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Diamond-based electrodes for organic photovoltaic devices

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ABSTRACT

The present paper demonstrates the possibility of replacing indium–tin oxide (ITO) with heavily boron-doped diamond (BDD). Plasma Enhanced Chemically Vapor Deposited BDDs layers of various thicknesses were prepared containing various boron concentrations in a gas phase. The dependence of the above-mentioned parameters on the optical and electrical properties of each BDD was studied in order to achieve optimal conditions for the effective application of diamond electrodes in organic electronics as a replacement for ITO. Bulk-heterojunction polymer–fullerene organic solar cells were fabricated to test the potency of BDD application in photovoltaic devices. The obtained results demonstrated the possibility of the aforementioned application. Even though the efficiency of BDD-based devices is lower compared to those using regular ITO-based architecture, the relevant issues were explained.

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1. Introduction

Today, the development potential of organic electronics can hardly be overestimated [1]. In an age of silicon devices, it is worth noting that, although silicon is inexhaustible, its purification, processing and utilization are not as ecologically friendly as could be desired. Organic electronic devices, meanwhile, are beginning to enter many fields of science and technology due to their many clear advantages. Owing to its low production temperatures, organic electronics does not require large energy consumption, which makes such electronics, along with the non-toxicity of its component materials, much more environmentally friendly in comparison with commonly used inorganic semiconductors. Similar to silicon, organic materials are practically unlimited, but much easier to utilize. Specifically, due to their particular properties, they are flexible, adaptive to many types of surfaces, and virtually unbreakable.

In spite of the fact that organic electronics seems to be the technology of the future, some technological steps remain un- or underdeveloped, which significantly thwarts the infusion of this technology. Considering organic solar cells, which seem to have particular potential, one of the limiting factors is the development of transparent electrodes. At first glance, the widely-used compound indium–tin oxide (ITO) seems to be the perfect candidate as a material for transparent electrode, as it exhibits superior transparency and a sheet resistance of below 100 Ω/sq. However, the declining availability of indium compels us to look for an alternative solution.

One possibility is to substitute ITO with carbon-based electrodes such as heavily boron-doped conductive diamond polycrystalline films [2]. Polycrystalline diamond films can be produced by a large variety of chemical deposition techniques, such as hot filament deposition [3], bias enhanced deposition [4], and Plasma-Enhanced Chemical Vapor Deposition (PECVD) with linear antenna delivery, where precursor gases are ionized to enhance their chemical reaction rates [5,6]. One of the advantages of the PECVD technique is the possibility to operate on a large variety of substrates at low deposition temperatures, which can be a crucial condition in the manufacture of semiconductors. With respect to photovoltaic devices, light-harvesting materials can be directly manufactured by the PECVD process [7,8]. Diamond is recognized to be a remarkable material due to its particularly attractive properties combining chemical resistance, optical transparency, thermal conductivity [9–13], and electrochemical properties [14–18]. Once successfully doped, diamond, which is generally recognized as an insulating material, becomes a wide-band gap semiconductor material with excellent potential due to the unique combination of its physical and electronic properties. The boron atom seems to be the only efficient dopant atom in diamond, which can be incorporated with high reproducibility and at a concentration high enough to be useful for

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electronic devices [15–19]. The physical properties of lightly-doped semiconductors are described in terms of band structures and impurity levels – the phenomenon of the formation of an impurity band was observed even at room temperature [17,18]. In the present paper, the possibility of replacing ITO with boron-doped diamond is described.

The addition of boron has a strong influence on the electrical conductivity of diamond layers [19–27]. For moderate boron concentrations – below 3×10^{20} cm^{-3} – standard conductivity values for diamond layers were found [19,28]. Higher boron concentrations typically result in conductive systems with electrical properties comparable to those of metallic materials; also, superconductivity was reported by Ekimov et al. [15] in heavily B-doped diamond. There are also other parameters which influence boron-doped diamond layer charge transport properties – namely, the electronic structures of boron defects, the morphology of the nanodiamond layer (the addition of boron has a strong influence on the morphology of the layers grown [20]), and the relative amount of sp^2 and sp^3 hybridized carbon in the nanodiamond layer [29–31]. The optimization of growth conditions at high boron/carbon ratios (up to 8000 ppm in the gas phase during growth) can lead to low sheet resistance comparable to, or even lower than those of the best ITO samples.

2. Experimental

2.1. Diamond electrodes fabrication

One of the main tasks was the proper patterning of the glass substrates for the subsequent measurements of the solar cells' power conversion efficiency (PCE). Prior to the growth of the diamond layer, glass substrates were screened with a pattern mask, which enabled the area on top of the substrates to be covered with electrode channels 8 mm in width. The substrates were then seeded with a nanodiamond particle solution which was sonicated before seeding in order to break up large clusters in dispersion. The mean size of the nanodiamond seeds in the solution was 5–10 nm as measured by dynamic light scattering (DLS) after sonication. Then, the glass substrates were cleaned in isopropyl alcohol (IPA) and subsequently dipped into the diamond dispersion. In order to produce a monolayer of nanodiamond seeds on the glass substrate surface, the seeding solution was deposited by spin coating for 30 s at 3000 rpm. This procedure resulted in homogenous coverage of the patterned substrates with a nucleation seed density of approximately 10^{10} cm^{-2} as measured by Atomic Force Microscopy (AFM).

After the patterned seeding on glass substrates was prepared, boron-doped diamond (BDD) nanocrystalline films were grown by a chemical vapor deposition technique. A SEKI ASTeX 5010 Microwave Plasma Enhanced CVD reactor was used to grow the BDD layers. Growth was performed in CH_4/H_2 plasma with a respective gas concentration ratio of 5%/95%. Boron doping was achieved by adding trimethylboron (TMB) to the gas mixture. The substrate temperature (700 °C) during the BDD growth process was monitored by a Williamson Pro 92-38 dual-wavelength infrared pyrometer. By varying the B/C concentrations (from 2000 to 8000 ppm) during the growth process, layers with different doping levels were obtained. In order to obtain optically transparent electrodes, BDD layers with various thicknesses were produced.

2.2. Diamond film characterization techniques

Several characterization techniques were applied to investigate the properties of the boron-doped diamond films. In order to reveal the sp^3/sp^2 ratio (the presence of amorphous and graphitic phases) throughout the layer, Raman spectroscopy measurements were carried out. Spectra were taken at room temperature using a Renishaw InVia Raman microscope under the following conditions: wavelength – 488 nm (25 mW); objective – ×50 Olympus; slit size – 65 μm; type of focus – spot focus; grating – 2400 l/mm.

The conductivity and Hall constant were measured by the differential van der Pauw method using a Keithley 6221 current source and two electrometers, a Keithley 6514 with nano-voltmeter, and a Keithley 2182A, which recorded the voltage difference between the electrometers. A pulsed (quasi-DC) measuring mode was used to compensate for parasitic thermoelectric signals. All electrical measurements were performed in dark at room temperature (296 ± 1 K). In all films investigated, the resistivity was measured with an error not exceeding 1–2%, while the Hall constant and mobility were determined with an accuracy of about 5%. For the electrical characterization of BDD films, titanium (20 nm)/gold (100 nm) triangle contacts were evaporated.

Surface roughness, morphology, and film thickness were investigated by AFM using an NTEGRA Prima NT-MDT system under ambient conditions. Samples were scanned using a HA_NC Etalon tip in semi-contact mode. A local contrast (LC) filter was applied to all images to better visualize each film's morphology.

The transmittance spectra of samples were characterized by a Varian Cary Probe 50 UV–vis spectrometer (Agilent Technologies Inc., Santa Clara, CA, U.S.A.). The integral value of transmittance was determined by integrating spectral data in the range 300–850 nm. Commercially available ITO/glass reference substrates were purchased from Sigma-Aldrich. The thickness of the reference ITO samples was 60-100 nm, with an electrical conductivity of ~50 Ω/sq, as measured by a 4 probes method.

2.3. Materials

P3HT (Luminescence Technology Corp.), $PC_{60}BM$ (Solenne, 99%), o-dichlorobenzene (Aldrich, 99.9%), ZnO nanoparticles dispersed in IPA (Gene's Ink), Ca (Aldrich, 99.995%), MoO_3 (Aldrich, 99.98%) and Ag (Aldrich, 99.99%) were used as received without further purification. The active layer films were prepared from a P3HT:$PC_{60}BM$ solution (1:0.8 by weight) in o-dichlorobenzene and were stirred for 12 h at 80 °C until complete dissolution. The concentrations of the pure P3HT and $PC_{60}BM$ solutions in o-dichlorobenzene solvent were 17 mg mL^{-1} and 13.6 mg mL^{-1}, respectively. All manipulations were carried out in a glove box under a nitrogen atmosphere unless otherwise stated.

2.4. Device fabrication

Inverted architecture P3HT:$PC_{60}BM$ devices were fabricated using the regular procedure [32–34]. After boron-doped diamond electrodes were grown, substrates were cleaned ultrasonically in 10% NaOH solution for 5 min, in deionized water for 15 min, and finally in IPA for 10 min to remove residual impurities. The cleaned substrates were covered by a ZnO suspension by spin-coating at 2000 rpm for 40 s at ambient atmosphere, and further annealed at 75 °C for 45 s. Prior to deposition, the ZnO was filtered through a 0.45 mm nylon filter.

100 nm polymer–fullerene heterojunction layers were prepared by spin coating 55 μl of P3HT:$PC_{60}BM$ solution at 1200 rpm for 15 s in a nitrogen atmosphere and then immediately placed into a closed petri dish for 60 min for solvent annealing. The devices were further thermally annealed at 130 °C for 10 min. Top electrodes (7 nm of MoO_3 and the subsequent evaporation of 100 nm of Ag) were deposited by vacuum evaporation.

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