



A preliminary development in hybrid *a*-silicon/polymer solar cells



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ABSTRACT

Amorphous undoped intrinsic silicon, B-doped silicon and P-doped silicon hybrid bilayer structures with poly(3-hexylthiophene) have been fabricated and their photovoltaic responses have been investigated. Open-circuit voltages and fill factors of the devices are moderate, but strongly dependent on the doping type of *a*-Si:H films. The highest available open-circuit voltage from the hybrid solar cells within this investigation is 1.23 V in a standard test condition. Both inorganic and organic semiconducting layers contribute to the photocurrent generation. The short-circuit currents appear to be limited by unbalanced charged carriers collected from different sides of the semiconductors, which indicates a way forward in device optimization.

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1. Introduction

Silicon, especially the single crystal form, has been one of the most successful materials to be used in solar cell application. A single silicon cell reaches 25% of power conversion efficiency (PCE) [1]. However, in order to produce single crystal silicon, an enormous amount of energy is consumed, due to expensive purification process and the layer thickness in hundred microns scale to harvest the light. This clearly contradicts the energy saving ideas of photovoltaic cells and requires an extended period of operation under the sun to repay the energy embedded in the cells. Due to the low cost manufacture, the organic and polymer solar cells may offer a real energy saving technology. Current development shows promising PCE over 10% [1,2]. However, these devices still suffer a low operational stability. There is still a long way towards commercialization and still much work to be done [3,4].

Hybrid organic–inorganic solar cells have received much attention recently due to their combination of high charge-carrier mobility of inorganic semiconductors with strong optical absorption

of organic semiconductors. In recent years, hybrid structures have been investigated for solar cell applications by different research groups. For instance, TiO₂ [5–7], ZnO [5,8,9], CdSe [10–12], GaAs [13,14] or their nano-structured materials [7–10,12–14] have been developed and their preliminary solar cell performance in combination with poly(3-hexylthiophene) (P3HT) [6–10,12–14], poly[2,6-(4,4-bis-(2-ethylhexyl)-4*H*-cyclopenta-[2,1-*b*;3,4-*b'*]dithiophene)-alt-4,7-(2,1,3-benzothiadiazole)] (PCPDTBT) [11], poly(2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylenevinylene) (MEH-PPV) [5] have been reported.

The very first report regarding Si–polymer solar cells was reported by Lewis et al. [15] The work described a device prepared by coating of poly-(CH₃)₃Si-cyclooctatetracene over crystalline Si (*c*-Si) surface with a conversion efficiency of 1.5%. In a later attempt with *c*-Si/conjugated (P3HT), the PCE was improved slightly to 2.46% [16]. A recent composite structure of silicon nanopillar arrays (SiNPs) on *c*-Si substrates with P3HT showed a promising performance (PCE ~ 9.2%) [17]. These works were all based on *c*-Si technology. It is obvious that the efficiencies are still far below that of *c*-Si solar cells.

The more attractive idea is to completely move away from *c*-Si technology. Low cost thin-film silicon technology by plasma enhanced chemical vapour deposition (PECVD) may be an alternative to develop in combination with wet-coating technology for polymer thin films, which have mutually complementary

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properties and especially suitable for cells having flexible substrates or substrates of any shape. It is also expected that the incorporation of polymer in the cell will bring in compatibility with plastic substrates for future flexible devices. The work has both technological and theoretical importance.

Hybrid organic and thin-film silicon devices have also been assessed in a couple of reports for their potential in solar energy application. Gowrishankar et al. [18] and Alet et al. [19] reported hybrid devices formed with amorphous silicon and P3HT heterojunction layers, which revealed their potential in solar energy application. However, the PCEs are low ($\leq 0.2\%$). They are lower than an optimized $p-i-n$ junction amorphous Si device (8–12%) or a typical P3HT:PCBM bulk-heterojunction device (2–6%). Obviously, theoretical challenges and practical problems still remain before hybrid device structures achieve a high enough performance. One may wonder how an excitonic effect in organics or polymers may be associated with the unbound photo electron–hole pairs in an amorphous semiconductor. Obviously, much more work is necessary before reaching a basic understanding.

In an amorphous semiconductor it is currently accepted that the open-circuit voltage depends on (1) the electrostatic field, i.e. the difference between the Fermi level of p layer and n layer in amorphous silicon solar cells ($p-i-n$ type); (2) any variations in electron and hole affinities across the device; (3) any variations in the effective density of states in the two bands; and (4) any variation in the photogenerated electron and hole concentrations across the device (the “Dember effect”). The Dember effect may be significant if the two carrier types have different mobilities, even if the two increased carrier densities are the same. This has been documented by Fonash [20]. In contrast, the source of photovoltage in polymer or organic solar cells is rather poorly understood and is empirically believed to relate to the difference between the HOMO level of the donor and LUMO level of the acceptor [21–23]. How are these levels related to open-circuit voltage in inorganic/organic hybrid solar cells? Underlying physics has yet to be understood as research in this area is rather limited. The investigation of hybrid structure may lead to novel cell structures and a better understanding of their photovoltaic responses for future device development.

The present work reports a preliminary investigation on hybrid solar cells using amorphous Si thin film and polymer. Hydrogenated amorphous silicon (a -Si:H) material was prepared by plasma enhanced chemical vapour deposition (PECVD) to form a hybrid structure with poly(3-hexylthiophene) (P3HT). Double-layered hybrid solar cells with undoped “intrinsic”, or boron doped (B-doped), or phosphorus doped (P-doped) hydrogenated amorphous silicon as the inorganic layer and P3HT as the organic polymer layer have been investigated. Performances and their relation to the morphology of interfacial structure, energy band gaps of intrinsic and doped silicon in these different structures as compared to P3HT have been discussed.

2. Experimental details

2.1. Material and film preparation

Indium Tin Oxide (ITO) coated substrates with a surface resistance of $10 \Omega/\text{sq}$ were etched with hydrochloric acid to make bottom electrode patterns which were then cleaned with acetone, isopropyl alcohol and de-ionized water in ultrasonic bath. Poly(3-hexylthiophene) (P3HT) (electronic grade) with a weight average molecular weight $\overline{M}_W = 39,000$ and a polydispersity of $\overline{M}_W/\overline{M}_N = 3.44$ (GPC, polystyrene standard) was purchased from Lumtec (Luminescence Technology Corp.).

P3HT films were deposited on top of the ITO substrates using spin-coating at different spinning rates to achieve different

Table 1

Experimental conditions in preparation of intrinsic, B-doped (p -type) and P-doped (n -type) a -Si:H layers using RF PECVD.

	SiH ₄ (sccm)	PH ₃ (sccm)	B ₂ H ₆ (sccm)	Pressure (mbar)	RF power (W)	Thickness (nm)
a -Si:H	2.0	0	0	0.5	30	190
a -Si:H,P	2.0	0.12	0	0.5	30	170
a -Si:H,B	2.0	0	0.10	0.5	30	230

thicknesses from a chlorobenzene solution at a concentration of 20 mg/ml. The layers were then annealed for 30 min at 110 °C to remove the solvent.

Hydrogenated amorphous silicon (a -Si:H) was prepared from silane using low power RF excited plasma enhanced chemical vapour deposition (RF PECVD). The intrinsic, B-doped (p -type) and P-doped (n -type) a -Si:H layers were prepared without doping or with the addition of 1% diborane in hydrogen for p -type doping, or 1% phosphine in hydrogen for n -type doping. The RF PECVD deposition was set for ~ 20 min in conditions as given in Table 1. Substrates were heated to 200 °C during the ~ 20 min deposition process, and the thickness of amorphous silicon is about 200 nm in the hybrid devices.

For optical measurement, the intrinsic, p -type or n -type a -Si:H layers were deposited on quartz slides. The devices were prepared directly on top of P3HT films. Finally, a top contact of aluminium was vacuum-evaporated through a metal mask. The active area was 0.9 cm^2 and the final active layers were about 290 nm in total thickness. Hybrid solar cells were prepared with a general structure of ITO/P3HT/Si/Al as shown in Fig. 1. In this work, three hybrid photovoltaic devices were prepared, i.e. intrinsic Device 1: ITO/P3HT/ a -Si:H/Al, p -type (B-doped) Device 2: ITO/P3HT/ a -Si:H,B/Al and n -type (P-doped) Device 3: ITO/P3HT/ a -Si:H,P/Al, and the thickness of P3HT has been varied and optimized.

2.2. Test and measurements

Optical densities of the films were measured using UV–VIS absorption spectra (Shimadzu UV-3101PC spectrophotometer or Perkin Elmer UV/VIS Lambda 950 spectrometer). Infrared absorption spectra of the hybrid structure (P3HT + Si layer), pure P3HT powder, and a -Si:H were measured using a Fourier transform infrared (FTIR) spectrometer (Nicolet 6700) with a total reflection attachment, where the hybrid specimen was removed from the substrate prior to measurement and the a -Si:H specimen was on a quartz substrate. Surface morphology was observed using a Leica DMR microscope equipped with a reflective differential interference contrast mode. A surface profiler (Ambios Technology XP-2) was used to measure thicknesses of the thin films and the devices. Photovoltaic current–voltage characteristics were measured using an I – V curve tracer (EKO Instruments Co Ltd., MP-160), a solar cell characterization system, under standard AM1.5 conditions using a solar simulator (Abet Technologies sun 2000). Photocurrent responsive spectra were taken using Bentham solar

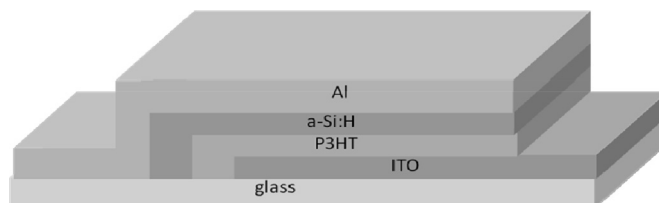


Fig. 1. A schematic diagram of the hybrid structure.

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