

Characterization of stability and challenges to improve lifetime in perovskite LEDs

Low stability of perovskite light-emitting diodes (PeLEDs) is the biggest obstacle to the commercialization of PeLED displays. Here, we cover the current status and challenges in analysing and improving the stability of PeLEDs and suggest some advice that will benefit the community to boost the operational lifetime of PeLEDs.

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Perovskite light-emitting diodes (PeLEDs) are promising candidates for the light source of next-generation displays due to their narrow emission spectra, colour-tunability, and low-cost manufacturing process¹. The biggest merit of employing metal halide perovskites (MHPs) as light-emitting materials stands out when it comes to reproducing the BT.2020 wide-colour gamut. Contrary to inorganic quantum-dot (QD) particles, for which the colour is tuned by the size of the particles, the colour of MHP nanocrystals can be tuned by the content of halogens at the size beyond the quantum confinement regime so that it could benefit from the size-independent narrow spectra with full width at half maximum ≈ 20 nm. In the same context, PeLEDs are more advantageous for displays satisfying BT.2020 than organic light-emitting diodes (OLEDs), which exhibit broad emission spectra arising from multiple vibronic transitions.

Since the reports of bright PeLEDs at room temperature in 2014, the efficiency has rapidly increased through seven years of intensive research and development around the world^{2,3}. The external quantum efficiencies (EQEs) of green and red PeLEDs, which are 23.4% and 23% respectively^{4,5}, are approaching the theoretical out-coupling efficiency ($\eta_{\text{out}} \approx 30\%$ for nanoparticle PeLEDs⁴) of thin-film LEDs assuming isotropic transition dipole of the emitter. However, the reported best operation lifetimes of PeLEDs are only in the range of several hundred hours at 100 cd m^{-2} , which are insufficient for display applications^{6,7}. Recent studies attempted to overcome the low device stability, but breakthroughs beyond the current slow progress have not been reported, mostly pointing out that the ion migration must be suppressed to realize long-term stability. Although stability is considered a critical issue among the PeLED community, many papers still do not report operational lifetime. Moreover, the conditions for the lifetime measurements among papers reporting the lifetime of PeLEDs vary, and

Table 1 | Lifetime comparison of PeLEDs and OLEDs

	PeLEDs			OLEDs		
	Blue	Green	Red	Blue	Green	Red
LT50 (hours) L_0 : shown below	12 ^a	255 ^b	317 ^c	-	-	-
LT50 (hours), $L_0 = 1,000 \text{ cd m}^{-2}$	0.4 ^d	11 ^d	112 ^d	11,000 ^e	400,000 ^e	250,000 ^e

^aLT50 (half lifetime) at L_0 (initial luminance) = 102 cd m^{-2} (ref. ⁸). ^bLT50 at $L_0 = 120 \text{ cd m}^{-2}$ (ref. ⁹). ^cLT50 at 30 mA cm^{-2} (about $L_0 = 500 \text{ cd m}^{-2}$, ref. ⁷). ^dEstimated value using an acceleration factor $n = 1.5$. ^eRef. ⁹.

include constant current, constant voltage, or a specific initial brightness (Table 1). This leads to difficulties in making a reliable comparison between devices and understanding the mechanisms underlying the degradation of PeLEDs.

To accelerate the commercialization of PeLED displays, more attention should be focused on the operational lifetime of PeLEDs. Here, we address the current status of the stability of PeLEDs, suggest some advice for reliable lifetime measurements, and cover challenges and the outlook to improve the lifetime of PeLEDs.

Current lifetime status of PeLEDs

The operational lifetimes of state-of-the-art PeLEDs are 1.5 h, 255 h and 317 h for blue (LT50 at $L_0 = 100 \text{ cd m}^{-2}$) green (LT50 at $L_0 = 120 \text{ cd m}^{-2}$) and red (LT50 at $L_0 = 500 \text{ cd m}^{-2}$) devices, respectively (Table 1)^{6–8}. These lifetimes are not only much shorter than the lifetime of current OLEDs⁹, but are also much shorter than the requirements for display applications, which are at least several tens of thousands of hours at $1,000 \text{ cd m}^{-2}$ for green and red OLEDs¹⁰. It might be frustrating considering the large discrepancy between the current lifetimes of PeLEDs and the minimum lifetime requirements for practical use. However, we should note that half lifetime (LT50) at the initial luminance (L_0) of 50 cd m^{-2} of the first OLED was only 100 h in 1987¹¹. It took about 20 years for the lifetime of OLEDs to reach the sufficient level for

display applications, which led to the rapid adoption of OLED displays in smartphones in the 2010s. Considering the relatively short period of PeLED research, much more progress can be made to improve the operational lifetime of PeLEDs.

There are only several papers reporting LT50 exceeding 100 h, and lifetimes of a few minutes are frequently observed. Ion migration induced by the electric field applied during the operation is thought to be the primary origin of the poor stability of PeLEDs^{12,13}. Interfacial reaction, electrochemical reaction and external factors such as moisture and temperature are also considered as the origin of device degradation. However, underlying mechanisms of degradation are not understood yet. This lack of understanding stems from the complicated situation occurring inside a device composed of various types of materials. Comprehensive investigation from a chemical and electrical perspective based on the luminance–time (L – T) curve of operating devices leads to a better understanding of the device degradation. Decay characteristics of L – T curves contain information about the degradation routes, and comparing L – T curves of different devices can lead to new ideas to improve device stability. However, L – T curves of reported PeLEDs are measured at various initial luminance, which hinders meaningful comparison between devices. It is understandable considering the overshoot that occurs at the

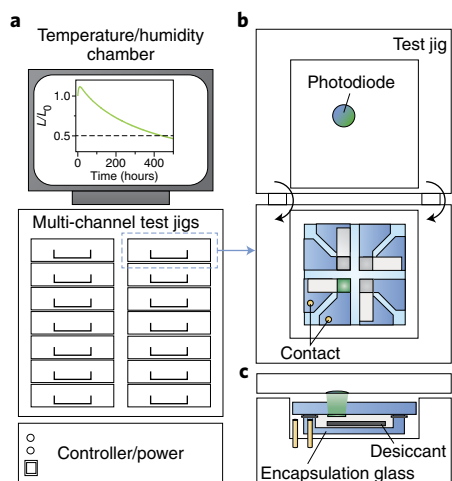


Fig. 1 | Lifetime measurement setup for PeLEDs.

a, Multi-channel test jigs in a temperature/humidity chamber. **b,c**, Top view (**b**) and side view (**c**) of a test jig and a device.

initial stage of lifetime measurement, which makes it difficult to accurately control the initial brightness. Moreover, luminance overshoot also leads to large variation of the operational lifetime, depending on which point is set as the initial luminance. A consensus stability testing protocol for organic solar cells was reported in 2011 and has guided the solar cell community for reliable stability measurements¹⁴. Recently, a consensus statement for reporting stability of perovskite photovoltaics has been announced to further unify and standardize the stability assessment¹⁵. Similar standards or protocols would support the PeLED community to boost stability, and we hope our Comment could also contribute to this goal.

How to measure lifetime of PeLEDs

The lifetime measurement setup and procedure are pretty much the same as those of OLEDs. Setup is composed of test jigs, a multi-channel test jig frame, a controller, a computer, and a temperature/humidity chamber where all components are placed (Fig. 1). A fabricated device is mounted to a jig with a photodiode and electrical contact. The photodiode is placed on top of the operating cell when the jig is folded, and the luminance decay of the device is measured by the photocurrent from the photodiode. The detailed procedure for lifetime measurement is described in Table 2. For the initial luminance of the lifetime measurement, we recommend both 100 cd m^{-2} and $1,000 \text{ cd m}^{-2}$ because the former would be more suitable to study the degradation mechanism considering

the current stage of PeLED's lifetime, and the latter would be more useful to reflect the operating condition of real display applications, which are more suitable for the comparison with OLED's lifetime.

Lifetime trends and analysis of PeLEDs

Luminance overshoot is frequently observed from PeLEDs, and the lifetime can be overestimated if it is not considered. We recommend setting the maximum luminance as the actual initial luminance of the lifetime test to determine the LT_{50} , as shown in Fig. 2a.

The decay characteristic of $L-T$ curves and fitting them with decay models gives information about the degradation mechanism of the device^{16,17}. Several functions used for the fitting of $L-T$ curves of LEDs are shown in Fig. 2a. Single exponential decay (model (1)) is the most basic function. However, if multiple origins cause the degradation, modified functions such as a stretched exponential function or a combination of exponential functions might better fit the $L-T$ curve (model (2) or (3)). Overshoot can also be fitted by adding an exponential function with a short lifetime and negative coefficient (model (4) or (5))¹⁸. An example of fitting the reported $L-T$ curve of a 3D bulk polycrystal and 3D/2D hybrid PeLEDs is shown in Fig. 2b¹⁹. The $L-T$ curve of the 3D PeLED¹⁹ cannot be well-fitted due to the huge overshoot and the catastrophic breakdown. In the bulk polycrystal perovskite of 3D PeLED, there are many defects and dangling bonds at the grain boundaries. When the electric field is applied to the 3D PeLEDs, the perovskite at the grain boundaries decomposes quickly, and thus the ions migrate along the grain boundaries, accelerating the decomposition. On the other hand, 2D shells covering the grain boundaries passivate the defects and suppress the ion migration, leading to the reduced overshoot and luminance decay without breakdown. The $L-T$ curve of the 3D/2D hybrid PeLED can be fitted well with a multi-exponential function (for example, three or more exponential functions) (model (4)), implying that multiple origins of $L-T$ decay characteristics exist, which can be attributed to overshoot, short-term degradation and long-term degradation, respectively. Although fitting $L-T$ curves with exponential models provides clues to understanding the degradation mechanisms, a comprehensive model based on possible physical mechanisms is required to unravel the exact underlying degradation mechanisms^{16,17}. Luminance overshoot of PeLEDs is known to be caused by initial facile migration of mobile ions mostly along grain boundaries without significant

ion migration inside a grain (intragrain migration) and accumulation of ions at the device interfaces that facilitates the charge injection due to a lowered effective injection barrier¹⁹. For comprehensive analysis in the regime of luminance decay, we may consider all possible physical degradation routes in the polycrystalline perovskite layer, which is caused by intragrain migration and ion migration from grain bulk to grain surface as well as the ion migration along grain boundaries¹⁹. By the formation of granular-like grains and 2D shells covering the grains in 3D/2D perovskites, ion migration along grain boundaries and ion migration from grain bulk (3D) to grain surface (2D) is expected to be suppressed compared to those in 3D perovskites. Meanwhile, the energy barrier of ion migration within a grain in 3D/2D perovskites would be similar to that in 3D perovskites so that luminance decay originating from intra-grain ion migration would occur in both 3D and 3D/2D perovskites. Because the intragrain ion migration could be much slower than the ion migration in the grain boundaries, the degradation time constant that originates from intragrain ion migration can be much longer than that in the grain boundaries. Because we need to consider the interfaces and electrodes to understand the degradation in the devices, the degradation pathways can be more complicated. Based on these possible physical mechanisms, a degradation model could be developed to further study the underlying degradation mechanism of PeLEDs.

Accelerated lifetime test of PeLEDs

The accelerated lifetime test (ALT) is useful for reducing the time spent on the lifetime measurement. The ALT method is especially useful for lifetime measurements of PeLEDs where setting an exact initial luminance is difficult due to the luminance overshoot. A lifetime at a specific initial luminance can be obtained from the inverse power law shown in Fig. 2c. The acceleration factor n can be determined from the $LT_{50}-L_0$ plot shown in Fig. 2c. The acceleration factor depends on the material and device structure. For example, acceleration factors in the range of $n = 1.4$ to $n = 2.0$ have been reported for typical OLEDs. For PeLEDs, an acceleration factor of $n = 1.5$ determined from the $LT_{50}-L_0$ plot in the initial luminance range of $1,000 \text{ cd m}^{-2}$ to $20,000 \text{ cd m}^{-2}$ was reported²⁰. In Lin et al's paper²⁰, the estimated LT_{50} at $L_0 = 100 \text{ cd m}^{-2}$ extrapolated from the $LT_{50}-L_0$ plot was 100 h, which was a largely overestimated value compared to the experimentally obtained LT_{50} of 46 h. Another paper²¹ also reported

Table 2 | Procedure for lifetime measurement of PeLEDs

Step	Procedure
1	Properly encapsulate fabricated devices using ultraviolet-cured resin. Spin-coated perovskite or transporting materials on the region where resin is applied must be removed from the substrate before the encapsulation. Inserting a desiccant in the encapsulated device is recommended.
2	Measure the current density–luminance (J – L) curve using a source meter and a spectrometer.
3	Find the current density at which the device operates at the desired initial luminance (L_0) (for example, 100 cd m ⁻² , 1,000 cd m ⁻²) for the lifetime measurement.
4	Check if the temperature/humidity chamber is working properly.
5	Mount the device to a test jig in the position where the fresh cell is placed under the photodiode.
6	Set the current density that corresponds to the desired initial luminance and start the measurement. Both 100 cd m ⁻² and 1,000 cd m ⁻² are recommended as initial brightness for lifetime measurement.
7	End the measurement when the luminance from the photodiode becomes lower than half of the initial luminance (L_0) and determine the half lifetime (LT50).
8	If the device shows luminance overshoot, set the maximum luminance as the initial luminance as shown in Fig. 2. Measuring LT50 at multiple initial luminance points in the range of 100–10,000 cd m ⁻² is necessary to extract the acceleration factor for the conversion of LT50 at non-standard initial luminance for the direct comparison of LT50 at 100 cd m ⁻² or 1,000 cd m ⁻² .
9	Measure the spectrum of the aged device to check the colour stability.
10	When performing an accelerated lifetime test at high current density, multiple initial luminances should be tested for the reliability of the acceleration factor. Also, check the temperature of the device as it can overheat.

an overestimated value of LT50 = 108 h at $L_0 = 100$ cd m⁻² calculated by using the acceleration factor $n = 1.5$ where the experimentally measured LT50 was 21 h. These reports imply that there may be other routes such as spontaneous degradation without an electric field including decomposition of perovskite layers, interfacial electrochemical reaction with electrodes caused by mobile ions (for example, corrosion, generation of insulating products and so on), or electrochemical degradation caused by moisture and oxygen infiltrated through imperfect encapsulation. Even under perfect encapsulation, such electrochemical reactions may occur in PeLEDs, which leads to the acceleration of the device. Therefore, unlike OLEDs, the solid buffer layers to prevent the migration of mobile ions that react with both the electrodes should be designed more carefully.

Causes of degradation in PeLEDs

The major origin of the degradation of PeLEDs is known to be the ion migration under an electric field during operation. Ion migration within perovskite results in the vacancy defect inside perovskite, accumulation of halide interstitial at the interface, and distortion of crystal structure^{19,21,22}. These lead to the formation of

trap states and nonradiative recombination centres in the perovskite, which results in the decrease of radiative recombination efficiency of the perovskite emitting layer.

An electric field applied to the device can also induce the ion migration across the interface and into the charge-transporting layers and electrodes. Migrated ions affect the conductivity of charge-transporting layers, which results in the shift of recombination zone or charge balance of the device. Joule heating during the operation of PeLEDs leads to the increase of temperature and may accelerate the degradation caused by ion migration^{23,22}. In a colour-tunable mixed-halide perovskite device, ion migration can result in phase segregation under electrical bias or illumination, which leads to the poor stability and colour purity of the devices^{24,25}. Under an electric field, an electrochemical reaction can occur in the perovskite layer, which leads to the decomposition of the perovskite emitting layer resulting in PbX₂ and halide gas products²⁶, or methylamine and hydrogen bromide gas products²⁷. These gases delaminate the layers in the device and lead to the formation of dark spots. When these gases become accumulated at the interfaces, pores are created, resulting in a catastrophic breakdown of PeLEDs.

Ion migration of halide ions across the interface and reaching the metal electrode may lead to the corrosion of cathodes. Degradations caused by delamination or electrode corrosion are irreversible, whereas moderate ion migration inside perovskite, such as interstitial accumulation, is reversible²². Besides the degradation caused by perovskite, PEDOT:PSS is also known to cause the degradation of PeLEDs. Acidic PEDOT:PSS could etch the ITO and water residue in the PEDOT:PSS could decompose the perovskite layer²².

Strategies to improve stability of PeLEDs

Strategies to improve stability could be divided into two categories: (1) passivation of defects, and (2) suppression of ion migration. During the operation of PeLEDs, defect state density increases due to the ion migration and decomposition of the perovskite layer. In particular, defects are highly accumulated at the grain boundaries. If defect-passivating additives are incorporated in the perovskite layer, additional defects formed during the device operation could also be passivated, suppressing the increase of the non-radiative recombination centre. The incorporation of defect-passivating additives such as lithium halides, phosphine oxide derivatives, and acetate derivatives successfully enhanced the operational stability of PeLEDs^{21,28,29}.

Suppressing ion migration is also an effective strategy to improve the stability of PeLEDs. Ion migration in PeLEDs can be mainly divided into three types: (1) ion migration within a grain, (2) ion migration along or across grain boundaries, and (3) ion migration across device interfaces^{19,22}. Ion migration within a grain could be suppressed by replacing the methylammonium cation with other cations like larger formamidinium cation or inorganic caesium cation, which increase the energy barrier for ion migration. Mixing A-site cations such as formamidinium, caesium and rubidium, which leads to triple- or quadruple-cation perovskites, is also an effective strategy to improve the stability and may also suppress phase segregation in mixed-halide perovskites²⁴. Also, incorporating a small amount of alkali cations such as potassium into interstitial sites of crystal lattice can also lead to an increased diffusion barrier of halide anions³⁰. Ion migration along or across grain boundaries can be suppressed by covering grain boundaries with bulky organic cations, ligands or small organic molecules^{13,19}. By adding a small amount of bulky organic cation to the perovskite precursor, 3D/2D hybrid perovskite, where 2D perovskites cover the surface of 3D perovskite along the grain boundaries, can be

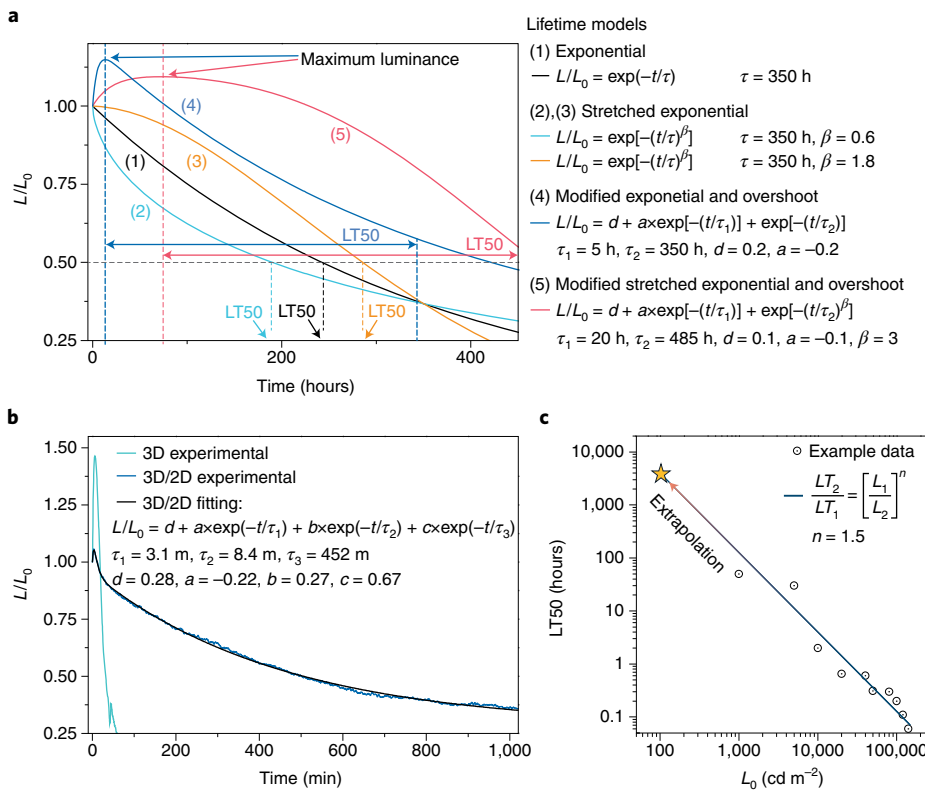


Fig. 2 | Lifetime characteristics of LEDs. **a**, Lifetime models for luminance–time (L - T) curve. **b**, Measured L - T curves of 3D and 3D/2D PeLEDs¹⁹ and its fitted curve. **c**, An example of an accelerated lifetime test (ALT) plot. Acceleration factor $n = 1.5$ was used. L_0 is initial luminance. LT50 is half lifetime. Panel **b** adapted with permission from ref. ¹⁹, Springer Nature Ltd.

fabricated¹⁹. The 2D perovskite blocks the ion migration across grain boundaries, leading to an improved operational lifetime¹⁹. By crosslinking the organic ligands that passivate the grain boundary can further improve the operational lifetime by enhanced blocking of ion migration across grain boundaries¹³. Ion migration across device interfaces can be suppressed by inserting interlayers such as alkali metal halides between perovskite and transporting layers⁶. Compact and robust inorganic transporting layers such as zinc sulfide also suppress ion migration across interfaces. By incorporating LiF interlayers combined with an all-inorganic device structure, a long operational lifetime of LT50 = 255 h at $L_0 = 120$ cd m⁻² has been reported⁶. Besides engineering materials, managing Joule heating with heat sinks or pulsed operation could also improve the operational lifetime of PeLEDs²³.

Challenges and outlook to improve lifetime of PeLEDs

Some selected PeLEDs with long lifetime, and their strategies to improve stability, are shown in Fig. 3^{6,13,19–21,28,29}. Given

the current status of the stability of PeLEDs, an all-inorganic device seems to be the most effective strategy for long-operational-lifetime devices. Compact and robust inorganic interlayers and transporting layers can suppress the ion migration across the device and prolong the device lifetime. Although the operational lifetime of the all-inorganic device is the longest among devices shown in Fig. 3, its lifetime is in the same order as other devices. This indicates that inorganic layers do not eliminate the fundamental cause of the degradation of PeLEDs; the intrinsic degradation of perovskite emitting layers. Moreover, the refractive index of inorganic layers such as zinc sulfide exceeds $n = 2.1$ in the visible wavelength range, which leads to the low out-coupling efficiency.

Reported stable perovskite solar cells exceed 1,000 h of T_{80} (operational lifetime to reach 80% of the initial efficiency) even under a strong illumination of air mass 1.5 global (AM 1.5G)¹⁵. Considering similar materials and structures used for perovskite LEDs and solar cells, we may infer that much shorter operational lifetimes of PeLEDs

originate from accelerated ion migration under a higher electric field than that in solar cells²². Although device lifetimes of PeLEDs have been improved by various strategies, it is challenging to solve the low intrinsic stability of perovskites arising from ion migration under an electric field during operation. It has been reported that the bulky cations of 2D or quasi-2D perovskites suppress ion migration in the out-of-plane direction¹². Only considering the suppression of ion migration, utilizing the emitting layer composed of 2D perovskites oriented in the in-plane direction of the substrate might be a good solution to improve the intrinsic stability of perovskites. However, 2D or quasi-2D PeLEDs with a long operational lifetime and high efficiency have not been reported yet. This is probably due to the low conductivity of bulky organic cations, high defect state density due to the high surface-to-volume ratio, low radiative efficiency arising from electron–phonon coupling³¹ and singlet–triplet exciton splitting³², susceptible lateral ends that are not passivated by organic cations, or non-horizontally oriented 2D or quasi-2D perovskites. Therefore, incorporating bulky organic cations with hole or electron transporting properties, controlling the number of 2D perovskite layers (n -value) to balance the radiative efficiency and the ion-migration suppression, and engineering the fabrication method to align the 2D or quasi-2D perovskites in the in-plane direction of substrates may lead to the increase of efficiency and lifetime of PeLEDs.

The core/shell-like grain structure in which 3D bulk grains are covered with shells such as bulky organic cations might be a better strategy to realize long-lifetime PeLEDs. In 3D/2D core/shell-like polycrystalline perovskites, unlike 2D perovskites, grain boundaries in all directions are passivated by the 2D shell so that ion migration can be suppressed in all directions¹⁹. Crosslinking the organic ligands¹⁴ or incorporating robust organic ligands may lead to further enhanced stability of core/shell-like polycrystalline PeLEDs.

Perovskite nanoparticles (PeNPs) covered by organic ligands can also be used as light-emitting materials, and high-efficiency PeLEDs based on PeNP have been realized recently⁴. However, the operating lifetime of these PeLEDs is generally much lower than that of polycrystalline perovskite-based PeLEDs. The reason may be that perovskite nanocrystals with high surface-to-volume ratios are more susceptible to form trap states during device operation, resulting in a faster drop in luminous efficiency. In

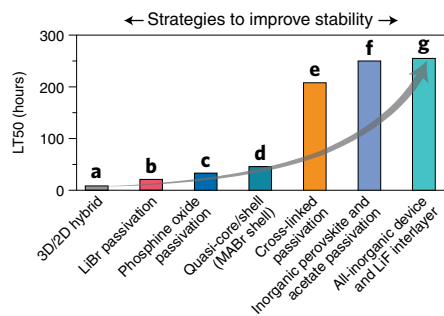


Fig. 3 | Some selected PeLEDs with long lifetime, and their strategies to improve stability. The strategies for each report are as follows. **a**, 3D/2D hybrid perovskite¹⁹. **b**, LiBr passivation²¹. **c**, Phosphine oxide passivation²⁸. **d**, MABr shell²⁰. **e**, Cross-linked passivation¹³. **f**, Acetate passivation²⁹. **g**, All-inorganic device and LiF interlayer⁶.

addition, alkyl ligands required for the synthesis process of nanocrystal are long and insulating, which results in the poor packing of PeNPs and poor charge injection and transport. Core/shell structured PeNPs in which organic ligands are replaced with inorganic shells such as chalcogenide or oxides might be a solution to overcome the poor stability of conventional PeNPs³³. Since inorganic shells are more robust and denser than alkyl ligands, they might better passivate surface defects and suppress ion migration during the device operation. In addition, charge transport through the inorganic shell could be facilitated by compositional engineering. For example, operational stability and charge

transport are simultaneously improved in PeLED based on CsPbI₃/Pbs core/shell PeNPs³⁴.

Concluding remarks

In this Comment, we covered the characterization and current status of operational stability of PeLEDs and challenges to improve operational lifetime for the commercialization of PeLEDs. We also provide a lifetime measurement procedure for the community to report reliable lifetime data and to suggest analysing lifetime trends to unravel the underlying degradation mechanism of PeLEDs. Implementing the lifetime measurement procedures and suggestions we provide will benefit the community to boost up the operational lifetime of PeLEDs. □

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