Three-dimensional Nitrogen and Boron Co-doped Graphenes for High-Performance All Solid-State Supercapacitors

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Abstract

Three-dimensional (3D) graphene-based frameworks, such as aerogels, foams, and sponges are an important class of new-generation porous carbon materials, which exhibit continuously interconnected macroporous structures, low mass density, large surface area and high electrical conductivity. These materials can serve as robust matrix for accommodating metal, metal oxide and electrochemically active polymers for various applications in ECs, batteries, and catalysis.[1-3] Herein we demonstrate a simplified prototype device of high-performance ASSSs based on 3D nitrogen and boron co-doped monolithic graphene aerogels (BN-GAs, Figure 1). The device possesses an electrode-separator-electrolyte integrated structure, in which the GAs serve as additive/binder-free electrodes and a polyvinyl alcohol (PVA)/H_2SO_4 gel as solid-state electrolyte and thinner separator. The nitrogen and/or boron doping in carbon networks can facilitate charge transfer between neighboring carbon atoms and thus enhance the electrochemical performance of carbon-based materials. The as-prepared GAs show 3D interconnected frameworks with a macroporous architecture, which are favorable for ion diffusion and electron transport in bulk electrode. Further, monolithic BN-GAs can be easily processed into thin electrode plates with a desirable size upon physical pressing. As a consequence, the resulting BN-GAs based ASSSs exhibit not only minimized device thickness, but also show high specific capacitance (~ 62 F g^{-1}), good rate capability, and enhanced energy density (~8.65 Wh kg^{-1}) or power density (~1600 W kg^{-1}) with respect to undoped (U-GAs), nitrogen doped (N-GAs), boron doped (B-GAs) GAs, or layer-structured graphene paper (GP) (Figure 2). Aerogels built of well-interconnected ultrathin graphene sheets can provide high surface area, 3D macroporosity and high electrical conductivity, which are favorable for enhancing solid-state ion and electron transport in supercapacitors.

References

[1] Wu, Z. S.; Winter, A.; Chen, L.; Sun, Y.; Turchanin, A.; Feng, X.; Müllen, K. Adv. Mater., **24**(2012)5130.
[2] Wu, Z. S.; Sun, Y.; Tan, Y. Z.; Yang, S. B.; Feng, X. L.; Müllen, K. J. Am. Chem. Soc., **134**(2012) 19532.
[3] Wu, Z. S.; Yang, S. B.; Sun, Y.; Parvez, K.; Feng, X. L.; Müllen, K. J. Am. Chem. Soc., **134**(2012) 9082.

Figures

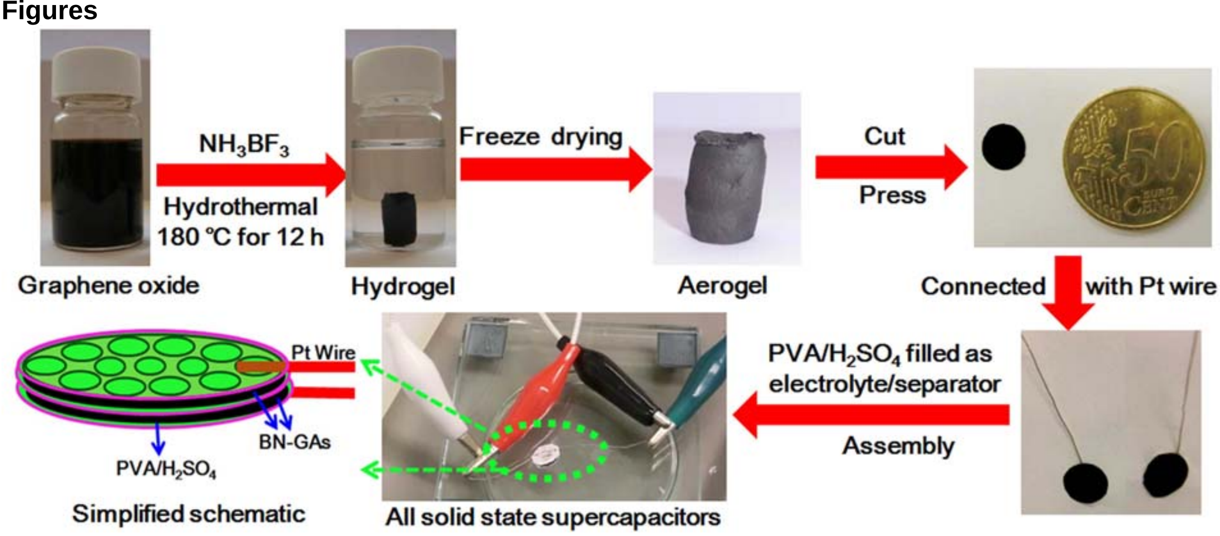

Figure 1. Fabrication illustration of ASSSs based on BN-GAs that were involved by a combined hydrothermal process and freeze-drying process. The as-fabricated supercapacitors with a diameter of 7 mm indicated by green dot ring and its simplified schematic of ASSSs based on aerogels (left down).

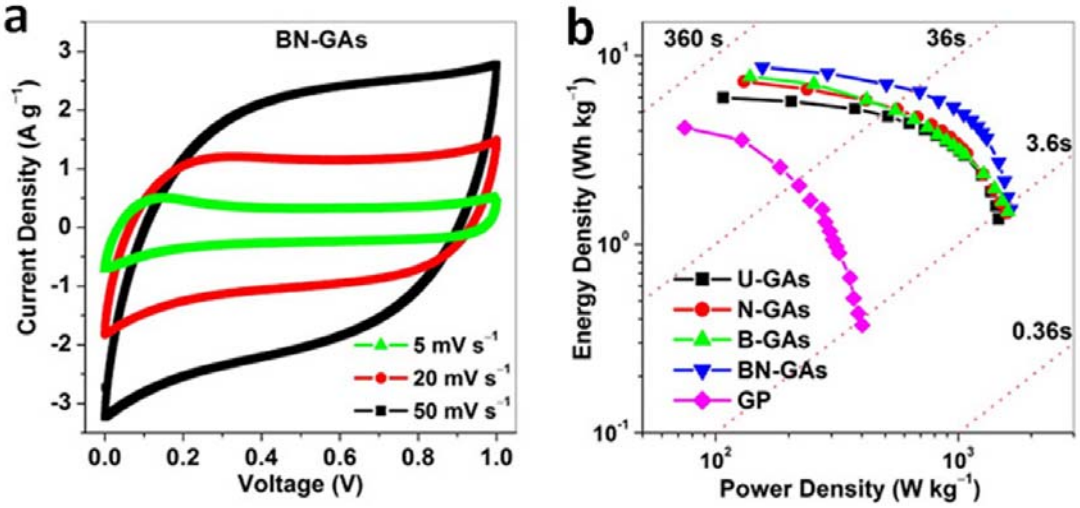

Figure 2. a) CVs of BN-GAs based ASSSs measured at the scan rates of 5, 20 and 50 mV s^{-1}. b) Ragone plot of ASSSs based on U-GAs, N-GAs, B-GAs, BN-GAs and GP, based on two-electrode mass of active materials.