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Cyclodextrin-assisted supramolecular host-guest inclusion for durable and sustainable optoelectronics

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Abstract

Metal halide perovskites and organic nonlinear optical materials have showcased enormous potential in many kinds of optoelectronic applications, such as solar cells, light-emitting diodes, and patterned displays. However, further enhancement of optoelectronic performances has been largely limited by the intractable issues of these materials including high defect densities, unstable crystallographic structure, harsh fabrication conditions, and unfavorable biocompatibility and environmental sustainability. Encouragingly, several recent works have demonstrated an effective supramolecular host-guest inclusion strategy could ideally address abovementioned concerns by nesting optoelectronic materials within the cavities of cyclodextrin molecules and their analogs. Specifically, the supramolecule hosts embedded with multiple functional groups and/or crosslinked networks could robustly interact with those optoelectronic materials, which play multifaceted roles in terms of chemical chelation, spatial confinement, structural stabilization, defect passivation, ion immobilization and compensation, thus resulting in comprehensive enhancement of optoelectronic performances and sustainability. The current



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challenging issues and potential solutions are also discussed to provide a roadmap for achieving more durable and sustainable optoelectronics toward practical applications and real commercialization.

Keywords: Perovskite, organic nonlinear optical material, host-guest inclusion, cyclodextrin, optoelectronics

INTRODUCTION

Over the past decades, the metal halide perovskites (MHPs) have witnessed unprecedentedly fast development and have been regarded as the new generation photovoltaic and luminescent materials due to their fascinating optoelectronic properties such as tunable bandgaps, decent light absorption coefficient, long carrier diffusion length, high color purity and high photoluminescence quantum yield (PLQY). Despite fruitful breakthroughs in MHPs-based optoelectronics, the inherent crystallographic imperfections of MHPs and their structural instability under light/heat/solvent stimuli have been huge obstacles to limiting the further device performance enhancement and practical application^[1]. The same situation is encountered for organic nonlinear optical (NLO) materials, which, beyond that, also suffer from harsh fabrication conditions.

It is well known that the solution-processed MHPs intrinsically contain multiple types of ionic defects, which inevitably alter the optoelectronic properties and performances of MHPs materials and devices. Because of low migration activation energy, several kinds of ions can easily migrate through the vacancy defects within the perovskite lattice, which would cause the decomposition of MHPs and result in the degradation of MHPs-based optoelectronics^[2]. Various small organic molecules with moderate molecular size and several functional groups have been designed for passivating both the bulk and surface defects of MHPs. For example, uracil was introduced to manipulate the film growth by Wang *et al.*^[3]. Though effective and promising, one can still expect the potential shortcomings such as limited chemical interaction sites, insufficient surface protection^[4] and extra processing steps^[5], not to mention their inferior capability of reducing lead leakage. Specifically, the limited chemical interaction sites would result in limited passivation efficacy of crystallographic imperfections and limited ion immobilization effect. Moreover, most of the reported organic molecules may have planar configurations and/or relatively short molecular chain length, which has negligible spatial confinement effect on MHPs. Superior to the conventionally employed molecules, several recent works have demonstrated the unique advantages of cyclodextrins (CDs) and their analogs, which possessed desirable inner/outer cavities embedded with ample functional groups that could form a supramolecular host-guest inclusion complex with MHPs and NLO materials. Encouragingly, such a supramolecular host-guest inclusion strategy can passivate the crystallographic imperfections, spatially confine the guest materials, inhibit the ion migration and capture the leaked toxic metal ions in a more effective manner, thus making MHPs and NLO materials-based optoelectronics more efficient, durable and sustainable. Interestingly, such a strategy has resulted in the fabrication of waterproof perovskite luminescent textiles (PLTs) that are durable in water for more than one year, light/thermal-stable Pb-based perovskite solar cells (PSCs) that are biocompatible and sustainable and flexible NLO membranes for realizing *in-vivo* noninvasive two-photon bioimaging.

This perspective summarizes the recent advancement of employing supramolecular host-guest inclusion strategy to improve the performance, durability and sustainability of MHPs or NLO materials-based optoelectronic devices, with particular emphasis on highlighting the multifaceted roles of host-guest inclusion strategy in defect passivation, spatial confinement, ion immobilization and compensation, and lead capture. An insightful perspective toward future optimization routes and research directions will be

provided, aiming to popularize the host-guest inclusion strategy and facilitate the development of more efficient, durable and sustainable optoelectronics.

MULTIPLE CHEMICAL INTERACTIONS AND DEFECT PASSIVATION

MHPs, with a general chemical formula of ABX_3 , are composed of monovalent A cations and corner-sharing metal halide $[BX_6]^{4-}$ octahedras, in which B and X are bivalent metals and monovalent halides, respectively. Due to the fragile chemical bonds between the cations and anions, ionic defects are easy to form and migrate within MHPs when subjected to external stress or stimuli, thus accelerating the perovskite degradation. In addition, severe ion migration can eventually lead to local stoichiometric variations of MHPs, which would change their defect chemistry and promote unwanted variations of optoelectronic properties. CDs are cyclic oligosaccharides linked by α -1,4-glycosidic bonds^[6]. Three major CD variants, namely, α -, β -, and γ -CDs, comprise six, seven, and eight D-glucose units, respectively. More interestingly, the CDs feature a distinct truncated cone structure with a hydrophilic exterior and hydrophobic interior. Multidentate hydroxyl groups embedded along the wall of internal and external cavities enable the formation of robust inclusion complexes with various guest molecules, such as perovskites, and simultaneously passivate the defects and enhance the optoelectronic performance of perovskites.

Yang *et al.* have innovatively introduced a self-crosslinking supramolecular complex consisting of 2-hydroxypropyl- β -cyclodextrin (HP β CD) and 1,2,3,4-butane tetracarboxylic acid (BTCA) into perovskite precursors, which could effectively bond with undercoordinated Pb^{2+} and I⁻ ions in perovskites via multiple chemical interactions, such as coordination bonding, hydrogen bonding and chelation, *etc.* [Figure 1A]^[7]. Such luxurious mutual interactions are beneficial to retard the formamidinium iodide (FAI) deprotonation and undesirable side reactions of forming s-triazine by-products upon the accelerated aging of perovskite precursors, and concurrently enhance the stability of both perovskite films and devices under the stress of moisture and heat, for instance, preventing the perovskite film surface from water invasion and preventing the component loss during thermal treatment [Figure 1B]. As a result, the target PSCs retained more than 96% of their initial efficiency after aging for ~4,000 h in ambient air with a relative humidity of $\sim 30\% \pm 10\%$ and $\sim 80\%$ under continuous thermal stress at 85 °C in a N_2 -filled glovebox, respectively. The defect passivation effect of this supramolecular host-guest inclusion strategy was verified by the space charge-limited current characterization, which showed a lower trap-filling limit voltage (V_{TFL} , 0.20 V) and a lower trap-state density (N_t , $2.832 \times 10^{15} \text{ cm}^{-3}$) of the HP β CD-BTCA-modified device than that of the control counterpart (a V_{TFL} of 0.33 V and a N_t of $4.673 \times 10^{15} \text{ cm}^{-3}$, Figure 1C and D). Similarly, Liu *et al.* incorporated another typical CD molecule, namely, 2-O-methyl- β -cyclodextrin (OM β CD), into perovskite films, which also exhibited enhanced crystallinity, improved morphology, and reduced crystallographic imperfections owing to the strong interactions of OM β CD with perovskite ions^[8]. Interestingly, compared to a linear oligosaccharide molecule, namely, maltotetraose (G4), with the identical number (14) of hydroxyl groups, OM β CD with seven primary and seven secondary hydroxyl groups evenly distributed around each rim of the cyclodextrin molecule is more favorable to form focal multi-site interactions with perovskite ions. Simultaneously, it provides robust intermolecular forces to capture and immobilize the migrated ions [Figure 1E and F]. Echoing the structural features, OM β CD exhibited superior defect passivation effect to its G4 counterpart, with a further 5.05% decrease of N_t for the former, indicating the synergistic roles of cyclic structures and embedded functional groups in achieving more robust and comprehensive defect passivation. Encouragingly, the OM β CD-modified PSCs showed an enhanced device efficiency of 24.22%, higher than that of the control (20.85%) and G4-modified device (22.68%). Overall, rational incorporation of CD supramolecules could comprehensively passivate both the bulk and surface defects of perovskite photovoltaic materials, thus enabling remarkably enhanced optoelectronic performances, such as low voltage deficit and high power conversion efficiencies (PCEs).

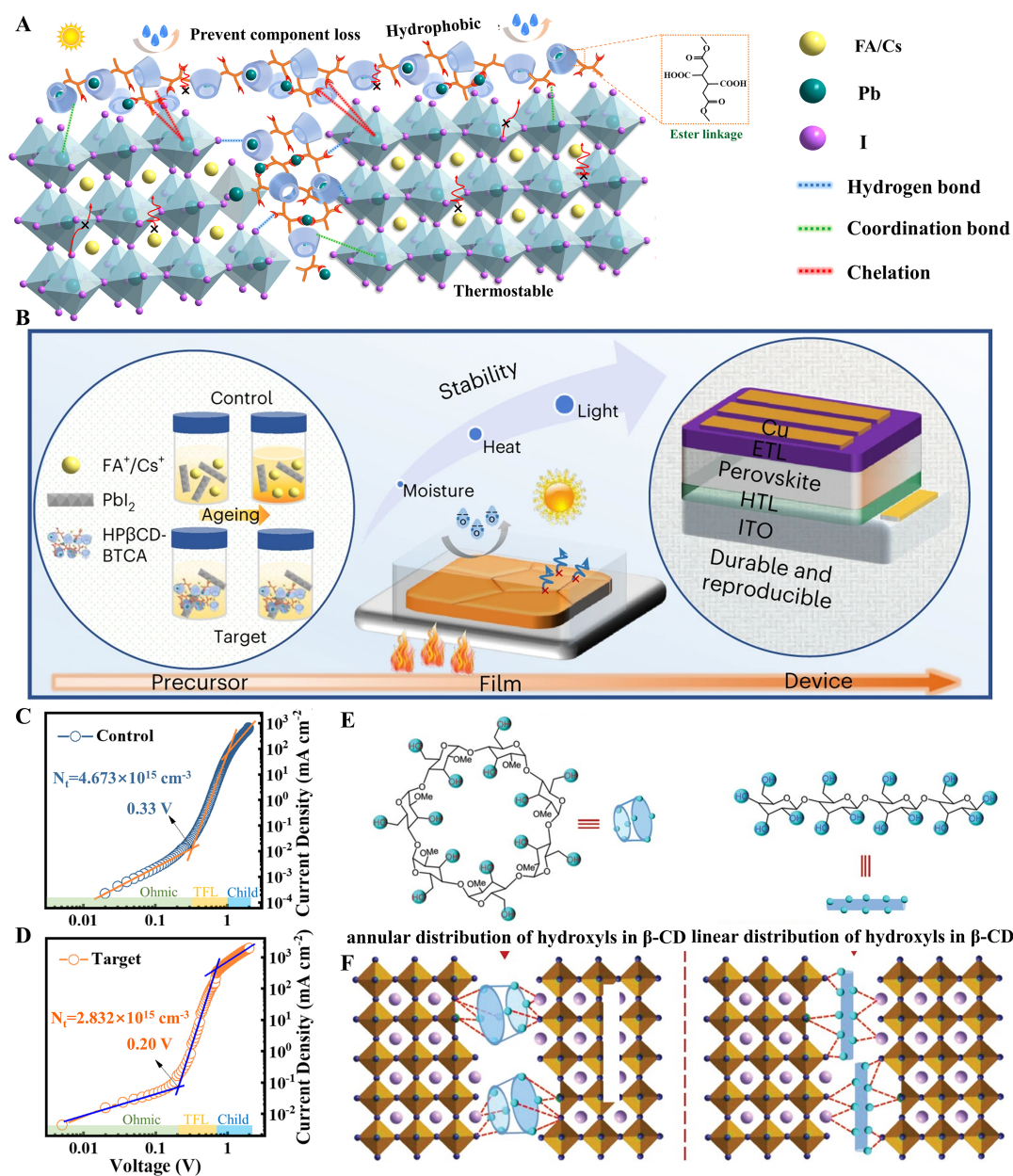


Figure 1. (A) Schematic diagram of multiple chemical interactions between crosslinked HP β CD-BTCA network and perovskites. (B) Schematic illustration of the HP β CD-BTCA-assisted stabilization of the whole fabrication process from precursor to film and to the final device. Space charge-limited current curves of the (C) control and (D) target HP β CD-BTCA-modified device. Reprinted with permission^[7]. Copyright © 2023 Springer Nature Limited. (E) Different distributions of hydroxyl groups in OM β CD and G4, and (F) their interactions with different ions in perovskites. Reprinted with permission^[8]. Copyright © 2023 Wiley-VCH GmbH.

SPATIAL CONFINEMENT

When the size of perovskite materials is reduced to the nanometer level comparable to their exciton Bohr radius, the quantum confinement effect is shown and the energy level is quantized, which would change the band gap of perovskites, thus producing some unique photoelectric properties. The abovementioned supramolecular host-guest inclusion strategy could also be employed to confine the sizes of perovskite nanocrystals, aiming to enhance the optical properties and durability of perovskite luminescent materials. Tian *et al.* have demonstrated that inclusion of CsPbBr₃ by HP β CD and super-hydrophobic

perfluorooctyltriethoxysilane (PFOS) could significantly improve the color purity of fibers with narrow-band photoluminescence (full-width at half-maximum < 17 nm), enhance the radiative recombination rate by more than 4-fold, and improve the PLQY by 25-fold to approximately 50%, relative to the control counterpart, which can be attributed to the quantum confinement effect, manifold defect passivation and reduced overlap of the absorption band edge and the PL emission peak [Figure 2A-C]^[9]. Specifically, unlike the irregular-shaped perovskite crystals disorderedly distributed and aggregated on the surface of the control perovskite-polymer composited fibers, the aggregation of CsPbBr₃ nanocrystals could be efficiently suppressed in the CsPbBr₃@HPβCD composites, in which the CsPbBr₃ nanocrystals were uniformly encapsulated and spatially separated by HPβCD clusters, thus confining the evolved CsPbBr₃ nanocrystals to much smaller sizes (i.e., 14.24 ± 6.02 nm) inside and/or at the surface of the polymer matrix [Figure 2D and E]. In this case, the quantum confinement effect was further reinforced, which resulted in blue shift of the absorption band edge [Figure 2B]. Ideally, more effective host-guest inclusion of perovskite nanocrystals can be realized through precise control of the cavity sizes of various CD molecules.

The spatial confinement effect induced by supramolecular host-guest inclusion could also be applied to organic NLO materials. Very recently, Tian *et al.* demonstrated the production of the large-area organic NLO membranes consisting of 4-N,N-dimethylamino-4'-N'-methylstilbazolium tosylate (DAST)@HPβCD host-guest supramolecular complexes with outstanding multiphoton absorption characteristics^[10]. DAST is a kind of stilbazolium derivative, featuring a structure of donor-π-acceptor (D-π-A)-type. Without spatial confinement and geometric restriction, the DAST molecules are easily burdened with serious aggregation-caused quenching (ACQ), trans-cis isomerization and/or intramolecular charge transfer (ICT) process-induced nonradiative decay, which, more often than not, showcased inferior luminescent performance and even lost their NLO activity when DAST materials are exposed in ambient air and/or polar solvent environment^[11]. With crosslinked HPβCD clusters incorporation, DAST molecules are inserted into the cavity of HPβCD hosts to form the DAST@HPβCD supramolecular host-guest inclusion complexes via robust interactions between these molecules, including hydrogen bonding interactions and electrostatic interaction [Figure 3A and B]. In this case, the HPβCD framework could spatially split the DAST molecules and successfully restrict the isomerization of DAST molecules, thus minimizing the ACQ effect, inhibiting the excited-state rotation from locally-excited (LE) state to twisted ICT (TICT) state [Figure 3C], and essentially restrain the TICT state-caused nonradiative decay [Figure 3D and E]. All these advantages contribute together to amplifying the PLQY of DAST@HPβCD fibers by ~78 fold, attaining a high value of 70.4%, which significantly outperformed that of the control counterpart (an extremely low PLQY of 0.9%). Further crosslink of DAST@HPβCD supramolecular framework boosted the PLQY to 73.5% [Figure 3F], which represented one of the highest reported values for the DAST materials and their derivatives. Beyond one-photon-excited fluorescence (1PEF), the DAST@HPβCD membranes showed outstanding NLO properties, which improved second-harmonic generation (SHG) signal, and could exhibit broadband two-photon-excited fluorescence (2PEF) (ca. 520-650 nm) when excited at a wavelength region from 770 nm to 1,000 nm and showcase three-photon-excited fluorescence (3PEF) when excited at 1,590 nm [Figure 3G]. The chemically and thermally stable crosslinked HPβCD matrix could achieve robust supramolecular encapsulation of DAST guests, in which the structural deformation of DAST molecules was successfully inhibited, even under high humidity or high temperature conditions, thus endowing the luminescent DAST@HPβCD membranes with outstanding ambient and thermal stability. Surprisingly, the DAST@HPβCD fibers retained 88% of their initial PLQY when stored in ambient air for 470 days. Such durable DAST@HPβCD fibers were, for the first time, applied to realize real-time, *in-vivo* noninvasive bioimaging of live *Escherichia coli* (*E. coli*) [Figure 3G]. All in all, the spatial confinement effect induced by supramolecular host-guest inclusion strategy is highly promising to greatly improve the optical and luminescent properties, along with durability of perovskite-based or organic NLO materials-based optoelectronics via regulating the crystal sizes, modulating the molecular orientation and fixing the

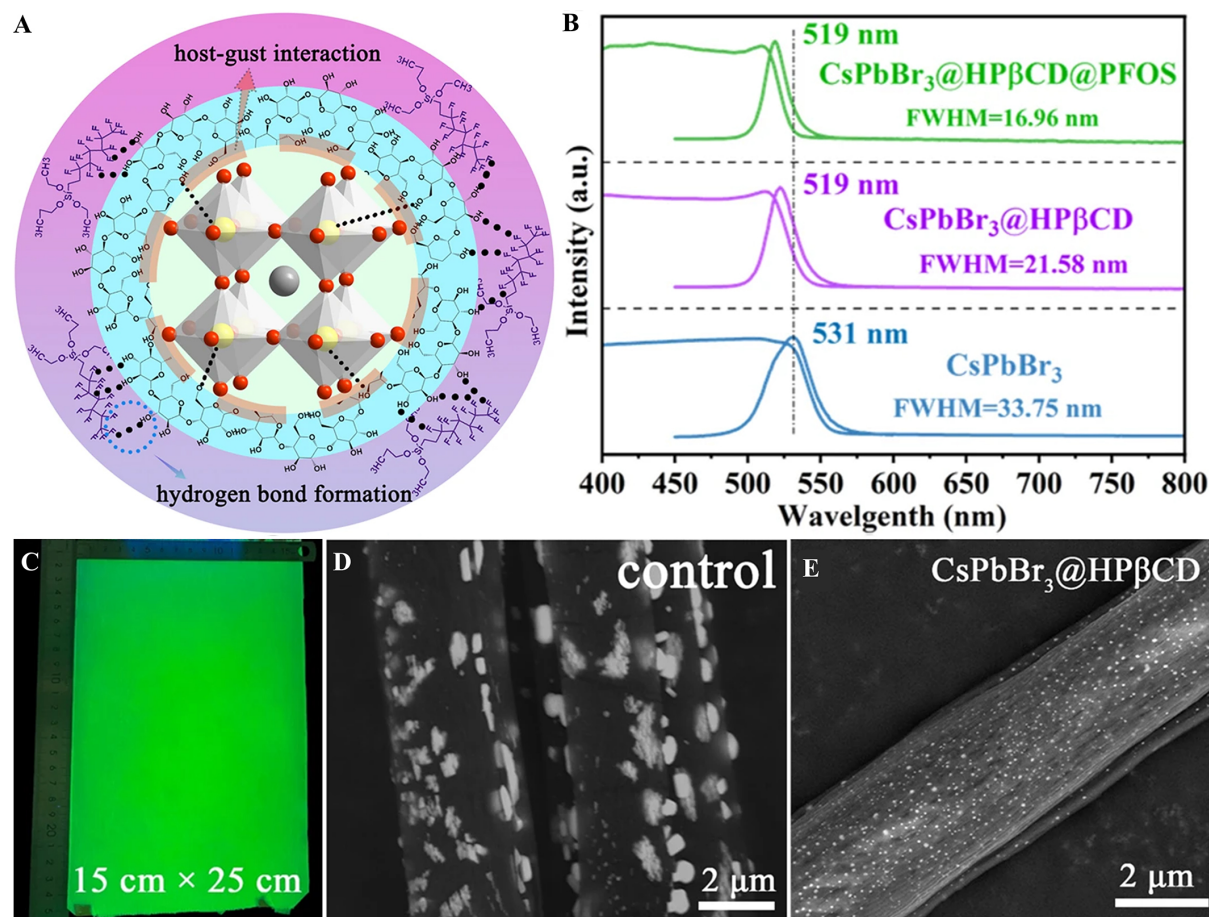


Figure 2. (A) Schematic illustration of CsPbBr₃@HPβCD@PFOS composites with host-guest interaction and hydrogen bond formation. (B) UV-visible absorption and PL spectra of CsPbBr₃, CsPbBr₃@HPβCD and CsPbBr₃@HPβCD@PFOS fibers. (C) Fluorescence image of large-area CsPbBr₃@HPβCD@PFOS fibrous membrane excited by UV light. TEM images of the (D) control CsPbBr₃ fibers and (E) CsPbBr₃@HPβCD fibers. Reprinted with permission^[9]. Copyright © 2023 Springer Nature Limited.

noncentrosymmetric arrangement, optimizing the excited-state population, and improving the light/heat/moisture stability.

ION IMMOBILIZATION AND COMPENSATION

Benefiting from the abovementioned multiple chemical interactions between host and guest molecules, the structural stability of MHPs is expected to be enhanced owing to the inhibited ion migration and retarded lattice deformation. Yang *et al.* have demonstrated the crosslinked HPβCD matrix with multiple positively charged hydrogen ions could interact intensely with negatively charged halide ions in perovskites via massive hydrogen bonding^[7]. Modified by 3D supramolecular framework, the target PSCs exhibited enhanced stability under the stress of light and heat, which preserved nearly 99% of their initial PCE after 700 h of continuous 1 sun illumination and retained 80% of the initial efficiency after 300 h upon continuous heating at 85 °C, while the control device showed poor stability. The remarkably extended stability of PSCs could be attributed to the ion immobilization effect induced by anchoring of HPβCD-BTCA crosslinking supramolecular framework, especially at the grain boundaries and on the film surface of perovskite film^[7]. It is anticipated that, relative to pristine HPβCD modification, the incorporation of cross-linking HPβCD-BTCA supramolecular network could further reinforce the ion immobilization effect

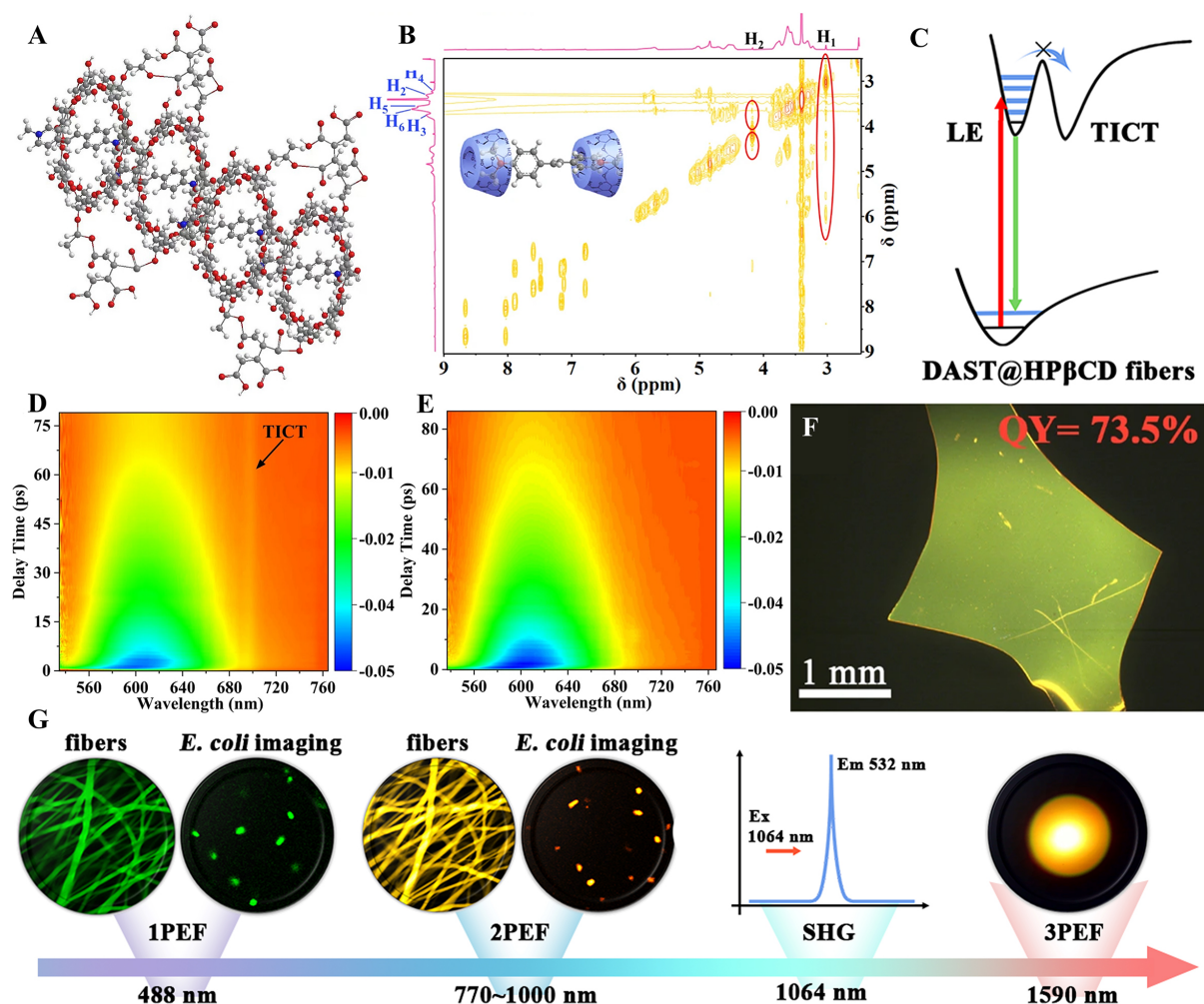


Figure 3. (A) Schematic showing the insertion of DAST guest molecules into the cavities of HP β CD hosts. (B) The correlation spectroscopy (COSY) NMR of DAST@HP β CD supramolecular host-guest inclusion complex. (C) Potential energy diagram and corresponding excited-state conversion of DAST@HP β CD fibers. The TA contour plots of (D) DAST and (E) DAST@HP β CD samples. (F) Fluorescence image of DAST@HP β CD thin film. (G) Schematic illustration of 1PEF/2PEF/3PEF and SHG properties of DAST@HP β CD fibers and their application in bio-imaging of *E. coli*. Readapted with permission^[10]. Copyright © 2023 Springer Nature Limited.

towards the perovskites owing to more luxuriant interactions to stabilize the ions and more robust physical blocking effect to inhibit the ions migration. Learning from the supramolecular design of Yang *et al.*^[7], Li *et al.* employed the β -cyclodextrin (β CD) as a typical I₂ trapper, in which the I₂ can be trapped into the hydrophobic cavity of bowl-like β CD, which beneficially increased the volatilization temperature of I₂ and inhibited its escape from perovskites^[12]. During device operation, the trapped I₂ could react with Pb⁰ to eliminate related defects, and favorably interact with FAI to form photoactive perovskites [Figure 4A]. It is worth noting that under International Submit on Organic Photovoltaic Stability (ISOS) protocol, the β CD-modified PSCs exhibited both high efficiency (> 23%) and good thermal stability, retaining over 80% of the highest efficiency after tracking at maximum power output point (MPP) for 300 h at 85 °C, while the control PSCs only retained about 56% of highest efficiency after 150 h [Figure 4B]^[12].

In addition to halide ion immobilization, supramolecular host-guest strategy can be employed to realize halide ion compensation, which is beneficial to reduce the halide deficiency, and mitigate the ion migration

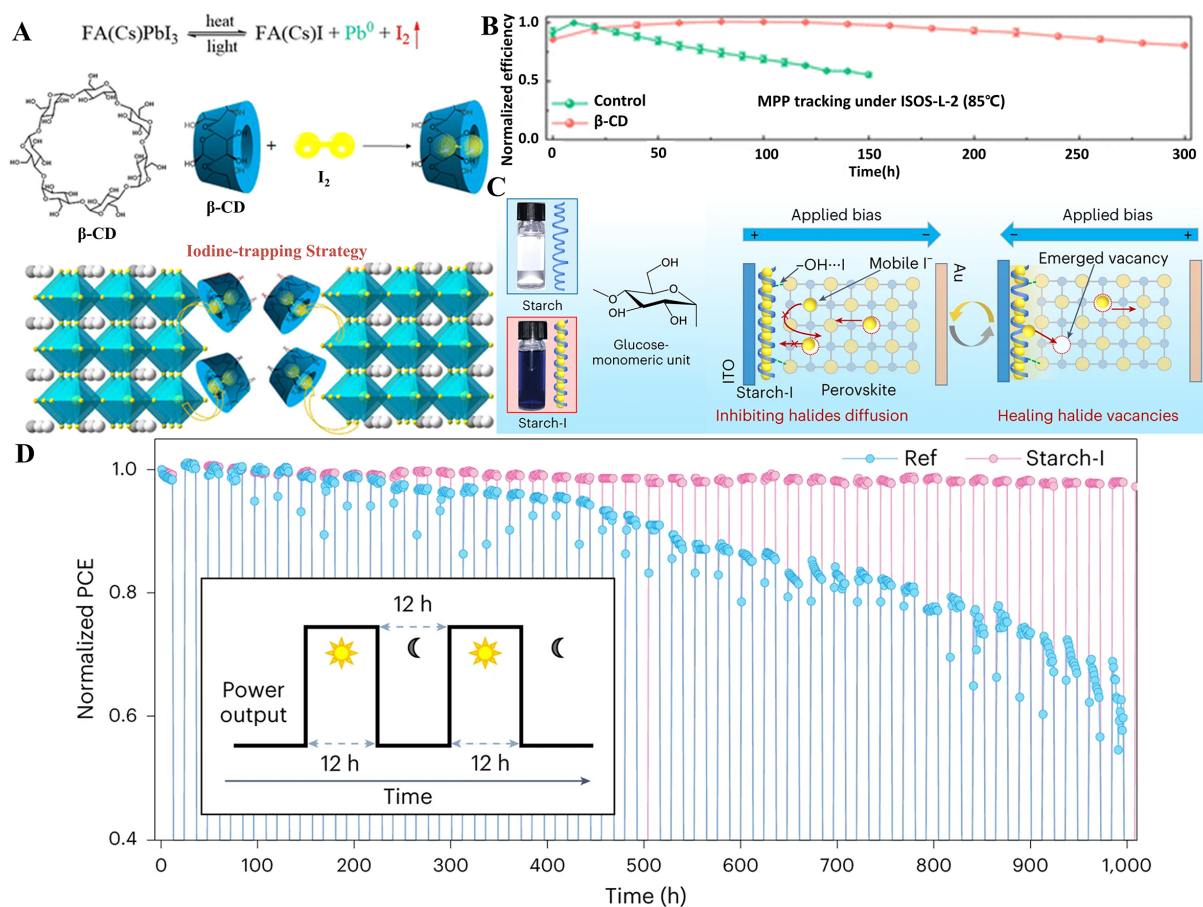


Figure 4. (A) Schematic diagram of iodine immobilization and regeneration of photoactive perovskites via β CD incorporation and modification. (B) Stability test of control and β CD-modified PSCs under ISOS protocol. Reprinted with permission^[12]. Copyright © 2023 Royal Society of Chemistry. (C) Molecular structure of Starch and Starch-I, along with proposed functions of the Starch-I buffer layer regarding halides diffusion inhibition and halide vacancies healing upon applied bias. (D) PCE tracking of the reference (Ref) and Starch-I devices upon day and night cycle operation. Reprinted with permission^[13]. Copyright © 2023 Springer Nature Limited.

and redistribution. Very recently, Zhang *et al.* innovatively introduced a starch-polyiodide supermolecule (Starch-I) as a bifunctional buffer layer at the buried interface of perovskite film, which simultaneously suppressed ion migration and promoted defect self-healing via modulating the halide migration kinetics by reserving sufficient iodide species^[13]. Starch is a polymeric carbohydrate with a molecular formula of $(\text{C}_6\text{H}_{10}\text{O}_5)_n$, which is composed of numerous glucose units. Similar to CDs, starch can spontaneously form a helix structure with a hydrophobic channel to host polyiodide anions in aqueous solution. When the I^- ions drifted towards the buried interface and the iodide vacancy defects emerged in the perovskite films, a Starch-I buffer layer acted as a barrier to restrain further I^- migration by continuously releasing extra I^- to neutralize these off-site defects [Figure 4C]. The modified PSCs presented improved stability by retaining 98% of their initial efficiency after operation for 42 diurnal cycles (12/12 h light/dark cycle, a total of > 1,000 h, Figure 4D). Encouragingly, the champion devices also delivered a high PCE of 24.3% (a certified value of 23.9%). It is worth noting the supramolecular host-guest inclusion strategy is promising to inhibit the notorious ion migration of ionic-typed perovskite compounds with soft lattices via ion immobilization, trapping and/or compensation, which is conducive to enhancing the thermal and operational stability of perovskite-based optoelectronic devices.

LEAD CAPTURE

Though promising, practical application of Pb-containing MHPs and long-term deployment of MHPs-based devices is confronted with the Pb toxicity matter, not to mention the case of unwanted Pb leakage from broken devices, which has become a serious problem with damaging impact on the environment and human health. Up till now, it is demonstrated that different approaches have been developed to reduce the Pb leakage through additional physical encapsulation or chemical absorption, but at the cost of introducing extra manufacturing processes. Yang *et al.* subtly minimized Pb leakage and lowered Pb toxicity via incorporating crosslinking HP β CD-BTCA supramolecular complex that enabled robust chemical coordination and multidentate chelation with Pb²⁺ ions [Figure 5A and B]^[7]. When the PSC devices were severely damaged, it was worth noting that Pb leakage rates significantly decreased from a high speed of 973 mg m⁻² h⁻¹ for the control device to 54 mg m⁻² h⁻¹ for the target PSC with built-in supramolecular complex, corresponding to a surprising Pb sequestration efficiency of 94.5% [Figure 5C]. This result validated the presence of the HP β CD-BTCA complex throughout the bulk perovskite film could effectively sequester the leaked lead ions and slow down the lead leakage rate, which showcased great potential for hindering the Pb leakage even when the perovskite photovoltaics confronted repeated damage and the encapsulation glass was harshly broken. Further physical encapsulation of completed target devices by a flexible polymer@HP β CD-BTCA-based sheet further realized a Pb sequestration efficiency of nearly 99%. Such severely damaged devices could still retain 97% of the initial PCE after 522 h dynamic water scouring, with merely < 14 ppb lead contamination in water, which is in compliance with the standards of the US Environmental Protection Agency.

The conventional Pb-based photovoltaics poses challenges due to the presence of toxic, pollutional, and bio-accumulative Pb, which could hinder the widespread adoption of this fascinating photovoltaic technology. Obviously, inhibiting the Pb leakage is still not safe enough to ensure the sustainable deployment of perovskite photovoltaics. Note that the biocompatible CD supramolecules have been demonstrated to be capable of constructing bio-safe host-guest inclusion complex-based fluorescence probes for realizing *in-vivo*, non-invasive bioimaging^[10]. Yang *et al.*, for the first time, evaluated the biocompatibility of the different perovskite compositions by evaluating their impacts on the growth of *E. coli*^[7] [Figure 5D]. The results indicated no difference in the *E. coli* growth between the target sample (with HP β CD-BTCA-modified perovskite) and the blank sample (no Pb contamination). In contrast, the control sample (with pristine perovskite contamination) revealed a remarkable inhibitory effect [Figure 5E], suggesting the reduced Pb toxicity for the former. It is worth noting that the biocompatibility of Pb-based perovskites with HP β CD-BTCA chelation is comparable to that of Pb-free perovskites, such as pure Sn-based compositions. In addition to preventing lead leakage and reducing lead toxicity, lead recycling and management is crucial for making perovskite photovoltaics safe and environmentally sustainable, especially for failing devices. Yang *et al.* have demonstrated a close-loop Pb recycling and management of PSCs^[7]. Specifically, the outstanding lead capture capability of the HP β CD-BTCA complexes enabled the collection of HP β CD-BTCA@PbI₂ composites from failed devices, which can be readily reused as additives in device fabrication. Interestingly, high-purity PbI₂ components can be separated and recycled effectively from HP β CD-BTCA@PbI₂ composites, which can be reused for fabricating high-efficiency PSCs as well. As a result, the fresh PSCs made either with 2.5 wt% recycled Pb-containing additives or with raw materials based on recycled PbI₂ both demonstrated PCEs approaching 20% [Figure 5F]^[7]. Overall, the supramolecular host-guest inclusion strategy perfectly mitigates the Pb leakage, reduces the Pb toxicity, and enables close-loop Pb recycling and management, which will, to a large extent, ease the concerns of end-users and promote the practical application of efficient, durable and sustainable perovskite photovoltaics and other optoelectronic devices.

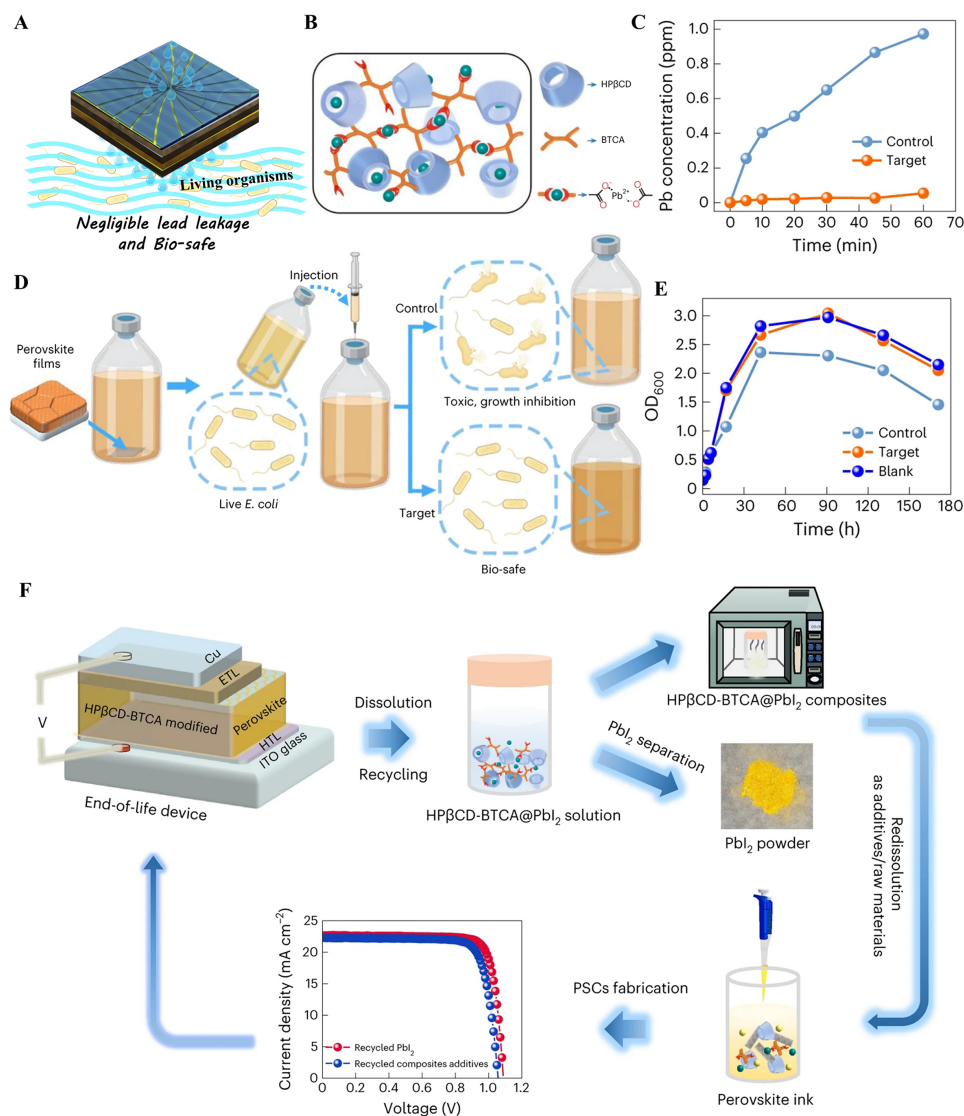


Figure 5. (A) Schematic showing the negligible Pb leakage and biocompatibility to living organisms of broken PSCs with supramolecular HP β CD-BTCA complex modification, and (B) schematic diagram of the lead capture mechanism of supramolecular HP β CD-BTCA complex. (C) Comparison of Pb sequestration for the damaged control and HP β CD-BTCA-modified PSCs. (D) Schematic illustration of Pb toxicity test procedures and (E) the corresponding results for the impacts of pristine perovskites and HP β CD-BTCA-chelated perovskites on the growth of *E. coli*. (F) Schematic illustration of Pb recycling and management in PSCs. Readapted with permission^[7]. Copyright © 2023 Springer Nature Limited.

SUMMARY AND OUTLOOK

In summary, the aforementioned breakthrough studies have highlighted the unique advantages and beneficial roles of host-guest inclusion between supramolecules host and optoelectronic materials guests (i.e., perovskites and organic NLO materials). The outstanding merits are multifaceted, including effective defect passivation, uniform nanoconfinement and robust ion immobilization, which synergistically boosted the optoelectronic performance of photovoltaics and luminescent devices. Encouragingly, the supramolecular host-guest inclusion strategy has enabled the realization of high-performance PSCs with champion efficiency of up to 24.22% with an extended lifespan of over 1,000 h. The perovskite-based luminescent films showed a champion PLQY of 49.7%, corresponding to an amplification of 25-fold. The NLO-based luminescent films showed a champion PLQY of 73.5%, corresponding to an amplification of

Table 1. Summary of the key performances of cyclodextrin-modified optoelectronics

Type of cyclodextrin and its derivatives	Guest materials	PCE (%)	PLQY (%)
HP β CD ^[7]	(Cs _{0.1} FA _{0.9})PbI ₃	22.14	
OM β CD ^[8]	FAPbI ₃	24.22	
β CD ^[12]	Cs _{0.05} FA _{0.95} PbI ₃	23.20	
Starch-I ^[13]	FAPbI ₃	23.90	
HP β CD ^[9]	CsPbBr ₃		49.7
HP β CD ^[10]	DAST		73.5

78-fold [Table 1]. Via *in-situ* chemical chelation, encapsulation and protection of toxic Pb-containing perovskites by biocompatible CD-like supramolecular host, not only the possibility of Pb exposure to environment has been minimized (i.e., nearly 99% Pb sequestration efficiency and 100% Pb recycling), but also the Pb toxicity has been favorably reduced to a bio-safe level. Regarding the roles of supramolecular inclusion strategy on the path towards commercialization of perovskite-based optoelectronics, incorporating the low-cost and biocompatible CDs could not only largely retain the high efficiency in a cost-effective manner but also release the concerns about the toxicity of perovskite materials to the global environment and human health. Undoubtedly, this will certainly expand the applicability of perovskite-based optoelectronic devices.

While fruitful results and advanced breakthroughs have been achieved, the relevant community was encouraged to seek future development. We thus provide an insightful perspective toward future research directions, aiming to take full advantage of MHPs and organic NLO materials and further extend their applications in various optoelectronics. First of all, the CDs and their derivatives have luxuriant structural constituents. As for the principles of selecting different CDs, one should consider their inner cavity size, types and numbers of functional groups, and their chemical interaction affinity with guest molecules, which is beneficial to achieve robust host-guest inclusion effect and improve the optoelectronic properties of resultant complexes. More advanced design of supramolecular host is desirable, for instance, exploring other functionalized CDs with different function groups or moieties that could play multifunctional roles for optimizing optoelectronic properties of guest materials. Secondly, the host-guest relationship and mutual interaction mechanisms between the supramolecular host and optoelectronic guest molecules should be further studied, aiming to excavate the fundamental understanding of complicated host-guest inclusion chemistry and guide the further advancement of the optoelectronic performance. Thirdly, one of the most challenging obstacles that the supramolecular inclusion strategy encounters in the process of commercializing perovskite-based optoelectronic devices is its compatibility to realize low-cost, high throughput fabrication of efficient, stable and reproducible optoelectronic devices. To tackle it, one should adapt the well-designed host-guest inclusion ingredients/compositions to those advanced fabrication techniques that are compatible with large-scale mass production (e.g., electrospinning, blade-coating, inkjet printing, roll-to-roll manufacturing, and slot-die coating). Note that the functional groups embedded in the CD could be positively engaged in the crystallization process of perovskites, which beneficially helped to improve the uniformity of large-area perovskite films. Last but not least, not just limited to PSCs and luminescent applications, such a promising supramolecular host-guest inclusion strategy should be extended to other optoelectronic applications, such as photodetectors, lasers, photocatalysis, X-ray imaging, etc.

DECLARATIONS

Authors' contributions

Literature review and manuscript draft: Chen H

Figure layout: Chen H, Chang X
Picture production: Chen H, Yang G
Manuscript revision: Lei BX, Wu WQ
Project supervision: Wu WQ

Availability of data and materials

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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