#### Engineering Carbon Nanostructures and Architectures for High Performance and Multifunctional Electrodes

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#### **High Performance and Multifunctional Electrodes**

#### **Multifunctional Electronics**





Flexible and transparent smart phone, flexible computer, Flexible electronic newspaper (Wearable Electronics)



Devices require mechanically flexible, functional, and high performance energy storage systems

#### **Electrode Materials**

- 3D Nanostructures
- Electro-Mechanical Stability
  - Optical Transparency etc.



# SP<sup>2</sup> Carbon Nanostructured Materials



#### Mechanical Properties

- Strong sp2 Carbon-Carbon covalent bonding
- High elastic modulus (1 TPa) and High strength

#### Electrical and Optical Properties

- High Mobility
- Highly conductive w/wo mechanical deformation
- High current density (10<sup>9</sup> A/cm<sup>2</sup>)
- Optically Transparent
- In-plane Properties of Graphitic Carbon
  - Good thermal conductivity (<3000W/mK)
  - Good chemical stability



#### Limitation of current CNT/graphene based networks

Built on weak van der Waals interactions between CNTs, CNTs-Graphene
 Lower mechanical strength, electrical and thermal conductivities due to a lower pulling resistance, electron and phonon scatterings at these "unconnected" junctions
 Transforming physical Junctions into covalently bonded sp<sup>2</sup> Chemical Junctions







J. Tour et al., Nature Communications, 2012



#### Restructuring sp<sup>2</sup> Lattice and Network Structure

A voltage-induced *electrical fusion* of SWCNTs







Restructuring sp<sup>2</sup> Lattice Structure

H. Jung et al, Nature Communications 2014









## **Engineering Nanostructure and Morphology**



J. Hao et al, Unpublished



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## **Engineering 3D Nanoscale Architecture**

#### **Carbon Nanocups**

#### **Graphitic nanostructures**

having smaller length/diameter (L/D) aspect ratio, *nanoscale cup morphology*, can effectively contain other nanomaterials and polymers, leading to multi-component hybrid

- **Energy Storage**
- Nanogram Quantity Container
- Multifunctional Sensors





# **Engineering 3D Nanoscale Architecture**

#### **Fabrication Process**

The length of nanochannels are controlled by second anodizing time.



H. Chun, et. al., ACS Nano (2009) H. Jung et al. Scientific Reports (2011)



## **Engineering 3D Nanoscale Architecture**

#### TOP view

**BOTTOM** view



#### **3D Carbon Nanostructured Film for Supercapacitor Electrodes**

- Electrically Conductive: Surface Conductivity: 117 S/m
- High surface area and highly disordered graphitic layers provides the effective permeation of the polymer electrolyte and their conformal packaging with electrodes.
- Unique nanoscale cup feature enables the easy access and faster transport of ions at the electrode/electrolyte interface resulting in higher power capability.
- High current carrying capability, substantial mechanical strength, and small effective electrode thickness (5-10 nm: 80-85% Transmittance at 550nm wavelength) allow us to build optically transparent and mechanically flexible reliable thin-film (solid state) energy storage devices.



## **Flexible and Transparent Supercapacitors**



(a) concave and (b, c) convex and (d-f) branched nanocup films (H. Jung et al. Scientific Reports 2012)



- CNC films: Outer graphene layers are acting as current collectors and the Innermost layer exposed electrolyte is acting an electrode.
- Polymer electrolyte (PVA-H<sub>3</sub>PO<sub>4</sub>) is acting as both electrolyte and separator.



## **Flexible and Transparent Supercapacitors**





(a) Cyclic voltammetry (CV) measured with  $10 - 500 \text{ mVs}^{-1}$ scan rates. (b) Galvanostatic charge/discharge (CD) results measured at a constant current density of 5  $\mu$ Acm<sup>-2</sup>. The capacitances by the geometrical area calculated from CD curves are 409  $\mu$ F cm<sup>-2</sup>. (c) The capacitance change as a function of temperature









# **Flexible and Transparent Supercapacitors**



Normalized capacitance as a function of cycle-number (10,000) and w/wo the mechanical deformation (45° bending). (SG: single layer graphene, RMGO: reduced multilayer graphene oxide, HGO: hydrated graphitic oxide, LSG-EC: laser-scribed graphene electrochemical capacitor)

Jung, Ajayan et al. Scientific Reports 2012

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