

Carbon dots based nanocomposite thin film for highly efficient luminescent solar concentrators

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ABSTRACT

Luminescent solar concentrators (LSCs) have attracted extensive interest due to their potential application in building integrated photovoltaics. As for the emitters, organic dyes and semiconductor quantum dots are generally used for LSC fabrication. However, carbon-dots as a new class of quantum dots used in LSC study are rare. Here, N-doped carbon-dots (N-CDs) are used as phosphors to be embedded in polyvinylpyrrolidone (PVP) thin films for LSC fabrication because of the good compatibility between them. The optical properties and performance of N-CDs/PVP thin film based LSCs with different layer numbers are investigated. The high performance of N-CDs/PVP thin film based LSCs with a power conversion efficiency of 4.97% has demonstrated that N-CDs embedded in PVP thin films is an effective approach for LSC fabrication which makes the industrial applications of LSCs more feasible.

1. Introduction

Luminescent solar concentrators (LSCs) consisting of waveguides and fluorophores can collect incident light and redirect it onto photovoltaic cells which are attached to the transparent slab edges [1–3]. Compared to traditional solar cells, LSCs require only small side areas covered by expensive photovoltaic cells which can significantly reduce the cost of photovoltaic energy production [4,5]. Furthermore, the semi-transparent property of LSC device endows it potential application in photovoltaic windows [6–8]. Despite their promise, there is still an outstanding challenge in finding suitable emitters. Although many types of luminescent fluorophores such as organic dyes [9–11] as well as semiconductor quantum dots (QDs) [12–14] have been employed in LSCs, issues (e.g., photobleaching [15,16], toxicity [17] and cost [18]) caused by these fluorophores greatly hinder the real-life application of LSCs.

Carbon-dots (CDs) as a new class of QDs were discovered in 2004 [19]. Their unique properties such as high stability, low toxicity, and high photoluminescence quantum yield endow them wide applications such as biosensors [20–25], and photocatalysis [26]. Owing to these merits, researchers recently have paid special attention to exploiting new functions and application of CDs. It can be anticipated that the high photoluminescence quantum yield property of CDs would make them a promising candidate for fabricating LSC devices. However, up to

date, the attempt on CDs especially N-doped carbon-dots (N-CDs) for LSC fabrication is rare [27,28]. Most recently, our group have demonstrated the feasibility of CDs used for LSC device fabrication [29]. Unfortunately, the nanoparticle aggregation within the matrix [30] is still a big problem which greatly limits the performance of LSCs. Thus, this compatibility issue needs to be fully addressed in order to improve the performance of LSCs.

The poor compatibility between quantum dots (QDs) and polymer materials is a great problem which obviously causes the QD aggregation during the matrix fabrication [31] leading to photoluminescence quenching of QDs. To avoid this issue, a variety of methods have been introduced such as usage of UV-curable polymer [6,32] and hydrophilic polymer [12]. Polyvinylpyrrolidone (PVP) is a kind of hydrophilic polymer [33] materials whose pyrrolidone ring is able to coordinate to the hydroxyl group [34] which can significantly improve the compatibility between quantum dots (QDs) and polymer materials.

In this study, we report for the first time that N-CDs are used as phosphors to be embedded in PVP polymer film for LSC fabrication because of the good compatibility between them. The optical properties and performance of N-CDs/PVP thin film based LSCs with different layer numbers and concentrations of N-CDs are systematically investigated. Our results show that N-CDs are able to be effective fluorophores for LSC fabrication which makes the industrial applications of LSCs more feasible.

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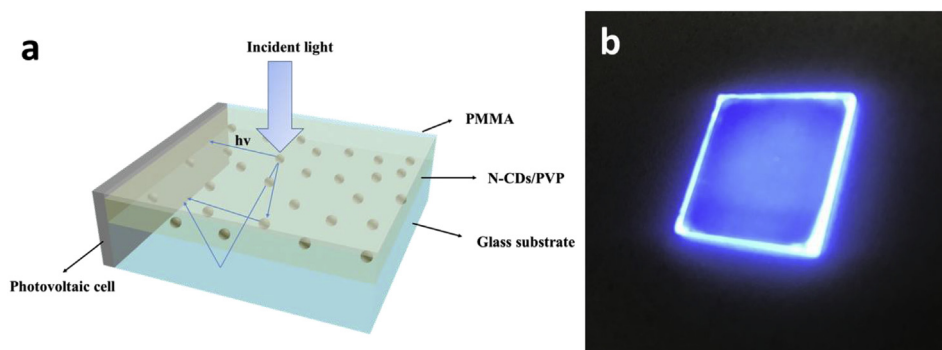


Fig. 1. (a) Schematic illustration of the LSC consisting of PVP film and N-CDs. (b) Photograph of N-CDs/PVP thin film based LSC sample under UV illumination. It is apparent that the emitted light is wave-guided to the slab edges through total internal reflection.

2. Results and discussion

The prepared N-CDs were first characterized by TEM. As shown in Fig. S1a, it can be clearly observed that these N-CDs exhibited nearly spherical shapes which had an average size of around 2.9 ± 1.3 nm. The diameter distribution of N-CDs is shown in Fig. S1b. As shown in Fig. S2, the FTIR absorption has a wide peak around 3400 cm^{-1} , which can be assigned as O–H and N–H stretching vibrations. The sharp peak at 1620 cm^{-1} is associated with C=O, C=C stretching vibrations and the peak observed around 1400 cm^{-1} is attributed to C–N bending vibrations. Meanwhile, the peak at 1082 cm^{-1} is ascribed to the C–O bending vibrations. These results suggest that plenty of hydroxyl groups and amine groups exist on the surface of the carbon nanodots.

Fig. 1a shows the schematic illustration of the LSC consisting of PVP thin film and N-CDs: When light is incident onto the face of the thin film LSC, it can be absorbed by N-CDs and redirected at a longer wavelength randomly. Because of a higher refractive index of the plate than that of air, most of the emitted photons is trapped inside the plate by total internal reflection and guided to the slab edges. Fig. 1b shows the photograph of PVP and N-CDs based LSC under UV illumination which clearly shows the concentrated light could emit from the four side edges of the slab.

Previous reports show that high concentration of the fluorophores within the polymer matrix is required to obtain optimal optical efficiency since intermolecular energy-transfer efficiency is a key limiting factor. However, higher concentrations can lead to larger losses generated by self-absorption and fluorophore aggregations [35,36]. In our experiments, different concentrations of N-CDs including 0.1%, 0.3%, 0.5%, and 0.7% were used to study. We prepared N-CDs/PVP thin film based LSC samples with different layer numbers controlled by spin-coating times. The UV absorbance spectra of N-CDs/PVP thin film LSC samples with different thin film layer numbers containing 0.7 wt% N-CDs are shown in Fig. 2a. It showed that there was an obviously broad adsorption peak from 350 nm to 400 nm. The absorbance intensity gradually increased with film layer numbers which should be ascribed to the fact that more N-CDs are embedded in the N-CDs/PVP thin film with higher layer number. Fig. 2b shows absorption spectra of N-CDs/PVP thin film based LSCs containing different concentrations with layer number 5, which also reveals the absorbance intensity increased with increasing the concentration of N-CDs. Our results also indicate that the visible light transmittance of LSCs with different layer numbers is excellent. Visible light transmittance could keep above 90% which clearly demonstrates the fabricated LSCs can realize semi-transparent photovoltaic windows. Reflectance spectra of the N-CDs/PVP thin film with different layer numbers (0.7 wt% N-CDs) were further measured as shown in Fig. 2c, which clearly shows a distinguishable absorption centered at ~ 325 nm which should be attributed to the N-CDs.

Fluorescence emission spectra of N-CDs/PVP thin film based LSCs with different layer numbers excited by light with different

wavelengths were investigated. As shown in Fig. 3a, regarding the 0.5 wt% N-CDs, the PL intensity increased with the increasing of layer numbers. When the layer numbers kept as 5, the PL intensity obviously increased with the increasing concentration of N-CDs (Fig. 3b). According to Fig. 3c, the peaks of N-CDs/PVP thin film based LSCs with different layer numbers all shifted from 420 nm to 560 nm when the excitation wavelength changed from 290 nm to 500 nm. The fluorescence peaks of N-CDs/PVP thin film based LSCs showing a red shift from 420 nm to 560 nm when the excitation wavelength increases indicates that the overlap of the absorption and emission spectra of LSCs becomes smaller since the absorption spectra peak position keep unchanged, which means Stokes shift become larger which is necessary for reducing reabsorption losses in LSC devices. The maximum fluorescence emission intensity of the N-CDs/PVP thin film appeared at 420 nm under 360 nm excitation. It is supposed that the fluorescence behavior depending on excitation wavelength is ascribed to the distribution of different emissive sites [26].

As shown in Fig. 4, the PL decays of the emissions from N-CDs in PVP thin films with different layer numbers had almost the identical lifetime of ~ 2.8 ns, which means the similar luminescent process. The PL lifetime of the N-CDs in ethanol was ~ 1.9 ns which is slightly shorter than that of N-CDs in PVP thin films, which indicates there is almost no effect on the PL dynamics of N-CDs before (in solution) and after they are embedded into the polymer thin films. The very small vary in the PL lifetime time from 1.9 ns to 2.8 ns can be explained by the difference in the environments of N-CDs [6].

The thicknesses of N-CDs/PVP thin film LSC samples with different layer numbers measured from SEM cross-sectional images are shown in Fig. 5. According to the insets in Fig. 5, the thickness of N-CDs/PMMA thin film LSC samples with layer number 1 was about $8\text{ }\mu\text{m}$. The thicknesses of N-CDs/PVP thin film LSC samples increased with increasing layer numbers.

The performance of the LSC devices in our study was evaluated by testing under AM 1.5G solar irradiation at $100\text{ mW}/\text{cm}^2$. Following parameters were measured and considered [37,38].

The geometric gain G is a ratio of the top surface area (A_{top}) to the edge surface area (A_{edge}) of the LSC slab, depending on the dimensions of the LSC slab.

$$G = A_{top}/A_{edge} \quad (1)$$

The optical efficiency (η_{opt}) which generally determines the performance of an LSC refers to the ratio of the power coming out of the slab edge of the LSC (P_{out}) to the power coming in (P_{in}) through the top surface, which can be simply defined as following when PV cells attached to the LSC slab edge.

$$\eta_{opt} = P_{out}/P_{in} \quad (2)$$

$$\eta_{opt} = \frac{I_{LSC}}{I_{SC} \times G} \quad (3)$$

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