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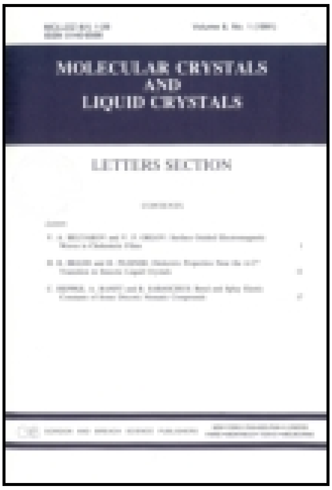

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Fabrication and Characterization of Reduced Graphene Oxide Counter Electrode for Dye-Sensitized Solar Cells

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Fabrication and Characterization of Reduced Graphene Oxide Counter Electrode for Dye-Sensitized Solar Cells

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To improve performance of dye-sensitized solar cell(DSSC), thermally reduced graphene oxide(rGO) was investigated as Pt-free counter electrodes(CE) material. The DSSCs with rGO were fabrcated by simple spin coating method under different temperature and ambient atmospheres like Ar, N_2 and air. When the rGO CE was reduced at 450°C under the Ar atmosphere, it was obtained the power conversion efficiency of 6.02%, which showed the high catalytic activity and performance of DSSC as much as Pt. This value means the thermally reduced rGO CEs under the Ar gas will be suitable as a substitute for Pt CEs.

Keywords Dye-sensitized solar cells; reduced graphene oxide; counter electrode

Introduction

Pt has been preferred as a CE material for DSSCs because of its high catalytic activity and corrosion-resistant to iodo species present in the electrolyte [1]. However, since Pt is a precious metal, it causes increasing the cost of the DSSCs. Thus, there have been many attempts to find cheap materials which could substitute for Pt. Among the substitute material for Pt, carbonaceous materials showed the remarkable results due to the high electronic conductivity, the corrosion-resistant to iodo species and low cost [2]. Under simulated sunlight, graphite, activated carbon, single-walled carbon nanotubes and multi-walled carbon nanotubes showed power conversion efficiency(η) of 6.67%, 3.9%, 4.5%, 6.07%, and 3.06%, respectively [3]. Like another carbonaceous materials, graphene was also used as a potential CE material for DSSCs, due to its exceptionally large surface area

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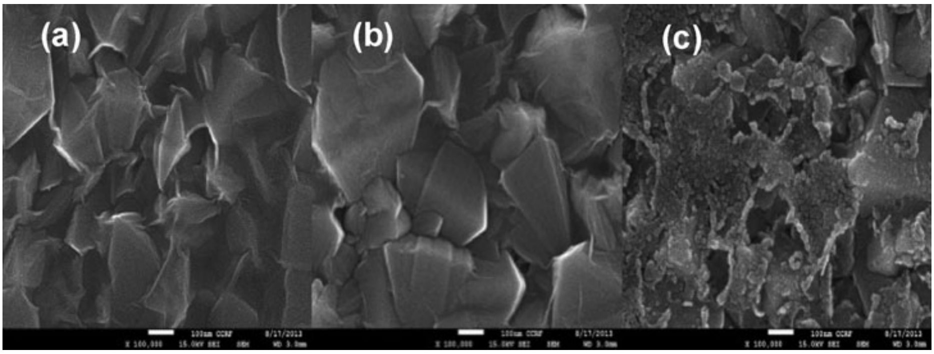

Figure 1. SEM images of thermally rGO CEs (a) under the Ar gas at 500°C, (b) under the N_2 gas at 500°C, (c) under the air at 500°C

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and high catalytic activity [2]. There were also many attempts using graphene. Choi et al. and Kim et al. fabricated GO CEs by electrophoretic deposition and obtained conversion efficiency of 5.69% and 5.87%, respectively [4, 5]. Zhang et al. also fabricated GO CEs by spray coating and obtained conversion efficiency of 5.36% [3].

In this study, thermally reduced graphene oxide(rGO) was used as a Pt-free CE material for DSSCs. To improve the performance of the DSSCs, the GO was thermally reduced under the different thermal conditions and ambient atmospheres.

Experiments

GO was prepared using a Hummer's method [6]. To make CEs, GO dispersed in water was spin-coated on FTO glass cleaned by acetone and isopropyl alcohol. And then, the coated GO film was heated at 300∼500°C for 15 minutes under the Ar, N_2 and air ambient atmosphere to obtain a reduced GO film. Photoelectrodes (PEs) were prepared as follows. TiO_2 paste(DSL 18NR-T, Dyesol Co.) was coated on FTO glass with 12 μm thickness by doctor blade method. The TiO_2 film was dried at 180°C for 10 minutes and immediately sintered at 450°C for 30 min. The TiO_2 PEs were sensitized by dipping them in N719 dye solution for 24 h. The DSSCs were assembled by sandwiching a HPE(Dyesol co.) electrolyte layer between the TiO_2 PEs and rGO CEs. The morphology of the rGO CEs was characterized using Field Emission Scanning Electron Microscopy (FE-SEM). The characteristics of the rGO CEs and DSSCs were used by cyclic voltammetry(CV), electrochemical impedance spectroscopy(EIS) and current density-voltage (I-V) measurements.

Results and Discussion

We prepared the rGO CEs on FTO glass with the different atmospheres and characterized. Figure 1 shows the FE-SEM images of the rGO CEs thermally treated with Ar(a), N_2(b) and air(c). As shown in Fig.1(a) and (b), graphenes were existed onto FTO surface and between FTO grains in the shape of a wrinkled sheets. Compared with rGO CEs prepared under the Ar and N_2 gas, graphene sheets were slightly more existed in Ar treatment as can be seen in Fig.1(a). The rGO CEs treated with air were burned out graphene residual particles due to oxygen in the air.

To characterize the catalytic activity of the rGO CEs, we performed CV and EIS measurement. Figure 2 shows the CV curves of the the I^-/I_3^- redox couple on both the Pt CEs

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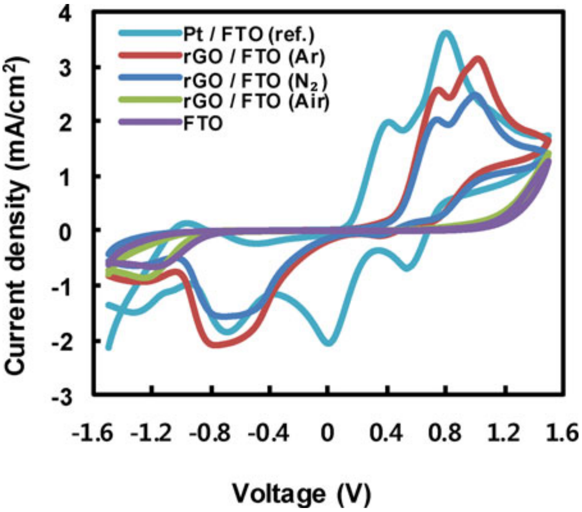

Figure 2. Cyclic voltammograms of bare FTO, Pt on FTO, various rGOs on FTO obtained under Ar, N_2 and air at a scan rate of 100 mV/s.

and rGO CEs at a scan rate of 100 mV s^{-1}. The reaction mechanism on the CE as follows [7].

$$3I_2 + 2e^- \rightarrow 2I_3^- \tag{1}$$

$$I_3^- + 2e^- \rightarrow 3I^- \tag{2}$$

The curves show two pairs of prominent redox peaks. The first reduction peak is due to reaction (2) and another reduction peak; the second reduction peak is due to the reaction (1). Thus magnitude of current density at the second reduction peak is directly proportional to the ability of the electrode to reduce the I_3^- species [8]. And that ability reflects the catalytic activity of the CEs. As can be seen in Fig.2, Pt on FTO showed highest and lowest redox peaks at the each range of voltage −0.2∼0.2 V and 0.4∼0.8 V. rGO CEs under the Ar and N_2 gas showed similar redox peaks and highest and lowest value of redox current density at the same range of voltage; each 0.8∼1.2 V and −0.8∼−0.4, but rGO CEs under the Ar showed higher and lower redox peaks when compared with rGO CEs under the N_2 gas. Compared Pt on FTO with rGO CEs under the Ar gas, Pt on FTO showed higher and lower redox peaks than rGO CEs under the Ar gas, but each value were closed. The rGO CEs under the air and the bare FTO showed similar redox peaks, as can be seen in Fig.1 (c), this means the graphene reacted on oxygen in the air and disappeared. As a result, we could know that rGO CEs under the Ar gas has a catalytic activity close to Pt CEs.

Figure 3 shows the EIS spectra(nyquist plots) for DSSCs using Pt CEs and rGO CEs. There are typical two semicircles in the nyquist plot. The first circle in the high frequency range is related to the interface between electrolyte and CE. And the second circle is related to the interface between TiO_2 and electrolyte [4, 5]. The charge transfer resistance(Rct) of the CE is determined by first circle, which values were about 6.12 Ωcm^2, 7.90 Ωcm^2, 12.46 Ωcm^2 and 17.51 Ωcm^2 corresponding to Pt CEs and rGO CEs under the Ar, N_2 gas and air, respectively. It can be seen the Rct of rGO CEs under the Ar gas was close to Pt CEs, it means rGO CEs under the Ar gas has catalytic activity close to Pt CEs.

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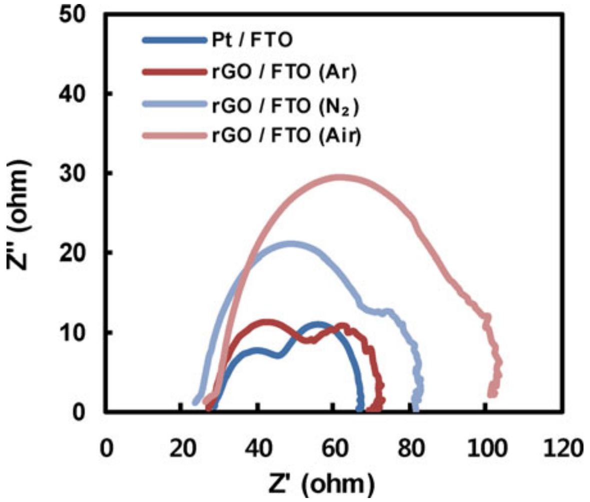

Figure 3. Nyquist plots of DSSCs at 1 sun illumination (100 mW/cm^2, AM 1.5) using Pt CEs and rGO CEs obtained under Ar, N_2 and air.

As shown CV and EIS data, I-V curves also show similar results as can be seen in Fig. 4. I-V characteristics of the each DSSCs were shown in Figs. 4 under 1 sun illumination (AM 1.5, 100 mW/cm^2). Based on these I-V curves, we summarized performance parameters; open circuit voltage(Voc), short-circuit current(Jsc), fill factor (FF), and power conversion efficiency (η) in Table 1. As can be seen in Fig. 4(a) and Table 1(a), the DSSCs with rGO CEs under the Ar gas showed power conversion efficiency of 5.89% which value was close to DSSCs using Pt CEs. The fill factor of the DSSCs with rGO CEs under the Ar gas was increased about 153% to compared with the rGO CEs under N_2.

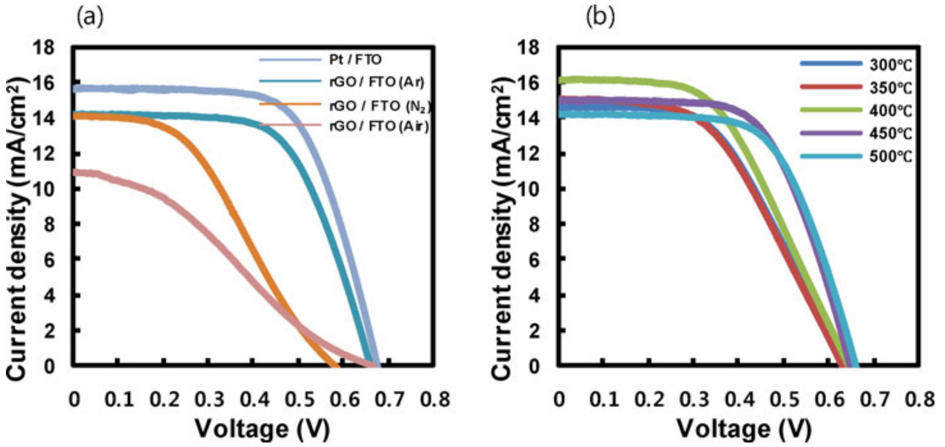

Figure 4. I-V curve characteristics measured at 1 sun illumination(100 mW/cm^2, AM 1.5) of DSSCs using (a) Pt CEs and rGO CEs obtained under Ar, N_2 and air. (b) rGO CEs obtained at 300∼500°C under Ar.

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Table 1. Solar cell performance of DSSCs using (a) Pt CEs and rGO CEs obtained under Ar, N_2 and air. (b) rGO CEs obtained at 300∼500°C under Ar.

sample	V_{oc}(V)	J_{sc}(mA/cm^2)	FF	η(%)
		(a)		
Air	0.67	10.94	0.31	2.23
N2	0.59	14.11	0.40	3.34
Ar	0.68	14.21	0.61	5.89
Pt	0.70	15.68	0.63	6.88
		(b)		
300°C	0.67	14.63	0.48	4.67
350°C	0.65	15.07	0.47	4.60
400°C	0.70	16.16	0.46	5.20
450°C	0.70	15.01	0.57	6.02
500°C	0.68	14.21	0.61	5.89

The maximum conversion efficiency was 6.02% and the optimum thermal reduction temperature of the rGO CEs treated with Ar gas was measured at 450°C in the range of 300°C∼500°C, as can be seen in Fig.4 (b) and Table 1 (b).

Conclusions

The DSSCs with thermally reduced GO CEs under the Ar gas showed higher catalytic activity than N_2 gas and air. The maximum power conversion efficiency was 6.02% which was attributed to improvement of fill factor of DSSCs rGO CEs under the Ar gas, which efficiency was close to one of Pt CEs, 6.88%, and even higher than existing recorded efficiency of 5.36% and 5.87% obtained by electrophoretic deposition and spray coating, respectively. Thus, rGO is expected to be a suitable CE material substitute for Pt.

Funding

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