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ABSTRACT

of doctoral thesis

Influence of graphitic precursor on the composition, morphology and structure of thermally reduced graphene oxides

In the view of the preparation method of graphene materials through graphite oxidation followed by exfoliation and reduction of graphite oxide is widespread, the study on the influence of the graphitic precursor on the composition, morphology and structure of resultant graphene materials, particularly reduced graphene oxides, has been performed. The method of reduced graphene oxide preparation used in this work involved the graphite oxidation and subsequent simultaneous exfoliation and reduction of the graphite oxide at high temperature. Ten different graphite precursors of natural and synthetic origin were the subject of this investigation. Three graphite samples were selected for further studies based on their structural characteristics determined by XRD and Raman spectroscopy. Among them, two graphites were natural, flake and shell type, and a synthetic graphite. The selected graphites were characterized by the largest size of the graphene sheets and the smallest number of structural defects.

The graphites were subjected to an oxidative treatment using different methods (A, B, C and D), including the original method (D) elaborated by the PhD candidate. The amount of oxygen introduced into the graphite structure was related to the graphite type and oxidation method, being in the range of 18 to 36 at.%. The distribution of oxygen functional species and the contribution of carbon in the sp^2 hybridization in the graphite oxides were determined by XPS. Introduction of the oxygen groups between the graphene planes and into the edges resulted in an increase of interlayer distance from 0,336 nm to 0,818 nm. Raman spectroscopy measurements revealed a higher contribution of disordered structure in the graphite oxides compared with pristine graphites. The graphite oxides obtained by A and D oxidative treatments exhibited both the highest oxygen content and interlayer spacing d_{001} as well as the most separated and corrugated graphene sheets as observed by SEM. The graphite oxides prepared by A, B and D methods were thermally exfoliated/reduced at a temperature of 900 °C. The resultant reduced graphene oxides exhibited two-three fold lower oxygen content (6,3-9,2 at.%) in

comparison with their respective graphite oxides. An increase of carbon contribution in sp^2 hybridization, from 37-53 % in the graphite oxides to 70-80 % in the reduced graphene oxides, was observed. The smallest number of the structural defects was revealed for the graphene materials obtained from the natural flake graphite, which was characterized by the largest graphene sheet size. The elimination of oxygen functionalities during exfoliation/reduction process was accompanied by a decrease in the interlayer distance from ~0,8 to ~0,4 nm for the graphite oxides and the reduced graphene oxides, respectively. The reduced graphene oxides presented a well-developed mesoporous structure. AFM studies revealed three-layer graphene flakes with a thickness of approximately 4 nm in all reduced graphene materials.

The research allowed developing preliminary criteria for selecting a graphite precursor for the production of the graphene materials with required textural and structural parameters using a thermal exfoliation/reduction of the graphite oxide.