

Critical role of functional groups containing N and S on graphene surface for stable and fast charging Li-S batteries

Jinhua Sun^a, Jang-Yeon Hwang^b, Piotr Jankowski^{c,d}, Linhong Xiao^e, Jaime Sanchez Sanchez^a, Zhenyuan Xia^a, Suyeong Lee^b, Alexandr Talyzin^f, Aleksandar Matic^g, Yang-Kook Sun^{*h}, Vincenzo Palermo^{*ai}, Marco Agostini^{*g}

^aDepartment of Industrial and Materials Science, Chalmers University of Technology, 41296 Göteborg, Sweden.

^bDepartment of Materials Science and Engineering, Chonnam National University, South Korea.

^cFaculty of Chemistry, Warsaw University of Technology, 00-664 Warsaw, Poland.

^dDepartment of Energy Conversion and Storage, Technical University of Denmark, Denmark.

^eDepartment of Chemistry, Umeå University, 90187 Umeå, Sweden.

^fDepartment of Physics, Umeå University, 90187 Umeå, Sweden.

^gDepartment of Physics, Chalmers University of Technology, 41296 Göteborg, Sweden.

^hDepartment of Energy Engineering, Hanyang University, Seoul 04763, South Korea.

ⁱInstitute of Organic Synthesis and Photoreactivity (ISOF), CNR, via Gobetti 101, 40129, Bologna, Italy
jinhua@chalmers.se

Lithium-sulfur (Li-S) batteries are considered one of the most promising energy storage technologies with the possibility to replace the state-of-the-art lithium-ion batteries. However, the migration of high-order lithium polysulfides (LiPs) to the lithium surface and the sluggish electrochemical kinetics pose challenges to their commercialization. Instead of doping carbon hosts with heteroatoms to enhance their interactions with LiPs, which is limited with relatively low doping content (<10%), we propose in this study the grafting of versatile functional groups with designable properties (e.g., catalytic effects) directly on the surface of the carbon host.[1] As model systems, nitrobenzene and benzene sulfonate with N/O and S/O atoms were grafted vertically on the surface of highly conductive expanded reduced graphene oxide (eRGO). The immobilized functional groups with high density are uniformly distributed, which provide anchor points for the soluble LiPs. The Density Functional Theory (DFT) calculations demonstrated that the presence of these functional groups improved the binding energy of eRGO with both elemental sulfur and Li_2S_n polysulfide species. As a result, a stable interface was formed by absorbing/depositing the LiPs on the vertical nitrobenzene or benzene sulfonate functional groups. The combination of experiments and DFT calculation showed an improved retention of Li_2S_n at the cathode side with enhancement in electrochemical kinetics. The practical power and energy densities Li-S batteries significantly improved after introducing vertical functional groups on carbon hosts.

References

[1] J.H. Sun, V. Palermo, et al., *Small*, (2017), 2007242.

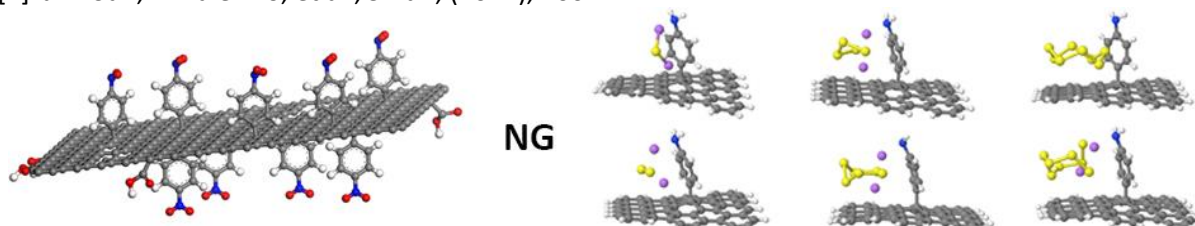


Figure 1. Molecular structure of functionalized graphene and its interaction with lithium polysulfides.