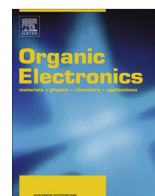




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# Direct determination of doping concentration and built-in voltage from extraction current transients

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## ABSTRACT

We have extended the analytical framework of the charge extraction by linearly increasing voltage (CELIV) theory to the case with doping, taking the effect of built-in voltage, diffusion and band-bending into account. Not taking the built-in voltage into account in dark CELIV will lead to an underestimation of the charge carrier density, while the mobility is overestimated. Furthermore, based on the analysis we propose to use CELIV in the doping-induced capacitive regime for direct determination of doping concentration and built-in voltage from extraction current transients in the time-domain of doped thin-film semiconductor devices. The analytical framework is confirmed numerically with a one-dimensional drift–diffusion model and experimentally on aged P3HT:PCBM bulk heterojunction solar cell devices. An excellent agreement between the experimental extraction current transients and the analytical prediction is found. The presented analytical treatment is not limited to sandwich-type thin-film devices, but is more general and the technique can also be extended to pn-junctions.

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## 1. Introduction

Organic semiconductors, such as conjugated polymers and small molecules, are intrinsically undoped materials. However, it is well known that polymer devices usually suffer from (photo) oxidation in the presence of oxygen and/or water, leading to an increased amount of dark (equilibrium) carriers [1–4]. This unintentional doping is believed to be caused by charged species/defects formed in the device, effectively acting as dopants [1,4–6]. Since unintentional doping of organic electronic devices is typically associated with degraded performance, it is vital to quantify and understand the relevant processes involved in order to increase device performance. Doping of organic materials can also be intentional by introducing electron or hole donating species into the organic layer(s) [7–9]. For

example, by doping separate charge transporting layers in organic solar cells and light emitting diodes charge selectivity can be ensured. Doping has also been utilized to deactivate traps in organic semiconductor devices and to reduce bias stress in organic transistors [10,11].

Experimentally, doping concentrations and built-in voltages are usually extracted from frequency-domain capacitance–voltage measurements using Mott–Schottky (MS) analysis [12,13]. It should be noted, however, that MS analysis was originally derived for infinitely thick devices and has been shown to fail on thin-film (organic) devices if the doping concentration is not high enough [14–16]. Taking this into account, Morfa et al. found a background hole doping concentration of  $N_p = 5 \cdot 10^{15} \text{ cm}^{-3}$  and a built-in voltage of  $U_{bi} = 0.66 \text{ V}$  in thick P3HT:PCBM devices, prepared and measured in nitrogen atmosphere [17]. On the other hand, Seemann et al. noted that by exposing P3HT:PCBM solar cells to oxygen and light for 2 h a doping concentration as high as  $N_p \approx 6.86 \cdot 10^{17} \text{ cm}^{-3}$

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could be determined, while  $U_{bi} = 0.77(\pm 0.02)$  V [4]. The increased density of mobile holes was attributed to immobilized superoxide anions, formed in the active layer.

The charge extraction by linearly increasing voltage (CELIV) method offers the possibility to simultaneously measure both the mobility and the amount of extracted equilibrium carriers and has, due to its experimental simplicity, become one of the most frequently used methods to investigate charge transport and recombination in organic semiconductor devices [18–21]. In the standard dark CELIV setup a ramp up reverse bias is applied over a diode structure, consisting of a (organic) semiconductor sandwiched between two electrodes, and the transient response is measured. The corresponding current transient obtained consists of two parts,

$$j(t) = \Delta j(t) + j_0 \quad (1)$$

a time-dependent extraction current  $\Delta j(t)$  and a geometric capacitive response  $j_0 = \frac{\epsilon \omega_0 A}{d}$ , where  $\epsilon$  is the relative permittivity,  $d$  is the (organic) semiconductor thickness and  $A = U_{max}/t_{pulse}$  is the voltage rise speed (rate of the ramp up pulse),  $U_{max}$  and  $t_{pulse}$  is the amplitude and duration of the ramp up voltage pulse, respectively. A schematic picture is shown in Fig. 1a). In the case of volume extraction,

the mobility  $\mu$  can be estimated from the maximum of  $\Delta j(t)$ , given by [19]

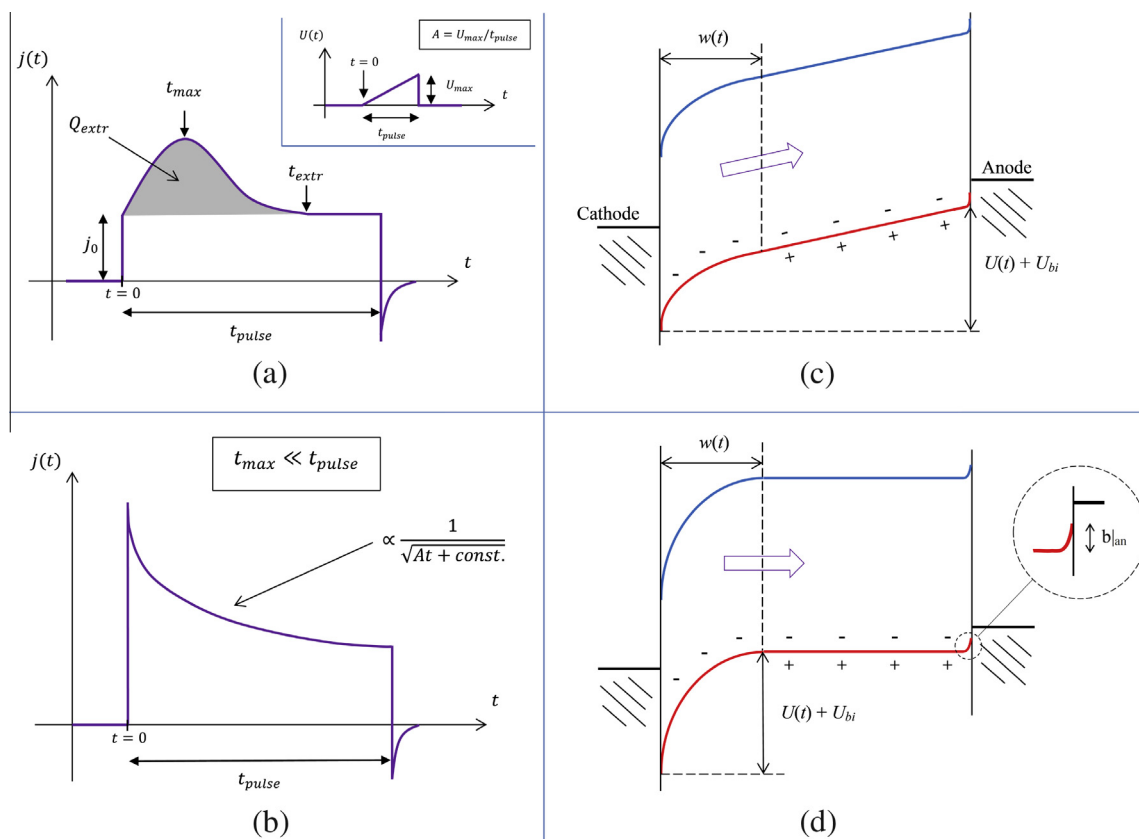
$$t_{max} = \frac{t_{max} |_{\Delta j/j_0=0}}{\sqrt{1 + 0.36 \frac{\Delta j(t_{max})}{j_0}}} \quad (2)$$

where  $t_{max} |_{\Delta j/j_0=0} = \sqrt{\frac{2d^2}{3\mu A}}$  is the exact analytical solution obtained by Juška et al. in the  $\Delta j(t) \ll j_0$ -limit [18]. The second term in the denominator of Eq. (2) is a numerical correction factor. The dark CELIV technique has also been used to estimate the doping concentration from the amount of extracted charge carriers in organic solar cell devices [3,4,22]. The density of mobile charge carriers  $n_{extr}$  is calculated from

$$Q_{extr} = \int_0^{t_{extr}} \Delta j(t) dt = \frac{1}{2} e n_{extr} d \quad (3)$$

where  $t_{extr}$  is the time at which all the carriers have been extracted (see Fig. 1a)).

The underlying CELIV-theory is, however, based on a drift-only model that neglects the influence of the built-in voltage and diffusion. In a doped device with a non-zero built-in voltage, a region depleted of free carriers is present in the semiconductor layer. Consequently, since the



**Fig. 1.** Schematic of the dark CELIV method on a p-doped device, taking the built-in voltage  $U_{bi}$  and the band-bending  $b|_{an}$  at the anode into account. Upon applying the ramp up pulse, holes are extracted at the anode. The standard case, assuming a large  $A$  (fast pulse), is depicted in (a). In (b) the doping-induced capacitive regime, corresponding to the case when a slow enough ramp up voltage pulse is applied such that  $t_{max} \ll t_{pulse}$ , is shown. In (c) and (d) the corresponding energy band diagrams of (a) and (b) are shown, respectively.

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