Dual-action carbon quantum dots with light assist in enhancing thermoelectric performance of polymer

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Experimental work

Prepare PP-CQDs composite films

To prepare PP-CQDs composite films, CQDs powders provided by Wang Zifei are firstly dispersed into deionized water to obtain the CQDs aqueous solution (0.85 mg/mL). And the synthesis of CQDs₁ and CQDs₂ was similar to his previous work.¹⁻³ Simultaneously, 1 mL PEDOT:PSS aqueous solution with 10% volume of DMSO is dispersed into 15 mL EtOH to obtain PEDOT:PSS/EtOH dispersion (0.85 mg/mL). And the previously prepared solution was sonicated for 30 min in order to maintain a uniform dispersion. The PEDOT:PSS/EtOH aqueous solution was coated to a pretreated glass substrate twice by spin-coating at 1500 rpm for 15 s. The substrates were rinsed with deionized water, acetone, and isopropanol and then treated with UV/ozone for 15 min before spin-coating. The spin-coated PEDOT:PSS film was dried at 60 °C for 5 min. Then, the CQDs aqueous solution was coated on the previously prepared PEDOT:PSS film. Finally, the above preparation process was repeated five times to obtain a uniform composite film. Finally, the spin-coated film was thermally annealed in a 150 °C oven for 15 min.

Instruments and characterizations

The FTIR spectra (Nicolet-460) and UV–vis spectroscopy (Analytikjena Specord 200) were used to characterize the structural information on as-prepared samples. The chemical compositions of PP-CQDs composite films were analyzed by X-ray photoelectron spectroscopy (XPS, Thermo Fisher Scientific: Escalab 250Xi). Ultraviolet photoelectron spectra (UPS) of the PEDOT:PSS, PP-CQDs₁ and PP-CQDs₂

films were recorded by a multifunctional X-ray Photoelectron Spec-troscopy (XPS)/ESCA (K-ALPHA+, Thermo Fisher Scientific)using the He I light source (21.2 eV). The Seebeck coefficient (*S*) is calculated from $S = -\Delta V / \Delta T$, where ΔV and ΔT are the TE voltage and temperature difference between the two sides of the samples. Here, ΔT is seted up by heating plate, ΔV and ΔT are monitored with Keithley 2700 system and Pt100 thermocouples, respectively. The thickness of the samples was measured with a profiler (Dektak XT, USA). A JEOL JEM 2100 TEM was used to examine the morphologies of CQDs.

In the test of PTE properties, the induced voltage is detected with Keithley 2700 system, the temperature is monitored with Pt100 thermocouples. Herein, to realize an asymmetric light illumination, a shadow mask with different size of aperture (square) is set aside to the sample, and the square part of the sample is illuminated through the aperture and the diameter of the dark box hole can be changed. Notably, there is a gap of 0.5 cm between the sample and shadow mask, and the sample is 50 cm far away the light source. The induced voltage is detected by attaching Ag electrodes to the sample, and temperature is tested by attaching two Pt100 thermocouples onto the two sides of the sample. A PLS-SXE300+ (Perfect Light Technology Co., Ltd. China) Xenon lamp is used as light source (wavelength: 320-2500 nm). During the photothermoelectric performance test, the irradiation area of the films were100 mm² and the irradiation power of the xenon lamp was 249.3 nW. The lasers were purchased from Changchun lei Laser Technology Co. Ltd. Several types of continuous-wave lasers served as the excitation light sources with different wavelengths, including UV (405 nm), visible

(532 and 635 nm) and near-infrared (808 and 1064 nm). The incident power at each wavelength was monitored by a calibrated power meter operating in the corresponding waveband. Lasers of different wavelength band were used to irradiate left side of the film and the incident beam diameters from different lasers were kept on the same level (about 2-3 mm).



Fig. S1 (A) The sheet resistance of PEDOT:SS film under different spraying times; (B) The sheet resistance and *S* of PEDOT:PSS film with 10 *wt*% DMSO added before and after annealing.

Fig. S1A shows the sheet resistance of PEDOT:SS film under different spraying times. It was found that the resistance of the PEDOT:SS film is relatively stable after 10 times of spraying, and the sheet resistance can be further reduced after the film is annealed (Fig. S1B), which may be attributed to the improvement of the microstructure of the film by annealing.



Fig. S2 The sheet resistance of PEDOT:PSS film after adding different amounts of DMSO.

Fig. S2 shows the sheet resistance of PEDOT:PSS film after adding different amounts of DMSO. When 10% DMSO was added to PEDOT:PSS, the sheet resistance of the film was the smallest and the Seebeck coefficient did not change much. Therefore, this condition was chosen to prepare the composite films.



Fig. S3 The FT-IR spectrum of CQDs₁ and CQDs₂.

Fig. S3 demonstrates that CQDs₁ and CQDs₂ have very similar IR spectra that revealed the existence of functional groups. Broad absorption bands at 3000–3500 cm⁻¹ are assigned to v(N-H) and v(O-H). The strong stretching vibration bands of C = O, C = N, and C-N are observed at 1646, 1562, and 1400 cm⁻¹, respectively.^{3, 4} The peaks located at 1178 and 1061 cm⁻¹ should be caused by P = O and P-O vibrations.^{1, 5, 6} CQDs₁ and CQDs₂ have very similar IR spectra that revealed the existence of functional groups. The presence of functional groups (C=N, C=O, NH, OH, etc) imparts excellent solubility in water without the need of further chemical modification of nanoparticle surface.⁸



Fig. S4 TEM image of $CQDs_1(A)$ and $CQDs_2(B)$; the size distributions of $CQDs_1(C)$ and $CQDs_2(D)$.



Fig. S5 XPS results for the C 1s (A), N 1s (B), O 1s (C), and P 2p (D) spectra of PP-

CQDs₁, respectively.



Fig. S6 The maximum saturation temperature difference of PP, PP-CQDs₁ and PP-CQDs₂ films under illumination. Here is the xenon lamp lighting and the illumination area is 100 mm^2 .



Fig. S7 The induced temperature difference of PP, $PP-CQDs_1$ and $PP-CQDs_2$ films under continuous direct heating and then lighting. Note that with the dotted line as the center, the left is the temperature change caused by direct heating, and the right is the temperature change caused by lighting.



Fig. S8 The induced voltage of PP-CQDs₁ film under continuous direct heating and then lighting.



Fig. S9 The relationship between the induced voltage of $PP-CQDs_1$ film and laser power (A), and the time-dependent induced voltages of the $PP-CQDs_1$ film is irradiated with different lasers (B).

	405 nm (K)	532 nm (K)	635 nm (K)	808 nm (K)	1064 nm (K)
РР	0.85	0.75	0.87	0.92	0.90
PP-CQDs ₁	0.98	0.96	0.92	1.1	0.93
PP-CQDs ₂	1.1	0.90	0.94	1.0	0.92

Table S1. The induced temperature difference of PP, PP-CQDs₁ and PP-CQDs₂ films were irradiated with different lasers.



Fig. S10 The *I*–*V* characteristics of PP (A) and PP-CQDs₁ (B) films under the avoid light and light illumination. Here is the xenon lamp lighting and the illumination area is 100 mm^2 .



Fig. S11 The I-V characteristics of PP-CQDs₁ (A) and PP (B) films were irradiated with different lasers.



Fig. S12 The saturated temperature difference for PP, PP-CQDs₁ and PP-CQDs₂ films at different illumination area. Here is the xenon lamp lighting.



Fig. S13 The temperature difference and induced voltage as a function of illumination time within 6 cycles light on and off for PP (A) and PP-CQDs₁ (B) film. Here is the xenon lamp lighting and the illumination area is 100 mm^2 .

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