Efficient cathodic exfoliation of graphite in aqueous electrolytes towards high quality graphene for energy and environmental applications

Sergio García Dalí
Juan I. Paredes, José M. Munuera, Silvia Villar-Rodil, Amelia Martínez-Alonso, Juan M.D. Tascón

Instituto Nacional del Carbón, INCAR-CSIC, C/Francisco Pintado Fe 26, Oviedo, Spain.

sergio.dali@incar.csic.es

The electrochemical exfoliation of graphite in aqueous medium stands out as an attractive, scalable approach for the production of graphenes for different applications, due to its simplicity, cost-effectiveness and environmental friendliness [1,2,3]. In particular, cathodic exfoliation in water should allow access to high quality, non-oxidized graphene flakes, as it avoids the intrinsic oxidizing conditions that typically plague the anodic route, but this possibility has been limited by a poor intercalation ability of aqueous cations [4]. Here, we demonstrate that with a proper choice of starting graphite and electrolyte, high quality graphene flakes can be obtained in substantial yields via cathodic delamination in water. Graphites having some pre-expanded edges and interlayer voids (e.g., graphite foil) were found to be critical for a successful exfoliation. Large differences in the efficiency of a range of aqueous quaternary ammonium-based electrolytes were observed, quantitatively compared and rationalized on the basis of their chemical structure. Graphene yields up to 40–50 wt% were attained with the most efficient quaternary ammonium-based cations (tetrapropylammonium and hexyltrimethylammonium). Hydrophobic sponges made up of cathodic graphene-coated melamine foam exhibited a notable capacity towards the sorption of oils and organic solvents from water with good re-usability. Hybrids comprised of cathodically exfoliated graphite and a small amount of vertically oriented cobalt oxide nanosheets displayed good electrochemical charge storage behavior. Overall, the ability to access graphene flakes in considerable yields by the aqueous cathodic route disclosed here should raise the prospects of cathodic exfoliation as a competitive method for the industrial manufacturing of high quality graphene for practical applications.

References

[1] S. Yang, M.R. Lohe, K. Müllen, X. Feng, Adv. Mater. 28 (2016), 6213-6221
[2] A.M. Abdelkader, A.J. Cooper, R.A.W. Dryfe, I.A. Kinloch, Nanoscale 7 (2015), 6944-6956
[3] M. Zhou, J. Tang, Q. Cheng, G. Xu, P. Cui, L. Qin, Chem. Phys. Lett. 572 (2013), 61–65
[4] G. M. Morales, P. Schifani, G. Ellis, C. Ballesteros, G. Martínez, C. Barbero, H. J. Salavagione, Carbon 49 (2011), 2809–2816

Figures

Figure 1: Digital photographs of: olive oil floating on water (left), a graphene-coated melamine foam adsorbing the oil phase (middle), and the final oil-free water after extraction of the sorbent (right).