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Concepts for solution-processable OLED materials at Merck

A. Hayer*, R. Anémian, T. Eberle, S. Heun, A. Ludemann, N. Schulte and H. Buchholz

Merck KGaA, Frankfurter Straße 250, 64293 Darmstadt, Germany

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Drastic performance increase and accelerated progress in organic light-emitting diodes from solutions are shown using solution-processable small molecules based on the core structures of vacuum-evaporable materials. Systematic modification of small-molecule materials toward better processability from solutions at identical electro-optical properties is shown. This is demonstrated to lead to a significant improvement in the device performance.

Keywords: OLED; material; soluble

1. Introduction

Organic light-emitting diodes (OLEDs) have drawn much attention due to their potential application in next-generation flat-panel displays [1] and lighting fixtures [2,3]. Displays based on vacuum-evaporated small molecules are already being mass produced, and can already be found in mobile applications such as cell phones [4,5] and even in some TV sets [6]. While solution-based materials offer the potential for greater ease of processing, reduced manufacturing costs, and very large-area application [7], the improvement of their power efficiency and device lifetime is still a challenge [8].

In this paper, it is shown how solution-processable small molecules based on the core structures of vacuum-evaporable materials can be used to accelerate performance progress. The materials designed for vacuum evaporation have been highly optimized in terms of electro-optical properties and device performance. However, the standard evaporable materials often cannot be used in solution-processed OLEDs due to factors such as insufficient solubility, high crystallization and/or phase separation tendency, and glass transition temperatures lower than the temperatures needed for solvent removal (Figure 1). While the relationship between molecular structure, film formation, and the ensuing OLED device performance has not yet been widely studied, it has been shown in a systematic study [9] that solution-processed films not only have morphologies and densities different from those of their evaporated counterparts, but also have an impact on efficiency and charge balance. It has been shown in another study [10] that bulky side groups, which effectively hinder aggregation, can indeed improve power efficiency.

2. Results and discussion

In this paper, the systematic modification of small-molecule materials toward better processability from solutions at identical electro-optical properties is shown, as well as an ensuing performance increase. The concept is based on the use of a constant molecular core combined with the adaptation of the periphery of the molecule without disturbing the electro-optically active unit (Figure 2).

As shown in Table 1, the aforementioned concept was successful for all classes of materials. For triplet hosts, singlet hosts, and triplet emitters, increased solubility and glass transition temperatures were achieved, while the energy levels remained unchanged. The improvement was particularly drastic for the singlet blue host shown here, where the standard evaporable material could not be dissolved at all in a concentration suitable for OLED processing. The effect of the improved processing on film formation in green, red, and blue OLEDs is shown in Figure 3.

Figure 4 shows that the improvement in processability can be used to achieve a significant improvement in device performance. Three examples based on triplet green hosts are shown. In the left example, two triplet green hosts based on the same core A combined with either periphery 1 or 2 are compared. Better processability is shown to lead to a significant improvement in device efficiency. In the middle example, a similar efficiency increase is shown to have been obtained for two hosts based on the same core B. In addition to this, the efficiency can be increased even further if a second processability-improving co-host is added (circles). Besides the improved performance at an identical composition, as shown in the left and middle examples, improved processability has another

*Corresponding author. Email: anna.hayer@merck.de

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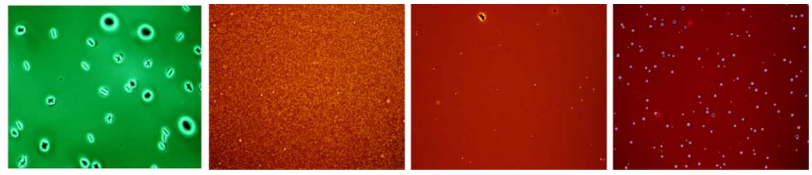

Figure 1. Low film quality in solution-processed films from standard evaporable OLED materials. From left to right: Crystallization (e.g. triplet green host), phase segregation (e.g. triplet red host), crystallization (e.g. triplet red host), and micelle formation (e.g. triplet red emitter). Note that no singlet blue example is given as its processability in the chosen standard evaporable host material is so low that it cannot be dissolved at all, and therefore, no film can be prepared from the solution.

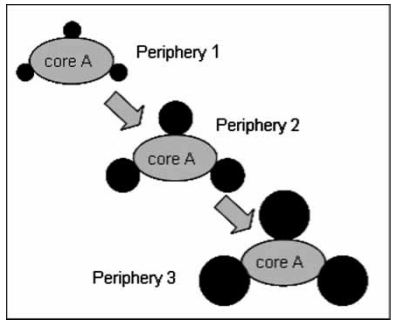

Figure 2. Adapted molecular periphery on a constant molecular core.

advantage: if all the components in a host/emitter mix are well soluble and easily processable, then the mixture can be concentration-optimized purely in terms of what gives the best electro-optical properties. The right example in Figure 4 shows that once a full range of relative concentrations of the components is accessible, the performance can be improved even further.

Efficiency is not the only aspect of performance that can be drastically improved. Figure 5 shows that the OLED device lifetimes were increased even more significantly (middle and right). Due to the new-material development strategy, which is universally applicable, a wide range of

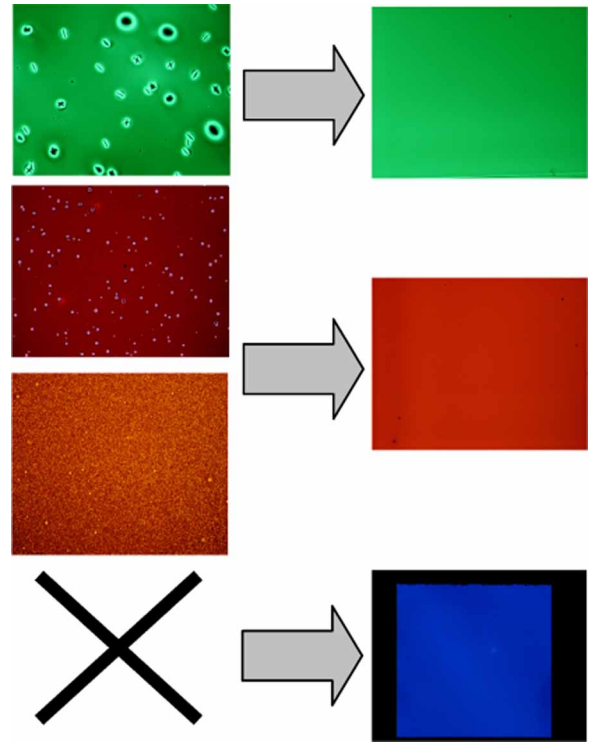

Figure 3. Improved film quality in the green (top), red (middle), and blue (bottom) OLEDs using the material strategy shown in Figure 1.

Table 1. Solubility, glass transition temperatures, and energy levels for triplet green hosts, singlet blue hosts, and triplet red emitters

	Periphery 1	Periphery 2	Periphery 3	
Triplet green hosts				
Soluble in toluene	At 70°C	At 50°C	At 20°C	⇒ Improved
Tg (°C)	97.9	109.5	159.6	⇒ Increased
HOMO (eV)	−6.32	−6.28	−6.31	⇒ Unchanged
LUMO (eV)	−2.79	−2.80	−2.80	⇒ Unchanged
Singlet blue hosts				
Solubility	Insoluble	Chlorobenzene	Toluene	⇒ Improved
Tg (°C)	148	177	174	⇒ Increased
HOMO (eV)	−5.61	−5.63	−5.63	⇒ Unchanged
LUMO (eV)	−2.70	−2.70	−2.71	⇒ Unchanged
Triplet red emitters				
Solubility	+	++		⇒ Improved
Tg(°C)	165	202		⇒ Increased
HOMO (eV)	−5.27	−5.34		⇒ Unchanged
LUMO (eV)	−2.67	−2.77		⇒ Unchanged

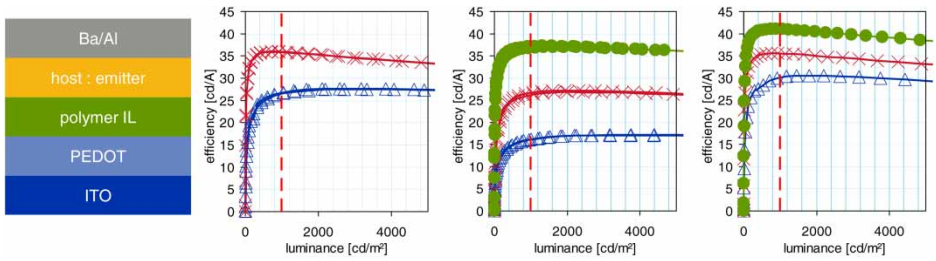

Figure 4. Device stack and efficiency results from three different triplet green OLED systems. Left: Standard evaporable host A (triangles) versus an A-based host with improved processability (crosses). Middle: Standard evaporable host B (triangles) versus a B-based host with improved processability (crosses) versus a B-based host mix with improved processability (circles). Right: Concentration optimization in a three-component mix. CIE color coordinates: $0.33 < x < 0.34$ and $0.62 < y < 0.63$ for all the samples.

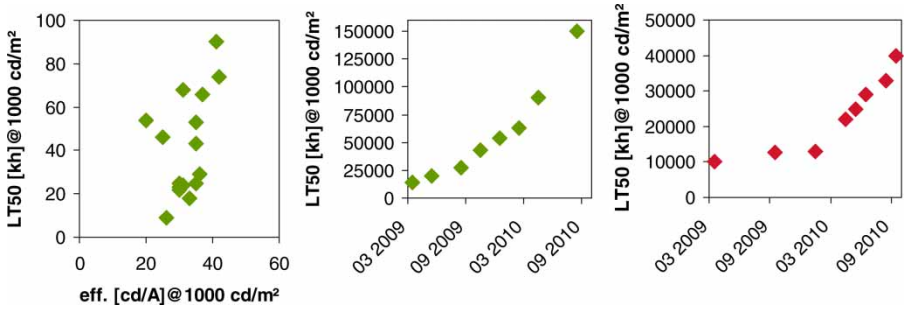

Figure 5. Impact of new host materials on OLED lifetime and efficiency. Left: Lifetimes correlated with efficiencies for a series of green host systems now accessible due to the new-material development strategy. Middle: Development of green OLED device lifetimes in recent months. Right: Development of red OLED device lifetimes in recent months. CIE color coordinates: $0.33 < x < 0.34$ and $0.62 < y < 0.63$ for all the green samples, and $0.65 < x < 0.66$ and $0.34 < y < 0.35$ for all the red samples.

host materials based on the core structures of state-of-the-art evaporable materials are now accessible. Consequently, optimization can be carried out within a wide range of material mixes, which, in turn, will lead to several compositions that combine good efficiency and lifetime (Figure 5, left).

While almost all the lifetime improvements shown in the middle and right parts of Figure 5 were achieved solely by optimizing the emissive layer, each graph contains one device indicating the potential of further optimization based on the adjacent layers: the highest lifetimes of 40.000 h at 1000 cd/m^2 in triplet red OLEDs and of 150.000 h at 1000 cd/m^2 in triplet green OLEDs were achieved using a new polymeric hole transport layer with improved hole injection characteristics.

3. Summary

A universal approach toward high-performance solution-processable materials was shown, based on the systematic improvement in processability at unchanged electro-optical properties using a constant core structure and varying the periphery. The applicability of the approach to triplet hosts, singlet hosts, and triplet emitters was shown. Excellent film formation ensued. Strong performance increase (efficiency and lifetime) based on this new material development concept was demonstrated. Lifetimes of up to 150.000 h in OLEDs processed from solutions were achieved.

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