

GhulamAbbas^{1,2},
Martin Kalbáč¹, Otakar Frank^{1*}, Matěj Velický^{1*}

¹J. Heyrovský Institute of Physical Chemistry of CAS, v.v.i, Dolejškova 2155/3, 183 23 Prague 8, Czech Republic

²Department of Physical and Macromolecular Chemistry, Faculty of Science, Charles University in Prague, Hlavova 2030, 128 43 Prague 2, Czech Republic

otakar.frank@jh-inst.cas.cz and matej.velicky@jh-inst.cas.cz

In-situ Raman spectroelectrochemistry of graphene in concentrated aqueous electrolytes

The ionic adsorption at electrode surface changes the chemical potential which has a substantial influence to modulate the electronic properties of material¹. The interpretation of the role of specific ion on electrolyte/material interface during electrochemical process is not a simple task due to the complex interplay between the material's properties and the electrochemical process. Therefore, a combination of *in-situ* Raman microspectroscopy with electrochemistry is an intriguing technique that can provide the valuable information about the electrochemical behavior of the layered material in an aqueous medium². We performed μ -droplet in-situ Raman spectroelectrochemistry (μ -SEC) measurements on monolayer graphene in concentrated aqueous electrolytes of different ionic type and ionic strength to demonstrate the charge transfer doping (electron, hole) with alteration in the structural properties of graphene.

We observed a shift in G peak position with gating of monolayer graphene by applied potential and monitored the position of the G peak minimum corresponding to the charge neutrality point (CNP). The different electrolytes have different CNP in graphene which could correspond to minimum in charge carrier density. However, the overall shift rate of the G peak ($\Delta\omega$) of graphene with potential was observed to be similar regardless of the ionic type and ionic strength. Also, we calculated the Fermi energy shift for 2.4M $\text{Al}(\text{ClO}_4)_3$ electrolyte and calculated the doping efficiency in graphene with reference to the gate voltage. The doping efficiency was about 60 % in the hole doping regime at the maximum gate voltage of 1.6. The decrease in doping efficiency is attributed to the potential drop across the electrolyte/graphene interface.

References

- [1] S. Pisana, M. Lazzeri, C. Casiraghi, K.S. Novoselov, A.K. Geim, A.C. Ferrari, F. Mauri, *Nat. Mater.* 6 (2007) 198–201.
[2] M. Jindra, M. Velický, M. Bouša, G. Abbas, M. Kalbáč, O. Frank, *J. Phys. Chem. Lett.* 13 (2022) 642–648.