Graphene-based Microsupercapacitors for On-Chip Micro Power Sources

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Given the rapid development of miniaturized electronic devices like microelectromechanical systems, nanorobotics, wireless sensor networks, micro medical devices, on-chip micro electrochemical energy storage systems are critically needed to power these devices and microsystems. Microbatteries have generally high energy storage density. However, due to the redox reactions between electrodes and electrolytes, they share the same disadvantages as normal batteries such as limited lifetime, low power density, and safety concerns. Microsupercapacitors, on the other hand, offer faster charge-discharge rate, higher power density and longer lifetime, but suffer from low energy density.

Here we report electrochemical microsupercapacitors using highly porous vacuum-annealed graphene (VAG) as electrode materials combined with photolithography techniques to pattern the interdigital electrodes. The VAG has a surface area of ~500 m^2/g measured by Brunauer-Emmett-Teller (BET) method with a C/O ratio of 5 measured by XPS. The removal of oxygen groups in graphene oxide by low-temperature vacuum annealing process leads to the broadening and shifting of the peak to $2\theta=23.60°$ and a reduced interlayer spacing of 3.76 Å in X-ray diffraction. From Raman spectrum, the intensity ratio I_D/I_G is as low as 0.91 indicating a less defect content than the other reported graphene materials. The reduced graphene material is deposited on a gold interdigital pattern by electrophoretic deposition at a deposition rate around 1 um/min. The resulted VAG electrodes was tested in both aqueous electrolyte and solid electrolyte. At various scan rates from 5-100 mV/s, the device shows outstanding electric double layer capacitor behavior. At the scan rate of 5 mV/s, it shows a stack capacitance of 11.30 F/cm^3 and an energy density of 1 mWh/cm^3 with solid electrolyte. The VAG based microsupercapacitor was charged and discharged at current from 10 A/cm^2 to 50 A/cm^2. VAG microsupercapacitors show good capacitive performance with nearly triangular curves. The Nyquist plots for VAG microsupercapacitors shows a sharp increase of imaginary part in the low frequency region, indicating a capacitive behavior. Other than the high capacitance, the cycling stability is also tested for 10,000 cycles and 93% of the initial capacitance is retained, indicating good electrochemical stability of the VAG material. To our knowledge, the specific energy density is the highest among reported microsupercapacitors.

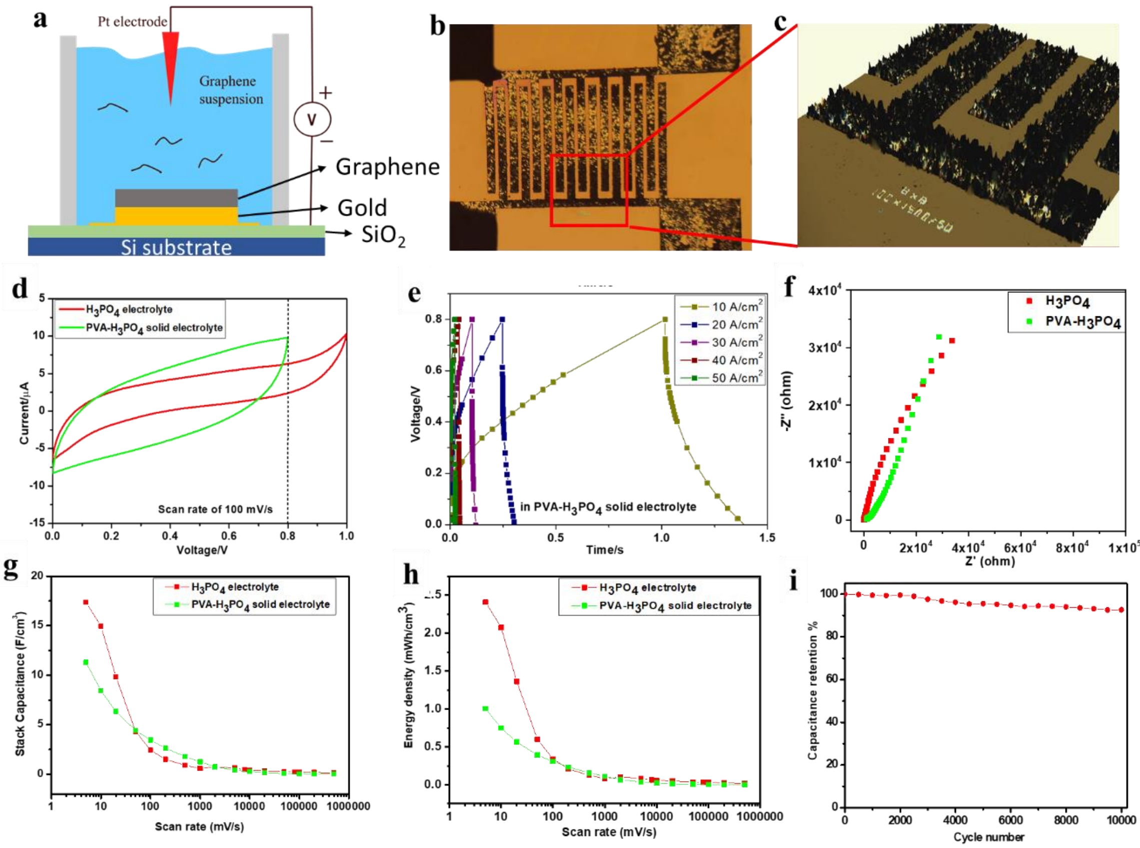

Figure (a) Schematic of the graphene deposition process, (b) The optical image of a deposited graphene interdigital electrodes, (c) Surface profile of the interdigital graphene microsupercapacitor, (d) Cyclic voltammetry scans of the VAG supercapacitor in aqueous and solid electrolyte, (e) Charging-discharging curves of VAG microsupercapacitor, (f) EIS plot of the VAG microsupercapacitor, (g) Stack capacitance as a function of scan rate, (h) Energy density plots vs scan rates, (i) Cycling stability of VAG microsupercapacitor for 10,000 cycles.