



Towards improved efficiency of bulk-heterojunction solar cells using various spinel ferrite magnetic nanoparticles



Alexander Kovalenko^{a,*}, Raghvendra Singh Yadav^a, Jan Pospisil^a, Oldrich Zmeskal^a, Daniela Karashanova^b, Patricie Heinrichová^a, Martin Vala^a, Jaromir Havlica^a, Martin Weiter^a

^a Brno University of Technology, Faculty of Chemistry, Materials Research Centre, Purkyňova 118, 612 00, Brno, Czech Republic

^b Institute of Optical Materials and Technologies, Bulgarian Academy of Sciences, 109, Acad. G. Bontchev Str., P.O. Box 95, 1113, Sofia, Bulgaria

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ABSTRACT

A detailed study of organic solar cells (OSC) doped with various ferromagnetic and superparamagnetic (Fe_3O_4 , ZnFe_2O_4 , NiFe_2O_4) nanoparticles (MNPs) is presented. Additionally to previously used magnetite nanoparticles, various magnetic moment spinel ferrites were applied. By impedance spectroscopy (IS) analysis it is shown how the doping with various MNPs influences solar cells' performance by the charge carrier effective lifetime extension. In this regard, we introduced a convenient illustrative method to define time constants from the impedance measurements. It is also shown that, photovoltaic performance of the solar cells directly depends on the magnetic moment and alignment of the superparamagnetic single-domain MNPs. Alignment of the MNPs within the OSCs' active layer results in MNPs dipole-dipole interaction, thus further-improves photovoltaic performance due to efficient charge collection at the short-circuit condition. OSC doping with ferromagnetic MNPs showed negative influence on the device performance, however in dark conditions, devices doped with CoFe_2O_4 showed higher forward current presumably due to leakage current through the large MNP aggregation or electron-polaron hopping.

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1. Introduction

Power conversion efficiencies (PCE) of organic solar cells [1–6] seemingly approaching their top empirical formulations to the efficiency limits of 10–12% [7], reporting top PCE values over 10% for single junction polymer [8–11] and over 9% [12,13] for small-molecular solar cells. Localized nature of the electronic states [14] and low dielectric constant ($\epsilon \sim 3$) [15–17] of organic semiconductors result in short exciton length (~ 10 nm) [18] and large Coulombic barrier [19] to dissociate the photo-excited charge carriers. Thus, PCE of organic solar cell strongly depends on the phase morphology in so-called bulk heterojunction (BHJ) [4–6] where two different semiconductors with the offset energy levels forming nanometric domains in the bulk, which is however, a challenging task. The behavior of the organic semiconductors can be hardly predicted in terms of forming interpenetrating phase-separation in BHJ solar cells, and most of the recent records of PCEs directly

associated with BHJ morphology improvement. Another way to improve OSC performance is to enlarge effective lifetime [20] by doping of the photoactive layer with magnetic nanoparticles, thus attain efficient photogenerated charge carriers' collection [21–23].

It has been reported, that there are two possible ways, how doping with MNP improves photovoltaic characteristics: on one hand [23] is a coercive electric field, due to dipole interactions [24] from Fe_3O_4 magnetic nanoparticles (MNPs) dispersed in BHJ layer show potential to strengthen the performance of BHJ polymer-fullerene OSCs. Moreover, alignment of the MNPs within the BHJ layer with an external magnetic field by placing the devices onto the magnet at elevated temperature has shown a further increment of the device performance [23], due to dipole reorientation of the MNPs, thus the coercive electric field of aligned MNPs was augmented. On the other hand it is spin-orbit coupling [20] which increases the efficiency of the exciton intersystem crossing process in the device, and thus extends efficient life-time. This correlates with the previous experiment where the reverse bias was applied to the OSC to create an internal 50–70 V/ μm electric field to ensure efficient charge collection at the short-circuit condition [25,26].

* Corresponding author.

However in conventional BH OSCs, charge collection electrode layers with offset in a work-function, provide a difference of less than 2 eV producing an internal electric field in the range of 20 V/ μm for a 100 nm OSC photoactive layer, which is below the requirements for an effective charge collection [26,27].

In the present paper, the influence of various magnetic nanoparticles, on the performance of organic bulk-heterojunction solar cells is described. Ferromagnetic (CoFe_2O_4) and three types of different superparamagnetic nanoparticles (Fe_3O_4 , ZnFe_2O_4 – high magnetization and NiFe_2O_4 – low magnetization) were dispersed in the blend of $\text{DPP}(\text{TBFu})_2:\text{TC}_{60}\text{BM}$ with 1% $\text{DPP}(\text{TBFu})_2:\text{MNPs}$ weight ratio. Magnetic nanoparticles of spinel ferrites were used in the present paper due to the possibility of fine-tuning the magnetic properties of nanoparticles by chemical manipulations. Generally spinel ferrites are of a great interest in fundamental science, especially for addressing the fundamental relationships between magnetic properties and their crystal chemistry and structure [30]. Synthesized by co-precipitation method spinel ferrite MNPs were stabilized with oleic acid. Here, oleic acid is used as surfactant which provides a steric stabilization of the nanoparticles against the van der Waals and magnetic attraction interactions and thereby prevents agglomeration [28]. This surfactant controls the growth of nanoparticles and prevents the Ostwald ripening process. Also, previous results [29] show that oleic acid capped nanoparticles, are able to prevent water adsorption, oxidation and are capable of being dispersed stably in organic solvents or mineral oils.

2. Experimental section

2.1. Materials

Oleic acid stabilized magnetic iron oxide nanoparticles dispersion in toluene (6.5 nm \pm 3.0 nm Fe-Oxide-Nucleus) (Aldrich) [6,6]-Thienyl C_{61} butyric acid methyl ester (TC_{60}BM) (Aldrich, 99%), chloroform (Aldrich, 99.9%), 3,6-bis[5-(benzofuran-2-yl)thiophen-2-yl]-2,5-bis(2-ethylhexyl)pyrrolo [3,4-c]pyrrole-1,4-dione ($\text{DPP}(\text{TBFu})_2$) and Al (Aldrich, 99.99%) were used as received without further purification. All the materials were weighted in ambient atmosphere and consequently, as an additional drying step, prior dissolving, materials for the active layer, were kept in vacuum chamber at 60 °C overnight, and then transferred to the nitrogen atmosphere. The solution for the photoactive layers was prepared by dissolving $\text{DPP}(\text{TBFu})_2:\text{TC}_{60}\text{BM}$ blend (1.5:1 by weight) in chloroform with a total concentration 20 mg/ml and then placed to the ultrasonic bath for 5 min until complete DPP dissolution. Then, the solution was filtered with 0.45 μm PVDF filter into the vial with MNPs (0.25, 0.5, 1% or 2% of $\text{DPP}(\text{TBFu})_2$ by weight). Subsequently vials were additionally sealed with parafilm and placed in the ultrasonic bath overnight to obtain stable dispersion. All further manipulations were carried out in a glovebox under a nitrogen atmosphere unless otherwise stated.

2.2. Spinel ferrites nanoparticles synthesis

Spinel ferrite AFe_2O_4 ($\text{A} = \text{Co}, \text{Zn}, \text{Ni}$) magnetic nanoparticles were synthesized by co-precipitation method in presence of oleic acid (OA). The stoichiometric amount of $\text{Co}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (or $\text{Ni}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ or $\text{Zn}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$) and $\text{Fe}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ were taken in 1:2 M ratio with distilled water in a beaker. This solution was heated to 50 °C and mixed by the magnetic stirring. A specified amount of oleic acid was added to the abovementioned mixture as a capping agent. A 200 mL of NaOH with 1 M was heated to 100 °C in another beaker. The mixture was added dropwise in NaOH solution at a rate of 4 mL/s with stirring. Within few minute, a precipitate was obtained. The precipitate was kept to a reaction

temperature of 120 °C for 45 min and continuous stirring for 2 h in order to complete the growth process. The obtained precipitate was washed by distilled water and ethanol to remove any impurities present in the precipitate. After washing, the material was dried at 45 °C for 4 h in ambient atmosphere. The obtained sample was designated as $\text{CoFe}_2\text{O}_4/\text{OA}$. Similar above synthesis process was followed for $\text{ZnFe}_2\text{O}_4/\text{OA}$, $\text{NiFe}_2\text{O}_4/\text{OA}$ sample. These obtained samples $\text{CoFe}_2\text{O}_4/\text{oleic acid}$, $\text{ZnFe}_2\text{O}_4/\text{OA}$, $\text{NiFe}_2\text{O}_4/\text{OA}$ were characterized and utilized in PV solar cells.

2.3. Device fabrication

Pre-patterned glass/ITO substrates were preliminary cleaned in ultrasonic bath firstly in the 5% NaOH solution at room temperature, then rinsed in water and consequently washed twice in deionized water and then cleaned in acetone to remove residual water. Finally, ultrasonic treatment in isopropanol bath was applied for 10 min. Following step was the deposition of PEDOT:PSS layer onto glass/ITO substrates. Clevios P PEDOT:PSS was deposited by means of spin-coating at 5500 rpm for 60 s with an immediate annealing for approximately 5–10 min at 150 °C in the ambient conditions followed by transfer to the nitrogen atmosphere and 10 min annealing at 110 °C to remove a residual moisture. $\text{DPP}(\text{TBFu})_2:\text{TC}_{60}\text{BM}$ heterojunction layer was deposited by dynamic spin coating dropping 25 μl of the prepared solution on pre-rotated at 2500 rpm substrates for 40 s. $\text{DPP}(\text{TBFu})_2$ (see Figure S1), which is one of the commonly used small-molecule diketopyrrolopyrrole (DPP) derivatives, was used as an electron donor due to its high reproducibility, high thermal and long-term stability and easy processing. Consequent to the active layer deposition, devices were annealed at 110 °C. Finally, 200 nm Al electrodes were deposited. After that step, to operate the solar cells in the ambient environment, devices were encapsulated with glass slides and Ossila epoxy resin and treated by UV lamp for 30 min. To align magnetic nanoparticles in the active layer ready-made devices (including reference undoped devices) were placed onto the neodymium magnet at 80 °C for 10 min.

2.4. Characterization techniques

Thickness of all the layers measured by the Decktak XT profilometer was in a range of 100 \pm 10 nm. PCE measurement were performed under standard condition by AAA certified Abet Sun solar simulator with an air mass (AM) 1.5G filter. The simulated

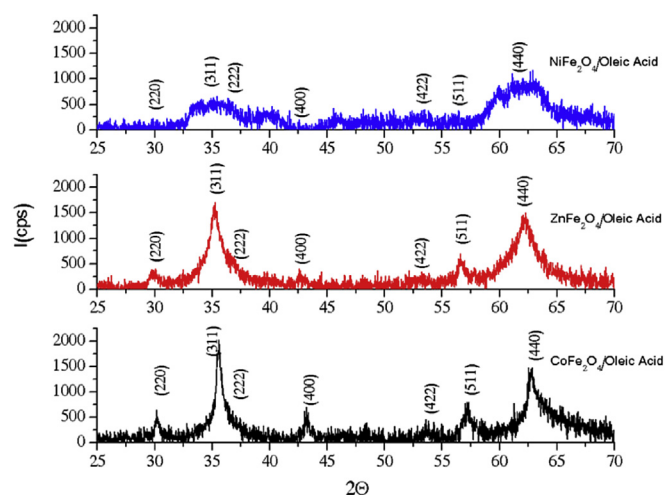


Fig. 1. X-ray Diffraction patterns of CoFe_2O_4 , ZnFe_2O_4 and NiFe_2O_4 nanoparticles synthesized by co-precipitation method in presence of oleic acid.

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