



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



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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.

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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 m²/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.

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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 m²/g) and high electrical conductivity (~10⁶ 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 m²/g) by up to 18%

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