Chapter 14

Ultralight Paper-Based Electrodes for Energy Applications

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Additional information is available at the end of the chapter

http://dx.doi.org/10.5772/intechopen.74098

Abstract

In this chapter, we briefly introduce our recent work related to the topic of ultralight paper-based electrodes for energy applications. Herein, the ultralight paper-based counter electrodes containing commercial poly(3,4-ethylenedioxythiophene):polystyrenesulfonate (PEDOT:PSS) and homemade graphene dots (GDs) are synthesized for preparing flexible dye-sensitized solar cells (DSSCs). Because the GDs/PEDOT:PSS composite can well fill the porosity of paper substrate, the flexible DSSC with GDs/PEDOT:PSS-coated paper electrode exhibits much higher cell efficiency than that of DSSC using paper electrode with Pt. The features of lightweight, low-cost, space-saving (high flexibility), high machinability (easy-cutting) and environmental friendly would make the GDs/PEDOT:PSS-coated paper electrodes highly potential in portable/wearable electronic applications.

Keywords: conducting polymer, flexible electronic, graphene dot, paper electrode

1. Introduction

Flexible and lightweight electronics have attracted much attention because of their high potential to be integrated into wearable commercial products [1]. Among all the flexible substrates, the printed paper substrate, composed of cellulose fibers, has been considered as the most promising one due to the following features: environmental friendly, low-cost, lightweight and easy for roll-to-roll processing [2]. Currently, the printed paper has been widely used as a substrate for transistors [3], displays [4], sensors [5], memories [6], batteries [7] and supercapacitors [8], etc. Furthermore, Professor Wang's group reported a paper-based triboelectric nanogenerator having origami configurations for harvesting mechanical energy [9].

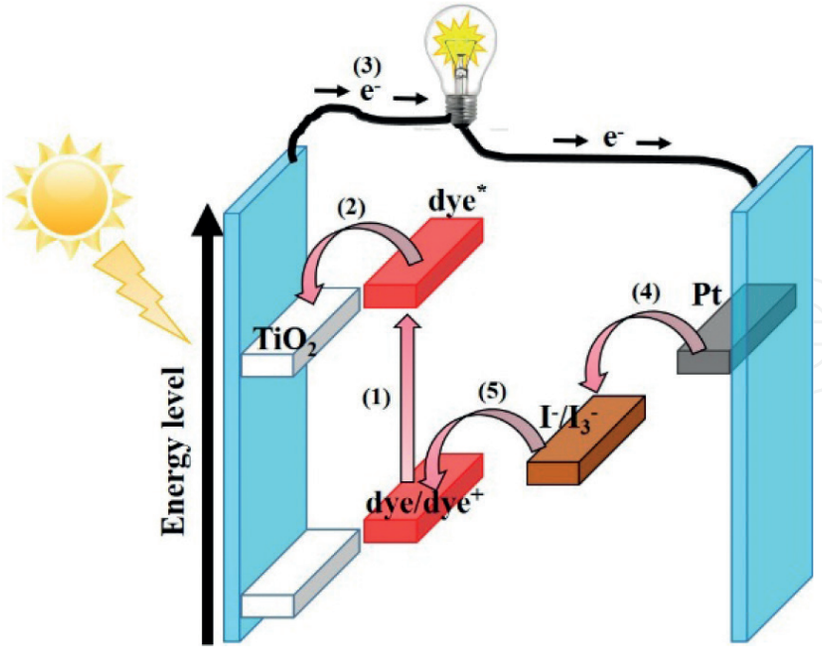

Figure 1. Schematic sketch of a DSSC.

Professor Hu's group developed highly flexible organic light-emitting diodes (OLEDs) on biodegradable and transparent paper substrates [10]. They also demonstrated a range of electronic devices on transparent paper substrate, including organic solar cell [11], touch screen [12] and thin film transistor [13].

For the applications in electrochemical devices, the main challenges of using printed paper as electrode substrates are how to make it to become conductive and electrochemical active via nonsintering processes, as well as to make a conductive layer on it uniformly and continuously because paper-based substrates are nonconductive, porous and rough due to its fibrous nature [14]. Up to now, the printed paper has not been used as the electrode substrates for the third-generation solar cell, namely dye-sensitized solar cell (DSSC). DSSCs have been extensively studied and widely developed in the last two decade since they do not rely on expensive fabrication processes and can be possibly prepared by using roll-to-roll methods [15]. Whereas, flexible DSSCs are handy, convenient for transportation and suitable for setting up at complex environments, which endow them high competitiveness in solar cell markets [16].

As shown in **Figure 1**, a traditional DSSC is consisted of a counter electrode (CE) with Pt catalyst, an I^-/I_3^--based electrolyte and a dye-sensitized TiO_2 photoanode. Briefly, the basic sequence of the working principle of a DSSC is shown below:

Activation

$$TiO_2 - dye \rightarrow TiO_2 - dye^* \quad (1)$$

Electron injection

$$TiO_2 - dye^* \rightarrow TiO_2 - dye^+ + e^-(TiO_2\ film) \quad (2)$$

$$e^{-}(TiO_2\ film) \rightarrow e^{-}(TCO) \tag{3}$$

Electron reception

$$I_3^{-} + 2\ e^{-}\ (Pt\ CE) \rightarrow 3I^{-} \tag{4}$$

Interception reaction

$$1.5\ I^{-} + TiO_2 - dye^{+} \rightarrow 0.5\ I_3^{-} + TiO_2 - dye \tag{5}$$

Under light illumination, a photo-excited electron is injected from the excited state of the dye (dye*) into the conduction band of the TiO_2. The injected electron percolates through the TiO_2 film by a driving chemical diffusion gradient, and is collected at a conductive glass substrate. After passing through an external circuit, the electron is reintroduced into the DSSC at the Pt CE, where I_3^- is reduced to I^-. Immediately, I^- regenerates the oxidized dye (dye^+) to complete the circle of a DSSC and to refresh the DSSC without other chemical side reactions.

However, the most used electrocatalyst on the CE of DSSCs is Pt, which is an expensive noble metal and is rare on earth. To further reduce the cost of fabrication of DSSCs on industrial scale, it is better to develop metal-free electrocatalytic materials for the CEs of DSSCs. Accordingly, conducting polymers (e.g., poly(3,4-ethylene dioxythiophene) (PEDOT) [17], poly(3,3-diethyl-3,4-dihydro-2H-thieno-[3,4-b][1,4]-dioxepine) (PProDOT-Et_2) [18], polypyrrole (PPy) [19], and polyaniline (PANI) [20]) and carbonaceous materials (such as carbon black (CB) [21], graphite [22], carbon nanotube (CNT) [23] and graphene [24]) have become the most promising electrocatalysts for the CE of DSSCs since they are metal element-free, have low material cost and possess good electrocatalytic activity. A water-dispersible conducting polymer, poly(3,4-ethylene dioxythiophene):poly(4-styrene sulfonate) (PEDOT:PSS), has attracted much attention as the catalytic CEs of DSSCs mainly due to its exceptional advantage of aqueous solution processibility [25]. Nevertheless, pristine PEDOT:PSS films are plagued by low conductivity (i.e., <1 S cm^{-1}) and poor electrocatalytic activity for the reduction of I_3^- due to the nonconductive counter anion, PSS^-, disturbing the conduction path of PEDOT inside the film as well as the poor catalytic surface area of its flat film [26]. Inert solvents [27] or carbon materials [28] have been employed to improve conductivity and catalytic surface areas of PEDOT:PSS films. For example, by introducing CB into the PEDOT:PSS-based CEs for their DSSCs, η can achieve to 7.01% [28]. Multiwall CNT-PEDOT:PSS composite CE for DSSCs exhibits η of 6.50% [29]. A catalytic film composited of graphene and PEDOT:PSS for the use of CE in a DSSC had reached 4.50% efficiency [30]; however, a perfect graphene sheet usually possesses limited active sites for electrocatalytic reaction in spite of its extraordinarily high electrical conductivity [31]. Several strategies are employed to increase the electrocatalytic active sites on graphene sheets, such as chemical functionalization [32], heteroatom doping (e.g., nitrogen-doped graphene) [33] and nanosized graphene pieces (e.g., graphene dots (GDs) [34]). Among these graphene nanostructures, GDs have attracted great attention and been widely applied in bioimaging [35], LEDs [36] and photovoltaics [37] due to their unique properties of quantum confinement and edge effects [38, 39]. Moreover, nanometer size and rich oxygen-containing group of GDs facilitate them to be well dispersed in most solvents [40],

which benefits for various solution-processable applications. Therefore, the incorporation of GDs in PEDOT:PSS enable us to fabricate an efficient electrocatalytic film by using a simple solution-coating method under low temperature (<100°C).

In this chapter, an all-metal-free CE-containing GDs-PEDOT:PSS and printed paper is developed for flexible DSSCs, and it exhibits higher performance and bending stability than those of a paper electrode with sputtered Pt. The concurrent advantage in low material cost, simple fabrication processes, highly bending durability, lightweight, space-saving, high machinability and environmental friendly makes the GDs-PEDOT:PSS-coated paper electrode playing a crucial role in lightweight electronic devices. Most importantly, this GDs-PEDOT:PSS composite ink can be used in the printable processes for mass production of flexible electrodes.

2. Experimental

2.1. Synthesis of graphene dots

The GDs solution was prepared by using deionized water and glucose as the solvent and source, respectively. First, the as-prepared glucose solution (2.5 mL) was transferred to a glass bottle with 4 mL volume and a tightened cover. The synthesis reaction was carried out in a microwave oven (595 W) for 9 min; the glucose molecules are pyrolyzed and then converted to GDs as shown in **Figure 2**. Subsequently, the reaction bottle was cooled to ambient temperature, and the water-soluble GDs solution was thus prepared. **Figure 3** is the photographs of GDs solution taken under the illumination of visible light and UV light, showing the excitation wavelength-dependent fluorescence property of GDs.

2.2. Preparation of paper-based counter electrodes

The mixing solutions composed of 50 V% PEDOT:PSS aqueous solution and 50 V% binary solution consisting of GDs (X) and ethanol (Y) (X/Y = 3/2) were used for preparing the GDs-PEDOT:PSS composite inks.

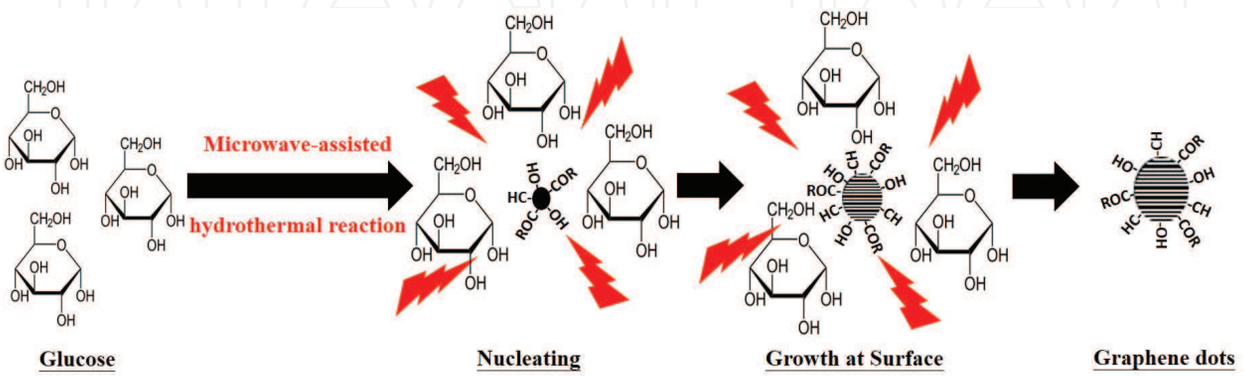

Figure 2. Preparation of graphene dots via the microwave-assisted hydrothermal technique.

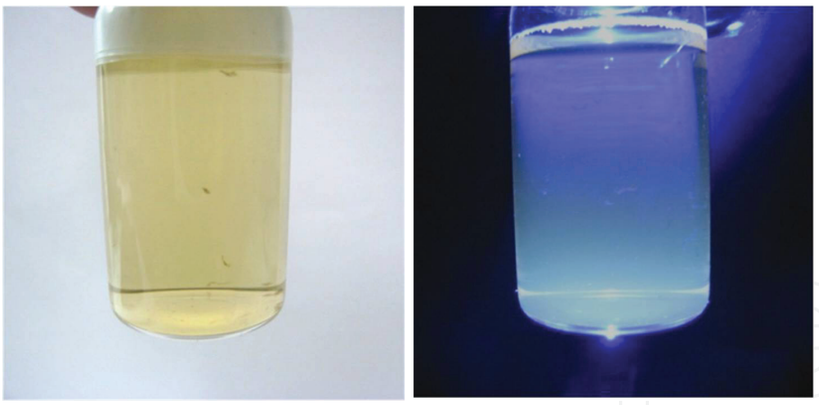

Figure 3. The GDs solutions under ambient light (left) and UV light (right) [41].

For the preparation of paper-based CEs, the commercial printing papers were used as the substrate of counter electrodes. The paper with a fixed coating area was immersed in the GDs-PEDOT:PSS solution (i.e., 30 V% GDs solution content) for 10 min, and it was took out and then dried under 60°C. The thus prepared GDs-PEDOT:PSS/paper electrode (left-hand side of **Figure 4**) was used as the CE for the studies on flexible DSSCs. For comparison purposes, the paper-based CE with sputtered Pt (right-hand side of **Figure 4**) was also prepared as the standard CE.

2.3. Fabrication of flexible dye-sensitized solar cells

For the preparation of the flexible photoanode of DSSCs, the dye-sensitized TiO_2 film was prepared on the conducting plastic substrate (13 Ω $sq.^{-1}$, ITO-PEN) according to the previous reports [42, 43]. First, the binder-free TiO_2 paste was synthesized by mixing 1 g TiO_2 powder (P25) with 6 mL binary solution consisting of tert-butanol and deionized water (volume ratio of 2:1) uniformly. Then, the surface of the ITO-PEN substrate was coated with a TiO_x compact layer by spraying an ethanol solution (10 mL) containing titanium tetraisopropoxide (0.028 g) on it. Using the binder-free TiO_2 paste, a 10-μm-thick film was coated on the treated ITO-PEN substrate through doctor blade technique [44]. Thereafter, an active area of 0.4 × 0.4 cm^2

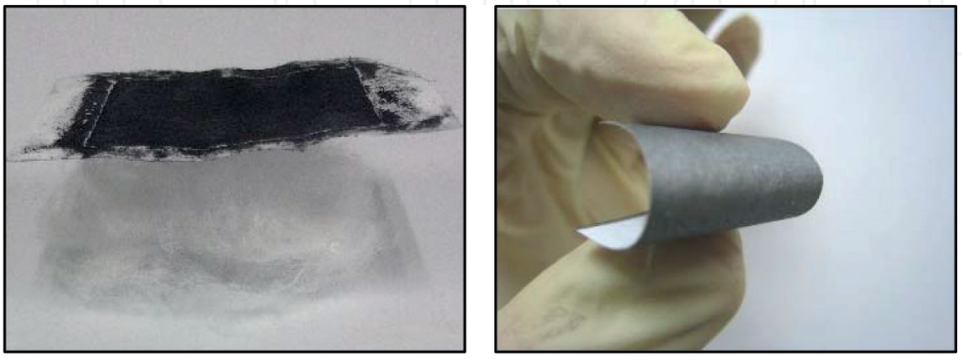

Figure 4. Pictures of CEs with 30 V% GDs-PEDOT:PSS composite (left) and with sputtered Pt (right) on paper substrates.

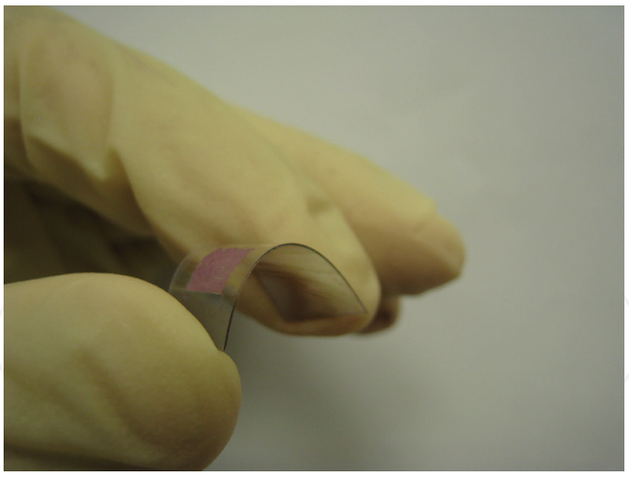

Figure 5. A flexible photoanode with dye-sensitized TiO_2 film on ITO-PEN substrate.

was selected from the TiO_2 films by scrapping. The as-prepared TiO_2/ITO-PEN electrodes were gradually heated to 120°C under ambient conditions, and subsequently annealed at the respective temperatures for 60 min. After annealing and cooling to 80°C, the TiO_2/ITO-PEN electrode was immediately immersed in a 5×10^{-4} M N719 dye solution for 60 min under 55°C. The thus prepared dye-sensitized TiO_2/ITO-PEN photoanode (**Figure 5**) was coupled with paper-based CE (i.e., GDs-PEDOT:PSS or sputtered Pt) as the flexible DSSC.

3. Results and discussion

GDs are edge-bound nanosized graphene pieces and exhibit unique electronic and optical properties due to the quantum confinement and edge effects [38, 39]. **Figure 6(a)** shows the transmission electron microscopy (TEM) image of the monodispersed GDs, which exhibit uniform diameters of ~3.50 nm. As shown in the inset of **Figure 6(a)**, the high-resolution transmission electron microscopy (HRTEM) image indicates high crystallinity of GDs with a lattice spacing of 0.246 nm corresponding to the interplanar separation of graphene (1120). **Figure 6(b)** presents the atomic force microscope (AFM) image of the monodispersed GDs; the inset of **Figure 6(b)** reveals that the average height of GDs is around 2.90 nm. As shown in **Figure 6(c)**, two absorption peaks centered at 228 and 282 nm are observed in the ultraviolet-visible (UV-visible) spectrum of the diluted GD solution, which consists with the result in the previous literature [36]. **Figure 6(d)** shows the photoluminescence (PL) spectrum of the GD solution. The broad emission peaks centered at around 450, 460 and 537 nm are observed when the sample is excited by 300, 400 and 500 nm, respectively, and they show the decrease of PL intensity. The excitation wavelength-dependent intensity and emission wavelength observed here is a common phenomenon for carbonaceous quantum dots [36, 45]. The elemental distribution of the GDs is analyzed by energy-dispersive X-ray spectroscopy (EDS), and the elemental mappings of C and O are shown in **Figure 7**. Obviously, the C content is much higher than O content; the atomic ratio of C/O is 95.32/4.68, which demonstrated C is the dominant element in the GDs.

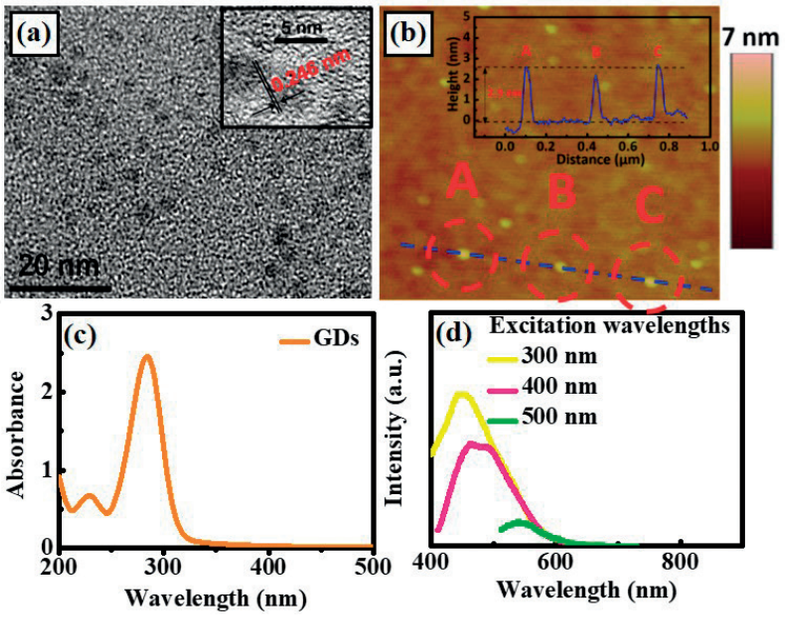

Figure 6. (a) TEM image with the corresponding HRTEM image (inset) of GDs; (b) AFM image of the GDs and its corresponding height profile; (c) absorbance spectrum of the GDs; (d) PL spectra of GDs [41].

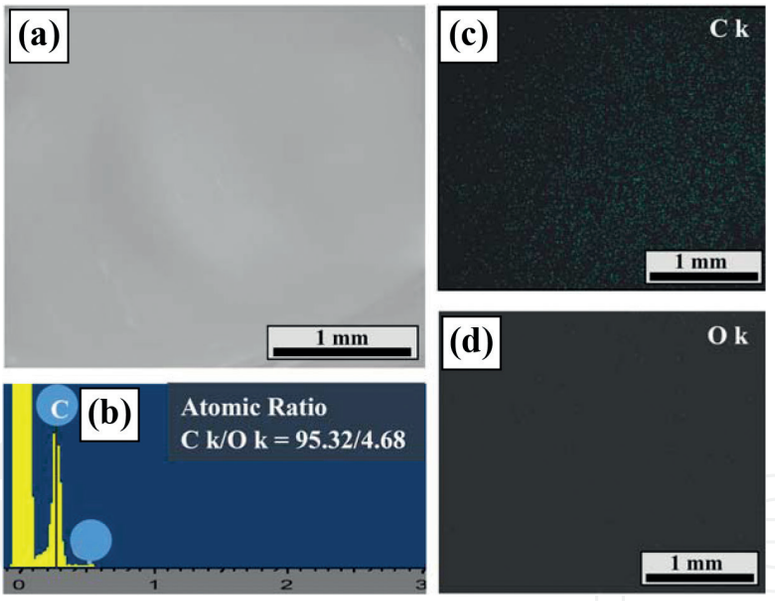

Figure 7. SEM image (a) and EDS spectrum (b) of a GD film. (c) Elemental C mapping of the image shown in (a). The elemental mappings of C (c) and O (d) of the GD film [46].

A mixing solution composed of 50 V% PEDOT:PSS solution, 30 V% GDs solution and 20 V% ethanol were used as the GDs-PEDOT:PSS composite ink. To explore the advantage of the GDs-PEDOT:PSS composite ink for the application in DSSCs, 30 V% GDs-PEDOT:PSS and Pt were separately coated onto the printed papers as the CEs of flexible DSSCs. **Figure 8(a)** and **(b)** shows the top-view SEM images of paper electrodes with sputtered Pt and with 30 V% GDs-PEDOT:PSS, respectively. The SEM images obviously show that the porosity of paper substrate

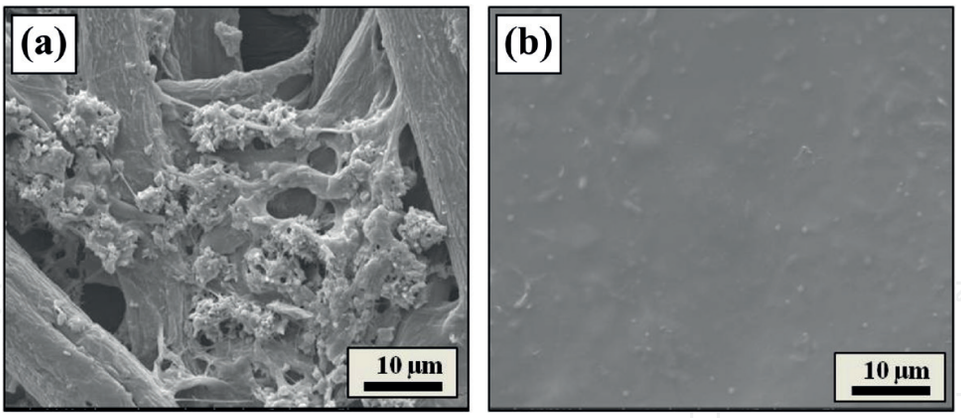

Figure 8. The top-view SEM images of paper substrates with (a) sputtered Pt and with (b) 30 V% GDs-PEDOT:PSS composite [46].

can be perfectly filled by GDs/PEDOT:PSS; however, that cannot be achieved by sputtering Pt, which means that the sputtered Pt layer could not provide a continuous electron transport route in a porous paper substrate. Above two paper, CEs (i.e., 30 V% GDs-PEDOT:PSS and sputtered Pt) are assembled with flexible dye/TiO_2/ITO-PEN photoanodes for studying the pertinent photovoltaic performance.

Figure 9 shows the photovoltaic parameters of the flexible DSSCs using various paper-based CEs with different bending times. As shown in **Figure 9(a)**, the flexible DSSC with GDs/PEDOT:PSS-coated paper CE (η = 4.91%) exhibits three times higher cell efficiency than that of cell using Pt-coated paper CE (η = 1.70%), since the GDs/PEDOT:PSS composite can well fill the porosity of paper substrate. It is worth to mention that the development of an all-metal-free CE is an effective

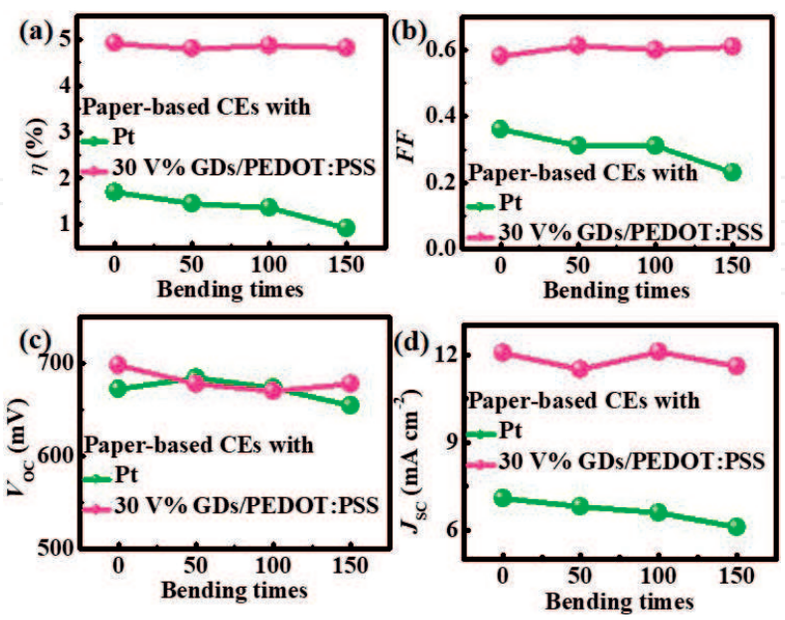

Figure 9. The photovoltaic parameters of the flexible DSSCs using various paper-based CEs with different bending times. (a) Cell efficiency (η); (b) fill factor (FF); (c) open-circuit voltage (V_{OC}); (d) short-circuit current density (J_{SC}) [46].

approach for reducing the cost of the DSSCs. Moreover, the bending test is carried out to study the durability of the paper CEs. Both GDs/PEDOT:PSS-coated paper electrode and Pt-coated paper electrode are bended for several times (0, 50, 100 and 150 times) and then assembled with the flexible photoanodes to measure and record their corresponding photovoltaic performances. **Figure 9(a)** shows the bending time dependence of cell efficiency for the flexible DSSCs with various paper CEs; their corresponding photovoltaic parameters are shown in **Figure 9(b)–(d)**. It obviously shows that the GDs/PEDOT:PSS-coated paper CE shows unfailing performance even though it was bended for 150 times; on the contrary, the Pt-coated paper CE lost its original performance drastically.

4. Conclusion

In summary, a GDs-PEDOT:PSS composite ink was synthesized for preparing the all-metal-free paper-based CEs for flexible DSSCs. The GDs-PEDOT:PSS/paper CE was fabricated through a low-cost and simple coating method (i.e., soak and dry), which could be easily scaled up to mass production. An all-flexible DSSC with GDs/PEDOT:PSS-coated paper CE exhibits much higher cell efficiency (4.91%) than that of cell using paper CE with sputtered Pt (1.70%), since the porosity of paper substrate can be well filled by GDs-PEDOT:PSS, which cannot be achieved by sputtered Pt. After bending for 150 cycles, the performance of GDs/PEDOT:PSS-coated paper CE is still perfectly preserved; on the contrary, the paper CE with sputtered Pt lost its initial performance drastically. In conclusion, this GDs/PEDOT:PSS-coated paper CE is lightweight, low-cost, space-saving (high flexibility), high machinability (easy-cutting) and environmental friendly, which shows the potential for the future applications on portable/wearable electronics.

Acknowledgements

This work was supported by the Ministry of Science and Technology (MOST) of Taiwan.

Conflict of interest

The authors declare no competing financial interests.

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