

Graphene Meets Ionic Liquids: Fermi Level Engineering via Electrostatic Forces

Gangamalliah Velpula¹, Roald Phillipson¹, Jian Xiang Lian², David Cornil³, Peter Walke¹, Ken Verguts^{3,4}, Steven Brems⁴, Hiroshi Uji-i^{1,5}, Stefan De Gendt^{3,4}, David Beljonne², Roberto Lazzaroni², Kunal S. Mali¹ and Steven De Feyter¹

¹Division of Molecular Imaging and Photonics, Department of Chemistry, KU Leuven, Celestijnenlaan, 200F, B-3001 Leuven, Belgium; ²Laboratory for Chemistry of Novel Materials, University of Mons, Place du Parc 20, 7000 Mons, Belgium; ³Molecular Design and Synthesis, Department of Chemistry, KU Leuven, Celestijnenlaan 200F, B-3001 Leuven, Belgium; ⁴imecvzw, Kapeldreef 75, B-3001 Leuven, Belgium; ⁵RIES, Hokkaido University, N20 W10, Kita-Ward, Sapporo 001-0020, Japan
gm.velpula@kuleuven.be, kunal.mali@kuleuven.be, steven.defeyter@kuleuven.be

Graphene, a single layer of carbon atoms in a two-dimensional (2D) hexagonal lattice, showcases key properties suitable for electrode materials such as high specific surface area, good electrical and thermal conductivities.¹ Ionic liquids (ILs) are a class of molten salts that are entirely composed of ions. These liquids possess exceptional properties appropriate for electrolytes, such as high electrochemical window, electrochemical stabilities and extremely low vapor pressure.² Graphene-based 2D materials are promising candidates for a number of different energy applications. A particularly interesting one is in next generation supercapacitors, where graphene is being explored as an electrode material in combination with ionic liquids as electrolytes. Because the amount of energy that can be stored in such supercapacitors critically depends on the electrode – electrolyte interface, there is considerable interest in understanding the structure and properties of the graphene/IL interface. Here, we present the changes in the properties of graphene upon adsorption of ILs and ILs – organic solvent mixtures using a combination of experimental and theoretical tools. For example, Raman spectroscopy reveals that imidazolium tetrafluoroborate ILs ($C_n\text{MIM BF}_4$) cause n-type doping of graphene, and the magnitude of doping increases with increasing cation chain length despite the expected decrease in the density of surface-adsorbed ions (Fig 1). Molecular modeling simulations show that doping originates from the changes in the electrostatic potential at the graphene/IL interface. The findings described here represent an important step in developing a comprehensive understanding of the graphene/IL interface.³

References

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Figures

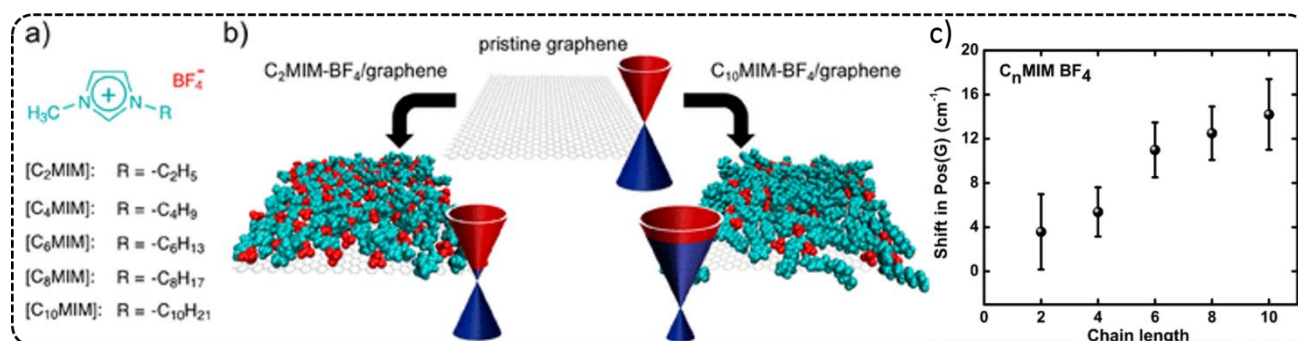


Figure 1: (a) Molecular structures of the ILs employed in this study. (b) Scheme showing the influence of the adsorption of ILs with increasing chain length on the Fermi level of graphene. (c) Shift in Pos(G) versus the number of carbon atoms present in the alkyl chain on the imidazolium cation.