

Evaluating Influence of Solvents in Graphene Synthesis via Electrochemical Exfoliation: Optimal and Suboptimal Performers

ON Malawalaarachchi^{1,2} and NPW Rathuwadu^{1#}

¹Institute for Combinatorial Advanced Research and Education, General Sir John Kotelawala Defence University, Sri Lanka

²Faculty of Graduate Studies, General Sir John Kotelawala Defence University, Sri Lanka

#rathuwadunpw@kdu.ac.lk

Electrochemical exfoliation is an eco-friendly technique for graphene synthesis, with the major challenge being restacking of graphene layers. This study's novelty lies in optimizing solvents to prevent restacking by considering solvent parameters, including Hansen and Hildebrand solubility parameters, surface tension, graphene dispersibility, and electrostatic and steric parameters, collectively. The optimized electrochemical exfoliation approach was applied to eleven different solvents using two natural graphite materials (KG and BG) and characterized structurally and electrochemically. The gravimetric analysis showed high crystallinity and purity of KG graphite. The peak at 270 nm in Ultraviolet-visible spectroscopy, a broad peak at $2\theta=26.45^\circ$ in X-ray diffraction, sheet-like nanostructures in scanning electron microscopy (SEM), and characteristic graphene peaks in Fourier transform infrared spectroscopy (FT-IR) confirmed the successful synthesis of graphene. The calculated mean lateral size of graphene flakes was $\sim 0.723 \pm 0.259 \mu\text{m}$. The Raman analysis confirmed the formation of high-quality graphene with a very low number of defects ($I_D/I_G=0.08$), and thickness of a few layers ($I_{2D}/I_G=0.45$). Electrochemical impedance spectroscopy showed the highest ion diffusion behaviour and lowest electron transfer resistance of the BG-graphene-5% acetonitrile (ACN) and KG-graphene-5% N-methyl-2-pyrrolidone (NMP) electrodes. Cyclic voltammetry confirmed the superior electrochemical performances of the KG-Graphene-NMP and BG-Graphene-ACN due to their highest peak currents of 88.04 (860.6%) and 74.16 (343.5%) μA , and lowest peak-to-peak separations of 73.2 and 70.8 mV, respectively. Comparing KG and BG, solvents with compatible surface tension, Hansen, and Hildebrand solubility parameters supported successful graphene synthesis; hence, the best solvent depends on both the compatibility of these parameters and the type of raw material used.

Keywords: *graphite, graphene, electrochemical exfoliation, solvent parameters*