Interfacial Capacitance and Defect Density Dependent Activity of Graphene
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Correlating the interfacial structure and property of an electrochemical system is vitally important for understanding and designing high performance electrochemical devices. The combination of spectroscopy and electrochemistry method, especially the in-situ methods, would allow us to gain a more clear understanding about the interface.

We first demonstrated the in-situ electrochemical Raman spectroscopy study of single layer graphene to unlock the physical origin of the electrochemical properties of graphene. The origin of the low interfacial capacitance of carbon-based materials is a long standing puzzle. The Raman parameters of single layer graphene, including the frequency and band width of the G band, the frequency of the 2D band and the intensity ratio of 2D to G bands (I_{2D}/I_{G}), show the similar potential dependent behavior to that of the capacitance curve (Fig. 1a). The clear correlation between the Raman parameters and the capacitance can be understood by the same physical origin of carrier concentration (n) of graphene.

We then showed that the combination of Raman spectroscopy and scanning electrochemical microscopy (SECM) helped us to understand the defect density dependent electrochemical activity of graphene. To fully understand the impact of defect on the electrochemical activity of graphene, we introduced controllable defects in single layer graphene by Ar⁺ ion bombardment. The heterogeneous electron transfer rate (k) of graphene was improved because of defect. The improvement could be understood by the increase of the local density of states. The defect density could be routinely determined by Raman spectroscopy. An optimal defect density has been found for the highly electrochemical activity. This study offers an important guide for balancing the activity and the conductivity of graphene, which is a challenge and key point for achieving high performance graphene (and also carbon)-based electrochemical devices.

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References: