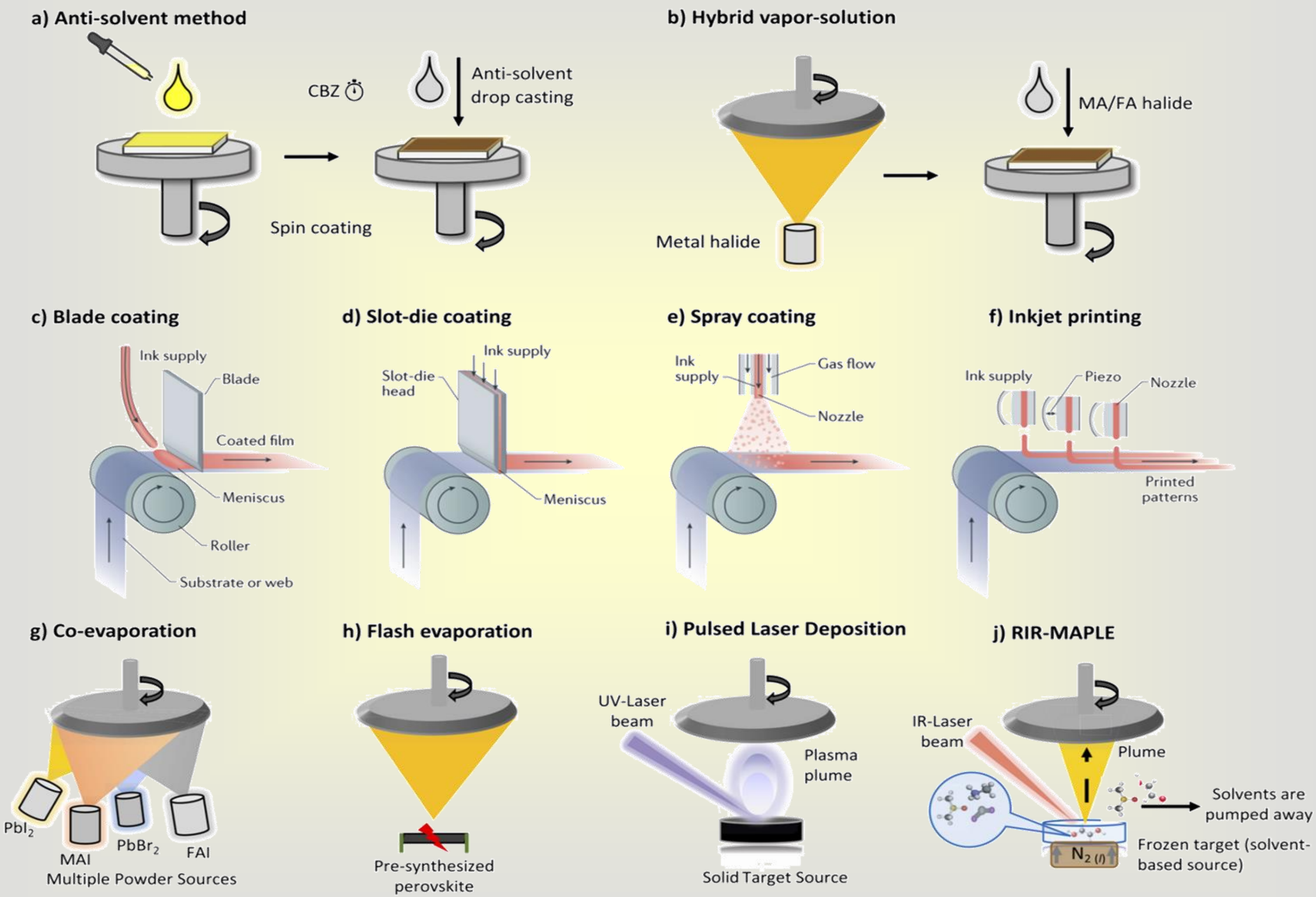


Various Solution Deposition Methods



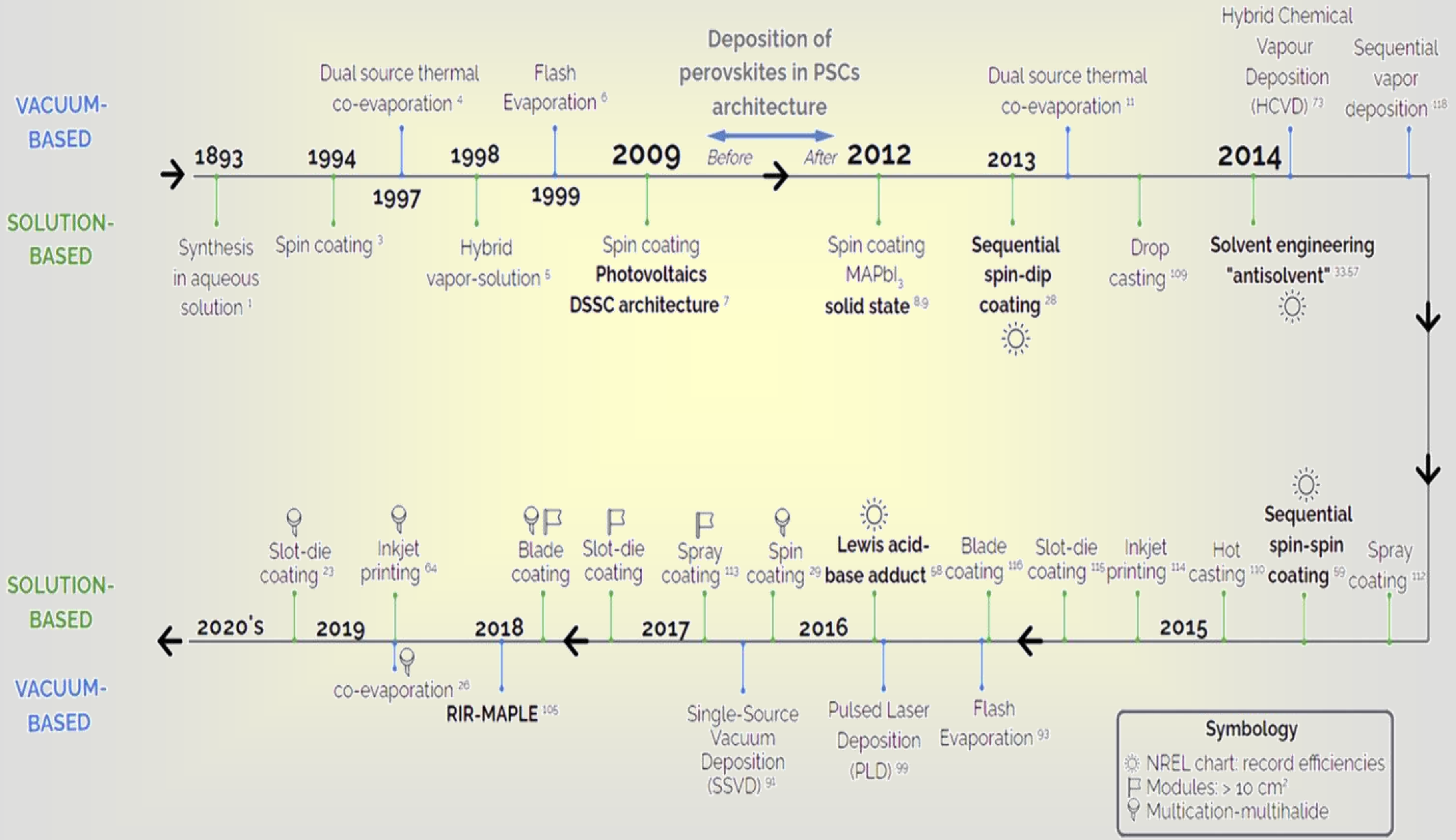
Chem. Soc. Rev. **2011**, *40*, 5406–5441.

Preparation of Halide Perovskite Thin Films



APL Mater. 8, 2020, 110903.

Timeline of thin-film fabrication methods for inorganic and hybrid halide perovskites



APL Mater. 8, 2020, 110903.

Control of Morphology for Perovskite Nanocrystals

Solvent Engineering to Attain PCE 16.2 %

nature materials

2014, *13*, 897-903

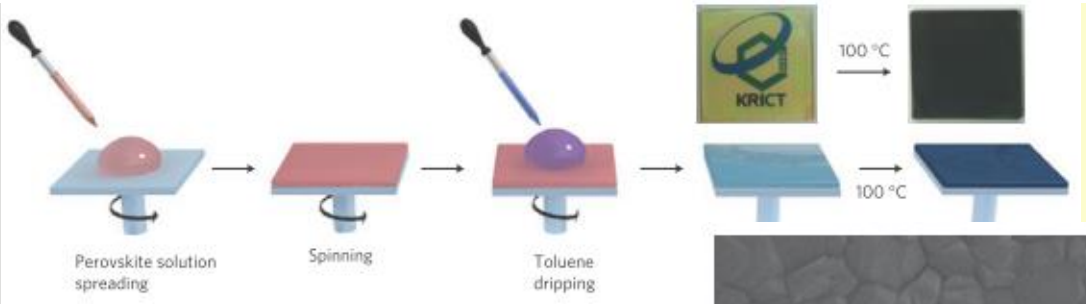
ARTICLES

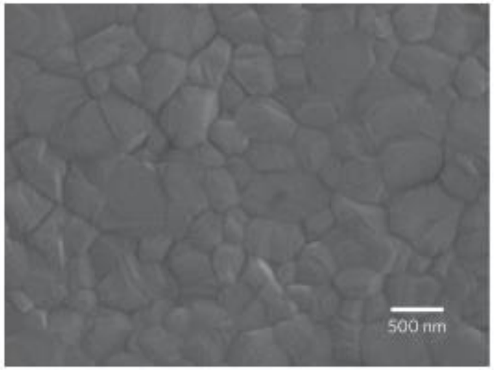
PUBLISHED ONLINE: 6 JULY 2014 | DOI: 10.1038/NMAT4014

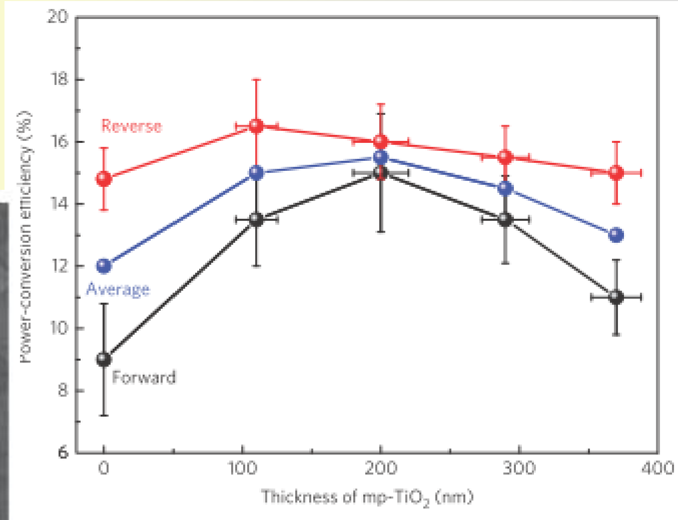
Solvent engineering for high-performance inorganic–organic hybrid perovskite solar cells

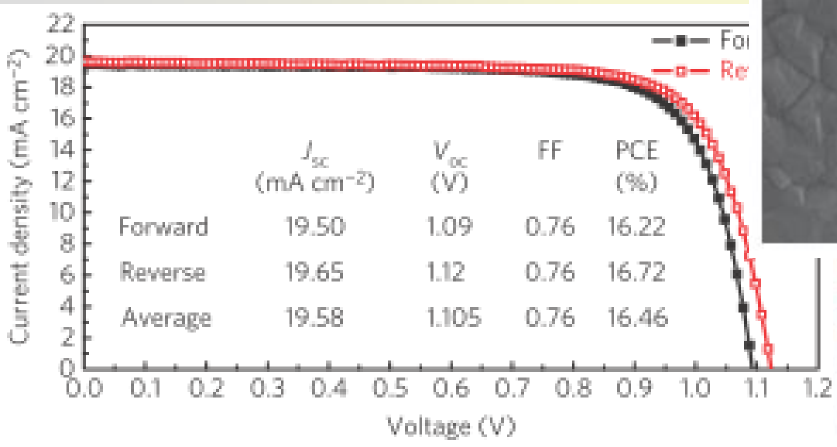
Nam Joong Jeon[1†], Jun Hong Noh[1†], Young Chan Kim[1], Woon Seok Yang[1], Seungchan Ryu[1] and Sang Il Seok[1,2*]

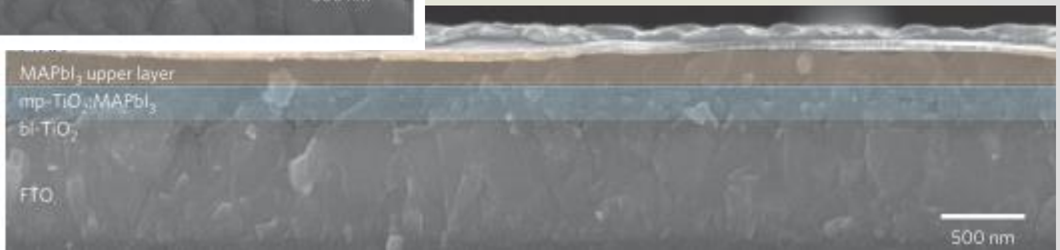
Sang Il Seok



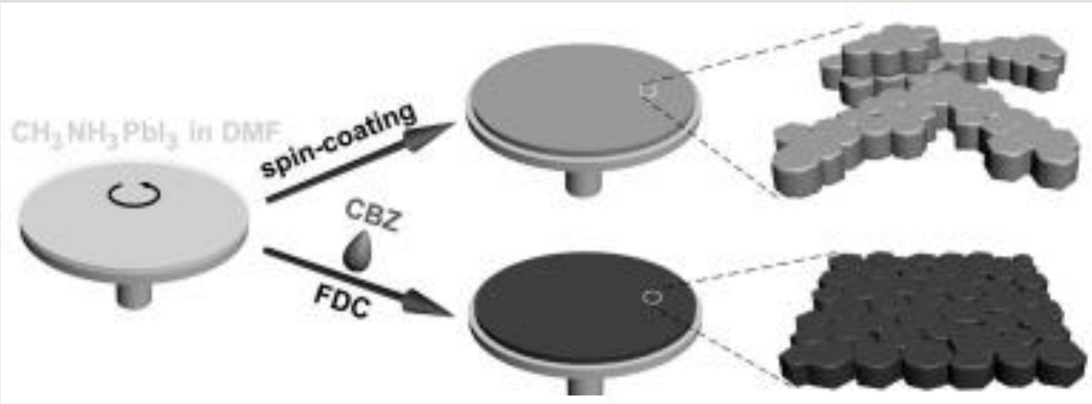








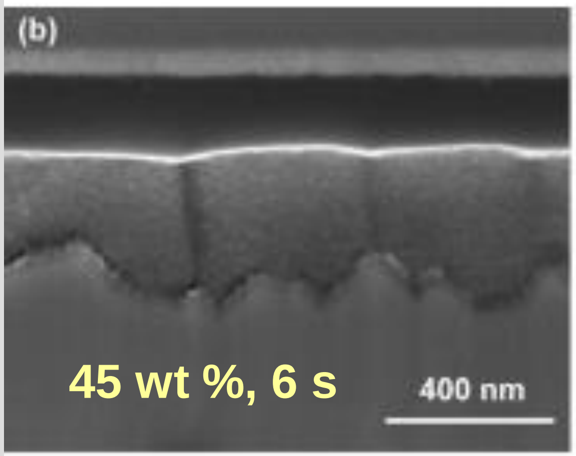
Solvent–Assisted Crystallization

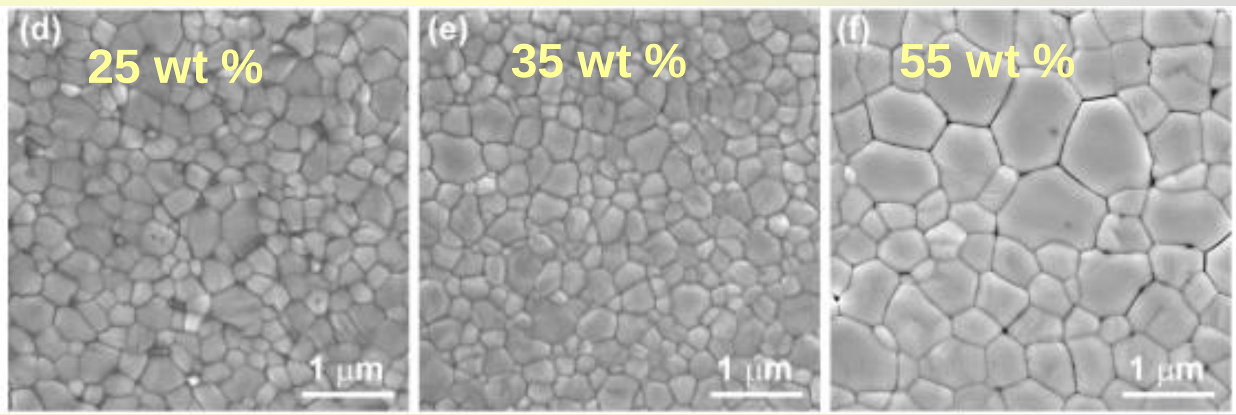


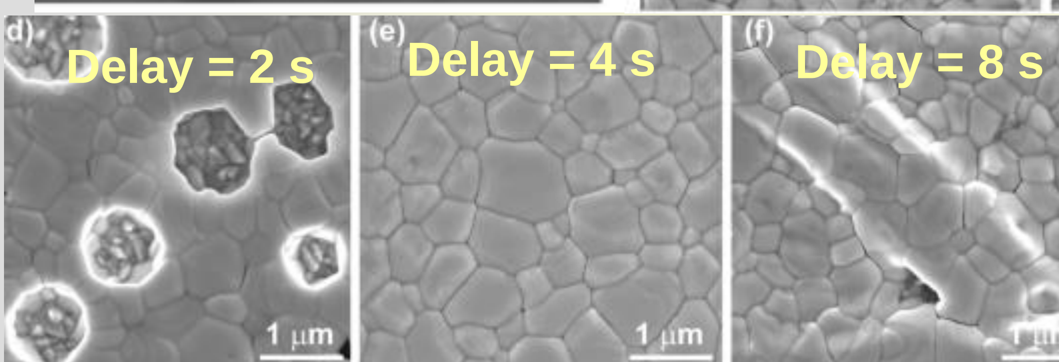
Yi-Bing Cheng

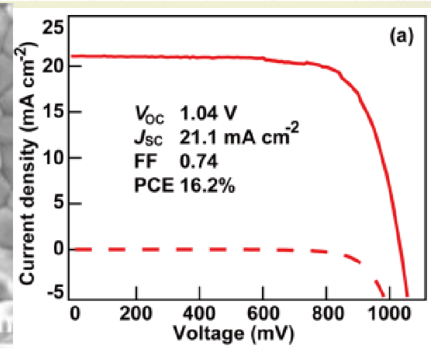
Leone Spiccia

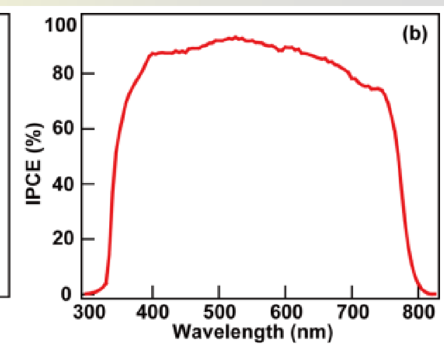
Angew. Chem. Int. Ed. **2014**, *53*, 9898-9903.





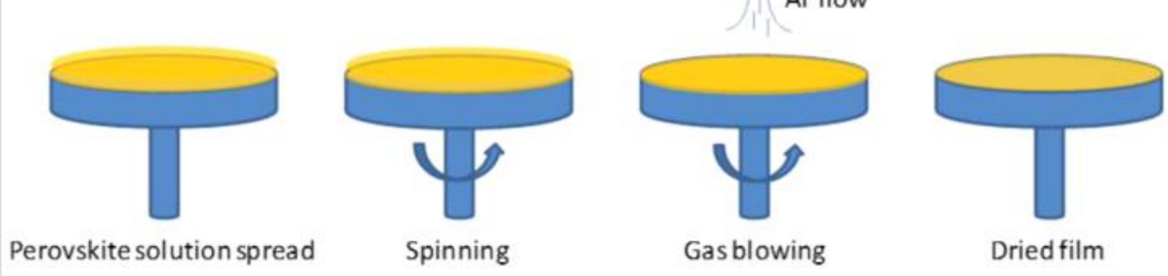






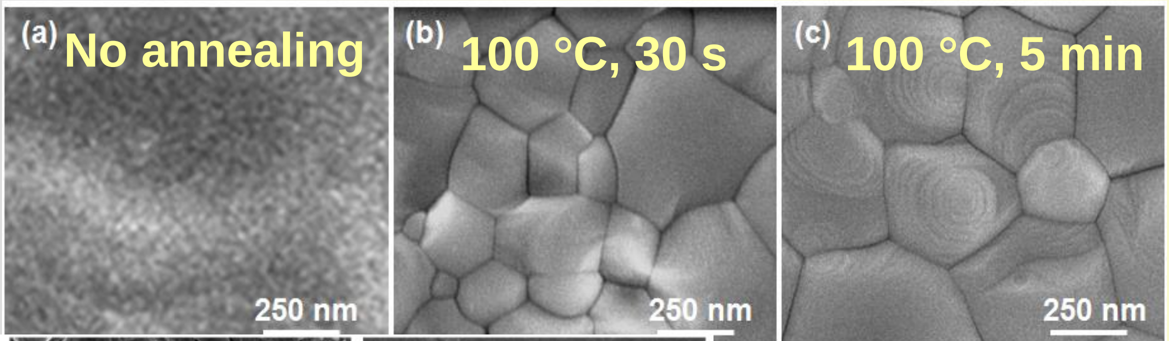
Gas–Assisted Crystallization

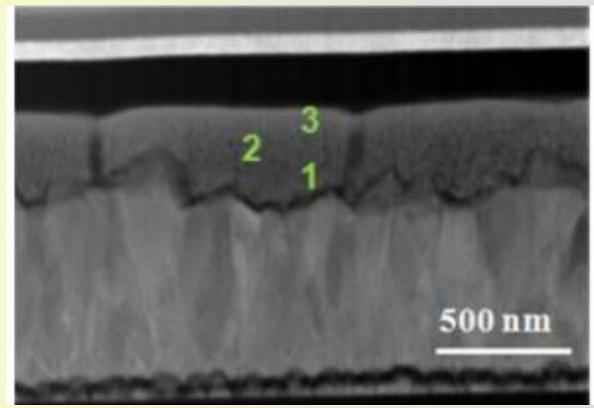
Nano Energy **2014**, *10*, 10-18.

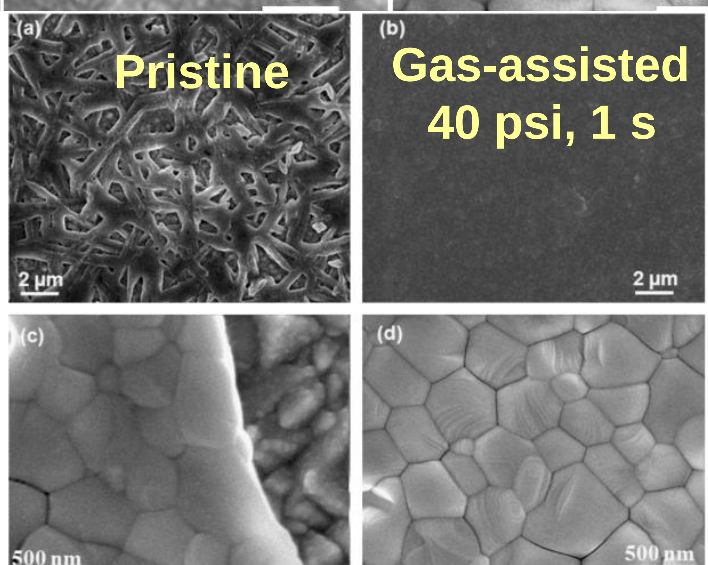


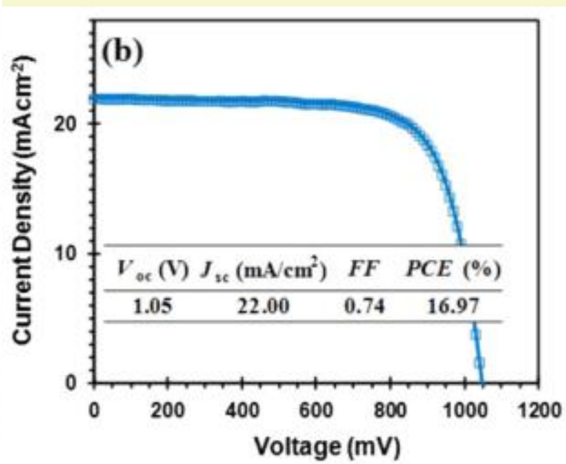
Yi-Bing Cheng

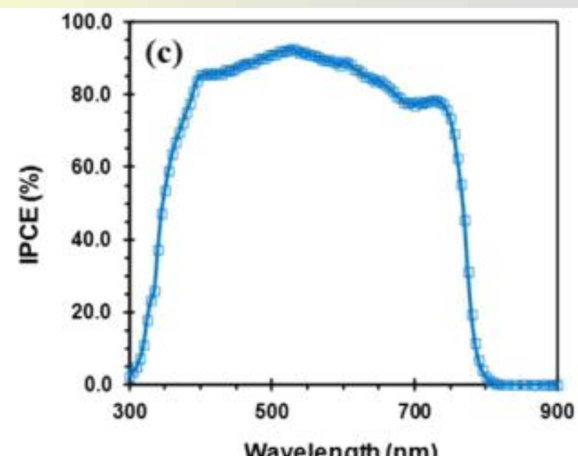
Leone Spiccia



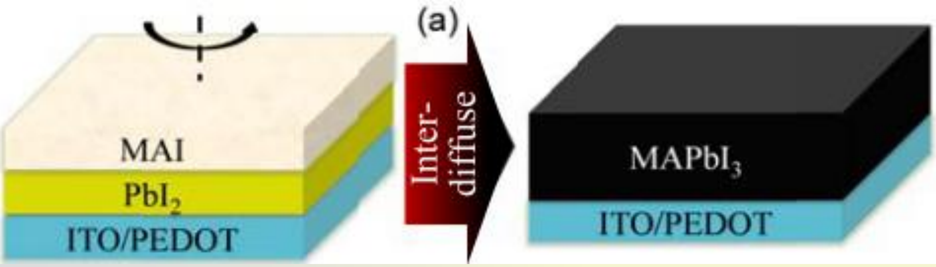




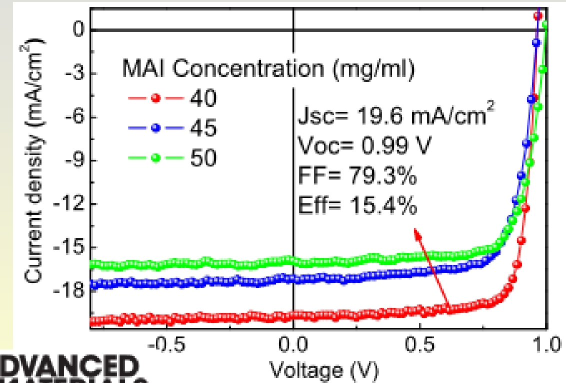


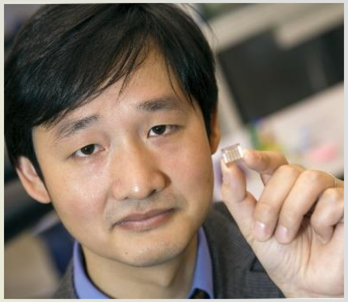


Interdiffusion and Solvent Annealing



Energy Environ. Sci. **2014**, *7*, 2619−2623.





Jinsong Huang

Materials Views
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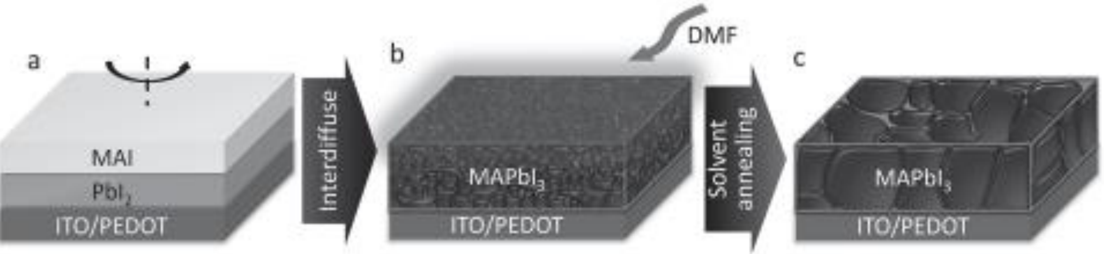
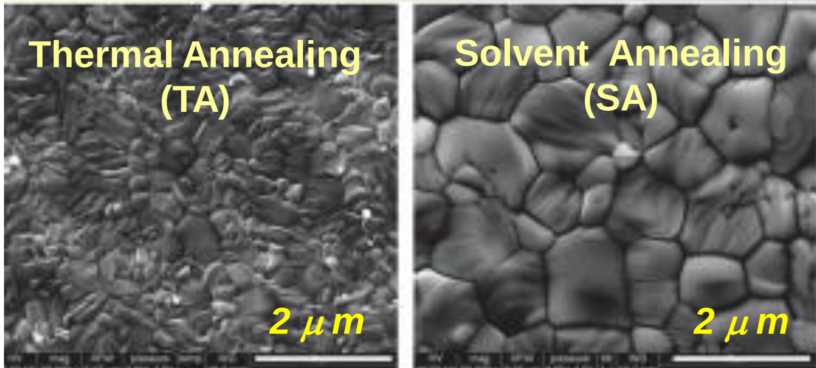
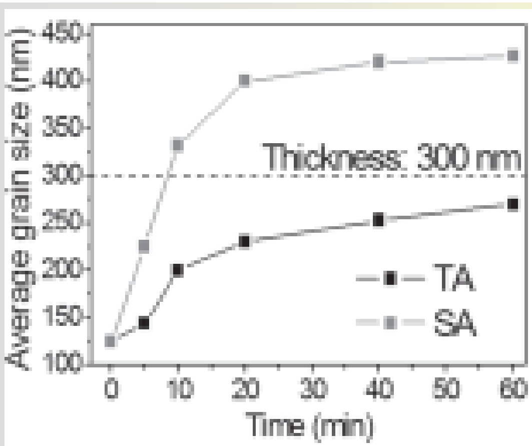
www.advmat.de

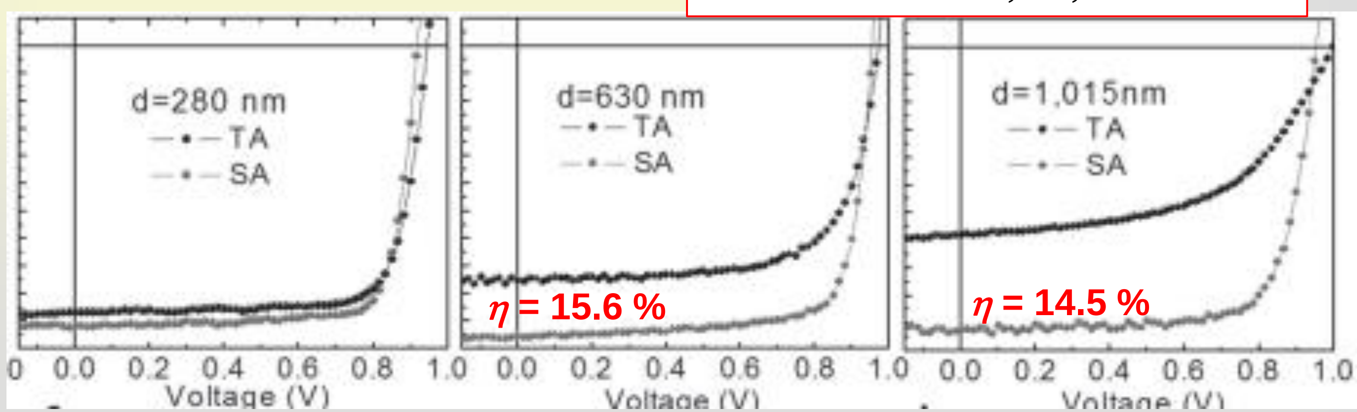
Solvent Annealing of Perovskite-Induced Crystal Growth for Photovoltaic-Device Efficiency Enhancement



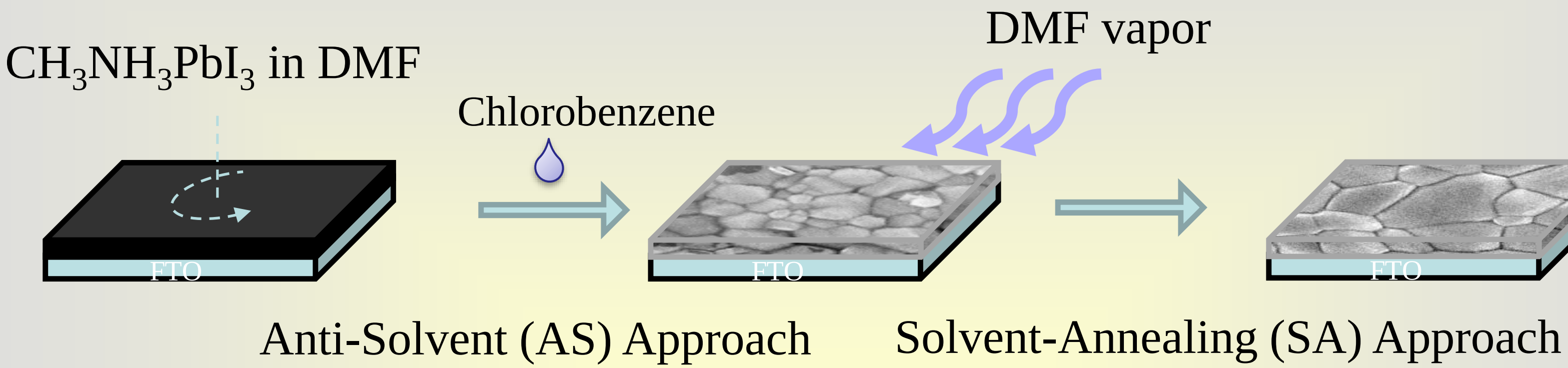


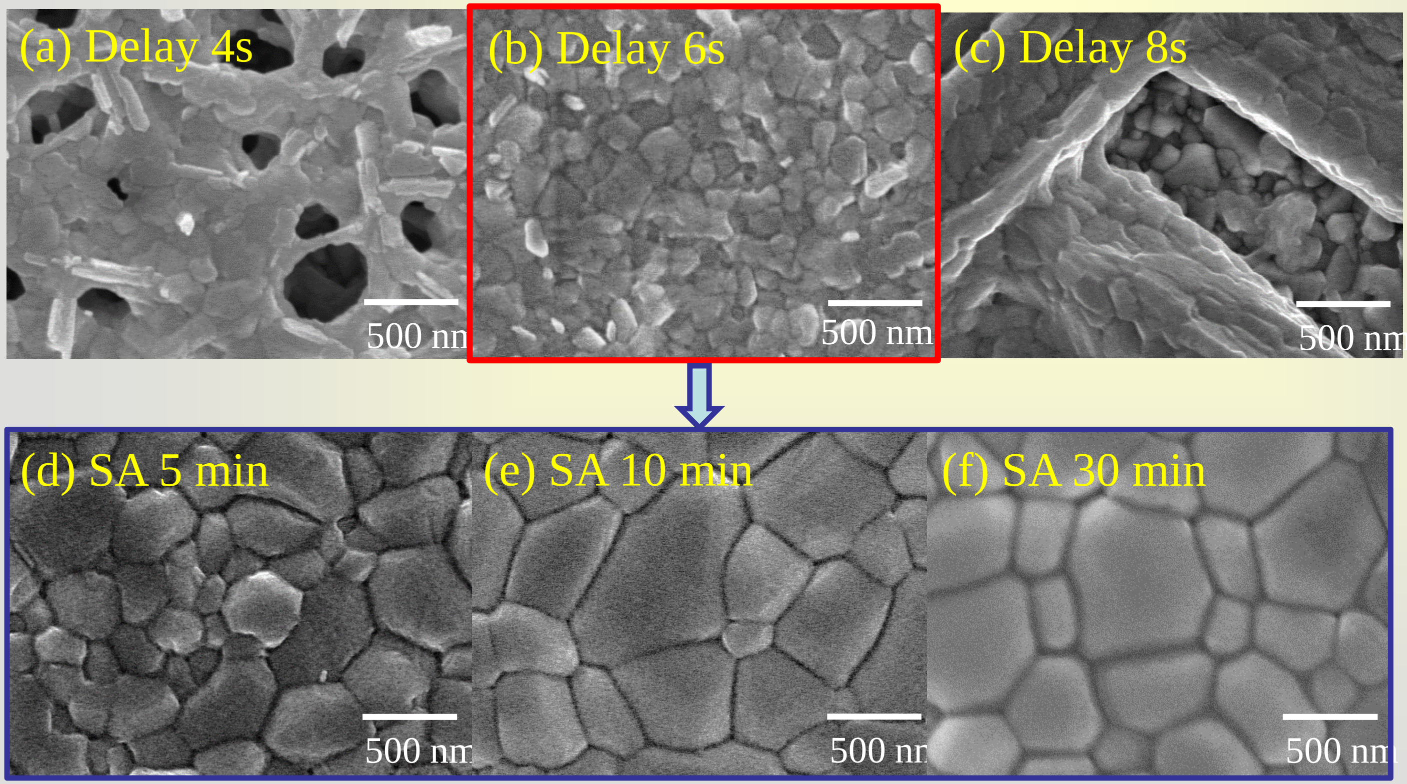
Adv. Mater. **2014**, *26*, 6503-6509.





A Sequential Anti-solvent and Solvent Annealing

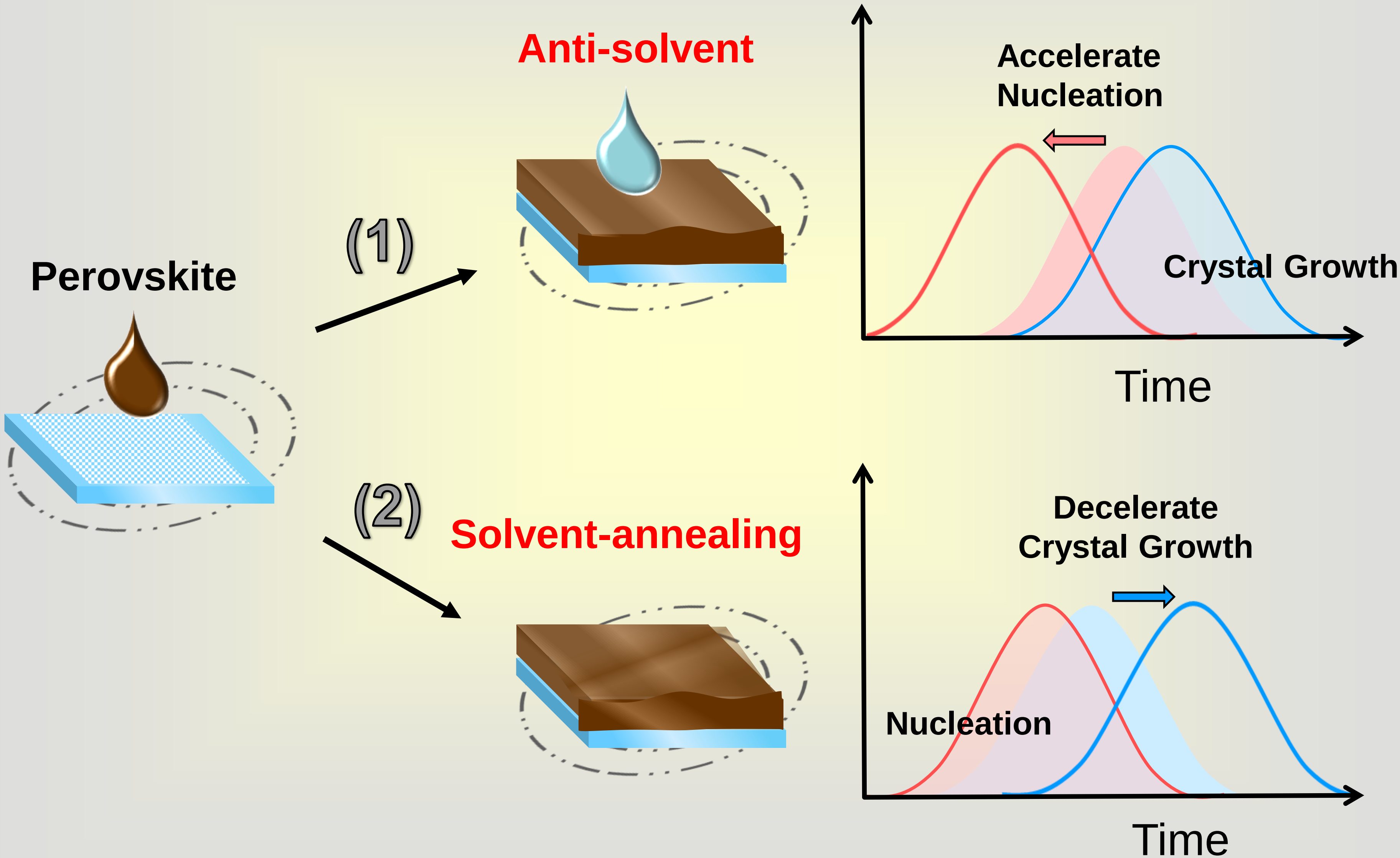




AS treatment in varied delay periods

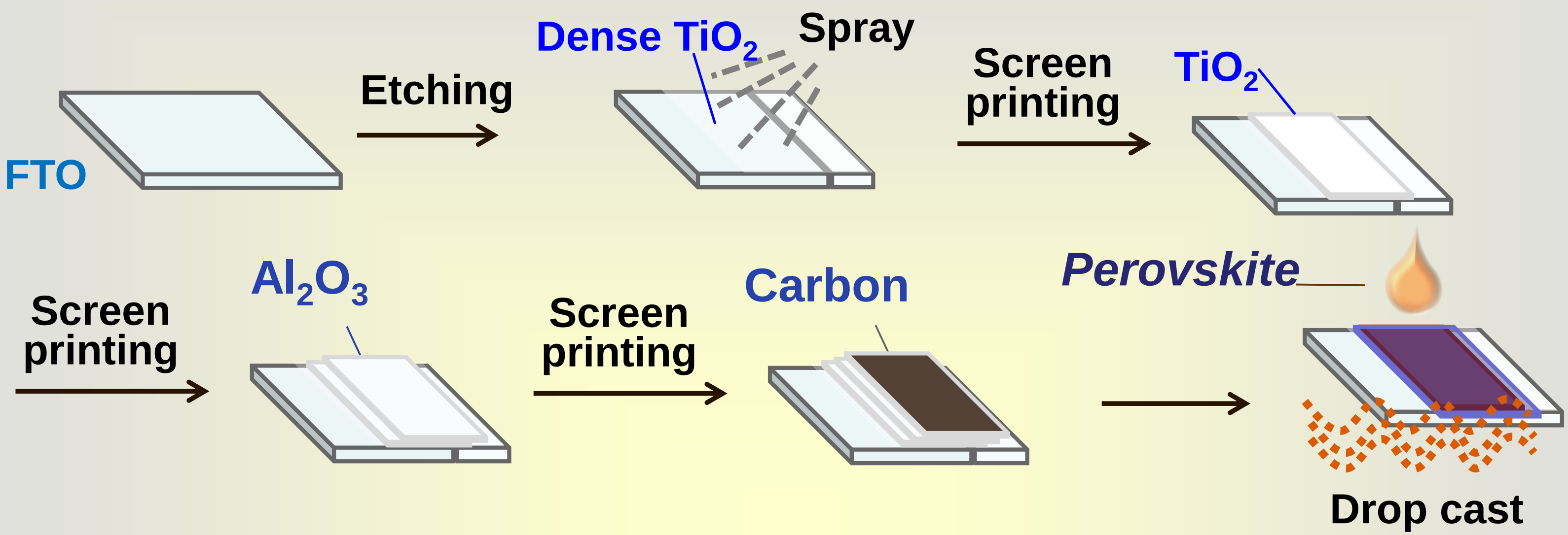
AS (6s) followed by SA in varied delay periods

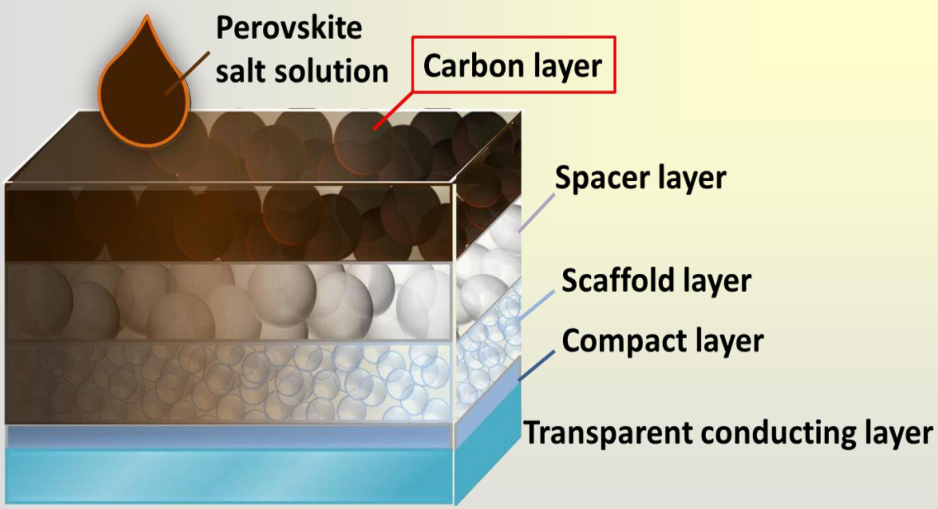
Nucleation and Crystal Growth



Stability Issue: Carbon-based HTM-free Perovskite Solar Cells

Fabrication of Carbon-based PSC

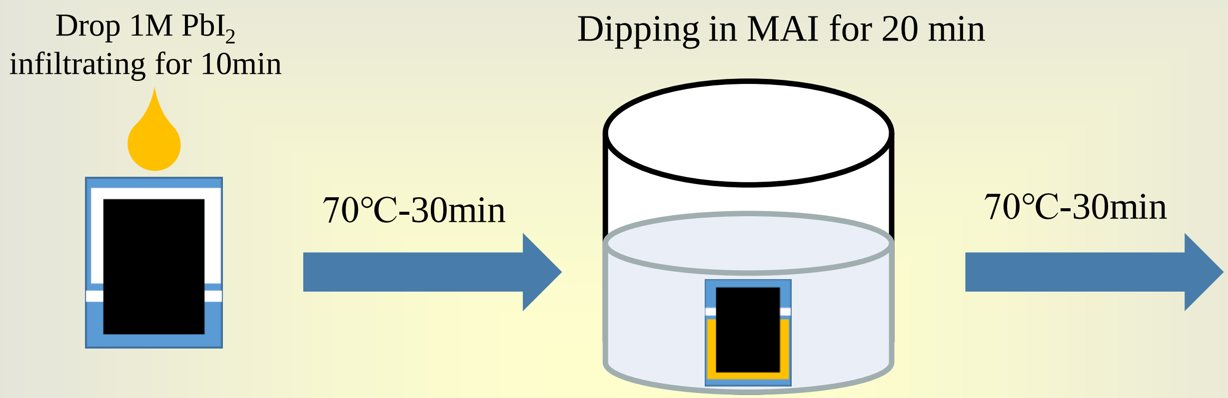




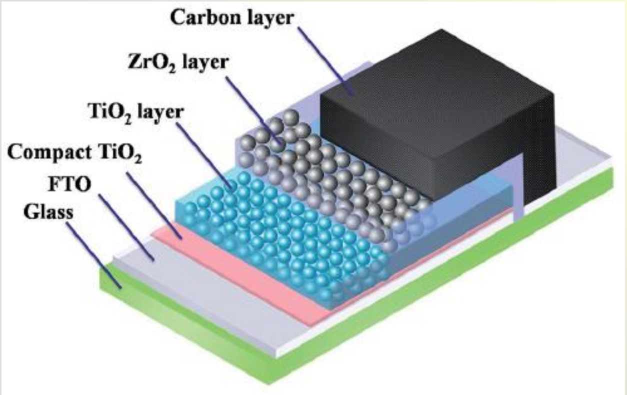
(1) Screen-printing and solution processing suitable for large-scale production.
(2) Using mesoporous carbon layer to replace Au electrode for low-cost production.
(3) No organic charge collection layers for excellent long-term stability.
(4) No liquid electrolyte for feasible device encapsulation.

Carbon-based PSC with a Multi-layer Configuration

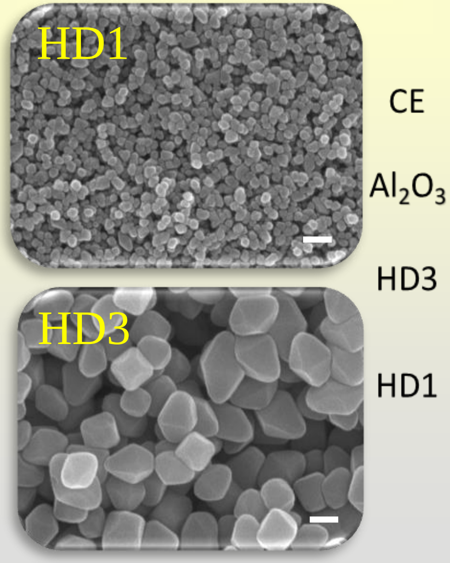
A sequential (two-step) approach



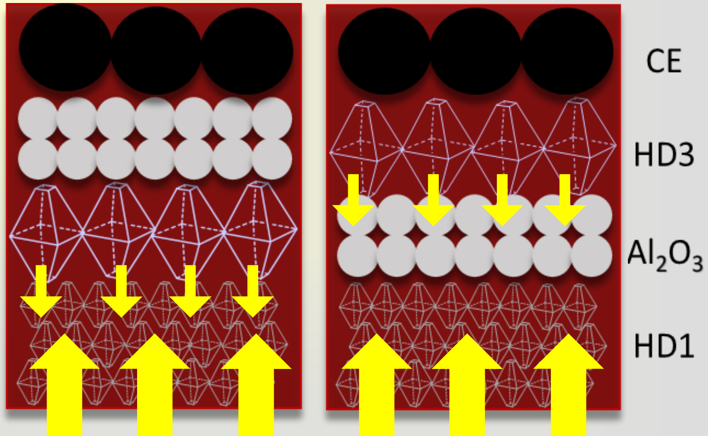
Han and co-workers, *Journal of Materials Chemistry A* **2014,** *2*, 17115-17121.



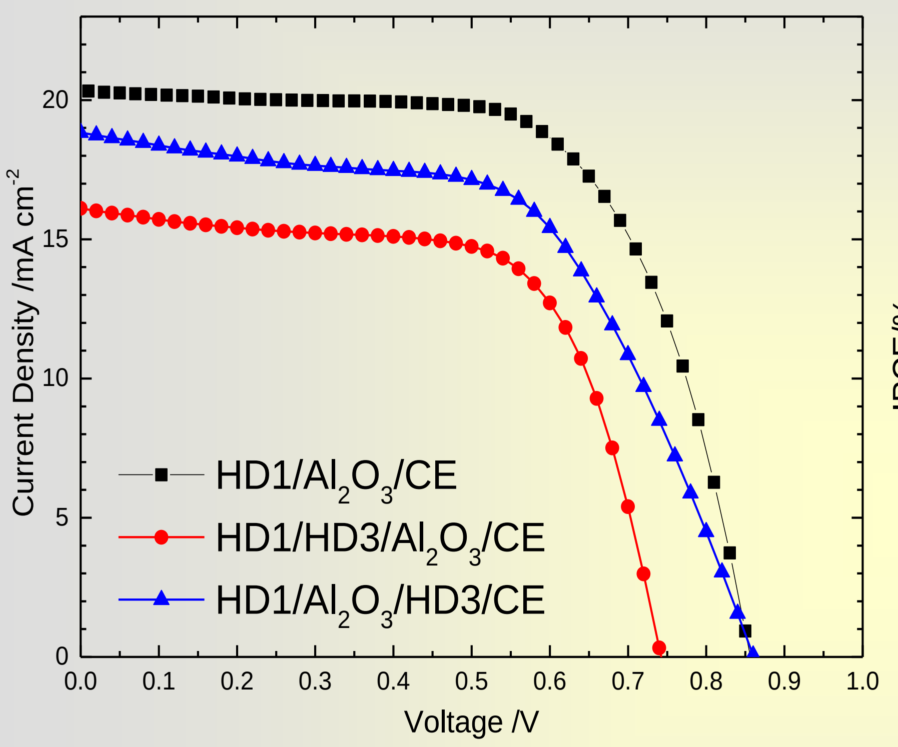
J_{SC} (mA cm^{-2})	V_{OC} (mV)	FF	η (%)
17.5	928	0.70	11.4

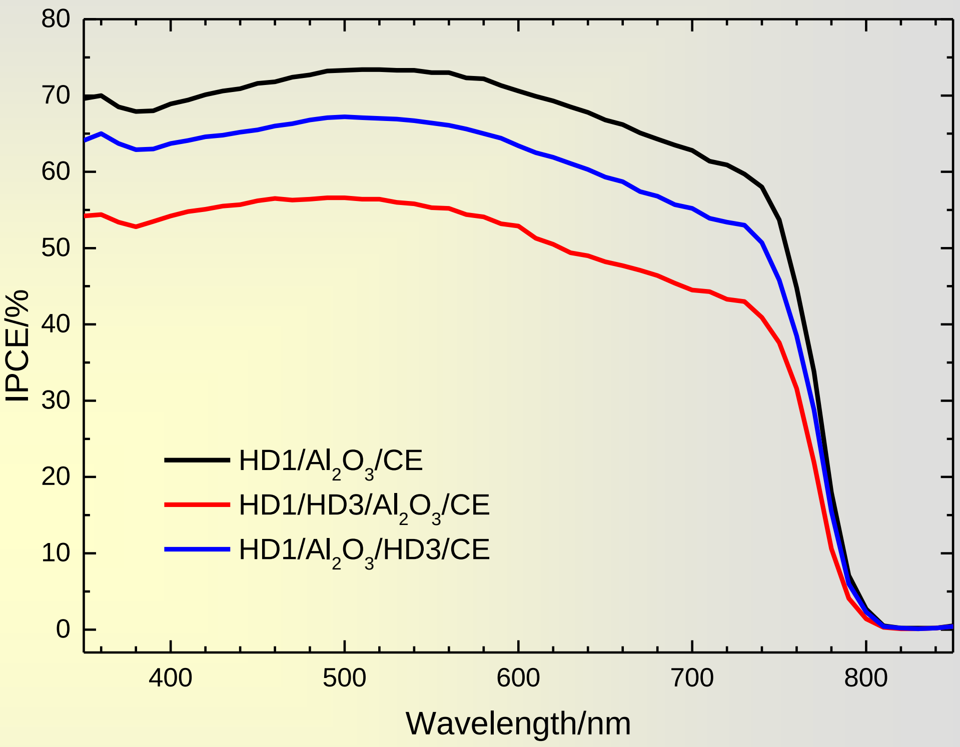


Our approach



Our Best Results with a HD1/Al_2O_3/CE Configuration

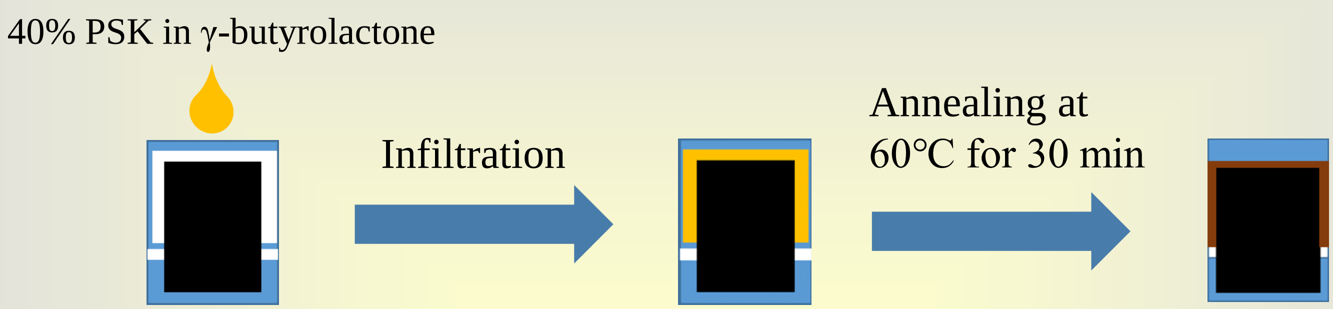


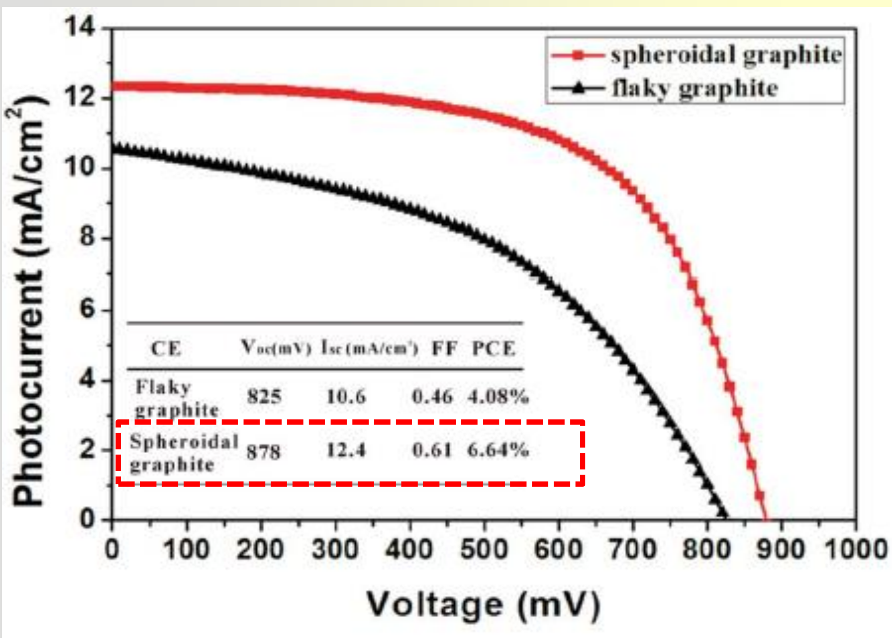


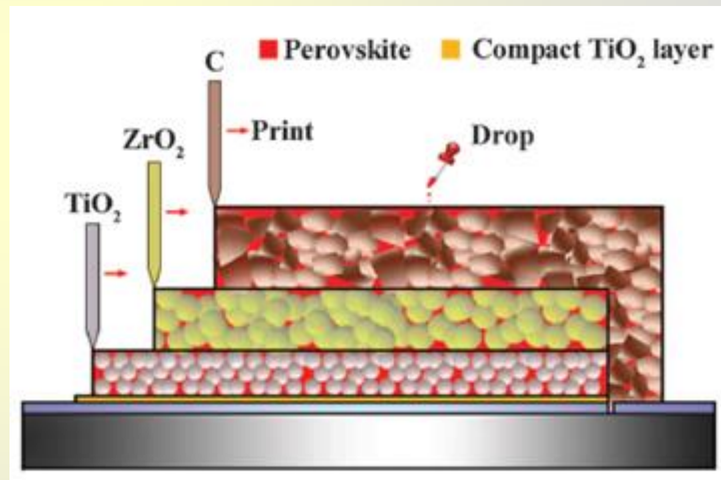
Film Configuration	J_{SC} /mA cm^{-2}	V_{OC} /mV	FF	η /%
HD1/Al_2O_3/CE	**20.357**	**857**	**0.646**	**11.3**
HD1/HD3/Al_2O_3/CE	16.112	742	0.653	7.8
HD1/Al_2O_3/HD3/CE	18.833	861	0.572	9.3

Carbon-based PSC with a Multi-layer Configuration

A one-step approach





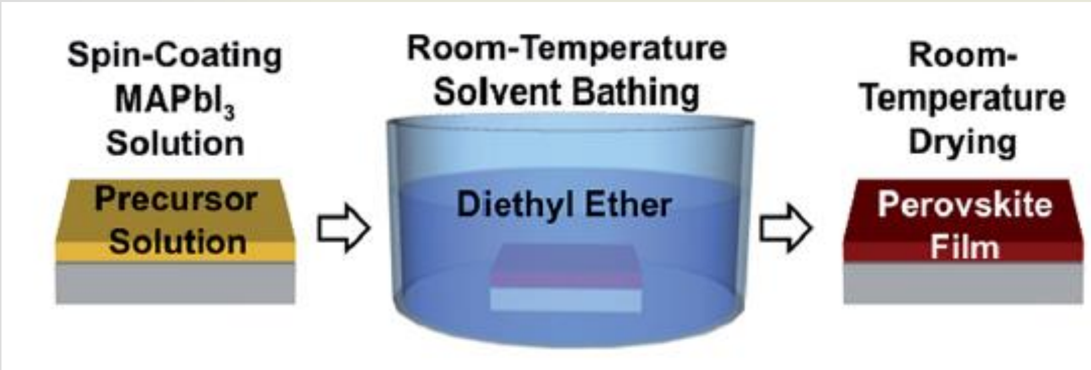


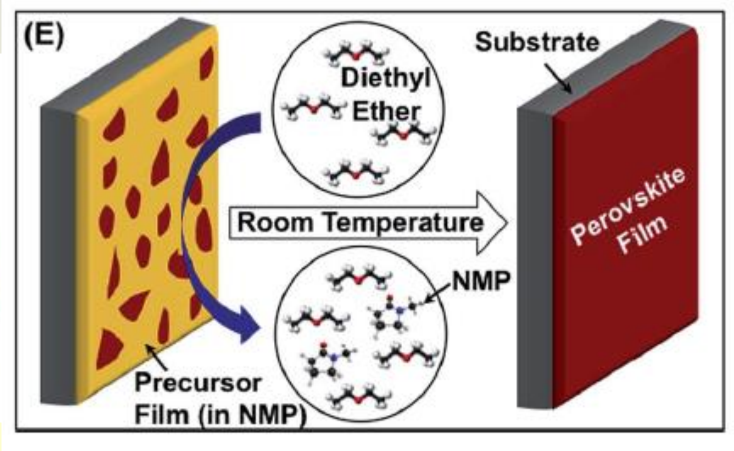
$(5\text{-AVA})_x(MA)_{1-x}PbI_3$			
J_{SC} (mA cm^{-2})	V_{OC} (mV)	FF	η (%)
22.80	858	0.66	12.84

Han and co-workers, *Sci. Rep.* **2013,** *3,* 3132.

Han and co-workers, *Science* **2014**, *345*, 295-298.

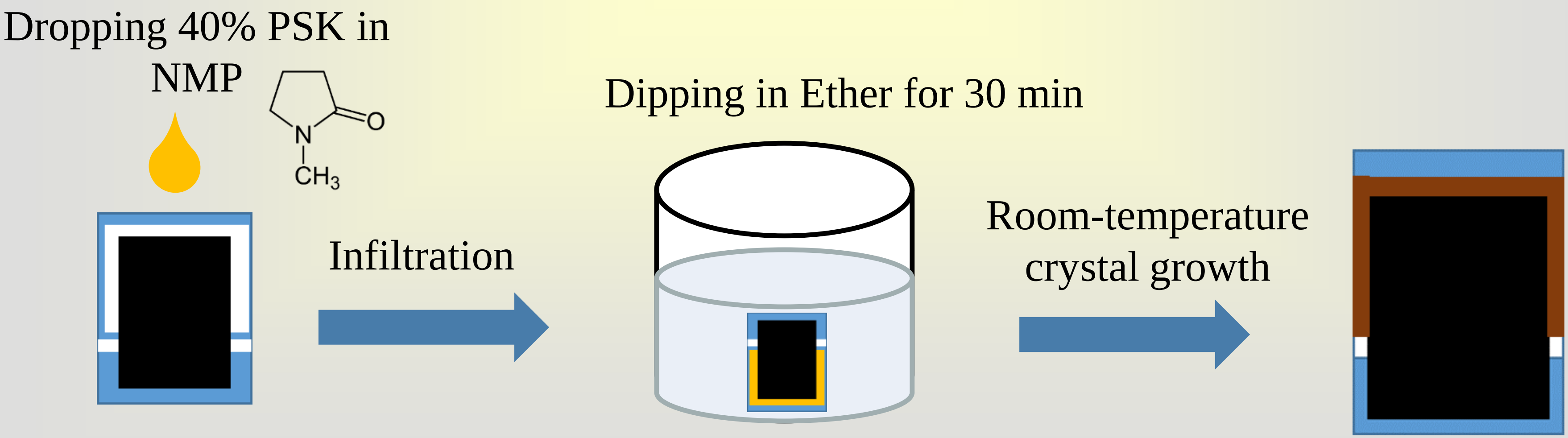
The Solvent-Extraction Crystal Growth Approach





Zhu and co-workers, *Journal of Materials Chemistry A* **2015**, *3*, 8178-8184.

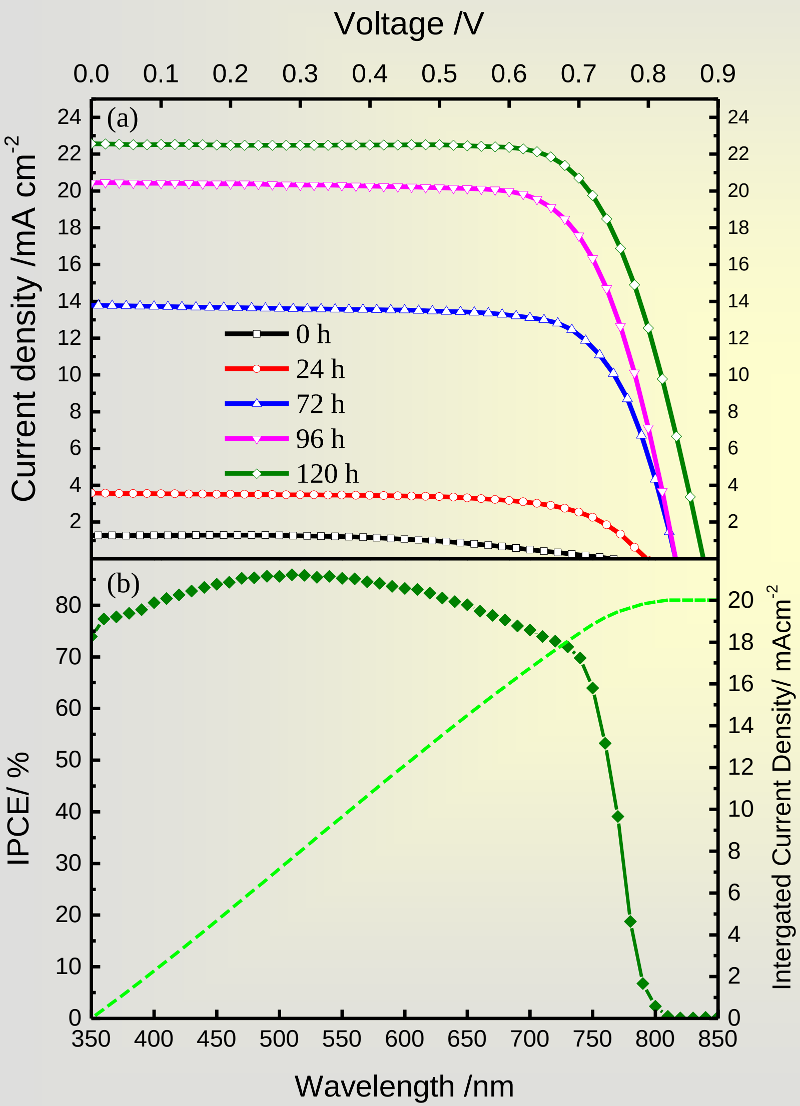
Our approach

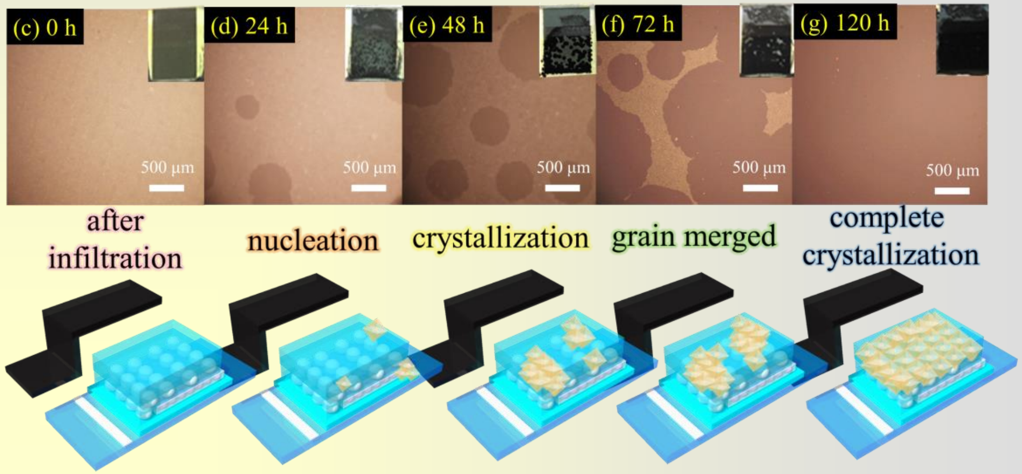


Chan et al, *Journal of Materials Chemistry A* **2016**, *4*, 3872-3878.

Solvent Extraction + Slow Crystal Growth

Slow Crystallization to Attain PCE 15 %

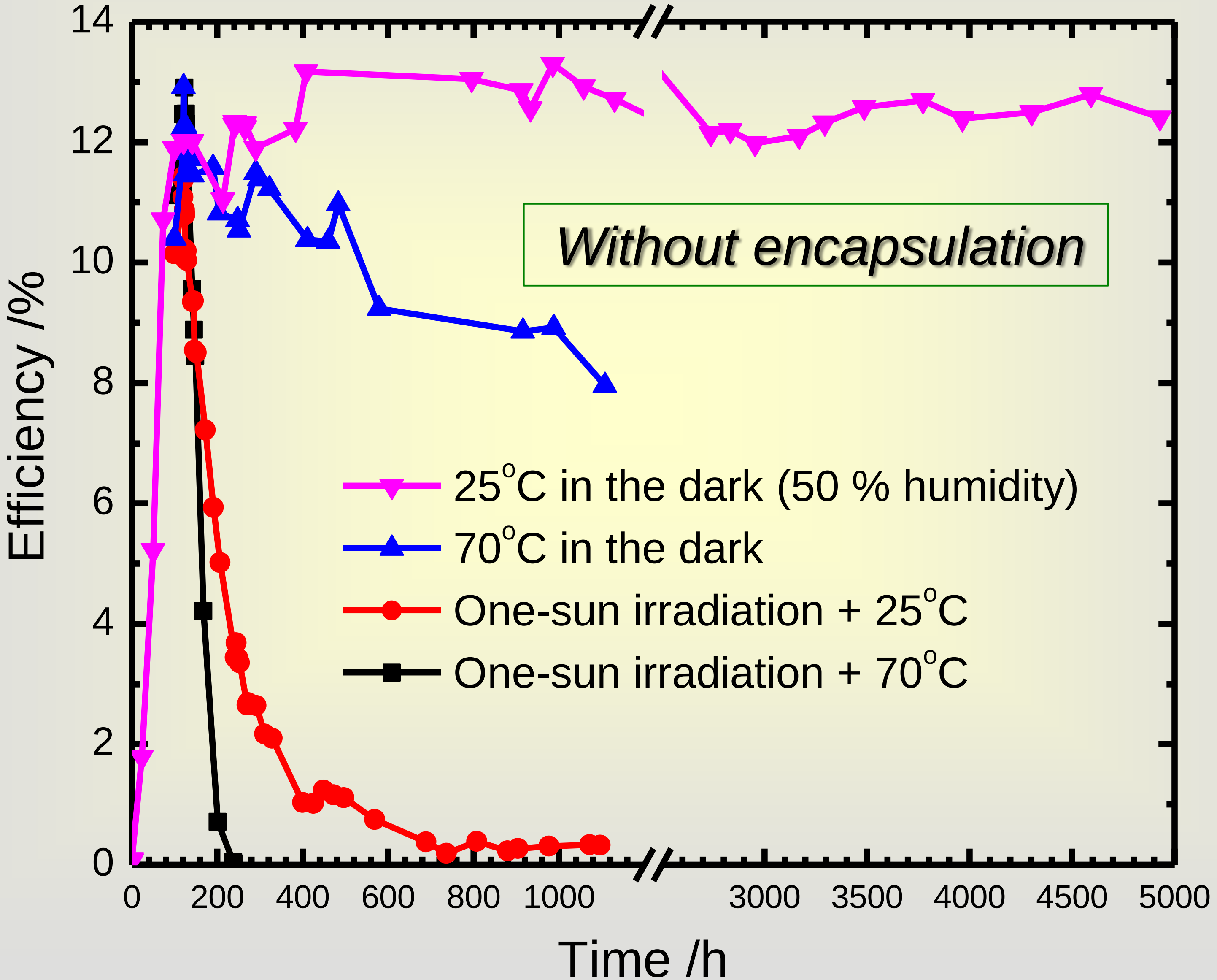




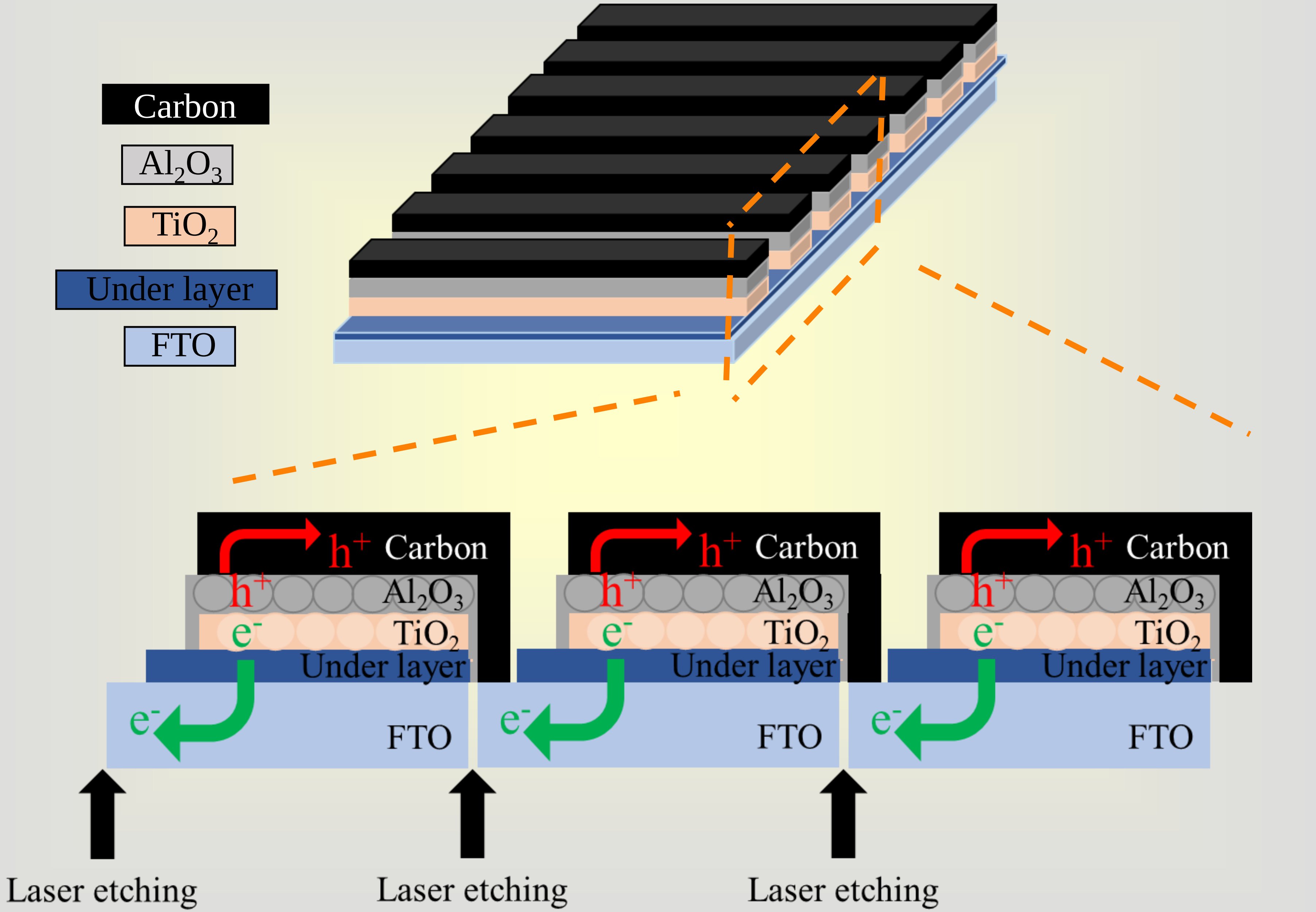
SC time /hours	J_{SC} /mA cm^{-2}	V_{OC} /mV	*FF*	η /%
0	1.27	750	0.511	0.5
24	3.57	798	0.676	1.9
72	13.77	840	0.745	8.6
96	20.43	841	0.734	12.6
120	22.43	893	0.747	15.0

J. Mater. Chem. A **2017**, *5*, 739-747.

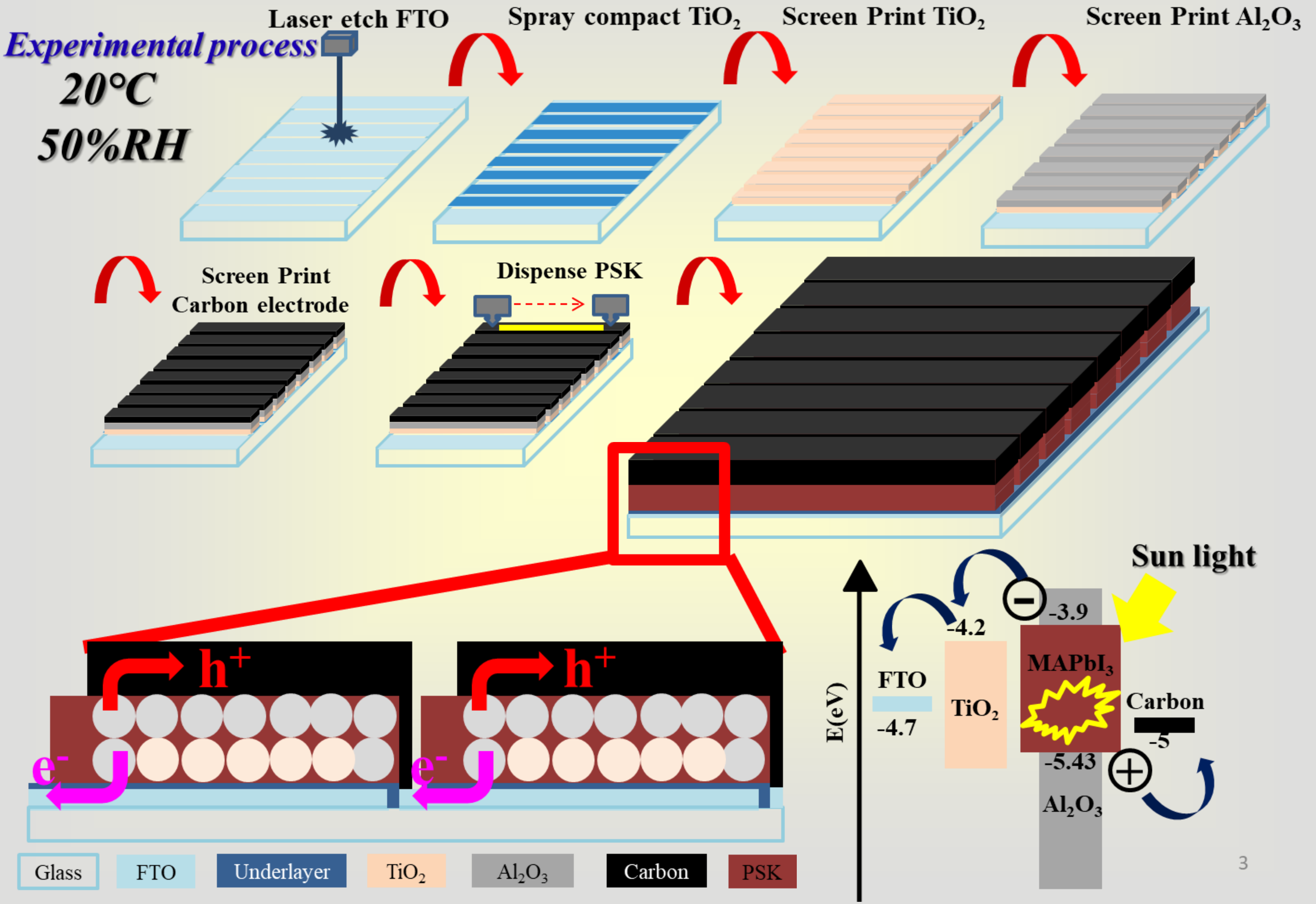
Long-term Stability Examination



Large-Scale Carbon-based PSC



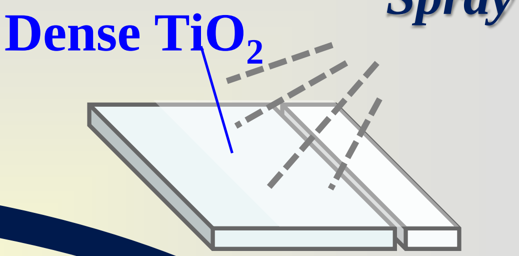
Carbon-Based Perovskite Modules



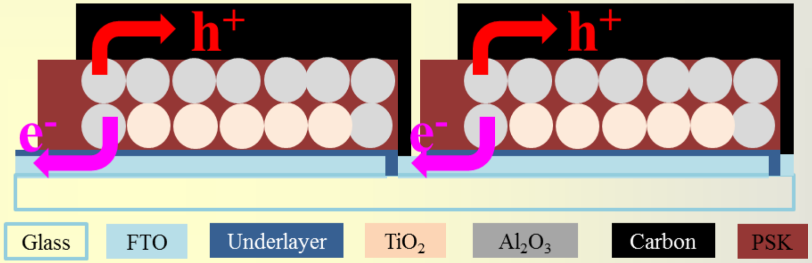
Advantages

- Module design flexible
- Stable materials
- Ambient condition process
- Cost effective

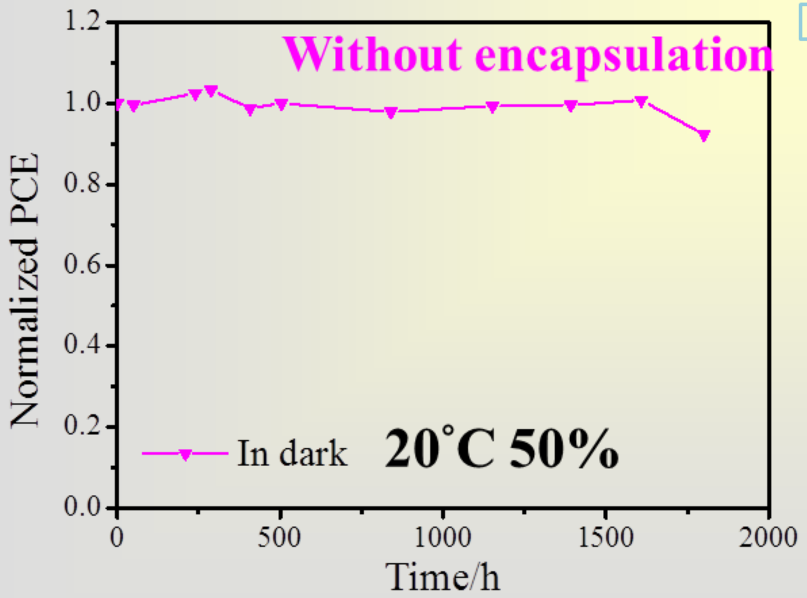
Etch



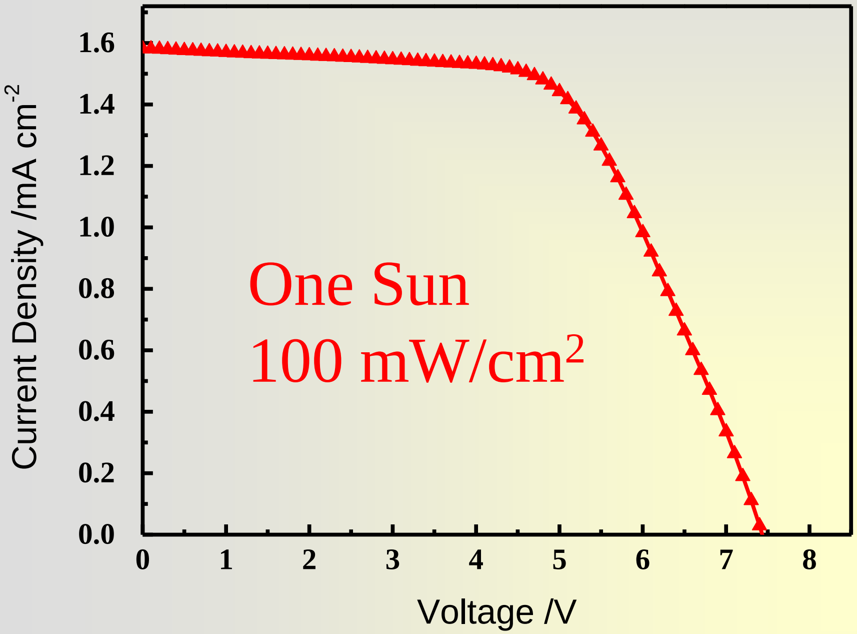
Screen Printing

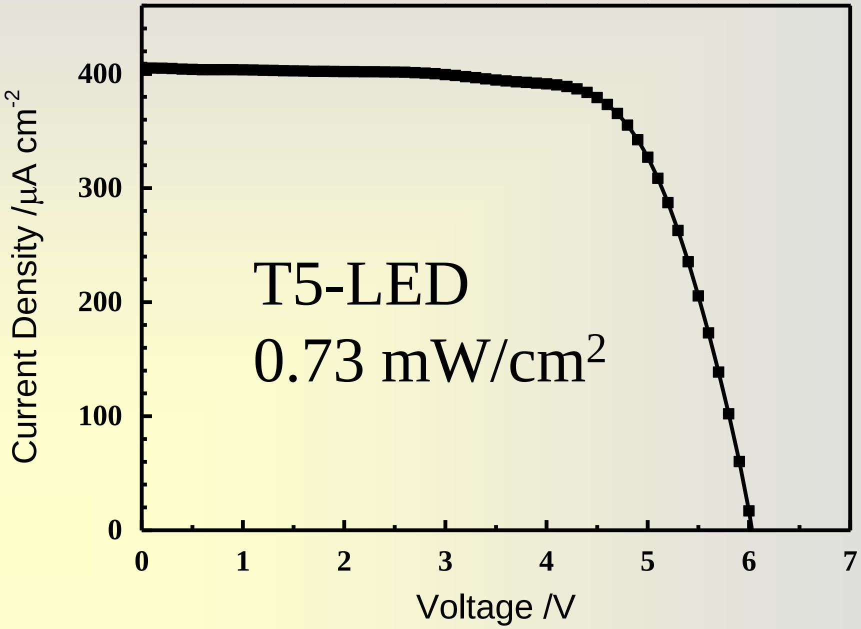


Inkjet printing



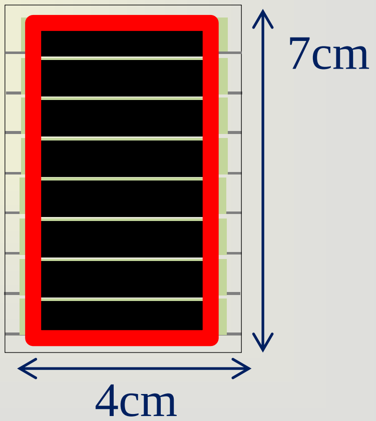
Carbon-Based Perovskite Modules





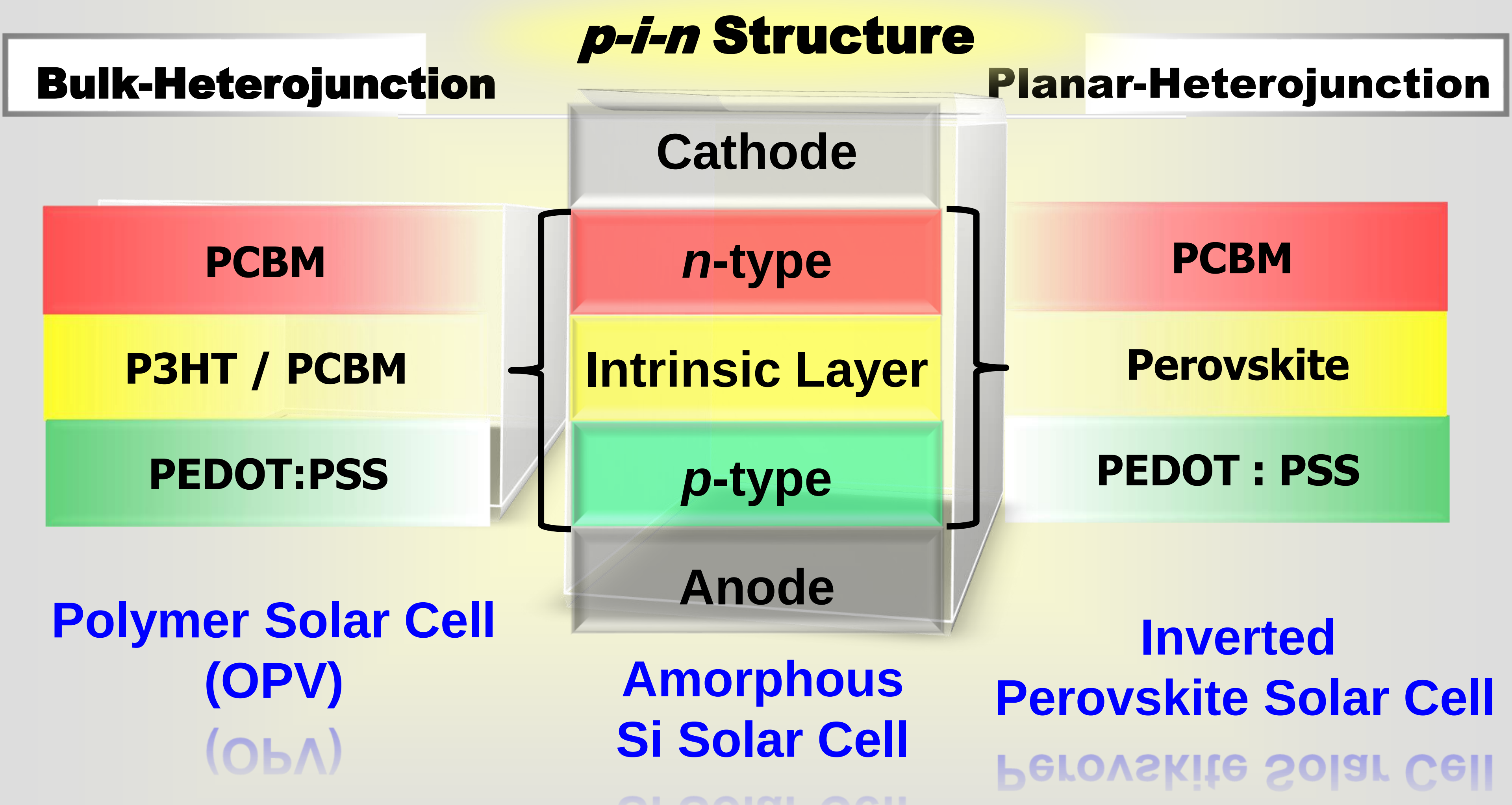
Mask (19 cm^2)	Voc /V	Isc /mA	FF	η/%
One Sun	7.5	23.9	0.61	7.2
T5 LED	6.0	0.41	0.70	12.5

Mask

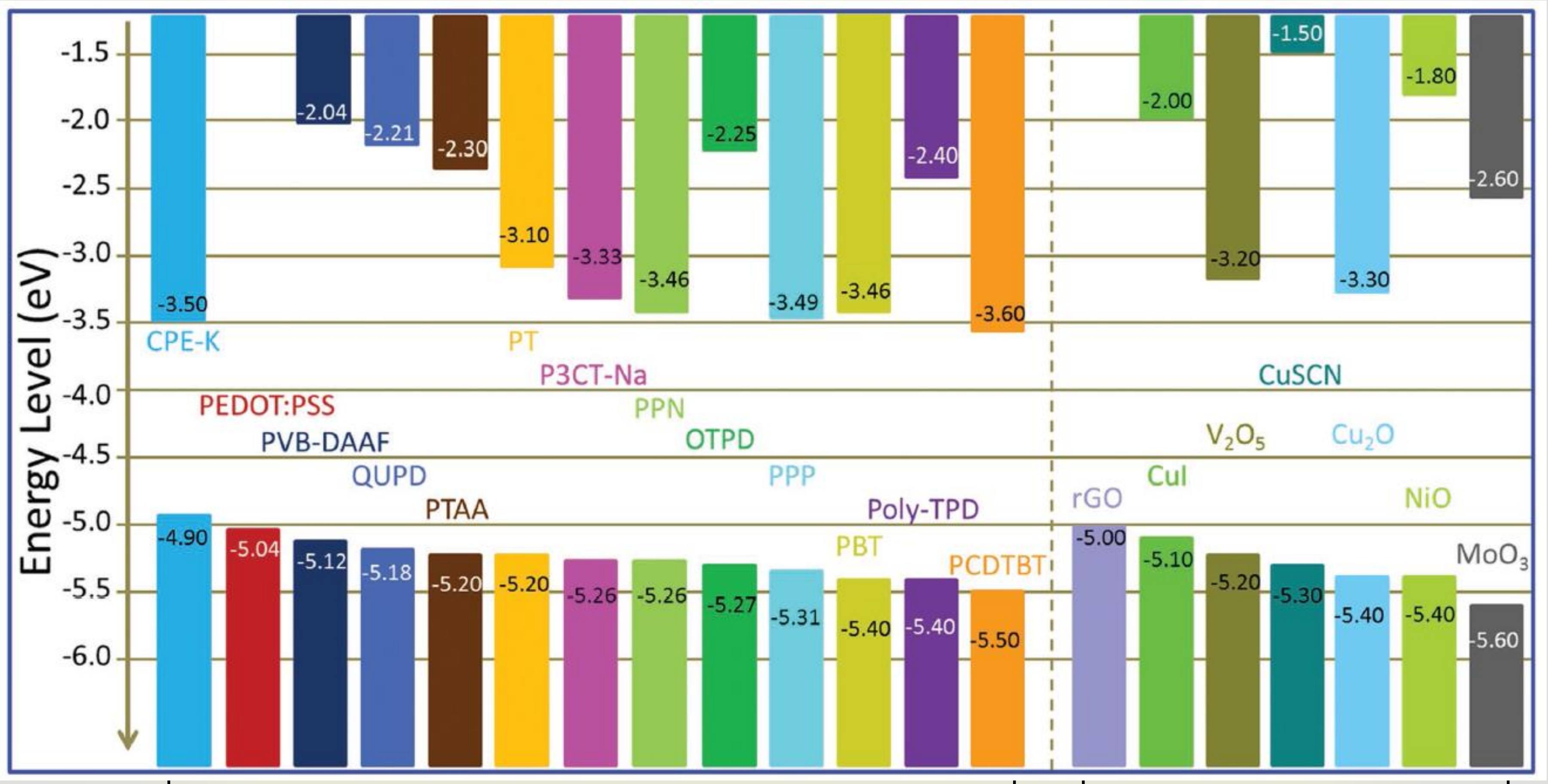


Inverted Planar Perovskite Solar Cells

Planar Heterojunction (PHJ) Structures



Varied Types of Hole Conductors



Organic hole conductor **Inorganic hole conductor**

Yan et el., *Adv. Energy Mater.* **2016,** 1600474.

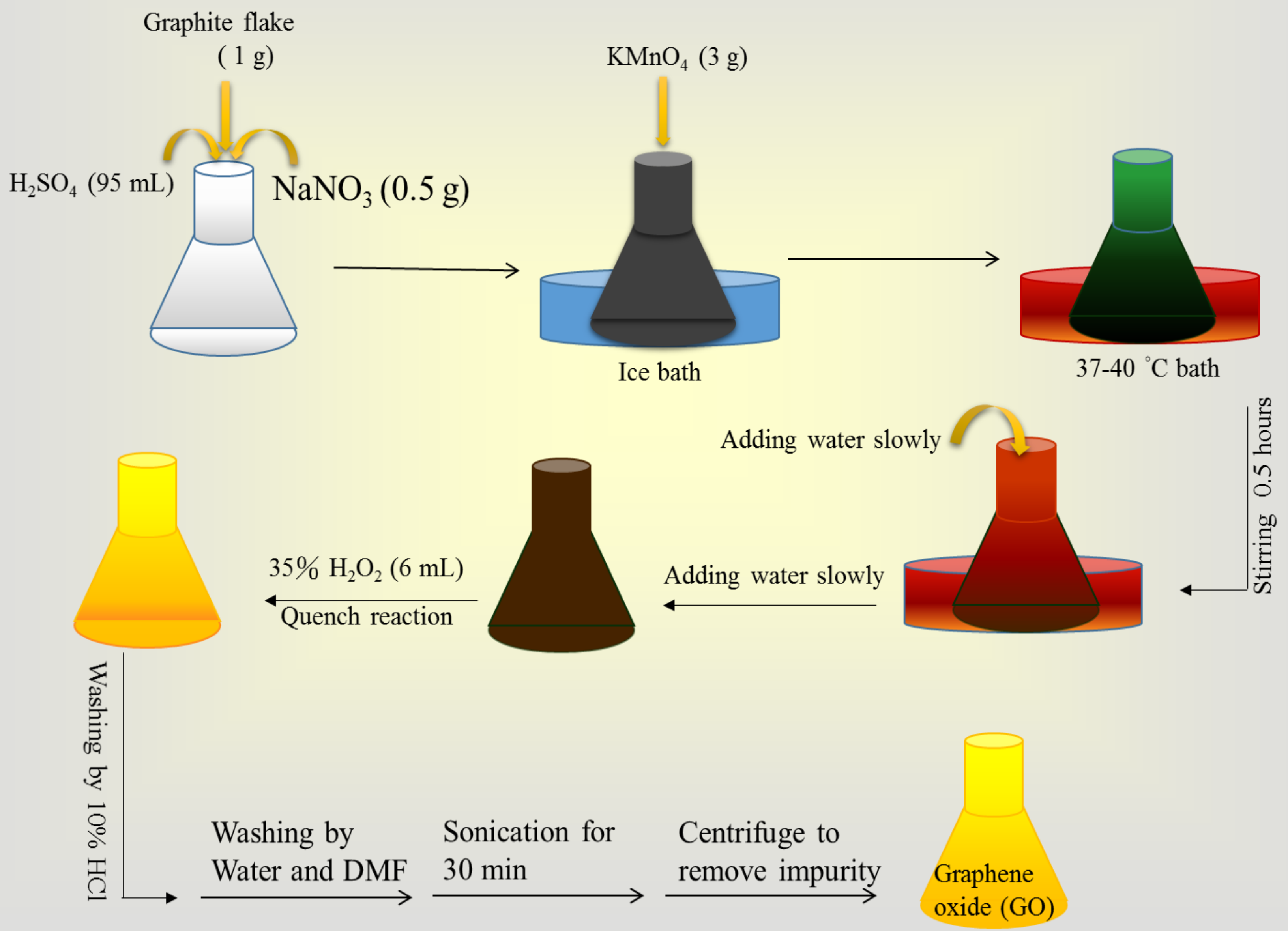
Graphene Oxide (GO)-based Inverted Perovskite Solar Cells

Advantages of Graphene Derivatives

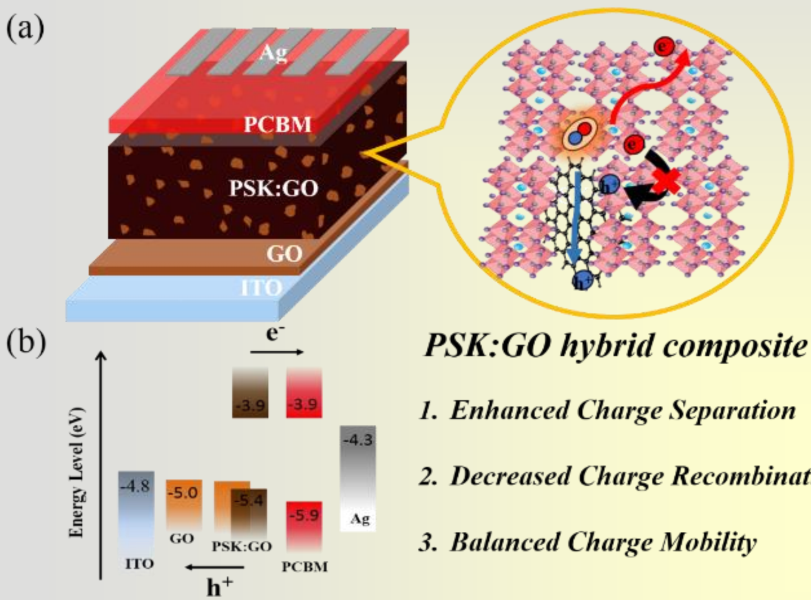
- **Excellent thermal and chemical stability**
- **High mechanical strength**
- **High transparency**
- **Large surface area**
- **Rapid charge mobility**
- **Easy to modify → GO, rGO, etc.**
- **Cost-effective solution-based processing**
- **Low-temperature fabrication**
- **Flexible devices**

Angew. Chem. Int. Ed. **2014**, *53*, 3588-3593;
JACS, **2013**, *135*, 10286-10289;
J. Mater. Chem. A **2014**, *2*, 1325-1331.

Synthesis of GO using Hummers Method

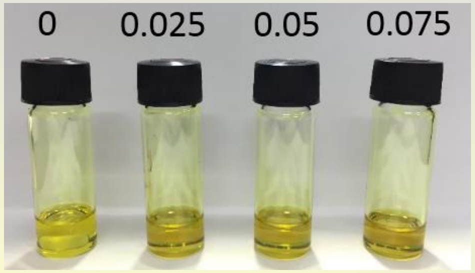


The First Inverted PSK/GO BHJ PSC

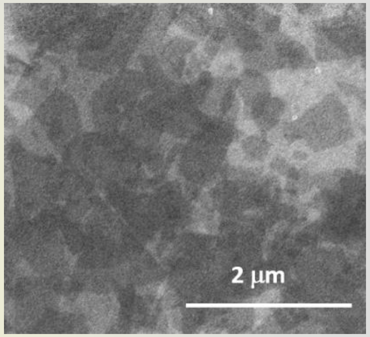


PSK:GO hybrid composite

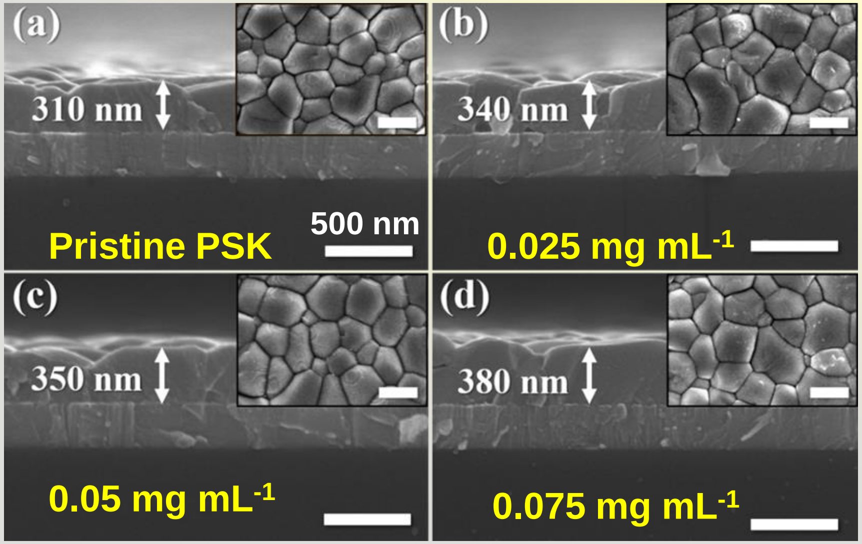
1. *Enhanced Charge Separation*
2. *Decreased Charge Recombination*
3. *Balanced Charge Mobility*

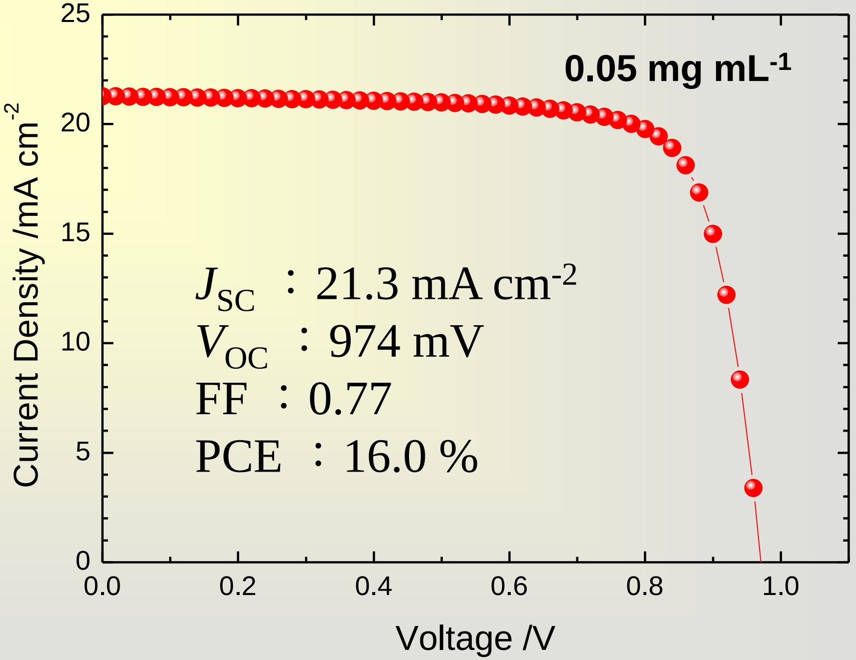


PSK:GO solution in mg mL^{-1}



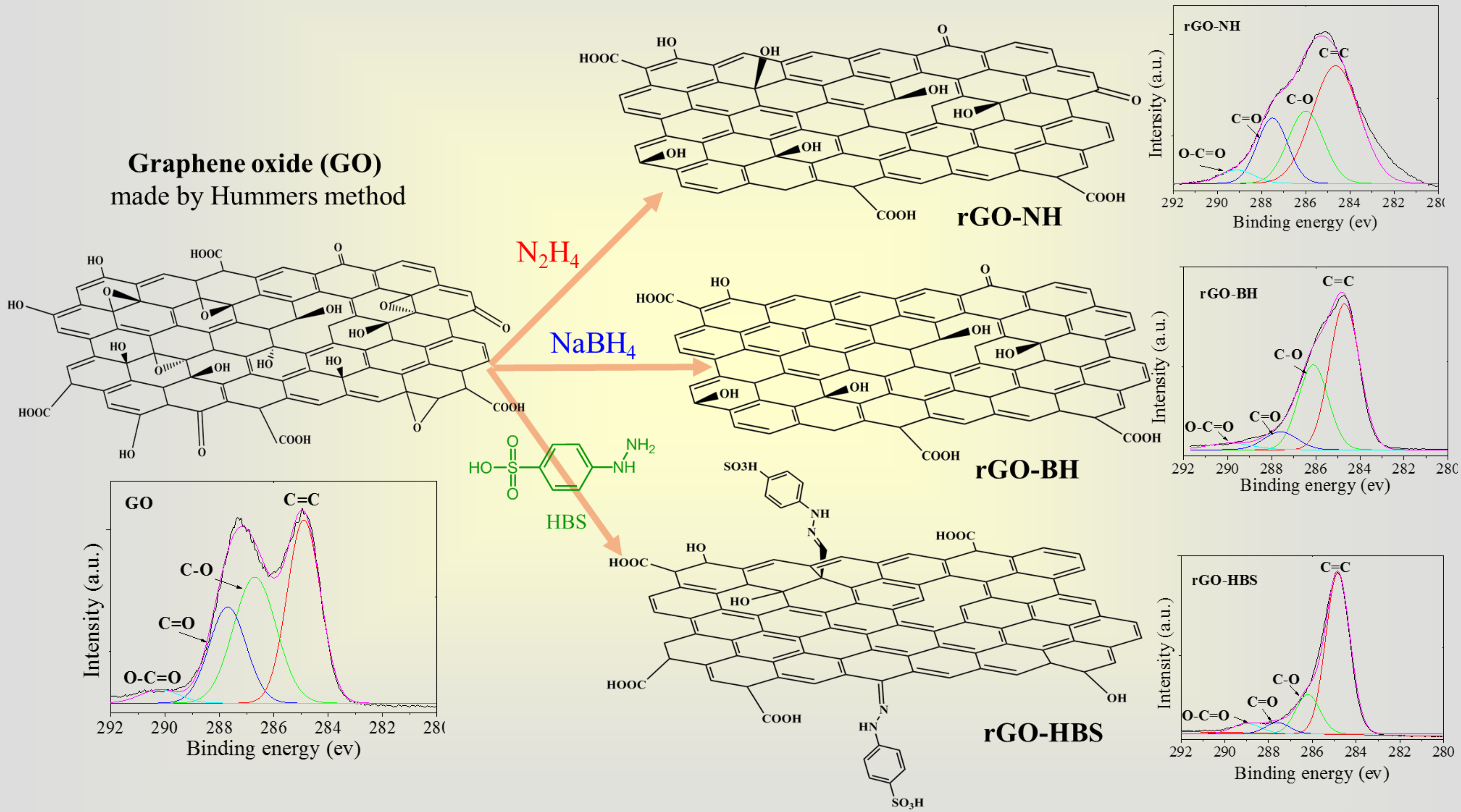
SEM of GO on ITO





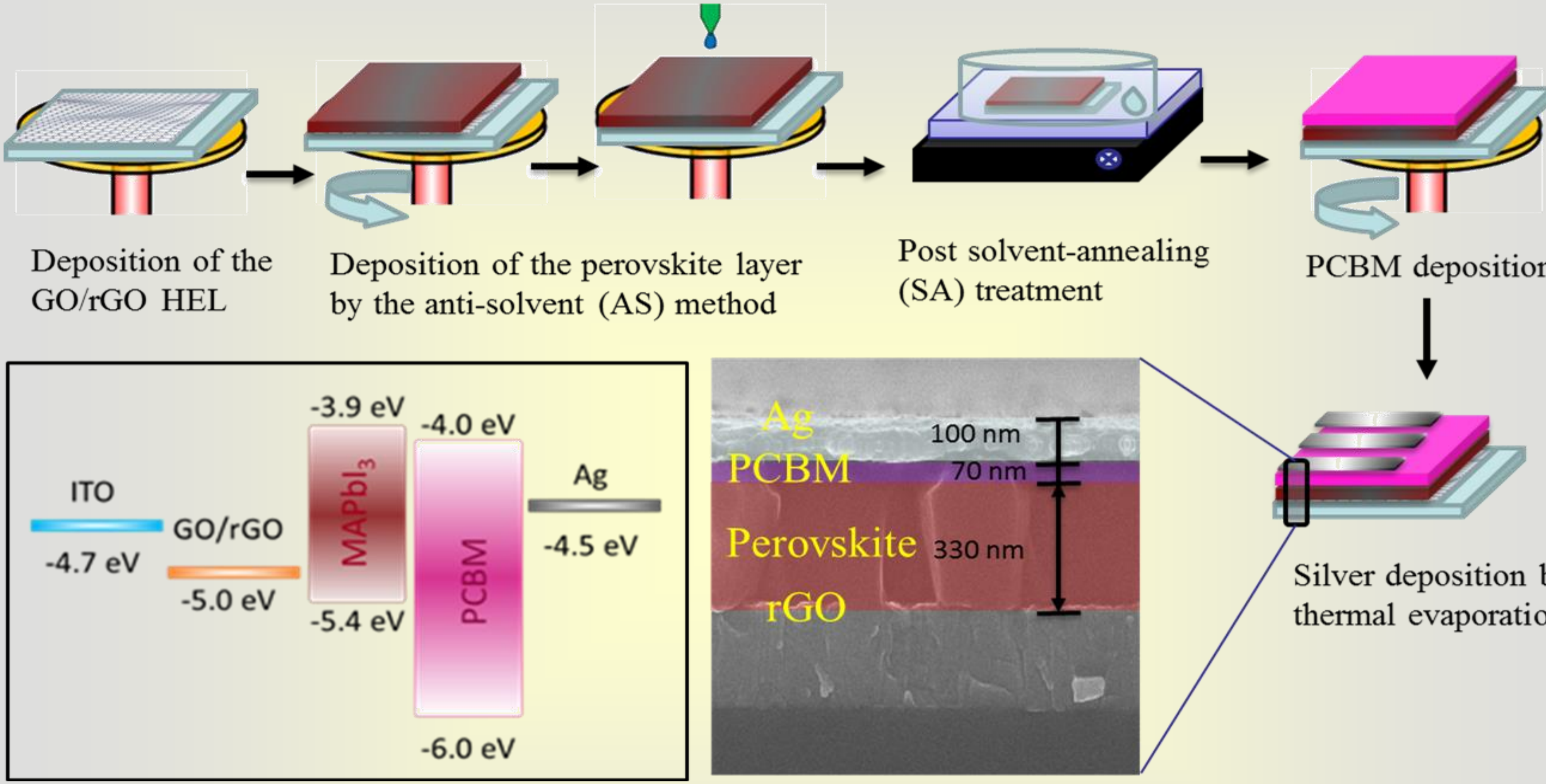
J. Mater. Chem. A **2017**, *5*, 13957-13965.

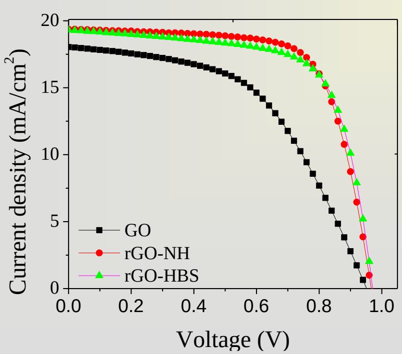
Using rGO as HEL for Flexible PHJ PSC

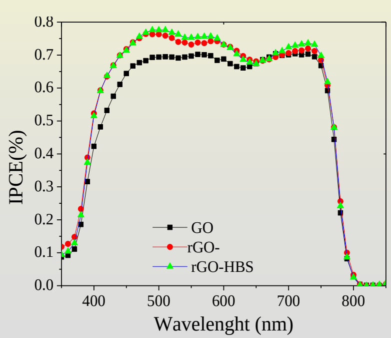


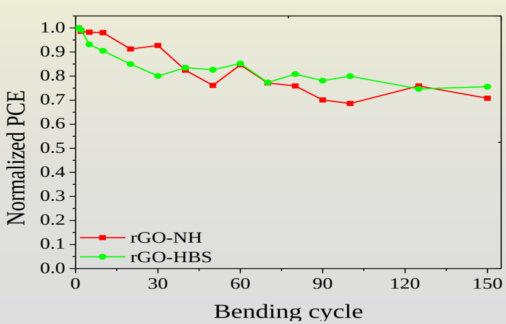
Adv. Energy Mater. **2018**, *8*, 1701640

Reduced Graphene Oxide for PHJ PSC

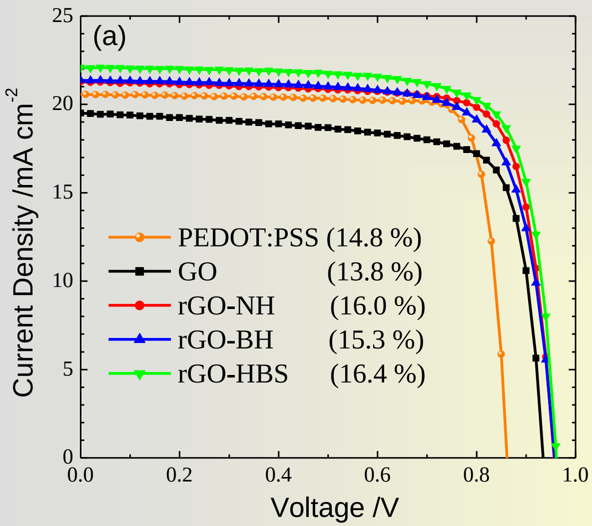


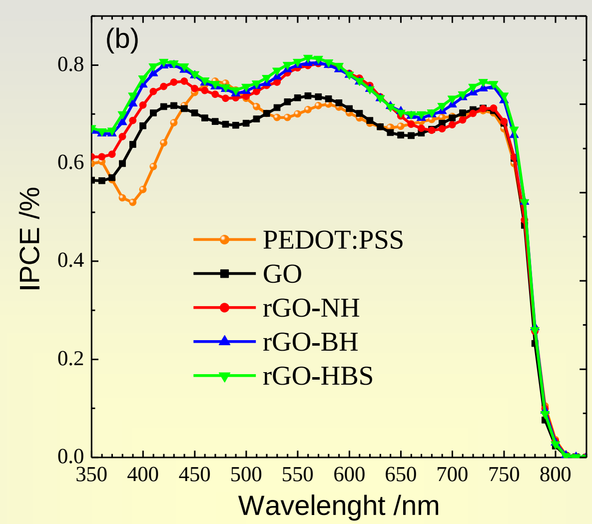


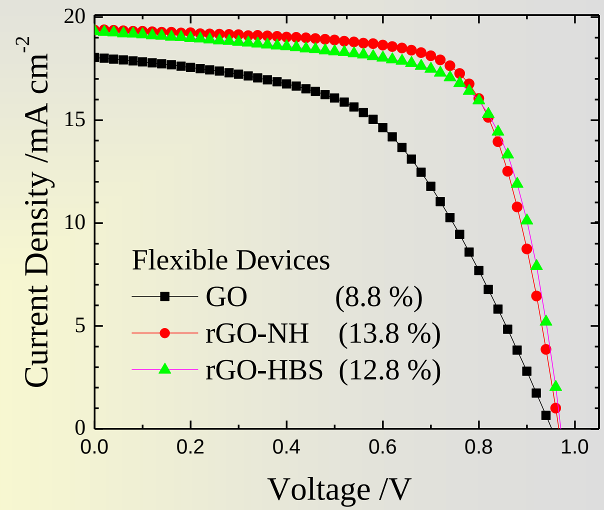


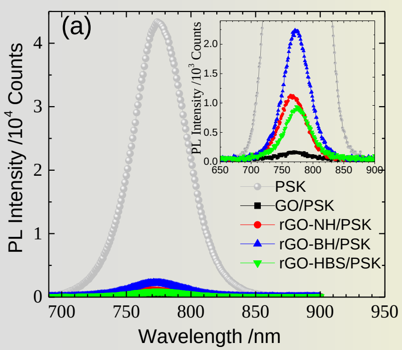


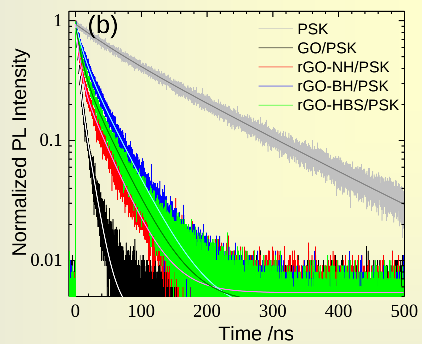
Anomalous Charge-extraction Behavior

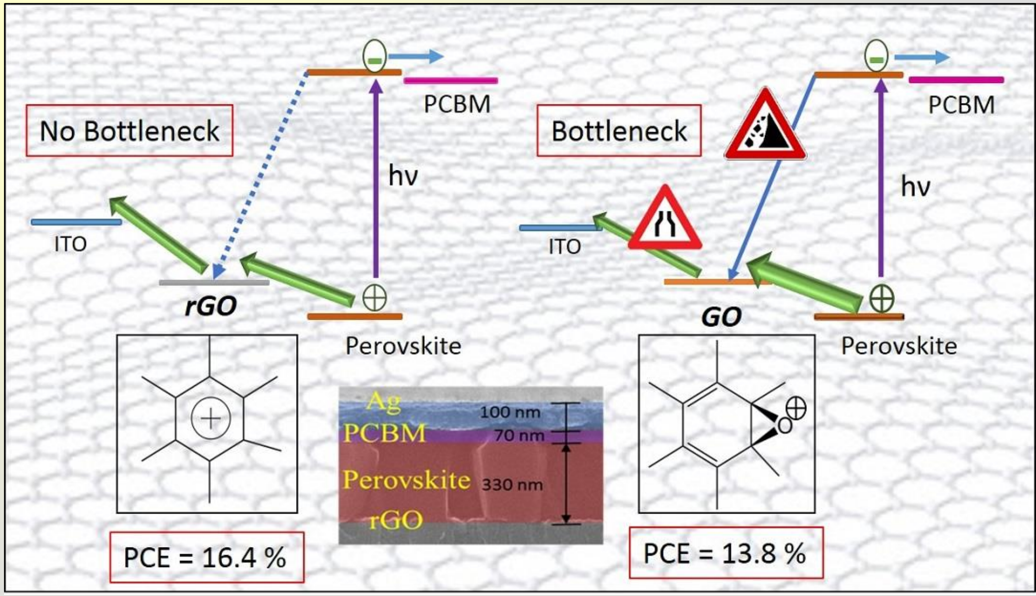


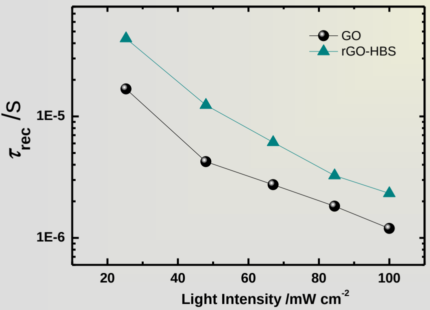


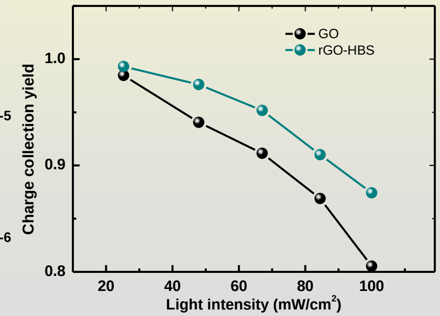






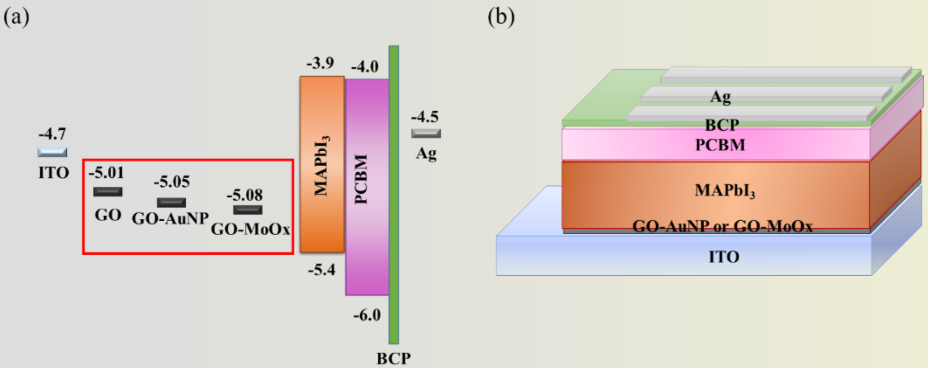


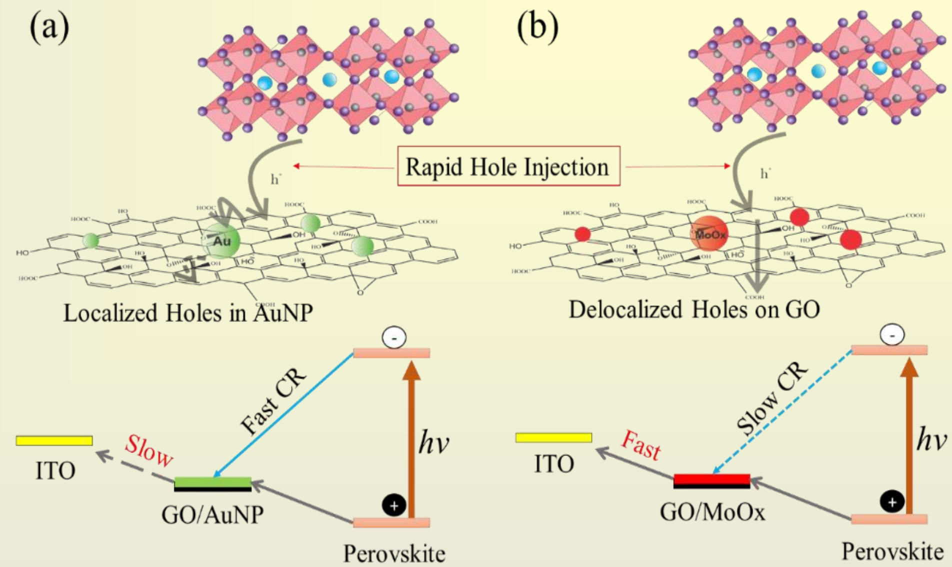


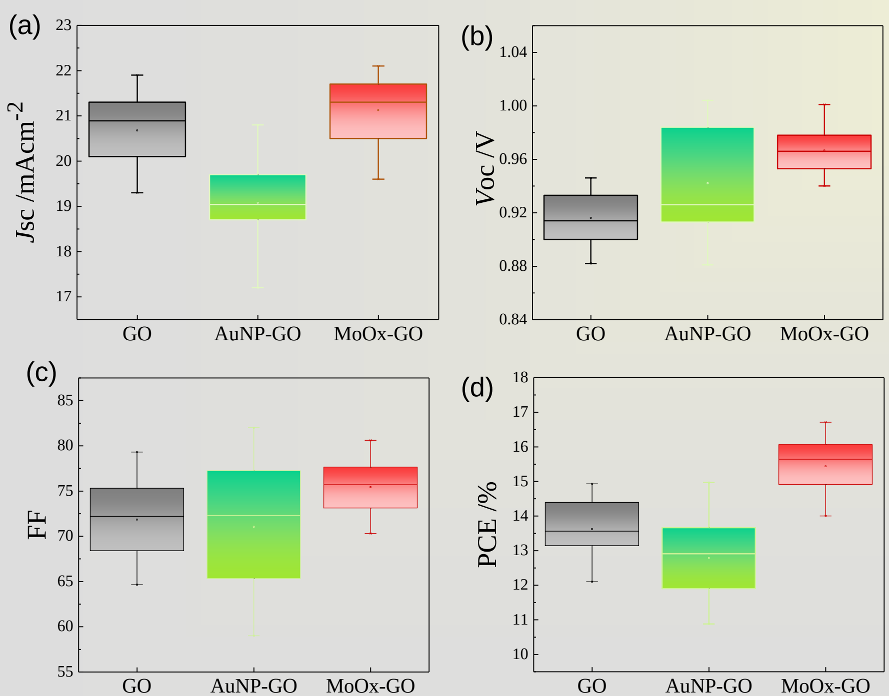


Adv. Energy Mater. **2018**, *8*, 1701640.

NP-modified GO as HEL for Planar PSC



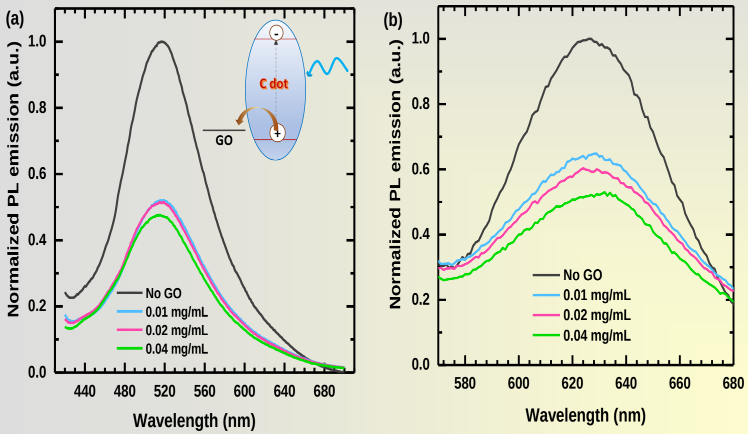


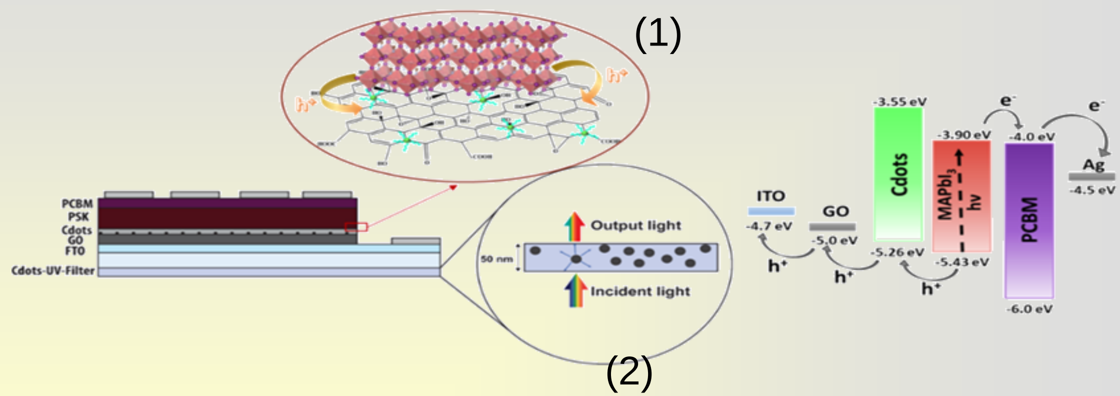


HEL	Jsc /mA cm^{-2}	Voc /V	FF	PCE /%
GO	21.0	0.913	0.751	14.4 (13.6 ± 0.8)
GO-AuNP	20.8	0.961	0.726	14.6 (12.8 ± 1.1)
GO-MoO_x	21.8	0.990	0.773	16.7 (15.4 ± 0.8)

Adv. Funct. Mater. **2018**, *28*, 1803200.

Carbon-Dot-modified GO as HEL for PSC

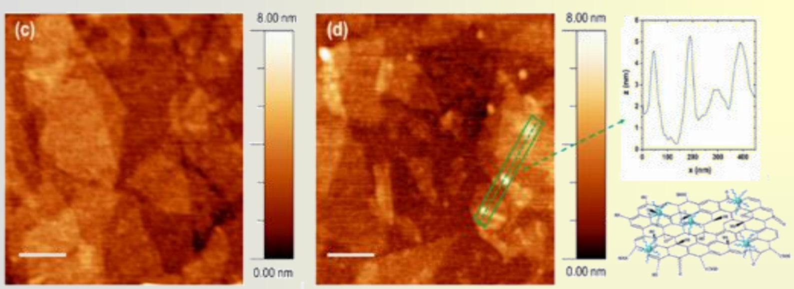


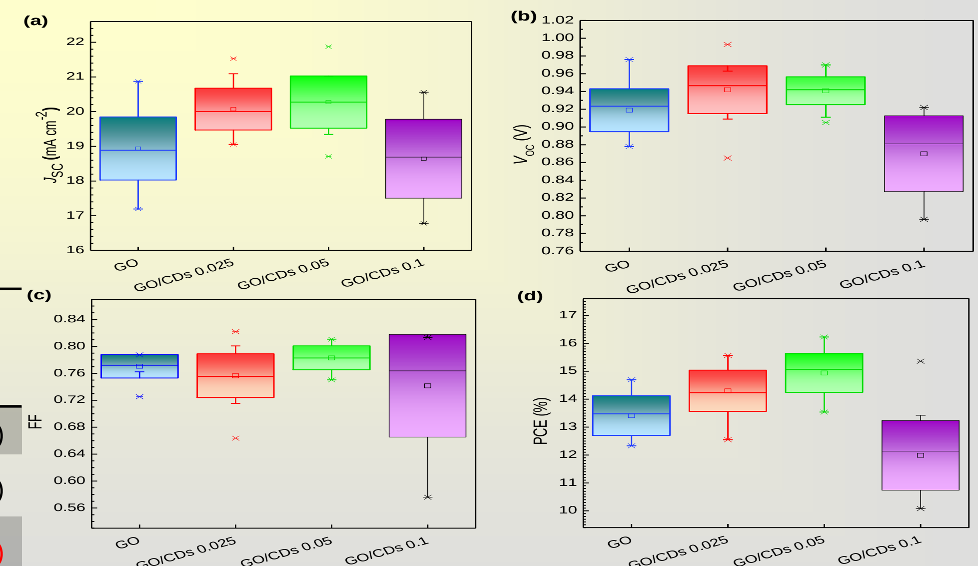


Two type of Carbon Quantum Dots :

(1) Hydrothermal: modifier of GO surface as HEL

(2) Microwave method: UV filter for back contact

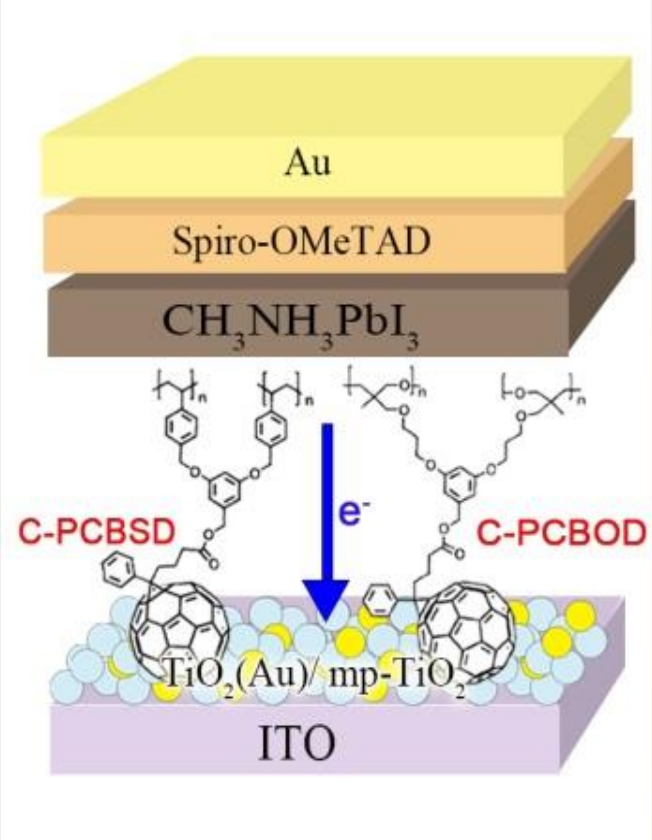


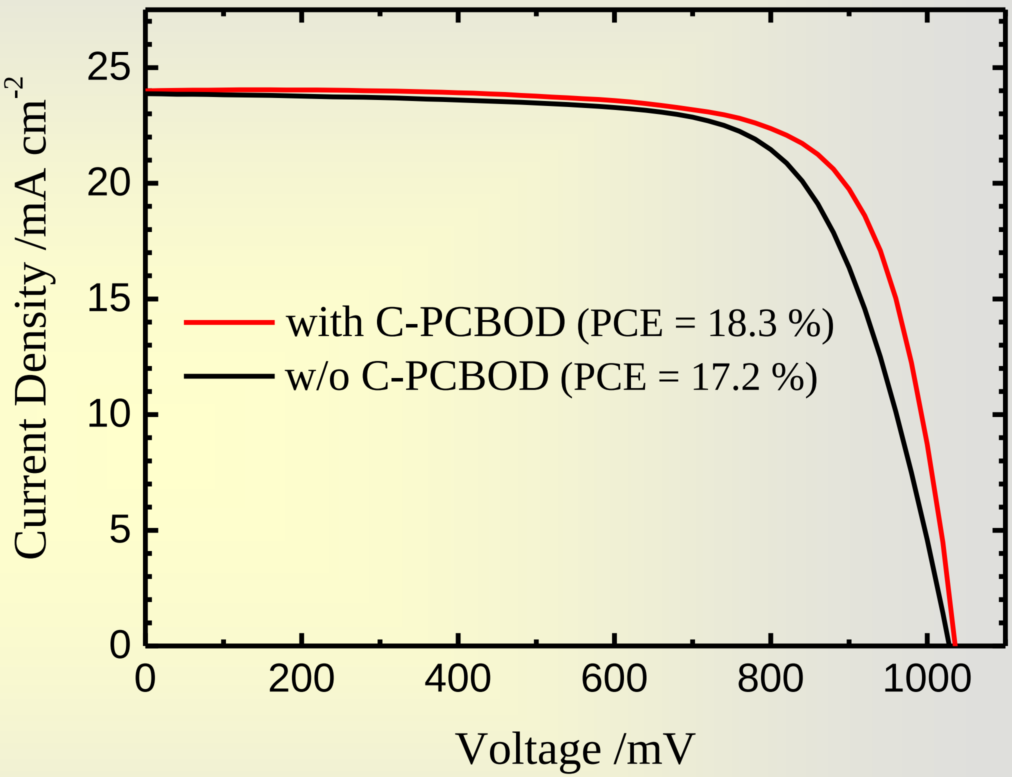


Devices	J_{SC} (mA cm^{-2})	Voc (mV)	FF	PCE (%)
GO	20.0	935	0.787	14.7 (13.4±0.7)
GO/Cdots 0.025	20.2	935	0.822	15.6 (14.3±0.7)
GO/Cdots 0.05	21.0	953	0.801	16.8 (14.9±0.7)
GO/Cdots 0.1	20.6	922	0.810	15.4 (12.0±1.4)

Nano Energy **2019**, *62*, 781-790.

Interfacial Engineering with Cross-Linked Fullerenes -- Our Best n-type PSC in 2017





Devices	V_{oc} / mV	J_{sc} / mA cm^{-2}	FF /%	PCE /%
w/o C-PCBOD	1042	24.7	66.6	17.2
with C-PCBOD	1046	24.6	70.5	18.3

ACS Appl. Mater. Interfaces **2017**, *9*, 38530-38536.

The Toxicity Issue: Go Lead-Free