

# Tailoring the properties of graphene aerogel cathodes for eco-friendly and high-performing Na-O<sub>2</sub> batteries

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Rechargeable Na-O<sub>2</sub> batteries have received a great deal of attention as an alternative to current lithium-ion batteries, mainly due to their potential to provide higher energy densities. Nevertheless, Na-O<sub>2</sub> batteries are still in an early stage of development, with several challenges that need to be addressed, including unsatisfactory kinetics at the air cathode. In this regard, the development of a suitable cathode material that is able to efficiently catalyze the oxygen reduction and evolution reactions (ORR & OER), while accommodating larger amounts of discharge products and minimizing pore clogging, is a point of major concern.

Carbon materials have been widely used as air cathodes due to their low cost, high surface area, chemical and mechanical stability, high electrical conductivity, well-developed porosity and intrinsic catalytic activity towards ORR/OER. In particular, graphene has arisen as a versatile and promising cathode, as it not only boasts these appealing features, but can also be readily processed into macroscopic structures (e.g., aerogels) that are useful for this purpose.

We have prepared graphene nanosheets by two different methods: graphite oxide route [1] and electrochemical exfoliation of graphite [2]. The ensuing colloidal graphene-based suspensions were processed into aerogels, which were then tested as Na-O<sub>2</sub> battery cathodes to study the impact of porosity, surface chemistry and preparation procedure on the battery performance. First, graphene aerogels with different pore sizes obtained by the graphite oxide route revealed a trade-off between pore texture and battery performance. This route, however, is not environmentally friendly and involves multiple steps, including a reduction step to tune the sheet conductivity. As an alternative, we have used a simpler, faster and more eco-friendly route to access high-quality graphene by electrochemical exfoliation of graphite, resorting to natural nucleotides as both exfoliating electrolytes and colloidal stabilizers. An aerogel prepared from this graphene suspension delivered a large discharge capacity and longer cycle life than those cathodes prepared by the graphene oxide route. The origin of such good performance is attributed to the participation of the nucleotide molecules in key chemical processes taking place at the battery cathode, including oxygen electrocatalysis and nucleation of the discharge products. The present work highlights not only the attraction of this electrochemical method over more traditional routes when it comes to manufacturing graphene, but also the direct potential benefits of the resulting graphene in novel electrochemical energy storage technologies.

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## References

- [1] M. Enterría, C. Botas, J.L. Gómez-Urbano, B. Acebedo, J.M. López del Amo, D. Carriazo, T. Rojo, N. Ortiz-Vitoriano, *Journal of Materials Chemistry A*, 42 (2018) 20778.
- [2] M. Enterría, J.L. Gómez-Urbano, J.M. Munuera, S. Villar-Rodil, D. Carriazo, J.I. Paredes, N. Ortiz-Vitoriano, *Small*, 17 (2021) 2005034.