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Extended Abstract

Silicon-Doped Graphene Nanoflakes: Controllable Doping and Application as Metal-Free Catalyst and Electrode Material †

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Doping of graphene-based nanomaterials is one of the most effective approaches for the modification of their physicochemical properties, defectiveness, and surface structure. Nitrogen is usually used as a dopant due to the simplicity of the synthesis technique and high stability of the produced materials, while boron, sulphur, and phosphorous are less popular dopants. Silicon doping is rather exotic because the formation of Si–C bonds is difficult, and mostly SiO_2 is produced. At the same time, the doping of graphene-based materials with silicon distorts carbon layers and generate unique out-of-plane defects.

In this study, Si-doped graphene nanoflakes (Si-GNFs) were synthesized by the pyrolysis of the hexane+tetramethylsilane mixtures over a MgO template using a tubular quartz reactor. To prepare the electrodes, 80 wt.% active materials, 10 wt.% super-P, and 10 wt.% polyacrylic acid binder were mixed homogeneously in an N-methylpyrrolidinone solvent to form a uniform slurry. The slurry was coated on the copper foils and then dried at 100 °C under vacuum for 16 h. Galvanostatic charge/discharge measurements were carried out at a potential range of 0.01–3.0 V using a Landt Instr. CT2001A battery tester.

The present study is devoted to the development of a simple and controllable technique for the synthesis of Si-doped graphene nanoflakes (Si-GNFs) with a small domain size (20–50 nm) and 3–10 monolayers of thickness (Figure 1) with different silicon content and high Si–C and C–Si–O_x bond concentrations up to 4.5 at.%.

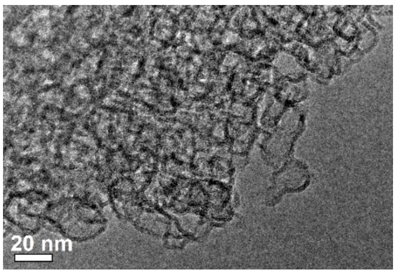

Figure 1. TEM image of Si-doped graphene nanoflakes (GNFs).

The produced materials were tested as metal-free catalysts in the dehydrogenation and dehydration of aliphatic alcohols and as electrode materials of Li-ion batteries. It was found that the defectiveness of Si-GNFs affected their catalytic and electrochemical properties. Si-GNFs electrodes showed a specific capacitance up to 650 mAh/g at 0.5 A/g after 300 cycles and capacity retention of ~65% at 5 A/g after 3000 cycles.

The developed technique allows for the synthesis of new type of graphene materials: Si-doped graphene nanoflakes with controllable doping level, localization, and defectiveness. By tuning the silicon concentration, one can change the catalytic properties and capacity of the materials.

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