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Design of New Materials for Use in Supercapacitor Electrodes by Tuning the Electronic Structure of Graphene

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Introduction

Today, generation and storage of energy is one of important areas of researches. Like an electrochemical battery, supercapacitors are used to store electrical energy. Supercapacitors have attracted a great deal of attention thanks to their great advantages such as high power density, no memory effect, long life cycle and good stability [1]. However, low specific energy is still their primary limiting factor for their usage as the electrical storage devices in industrial and commercial applications. Graphene and their derivatives are properly suited to be used in supercapacitor applications due to their many advantages such as large electrical conductivity and high surface area [2]. Recently, it has found that for increasing the specific energy of graphene-based supercapacitors, we must adjust their electronic structure. In other word, deficit in the density of states (DOS) near the Fermi level will reduced the ability to store electrical charge. In fact, Electronic structure is associated with the ability of electrode to store electrical charge by quantum capacitance concept. The quantum capacitance is an accepted concept in the field of graphene-based transistor devices. However, in the field of electrochemical supercapacitors, the impact of quantum capacitance on device performance has just recently begun to attract attention [3]. Therefore, in this work we tried to optimize the electronic structure of graphene with making defects in functionalized graphene with several divalent functional groups.

Method

Density functional theory (DFT) calculations were employed within the plane wave ultra-soft Pseudo-potential scheme, as utilized in the Quantum-ESPRESSO package. All the DFT calculations reported in this work have been performed using the (GGA-PW91) exchange-correlation functional. All numerical calculations have been done using a student version of MATLAB.

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Results and Discussion

Structures that we have considered, including mono-vacancy, di-vacancy and SW structural defects in functionalized graphene with $\succ O, \succ NH, \succ CH_2$. At first, the electronic structure and DOS of considered structure are calculated. Then the quantum capacitance of them was calculated using the obtained DOS. Finally, the stored charge on the supposed electrode was obtained. With the creation of structural defects on graphene, the quasi-localization of the p_z states appear near the defect. As a result, the impurity states arise near the Fermi level. Functionalized graphene with functional group of $\succ O, \succ NH, \succ CH_2$, Show π-derived impurity states. The bond length between two neighboring carbon atoms in functional site increase to 1.60 Å, which larger than the C-C bond length of 1.42 Å of graphene with sp^2 hybridization. This means a weakening of the bond that leading to fractional recovery of the π-electron system of graphene. As a result, the π-derived states appear on both sides of the Fermi level. Very amazing and interesting results were obtained. The curves of stored charges for combination of SV(5-9) and SW(55-77) structural defect and –NH functional group are shown in fig. 1. Other groups have good results as well, but due to the limited length of the paper, we could not insert their curves. In short, our results show that with creation of structural defects on functionalized graphene, a significant improvement in electrode capacity will achieved. Some structural defects increase the capacity at the high voltage magnitudes. In contrast, some other structural defects increase the capacity at the lower voltage magnitudes. So, we could design the asymmetric Supercapacitors with high capacity using clever combination of functionalized graphene and structural defects.

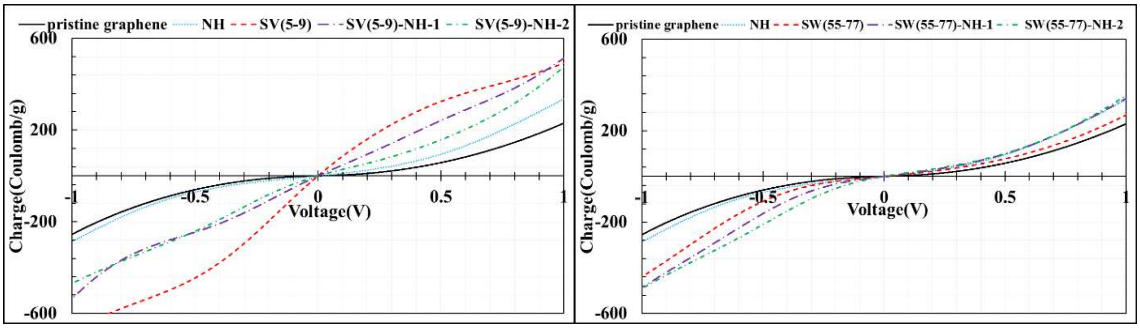

Fig. 1. Stored charge per unit weight for functionalized graphene with –NH functional group and SV(5-9), SW(55-77) structural defects along with pristine graphene for comparison.

References

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