



How do 10-camphorsulfonic acid, silver or aluminum nanoparticles influence optical, electrochemical, electrochromic and photovoltaic properties of air and thermally stable triphenylamine-based polyazomethine with carbazole moieties?



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ABSTRACT

Organic (10-camphorsulfonic acid, CSA), organic-inorganic (Ag-poly(vinylpyrrolidone), Ag-PVP with 10 and 20 nm size of Ag) or inorganic (Al, 18 nm) compounds were applied as new components of active layer in bulk heterojunction polymer solar cells based on a new polyazomethine (PAZ-Car-TPA) resulting in significant change of optical and electrical properties. Moreover, colloidal Ag (100 nm) and Ag-PVP in aqueous solution (10 nm) were tested as an addition to the hole transporting layer based on PEDOT:PSS in polyazomethine solar cells. CSA added to PAZ-Car-TPA decrease its energy gap from 1.91 to 1.20 eV and causes a significant bathochromic shift of the maximum of absorption band along with the change of the polymer color from yellow to red (electrochromic behavior). Photoluminescence maximum of PAZ-Car-TPA protonated with CSA showed 120 nm redshift from 500 to 620 nm in comparison to undoped PAZ-Car-TPA. HOMO-LUMO of PAZ-Car-TPA and its mixture with CSA were analyzed by cyclic voltammetry and quantum mechanical calculations using Density Functional Theory method. Refraction index and extinction coefficient of PAZ-Car-TPA and its mixtures with PC₇₁BM as well as Ag or Al nanoparticles were investigated taking into consideration various thickness of polymer layer. The power conversion efficiency of the ITO/PEDOT:PSS/PAZ-Car-TPA:PCBM:CSA/Al device was five time higher than that of the device based on PAZ-Car-TPA:PCBM. Such an enhancement was found to be primarily due to the increase of the short-circuit current, suggesting that the charge collection increases upon the incorporation of CSA in the active layer. Moreover, presence of Ag-PVP (20 nm) or Al (18 nm) in device increased short circuit current of the constructed polymer solar cells. Additionally, devices were tested by external quantum efficiency measurements and electrochemical impedance spectroscopy in dark and under illumination. The polymer solar cell with PAZ-Car-TPA as donor in active layer showed good stability over 600 hours, when stored in a climatic chamber.

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1. Introduction

The triphenylamine units (TPA) are commonly used in various kind of polymers such as poly(vinylene)s, poly(amide)s, poly(imide)s, poly(azomethine)s, poly(arylate)s, poly(urethane)s and poly(ester)s [1]. New functional materials based on the synergistic

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effect of both of the units in polymer structure are created. The introduction of triphenylamine in polymers and small compounds improves their hole-transporting ability and reduces crystallization propensity [1]. Polyazomethines (PAZ) with TPA moieties in the main or side chain have been investigated mostly as luminescence materials [1–10]. Some scientists have proposed PAZ with TPA units as donor material in bulk heterojunction (BHJ) polymer solar cells [11–18]. The most important parameter of solar cells is their power conversion efficiency (PCE), defined as $PCE = \frac{V_{oc} I_{sc} FF}{P_{in}}$, where V_{oc} is the open circuit voltage, I_{sc} is the short circuit current, FF is the fill factor and P_{in} is the input optical power. The achieved PCE for PAZ with TPA unit has been in the range of 0.02–0.56%, with the highest value found for PAZ with TPA and *p*-terphenyl moieties [18].

Moreover, polymers with carbazole moieties, e.g. poly(2,7-carbazole) derivatives have been also investigated as components of solar cells [19]. Unfortunately, poly(2,7-carbazole) has a relatively large energy gap (E_g approximately 3.0 eV) [19]. However, the E_g of poly(2,7-carbazole) can be reduced by adding an acceptor unit to the structure of polymer to create donor-acceptor polymer.

One of important recent developments in organic solar cells is connected with increased value of PCE by introducing different nanoparticles (NPs). This kind of organic solar cells is classified as the fourth generation of photovoltaic devices and includes application of NPs such as carbon nanotubes, graphene flakes and their chemical modifications, quantum dots, various oxides e.g. TiO_x , MoO_3 , V_2O_5 or ZnO as well as metals (Ag, Au, Pt). NPs can be applied as a component of (i) active layer, (ii) hole transporting layer, (iii) electrodes and (iv) interlayer in organic solar cells [20–31]. In our last work, we tested polymer solar cell with 10-camphorsulfonic acid (CSA) in active layer based on PAZ with vinylene moieties, and received PCE of 0.32% [32].

In this work different architecture of polymer solar cells are analyzed varying the placement and the type of NPs in polymer devices. Below we report on polymer solar cells based on polyazomethine with triphenylamine and carbazole moieties used as a donor, and fullerene derivatives applied as an acceptor. The optical and electrical properties as well as morphology were studied in PAZ-Car-TPA films as a function of PCBM, $PC_{71}BM$, CSA, Al or Ag additives. To the best of our knowledge, this is the first time that polymer solar cells with Al and Ag nanoparticles in active layer based on polyazomethine are analysed with special emphases on the study of refraction index and extinction coefficient.

2. Experimental Section

2.1. Materials and synthesis procedure

4,4'-diformyltriphenylamine, 3,6-diaminocarbazole, *N,N*-dimethylacetamide (DMA), chloroform, 10-camphorsulfonic acid (CSA), poly(3-hexylthiophene) (P3HT), [6,6]-phenyl C_{61} butyric acid methyl ester (PCBM) and Ag NPs stabilized by poly(vinylpyrrolidone) (PVP) in aqueous solution (10 nm) were purchased from Sigma-Aldrich and used as received. Nanopowders of Al (18 nm) and Ag (20 nm) were purchased from US Research Nanomaterials Inc, while Ag (10 nm) nanopowder from nanoComposix Europe. Small silver NPs (Ag, 99.99%, 10 nm) were coated with 97 wt% poly(vinylpyrrolidone) (PVP), while larger ones (Ag, 99.99%, 20 nm) were coated with ~0.2 wt% PVP.

Indium tin oxide (ITO) and poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) with a viscosity of 5–12 cP were purchased from the Ossila. There were six pixels on the ITO glass substrate. The active area of the device was 27 mm². The surface resistance of ITO was approximately 20 ohm/square.

Synthesis of PAZ-Car-TPA and colloidal silver obtained by electrical method is described in the Supplementary information.

2.2. Characterization

The obtained materials were characterized using several techniques.

The UV-vis spectra of the thin films deposited on a glass substrate from a chloroform solution by spin-coating method were recorded on a Jasco V670 spectrophotometer.

Time resolved photoluminescence (PL) was measured at room and liquid helium (4 K). The third harmonic of Ti:sapphire laser (300 nm) was applied for PL excitation. PL kinetics were measured with use of a Hamamatsu streak-camera.

Electrochemical measurements were carried out using Eco Chemie Autolab PGSTAT128n potentiostat with glassy carbon electrode (diam. 2 mm), platinum coil and silver wire as working, auxiliary and reference electrode, respectively. Potentials were referenced with respect to ferrocene (Fc), which was used as the internal standard. Cyclic and differential pulse voltammetry experiments were conducted in a standard one-compartment cell, in CH_2Cl_2 (Carlo Erba, HPLC grade), under argon atmosphere. 0.2 M Bu_4NPF_6 (Aldrich, 99%) was used as the supporting electrolyte.

Refraction index (n) and extinction coefficient (κ) of PAZ-Car-TPA- $PC_{71}BM$ films deposited on SiO_2 covered with 100 nm-thick Al layer were determined in spectroscopic ellipsometry measurements assuming a layered model of the samples with two interfaces: air/polymer blend and polymer/aluminum. Ellipsometric measurements were performed in the spectral range from 0.6 to 5.5 eV (2200–225 nm) for different incidence angles between 40° and 80° using a rotating-analyzer ellipsometer equipped with auto-retarder (V-VASE, J. A. Woollam Co., Inc.). Optical parameters of the 100 nm-thick Al film on SiO_2 were measured a few days after deposition in e-beam evaporator (PVD75, Lesker). The effective optical constants of PAZ-Car-TPA- $PC_{71}BM$ films were determined for each spectral point by the least-squares regression algorithm of the commercial software (WVASE32[®]). The ellipsometric spectra were initially fit using values of the active layer thickness estimated from profilometer measurements. The influence of surface roughness on the optical response of spin-coated polymer films was corrected using the experimental RMS roughness values to model the intermediate layers composed of air/polymer blend and polymer blend/aluminum voids, respectively.

Conductivity (σ) of PAZ-Car-TPA and PAZ-Car-TPA:CSA layers was measured for ITO/PAZ-Car-TPA/Al structure at 25, 50 and 75 °C for 100, 200 and 500 mV bias. The thickness of the PAZ-Car-TPA layer was determined using the AFM technique by measuring the height of the edge made by using a sharp copper blade.

The AFM measurements were performed using intermittent contact mode in air at room temperature (25 °C, RH = 35%). The Innova instrument from Bruker (former Veeco) was used. Typical probes were utilized to perform the measurements (about 40 N/m and <10 nm tip radius).

2.3. Construction and characterization of polymer solar cells

The solar cells were constructed in a glove box on ITO-coated glass substrate with a ITO/PEDOT:PSS/PAZ-Car-TPA:PCBM/Al structure. First, the ITO-coated glass substrate was cleaned with deionized water and then ultrasonicated in isopropanol for approximately 20 minutes. PEDOT:PSS or PEDOT:PSS with 5 μ l of colloidal Ag (or 10 nm Ag-PVP in water solution) were spin cast (2500 rounds per minute, 25 s) from an aqueous solution to form a film on the ITO substrate. Then, a mixture of PAZ-Car-TPA:PCBM (or $PC_{71}BM$) in a chloroform solution with a weight ratio varying from

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