Interfacial phenomena governing performance of graphene electrodes in aqueous electrolytes

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Abstract

Water intercalation between graphene and its supporting substrate is known to occur in graphene devices operating in aqueous media[1,2]. Due to the importance of graphene-based technology in the biomedical field[3,4], it is crucial to understand the impact that this phenomenon may have on the devices' performance.

Here, we use as-transfer graphene electrodes on insulating and conducting substrates to assess the influence of intercalated water on its electronic properties, by applying a combination of electrochemical impedance spectroscopy (EIS), Raman spectroelectrochemistry and theoretical modelling.

Results reveal an intriguing behaviour of the measured capacitance of the graphene/conductive substrate system, which is one order of magnitude higher than that of the graphene/non-conductive substrate one. Interestingly, Raman spectroelectrochemistry demonstrates that the modulation of graphene surface charge by the application of an external voltage is independent of the substrate conductivity nature. These observations are explained by considering the influence of intercalated water between graphene and the substrate, and the impact of the conductivity nature of the substrate to the measured capacitance by EIS. Detailed numerical simulations are used to give insight on the impact of the intercalated water properties on the experimental measurements.

This work represents an important advance towards the understanding of the electronic and electrochemical performance of graphene-based devices operating in electrolytes and of the methods to measure it.

References

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