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# A comparative evaluation of catalytic activities of carbon molecular sieve counter electrode toward different redox couples in dye-sensitized solar cells



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## ARTICLE INFO

### Article history:

Received 1 December 2015

Received in revised form 1 March 2016

Accepted 17 March 2016

Available online 30 March 2016

### Keywords:

solar cells

carbon molecular sieve

counter electrode

redox couple

catalyst

## ABSTRACT

Carbon molecular sieve (CMS) has been proposed as counter electrode (CE) catalyst in dye-sensitized solar cells (DSCs) for the first time. We evaluate the catalytic activities of CMS for the regeneration of different redox couples of  $I_3^-/I^-$ ,  $T_2/T^-$ , and  $Co^{3+/2+}$ , and compare them with those of the traditional Pt CE, which is very expensive. For the  $I_3^-/I^-$  redox couple, the CMS shows a relatively lower catalytic behavior than Pt due to large charge transfer resistance and low mass transport rate. Toward the organic  $T_2/T^-$  redox couple, the CMS is a much better catalyst than Pt, and the DSCs exhibits a power conversion efficiency (PCE) of 5.30%, which is a great enhancement of 42.09% over the Pt CE based DSCs. Cyclic voltammograms depict that CMS displays a high peak current density and a low peak-to-peak separation, indicating a high catalytic activity of CMS toward  $T_2/T^-$  redox couple. For the  $Co^{3+/2+}$  redox couple, CMS presents a competitive catalytic activity relative to Pt and the DSCs shows a PCE of 8.73%.

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## 1. Introduction

Dye-sensitized solar cells (DSCs) have attracted large attention due to their environmental friendliness, easy fabrication procedure, and great commercial potential [1,2]. In DSCs, the counter electrode (CE) works as a catalyst for regenerating the redox couple to complete the cycle of electron transfer [3,4]. In this respect, Pt is generally used as a CE because of its inherent high catalytic activity and excellent conductivity [5]. However, Pt, a noble metal, is very expensive and the reserves is also limited. In addition, the corrosion effect of an electrolyte on a Pt electrode can shorten the working life of DSCs [6,7]. Therefore, much effort has been devoted in search of alternative catalytic materials that can replace Pt. To this end, several new CE catalysts have been used in DSCs, such as carbon materials [8–10], organic polymers [11–13], and metal compounds [14–20]. Among these Pt-free materials, carbon is a promising CE catalyst for commercial applications due to its advantages of rich reserves, low cost, high conductivity and catalytic

activity, good stability, and easy availability. Till now, several kinds of carbon material CEs have been used in DSCs, such as activated carbon (Ca), carbon black (Cb), carbon fiber (Cf), mesoporous carbon, carbon nanotubes (CNTs), graphene, and carbon spheres, and all of these materials show decent catalytic activities [21–29].

Besides the above-mentioned carbon materials, carbon molecular sieve (CMS) is also an attractive functional material due to its thermal and chemical stabilities. CMS also has good stability under high pressure which made it a widely-studied material in energy-efficient separations of gas mixtures [30–33]. Moreover, CMS owns high conductivity, mesoporous structure, and well-developed synthetic methods which make them around 5000 times cheaper than Pt. All these advantages therefore make CMS a reasonable alternative to Pt. However, no study to-date has been done on the application of CMS as a CE catalyst in DSCs. In the current paper, we report a facile method to prepare a CE using low-cost commercial CMS. To the best of our knowledge, for the first time CMS has been applied as CE in DSCs, and the difference in the catalytic activities on regeneration of redox couples, e.g. iodide ( $I_3^-/I^-$ ), polysulfide ( $T_2/T^-$ ), and cobalt ( $Co^{3+/2+}$ ) has also been evaluated. We have characterized morphology and inner structure of CMS using TEM, SEM, and  $N_2$  adsorption-desorption isotherm, crystallinity by XRD, etc. and studied the redox behaviors using cyclic voltammetry

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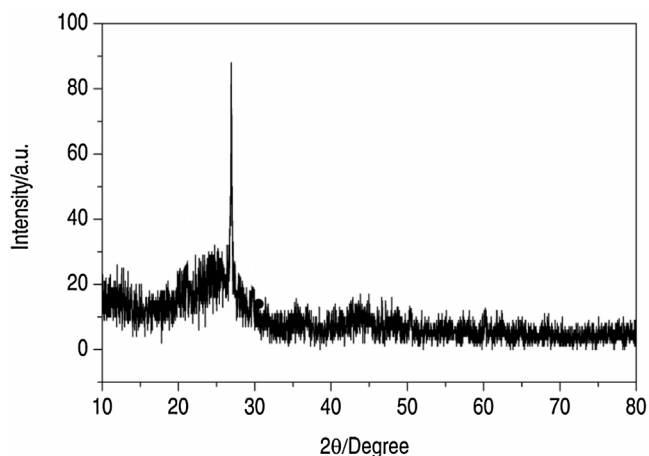


Fig. 1. XRD patterns of carbon molecular sieve (CMS).

(CV), electrochemical impedance spectroscopy (EIS), and Tafel curve technologies. Finally, the photovoltaic performance of the  $I_3^-/I^-$ ,  $T_2/T^-$ , and  $Co^{3+/2+}$  electrolytes based-DSCs using CMS CEs were checked by photocurrent density-voltage curves.

## 2. Experimental

### 2.1. Fabrication of device

The commercial CMS (CMS-260, special for nitrogen generator) was purchase from Hengxin filter plant, Gongyi Henan, China. The CMS CE was prepared by spray-coating method. The details are listed as follow: 200 mg of CMS was dispersed in 8 mL of isopropanol with ball-milling for 4 h, producing a suspension. Then, the suspension was sprayed on FTO glass followed by sintered under  $N_2$  atmosphere at  $450^\circ C$  for 30 min, obtaining CMS electrode. The thickness of CMS film was controlled around  $20\ \mu m$ . Pt electrode was achieved with thermal decomposition method as follows: isopropanol solution containing chloroplatinic acid (0.5 wt%) was sprayed on FTO glass for

5 s and sintered at  $450^\circ C$  for 30 min, obtaining the Pt electrode. As for the photoanode, a  $12\ \mu m$ -thick  $TiO_2$  film was pre-heated to  $80^\circ C$  and immersed in a  $5 \times 10^{-4} M$  solution of N719 dye in acetonitrile/tert-butylalcohol (1: 1 volume ration) for 16 h and the N719 sensitized photoanode was achieved for the  $I_3^-/I^-$  and  $T_2/T^-$  electrolyte based DSCs. A  $5\ \mu m$ -thick  $TiO_2$  film was pre-heated to  $80^\circ C$  and immersed in a  $2 \times 10^{-4} M$  solution of YD2-o-C8 dye in ethanol for 20 h and the YD2-o-C8 sensitized photoanode was achieved for  $Co^{3+/2+}$  electrolyte based DSCs. Three types of redox electrolytes were used in this work: iodide electrolyte contains 0.06 M LiI, 0.6 M 1-propyl-3-methylimidazolium iodide, 0.03 M  $I_2$ , 0.5 M 4-tert-butylpyridine (TBP), and 0.1 M guanidiniumthiocyanate in acetonitrile; sulfide electrolyte contains 0.4 M  $Na^+T^-$  (5-mercapto-1-methyltetrazole sodium salt), 0.4 M di-5-(1-methyltetrazole) disulfide ( $T_2$ ), 0.05 M  $LiClO_4$ , and 0.5 M TBP in 6:4 (v/v) acetonitrile/ethylene carbonate; cobalt electrolyte is composed of 0.25 M Co(II) tris(bipyridyl) tetracyanoborate, 0.05 M Co(III) tris(bipyridyl) tetracyanoborate, 0.5 M TBP and 0.1 M Bis(trifluoromethanesulfonylimide) lithium Salt (LiTFSI) in acetonitrile. The DSCs consisted of a photoanode, a CE, and an electrolyte. The symmetrical cell was fabricated with two identical CEs sandwiching the electrolyte for EIS and Tafel polarization tests. The cells were sealed with a hot-melt Surlyn film.

### 2.2. Measurements

X-ray diffraction (XRD) measurement was carried out using an automatic X-ray powder diffractometer (D/Max 2400, Rigaku). The morphology of CMS was characterized by scanning electron microscopy (SEM, S-4800, Hitachi, Japan), transmission electron microscopy (TEM, H-7650, Hitachi, Japan), and high resolution transmission electron microscopy (Tecnai G2 F30, Philips-FEI).  $N_2$  adsorption-desorption isotherm, carried out using an Antosorb-1 apparatus (Antosorb-1, Quantachrome, USA), and the surface area was calculated by the Brunauer-Emmett-Teller (BET) method. The current density-voltage performance of the DSCs was investigated under simulated AM 1.5 illumination ( $I = 100\ mW\ cm^{-2}$ , PEC-L01, Pecell, Yokohama, Japan). The active area of the

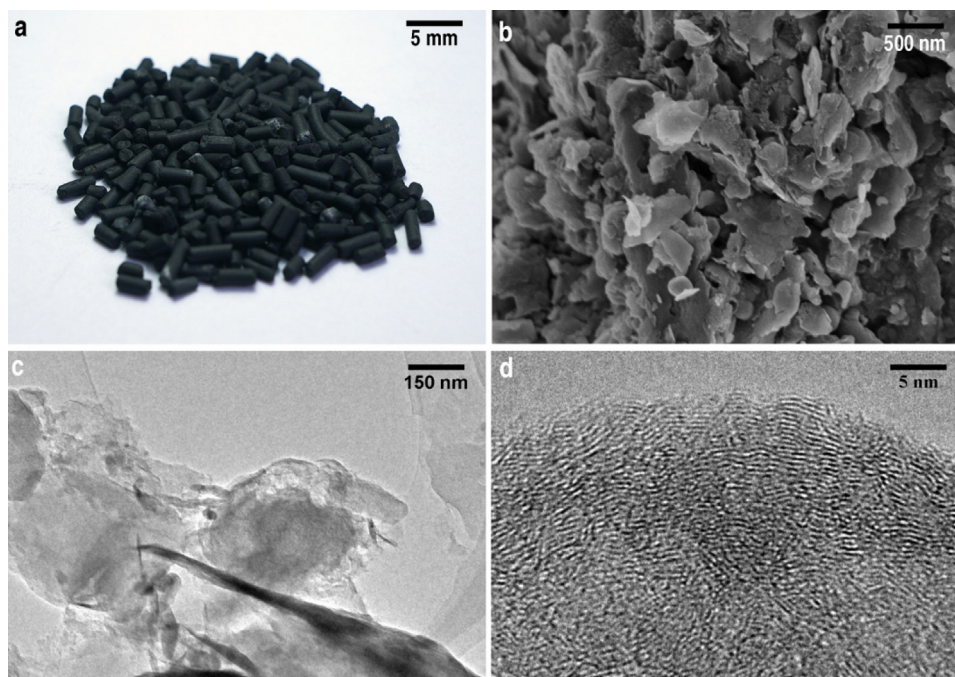


Fig. 2. (a) Photograph, (b) SEM, (c) TEM and (d) HR-TEM images of CMS.

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