From Molecule to Device:

Predictive Simulations for OLEDs

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ABSTRACT

Simbeyond's progress towards achieving a complete end-to-end predictive device simulator from the molecule up to the device scale is briefly presented.

1 Introduction

Despite the successes of OLED applications, the research and development (R&D) processes still mainly rely on costly and labor-intensive experiment-driven trial-and-error approaches. OLED device manufacturers, chemical material suppliers and technology institutes are urgently looking for a breakthrough in their R&D approach, which should result in less development cycles and therefore in a significant reduction in time-to-market and increase in R&D cost effectiveness.

With the help of predictive virtual experiments, R&D time for OLED materials and devices can be effectively reduced by up to 40% and R&D cost effectiveness can be increased by up to 150%. Significant prototype and experiment based parts of the R&D process can be replaced by computer simulations. Experimentally inaccessible physical processes can be easily studied with only minimal effort required by the modeler. A wide range of possibilities for stack optimization then becomes available. At Simbeyond, we developed such predictive simulation software. With our Bumblebee[1] software one can simulate the performance of OLED device stacks with complex 3D morphologies from the nanosecond up to its full lifetime. All relevant physical processes can be studied, including charge transport, exciton dynamics, loss processes, final emission[2] and even degradation scenarios[3,4]. Using Bumblebee, both the material and stack properties can be optimized simultaneously, thus revolutionizing OLED R&D.

We will discuss our vision about a fully integrated multiscale toolchain for OLEDs and then present a concrete example of a predictive device simulation for OLEDs.

2 Multiscale toolchain

In Bumblebee, input parameters such as molecular energy levels or excitonic lifetimes are currently needed. These material specific parameters can be obtained from experiment, literature libraries, material suppliers, or can be calculated directly using ab initio methods. In a future product, quantum chemistry simulations will be seamlessly integrated within an automatic simulation workflow that links the molecular scale to the device scale (Fig. 1). Such a multiscale toolchain will allow the modelers to simulate all the relevant scales required for a fully-predictive simulation workflow. The molecular properties required for device simulations are consistently scaled up all the way up to the material morphology of the layers of the stack.

This multiscale toolchain, which we are developing in collaboration with the Amsterdam-based company SCM, will enable efficient collaboration between chemical material suppliers and display manufacturers. It will ultimately reduce the number of OLED device development cycles and consequently increase R&D cost effectiveness.

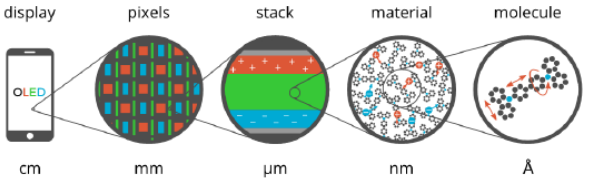

Fig. 1 Multiscale OLED device simulations. Schematic representation of the different scales that are relevant for OLED displays simulations.

3 Results: From material properties to device efficiency and lifetime

We present here an example of a predictive simulation study of a device based on the stack that was experimentally characterized by Furukawa et. al.[5]. Simulation input parameters for the stack and the materials have been taken from the literature. Typical values have been used for material parameters that were not known.

In Fig. 2, a visualization snapshot of a typical simulation is shown. Bumblebee can simulate any morphology and provides time and 3D spatially resolved views of your device functioning.

The simulated stack presented in Fig 3, is based on a thermally activated delayed fluorescence (TADF) emitter,

4-CzIPN-Me. The emissive layer is made of a host-guest mixture with 93.7% of mCBP and 6.3% of the TADF dye. A steady state analysis of the device will now be performed and compared with the experimental results. The evolution of current density as a function of voltage is presented in Fig. 4. The conclusion that we can draw from this result is that there is a good match between the experimental and simulated results at all voltages.

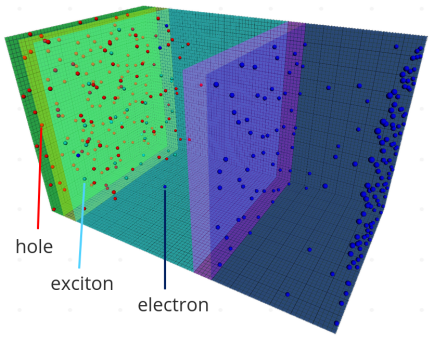

Fig. 2 Visualization of a Bumblebee OLED device simulation - Digital twin of the device. Holes, electrons and excitons are indicated. Bumblebee simulates charge transport and excitonic processes on each molecular site in 3D. The interplay among all these processes is simulated in time.

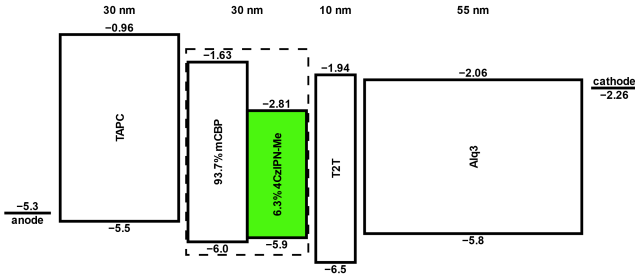

Fig. 3 Energy diagram of the simulated stack.

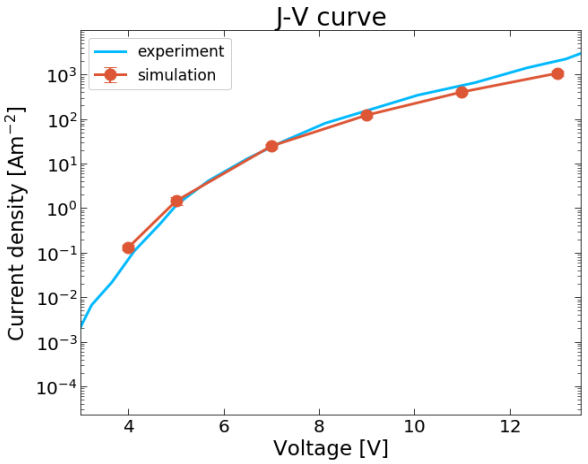

Fig. 4 Current density curve

Fig. 5 presents the evolution of the External Quantum Efficiency (EQE) as a function of current density. Those results were obtained by assuming a light outcoupling of 25% at all current densities. The results show a good matching between experimental and simulated results at both the lower and higher current densities. Some differences are seen at intermediate current densities. Those differences can be explained by the assumption of the constant light outcoupling.

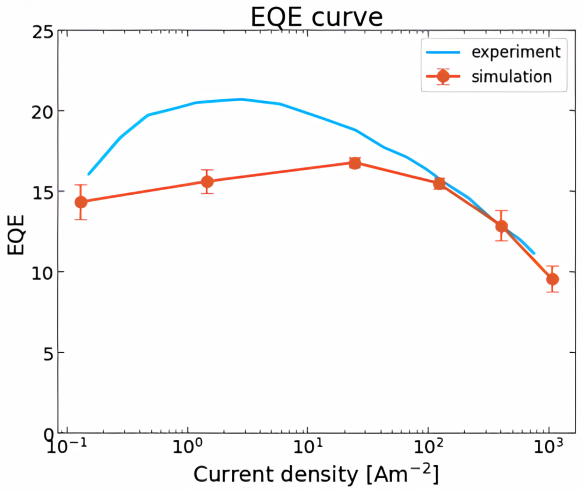

Fig. 5 External quantum efficiency curve

Another interesting result that can be obtained with Bumblebee is the dependence of the operating voltage on the loss mechanisms in the stack. These results are shown in Fig. 6. The first conclusion that we can draw is that the impact of the non-radiative decay loss is mostly unchanged at all voltages. On the other hand, we can see that both the quenching and annihilation are getting responsible for most of the losses at higher voltages, which is expected due to higher exciton densities. A result that was not expected is the importance of the quenching at lower operating voltages. This can most

likely be explained by a charge accumulation at the interface with the EML, that leads to high exciton-polaron quenching rate.

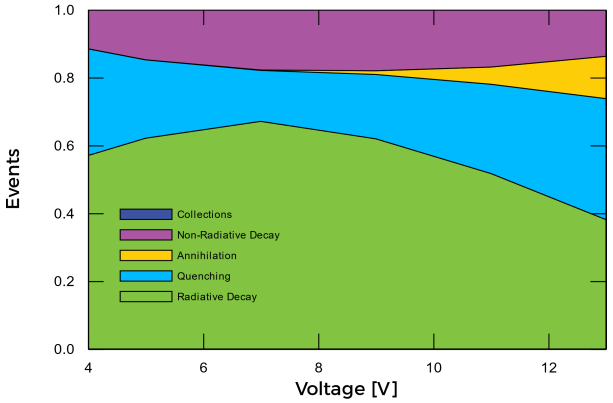

Fig. 6 Excitonic events contributing to the device efficiency and losses at each voltage.

Now that the analysis of the steady state has been discussed, the lifetime of the device will be studied. Fig 7 displays two main lifetime results: the evolution of radiative rate decay and the voltage shift. The influence of different parameters on the device lifetime and driving voltage has been studied. First, we focused only on the degradation of the emitter in the EML and varied the properties of the degraded products. Second, we refined our study including the material degradation in the other layers. Four scenarios for the degraded products were considered. Either the degraded sites became an electron trapping, a hole trapping, a full-trapping site (electron and hole) or inactive site.

The first conclusion that we can draw from these simulations is that the device lifetime may depend strongly on the properties of the degradation product. We observe that inactive sites do not significantly contribute to the decay in light emission. This suggests that degraded sites must have at least some carrier trapping character. Simulations predict negligible difference in radiative rate decay between electron and hole trapping degradation products, while a slightly worse lifetime is predicted for this OLED stack when the emitters degrade into fully-trapping sites (yellow curve, electron+hole trapping). On the other hand, enabling degradation of the transport layers in the simulation (in purple) has a mild effect on the radiative rate decay curve when comparing with the scenario where only the TADF emitters can degrade (in yellow). This result is understood by considering that degraded sites in the transport layers mainly affect charge transport, while their impact on the light emission is only indirect.

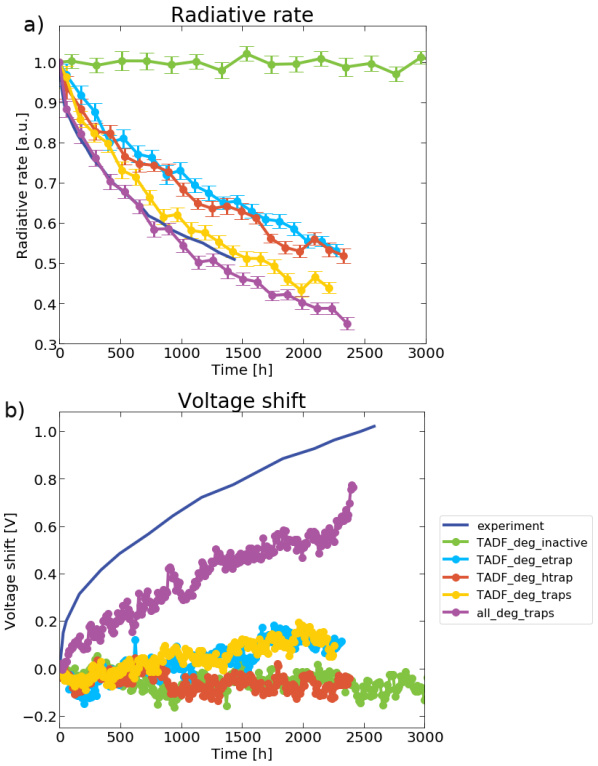

Fig. 7 Lifetime prediction for different degradation scenarios. a) decay in radiative rate due to degradation and b) voltage shift during degradation. The degradation products properties are varied.

The relative trends are quite different when looking at the effects on the driving voltage. The voltage shift predicted by Bumblebee shows that both inactive and hole trapping sites cannot be responsible for the voltage shift observed in the experiment. The increase in voltage shift seen in experiment can most likely be attributed to the formation of electron trapping. It is clear however that in the case where the emitter material is the only degrading material in the stack, we cannot fully explain the experimentally measured voltage shift. When the transport layers are included in the degradation scenario, it results in a major impact on the voltage shift. The voltage shift curve obtained with this scenario is getting much closer to the experimentally observed curve.

4 Discussion: Understanding the remaining differences between experimental and simulated results

The overall experimental trend is reasonably well reproduced for both the radiative decay rate and voltage shift. However, there is still a quick initial change of the experimental voltage shift curve that is not well reproduced by our modeling.

A possible explanation for the presence of seemingly two regimes in the voltage time dependence (a quick component and a slower component) can be found when considering an extrinsic origin for this effect, such as the presence of water contamination. In fact, water

may accelerate the degradation of transport layers up to a certain extent at the beginning of the lifetime measurements. After which, the normal intrinsic degradation mechanisms become dominant. Experimental evidence of this behavior has been reported by H. Yamamoto et. al, showing that ultra-high vacuum fabrication of devices with a lower concentration of water contaminants in the stack resulted in a reduced luminance and voltage change during the first stages of device degradation[6]. They argued that electrochemical reduction of water impurities early in the lifetime tests may result in the formation of traps causing a sudden change in the operating voltage that later saturates. This effect might be responsible for the initial drop that is observed in the stack that we studied.

5 Conclusion: Added-value of simulation-assisted processes with Bumblebee

We presented here new and valuable insights into OLED device degradation and lifetime, and provided a concrete case study explaining how Bumblebee can bring added value to your R&D. The results presented here are only a small example of what you can achieve with Bumblebee. However, you can already learn why Bumblebee 3D simulations of OLED stacks are necessary to understand the subtle contributions that each process has in determining the stack performance and lifetime.

References

[1] Bumblebee is the state-of-the art Kinetic Monte Carlo simulation code of Simbeyond B.V, https://simbeyond.com/bumblebee/

[2] Mesta, M., Carvelli, M., De Vries, R.J., Van Eersel, H., Van Der Holst, J.J., Schober, M., Furno, M., Lüssem, B., Leo, K., Loebl, P. and Coehoorn, R., 2013. Molecular-scale simulation of electroluminescence in a multilayer white organic light-emitting diode. *Nature materials*, *12*(7), pp.652-658.

[3] Coehoorn, R., Van Eersel, H., Bobbert, P. and Janssen, R., 2015. Kinetic Monte Carlo study of the sensitivity of OLED efficiency and lifetime to materials parameters. Advanced Functional Materials, 25(13), pp.2024-2037.

[4] Gottardi, S., Barbry, M., Coehoorn, R. and Van Eersel, H., 2019. Efficiency loss processes in hyperfluorescent OLEDs: A kinetic Monte Carlo study. Applied Physics Letters, 114(7), p.073301.

[5] Furukawa, T., Nakanotani, H., Inoue, M. and Adachi, C., 2015. Dual enhancement of electroluminescence efficiency and operational stability by rapid upconversion of triplet excitons in OLEDs. Scientific reports, 5(1), pp.1-8.

[6] Yamamoto, H., Brooks, J., Weaver, M. S., Brown, J. J., Murakami, T., & Murata, H., 2011. Improved initial drop in operational lifetime of blue phosphorescent organic light emitting device fabricated under ultra high vacuum condition. Applied Physics Letters, 99(3), 138.