

Degradation of organic light emitting diode: Heat related issues and solutions



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ABSTRACT

Degradation of organic light emitting diodes (OLEDs) is the most serious obstacle towards their commercialization. OLED degrades due to various internal and external mechanisms. External degradation is mainly caused due to the instability of low work function cathode, pin-hole formation during fabrication which provides a path for oxygen and moisture infiltration. Operation of OLED also leads to degradation with major causes being morphological instability of organic layers, trap formation, indium or oxygen diffusion from anode, interface deterioration etc. Heat generation in the OLED also acts as a source of degradation. Most of the heat is generated instantaneously upon biasing of OLED due to resistive or Joule heating as a consequence of high resistance of organic layers and non-radiative recombination. Generated heat can be reduced by reducing the effects of the generation sources such as improving conductivity of organic layers by doping, using additional layers to improve charge injection, employing emissive layers with low recombination losses etc. However, these ways can only reduce the heat up to a certain amount. To further improve the lifetime of OLED, the generated heat can be dissipated by employing heat sinks using either thermally conducting substrate or encapsulation etc. We present a review on OLED degradation with a particular focus on heat generation, its consequences and ways of reduction.

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1. Introduction

Organic light emitting diodes (OLEDs) are currently commercialized as information displays for smartphones and large-screen TVs and promise large market opportunities for flat panel, flexible displays and solid-state lighting sources [1–5]. OLEDs have striking features as high color purity, ease of fabrication, low power consumption, color purity and low cost [6–11]. Organic semiconductors (OSCs), building blocks of OLEDs, are cheap and abundant [12–16]. Current state-of-the-art OLEDs are performing at over 100 lm/W efficiency at lab scale and 30 lm/W at industrial applications due to significant improvement from the first observation of electroluminescence in OSCs [17–24]. The main challenge ahead is the lifetime of OLEDs due to several internal and external degradation sources. Light emitting diodes based on both inorganic and organic semiconducting materials suffer from internal

energy losses and these losses convert into heat [25,26]. The generated heat can then act itself as a source of degradation and decrease the lifetime of device [27–30]. Therefore, reduction of heat and dissipation is required in order to increase lifetime of OLEDs. To date this issue has been more widely discussed in inorganic LEDs in comparison to their organic counterparts [31–34].

Degradation of OLEDs is a widely reviewed topic and several mechanisms responsible are described. A typical OLED is a multilayer device with a stack of organic layers sandwiched between two electrodes. Most efficient structure use separate organic layer for injection and transport for electrons and holes. An emission layer with a host-guest system having a phosphorescent guest and compatible host is used and to block un-recombined charge carrier, typically, electron and hole blocking layer is employed. Charge carriers are injected into the transport layers upon application of a forward bias by electrodes with the aid of injection layers. Transport layers are generally opted for high mobilities for respective carriers and then the operation of emission layer is efficient exciton formation and decay. Un-recombined carriers are then stopped by blocking layers. Blocking

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layers are sometimes also selected to stop the exciton from diffusing into the transport layer. Generated photon upon exciton decay then out-couples from the device. Degradation of OLEDs is caused by several different mechanisms both due to internal and external sources.

2. Degradation in OLEDs

2.1. External degradation

In the early days of research, the future of OLEDs was doubtful due to instability of OSCs under ambient conditions. Other than the instability of OSCs, the other issue has been the requirement of low work function electrodes as cathode which requires control on the deposition environment as an exposure of oxygen and moisture could cause degradation [35]. Exposure of device to ambient conditions would cause the cathode degradation and generation of non-emissive areas—often termed as dark-spots in the OLED community.

Fig. 1a presents the optical photographs of operating OLEDs showing such dark-spots. A number of studies [35–40] were thus performed for finding the origins of dark-spot growth during the initial days of OLED research. Dark-spot are originated from pre-existing particle defects on the substrate and subsequent deposition of cathode leading to pin-hole formation in the metal thin film and provide an opportunity to moisture and oxygen to diffuse through it causing localized delamination of cathode and formation of dark-spot. Study of degrading OLED by Schaer et al. [29] showed that these dark-spots form like a bubble (Fig. 1b). An electrical bias applied to device accelerates the growth in the size of dark-spots and stopping the current stops the process. It was also reported that gases evolve by the electrochemical processes at cathode/organic interface which cause the formation of these bubbles [41–43]. Indeed, the formation of dark-spot is related to delamination of cathode and not related to underlying organic layer. Therefore, redeposition of the cathode after peeling of the degraded one restores the performance. It is observed that formation of new dark-spots during operation is less probable than the widening of the dark-spot already present. As an example,

dark-spots ‘1’ and ‘2’ broadened in size upon degradation of OLED. It is also observed that the absolute number of dark-spots does not change during the operation [29].

When the device is fabricated, presence of oxygen and moisture inside the vacuum system degrades the organic layers [44–46]. Additionally, presence of small particles inside the device may also cause the degradation of device, if they puncture through the organic layers and come in contact with aluminum [47]. Further, the widely used electron injection layer (LiF) has a hydrophilic nature, as a result, absorbs moisture and degrade the device [48]. Appearance of dark-spots is dependent on the device growth conditions; however, broadening of dark-spots can be controlled by a proper encapsulation. Fig. 1c presents the optical images of an un-encapsulated and encapsulated OLED under operation at different time. A proper encapsulation slows down the appeared dark-spots during the operation and thus is effective in slowing down the degradation. Proper encapsulation of OLED prevents the corrosion of low work function cathode which was the initial concern of researchers. However, OLEDs have several other sources of degradation and such degradation is often referred as operational degradation.

2.2. Operational degradation

Operational degradation refers to the losses of luminance efficiency with time due to application of field. Here, we aim to briefly describe the possible operational degradation mechanisms and detailed knowledge of operational degradation can be found in previously published review [35]. The main causes of operational degradation are (i) thermal stability of organic layers, (ii) traps and luminescence quencher formation, (iii), anode instability and (iv) interface degradation [35]. Here, we will discuss briefly about these sources and their effects on the device performance. Thermal stability is determined by the glass transition temperature (T_g) of organic layers. *N,N'*-Bis(3-methylphenyl)-*N,N'*-diphenylbenzidine (TPD) was used as the hole transport layer (HTL) in early OLEDs, and it has a low T_g ($\approx 60^\circ\text{C}$) [49]. Similarly, 4, 7-diphenyl-1, 10-phenanthroline (BPhen); often used as electron transport material (ETM), has a low T_g value of 65°C . Device temperature often increases over T_g value of such compounds during operation, which leads to its crystallization and degrades the device [50]. Therefore, to avoid the crystallization of organic layers, molecules with high T_g values are required. This transpired the synthesis of novel transport materials and at later stages hole transport materials (HTMs) and ETMs with high T_g found successful. Such degradation only occurs for a device under operation.

Second, bulk trap states are formed during the operation of device in the form of non-luminescent centers inside the emissive layer. These trap states decrease the conductivity of organic layers which also lead to an increase in operating voltage [51]. Indeed, the formation of traps upon operation of OLEDs is often observed in case of electrons than the holes. Study by different groups [51,52] on poly(phenylene vinylene) (PPV) inferred that the trap formation in OLED is more often caused by electron. Third, prominent mechanism of operation degradation is interface deterioration. Polymer based LEDs (PLEDs) have only two interface, one for electron injection and other for hole injection. Any deterioration of these interfaces will lead to interface degradation. Contrary to PLEDs, small molecule OLEDs (SMOLEDs) have many interfaces depending upon the device structure, and the interface degradation is a severe problem in these devices. The first report on SMOLEDs with a simple HTL/ETL interface for light emission was found to have very short life time of nearly 50 h due to existence of barrier for holes at the interface of Alq_3 /diamine used in the device. Presence of such barrier at the organic interfaces originates traps and degrades the device under operation [53].

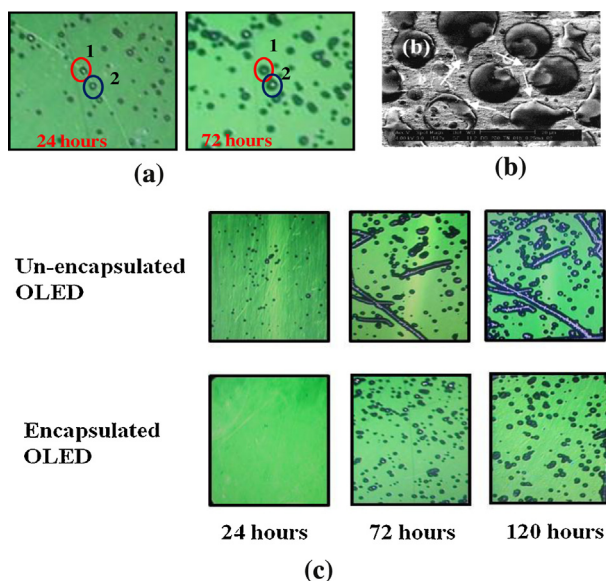


Fig. 1. (a) Optical image of operating OLED (under a current density of 10 mA/cm^2) showing the dark-spots and their broadening upon degradation. (b) SEM image of the bubbles formed in cathode at dark-spot areas. [printed with permission, Adv. Funct. Mater. 2001, 11, 116] (c) Optical images of un-encapsulated and encapsulated OLED at different times.

The fourth mechanism is anode degradation. Requirement of one transparent electrode in OLEDs has led to over dependency on indium tin oxide (ITO). It is discovered by several researchers, indium is released from ITO into organic layers during the device operation. Later it is observed that diffusion of oxygen from ITO is more problematic than diffusion of indium with diffusion of former causing a significant change in work function of ITO. Indium diffusion into organic layers forms non-radiative centers while oxygen diffusion significantly increases the operating voltage [54–56]. Anode degradation is somewhat overcome by use of hole injection layer (HIL) (for reducing indium diffusion) and UV-ozone or oxygen plasma treatment (for oxygen diffusion). Additional to these degradation sources, operating OLED generate significant amount of heat due to high resistance of organic layers and all the above mentioned sources of degradation and then becomes a source of degradation.

3. Heat generation and consequences

Once the OLED is biased, heat is generated due to potential drop at the interfaces and due to high resistances of organic layers. Generally, resistances caused by interfaces and bulk are the major sources of heat generation. Additional to resistive losses, non-radiative recombination of exciton also leads to the heat generation. However, proper use of host-guest system based emissive layers has reduced such losses to its minimal. Resistive and recombination losses cause the heat generation instantly upon application of bias. While the degradation of OLED then add to heat upon operation. Additional to all above mentioned sources of degradation, heat is also a source of degradation. Generated heat increases the temperature of OLED and thus assists to non-radiative losses inside the device. Excitons are generally dissociated due to higher operating temperature of OLEDs [57,58]. The geminate pair dissociation rate is dependent on temperature as [59]

$$k_d(T) = a \times T^{3/2} \times \exp\left(-\frac{\Delta E}{kT}\right) \quad (1)$$

where a is temperature independent term [58], T the temperature, k Boltzmann's constant and ΔE is binding energy of geminate pair. From this equation, it is evident that the geminate pair dissociation rate is higher for higher temperature. Therefore, generated heat lead to exciton dissociation also higher device temperature will assist to operational degradation. Before reducing the heat losses, probing is the first complicated target as it being a form of energy direct measurement of heat is not possible.

3.1. Heat based measurements in OLEDs

Heat generation inside the OLED elevates the temperature of device, which can be efficiently probed and temperature measurements can be efficiently used to calculate the amount of heat generated inside the device by solving the heat conduction equation. Therefore, the probing of heat generated requires efficient temperature measurement of OLEDs. Several techniques have been used for temperature measurements in microelectronic devices.

3.2. Temperature measurements

Temperature measurements in microelectronic devices have remained a topic of scientific and industrial importance. Several techniques have been used to efficiently probe the temperature of these devices, most effective being use of thermocouples, Raman spectroscopy and IR-thermography, in context of OLEDs.

(a) Thermocouple: use of radiation thermometer or a thermocouple is a contact method to measure the temperature of a surface. In this technique, the device is placed on a thermocouple and it directly measures the temperature of the device surface. The constraints for this technique are that the device should be fabricated on thermally conducting substrates, it should efficiently transfer the heat to thermocouple, and the substrate should be thin enough to avoid lateral cooling of the substrate. These constraints make it difficult to use this technique for OLEDs which are fabricated on glass or plastic substrates with poor thermal conductivity. Also, this is a contact method which is an added disadvantage [60].

(b) Raman Spectroscopy: Raman spectroscopy has been used to efficiently probe the temperature of any material by using the intensity ratio of anti-stokes and stokes Raman lines. It is a non-contact method. The intensity ratio between anti-stokes and stokes Raman scattering of vibration depends on the temperature of sample at equilibrium by the equation [61,62]

$$\frac{I_i^{AS}}{I_i^{S}} = K_\alpha \left[\frac{(v_0 + v_i)}{(v_0 - v_i)} \right]^4 \exp\left(-\frac{hv_i}{kT}\right) \quad (2)$$

where v_0 is the frequency of incident light, v_i is the Raman shift for the i^{th} vibration, k is the Boltzmann constant and K_α is the ratio of the squares of Raman tensor of antistokes scattering to those of stokes scattering and generally it is unity. There are few reports on the temperature measurements of OLEDs by this technique [61,62]. However, the maximum error for this method is $\pm 6^\circ\text{C}$, which is quite significant. Also in resonant condition, the value of K_α is not unity resulting in further errors.

(c) IR Thermography: Infrared (IR) thermography is a widely used technique due to its high accuracy and non-contact measurement nature. It works on a principle that every heated object emits radiation in infrared region. Modern IR cameras or imagers can easily resolve temperature differences of 0.1°C or less [63]. Wavelength corresponding to the maximum intensity depends on temperature. The Wien displacement law relates the temperature, T (in Kelvin) with the peak radiation wavelength, λ_{peak} , emitted by a body:

$$\lambda_{\text{peak}} = \frac{2897.8}{T} \quad (3)$$

These features make IR-thermography a suitable technique for the temperature measurements. There are few reports on temperature measurements on OLEDs by using IR-thermography. Xiangfei Qi and S.R. Forrest [64] have utilized IR imaging to probe the temperature of a phosphorescent OLED (PHOLED) with iridium (III) tris(2-phenyl-pyridinato- $N, C^{2'}$) ($\text{Ir}(\text{ppy})_3$) as emissive layer. IR-thermography is effectively used to observe in different cases effectively to measure the device temperature.

3.3. Heat analysis

A complete analysis of heat requires the calculation of heat losses based on the temperature measurements inside the device. When a thin film with uniform thickness is subjected to an input flux, Q , on one of its surface, the one dimensional heat conduction equation or Fourier's law states that the heat flux is proportional to the negative of local temperature gradient [65],

$$-\frac{\partial T(x, t)}{\partial x} = \frac{1}{K} Q(x, t) \quad (4)$$

Continuity equation states

$$\frac{\partial T(x, t)}{\partial t} = \frac{1}{C} \frac{\partial Q(x, t)}{\partial x} \quad (5)$$

where $T(x,t)$ is the temperature of the thin film at point x and time, t , $Q(x,t)$ is the heat flux, K is the thermal conductivity of the thin film and C is the volumetric heat capacity, which is a product of specific heat at constant volume, c_v , and density, ρ . Eqs. (4) and (5) can be combined to yield

$$\frac{\partial T(x,t)}{\partial t} - \alpha \frac{\partial^2 T(x,t)}{\partial x^2} = 0 \quad (6)$$

where $\alpha = K/(c_v \times \rho)$ is the heat diffusivity of the material. Laplace transform of Eqs. (5) and (6) yields

$$-\partial \frac{\hat{T}(x,s)}{\partial x} = \frac{1}{K} \hat{Q}(x,s) \quad (7)$$

$$-\partial \frac{\hat{Q}(x,s)}{\partial x} = Cs \hat{T}(x,s) \quad (8)$$

where, $\hat{T}(x,s)$ and $\hat{Q}(x,s)$ are the Laplace transforms of temperature and heat flux, respectively and s is the Laplace variable. From these equations, the source temperature can be correlated with the ambient temperature by the following equation [64,66]

$$\begin{bmatrix} \hat{T}_{SRC} \\ \hat{Q}_{SRC} \end{bmatrix} = [T(\theta_i)] \begin{bmatrix} \hat{T}_0 \\ \hat{Q}_0 \end{bmatrix} \quad (9)$$

where, $\hat{T}_{SRC}$ and $\hat{T}_0$ are Laplace transforms of the source and ambient temperatures, respectively, and $\hat{Q}_{SRC}$ and $\hat{Q}_0$ are Laplace transforms of heat flux of source, heat fluxes dissipated through the layer. The conduction matrix is given as

$$[T(\theta_i)] = \begin{bmatrix} \cosh(\theta_i) & Z_i \sinh(\theta_i) \\ \frac{\sinh(\theta_i)}{Z_i} & \cosh(\theta_i) \end{bmatrix} \quad (10)$$

where, $\theta_i = L_i \sqrt{C_i s / K_i}$ is the operational propagation coefficient characterized by K_i (thermal conductivity), C_i (volumetric heat capacity), L_i (thickness of the layer) and $Z_i = \sqrt{1/(K_i C_i s)}$ is the characteristic thermal impedance of the layer.

This method has particular advantage in case of multilayer structure, as shown in Fig. 2, because conduction matrix can be evaluated easily for each layer and the value of outgoing flux can be easily correlated with the incoming flux by using the temperature measurements. Qi and Forrest have used these heat calculations to evaluate the estimated heat losses inside the OLEDs using temperature measurements [64]. They observed that about 94–95% of the supplied power is lost as heat in a simple multi layer OLED. Further, the method is extensively used by Tyagi et al. [67] in conjunction with the temperature measurements on a degrading OLED. They have evaluated the heat generated during the degradation of OLED. They observed the effect of glass

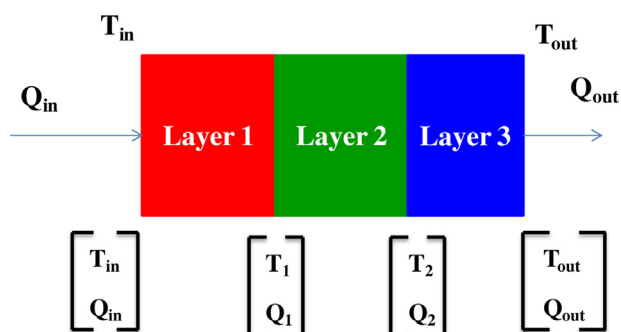


Fig. 2. Typical OLED geometry used for heat calculations, showing temperature and heat at different positions.

encapsulation on generated heat inside the device with lifetime. As expected, the encapsulation reduces the rate of heat generation with operation as a consequence of slow degradation.

Yoshioka et al. [68] have recently reported new strategy for the improved prediction of OLED lifetimes by employing an analysis of luminescence decay curves and decoupling of the self or Joule heating effects from coulombic effects. They have used Raman spectroscopy and temperature dependent current-voltage characteristics in order to evaluate the organic layer temperature. This temperature measurement combined with accelerated life test enabled the nearly accurate prediction of lifetime by only testing a device for 1/40th time of its lifetime (40,000 h). They have tested the developed strategy in two cases—one with negligible self-heating and other with significant. Lifetime testing for less than 1000 h was required for both cases for approximating the lifetime of nearly 40,000 h.

4. Resistive losses and Joule heating

OLEDs consist of multiple organic layers and their interfaces with each of them causing electrical resistances. Fig. 3 presents a simple resistive equivalent of OLED showing the resistance originated due to metal/organic, organic/organic interface and internal resistance of the organic layers. All these resistances contribute towards heating (Joule heating) which is proportional to the resistance.

Organic layers are generally selected in such a way to minimize the interface barriers at each organic/organic interface. Therefore, the main source of Joule heating inside an OLED is the interface resistances at metal/organic interface and the internal resistances of each organic layer. In a further simple representation, OLED can be considered as the series combination of three resistances only as shown in Fig. 4.

Here, two resistances R_{s1} and R_{s2} represent the interface resistances at anode/HTL interface and cathode/ETL interface, respectively; while R_b represents the combined intrinsic resistance of organic layers. When the energy barrier at metal/organic interface is zero, the value of interface resistance and power loss is minimum and increases drastically by increasing the value of interface barrier [69–74]. Kwak et al. [75] used IR-thermography in order to observe the effect of electron injection layer (LiF) on the temperature rise in the device having configuration ITO/ α -NPD/Alq₃/Al. Introduction of LiF injection layer at Al/Alq₃ interface reduced the temperature rise from 110°C to 29°C as a direct consequence of reduced interface barrier and thereby interface

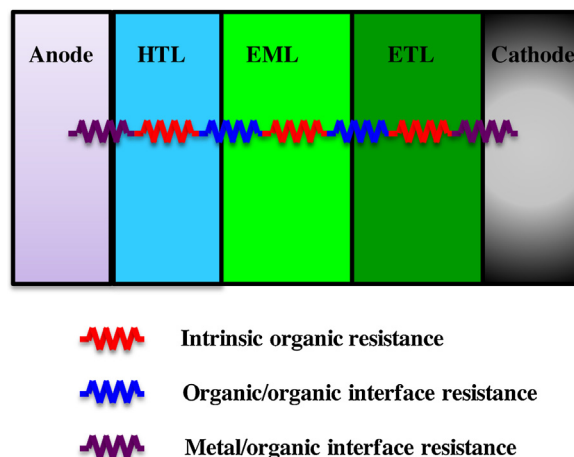


Fig. 3. Schematic representation of different electrical resistances originating inside the OLED structure.

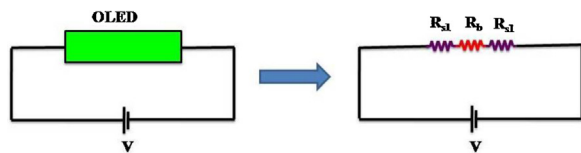


Fig. 4. Simplified resistive representation of an OLED showing the two interface resistances and one combined intrinsic resistance.

resistance. In their study, they fabricated device with four structures (i) Reference: ITO/ α -NPD/Alq₃/Al, (ii) Device A: ITO/ α -NPD/Alq₃/LiF/Al, (iii) Device B: ITO/ α -NPD/Alq₃/TPBi/LiF/Al and (iv) Device C: ITO/ α -NPD/Alq₃/Bphen/LiF/Al. Device temperature significantly reduced upon introduction of an electron transport layer (TPBi or Bphen) as can be seen from Fig. 5(a and b). Further, introduction of an F₄-TCNQ layer at ITO/ α -NPD interface [76] in a device structure ITO/ α -NPD/5% Ir(ppy)₃ doped CBP/BCP/Alq₃/LiF/Al reduces the temperature rise by half as shown in Fig. 5c. Introduction of interface layer thus reduces the Joule heating and significantly reduce the temperature rise of the device.

Even after the introduction of interface layer, there is significant amount of temperature rise in the device as can be seen from Fig. 5c. This heat is contributed by the bulk resistances of the device. As the OSCs are devoid of free carrier concentration, intrinsic resistances of these materials are very high. Hopping between localized energy states govern the charge transport in the OSCs with the required energy provided by the thermal energy present in the system [77]. In general, this hopping process does not have any preferred direction and the overall displacement of charge carrier is zero in equilibrium. However, the application of electric field in a particular direction breaks the equilibrium of the solid and charge carriers start to jump with a preferred direction and displacement in that direction. The hopping process depends on the difference between the energy states and overlap between the initial state and final state. A faster movement of charge carriers can result from smaller differences between the energy states and higher overlap. The hopping rate from one state to another state is given by Miller and Abraham equation [77]:

$$v_{ij} = v_0 \exp\left(\frac{\gamma \Delta R_{ij}}{a}\right) \exp\left(-\frac{\varepsilon_j - \varepsilon_i}{kT}\right); \text{ if } \varepsilon_j < \varepsilon_i \quad (11)$$

$$v_{ij} = v_0 \exp\left(\frac{\gamma \Delta R_{ij}}{a}\right); \text{ if } \varepsilon_j > \varepsilon_i \quad (12)$$

where v_0 is the attempt to jump frequency, γ is the inverse localization length, R_{ij} is the distance between two localized states, ε_i and ε_j are the energies of site i and j and a is the intermolecular spacing. The first term on the right side of Eq. (11) give the tunneling probability between two states and the second the energy dependence of carrier hop. Jumps upward in energy are impeded by the energy barrier between the initial and final state, while downward jumps are independent of energy barrier [77]. Value of v_0 is 10^{12} and the extremely high value of inverse localization length (γ) originates the high low values of hopping rate. Additionally, the hopping rate is impeded by the discrete energy states present in case of OSC with the difference between the energy states about or more than the room temperature thermal energy ($k_b T \sim 26$ meV). Thermal energy induces the hopping of charge carrier and the energy barrier and high inverse localization length in OSC cause the high resistance values. Table 1 shows the typical values of the resistances for the commonly used transport layers and relative values of power loss across them. It can be seen from this table that these transport layers also cause significant power loss inside OLEDs. The resistance of emissive

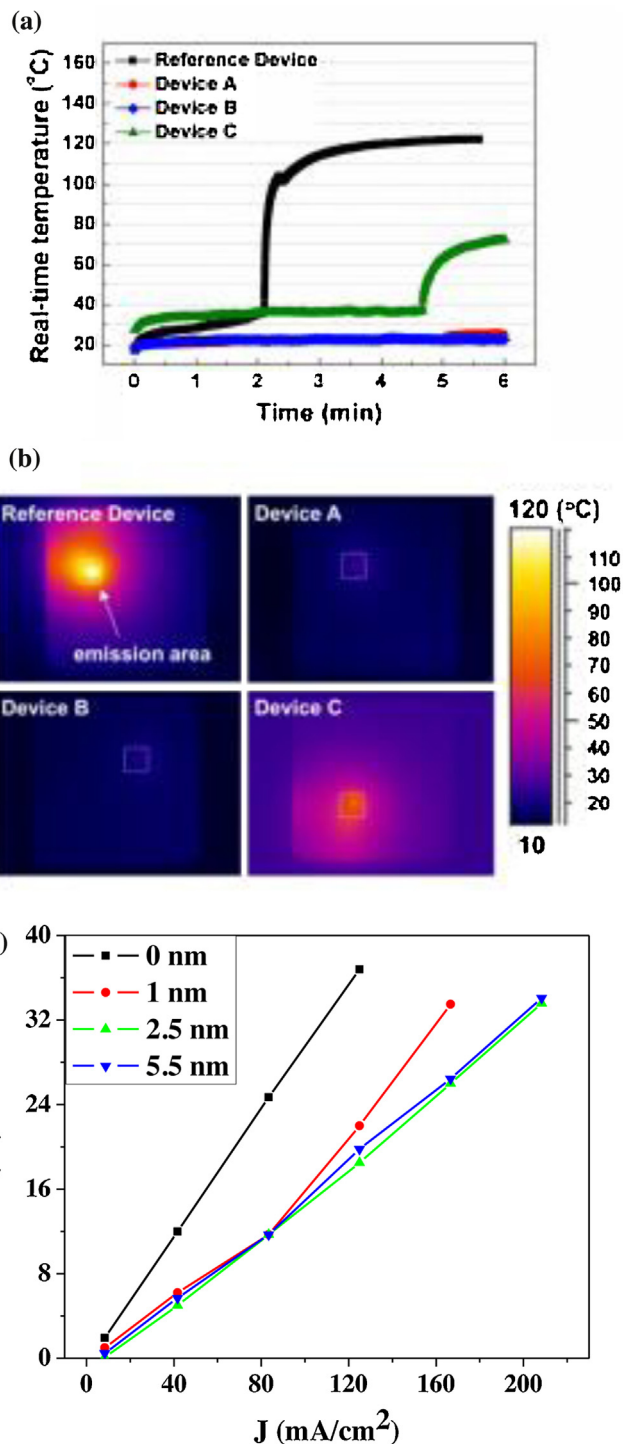


Fig. 5. (a) Real-time temperature of OLEDs and (b) the IR images at 6 min after the operation (printed with permission Opt. Exp. 21, 29558, 2013). (c) Temperature rise as a function of current density for OLEDs with F₄-TCNQ interface layer having different thickness (0, 1, 2.5 and 5.5 nm).

layer can only be modified by changing the material because doping inside the emissive layer induces exciton dissociation. Therefore, it will increase the recombination losses. Overall, the Joule heating can be quantitatively given as $I^2(R_{s1} + R_{s2} + R_i)W$ in an OLED with a current I flowing through it.

Use of low resistance transport layer directly reduces the generated heat as observed by Kwak et al. [75]. Replacing Bphen transport layer by TPBi in device (ITO/ α -NPD/Alq₃/Bphen/LiF/Al

Table 1Resistance for the commonly used transport layers and the approximate power loss caused by these resistances at 100 mA/cm².

Material	Resistance	Approximate power loss (at 100 mA/cm ²)	Reference
Alq ₃	10 KΩ	100 W/cm ²	[78,79]
<i>N,N'</i> -Di(1-naphthyl)- <i>N,N'</i> -diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB)	0.36 KΩ	3.6 W/cm ²	[79]
Poly(<i>p</i> -phenylenevinylene) (PPV)	0.78 KΩ	7.8 W/cm ²	[80]
Poly(2-methoxy-5-(2'-ethyl-hexoxy)-1,4-phenylene-vinylene) (MEH-PPV)	1 KΩ	10 W/cm ²	[81]
<i>N,N'</i> -Dicarbazolyl-4,4'-biphenyl (CBP)	2.22 KΩ	22.2 W/cm ²	[82]
α-NPD	0.5 KΩ	5 W/cm ²	[83]
<i>N,N'</i> -Bis(3-methylphenyl)- <i>N,N'</i> -diphenylbenzidine (TPD)	51 KΩ	510 W/cm ²	[84]

led to a temperature reduction from 74 °C to 22 °C (Fig. 5a). However, in this case the low T_g value (55 °C) of Bphen also plays a role. High resistance of OSCs not only led to reduced performance of OLEDs, but also generates heat and thus expected to become a source of degradation.

Joule or resistive heating can be reduced by decreasing the resistances of the layers and interfaces used in OLEDs. The major resistances are caused by the metal/organic interface and the bulk resistances of these layers. Therefore, the interface resistances and bulk resistances can be minimized to reduce the Joule heating. Interface resistances can be minimized by introducing the interface layers at anode/HTL interface and cathode/ETL interface or by developing new organic materials with matched HOMO/LUMO value to the metal Fermi levels [84–93]. Introduction of interface layers have by far remained the better choice to reduce the interface resistance. Table 2 shows some of the widely used interface layers introduced at anode/HTL interface.

It can be seen from the table that several injection layers have been used for the reduction of barrier at this interface and the OLED efficiency has been significantly improved. Metal oxides such as MoO_x, WO₃ etc. [86–88] have been found very effective to improve this interface; however, their deposition is very difficult and depend on the deposition conditions such as pressure inside the chamber. Organic acceptor layers such as 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F₄-TCNQ), 3,6-difluoro-2,5,7,7,8,8-hexacyano-quinodimethane (F₂-HCNQ) have been found effective and also can be easily deposited [76,90–93]. Out of these materials, F₄-TCNQ is the widely used one. p-doped HTLs were also found to be effective for reducing the interface barrier [84,85], however, the thickness of these layers are in the range of 10–30 nm and therefore, introduction of these layers increase the bulk resistance.

Heat related studies on these interface modified OLEDs have not by far emphasized. Temperature measurement of F₄-TCNQ modified green OLED (ITO/F₄-TCNQ/α-NPD/5% Ir(ppy)₃ doped CBP/BCP/Alq₃/LiF/Al) revealed that the thickness of F₄-TCNQ plays an important role in reduction of device temperature. Simple introduction of 1 nm F₄-TCNQ reduces the temperature by half and using calculation based on transverse matrix approach showed that the generated heat reduced by more than two times using the interface layer. However, in this case, diffusion of F₄-TCNQ is found to affect the life time and the generated heat (with operation) as

can be seen from the Fig. 6. And the superior performance is only observed in case of F₄-TCNQ thickness higher than 2.5 nm. In this OLED, the generated heat is reduced more than two times during the operation of device checked up to 168 h [76].

Table 3 shows the effectively used injection layers used at cathode/ETL interface and the typical improvement in the performances of OLEDs by their introduction. Alkali metal fluorides, carbonates and chelates have been used as interface layers for this interface and were found to be effective in improving the performances of OLEDs by reducing the barrier at cathode/ETL interface [94–99]. Bulk resistances of the organic materials can be decreased by doping of transport layers with suitable dopants [100]. Organic materials with high lying LUMO values or transition metal oxides have been used as dopants for decreasing the resistances for holes in HTLs. Hole conductivity of these materials has been found to improve by two or three orders of magnitudes due to the doping of metal oxides and electron acceptors such as F₄-TCNQ [100]. F₄-TCNQ has been found to be the most effective p-type dopant due to low evaporation temperature and its miscibility with organic materials. At the same time metal oxides have very high evaporation temperatures as compared to organic materials. Increasing the electron conductivity of ETLs is rather difficult because the n-type doping of ETLs require organic materials with low lying HOMO values, which are highly reactive and unstable

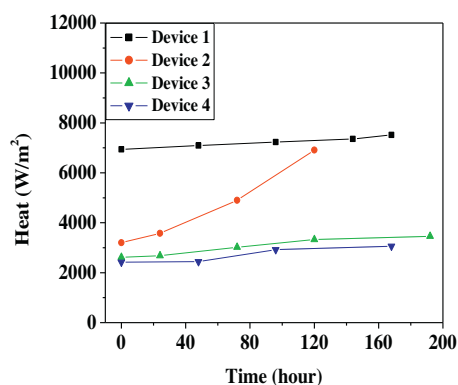


Fig. 6. Heat calculated for F₄-TCNQ modified devices having 0 (device 1), 1 (device 2), 2.5 (device 3) and 5.5 nm (device 4) thickness as a function of time.

Table 2

Typical reported performance improvement in OLEDs by insertion of effective injection layer at the anode/HTL interface.

Injection layer	Performance improvement	Reference
p-doped HTLs (F ₄ -TCNQ as dopant)	Reduction in operating voltage to almost half and 30–70% increase in efficiency	[84,85]
MoO ₃	Formation of efficient ohmic contact, reduction in turn-on voltage to 2.4 V and increase in efficiency by more than 50%	[86,87]
WO ₃	No barrier at interface and increase in efficiency by two times	[88]
Polyaniline-poly (<i>p</i> -styrenesulfonic acid)	Operating voltage reduction by 15–30% and luminance improvement by about 30–50%	[89]
PANI-PSS		
F ₄ -TCNQ	Turn on voltage reduction to 2.5 V and efficiency improvement more than 2 times	[76,90,91]
Organic acceptors such as F ₂ -HCNQ etc.	Turn on voltage reduction to 2.5 V and efficiency improvement by more than 50%	[92,93]

Table 3

Typical reported performance improvement in OLEDs by insertion of effective injection layer at the cathode/ETL interface.

Injection layer	Performance improvement	Reference
Alkali metal fluorides such as LiF, CsF etc	Increase in efficiency by four times	[94,95]
Alkali metal carbonates such as Cs_2CO_3 , Rb_2CO_3 etc.	Formation of efficient ohmic contact and increase in efficiency by more than 3 times	[96,97]
Alkali metal chelates such as Liq, Naq etc.	Efficiency improvement by more than three times	[98,99]

[101,102]. Therefore, alkali metals, alkali metal halides and carbonates have been used to reduce the resistance for electron flow in ETLs and conductivity increase of around one to two orders of magnitude has been obtained [103–105]. Fig. 7 presents a pictorial summary of different major sources of heat generation.

5. Heat dissipation

Additional to reduction of heat losses, dissipation of generated heat is an effective approach to increase the lifetime of OLED. Efficient heat dissipation and device temperature reduction have already been achieved in case of inorganic devices by proper thermal management of these devices [106–112]. Inorganic devices are fabricated on thermally conducting substrates such as Si, Ge and other semiconductors; therefore, the substrate itself works as a heat sink [106,107]. Additionally, several different types of heat sinks are already commercialized for these devices [108–112]. Since these devices are fabricated on thermally conducting substrates, they can be externally cooled by cold water flow, liquid nitrogen flow etc [111,112]. This technique is more common for solar panels.

Achieving similar thermal management in organic devices is rather more difficult because these devices are fabricated on thermally non-conducting glass substrates. Thermally conducting substrates are generally opaque to visible light; therefore, bottom emitting OLEDs cannot be fabricated on these substrates and only TOLEDs can be realized [113]. The other possibility is to incorporate heat sink in the encapsulation of these devices. Glass encapsulation is common to modern day OLEDs. However, it has very poor thermal conductivity ($\sim 1.05 \text{ W/mK}$) and cannot dissipate the generated heat inside the device [113]. Use of thermally conducting materials for encapsulation is restricted due to their higher electrical conductivities, which could lead to device failure. There are some reports mentioning techniques for the thermal management of OLEDs. Table 4 lists these techniques and their results on the performance of OLEDs.

These techniques include liquid getter filled thin film encapsulation, use of thick cathode working as heat sink, immersing the whole device inside a thermally conducting liquid and fabricating device on thermally conducting substrates. Chung et al. [113] fabricated top-emitting OLEDs over conducting substrates (silicon and stainless steel). Fig. 8 presents the IR images showing the

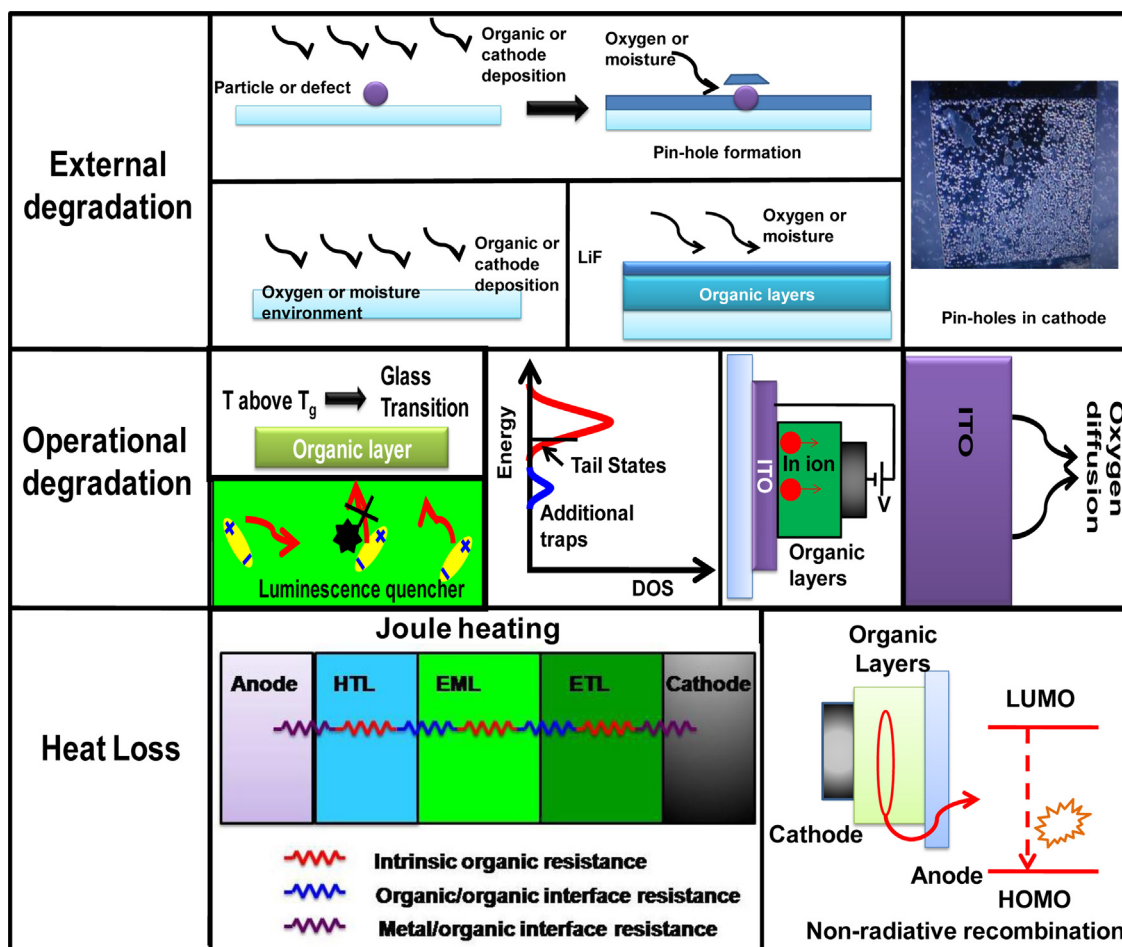
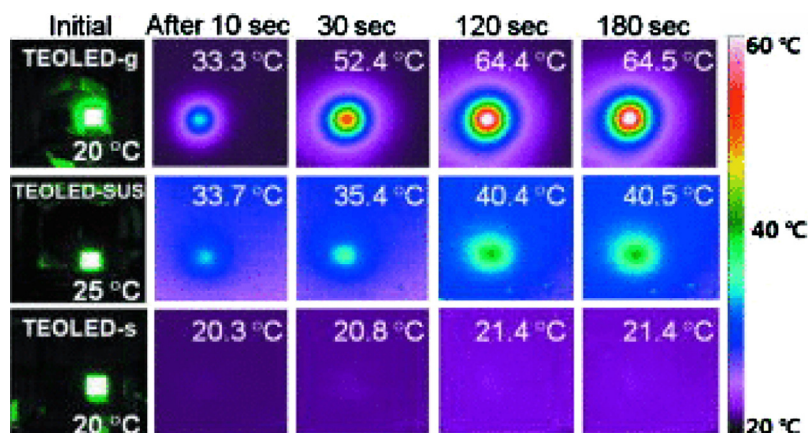
**Fig. 7.** Pictorial representation of different heat generation sources.

Table 4

Effective techniques used to reduce the temperature of operating OLEDs and their effect on device temperature, efficiency and lifetime.

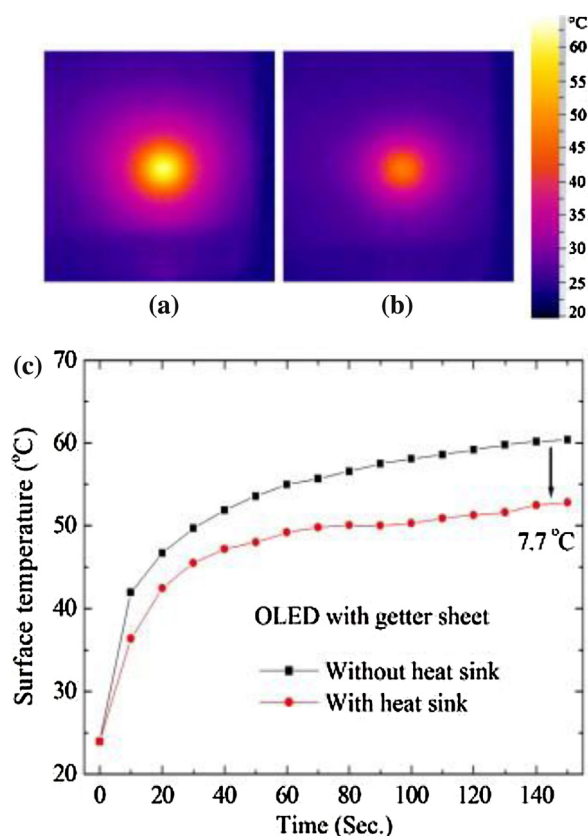
Technique	Temperature reduction	Efficiency improvement	Lifetime improvement	Reference
Liquid getter filled thin film encapsulation	7.7 °C at 8 V	Not reported	>800 h	[114,115]
Thick cathode working as heat sink	8.5 °C and 11.5 °C at 6 and 9 V	Not reported	>3 times	[116]
Hydrofluoroethers as heat transfer fluid	Not measured	70% enhancement	16% improvement	[117]
TOLEDs on thermally conducting substrates	24.1 °C and 43.1 °C for SUS and silicon substrate	0.21 Cd/A higher at 1000 Cd/m ²	>3 times for SUS and >6 times for silicon	[113]

**Fig. 8.** IR images for top emitting OLEDs fabricated over glass (TEOLED-g), TEOLED-SUS and TEOLED-s after different interval of operation showing the device temperature. (printed with permission Appl. Phys. Lett. 94, 253302, 2009).

device temperature evolution observed in these OLEDs with lowest temperature for OLED on silicon. Fabricating OLED on conducting substrate led to an increase of lifetime by more than 6 times in silicon while more than three times for OLED on stainless steel.

A thick aluminum cathode as heat sink (600 nm) was used by Choi et al. [116], which has reduced the device temperature from 44 °C (observed for 150 nm thickness) to 35.9 °C at 6 V. The device temperature has further been reduced to 35.5 °C by using Al (150 nm)/Cu (150 nm) as cathode utilizing superior thermal conductivity of Cu in place of Al. Lifetime has thus been improved by more than 3 times using both only Al and Al/Cu with the highest enhancement for Al/Cu. Ham et al. utilized liquid getter (AquaDry ODF-U) filled between the device and the encapsulation in order to reduce the device temperature. They have observed a reduction in device temperature by 7.7 °C at 8 V (Fig. 9). Further, they have reduced the device temperature by using attaching a polynetwork flexible heat sink (PN-FHB050N) over glass encapsulation. They observed enhancement in lifetime more than 800 h. Zakhidov et al. [117] have used high thermal conductivity hydrofluoroether liquid for heat dissipation and immersed OLED in this liquid which provided convective cooling of the device and device temperature is reduced by 8% (Fig. 10).

Using a thermally conducting encapsulation was found to be a more effective and cheap approach [118]. Such encapsulation include electrically insulating thin mica sheet (for electrical insulation) attached to metal sheet (Al, Cu and Mo) by thermally conducting epoxy. Using sufficiently thin sheet of mica does not affect the heat dissipation capabilities of encapsulation and effectively reduce the temperature. Temperature rise of OLED is reduced from 12.8 °C (for glass) to 7.1, 5.7 and 6.3 °C (Fig. 11) for thermally conducting encapsulation using Al, Cu and Mo metal sheet, respectively for a white organic light emitting diode (ITO/0.4 wt% F₄-TCNQ doped α -NPD/5 wt% BCzVBi doped CBP/5 wt% Ir(ppy)₃ doped CBP/0.75 wt% Ir(btp)₂acac doped CBP/BAIq/LiF/Al). As a consequence of device temperature reduction lifetime is

**Fig. 9.** IR image for the reference OLED (a) and OLED with liquid getter filled between the device and encapsulation (b). (c) Surface temperature as a function of time showing the effective ness of heat sink together with filling of liquid getter. (printed with permission, Organic Electronics 12, 2174, 2011).

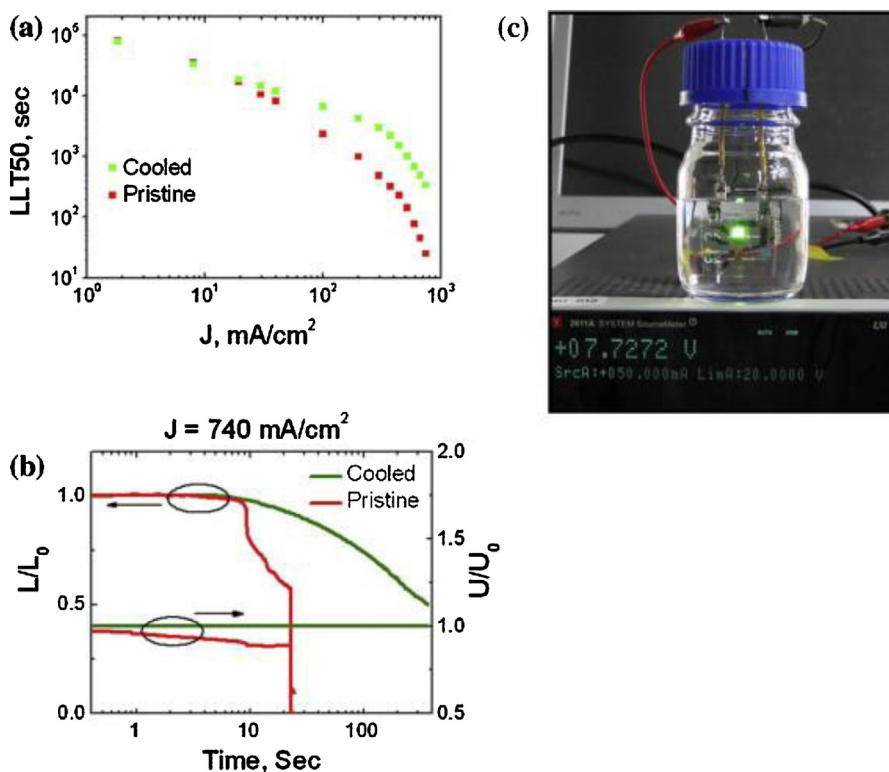


Fig. 10. Time required for reaching the 50% of initial luminance value (LLT50) dependence on applied current density. (b) Relative luminance and voltage drop (normalized with initial values) with time at constant DC current density of $740 \text{ mA}/\text{cm}^2$ for HFE7100 liquid cooled (green) and reference (red) OLEDs. (c) OLED in HFE7100 liquid. (printed with permission Organic Electronics 13, 356, 2012). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.).

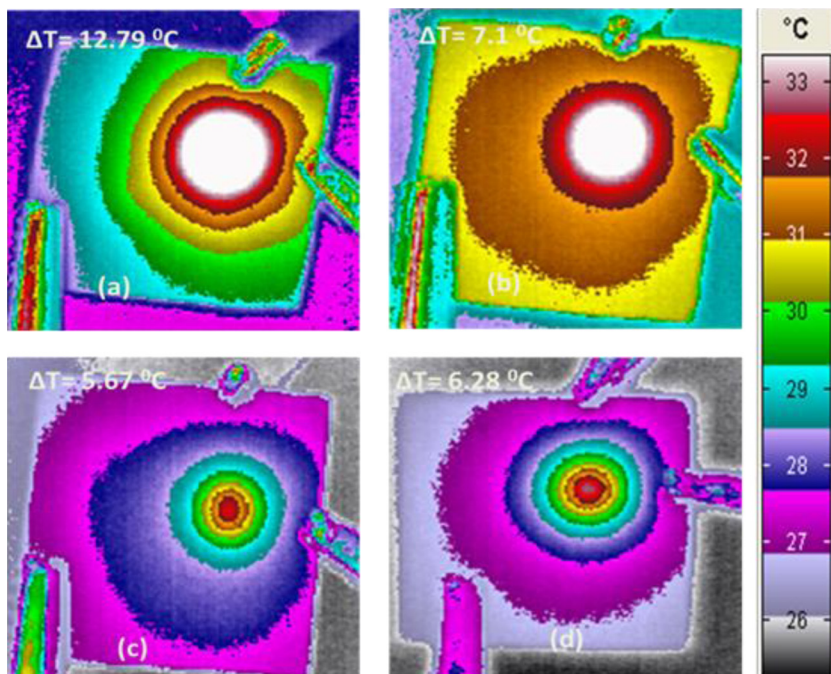


Fig. 11. IR images for the white OLED with glass (a), thermally conducting encapsulation having Al (b), Cu (c) and Mo (d) sheet at 17 V. (printed with permission Appl. Phys. Lett. 106, 13301, 2015).

increased by more than 60% by using thermally conducting encapsulation with the highest observed in case of Cu. Additionally, these encapsulation increase the efficiency of OLED due to reduced exciton dissociation.

6. Conclusion

Heat generation inside OLED reduces the lifetime significantly and relatively less discussed topic in context. In this review, we have presented the possible causes of heat generation including

the internal resistances and operation degradation. Analysis of generated heat required efficient temperature measurement for which IR-thermography has been efficiently used in the literature. Solving heat conduction equation then gives an efficient measure of generated heat. Consequences of generated heat of lifetime are then discussed. Heat dissipation and thermal management in OLEDs is then an efficient way to improve their lifetime.

Acknowledgements

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