A Comparative Study on Supercapacitors Formed with Different Graphene-Based Hybrid Nanostructured Materials

Tasnim Mahjabin and Md. Abdullah Al Amin

Abstract The development of high-performance energy-storage devices is crucial for satisfying the rapidly increasing demands for new applications that require high-power, high-energy, and cost-effective energy-storage systems (ESSs). The supercapacitors have high power densities, long lifetimes, short charging times, and excellent safety characteristics for fulfilling these requirements. If hybridization of different chemical substances is made, then even better properties can be achieved. In this review article, different hybrid nanostructures for advanced supercapacitors are going to be discussed and compared to find the one which will demonstrate optimum properties. Here, graphene with its excellent electrical properties and porous carbon nanostructure resulting in large specific surface area will be kept as the constant part, while various other chemical substances are going to be added to produce different supercapacitors. These will include: hybrid of (1) graphene-wrapped $Li_4Ti_5O_{12}$ and activated carbon, (2) copper oxide nanowire/graphene, (3) oxygen-deficient TiO_2/graphene.

Keywords Supercapacitor · Graphene · Nanostructure

Introduction

At present, studies on practical energy-storage systems have given greater importance on the development of high-performance electrochemical energy devices, such as batteries and supercapacitors. The supercapacitors have been greatly analyzed because of their high power densities, long lifetimes, short charging times, excellent safety properties, and eco-friendly nature [1–4]. The basic principle of supercapacitor is to store electrical energy through the electric double-layer capacitance formed by the charge separation on the interface between the electrolyte and the bath solution [5]. The supercapacitors can be mainly categorized under two basic types the basis of the energy-storage mechanism of the electrode materials, such as: (i) electrochemical double-layer capacitors (EDLCs) and (ii) pseudocapacitors. EDLCs physically store

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charges through reversible ion adsorption at the electrode–electrolyte interface so that they have greater advantages over batteries, such as fast charging/discharging, higher longevity (long lifetimes), and simple and stable structures [6, 7]. And, the second one is the pseudocapacitor which chemically stores charges through redox reactions between the electrodes and electrolytes using metal oxides and/or conducting polymers, which enable to induce high energy densities [8, 9]. A hybrid capacitor can be made with a battery and a supercapacitor or a metal oxide with supercapacitor, and in both cases, enhanced properties can be achieved.

The hybrid of graphene-wrapped $Li_4Ti_5O_{12}$ and activated carbon for supercapacitors has drawn much attention in the present era. Lithium-ion batteries (LIBs) can provide the highest energy density (about 150 $Whkg^{-1}$) through Faradaic lithium insertion reactions among the prevailing energy-storage devices [10–12]. Nevertheless, Faradaic lithium de/intercalation reactions involve the solid-state diffusion of ions in a crystal, for which low power density is often resulted [10, 11]. On the other hand, supercapacitors, one of the most important types of energy-storage device, can provide high power densities (about 3 $kWkg^{-1}$), because of non-Faradaic surface reactions, at the expense of the energy density (about 5 $Whkg^{-1}$) [12, 13]. So, combination of these two can result in synergetic properties which can surmount the shortcomings of each other to a significant extent.

Recent advances have also been focused on the production of supercapacitors formed with hybrid of copper oxide nanowire/graphene. The abundance of copper oxide (CuO) in the earth has made it easily available, and it is one of the low-cost materials with a high capacitance, suitable for use in energy-storage devices [14, 15]. Because of its non-toxicity and simple preparation in the form of a nanostructure, it has been widely used in supercapacitors. Bulk CuO has an insufficient electrical conductivity and small surface area for commercial energy-storage electrodes, and so, the nanostructure of CuO has been preferred to deliver a facile pathway for electrolyte ion penetration. Different morphologies of CuO nanostructures have been found, such as nanoparticles (NPs) [15, 16], nanosheets [17–19], nanoflowers (NFs) [20], and nanowires (NWs) [21, 22]. They maximize the surface area for access of ions from the electrolyte. In order to further increase the electrochemical performances of supercapacitors, hybridization with electrically conducting materials for electrodes could be employed to effectively increase the capacitance, as the synergetic effect of the EDLC and pseudocapacitance is expected to enhance the capacitance [2]. Besides, graphene oxide (GO) or its reduced form, reduced graphene oxide (RGO), is the most suitable for electrochemical applications, such as energy conversion and storage, and electrochemical sensors and catalysts, as can be utilized as a platform to develop different hybrids because of its superior functionality [23, 24]. So, synergetic combination of one-dimensional copper oxide nanowires and two-dimensional RGO sheets to fabricate a highly porous and electrically conductive three-dimensional hybrid nanostructure can be really efficient for high-performance supercapacitor electrodes with increased capacitance.

Another type of supercapacitor material which has also gained much importance is the hybrid of oxygen-deficient TiO_2/graphene. In general, pseudocapacitance materials (TiO_2, MnO_2, Fe_2O_3, Co_3O_4, $ZnFe_2O_4$, etc.) show higher capacitance through surface redox reactions than electric double-layer capacitors (EDLCs) [25–29]. Owing to its high stability, low cost, excellent electrochemical stability, and non-toxicity, TiO_2 has gained considerable attention to be utilized supercapacitors [30–33]. Nevertheless, one major issue to consider is the poor electrical conductivity that limits its power density and rate capability [53]. In order to overcome this problem and improve its electrochemical performance, most investigations have been focusing on designing nanostructured electrodes and preparing composite electrodes. Anodized TiO_2 nanotube arrays have been developed to provide a direct pathway for electron transport along the nanotubes to the Ti foil substrate to enhance the electrochemical performance [34, 35]. Combining TiO_2 with highly conductive carbon materials such as carbon nanofibers and graphene can also result in outstanding properties [36–38]. At present, improving the electrical properties of TiO_2 through doping or creating defects has been developed, and these are capable of charging and discharging at a very high rate [33, 35, 39].

Various reinforcing materials for supercapacitor electrodes have been studied among which carbon materials have been extensively investigated because of their outstanding mechanical and electrical properties [44–46]. Generally, it has been found that carbon-based materials, including activated carbon, carbon nanotubes, and graphene, exhibit superior electrical conductivities and large physical surface areas and thus are ideal active electrode materials for supercapacitors [47, 48]. Particularly, graphene has attracted immense interest for energy-storage applications owing to its outstanding mechanical, electrical, and thermal properties as well as intrinsically large surface area [49, 50]. For these reasons, in most of the cases, graphene material has been used as the reinforcing part as can be seen in the as-mentioned supercapacitors.

All the above-mentioned supercapacitors have been produced recently in various researches. These are going to be discussed and studied thoroughly in this review work to find the most suitable one.

Process Mechanisms

Conventional electric double-layer capacitors (EDLCs) use only non-Faradaic surface reactions at both electrodes, but hybrid supercapacitor of graphene-wrapped $Li_4Ti_5O_{12}$ and activated carbon has been produced to adopt Faradaic lithium intercalation reactions at one electrode and non-Faradaic surface reactions at the other electrode (Fig. 1a and b) [12, 41]. Thus, the overall energy density of the system can be increased while maintaining a high power density [40, 41].

Highly porous and capacitive 3D CNSs based on CuO-NW/graphene hybrids for high-performance supercapacitor electrodes are made by the process mechanism shown in Fig. 2. Here, preparation of the 3D porous CNS was carried out by a simple and efficient solution-based hydrothermal technique, wherein GO sheets were easily self-assembled. During the hydrothermal process, GO was successfully reduced to RGO by losing its oxygen-based functional groups because of the high-temperature annealing. Hence, the resulting 3D CNS exhibited an electrically conducting RGO network. Hybridization of hydrothermally synthesized CuO-NWs with the 3D RGO CNS was performed in order to increase the energy-storage performance.

The preparation process mechanism of oxygen-deficient TiO_2/graphene hybrid is described in Fig. 3. Here, graphene oxide (GO) and as-prepared TiO_2 nanocrystals were dispersed in deionized water, then the suspension was irradiated with laser. During the irradiation process, the oxygen-deficient TiO_2 and reduced graphene oxide

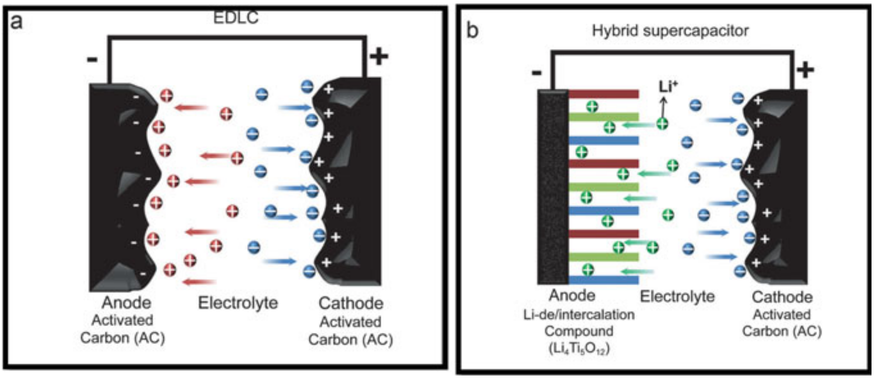

Fig. 1 Electrochemical reaction mechanism of **a** EDLCs and **b** Hybrid supercapacitors. Adapted with permission from Ref. 1(1), pp.125–130. Copyright 2013, John Wiley and Sons

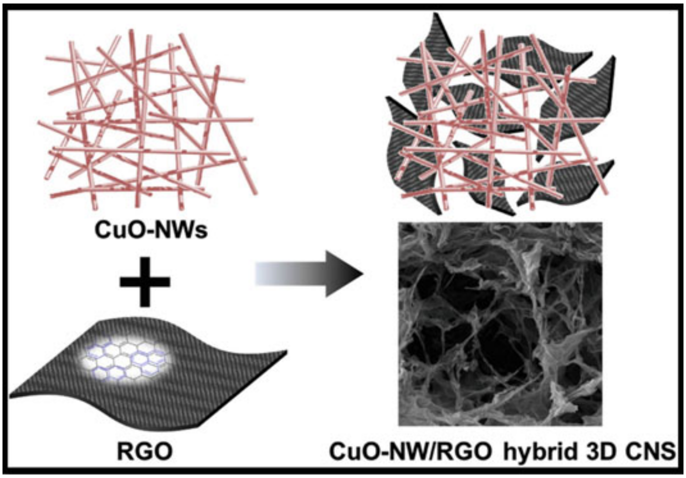

Fig. 2 Schematic of the CuO-NW/RGO hybrid 3D CNS. Adapted with permission from Ref. 178. Copyright 2019, Elsevier

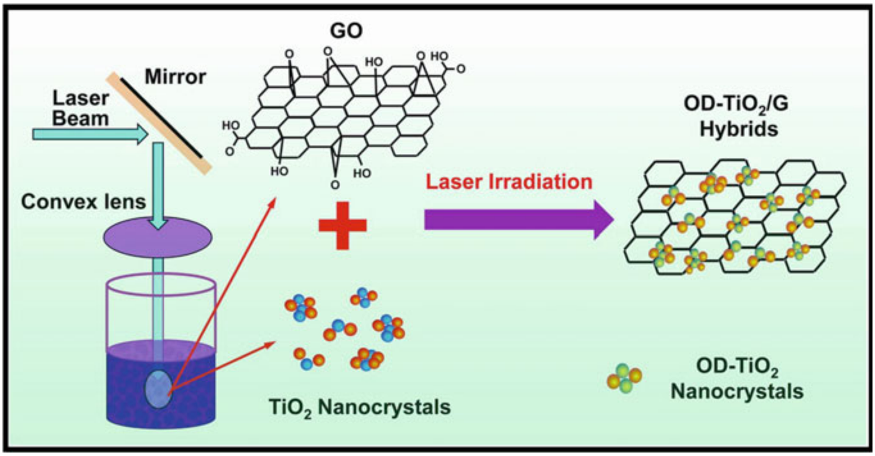

Fig. 3 Schematic illustration for the preparation of OD TiO2/G hybrids. Adapted with permission from Ref. 431. Copyright 2019, Elsevier

(LRGO) were produced with the help of the electrons generated from water in the extreme non-equilibrium reaction conditions, whereas the LRGO with numerous defect sites induced by laser irradiation provides efficient nucleation sites for anchoring the oxygen-deficient TiO_2 nanocrystals [42, 43].

Results and Discussion

All the three hybrid supercapacitors showed excellent properties when experimented in different researches. A brief comparison of their properties is given below:

Properties	Graphene-wrapped $Li_4Ti_5O_{12}$ and activated carbon	Copper oxide nanowire/graphene	Oxygen-deficient TiO_2/graphene
Energy density	50 $Whkg^{-1}$	50.6 $Whkg^{-1}$	14.1$Whkg^{-1}$
Power density	2500 Wkg^{-1}	200 Wkg^{-1}	8.5 $kWkg^{-1}$
Method	Graphene nanosheet coating method, which uses electrostatic attraction between negatively charged graphene and positively charged LTO particles within a specific pH range	Self-assembly technique through a simple hydrothermal treatment	Graphene oxide (GO) and as-prepared TiO2 nanocrystals were dispersed in deionized water, then the suspension was irradiated with laser

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Properties	Graphene-wrapped $Li_4Ti_5O_{12}$ and activated carbon	Copper oxide nanowire/graphene	Oxygen-deficient TiO_2/graphene
Cost effectiveness	Low cost	Cheaper as CuO is abundant in earth	Low cost but not as low as the other two
Longevity/stability	Higher longevity	Greater stability	High stability
Non-toxicity	Non-toxic	Non-toxic	Non-toxic
Application	Hybrid electrical vehicle (HEV) application	Energy-storage applications	Long-term compact energy storage

Refs. [51–53]

So, from the above-mentioned data in the table, it can be found that graphene-wrapped $Li_4Ti_5O_{12}$ and activated carbon-based hybrid supercapacitor is going to be the most effective one as it has both high energy density and high power density. It can store a high amount of energy as per requirement and can also release energy at a greater rate. This is a new high-energy and high-power hybrid supercapacitor based on a graphene-wrapped LTO anode and an AC cathode where the graphene nanosheet wrapping significantly improved the rate capability of the LTO anode, which contributed in surmounting the intrinsic kinetic imbalance between non-Faradaic capacitive electrodes (AC) and Faradaic lithium intercalation electrodes (LTO) [51]. It has high specific energy of up to 50 $Whkg^{-1}$ and can even maintain an energy of approximately 15 $Whkg^{-1}$ at a 20 s charge/discharge rate [51]. Besides, the other two aforementioned supercapacitors have also shown great importance. The combination of one-dimensional CuO-NWs and two-dimensional RGO sheets to produce the 3D porous hybrid nanostructure resulted in a hybrid nanostructure that had a large surface area, increased thermal stability, and high crystallinity [52]. It also showed high energy density, power density, outstanding cycling stability (capacitance retention of 91.2% after 5,000 CD cycles), high specific capacitance (364 F g^{-1}), and increased electrothermal conductivity [52]. On the other hand, oxygen-deficient TiO_2/graphene-based supercapacitor also showed promising characteristics as can be seen from the table. It also depicted the use of laser irradiation method in the in situ preparation of OD-TiO_2/G and found the enhancement of electrical conductivity associated with TiO_2 [53].

Conclusion

For emerging large-scale ESSs, bridging this performance gap remains a key issue. Utilizing the synergetic effect of different materials, battery, supercapacitors can reduce this gap to a considerable extent. Graphene nanosheets and nanoparticles have played a significant role in this regard. The supercapacitors of hybrid of graphene-wrapped $Li_4Ti_5O_{12}$ and activated carbon are capable of delivering a high specific energy of up to 50 $Whkg^{-1}$ and can even maintain an energy of approximately 15 $Whkg^{-1}$ at a 20 s charge/discharge rate. The hybrid supercapacitors of copper oxide nanowire/graphene showed high specific capacitance (364 F g^{-1}), outstanding cycling stability (capacitance retention of 91.2% after 5,000 CD cycles), and high energy density (50.6 $Whkg^{-1}$ at 200 Wkg^{-1}). Moreover, the supercapacitor formed with hybrid of oxygen-deficient TiO_2/graphene delivered a maximum energy density of 14.1 $Whkg^{-1}$ and a maximum power density of 8.5 $kWkg^{-1}$. Comparing all the three supercapacitors, the one with the hybrid of graphene-wrapped $Li_4Ti_5O_{12}$ and activated carbon showed optimum properties.

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