Analysis of Self-Heating and Negative Capacitance in Organic Semiconductors Devices

Evelyne Knapp*, Beat Ruhstaller***

***Zurich University of Applied Sciences, Institute of Computational Physics, Winterthur, Switzerland**

****Fluxim AG, Winterthur, Switzerland**

Abstract

A numerical model for charge transport in organic semiconductor devices that accounts for self-heating is presented. In admittance spectroscopy this model reproduces the negative capacitance in bipolar, and more importantly, in single carrier devices. We show that self-heating is crucial not only in large-area OLEDs, but also in small-area devices.

Author Keywords

OLED; self-heating; negative capacitance; admittance spectroscopy; simulation

1. Introduction

Admittance spectroscopy is a popular and powerful characterization technique for organic semiconductor devices. Numerous publications exploit this technique [1, 2, 5, 9, 10, 11, 14, 15]. Negative capacitance values are often observed in admittance spectroscopy at high bias and low frequency as shown in Figure 1 for an OLED and have puzzled scientists ever since.

Negative capacitance measurements are well known from Si diodes and other devices. Numerous explanations [3] and references therein to parasitic effects [4] were proposed as origin.

First reports of negative capacitance in organic LEDs appeared about a decade ago [5, 6]. In general the same device also exhibits positive capacitances at different measurement conditions such as frequency, applied voltage or temperature. The occurrence of negative capacitance implies that an increase of voltage leads to a reduction of charges on the electrodes. A wide range of explanations for the negative capacitance have been suggested for organic semiconductors [7, 8]. However, these numerous explanations for the origin of negative capacitance are restricted to bipolar devices and therefore cannot explain the occurrence in hole-only devices or electron-only devices. Little work has been dedicated to unipolar devices so far. In references [9, 10] single carrier devices were fabricated and measured exhibiting negative capacitance. The authors attribute the observed negative capacitance effects to interfacial states.

Only recently, another origin for the negative capacitance in organic semiconductor devices has been identified by Okumoto and Tsutsui [11] for a hole-only and bipolar device, namely the temperature. Self-heating of the device changes the capacitance value of the structure. Okumoto and Tsutsui have used a copper block on top of the hole-only device and found a reduced negative capacitance value indicating that the temperature plays an important role. So far, self-heating of organic semiconductor devices is exclusively discussed in large-area devices [12]. Only recently the influence of self-heating in small devices has been pointed out [13].

In this contribution we investigate the occurrence of a negative capacitance in single carrier devices by simulating admittance measurements with the aid of a one dimensional drift-diffusion model that is extended by a heat generation and transport equation. We also shed light on the time-dependent as well as steady-state behaviour of a device with self-heating.

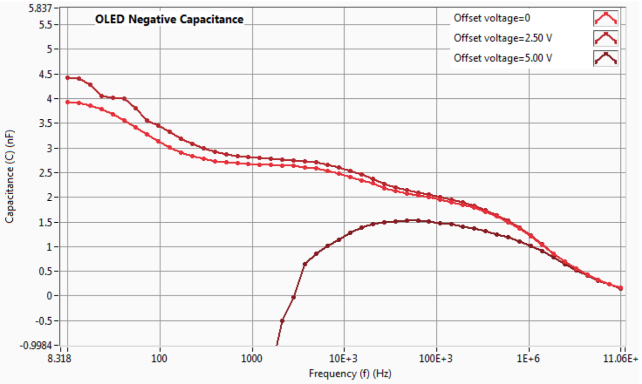

Figure 1. Capacitance vs. frequency data of a bipolar (OLED) device at different bias applied voltage. By increasing the bias the capacitance becomes negative in the low frequency regime. The measurements were performed with the all-in-one instrument Paios [17].

We thus hope to contribute new insights to the debate over the origin of negative capacitance for organic semiconductor devices and to the implications that self-heating has on OLED applications.

2. Mathematical Model

In our modelling approach we simplify a multilayer OLED to a single layer and hole-only device as illustrated in Figure 2 in order to illustrate that the key observations can be made even in a simple model device.

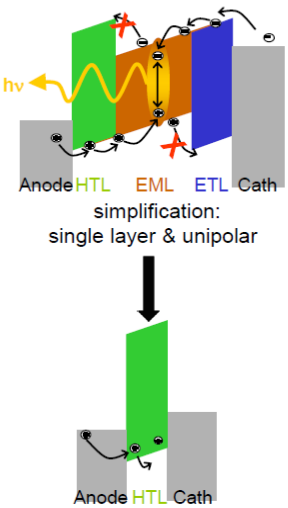

Figure 2. Model simplifications leading to a unipolar, single-layer device.

In the organic material we then solve the one dimensional drift-diffusion model for the hole concentration and the electrical potential. A more detailed description of this model can be found in reference [14, 15]. This model is denoted as 1D-DD in the following.

In a next step we extend the classical drift-diffusion model by the heat equation where T is the temperature, c the specific heat capacity, ρ the density and k the thermal conductivity of the organic semiconductor material:

$$c\rho\frac{\partial T}{\partial t} = \nabla \cdot (k\nabla T) + \frac{J_p^{\,2}}{q\mu p}.$$

The heat source on the right side is Joule heating and depends on the hole current J_p, the elementary charge q, the hole density p and the hole mobility μ. For the boundary we use convective boundary conditions where the heat flux density F is given in terms of the heat transfer coefficient h and the ambient temperature T_{ref}:

$$F = -k\nabla T = h(T_{ref} - T).$$

The 1D-DD plus the heat equation is denoted as 1D-EDD in the remainder of this paper. Please note that we assume for simplicity a constant mobility although organic semiconductors exhibit a temperature dependent mobility. It is expected that a temperature dependent mobility would further enhance self-heating effects. But the main characteristics of self-heating become apparent even when assuming constant mobility.

3. Steady-State Simulations and Measurements

In the following the two models are used to simulate a current-voltage curve. Model parameters that are used in the following simulations are listed in **Table 1** at the end of the paper.

The 1D-DD model is displayed in black in Figure 3, while the 1D-EDD is shown in red. At high voltage the current is heating up the device leading to an enhanced carrier diffusion and thus higher current density.

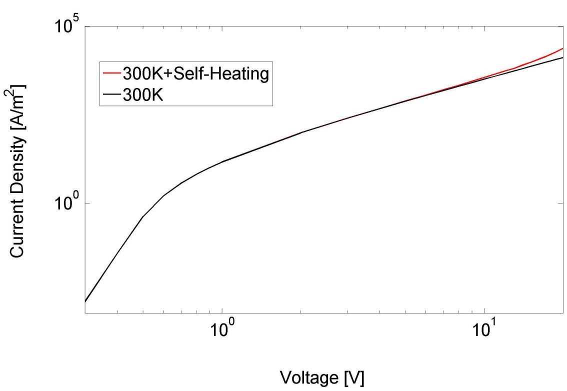

Figure 3. Current-voltage curves for the two models. At high bias the two models show a different behavior.

In Figure 4 a current-voltage measurement of a bipolar OLED is shown. The black line represents a fast measurement where the device is not heating up in time to be noted on the current-voltage curve. The red line shows a slow current-voltage curve measurement where the device has time to heat up. The experimental data in Figure 4 is in good qualitative agreement with the simulation in Figure 3.

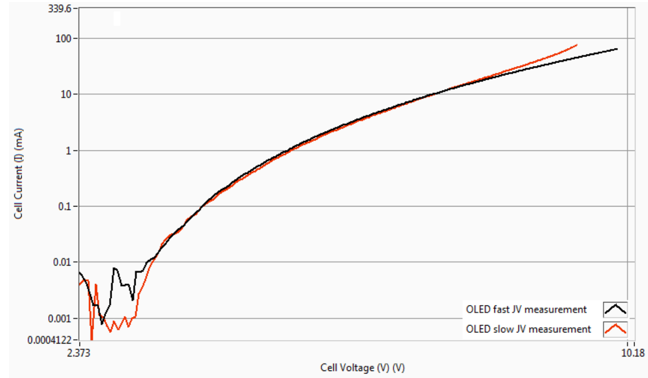

Figure 4. Experimental current-voltage curves for a "slow" (in black) and "fast" (in red) measurement. The fast measurement does not allow the device to heat up to its steady-state temperature.

To explain the enhanced current density at higher temperature two charge carrier profiles at different temperatures are illustrated and compared in Figure 5. We show the results for V_{app} = 6 V at 150 K and 300 K. The temperature difference is chosen to be large such that the influence on the charge carrier profiles can be recognized. The higher temperature leads to stronger diffusion and thus to more charge carriers resulting in a higher current density.

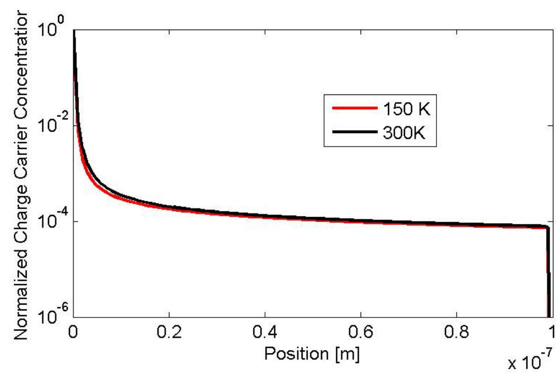

Figure 5. Charge carrier profiles for low and high device temperature. High temperature leads to large charge carriers and more current.

4. Dynamic Simulations

In this section we apply our modelling approach to study the impact of the device temperature in dynamic characterization like admittance spectroscopy and dark injection transient current experiments.

4.1. Negative Capacitance

As shown in Figure 6 the 1D-DD model (given in black) is used to calculate the capacitance of the hole-only device at V_{app} = 6 V with numerical methods that we previously described [14,15]. The result is in agreement with analytical solutions for the simplified drift-only problem [16] (not shown here). In a second step we use the 1D-EDD and calculate the capacitance for different applied voltages. We observe a marked decrease of capacitance and change of sign below a certain frequency. In the logarithmic representation of Figure 6 the absolute and therefore positive values of capacitance are plotted. The difference in capacitance between the 1D-EDD and the 1D-DD model is getting more and more pronounced with increasing voltage. This clearly implies that more Joule heating and thus an increasing device temperature is responsible for the negative capacitance effect.

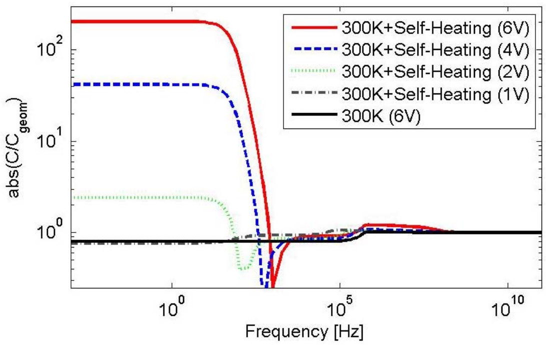

Figure 6. Absolute values of the negative capacitances for different applied voltages with the 1D-EDD model. The black line is simulated with the 1D-DD model.

Negative capacitance values were also recently reported by Okumotot and Tsutsui [11] where a hole-only device as well as a bipolar device were measured. A change in the capacitance was obtained in reference [11] by adding a copper block to the device which serves as a heat sink and thus helps cooling the device.

In Figure 7 we simulate the organic part of the device structure of reference [11]. We achieve a very good agreement between the measurements [11] and our simulations. The influence of the copper block is clearly visible. The device temperature is lower and the negative capacitance effect less pronounced than in [11]. In our simulation we modeled the Cu block cooling by increasing the heat coefficient h in the boundary conditions.

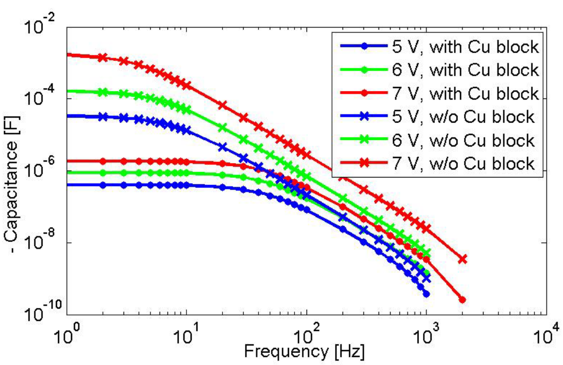

Figure 7. Simulation of a hole-only device (organic layer is 150 nm, rest of parameters as in **Table 1**) at different voltages with and without cooper block that serves a heat sink. The copper block is simulated by changing the heat transfer boundary conditions.

4.2. Dark Injection Transient Currents

We now perform another computer experiment where a voltage step of 0.1 V at 6 V is applied. When we look at the current density response a cusp is visible at around 1e-6 s. This is in agreement with formula [16]:

$$t_{DI} = 0.786 \frac{L^2}{\mu(V - V_{bi})}.$$

In the case of the 1D-DD model the steady-state current density is reached almost immediately after the cusp in **Figure 8**, while for the 1D-EDD the current density still increases till it reaches its steady-state value at around 1e-2 s, which is a time scale that is almost 4 orders of magnitude larger than the carrier transit time. The slow process of heating up the device shows its signature in the dark injection transient current experiment at long times as well as in the admittance spectroscopy at low frequency.

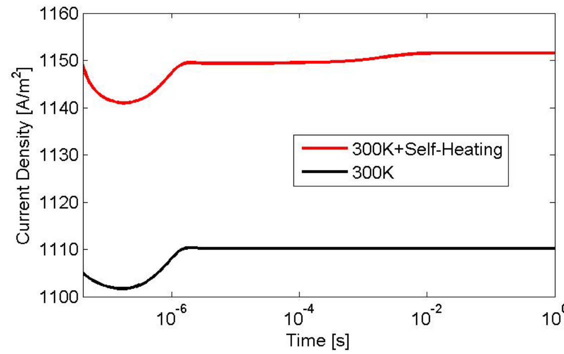

Figure 8. Dark injection transient currents results for the 1D-DD model are shown in black and for the 1D-EDD model in red. The self-heating effect starts at around 1e-2 s as shown in the red curve.

Table 1. Simulation parameters are displayed for the thermal and the drift-diffusion model. Further, device configurations are listed.

	parameter	value	units
electrical	μ	$1e-9$	$m^2V^{-1}s^{-1}$
	N_0	$1e27$	m^{-3}
	ϵ_{rel}	3	
thermal	k	$2e-1$	$WK^{-1}m^{-1}$
	c	1.7e3	$Jkg^{-1}K^{-1}$
	ρ	$1.2e3$	kgm^{-3}
device	V_{bi}	0.8	eV
	L	$100e-9$	m
	$p(0)$	N_0	m^{-3}
	$p(L)$	N_0	m^{-3}

5. Conclusion and Outlook

In this paper we have presented an extended modelling approach for OLED simulations that accounts for self-heating of the device through Joule heating.

We have demonstrated the influence of the device temperature on the current-voltage curve and compared it with measurements. Moreover, the dynamic behavior of a hole-only device was discussed in terms of the negative capacitance. Our simulations confirm self-heating as the origin for the negative capacitance effect in single-carrier devices and most likely also in multilayer, bipolar OLEDs. In the simulated dark injection transient current a current density rise due to the self-heating occurs at a point in time that is much later than the dark injection charge transit time. Therefore we have successfully and consistently described the effect of self-heating in steady-state, AC and transient operation.

We conclude that in order to get accurate simulation results at typical operating conditions i.e. current densities of organic semiconductor devices it is mandatory to model the heat generation and transport, too.

6. Acknowledgements

E. Knapp acknowledges financial support from the Swiss National Science Foundation for granting a Marie-Heim-Vögtlin Fellowship. The authors would like to thank Martin Neukom from Fluxim for carrying out the measurements.

7. References

[1] N. D. Nguyen, M. Schmeits, and H. P. Loebl, "Determination of charge-carrier transport in organic devices by admittance spectroscopy: Application to hole mobility in α-NPD", Phys. Rev. B **75**, 075307 (2007).

[2] J. M. Montero, J. Bisquert, G. Garcia-Belmonte, E. M. Barea, H. J. Bolink, "Trap-limited mobility in space-charge limited current in organic layers," Organic Electronics 10, 305–312, (2009).

[3] M. Ershov, H. C. Liu, L. Li, M. Buchanan, Z. R. Wasilewski,and A. K. Jonscher, "Negative capacitance effect in semiconductor Devices" IEEE Trans. Electr. Dev., Vol 45, No. 10, (1998).

[4] S.A Butcher, T.L Tansley, D Alexiev, "An instrumental solution to the phenomenon of negative capacitances in semiconductors," Solid-State Electronics, Vol. 39, Issue, 333-336, 3 (1996).

[5] H. C. F. Martens, J. N. Huiberts, and P.W. M. Blom, "Simultaneous measurement of electron and hole mobilities in polymer light-emitting diodes," Appl. Phys. Lett. 77, 1852 (2000).

[6] H. L. Kwok, "Modeling negative capacitance effect in organic polymers,"Solid-State Electron. 47, 1089 (2003).

[7] H. H. P. Gommans, M. Kemerink, and R. A. J. Janssen, "Negative capacitances in low-mobility solids", Phys.Rev. B 72, 235204 (2005).

[8] E. Ehrenfreund et al., „Negative capacitance in organic semiconductor devices: Bipolar injection and charge recombination mechanism", Appl. Phys. Lett.91, 012112 (2007).

[9] G. Garcia-Belmonte, J. Bisquert, P. R. Bueno, and C. F. O. Graeff, "Impedance of carrier injection at the metal-organic interface mediated by surface states in electron-only tris(8-hydoxyquinoline) aluminium (Alq_3) thin layers", Chemical Physics Letters 455, 242-248 (2008)

[10] G. Garcia-Belmonte et al.," Kinetics of interface state-limited hole injection in -naphthylphenylbiphenyl diamine (α -NPD) thin layers," Synthetic Met. (2008)

[11] H. Okumoto and T. Tsutsui,"A source of negative capacitance in organic electronic devices observed by impedance spectroscopy: Self-heating effects," Appl. Phys. Express 7, 061601 (2014).

[12] A. W. J. Gielen et al., "The electro-thermal-mechanical performance of an OLED: A multi-physics model study," Thermal, Mechanical and Multi-Physics simulation and Experiments in Microelectronics and Microsystems, 2009. EuroSimE (2009)

[13] Axel Fischer et al.," Feel the Heat: Nonlinear Electrothermal Feedback in Organic LEDs", Advanced Functional Materials, (2014).

[14] E. Knapp, B. Ruhstaller, "The role of shallow traps in dynamic characterization of organic semiconductor devices", J. Appl. Phys. 112 (2), 024519-024519-6 (2012)

[15] E. Knapp and B. Ruhstaller, "Numerical impedance analysis for organic semiconductors with exponential density of localized states", Appl. Phys. Lett. 99, 093304 (2011)

[16] A. Many and G. Rakavy, "Theory of Transient Space-Charge-Limited Currents in Solids in the Presence of Trapping,"Phys. Rev. 126, 1980 (1962).

[17] Paios instrument provided by Fluxim AG, www.fluxim.com