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Review

Energy systems and green sourced nanomaterials—A today's outlook

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Abstract: Owing to current growing demands of environmental friendly energy devices, innumerable green materials/nanomaterials have been applied to design the desired high tech devices. Amongst energy devices, supercapacitors have been ranked distinctively for efficient energy storage competence. Principally, green nanocomposites derived from green or ecological polymers and green nanoparticles have been scrutinized for supercapacitor components. Concerning this, current review has been planned to sketch the energy storage application of green nanocomposites, predominantly for supercapacitors. In this concern, mostly synthetic green polymers (such as polyaniline, polypyrrole, etc.) and their blends with natural polymers (like chitosan) having fine biodegradability, non-toxicity, low cost, and superior device end performance have been found as the noteworthy materials. Additionally, green nanofillers like carbon nanoparticles (carbon nanotube, graphene, etc.) and metal nanoparticles have been processed with green polymers via ecological techniques, like in situ, solution, sonication, mixing, hydrothermal, exfoliation, reduction, etc., to form the anticipated energy device components. In consequence, the designed ecological nanocomposites expectedly had the advantages of low price/weight, superior mechanical/heat resilience, electron transference, capacitance, power/charge density, charge-discharge, sustainability as well as environmentally friendliness for energy related methodological systems. Incidentally, the design and performance challenges towards the application of ecological nanocomposites in energy storage devices have been conversed.

Keywords: green; polymer; nanofiller; nanocomposite; supercapacitor; environmental; high performance

1. Introduction

With the advancements in the materials and nanotechnological breakthroughs, green materials/nanomaterials having facile biodegradability and sustainability features have been the focus of recent technical researches [1,2]. For nanocomposites, essential environmental materials like natural or synthetic green polymers have been investigated as matrix materials [3,4]. Further, numerous nanoparticles of inorganic (metal, ceramics) and organic (carbon, polymers) types have been studied for advanced green nanocomposites [5]. In consequence, the nanocomposites designed from green matrices and nanofillers have been reported for valuable structural, mechanical, biodecomposibility, and sustainability features for progressive applications [6,7]. Besides, one important aspect of the green nanomaterials has been identified as the utilization of environmentally safe processing techniques to avoid any probable green house effects [8]. Ultimately, carefully designed green nanocomposites have been scrutinized for demanding industrial level employments, including energy sector, electronics, engineering, biomedical, etc. [9]. Use of environmentally safe nanomaterials have recently gained much curiosity in the energy/electronics fields [10]. In this concern, various combinations of eco polymers and eco nanoparticles have been focused for advanced energy devices and systems [11]. Furthermore, use of advanced green synthesis techniques has been engrossed to form the green nanomaterials for high tech energy applications [12]. Especially, in the field of energy storing devices, like supercapacitors, interesting field researches have been observed concentrating the use of green nanomaterials, instead of traditional non-ecological materials designs in practice [13,14].

Accordingly, disciplined research attempts have been observed for the synthesis of ecological or green materials or nanocomposites for energy storage devices [15]. For example, research reports have been detected on conducting polymers and graphene-based nanomaterials using green methods for supercapacitor electrodes having environmental friendliness, low price, superior surface area, capacitance, and charge-discharge properties [16]. Lots of attempts have been reported in recent years (2020–2024) on supercapacitor electrodes derived from conducting polymers (polyaniline, polypyrrole, polythiophene, and polythiophene) and carbon nanoparticles with sustainability and ecofriendliness for application in supercapacitor electrodes [17,18]. Çıplak et al. [19] formed ecological polyaniline/graphene/gold nanoparticles, by green in situ polymerization method, for supercapacitor electrode applications. Consequently, the environmentally friendly supercapacitor electrode showed reasonable specific capacitance of 212.8 Fg^{-1}. In recent years, Arthisree et al. [20] formed environmentally friendly high performance supercapacitor electrodes using polyacrylonitrile, polyaniline, and quantum dots. Consequently, the polyacrylonitrile/polyaniline/quantum dot nanocomposite showed high specific capacitance of up to 600 Fg^{-1}. More recently, Zhao et al. [21] used carboxyl substituted dipyridophenazine and MXene based green nanocomposites to design electrodes for efficient energy storing devices. The resulting nanomaterial had reasonably high reversible capacity of 172.6 mAh cm^{-3} in 4000 cyclic performances. The carboxyl substituted dipyridophenazine/MXene were suggested for ecofriendly flexible portable energy storing device applications. Hence, lots of recent researches have been seen on the green and sustainable nanocomposite electrodes for environmentally friendly charge storage devices [22].

Along these lines, this novel review has thrown light upon the existing designs and demanding necessities regarding the implementations of green nanocomposites for energy storage systems. In this concern, essential green polymers and nanofiller based designs have been argued here. Effectiveness of green nanomaterials to attain low weight, cheap, robust, and sustainable green supercapacitor components, having high specific capacitance, power/charge density, recyclability, etc., have been surveyed for commercial energy devices applications. It seems that future of energy storage systems, like supercapacitors, relies upon more precise research endeavors to identify high-tech green/sustainable designs having least processing and performance related challenges.

2. Concept of green polymers, nanofillers and nanocomposites

Recognition of natural or green polymers can be related to centuries back with the wood, cotton, and natural rubber like materials [23,24]. Later, natural green polymers like cellulose, chitosan, starch, nylons, etc. and synthetic green polymers like poly (vinyl alcohol), poly (ethylene glycol), poly (lactic acid), etc., have been foreseen [25,26]. Green polymers usually own biodegradability, sustainability, and environmentally friendly features [27,28]. Moreover, synthetic green polymers must be processed via ecological routes to maintain the environmental demands [29]. Further, green polymers have been observed practically beneficial for a number of industrial applications due to their increasing demand for engineering structures, adhesive/coating, and biomedical industries [30–32]. Besides, for reinforcing green polymeric matrices, numerous ecologically beneficial green nanofillers have been adopted including the polymeric nanoparticles, metal nanoparticles, carbon nanoparticles, and inorganic nanoparticles like nanoclays [33–35]. **Figure 1** shows few examples of essential ecological or green nanoparticles and also the green polymers used for environmentally friendly nanocomposites.

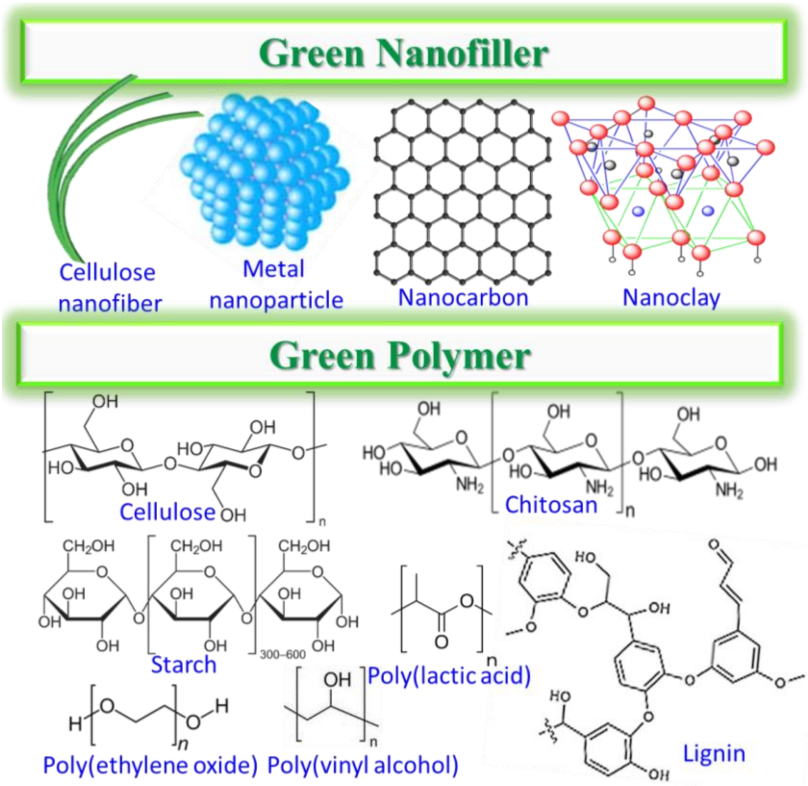

Figure 1. Few green polymers and green nanofillers.

For green nanocomposites, starch has been adopted as a low-cost natural polymer having facile biodegradation and environmentally friendly nature [36,37]. For example, Cheviron et al. [38] formed green nanocomposites based on starch and silver nanoparticles and explored for antimicrobial packaging applications. Similarly, the nanocomposites of lignin, cellulose, and allied natural green polymers have been reported [39,40].

Chitosan is an important green polymer used as matrix material for nanocomposites [41]. Regarding green metal nanoparticle-based system, Hashem et al. [42] prepared the chitosan/gold nanoparticle hybrid using a green chemical reduction method. **Figure 2A** shows the transmission electron microscopy image of the chitosan/gold nanoparticle hybrid having gold nanoparticles of sizes in the range of

20–120 nm in the matrix. **Figure 2B** depicts the particle size distribution of the chitosan/gold nanoparticle hybrid with average nanoparticle sizes of ~ 200 nm, owing to the insertion in the polymeric matrix and coating with the polymer. In addition, **Figure 2C** shows the X-ray diffraction patterns of the chitosan/gold nanoparticle hybrid with characteristic diffraction peaks for chitosan (22.8°) and gold nanoparticle peaks at 37.9° and 44.1°–77.4° due to crystallinity of the nanocomposite sample. Carlo et al. [43] also loaded gold nanoparticles in chitosan matrix using a green solution route based on $HAuCl_4$ salt and caffeic acid. The chitosan/gold nanoparticle nanocomposites had fine biodegradability and biocompatibility features.

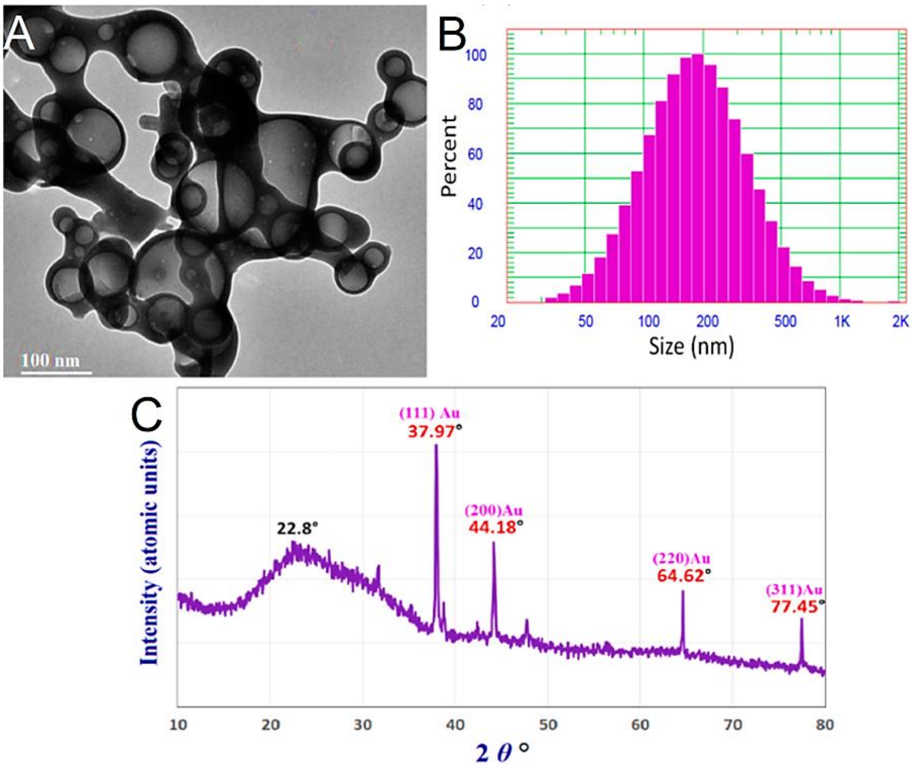

Figure 2. (A) Transmission electron microscopy image of the chitosan/gold nanoparticle hybrid; **(B)** particle size distribution of the chitosan/gold nanoparticle hybrid; **(C)** X-ray diffraction patterns of the chitosan/gold nanoparticle hybrid [42]. Reproduced with permission from MDPI.

Poly (ethylene glycol) is a synthetic green polymer having water solubility, biodegradability, and non-toxicity properties and it is widely applied as a matrix for nanofillers [44]. Including metal nanoparticles or carbon nanofillers in poly (ethylene glycol) matrix has been found to enhance the heat stability and mechanical properties of the resulting nanomaterials [45]. For example, Nguyen et al. [46] applied green plasma assisted chemical reduction approach for the formation of the poly (ethylene glycol) functional gold nanoparticle nanocomposite. **Figure 3A** shows a simple synthetic process for plasma assisted formation of the poly (ethylene glycol) functional gold nanoparticle nanocomposite. The as prepared poly (ethylene glycol) functional gold nanoparticle sample was scanned for the scanning electron microscopy images of different resolutions, as given in **Figures 3B,C**. According to the micrographs, gold nanoparticles had fine dispersions in the polymer and own uniform quasi spherical shapes.

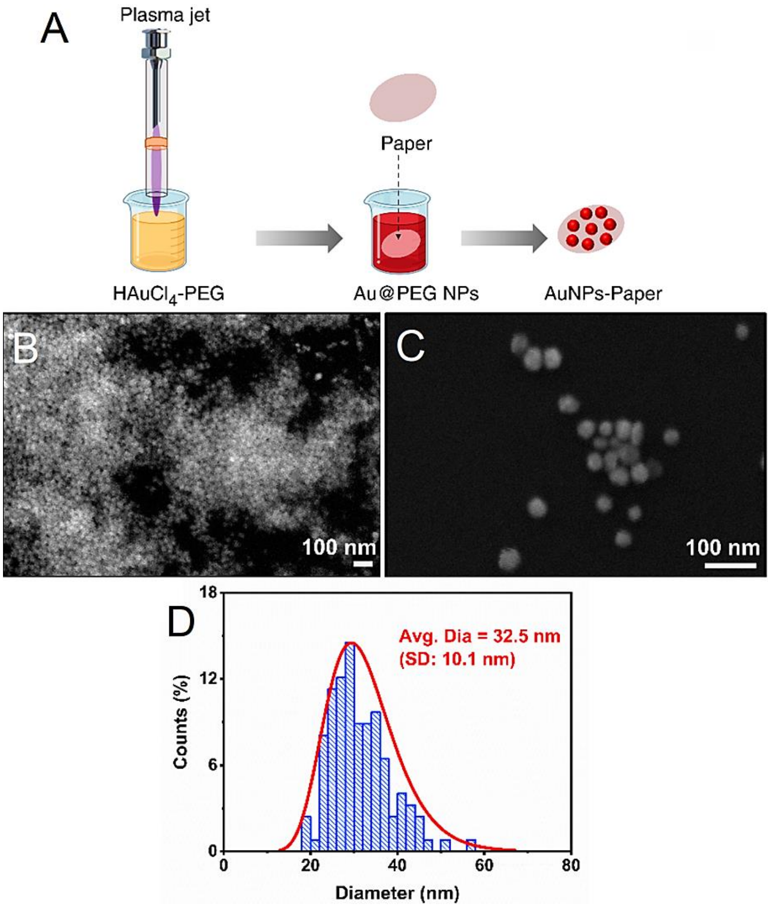

Figure 3. (A) Process for the plasma assisted synthesis of poly (ethylene glycol) functional gold nanoparticles (Au@PEG NPs); scanning electron microscopy images of Au@PEG NPs: **(B)** at 60,000 × resolution; and **(C)** at 160,000 × resolution; **(D)** the particle size distribution of Au@PEG NPs, with average particle size of 32.5 nm (standard deviation, SD, of 0.1 nm [46].
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Figure 3D illustrates the particle size distribution plots of the poly (ethylene glycol) functional gold nanoparticles obtained by Image J software showing a uniform symmetrical gold nanoparticle distribution and average size of around 32.5 nm. These results indicated the success of the green plasma-based technique applied to form the green poly (ethylene glycol)/gold nanoparticle hybrids. Similarly, countless reports have been observed on the poly (ethylene glycol) hybrids, e.g., the poly (ethylene glycol)/halloysite nanotubes [47], poly (ethylene glycol)/turmeric nanofibers [48], poly (ethylene oxide)/silver nanoparticles [49], poly (ethylene oxide)/silica nanoparticles [50] for applications in devices, packaging, antimicrobials, and biomedical fields [51].

Among other metal (gold/silver nanoparticles) or inorganic nanoparticles (nanoclays) filled green hybrids, poly (vinyl alcohol) [52–54] and poly (lactic acid) [55–57] based nanomaterials have been reported for low cost, fine biodegradability, non-lethality, and environmentally friendliness for wide ranging applications as antibacterial, wound healing, biomedical, and technical relevance.

Besides, various nanocarbons including carbon nanotube, graphene, graphene oxide, and other modified graphene forms have been applied as beneficial nanofillers for the formation of the green polymers and nanocomposites.

3. Green nanocomposites for energy systems

Concept of using green or ecological materials can be related to the ancient development of wind turbine blades using natural composites, instead of heavy metal-based structures [58]. Further research efforts on ecological composites exposed uses in sophisticated devices, like optoelectronics [59]. Similarly, numerous environmental materials, e.g., natural polymer, have been recognized with the potential for energy conversion systems, like batteries [60]. Consequently, natural/green polymers and related materials have been explored for light emitting diodes [61], solar cell devices [62], thermoelectric generators [63], and related thermoelectric devices [64]. In these systems, green polymers have been suggested to develop donor-acceptor type associations for energy conversion purposes [65]. Incidentally, the optical, electron conduction, and energy conversion features of these ecological materials depicted their demand in today’s high-tech energy conversion systems, like supercapacitors.

Among efficient energy conversion systems, supercapacitors have been enormously focused by the researchers due to high energy outputs [66]. Advancements in the field of supercapacitors have devised the use of effective green materials, such as polymers, composites, and nanocomposites [67]. Consequently, green or ecological polymers, natural fillers, green or green derived nanoparticles have been investigated to form the ecological composites and nanocomposite designs for energy storage devices [68]. In this concern, ecofriendly/green synthesis methods have also been studied for the formation of environmentally safe materials or nanomaterials [69].

Out of huge variety of polymers, naturally occurring and synthetic polymers having ecofriendly, biodegradability, and sustainability properties have been focused for green energy applications [70]. Few examples may include cellulose, chitosan, chitin, lignin, starch, and countless other eco polymers [71]. Particularly, for supercapacitor electrodes, natural/green polymers offer the advantages of facile processing and superior performance, in addition to sustainability aspects [72]. Despite of using green polymers, several non-green materials have been processed using ecological routes to attain environmentally safe materials for supercapacitors or energy devices [73].

For superior supercapacitor device performance, green polymers have been converted to green nanocomposite by adopting several inorganic (metal nanoparticles or metal oxide nanoparticles) and carbon nanomaterials [74,75]. Amidst carbon nanoparticles, carbon nanotube, graphene, and modified carbon nanotube or graphene nanostructures have been investigated [76]. Additionally, use of green methods with safe chemical and reagents have been practiced to form the desired ecological nanocomposite for energy storage [77].

4. Green energy devices—Supercapacitors

For energy devices, carbon nanoparticles have been processed with polymers or inorganic matrices via environmentally safe chemicals and methods to attain the desired ecological device materials [76]. In this way, use of hazardous organic solvents and reagents can be avoided to form green nanocomposites [78]. Subsequently, researchers are continuously struggling to fabricated advanced supercapacitor devices with high capacitance, power density, and cyclic performance using green polymers, green nanocarbons, and green synthesis routes [79].

Carbon nanotube based green nanocomposites have been reported for supercapacitor assemblies [80]. Here, Jyothibasu et al. [81] used modest low cost in situ chemical polymerization, sonication, and filtration routes to form green polypyrrole/kapok fiber/carbon nanotube nanocomposite. **Figure 4A** shows the cyclic voltametric curves of the polypyrrole/kapok fiber/carbon nanotube nanocomposite, which was bent at several angles of 0° to 180° (40 mV s^{-1}). The obtained nanocomposite based flexible supercapacitor is also shown as an inset with digital photograph in bent form. The nanocomposite-based supercapacitor electrode had symmetric cyclic performance in cyclic voltametric curves. **Figure 4B** displays electrochemical impedance spectroscopy spectra of the polypyrrole/kapok fiber/carbon nanotube nanocomposite based flexible supercapacitor at various bending angles. The results showed an increase in the areal capacitance from 128.4 mF cm^{-2} (0°) to 139.4 mF cm^{-2} (180°). Ultimately, the polypyrrole/kapok fiber/carbon nanotube nanocomposites connected in series were used to light the red-light emitting diode, as given in **Figure 4C**. Moreover, the polypyrrole/kapok fiber/carbon nanotube nanocomposite based flexible solid state symmetric supercapacitor had constant cycling stability over 2500 cycles (25 mA cm^{-2}) and capacitance retention of 97.4% (**Figure 4D**). Hence, low price, green and sustainable high performance flexible polypyrrole/kapok fiber/carbon nanotube nanocomposite-based supercapacitor has been developed.

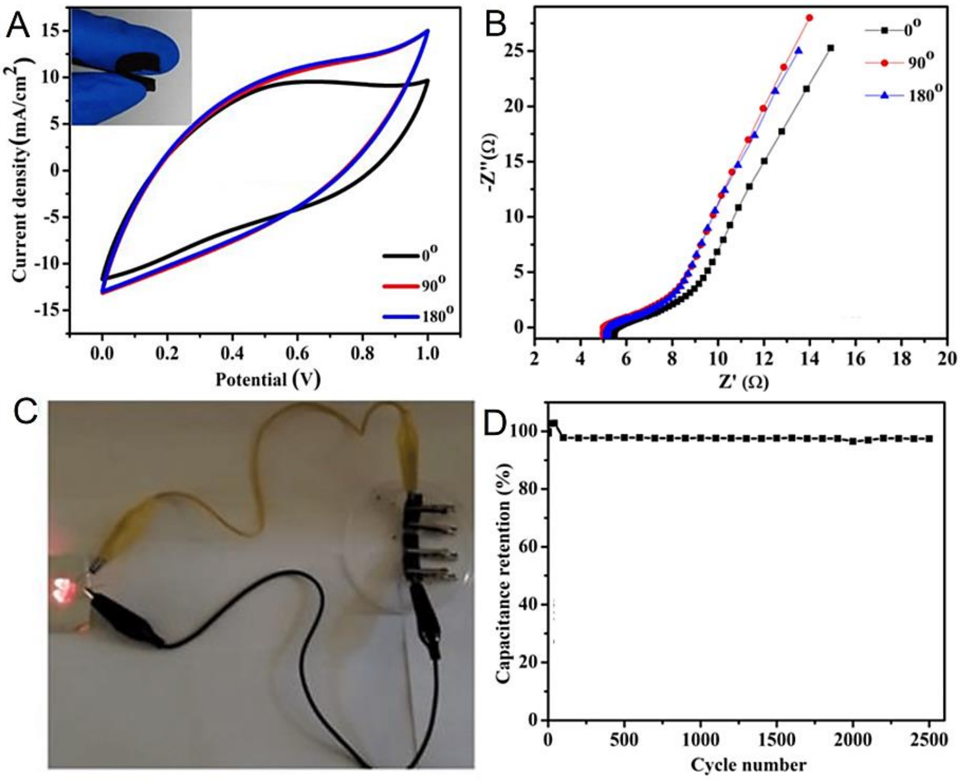

Figure 4. (A) Cyclic voltametric curves of the polypyrrole/kapok fiber/carbon nanotube nanocomposite bent at various angles (40 mV s^{-1}), inset is digital photograph of flexible supercapacitor in a bending state; **(B)** electrochemical impedance spectroscopy spectra of the polypyrrole/kapok fiber/carbon nanotube nanocomposite based flexible supercapacitor at various bending angles; **(C)** digital photograph of red light emitting diode powered by three polypyrrole/kapok fiber/carbon nanotube nanocomposites based supercapacitors connected in series; **(D)** cycling stability (2500 cycles, 25 mA cm^{-2}) of the assembled polypyrrole/kapok fiber/carbon nanotube nanocomposite based flexible all-solid-state symmetric supercapacitor [81].

Recently, graphene has been a hot focus of research for technical applications, due to its inherent nanocarbon nanostructure [82]. Predominantly, nanocomposite forms of graphene have been reported efficient for energy conversion device applications [83]. Here, adding minute graphene contents have shown remarkable increase in the physical properties of the nanocomposites [84]. For supercapacitors, graphene or modified graphene has been combined with inorganic metal or metal oxide nanoparticles to form the supercapacitor electrodes [85]. For example, Ezeigwe et al. [86] fabricated a supercapacitor electrode based on the graphene and zinc oxide-based nanomaterial. They used green liquid phase exfoliation and solvothermal_techniques to synthesize the graphene/zinc oxide hybrid. In these methods water and ethanol solvents were used as environmentally safe reagents. The resulting graphene/zinc oxide hybrid had reasonable specific capacitance of about 236 Fg^{-1} for supercapacitor electrode application. Additionally, non-conjugated polymers, like poly (vinyl alcohol) and poly (ethylene glycol) have been used for supercapacitor electrodes [87]. In this regard, Nayak et al. [88] used poly (vinyl alcohol) as a green polymer with graphene for supercapacitor electrodes. Consequently, they designed the poly (vinyl alcohol)/graphene/WO_3 hybrid as a green electrode nanomaterial for solid state asymmetric supercapacitor application. This device had energy density of 6–25 W h kg^{-1} over 4000 charge-discharge cyclic recital. The solid-state asymmetric supercapacitor was capable of lighting the red-light emitting diode, as shown in **Figure 5**.

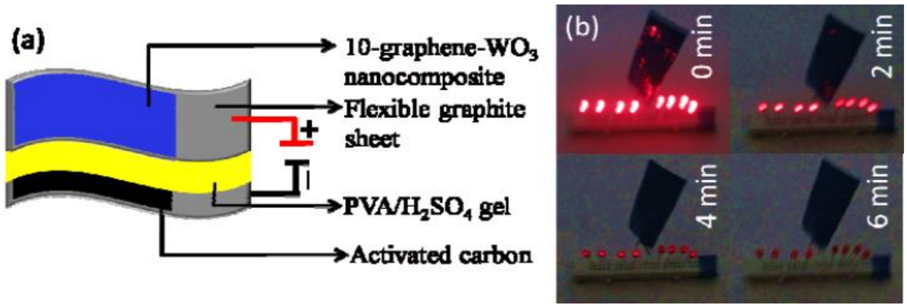

Figure 5. (a) Diagram of a flexible solid state asymmetric supercapacitor device set-up of poly (vinyl alcohol)/graphene/WO_3 hybrid; **(b)** picture of real asymmetric supercapacitor device of poly (vinyl alcohol)/graphene/WO_3 hybrid applied to light red light emitting diodes after charging [88].

Majority of the research attempts on polymer/nanocarbon nanomaterials for supercapacitor electrodes have reported the use of conjugated polymers [89]. According, conducting polymer/graphene nanocomposites have been prepared having superior electrochemical performance for supercapacitors [90]. For this purpose, the conducting polymer/graphene nanocomposites have been fabricated through facile green methods [91]. Consequently, conducting polymer/graphene nanocomposite-based electrodes were found to have low cost, durability, superior surface area, specific capacitance, energy/power density, and cyclic performance [92]. A number of conductive polymers have been used with graphene to form supercapacitor electrodes, namely polyindole, polyaniline, polypyrrole, polythiophene, and their derived forms [93]. Ramesh et al. [94] used a green hydrothermal route for the formation of the polyindole/cobalt (II, III)/nitrogen doped graphene oxide hybrids aiming for ecological supercapacitor electrodes. **Figure 6A** shows the process for the formation of cobalt (II, III)/nitrogen doped graphene oxide and polyindole/cobalt (II, III)/nitrogen doped graphene oxide hybrids involving oxidative polymerization and hydrothermal routes. **Figures 6B,C** shows the scanning electron microscopy images of polyindole/cobalt (II, III)/nitrogen doped graphene oxide. A homogeneously dispersed microstructure with cobalt oxides nanoparticles of about 10–20 nm were seemed to be distributed on the hexagonal nanosheets of nitrogen doped graphene oxide. **Figure 6D** depicts the specific capacitance vs. current density of the polyindole/cobalt (II, III)/nitrogen doped graphene oxide hybrid. Here, highest specific capacitance of around 680 F g^{-1} was attained at the current density of 0.5 Ag^{-1} in 5000 cyclic durations. Hence, the green synthesized polyindole/cobalt (II, III)/nitrogen doped graphene oxide hybrids had fine potential for supercapacitor electrode utilization.

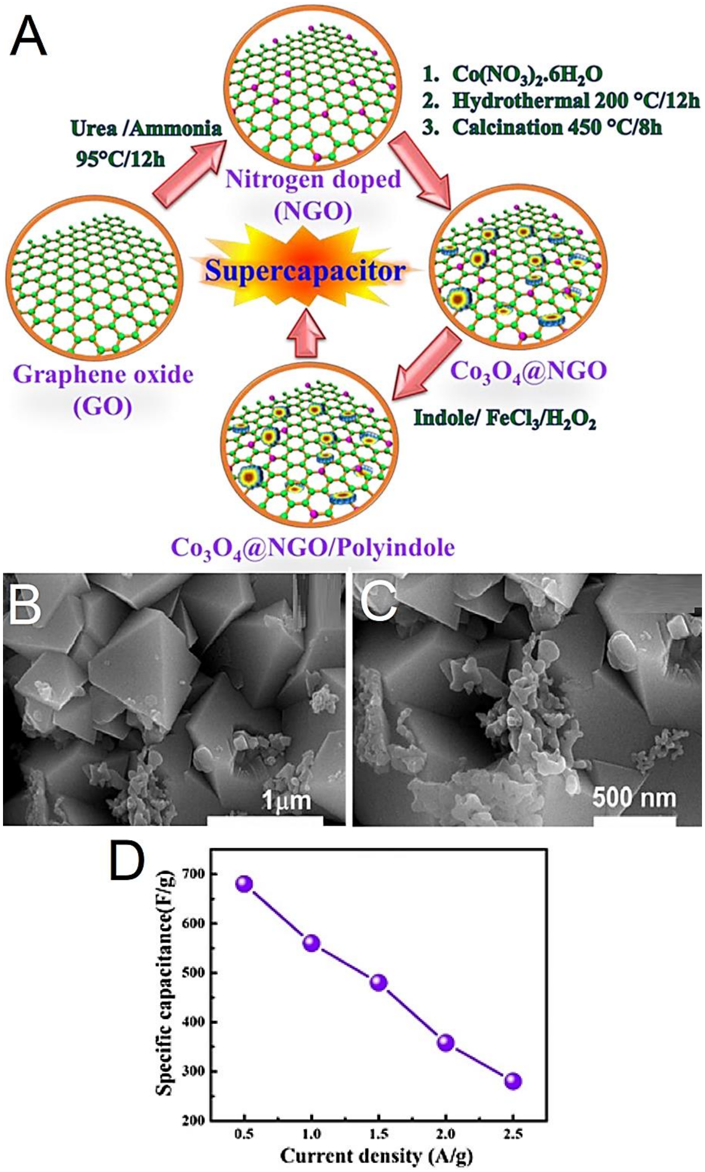

Figure 6. (A) Schematic for the formation of cobalt (II, III)/nitrogen doped graphene oxide (Co_3O_4@NGO) and polyindole/cobalt (II, III)/nitrogen doped graphene oxide (PIN/Co_3O_4@NGO) hybrid; **(B** and **C)** scanning electron microscopy images of PIN/Co_3O_4@NGO hybrid; **(D)** specific capacitance vs. current density of the PIN/Co_3O_4@NGO hybrid [94].

Gul et al. [95] devised green nanocomposites of the polyaniline/graphene oxide co-doped dodecyl benzene sulfonic acid hybrid and the polyaniline/graphene oxide co-doped camphor sulfonic acid hybrid via in situ and doping processes for the supercapacitor electrodes. **Figure 7A** shows the scanning electron microscopy micrographs of the polyaniline/graphene oxide co-doped dodecyl benzene sulfonic acid hybrid with fine nanoparticle dispersion and porous morphology. **Figure 7B** displays the scanning electron microscopy micrographs of the polyaniline/graphene oxide co-doped camphor sulfonic acid hybrid having similar nanoparticle distribution and porous morphology.

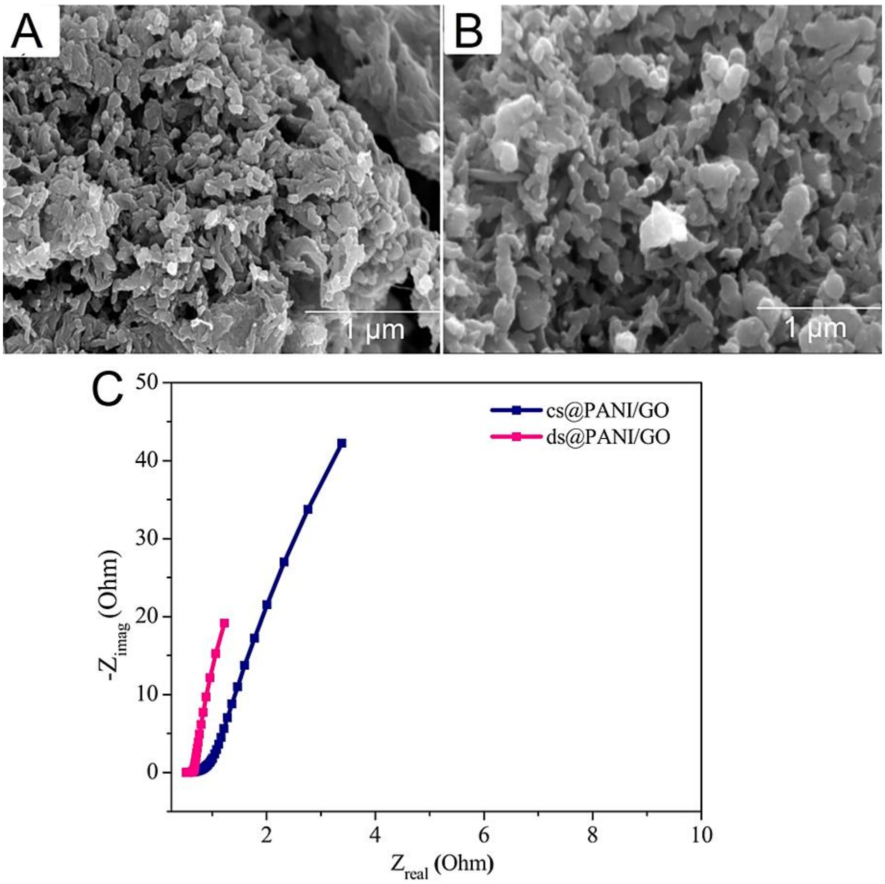

Figure 7. Scanning electron microscopy micrographs of **(A)** polyaniline/graphene oxide co-doped dodecyl benzene sulfonic acid (ds@PANI/GO) hybrid; **(B)** polyaniline/graphene oxide co-doped camphor sulfonic acid (cs@PANI) hybrid; **(C)** Nyquist plots of ds@PANI/GO and cs@PANI/GO hybrids [95].
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Figure 7C expresses the Nyquist plots of the hybrids with a compressed semicircle behavior in high frequency region by both the polyaniline/graphene oxide co-doped dodecyl benzene sulfonic acid and polyaniline/graphene oxide co-doped camphor sulfonic acid hybrids showing low resistance electrical conductivity properties, Consequently, the green supercapacitor device had high power density of > 1700 Wkg^{-1}, specific capacitance of 97–150 Fg^{-1}, and capacitance retention of 93–97 %. Li et al. [96] fabricated the cellulose/polyaniline blend and cellulose/polyaniline/graphene oxide hybrid via facile green in situ polymerization, as shows in **Figure 8a**.

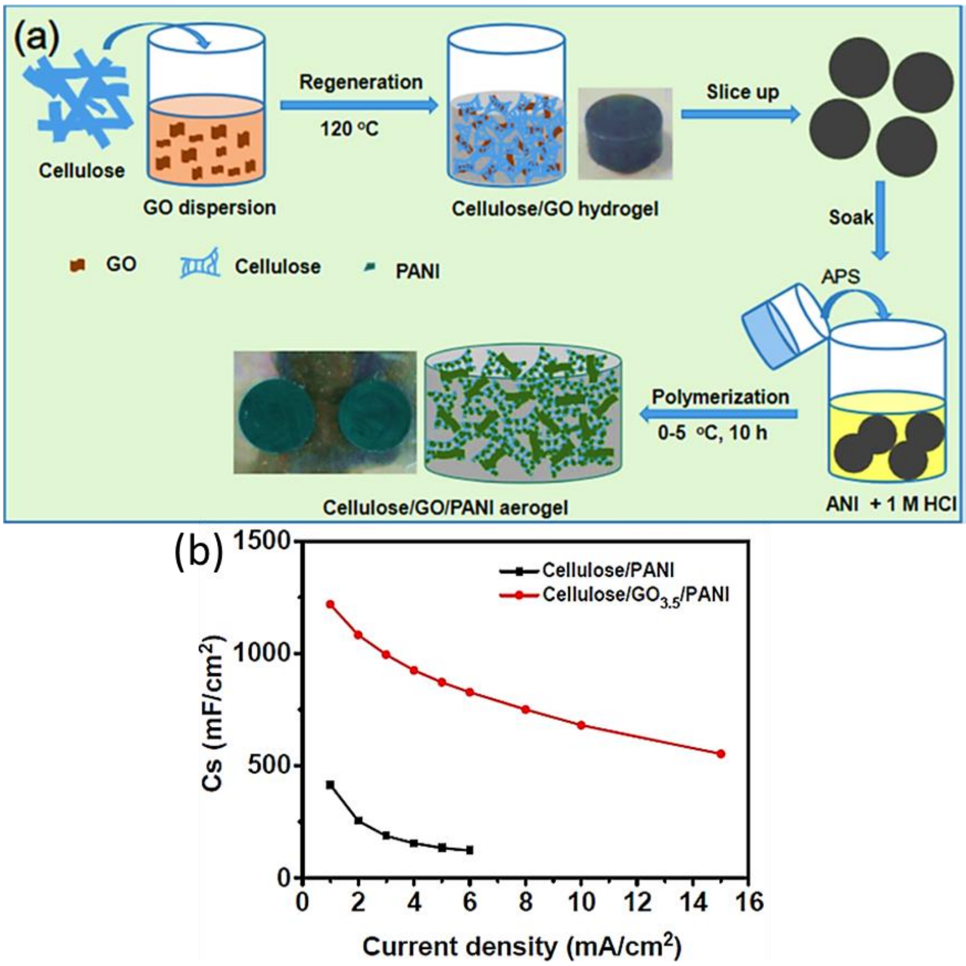

Figure 8. (a) Synthetic route for the cellulose/polyaniline/graphene oxide; **(b)** plots of areal specific capacitance vs. current for the cellulose/polyaniline and cellulose/polyaniline/graphene oxide [96].

GO = graphene oxide; PANI = polyaniline; ANI = aniline Reproduced with permission from MDPI.

Figure 8b presents the areal specific capacitance of the cellulose/polyaniline blend and cellulose/polyaniline/graphene oxide hybrid. As compared to the blend sample, the cellulose/polyaniline/graphene oxide hybrid with 5 wt.% nanofiller had superior areal specific capacitance, power density, and electrical conductivity of around 1218 $mFcm^{-2}$, 1201 μWcm^{-2}, and 1.15 Scm^{-1}, respectively. Thus, the green cellulose/polyaniline/graphene oxide hybrid revealed high performance for practical ecological supercapacitor applications.

Henceforth, several successful design combinations have been proposed for green energy storage devices, like supercapacitors, employing suitable green materials and methods [97–99]. In this concern, environmentally friendly supercapacitor electrodes based on conjugated polymers, such as polyaniline, polypyrrole, etc., and green polymers like cellulose, chitosan, etc., have been developed [100–102]. Moreover, carbon nanotube and graphene have been used as green nanocarbons for the ecological supercapacitor electrodes [103,104]. Moreover, facile green processing techniques, such as in situ method, sonication, solution mixing, hydrothermal method, etc., have been adopted to form the ecological polymer/nanocarbon nanomaterials for energy maneuvers [105]. Consequently, the ensuing supercapacitor electrodes had fine electron conduction and charge transport features for high end green energy devices [106–108]. In this way, superior surface area, electrochemical features, electrical conductivity, specific capacitance, and power/charge density have been attained for the ecological supercapacitor devices [109].

5. Perspectives and encounters

Conventionally, industrial level energy systems have been designed using expensive materials and high-priced sophisticated techniques to attain desired high energy outputs. For example, commercially employed supercapacitor devices usually comprise of high cost electrodes and catalysts prepared via non-environmental techniques. Moreover, commercial supercapacitors lack sustainability and biodegradability properties, in turn causing green house effects. In this concern, recent research has been continuously turned toward the development of low cost, sustainable, biodegradable, non-toxic, and environmentally friendly materials for supercapacitor assemblies. Use of green techniques may further enhance the potential of green nanomaterials towards supercapacitors. Additionally, green materials derived supercapacitors must have the advantages of superior mechanical/thermal stability, electron conduction, specific capacitance, capacitance retention, energy/power density, charge-discharge, durability over repeated cyclic life, and other desirable electrochemical features. For this purpose, choice of green polymer, green nanofiller, as well as green processing route have found indispensable to fabricate high-tech energy device materials. **Table 1** presents essential comparison of important design, properties and application of various past and present green nanomaterials used for supercapacitors.

According to the literature presented in above sections, it is important to highlight the specific performance metrics (such as specific capacitance, capacitance retention, energy density, power density, charge-discharge, etc.) of green nanocomposites needed for high performance supercapacitor devices. Specifically, Pawar et al. [110] recently used a green chemical bath deposition method for the formation of polyaniline and reduced graphene oxide derived nanocomposite for supercapacitor electrodes. The subsequent polyaniline/reduced graphene oxide nanocomposites showed considerable high specific capacitance and capacitance retention of > 1130 Fg^{-1} and > 80 %, respectively, in 5000 charge-discharge cyclic route. In addition, polyaniline/reduced graphene oxide had superior power density and energy density around 732 W kg^{-1} and 23 Wh kg^{-1}, respectively. Thus, supercapacitor performance matrices were analyzed via specific capacitance, capacitance retention, charge-discharge, energy density, and power density features. Very recently, Mupit et al. [111] used green chemical exfoliation and in situ methods for the formation of environmental friendly polyaniline/graphene nanocomposites. These supercapacitor electrodes had high specific capacitance of > 300 F g^{-1}. Also, in a recent attempt by Tale et al. [112], nanocarbons and manganese dioxide based green nanomaterials were prepared for supercapacitor electrodes. The ensuing electrodes showed superior specific capacitance and capacitance retention of about 1900 Fg^{-1} and 98 %, relatively, over 6000 charge/discharge cycles.

Presently used environmentally friendly methods for green nanomaterials include in situ polymerization, solution mixing, and similar simple techniques [113,114]. These lab-scale synthesis practices for green nanomaterials have advantages of inexpensiveness, non-toxic fabrication, and easy processing. However, large scale or industrial level production of green nanomaterials using solution or in situ methods face countless challenges [115]. Most importantly, these methods may result in poor material compatibility, dispersion, and homogeneity issues, so hindering the scalability, high performance, and applicability of these nanomaterials on commercial level [116]. Here, it is suggested to use advanced sophisticated techniques like spinning, coating, three dimensional printing, etc., to overcome the potential barriers towards scalability or commercial adoption of green nanocomposites. Use of advanced processing techniques for green nanomaterial ensure well-controlled and optimized conditions for industrial scale production of energy systems [117].

In real world, field researchers are constantly striving to integrate green nanohybrids into supercapacitor electrodes (especially as anodes) to gain superior charge storage, cyclic rate, and performance endurance [118]. For practical industrial applications, recent designs of green derived supercapacitor components had sustainability, long life, and potential to store larger amount of electricity for rapid charging, compared with out-of-date traditional commercial supercapacitors [119]. Continuing research on conducting polymers based green or environmentally friendly nanomaterials showed important real life uses in micro-supercapacitors. Presently, these green micro-supercapacitors have been found commercially beneficial for applications in modern smartphones, portable computers or laptops, electric vehicles, and other sustainable energy/electronic devices [120,121]. It is important to mention that restricted attempts have been reported to deal with the current market trends of green nanomaterials based energy systems so far [122]. Nevertheless, the available literature up till now specified future potential estimates of green nanomaterials for forthcoming breakthroughs of their possible marketplace [123].

Table 1. Comparison of past and present green nanomaterials for supercapacitors.

Matrix	Nanoadditive	Green method	Property/Application	Ref.
Polyaniline/graphene oxide/gold nanoparticles and polyaniline/	Reduced graphene oxide/gold nanoparticles	Green in situ synthesis; Cetraria Islandica L. Ach lichen	Supercapacitor electrode has scan rate of 5–200 mV s^{-1}; specific capacitance and capacitance retention 212.8 F g^{-1} and 86.9%, correspondingly	[19]
Polyacrylonitrile/polyaniline/	Graphene	Drop casting; screen printing	Electron conductivity 2.362×10^{-6} S m^{-1}	[20]
Cellulose/polyaniline	Graphene oxide	In situ technique	Electron conductivity and areal specific capacitance of 1.15 S cm^{-1} and 1218 mF cm^{-2}, respectively; energy density of 1201 μW/cm^2	[96]
Polyaniline	Reduced graphene oxide	Chemical bath deposition	Supercapacitor electrodes; specific capacitance 1130 F g^{-1}; capacitance retention 82 %; 5000 cycles	[110]

Table 1. (*Continued*).

Matrix	Nanoadditive	Green method	Property/Application	Ref.
Polyaniline	Graphene oxide	Chemical exfoliation; in situ technique	Supercapacitor electrode; specific capacitance > 300 F g^{-1}	[111]
Polyaniline	Graphene; graphene oxide; manganese dioxide	In situ method	Supercapacitor electrode; specific capacitance > 1882 F g^{-1}; capacitance retention ~ 98 %; 6063 cycles	[112]
Polyaniline	Reduced graphene oxide; amine functional graphene	In situ process	Electrical conductivity 8.66 S cm^{-1}; specific capacitance 250 F g^{-1}	[124]
Polyaniline	Sulfonated graphene	In situ polymerization	Specific capacitance 478 F g^{-1}	[125]
Polyaniline	Aniline-functional-graphene	In situ method	Supercapacitor electrodes; specific capacitance 100–440 F g^{-1}; capacitance retention 73–80%	[126]
Bacterial cellulose	Carbonization	Solution process	Supercapacitor electrodes; specific capacitance and capacitance retention 216 F g^{-1} and 97.6 %, respectively	[127]
Bacterial cellulose	Carbon nanotube	Solution process	Supercapacitor electrodes; specific capacitance and capacitance retention 50.5 F g^{-1} and 99.5%, respectively	[128]
Bacterial cellulose	Polypyrrole	In situ Method	Supercapacitor electrodes; specific capacitance and capacitance retention153 F g^{-1} and 93.0 %, respectively	[129]
Bacterial cellulose	Polypyrrole; Carbon nanotube	In situ Method	Supercapacitor electrodes; specific capacitance and capacitance retention 228 F g^{-1} and 88.0%, respectively	[130]
Bacterial cellulose	Polypyrrole; Graphene	In situ Method	Supercapacitor electrodes; specific capacitance and capacitance retention 4.16 F cm^{-2} and 91.5%, respectively	[131]
Bacterial cellulose	Polypyrrole; reduced graphene oxide	In situ Method	Supercapacitor electrodes; specific capacitance and capacitance retention 3.66 F cm^{-2} and 73.5%, respectively	[132]
Bacterial cellulose	Reduced graphene oxide	Solution process	Supercapacitor electrodes; specific capacitance and capacitance retention 2106–2544 mF cm^{-2} and 100%, respectively	[133]
Poly (vinylpyrrolidone)/recycled poly (ethyelene terephthalate)	Graphene	Screen printing technique	Specific capacitance and capacitance retention of 201 F g^{-1} and 97%, respectively	[134]
Polyaniline	Olive stones functional reduced graphene oxide	In situ self assembly	Specific capacitance 582.6 F g^{-1}; capacity retention 97 %; 3000 cycles	[135]

For future high performance and applicability of green nanomaterials in advance energy storage systems, it is suggested to define precise research directions by intensive repeated research efforts on the synthesis and analysis of these materials. In this regard, wide-ranging experimental as well as theoretical studies need to be performed on nontoxic, ecofriendly, sustainable, and biocompatible nanocomposites with predefined property improvement phenomenon for progresses in this field. According to literature analysis, it can also be suggested that future of green nanocomposites relies on scalability and large scale production of these materials for real world applications by considering the necessary environmental and economic concerns. Moreover, future research on green nanomaterials have been found essential to reveal further utilizations towards the green/sustainable microelectronics, solar cells, light emitting diodes, and related high end devices.

6. Conclusions

In brief, this review manuscript argues ecologically essential polymers, nanofillers, and nanocomposites viable for applications in significant energy devices, particularly the supercapacitor components. For this application, conducting and synthetic green polymers have been filled with carbon nanoparticles like carbon nanotube and graphene. In this regard, ecological and low cost method have been applied to attain high performance environmental friendly energy materials for supercapacitor related systems. Typically, in situ polymerization, electrochemical polymerization, solution processing, and similar facile synthesis techniques have been used for the synthesis of green nanomaterials for energy devices. Mixing of polymers with nanomaterials through green methods considerably improved the processability, structural stability, ecofriendliness, and life-long performance of the resulting components for energy devices. In this regard, electrical conductivity, mechanical stability, heat conduction, capacitance, and cyclic performances of green nanomaterials have been investigated. Potential of green nanomaterials for energy devices points towards essential future industrial level utilizations, nevertheless continuous research efforts must be performed to introduce new design varieties and to overcome the fabrication and performance hindering challenges. After considering the literature surveys presented in the above sections of this novel review, it can be suggested that future research on new design combinations, property/performance investigation, and minimization of challenges for green nanocomposites can lead to promising commercial level energy devices. In this way, advanced potentials of high performance green nanomaterials based energy storing systems can be explored for utilizations in modern electronics, space/defence structures, and biomedical equipment.

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