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Ionic Liquids – Graphene Interface: Effect of ions of Ionic liquids on the Fermi Level

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Electrochemical energy storage devices receive significant attention due to their sustainability and reliability. In order to meet the energy demand, it is imperative to fabricate high-energy and high power-density electrochemical energy storage devices.

Graphene-based supercapacitors using ionic liquids (ILs) as electrolytes are an important class of energy storage devices.[1] The research activities in this field focus on the energy density of these devices. Since energy storage and conversion take place at the graphene – ILs interface, it is important to develop experimental and theoretical procedures to understand the interfacial nanostructure in graphene-based supercapacitors where ILs are used as electrolytes.

In the first part, we will focus on how the adsorption of a homologous series of 1-alkyl-3-methylimidazolium tetrafluoroborate ($C_nMIM\ BF_4$, n = 2, 4, 6, 8 and 10) ionic liquids (cation size) influences the interfacial nanostructure and its impact on graphene electronic structure using experimental and theoretical studies. Raman spectroscopy showed that adsorption of the ILs led to n-type doping of graphene and the magnitude of such doping increased as the length of the alkyl chain attached to the cation increased (cation size).[2]

In a second part, we will focus on the interfacial nanostructure of 1-butyl-3-methylimidazolium-based ILs with different anions (X = SCN, BF_4, DCA, PF_6, TFO and TFSI) and its impact on the electronic structure of graphene using molecular dynamics (MD) simulations and Raman spectroscopy. The latter reveals that small anions cause n-type doping of graphene and the magnitude of such doping decreases as the size of the anion increases. This contrasts the trend observed for cations. MD simulations provide insight in these opposing trends.

References

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[2] Velpula, G.; Phillipson, R.; Lian, J. X.; Cornil, D.; Walke, P.; Verguts, K.; Brems, S.; Uji-i, H.; De Gendt, S.; Beljonne, D.; Lazzaroni, R.; Mali, K. S.; De Feyter, S. Graphene Meets Ionic Liquids: Fermi Level Engineering via Electrostatic Forces. *ACS Nano* **2019**, *13*, 3512-3521.

Figures

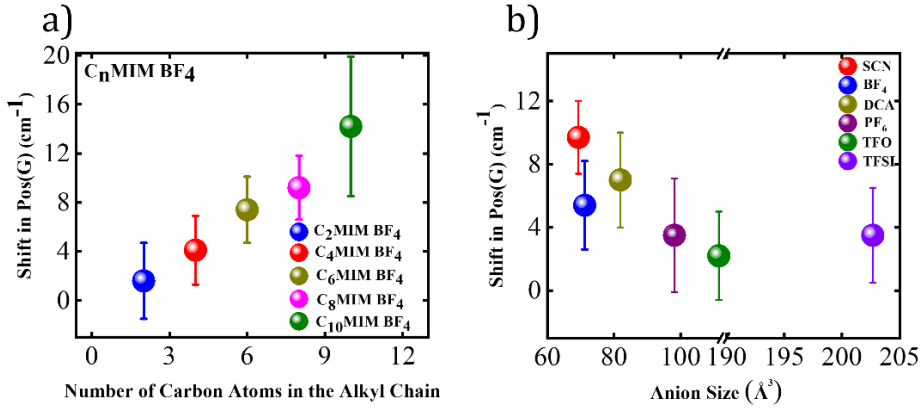

Figure 1: (a) Shift in Pos(G) versus the number of carbon atoms present in the alkyl chain on the imidazolium cation. (b) Shift in Pos(G) versus anion size.