Real-time imaging of Na+ reversible intercalation in "Janus" graphene stacks for battery applications

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Abstract (Century Gothic 11)

Sodium, in contrast with other group I and group II metals, cannot intercalate effectively in graphite. Fostering the sodium intercalation process would enable the use of this cheap, abundant element for applications in rechargeable batteries. Here, we report an artificial graphite-like anode for sodium ions storage, formed by stacked graphene sheets functionalized with an aminobenzene derivative only on one side, named as Janus graphene.[1] The asymmetric functionalization allows reversible intercalation of Na+ into such unique structure (Figure 1). This process can be easily monitored by operando Raman spectro-electrochemistry and visualized by imaging ellipsometry. The stacked Janus graphene with planar geometry has only one chemical group present, negligible local curvature, uniform inter-sheet pore size, controllable density of functional groups and minimal amount of edges. This material can store sodium ions differently from both graphite and stacked graphene films. Density Functional Theory (DFT) calculations demonstrate that Na^+ preferably occupies sites close to the $-NH_2$ group forming a synergic ionic bond to the graphene sheet, making the interaction process energetically favourable. Estimates based on electrochemical methods suggest a potential sodium storage as high as $C_{6.9}Na$, comparable to what is currently achieved in standard lithium ion batteries.

References

[1] J.H. Sun, et al., Sci. Adv., (2021), 7, eabf0812.

Figures

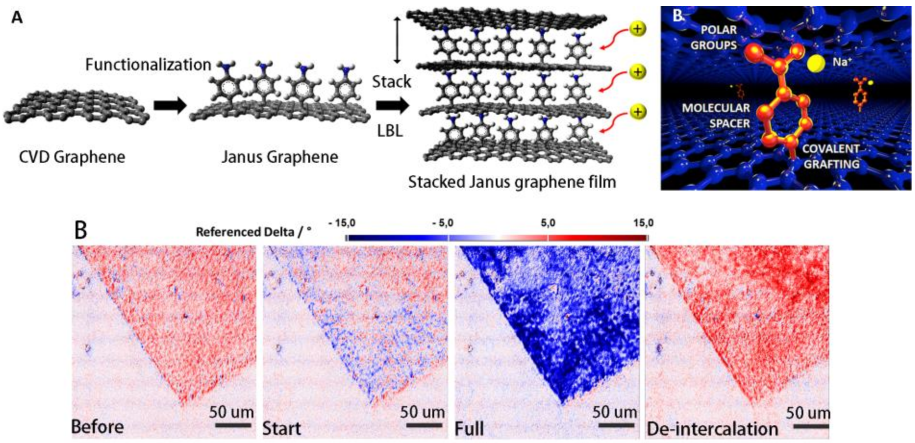

Figure 1: (A) Preparation of Janus graphene. (B) Ellipsometry imaging of sodium ions in/de-intercalation.