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## Additive manufacturing of heterogeneous nanostructures for energy storage

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This work presented an innovative additive process for precisely fabricating layered ordered heterogeneous three-dimensional (3D) nanostructures. The vertically aligned polyaniline (PANI) nanowires arrays and monolayer graphene sheets were layer-by-layered deposited to specific substrate for tailored structures. Driven by external voltage, aniline molecules and graphene oxide were alternatively assembled for hierarchical porous three-dimensional nanostructures while graphene oxide was in-situ reduced to graphene during the assembly process. The process variables and manufacturing mechanism were studied. Scanning electron microscopy and atomic force microscope results indicated monolayer graphene sheets serve as the transition nodes for the neighbored nanowire arrays. The assembly mechanism was investigated by comparative experiments and X-ray photoelectron spectroscopy, indicating the presence of  $\text{Cl}^-$  ion and its oxidation were critical for the successful assembly. As-produced 3D nanostructures were used as the electrodes of an ultra-capacitor, and an unusual electrochemical behavior was discovered. In organic electrolyte, the capacitance increases as the stack of nanowire arrays increases. However, it shows an opposite trend in an aqueous electrolyte. Further analysis found that the distinctive electrochemical behavior originates from the electrode/electrolyte interactions and the dependence on the diffusion and charge transferring process. The specific energy density was as high as 137 Wh/Kg while power density is in excess of 1000 W/Kg. This work pointed a simple pathway to tailor architecture and electrochemistry for robust design of high-performance ultra-capacitor at a limited lateral size.

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