## **High-Voltage Electrical Double-Layer Capacitors**

## Kun-Ping Huang<sup>1</sup>

Yu-Wen Chi<sup>1,2</sup>, Chi-Chang Hu<sup>2</sup>

<sup>1</sup>Mechanical and Mechatronics Systems Research Laboratories, Industrial Technology Research Institute, 195, Sec. 4, Chung Hsing Road, Chutung, Hsin-Chu 31040, Taiwan, R.O.C. <sup>2</sup>Department of Chemical Engineering, National Tsing Hua University, 101, Section 2, Kuang-Fu Road, Hsin-Chu 30013, Taiwan

## kphuang@itri.org.tw

Here we propose a guideline, "choosing a matching pair of electrode materials and electrolytes", to effectively extend the cell voltage of EDLCs according to three general strategies. Based on the new strategy proposed in this work, materials with an inert surface enable to tolerate a wider potential window in commercially available organic electrolytes in comparison with activated carbons (ACs). The binder-free, vertically grown graphene nanowalls (GNW) and nitrogen-doped GNW (NGNW) electrodes (Figure 1) respectively provide good examples for extending the upper potential limit of a positive electrode of EDLCs from 0.1 to 1.5 V (vs Ag/AgNO3) as well as the lower potential limit of a negative electrode of EDLCs from -2.0 V to ca. -2.5 V in 1 M TEABF4/PC (propylene carbonate) compared to ACs (Figure 2). This newly designed asymmetric EDLC exhibits a cell voltage of 4 V, specific energy of 52 Wh kg-1 (ca. a device energy density of 13 Wh kg-1), and specific power of 8 kW kg-1 and ca. 100% retention after 10,000 cycles chargedischarge, reducing the series number of

EDLCs to enlarge the module voltage and opening the possibility for directly combining EDLCs and LIBs in advanced applications. References  Yu-Wen Chi, Chi-Chang Hu, Hsiao-Hsuan Shen, and Kun-Ping Huang, Nano Lett., 16 (2016) 5719

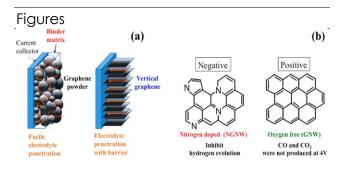
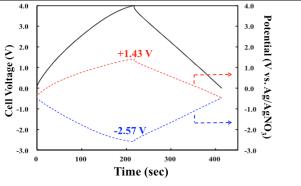


Figure 1: (a) In comparing with AC powdercoated electrodes, GNW/Ti and NGNW/Ti display a binder-free, vertical structure, favoring the penetration of electrolytes and electron transport in the whole graphene matrix. (b, oxygen-free, binder-free GNWs right) The circumvent the issue of oxygen-functional group removal, which are inert to the irreversible oxidation of organic electrolytes, enlarging the upper limit of working potential window. (b, left) The uniform N doping on the binder-free, vertical NGNWs significantly depresses the irreversible reduction of residual water and organic electrolyte at the negative potential end, further enlarging the working potential window.



**Figure 2:** The binder-free, vertically grown graphene nanowalls (GNW) and nitrogendoped GNW (NGNW) electrodes respectively provide good examples for extending the upper potential limit of a positive electrode of EDLCs from 0.1 to 1.5 V (vs Ag/AgNO3) as well as the lower potential limit of a negative electrode of EDLCs from -2.0 V to ca. -2.5 V in 1 M TEABF4/PC