Perspective on perovskite indoor photovoltaics

Cite as: Appl. Phys. Lett. 122, 130501 (2023); doi: 10.1063/5.0147747 Submitted: 25 February 2023 · Accepted: 16 March 2023 · Published Online: 28 March 2023







Chun-Hao Chen, 🕞 Zhao-Kui Wang, 1 🕞 and Liang-Sheng Liao 🕞

AFFILIATIONS

Institute of Functional Nano & Soft Materials (FUNSOM), Jiangsu Key Laboratory of Advanced Negative Carbon Technologies, Soochow University, Suzhou 215123, China

^{a)}Author to whom correspondence should be addressed: zkwang@suda.edu.cn

ABSTRACT

The concept of the Internet of Things (IoT) is a future development opportunity for mankind, which is a system that realize the interaction of various electronic devices through wireless communication. With the rise and development of this concept, the energy demand gap of self-powered equipment in IoT has emerged. The construction of an off-grid power system will make the wireless network of IoT easy to integrate and meet the higher requirements of power supply equipment in terms of size, weight, energy-consumption, and cost. Indoor photovoltaics (IPVs) can provide stable and long-term power guarantee by collecting indoor light, which are perfectly matched with IoT. Photovoltaic cells based on a-silicon, dye, organic compounds, and halide perovskite have been proved to be suitable for IPVs. Among them, perovskite indoor photovoltaics (PIPVs) have attracted much attention due to its advantages of tunable bandgap, high output voltage, flexible preparation, and low cost. In addition, the indoor stability and toxicity of PIPVs are also discussed in this Perspective.

Published under an exclusive license by AIP Publishing. https://doi.org/10.1063/5.0147747

I. INTRODUCTION

The Internet of Things (IoT) is a network based on the Internet, traditional telecommunications networks, and other information carriers, enabling all ordinary physical objects that can be independently addressed to achieve interconnection. 1,2 This concept has developed rapidly since it was put forward at the end of the 20th century. In this process, the number of self-powered devices (such as sensors, terminal devices, and health monitoring) in IoT has expanded rapidly, and the gap in energy demand has become increasingly obvious. Since the IoT system operates through the wireless network, the construction of an off-grid power system can realize the terminal integration and improve the operation stability and independence of this system. At this stage, the main portable power supply is still batteries. However, low power-mass ratio and intermittent battery replacement will lead to instability and high energy consumption of the IoT system. Therefore, we put forward higher requirements on the size, weight, energy consumption, and cost of the off-grid power supply equipment.³ Because indoor photovoltaics (IPVs) can meet these requirements, it has become an important energy support technology for the IoT.4-7 Amorphous silicon solar cells (a-Si),8 dye-sensitized solar cells (DSSCs), 6,9-11 organic photovoltaics (OPVs), 5,12,13 and perovskite solar cells (PSCs)¹⁴ have been proved to be suitable for IPVs.

Due to the huge differences in intensity $(300 \,\mu\text{W/cm}^2)$ and 100 mW/cm²) and wavelength (350–750 and 150–4000 nm) between indoor and outdoor light sources, and mature photovoltaic devices (PVs) that work efficiently under AM 1.5G do not always perform well under indoor conditions. Therefore, unlike the ideal bandgap of outdoor PSCs (~1.34 eV), the absorption layer with a bandgap of 1.8-2.0 eV can completely cover the indoor spectrum and produce high optical voltage, which is considered as the best choice for IPVs. 15,16 Because a relatively stable voltage needs to be realized to drive the self-powered equipment in the IoT system. The IPVs with high voltage output has competitive advantages by reducing the number of series units and reducing costs. Perovskite photovoltaics are ahead of other mainstream IPVs because it can achieve an optical voltage of more than 1.2 V under 1000 lux light source 17,18 (~ 0.7 V for a-Si, $^8 \sim 1.0$ V for DSSCs, and ~1.1 V for OPVs⁵) The tunable bandgap of perovskite materials has become the key factor for achieving high power conversion efficiency (PCE) of PSCs in both indoor and outdoor conditions. 19-21 Especially, compared with the widely deployed a-Si cells, the voltage of perovskite indoor photovoltaics (PIPVs) under indoor light has increased by more than 70%. In addition, in line with outdoor PSCs, low cost and flexible preparation are also inherent advantages for perovskite indoor photovoltaics (PIPVs). 22,23 The cost of PSCs is estimated to be about 1/3 of that of a-Si solar cells.²⁴ The toxicity of lead is particularly prominent in indoor photovoltaic applications, which is a key issue to be solved before putting into the market.^{25,26} It is worth noting that the instability of outdoor PSCs is mainly caused by the strong fluctuation of device temperature and device damage under adverse conditions (such as hail), which can be effectively

avoided in indoor condition. Therefore, the indoor stability of perovskite photovoltaics is greatly improved.²⁷ PIPVs are easier to meet the requirements of entering the market.

II. TUNABLE BANDGAP AND HIGH VOLTAGE OF PIPVS

The fundamental difference between indoor and outdoor photovoltaic power generation is the huge difference in the intensity and wavelength of the received light source. IPVs work under low intensity diffuse light mainly in the visible spectrum (350–750 nm), provided by compact fluorescent lamps (CFLs), light emitting diodes (LEDs), and other artificial light sources as shown in Fig. 1(a).⁴ While outdoor PSCs work under AM 1.5G, with spectrum from ultraviolet (150 nm) to mid-infrared (4000 nm). Therefore, it is critical to accurately evaluate the photovoltaic performance of IPVs.²⁸ In our daily life, the intensity of various indoor light sources we use is about 200–1000 lux. The following formula can be used to calculate the illuminance of the indoor light source using the corresponding incident power²⁹

$$E_{\nu} = K_m \times \int_0^{\infty} E_{\lambda} \times V(\lambda) d_{\lambda},$$

where $V(\lambda)$ indicates CIE spectral brightness factor of human light vision, K_m equals 683 lm/W. In order to calibrate the indoor light source, Wong *et al.* proposed to use the universal LED light meter of the National Institute of Standards and Technology (NIST)-traceable calibration.²⁹ Hou *et al.* suggested that accurate IPVs testing as shown in Fig. 1(b) should include the following elements.³⁰ (1) The instability of the intensity of the indoor photovoltaic measuring light source should be less than 2%. (2) Optical mask is required for testing. (3) Spectrometer rather than illuminometer is more suitable for calibration

of light source, with improved accuracy. (4) The difference between external quantum effiency (EQE) integrated current density and actual current density should not exceed 5%. The incident power density of indoor light sources (CFL and LED) with 1000 lux illuminance is about $300 \, \mu \text{W/cm}^2$, so the current density (I_{sc}) limit of IPVs is estimated to be about $140 \, \mu \text{A/cm}^2$. In some cases, PIPVs show that the current density exceeds the limit, which is attributed to the inaccurate test method. Among them, the non-parallelism of indoor light and stray light are the main reasons for the measurement error. In addition, larger area devices can obtain more accurate PCE and I_{sc} .

Due to the difference between indoor and outdoor spectra, mature perovskite photovoltaic devices with high performance under 1 sun do not always perform well under indoor light. Different from the bandgap of 1.1-1.6 eV of high-performance outdoor PSCs, the optimal bandgap of PIPVs is 1.8-2.0 eV. PIPVs with this range of bandgap can fully absorb the indoor full-spectrum light source and produce high optical voltage. Unlike the restrictedly adjustable bandgap width of a-Si, DSSCs, and OPVs, perovskite materials have a wide range of precisely tunable bandgap, making them particularly attractive in IPVs. The perovskite crystal is described as the octahedral structure of ABX₃, where A is the monovalent cation [CH₃NH₃⁺ (MA), $HC(NH_2)_2^+$ (FA), Cs], B is the divalent metal cation (Pb, Sn), and X is the halide anion (I, Br, Cl). As shown in the Fig. 1(c), the bandgap of perovskite can be continuously adjusted from infrared (1.15 eV) to ultraviolet (up to 3 eV) through the composition engineering of mixed cations or anions.³² At present, the mainstream system of outdoor PSCs is FAPbI3-based narrow bandgap perovskite with a bandgap of about 1.5 eV.33 Due to its mature preparation process, many works have applied photovoltaic devices of this system into

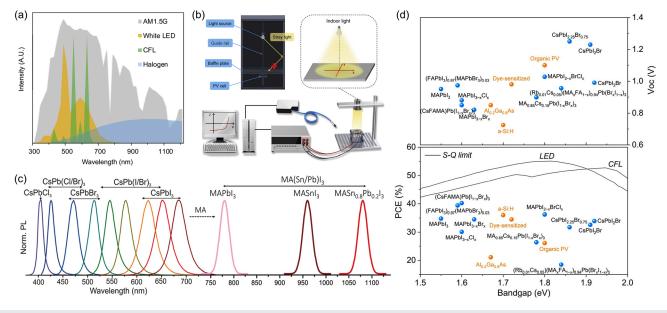


FIG. 1. (a) The different light spectra of AM 1.5G, white LED, CFL, and halogen. Reprinted with permission from Mathews *et al.*, Joule 3, 1415 (2019). Copyright 2019 Elsevier Inc. (b) The typical setup for indoor PCE measurement and schematic illustrations. Reprinted with permission from Cui *et al.*, Joule 5, 1016 (2021). Copyright 2021 Elsevier Inc. (c) The tunable bandgap of perovskite from infrared to ultraviolet through the composition engineering of mixed cations or anions. Reprinted with permission from Saliba *et al.*, Angew. Chem., Int. Ed. 57, 2554 (2018). Copyright 2018 Wiley-VCH. (d) PCE and Voc summary of PIPVs in comparison with other IPV technologies. Reprinted with permission from Guo *et al.*, ACS Energy Lett. 8, 90 (2023). Copyright 2022 American Chemical Society.

IPVs and achieved high PCE. However, its narrow bandgap characteristic determines the low optical voltage in indoor photovoltaic applications. Therefore, appropriate perovskite bandgap width needs to be developed and used in indoor photovoltaic scenes. The performance of PIPVs and other IPVs are summarized in Fig. 1(d).²⁷ The MA/FAbased system used by the mainstream outdoor perovskite solar cells is not prominent in the indoor voltage performance due to the bandgap limitation. Taking into account the matching of bandgap and indoor light source spectrum (350-750 nm), CsPbBrI₂ (1.89 eV) and CsPbI₃ (1.7 eV) can be used to prepare high performance PIPVs. 34,3 Compared with other IPV devices [such as a-Si (0.73 V, 36% PCE), DSSCs (0.98 V, 35% PCE), and OPVs (1.10 V, 26.1% PCE)], 5 PIPVs achieve an open-circuit voltage of more than 1.20 V due to the matched bandgap width under 1000 lux indoor illumination. Guo et al. employed the perovskite system of CsPbI_{2,25}Br_{0,75} under indoor applications.¹⁸ After using 2,5-thiophenedicarboxylic acid to passivate the surface defects of perovskite, a recorded photovoltage of 1.23 V with the PCE of 32.41% was obtained under 1000 lux illuminance [Fig. 2(a)]. In 2020, Kim et al. employed high-gap triple layers to improve the performance of a-Si solar cells.8 As a result, a recorded a-Si indoor PCE of 36.0% (under 3000 lux) with the voltage of 0.730 V was achieved. Under indoor light, there is a huge gap between the voltage of a-Si solar cells and that of PIPVs. Because indoor small electrical appliances always need a specific voltage to realize the drive (e.g., 1.5

and $5.0\,\mathrm{V}$), stable photovoltage of IPVs is particularly critical. PIPVs have competitive advantages by reducing the number of series units and reducing costs.

III. FLEXIBLE PIPVS

Another important advantage of PIPVs is that perovskite thin films can be deposited by solution method. Low cost and high efficiency production are realized on the flexible polymer substrate. Flexible perovskite photovoltaic products provide a series of new application possibilities based on the IoT system due to its unique advantages, such as low weight, high power-mass ratio, and high Compared with rigid devices, flexible PSCs can provide a more suitable surface for IoT self-powered devices. Through this technology, customers' needs in specific applications can be met and customization can be provided. Similarly, flexible OPVs can also be used for indoor applications. However, due to their large open-circuit voltage loss and non-optimal absorption bandgap, their PCE always hovers below 30%.³⁸ There remains a gap between their efficiency and PIPVs'. Typically, Cui et al. designed a non-fullerene receptor to obtain the absorption spectrum matching with the indoor spectrum.⁵ The lower energy loss makes the device obtain the champion PCE of 26.1% under 1000 lux (2700 K) with a good stability. On the other hand, Chen et al. studied the flexible PIPVs for the first time.³⁹ A three-dimensional cross-linking agent called borax was employed to

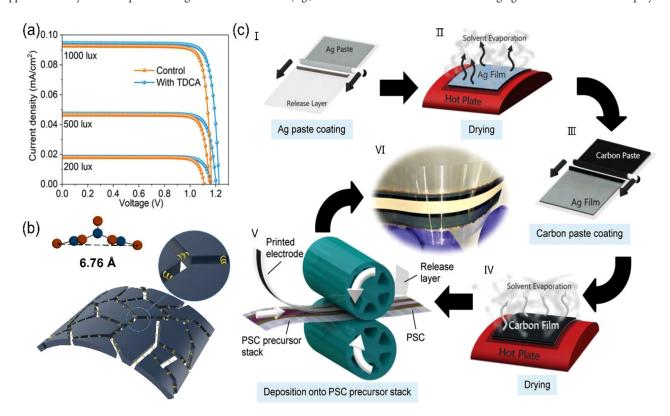


FIG. 2. (a) *J–V* curves measured under an LED light with different intensities in the reverse scan. Reprinted with permission from Guo *et al.*, Adv. Funct. Mater. **32**, 2207554 (2022). Copyright 2022 Wiley-VCH. ¹⁸ (b) Schematic diagram after bending of the flexible films with stretch forms of borax. Reprinted with permission from Chen *et al.*, Adv. Mater. **34**, 2200320 (2022). Copyright 2022 Wiley-VCH. ³⁹ (c) Flow-chart illustrating the fabrication steps of the flexible printed electrodes. Reprinted with permission from Sutherland *et al.*, Adv. Energy Mater. **12**, 2202142 (2022). Copyright 2022 Wiley-VCH. ⁴³

achieve all-round stress release for perovskite films as shown in Fig. 2(b). The mechanical stability was enhanced under large temperature changes and external forces. A recorded flexible PIPVs PCE of 31.85% was obtained under 1062 lux illuminance (LED, 2956 K). In order to further reduce the cost of flexible perovskite photovoltaic devices, flexible manufacturing technology based on large areas need to be developed. The continuous roll-to-roll (R2R) process with fast fabrication speed and low cost meets the requirement for IoT system. ^{40–42} In 2022, Sutherland *et al.* introduced a new vacuum-free R2R compatible method to deposit electrodes based on conductive pastes. ⁴³ Except for transparent conductive electrodes, all functional layers are produced by vacuum-free deposition [Fig. 2(c)]. The devices obtained a record PCE of up to 16.7% with outstanding operational and mechanical stability. This large-area flexible PIPV preparation process shows competitive cost advantages for the application in the IoT system.

IV. STABILITY ISSUES OF PIPVS

The stability of PSCs is still an important issue that needs to be studied and optimized. The effect of water and oxygen on PSCs can be almost eliminated by the advanced encapsulation technology. However, the photo-induced damage to perovskite caused by sunlight is inevitable. Brennan *et al.* concluded that the light-induced structural instabilities of APb(I_{1-x}Br_x)₃ attribute to the reversible halide phase segregation as shown in Fig. 3(a). The fundamental reason for the bandgap difference is the physical change of the Pb–X bond distance. Accordingly, increasing the proportion of bromine will reduce the average bond distance of Pb–X, resulting in lattice shrinkage and corresponding increase in the bandgap. The halide phase segregation is reversible in the dark, with the recovery of the absorbance and

photoluminescence spectra. As a result, the phase segregation of perovskite under indoor light can be greatly reduced. All-inorganic perovskite CsPb(I_{1-x}Br_x)₃ can also effectively suppress the photoinduced segregation. Additionally, Liang et al. revealed the origins and influences of Pb0 induced by light and x-ray [Fig. 3(b)]. 46 On the other hand, the sharp increase in the operating temperature of the devices caused by sunlight will lead to the decline of the dynamic stability, causing the attenuation of the device performance.⁴⁷ However, the intensity of indoor light source is far lower than that of sunlight. As a result, the impact of light source on perovskite has also changed greatly due to different application scenarios. Under indoor conditions, the temperature of PIPVs will not increase as it does outside during normal operation. Because PIPVs have no radiation (such as ultraviolet), temperature, and humidity fluctuations when working in the indoor environment, whose stability can be improved and working time can be greatly extended. For example, Guo et al. found the intense redshift in both photo-luminescence (PL) and UV-vis absorption under 1 sun irradiation for 20 min [Fig. 3(c)] did not appear under LED light [Fig. 3(d)].²⁷ Therefore, the indoor stability of perovskite photovoltaic is greatly improved due to its mild application environment. In addition, Kim et al. revealed the effect of chloride ion on the performance of PIPVs. 48 By reducing the bulk defect density of the perovskite layer, the ion migration behavior has been proved to be greatly reduced. PIPVs are easier to meet the requirements of entering the market.

V. TOXICITY OF LEAD

Since PIPVs is more likely to contact with humans, the toxicity of PSCs is also an important factor in the final market selection. In 2011,

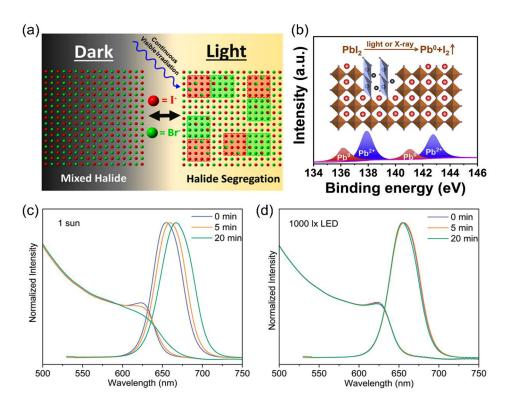


FIG. 3. (a) Schematic diagram of the phase segregation of perovskite under illumination. Reprinted with permission from Brennan et al., ACS Energy Lett. 3, 204 (2018). Copyright 2017 American Chemical Society. (b) Schematic diagram on the origins and influences of Pb⁰ for PSCs. Reprinted with permission from Liang et al., Joule 6, 816 (2022). Copyright 2022 Elsevier Inc. (b) Land UV—vis spectra variation of perovskite films under the illumination of (c) sunlight and (d) LED light. Reprinted with permission from Guo et al., ACS Energy Lett. 8, 90 (2023). Copyright 2022 American Chemical Society.

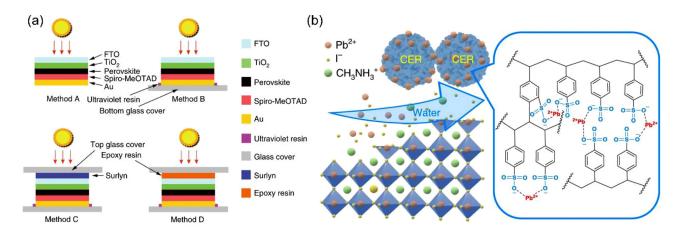


FIG. 4. (a) Schematic showing the four different encapsulation methods. Reprinted with permission from Jiang *et al.*, Nat. Energy **4**, 585 (2019). Copyright 2019 Springer Nature Limited. (b) Schematic of how CERs prevent lead leakage. Reprinted with permission from Chen *et al.*, Nat. Energy **5**, 1003 (2020). Copyright 2020 Springer Nature Limited. (a) Schematic of how CERs prevent lead leakage. Reprinted with permission from Chen *et al.*, Nat. Energy **5**, 1003 (2020). Copyright 2020 Springer Nature Limited.

the European Union issued the "Directive on Restricting the Use of Certain Hazardous Substances in Electrical and Electronic Equipment (RoHS)," which stipulates that the maximum concentration of lead in any electronic equipment is limited to 0.1% by weight. 49 However, the content of lead in perovskite is more than 10%. Moreover, the areal density of lead in common PSCs reaches 0.07 mg/cm^{2.50} The higher the level of lead exposure and the younger the person (especially the fetus), the greater the toxicity is to the body. Timely monitoring of lead concentration in blood has become an important means to prevent and control lead poisoning internationally. When the blood lead concentration of children exceeds 100 µg/L, it should be diagnosed as hyperlipemia. More than 200 µg/L should be diagnosed as lead poisoning in children.⁵¹ After calculation, the lead leakage from a 1.4 cm² device without encapsulation will cause the blood lead concentration of young children to exceed the threshold.⁵⁰ In fact, the area of perovskite photovoltaic modules will be much larger than this order of magnitude. Therefore, isolation strategies between PSCs and humans, including space distance limitation and effective packaging strategies, should be developed. Jiang et al. investigated three encapsulation methods [Fig. 4(a)] for PSC modules and conducted the simulated hail damage experiment to test the lead leakage rate.⁵² Among them, epoxy resin-based polymers added between the perovskite layer and the top glass cover can effectively reduce the lead leakage from 30 to 0.08 mg h⁻¹ m⁻². In addition, Chen et al. employed the low-cost cation-exchange resins (CERs) to trap lead in PSCs as shown in Fig. 4(b).⁵³ The Pb leakage rate of the treated devices was reduced by 62-fold to 14.3 ppb. Especially, Yang et al. applied lead-free tin-based perovskites to PIPVs first in 2021. 54 They introduced catechin into perovskite layers to inhibit oxidation of Sn2+, resulting in FA0.75MA0.25SnI2Br films with lower Sn4+ ratio. The final optimized devices achieved a maximum PCE of 12.81% under 1000 lux. This work further advanced the potential of lead-free PSCs for IPV applications.

VI. CONCLUSIONS AND OUTLOOK

The IoT has developed rapidly in recent years, accompanied by a huge energy gap of self-powered equipment in the system. IPVs have become the key technology supporting the energy supply for the IoT system. PIPVs have attracted much attention due to its advantages such as tunable bandgap, high optical voltage, and flexible preparation. For the future development of this technology, the following points need to be followed and optimized. First, take advantage of the adjustable bandgap of perovskite materials and control it in the range of 1.8–2.0 eV to obtain higher indoor optical voltage. Second, further promote the flexible large-area R2R printing technology to reduce the preparation cost. In adition, develop a customized service system for flexible devices to meet the needs of customers in specific applications. Third, further solve the lead toxicity of perovskite, including but not limited to more effective encapsulation technology and the development of lead-free perovskite materials.

The authors acknowledge the financial support from the National Natural Science Foundation of China (Nos. 62075148, 52073197, and 52273189), the Natural Science Foundation of Jiangsu Province (Nos. BE2022026-2, BK20201413, and BK20211314), and the Suzhou science and technology plan project (Nos. N321461821 and ST202212). This work was supported by the Suzhou Key Laboratory of Functional Nano & Soft Materials, the Collaborative Innovation Center of Suzhou Nano Science & Technology, the 111 Project, the Joint International Research Laboratory of Carbon-Based Functional Materials and Devices, and the Soochow University Tang Scholar.

AUTHOR DECLARATIONS Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Chun-Hao Chen: Data curation (lead); Investigation (lead); Methodology (lead); Writing – original draft (lead). Zhao-Kui Wang: Conceptualization (lead); Funding acquisition (lead); Investigation (equal); Supervision (lead); Writing – review & editing (lead). Liang-Sheng Liao: Conceptualization (supporting); Supervision (equal); Writing – review & editing (supporting).

DATA AVAILABILITY

The data that support the findings of this study are available within this article.

REFERENCES

- ¹L. Atzori, A. Iera, and G. Morabito, Comput. Networks 54, 2787 (2010).
- ²F. Xia, L. T. Yang, L. Wang, and A. Vinel, Int. J. Commun. Syst. **25**, 1101 (2012).
- ³S. Li, L. D. Xu, and S. Zhao, Inf. Syst. Front. 17, 243 (2015).
- ⁴I. Mathews, S. N. Kantareddy, T. Buonassisi, and I. M. Peters, Joule 3, 1415 (2019).
- ⁵Y. Cui, Y. Wang, J. Bergqvist, H. Yao, Y. Xu, B. Gao, and J. Hou, Nat. Energy 4, 768 (2019).
- ⁶M. Freitag, J. Teuscher, Y. Saygili, X. Zhang, F. Giordano, P. Liska, and A. Hagfeldt, Nat. Photonics 11, 372 (2017).
- ⁷J. K. W. Ho, H. Yin, and S. K. So, J. Mater. Chem. A 8, 1717 (2020).
- ⁸G. Kim, J. W. Lim, J. Kim, S. J. Yun, and M. A. Park, ACS Appl. Mater. Interfaces 12, 27122 (2020).
- ⁹D. Zhang, M. Stojanovic, Y. Ren, Y. Cao, F. T. Eickemeyer, E. Socie, N. Vlachopoulos, J. E. Moser, S. M. Zakeeruddin, A. Hagfeldt, and M. Grätzel, Nat. Commun. 12, 1777 (2021).
- ¹⁰D. Song, M. Li, Y. Li, X. Zhao, B. Jiang, and Y. Jiang, ACS Appl. Mater. Interfaces 6, 7126 (2014).
- ¹¹S. Mathew, A. Yella, P. Gao, R. Humphry-Baker, B. F. E. Curchod, N. Ashari-Astani, I. Tavernelli, U. Rothlisberger, M. K. Nazeeruddein, and M. Gratzel, Nat. Chem. 6, 242 (2014).
- ¹²N. Gasparini, A. Salleo, I. McCulloch, and D. Bara, Nat. Rev. Mater. 4, 229 (2019).
- ¹³J. Zhao, Y. Li, G. Yang, K. Jiang, H. Lin, H. Ade, W. Ma, and H. Yan, Nat. Energy 1, 15027 (2016).
- ¹⁴M. Saliba, T. Matsui, K. Domanski, J.-Y. Seo, A. Ummadisingu, S. M. Zakeeruddin, J.-P. Correa-Baena, W. R. Tress, A. Abate, A. Hagfeldt, and M. Grätzel, Science 354, 206 (2016).
- ¹⁵ K. L. Wang, Y. G. Yang, Y. H. Lou, M. Li, F. Igbari, J. J. Cao, J. Chen, W. F. Yang, C. Dong, L. Li, R. Z. Tai, and Z. K. Wang, eScience 1, 53 (2021).
- ¹⁶M. Li, F. Igbari, Z. K. Wang, and L. S. Liao, Adv. Energy Mater. 10, 2000641 (2020).
- T. Guo, A. K. Jena, I. Takei, M. Ikegami, A. Ishii, Y. Numata, N. Shibayama, and T. Miyasaka, Adv. Funct. Mater. 31, 2103614 (2021).
 T. Guo, S. Zhao, N. Shibayama, A. Kumar Jena, I. Takei, and T. Miyasaka, Adv.
- ¹⁸Z. Guo, S. Zhao, N. Shibayama, A. Kumar Jena, I. Takei, and T. Miyasaka, Adv. Funct. Mater. **32**, 2207554 (2022).
- ¹⁹M. Li, C. Zhao, Z. K. Wang, C. C. Zhang, H. K. H. Lee, A. Pockett, J. Barbé, W. C. Tsoi, Y. G. Yang, M. J. Carnie, X. Y. Gao, W. X. Yang, J. R. Durrant, L. S. Liao, and S. M. Jain, Adv. Energy Mater. 8, 1801509 (2018).
- ²⁰C. Dong, X. M. Li, C. Ma, W. F. Yang, J. J. Cao, F. Igbari, Z. K. Wang, and L. S. Liao, Adv. Funct. Mater. 31, 2011242 (2021).
- ²¹N. J. Jeon, J. H. Noh, W. S. Yang, Y. C. Kim, S. Ryu, J. Seo, and S. I. Seok, Nature 517, 476 (2015).
- ²²K. L. Wang, X. M. Li, Y. H. Lou, M. Li, and Z. K. Wang, Sci. Bull. 66, 347 (2021).
- ²³K. Wojciechowski and D. Forgács, ACS Energy Lett. 7, 3729 (2022).
- ²⁴J. Ling, P. K. K. Kizhakkedath, T. M. Watson, I. Mora-Seró, L. Schmidt-Mende, T. M. Brown, and R. Jose, Sol. RRL 5, 2100401 (2021).
- ²⁵W. Ke and M. G. Kanatzidis, Nat. Commun. **10**, 965 (2019).
- ²⁶A. Abate, Joule 1, 659 (2017).

- ²⁷Z. Guo, A. K. Jena, and T. Miyasaka, ACS Energy Lett. **8**, 90 (2023).
- ²⁸K. L. Wang, Y. H. Zhou, Y. H. Lou, and Z. K. Wang, Chem. Sci. 12, 11936 (2021).
- ²⁹A. Venkateswararao, J. K. W. Ho, S. K. So, S. W. Liu, and K. T. Wong, Mater. Sci. Eng., R 139, 100517 (2020).
- 30Y. Cui, L. Hong, T. Zhang, H. Meng, H. Yan, F. Gao, and J. Hou, Joule 5, 1016 (2021).
- ³¹H. Y. Lin, C. Y. Chen, B. W. Hsu, Y. L. Cheng, W. L. Tsai, Y. C. Huang, C. S. Tsao, and H. W. Lin, Adv. Funct. Mater. 29, 1905163 (2019).
- ³²M. Saliba, J. P. Correa-Baena, M. Grätzel, A. Hagfeldt, and A. Abate, Angew. Chem., Int. Ed. 57, 2554 (2018).
- ³³C. H. Chen, F. Hu, Z. H. Su, Y. J. Yu, K. L. Wang, Y. R. Shi, J. Chen, Y. Xia, X. Y. Gao, Z. K. Wang, and L. S. Liao, Adv. Funct. Mater. 33, 2213661 (2023).
- ³⁴M. A. Green, Nat. Energy **1**, 15015 (2016).
- ³⁵C. C. Chueh, C. Z. Li, and A. K. Y. Jen, Energy Environ. Sci. **8**, 1160 (2015).
- ³⁶Z. Chen, Q. Cheng, H. Chen, Y. Chen, Y. Wu, J. Ding, X. Wu, H. Yang, H. Liu, W. Chen, X. Tang, X. Lu, Y. Li, and Y. Li, Adv. Mater. 35, 2300513 (2023).
- ³⁷L. Li, Y. Wang, X. Wang, R. Lin, X. Luo, Z. Liu, K. Zhou, S. Xiong, Q. Bao, G. Chen, Y. Tian, Y. Deng, K. Xiao, J. Wu, M. I. Saidaminov, H. Lin, C. Q. Ma, Z. Zhao, Y. Wu, L. Zhang, and H. Tan, Nat. Energy 7, 708 (2022).
- ³⁸H. S. Ryu, S. Y. Park, T. H. Lee, J. Y. Kim, and H. Y. Woo, Nanoscale **12**, 5792 (2020)
- ³⁹C. H. Chen, Z. H. Su, Y. H. Lou, Y. J. Yu, K. L. Wang, G. L. Liu, Y. R. Shi, J. Chen, J. J. Cao, L. Zhang, X. Y. Gao, and Z. K. Wang, Adv. Mater. 34, 2200320 (2022).
- Zuo, D. Vak, D. Angmo, L. Ding, and M. Gao, Nano Energy 46, 185 (2018).
 J. E. Kim, S. S. Kim, C. Zuo, M. Gao, D. Vak, and D. Y. Kim, Adv. Funct. Mater. 29, 1809194 (2019).
- ⁴²Y. Y. Kim, T. Y. Yang, R. Suhonen, M. Välimäki, T. Maaninen, A. Kemppainen, N. J. Jeon, and J. Seo, Adv. Sci. 6, 1802094 (2019).
- ⁴³L. J. Sutherland, D. Vak, M. Gao, T. A. N. Peiris, J. Jasieniak, G. P. Simon, and H. Weerasinghe, Adv. Energy Mater. 12, 2202142 (2022).
- ⁴⁴J. Li, R. Xia, W. Qi, X. Zhou, J. Cheng, Y. Chen, G. Hou, Y. Ding, Y. Li, Y. Zhao, and X. Zhang, J. Power Sources 485, 229313 (2021).
- 45 M. C. Brennan, S. Draguta, P. V. Kamat, and M. Kuno, ACS Energy Lett. 3, 204 (2018).
- 46J. Liang, X. Hu, C. Wang, C. Liang, C. Chen, M. Xiao, J. Li, C. Tao, G. Xing, R. Yu, W. Ke, and G. Fang, Joule 6, 816 (2022).
- 47T. Moot, J. B. Patel, G. McAndrews, E. J. Wolf, D. Morales, I. E. Gould, B. A. Rosales, C. C. Boyd, L. M. Wheeler, P. A. Parilla, S. W. Johnston, L. T. Schelhas, M. D. McGehee, and J. M. Luther, ACS Energy Lett. 6, 2038 (2021)
- ⁴⁸J. Kim, J. H. Jang, E. Choi, S. J. Shin, J.-H. Kim, G. G. Jeon, M. Lee, J. Seidel, J. H. Kim, J. S. Yun, and N. Park, Cell Rep. Phys. Sci. 1, 100273 (2020)
- ⁴⁹See https://ec.europa.eu/growth/single-market/european-standards/ for RoHS.
- 50 C. H. Chen, S. N. Cheng, L. Cheng, Z. K. Wang, and L. S. Liao, Adv. Energy Mater. 13, 2204144 (2023).
- ⁵¹M. E. Markowitz, P. E. Bijur, H. A. Ruff, K. Balbi, and J. F. Rosen, Environ. Health Perspect. 104, 968 (1996).
- ⁵²Y. Jiang, L. Qiu, E. J. Juarez-Perez, L. K. Ono, Z. Hu, Z. Liu, Z. Wu, L. Meng, Q. Wang, and Y. Qi, Nat. Energy 4, 585 (2019).
- ⁵³S. Chen, Y. Deng, H. Gu, S. Xu, S. Wang, Z. Yu, V. Blum, and J. Huang, Nat. Energy 5, 1003 (2020).
- 54 W. F. Yang, J. J. Cao, C. Dong, M. Li, Q. S. Tian, Z. K. Wang, and L. S. Liao, Appl. Phys. Lett. 118, 023501 (2021).