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Covalently Bonded Polyaniline and *para*-phenylenediamine
Functionalized Graphene Oxide: How the Conductive Two-dimensional
Nanostructure Influences the Electrochromic Behaviors of Polyaniline

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Abstract: Polyaniline (PANI) was attached onto the reduced graphene oxide (rGO) sheets through copolymerization of aniline with a *para*-phenylenediamine (PPD) functionalized graphene oxide (GO-PPD) using the poly(styrene sulfonate) (PSS) as the macromolecular dopant agent to produce a water-dispersible electrochromic material. The structures and the morphologies analysis confirm that the final electrochromic materials (rGO-PANI) are the mixture of PANI/PSS and the covalently bonded rGO-PANI (rGO-PANI hybrid). The rGO-PANI hybrid can be found to form a parallel arrangement to the substrate in the spin-coated film. This parallel arrangement of the layered two-dimensional nanostructure of rGO-PANI hybrid may narrow the ion transportation pathways of the exchanged ions, which will result in a high charge transfer resistance and slow switching speed. Meanwhile, with the conductive rGO added, the electrical conductivity of the electrochromic layer will be increased, which will benefit to low charge transfer resistance and high optical contrast. So the conductive two-dimensional nanostructure has a double-face influence on the electrochromic performances of PANI, which include a positive influence on the electrical conductivity and a negative influence on the ion diffusion. The overall influences depend on the loading amount of GO-PPD. With 4 wt.% GO-PPD feeding,

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