INVESTIGATION OF TIME-OF-FLIGHT RELIABILITY IN EVALUATING BIMOLECULAR RECOMBINATION IN BULK HETEROJUNCTION DEVICES

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Due to low charge carrier mobilities in disordered organic materials, compared to crystalline semiconductors, the concentration of charge carriers in third generation organic thin film solar cells must be much higher to achieve comparable efficiencies to their non-organic counterparts. Previous studies have demonstrated that for these low-mobility organic materials, the primary mechanism limiting current density is Langevin-type bimolecular recombination. This type of recombination can be reduced by taking advantage of the morphology of bulk heterojunctions (BHJ), which is comprised of bicontinuous interpenetrating networks of acceptor and donor materials, where carriers are spatially separated. This makes the encounter probability for the carriers less likely and reduces Langevin recombination. This in turn makes the reduction factor an important testing parameter to achieve high efficiency organic thin film solar cells.

There are several methods to measure the bimolecular recombination coefficients in BHJ, including Transient Photovoltage (TPV), Transient Photocurrent (TPC) and integral mode Time-Of-Flight (Q-TOF) techniques. TPV and TPC techniques provide valuable insights into charge carrier dynamics by analyzing voltage and current decay after a small perturbation, however their reliance on low-intensity probing makes them susceptible to noise, and their applicability is restricted to open-circuit and short-circuit conditions [1]. Additionally, in multilayered devices, distinguishing capacitance contributions from different layers complicates the analysis, potentially leading to inaccuracies. In contrast, the Q-TOF technique, introduced in [2], can offer more direct measurements of charge transport and recombination in operational multilayered devices under applied electric field that are less affected by multilayer capacitance contributions, making it a more robust method for characterizing carrier dynamics in complex device architectures. In this study we are investigating the necessary experiment conditions introduced in the original paper, including high load resistance, high light intensity and bulk charge carrier generation ($\alpha d \leq 1$), as well as the effect of diffusion and carrier mobility ratio on the calculated bimolecular recombination coefficient value.

In order to better assess the reliability of Q-TOF technique the experiment was numerically modelled as a one-dimensional system by solving Poisson and current continuity equations utilizing the finite difference method. Using these numerical calculations, working limits for Q-TOF technique were set.

Obtained results demonstrated that diffusion lowers the accuracy of the method by approximately 20%, while photogeneration profile had negligible impact on the techniques accuracy (up to $\alpha d = 10$). In addition, different carrier mobility ratios introduce a deviation in calculated bimolecular recombination values.

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