Tuning Graphene Oxide via N/S Doping and Thermal Treatment for Energy Storage

Krishna Kumar M, Dhana Shree N
Department of Physics and Electronics, Christ University, Bengaluru-560 0269, India
krishnakumar.m@christuniversity.in

Abstract

This study aims to systematically elucidate the influence of heteroatom doping and controlled thermal treatment on the structural ordering, defect chemistry, surface composition, and electrochemical performance of graphene oxide for advanced energy storage applications. Specifically, the objective is to establish a quantitative correlation between nitrogen (N) and nitrogen-sulphur (N,S) co-doping, thermally induced deoxygenation at 350°C, and the resulting modulation of defect density, electronic conductivity, and capacitive behaviour in reduced graphene oxide (rGO). Graphene oxide (GO) was synthesized and subsequently subjected to pressurised reduction, enabling simultaneous incorporation of N and N,S heteroatoms. X-ray diffraction confirmed the oxidation of graphite through the characteristic (001) reflection and its evolution toward graphitic (002) ordering after reduction and annealing. It is identified that, a significant mass loss associated with oxygen functional groups and established 350°C as an effective temperature for deoxygenation without structural degradation. Raman spectroscopy revealed with systematic variation in the ID/IG ratio indicating defect generation, partial restoration of sp^2 domains, and successful heteroatom incorporation within the two-dimensional carbon lattice and demonstrated a substantial increase in the C/O atomic ratio. Morphological analysis confirmed the preservation of layered sheet-like architecture with enhanced exfoliation upon annealing. Electrochemical measurements in KOH electrolyte demonstrated that synergistic heteroatom doping and thermal reduction markedly improve charge transport and electric double-layer capacitive behaviour. The N,S-rGO-350°C electrode exhibited a maximum specific capacitance of 1059.8 Fg^{-1} at 5 mVs^{-1}, surpassing undoped counterparts with minimal IR drop, energy density of 26.2 $Whkg^{-1}$ and a power density of 625 Wkg^{-1} and maintaining 97.9% capacitance retention over 5000 cycles. These results demonstrate that defect engineering effectively tailors the electronic structure and ion-accessible surface of graphene oxide, leading to the use of high-performance electrode applications.

References

[1] D. Zhan, J. Yan, L. Lai, Z. Ni, L. Liu, Z. Shen, Engineering the Electronic Structure of Graphene, Adv. Mater. 24 (2012) 4055-4069.
[2] R. Tarcan, O. Todor-Boer, I. Petrovai, C. Leordean, S. Astilean, I. Botiz, Reduced graphene oxide today, J. Mater. Chem. C. 8 (2020) 1198-1224.

Figures

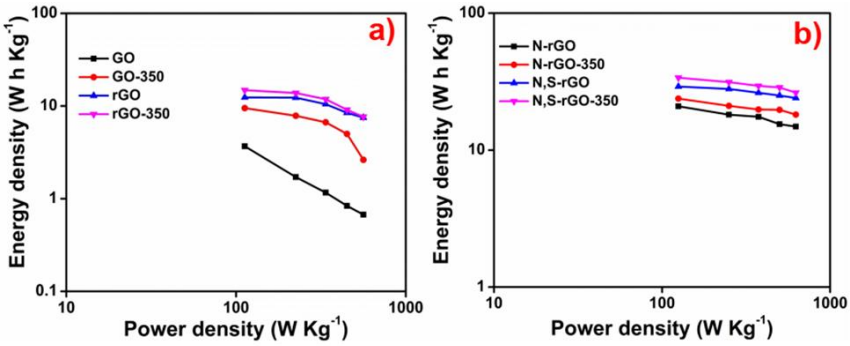

Figure 1 Fig.21 Energy density and power density of a) GO, GO-350, rGO and rGO-350 b) N-rGO, N-rGO-350, N,S-rGO and N,S-rGO-350