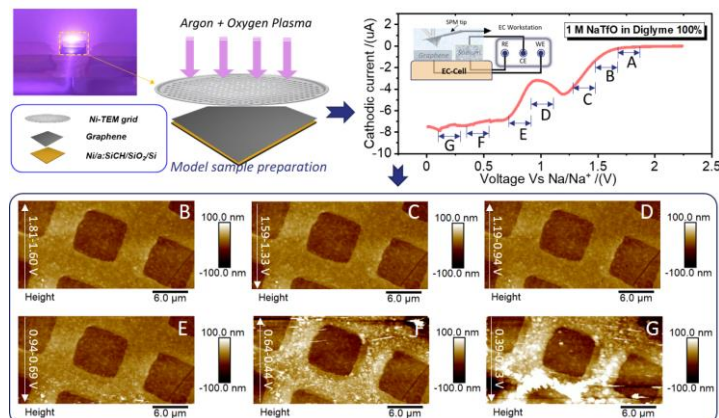


Visualization of Na-diglyme co-intercalation induced few-layer graphene expansion and SEI formation using *operando* electrochemical atomic force microscopy

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Diglyme solvated sodium-ion complexes enable the superfast co-intercalation of charge carriers (Na^+) into graphite carbon interlayers,^[1] providing unprecedented prospects for the application of low-cost graphite carbon as sodium-ion battery anode materials. Insights into this novel co-intercalation process are essential for enhancing the electrochemical performance of co-intercalation-based energy storage systems.^[2, 3] Meanwhile, the paradox role of the co-existence of solid electrolyte interphase (SEI) and solvent co-intercalation behaviors needs to be further clarified.^[4, 5] This presentation focuses on the real-space *operando* observation of the SEI formation, as well as Na-diglyme co-intercalation induced carbon-interlayers expansion in few-layer graphene as sodium anode electrodes. The few-layer graphene grown on the Ni current collector was patterned by Ar/O₂ plasma to serve as a model anode electrode. The co-intercalation phenomenon was then directly observed by monitoring the interlayer spacing expansion using *operando* electrochemical atomic force microscopy (EC-AFM). The electrolyte decomposition was clearly observed on the few-layer graphene surfaces, and the anisotropic chemical components of SEI formed on graphite edge and basal planes were confirmed by XPS. The characterization results indicate that the SEI formed on the graphite edge planes cannot act as a



physical 'barrier' to fully seal the edge sites and prevent the solvent co-intercalation into the carbon-interlayers. This is due to the huge interlayer spacing expansion and contraction rate (300%) upon the intercalation/deintercalation of sodium-diglyme complex as confirmed by *operando* electrochemical EC-AFM characterisations.

Fig. 1. Model sample (Few-layer graphene grid) preparation and electrochemical AFM characterization. (This work was supported by the Faraday Institution (grant number FIRG018), EU Graphene Flagship Core 3 project and EPSRC project EP/V00767X/1 and EP/P009050/1.)

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