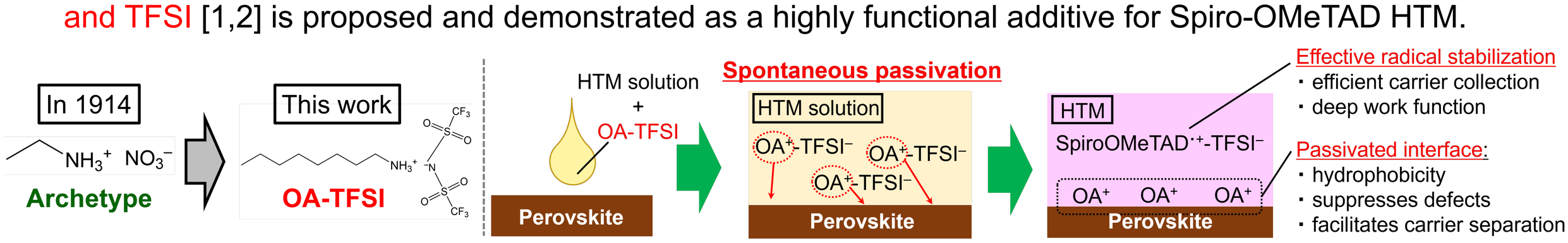
Aliphatic Primary Ammonium Bis(trifluoromethylsulfonyl)imide as a Highly Functional Additive for Hole Transport Material Layer in Perovskite Solar Cells

Introduction

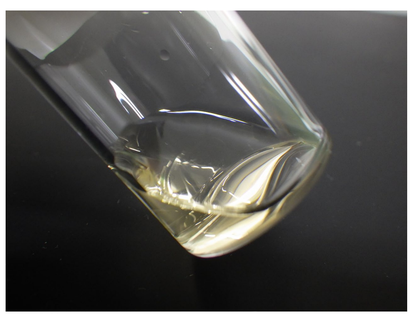
- Room-temperature ionic liquids (RTILs) based on bis(trifluoromethylsulfonyl)imide (TFSI) anion have been reported as promising Li-free additives for hole transport materials (HTMs) in perovskite solar cells (PCSs).
- However, cation designs of RTILs for PSC applications so far have been limited within currently major ones (e.g., imidazoles), so their functions have been confined particularly in controlling HTM/perovskite interface.
- In this work, an RTIL comprising an archetypal aliphatic primary ammonium (i.e., *n*-octylammonium: OA) and TFSI [1,2] is proposed and demonstrated as a highly functional additive for Spiro-OMeTAD HTM.



Methods

OA-TFSI

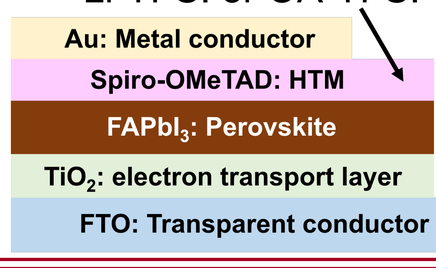
- **Synthesized by an ion exchange method**
- **Melting point: < 190 K**

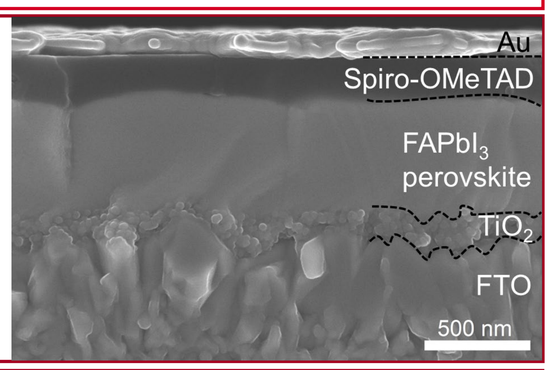


[Cell configuration]
FTO/ TiO_2/ $FAPbI_3$ perovskite / Spiro-OMeTAD/ Au
(active area: 0.12 cm^2)

[Deposition method]
Spin-coating in dry air
(one-step for perovskite)

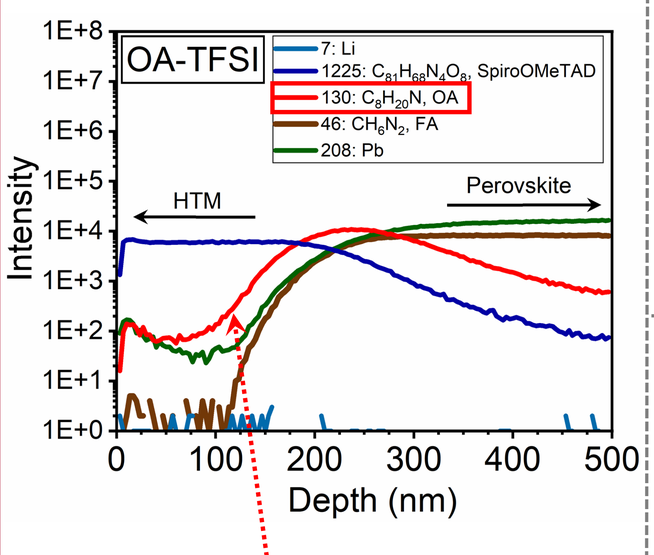
[HTM additive]
Li-TFSI or OA-TFSI





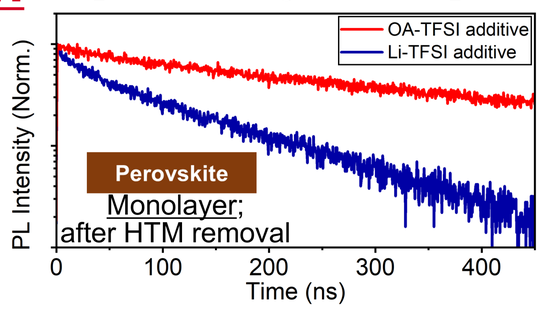
Results and discussion

[Compositional depth analysis]



OA cations spontaneously covered perovskite surface under the HTM deposition

[Carrier dynamics]



OA passivation suppressed defects over perovskite surface

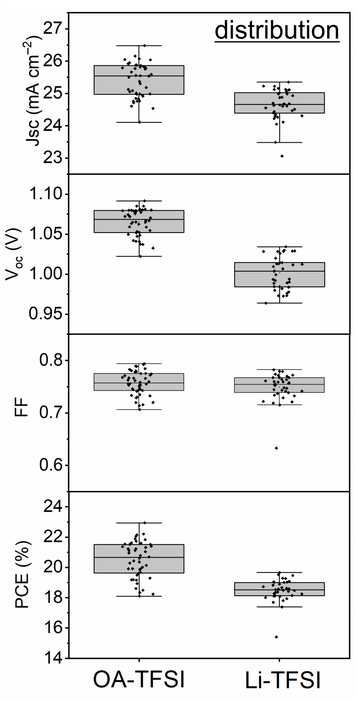
[HTM radical concentration]

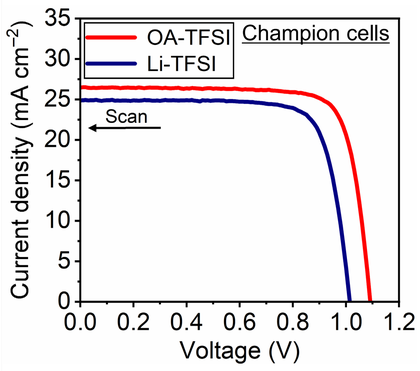
Additive (optimal amount)	Work function (eV)
12 mM OA-TFSI	5.56
48 mM Li-TFSI	5.49

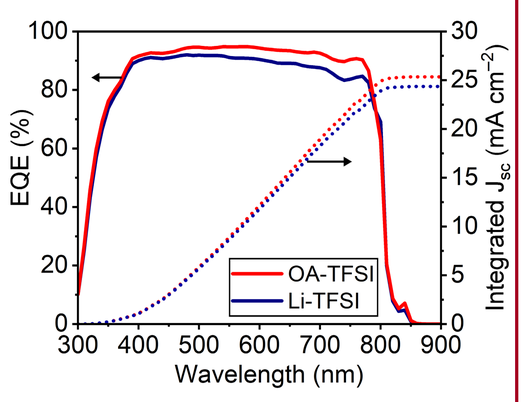
∗ measured by photoelectron yield spectroscopy

The deeper work function suggests higher HTM radical concentration

[Photovoltaic performances]





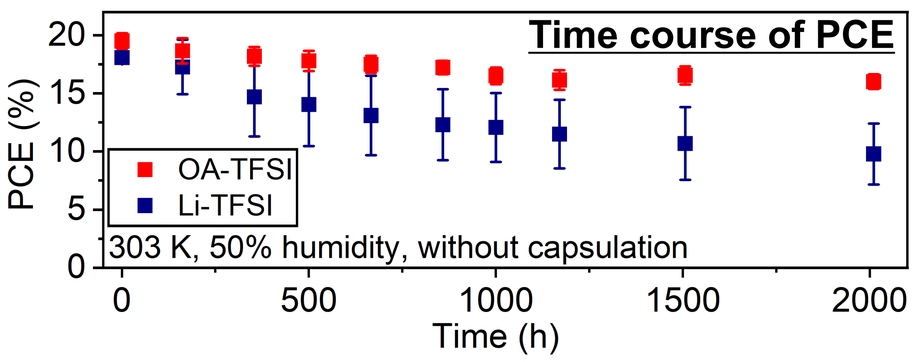


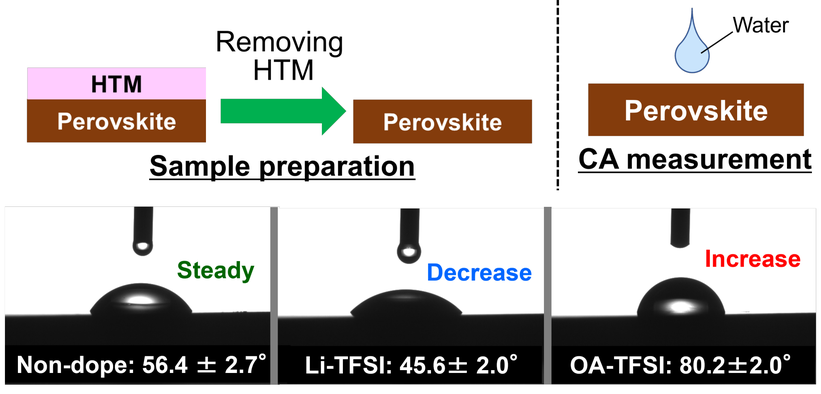
Sample	Scan	Jsc (mA/cm^2)	Voc (V)	FF	η (%)
OA-TFSI	Backward	26.5	1.09	0.79	22.9
	Forward	26.5	1.07	0.72	20.4
Li-TFSI	Backward	24.9	1.01	0.78	19.7
	Forward	25.1	1.02	0.70	17.8

∗ Quasi-steady-state PCE for OA-TFSI sample: 22.2%

OA-TFSI additive enhanced initial photovoltaic performances owing to the improved hole collection and its passivation effects

[Hydrophobicity of perovskite surface]



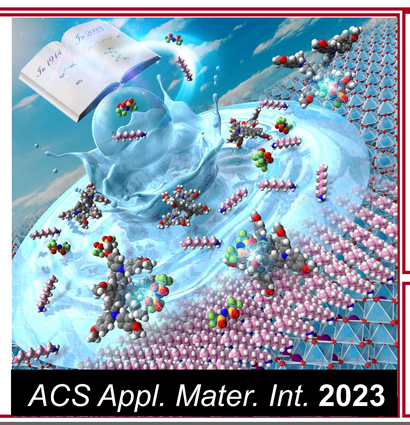


Pristine perovskite (before HTM deposition): 58.8± 1.3°

The spontaneous passivation by OA-TFSI rendered perovskite hydrophobic, and thereby improved long-time PCE stability in the presence of humidity

Conclusion

OA-TFSI exhibited crucial benefits simultaneously: (i) spontaneous perovskite passivation under HTM deposition process, and (ii) effective stabilization and generation of cationic Spiro-OMeTAD radicals, improving PCS performances involving their stability.



References

[1] N. Nishimura et al., *Chem Rxiv* **2023**, preprint, DOI: 10.26434/chemrxiv-2023-hsh8k.

[2] Y. Kim et al., *Energy Environ. Sci.* **2023**, *16*, 2226−2238.

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