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REVIEW ARTICLE

Graphene and nanocomposites—Imprints on environmentally sustainable production and applications based on ecological aspects

Ayesha Kausar[1,2,*], Ishaq Ahmad[1,2]

[1] *NPU-NCP Joint International Research Center on Advanced Nanomaterials and Defects Engineering, Northwestern Polytechnical University, Xi'an 710072, Shaanxi Province, China*

[2] *UNESCO-UNISA Africa Chair in Nanosciences/Nanotechnology, iThemba LABS, Somerset West 7129, South Africa*

* **Corresponding author:** Ayesha Kausar, dr.ayeshakausar@yahoo.com

ABSTRACT

Graphene, an innovative nanocarbon, has been discovered as a significant technological material. Increasing utilization of graphene has moved research towards the development of sustainable green techniques to synthesize graphene and related nanomaterials. This review article is basically designed to highlight the significant sustainability aspects of graphene. Consequently, the sustainability vision is presented for graphene and graphene nanocomposites. Environmentally sustainable production of graphene and ensuing nanomaterials has been studied. The formation of graphene, graphene oxide, reduced graphene oxide, and other derivatives has been synthesized using ecological carbon and green sources, green solvents, non-toxic reagents, and green routes. Furthermore, the utilization of graphene for the conversion of industrial polymers to sustainable recycled polymers has been studied. In addition, the recycled polymers have also been used to form graphene as a sustainable method. The implication of graphene in the sustainable energy systems has been investigated. Specifically, high specific capacitance and capacitance retention were observed for graphene-based supercapacitor systems. Subsequently, graphene may act as a multi-functional, high performance, green nanomaterial with low weight, low price, and environmental friendliness for sustainable engineering and green energy storage applications. However, existing challenges regarding advanced material design, processing, recyclability, and commercial scale production need to be overcome to unveil the true sustainability aspects of graphene in the environmental and energy sectors.

Keywords: graphene; sustainability; environmentally friendly; recycled; energy

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1. Introduction

Sustainable carbon materials and nanomaterials have been the focus of recent technical research, especially in the high-tech energy and environment sectors[1]. In addition to sustainability, carbon nanomaterials have countless structural and physical benefits like strength, durability, and recyclability[2,3]. Graphene can be marked as the most competent nanocarbon owing to two dimensional nanosheet nanostructure and methodological aspects[4]. Resourceful approaches have been discovered for the formation of graphene[5]. However, sustainable synthesis paths have been preferred for graphene employing less cost, environmentally safe solvents, and chemicals[6]. The properties of graphene have been further improved by its structural modification, doping, or nanocomposite formation[7]. Furthermore, graphene has been used to recycle waste plastic resins. For sustainable graphene nanocomposite formation, the use of green fabrication strategies and green polymers has been favored[8]. High performance sustainable graphene nanocomposites have been found functional in energy, electronics, engineering, biomedical, and environmental sectors[9]. To enhance the sustainability aspects of existing energy devices like supercapacitors, green electrode materials have been used for easy recyclability as well as lower pollution impacts[10]. Hence, supercapacitors based on sustainable, biodegradable, and recyclable materials have been developed[11]. In this way, sustainable graphene and derived nanomaterials have found wide scope for green energy and environmental applications.

To the best of our knowledge, this article is novel in terms of design, framework, and assembled literature regarding sustainable green based nanomaterials. Here, essential green routes and matrices have been covered for the formation of sustainable graphene nanomaterials. The resulting graphene has been further applied to recycle industrial waste plastics. The use of sustainable graphene nanocomposites has also been essentially explored for energy storage devices like supercapacitors. Consequently, this novel article presents a combination of green technologies for the production of graphene, the formation of sustainable nanomaterials, and the practical application of green graphene. Such collective research directions have not been presented in any of the reported review articles of ecologically derived or resulting ecological nanocomposites using green graphene. Thus, in this article, the design and properties of waste-derived graphene, the resulting sustainable nanomaterials, and green graphene based applications have been scrutinized. Accordingly, the formation of green derived graphene has been highlighted to promote future research in this direction. The need for this review article also rises due to remarkably increased research reports on graphene and graphene nanocomposite research for environmentally sustainable production of graphene and practical and ecological features. Therefore, this review can be claimed to be novel as the most demanding to grow future research in the field of ecological graphene nanomaterials. Consequently, this innovative review article will definitely be beneficial for field-related scientists and researchers dealing with sustainable research on green graphene. Hardly any recent topical comprehensive review reports have been observed on these nanomaterials. Future of sustainable engineering recycled resin industries and energy sectors deficiently rely on overcoming the challenges of producing green graphene nanomaterials using sustainable practices.

2. Environmentally sustainable production of graphene and derived materials

Environmentally sustainable graphene and derived materials have been examined[12–14]. To meet marketable demands to manufacture graphene, numerous safe techniques have been applied[15]. Unfortunately, effective graphene synthesis approaches use toxic chemicals and reagents for synthesis[16–18]. Any technique employing noxious solvents is not sustainable or ecological. A few sustainable, environmental, and safe methods may be listed such as mechanical exfoliation of graphite[19], chemical vapor deposition with green precursors[20], and others[21,22]. A very modest green way of forming graphene is the ball milling method[23]. Hence, the existing graphene synthesis techniques can be modified towards sustainability through using ecofriendly precursors, solvents, chemicals, and sustainable pathways[24–26].

Efforts have been observed regarding the use of sustainable carbon sources for graphene synthesis[27]. Ruan et al.[28] used biscuits and chocolate as carbon sources to form graphene using chemical vapor deposition technique. Kalita et al.[29] employed camphor plant extracts as green carbon source for graphene. Zhang et al.[30] picked glucose as carbon precursor for graphene. All these green methods can be safely used for mass level graphene synthesis.

Tavakoli et al.[31] adopted a green sustainable method for the formation of graphene from graphene oxide using pomegranate juice. Graphene oxide was formed using green Hummer’s process from graphite precursor. **Figure 1** shows graphene formation from graphene oxide in the presence of pomegranate juice (naturally having anthocyanins as reducing agent). Thus, due to electron deficient nature, pomegranate juice better reduced the graphene oxide to graphene. Gu et al.[32] synthesized green sustainable nanocomposites of chitosan, reduced graphene oxide, and silver nanoparticles. **Figure 2** illustrates the mechanism for the formation of reduced graphene oxide and silver nanoparticles based nanomaterial due to electrostatic interactions. These associations led to formation of a compatible nanostructure. According to transmission electron microscopy based morphology studies, reduced graphene oxide/silver nanoparticles revealed wrinkled graphene nanosheet on which silver nanoparticles were found homogeneously scattered. The silver nanoparticles of 15 nm were observed in the micrographs. The silver nanoparticles were also uniformly coated with polymers. Fine dispersion revealed also effectiveness of the green method Thus, the morphology studies supported the formation of a compatible matrix-nanofiller nanostructure. In addition, the green nanomaterials revealed antibacterial effects towards the *E. coli* and *S. Aureus* bacterial strains.

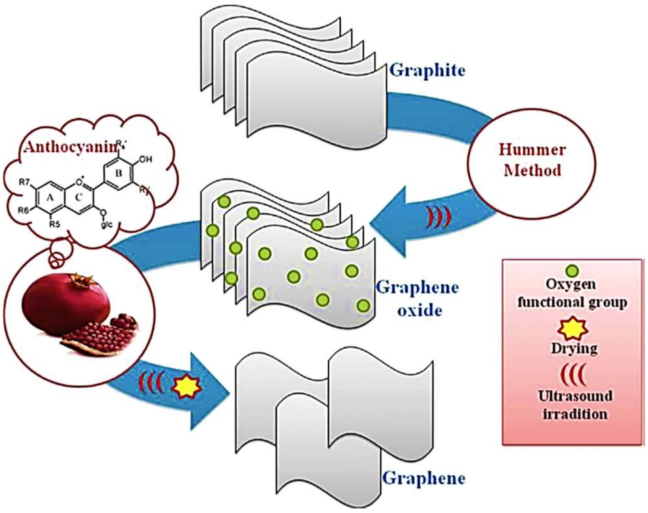

Figure 1. Mechanism of synthesizing graphene nanosheets by using pomegranate via green route[31]. Reproduced with permission from Elsevier.

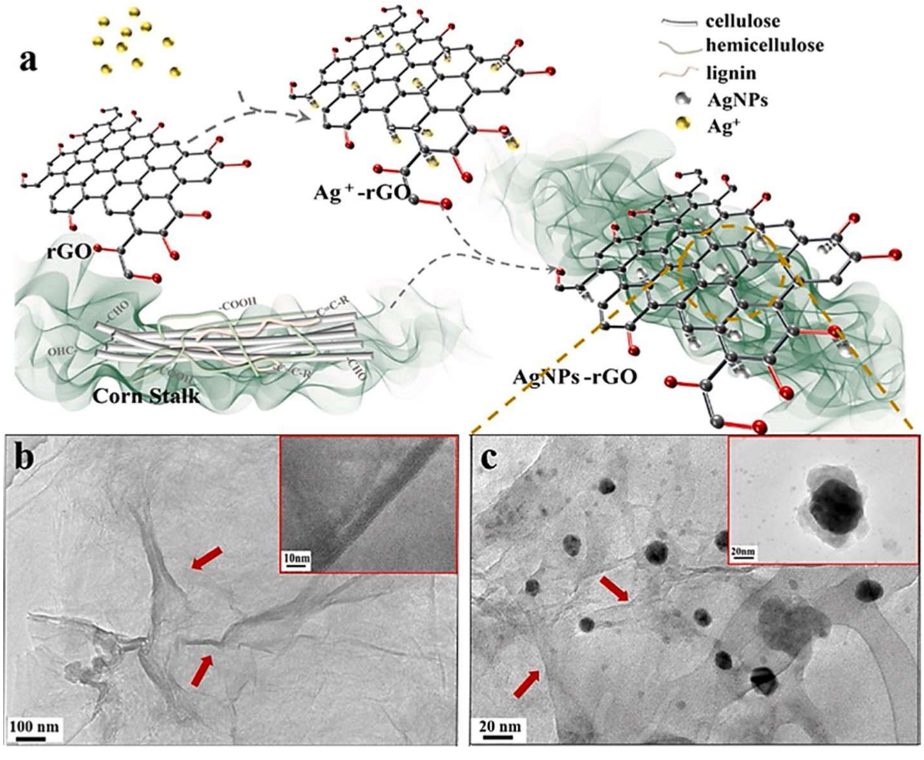

Figure 2. (a) Preparation mechanism of rGO/AgNPs; and **(b)** and **(c)** TEM images of GO and rGO@AgNPs[32].
Red arrows = two dimensional nanosheets of GO and rGO; TEM = transmission electron microscopy; GO = graphene oxide; rGO = reduced graphene oxide; rGO@AgNPs = reduced graphene oxide@silver nanoparticles. Reproduced with permission from Elsevier.

Upadhyay and co-workers[33] extracted reduced graphene oxide from vitis vinifera of grapes. This green method was low cost and facile for commercial graphene production (**Figure 3**). Starting materials was graphene oxide which was formed by Hummer's technique. This process also used green materials for synthesis. According to X-Ray diffraction studies, differences in the graphite, graphene oxide, and reduced graphene oxide were analysed. Structures of graphene oxide and reduced graphene oxide were confirmed through peaks at 10.4° and 23.7°, respectively. Transparent graphene nanosheet nanostructure was further verified using the transmission electron microscopy analysis. Graphene was observed as thin transparent nanosheet which was lightly wrinkled but has fine even surface morphology. Li et al.[34] formed the cellulose, polyaniline, and graphene oxide-based nanocomposite through green in situ polymerization. The 3.5 wt.% nanofiller loading was used in these nanocomposites. This method involves simple solution and in situ polymerization techniques (**Figure 4**). In addition, green starting materials and Hummer's method were also employed for the formation of sustainable materials. X-ray photoelectron spectroscopy was used to study the chemical bonding and structure of the nanomaterial (**Figure 5A**). In addition to C and O elements, nitrogen, sulfur, and chloride were also identified in the study. According to cyclic voltametric curves (**Figure 5B**), the cellulose/graphene oxide/polyaniline nanomaterial had significant electrical response, relative to cellulose/graphene oxide and cellulose/polyaniline. The superior cyclic response of the cellulose/graphene oxide/polyaniline was observed due to the synergistic effects of the conjugated polymer and graphene oxide with green polymer. Gas adsorption-desorption isotherms of cellulose, polyaniline, and graphene oxide based nanocomposite aerogel were studied using Brunauer-Emmett-Teller (BET) technique as given in **Table 1**. The surface area, pore volume, and pore sizes of cellulose/graphene oxide/polyaniline nanocomposite were observed as 66.7, 0.37 cm^3/g, 22.5 nm, respectively. These values were observed lower than the cellulose/graphene oxide and cellulose/polyaniline nanocomposites. These nanocomposites have been used as efficient ecological nanocomposites.

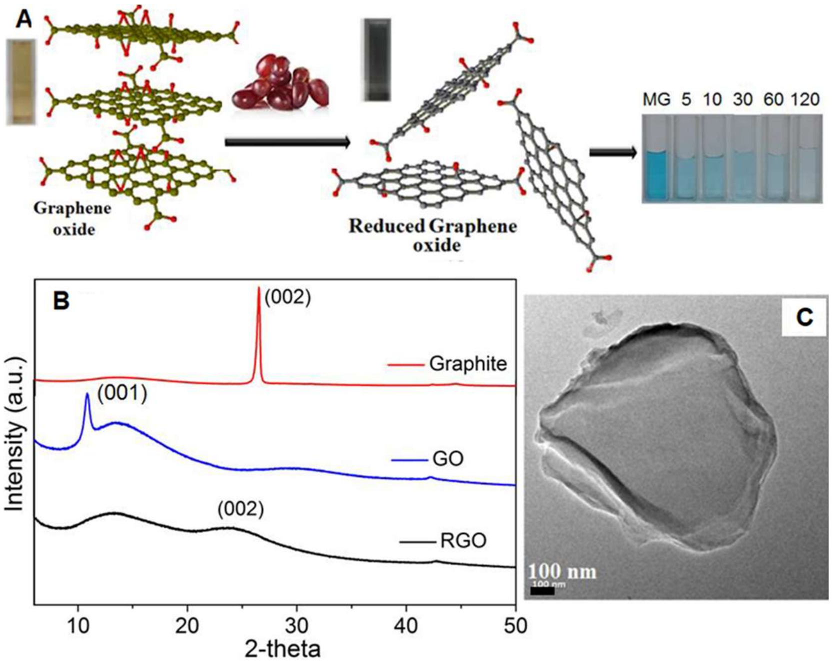

Figure 3. (A) Geen synthesis of reduced graphene oxide; **(B)** X Ray diffraction of graphite, GO, and RGO; and **(C)** TEM image of RGO sample[33].
GO = graphene oxide; RGO = reduced graphene oxide; TEM = transmission electron microscopy. Reproduced with permission from Elsevier.

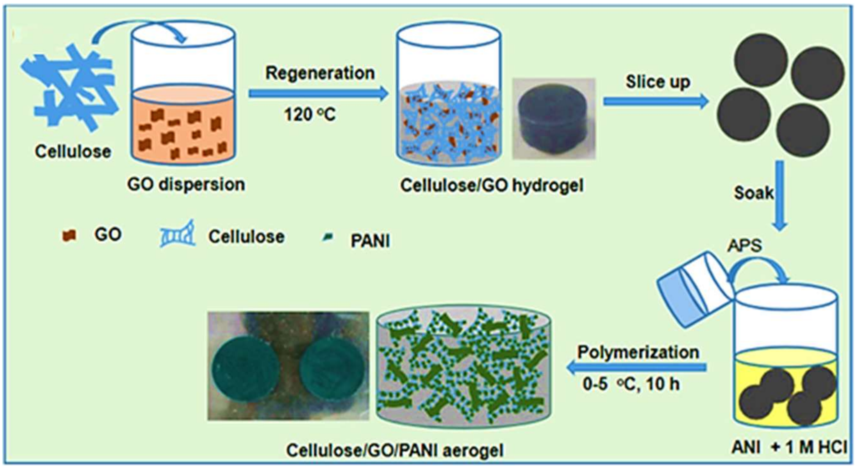

Figure 4. Synthetic route to cellulose/graphene oxide/polyaniline nanocomposite[34].
GO = graphene oxide; PANI = polyaniline; ANI = aniline; HCl = hydrochloric acid. Reproduced with permission from MDPI.

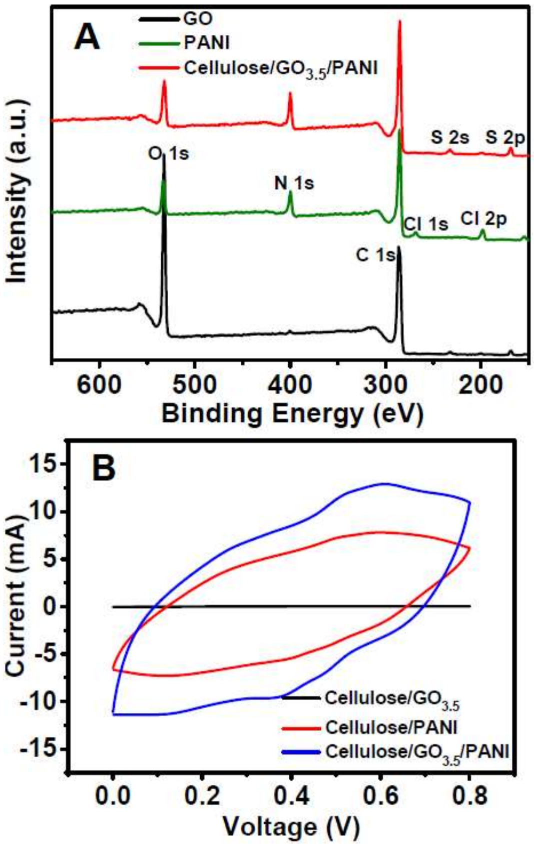

Figure 5. (A) X-ray photoelectron spectroscopy spectra of pristine graphene oxide (GO), neat polyaniline (PANI), and the cellulose/GO/PANI nanocomposite; and **(B)** typical cyclic voltammogram (CV) curves of the cellulose/GO, cellulose/PANI, and cellulose/GO/PANI electrodes at 50 mV/s[34]. Reproduced with permission from MDPI.

Table 1. Brunauer-Emmett-Teller (BET) analysis of the regenerated cellulose, cellulose/GO, cellulose/PANI, and cellulose/GO/PANI[34].

Sample	**S_{BET}**	**Pore volume (cm^3/g)**	**Pore size (nm)**
Cellulose	137.6	0.38	10.9
Cellulose/GO	147.0	0.46	12.5
Cellulose/PANI	68.7	0.36	20.8
Cellulose/GO/PANI	66.7	0.37	22.5

GO = graphene oxide; cellulose/GO/PANI = cellulose/graphene oxide/polyaniline nanocomposite. Reproduced with permission from MDPI.

Gao et al.[35] used Vitamin C and amin reducing agent/stabilizer to fabricate green graphene. The Vitamin C was proved as an active reducing agent to form graphene in large quantity. No harmful solvent was used in this technique[36]. Ren et al.[37] formed a nanocomposite based on chitosan matrix, reduced graphene oxide, and gold nanoparticles. Homogeneous dispersion of gold nanoparticles was observed. Effect of increase in the molecular weight of chitosan was studied on the size of gold nanoparticles. Sayed et al.[38] used green sonication route to form the chitosan, erythritol, and graphene oxide derived nanocomposites. The nanocomposite was applied to detect the Hg^{2+} ions and methylene blue dye. The removal efficiency was observed in the range of 186–205 mg g^{-1}. Sharif and researchers[39] formed ecological chitosan/graphene oxide nanocomposites using green solution method. The interfacial, load transfer, and physical properties were studied and found to enhance with the nanofiller loading levels. Meera and colleagues[40] developed green nanocomposites based on the carboxymethyl chitosan, cashew gum, and boehmite nanoparticles. Including 7 wt.% boehmite nanoparticles augmented the AC conductivity of the nanocomposites. The nanocomposites were found suitable to be applied in the green eco-electronic devices.

3. Graphene for sustainable energy

Since decades, sustainable energy devices and systems have been researched and industrialized[41–43]. One of simple example is the wind turbines with green natural composite based blade materials by replacing the metal blades[44,45]. The sustainable composite materials have low weight, low cost, and high mechanical properties. For sophisticated energy devices like supercapacitors, solar cells, etc., use of sustainable materials has been concerned[46]. Among energy storage strategies, supercapacitors have been categorized as most effective devices and so focused for the use of sustainable materials. In this concern, numerous sustainable green polymers (starch, cellulose, chitosan, etc.) have been used to form supercapacitor electrodes[47–49]. To further enhance the performance of supercapacitor electrodes based on green polymers, matrices have been reinforced with nanoparticles to form high performance nanocomposites. Hence, sustainable synthesis techniques as well as green materials have been used to form sustainable supercapacitors[50]. Sustainable fabrication strategies and materials for supercapacitor electrodes not only produce safe energy but also minimize the environmental risks[51]. Nevertheless, limited research attempts have been observed in literature regarding the sustainable supercapacitor materials[52]. In this context, attaining high efficiency using sustainable supercapacitors has been found efficient. Two-dimensional graphene nanosheets have been considered as best choices for energy storage devices like supercapacitors[53–55]. In addition to superior charge storing competence, graphene owns the advantageous properties like fine microstructure, durability, conductivity, strength, thermal stability, and other physicochemical properties. The functionalized or doped graphene nanosheets have revealed further enhanced electrical conduction and specific capacitance values[56]. Conversion of graphene to high performance nanocomposites has also formed active materials for efficient supercapacitor electrodes. Zhang et al.[57] adopted green techniques like freeze drying and hydrothermal method for the formation of three-dimensional graphene hydrogel or nitrogen doped (N-doped) graphene hydrogel. **Figure 6** shows simplistic routes towards the formation of hydrogels. In green freeze drying and hydrothermal techniques, water solvent was used[58,59]. Then, ammonium bicarbonate was used to form N-doped graphene hydrogel with density of 0.034 g cm^{-3}. **Figure 7** illustrates the cycling performance of neat graphene aerogel and N-doped graphene aerogel at current densities of 0.1 A g^{-1} and 10 A g^{-1}, above > 200 cycles. At 100th cycle, specific capacity of 209 mA h g^{-1} was observed with 93.4% capacitance retention for N-doped graphene aerogel (0.1 A g^{-1}), whereas the specific capacity was decreased at higher current density. Under same conditions, the values were comparatively lesser for the pristine graphene aerogels. N-doping was so found effective to improve the supercapacitor performances.

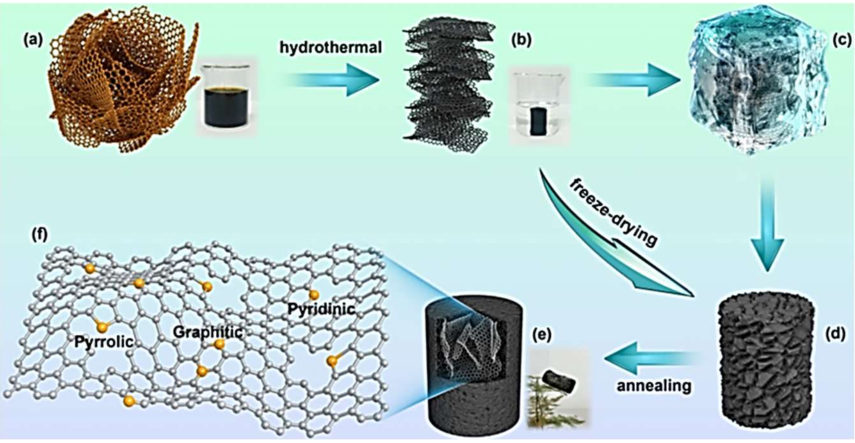

Figure 6. Schematic illustration of synthesis steps for the N-doped graphene aerogel: **(a)** photographic images of graphene oxide suspension (5 mg mL^{-1}); **(b)** freeze-dried graphene hydrogel obtained after hydrothermal reaction, **(c)** freeze-drying treatment; **(d)** 3D graphene aerogel; and **(e)** N-doped graphene aerogel; and **(f)** Schematic illustration of a N-doped graphene aerogel sheet with three nitrogen doping types[57]. Reproduced with permission from springer nature.

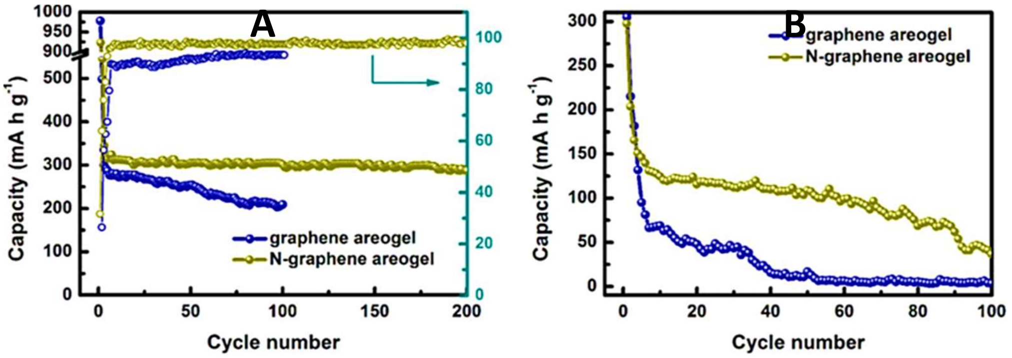

Figure 7. Comparison of cycling performance of graphene aerogel and N-doped graphene aerogel at a current density of **(A)** 0.1 A g^{-1}; and **(B)** 10 A g^{-1}[57]. Reproduced with permission from springer nature.

Çıplak and group[60] reinforced polyaniline with graphene oxide, reduced graphene oxide, and gold nanoparticles to form the nanomaterials aiming the sustainable supercapacitor electrodes. The nanocomposites were prepared using green in situ polymerization technique. An ecological method was also used to form graphene oxide. **Figure 8** illustrates the specific capacitance scans for unfilled polyaniline, polyaniline/graphene oxide-gold nanoparticle, and polyaniline/reduced graphene oxide-gold nanoparticle nanocomposites. The specific capacitance results revealed that the polyaniline/reduced graphene oxide-gold nanoparticles had value of 212.8 Fg^{-1}, which was observed 64 % higher than the unfilled matrix. Superior specific capacitance results were obtained due to better π-π interacted nanostructure supporting the high electron conduction through the system. Arthisree et al.[61] fabricated the polyaniline, polyacrylonitrile, and graphene quantum dot based sustainable nanocomposites as supercapacitor electrode material. **Figure 9** shows the design of supercapacitor based on nanocomposite with 1.5 wt.% loading and image of voltage generation for the optimal nanocomposite designed device. The specific capacitance of the sustainable electrode was recorded between 100 to 600 Fg^{-1}[62]. The supercapacitor represented reasonable output power of 1.4 V. Fine performance was due to synergistic effects in the matrix-nanofiller and electron conductivity features[63]. Hence, the formation of sustainable nanomaterials has contributed towards the green charge storage devices[64]. Cellulose matrix has been explored as low cost green material for energy systems[65–67]. Green supercapacitor electrodes based on cellulose and graphene have high electron conduction, charge transportation, and capacitance values[68]. Three dimensional cellulose/graphene oxide sponges had high capacitance performance[69–71]. Conductive polymers have been widely used in supercapacitors[72]. Green prepared graphene and synthesis route has been applied to form conductive polymer/graphene supercapacitor electrodes[73–75]. High performance electrodes have high surface area, specific capacitance, charge density, and electrochemical properties[76].

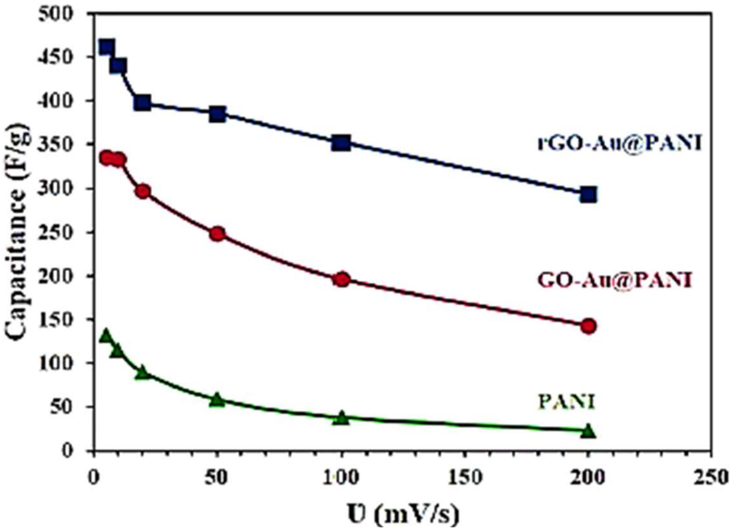

Figure 8. The specific capacitance dependence on the scan rate (5–200 mVs^{-1}) for neat PANI, GO-Au@PANI, and rGO-Au@PANI[60].
PANI = polyaniline; GO-Au@PANI = graphene oxide-gold nanoparticle@polyaniline; rGO-Au@PANI = reduced graphene oxide-gold nanoparticle@polyaniline. Reproduced with permission from Elsevier.

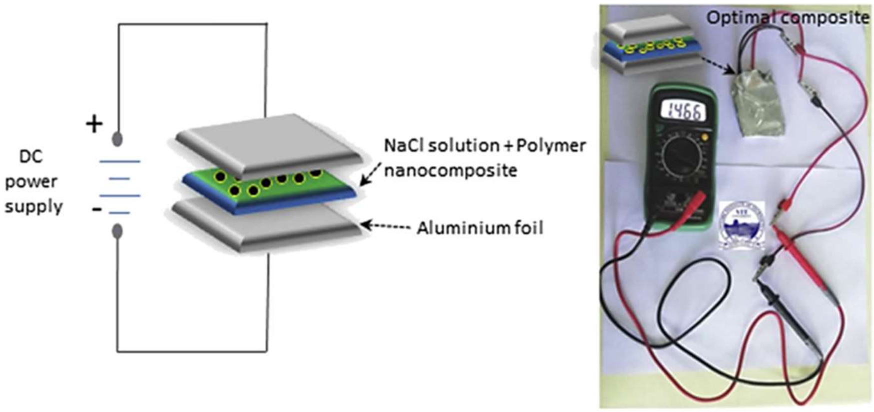

Figure 9. Schematics of PAN/PANI@G-1.5 wt.% based supercapacitor along with the characteristic digital photograph of optimal nanocomposite for voltage generation[61].
PAN/PANI@G = polyacrylonitrile/polyaniline/graphene quantum dot. Reproduced with permission from Elsevier.

4. Graphene for conversion of recycled resin to graphene or sustainable materials

Globally, plastic based industries are generating greenhouse emissions and environmental pollution, since decades[77–79]. According to worldwide surveys, 200 million tons of plastic waste can be generated annually[80–82]. Out of which only 10% of plastics is usually recycled and rest causes ecological hazards[83]. Consequently, there is stern need of developing recycling strategies for waste plastics. However, the recycled plastics usually have low mechanical and physical properties for further technical uses and advanced techniques must be invented[84]. For the formation of graphene, various sustainable and non-toxic carbon sources have been employed[27]. El Essawya et al.[85] established a route to form graphene using the recycled poly(ethylene terephthalate) (basically waste bottles). This method is commonly referred as waste-treats-waste[86,87]. Here, synthesis of graphene using poly(ethylene terephthalate) waste bottles is given in **Figure 10**. The waste bottles were treated with very high temperature of 800 °C for 1 h. Resulting dark colored product was ground to form graphene. The as prepared graphene was used to treat the methylene blue dye.

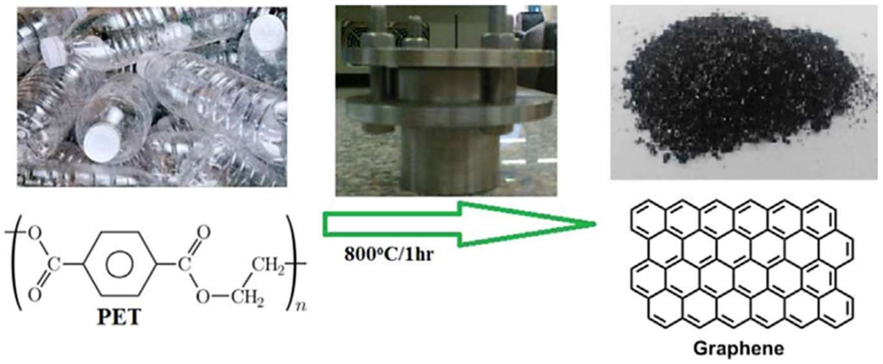

Figure 10. Schematic representation for the synthesis of graphene from PET recycled bottles[85]. PET = poly(ethylene terephthalate). Reproduced with permission from Elsevier.

Numerous reinforcing agents and compatibilizers have been used to elevate the features of recycled polymer resins[88]. Furthermore, blends of recycled polymers have been prepared, as a solution to attain enhanced properties[88]. To improve the performance and applications of recycled polymers, effective nanofillers used are graphene, few-layer graphene, graphene nanoplatelets, and other graphene derivatives[89]. Among these nanocarbons, few layer graphene has been found to improve the heat constancy, strength, and chemical stability features[90]. Moreover, doped graphene like sulfur doped graphene has been filled in recycled polymers to enhance the physical characteristics[91,92].

Few layers graphene has been industrialised on commercial scale for desired sustainability applications[93]. Among recycled polyethylene resins, high density polyethylene has been efficiently recycled on large scale[94,95]. To augment the mechanical or thermal features of recycled polyethylene resins, commercial high-density polyethylene has been added through melt mixing[96]. The blend of recycled and non-recycled high-density polyethylene has been filled with different graphene nanoparticles like few layer graphene and graphene nanoplatelets using melt compounding technique. Specifically, few layer graphene has been used to improve the morphological, rheological, thermomechanical, and chemical resistance features of polyethylene resins[97–99].

Diallo et al.[100] researched on polyethylene resin which has been used to form recycled resin with reasonable physical features for industrial purposes. The recycled polyethylene/polypropylene blend as well as few-layer graphene filled recycled polyethylene/polypropylene nanocomposites have been prepared. Influence of adding nanofiller on the mechanical, thermal, and rheological characters were explored. Inclusion of few layers graphene was effective to form large amount of recycled polyethylene resin. Sultana et al.[101] also investigated the effect of adding few-layer graphene on the processing of recycled polyethylene. The recycled polyethylene/polypropylene blend filled with few-layer graphene was also formed. Consequently, the morphology and mechanical properties of the resulting nanomaterials were explored. The nanocomposites were developed using the melt extrusion method. **Figure 11** displays scanning electron micrographs of unfilled recycled polyethylene/polypropylene blend as well as recycled polyethylene/polypropylene/few-layer graphene nanocomposites with varying nanofiller contents.

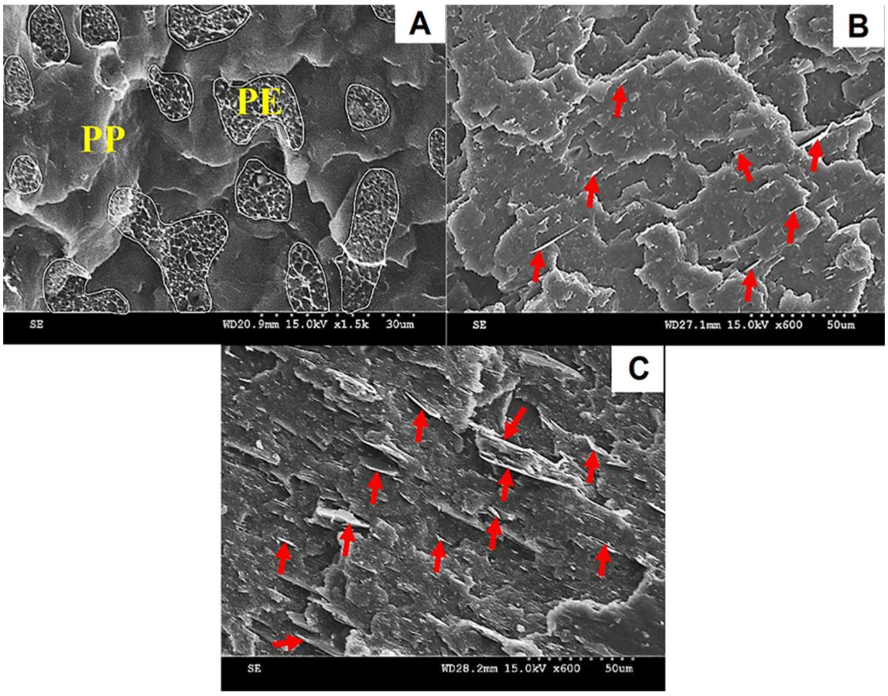

Figure 11. Scanning electron microscopy images of the fractured surface of the **(A)** neat PE/PP; **(B)** R-(PE/PP)/FLG = 96/4; and **(C)** R-(PE/PP)/FLG = 90/10.
The arrows indicate a few of the FLG, visible on the fractured surface of the R-(PE/PP)/FLG composites[101]. PE/PP = polyethylene/polypropylene; R-(PE/PP)/FLG = recycled polyethylene/polypropylene/few-layer graphene; FLG = few-layer graphene. Reproduced with permission from MDPI.

Neat blend matrix formed a phase separated structure with globular areas. Whereas the nanocomposite did not revealed phase separated structure and instead waves like pattern was observed on the fractured surface. Increasing the amount of nanofiller affected the surface morphology of the blend matrix. Graphene layers can be seen dispersed in the recycled blend matrix. It can be analysed that the nanocomposite with higher graphene loading had better consistent morphology at fractured surface due to compatible morphology. In other words, with the rising nanofiller contents compatible nanostructure was formed and homogeneous microstructure was observed. **Figure 12** shows the tensile and flexural strength, tensile and flexural modulus and impact strength of the nanocomposites vs. rising graphene contents. The mechanical results were well supported by the morphology studies. All the tensile and flexural strength and modulus and impact strength properties were found to enhance with the few layer graphene loadings. Conclusively, 10 wt.% nanoparticle addition led to superior mechanical profile due to fine interactions in the nanocomposite and load transfer effects and resistance to crack propagation effects. Upsurge in tensile strength, flexural strength, and impact strength were found as 9%, 23%, and 9%, respectively, relative to pristine polymers. **Table 2** displays change in the mechanical features of 4 wt.% nanofiller loaded samples to analyse the performance of the samples. In this way, the recycled resins have been modified for industrial uses towards sustainability applications[102,103].

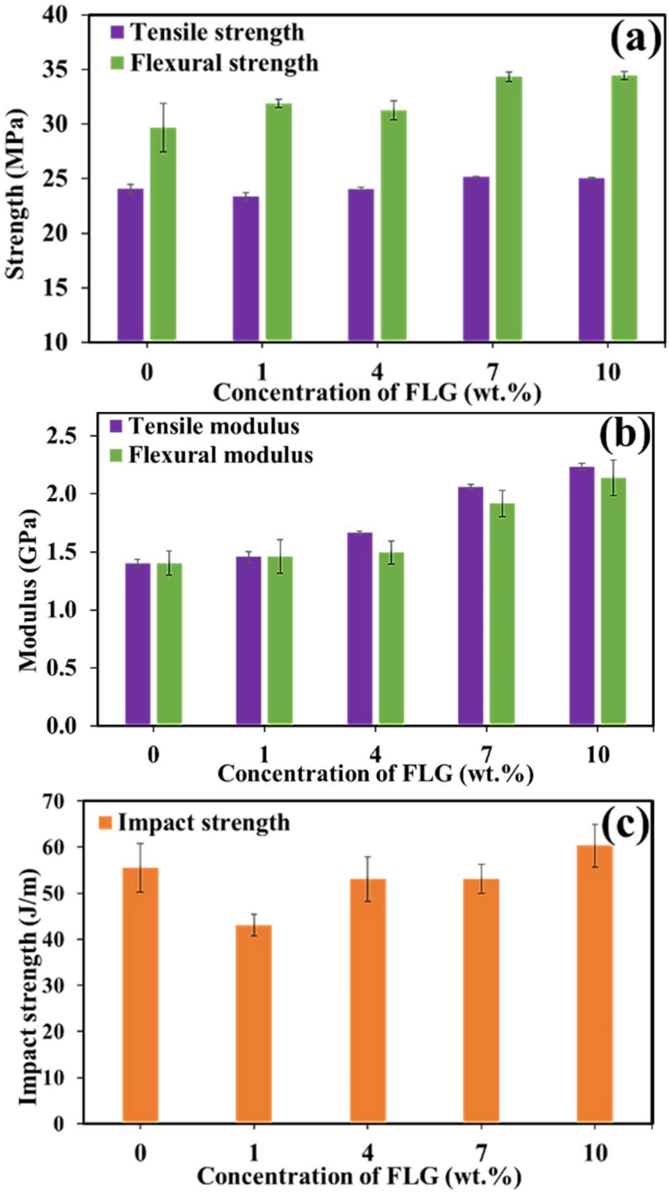

Figure 12. **(a)** Tensile strength and flexural strength; **(b)** tensile modulus and flexural modulus; and **(c)** impact strength of R-(PE/PP)/FLG composites as a function of FLG concentration[101].
R-(PE/PP)/FLG = recycled polyethylene/polypropylene/few-layer graphene; FLG = few-layer graphene. Reproduced with permission from MDPI.

Table 2. Mechanical property variation (%) in compounds showing a significant change in properties compared to the neat R-(PE/PP).

Properties	**Neat polymer blend**	**4 wt.% nanofiller**
Tensile strength	24 MPa	+9%
Tensile modulus	1.4 GPa	+37%
Flexural strength	30 MPa	+23%
Flexural modulus	0.9 GPa	+34%
Elongation at break	34%	−80%

5. Challenges and future

All above discussed green approaches for graphene and nanocomposite synthesis offer remarkable opportunities towards the future sustainable engineering and supercapacitor expertise (**Table 3** and **Figure 13**). Furthermore, the continuous ongoing research efforts may resolve the underlying design, feature, and performance challenges in these fields. Important challenges have been encountered for large scale fabrication of graphene and derived forms through green routes. Some attempts have been performed on using safe, recyclable, and green bio-based carbon sources to form graphene. However, these methods have not been applied for graphene production on commercial scale. Although various studies have been reported for the formation of graphene from sustainable sources, formation of high quality graphene is still uncertain[104]. Consequently, the graphene obtained from ecological sources may have defects, impurities, non-homogeneous morphology, and oxidized surface, relative to the graphene obtained through frequently used techniques. In addition, strength and charge transportation properties of graphene nanostructure have been greatly affected using waste derivation method. Hence, it has been found challenging to use the waste derived graphene for technical applications. These challenges also hinder the large scale production of useful graphene nanostructure. In this regard, further research efforts have been found desirable for the production of high quality graphene through green methods, especially from waste and biomass. Continuous research on sustainable graphene materials have led to the formation of sustainable and recyclable supercapacitor electrode materials with superior reliability, capacitance, charge density, power density, charge-discharge, and cyclic performance. For recyclable electrodes, sustainable polymers and green synthesis routes have been applied. Nevertheless, limited material designs and fabrication methods have been explored so far for sustainable graphene based energy devices. Furthermore, sustainable supercapacitors have yet not been used for industrial scale energy applications. Future research may expand the use of graphene nanomaterials towards the more sustainable energy and electronic devices like micro-supercapacitors and microelectronics.

Here, research progress in the field of ecologically sustainable production of graphene and graphene nanocomposite membranes need to be analyzed according to the demands for the production of high quality of graphene keeping in view the crucial foremost difficulties in this field and also according to the end applications focused using green graphene Research progress in this field need to be categorized according to the possible waste or ecological sources used for the production of green graphene. Design and characteristics of sustainable and ensuing graphene nanocomposite need to be studied comprehensively. Mechanisms for the conversion of ecological sources to green graphene also need to be analyzed. Consequently, focused research efforts have been required on the structure, microstructure, mechanical, and physical properties of the green graphene and nanocomposites.

In the nanocomposite form, future studies must focus the dispersion patterns of green graphene in nanomaterials. As green graphene is not pure and high quality having lots of surface defects, therefore, it has been found indispensable to study the matrix-nanofiller interactions, miscibility, interfacial, and miscibility effects in the resulting nanomaterials. Research must focus the advanced methods to deal with the structural issues in the nanomaterials arising due to the use of waste derived graphene nanostructures. The durability of the green graphene based nanomaterials must also researched to find out improved methods for production. In the waste derived graphene production methods, optimum parameters and identification of perfect graphene design need to be investigated. Major challenges identified in this sector are towards the commercial scale production of the green graphene. In addition, there is lack of targeted research in the area of conversion of plastics or recycled wastes into graphene. More focused research efforts have been certainly desirable in these discussed research directions to form high performance sustainable graphene nanostructures.

Table 3. Features of sustainable graphene and graphene based nanocomposites.

Graphene, derivative, or nanocomposites	**Green fabrication techniques**	**Properties/potential**	**Ref.**
Reduced graphene oxide	Vitis vinifera from grape extracts for graphene oxide reduction; Hummer's method	X-Ray diffraction peaks for graphene oxide and reduced graphene oxide at 10.4° and 23.7, respectively	[33]
Reduced graphene oxide	Pomegranate juice for graphene oxide reduction	Mechanism studies	[31]
Reduced graphene oxide	Vitamin C as reducing agent	Large scale production	[35]
Graphene	Chemical vapor deposition; Biscuits/chocolate carbon source	Green technique	[28]
Graphene	Camphor plant extracts as green carbon source	Mass level synthesis	[29]
Graphene	Glucose carbon source	Efficient carbon source	[30]
Chitosan/reduced graphene oxide/gold nanoparticles	Chitosan as as stabilizer and reducing agent	Low molecular weight chitosan enhance kinetic rate constant of 0.21 min^{-1}	[37]
Chitosan/erythritol/graphene oxide	Sonication	Methylene blue dye and Hg^{2+} ions removal efficiency of 186.23 mg g^{-1} and 205 mg g^{-1}, respectively.	[38]
Carboxymethyl chitosan/cashew gum/boehmite nanoparticles	Water solvent	AC conductivity and low activation energy	[40]
Epoxy/chitosan encapsulated graphene oxide	Biodegradable chitosan	Interface formation between epoxy and chitosan-graphene oxide; elastic modulus enhanced by 65%, load transfer	[39]
Chitosan/reduced graphene oxide/silver nanoparticles	Electrostatic method; corn stalk for silver nanoparticles	Morphology; antibacterial activities against the *E. coli* and *S. Aureus* bacterial strains	[32]
Polyaniline/graphene oxide/gold nanoparticles and polyaniline/reduced graphene oxide/gold nanoparticles	Cetraria Islandica L. Ach lichen for reducing graphene oxide; green in situ synthesis	Supercapacitor electrode has scan rate of 5–200 mVs^{-1}; specific capacitance and capacitance retention of 212.8 Fg^{-1} and 86.9%,	[60]
Cellulose/graphene oxide/polyaniline	In situ technique	Electron conductivity 1.15 Scm^{-1}; areal specific capacitance 1218 $mFcm^{-2}$; energy density 1201 μW/cm^2	[34]
Chitosan/reduced graphene oxide/silver nanoparticles	Green materials and methods; phosphate buffer saline	Anti-bacterial packaging against *E. coli* and *S. aureus* bacterial strains.	[32]
Three dimensional graphene hydrogel; nitrogen doped graphene hydrogel	Freeze drying; hydrothermal	Green methods; current densities 0.1-10 A g^{-1}, specific capacity 209 mA h g^{-1}; capacitance retention 93.4%	[57]
Polyaniline; graphene oxide, reduced graphene oxide, gold nanoparticles	Green in situ polymerization	Specific capacitance 212.8 $Fg^{-1;}$ capacitance retention 64 %	[60]
Polyaniline, polyacrylonitrile, graphene quantum dot	In situ; Solution	Specific capacitance 100-600 Fg^{-1}	[61]
Recycled poly (ethylene terephthalate) to form graphene	Waste-treats-waste	High temperature stability 800 °C; methylene blue dye adsorption	[85]
Recycled polyethylene/few-layer graphene	Melt extrusion technique	Tensile strength, flexural strength, and impact strength were found as 9%, 23%, and 9%, respectively	[101]

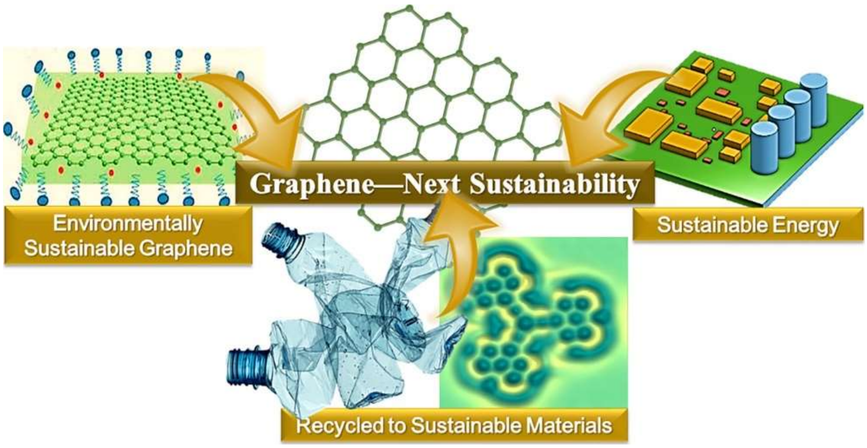

Figure 13. Graphene nanostructures in sustainable applications.

An important ecological application of graphene has been reported for the energy storage devices. A benign solution for commercialization of energy storage technology is to use the green processing approaches. Incidentally, ecological friendly solvent, process, and materials have been preferred along with the green formed graphene. Materials compatible with the green solvents like water, ionic liquid, etc. need to be used. Applying green solvents depicted low noxiousness and facile processability effects. However, there are challenges in utilization of green nanotechnologies for energy storage devices. For example, using water or non-toxic solvents for sustainable materials may not be suitable for all type of efficient energy designs. Conjugated systems have been found most efficient for energy devices; however, the green synthesis techniques cannot be adopted for all combinations of conducting graphene nanocomposites. Applying green solvents like water may result in poor dispersion and conductivity properties, so decreasing the supercapacitor performance. Future research is desirable is needed in this regard to form some novel ionic liquid based green solvents in place of water or toxic chemicals. Comprehensive future efforts are required to use green polymers like cellulose and green graphene to form the supercapacitor electrodes. Moreover, future research on functionalization of graphene to form green nanostructures must be focused.

6. Conclusions

This state-of-the-art overview fundamentally sheds light on the ecological aspects of graphene, which of course define the next technological sustainability vision. Here, three sustainability characteristics of graphene have been engrossed, including the (i) environmentally sustainable production of graphene and nanocomposites; (ii) applying graphene to convert waste industrial resins into valuable recycled polymers; and (iii) using graphene in sustainable energy devices and systems. The first important stage is the identification of sustainable techniques to form green graphene on a large commercial scale. Then, there is a need to identify sustainable techniques for the formation of graphene-based nanocomposites. Green synthesized graphene and nanocomposites have been efficiently applied to form recycled engineering resins and electrodes for energy devices. However, the sustainability prospects of graphene need to be further researched to discover safe synthesis strategies and degradable material designs.

Conflict of interest

The authors declare no conflict of interest.

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