Fully Organic PIN OLEDs with High Power Efficiency and Long Lifetime for the Use in Display and Lighting Applications

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Abstract

Power efficiency, lifetime and stable manufacturing processes are the crucial parameters for the success of organic light emitting diodes (OLEDs) in display and lighting applications. Highest power efficiencies of PIN-OLEDs for all principal colours and for bottom and top emission OLED structures have been demonstrated. The PIN structure, which means the incorporation of intentionally doped charge carrier transport layer in a suitable OLED layer setup, lowers the operating voltage to achieve highest power efficiencies. Up to now the n-doping of the electron transport layer has been done by alkali metal co-deposition. This has main draw-backs in terms of manufacturability, since the handling of large amounts of pure Cs is a basic issue in production lines. Here we present in detail results on PIN-OLEDs comprising a newly developed molecular n-dopant. All the previous OLED performance data based on PIN-OLEDs with alkali metal doping could be reproduced and will be further improved in the future. Hence, for the first time, a full manufacturing compatible PIN-OLED is available.

1. Bottom and top emission PIN-OLEDs using alkali metal doped electron transport layers: performance and lifetime

The increase of efficiency and lifetime of OLEDs is of great interest since these parameters have to be improved in respect to the current state-of-the-art level to make OLEDs a superior choice for active-matrix displays whether for mobile phone or mobile video or for TV use. Furthermore utmost power efficiencies are important for future lighting applications.
A first step towards very high power efficiencies was made with the realization of the first PIN-OLEDs. Here, the acronym PIN refers to an OLED structure with a p-doped hole transport layer, an intrinsically conductive emission zone and an n-doped electron transport layer. Three years ago a green emitting PIN OLED was published that reached 100 cd/m² at a voltage of 2.6 V only [1]. Since that time performance of PIN OLEDs improved very rapidly. Very recently, power efficiencies exceeding 100 lm/W were published [2].

Moreover, also the lifetimes of PIN OLEDs increased drastically and now partly even outperform the reference OLEDs without charge carrier doping. The here presented results are based on OLEDs combining the new p-doped hole transport material systems, the n-doped electron transport systems (both developed at Novaled in co-operation with TU Dresden, Institute for Applied Photophysics) and emitter material systems from state-of-the-art material makers. It could be shown that basically all emitter systems can be combined with the PIN approach. Lately, a green PIN-OLED processed by Novaled comprising the phosphorescent emitter $Ir(ppy)_3$ (Tris-phenylpyridine-iridium), a full molecular

hole transport system (matrix material Novaled NHT-5, p-dopant Novaled NDP-2) and a alkali metal doped electron transport system (matrix: BPhen, dopant: Cs) has reached a projected lifetime of about 18,000 hours at an initial brightness of 500 cd/m² (see fig. 1), thus beating the highest lifetime reported so far for an $Ir(ppy)_3$ based OLED of 10,000 hours at 600 cd/m² [3]. This PIN OLED is dc-driven, the measurement is performed on an encapsulated device at room temperature of about 25°C. The encapsulation is made via glass lid and it contains a state of the art getter material. Recent status of lifetime measurement even tend into direction of 25,000h-30,000 hours for this emitter system in a PIN-OLED structure. This major improvement in PIN-OLED lifetime is based on careful device structure design and optimization as well as a careful control over the alkali metal doping process.
Also for red phosphorescent and (deep) blue fluorescent emitter systems, state-of-the-art lifetimes were obtained: 30,000 hours and 6,000 hours at 500 cd/m² initial luminance respectively [4]. Lifetimes of blue diodes are still improved every month. These results show that in terms of efficiency and lifetime PIN OLEDs are the first choice for the production of RGB displays.

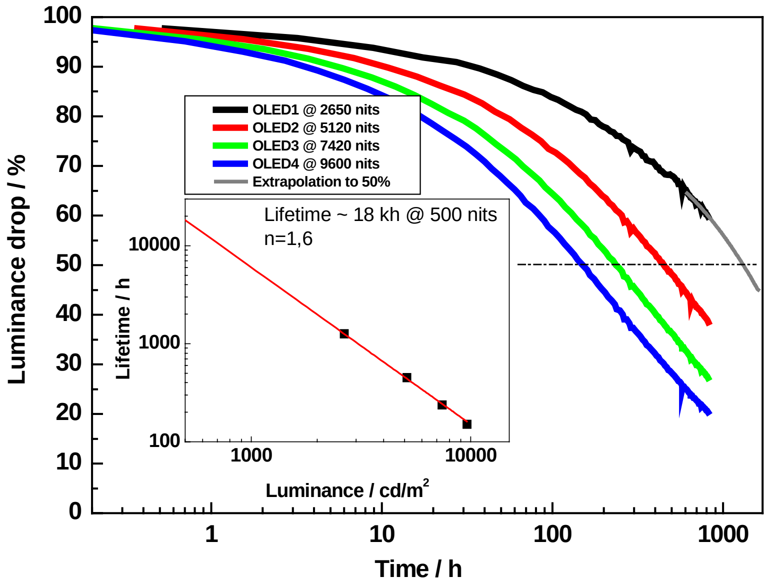


Fig. 1: Lifetime of a green PIN-OLED comprising $Ir(ppy)_3$ as emitter dopant, Cs as n-dopant and NDP-2 as p-dopant. Four identical diodes were measured at four different initial luminances. Their lifetimes (i.e. the time until their brightness drops to 50%) are plotted in a double logarithmic graph (see inset). This graph allows to estimate the lifetime at any given starting luminance. Recent measurements even tend in the direction of 25-30khours lifetime.

The following table 1 gives an overview about the achieved performance values of PIN-OLEDs based on the above described charge carrier transport system.
The basic layer structure of the bottom-emission PIN-OLED consists of a transparent anode (ITO) / p-type doped hole transport layer (HTL featuring Novaled's transport and doping materials) / interlayer on hole side / emission layer (e.g. matrix with phosphorescent or fluorescent emitters) / interlayer on electron side / n-type doped electron transport layer (ETL) / metallic cathode (Al). In contrast to that the top emission OLED have a highly reflective bottom electrode and a semi-transparent top electrode, the

remaining part of the OLED structure stays basically the same, except that the thicknesses of the transport layers are newly adjusted to optimize optical effects in the devices.
The p-type doped HTL comprises a matrix material and as dopant a strong acceptor material, the ETL on the other side contains a matrix and a strong donor material. The conductivity of both transport layers is in the range of 10^{-5} S/cm, which translates into a voltage drop over a 100 nm thick transport layer of only 0.1 V at a current density of 100 mA/cm². Therefore, the driving voltage of PIN OLEDs is very close to the energetic limit of the particular emission colour. The two interlayers prevent exciplex formation thus maintaining and increasing the current efficiency in PIN-OLEDs. Due to the high conductivity of the transport layers, no injection layers between the electrodes and the organic stack are necessary. Even a rather free choice of electrode materials is possible, e.g. ITO pre-treatment is not needed.
After processing of the organic stack and the top electrode, all devices were encapsulated with an UV-curable epoxy glue and a cavity encapsulation glass that includes a getter (in case of top emission no getter is used). After the encapsulation the device characterization takes place under ambient conditions. This includes measurement of LIV-curves, total light output in an integrating sphere, measurement of emission spectra and lifetime (results see above).

Color	**CIE (x/y)**	**Volts**	**cd/A**	**lm/W**	**Q.E.(%)**
		at 1000 cd/m²			
Green bottom phosphorescent	0.30 / 0.62	2.9	83	77	19
Green top phosphorescent	0.30 / 0.65	2.9	78	73	19
Red bottom phosphorescent	0.69 / 0.31	2.9	6.4	6.7	11
Red top phosphorescent	0.68 / 0.31	2.9	8.4	9.1	11
Blue bottom fluorescent	0.15 / 0.24	3.3	5.8	5.6	3
Blue top fluorescent	0.14 / 0.23	3.4	9	8.4	5.5

Tab. 1: Overview of top vs. bottom emitting red, green, and bluePIN-OLEDs based on molecular hole transport system and alkali metal doped n-transport system. The emitter systems in the bottom vs. top comparisons are identically. The current efficiency values of the green PIN OLEDs are due to an improved emitter layer structure. All samples are of the normal type that means having the anode on the substrate. The blue emission colour is a more light blue – suited for mobile phone applications. We have as well realized PIN-OLEDs with more saturated blue colour (coordinates 0.15 / 0.15) and quantum efficiencies in the same range as stated in this table.

The best lifetimes of the bottom emission OLEDs have been given above. Due to latest improvements of the top emission PIN-OLED structure we have been able to extend the lifetime of the top emission OLEDs up to a comparable level of the comparison bottom emission PIN-OLEDs. In figure 2 an example for a still running lifetime test is given. As a very attractive feature for active-matrix driven OLED video displays, no initial drop of the brightness is observed under conditions of normal operation.

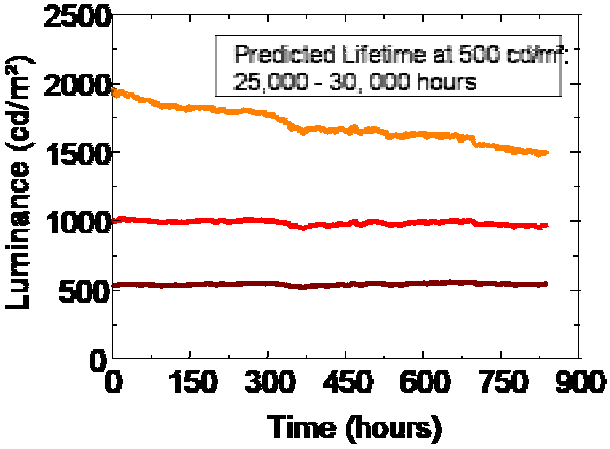


Figure 2: Lifetime measurement of a red phosphorescent top-emission PIN-OLED. Below 1,000 cd/m^2 starting brightness no degradation after 800hours measurement is seen. An estimate of the lifetime would yield around 25-30khours lifetime (at 500cd/m^2 starting brightness). The non-occurrence of an initial drop is a very important feature for RGB video displays since the white colour point would not shift in such a case. The other basic colours green and blue are currently under investigation. First results indicates similar results.

As a summary very high power efficiencies and state-of-the-art lifetime can be achieved with a proper PIN-OLED technology, both for top and bottom emission. This can, in contradiction to prior established opinions, even be achieved with alkali metal doping of the electron transport layer.

2. Performance of PIN OLEDs comprising molecular p- and n-dopant

All OLEDs from chapter 1 still contain Cs as n-dopant. For large scale production the use of Cs is a major drawback due to difficult process control and security issues of Cs handling.
For the hole transport side of the PIN-OLED, the newly developed molecular p-dopant NDP-2 fulfils all requirements for large scale production, e.g. low vapour pressure, good controllability and high evaporation temperature.
To overcome the drawback on the electron transport side, a molecular n-dopant has been developed by Novaled that allows for large scale processing of stable, highly efficient, long-living PIN-OLEDs. Results of diodes comprising this new dopant (named NDN-1) and the comparison to Cs-doped reference diodes are given here. The new molecular n-dopant is co-deposited with its matrix material (Novaled NET-5) in the normal way, its deposition temperature is around 200°C, its molecular mass above 500g/mol.

The layer structure of the following samples is similar to the one described above, differences occur partly in the emission layer material setup.

The figures 3 and 4 show a comparison on red, green and blue PIN-diodes, prepared both with Cs and with NDN-1 as n-dopant. Obviously, the efficiencies as well as the jV-curves are similar for the two n-dopants. However, the lifetime (shown in figure 4 for the blue OLEDs) seems to be higher for the molecular n-dopant. Further investigations to confirm these results are underway.

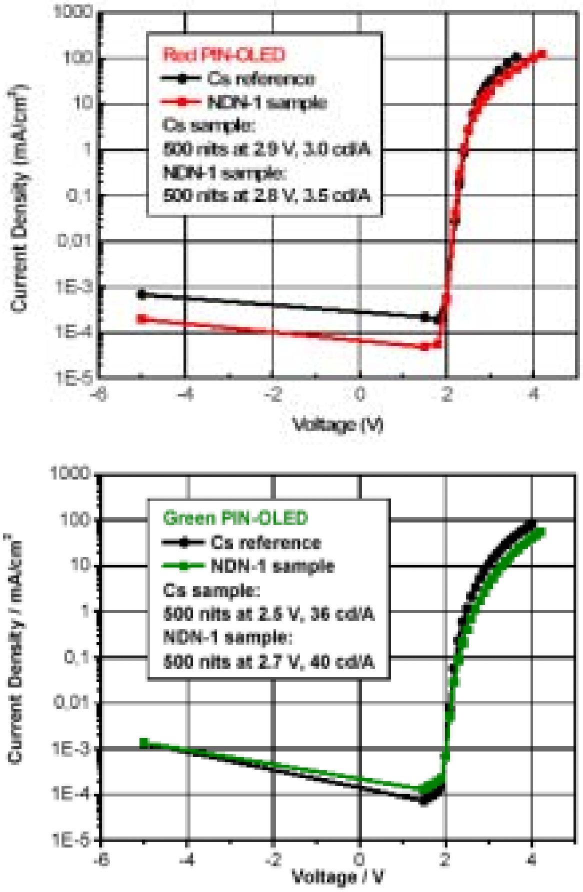
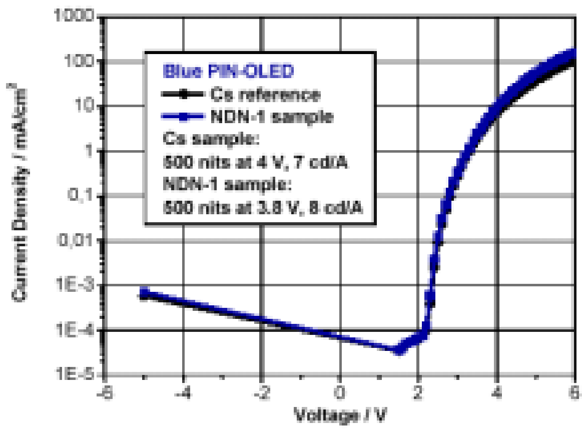


Fig. 3: Current density vs. Voltage graphs of red (top) and green (bottom) phosphorescent PIN-OLEDs: Comparison of devices doped with the new organic n-dopant NDN-1 (red and green curves) and Cs-doped reference diodes (black curves). The inset states the current density and operating voltage at 500 nits brightness. The here used emitter material systems differ from the best ones reported in chapter 1.



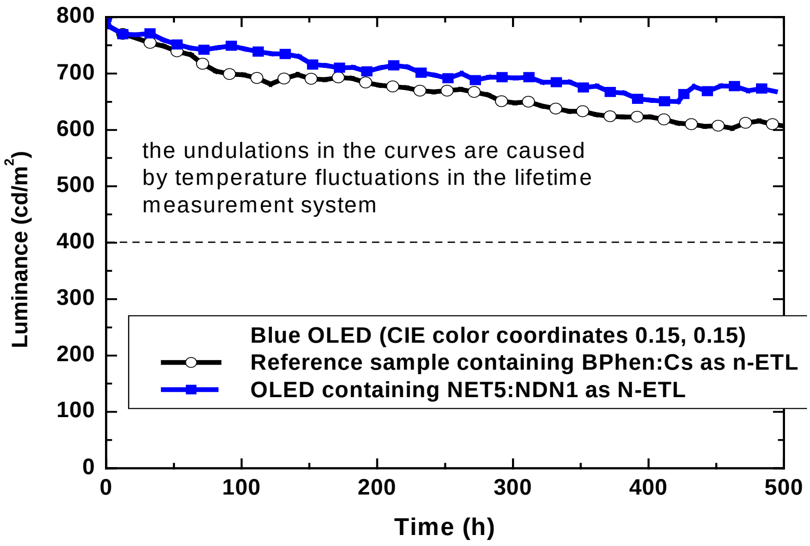
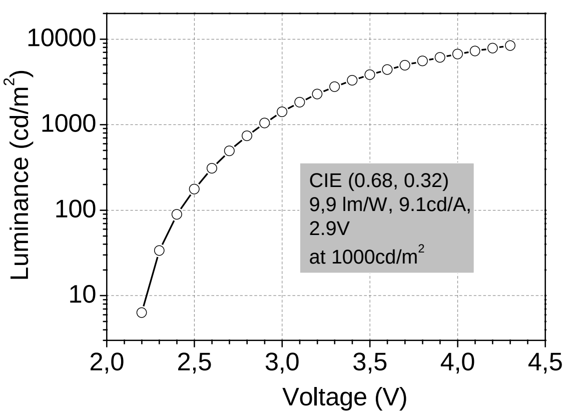


Fig. 4: Comparison of a blue fluorescent PIN-OLED doped with the new organic n-dopant NDN-1 (blue curve) and a Cs-doped reference diode (black curve): jV-curve (top) and lifetime measurement (bottom). The NDN-1 containing sample has similar LIV-curve and even higher lifetime. The here used emitter material systems differ from the best ones reported in chapter 1.

Now we are improving the performance of our PIN-OLEDs containing the new molecular n-dopant further. As an example figure 5 shows latest results for a red phosphorescent PIN-OLED.



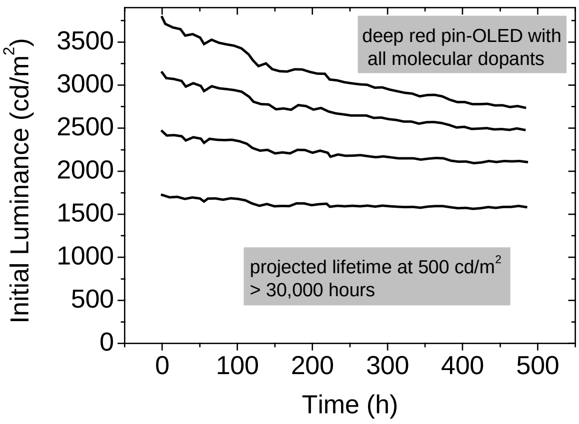


Fig. 5: Results for a red phosphorescent bottom emission PIN-OLED with only molecular dopants (NDP-2 resp. NDN-1, top figure: L-V curve, below: lifetime measurements). Performance in this case is even better than our best PIN-OLED containing Cs as n-dopant. Lifetime tests are still running but indicating very high stability. Due to recent improvements in the device structure also here no initial drop is observed.

3. Summary

With the integration of a molecular n-dopant in red, green, and blue PIN OLEDs the last step towards a production ready RGB PIN-OLED stack has been done. The Novaled material set consisting of molecular p- and n-dopants, their respective matrix materials and interlayer materials allows to implement both fluorescent and phosphorescent emitting systems into the OLED stack, thus providing world-record efficiencies and state-of-the-art lifetimes that are already inline with the requirements for many display applications.

Furthermore, the PIN approach enables the production of top emitting structures with same performance and lifetime as standard bottom emission PIN OLEDs, which is of particular interest for active matrix display applications since this top emitting design drastically increases the overall aperture ratio of the display, decreases the current load at OLED level, hence increases system lifetime, and gives more freedom for the design of the TFT circuitry.

Further development steps will include even stronger dopants to be able to simplify the OLED layer structure. The PIN-OLED approach is currently under test on a large area vertical Inline deposition system (made by Applied Films, Germany; located and operated at Fraunhofer-Institute for Photonic Microsystems, Germany) to show its compatibility with this next generation advanced manufacturing technology.

4. Acknowledgment

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5. References

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