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BOOK OF ABSTRACTS

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ALKALI METAL CATIONS IN ELECTROCHEMICAL REDUCTION OF GRAPHENE OXIDE: EFFECTS ON CAPACITIVE PERFORMANCE

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It is well established that different heteroatom functionalities on graphene basal plane and edges, as well as vacancies on the basal plane, represent active sites for processes important for the application of these materials in electrochemical systems. Presence of these moieties above certain level can significantly reduce electrical conductivity, which is inherent to the graphene due to delocalized π-electron system and is essential for the use of graphene-based materials in supercapacitors. Simple and precise control of the number of oxygen functionalities and their distribution over the basal plane, which is in most cases directly achieved through control of the reduction conditions (mainly time and temperature for thermal and potential for electrochemical reduction) of chemically prepared graphene oxides opens up possibilities for the achievement of optimal sp³/sp² carbon atom ratio (ie. graphene oxide phase/graphene phase ratio) at which the highest specific capacitances, as well as other important features, can be achieved [1-3].

Satisfactory control of the extent of reduction for graphene oxide films can be achieved through simple control of reduction potential and time. Here, we present some experimentally observed trends in capacitive behavior of chemically prepared graphene oxide after its potentiostatic reduction at different potentials in aqueous LiCl, NaCl and KCl, as well as differences in irreversible reduction behavior in these electrolytes. In all cases, the highest increase in capacitance is observed at moderate reduction potentials, which is in accordance with previous reports [4,5] as well as with the observation that pseudofaradaic processes on oxygen functionalities contribute to the total capacitance [6]. Furthermore, we observed that the potential of voltammetric reduction peak shifts to the more positive values when the electrolyte cation is changed in the Li⁺ \rightarrow Na⁺ \rightarrow K⁺ direction, while the maximum capacitance increase upon reduction decreases for the same direction of change in electrolyte cation.

Electrochemical quartz crystal nanobalance measurements indicate sharp increase in mass of the electrode film in the very first moments of potentiostatic reduction, which is proportional to the applied potential. However, as reduction proceeds, mass gradually decreases due to the removal of significant quantities of oxygen from the material. Initial mass gain is discussed in terms of electrolyte cation influx, necessary for charge compensation. Mass changes observed during voltammetric reduction indicate significant differencies in the reduction process, when different cations are employed. All experimental observations are discussed in terms of hydrated cation radius, and some correlations are presented.

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