Review

Green-Synthesized Graphene for Supercapacitors—Modern Perspectives

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Abstract: Graphene is a unique nanocarbon nanostructure, which has been frequently used to form nanocomposites. Green-synthesized graphene has been focused due to environmentally friendly requirements in recent technological sectors. A very important application of green-synthesized graphene-based nanocomposite has been observed in energy storage devices. This state-of-the-art review highlights design, features, and advanced functions of polymer/green-synthesized graphene nanocomposites and their utility in supercapacitor components. Green graphene-derived nanocomposites brought about numerous revolutions in high-performance supercapacitors. The structural diversity of conjugated polymer and green graphene-based nanocomposites has facilitated the charge transportation/storage capacity, specific capacitance, capacitance retention, cyclability, and durability of supercapacitor electrodes. Moreover, the green method, graphene functionality, dispersion, and matrix–nanofiller interactions have affected supercapacitance properties and performance. Future research on innovative polymer and green graphene-derived nanocomposites may overcome design/performance-related challenging factors for technical usages.

Keywords: green; graphene; nanocomposite; polymer; supercapacitor

1. Introduction

Graphene is a one-atom thick, two-dimensional nano-allotrope of carbon [1]. Graphite is a common carbon structure consisting of layers of graphene [2–5]. The word 'graphene' is derived from graphite with the suffix '-ene'. Graphene nanosheets are stacked together through weak dispersion forces. Owing to unique structure and exclusive properties, graphene nanosheets have been considered as a remarkable candidate for functional nanocomposites [6,7]. Graphene has been synthesized using numerous top-down and bottom-up methods [8]. The most common, inexpensive, and facile method initially used for the production of graphene include the mechanical and thermal exfoliation of graphite [9–11]. Isolated graphene is a free-standing nanosheet. For large-scale synthesis of graphene, due to its large surface area and superior chemical and physical properties, advanced techniques like the chemical vapor deposition (CVD) technique have been frequently adopted [12]. According to recent requirements of eco-friendly materials and methods, green technologies have been used to develop graphene and graphene-based materials [13]. Consequently, graphene has been investigated as an important material in

supercapacitors [14]. Specifically, green-synthesized graphene and derived nanocomposites have been applied in supercapacitor applications [15,16]. In supercapacitors, the conjugated polymers such as polyaniline, polypyrrole, polythiophene, etc., have been widely used due to their high electrical conductivity, heat stability, and durability properties [17]. Particularly, conjugated polymer-based nanocomposites have been used for supercapacitor components [18]. In this regard, carbon nanofillers have enhanced the electron transportation and physical properties of conjugated polymers for technical applications. Among nanocarbons, graphene nanocomposites have been effectively investigated for energy storage devices [19,20]. In supercapacitor electrodes, graphene-derived materials revealed high surface area, electrical conductivity, and electrochemical properties [21].

In this review, the design of green-synthesized graphene, related nanocomposites, and resulting supercapacitor components have been studied. Green methods have efficiently contributed to the fabrication of green and ecologically safe graphene-based materials. The electrical conductivity, specific capacitance, capacitance retention, recyclability, and durability of supercapacitor electrodes/electrolytes depend upon the matrix–nanofiller interactions, graphene contents, functionality, and dispersion in the matrix. Thus, using green polymer/graphene nanomaterials has positively affected supercapacitor performances. For high-performance supercapacitor devices, the design of advanced green-synthesized graphene-based materials must be examined.

2. Graphene

Graphene is a nanosheet made up of sp^2 hybridized carbon atoms [22]. It is a honeycomb lattice nanostructure consisting of sp^2 hybridized carbon atoms [23]. Graphene can be considered as a derived form of graphite (having stacking graphene layers) [24]. Graphene possess van der Waals interactions, which may cause wrinkling and restacking of the nanosheet [25]. Graphene has high surface area, electron transport, thermal conductivity, Young's modulus, and strength properties [26]. Due to its unique structure and properties, graphene has gained a distinct position among nanocarbon nanoparticles [27]. Graphene has been applied to synthesize various nanomaterials [28]. Moreover, it can be functionalized or modified to form graphene oxide, reduced graphene oxide, and functional graphene structures [29]. Graphene, graphene oxide, reduced graphene oxide, and other graphene derivatives have been employed as effective nanofillers to form polymeric nanocomposites [30]. Numerous application areas have been identified for graphene and derived materials such as energy devices, electronics, engineering, biomedical, and nanocomposites [31–33].

3. Graphene in Supercapacitors

Supercapacitors have been identified as the most efficient energy storage devices [34–36]. Supercapacitors have the advantages of efficient charge storage, reversibly, and chemical stability [37–39]. In these charge storage devices, nanocarbon nanomaterials have been used due to their high electron transportation, optimum porosity, and durability properties [40]. Graphene has high surface area, electrical conductivity, and mechanical stability properties, in addition to superior charge storage to be utilized in supercapacitor components [41]. Due to optimum porosity and pore size distribution in graphene-based electrodes, electrolyte has easy accessibility to the electrode surface. Yan et al. [42] developed highly corrugated graphene sheets and thermally exfoliated graphene sheets for supercapacitor electrodes. Graphene oxide was prepared using the green Hummer's method using eco-friendly reagents. The thermal reduction was adopted as a green method for the conversion of graphene oxide to thermally exfoliated graphene sheets at a high temperature of 900 °C (Figure 1). Liquid nitrogen treatment was used for the conversion of thermally exfoliated graphene sheets to highly corrugated graphene sheets. The highly corrugated graphene, synthesized using green method, has the least agglomeration and restacking tendencies. Thus, in a supercapacitor electrolyte, the corrugated graphene has a high surface area ($517.9 \ \mathrm{m^2 \ g^{-1}}$), leading to high specific capacitance of $349 \ \mathrm{Fg^{-1}}$, relative to the thermally

exfoliated graphene (183 Fg^{-1}) (Figure 2). It was observed that the capacitive performance of the green-synthesized graphene was considerably higher than the environmentally hazardous hydrazine vapor-reduced graphene (155 Fg^{-1}) [43].
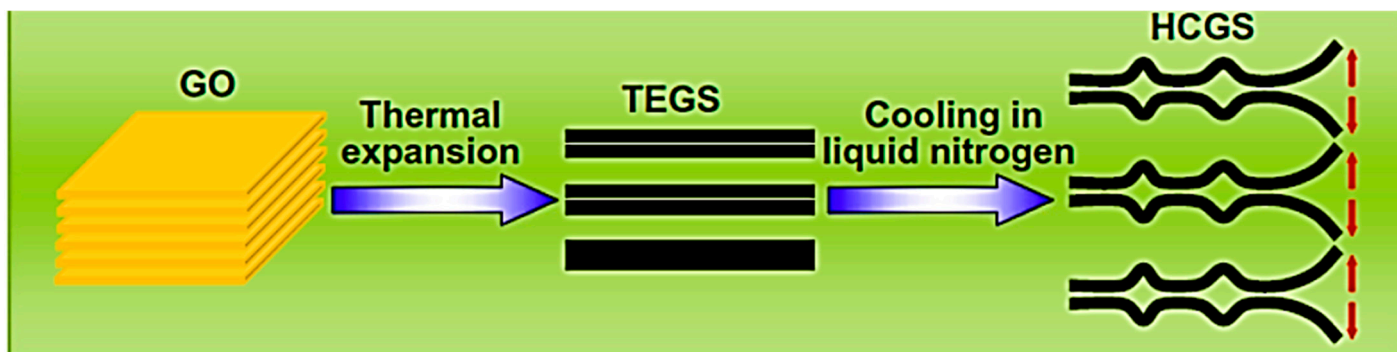

Figure 1. Schematic of formation of TEGS and HCGS [42]. GO = graphene oxide; HCGS = highly corrugated graphene sheets; TEGS = thermally exfoliated graphene sheets. Reproduced with permission from Elsevier.
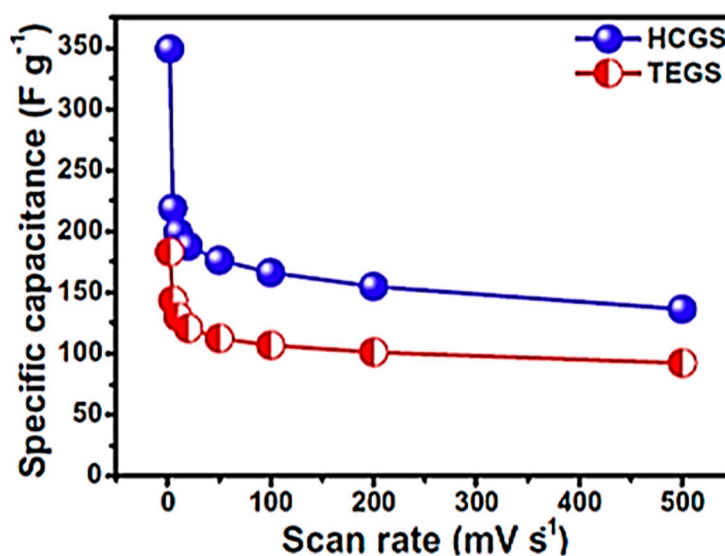

Figure 2. Specific capacitance of HCGS and TEGS electrodes at different scan rates in 6 M KOH solution [42]. HCGS = highly corrugated graphene sheets; TEGS = thermally exfoliated graphene sheets. Reproduced with permission from Elsevier.

4. Green Synthesis of Graphene

The industrial scale mass production of graphene has been focused due to its technical applications [44]. In this regard, chemical reduction oxidation techniques have been investigated for the large-scale synthesis of graphene. However, using hydrazine and other toxic regents in graphene production has produced human health hazards and environmentally harmful effects [45,46]. Alternatively, various facile, safe, green, and low-cost methods have been focused upon for the graphene production. Most importantly, micromechanical exfoliation [47] and chemical vapor deposition [48] have been adopted as green synthesis approaches to form graphene. Green synthetic protocols have been used to synthesize safe and green graphene [49]. Green techniques involve the use of green synthesis routes following green chemistry, eco-friendly reagents and solvents, and green energy consumption methods for graphene production [50,51]. Incidentally, green, inexpensive, and effective biomass-reducing agents have been used to form reduced graphene oxide (RGO) [52]. Green methods have the advantages of low-cost production and ecofriendly processing [53]. Micromechanical exfoliation has been used as a low-cost

and green methodology for graphene synthesis [54]. This method may involve using graphite interlayer compounds for the exfoliation of graphene nanosheets [55]. This is not only an inexpensive and eco-friendly technique, but also yields high-quality graphene [56]. However, micromechanical exfoliation has limitations for the large-scale production of graphene. Moreover, chemical vapor deposition (CVD) synthesis has been used to form green planar graphene nanostructures [57]. In CVD, transition metal catalysts have been used to synthesize graphene with controlled thickness, low surface defects, and surface properties [58]. This method has the advantages of low temperature processing and no use of corrosive solvents [59].

Graphene has been prepared using renewable carbon sources [60]. The synthesis of graphene and its derivatives usually requires expensive and toxic reagents. Consequently, research efforts have been focused towards the use of sustainable routes [61]. The synthesis, performance, and applications of graphene-derived nanomaterials prepared from renewable carbon sources and reduction by green alternatives have been of specific interest [62]. In this regard, the graphitization of waste carbon precursors has been used to attain high purity graphene derivatives. Here, the graphitization of waste carbon precursors has been performed using CVD, hydrothermal, laser ablation, etc. Green reduction of graphene oxide has been carried out using plant extracts from fruits, leaves, etc., containing a high concentration of phenolic compounds [63,64]. Green extracted phenolic compounds have been used a as carbon source for graphene synthesis [65–67]. Renewable carbon sources are usually inexpensive, non-toxic, non-flammable, and environmentally friendly [68–70]. Different hydrocarbon sources have been used for graphene synthesis [71–73]; Nasir et al. [61] produced graphene oxide using oil palm leaves, palm kernel shells, and empty fruit bunches. Carbonization of the green materials was performed at 400–900 °C in furnace, under a nitrogen atmosphere. The graphene oxide was synthesized from as-carbonized materials. The surface area of graphene oxide, obtained by the Brunauer—Emmett—Teller technique, was 117 $m^2\,g^{-1}$. Salifairus et al. [74] used palm oil as a natural carbon precursor to prepare graphene. The thermal chemical vapor deposition was applied to obtain graphene. Likewise, Qu et al. [75] performed the synthesis of graphene sheets from alfalfa plants. Kalita and co-workers [76] synthesized large surface area graphene nanosheets using camphor plant derivatives. Zhang Kalita and co-workers [77] prepared high-quality graphene using glucose and ferric chloride. Nguyen and co-researchers [78] applied biscuits, chocolate, grass, and plastic on copper foil in a CVD technique for graphene synthesis. However, these techniques need further developments for the commercial-level production of graphene.

Graphene synthesis from biomass like sugars, Escherichia coli, microorganisms, etc., have gained noteworthy research attention [79]. Eco-friendly reducing agents have been used for the safe production of graphene [80]. Upadhyay and co-workers [81] employed grape (*Vitis vinifera*) extracts as a green reducing agent for the synthesis of reduced graphene oxide. This method was proved inexpensive and feasible for large-scale production. Graphene oxide was widely prepared using the modified Hummer's method [82]. Graphene oxide was then converted to reduced graphene oxide using a green route employing *Vitis vinifera* with refluxing (95 °C), filtering, and drying (Figure 3). Figure 4 shows the X-ray diffraction of graphite, graphene oxide, and reduced graphene oxide. The formation of graphene oxide (from graphite) was identified by the appearance of a peak at 2θ = 10.4°. The reduced graphene oxide was characterized by the appearance of a broad peak at 23.7°, indicating the presence of few layered graphene. Transmission electron microscopy image revealed a graphene structure with a few layers in the reduced graphene oxide sample. The green-synthesized nanosheet was found to be slightly wrinkled and corrugated.
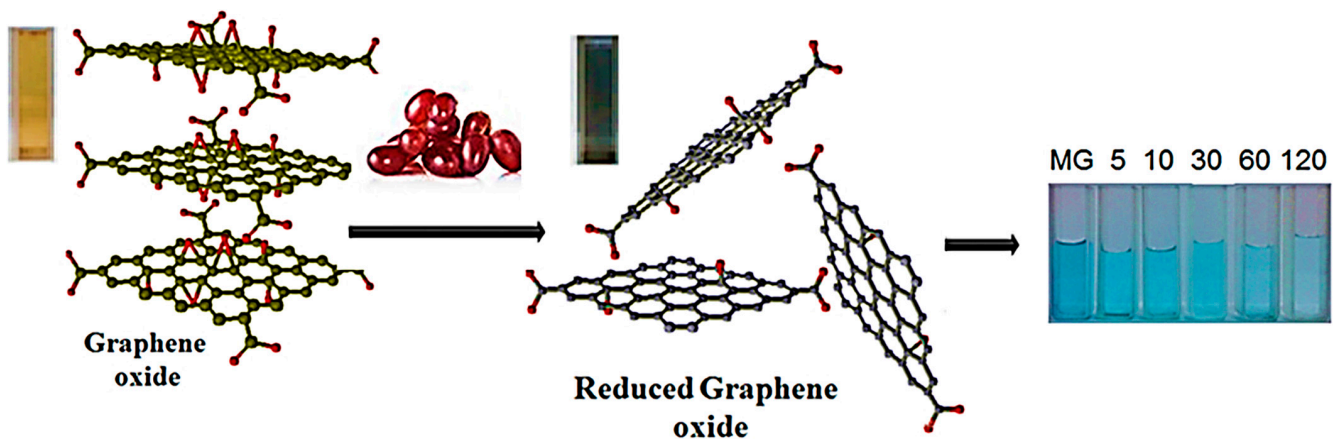
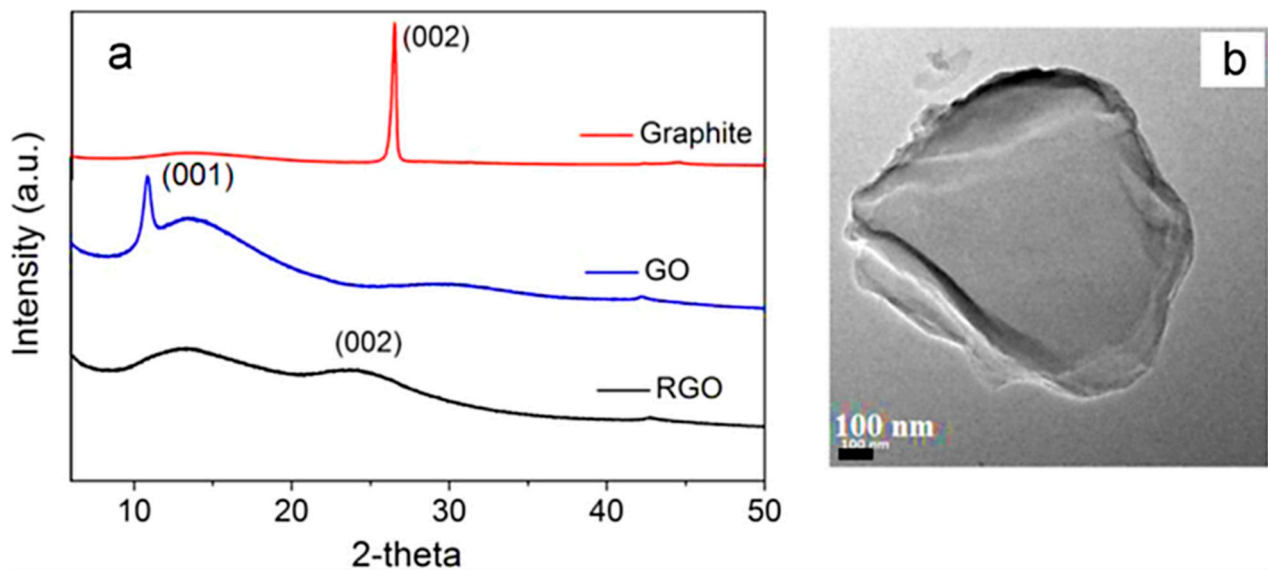**Figure 3.** Green synthesis of reduced graphene oxide [81]. Reproduced with permission from Elsevier.

Figure 4. (**a**) X-ray diffraction of graphite, GO, and RGO and (**b**) TEM image of RGO sample [81]. GO = graphene oxide; RGO = reduced graphene oxide; TEM = transmission electron microscopy. Reproduced with permission from Elsevier.

Tavakoli et al. [83] used pomegranate juice to synthesize a graphene nanosheet from graphene oxide. First, Hummer's method was used to form graphene oxide from a graphite precursor. Next, the graphene oxide was converted to reduced graphene oxide in the presence of pomegranate juice (having anthocyanins as reducing agent). Consequently, the anthocyanins reducing agent (electron deficient) removed the oxygen functional groups from the graphene oxide surface to generate graphene nanosheets. Figure 5 presents a mechanism for the formation of graphene from graphene oxide via a green route.

Vitamin C has been applied as a green bio-precursor for the reduction of graphene oxide to graphene [84]. Vitamin C has been successfully used as a green alternative to the environmentally hazardous hydrazine during graphene synthesis. This green and pollution-free method yielded high-quality and wrinkle-free graphene, with high electronic conductivity [85]. L-ascorbic acid has been effectively used as a source of vitamin C-based green reducing agent to form graphene. Gao et al. [86] adopted a green method for graphene production using the eco-friendly Vitamin C (reductant) and amino acid (stabilizer). Accordingly, this method avoids using any toxic reagents for graphene production. The resulting graphene has high electron transportation. Thus, Vitamin C sources have been found as safe reducing agents to yield high-quality graphene at a large scale, thus avoiding the toxic reagents like hydrazine [87].
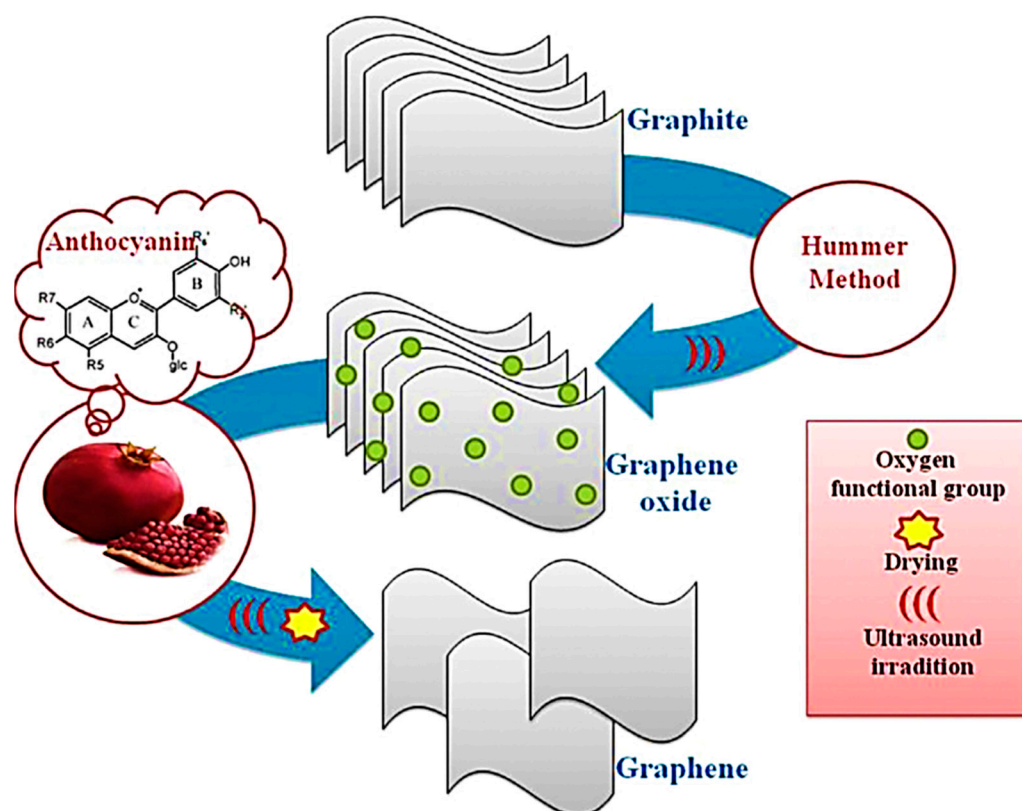
Figure 5. Mechanism of forming graphene nanosheets by using pomegranate via a green route [83]. Reproduced with permission from Elsevier.

5. Green-Synthesized Graphene-Based Nanocomposites for Supercapacitors

Flexible supercapacitors based on polymers and polymeric nanocomposites have been explored for high-specific capacitance and power density performance [88–90]. Flexible graphene-based supercapacitors have been applied in the advanced electronic devices [91–93]. Here, the mechanical flexibility of materials was desirable to form the graphene-based flexible supercapacitors [94]. Bressi et al. [95] surveyed fossil resources for the development of graphene and derivatives, according to growing environmental impacts. Several green routes have been used for exploiting renewable resources towards facile, low-cost, and eco-friendly synthetic processes [96,97]. Subramanya et al. [98] used a facile green one-pot method for the synthesis of few-layered graphene nanosheets. In this green method, graphene was obtained directly from graphite in an aqueous medium, using a regenerative catalyst. The as-synthesized graphene had large capacitance of 219 Fg^{-1} and high energy density of 83.56 $W\,h\,kg^{-1}$. Excellent cyclability—over 3000 cycles—was observed for the green-synthesized graphene-based electrodes. Liu et al. [99] used a facile green method for the formation of graphene oxide and graphene oxide sheets—Prussian blue nanocomposite. This green method involves using the redox reaction of $FeCl_3$ and $K_3[Fe(CN)_6]$ in an aqueous solution. The cyclic voltammograms of graphene oxide are given in Figure 6. The green-derived electrodes were tested in the electrochemical cell with H_2O_2. The cathodic peak current and anodic peak current increased and decreased, respectively. The changes were observed due to the electrochemical catalytic reaction between the electrode and H_2O_2. The electrode had higher sensitivity toward the electro-catalytical reduction of H_2O_2. The exceptional electrochemical properties and green preparation revealed potential in the field of electrochemical sensors [100].
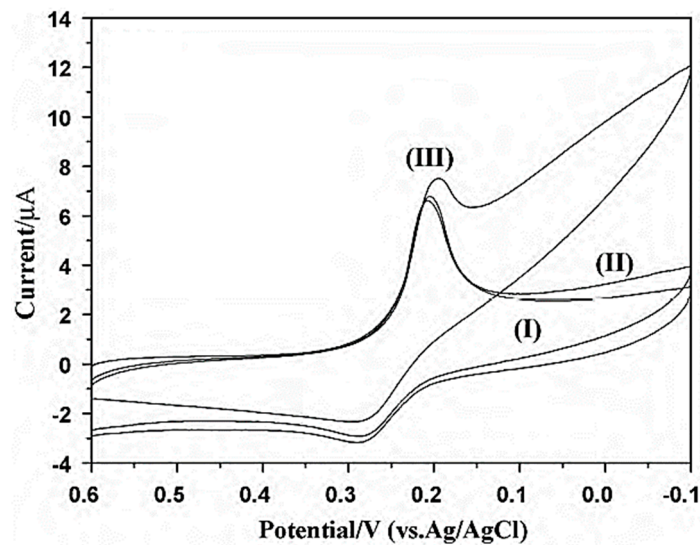
Figure 6. Cyclic voltammograms of green-synthesized graphene oxide nanomaterial [99]. (I) = 0 mM; (II) = 0.1 mM; and (III) = 0.5 mM of H_2O_2 at scan rate of 50 mVs^{-1}. Reproduced with permission from Elsevier.

Liu et al. [101] used the green electrochemical synthetic route with series of electro-chemical processes to form an expandable graphene sheet and platinum nanoparticle-deposited expandable graphene sheet. The conductive indium tin oxide glass electrode was used as a substrate. The catalytic activity and stability of the synthesized electrodes were measured using cyclic voltammetry. The as-synthesized nanocomposite had high surface area, electrical conductivity, catalytic activity, and good stability. CV curves were scanned to analyze the comparative performance of the Platinum/expandable graphene sheet and Platinum/glass carbon composite electrode (Figure 7). The electrodes were cycled repeatedly to achieve a steady state. The Platinum/expandable graphene sheet had a forward peak current density of 7.41 mAcm^{-2} at 0.92 V. The more positive current density of the Platinum/expandable graphene sheet was credited to the residual oxygen containing functional groups on the surface.

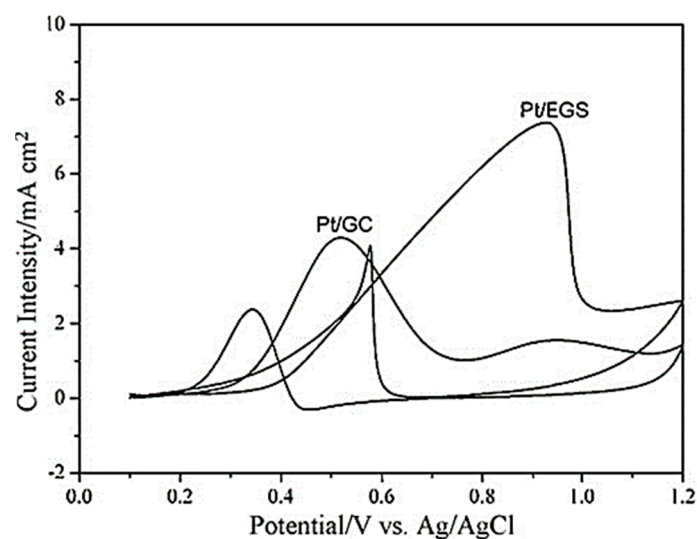**Figure 7.** CV curves of Pt/EGS and Pt/GC electrodes in mixed solution of 0.5 M CH_3OH and 0.5 M H_2SO_4 at 5 mVs^{-1} [101]. Pt/EGS = Platinum/expandable graphene sheet; Pt/GC = Platinum/glass carbon composite electrode. Reproduced with permission from Elsevier.

Conducting polymers have been efficiently applied as electrodes, revealing high electrochemical performance [102–104]. Conducting polymer/graphene nanocomposites have electroactive regions for the diffusion of ions in supercapacitor electrodes [105,106]. Polyaniline-based electrodes have a high specific surface area of 2600 $m^2 g^{-1}$ [107]. Wu et al. [108] designed a polyaniline nanofiber/graphene nanocomposite-based supercapacitor electrode. The electrode had a high specific capacitance of 210 Fg^{-1}. The specific capacitance of the green-synthesized electrode was found to be comparable to that of the non-green-synthesized electrode reported in the literature [109].

Graphene has gained increasing interest for the energy storage applications [110–112]. Graphene oxide and reduced graphene oxide have also been focused for charge storage due to their low-cost, high surface area, and high conductivity properties [113–115]. Green-synthesized graphene-based materials have been applied for eco-friendly energy storage electrodes [116–118]. To form graphene using the green route, Cetraria Islandica L. Ach extract has been used to improve the dispersibility and chemical reduction of graphene oxide nsnosheets [119–121]. The green method also has the advantages of cost-effectiveness and producing high-quality graphene. Çıplak et al. [122] designed the polyaniline, graphene oxide-gold@polyaniline, and reduced graphene oxide-gold@polyaniline nanocomposites for supercapacitor application. In the green method, Cetraria Islandica L. Ach lichen extract was used for the reduction of graphene oxide to reduced graphene oxide. Through in-situ polymerization, aniline monomer and gold nanoparticles were reacted to form the graphene oxide-gold@polyaniline and the reduced graphene oxide-gold@polyaniline nanocomposites (Figure 8).
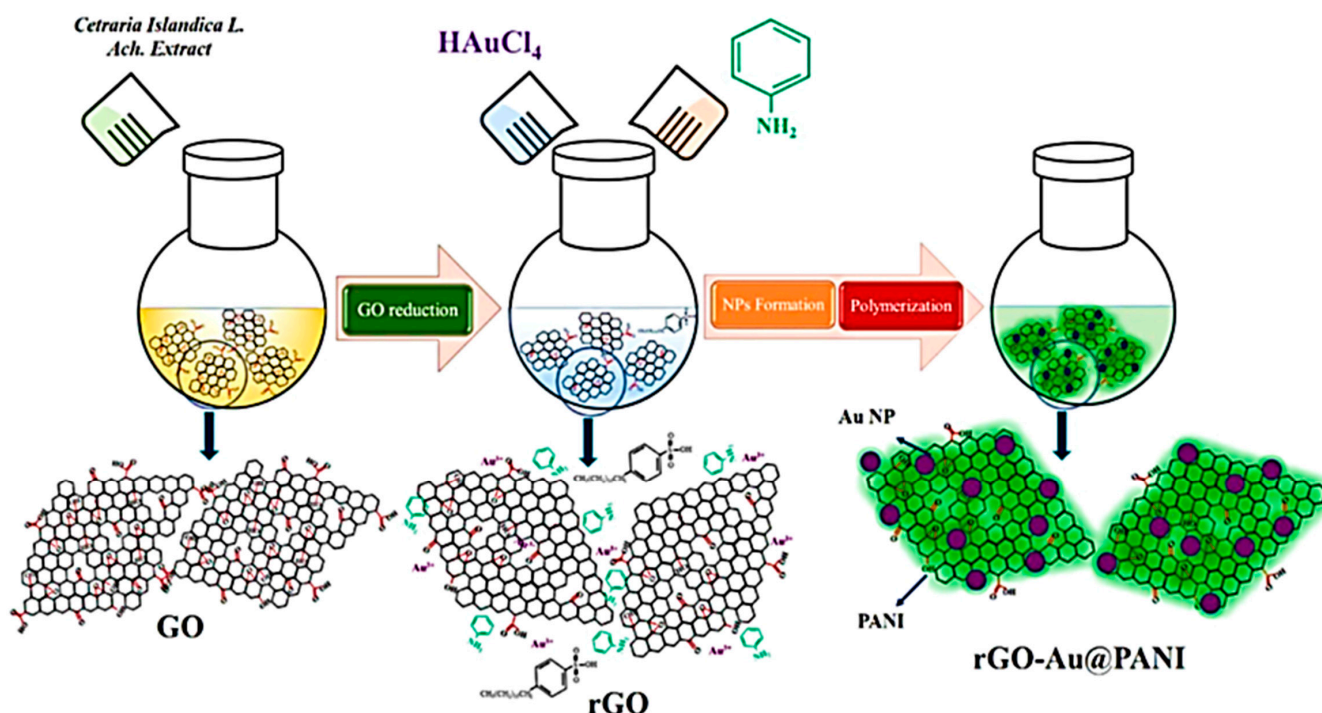
Figure 8. Schematic representation of preparation of rGO-Au@PANI nanocomposite [122]. PANI = polyaniline; GO = graphene oxide; rGO = reduced graphene oxide; rGO-Au@PANI = reduced graphene oxide-gold@polyaniline. Reproduced with permission from Elsevier.

The π–π interactions were developed between the polyaniline graphene nanosheets. Figure 9 shows CV curves for the polyaniline, reduced graphene oxide, and related nanocomposites, at scan rate of 10 mV/s in 1 M H_2SO_4 electrolyte. The integrated area in the CV curves was directly proportional to the specific capacitance of the electrode. Moreover, the diagram shows the capacitance performance of neat polyaniline and the related nanocomposites, in the range of a 5–200 mV/s sweep rate. It was observed that the

reduced graphene oxide-gold@polyaniline nanocomposite had higher specific capacitance of 212.8 Fg^{-1} (1 Ag^{-1}) and capacitance retention of 86.9%, relative to graphene oxide-gold@polyaniline nanocomposite and pristine polyaniline. The results suggested that the use of green reduced graphene oxide has enhanced the contribution of polymer chains to the charge diffusion, and so improved the rate capability of the supercapacitor electrode, compared with the literature non-green-synthesized polyaniline/graphene supercapacitor electrode [123].
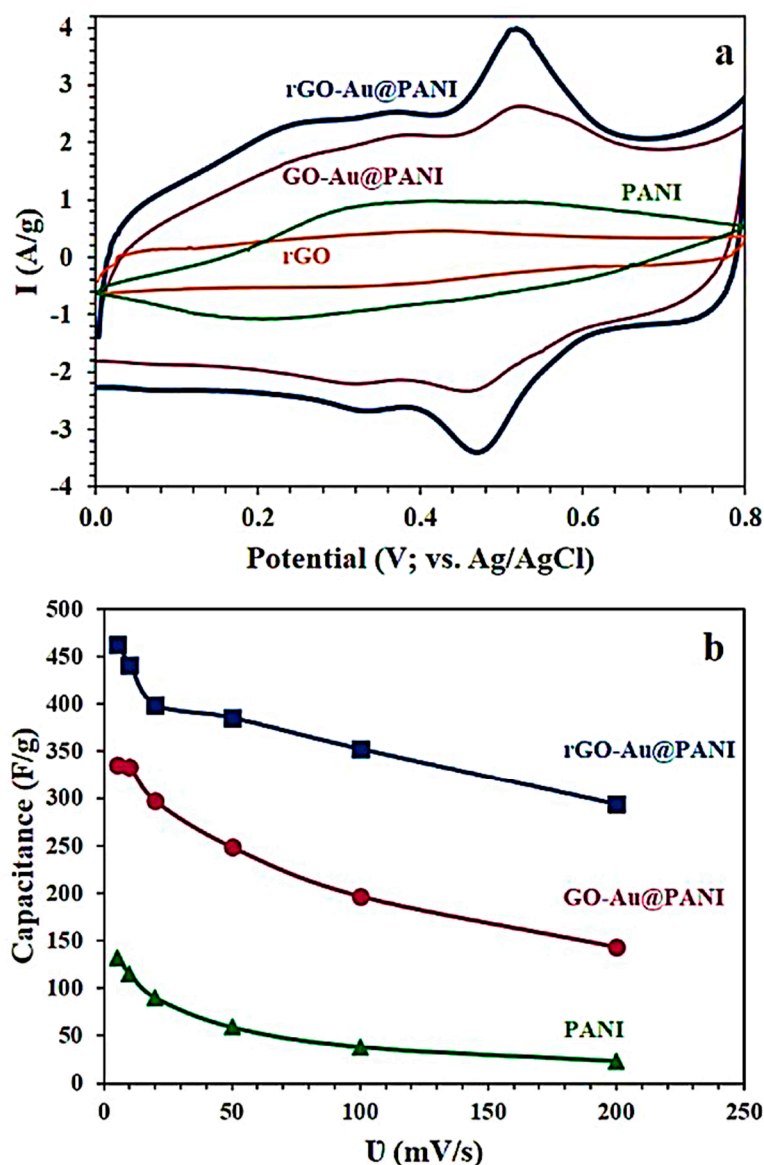
Figure 9. (**a**) CV curves of rGO, pristine PANI, rGO-Au@PANI, and GO-Au@PANI at scan rate of 10 mV/s in 1 M H_2SO_4 electrolyte; CV curves and (**b**) Variation of specific capacitance vs. scan rate (5–200 mV/s) for rGO-Au@PANI, GO-Au@PANI, and PANI [122]. rGO = reduced graphene oxide; rGO-Au@PANI = reduced graphene oxide-gold@polyaniline; GO-Au@PANI = graphene oxide-gold@polyaniline; PANI = polyaniline. Reproduced with permission from Elsevier.

Arthisree et al. [124] prepared the polyacrylonitrile/polyaniline@graphene nanocomposite via green route for supercapacitor application. A graphene quantum dot was prepared using a green approach (Figure 10). Afterwards, different graphene quantum dot contents were reinforced in the nanocomposites. The nanocomposite electrode was fabricated using eco-friendly drop casting and screen-printing techniques. In the nanocomposite, the proton-coupled electron-transfer was observed between polyacrylonitrile, polyani-

line, and the graphene quantum dot. According to current-voltage measurements, the nanocomposite with 1.5 wt.% nanofiller contents had higher electrical conductivity of 2.362×10^{-6} Sm^{-1}, relative to neat polymers and the nanocomposite with lower nanofiller contents (Figure 11). The higher electrical conductivity led to enhanced carrier density and specific capacitance properties. The CV analysis were performed to study the electrochemical characteristics of the prepared nanocomposites (Figure 12). The study was carried out in 0.1 M H$_2$SO$_4$ electrolyte, at a scan rate of 50 mVs^{-1}. The difference in the current signals of the graphene nanostructure, polyacrylonitrile/polyaniline blend, and nanocomposites can be distinguished. The CV curve of the graphene nanostructure revealed an excellent electrical double-layer capacitor [125]. The CV curve of the blend had a larger conducting area than that of the pure graphene nanostructure. The reason seems to be the high surface area and pseudocapacitance of the conjugated polymer [126]. The CV scans of the nanocomposites revealed enhancement in the area of cathodic and anodic portions, with increasing nanofiller loading. The results suggested the higher pseudocapacitance mechanism of the nanocomposite material with increasing graphene loading levels [127].
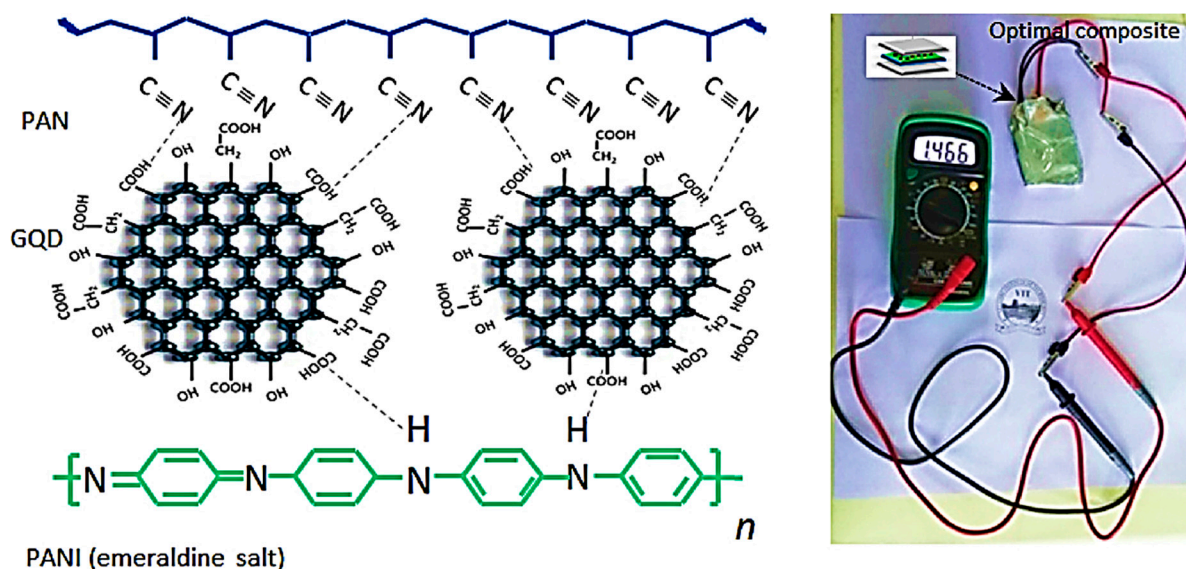

Figure 10. Illustration of PAN/PANI@G nanocomposite and its proton-coupled electron-transfer reaction responsible for the pseudocapacitance activity [124]. PAN/PANI@G = polyacrylonitrile/polyaniline@graphene; GQD = graphene quantum dot; PANI = polyaniline. Reproduced with permission from Elsevier.

Li et al. [128] designed the cellulose/polyaniline and cellulose/graphene oxide /polyaniline nanocomposite. A green method was used to form the nanocomposite using the in-situ polymerization, as illustrated in Figure 13. The areal specific capacitance of the cellulose/polyaniline and cellulose/graphene oxide/polyaniline nanocomposite is given in Figure 14. The cellulose/graphene oxide/polyaniline nanocomposite with 3.5 wt.% nanofiller formed a three-dimensional structure revealing high electrical conductivity of 1.15 Scm^{-1} and a significantly high areal specific capacitance of 1218 mFcm^{-2}. Moreover, the cellulose/graphene oxide/polyaniline nanocomposite had higher energy density (258.2 µWh/cm^2) and power density (1201.4 µW/cm^2) for the high-performance supercapacitors. Figure 15 depicts the CV performance of the cellulose/polyaniline, cellulose/graphene oxide, and cellulose/graphene oxide/polyaniline nanocomposites. Inclusion of 3.5 wt.% nanofiller contents in cellulose/graphene oxide/polyaniline nanocomposite revealed promising electrical properties. The enhanced performance was observed due to the synergistic effect of the graphene oxide and polyaniline as conducting fillers in the cellulose matrix [129]. A comparable supercapacitor electrode prepared by Luo et al. [130], using a non-green route, had a lower specific capacitance of 645 Fg^{-1}. The higher capacitance indicated the effectiveness of the green method used. Similar other

attempts of using non-green synthesis methods have revealed a lower specific capacitance of 486 Fg^{-1} [131].
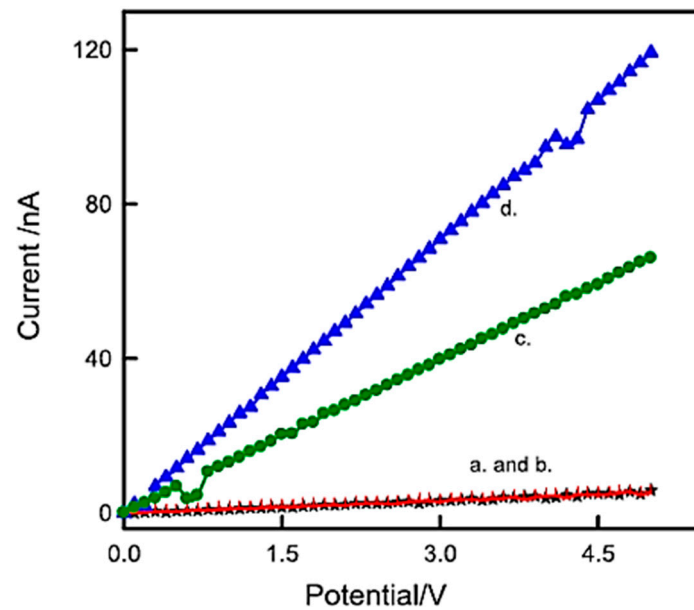
Figure 11. I-V characteristics of various polymer nanocomposite systems (**a**) PAN/PANI blend; (**b**) PAN/PANI@G-0.5 wt.%; (**c**) PAN-PANI@G-1 wt.%; and (**d**) PAN-PANI@G-1.5 wt.% [124]. PAN/PANI = polyacrylonitrile/polyaniline; PAN/PANI@G = polyacrylonitrile/polyaniline@graphene. Reproduced with permission from Elsevier.
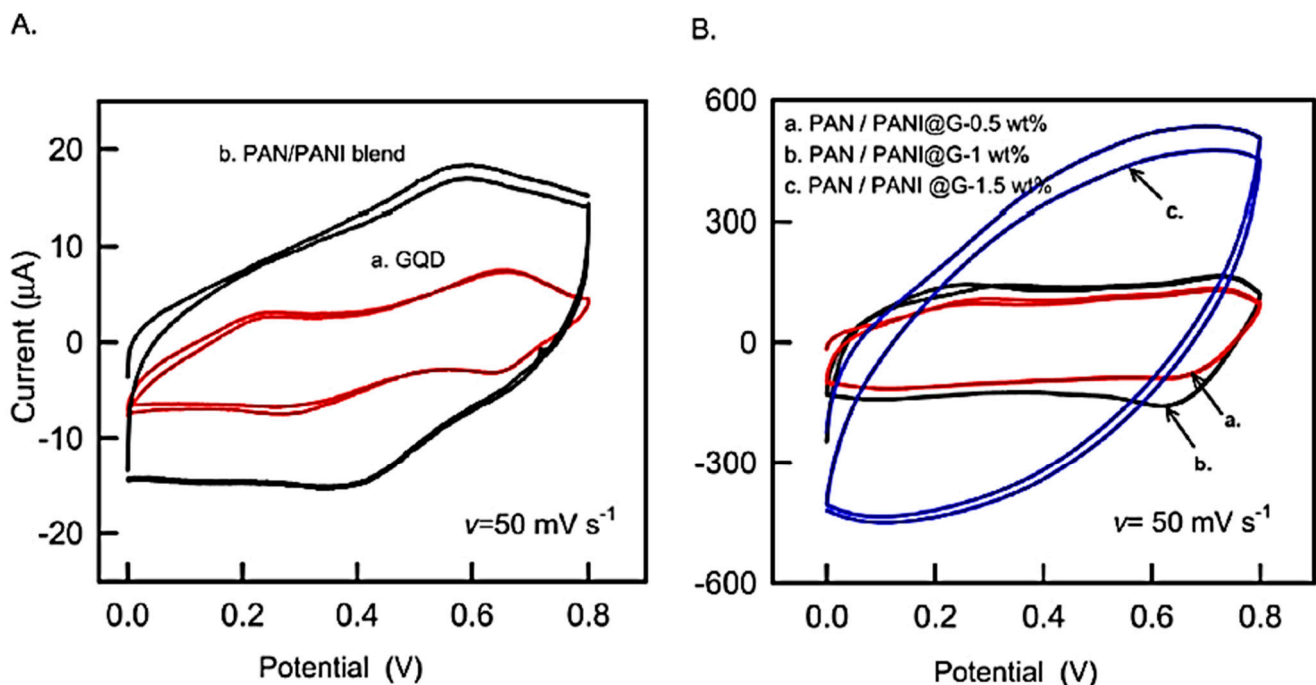
Figure 12. (**A**) CV response of a. GQD, b. PAN-PANI blend and (**B**) CV response of PAN/PANI@G-0.5 wt.%, PAN/PANI@G 1 wt.%, PAN/PANI@G 1.5 wt.% nanocomposites [124]. PAN/PANI = polyacrylonitrile/polyaniline; PAN/PANI@G = polyacrylonitrile/polyaniline@graphene. Reproduced with permission from Elsevier.
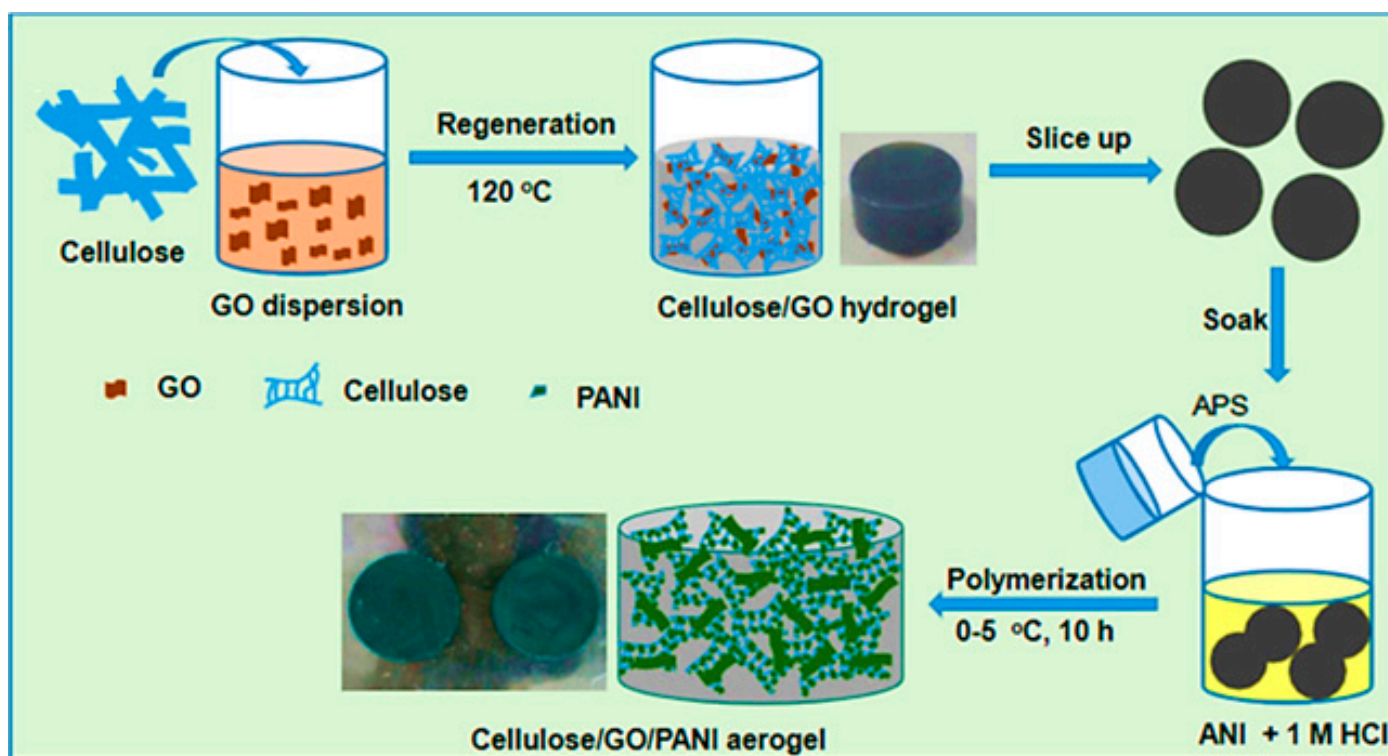
Figure 13. Synthetic pathway of the cellulose/graphene oxide/polyaniline [128]. GO = graphene oxide; PANI = polyaniline; ANI = aniline. Reproduced with permission from MDPI.
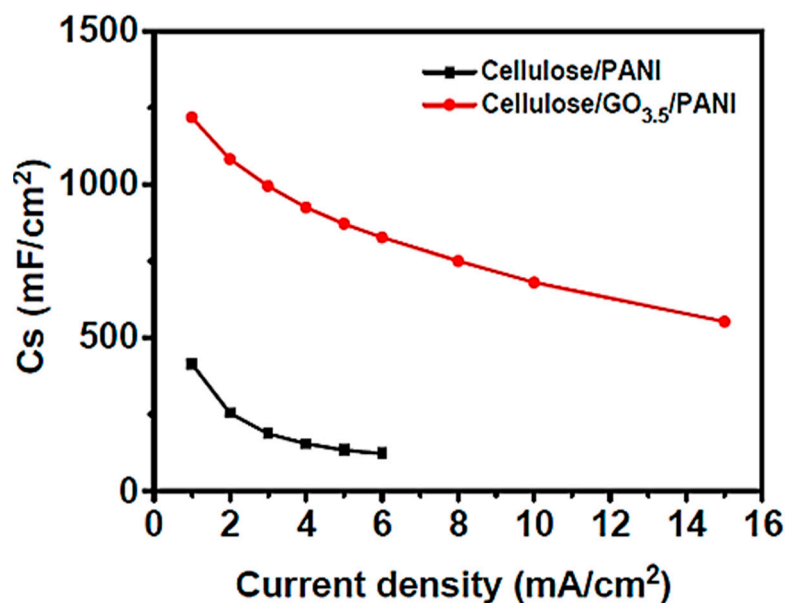
Figure 14. Plots of areal specific capacitance vs. current for cellulose/polyaniline and cellulose/graphene oxide/polyaniline [128]. Reproduced with permission from MDPI.

Polypyrrole has been applied as an important conducting polymer in supercapacitors [132–134]. Research attempts have been observed on polypyrrole/graphene nanomaterials for supercapacitor applications [135]. Sahoo and co-workers [136] designed polypyrrole/graphene nanocomposites for a supercapacitor electrode. The polypyrrole/graphene nanocomposite-based electrode revealed reasonable electrical conductivity (1.45 Scm^{-1}) and specific capacitance (466 Fg^{-1}), due to the synergetic effect of conjugated polymer and graphene nanofiller. Biswas and Drzal [137] reported on the polypyrrole/graphene

nanocomposite electrode. The specific capacitance of 165 Fg^{-1} was attained along with good electrochemical cyclic stability. Lim and co-researchers [138] fabricated the polypyrrole/graphene-based electrode via electrochemical polymerization. The capacitance of polypyrrole was found to improve with the graphene addition [139]. Zhou and co-workers [140] used the electrochemical co-deposition technique for the polypyrrole/graphene oxide. The electrode depicted an areal capacitance of 152 mFcm^{-2}.

Figure 15. Typical cyclic voltammogram (CV) curves of cellulose/polyaniline, cellulose/graphene oxide, and cellulose/graphene oxide/polyaniline electrodes at 50 mV/s. [128]. Reproduced with permission from MDPI.

In this regard, using the green synthesis approaches may overcome the challenges for forming eco-friendly polypyrrole/graphene nanocomposites having high specific capacitance and cycle stability. Consequently, Pourbeyram et al.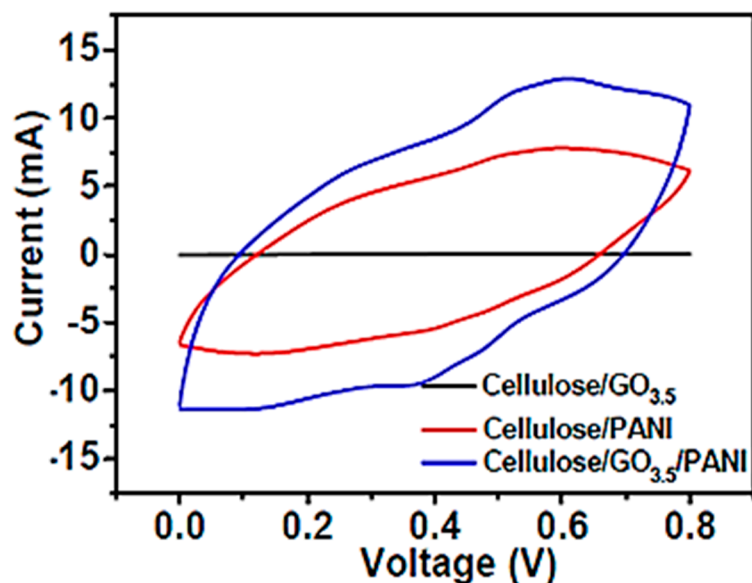 [141] synthesized the green polypyrrole/graphene nanocomposite nanofiber. The green method involved the direct interaction of polypyrrole and graphene without using the reducing/oxidative agents, surfactants, and organic solvent. The green method was proved low cost and ecological friendly. The electrode obtained using green route revealed high specific capacitance of 480 Fg^{-1} and capacitance retention of 94%, over 5000 charge-discharge cycles. Authors [141] applied green one-step electro-polymerization to deposit polypyrrole on the graphene paper. The supercapacitor electrode was developed using the eight deposition cycles. The green-synthesized supercapacitor electrode revealed an areal capacitance of 128.9 mF/cm^2. After 5000 galvanostatic charge–discharge cycles, the cyclic stability of 85% was attained. Moreover, the high energy density (16.1 mWh/cm^2) and power density (180 mW/cm^2) were observed for the electrode. Liu et al. [142] prepared the green graphene oxide in an aqueous solution. Then, graphene oxide was chemically reduced using ethylene glycol, which is an ecologically safe material. The polypyrrole/ethylene glycol-reduced graphene oxide exhibited a specific capacitance of 420 Fg^{-1}, which was considerably higher than the neat polymer electrode (159 Fg^{-1}). Moreover, a capacitance retention of 93% was observed. Zhang et al. [143] applied the one-pot green hydrothermal method to develop graphene hydrogel. The oxidative polymerization of polypyrrole was carried out on graphene hydrogel through a green route without using any toxic reagents. The green-synthesized polypyrrole/graphene hydrogel revealed a high specific capacitance of 375 Fg^{-1} and capacitance retention of 87% over 4000 charge–discharge cycles. Thus, the overall electrochemical performance of the supercapacitor electrodes was enhanced due to the double layer capacitance of green graphene and pseudo-capacitance of polypyrrole [144–146]. Contrarily, Xu et al. [147] used a non-green method for the formation of polypyrrol/reduced graphene

oxide nanocomposite electrode. The lower specific capacitance of 336 Fg^{-1} was attained. The use of green synthesis methods for polypyrrol/graphene nanocomposites has been found effective to enhance their capacitance properties due to better matrix–nanofiller interactions and interface formation.

Green polymers such as biodegradable poly(vinylpyrrolidone) (PVP) binder [148] and recyclable poly(ethyleneterephthalate) (PET) [149] have been preferred for the supercapacitor electrodes. In addition, a green screen-printing technique was applied for the deposition of graphene on these polymers to form the desired electrodes [150–152]. To avoid the leakage of liquid electrolyte, phosphoric acid-doped biodegradable PVP has also been used as a solid polymer electrolyte in the supercapacitors [153]. Consequently, fine interactions and interface formation were observed between the PVP/electrode and PVP electrolyte in the system [154–156]. Sudhakar et al. [157] adopted a green method to form reduced graphene oxide using calcium carbonate. Then, an ecologically safe screen-printing method was applied to form the supercapacitor electrodes using biodegradable PVP binder and recyclable PET. The PVP was also used as an electrolyte in the system. The low-cost environmentally friendly supercapacitor electrode exhibited a specific capacitance of 201 Fg^{-1} and capacitance retention of 97% over 2000 cycles. The performance of green electrodes was found comparable to the literature-reported electrodes prepared through non-green method [158,159]. The specific capacitance of a green-synthesized PVP binder-based electrode was found comparable to that of a non-green fabricated supercapacitor electrode (~200 Fg^{-1}) [160]. Table 1 shows specifications of important green synthesized nanocomposites.

Table 1. Specifications of green synthesized nanocomposites.

Polymer	Graphene	Green Method	Properties/Applications	Ref
Polyaniline	Graphene oxide-gold@polyaniline; reduced graphene oxide-gold@polyaniline	Cetraria Islandica L. Ach lichen-based method	Specific capacitance 212.8 Fg^{-1}	[122]
Polyacrylonitrile	Polyaniline@graphene	Green drop casting; screen-printing techniques	Electrical conductivity 2.362×10^{-6} Sm^{-1}	[124]
Cellulose/polyaniline	Graphene/graphene oxide	Green in-situ method	Electrical conductivity 1.15 Scm^{-1}; specific capacitance 1218 $mFcm^{-2}$	[128]
Polypyrrole	Graphene paper	Green one-step electro-polymerization	Areal capacitance 128.9 mF/cm^2; cyclic stability 85%	[141]
Polypyrrole	Reduced graphene oxide	Green reduction method using ethylene glycol	Specific capacitance 420 Fg^{-1}; capacitance retention 93%	[142]
Polypyrrole	Graphene hydrogel	one-pot green hydrothermal method; green oxidative polymerization without using toxic reagents	Specific capacitance 375 Fg^{-1}; capacitance retention 87%	[143]
Poly(vinylpyrrolidone); poly(ethyleneterephthalate)	Graphene	Biodegradable poly(vinylpyrrolidone); recyclable poly(ethyleneterephthalate); green screen-printing technique	Supercapacitors; electrode binder	[148]

Table 1. *Cont.*

Polymer	Graphene	Green Method	Properties/Applications	Ref
Poly(vinylpyrrolidone)	Reduced graphene oxide	Green method for reduced graphene oxide; ecologically safe screen-printing method	Specific capacitance 201 Fg^{-1}; capacitance retention 97%	[157]

6. Prospects and Summary

Green-synthesized graphene, graphene oxide, and reduced graphene oxide have been used as nanofillers for conjugated polymers like polyaniline, polypyrrole, and poly(vinylpyrrolidone) [161]. Various green routes have been adopted to form green graphene and modified graphene nanofillers. Using green reducing agents efficiently converted the graphene oxide to reduce graphene oxide. Environmentally harmless reducing agents have been used to prevent the toxic effects of chemicals like hydrazine. Moreover, green in-situ polymerization, electro-polymerization, printing, hydrothermal, and other methods have been used for developing polymer/graphene nanocomposite electrodes. The ecofriendly polymer/graphene nanocomposite-derived supercapacitor electrode revealed high electron transport, capacitance, durability, and overall enhanced electrochemical performance [162–164]. In addition to the green-synthesized graphene, various green polymers need to be applied for the fabrication of supercapacitor electrodes [165]. Moreover, precise control over the processing parameters may further enhance the supercapacitor performance. The morphology of green-synthesized graphene-based supercapacitor electrodes must be investigated in detail to gain insight of the nanostructure. Consequently, the formation of a homogeneous nanoporous structure may facilitate the high specific capacitance, energy density, and cyclic performance of green electrodes [166].

In this article, various strategies have been reviewed for the formation of green-synthesized graphene and green polymer/graphene nanocomposites. Low-cost, environmentally friendly, and flexible polymer/graphene nanocomposite-based supercapacitors have been designed. Using green-synthesized graphene, graphene oxide, and reduced graphene oxide in the supercapacitor electrodes revealed high specific capacitance, electrical conductivity, charge density, and cycling stability features. Specifically, the synergistic effects of green-synthesized graphene and conjugated polymers promoted the electron and charge transport properties in the supercapacitor electrodes. Future innovations in green design strategies may reveal exclusive structure, morphology, and high performance of polymer/graphene nanocomposites for supercapacitor electrodes, electrolytes, and other components.

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