

Scalable production of graphene for inks and polymer composites

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The largest and most promising markets for large-scale graphene applications are polymer composites and conductive inks. Polymer composites, paints and coatings require thin graphene flakes with large lateral sizes to enable low electrical percolation thresholds and improved mechanical properties such as increased strength. Conductive inks require dispersions of graphene with tunable lateral size in a range of μm to nm , depending on the printing technology. However, graphene is typically only dispersible in toxic, environmentally hazardous organic solvents, a problem compounded by the fact that only low concentrations are achievable ($<1 \text{ mg/ml}$). While optimizing the processability of graphene with these solvents is undoubtedly interesting and one of our ongoing investigations, giving graphene hydrophilicity will make it compatible with different polymer matrices and allow water-based inks. Apart from using surfactants, this can be achieved by functionalization of the graphene surface with hydrophilic groups.

However, no routes of graphene production fulfil all of these requirements while still being readily scalable. For example, graphene oxide (GO) exhibits large lateral flake-sizes and dispersibility, but lacks conductivity. On the other hand, liquid phase exfoliated few-layer graphene (LPEG) possesses extremely small lateral sizes ($\sim 200 \text{ nm}$). Moreover, the surface functionalization of these materials needs an additional production step, which

reduces the potential of graphene being used in products in industry and consumer environments. In contrast to these methods, the electrochemical exfoliation of graphite can produce few layer, large lateral size graphene flakes with low defect- and high conductivity¹. Recently, we have further developed this facile, inexpensive and scalable production method towards the incorporation of different functionalizing agents into the exfoliation cell, without adding steps to the process.

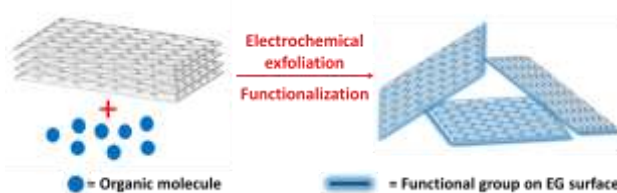


Figure 1: One-step electrochemical exfoliation and in-situ functionalization

We improved the quality and yield of the process by statistical parameter optimization. In this way, we were able to achieve functionalized graphenes, suitable to produce not only stable dispersions in DMF with stability of up to 3 months but also $\sim 1 \text{ mg/ml}$ EG inks in water without surfactants and a variety of high-concentration, high-viscosity pastes. Notably, even upon hydrophilic functionalization, the graphene retains an outstandingly high electrical conductivity of up to 85.000 S/m after filtration-deposition, without any post-treatment. The simplicity of the process makes it a prime candidate for upscaling, as parameter optimization shows ever-increasing production rates. If successful, the possibility to produce processable, few-layer, large-flake graphene with tailorable, defined functional surface groups on a large scale, can open up a completely new chapter towards graphene's commercial impact.

References

- [1] Yang, S.; Lohe, M. R.; Müllen, K.; Feng, X. *Adv. Mater.* 2016, 28(29), 6213-6221