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Synthesis and processing of strong light absorbent iron pyrite quantum dots in polymer matrix for efficiency enhancement of bulk-heterojunction solar cell



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ABSTRACT

Light absorbent FeS₂ quantum dots (QDs) with an average size of ~5 nm were synthesized by the colloidal method using a dodecylamine as a solvent and structure directing ligand at 215 °C in a Schlenk flask. X-ray diffraction pattern and UV–vis spectra analysis show a phase pure pyrite with a band gap of 0.99 eV. These QDs were blended in a polymer matrix comprising of poly(3-hexylthiophene-2,5-diyl):[6,6]-phenyl-C61-butyric acid methyl ester (P3HT:PCBM, 10 mg:10 mg) in bulk-heterojunction solar cell with loading of 10 wt%, 20 wt%, and 40 wt% of P3HT in order to harness excess light to enhance the efficiency and balance carrier mobility in our quest to devise efficient solar cells. An enhanced efficiency of 3.62% with the 20 wt% of QDs addition was achieved from the normal BHJ device of efficiency 2.32%. However, addition of 40 wt% QDs shows a decreased efficiency of 1.77%. The extended light harvest induced electronic mobility and reduced recombination of electron-hole may contribute to higher efficiencies. The morphology and optoelectronic properties are characterized by TEM, XRD, UV–vis, EQE and I–V curve techniques.

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1. Introduction

Semiconducting nanocrystals exhibit quantum confinement when the dimension (1–100 nm) of confinement is less than the de Broglie wavelength; then the crystal possesses strong size dependent optoelectronic properties enabling them to utilize in myriad of fields such as photovoltaics, photocatalysis, biological labeling, sensors, light emitting diodes and lasers [1–4]. At present, the harnessing of solar light is quite low which amounts less than 0.1% of the global energy demands [5]. The major glitch for the large-scale utilization of solar energy lies in their high cost and inadequate efficiencies of the existing solar cells. New strategies and innovations are needed to harvest maximum incident solar photons with greater efficiency and economic viability [6,7]. Recently, iron pyrite (FeS₂, fool's gold) is getting plenty of interest owing to its promising attributes such as strong light absorption, abundant in existence, non-toxicity, high absorption coefficient ($5 \times 10^5 \text{ cm}^{-1}$ for $h\nu > 1.3 \text{ eV}$) and a very suitable band gap (0.95 eV) [8,9] which may translate to an estimated 0.000002 c/W of material extraction cost. It was placed by Wadia et al. in the

top 23 promising potential candidates for future solar electricity generation [10].

Generally, a band gap of 1.1 eV is required to get 77% cover of the AM1.5 (air mass) solar photon flux (assuming complete absorption of the solar emission intensity), [11] whereas most solution processable semiconducting polymers (PPVs, poly(3-hexylthiophene) (P3HT)) have bandgaps larger than 1.9 eV, [12] which covers just 30% of the AM1.5 solar photon flux. Moreover, the short carrier diffusion length (1–10 nm) in bulk-heterojunction solar cells inhibits efficient utilization of photogenerated electron–hole (e^-h^+) [13]. Due to the low charge-carrier mobility of the polymers, increased thickness of the active layer is required, which, in turn, results in an absorption of only ~60% of the incident light. To achieve a substantial energy conversion efficiency, strong light absorption in visible to infrared spectra and the efficient utilization of excited electron–hole pairs are needed. Several kinds of additives in bulk heterojunction (BHJ) solar cells have been already reported in the literature such as alkanedithiol [14], solution based TiO_x [15], and nitrobenzene [16] to improve the device performances. However, controlled addition of soluble inorganic quantum dots (FeS₂) in matrix of active layer is not studied so far, and most of the existing studies are focused on the hybrid type solar cell by mixing a p-type P3HT polymer and n-type inorganic structures (CdSe, CdS, CIS, etc.) owing to their high absorption coverage in visible and infrared regions and induced charge mobility [17].

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For a pure n-type fullerene (PCBM) the electron mobility ($2.0 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) was found to be ~ 4000 times higher than those of hole mobility (PPV, P3HT) [18,19]. Since the FeS_2 synthesized at low temperature is reported to be a p-type semiconducting material [20], an important question is whether the mobility can be modified in addition to the light coverage. In this aspect, we devised a novel method of blending a composite of p-type conjugated polymers (P3HT) and controlled addition of pyrite QDs to modify electronic interaction besides the mechanical reinforcement of the active layer film. The rationale behind this approach is to utilize the extended wavelength absorption by QDs in conjunction to polymeric properties, which might ballistically transfer the generated excitons to boost up the device efficiencies. Here we report a pure iron pyrite quantum dot with excellent light absorption coverage as an additive in the active layer matrix of BHJ solar cell to enhance the efficiency up to 3.62%.

2. Experimental

Synthesis of FeS_2 QDs nanocrystals: Iron pyrites (FeS_2) quantum dot was prepared by colloidal synthetic route. In a typical synthesis, 20 ml of dodecylamine (90%, Showa Chemicals) is injected in a 100 ml Schlenk flask, then 500 mg of $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$ (Aldrich) was mixed into it in the argon atmosphere with a mild stirring at 95°C for 2 h to degas it and then in another Schlenk flask 500 mg of sulfur (99.9%, Sigma-Aldrich) was mixed in 20 ml of dodecylamine (Showa chemicals) and degassed in argon atmosphere at 95°C for 2 h. Then it was injected into the first flask at a temperature of 215°C ; the reaction was carried out for 1 h with vigorous stirring. Then few drops of chloroform (Showa Chemicals) were added to prevent coagulation of FeS_2 at suitable temperature and lastly it was washed with methanol (99.5%, Showa Chemicals) four times using a centrifuge at 4000 rpm for 10 min.

Solar cell fabrication: Reagent grade chemicals of [6,6]-Phenyl- C_{61} -butyric acid methyl ester (PCBM), Poly (3-hexylthiophene-2, 5-diyli) (P3HT) were purchased from Aldrich company. The ITO (resistivity 25 Ω/sq , Aldrich) glass was masked, patterned and cleaned successively by a 10% detergent, then with ethanol and further ultra-sonicated thrice in acetone; it was cleaned further with O_2 plasma (40 W, 7.5×10^{-5} Pa) for 10 min. Highly conducting poly(3,4-ethylenedioxyethiophene):polystyrene sulfonic acid (PEDOT:PSS) was then spin casted (3000 rpm for 30 s) after passing through a $0.80 \mu\text{m}$ filter. After that it was annealed at 120°C for 30 min in vacuum. The substrates were then moved into a nitrogen-filled glove box where the active layer consisting of

P3HT:PCBM: FeS_2 QDs (10 mg:10 mg:1 mg) in the 1 ml chloroform solvent (sonicated for 15 min and solution was magnetically stirred for 48 h) was spin-casted at 1000 rpm for 30 s on to the PEDOT:PSS coated layer. Similarly a 20 wt% and 40 wt% of QDs based active layer were also prepared. Then with the use of cotton buds excess spillover was removed. The coated substrates were then moved to a thermal evaporator where aluminum electrodes (80 nm) were evaporated to complete the device (ITO/PEDOT:PSS/P3HT:PCBM: FeS_2 QDs/Al) and finally post-annealing was carried out at 120°C for 15 min which has a cell active area of 0.02 cm^2 . The morphology of the samples was imaged with a transmission electron microscope (Philips F20Teca) at 200 kV). X-ray diffraction (XRD) was analyzed by a Rigaku D/Max-RB diffractometer using $\text{Cu K}\alpha$ at 40 kV and 100 mA. Absorption spectra dispersed in toluene were recorded on a Shimadzu UV-3600 spectrophotometer and J - V characteristics and external quantum efficiency were measured by Bunkoh-Keiki CEP-015 system under AM 1.5 G irradiation ($100 \text{ mW}/\text{cm}^{-2}$).

3. Results and discussion

The morphological evaluation and the size of the synthesized pyrite quantum dots were analyzed by a transmission electron microscopy (TEM) and are represented in Fig. 1(a, b). It clearly shows that the synthesized nanocrystals are ~ 5 nm in diameter with the morphology matching with quantum dots (QDs) where a good monodispersity of the dots was observed. Corresponding high resolution TEM image (b) was measured showing a fringe spacing value of 0.3 nm, which is close to the space between (200) planes of bulk cubic iron pyrite (d_{200}), confirming the excellent defect free quantum dots. Selected area diffraction pattern of iron pyrite QDs (inset b) shows a ring pattern which reflects a well crystalline nature.

Optical properties of the synthesized quantum dots were analyzed by UV-vis spectra which are shown in Fig. 2(a). The figure clearly indicates that the absorption onset is at 1289 nm on extrapolating the spectral curves with the estimated bandgap of 0.99 eV. It is interesting to observe the shoulder at 2.38 eV which shows monotonic absorption rise. The nanocrystal shows visible and infra-red light absorption throughout indicating a promising material for extended light harvesting in photovoltaic application. Fig. 2(b) shows an X-ray diffraction (XRD) pattern which depicts that the synthesized nanocrystals show structural phases typical to cubic FeS_2 structure without a trace of detectable impure marcasite, pyrrhotite, and greigite peaks. The dominant

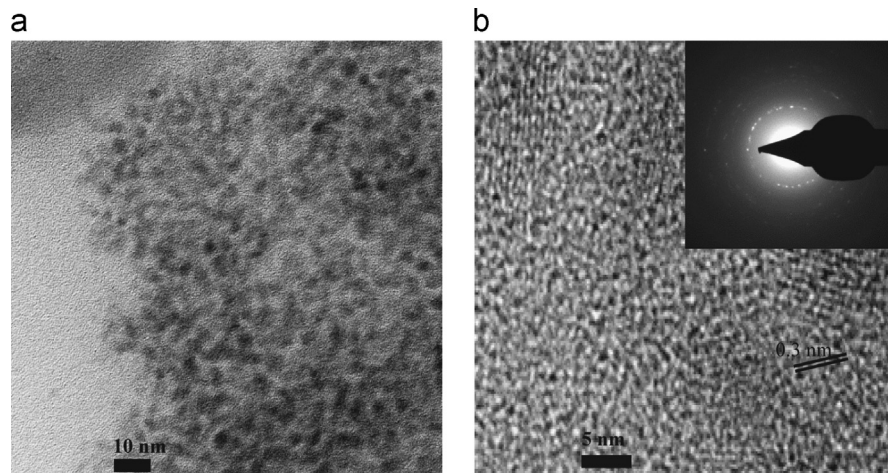


Fig. 1. TEM image of the synthesized FeS_2 QD sample with (a) low and (b) high resolution transmission images; inset: selected area electron diffraction pattern (SAED) of iron pyrite QDs nanocrystals.

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