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Supplementary Information

Intrinsic Stability of Perovskite Materials and their Operational Stability in Light-Emitting Diodes

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Emission layer (ref)	**Device structure**	**EQE (%)**	**Lifetime**	**Reason for instability**	**Strategies to overcome instability issue**	**EL Peak (nm)**	**Stability condition (cdm^{-2} or mAcm^{-2})**
CsPbBr$_3$ QDs(1)	ITO/PEDOT:PSS/PTAA/QDs/TPBi/LiF/Al	18.7	15.8 h	Surface and vacancy defects promote non-radiative recombination	Bilateral passivation of QDs EL	516	100 cd m^{-2}
GuI: CsPbI$_3$ (2)	ITO/PEDOT:PSS/ CsPbI$_3$ NCs / TPBi/LiF/Ag	13.8	20 min	Surface defects	GuI effectively passivates the halide vacancy defects of perovskites and increase the exciton confinement	695	25 mA cm^{-2}
Cd-CsPb(Br$_x$Cl$_{1-x}$)$_3$ QDs(3)	ITO/PEDOT:PSS/poly-TPD/anion-	14.6	12 min	Deep donor defects by Halide vacancies	Doped Cd ions reduces the formation energy of halogen vacancies filled by the	490	134 cd m^{-2}

	exchange QDs/TPBi/LiF /Al				external bromine ions. Free bromine ions in solution passivates the halogen vacancies, surface long-chain ligands also passivate the perovskite surface during the anion exchange process		
FABr induced formation of $CsPbBr_3$&Cs_4PbBr_6 with Type-I junction perovskite films(4)	ITO/NiMgLiO$_x$/$CsPbBr_3$&Cs_4PbBr_6/MABr &FABr/PMMA/B_3PYMPM/LiF/Al	22.3	59 h	low charge transport of Cs_4PbBr_6	No lattice mismatch between $CsPbBr_3$ and Cs_4PbBr_6. Cs_4PbBr_6 effectively passivates the Cs^+ and Br^- vacancy defects in $CsPbBr_3$ emission layer.	527	130 cd m^{-2}
Quasi-2D $(PEA/m\text{-}F\text{-}PEA)_xNMA_{1-x})_2CsPb_2I_7$(5)	ITO/the modified PEDOT:PSS /poly-TPD /PVK /quasi-2D perovskite /TPBi /LiF Al	25.8	35 min	Uncontrolled phase distribution and defect states	Controlled phase formation of multiple cation quasi-2D structure growth and passivation of Pb and I vacancy defects.	680	100 cd m^{-2}
PEA-$CsPbBr_3$ NCs (6)	ITO/PEDOT:PSS/ TFB with BPQDs/quasi-2D perovskite with PEA-$CsPbBr_3$NCs /TPBi/LiF/Al	25.32	174 min	Defects and unbalanced charge transfer	Leakage current reduction and passivation of lead and halide defects	532	3000 cd m^{-2}
EDA$Cs_{n-1}Pb_nX_{3n+1}$ (n = 1–	ITO/ZnO/PEIE /perovskite	17.03	15.4 h	Defects, Ion migration and strain	$EDABr_2$ preferentially bind with Pb-Br octahedra forms a quasi 2D phase with Br rich	671	20 mA cm^{-2}.

4) quasi-2D perovskites(7)	/TFB/MoO$_3$/Au				capping layer around 3D perovskite structure, Which reduces the lattice distortion, strain and defect formation. Moreover, it enhances the diffusion barrier of halide ions long the grain boundaries which results in mitigation of ion migration.		
PMA : β-CsPbI$_3$(8)	(ITO)/PEDOT:PSS/poly-TPD/β-CsPbI$_3$ NCs/ TPBi/LiF/Al	17.8	317 h	Phase change of CsPbI$_3$ from cubic to orthorhombic structure. Weak bonding between perovskite surface and long-chain capping ligands	PMA enhances the exciton binding energy by forming strong Pb-O bond with β-CsPbI$_3$ NCs, which inhibits the phase change and enhances the formation energy of Pb$_{Cs}$ deep defects.	689	30 mA cm^{-2}
(Cs$_{0.83}$Rb$_{0.17}$)$_{0.95}$K$_{0.05}$PbBr$_3$(9)	ITO/LiF/perovskite/LiF /ZnS/ZnSe /Ag	11.05	255 h	Ion migration Joule heating Trap states	Alkali-metal ions (Rb$^+$ and K$^+$) passivates the trap states in CsPbBr$_3$. Ultrathin LiF layers on both side of perovskite layer suppress the ion migrations and interfacial exciton quenching with ETL/HTL and facilitates the charge injection via tunneling effect. Inorganic ETLs mitigates the device degradation caused by the Joule-heating effect and	519	545 mA cm^{-2}

					supports the charge injection through better band alignment at interface.		
TFA-derived CsPbBr$_3$(10)	ITO)/PEDOT: PSS/TFA:CsPbBr$_3$/TPBI/LiF /Al.	10.5	250 h	Grain boundary and large crystal size induces trap states.	TFA$^-$ anions interact with Pb^{2+} ions and bound the perovskite crystal surface which stimulates the fast crystallization of CsPbBr$_3$ resulting small grain size perovskites.	520	100 cd m^{-2}
Quasi-core/shell CsPbBr$_3$/MABr(11)	ITO/PEDOT:PSS/perovskite/B$_3$PYMPM/LiF/Al	20.3	104.56 h	Defect states	The MABr shell passivates non-radiative defect sites and balance the charge injection	525	167 mA cm^{-2}
FA$_{0.9}$GA$_{0.1}$PbBr$_3$ (12)	ITO/polyethylenimine ethoxylated /FA$_{0.9}$GA$_{0.1}$PbBr$_3$ PNCs with or without TBTB/TPBI/LiF/Al	23.4	132 min	Liquid like dipoles of organic cation leads to low charge recombination.	Zero-dipole guanidinium cation (CH$_6$N$_3^+$) supports lattice-stabilization by forming hydrogen bonds and enhances the radiative recombination due to increased excitonic confinement, defect suppression and bulk entropy stabilization.	540	100 cd m^{-2}
MAPbI$_3$(13)	Sapphire/ITO/poly-TPD/MAPbI$_3$/PCBM/ZnMgO/AI	-	10 h And 5 h	Large device area increases the Joule heating	Reducing the device area mitigates the Joule heating which in result improves the device stability.	757	500 mA cm^{-2} And 1000 mA cm^{-2}
CsPbBr$_x$Cl$_{3-x}$(14)	ITO/PEDOT:PSS / C$_{51}$H$_{63}$N/TFB/PFI/Emission	2.75	1.1 h	Cl$^-$ anion migration during device operation produces chlorine deficient	Mitigation of anion drift pathways improves the operational stability of the LED device.	460	1 mA cm^{-2}

	film/PO-T2T/Liq /Al			region in chlorine-based perovskites which results unstable electroluminescence and device instability.			
$PEA_xPA_{2-x}(CsPbBr_3)_{n-1}PbBr_4$(15)	ITO/NiO_x/ $PEA_xPA_{2-x}(CsPbBr_3)_{n-1}PbBr_4$ /TPBi/LiF/Al	7.51	1.1 h	Defect and trap states generated during the crystallization of perovskite enhances the non-radiative recombination	Quasi-2D perovskite synthesized with an excess ammonium salt passivates the defects efficiently	450	1.5 mA cm^{-2}
$5AVA_x$–$FA_{0.47}Cs_{0.53}Pb(I_{1-y}Br_y)_3$(16)	ITO/ZnO-PEIE/ $5AVA_x$–$FA_{0.47}Cs_{0.53}Pb(I_{1-y}Br_y)_3$./ TFB/$MoO_x$/Au	8.7	8 h	Phase separation and poor charge transportation	5AVA additive improves the crystallinity, reduces the defect densities and facilitates charge transport of the perovskites.	690	100 mA cm^{-2}
NMAI-$FA_{1-x}Cs_xPbI_7$(17)	ITO/PEIE-modified zinc oxide /$NF_{1-x}Cs_xPI_7$/TFB/MoO_x/Au	7.8	31 h	Formation of interfacial barrier due to ion migration	Cs incorporation in FA based perovskites induces the contraction of cuboctahedra volume and increases the interaction between FA and iodide which in result reduces the defect densities. This process stabilizes the emission perovskite layer by mitigating the ion migration	783	10 mA cm^{-2}
4-F-PMAI :$CsPbI_3$(18)	ITO/poly-TPD/$CsPbI_3$/TPBi/LiF/Al	14.8	20 h	Phase change of $CsPbI_3$ from cubic to orthorhombic structure.	4-F-PMAI improves the stability of the α-$CsPbI_3$ phase by passivating the $CsPbI_3$ nanocrystals through in situ growth.	692	5 mA cm^{-2}

PEA_2 Cs_{n-1} Pb_nBr_{3n+1}(19)	AgNWs/AgNPs/PEDOT:PSS/$Ti_3C_2T_x$ MXene /PEDOT:PSS/PEA_2Cs_{n-1}PbnBr_{3n+1}/TPBi/LiF/Al	16.5	39.6 min	ITO electrode is brittle in nature, not suitable for flexible devices	Multi-dimensional composite electrode increases conductivity, improves charge balance, facilitates fast heat dissipation, enhances the quality, and coverage of perovskite thin films	520	0.5 mA cm^{-2}
$MAPbBr_3$(20)	ITO/$MAPbBr_3$ /Au	-	54 h	Ion migration	Freeze the ionic movement and eliminate their thermally activated relaxation processes by lowering the temperature below -110 °C	548	1 mA cm^{-2}
MABr:$CsPbBr_3$ Nano wires(21)	ITO/PEDOT:PSS/MABr modified $CsPbBr_3$ /TPBi / LiF/Ag,	16	37 min	Perovskite nano wires shows lateral invasion of water and oxygen	PAM templates prevent the lateral diffusion of water and oxygen molecule which in result mitigate perovskite nanowire degradation	530	20 mA cm^{-2}
$CsPbBr_{0.6}I_{2.4}$(22)	ITO/ZnMgO/ perovskite films with PEOXA polymer/poly-TPD/MoO_3/Ag	6.55	0.5 h	α-$CsPbBr_{0.6}I_{2.4}$ cubic perovskite transforms to δ- orthorhombic non-perovskite phase	PEOXA polymer stabilizes the α-phase of perovskite by forming bond with Pb^{2+} in PbI_2 or $PbBr_2$.	668	338 cd m^{-2}
PEA)$_2$SnI_4 (*n* = 1 QW) and (PEA)$_2$CsSnI_7 (*n* = 2 QW)(23)	ITO/PVK/perovskites multiple Quantum well structures/TmPyPB/LiF/Al	3	2h	Easy oxidation of Sn^{2+} and nonuniform film morphology results device instability	Formation of quantum wells with wider well width suppress the Auger recombination and facilitates the formation of uniform and highly emissive perovskite films and mitigate the oxidation of Sn^{2+} cations	920	10 mA cm^{-2}

$CsPbBr_3$ (24)	ITO/PEDOT:PSS/PVK/Perovskite/ TPBi/LiF/Al	10.59	5.8 h	The acidity and hygroscopicity of PEDOT:PSS degrades the perovskite film which causes operational instability of device	PVK induces balanced carrier injection, fewer interfacial defects and passivates the perovskite film from environmental moisture and PEDOT:PSS. Moreover, PVK mitigate the interfacial exciton quenching by restricting the ion migration.	516	
$(PEABr)_{0.4}CsPbBr_3$(25)	(ITO)/poly-TPD/LiF/$(PEABr)_xCsPbBr_3$/TPBi/LiF/Al	12.1	34 min	Decomposition of PEABr decompose into phenylethyl-amine and HBr under the applied electric field which generates mobile ions. These ions migrate toward the charge injection interfaces and induces defect states. Hence low operational stability	PEABr enhances the device efficiency but induces the ion migration.	518	10 mA cm^{-2}
$FACsRbPbI_3$(26)	ITO/ZnO/PEIE/$FACsRbPbI_3$/TFB/MoO_3/Au	15.84	60 h	Ion migration	Cs^+ reduces the I^- vacancies in the bulk and reduces the ion migration inside the grains. Rb^+ passivates the vacancy defects at the grain boundaries	405	10 mA cm^{-2}
$CsPbBr_3@Cs_4PbBr_6$(27)	$CsPbBr_3@Cs_4PbBr_6$ MCs on the surface of commercial LED chips	-	43.1	Thermal energy during device operation induces non-radiative recombination results PL quenching and device instability	Annealing of perovskite in Oleic acid and oleic reduces surface defects and passivates perovskites from the external environment. Core-shell $CsPbBr_3@CsPb_2Br_5$ MPs and	522	30 mA cm^{-2}

					$CsPbBr_3@Cs_4PbBr_6$ MCs, forms Type-I band alignment confines the charge carriers effectively.		
TFA: $CsPb(Cl_x Br_{1-x})_3$(28)	ITO/PEDOT:PSS/Perovskite/TPBi/LiF/Al	1.18	7.74 min	Low solubility of inorganic Cl sources results non uniform and insufficient coverage of perovskite film on substrate	Halide segregation is passivated by the anion exchange method and the TFA passivated grain boundaries	490	20 mA cm^{-2}
$PEA_{0.2}Cs_{0.4}MA_{0.6}Pb_{(1-y)}Mn_yBr_3$ $PEA_{0.2}Cs_{0.4}MA_{0.6}Pb_{(1-y)}Mn_yBr_{0.9}I_{2.1}$ and $PEA_{0.2}Cs_{0.4}MA_{0.6}Pb_{(1-y)}Mn_y(BrCl)_3$(29)	ITO/PEDOT:PSS/perovskite/TPBi/LiF/Al	3.2 5.1 0.58	23 min 5 h 24 min	Ion migration within the perovskite	Mn doping improves the formation energy and exciton binding energy of perovskites resulting reduces the ion migration and joule heating.	Green Red Sky blue	3 mA cm^{-2}
$CsSb_2Br_9$(30)	ITO/ZnO/PEI/$Cs_3Sb_2Br_9$/TCTA/MoO_3/Al	0.206	6 h	Toxicity of Pb. Pb-based perovskite LEDs are instable against heat and environment moisture	Sn-based LEDs are more stable in environmental effects	408	T90 @ 70 mA cm^{-2}
PMA, PEA, PBA, EA, PA, BA doped	(ITO)/PEDOT:PSS/perovskites/TPBi/LiF/Al	4.91 9.35 4.35 2.72	45 Min 114 5 20 18	Ion migration degrades the morphology and crystallinity of perovskite materials	Alkyl chain passivates the perovskite surface and improves the operational lifetime.	512 515 506 508 509	9.9 0.7 4.2 17.9 4.3

CsMAPbBr$_3$(31)		6.33 6.66	18min			512	7.9
Mg^{2+}-doped CsPbCl$_3$ NCs(32)	ITO/PEDOT:PSS/PVK/ Mg^{2+}-doped CsPbCl$_3$ NCs/TPBi/LiF/ Ag	0.1		Trap and defect states between the bandgaps due to halide vacancies	Excessive use of Cl$^-$ anions in precursor reduces Cl deficiencies and produce defect free high-quality film 2p orbitals of Mg^{2+} participate in conduction and valence band formation which changes the n-type CsPbCl$_3$ perovskite into ambipolar which supports the balanced charge injection. Mg^{2+} doping enhances the tolerance factor hence better crystal stability.	402	135 cd m^{-2}
CsEuBr$_3$(33)	ITO/LiF(insulator)/CsEuBr$_3$/LiF(insulator)/TPBi/LiF/Al	6.5	50 min	Toxicity of Pb metal	Eu^{2+} based LEDs shows blue emission with narrow linewidth, by fast intersystem crossing from 5d-4f to 4f-4f configuration	448	15.9 cd m^{-2}

ITO = Indium tin oxide, PEDOT:PSS = poly(3,4-ethylenedioxythiophene):poly-(styrene sulfonate), PTAA = poly[bis(4-phenyl)(2,4,6-trimethylphenyl) amine], TPBi = 1,3,5-Tris(1phen-y-1H-benzimidazol-2-yl) benzene, TFB = poly(9,9-dioctylfluorene-*alt*-N-(4-*s*-butylphenyl)diphenylamine), PFI = Nafion perfluorinated resin, PO-T2T = (1,3,5-triazine-2,4,6-triyl)tris(benzene-3,1-diyl)tris(diphenylphosphine oxide), ZnO-PEIE = polyethylenimine ethoxylated modified zinc oxide, MoO$_x$ = molybdenum oxide, Au = Gold, 5AVA = 5-aminovaleric acid, NMAI = 1-naphthylmethylamine iodide, FA = formamidinium, poly-TPD = poly[*N*,*N′*-bis(4-butyl-phenyl)-*N*,*N′*-bis(phenyl)-benzidine], LiF = lithium fluoride, PVK = poly(9-vinylcarbazole) poly(methyl methacrylate) (PMMA) ethylenediammonium dibromide (EDABr$_2$), TmPyPB = 1,3,5-tri(m-pyrid-3-yl-phenyl)benzene, 4-F-PMAI = 4-fluorophenylmethyl-ammonium iodide.

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