Evaluation of the effect of structural defects on the nitrogen-doped graphene electrode on the quantum capacitance

Ehsan Targholi*, **S. Morteza M. Khoshdel,*** **Mohmmadsafi Rahmanifar†** and **Hamideh-sadat Dashtmazari‡**
**Industrial Electrochemistry Lab, Department of Chemistry,*
Iran University of Science and Technology, Tehran, Iran
†Faculty of Basic Sciences, Shahed University, Tehran, Iran
‡ Department of Chemistry, Alzahra University, Tehran, Iran
Corresponding author E-mail: mmousavi@iust.ac.ir

Abstract

Supercapacitors have attracted a great deal of attention as an electrical storage device. The main materials that used in supercapacitor electrodes are carbons, metal oxides, and conducting polymers. Graphene-based material plays an important role in supercapacitor electrode materials thanks to their low production cost, high surface area and excellent conductivity. However, there is a major limitation of low energy density in graphene-based electrode compared to other material such as metal oxides. Recently, scientists have found that this limitation is due to the quantum capacitance. In fact, the total capacity of supercapacitors considered as the sum of two series capacitance of double-layer capacitance and electrode capacitance (quantum capacitance). Thus, Deficiencies in each of them will make a reduction in total device capacitance. In this paper, we explored the effect of nitrogen doping and structural defect as simultaneously on quantum capacitance. The mixed systems that we used are N-doped graphene with some structural defects such as Divacancy (5-8-5), Monovacancy (5-9) and Stone-Wales defect. Density functional theory (DFT) calculations were performed within the plane-wave pseudopotential formalism, as implemented in the Quantum-ESPRESSO code. Integrated quantum capacitance is given with equation 1.

$C_q^{int}(V) = \frac{1}{Ve}\int_0^V C_q(V')dV'$ (1)

Where, C_q is the differential quantum capacitance and V is the voltage that referenced to the zero-bias Fermi level. Our results show that good enhancement has been reached with doping and making structural defect in graphene sheet as an individually or simultaneously. This Significant enhancement is mainly due to the additional impurity states near the Fermi level. In such a mixed system, each type of mixed systems alter the electronic structure and create unique DOS and subsequently quantum capacitance. Exploring of projected DOS suggest that p_z states near the defect and p orbitals of heteroatom (N) contribute to the induced impurity states. In practice, a fabricated N-doped graphene electrode may have many types of this mixed system, which is outside the range of this work. Our findings suggest that, under charging conditions, these mixed systems will be more effective at positive bias. These results represent design strategies for improving the performance of supercapacitors.

Keywords: Supercapacitors, Nitrogen doped graphene, Quantum capacitance

References

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