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Enhancement of the photoelectric performance in inverted bulk heterojunction solid solar cell with inorganic nanocrystals

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HIGHLIGHTS

- Solid solar cells based on FeS_2 or PbS NCs showed power conversion efficiency (PCE) of 3.0% and 3.11%, respectively.
- The FeS_2 NCs/polymer solar cells showed good time and thermal stability when exposed in air condition.
- Ternary solid solar cells based on PbS NCs exhibited a higher short circuit current density (J_{sc}).

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ABSTRACT

Nanocrystal/polymer solid solar cells have the advantages of low-cost, simple process, and flexible manufacture. In this work, ternary solid solar cells based on FeS_2 and PbS nanocrystals exhibited photovoltaic conversion efficiency of 3.0% and 3.1%, respectively. As a kind of semiconductor with optical absorption in the visible and near-infrared regions, FeS_2 nanocrystals matched well with the solar radiation spectrum. Furthermore, PbS Nanocrystals could increase the number of electrons, due to its multiple exciton effect. Additionally, the FeS_2 nanocrystals solar cells showed high stability, with 83.3% of its initial efficiency remained after 15 weeks of exposure in air, and kept good stable performance at 20–80 °C. The photovoltaic conversion efficiency fluctuation magnitudes were also found to be smaller than quantum-dot sensitized solar cell under the same conditions.

1. Introduction

Renewable energy is regarded as an essential supplement to the fossil fuels. Among the various renewable energy resources such as solar energy, wind energy, geothermal energy, biological energy, hydropower, hydrogen energy and wave energy [1], solar energy with the excellent advantages of abundance and non-polluting nature is supposed to be a great potential candidate [2]. Exploitation of solar energy is aimed to convert sunlight/solar radiation into electricity via photovoltaic devices like solar cells. In 1954, Yamaguchi et al. [3] successfully invented a silicon solar cell with a photovoltaic conversion efficiency (PCE) of 6%, which was regarded as the first generation of solar cells. The second generation of solar cell, the copper indium gallium selenium (CuInGaSe) thin film solar cell, which was developed in early 1989 with its PCE reached up to 13% [4]. However, the further development of these two generation solar cells were restricted owing to the high cost of pure silicon and pollution issues. In recent years, Ito et al. [5] reported a bifacial dye-sensitized solar cell structure with a porous TiO_2 layer that provided high photo-energy conversion efficiency. Near infrared absorption of $CdSe_xTe_{1-x}$ alloyed quantum dot [6] was used to prepare the quantum dot-sensitized solar cells (QDSSCs). Polymer solar cells based on P3HT, porphyrin-modified ZnO nanorods [7] and self-assembled monolayers [8] have been extensively investigated. Nevertheless, DSSCs and QDSSCs show poor stability since their liquid electrolyte leaks, and polymer solar cells with low efficiency for the slow electron transmission. Solid cells combined semiconductor NCs and polymers are one of the most promising alternatives to conventional silicon solar cells.

In the system of NCs/polymer solid solar cells, both the semiconductor NCs and the polymers were served as sunlight absorbers and exciton generators as well as electron donor. Electron transmission was much faster in NCs than that in polymers. By tuning the size, composition and shape of NCs, the absorption wavelength could be matched with the solar radiation spectrum. Moreover,

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Nomenclature

Abbreviation

PCE	photovoltaic conversion efficiency
NC	nanocrystal
DSSC	dye-sensitized solar cell
QDSSC	quantum dot-sensitized solar cell
FF	fill factor
LUMO	lowest unoccupied molecular orbital
P3HT	poly (3-hexylthiophene-2,5-diyl)
PCBM	[6,6]-phenyl C61 butyric acid methyl ester
PEDOT:PSS	poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate)
PbS	lead (II) sulfide
FeS_2	ferrous disulfide

Symbols

V_{oc}	open voltage
J_{sc}	short circuit current density
J–V	current density versus voltage
P_{in}	incident irradiation intensity
M_n	molecular weight

Greek symbol

α	photovoltaic conversion efficiency fluctuations coefficient
η	photovoltaic conversion efficiency

polymer materials like P3HT have achieved great progress in stability and overcome the disadvantages of materials such as polyurethane benzene phosphite vinyl (PPV), whose branching chains are prone to oxidetively decompose under illumination. Solid thin solar cells were prepared with materials like P3HT-capped CdSe superstructures [9], which could enhance the light harvest ability of active layer. TiO_2 NCs as photoelectrode was modified by metal-free organic dye [10]. Thus, combining the strengths of NCs and polymers showed great potential for achieving high efficiency and stability.

In the past five years, Yuan et al. [11] have conducted lots of research on the fabrication of pure and high-quality FeS_2 NCs for solar cells via facile solvothermal and hydrothermal reaction processes. Compared with the typical CdSe, CdTe, and CdS NCs materials, FeS_2 NCs were not only nontoxic, but also of low band gap (about 1.0 eV) and cost performance. Somnath et al. [12] reported inorganic solar cell based on ITO/PEDOT:PSS/MEH-PPV:FeS_2/Al structure, which had the PCE of 0.064% and V_{oc} of 0.72 V. Bi et al. [13] used a kind of FeS_2 NCs to improve the stability and photo response in an ITO/FeS_2 NCs/Al device. Lin et al. [14] enhanced solar cell performance by adding FeS_2 NCs into P3HT:PCBM and a PCE of 2.3% was achieved. Because of the low band gap, variable sizes and shapes of PbS NCs, which showed potentially applicable in solar cells [15].

In this study, the ternary inverted bulk heterojunction solar cells with structure: FTO/TiO_2/P3HT:PCBM:NCs/PEDOT:PSS/Ag were fabricated. The performance of devices was measured with a Keithley 2400 source measurement unit and a solar simulator. PbS and FeS_2 NCs were used to add into the active layer with different concentration, respectively. TiO_2 and PEDOT:PSS layer were introduced to act as the electron selective layer and hole selective layer, respectively. The results showed that FeS_2 NCs and PbS NCs in active layer could enhance the performance of devices, and a high efficiency of 3.11% and 3.0% were achieved, respectively. The FeS_2 ternary solar cell showed high stability in long-term and high temperature environment, and PCE fluctuations of FeS_2 NCs/polymer solar cell were smaller than QDSSCs under different irradiation intensities. These results indicated that the moderate NCs could improve the performance of inverted bulk heterojunction solid solar cells by improving the conductivity of the device.

2. Experimental

2.1. Materials and preparation of FeS_2 and PbS NCs

[6,6]-Phenyl C61-butyric acid methyl ester (PCBM) (99.5%) was purchased from Nano-C, USA. Two kinds of molecular weight (15,000–45,000, and 54,000–75,000) of regioregular poly (3-hexylthiophene-2,5-diyl) (P3HT) (90%) were purchased from Sigma Aldrich, USA. Poly (3,4ethylenedioxythiophene)-poly (styrenesulfonate) (PEDOT:PSS) was purchased from Sigma Aldrich. All materials were used as received without any further purification. FeS_2 NCs were synthesized via a solvothermal reaction process which was similar with the literature [11]. The as-prepared FeS_2 NCs were dispersed in chloroform and washed with excess acetone to remove organic impurities before using. High-quality PbS NCs was synthesized by the aqueous synthesis method [15], then dispersed in acetone, and washed with excess ethanol before using.

2.2. Active layer solution preparation

The active polymer solution was prepared by dissolving 40 mg P3HT and 40 mg PCBM in 2 mL chlorobenzene. Subsequently, a certain amount of FeS_2 or PbS NCs was added to prepare active layers solutions with different concentrations. The high and small M_n of P3HT were used to prepare PbS NCs and FeS_2 NCs/polymer solar cells, respectively. The hybrid solutions were treated with ultrasonication for more than 1 h to ensure that the NCs were uniformly dispersed, and the hybrid solution was placed for 24 h at room temperature.

2.3. Experimental setup and device fabrication

The NCs/polymer solar cells were prepared with an inverted structure, which was shown in Fig. 1. Where TiO_2 and PEDOT:PSS acted as the electron selective layer and hole selective layer, respectively. The preparation of NCs/polymer solar cells processes were shown in Fig. 2, firstly, the glass substrate pre-coated with

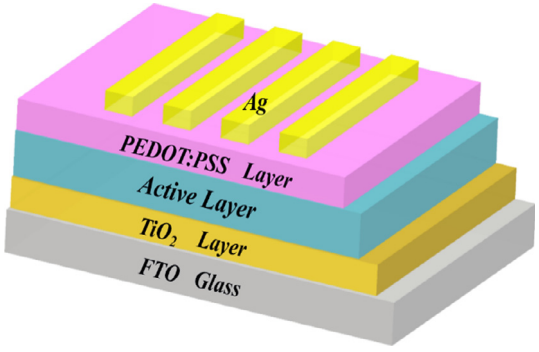

Fig. 1. Schematic architecture of the hybrid inverted solar cell.

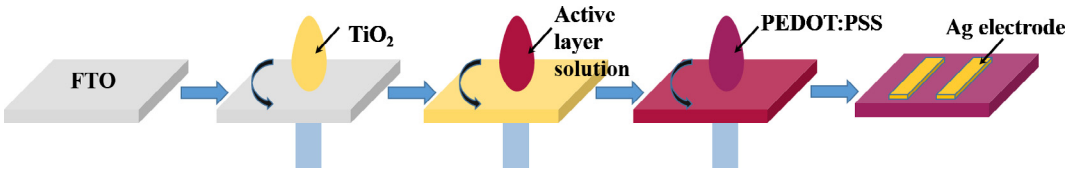

Fig. 2. Schematic diagram of the hybrid inverted solar cell preparation process.

fluorine tin oxide (FTO) (with size of 2 cm × 2 cm, 15 Ω/square) was cleaned in an ultrasonic bath with deionized water, acetone and isopropyl alcohol for 15 min, respectively. Then it was dried in a drying oven. Secondly, TiO_2 solution was spin-coated with 5000 rpm onto the FTO substrate, and sintered at 500 °C for 1 h. Thirdly, the active layer solution was spin-coated on the top of TiO_2 layer, and the blend films were placed on a hot plate and annealed at the temperature of 150 °C for 15 min. Subsequently, the PEDOT:PSS layer was spin-coated on the active layer and annealed at 160 °C for 5 min. Finally the Ag electrode was prepared by thermal evaporation as the counter electrode with an active area of 0.1 cm^2.

2.4. Characterization

The current density versus voltage (*J–V*) characteristic of the solar cells was measured with a Keithley 2400 source measurement unit and a solar simulator (Abet Technologies, USA) under AM 1.5 (1000 W/m^2) irradiation. The power conversion efficiency of solar cells could be calculated by Eq. (1), where V_{oc}, J_{sc}, *FF*, P_{in} are open voltage, short current density, fill factor and the incident irradiation intensity, respectively. The time stability of the solar cell was evaluated by continuous *J–V* measurement for 15 weeks. The thermal stability of the solar cells was investigated by putting devices in different temperature from 20 to 80 °C and the *J–V* characteristic was recorded. Additionally, the magnitudes of PCE fluctuation in a different irradiation intensity from 200 W/m^2 to 1500 W/m^2 were calculated. Surface morphology of active layer films were characterized by using atomic force microscope (AFM, Veeco) in non-contact mode.

$$\eta = \frac{V_{oc} J_{sc} FF}{P_{in}} \times 100\% \quad (1)$$

3. Results and discussion

3.1. The effects of FeS_2 and PbS NCs in the solid solar cell

The *J–V* characteristics of solar cells which were prepared with different concentrations of FeS_2 are showed in Fig. 3(a). The open voltages (V_{oc}) of all the prepared devices were held around 0.6 V. Moreover, obvious enhancement in short circuit current density (J_{sc}) was observed at the FeS_2 concentration of 1.25 mg/mL. In comparison, the best PbS NC cell exhibited a short circuit current density (J_{sc}) of 16.03 mA/cm^2, an open circuit voltage (V_{oc}) of 0.56 V, and a fill factor (FF) of 34.63%. The efficiency of 3.11% was obtained at a PbS NC concentration of 2.4 mg/mL. Fig. 3(b) shows the *J–V* characteristics of PbS solar cells with different PbS NC concentrations.

The performance of solar cells with different concentrations of high quality FeS_2 NCs are listed in Table 1. According to the results, the addition of FeS_2 NCs from 0 to 1.25 mg/mL increased the J_{sc} from 8.84 to 10.11 mA/cm^2, which resulted in the improvement of the PCE from 2.8% to 3.0%. The performance of device was higher than the literature reported [14]. This was attributed to the improved light harvesting capability of the FeS_2 NCs in the solar

Table 1
Photovoltaic parameters of solid solar cells fabricated with different concentrations of FeS_2 NCs.

Concentration (mg/mL)	J_{sc} (mA/cm^2)	V_{oc} (V)	FF (%)	PCE (%)
0	8.84	0.61	51.9	2.8
1.25	10.11	0.60	50.2	3.0
2.5	9.12	0.59	47.6	2.6
3.75	8.51	0.59	48.5	2.4
5	8.10	0.58	44.3	2.1
6.25	7.91	0.58	40.7	1.9
7.5	7.84	0.59	41.5	1.9

Table 2
Photovoltaic parameters of solid solar cells fabricated with different concentrations of PbS NCs.

Concentration (mg/mL)	J_{sc} (mA/cm^2)	V_{oc} (V)	FF (%)	PCE (%)
0	16.17	0.553	33.91	3.03
0.975	15.53	0.565	33.76	2.97
1.6	14.56	0.566	34.32	2.83
2	14.67	0.563	34.94	2.89
2.4	16.03	0.560	34.63	3.11
3	13.50	0.565	34.62	2.64
3.5	16.38	0.562	32.48	2.99

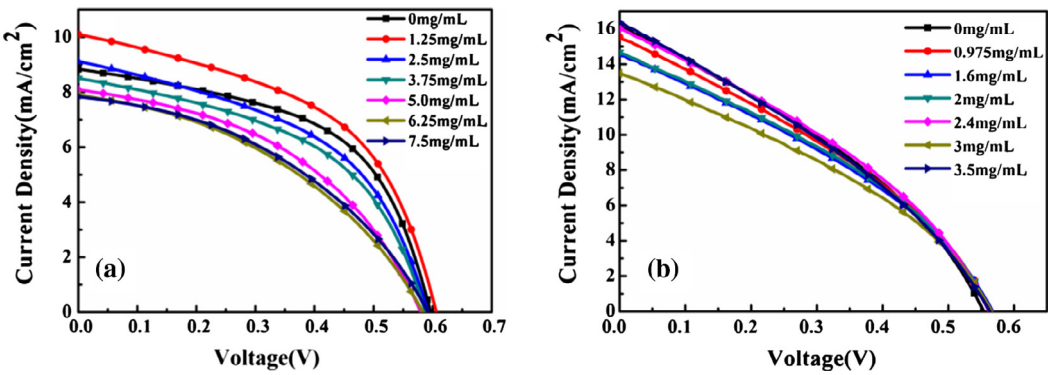

Fig. 3. *J–V* characteristics of fabricated solar cells containing different concentrations of (a) FeS_2 NCs, (b) PbS NCs.

cell. However, when the NCs concentration was further increased from 2.5 mg/mL to 7.5 mg/mL, the J_{sc} showed a decreasing trend, and the fill factor (*FF*) and the PCE also went down.

The devices performance with different concentrations of high quality PbS NCs are listed in Table 2. Compared with FeS_2-based solar cells, the PbS NCs/polymer solar cells possessed relatively higher J_{sc}, PCE and lower FF values. The V_{oc} values of both two kinds of solar cells were basically flat. With the introduction of low concentration (0.975 mg/mL) of PbS NCs, a decreased PCE was found. It was because the nanocrystal surface defect effect was greater than the multiple exciton effects [16]. With the increase of the concentration, the multiple exciton effects gradually dominated the performance of device. However, too much PbS NCs addition would influence the performance of film, accordingly, the PCE of the PbS NCs/polymer solar cells firstly increased and then declined. The best PCE of 3.11% was obtained at a PbS NCs concen-

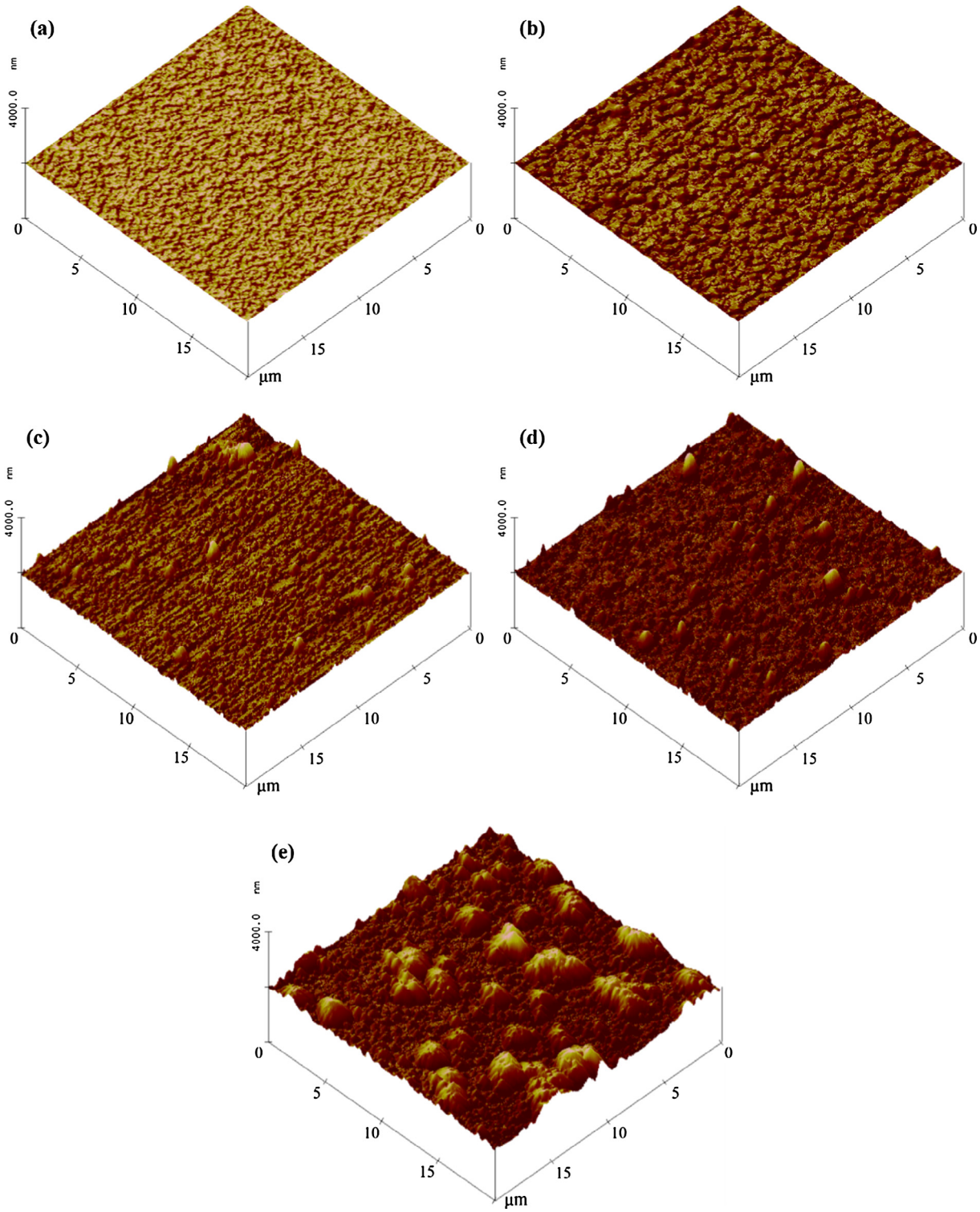

Fig. 4. AFM image of P3HT:PCBM thin films with different addition amount of FeS_2 NCs (a) 0 mg/mL, (b) 1.25 mg/mL, (c) 2.5 mg/mL, (d) 3.75 mg/mL.

tration of 2.4 mg/mL. Comparing with the non-PbS NCs organic solar cell, the power conversion efficiency of the cells with PbS NCs was improved.

In order to further analyze the effect of FeS_2 NCs, AFM was used to observe the evolution of morphology of P3HT:PCBM:FeS_2 thin films. Blend films with different addition amount of FeS_2 NCs were placed on a hot plate and annealed at the temperature of 150 °C for 15 min, after annealing treatment, the blend films not only removed excess organic solvent to increase the degree of phase separation, but also decreased the roughness of the film, the corresponding results are presented in Fig. 4. The AFM images show the 3-D topography and the roughness of active layer films. The r.m.s roughness of 0 mg/mL film was 5.338 nm, and the r.m.s roughness of 1.25 mg/mL film, 2.5 mg/mL film, 3.75 mg/mL and 5 mg/mL film were 14.120 nm, 50.068 nm, 70.157 nm, and 165.510 nm, respectively. The texture of 0 mg/mL film was much smoother than other films. However, the best device was obtained with a small amount of 1.25 mg/mL NC added into the active layer, the r.m.s roughness of film increased a little. Due to the low extent of surface roughness increase, the charge transport distance would effectively reduce and the internal reflection of light and light absorption would dramatically enhance. Thus, the short current density and the performance of device had also increased [17]. When more NCs added, the performance of the blend films would degraded, which was due to the defects in thin film increased and the charge was captured rather than to the corresponding electrode. The results indicated that a certain amount of NCs could help to increase the efficiency of the device.

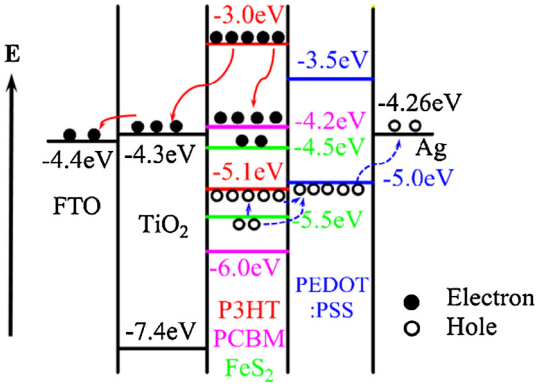

Fig. 5. Schematic diagram of the energy levels in the FeS_2 NCs/polymer solar cells.

Table 3
Photovoltaic parameters of FeS_2 NC/polymer solar cell after different operating time.

Duration time (week)	J_{sc} (mA/cm^2)	V_{oc} (V)	*FF* (%)	PCE (%)
0	10.11	0.60	50.2	3.0
1	9.66	0.60	50.3	2.9
2	9.28	0.59	48.8	2.7
3	8.84	0.60	53.1	2.8
5	8.71	0.60	52.4	2.7
7	8.56	0.59	52.5	2.7
10	8.45	0.59	52.5	2.6
15	8.19	0.60	50.6	2.5

In the irradiation condition, both the polymer P3HT and FeS_2 NCs could generate excitons. The excitons separated into electrons and holes at the donor–acceptor interfaces. As shown in Fig. 5, electrons were able to transfer from P3HT to TiO_2 and PCBM. Because the LUMO of FeS_2 (−4.5 eV) was lower than that of TiO_2 (−4.3 eV), the electrons generated by FeS_2 NCs could not reach TiO_2 layer, which resulted in a lower efficiency of electron collection. Holes transportation functioned inversely, the holes were generated by the FeS_2 NCs could be transferred to PEDOT:PSS and P3HT, which means that many more holes could be converted into photocurrent. When the solid solar cell contained fewer FeS_2

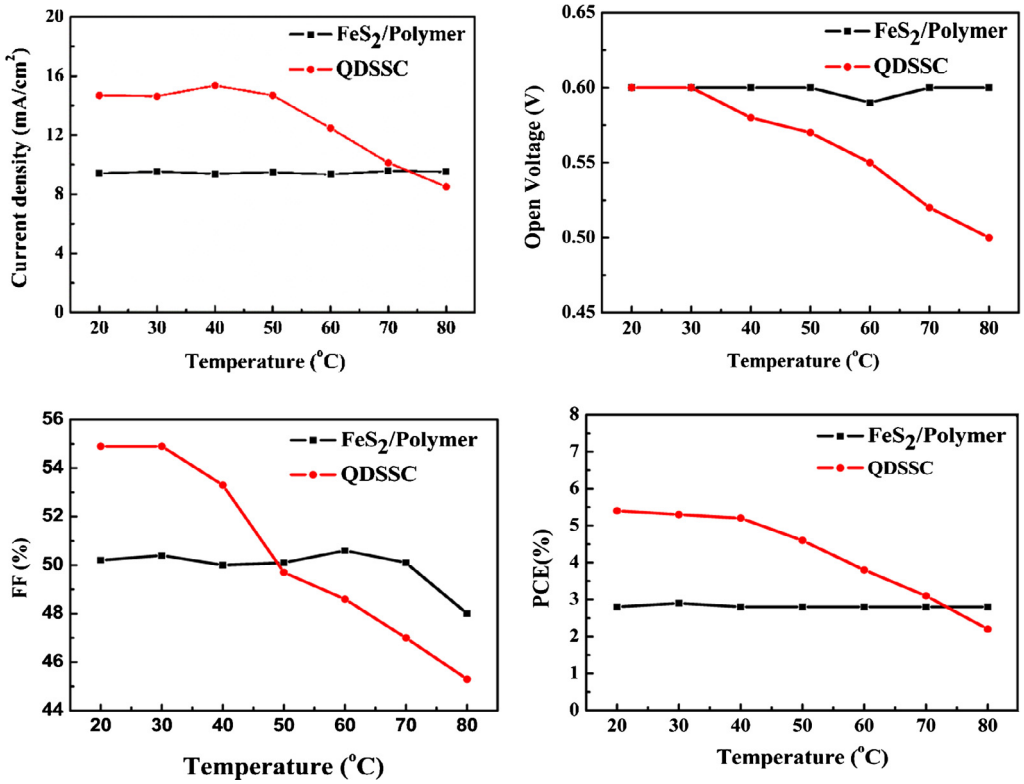

Fig. 6. Thermal stability comparison of FeS_2 NC/polymer solar cell and QDSSC.

NCs (i.e., 1.25 mg/mL), the FeS_2 NCs advantages of near-infrared absorption and rapid transmission of carriers were greater than its deficiency in electronic transmission. Therefore, the FeS_2 NCs/polymer solar cell displayed better performance than the polymer solar cell. When the concentration of FeS_2 NCs in the solar cell was higher, its deficiency was more dominant than its advantages, resulting in a large number of electrons unable to transfer. Thus, J_{sc} decreased gradually, which would also cause the PCE to decline. This is the reason why the present solid solar cells based on FeS_2 NCs exhibited the optimized efficiency at a certain concentration of FeS_2.

3.2. Time and thermal stabilities of FeS_2 NC/polymer solar cell

Although there are many reports on the high PCE of QDSSCs, the liquid electrolyte systems of QDSSCs used to have a common and severe problem of leakage that lead to a short working life. The time stability and thermal stability of the NCs/polymer solar cell with a FeS_2 NCs concentration of 1.25 mg/mL were investigated. The solar cells were stored in a petri dish at the ambient temperature without further seal. The results are summarized in Table 3. During the measurement period of 15 weeks, the J_{sc} of the cell decreased gradually from 10.11 mA/cm^2 to 8.19 mA/cm^2. The PCE declined from 3.0% to 2.5%. That means 83.3% of the original efficiency of the cell remained after 15 weeks. Compared with other literatures [12,18], the FeS_2 NCs solid solar cells exhibited higher time stability.

The thermal stability of the cell in the range of 20–80 °C is illustrated in Fig. 6. Fig. 6 showed that only the FF of FeS_2 NCs/polymer solar cell slightly decreased when the temperature raised from 70 °C to 80 °C. The J_{sc}, V_{oc}, and PCE values of the FeS_2 NCs/polymer solar cell almost remained the same at every temperature, while those of the QDSSCs showed a sharp decline with increasing temperature. Because of its liquid electrolyte, the QDSSCs was more suitable for operation between 20 and 40 °C. While the FeS_2 NCs/polymer solar cell displayed a wider working temperature range from 20 °C to 80 °C and an excellent stability.

3.3. PCE fluctuation of fabricated solar cells under different irradiation intensity

Irradiation intensity is not constant during the daytime. Generally, the standard irradiation intensity was 1000 W/m^2, which was corresponded to the standard PCE (η_{1000}) value of 2.4% in this paper. The influence of irradiation intensity changing from 200 W/m^2 to 1500 W/m^2 was investigated. The device could absorb more light and generate more excitons with increasing light intensity. It is the reason why the PCE was found to be proportional to the light intensity (as shown in Table 4). The prepared solar cell of PCE fluctuations coefficient (α) was calculated and the fluctuation coefficient (α) was described as Eq. (2). Comparing with that of the QDSSCs, the PCE fluctuation magnitudes of the QDSSCs were always larger than those of the FeS_2 NCs/polymer solar cell at different irradiation intensity.

$$\alpha = \frac{\eta - \eta_{1000}}{\eta_{1000}} \times 100\% \quad (2)$$

In summary, the NCs/polymer solar cells showed better photovoltaic conversion efficiency, which was affected slightly by irradiation intensity, and almost had no influence with temperature. Meanwhile, the substrate FTO glass can be replaced by flexible substrate, and it makes the solar cell a flexibility characteristic. Thus, the solar cells can be used in the solar backpack, solar cars, building integrated photovoltaic (BIPV) and so on. Due to these characteristics, the solar cells can inherit in the windows, roof, and wall or inside the wall.

Photovoltaic cells based on polymer and inorganic nanoparticles bulk heterojunction composites, which have the potential for renewable energy resources. it is because not only their lightness, low cost and simple fabrication processing in large area, but also the electron transport in nanoparticles is indeed faster than the electron mobility in polymers. The inorganic nanoparticles usually have higher holes transport properties, at the same time, it can also improve the light absorption in the active layer due to the specific advantage for light harvesting. Hybrid solar cells based organic/inorganic has gained a lot of attention in market of solar cells, but there still have many problem to solve, such as low efficiency, suitable material, and inorganic nanoparticles this area still needs to be explored deeply.

Table 4
Comparison between PCE fluctuation magnitudes of FeS_2 NC/polymer solar cell and QDSSCs.

Intensity (W/m^2)	FeS_2 NC/polymer solar cell		QDSSCs	
	PCE (%)	α (%)	PCE (%)	α (%)
200	1.0	−58.3	1.8	−66.0
350	1.5	−37.5	3.1	−41.5
500	1.9	−20.8	4.1	−22.6
800	2.2	−8.3	4.8	−9.4
1000	2.4	0.0	5.3	0.0
1200	2.5	4.2	5.8	9.4
1500	2.7	12.5	6.3	18.9

4. Conclusion

FeS_2 or PbS NCs based solid solar cells were prepared with an inverted structure. The addition of NCs into polymer solar cells resulted in an enhanced light harvesting ability and a correspondingly obvious increased in J_{sc}. High loading of FeS_2 NCs or PbS NCs were found to reduce electron transmission ability. The solid solar cell exhibited an optimal PCE of 3.0% at a FeS_2 NC concentration of 1.25 mg/mL, and a PCE of 3.0% with a PbS NCs concentration of 2.4 mg/mL. The optimized FeS_2 NCs-based solar cell kept high stability 83.3% of its initial efficiency after exposing in air for 15 weeks. And the device exhibited good stable performance between 20 °C and 80 °C. Furthermore, the PCE fluctuation magnitudes of the cells showed better than those of a QDSSC under the same conditions.

Acknowledgements

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