Surface Charge Storage Properties of Selected Graphene Samples in pH-neutral Aqueous Solutions of Alkali Metal Chlorides - Particularities and Universalities

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doi: 10.20964/2016.10.47

Received: 10 July 2016 / Accepted: 16 August 2016 / Published: 6 September 2016

In order to provide deeper understanding of surface charge storage mechanism in graphene materials, we analyzed the relative contribution of double layer capacitance and pseudocapacitance in overall capacitance of three different graphene-based materials in pH-neutral aqueous solutions of alkali metal chlorides. Both cyclic voltammetry at various sweep rates and DFT calculations were used for the analysis. Selected materials differ mutually to a great extent in specific surface area, surface concentration of oxygen functional groups and level of structural disorder. Measured capacitances were found to range between approx. 19 F g^{-1} , for graphene nanoplatelets, and 400 F g^{-1} , for industrial graphene. In general, measured capacitance changed directly proportionally to the cation size. Despite the differences in physical and chemical properties of graphenes subjected to investigations, similar contribution of pseudocapacitance to the overall capacitance at potentials lower than $0.4 \text{ V vs. Ag/AgCl}$ was evidenced. The contribution of the pseudocapacitance increased proportionally to the concentration of oxygen functional groups. At high sweep rates, we observed the following general relationship between the capacitance retention and the fraction of pseudocapacitance: the capacitance retention is more pronounced for lower contribution of pseudocapacitance. The overall capacitive response is primarily dominated by (i) specific surface area and (ii) the concentration of oxygen functional groups. The DFT calculations revealed that the strength of the interaction of alkali metal with the oxygen functional group depends critically on the type of surface group. In order to achieve top capacitive performance of graphene-based materials one should carefully optimize the concentration of oxygen functional group so that the conductivity is not compromised while keeping specific surface area as high as possible.

Keywords: graphene, capacitance, pseudocapacitance, oxygen functional groups

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