IUMRS-ICA 2026
ICC Tainan, Taiwan
Conference: October 31–November 4, 2026

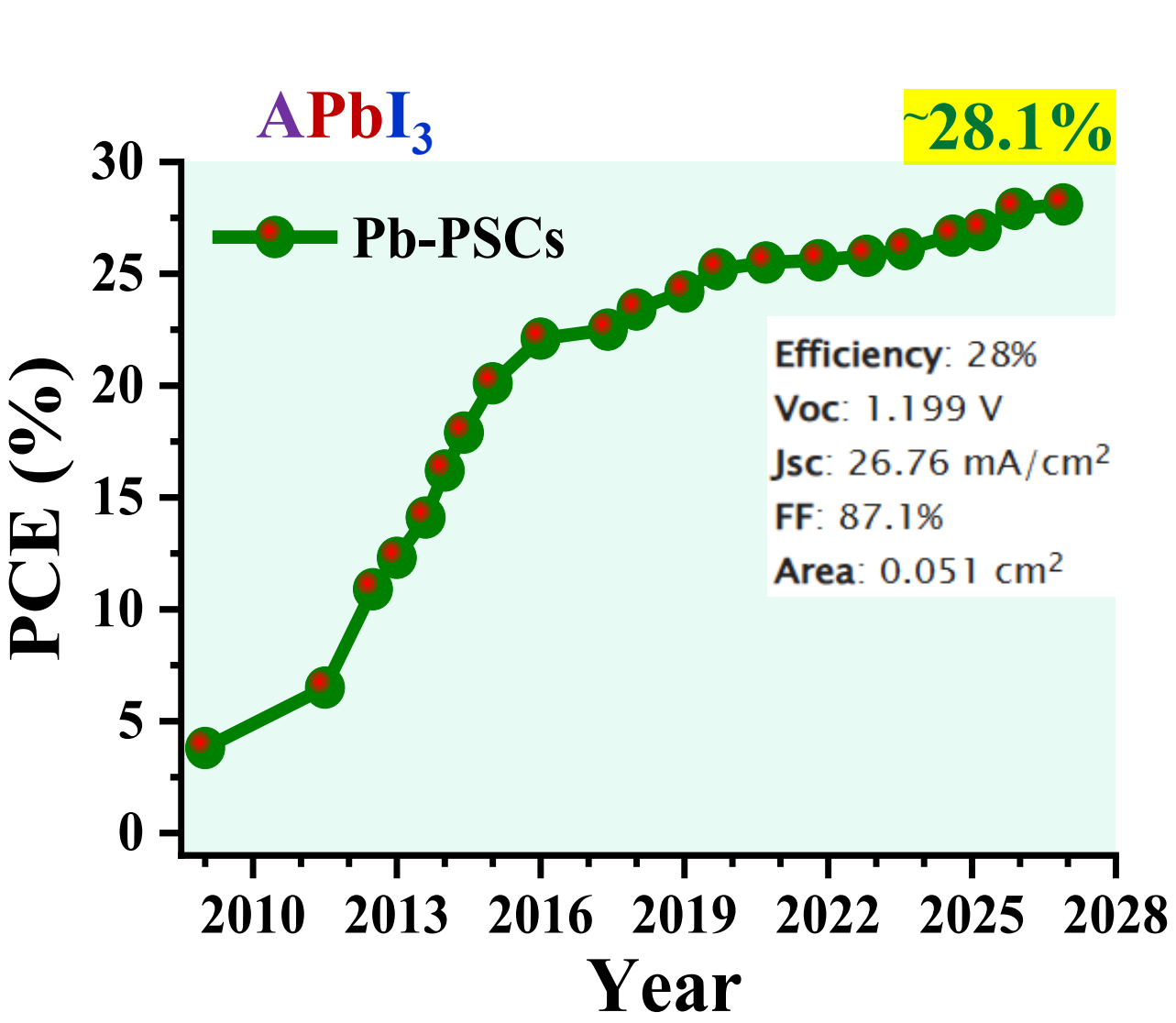
Multifunctional Additives Engineering of Perovskite Solar Cells

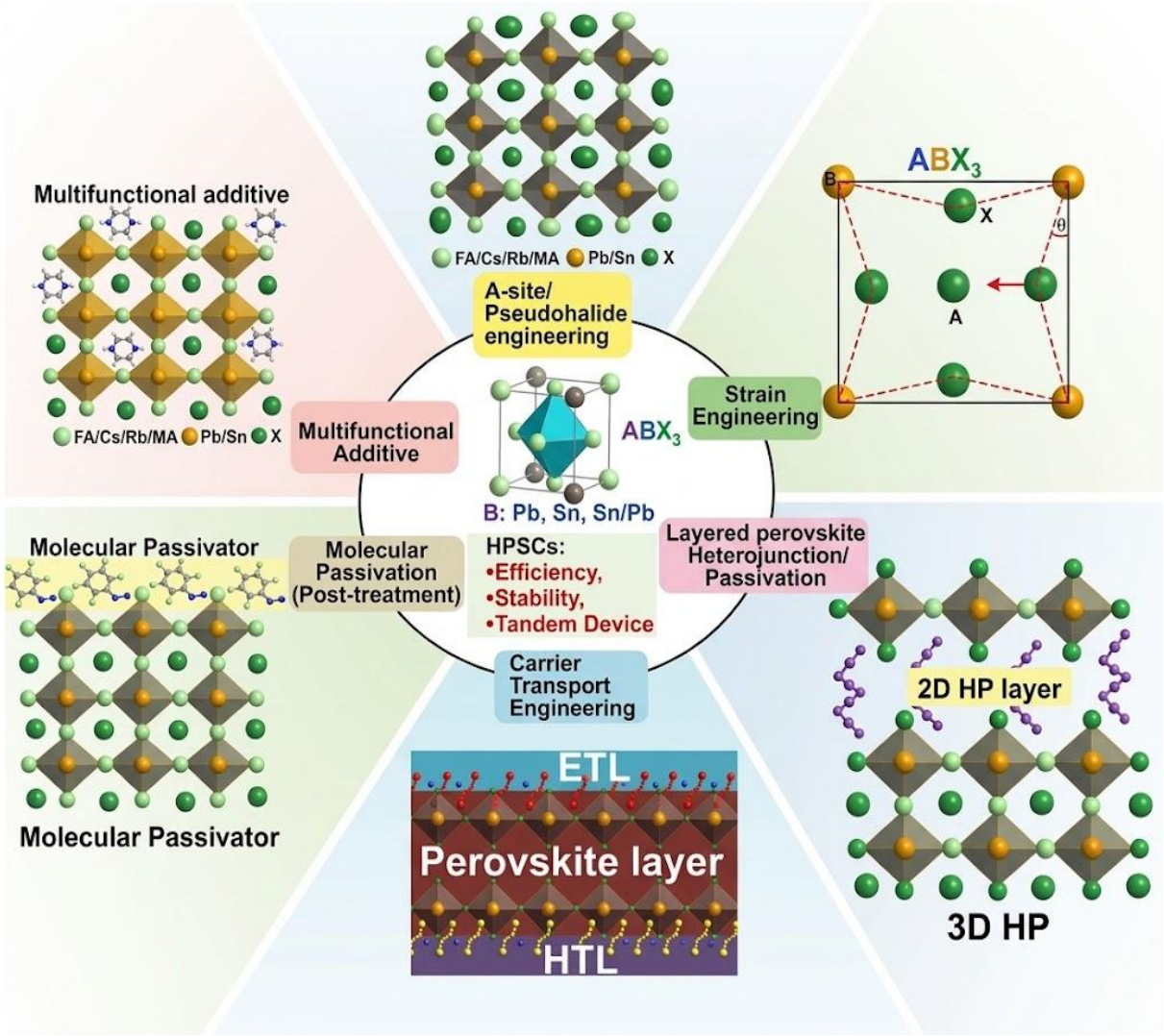
*Dhruba B. Khadka[1], Masatoshi Yanagida[1], Yasuhiro Shirai[1]

[1]Photovoltaic Materials Group,
Research Center for Energy and Environmental Materials (GREEN),
National Institute for Materials Science (NIMS), Tsukuba, Japan

C2 - Polymer Physics and Optoelectronic Applications
11:30 ~ 11:50; Venue: Nusantaria E

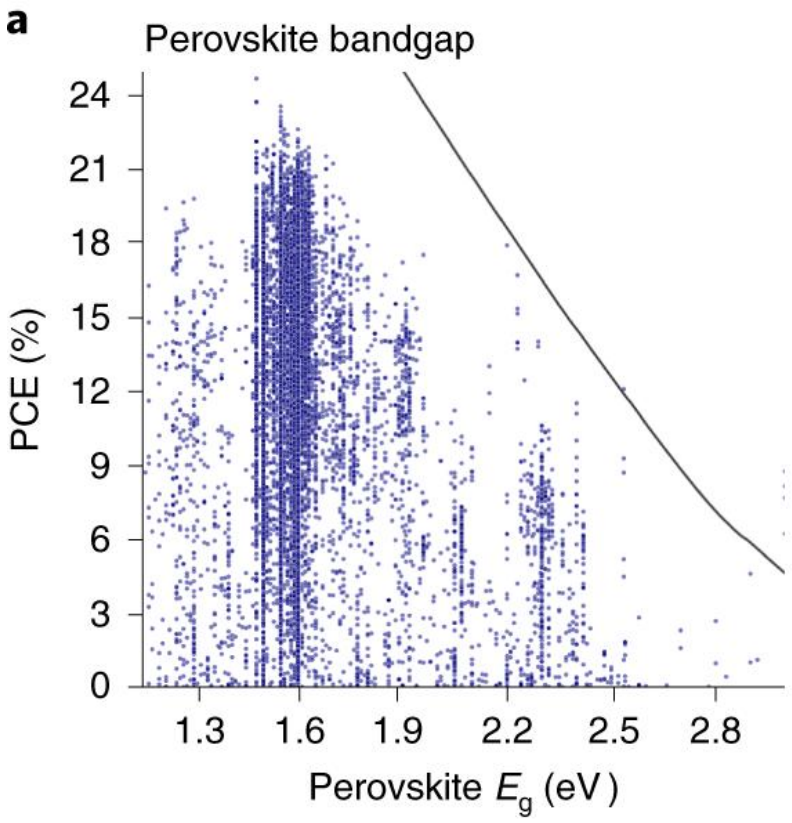
Device efficiency progress and stability

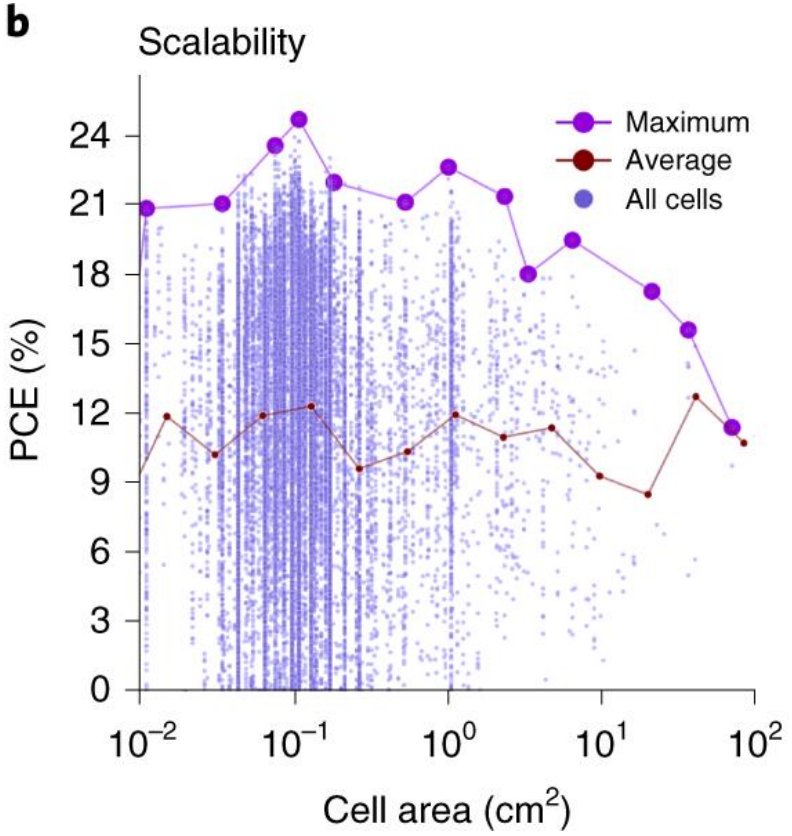


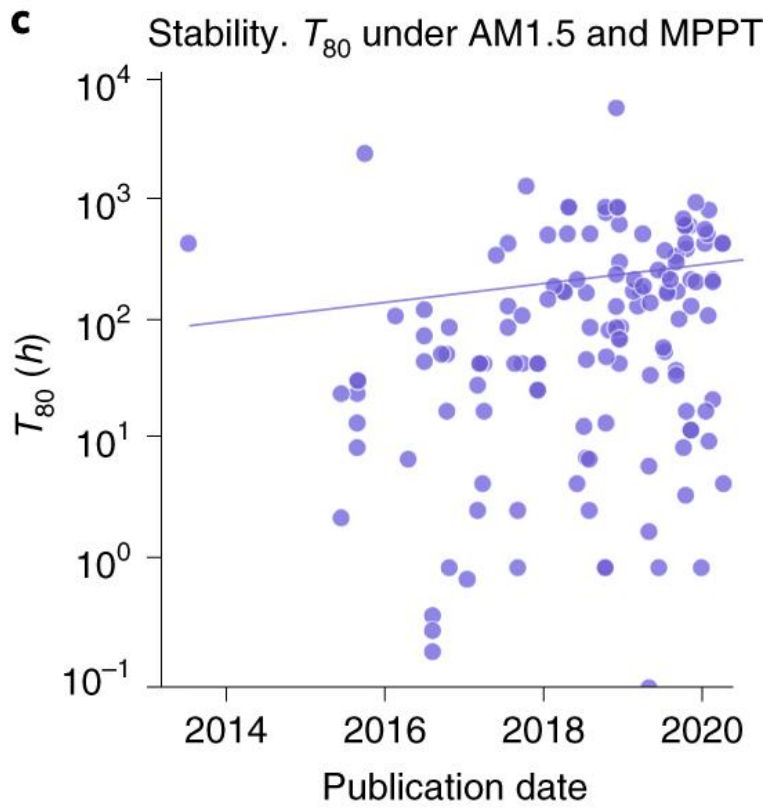


Khadka et al. Sol. RRL 2023, 2300535 (review article)

Device efficiency progress and stability







An open-access database and analysis tool for perovskite solar cells based on the FAIR data principles

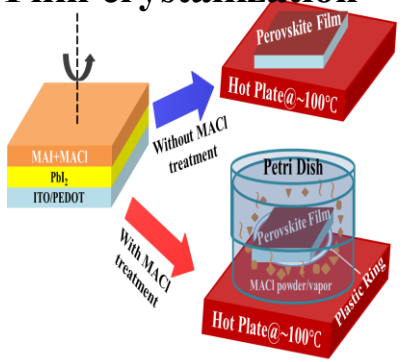
T. Jesper Jacobsson, Adam Hultqvist, Alberto García-Fernández, Aman Anand, Amran Al-Ashouri, Anders Hagfeldt, Andrea Crovetto, Antonio Abate, Antonio Gaetano Ricciardulli, Anuja Vijayan, Ashish Kulkarni, Assaf Y. Anderson, Barbara Primera Darwich, Bowen Yang, Brendan L. Coles, Carlo A. R. Perini, Carolin Rehermann, Daniel Ramirez, David Fairen-Jimenez, Diego Di Girolamo, Donglin Jia, Elena Avila, Emilio J. Juarez-Perez, Fanny Baumann, ... Eva Unger + Show authors

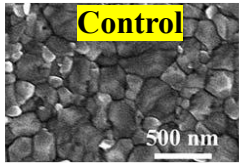
Nature Energy **7**, 107–115 (2022) | Cite this article

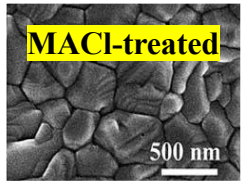
Save article

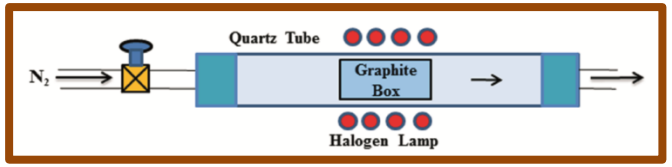
Our-reports-PSCs- device engineering

Film crystallization









Article | February 12, 2016 Subscribed

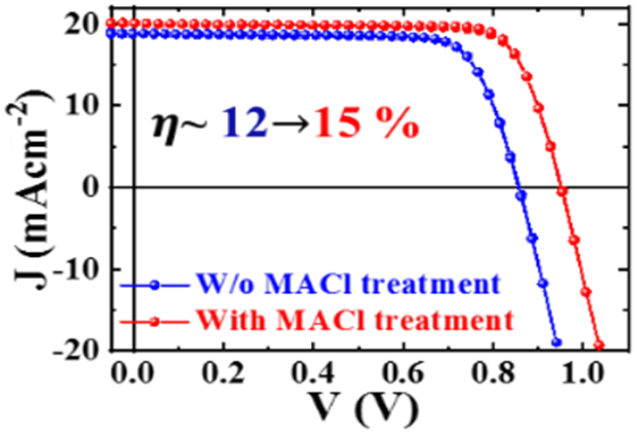
Effects of Ge Alloying on Device Characteristics of Kesterite-Based CZTSSe Thin Film Solar Cells

Dhruba B. Khadka; SeongYeon Kim; JunHo Kim *

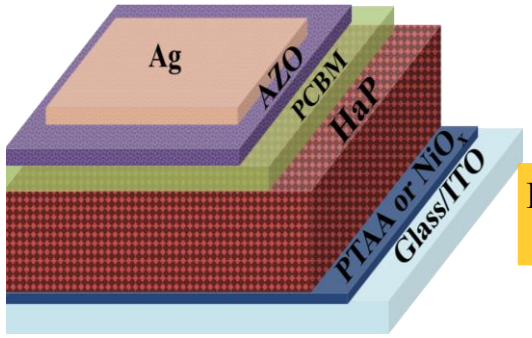
+ Author & Article Information

J. Phys. Chem. C (2016) 120 (8): 4251−4258.

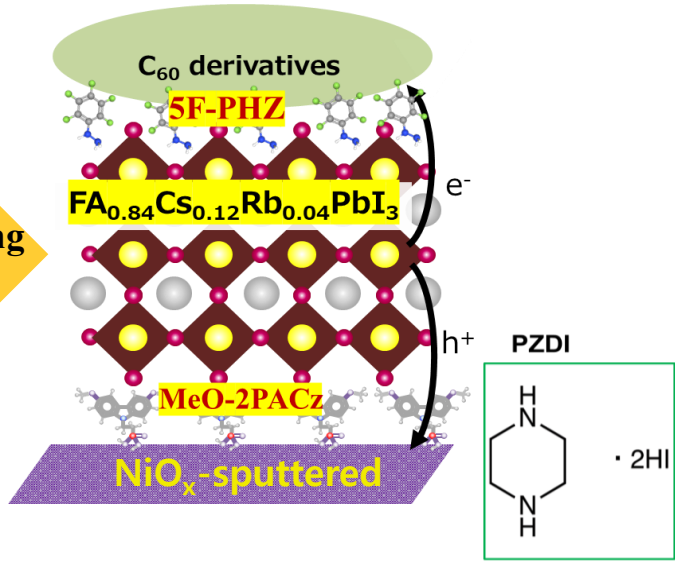
https://doi.org/10.1021/acs.jpcc.5b11594 Article history

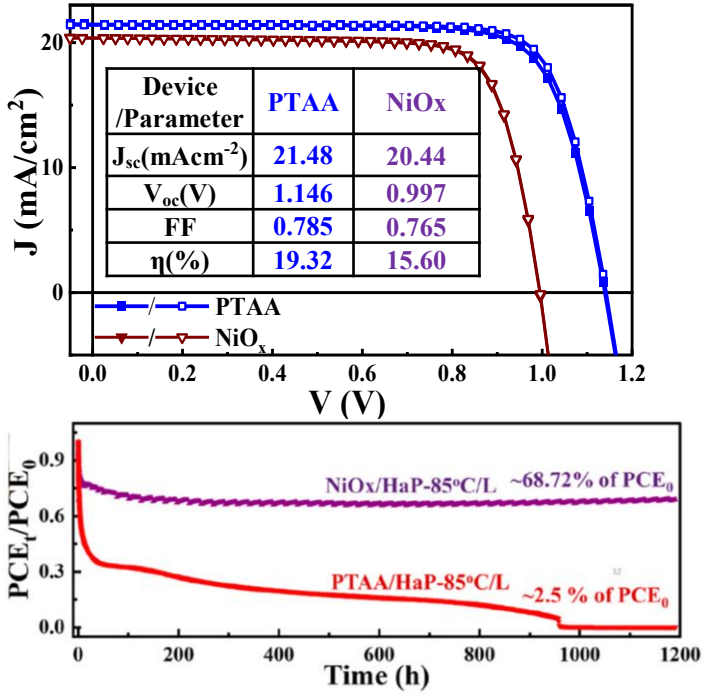


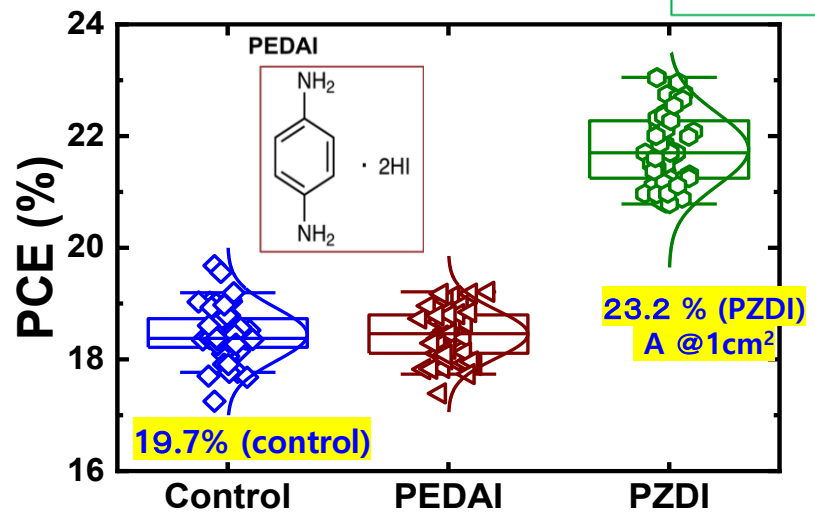
Khadka et al., Sustain. Energy Fuels, 2017, 1, 755



HTL/ETL- engineering – MA-free PSCs







- Khadka et al. ACS Appl. Energy Mater. 2021, 4, 10, 11121
- Khadka et al. ACS Appl. Mater. Interfaces, 2018, 11, 7055
- Khadka et al, ACS Appl. Mater. Interfaces, 2018,10, 22074
- Khadka et al., J. Mater. Chem. C, 2018, 6, 162-170

Khadka et al. Adv. Energy Mater. 2022, 12, 2202029

Khadka et al. Nat Commun 15, 882 (2024)

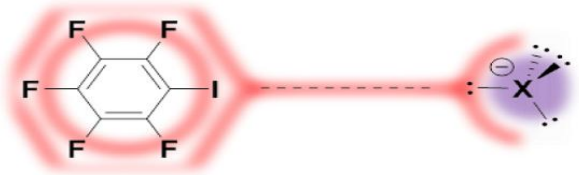
Molecular- passivation: Fluoroarene molecule?

Supramolecular Halogen Bond Passivation of Organic–Inorganic Halide Perovskite Solar Cells

Antonio Abate[†], Michael Saliba[†], Derek J. Hollman[†], Samuel D. Stranks[†], Konrad Wojciechowski[†], Roberto Avolio[‡], Giulia Grancini[§], Annamaria Petrozza[§], and Henry J. Snaith[*†]

Nano Lett. 2014, 14, 6, 3247–3254

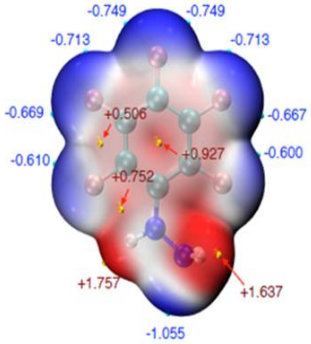
*Gratzel and co-workers, Sci. Adv.*5, 2019, eaaw2543

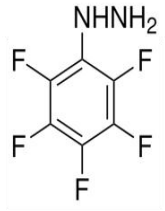


Halogen bond donor (IPFB, Lewis Acid)

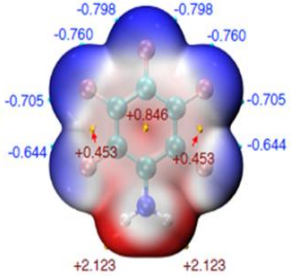
Halogen bond acceptor (X^-, Lewis Base)

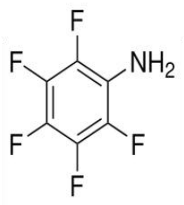
Pentafluorophenylhydrazine



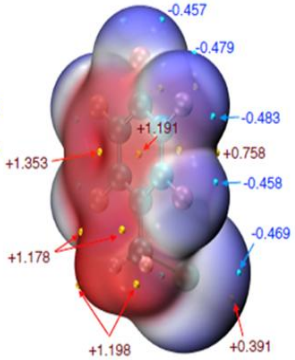


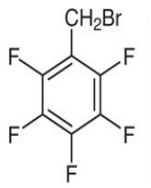
Pentafluoroaniline



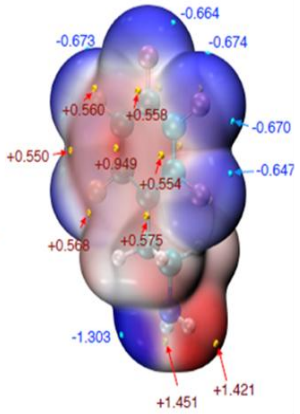


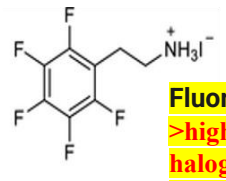
Pentafluorobenzyl bromide



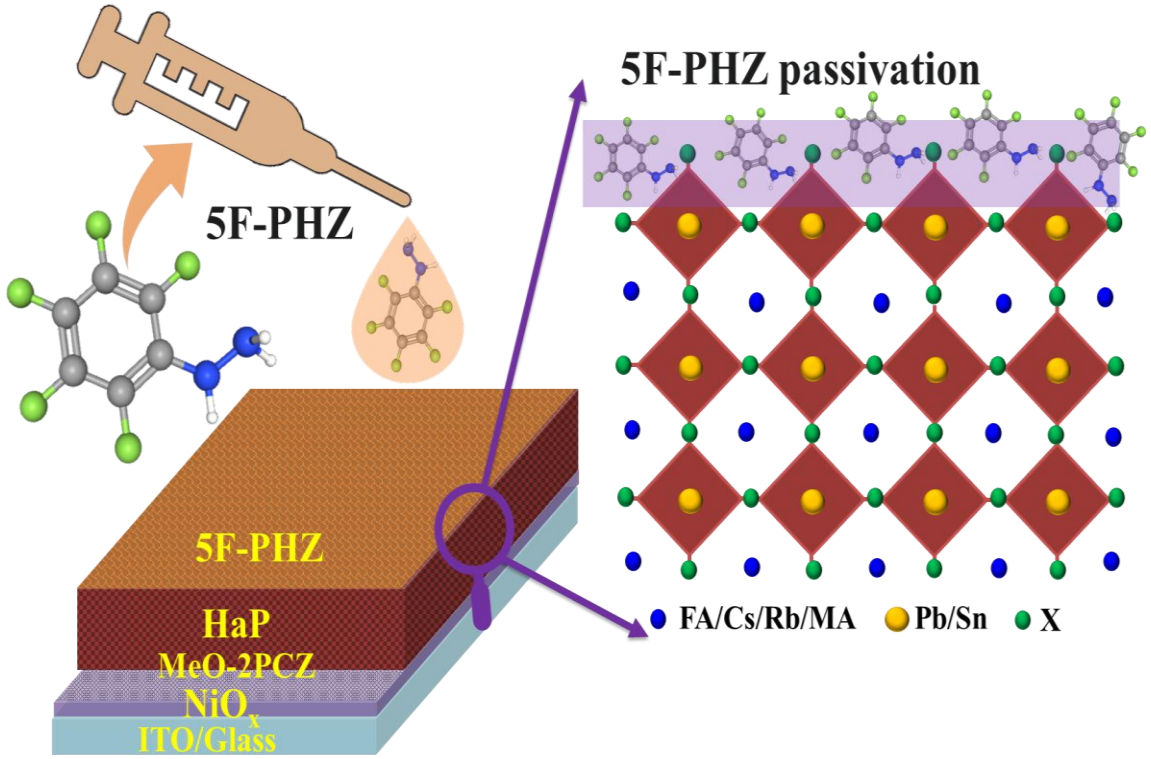


Pentafluorophenylethylammonium



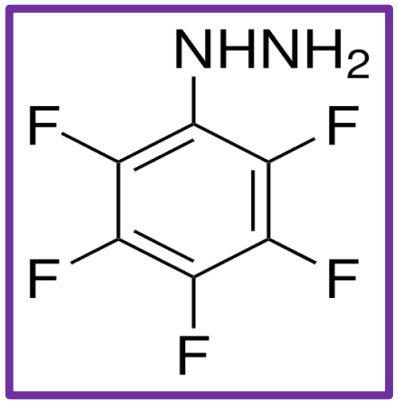


Fluoroarene derivatives
>high electronegativity of fluorine- stronger halogen bonding
>hydrophobic nature

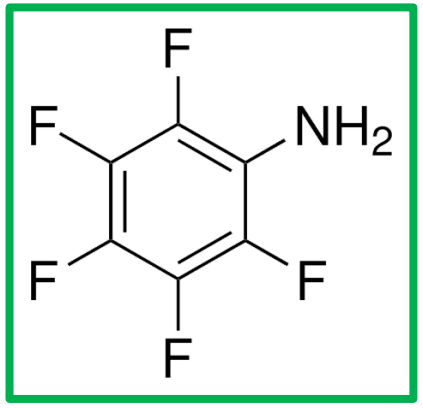


- ✓ Two-step → One-step method; ~270nm → ~500nm
- ✓ $MAPbI_3$ → $FA_{0.84}Cs_{0.12}Rb_{0.04}PbI_3$

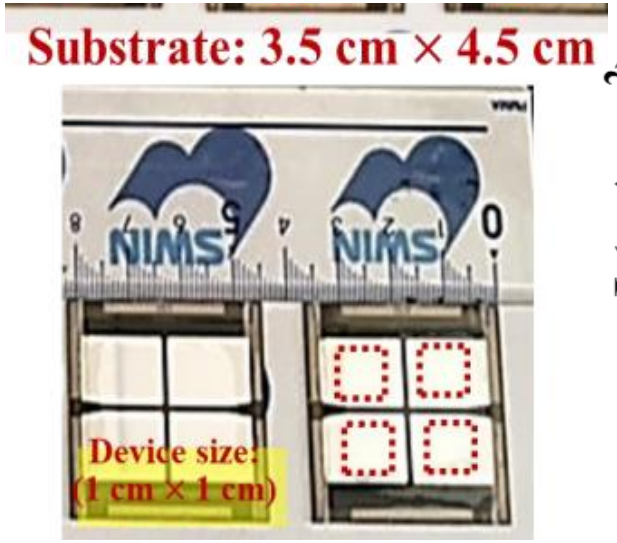
PSCs: Passivation on top/bottom of HP layer

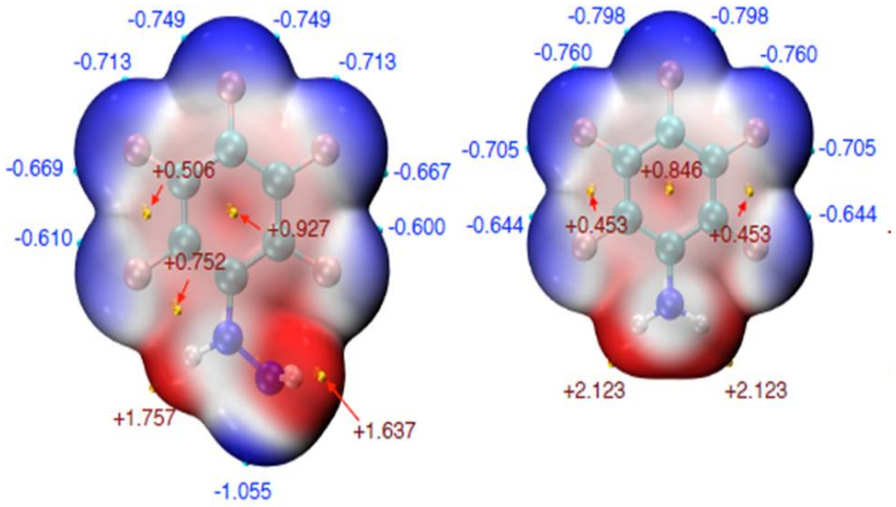


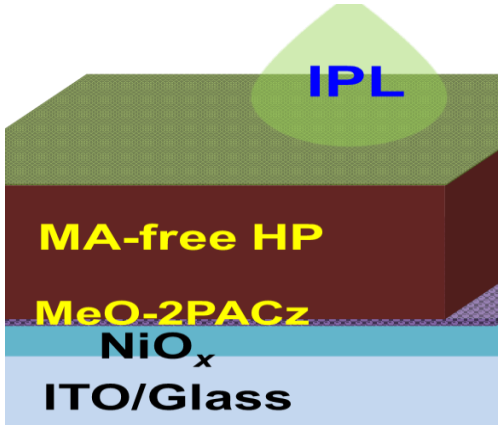
Pentafluorophenylhydrazine (5F-PHZ)

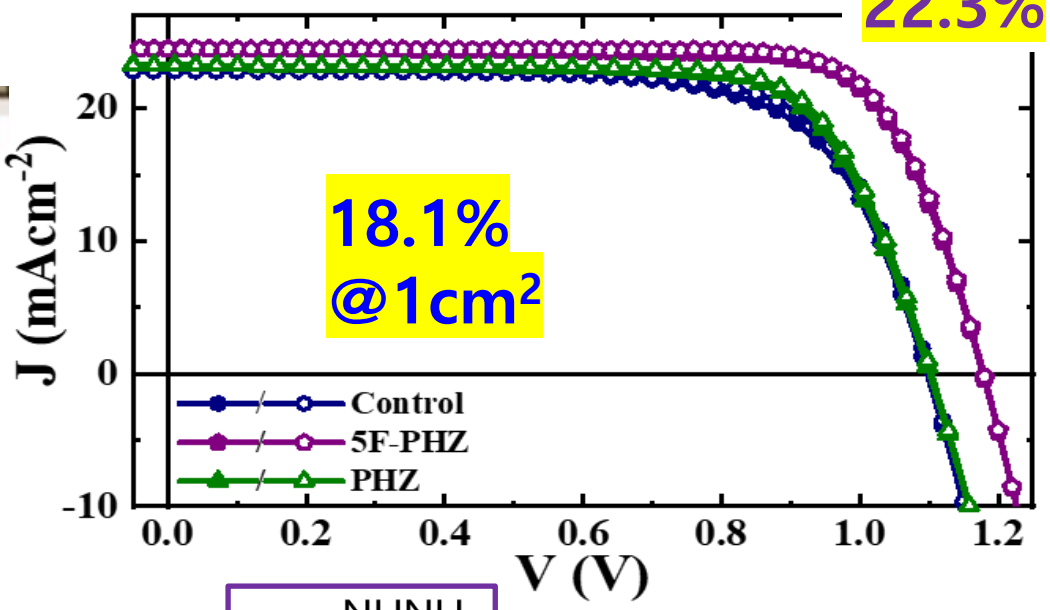


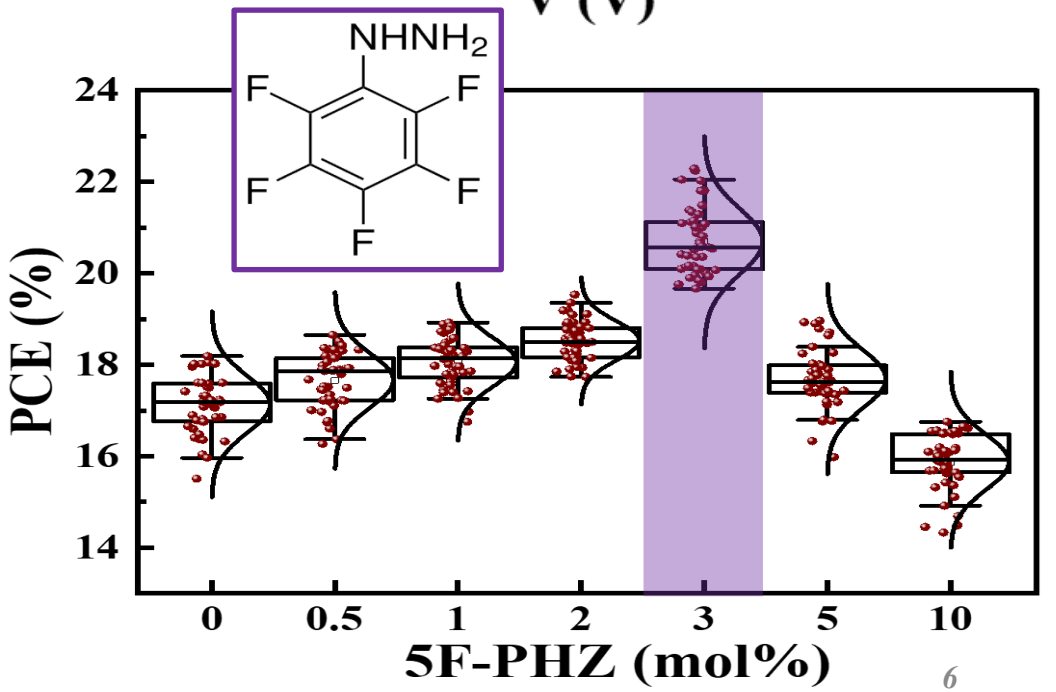
Pentafluoroaniline (PHZ)







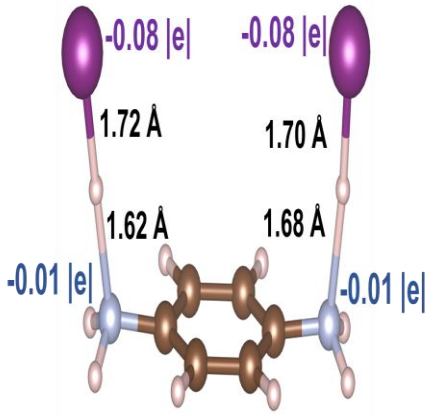




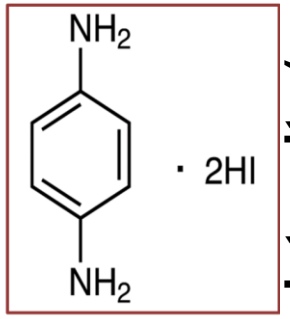
➢ **Khadka et al. Adv. Energy Mater. 2022, 12, 2202029**

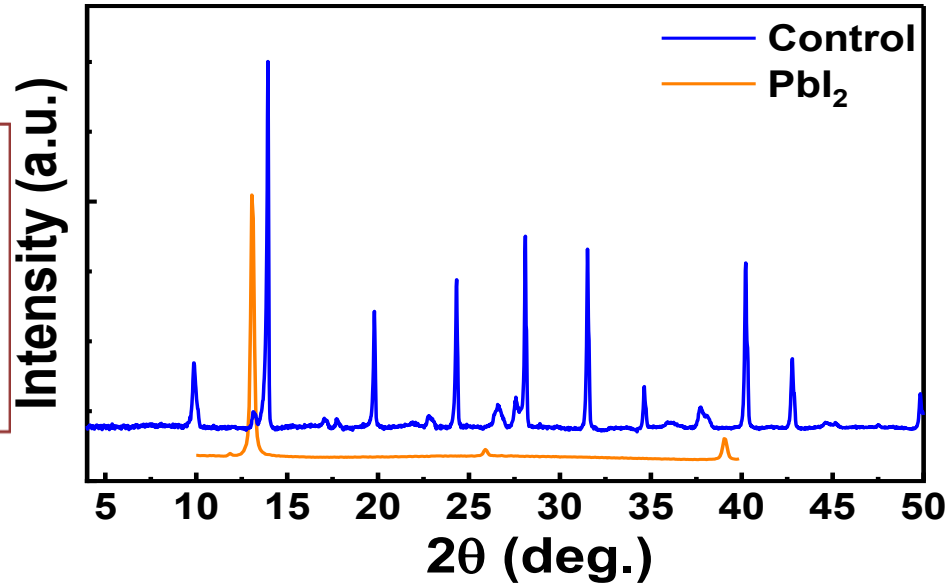
5. PSC: Diamine-based Molecular Passivation?

Aryl amine

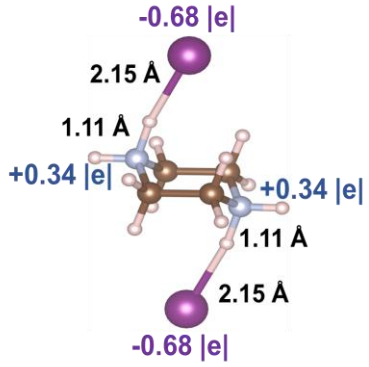


PEDAI

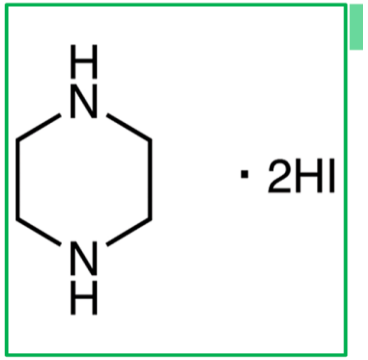


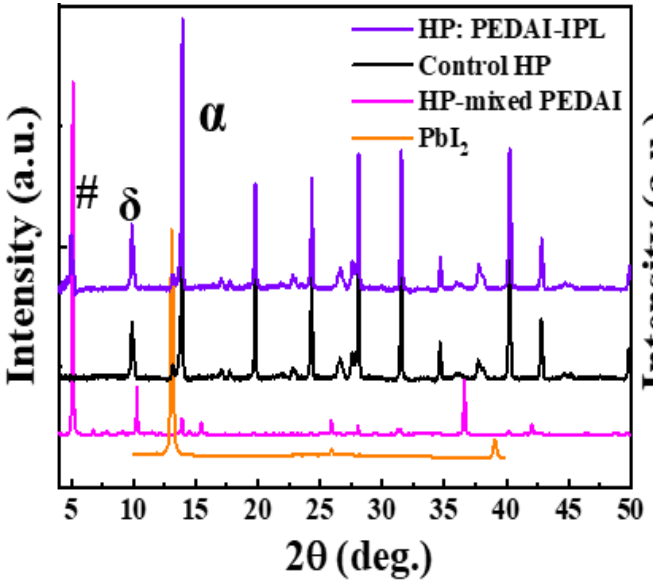


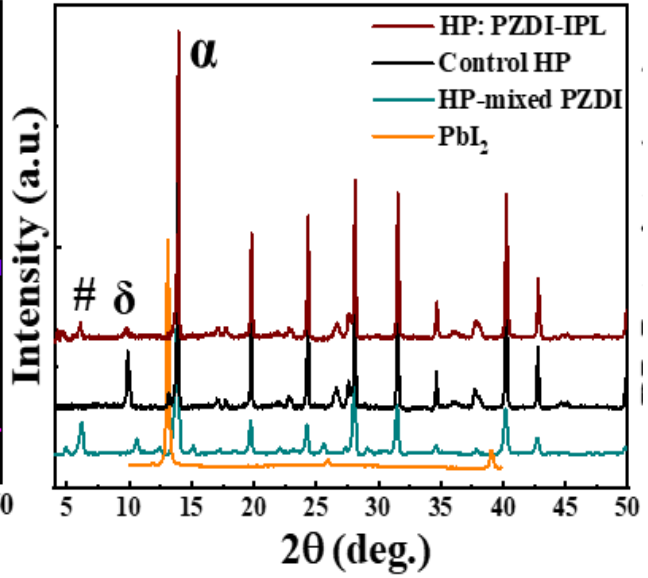
Alkyl amine

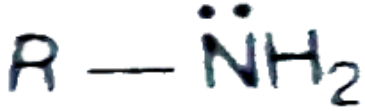


PZDI







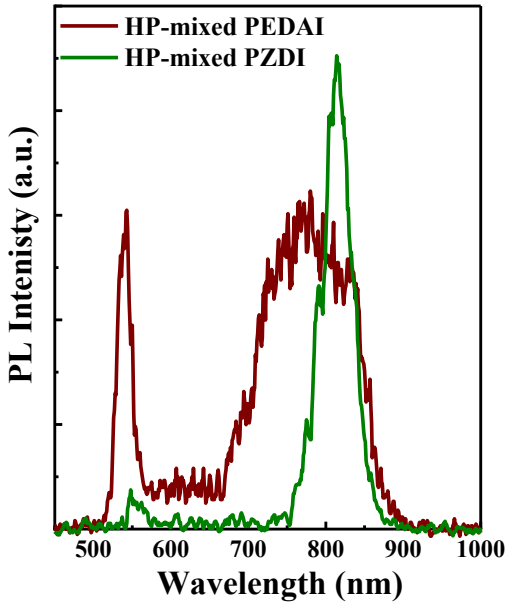


Alkyl amine
(more basic)

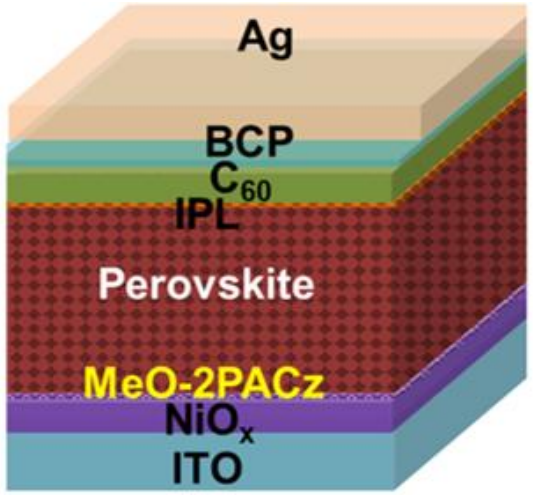


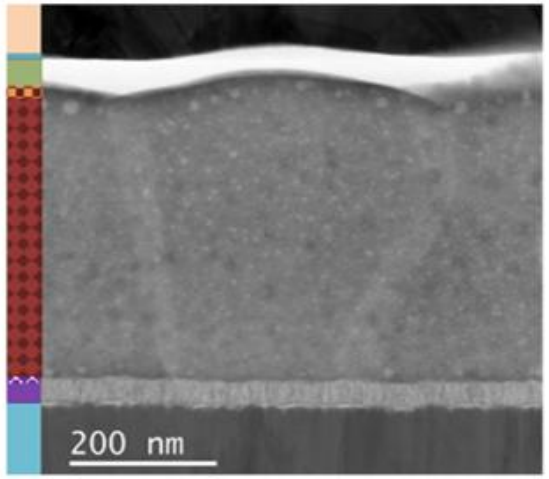
Aryl amine
(less basic)

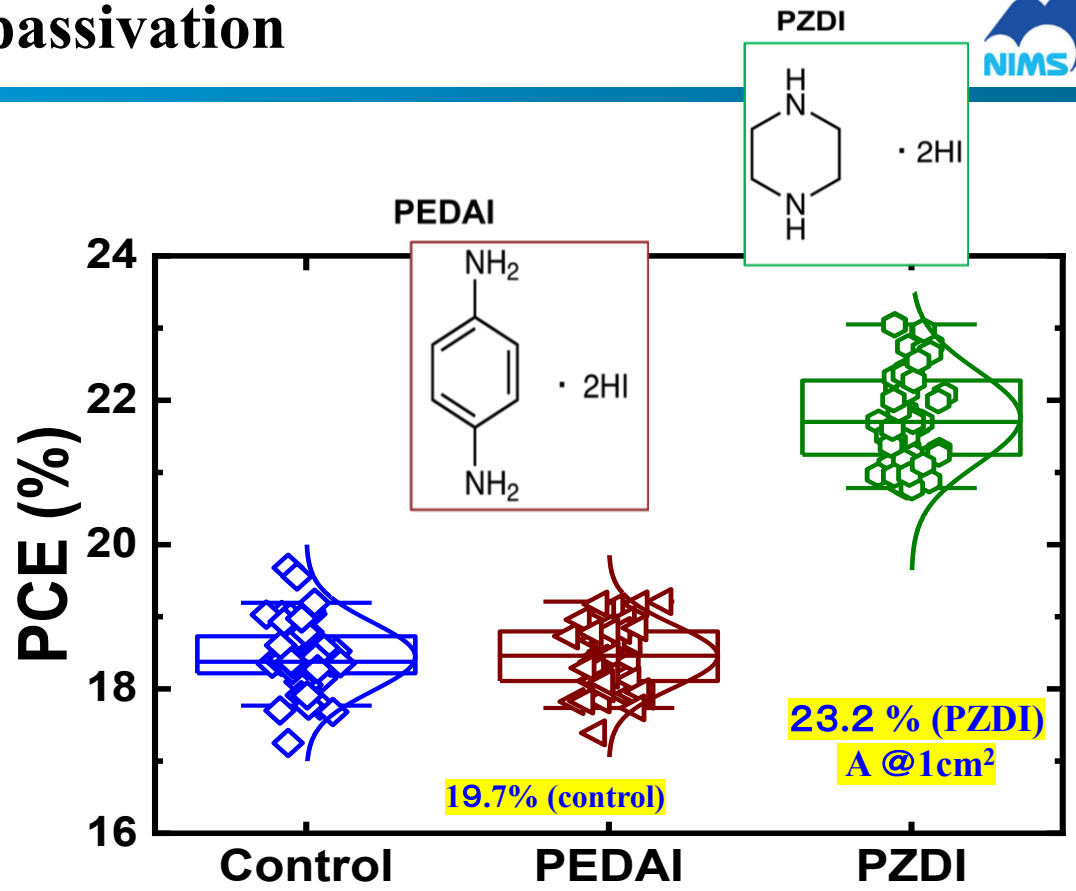
Aryl amine: less basicity
> Delocalization of the lone pair of electrons of the N atom to the benzene ring.

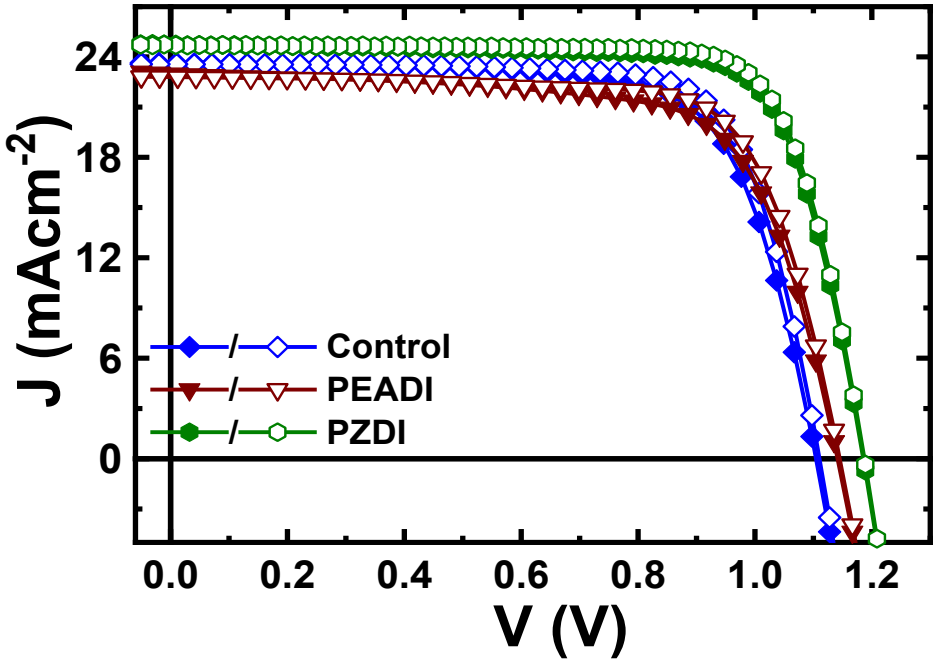


PSC: diamine molecular passivation









Condition	J_{SC} (mAcm^{-2})	V_{OC}(V)	FF	PCE (%)	V_{OC} deficit (V)
Control	23.56	1.112	0.750	19.65	0.402
PEDAI	22.79	1.145	0.736	19.21	0.381
PZDI	24.78	1.188	0.787	23.17	0.327

➢ *Khadka et al. Nat Commun 15, 882 (2024)*

PSCs: Perovskite System Passivation Method

➢ **Passivation method is applicable to different band gap perovskite devices**

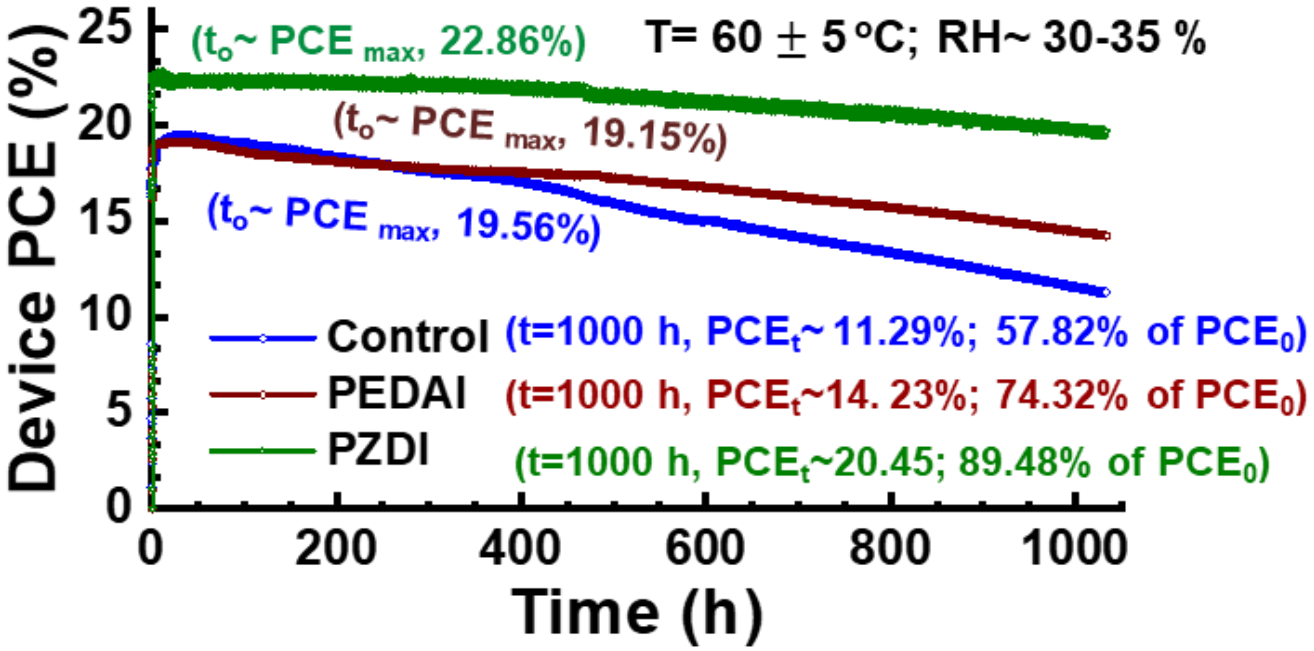
Perovskite composition	Condition	E_g^a eV	Jsc (mAcm^{-2})	*Voc* (V)	*FF*	*PCE* (%)	E_g^a/q -VOC (V)
$Rb_{0.04}Cs_{0.12}FA_{0.84}PbI_3$ (RG-HaP)	Control	1.513	23.56	1.112	0.750	19.65	0.402
	PZDI	1.515	24.78	1.188	0.787	23.17	0.327
RbCsFAPb(I0.64,Br0.36)3 (WG-HaP)	Control	1.722	18.80	1.274	0.732	17.53	0.448
	PZDI	1.725	19.32	1.321	0.761	19.42	0.404
Cs0.05MA0.1FA0.85(Pb0.5,Sn0.5)I3 (NG-HaP) : PEDOT:PSS for HTL	Control	1.263	31.15	0.785	0.730	17.85	0.478
	PZDI	1.265	31.46	0.860	0.751	20.32	0.405

✓ Highly efficient semi-transparent devices with wide-bandgap perovskite devices (V_{OC}-1.45V, PCE>18%)

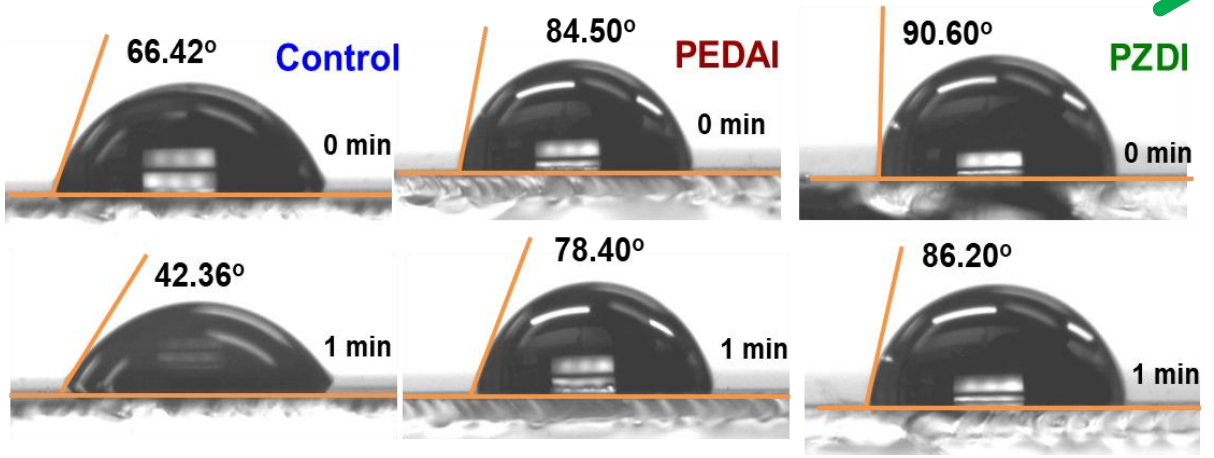
✓ Possibility achieve all-perovskite tandem devices with wide- and narrow-band gap devices

PSC- surface passivation: device stability/ degradation

PCE>20% after 1000 h

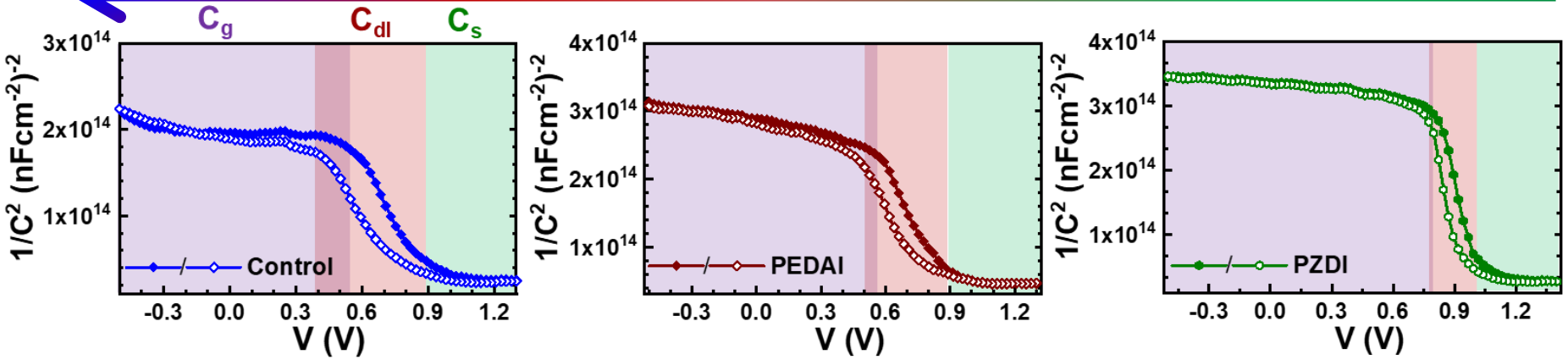


Moisture tolerance



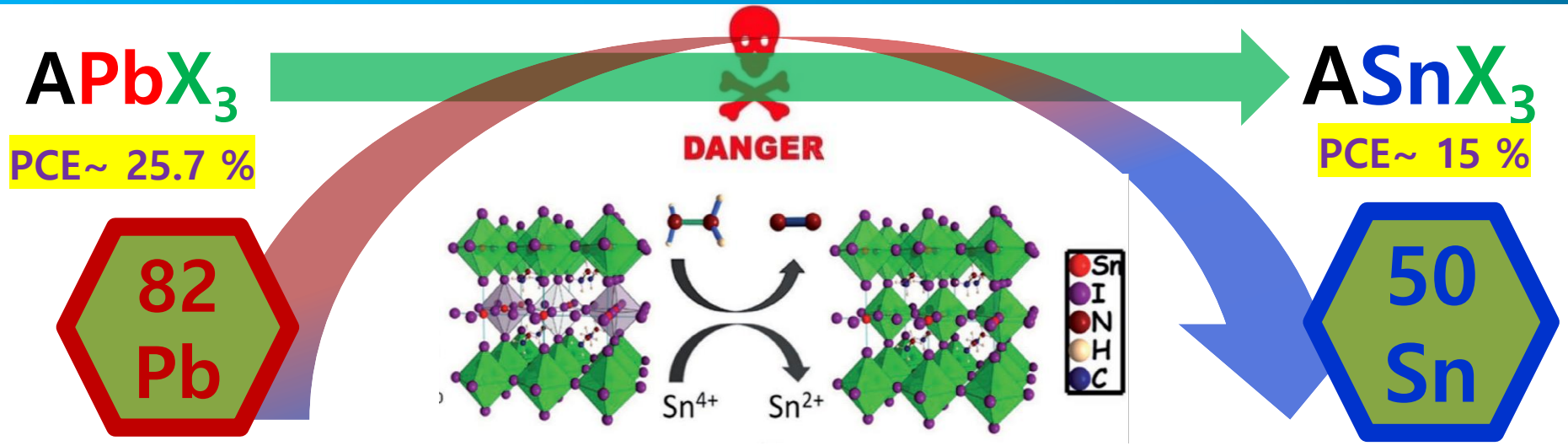
➢ **Mott-Schottky (*M-S*) plot**

Interfacial deterioration



➢ **Overlapped shaded region s-** lagging of the *M-S* curve- interfacial deterioration.

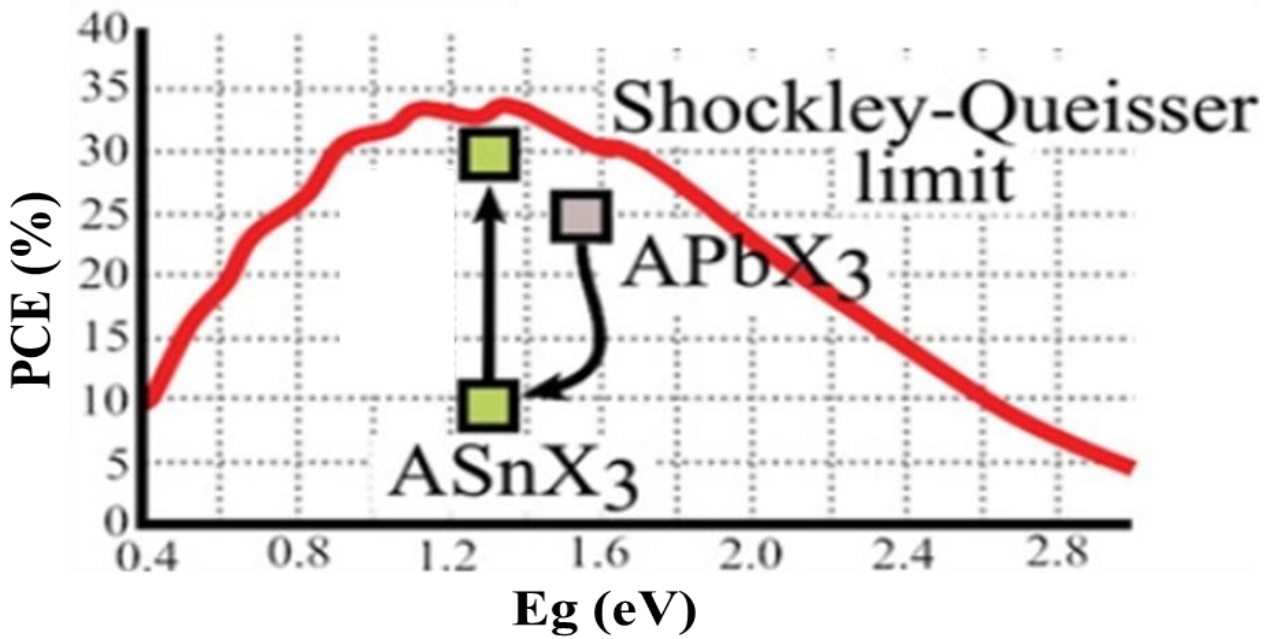
Sn-based materials for solar cells



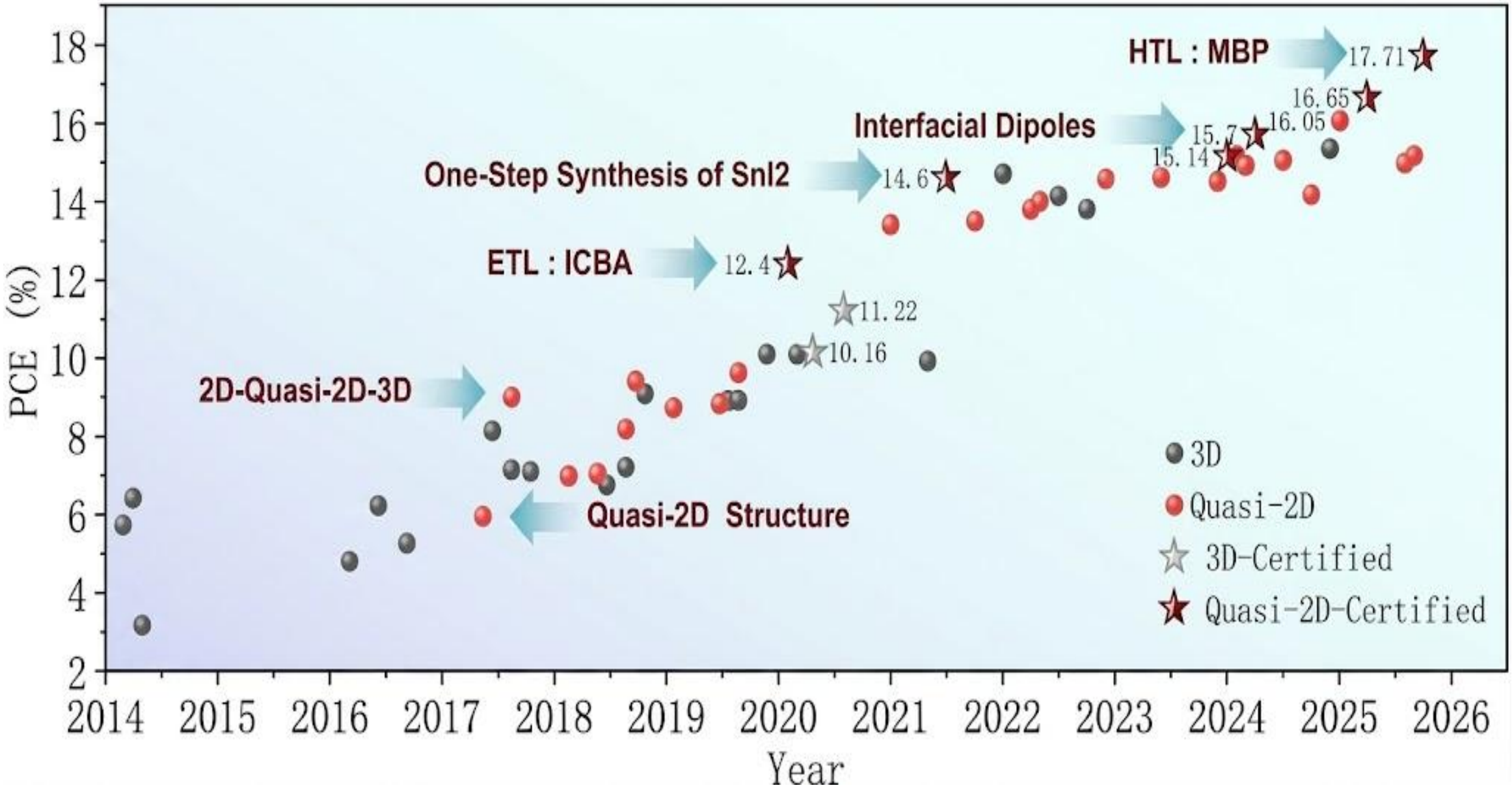
Toxic, stable Pb^{2+}

Oxidation issue

Non-toxic, less stable Sn^{2+}

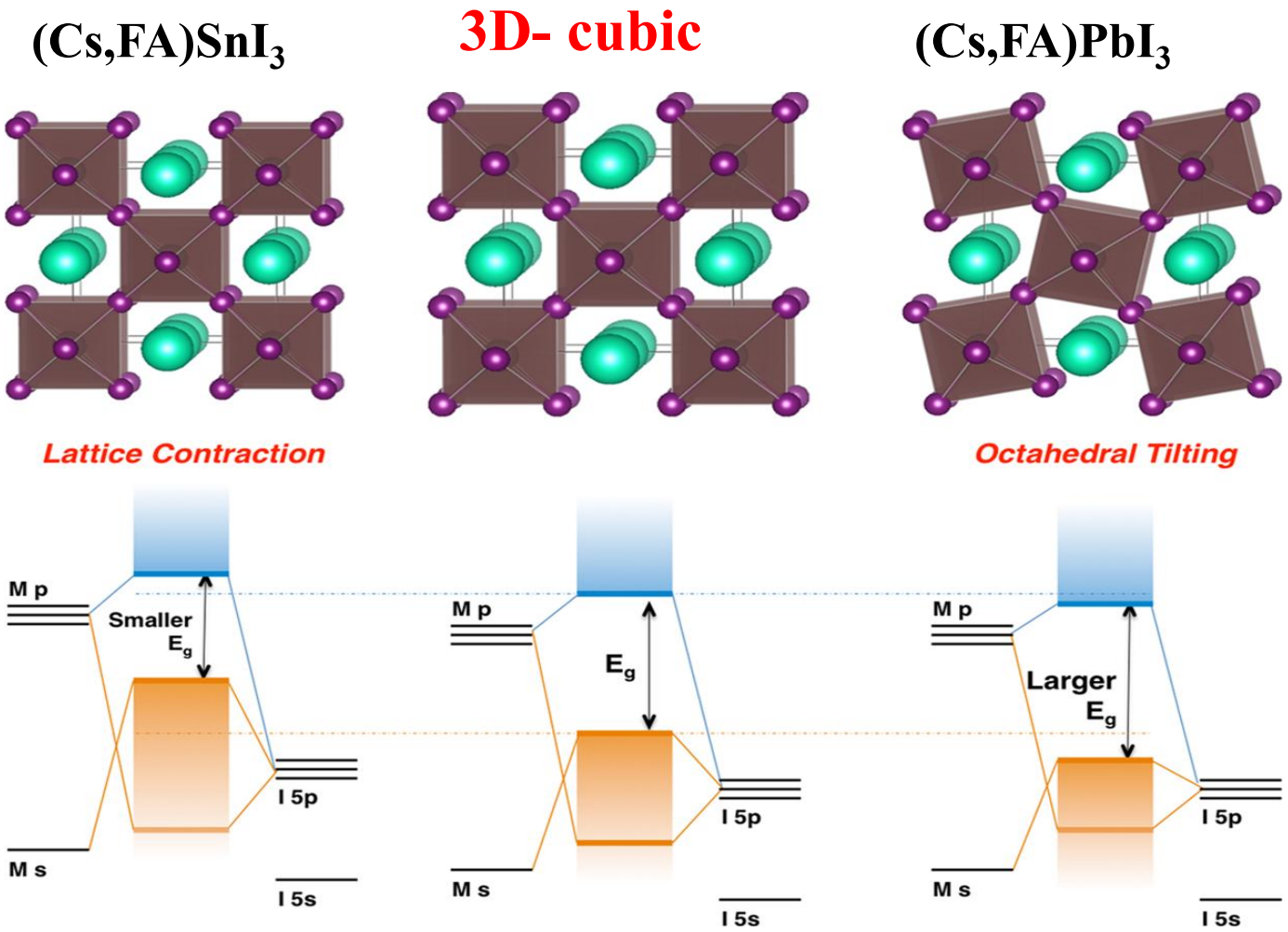


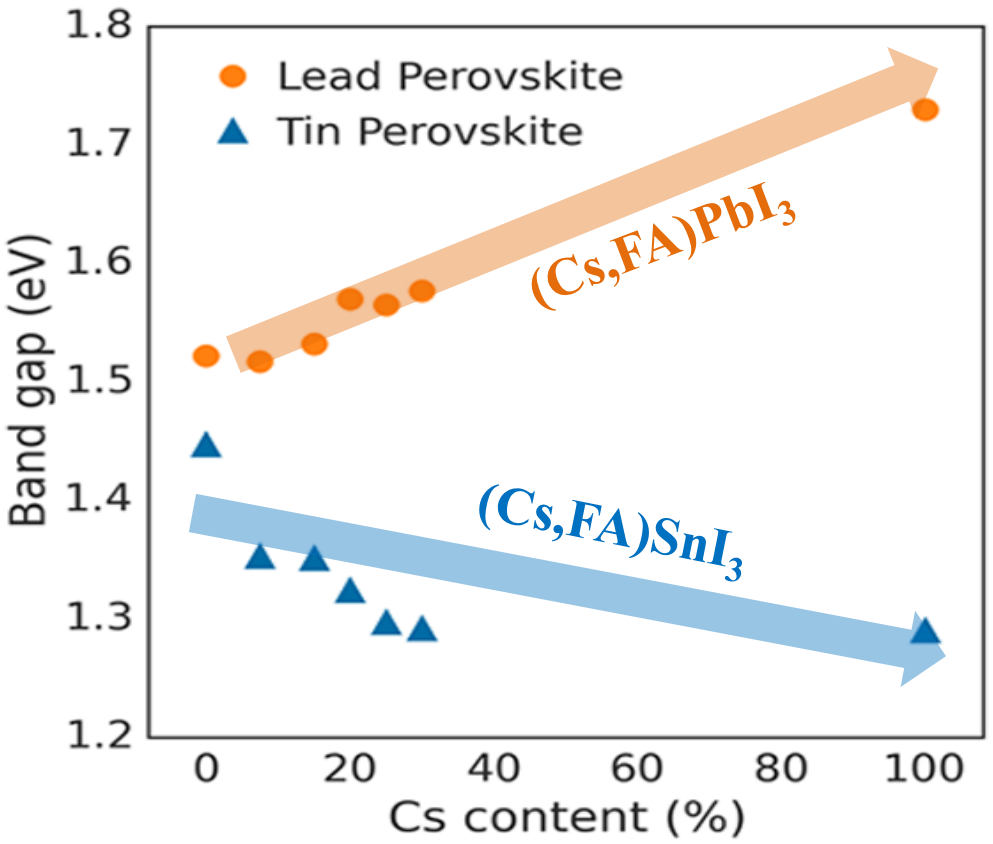
Strategies for Tin Perovskite Solar



- *ACS Energy Lett.* 2026, 11, 180

Effect of A-site in halide perovskite



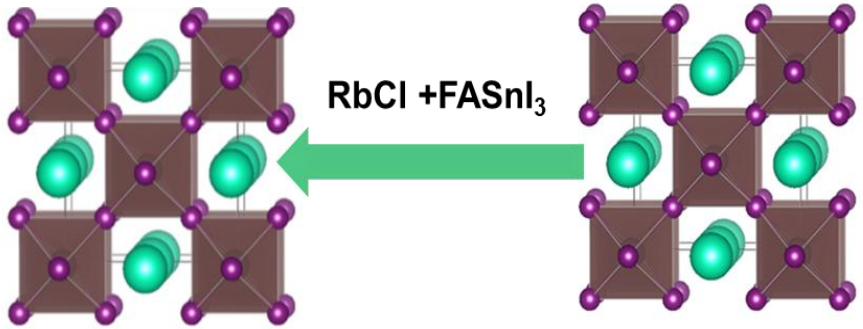


- **Structural regulation- improve the structural stability of $FASnI_3$ perovskite.**

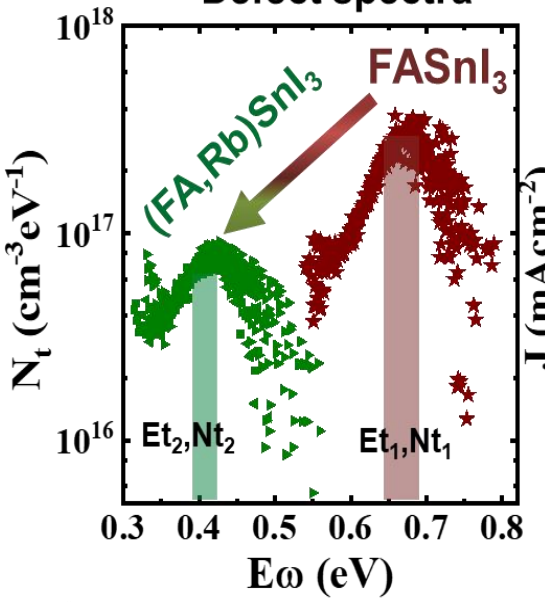
➢ McGehee and co-workers, J. Am. Chem. Soc. 2017, 139, 11117−11124

Our reports on Sn-PSCs

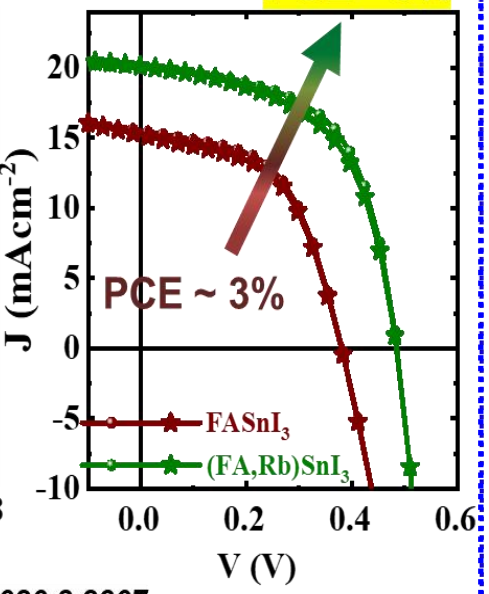
Structural regulation



Defect spectra

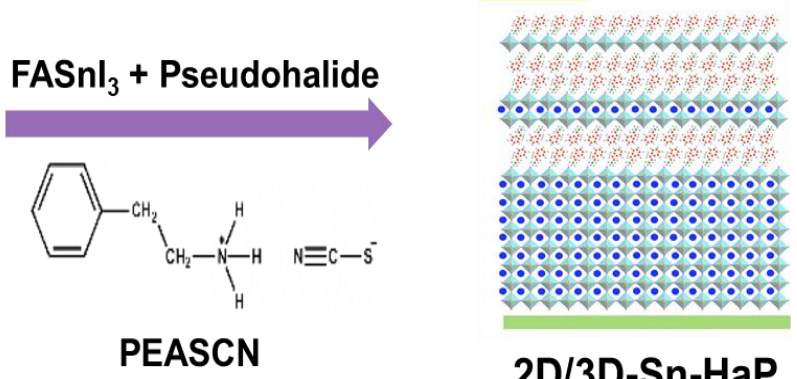


PCE~ 6%

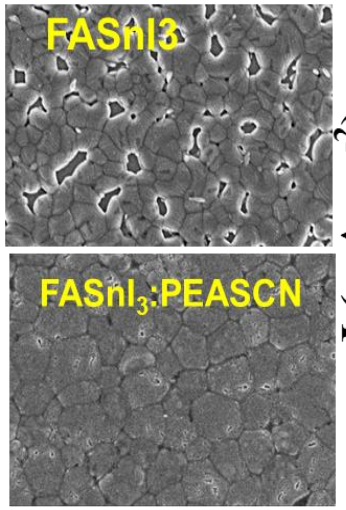


Khadka* et al. Mater. Chem. C,2020,8,2307

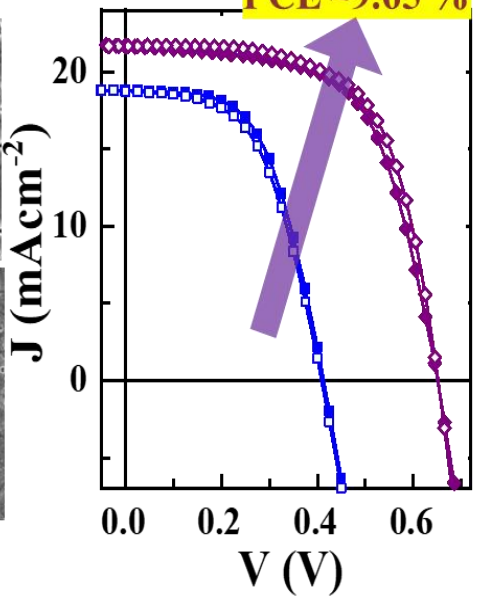
Pseudohalide additive effect



Film growth

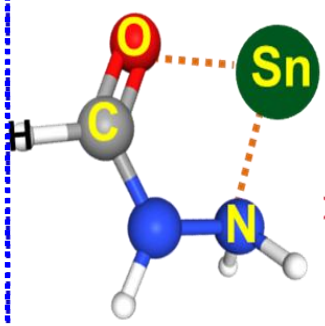


PCE ~9.65 %



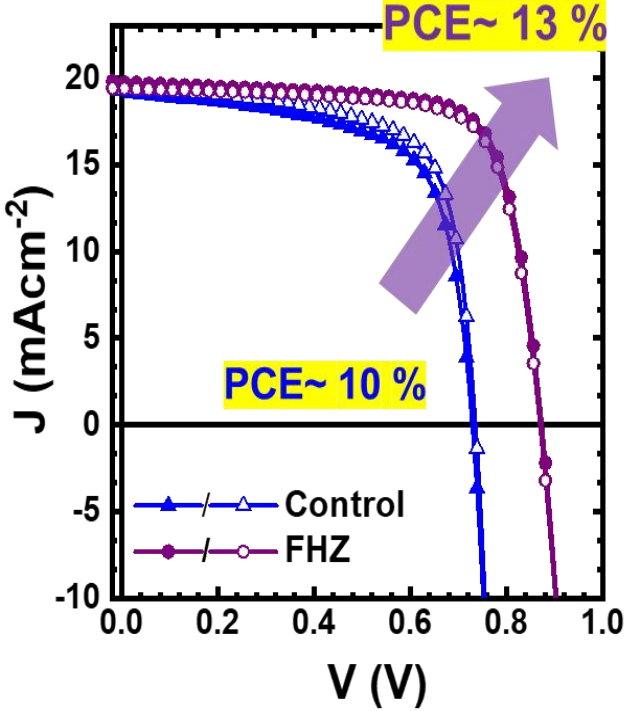
Khadka* et al. *ACS Appl. Energy Mater. 2021, 4, 12819*

Multi-functional ligand-additive



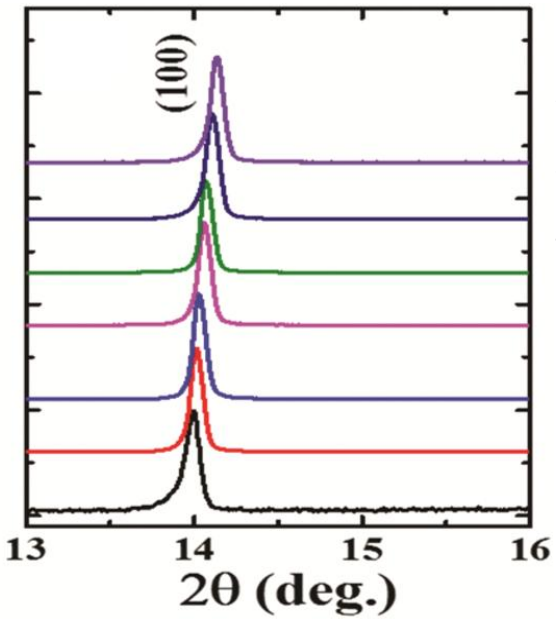
Formohydrazide (FHZ)
>Bidentate ligand

PCE~ 13 %

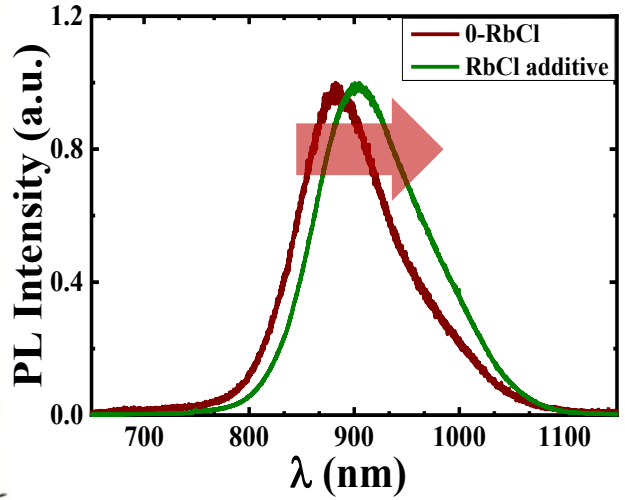


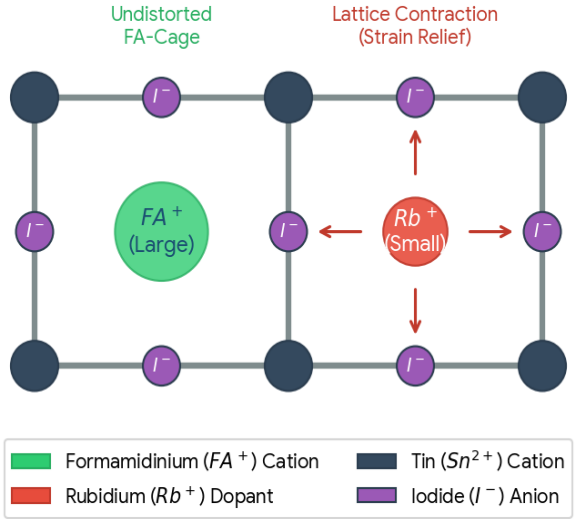
Khadka* et al. *Chem. Mater. 2023,35, 4250*

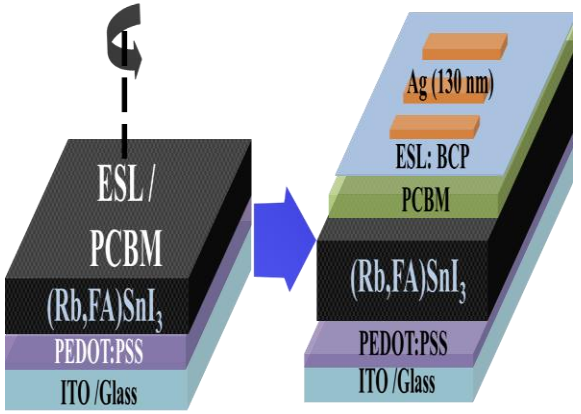
Structural regulation: Rb incorporation: A-site



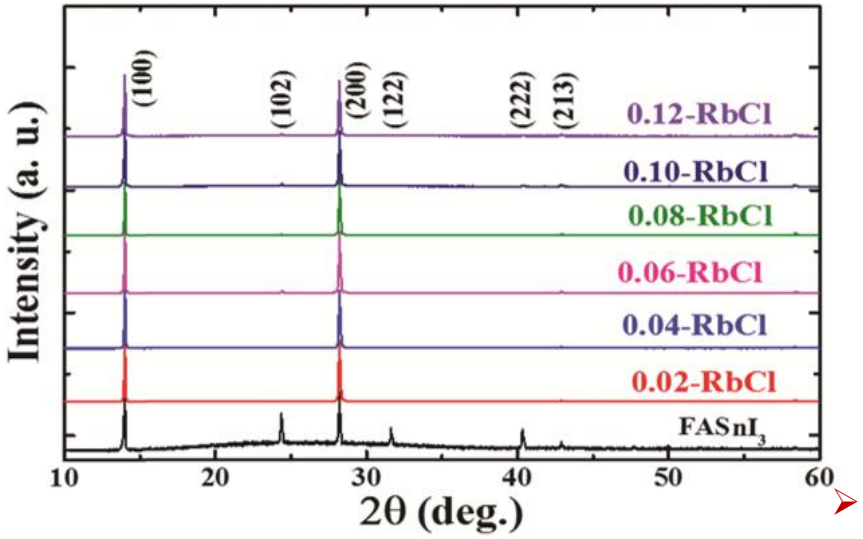
Rb in $FASnI_3$; E_g-red shifting

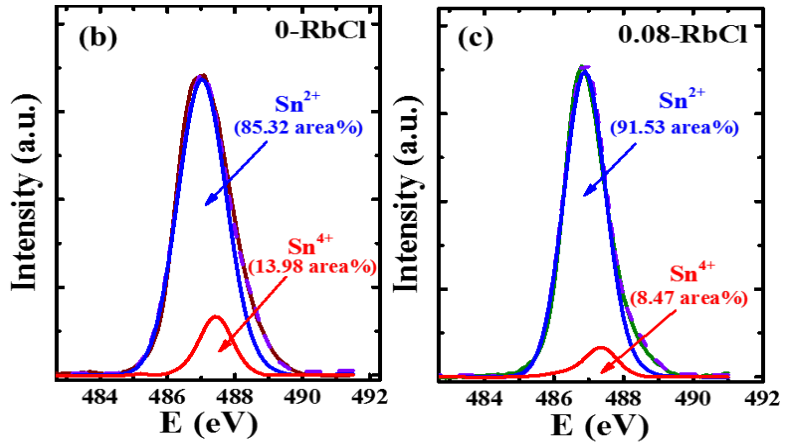


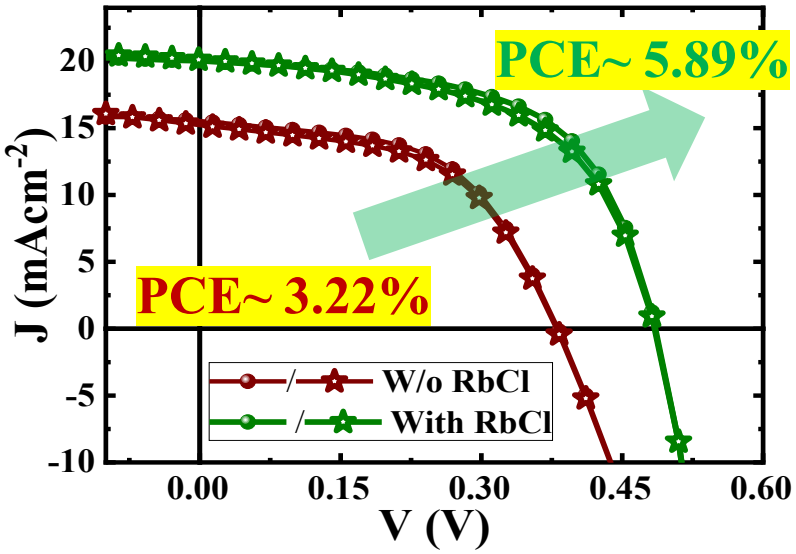




- ***Suppression of oxidation:***
- ***Attenuation of recombination***

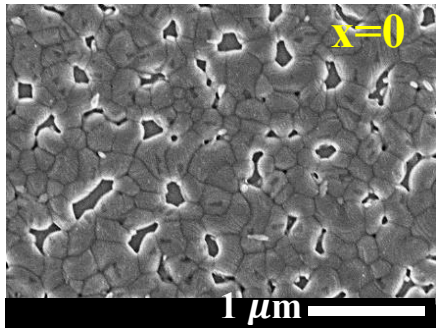


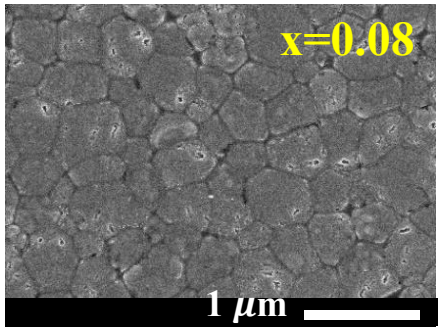


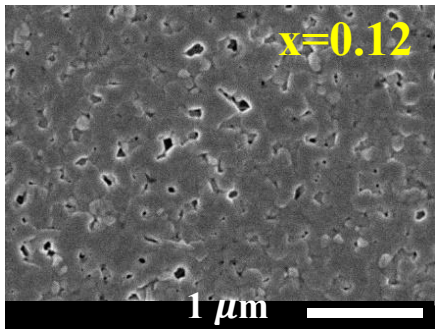


➢ *Khadka et al. J. Mater. Chem. C*, 2020,**8**, 2307-2313

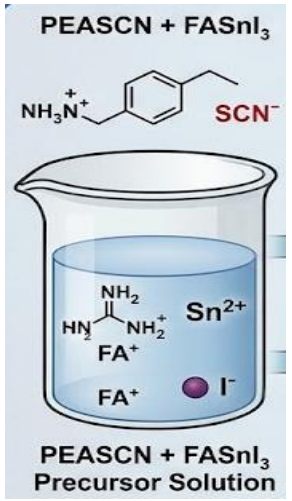
Impacts of $FASnI_3$+PEASCN: Film growth properties



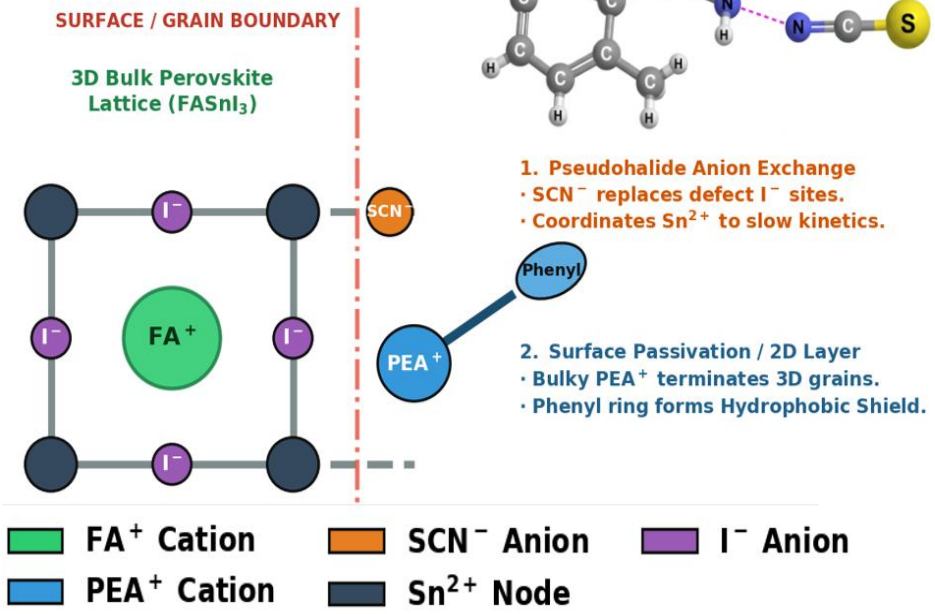


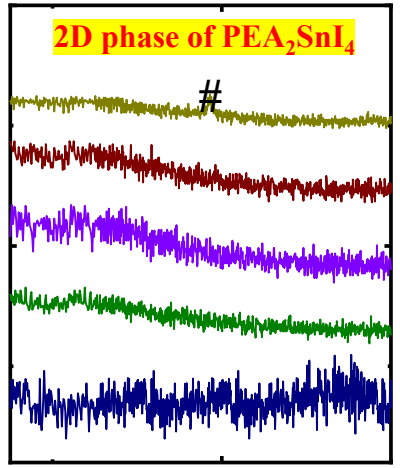


>PEASCN additive: better coverage and compact film



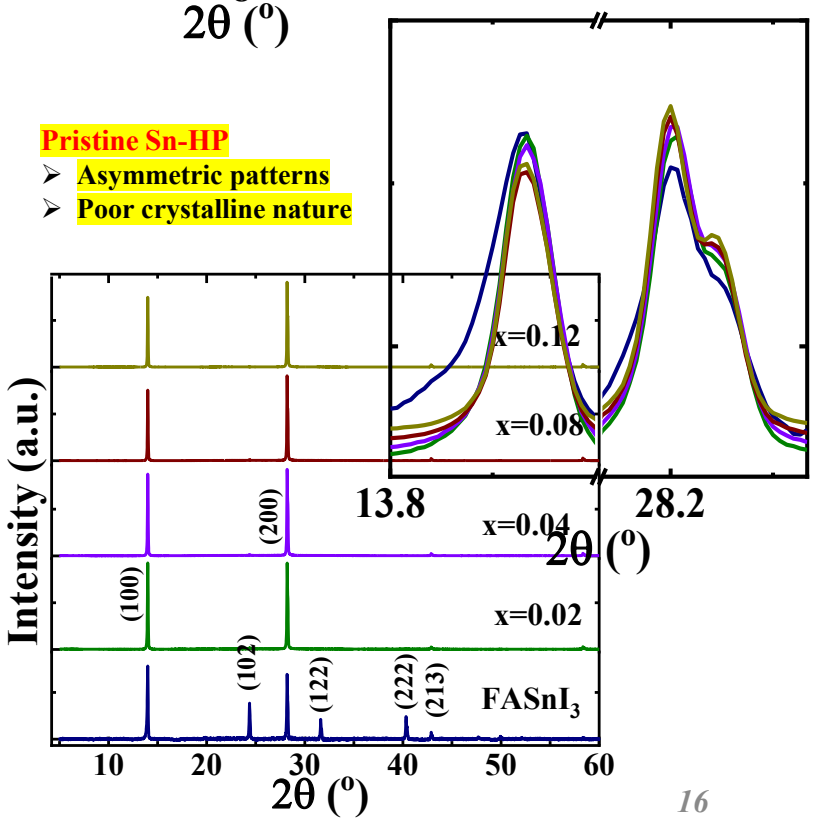
$(FA_{1-x}PEASCN_x)SnI_3$;
(PEASCN additive in latti

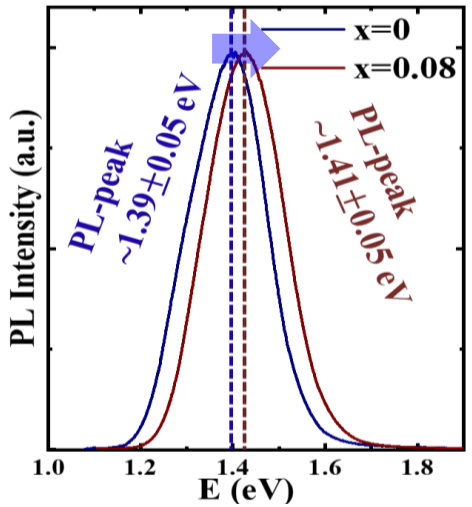




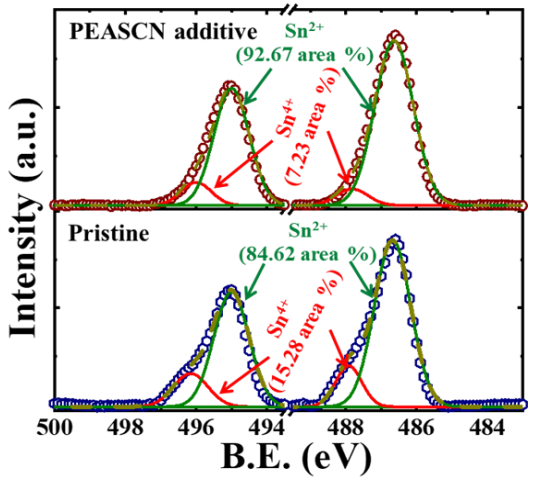
-Slight incline in 2θ,
-Incorporation of SCN^- into the crystal lattice.

Pristine Sn-HP
- Asymmetric patterns
- Poor crystalline nature



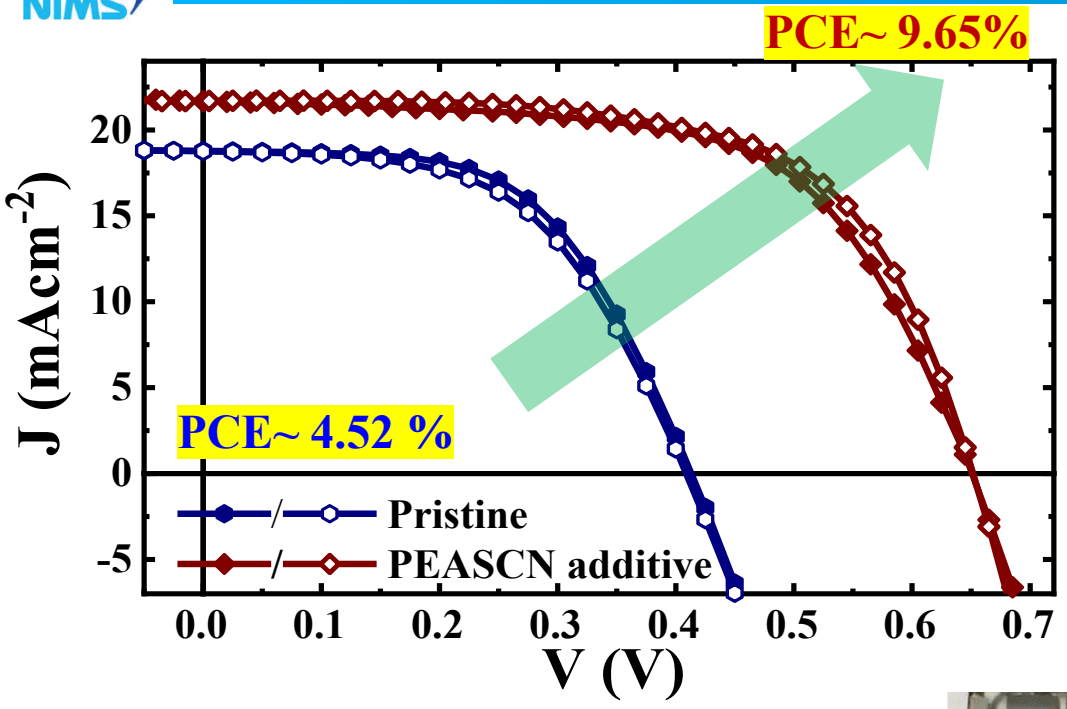


Suppression of oxidation

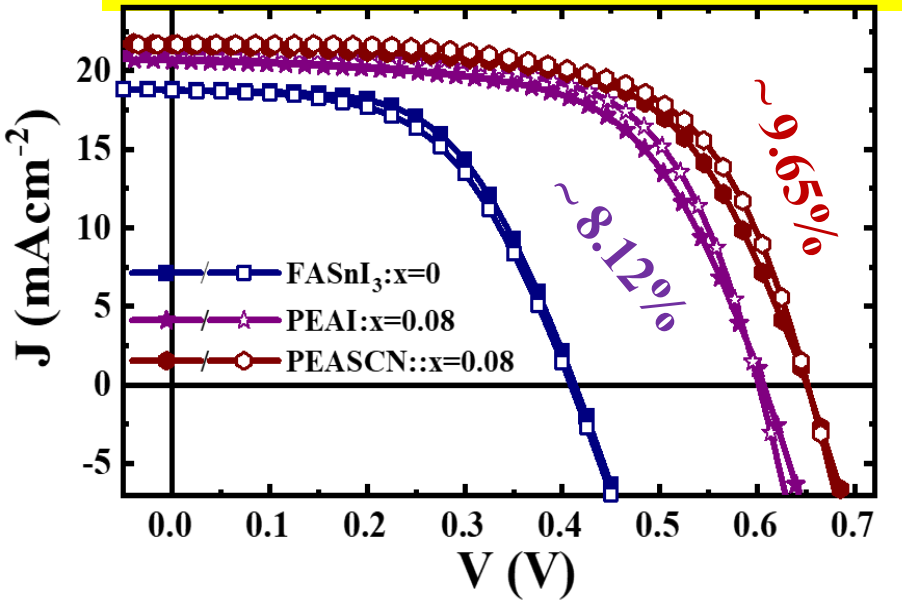


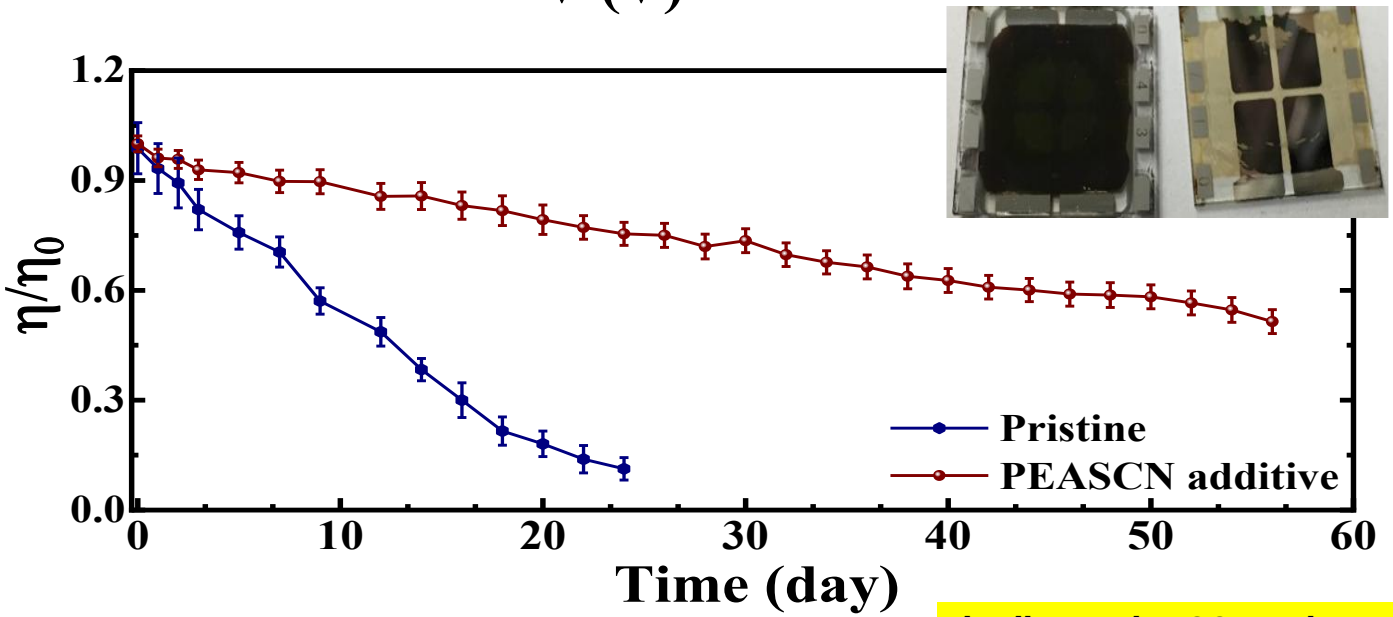
Khadka et al. ACS Appl. Energy Mater. 2021, 4,12819

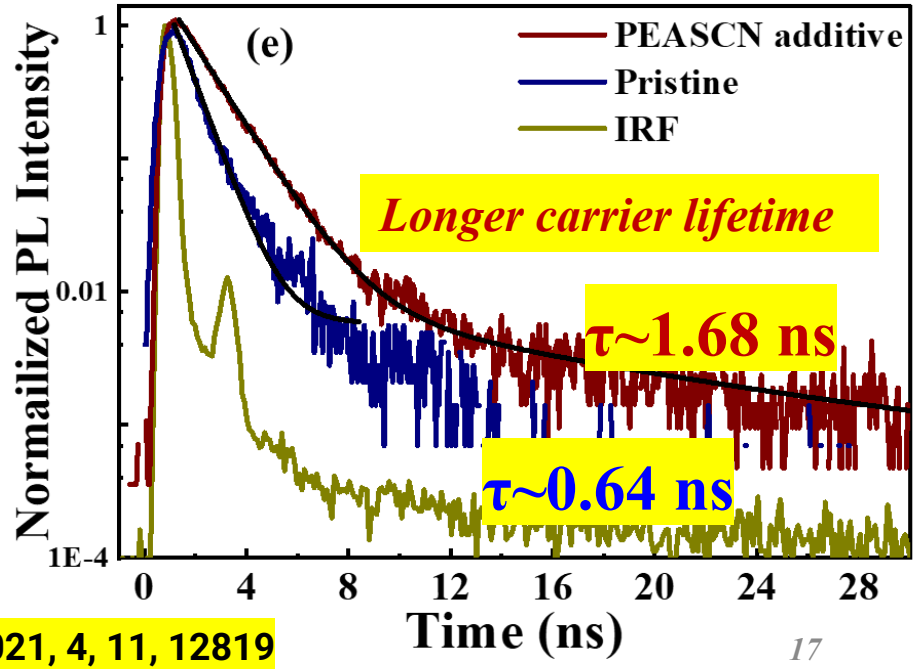
Impacts of $FASnI_3$+PEASCN: device performance



To examine the role of SCN^-; pseudohalide





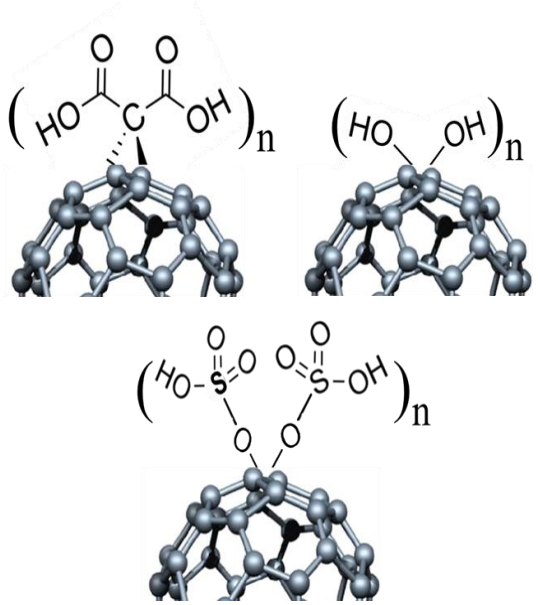


Khadka et al. ACS Appl. Energy Mater. 2021, 4, 11, 12819

Effect of Functional Fullerene-derivatives

Sn-perovskite

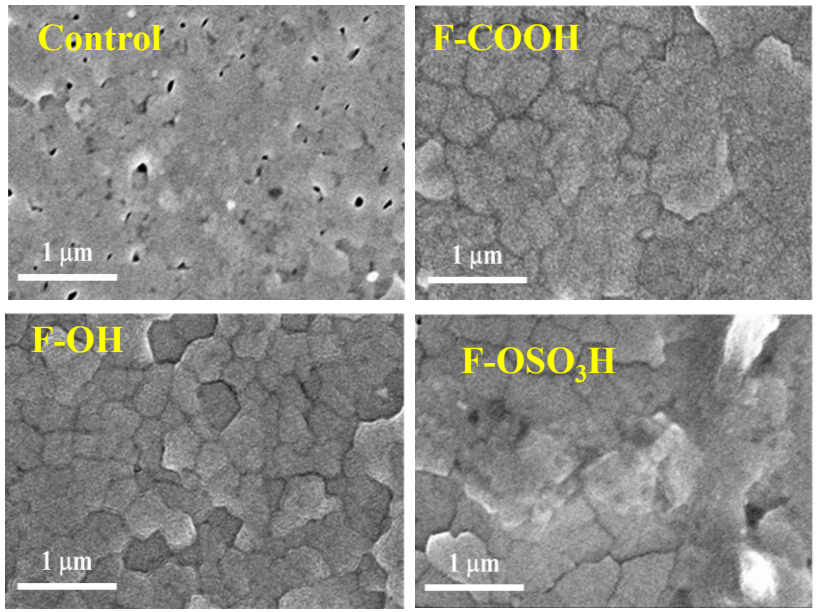
(FA, MA, PEA)SnI_3)

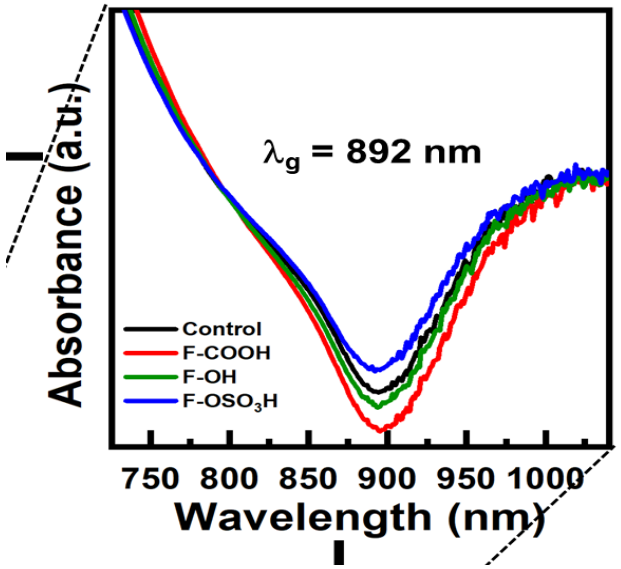


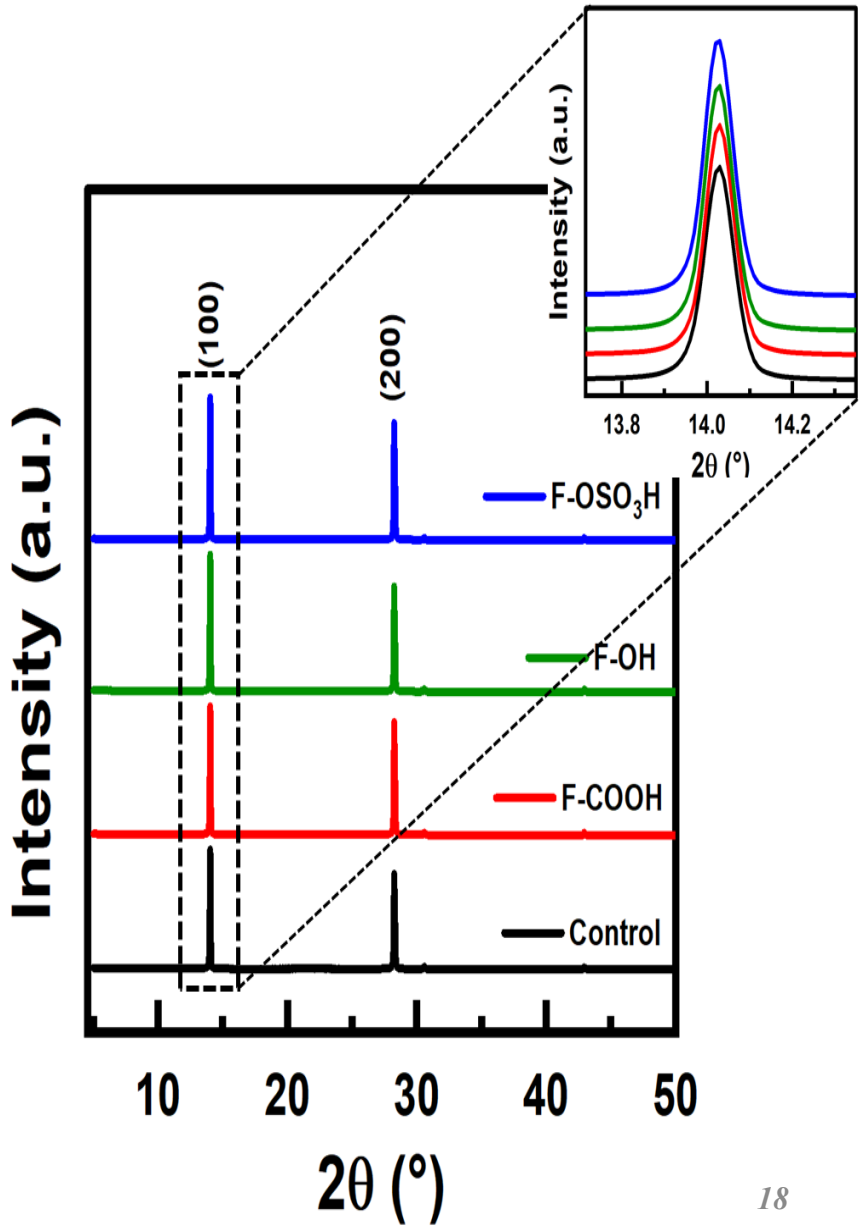


Prof. Takeoka Yuko,
Sophiya University

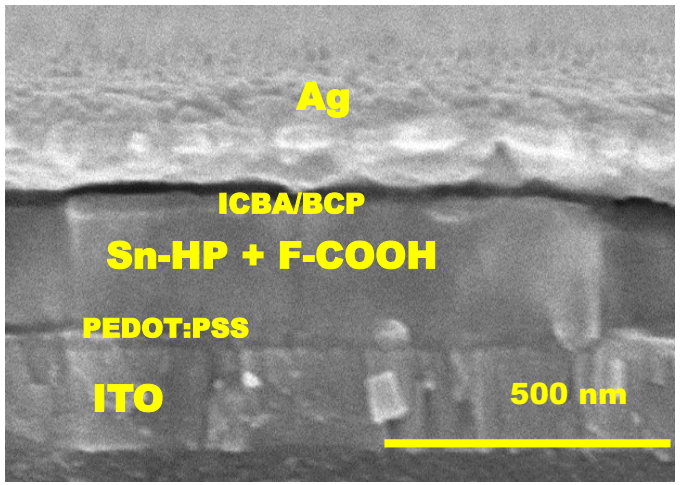
Film- morphology

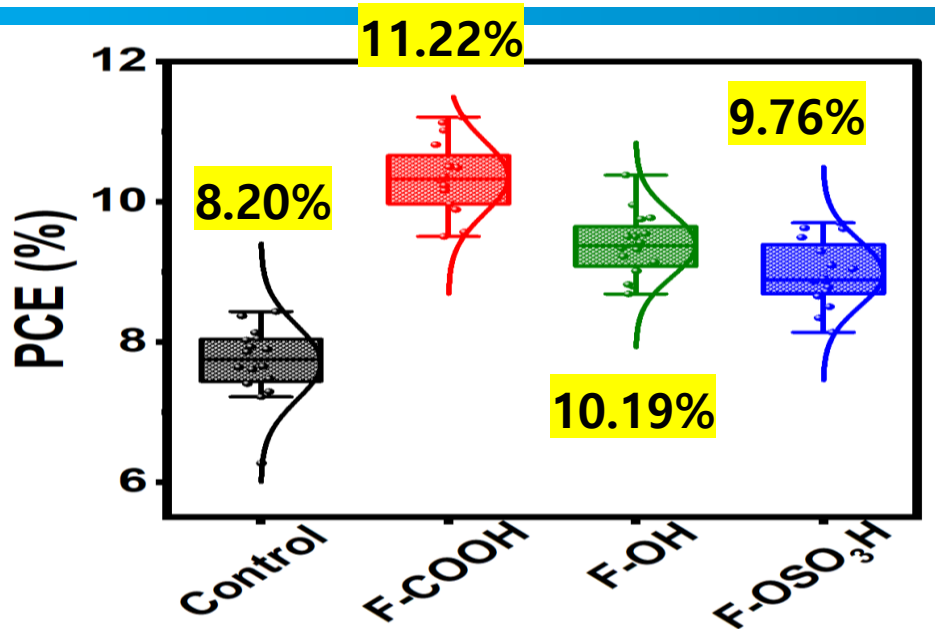


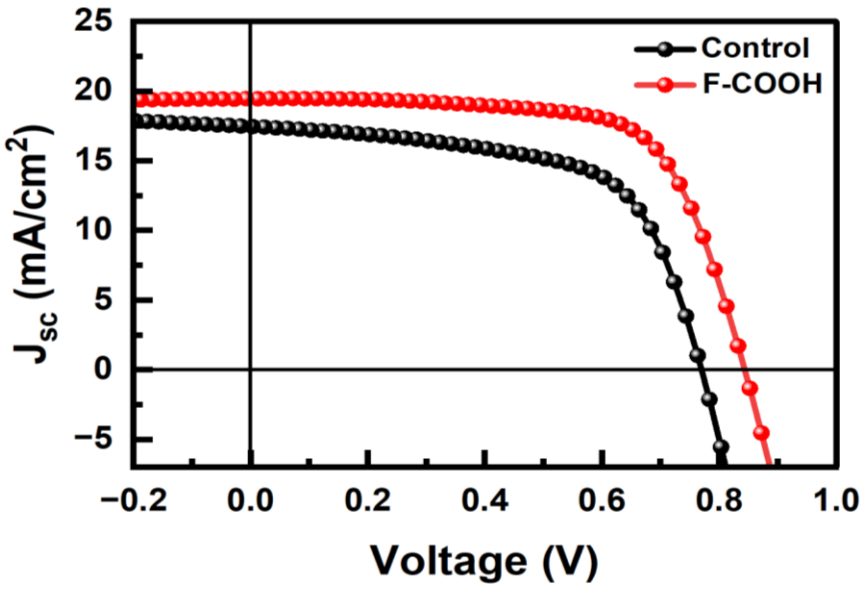


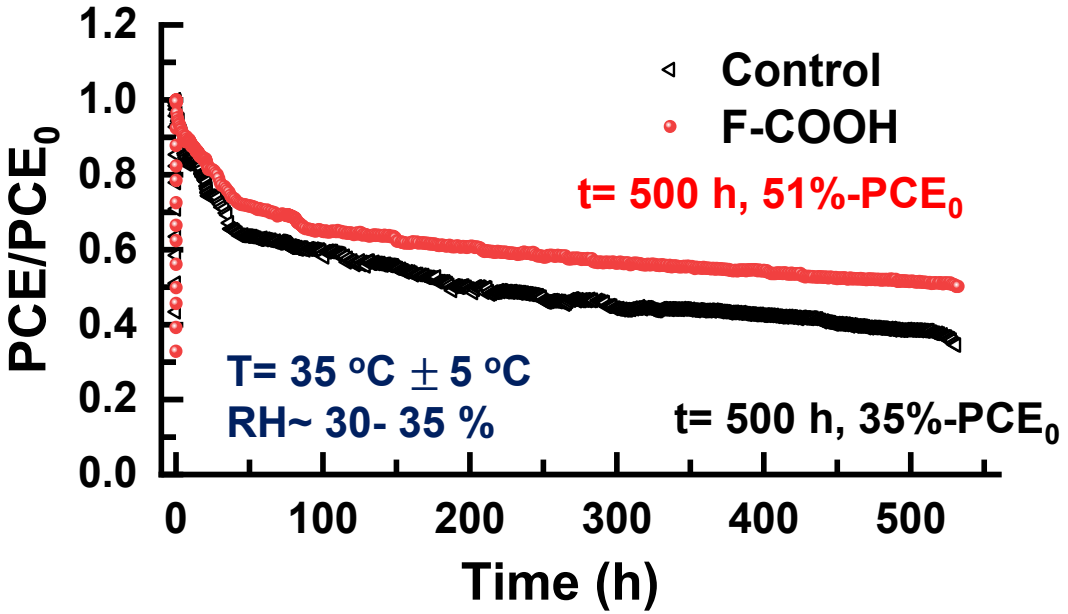


Effect of Fullerene derivatives: Sn-PSCs









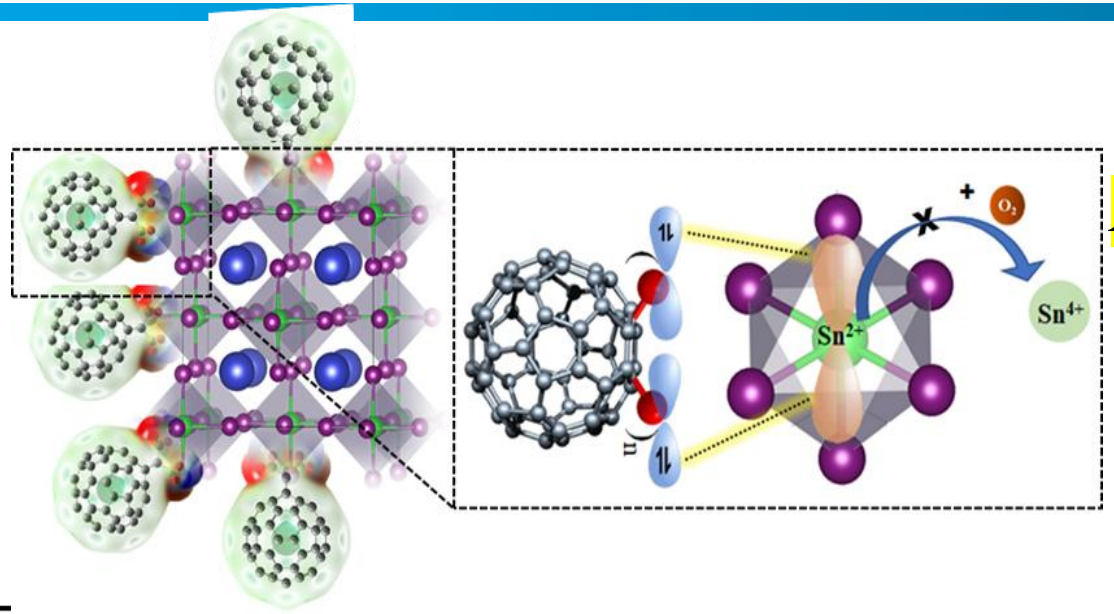
- ✓ J_{SC} -17.44 to 19.31 mA/cm²
- ✓ V_{OC} – 0.769 to 0.841 V
- ✓ FF- 0.613 to 0.691

Khadka and co-workers, J. Mater. Chem. A,2025, 13, 23487

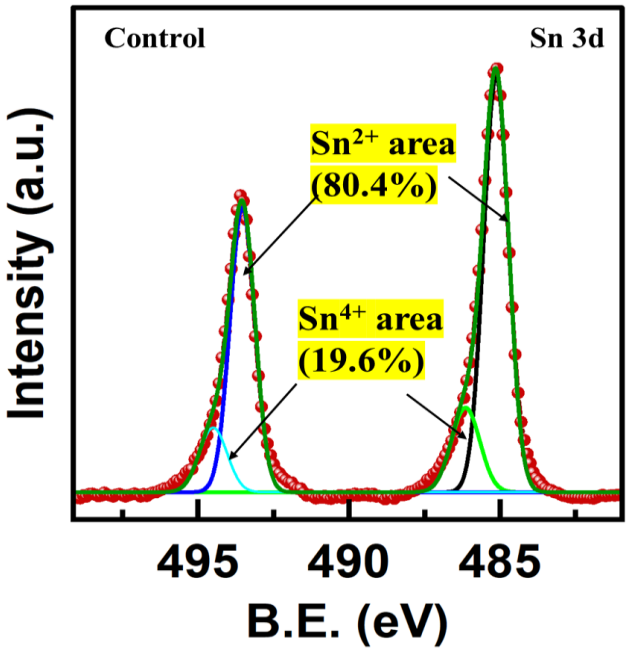
Effect of Fullerene-derivatives – XPS-Surface chemistry

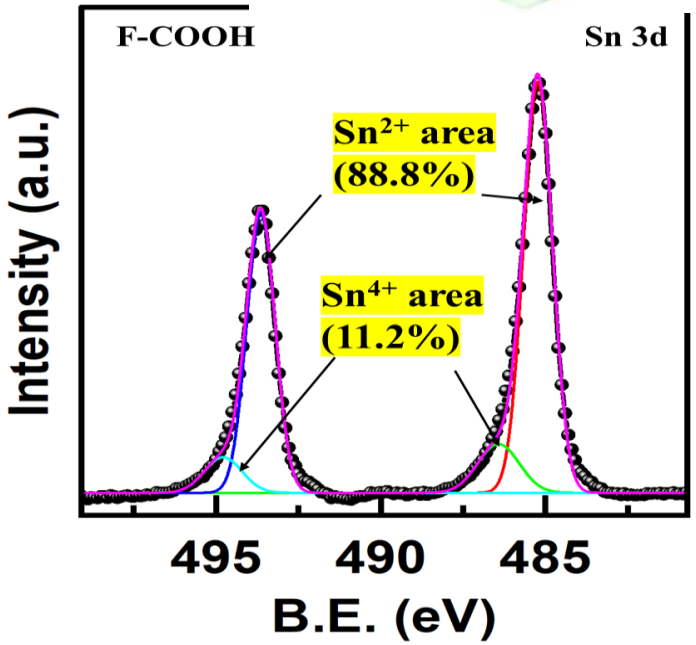
Fullerene- functional molecules:

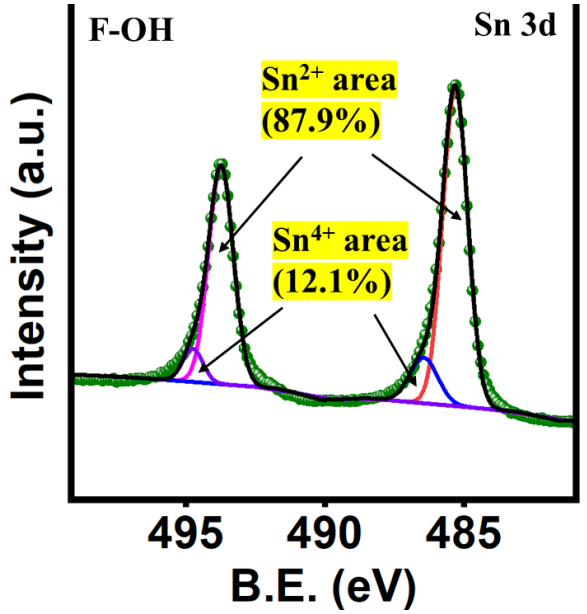
> Functional groups interact with Sn^{2+} of the $[SnX_6]^{4-}$ octahedra, reducing the e xtent of its oxidation to Sn^{4+}

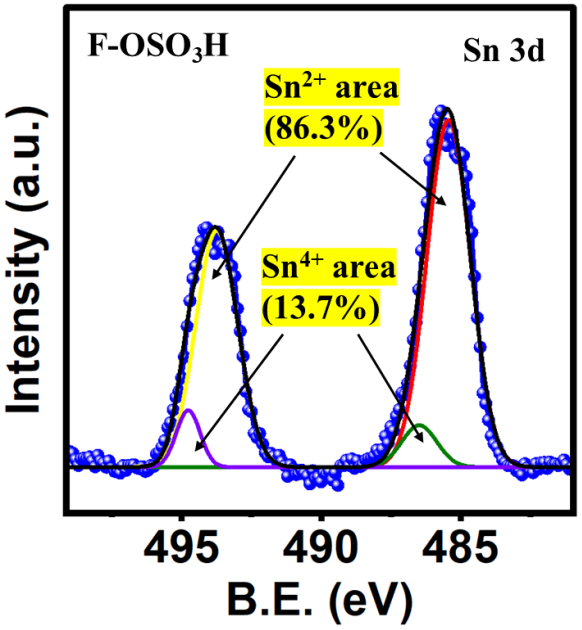


XPS: Suppression of oxidation

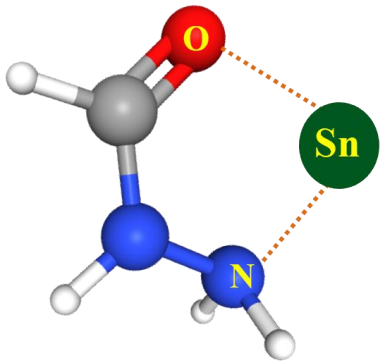




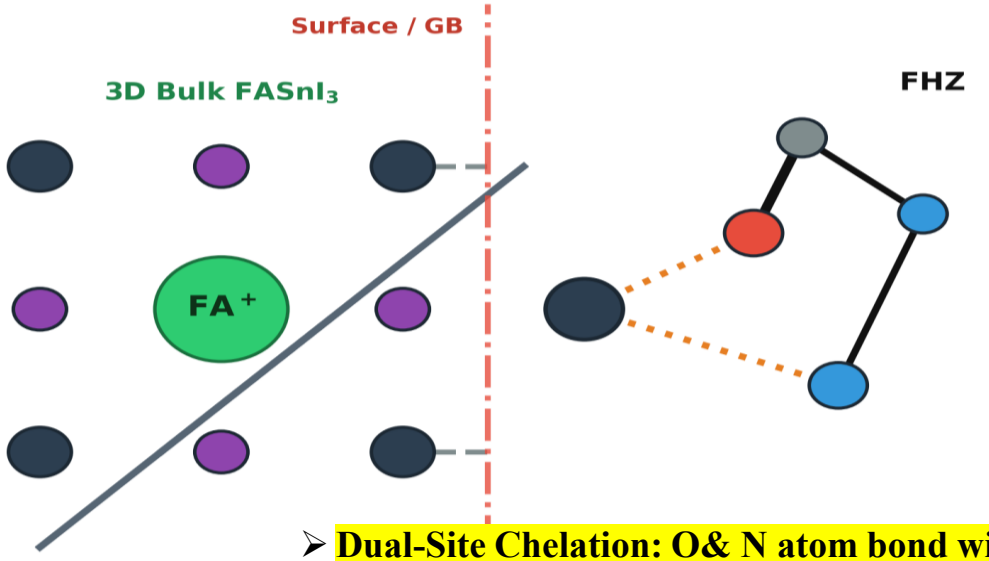




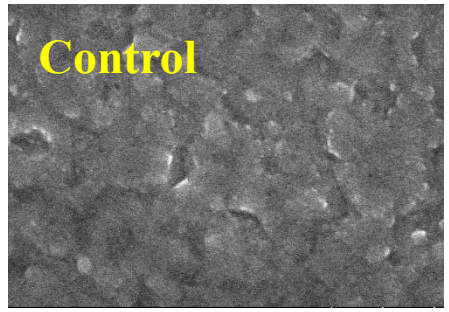
Effect of bidentate ligand: Sn-PSCs

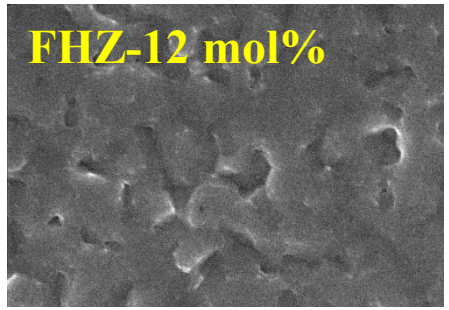


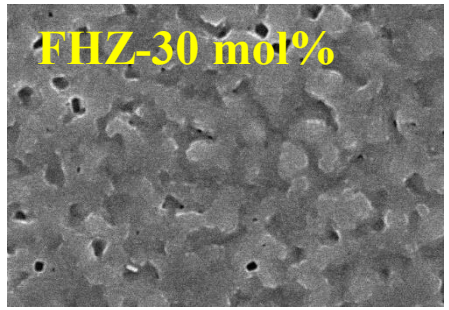
Formohydrazide (FHZ)



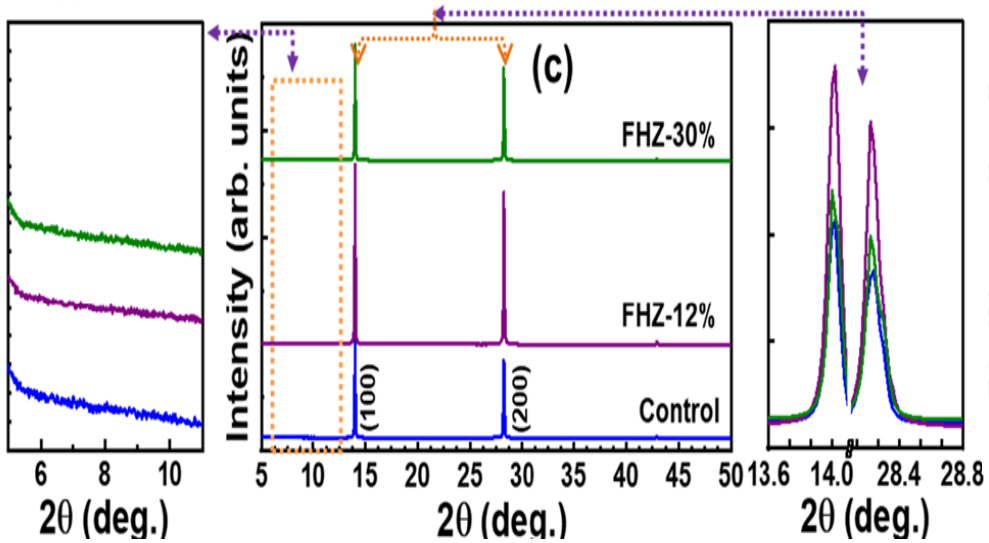
FHZ additive: better coverage and compact film

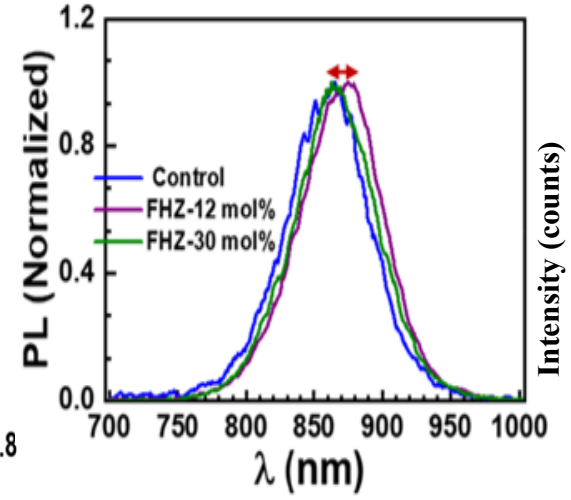


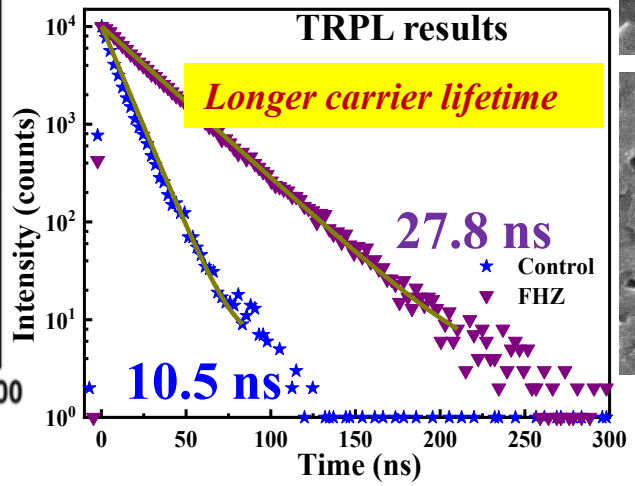




- Dual-Site Chelation: O& N atom bond with Sn^{2+}
- Defect Passivation: Eliminates surface Sn vacancies
- Anti-Oxidation: Suppresses Sn^{2+} -> Sn^{4+} High



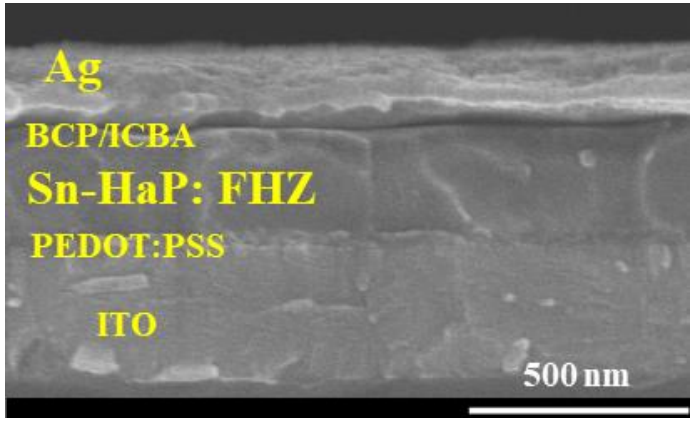


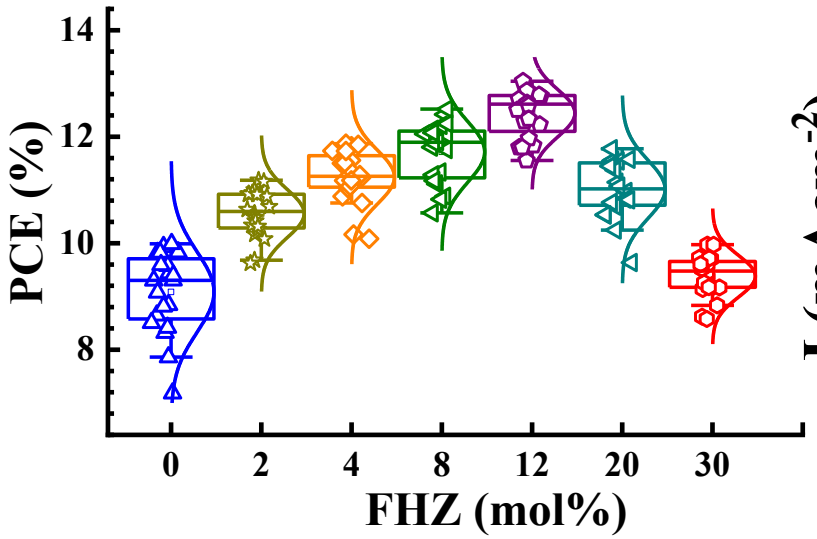


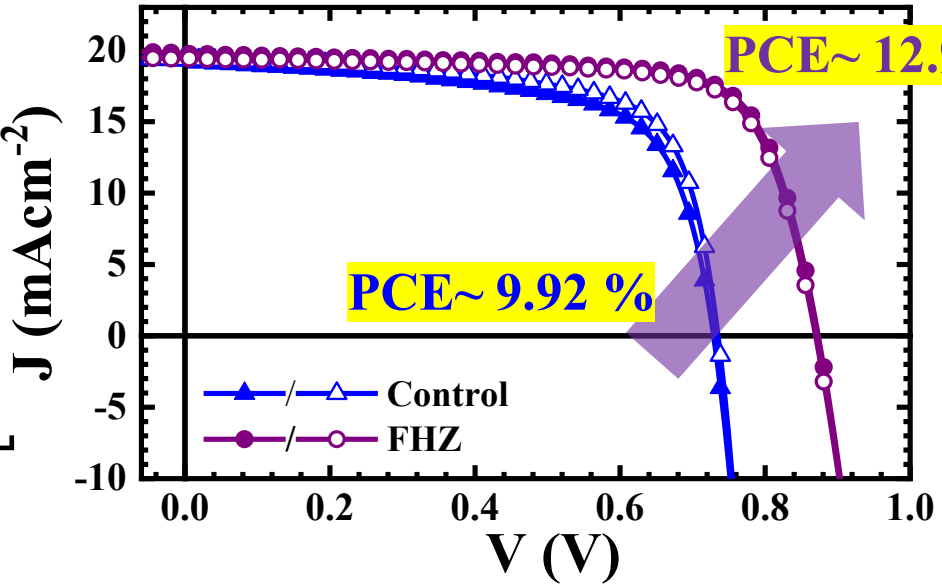
Sn-HP- FHZ added: improved crystallinity

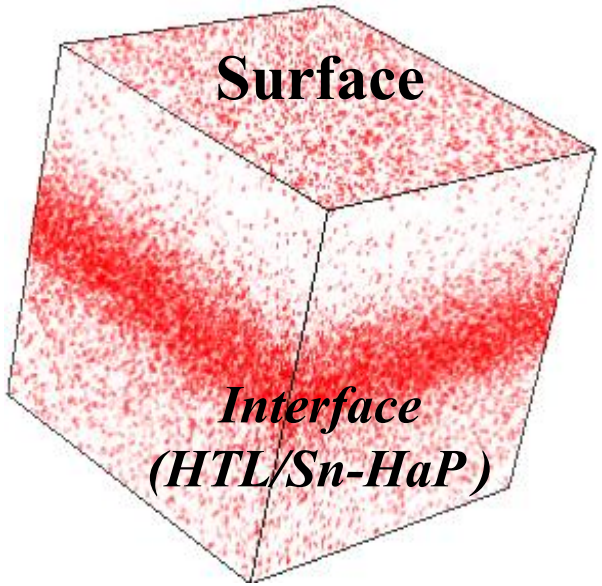
-No change in 2θ,
-Not incorporated into the crystal lattice.

Sn-PSCs+FHZ: device structure/results

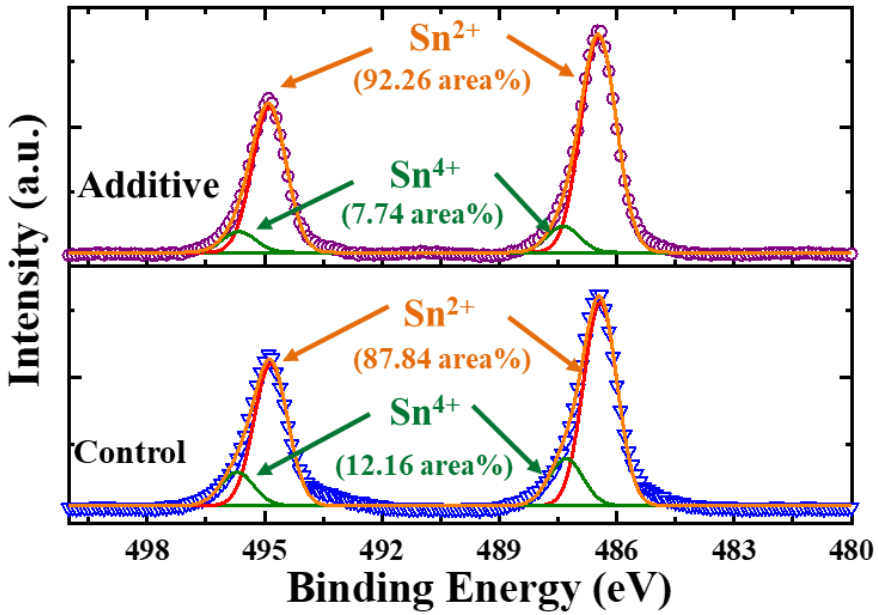


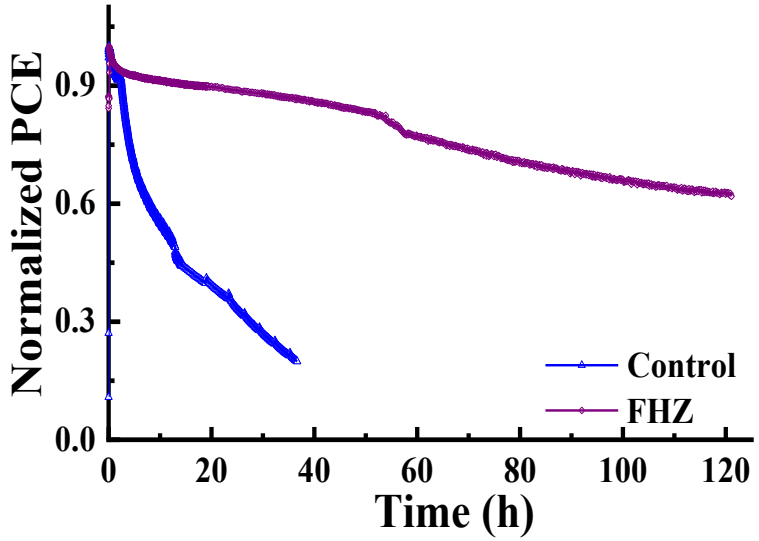






Suppression of oxidation





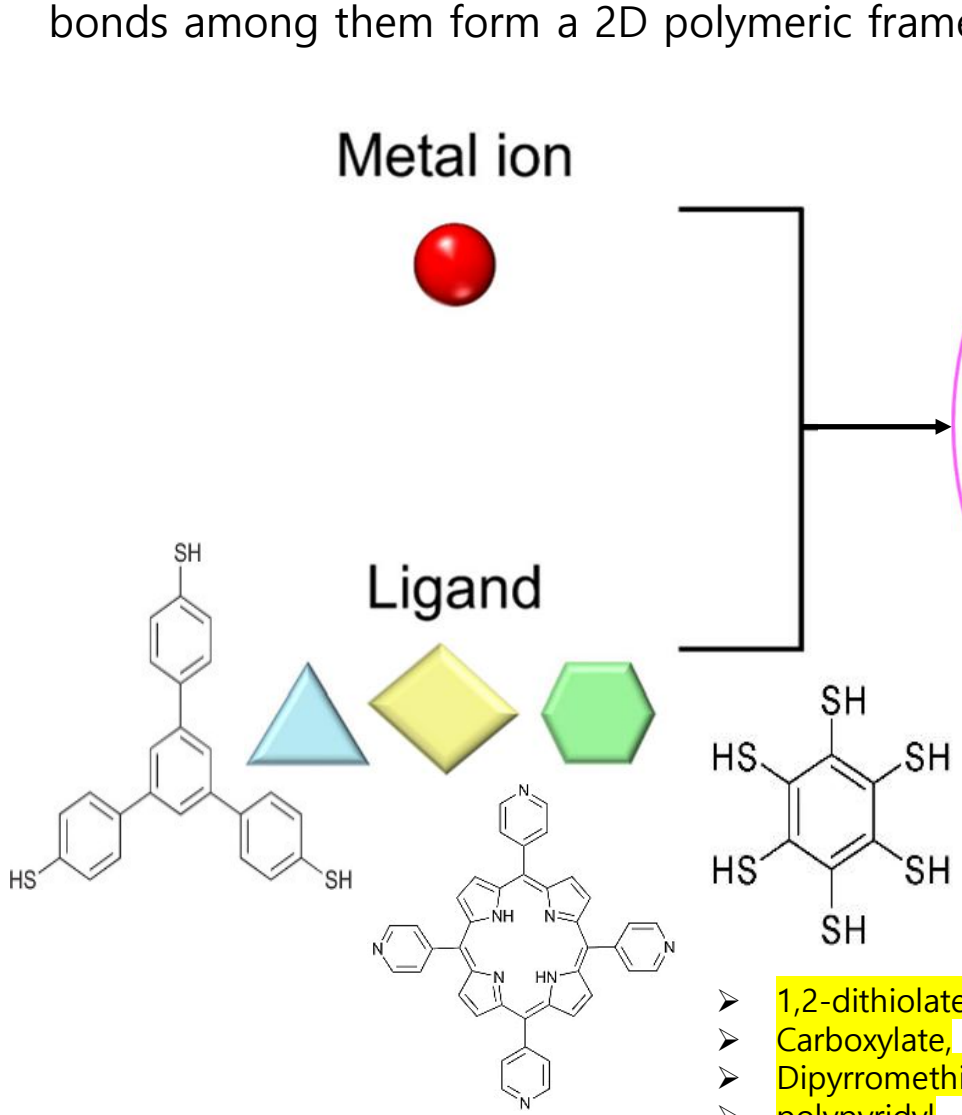
Khadka et al. Chem. mater. 2023,35,4250

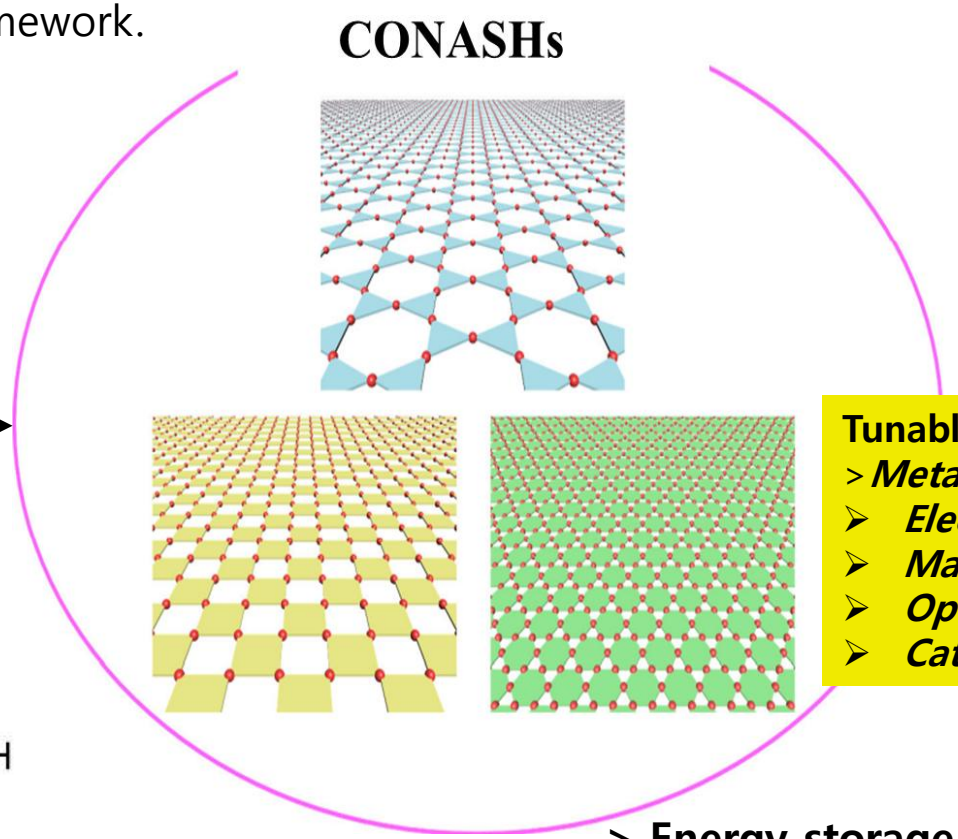
Coordination nanosheets (CONASHs)

Coordination nanosheets (CONASHs)

>Molecule-based nanosheets.

>Organic ligand molecules and metal ions/atoms/clusters, and the coordination bonds among them form a 2D polymeric framework.







Hiroshi Nishihara

Hiroshi Nishihara and coworkers, Chem. Commun.,2017, 53,5781

Tunable chemical and physical properties

>Metal nodes and organic ligands

- ***Electronic conductivity***
- ***Magnetic properties***
- ***Optical response***
- ***Catalytic activity***

- 1,2-dithiolate and its derivatives
- Carboxylate,
- Dipyrromethine,
- polypyridyl

> Energy storage and conversion

CNSs are used in:

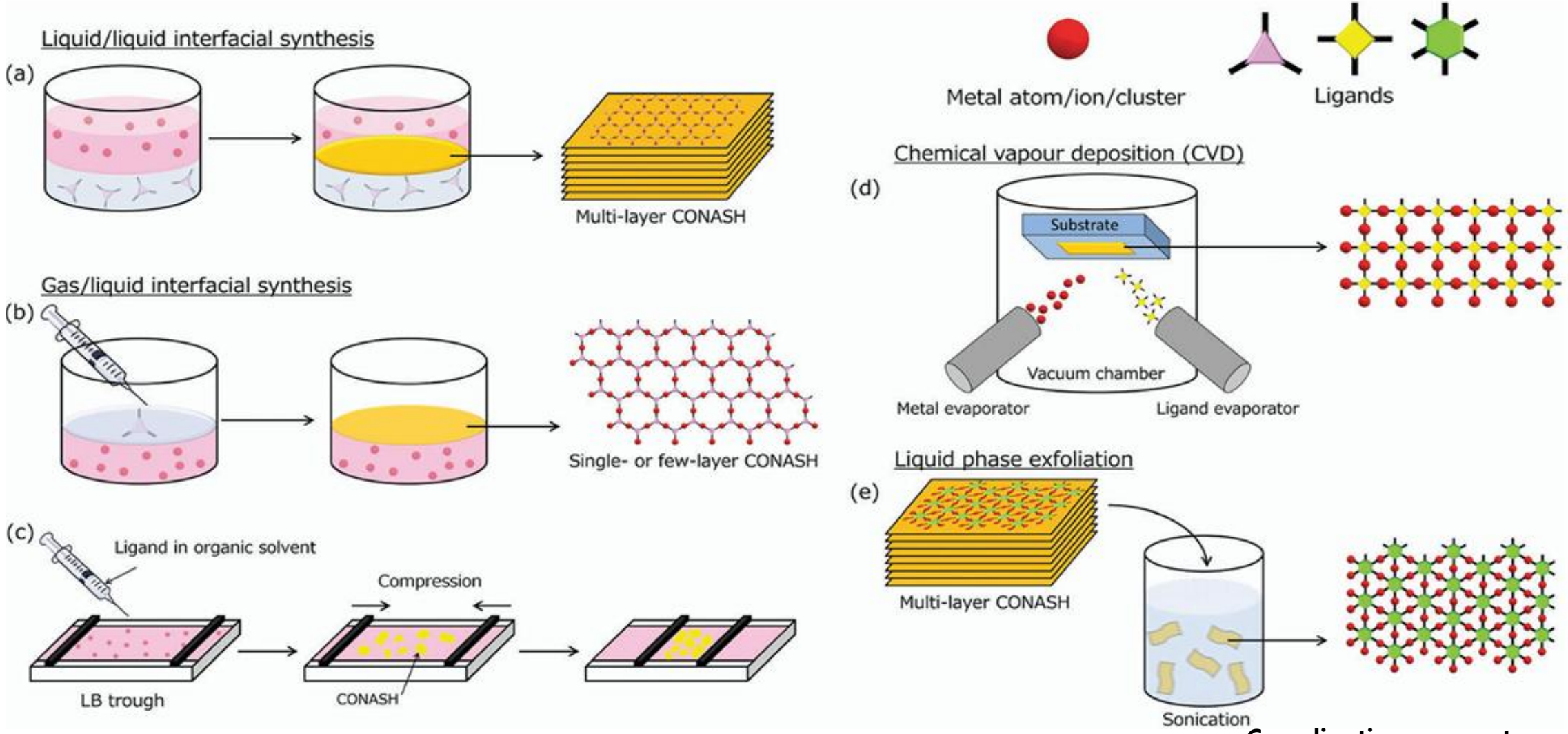
- ***Lithium-/sodium-ion batteries***
- ***Supercapacitors***
- ***Fuel cells***
- ***Solar energy harvesting***

➢ **2D frameworks facilitate fast ion/electron transport.**

Synthesis methods of CONASHs

Coordination nanosheets (CONASHs)

>molecule-based nanosheets> Organic ligand molecules and metal ions/atoms/clusters

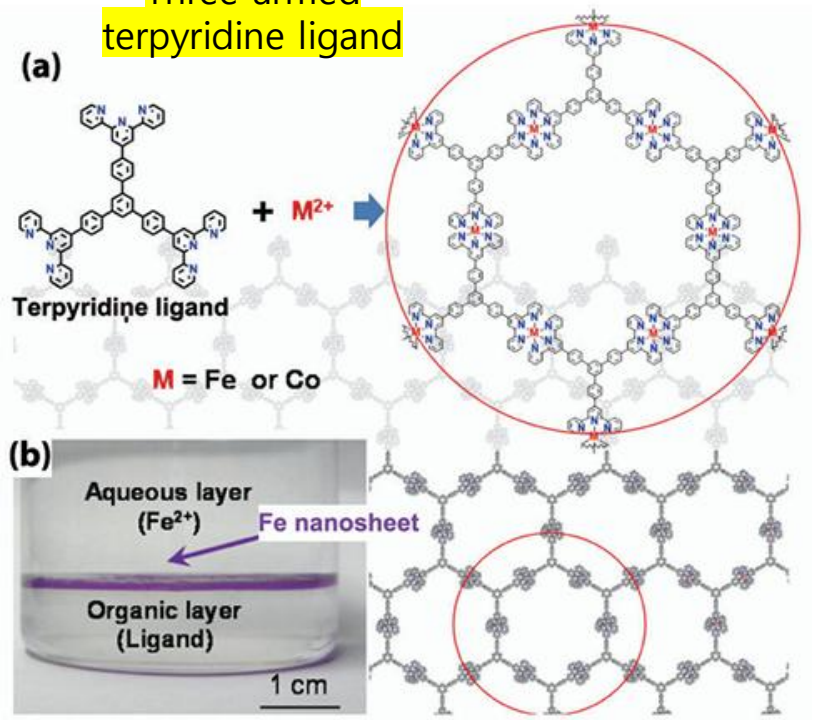


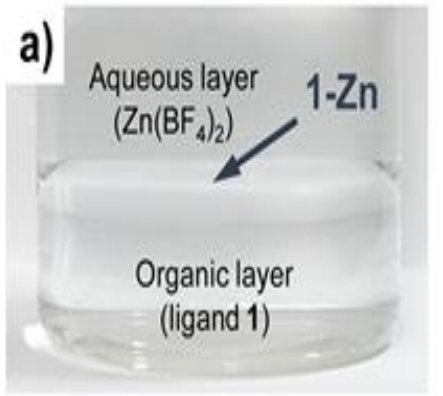
Coordination symmetry-CONASHs

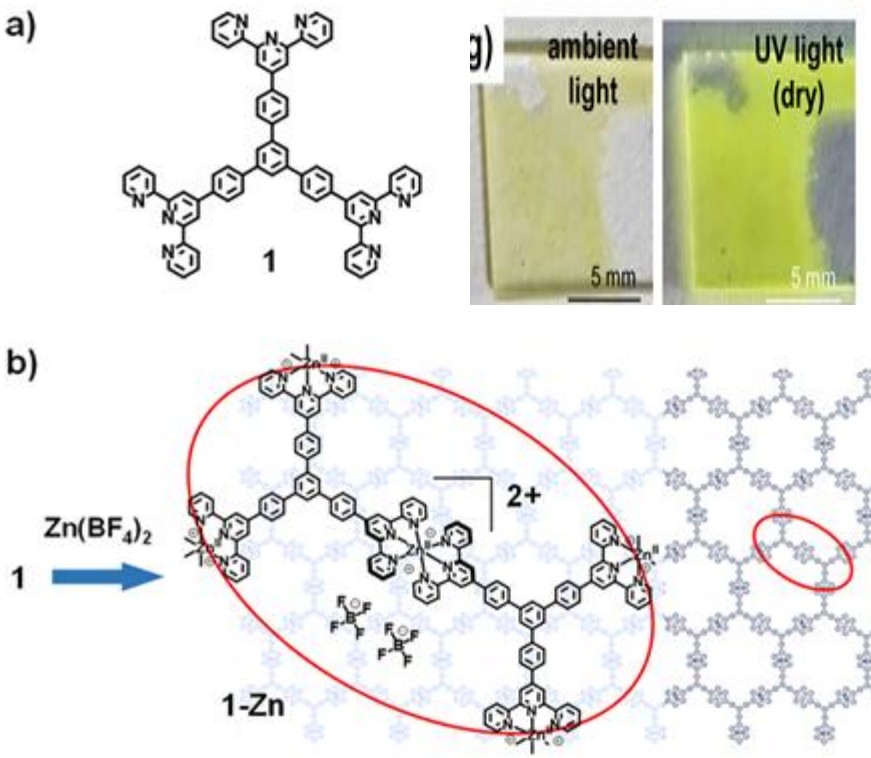
Hiroshi Nishihara and coworkers, Chem. Commun.,2017, 53,5781

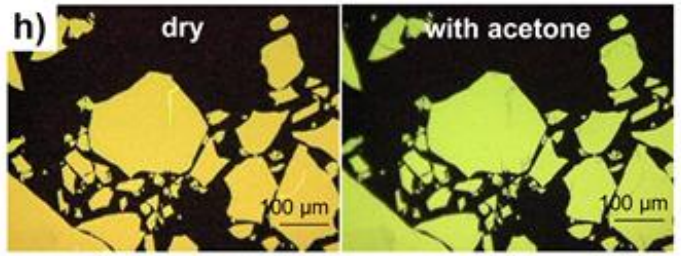
An example of CONASHs

Three armed terpyridine ligand









JACS
Article
pubs.acs.org/JACS

Coordination Nanosheets Based on Terpyridine–Zinc(II) Complexes: As Photoactive Host Materials

Takamasa Tsukamoto,[†,‡] Kenji Takada,[†] Ryota Sakamoto,[*,†,§] Ryota Matsuoka,[†] Ryojun Toyoda,[†] Hiroaki Maeda,[†] Toshiki Yagi,[†] Michihiro Nishikawa,[∥] Naoaki Shinjo,[†] Shuntaro Amano,[†] Tadashi Iokawa,[†] Narutaka Ishibashi,[†] Tsugumi Oi,[†] Koshiro Kanayama,[†] Rina Kinugawa,[†] Yoichiro Koda,[†] Toshiyuki Komura,[†] Shuhei Nakajima,[†] Ryota Fukuyama,[†] Nobuyuki Fuse,[†] Makoto Mizui,[†] Masashi Miyasaki,[†] Yutaro Yamashita,[†] Kuni Yamada,[†] Wenxuan Zhang,[†] Ruocheng Han,[†] Wenyu Liu,[†] Taro Tsubomura,[∥] and Hiroshi Nishihara[*,†]

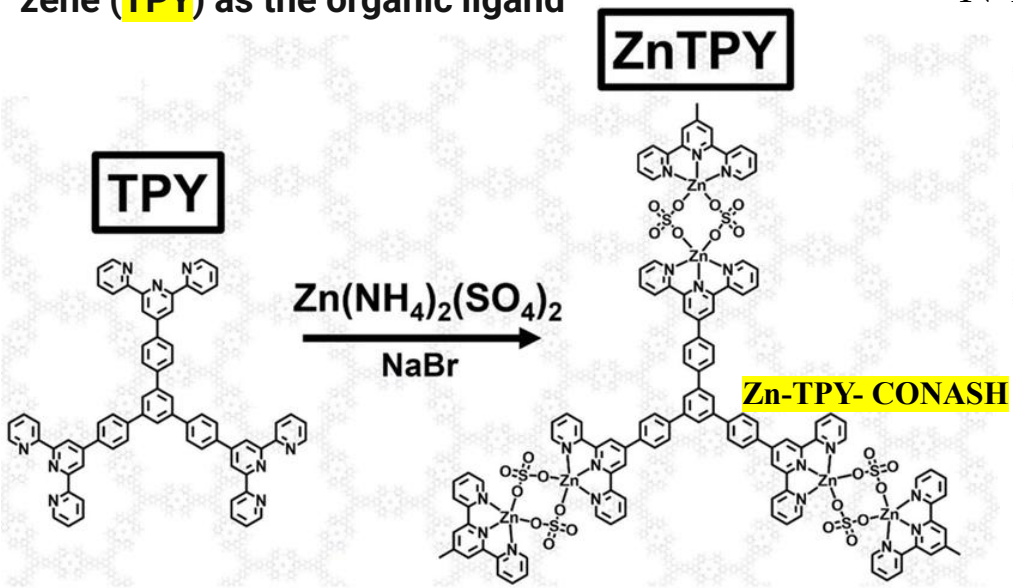
[†]Department of Chemistry, Graduate School of Science, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan
[‡]Japan Society for the Promotion of Science (JSPS), Ichibancho, Chiyoda-ku, Tokyo 102-8471, Japan
[§]JST-PRESTO, 4-1-8 Honjcho, Kawaguchi, Saitama 332-0012, Japan
[∥]Department of Materials and Life Science, Seikei University, 3-3-1 Kichijoji-kitamachi, Musashino-shi, Tokyo 180-8633, Japan

Supporting Information

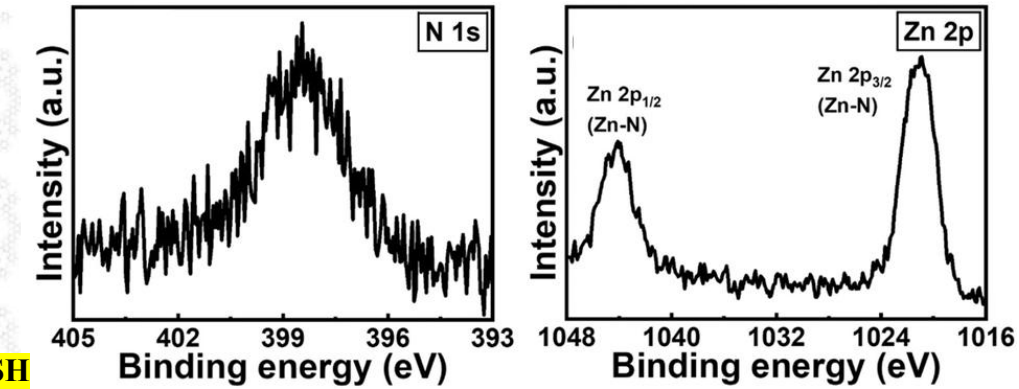
Hiroshi Nishihara and coworkers, J. Am. Chem. Soc. 2017, 139, 15, 5359

Zn-TPY-CONASH-Synthesis

1,3,5-tris[4-(4'-2,2':6',2"-terpyridyl)phenyl]-benzene (TPY) as the organic ligand



XPS spectra of as-synthesized ZnTPY CONASHs
>N 1s and Zn 2p core levels

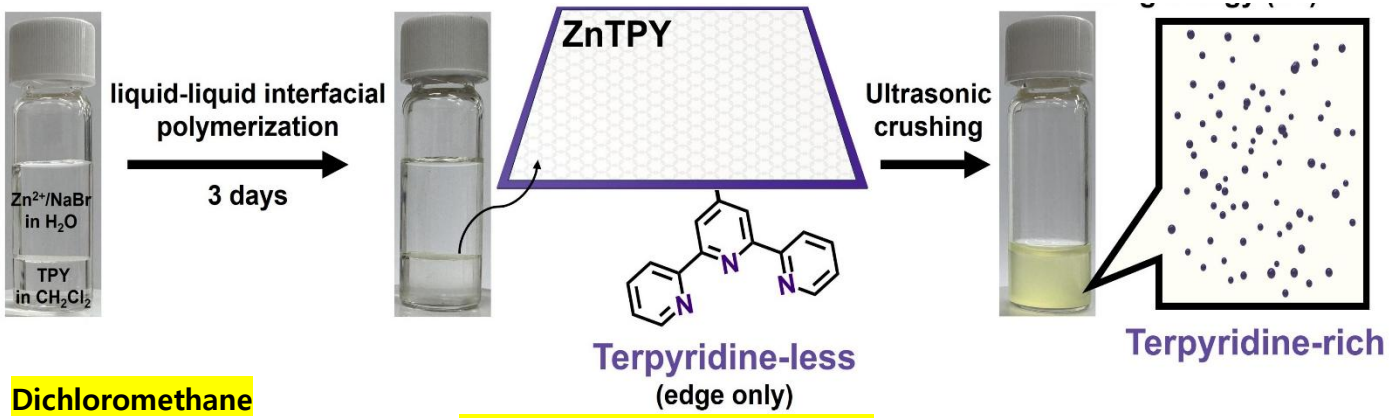




Dr. Ying-Chiao Wang
NIMS Alumni
(National Sun Yat-sen University)

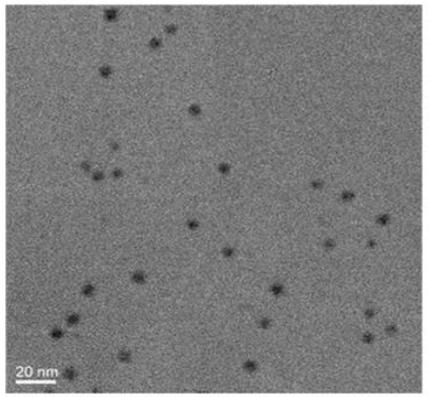
> Formation of ZnTPY CONASHs

- Liquid/liquid interfacial synthesis

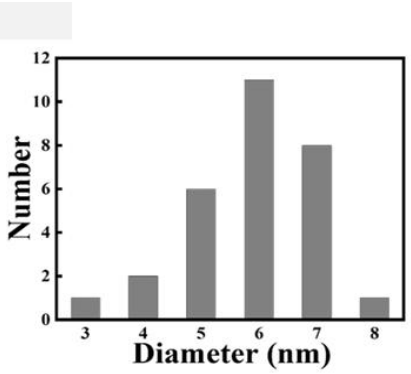


Dichloromethane

Zn-TPY-CONASH/DMF suspension



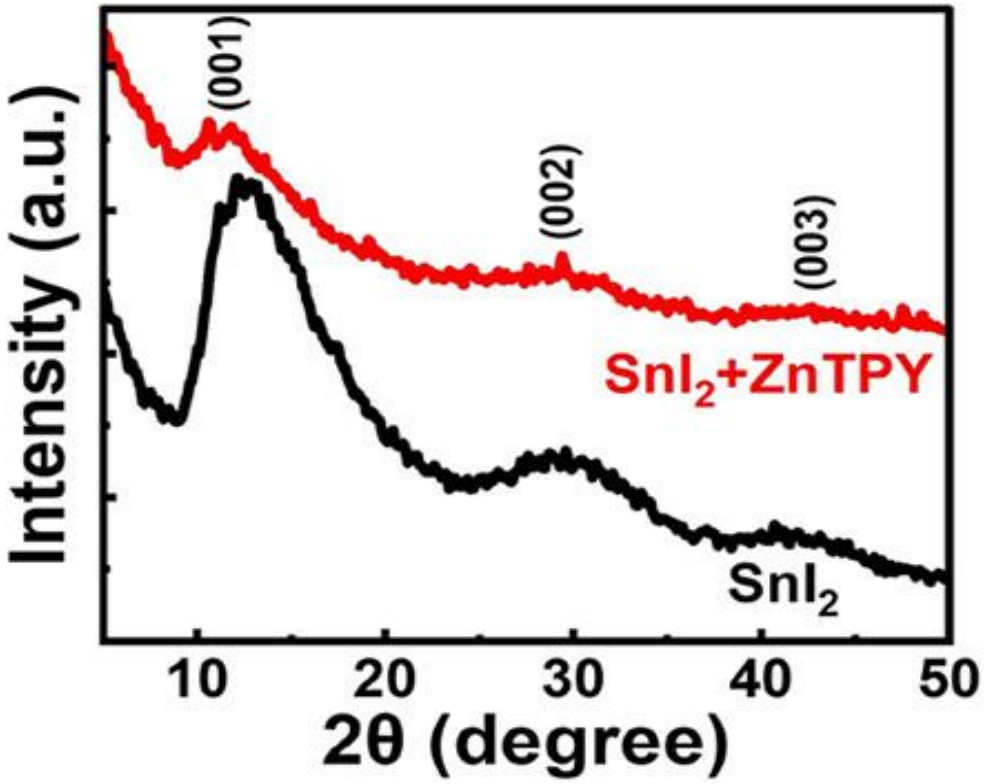
TEM image

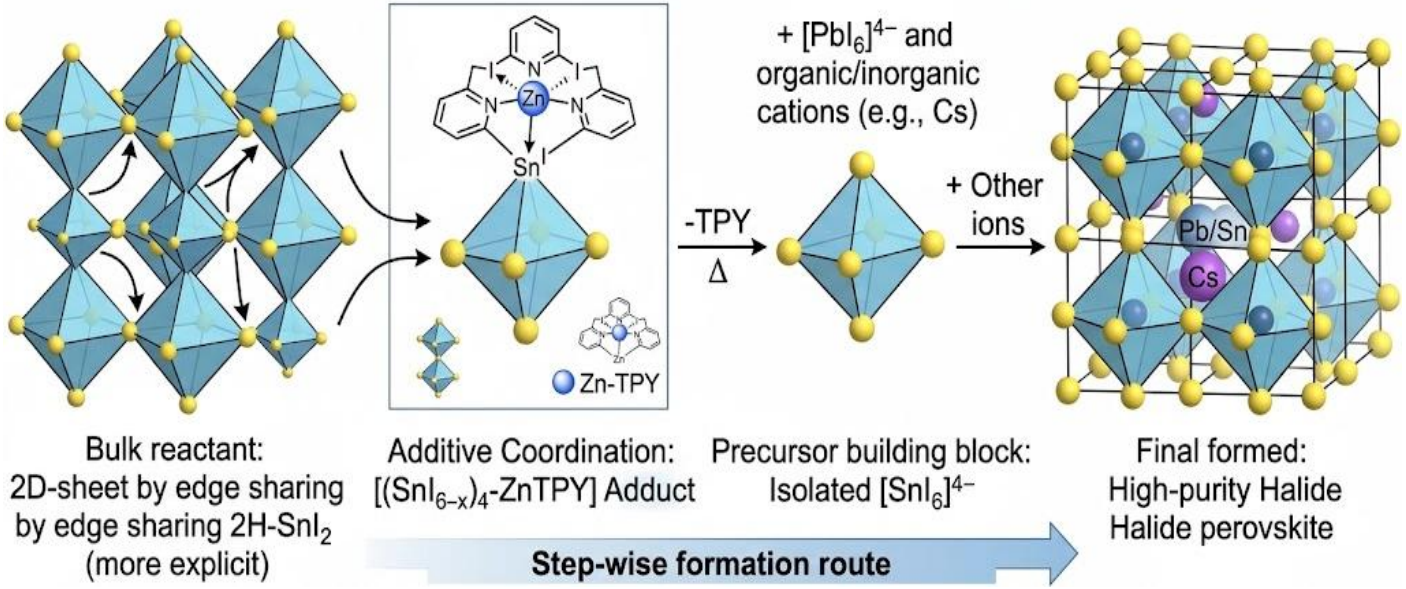


Khadka et al. ACS Appl. Mater. Interfaces 2025, 17, 18, 26813

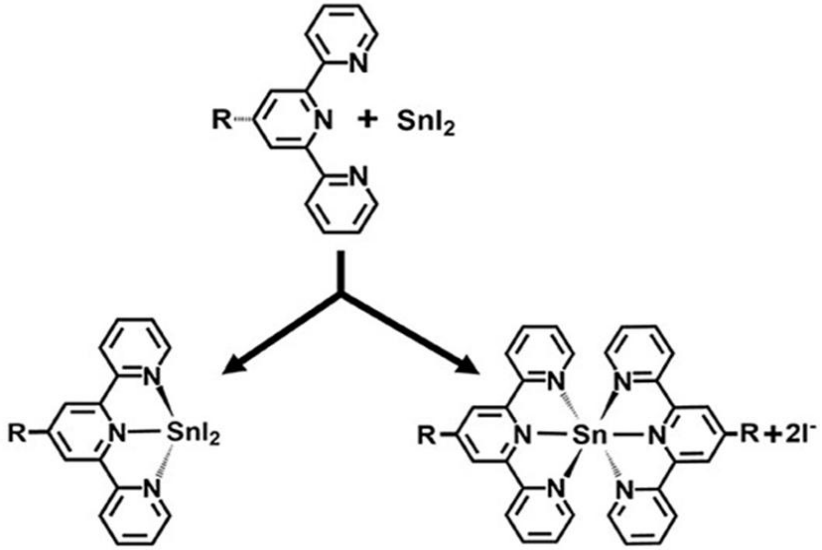
Growth of CONASH/Halide Perovskite

XRD- SnI_2 and Zn-TPY





Two possible routes of the formation of Zn-TPY: SnI_2 adducts



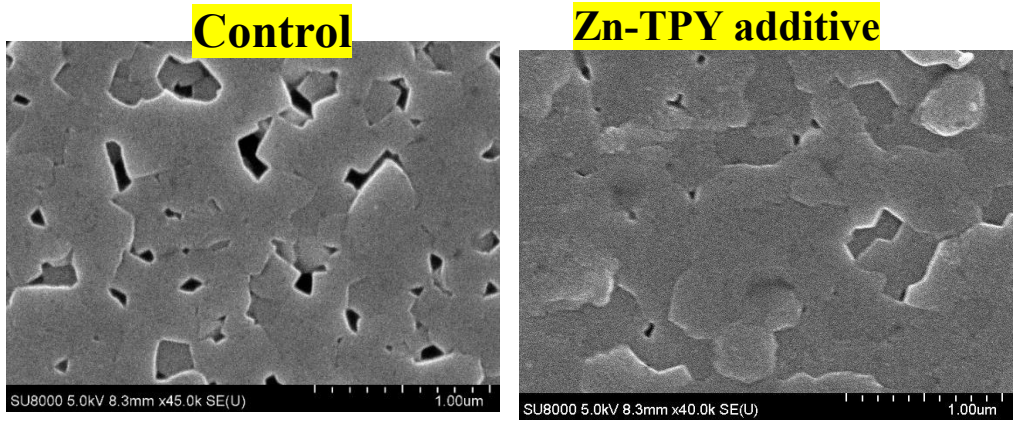
Diffraction peak-(001) – hexagonal 2H polytype SnI_2

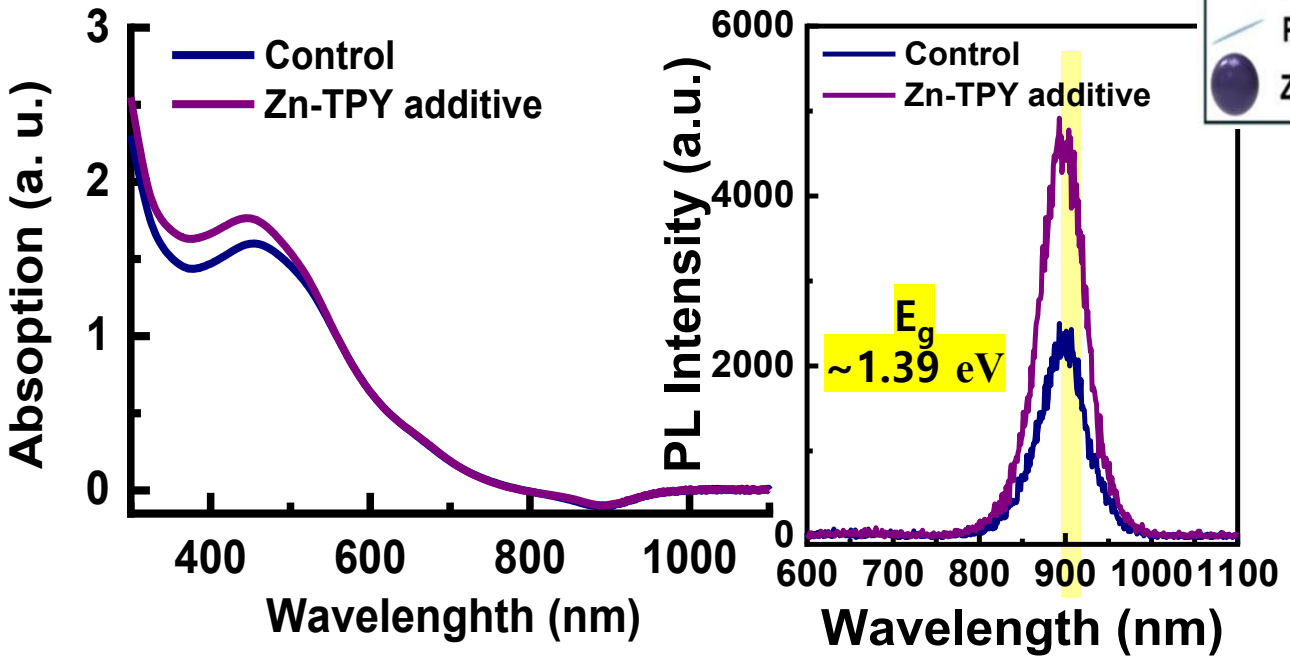
>Insertion of ZnTPY into SnI_2 –promote octahedral unit

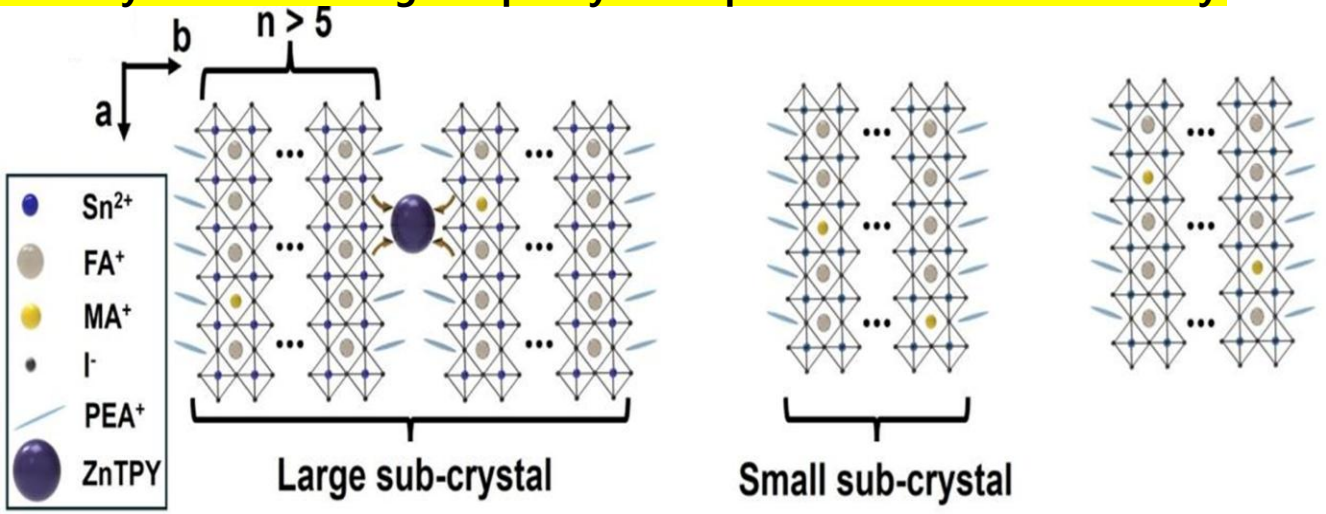
Khadka et al. ACS Appl. Mater. Interfaces 2025, 17, 18, 26813

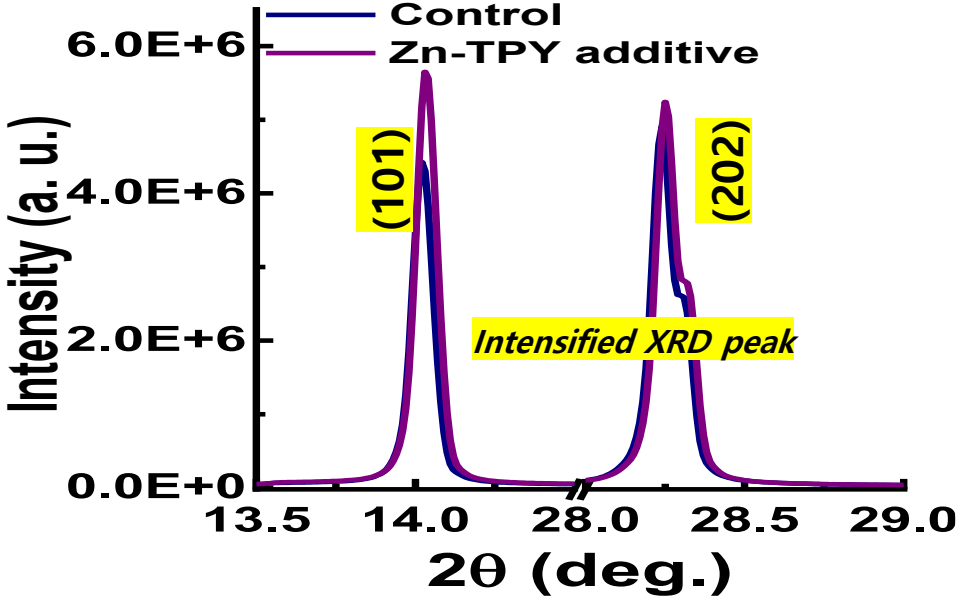
Effect of CONASH- Sn-perovskite film

- ZnTPY additive acts as a "molecular scissor" in a 2D/3D Sn-HP
- Convert-large layered sub-crystals into smaller
- Regulate crystallization>higher quality films-passivated defects>stability.





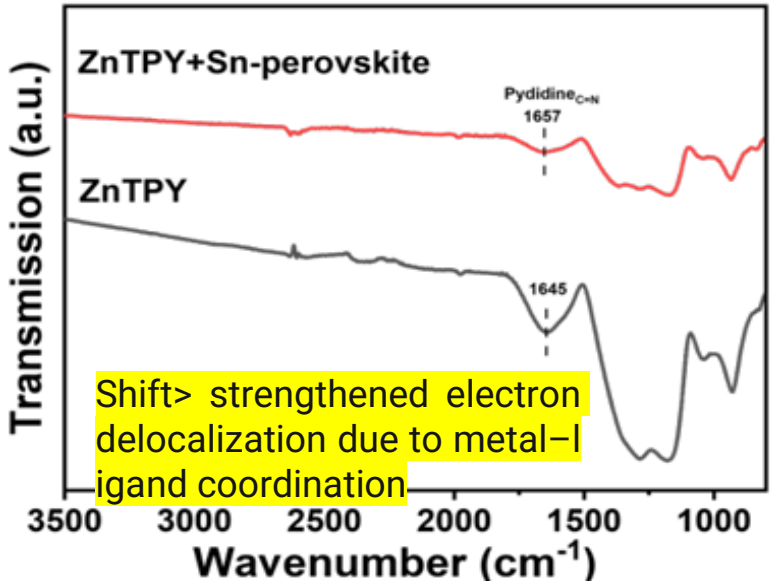




- *Khadka/Wang (NSYSU) et al. ACS Appl. Mater. Interfaces 2025, 17, 18, 26813*

Interaction of CONASH and Sn- Perovskite

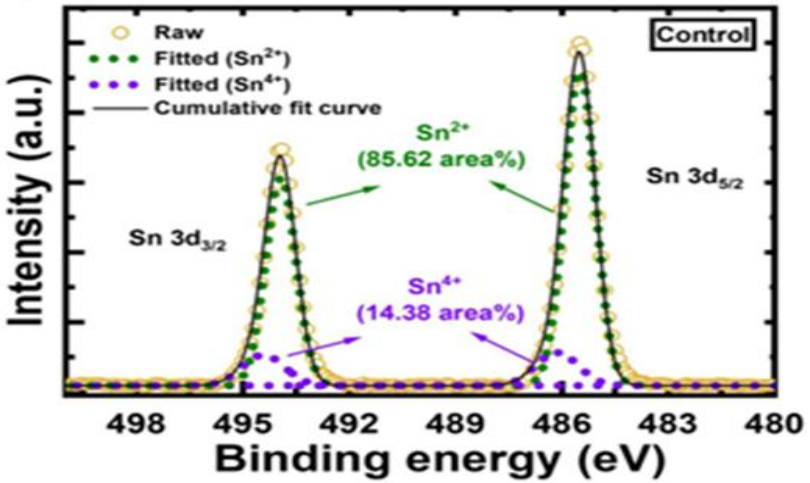
FTIR spectra of ZnTPY and the ZnTPY-Sn perovskite

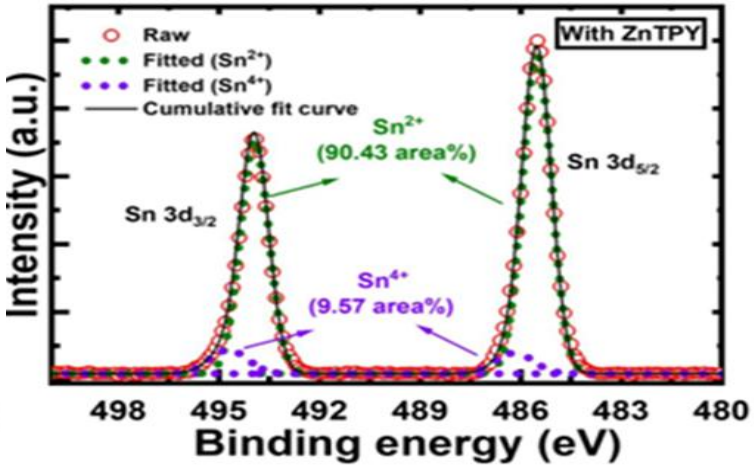


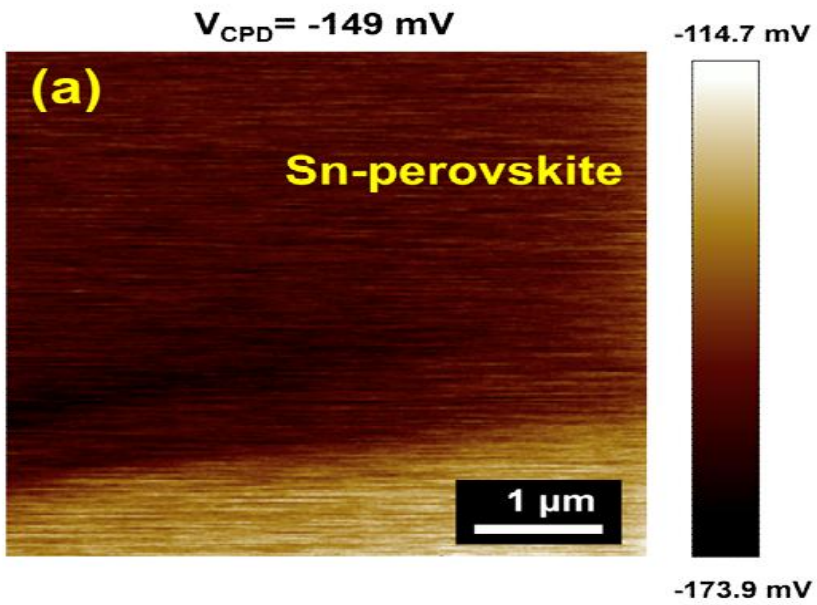
Shift> strengthened electron delocalization due to metal–ligand coordination

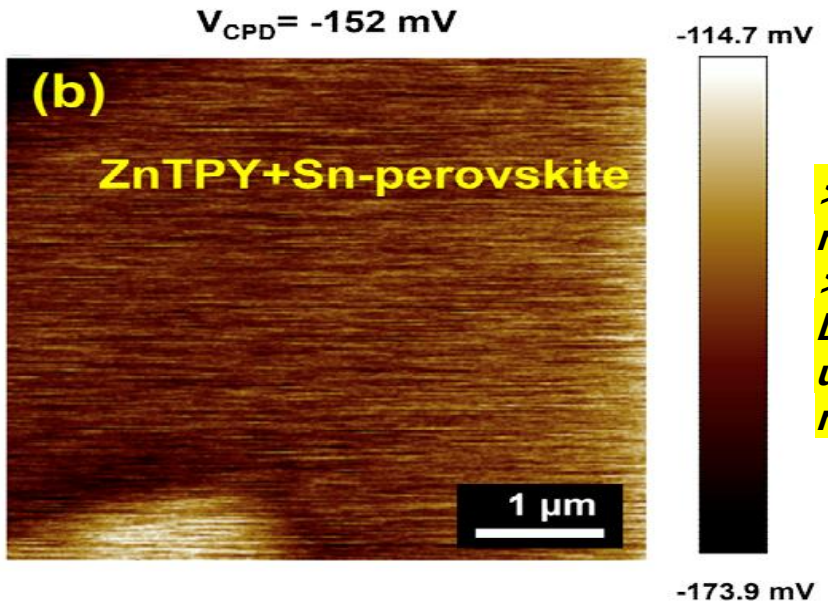
Chemical interaction –XPS Sn-perovskite and Zn-TPY

Sn^{4+} suppressed with Zn-TPY additive





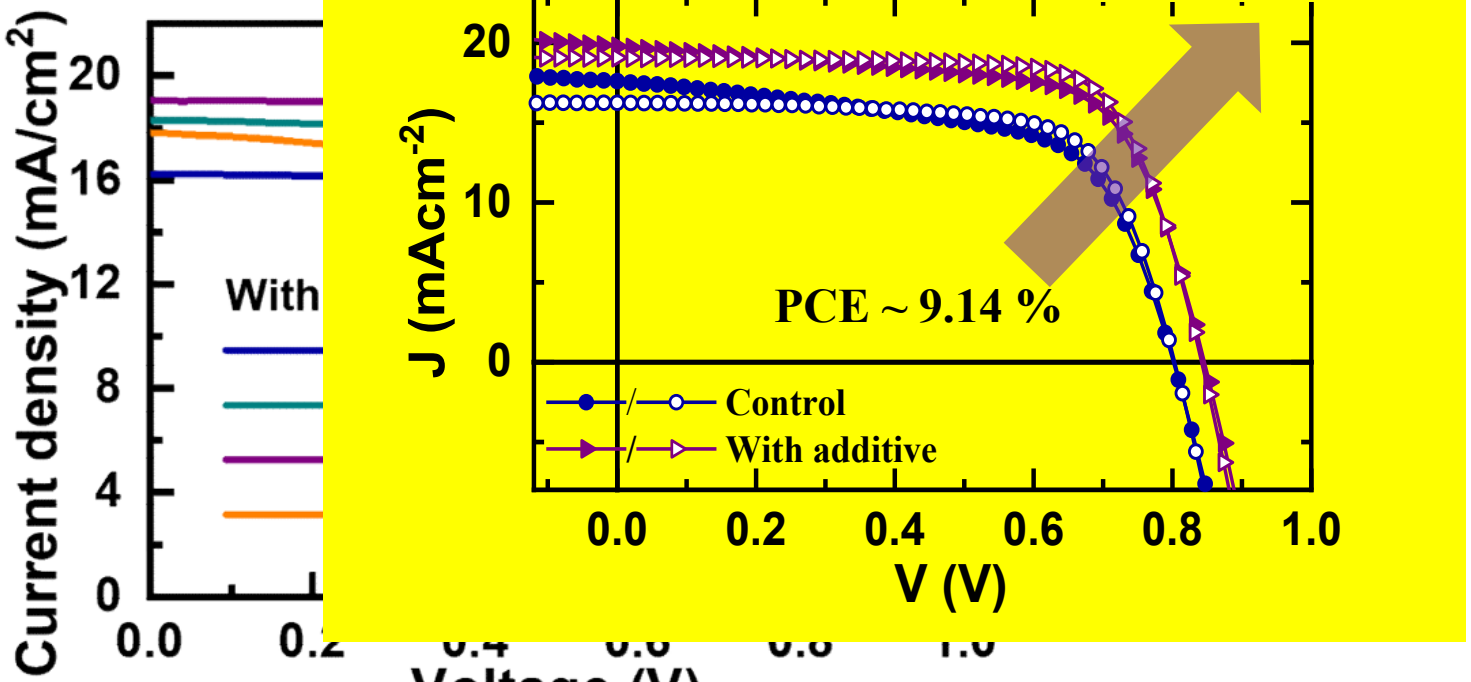




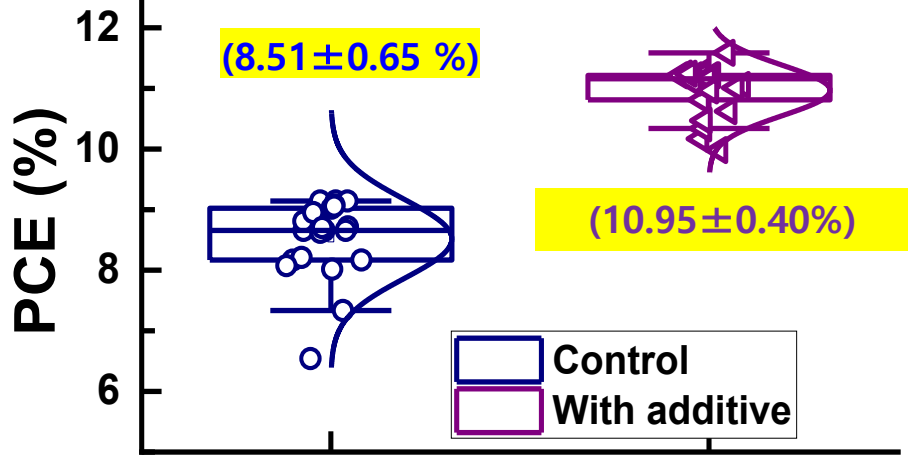
>KPFM images of Sn-perovskite films

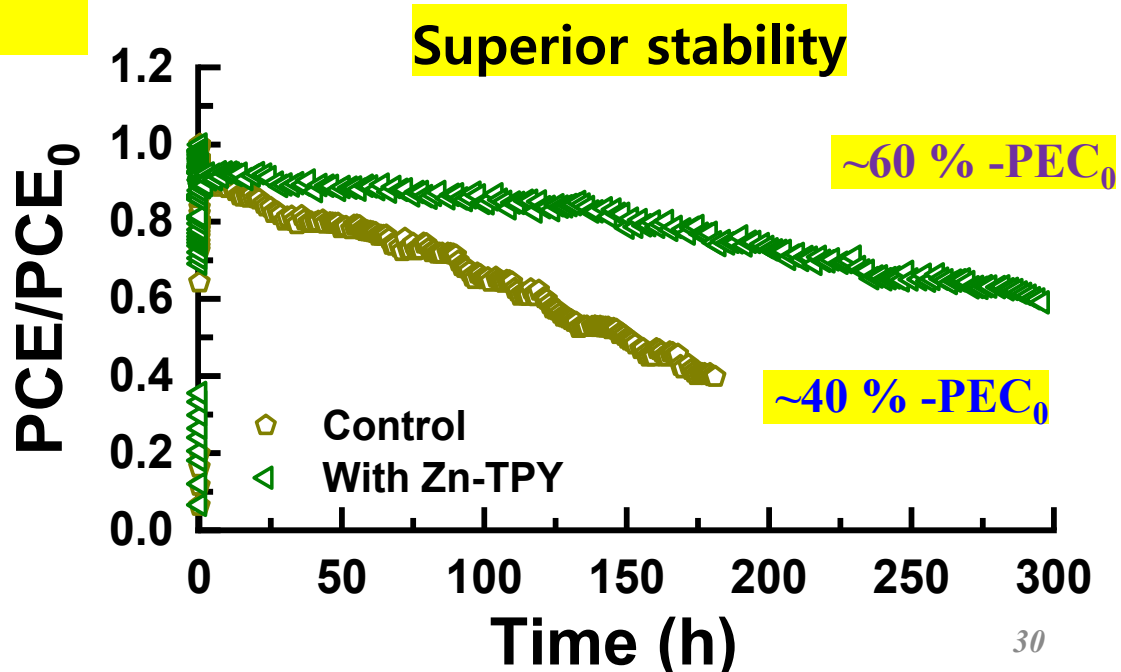
>Contact potential difference (CPD) - indicate surface energy modulation with the incorporation of Zn-TPY

Effect of CONASH- Sn-PSCs

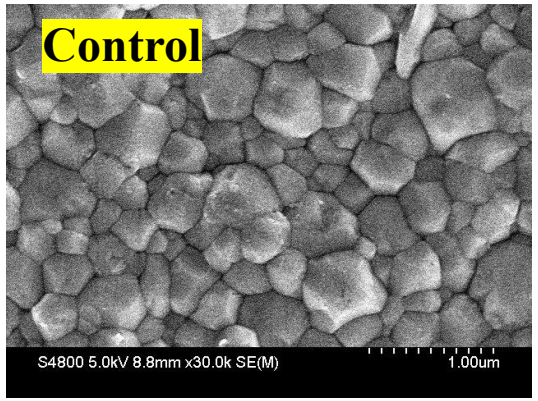


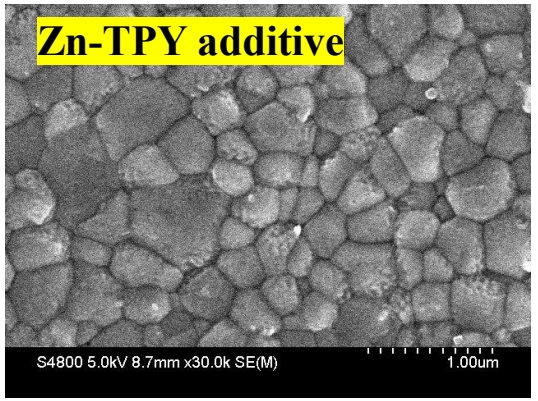
Devices	J_{SC} (mA cm^{-2})	V_{OC} (V)	FF (%)	PCE (%)
Control FS/RS)	16.25/ 17.68	0.807/ 0.801	69.70/ 61.12	9.14/ 8.66
ZnTPY (FS/RS)	19.06/ 19.83	0.842/ 0.848	72.19/ 66.70	11.59/ 11.2

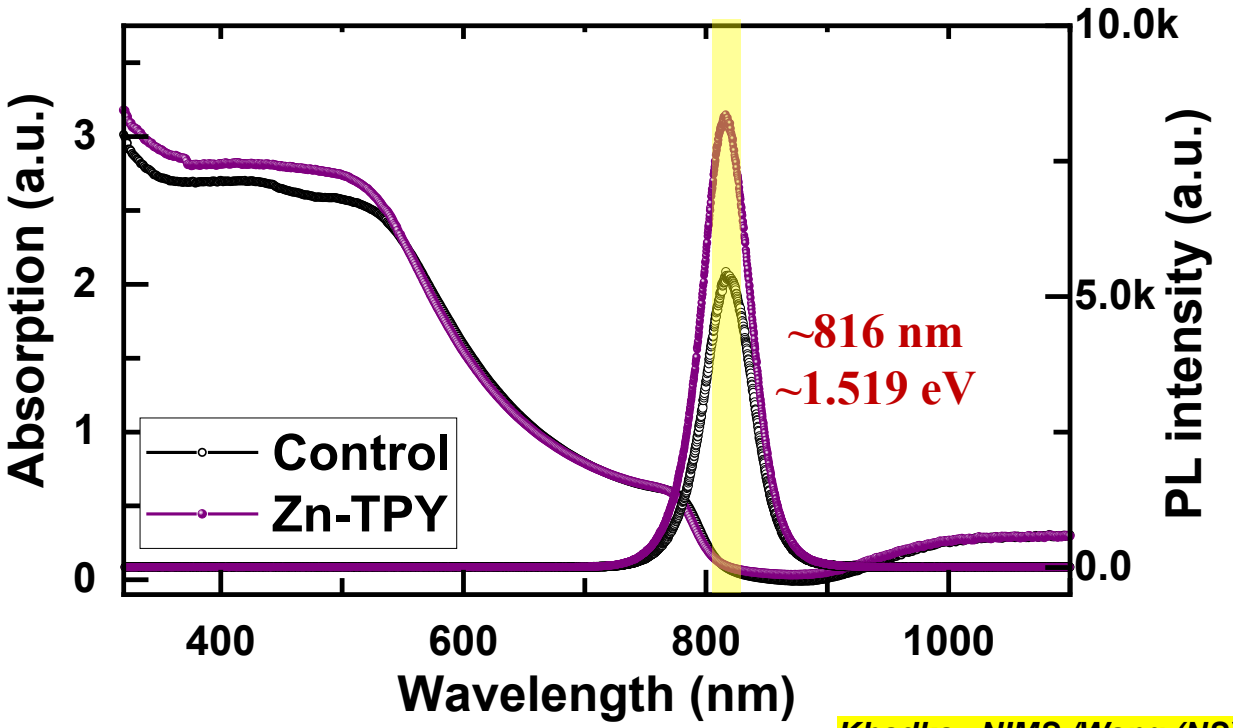




Effect of CONASH- Pb-Perovskite film

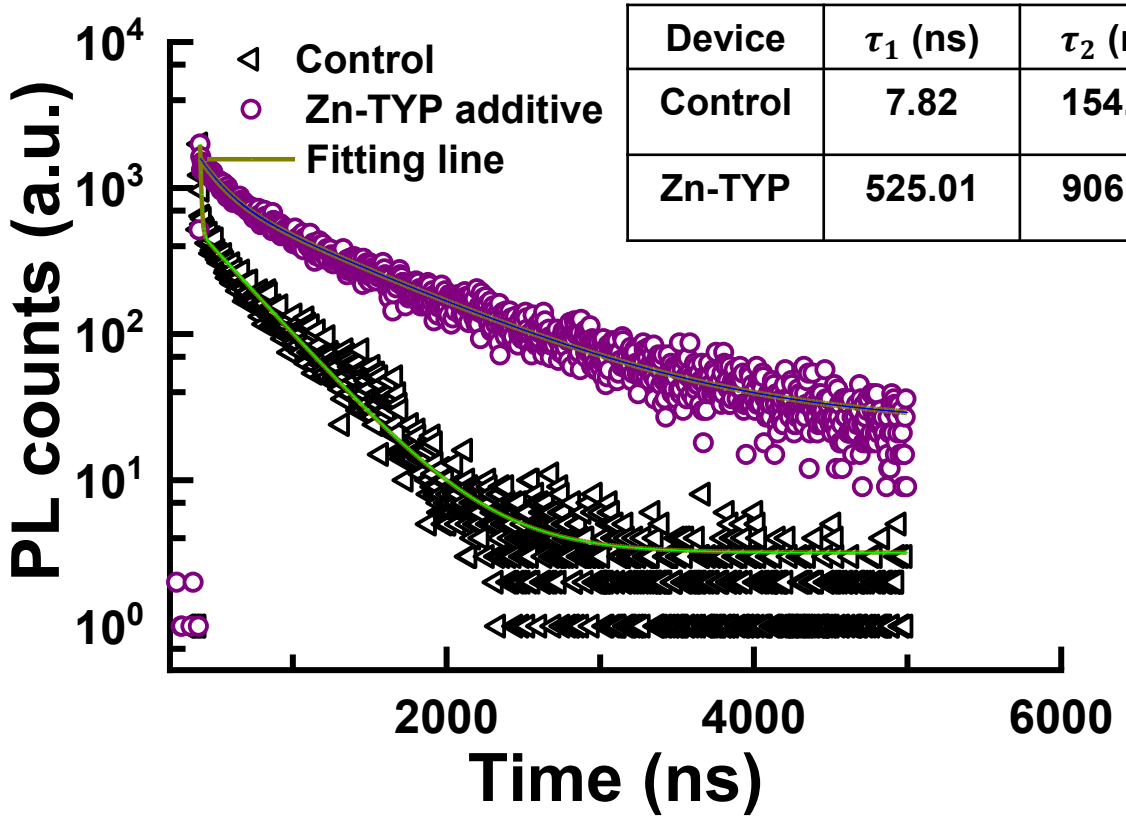






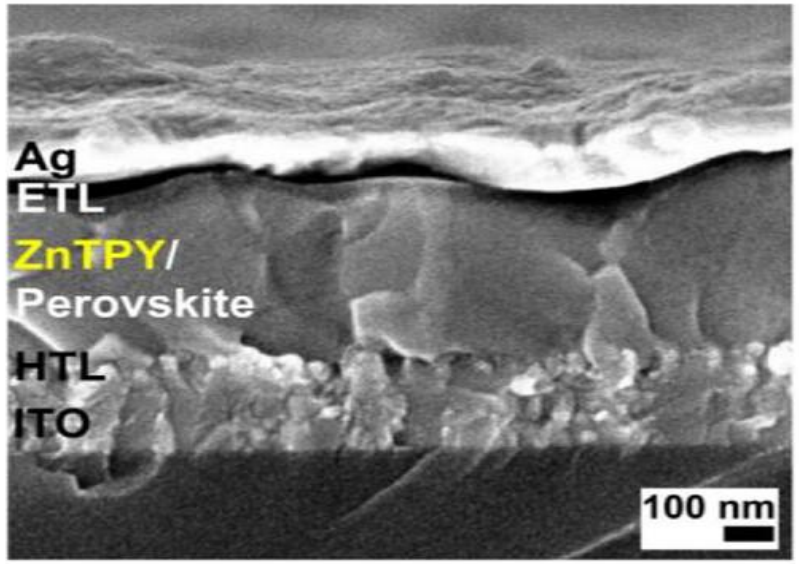
TRPL data

Device	τ_1 (ns)	τ_2 (ns)
Control	7.82	154.08
Zn-TYP	525.01	906.11

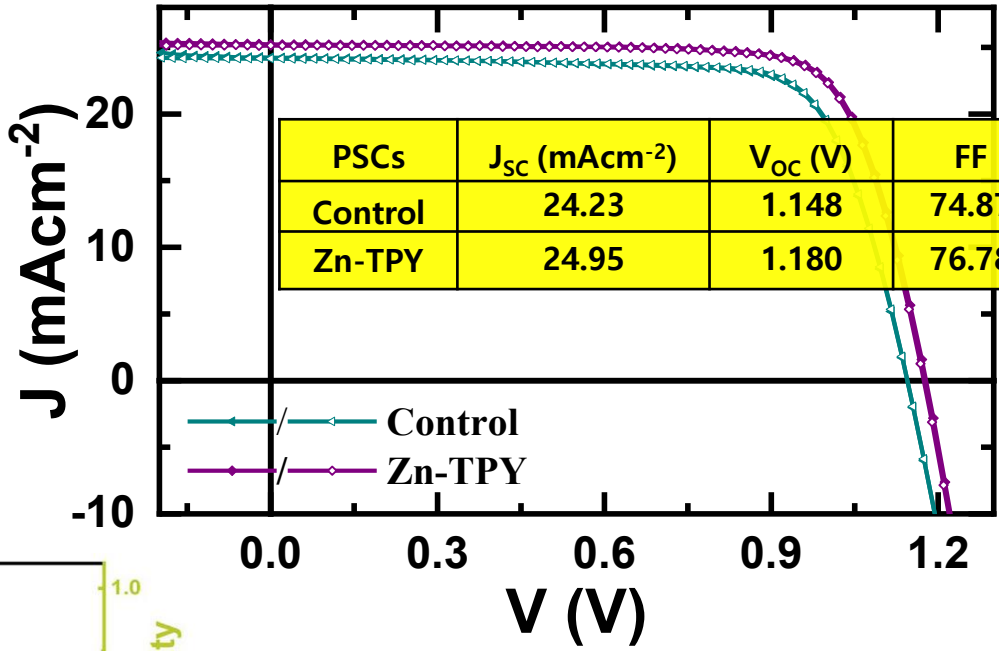


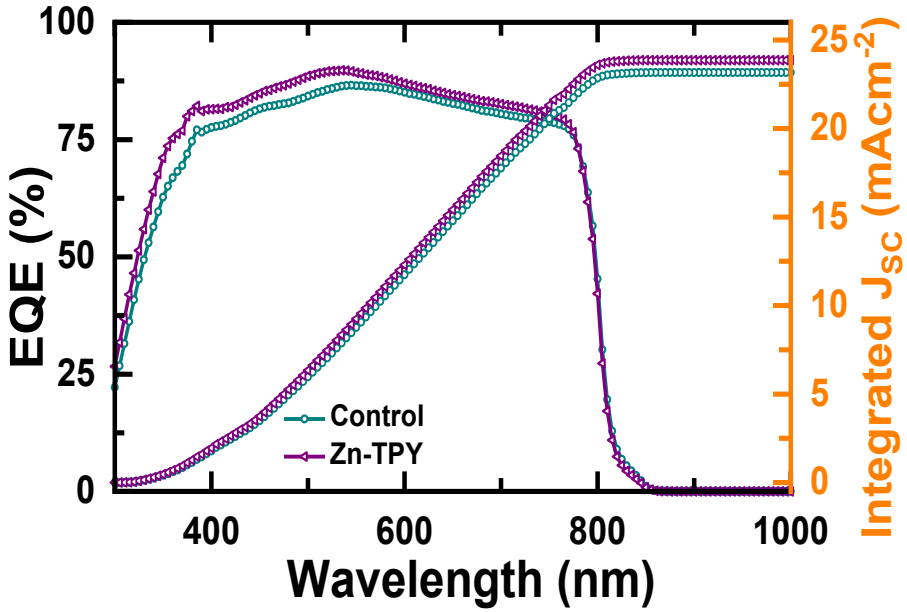
Khadka –NIMS /Wang (NSYSU) et al. Small (2026): e76016

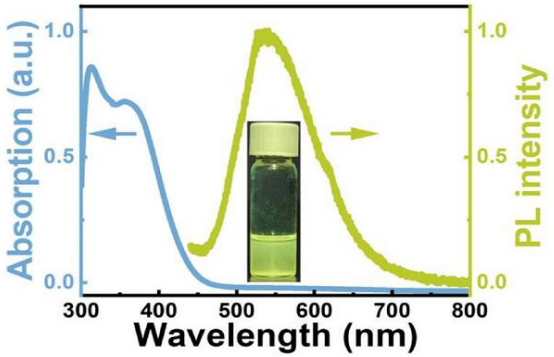
Effect of CONASH- Pb-PSC Device

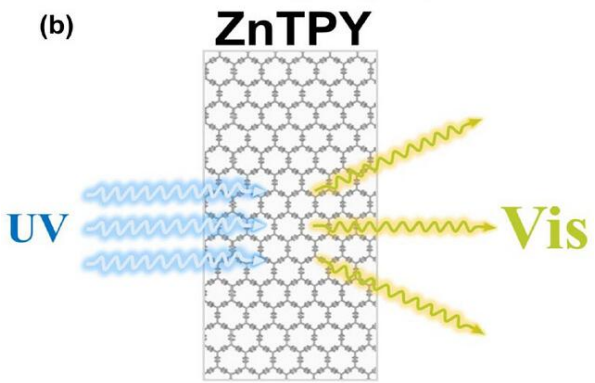


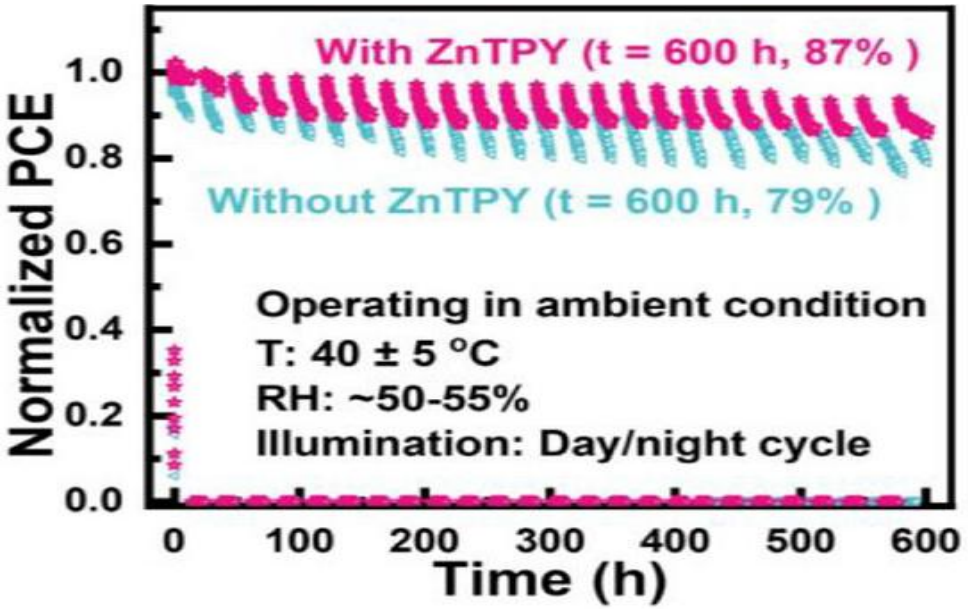
PSCs	J_{SC} (mAcm^{-2})	V_{OC} (V)	FF	PCE (%)
Control	24.23	1.148	74.87	20.83
Zn-TPY	24.95	1.180	76.78	22.60



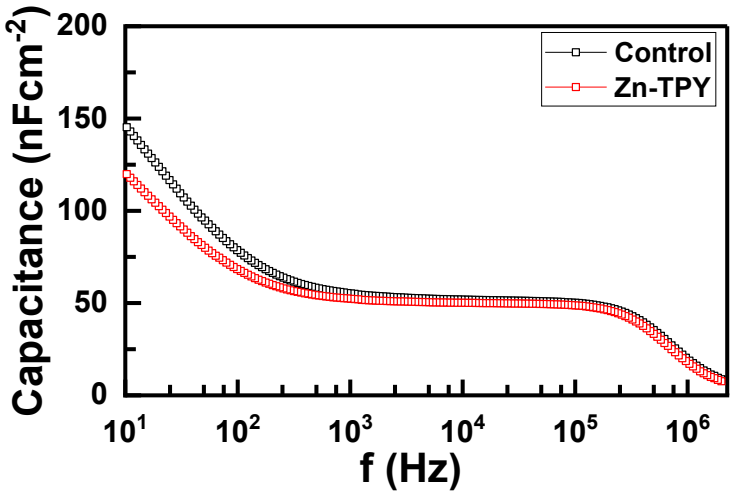






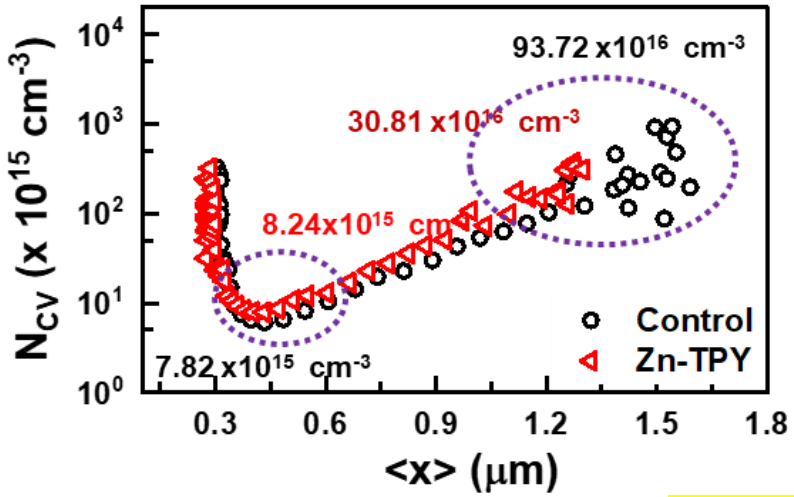


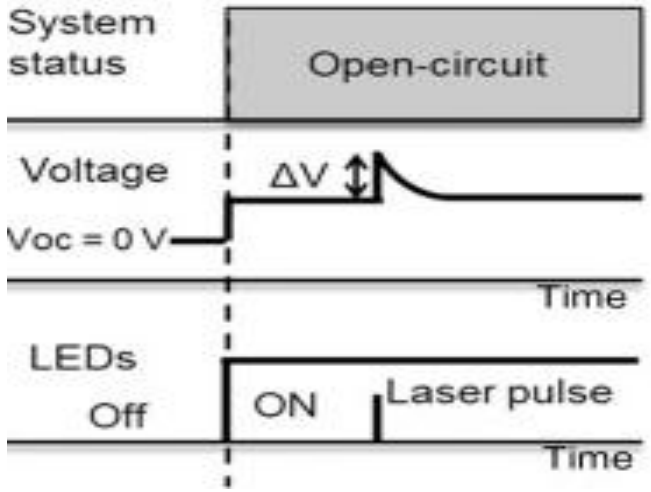
CONASHs-Pb-PSCs: Capacitance analysis/ Transient –photo characteristics



$$C(V)^{-2} = -\frac{2}{q\varepsilon_0\varepsilon_s N_A(x)}(V + V_D)$$

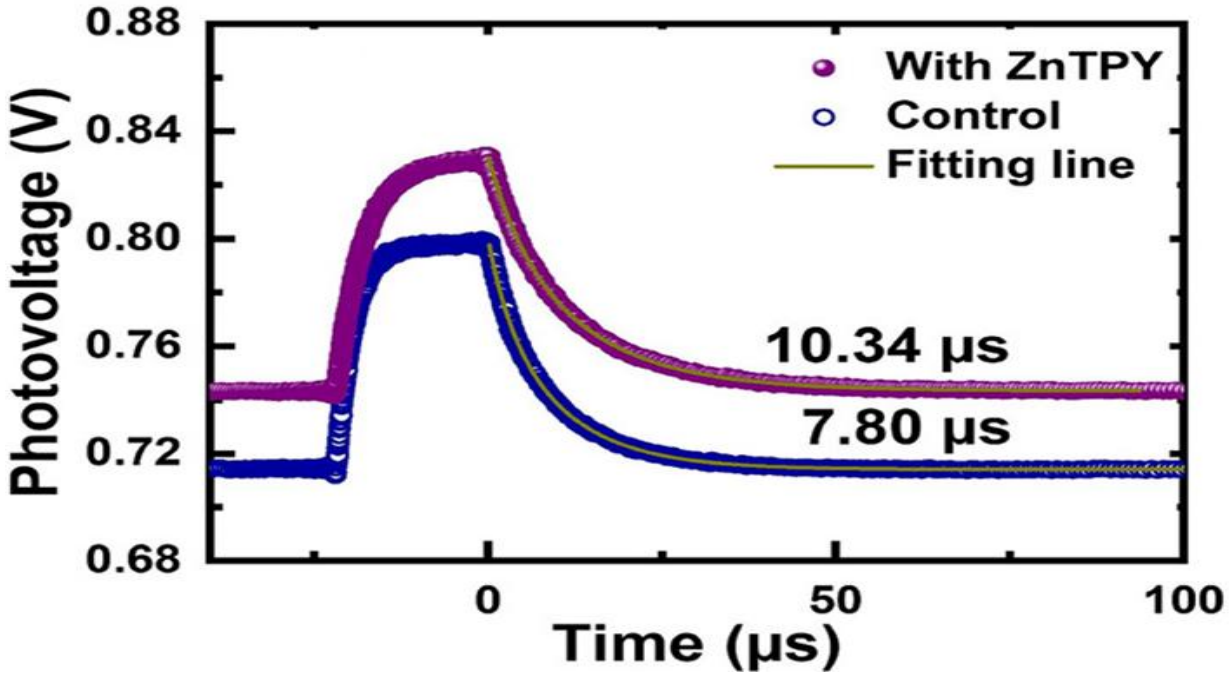
Suppression of defect profile with Zn-TPY additive





Transient Photovoltage Analysis

$$V(t) = V_0 e^{-t/\tau}$$



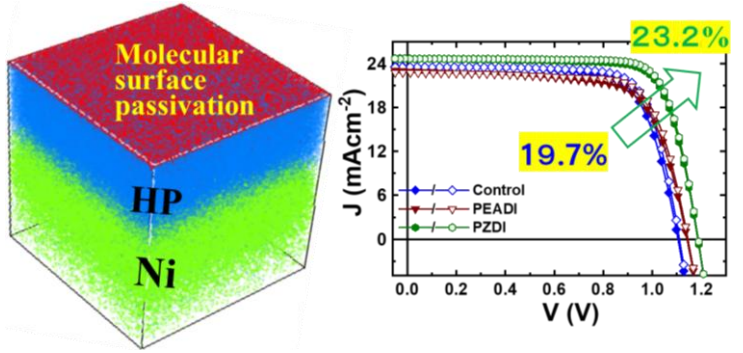
Khadka –NIMS /Wang (NSYSU) et al. Small (2026): e76016

Longer lifetime: Defect mitigation

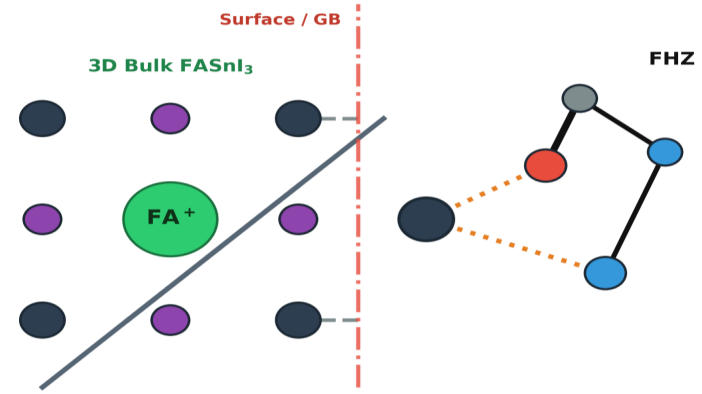
Note-Summary

Pb-PSCs: Charge-modulated amine/Flouroarene affect

- ***Modulates surface chemistry and surface charge modulation***
- ***Forms interfacial embedded 2D crystallite***



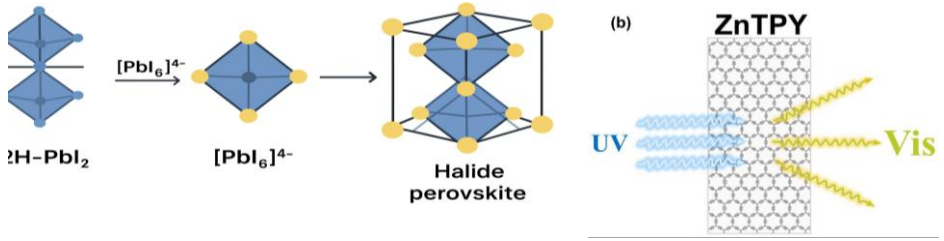
- Fullerene functional derivatives: modulate film growth and *control the e xtent of oxidation of Sn^{2+}*
- **Hydrazide additive in $FASnI_3$** > *Facilitates uniform morphology with b etter crystal quality*



- **CONASH in Sn/Pb- Perovskite**

>Precursor modulation: forming a well-dispersed Zn-TPY: PbI_2 or SnI_2 complex

>form heterogeneous nucleation ($[SnI_{6-x}]^{4-}$ octahedral) > Sn/Pb-perovskite film growth



$[(PbI_{6-x})^{4-}$–ZnTPY]

Acknowledgement





Thank you for your attention

Information of Invited Speaker

