

## Graphene-based Materials for Supercapacitor Applications

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### Abstract

High quality graphene sheets with superior physical properties such as large surface area and high electrical conductivity allowed the development of new engineered carbons for energy storage devices. Towards this goal, high surface area graphene oxide materials were prepared using various pristine graphite and oxidation methods. Natural graphite with different sizes and microwave expanded graphite were oxidized using modified Staudemayer or Hammers methods. The resulting graphite oxide paper was exfoliated and reduced via microwave treatment leading to few layers graphene/graphene oxide nanostructures (EGO). Impregnation in KOH and high temperature treatment in vacuum was also applied to increase the porosity and diminish the oxygen content in the final materials (t-EGO).

The obtained materials were further processed to create metal and metal oxide/graphene heterostructures [1, 2]. Graphene-based materials were either decorated with silver (Ag) nanoparticles or intercalated with iron carbide (Fe<sub>3</sub>C) nanostructures.

The structural, morphological and electrical properties of the bare and composite graphitic materials were investigated. The XRD and TEM analysis confirmed the oxidation of graphite and further exfoliation to few layers graphene, respectively. It was also established that the oxidation route and especially the type of the pristine graphite (flake size, pretreatment) influence significantly the level of oxidation, the specific surface area and the electrical properties of the produced graphene. Liquid N<sub>2</sub> adsorption-desorption isotherms demonstrated that small flakes (100 mesh) natural graphite and expanded graphite resulted in large specific surface area (>900 m<sup>2</sup>/g up to ~2400 m<sup>2</sup>/g) of the prepared EGO. The use of larger flakes (10 mesh) natural graphite led to lower BET surface but higher capacitance (~629 F/g) assessed by CV measurements. The Raman results revealed that KOH acted more as a reducing agent diminishing the defects than as surface modifier for high porosity. It was also established that the decoration of EGO and t-EGO with Ag nanoparticles (~40 nm) through photo-deposition decreased the structural defects of the graphene. However, the procedure did not improve the capacitance of the resulting materials. The activated t-EGO/Ag composite exhibited supercapacitor's behavior with lower capacitance than the bare graphene, while the non-activated EGO/Ag acted as a resistant. The outcome was related with the deposition of Ag not between but onto the surface and the edges of the graphene layers and the decrease of the materials porosity.

In the case of graphene oxide/Fe<sub>3</sub>C hybrids it was established that the immobilization of Fe-based intercalant (IFe) was governed by the pH of the aqueous graphite oxide dispersion following nucleophilic substitution or ion exchange path. Subsequent thermal annealing resulted in formation of pillaring Fe<sub>3</sub>C nanoparticles encapsulated in a graphite shell. It is suggested that the graphite shell prevent the aggregation between both adjacent Fe<sub>3</sub>C nanoparticles, and bundles of neighboring multi-layer graphenes. The exhibited properties of the obtained hybrid materials make them appropriate for magnetic and supercapacitor applications. These materials have also the advantage of being low cost, low toxicity and environmentally friendly.

### References

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