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

Vincenzo Palermo

Jinhua Sun¹, Matthew Sadd², Philip Edenborg³, Henrik Grönbeck³, Peter Thiesen⁴, Zhenyuan Xia¹, Vanesa Quintano⁶, Ren Qiu⁵, Aleksandar Matic², Vincenzo Palermo^{1,6,*}

¹Materials and Manufacture, Department of Industrial and Materials Science, Chalmers University of Technology, Göteborg, Sweden. ²Materials Physics, Department of Physics, Chalmers University of Technology, Göteborg, Sweden.

³Department of Physics and Competence Centre for Catalysis, Chalmers University of Technology, 412 96 Göteborg, Sweden.

⁴Accurion GmbH, Stresemannstraße 30, Göttingen 37079, Germany.

⁵Microstructure Physics, Department of Physics, Chalmers University of Technology, Göteborg, Sweden.

⁶Institute of Organic Synthesis and Photoreactivity (ISOF), National Research Council of Italy (CNR), Via P. Gobetti 101, I-40129 Bologna, Italy.

mailto:Vincenzo.palermo@isof.cnr.it

Sodium, in contrast to other metals, cannot intercalate in graphite, and this hinders the use of this cheap, abundant element in rechargeable batteries. Here, we report a nanometric graphite-like anode for sodium ions storage, formed by stacked graphene sheets functionalized only on one side, termed Janus graphene. The asymmetric functionalization allows reversible intercalation of Na⁺, as we monitored by *operando* Raman spectro-electrochemistry and visualized by imaging ellipsometry, both in real time. Our Janus graphene has only one chemical functional group, uniform pore size, controllable functionalization density, very few edges; it can store Na⁺ differently from both graphite and stacked graphene. Density Functional Theory (DFT) calculations demonstrate that Na⁺ preferably rests close to the -NH₂ group forming a synergic ionic bond to graphene, making the interaction process energetically favourable. We estimate a sodium storage up to $C_{6.9}$ Na, comparable to what currently achieved in standard lithium ion batteries. Our approach provides a new concept to design carbon-based materials as anode for the application of sodium ion battery.

FIGURES

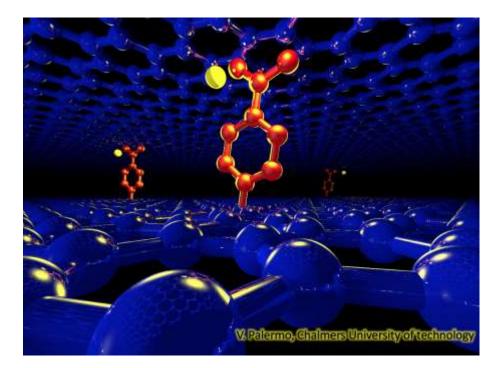


Figure 1: Cartoon showing Na+ ions intercalated in between graphene sheets with aminobenzene spacers.

SmallChem2021