Graphene/Carbon Nanotubes hybrid for Advanced Flexible Supercapacitors

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The research on flexible electronics is raising huge interest because of its plethora of applications. [1, 2] In particular, flexible energy storage systems (ESSs) will have a fundamental role as energy sources for wearable electronics, medical devices just to cite a few. [1, 2] Amongst all the ESSs, supercapacitors (or electrochemical double layer capacitors -EDLCs-) are promising devices as they bridge the gap between batteries and conventional capacitors. In fact, batteries suffer from a limited number of life cycles, low power density and high recharge time. [3] As concerns conventional capacitors, they have high power density, almost unlimited life cycles and fast charge, but at the expenses of low energy density if compared with the one of batteries. The performances of supercapacitors lie in between those mentioned: they show a higher energy density compared to conventional capacitors, and higher charge/discharge rates, life – time and power density than batteries. [4] Supercapacitor electrodes are usually built by using activated carbon (AC),[4] which is characterised by high surface area due to a large porosity. However, only a fraction of this surface area is accessible to the electrolyte ions, since they cannot penetrate in pores smaller than the ions dimension. In order to solve this drawback, other carbon-based compounds have been proposed as active materials for EDLCs, including carbon nanotubes (CNTs) and graphene. [2] Herein, we exploit hybrid graphene/CNTs compounds in order to avoid re-aggregation effects, i.e., both the bundle of CNTs and restacking of graphene, which are detrimental for the electrochemical performances of the electrodes, limiting the specific surface area of these materials. [7] The hybrids are produced in form of dispersion in N-methyl-2-pyrrolidone used as solvent. The CNTs are de-bundled by means of ultrasonication method [8,9], while graphene is produced by wet-jet milling exfoliation of graphite. [10] The latter enables an high-throughput production of graphene (> 24 g h^{-1}) with a yield of ≈100%,[10] overwhelming time consuming issues affecting other solution-based processes. [11] The final dispersion is obtained by mixing the dispersion in a 1:1 ratio. The so-produced functional ink is compatible with scalable supercapacitor manufacturing, such as printing and vacuum filtration methods. [12] Our production procedure allows us to fabricate large-area (cm^2) supercapacitor with high-areal capacitance (>100 mF cm^{-2}) on flexible metal-free substrate.

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