

Vapor-assisted pre-solvation for stable and solution-processable quantum dot conductive ink

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ABSTRACT: The development of high-performance, stable quantum dot (QD) inks is critical yet challenging for advancing the efficiency and scalability of next-generation solution-processed optoelectronic devices. Inorganic lead iodide ligands passivated lead sulfide QDs (PbS-PbI₂) have been attractive for their superior surface passivation and strong inter-dot coupling. However, the state-of-the-art inks typically require strongly coordinating alkylamine solvents to dissociate and solvate the PbI₂ ligand, yet these solvents can continuously etch the QDs, leading to rapid ink degradation within a few hours. Herein, an amine-vapor pre-solvation strategy is proposed using a facile butylamine (BA) vapor (< 10 min) pretreatment of QDs, enabling stable dispersion of QDs in BA-free solvents (e.g., N-methyl-2-pyrrolidone (NMP) and propylene carbonate). The resulting QD inks remain stable for > 60 days with no variation in QD size, while QD films exhibit reduced trap-related losses. Solar cells processed from 60-day-aged NMP inks retain 90% of their initial power conversion efficiency (12.1%), whereas BA-based inks degrade within hours, reducing their device efficiency from 11% to 0.3% after only 4 h of aging. The shelf-stable NMP ink further enables large-area blade-coating of uniform PbS-PbI₂ films, manifesting the potential in scalable manufacturing of high-performance QD optoelectronic devices.

KEYWORDS: quantum dots, ink, stability, solution processability

1 Introduction

Quantum dot (QD) inks have attracted much attention as outstanding materials for solution-processed optoelectronics, where high performance and stability are crucial for scalable manufacturing [1–5]. Over the past decade, lead sulfide (PbS) QDs have been distinguished by their size-tunable optical bandgaps, surface-controllable doping, and potential solution processability, making them great candidates for high-performance optoelectronic devices [6–8]. In particular, compared with long-chain organic

ligands that hinder carrier transport, short-chain inorganic PbI₂ ligands endow PbS QDs with more effective surface passivation afforded by ionic species and stronger inter-dot electronic coupling enabled by the close QD packing [9–13]. Accordingly, PbS QDs capped with PbI₂ ligands (PbS-PbI₂) inks have emerged as promising materials to boost the performance of infrared photodetectors, colloidal quantum dot photovoltaics, and field-effect transistors [14–18].

Nevertheless, a key limitation for PbS-PbI₂ inks is the weak dissociation and poor solvation of the inorganic PbI₂ ligand in common solvents, which reduces dispersibility and results in colloidal destabilization [19–23]. To date, butylamine (BA) has remained among the few viable solvents for dispersing PbS-PbI₂ QDs at high concentration, because its strong coordination to lead cation can promote the dissociation of PbI₂ into ionic species, such as PbI⁺ and PbI₃[−] in solution, thereby providing electrostatic stabilization of PbS-PbI₂ inks [6, 24–27]. However, the BA solvent

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can concurrently coordinate with surface lead sites on PbS QDs, inducing surface etching of the QDs. This undesirable interaction causes rapid colloidal destabilization within a few hours and progressive chemical degradation over several days [28]. To mitigate the adverse effects of amine-based solvents, additional ligands, such as mercaptopropionic acid [29] and phenethylammonium iodide [30], were introduced onto the QD surface to provide assisted chemical protection against solvent-induced etching. A mixed-solvent strategy has also been adopted to improve ink stability by reducing the dosage of BA in inks to no more than 20% [21, 31]. While these approaches offer improved short-term stability, the presence of BA still limits the stability of PbS-PbI₂ QD ink to approximately one week. Therefore, developing an alternative method to promote concentrated PbS-PbI₂ QDs in BA-free solvents is a critical challenge toward long-term-stable, high-performance PbS-PbI₂ QD inks.

Herein, an amine vapor-assisted pre-solvation is proposed to ensure PbS-PbI₂ solvation in BA-free solvents. The facile vapor pretreatment assisted the dissociation of the PbI₂ ligand, and the excess amine was subsequently removed, thereby avoiding the prolonged etching of PbS QDs and enabling stable dispersion in various BA-free solvents. Compared with the poor colloidal stability (< 24 h) of conventional BA inks, BA-free inks (e.g., N-methyl-2-pyrrolidone (NMP)) extend storage lifetime to 60 days while preserving superior optoelectronic properties. Solar cells fabricated from this highly dispersible ink achieve a power conversion efficiency (PCE) of 12.1%, and retain 90% of the initial PCE (10.8%) after storage. This performance is markedly superior to that of devices from BA-based inks, which degrade from 11.0% to 0.3% after aging. Furthermore, this strategy enhances thermal stability (up to 80 °C) of QD inks and enables blade-coating of large-area QD films (100 cm²).

2 Experimental section

2.1 Materials

N-Hexane (> 99%), NMP (99.5%, for HPLC), n-amylamine (AA, 99%), hexylamine (HA, 99%), isopropanol (IPA, 99.8%), and ethyl acetate (EA, 99.5%) were purchased from Acros. Zinc acetate dehydrate (99.995%), lead acetate trihydrate (99.99%), n-octane (> 99%), propylene carbonate (PC, > 99.7%), chlorobenzene (CB, > 99.5%), acetic acid (Acac, > 99.8%), and ammonium acetate (NH₄Ac, 99.9%) were purchased from Aladdin. Ethanolamine (99%, GC&T), 2-Methoxyethanol (99%), lead(II) iodide (PbI₂, for perovskite precursor, 99.99%), lead(II) bromide (PbBr₂, for perovskite precursor, 99%), and N,N-dimethylformamide (DMF, GC, 99.5%) were purchased from TCI. 1-Octadecene (ODE, > 90%), oleic acid (OA, 90%), hexamethyldisilathiane ((TMS)₂S, synthesis grade), n-butylamine (BA, GC&T, 99%), methanol (MeOH, > 99.9%), trichloromethane (TCM, 99%), and 1,2-ethanedithiol (EDT, 98%) were purchased from Sigma-Aldrich. Acetonitrile (ACN, 99.9%, extra dry with molecular sieves, water < 50 ppm) was purchased from Innochem. Toluene (99%), acetone (Ace, 99.5%), and pyridine (Py, AR) were purchased from Sinopharm. All the chemicals were used as received.

2.2 Synthesis of ZnO

The ZnO was prepared by the sol-gel method. The precursor solution of zinc oxide was obtained by dissolving 5 g zinc acetate dihydrate and 135 μ L ethanolamine in 5 mL of 2-methoxyethanol

at room temperature under stirring in the dark for about 12 h. The solution was spin-coated on polymer films or cleaned fluorine-doped tin oxide (FTO) at 2000 rpm for 30 s. The resulting precursor film was annealed at 200 °C for half an hour.

2.3 Preparation of PbS QD inks

We used the hot-injection method in nitrogen to prepare long-chain ligand-covered PbS QDs. Lead acetate trihydrate (0.6558 g), OA (1.7 mL), and octadecene (15 mL) were added to a flask (250 mL). The mixing solution was degassed at 80 °C for two hours. Then, a rapid infusion of 0.5 mL (TMS)₂S dispersed in 5 mL ODE was injected into the flask to react for about a minute. The solution was then free-cooled to room temperature. The colloidal quantum dots (CQDs) (first exciton absorbance peak, 926 nm) were cleaned twice with acetone and redispersed in n-octane (50 mg·mL⁻¹). The long oleic acid ligands on the surface of QDs were replaced by short halogen ligands through liquid ligand exchange. 0.2 M PbI₂, 0.04 M PbBr₂, and 0.05 M NH₄Ac were stirred in 5 mL DMF until the solution dispersed, and 5 mL 40 mg·mL⁻¹ PbS-OA n-octane solution was added and stirred for 5 min. Next, we used n-octane to clean it three times and got clean short-chain quantum dot powder by the antisolvent toluene.

The QD powder was dispersed in butylamine at a concentration of 200 mg·mL⁻¹ to obtain butylamine-based quantum dot ink. For vapor treatment, 0.06 g of QD powder was placed in a sealed glass vessel containing 5 mL of BA with no direct contact. After exposure at room temperature for 10 min, the powder was vacuum-dried for 3 min to remove excess BA. The treated powder was then dispersed in various BA-free solvents. The ink concentration can be adjusted according to the desired film thickness or coating method. For instance, for spin-coating QD films, a concentration of 800–1500 mg·mL⁻¹ was typically used. Low-concentration (< 2 mg·mL⁻¹) quantum dot solar ink was also prepared for absorption spectroscopy, photoluminescence spectroscopy, and transmission electron microscopy (TEM) characterization. The inks were stored in sealed vials under ambient air, protected from light, at 20–23 °C, 15%–18% RH.

2.4 Device fabrication

The device structure of lead sulfide colloidal quantum dot solar cell was FTO/ZnO/PbS-I/PbS-EDT/Au. The BA QD ink was then spin-coated on the ZnO films at 2000 rpm for 30 s. The BA-free QD ink was spin-coated on the ZnO films at 1000 rpm for 100 s or blade-coated at 50 mm·s⁻¹. The blade-coated QD film was prepared via the BA-free ink at a speed of 50 mm·s⁻¹, and the substrate was heated to 80 °C. EDT-capped quantum dots were deposited by the solid-state ligand exchange process. PbS-OA (50 mg·mL⁻¹) was spin-coated (2500 rpm for 10 s) on the PbS-PbI₂ films, followed by the EDT in acetonitrile (0.02%, v/v) for 30 s and cleaning with acetonitrile three times. This procedure was repeated twice for a suitable hole transport layer. Finally, Au electrodes are covered on the top of the device via metal evaporation. The active area of the solar cells was 0.0314 cm² for the J–V characterization.

2.5 Characterization

PE 1050⁺ (PerkinElmer LAMBDA) was used to collect ultraviolet–visible (UV–vis) near IR spectrum. Photoluminescence (PL) spectra were obtained by a 325 nm laser from a J-Y Horiba UV-lamb micro-Raman spectrometer. X-ray photoelectron spectroscopy (XPS) was measured with an Thermo Scientific K-

Alpha system equipped with an Al K α source, and the peaks were all corrected by the reference carbon peak at 284.6 eV. The morphological properties of the films were obtained by a FEI Quanta 250 scanning electron microscope (SEM). The grazing-incidence small-angle X-ray scattering (GISAXS) measurements were carried out at the Xeuss 2.0 instrument with a Cu-K α X-ray source and single pixel size of 172 μm . The detector was Pilatus 3R 300 K, the sample-detector distance was 1008 mm and the incident angle was 0.2°. Scanning probe microscope characterizations, including atomic force microscopy (AFM), Kelvin probe force microscopy (KPFM), were measured by Bruker Dimension Icon with ScanAsyst. Space-charge-limited current (SCLC) measurement was measured by a Keithley 2400 source meter with an electron-type device (FTO/ZnO/PbS-PbI₂/Al). The density of trap states (N_{trap}) could be calculated using Eq. (1)

$$N_{\text{trap}} = \frac{2\varepsilon_0\varepsilon_{\text{QD}}V_{\text{TFL}}}{eL^2} \quad (1)$$

where e , L , and V_{TFL} are the elementary charge, the thickness of QD film (250 nm), and trap-filled limit voltage, respectively. J - V plots, light-intensity dependence of J_{sc} and V_{oc} characteristics were measured by a Keithley 2400 source meter with a 3A steady-state solar simulator under simulated 100 $\text{mW}\cdot\text{cm}^{-2}$, AM1.5G illumination (Enli Technology SS-F5-3A). We fabricated 12 identical solar cells each time using the same batch of chemicals. The active area of solar cells was 0.0314 cm^2 with a 2.5 mm diameter photomask, and the step of voltage was 5 mV. External quantum efficiency (EQE) was evaluated through a Zolix Instruments system and a standard photodetector. Device-performance retention was calculated for the best-performing PCEs, and average values and standard deviations across 12 devices were used to evaluate reproducibility. All the same measurements have been repeated 3 times.

3 Results and discussion

3.1 Dissolution in BA-free solvent

We demonstrate that a short aliphatic BA vapor exposure treatment is sufficient to disperse PbS-PbI₂ QD powders in BA-free polar solvents without introducing liquid amines into the ink. Specifically, the QD powder was exposed to vapor from 5 mL of BA in a sealed glass vessel at room temperature for 10 min, followed by brief vacuum drying to remove excess BA (Fig. 1(a) and Fig. S1 in the Electronic Supplementary Material (ESM)). This simple pretreatment enabled QDs to disperse in various BA-free solvents (e.g., NMP, DMF, propylene carbonate (PC) (Fig. 1(b) and Fig. S2 and Table S1 in the ESM), whereas untreated powders remained insoluble under identical conditions. X-ray diffraction (XRD) was used to probe the BA-induced structural changes in the PbI₂ ligand shells and revealed the broadening of the (001) diffraction peak of PbI₂ alongside the appearance of a feature associated with an intermediate phase (Fig. S3 in the ESM). This behavior may originate from an increased interlayer distance in the PbI₂ structure due to the intercalation of BA molecules, which reduces the crystallinity of PbI₂ [32]. In addition, GISAXS characterization (Fig. 1(c) and Fig. S4 in the ESM) revealed that the PbS-PbI₂ inter-dot center-to-center distance peak shifted slightly from 4.12 to 4.17 nm after vapor treatment (Fig. 1(d)). Significantly, the full width at half maximum (FWHM) of the distance distribution increased from 4.69 nm (Pristine) to 6.15 nm (Vapor). This broadened distribution suggests increased disorder in interdot packing after vapor exposure of QD powders [33–35]. Accordingly, we attributed these structural features to amine vapor-induced PbI₂ ligand dissociation, thereby enabling subsequent solvation even in weakly coordinating BA-free solvents. Notably, the amine is introduced only as a short vapor pretreatment and is removed prior to solvation, which is confirmed by significantly reduced BA-related

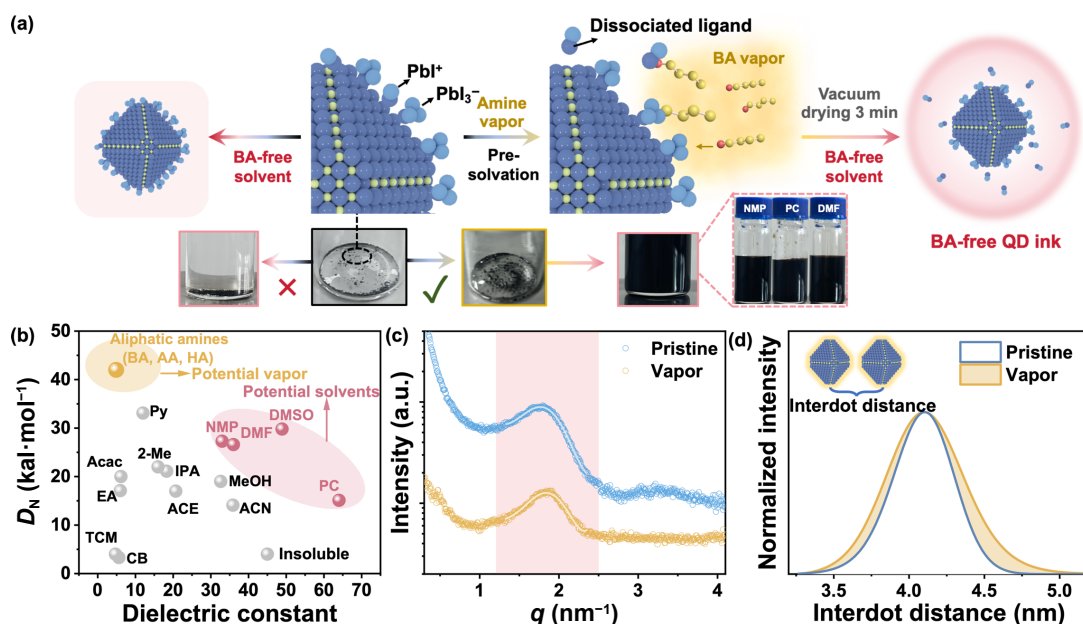


Figure 1 Amine-vapor-assisted pre-solvation and disperses quantum dots in BA-free solvents. (a) The ionic ligand (PbI⁺, PbI₃⁻) of PbS-PbI₂ QD powder dissociated after BA vapor treatment, and then dispersed QDs in BA-free solvents (right). QD powder cannot be directly dispersed in BA-free solvents (left). Photographs at the bottom show the transition of the QD powder during vapor treatment and directly adding BA-free solvent. (b) Potential vapor (yellow region) and BA-free solvents (pink region) to disperse the QD powder. Dielectric constant and Gutmann donor number (D_n) of the solvents used to identify differences in QD solubility are summarized in Table S1 in the ESM. (c) Azimuthal integrated intensity as a function of scattering vector q . (d) Derived QD inter-dot distance based on modeling of the GISAXS data with spheres.

signals in XPS and Fourier transform infrared (FTIR) spectra, enabling QDs dispersed in BA-free polar solvents without additives (Fig. S5 in the ESM). This method minimizes prolonged amine coordination with PbS-PbI₂ QDs, thereby preserving the PbI₂-passivated QD surfaces. Previous strategies, by contrast, typically require large quantities (20 wt.%–100 wt.%) of highly reactive amines [36] or ammonium salts (e.g., methylammonium iodide (MAI) [37–39] or ammonium iodide (NH₄I) [40]) as additives to promote the solvation of the QD, and because these BA or ammonium salts remain in the ink, they can strongly interact with the QD surface, thereby altering and potentially compromising its surface chemistry.

We further investigated a series of aliphatic amines with different carbon chain lengths to evaluate their ability to induce vapor pre-solvation. Amines with 4–6 carbon atoms (e.g., amylamine, hexylamine) showed the most effective pre-solvation, enabling QD dispersion in BA-free solvents (Fig. S6 in the ESM). Shorter chain amines with 1–3 carbon atoms (e.g., methylamine, ethylamine) were less suitable because their high volatility makes vapor exposure difficult to control, whereas longer straight-chain amines with more than 6 carbon atoms (e.g., octylamine, decylamine) were not examined because their high boiling points (> 150 °C) limit vapor availability and their extended alkyl chains may suppress interdot electronic coupling. Therefore, we focused on amines with 4–6 carbon atoms for subsequent studies, and additional parameters for these aliphatic amines are provided in Table S2 in the ESM.

3.2 Months-long stability

The BA-free quantum dot inks exhibit markedly improved storage stability compared with conventional BA inks. Upon storage in BA solvent, the QDs exhibit pronounced structural degradation, as shown by substantial broadening of the (111) and (200) diffraction peaks after 4 h (Fig. 2(a)), which becomes more severe after 60 days (Fig. 2(b)), suggesting continuous BA-induced etching of QDs.

Consistently, the first excitonic absorption peak at 950 nm vanishes after aging (Fig. 2(c) and Fig. S7 in the ESM). In contrast, QDs stored in NMP under the same conditions exhibit no broadening or shift in XRD peaks (Fig. 2(b)), confirming the preservation of crystallinity. As a result, the BA-free QD ink maintained a stable first excitonic absorption peak intensity (1.15 ± 0.03) and FWHM (91.35 ± 2.06 meV) after 60 days of ambient storage (20–23 °C, 15%–18% RH), demonstrating exceptional storage stability. The BA-free QDs maintain monodispersity over 60 days with a narrow size distribution and a constant average diameter of 3.74 ± 0.02 nm, as confirmed by TEM (Figs. 2(d) and 2(e)). Furthermore, the films prepared from this stored ink showed no change in root-mean-square (RMS) roughness (2.4 nm), as measured by AFM, whereas those from stored BA ink exhibited degraded morphology with increased pinholes and roughness increasing from 2.3 to 7.1 nm (Fig. 2(f) and Fig. S8 in the ESM). This morphological consistency following ink storage is further supported by negligible changes in surface potential and inter-dot distance in films prepared from stored BA-free inks (Fig. S9 in the ESM).

We compared the BA-free ink with a representative previously reported mixed solvent method for QD inks based on BA:NMP (1:4, v/v). A clear difference emerged within days under the same storage conditions between these two inks. The mixed-solvent ink showed a decrease in the intensity of the first excitonic peak and a blueshift after being stored for 4 days (Fig. S10 in the ESM). Whereas our BA-free ink maintains the excitonic peaks unchanged. We also compare our results with other BA-free inks reported in the literature. The BA-free inks, such as PbS-NH₄I in PC or PbS-MAPbI₃ in DFP, extended ink stability beyond one week but still showed gradual performance loss (approximately 25%) and spectral blue shifts in the first exciton absorption peak, along with an increase in FWHM after two weeks (Table S3 in the ESM). Conversely, the BA-free ink maintained stable optical properties and size distribution over 60 days, highlighting its favorable long-

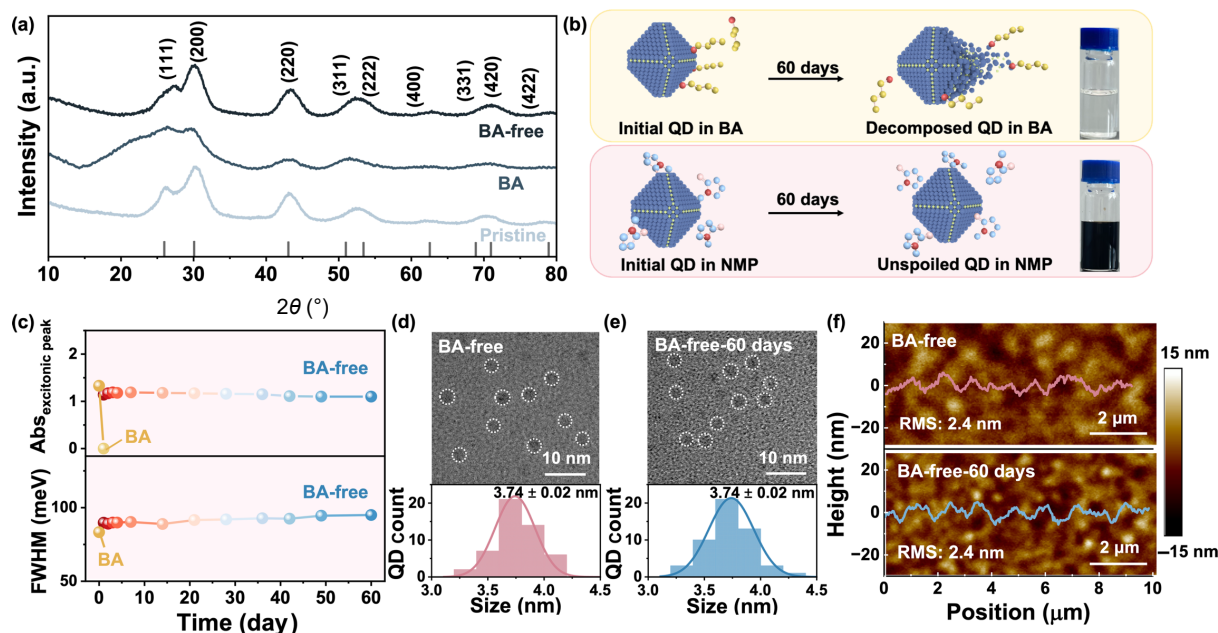


Figure 2 Months-long stability of PbS-PbI₂ ink with BA-free solvent. (a) XRD patterns of the pristine QD powder after ligand exchange, and of the same powder after being dispersed in BA and BA-free solvent for a period of time, respectively. (b) Schematic illustration of the etching of QDs in a BA ink after storage for 60 days, during which the ink transforms from a black dispersed colloid into a white aggregate. In contrast, the BA-free ink remains dispersed as a black colloid. (c) Variation of the first excitonic peak intensity and FWHM with storage time for BA and BA-free QD inks. TEM images and corresponding size distribution (with Gaussian fitting) of (d) fresh BA-free QD ink and (e) QD ink aged for 60 days. (f) AFM images and height curves at different positions of BA-free QD films prepared before and after aging the ink.

term storage stability. Further discussion of the stability of our BA-free ink is provided in Fig. S11 in the ESM.

3.3 Remarkable optoelectronic properties

We then investigated the optoelectronic properties to explore trap-related recombination losses in the films. Figures 3(a)–3(c) present the bleach signal and its kinetics from femtosecond transient absorption (TA) measurements on QD films. Upon excitation with a 750 nm pump laser, the TA spectra at different delay times showed a bleach signal around 950 nm in all samples (Fig. 3(a)). The BA-free QD films demonstrated superior properties, with a maximum bleach redshift of only 8.9 meV, much less than the 14.3 meV shift observed in BA QDs (Fig. 3(b)). Additionally, they exhibited a longer average carrier lifetime (τ_{avg}) of 3193 ps, compared to 2630 ps for the BA films, as determined by multi-exponential fitting of the bleach kinetics (Fig. 3(c)). Likewise, the PL lifetime of BA-free QD was determined to be 148 ns, longer than that of the mixed-BA QDs with a PL lifetime of 124 ns (Fig. S12 in the ESM). Collectively, the decreased spectral shift and extended carrier lifetimes suggest that trap-assisted non-radiative recombination is suppressed in the BA-free QD films [41–43].

To further evaluate the influence of BA-induced trap-state density in the films, we fabricated electron-only devices with an FTO/ZnO/PbS-PbI₂/Al architecture and characterized them using SCLC measurements. As shown in Figs. 3(d) and 3(e), the BA film exhibits a trap state density of $2.0 \times 10^{16} \text{ cm}^{-3}$ which was calculated by the trap-filled limit voltages (V_{TFL}), whereas the BA-free film shows a markedly lower value of $1.4 \times 10^{16} \text{ cm}^{-3}$. This reduction in trap density is consistent with the extended average carrier lifetime and PL lifetime, suggesting a suppressed trap-induced recombination in the BA-free solid. The mobility was further calculated to be $2.0 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for the BA-free and $1.5 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for the BA films, as obtained from SCLC measurements. Ultimately, these results indicate that the BA-free QDs exhibit reduced trap-related recombination losses, resulting in improved optoelectronic performance of the films [44–48].

3.4 Solution processability

Devices fabricated with BA-free inks with an improved photovoltaic performance. The device structure was FTO/ZnO/PbS-PbI₂/PbS-EDT/Au, as shown in the cross-section (Fig. 4(a)). Figure 4(b) presents the current density–voltage (J – V) curves measured under AM 1.5G illumination, with the average and best-performing photovoltaic parameters summarized in Table 1. Statistical analysis of 12 devices per condition showed the device using the BA-free inks achieved a PCE of 12.1%, with a short-circuit current density (J_{sc}) of 29.4 mA cm^{-2} , an open-circuit voltage (V_{oc}) of 0.63 V, and a fill factor (FF) of 65%. In comparison, devices from BA ink showed lower J_{sc} (27.0 mA cm^{-2}), V_{oc} (0.61 V), FF (67%), and a final PCE of 11.0%. The EQE of the BA device was lower than that of the BA-free device across the spectral range from 350 to 1200 nm (Fig. 4(c)). To evaluate carrier recombination in BA-free devices, we analyzed the ideality factor (n) from the light-dependent J – V measurement. The n value decreased from 1.75 for the BA device to 1.62 for the BA-free device (Fig. S13 in the ESM), consistent with reduced trap-assisted recombination [49]. The lower dark current density (J_0) in our device is further consistent with a reduction in trap density [50]. The lower trap density in BA-free films suppresses non-radiative recombination, consistent with the higher device parameters, including improved J_{sc} , V_{oc} and EQE.

Devices fabricated from the BA-free ink after storage exhibited higher PCEs than those from the aged BA ink. As shown in Fig. 4(b), solar cells fabricated from the BA-free ink stored for 60 days (BA-free-60 days) achieved a PCE of 10.8%, retaining approximately 90% of the initial efficiency (12.1%). The J_{sc} of the aged device was 28.9 mA cm^{-2} , nearly identical to that of the fresh device (29.4 mA cm^{-2}), as confirmed by their consistent EQE curves (Fig. 4(c)). In contrast, devices with BA ink stored for only 4 h (BA-1 day) exhibited a significant decrease in PCE to 0.3%, demonstrating its rapid performance decay upon storage and highlighting the superior performance of the BA-free QD ink compared to BA ink at storage durations. Importantly, the strategy was also applicable to larger QDs with a first excitonic peak at

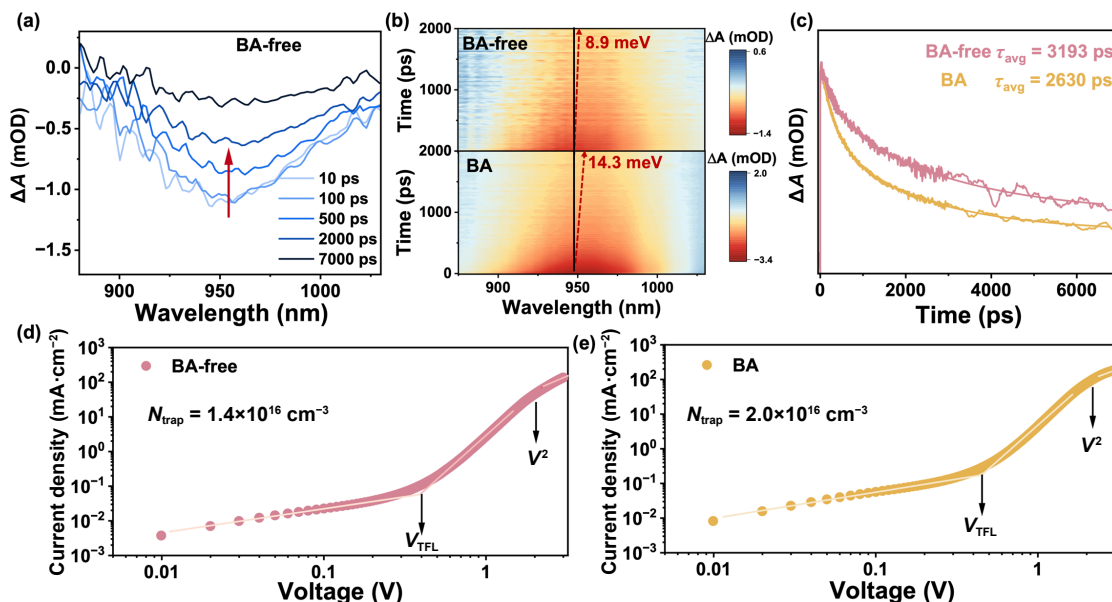


Figure 3 Remarkable optoelectronic properties of BA-free ink. (a) Transient absorption spectra at various delay times for BA-free QD. (b) Transient absorption map showing the red-shift of the maximum bleach signal over time for BA-free and BA QDs. (c) Kinetic traces at the exciton bleach peak of 950 nm QD films, where the solid lines represent multi-exponential fits. SCLC plots of (d) BA-free QD film and (e) BA QD film.

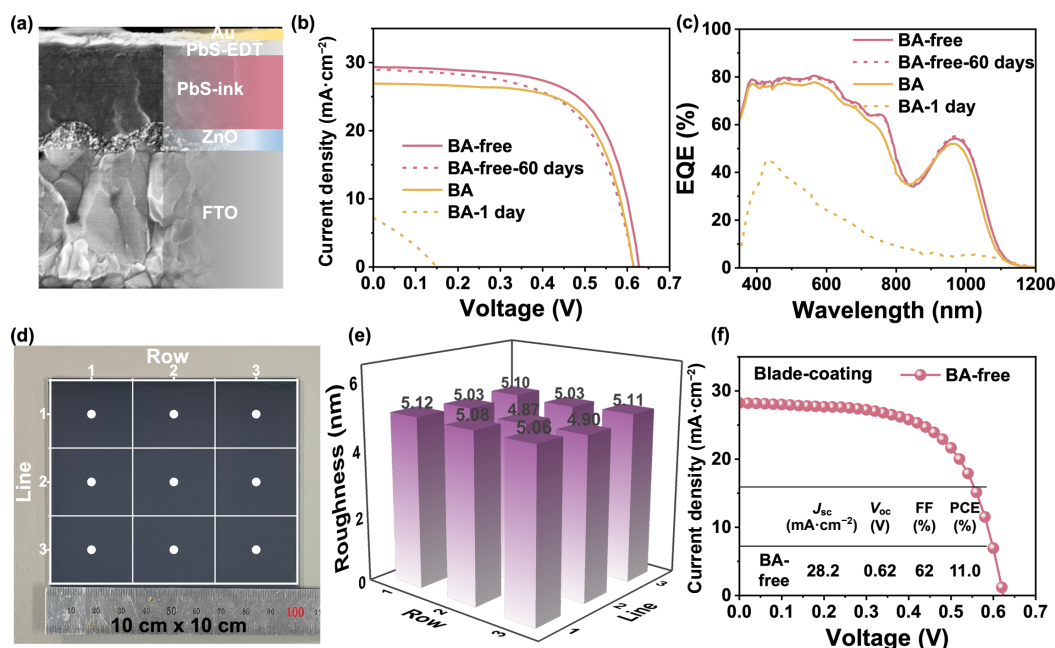


Figure 4 High photovoltaic performance and solution processability of PbS-PbI₂ QD inks with BA-free solvent. (a) Cross-sectional image of the QD solar cells. (b) *J*-*V* curves and (c) EQE curves of devices fabricated from BA-free and BA inks, where solid and dashed lines correspond to devices made with fresh ink and ink aged for 60 days, respectively. (d) Large-area (100 cm²) QD film prepared via blade-coating the BA-free ink. (e) Surface roughness was measured at nine different positions (the white dots in (d)) on the BA-free film by AFM measurement. (f) *J*-*V* curve of the BA-free QD solar cells fabricated by the blade-coating method.

Table 1 Best-performing and average photovoltaic parameters of solar cell^{a,b}

	J_{sc} (mA·cm ⁻²)	V_{oc} (V)	FF (%)	PCE (%)
BA-free	29.1 ± 0.82 (29.4)	0.63 ± 0.01 (0.63)	64 ± 1.64 (65)	11.7 ± 0.36 (12.1)
BA-free-60 days	28.6 ± 1.06 (28.9)	0.62 ± 0.01 (0.61)	64 ± 2.30 (61)	10.3 ± 0.47 (10.8)
BA	26.89 ± 1.05 (27.0)	0.62 ± 0.01 (0.61)	63 ± 3.51 (67)	10.8 ± 0.59 (11.0)
BA-1 day	6.06 ± 0.92 (7.2)	0.06 ± 0.05 (0.15)	26 ± 5.61 (30)	0.2 ± 0.35 (0.3)

^aThe individual parameters of the best-performing device are shown in parentheses. ^bStandard deviation was based on 12 solar cells for each consideration.

1100 nm, for which surface control is typically more demanding, and delivering a PCE of 11.0% (Fig. S14 in the ESM).

Beyond improved photovoltaic performance and storage stability, the low volatility and thermal stability of the BA-free ink enable scalable blade-coating deposition (Fig. 4(d)). The controlled fabrication of large-area, high-quality films is critical for QD devices from lab-scale to industrial manufacturing. Conventional large-area processes are often limited by the rapid evaporation of BA solvents, leading to local QD aggregation and film voids and ultimately resulting in poor film uniformity and significant variations in device performance [51]. In contrast, the low volatility of the BA-free ink slows solvent evaporation during blade coating, supporting controlled film deposition for orderly QD self-assembly [52]. To improve film uniformity, blade-coating was performed on a heated substrate at 80 °C, requiring the ink to remain thermally stable during processing. This requirement is met by the BA-free ink, as shown by its unchanged first excitonic absorption near 930 nm and photoluminescence near 1100 nm after heating at 80 °C for 15 min (Fig. S15 in the ESM). Benefiting from these favorable stabilities, the large-area (100 cm²) blade-coated film from the BA-free QD ink exhibited consistent roughness across nine measured points, with an average RMS roughness of 5.03 ± 0.09 nm, indicating a remarkably flat surface for this large-area film with negligible influence from residual QD aggregation (Fig. 4(e) and Fig. S16 in

the ESM). Blade-coated devices with an active area of 0.0314 cm² achieved a PCE of 11.0% (Fig. 4(f)), which is comparable to that of spin-coated devices and competitive with reported blade-coated PbS QD photovoltaics [8, 31, 38, 53]. Notably, blade coating also consumes less QD ink than spin coating (Table S4 in the ESM).

Last but not least, the BA-free ink will not cause the etching of ZnO electron transport layer (ETL), adapting better to ZnO ETL than the conventional BA-based ink (Fig. S17 in the ESM) [54, 55]. Moreover, unencapsulated BA-free devices under continuous AM 1.5G illumination showed a 30% improvement in PCE decay during the first 12 h of maximum power point tracking (Fig. S18 in the ESM) compared with the BA device. The devices also exhibited excellent long-term storage stability (Fig. S19 in the ESM), indicating greater application potential.

4 Conclusion

In summary, we proposed an amine-vapor-assisted pre-solvation strategy to enable stable dispersion of PbI₂-capped PbS QDs in BA-free solvents. By restricting BA to a brief, removable pretreatment, this strategy avoids prolonged BA exposure during ink preparation, allowing the resulting BA-free inks to retain their structural and optoelectronic properties for over 60 days under ambient conditions. Moreover, the BA-free QD films exhibit reduced trap density and suppressed non-radiative recombination. As a result,

solar cells fabricated from fresh BA-free inks achieve a PCE of 12.1%, while devices based on inks stored for 60 days retain 90% of their initial PCE (10.8%). In contrast, devices fabricated from BA inks undergo rapid performance loss upon storage, with the PCE decreasing from 11.0% to 0.3% after 4 h. Benefiting from the low volatility and thermal stability of the BA-free ink, large-area films can also be fabricated by blade-coating, yielding devices with a PCE of 11.0%. These results establish BA-free inks as a promising route toward stable and scalable solution processing for QD optoelectronics and potentially extend amine-free solution processing beyond PbS QDs.

Electronic Supplementary Material: Supplementary material (experimental details, additional characterization data (Figs. S1–S19), and tables (Tables S1–S4)) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908957>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

X. F. L.: Data curation, writing manuscript, methodology, experimental design. S. J.: Data curation, validation. Z. H. W.: Conceptualization, formal analysis. S. J.: Data curation, formal analysis. Z. X. G.: Methodology. Y. L. W.: Project administration, supervision, resources, funding acquisition, formal analysis. C. W.: Conceptualization. H. L.: Formal analysis, methodology. X. C. G.: Data curation. Y. L. L.: Validation. J. B. L.: Data curation, methodology. L. L. W.: Formal analysis. H. K.: Project administration, supervision, resources, funding acquisition, formal analysis. X. T. Z.: Project administration, supervision, resources, funding acquisition, formal analysis. Y. C. L.: Funding acquisition, resources. All the authors have approved the final manuscript.

Use of AI statement

None.

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