Chemical Engineering Science 231 (2021) 116301

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

Porous reduced graphene oxides derived by selective removal and formation of oxygen functional groups and their electrochemical capacitances

Sang Youp Hwang [a,1], Cheol-Ho Lee [b,1], Hae Ri Lee [a], Su-Young Son [a], Sungho Lee [b], Han-Ik Joh [a,*]

[a] Department of Energy Engineering, Konkuk University, 120 Neungdong-ro, Gwangjin-gu, Seoul 05029, Republic of Korea
[b] Carbon Composite Materials Research Center, Korea Institute of Science and Technology, Chudong-ro 92, Bongdong-eup, Wanju, Jeollabukdo 55324, Republic of Korea

HIGHLIGHTS

- We synthesized rGOs with a porous structure using a facile synthetic method.
- The synthetic method was realized by low-temperature treatments (air and nitrogen).
- Air-activation process led to the formation of pore-forming groups, resulting in porous rGO.
- The as-synthesized porous rGO exhibits an improved surface area and capacitance.

ARTICLE INFO

Article history:
Received 22 April 2020
Received in revised form 19 October 2020
Accepted 10 November 2020
Available online 16 November 2020

Keywords:
Graphene oxide
Reduced graphene oxide
Porous structure
Pore forming
Electrode materials
Capacitance

ABSTRACT

In this study, reduced graphene oxides (rGOs) with a porous structure were synthesized through a facile sequential low-temperature treatment at 150 and 250 °C in air and nitrogen atmosphere, respectively, without toxic chemicals. For the first treatment under oxygen-rich conditions, competitive oxidation and reduction reactions between various kinds of oxygen functional groups were observed, leading to the formation of preferential pore-forming groups such as carboxyl. Weakly bound groups on the GO surface (such as hydroxyl and carboxyl groups) were removed in the second step, leading to the formation of pores and improving electrical conductivity. The rGO suitable for use as an electrode material had a surface area of 636.6 m^2/g and a capacitance of 191.3 F/g. Therefore, we believe that this mild treatment could be a potentially cost-effective, efficient, and environmentally friendly strategy to synthesize electrode materials.

1. Introduction

Graphene has been studied quite extensively with regard to electrochemical applications due to its abnormal properties such as large specific surface area (SSA: 2630 m^2/g) and high electrical conductivity (~10^6 S/m) (Geng et al., 2011; Zhu et al., 2011; Du et al., 2008; Marsden et al., 2018). However, only single-layer graphene with perfect crystallinity exhibits these ideal properties. Graphene oxide (GO) exfoliated from graphite shows good mass productivity and good dispersity in polar solvents, but it suffers from disadvantages such as poor electrical conductivity and many artificial defects (Rao et al., 2018). Reduced graphene oxide (rGO) was developed to overcome the drawbacks of GO and is regarded as a potential electrode material. With regard to the fabrication process of electrochemical devices, rGO has many suitable properties, such as its moderate solution process and compatibility with certain solvents. However, rGO typically contains both oxygen functional groups and defects at the edges and basal planes of the graphene sheets, leading to insufficient electrical conductivity and SSA associated with restacking (Kim et al., 2013). Therefore, to allow rGO to be used as an electrode material, active rGO (arGO) was developed to enhance the electrical conductivity and SSA through the reduction of the functional groups and the formation of a porous structure, respectively.

Previous studies have proposed strategies to control the pore structure and SSA. Zhu et al. (2011) noted that microwave-exfoliated GO can be chemically activated by KOH at 53 kPa and 850 °C for 1 h. The chemically activated microwave-exfoliated GO exhibited micro- and meso-pore sizes of ~1 and 4 nm, respectively. KOH activation increased the SSA (3100 m^2/g) by up to 18%

* Corresponding author.
E-mail address: hijoh@konkuk.ac.kr (H.-I. Joh).
[1] These authors contributed equally to this work.

https://doi.org/10.1016/j.ces.2020.116301
0009-2509/