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Advances in Energy and Materials Research https://jaem.qom.ac.ir/

Recent advances and research outcomes concerning the application of graphene and its nanocomposites in supercapacitors

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Article Info

Article type:
Research Article

Article history:
Received 26 Dec 2024
Received in revised form 17 Jan 2025
Accepted 20 Feb 2025
Published online 27 Mar 2025

Keywords:
Supercapacitors, Renewable energy, Graphene based nanocomposites, Conductive polymers, Activated carbons, Transition metal oxides

ABSTRACT

The importance of introducing renewable and clean energy sources has never been more important, given the limited fossil fuel resources and concerns in recent years about the increase in global temperatures. Graphene and its composites have received much attention in recent years, and their applications in electrochemical energy storage and conversion systems are expanding. This review, by categorizing the results of recent research in the field of using graphene-based composites and combining them with innovative materials such as conductive polymers (CPs), activated carbons (ACs) extracted from biomass, and transition metal oxides (TMOs) nanoparticles, provides a good comparison and facilitates the selection of appropriate options in future research. The morphology of the electrode active materials used in the electrode structure and their discussion are among the other issues examined in the present work. It appears that binary and ternary nanocomposites, which combine nanomaterials and leverage the advantages of each, through a synergistic effect mechanism, will create a promising future for clean, affordable, and renewable energy systems.

Cite this article: M. Bigdeloo, (2025). Recent advances and research outcomes concerning the application of graphene and its nanocomposites in supercapacitors, *Advances in Energy and Materials Research*, *2* (1), 3-10.
https://doi.org/10.22091/jaem.2025.14185.1032

DOI: 10.22091/jaem.2025.14185.1032
Publisher: University of Qom.

1. Introduction

The environmental pollution and non-renewability are the main factors of non-reliance on fossil fuels [1, 2]. For several decades, scientists have been seriously looking for clean and environmentally friendly energy sources to replace fossil fuels. Various forms of renewable energy, such as solar cells, wind energy, and energy extraction from biomass, have been accompanied by significant advances in recent years [3, 4].

Supercapacitors (SCs) are a new type of electrochemical capacitors that have a much higher energy density (close to batteries) compared to physical capacitors [5]. In physical capacitors, only two parameters, the distance between the plates and their surface area, determine the specific capacitance of the system (often in the microfarad range), while in SCs, variables such as the conductivity of the electrolyte, the type of electroactive material, the resistance in the Helmholtz layer and the mass of the electroactive material are involved in the efficiency of the system (possibility of achieving Cs in the range of several thousand farads) [6].

With this purpose in mind, it will be possible to classify SCs based on electroactive materials, configuration, and electrolyte. In the first category, based on electroactive materials, SCs were classified into three categories: electrochemical double-layer capacitors (EDLCs), pseudo-capacitors, and hybrid SCs [7]. In EDLCs, only carbon-based materials such as graphene, activated carbon, etc., are employed. In this group, charge storage is carried out cumulatively (non-Faradaic) at the electrode-electrolyte interface, and most of the system will be under diffusion control, and Fick's laws are valid in this category. In the second category (pseudocapacitors), electroactive materials such as CPs and TMOs are used, which are called pseudocapacitors due to the similarity of these materials and the energy storage mechanism in them to batteries. Charge storage is carried out through redox reactions (oxidation and reduction half-reactions) and electron transfer (Faradaic). The last category, which includes the simultaneous use of both of the above categories and the benefits of both of the previous categories, is called hybrid SCs. Fig. 1 schematically illustrated a summary of this classification.

The primary limitation of utilizing carbon materials lies in their low specific capacitance. Recently, Ahmad et al. employed asparagus waste and transformed it into activated carbon through a combination of chemical and physical techniques. The principal advantages of this research included the development of environmentally friendly and cost-effective active electrode materials. Ultimately, the target material demonstrated a specific capacitance of 160 F g^{-1}, along with an energy density of 31 Wh kg^{-1} and a power density of 0.56 kW kg^{-1} [8].

Key findings of this study indicated that the integration of CPs, graphene, and other carbon-based materials enhances energy storage properties, particularly the structural stability of electroactive materials during prolonged charge-discharge cycles. Building on these properties, Bigdeloo et al. employed a ternary nanocomposite comprising ketoconazole-functionalized graphene oxide, activated carbon derived from canola waste, and poly-orthoaminophenol as the electrode material. The results were exemplary, with the specific capacitance within a two-electrode system reaching an outstanding 1056 F g^{-1}. Furthermore, the stability of the specific capacitance was assessed over 5000 charge-discharge cycles, retaining 96.2% of the initial capacitance [9].

Transition metal oxides (TMOs) have garnered substantial interest recently, given their pseudo-capacitive properties and their ability to impart battery-like characteristics to SCs. The co-utilization of multiple metal oxide nanoparticles and their synergistic effects are expected to enhance energy storage capabilities, thereby advancing supercapacitor technology. Recently, Saroha et al. synthesized a binary nanocomposite composed of CuO and TiO_2. Among the notable outcomes was achieving a specific capacitance of 1394.88 F g^{-1} at 5 mV s^{-1}. Additionally, no loss of initial capacitance was observed after 4000 charge-discharge cycles. Finally, a charge transfer resistance of 6.52 Ω demonstrated the high conductivity and minimal resistance of these materials [10].

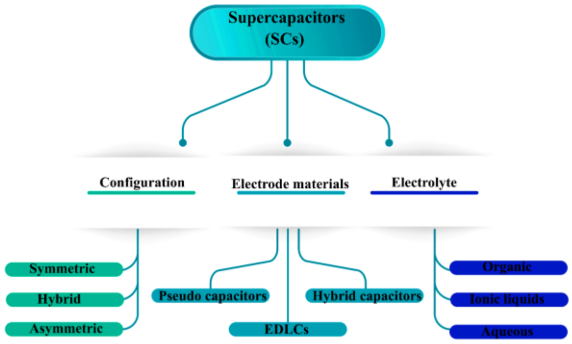

Fig. 1. Classification of types of supercapacitors.

2. SCs Electrode materials

This review aims to objectively examine and compare the electrochemical results and charge storage characteristics in supercapacitor systems based on three electroactive materials: conducting polymers (CPs), activated carbons (ACs), and transition metal oxides (TMOs).

2.1. Composites based on graphene / CPs

Materials are combined for specific purposes, for instance, graphene, due to its high specific surface area, provides a suitable substrate for the transport of ions and electrons and the accumulation of charge. CPs are added to graphene with two main purposes: to create structural stability and to enhance the flexibility properties. The structural stability created for electroactive materials when using CPs is due to the coating of a very thin layer of polymer (slurry state) on the materials. Similarly, CPs, since they have a stronger elasticity than carbon materials, enhance the flexibility properties of the electrode. On the other hand, the decrease in conductivity due to the relatively high band gap of CPs is the most important weakness of using CPs. Polyaniline, polythiophene and polyorthoaminophenol are among the most important CPs used in SC systems. In order to prevent the thickening of the polymer layer, the polymer is mainly deposited on the electrode surface through the electropolymerization method.

With the same strategy, Dayong Gui et al. first synthesized graphene oxide via the advanced Hammers method. Then, they produced a polyaniline/graphene oxide-based composite via electrochemical polymerization [11]. The porous morphology and suitable surface area were the most important features of this nanocomposite. Achieving a specific capacitance of 355.2 F g^{-1} at a current density of 0.5 A g^{-1} was the main result of the research. In addition, the structural stability of the polyaniline-based composite was confirmed by maintaining a constant and unchanged specific capacitance after 1000 continuous charge-discharge cycles. Finally, acceptable resistance values were observed both in solution and in the electrolyte through Nyquist plots. This research well demonstrated the potential of using graphene/CP-based composites even in other electrochemical energy storage and conversion systems.

In this regard, Akram Alabadi et al. doped sulfur on graphene oxide nanosheets. A very thin polythiophene layer was coated on the electroactive material structure through the electrochemical method [12]. The synergistic effect between polythiophene and graphene oxide enhanced the energy storage properties. Using an alkali aqueous electrolyte, the specific capacitance of the target material was measured to be 296 F g^{-1} at 0.3 A g^{-1}. Furthermore, the expansion of the research towards scaling up and fabricating a two-electrode system led to the achievement of an energy density of 148 W h kg^{-1} at a power density of 41.6 W kg^{-1}. Finally, the retention of 91.86% of the initial capacity after 4000 alternating charge-discharge cycles well demonstrated the robustness of the composite.

In another study, Kahriz et al. deposited a thin film layer of poly ortho aminophenol on a specific type of functionalized graphene oxide [13]. The layered structure of the studied graphene facilitated and accelerated the electron transport. The order and stability in the arrangement and the desired distance between the nanosheets were provided by the poly ortho aminophenol slurry. The specific capacitance calculated in this study reached a remarkable number of 8100 mF.cm−2 at 5 mA.cm−2. In addition, only 12% of the initial capacitance was lost after 1000 charge-discharge cycles for this composite. This study paved the way for further development of the use of poly ortho aminophenol in the structure of SCs.

To better understand how each of the CPs is combined with different synthesized graphene oxides, Table 1 presents a summary of recent research results in this field.

Table 1

Summary of Electrochemical results of graphene/CPs Composites for SC electrodes in recent investigations.

Electrode materials	Synthesis method	specific capacitance	Power density	Energy density	Capacitance retention	Ref.
Graphene–PANi hybrid	rapid-mixture polymerization	489 Fg^{-1}	33.9 kW kg^{-1}	-	96%	[14]
Symmetric PPy-GO fiber	wet-spinning	100 ($mFcm^2$)	0.5 ($mW.cm^2$)	9.7×10^{-3} ($mWh.cm^{-2}$)	-	[15]
PANI/CNTs/graphene	interfacial polymerization	160 Fg^{-1}	25 kW kg^{-1}	20.5 Wh kg^{-1}	91%	[16]
PPy@rGO	electrochemical polymerization	15.9 ($mFcm^2$)	0.36 W cm^{-2}	4.3 mW h cm^{-2}	82%	[17]
graphene nanoplatelets/polythiophene	in situ chemical polymerization	673 F g^{-1}	23.55 W kg^{-1}	2.25 W h kg^{-1}	84.9%	[18]
PANI/GF	wet-spinning / chemical reduction	87.8 ($mFcm^2$)	-	12.2 μW h cm^{-2}	100%	[19]
FGO/POAP	electrochemical polymerization	965 F g^{-1}	943.1 W.kg^{-1}	44.3 Wh.kg^{-1}	94.4 %	[20]

2.2. Composites based on graphene / ACs

The most important feature of activated carbons, and specifically activated carbons extracted from biomass and biowastes, is their low final price, which is due to their availability and abundance in nature. In addition to waste valorization, helping to preserve the environment based on a circular economy is another benefit of using biomass. When using biomass, carbonization is first performed using thermal methods, and then carbon activation is carried out using various methods, such as a stirrer with KOH. Recently, activated carbons extracted from waste coffee beans [21], cotton [22], rapeseed [23], sunflower seeds [24], and grape seeds [25] have been used in the SC system. The main disadvantage of using these carbons in SC systems is their low specific capacitance, which requires scientists to combine them with materials with stronger conductivity.

Alberto Adan-Mas et al. After carbonizing waste coffee beans, they were chemically activated. The achievement of a substrate with a specific surface area of 2330 m^2 g^{-1} was remarkable [26]. Despite having a high specific surface area, which was very favorable for ion transport, the absence of transport elements resulted in a typical specific capacitance of 84 F g^{-1} at 1 A g^{-1}. Finally, the electroactive materials used in this study retained 85% of the initial capacity after 5000 continuous charge-discharge cycles.

In an innovation, Oladepo Fasakin et al. used cocoa pod husks to obtain activated carbon. They used a tube furnace with KOH as an activating agent [27]. Morphological studies of this material confirmed a sponge-like structure with high porosity and high specific surface area, which was very suitable for SC electrodes. The Coulombic efficiency was measured to be 99.6% by a galvanostatic charge-discharge test. The cyclic stability was also 72% after 5000 cycles. In this study, a specific capacitance of 168 F g^{-1} at 0.5 Ag^{-1} was obtained. Finally, an energy density of 19 Wh kg^{-1} was calculated at a power density of 453 W kg^{-1}. The results of this work proved that cocoa pod husks are a suitable choice for an electroactive material in energy storage and can be used as a practical substrate in further research.

In a similar study, Veeramaniab et al. used Bougainvillea spectabilis as a precursor material and converted it into a graphene sheet-like porous activated carbon by thermochemical activation methods [28]. Accurate electrochemical measurements such as cyclic voltammetry, galvanostatic charge-discharge, and differential pulse voltammetry were performed to investigate the energy storage properties. At a current density of 1.6 A g^{-1}, which was relatively high in bio-based systems, a specific capacitance of 233 F g^{-1} was calculated to be acceptable. In addition, an energy density of 7.2 W h kg^{-1} in a symmetrical two-electrode system indicated the successful synthesis of electroactive materials. Finally, this study introduced a suitable option for converting biomass into valuable materials for energy storage systems.

In a recent study, Ming-Che Cheng et al. used jute fiber, a low-value material. Simultaneous KOH activation was performed under a nitrogen plasma atmosphere [29]. The final AC had high amounts of heteroatoms with positive functional groups in the SC system. In the final step of the synthesis, this AC was composited between graphite layers. The present nanocomposite exhibited a remarkably high specific capacitance of 503 F g^{-1} at a current density of 0.5 A g^{-1}. The loss of only 8% of the initial specific capacitance after 5000 consecutive charge-discharge cycles indicated a good stability of the electroactive material structure. Furthermore, the energy density of 30.5 W h kg^{-1} gave more hope for overcoming the main weakness of the SC system.

Table 2 summarizes the results of recent research on the use of ACs as electroactive materials for SCs to better understand how the type of starting material and synthesis method affect the performance of the SC system.

Table 2

Summary of Electrochemical results of graphene/ACs Composites for SC electrodes in recent investigations.

Electrode materials	Synthesis method	specific capacitance	Power density	Energy density	Capacitance retention	Ref.
Auricularia biomass/GO	Chemical routes/modified Hummers method	256 F g^{-1}	-	-	92%	[30]
cotton waste/graphene aerogel	Chemical activation/modified Hummers method	305 F g^{-1}	-	-	94%	[31]
Lignin/GO	hydrothermal method	94 F g^{-1}	150 Wkg^{-1}	13.4 $Whkg^{-1}$	91%	[32]
coffee grounds-derived AC (cAC)/RGO	Chemical routes	440 F g^{-1}	438 W kg^{-1}	187.3 Wh kg^{-1}	81.4%	[33]
HRGO/BC	solution oxidation method	65.9 Fg^{-1}	112.9 Wkg^{-1}	9.2 $Whkg^{-1}$	88%	[34]
CeCoSx-SA/GF	Chemical routes	873.3 Fg^{-1}	801 Wkg^{-1}	29.58 $Whkg^{-1}$	87.1%	[35]
BPC/graphene	chemical self-assembly	176 F g^{-1}	5000 W kg^{-1}	6.11 Wh kg^{-1}	99.9%	[36]

2.3. Composites based on graphene / TMOs

TMOs have properties such as a narrow band gap, excellent conductivity, and low resistance [37-39]. These properties can well compensate for the weakness of SC systems (relatively low energy density). These materials, by following the Faradaic mechanism, transfer ions and electrons in the Helmholtz layer [40]. More precisely, charge storage is provided through oxidation and reduction half-reactions. The similarity of the charge storage mechanism in this SC group led the scholar to call this group a pseudocapacitor. Metal oxides such as NiO [41], RuO_2 [42], MnO_2 [43], Co_3O_4 [44], and V_2O_5 [45] were among the materials used in recent studies. Transition metals and metal oxides are very popular in recent research. Recently, nanoparticles extracted from these groups, such as bimetals, have attracted more and more attention by displaying excellent energy storage [46].

In 2016, Murugan Saranya et al. deposited zinc oxide between graphene sheets via the solvothermal method and synthesized Graphene-zinc oxide (G-ZnO) nanocomposite [47]. ZnO nanoparticles with crystal dimensions of approximately 50 nm were deposited between graphene sheets. In this study, KOH 6M was used as the electrolyte. Considering the low resistivity of ZnO and its high conductivity, it was important that the electrolyte also had ionic resistance. The solution resistance was measured to be 12.2 Ω. The specific capacitance was calculated from galvanostatic charge-discharge curves to be 122.4 F g^{-1}.

In light of this research, Jiuzeng Jin and colleagues synthesized a novel nanocomposite utilizing a one-step solvothermal method. To promote the acceleration of electron transport via synergistic effects, two metals, zinc (Zn) and nickel (Ni), were incorporated within a bimetallic framework. The markedly enhanced conductivity observed in this combination necessitated the use of a substrate with a high specific surface area, which was provided by graphene. The resulting nanocomposite exhibited distinctive properties, including a high specific surface area, a porous morphological structure, and low electrical resistance. Achieving an exceptional specific capacity of 1644 F g-

[1] signifies considerable advancement. Additionally, a high energy density of 47.4 Wh kg^{-1} at a power density of 825 W kg^{-1} was determined. The primary limitation of this study pertains to the comparatively low structural stability, as the material retained approximately 85.1% of its initial capacity after 3000 cycles.

Table 3 summarizes the results of recent research on the use of TMOs in SC systems.

Table 3

Summary of Electrochemical results of graphene/TMOs Composites for SC electrodes in recent investigations.

Electrode materials	Synthesis method	specific capacitance	Power density	Energy density	Capacitance retention	Ref.
MnO_2/RGO	filtration deposition and thermal reduction	333.9 F g^{-1}	1716.9 Wkg^{-1}	23.5 Wh kg^{-1}	87%	[48]
Ni–Mn@C/rGO	one-pot hydrothermal	1674 F g^{-1}	444.4 Wkg^{-1}	24.1 $Whkg^{-1}$	90%	[49]
bimetallic sodium–zinc/rGO	Chemical routes	435.2 F g^{-1}	-	-	92%	[50]
Co_3O_4/GO	thermal treatment	114.1 F g^{-1}	225 W kg^{-1}	35.7 Wh kg^{-1}	95%	[51]
2D Zn/Cu-BTC/GO	in-situ solvothermal	1284.9 Fg^{-1}	10 $KWkg^{-1}$	60.55 $Whkg^{-1}$	92%	[52]
$ZnCo_2O_4$/rGO/sponge	hydrothermal reaction and thermal annealing	1116.6 Fg^{-1}	Wkg^{-1}	$Whkg^{-1}$	93.4%	[53]
CoFe-MOF/rGO	Solvothermal	2069.1 F g^{-1}	700 W kg^{-1}	75.8 Wh kg^{-1}	91.3%	[54]

According to the results of recent research, it still seems that the best option for electroactive materials in SC systems is to use several nanomaterials with specific purposes together in the form of a multiple nanocomposite. For better understanding, in studies that used CPs, the structural stability was usually high because the CP, like a slurry, maintained the electrode structure. Also, when researchers intended to have a high specific surface area, they used ACs. Similarly, if the goal was to achieve high conductivity and low resistance, TMOs were the best option. These features make the simultaneous use of these three categories in the form of a nanocomposite the best choice, and it seems that the future of these systems depends on the best combination of these materials with each other.

3. Conclusion

This review, by categorizing the results of recent research in the field of using graphene-based composites and combining them with innovative materials such as conductive polymers (CPs), activated carbons (ACs) extracted from biomass, and transition metal oxides (TMOs) nanoparticles, provides a good comparison and facilitates the selection of appropriate options in future research. The morphology of the electrode active materials used in the electrode structure and their discussion are among the other issues examined in the present work. It appears that binary and ternary nanocomposites, which combine nanomaterials and leverage the advantages of each, through a synergistic effect mechanism, will create a promising future for clean, affordable, and renewable energy systems.

Conflicts of interest

There are no conflicts to declare.

Acknowledgment:

The financial support of this work by the *Amirkabir University of Technology*, is gratefully acknowledged.

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