Review Article

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Recycling waste sources into nanocomposites of graphene materials: Overview from an energy-focused perspective

Abstract: In line with the global recognition of waste-to-wealth concept aiming for circular economy, scientific articles are published in greatly increasing number on the eco-friendly and sustainable utilization of carbon nanocomposites. However, control on the structure and properties of waste-derived carbon nanomaterials still requires substantial future research. In this review, recycling materials into nanocomposites containing graphene are narrated by overviewing all the 120 publications currently available in the literature including their pioneering study in 2012 and their recent developments until 2022, focusing on energy-related aspects of functional graphene-based nanocomposites. Interestingly, almost all currently available sources report on composites in which graphene is a high value-added filler or matrix, and only the other phase originates from wastes. Flexibility of process parameters of pyrolysis methods enables the synthesis of biomass-derived graphene composites for virtually any kind of industrial applications. Biomass often acts both as carbon and SiO_2 source, while only a few percentages of graphene material induce significant changes in their physicochemical properties. Utilization of wastes for energetic composites increases abruptly due to their outstanding price-to-value ratio and reusability. Future perspectives and current green chemistry or human health related challenges are also discussed to pave ways for new developments using unexplored waste sources.

1 Introduction

The growing consumption of durable and nondurable consumer goods and the associated production of waste materials is alarming global population for taking urgent steps towards more efficient measures in environmental protection [1]. One of the fastest growing wastes is waste electrical and electronic equipment (WEEE) [2]. According to a recent report by Shittu *et al.*, the global WEEE production was 54 million tons (MT) in 2019 which is a rise of 45 MT since 2016 [3]. This escalation is expected to upsurge to 75 MT by 2030 [4]. The concerns are thoughtful due to human health influences and loss of bio-resources as climate changes. In a recent review about systematic waste management, 366 articles were evaluated according to correlations between country income groups and different issues that indicate possible future trends in the use of municipal solid wastes [5].

Waste materials are used as raw materials in the production of value-added products. Development of perfect industrial symbiosis and cost benefit analysis in meeting the demand of sustainable green energy and eco-friendly recycling of waste materials has been one of the most perplexing waste-related issues in recent decades [6]. Plastic waste, which is the third most abundant waste source on the planet after food and organic waste, has recently emerged to be one of the world's most serious public health and environmental issues [7]. The total volume of plastic waste is growing in lockstep with the global population and per capita consumption. The amount of waste produced is inextricably linked to consumption and production patterns [8], and may differ substantially

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even in the same country. For example, Malaysian household waste generation varies by region and economic status, ranging from 0.85 to 1.5 kg per person per day [9].

To deal with the global concerns on waste generation, there is an ongoing demand for the reconsideration and improvement of waste management procedures. One notable example is to introduce economical fabrication methodologies of waste-derived energetic carbon nanomaterials such as graphene, carbon nanotubes (CNTs), and graphene quantum dots (GQDs) for energy storage applications [10]. These energetic substances are classified according to four categories based on their dimensionality features according to the common terminology used in materials science and nanotechnology: zero-dimensional (0D), one-dimensional (1D), two-dimensional (2D), and three-dimensional (3D) nanomaterials. Zero-dimensional energetic nanomaterials [11] include carbon quantum dots and spherical or roughly iso-dimensional carbon nanoparticles (NPs), which show exciting and indispensable prospects in the energy conversion and storage including photo- and electrocatalysis or light-emitting diode and photovoltaic cell fabrication [12]. 1D energetic nanomaterials [13], as the second category, include nanowires, nanorods, nanobelts, and nanotubes. The third category is that of (2D) materials characterized by a typical layered structure [14]: nanosheets and nanomembranes. Finally, 3D energetic nanomaterials [15], constitute structural hierarchies of the previously mentioned nanomaterials as base units. Flower balls made of nanosheets and hollow microspheres made of nanorods are noteworthy examples of such 3D hierarchical structures [16]. Thus, graphene materials are suitable for the synthesis of composite materials with different structural, chemical, and morphological properties and can be produced and used in a wide variety of ways. Figure 1 shows distinctive examples highlighting schematically the four generic types of synthesis methods for the production of waste graphene-based nanocomposites. It is remarkable that most synthesis strategies found in the literature involve two common steps: (i) formulation of a composite liquid dispersion containing both composite counterparts in suspended form and (ii) ultrasonication (either bath or tip sonication). Usually the forthcoming steps differ: the liquid is usually removed (*e.g.*, by spray evaporation, drop casting, or centrifugation/drying) or it is kept during the formation of the composite counterpart (hydrothermal treatment of carbon sources and metal salt precursors).

Graphene has an atom's thickness with a high surface area to volume ratio [17], as well as exceptional mechanical, chemical, thermal, and electrical properties [18]. Recent price checks in the market have revealed the worldwide concerns on the production of commercial graphene. Large-scale production and supply of cost-effective graphene-based nanomaterials with desired properties for specific applications are still an obstacle for its endorsement with high-end consumers and industries [19]. A plentiful of studies have reported on the preparation of nanocomposites of graphene-based materials (chemically derived graphene or graphene oxide, GO) associated with inorganic compounds [20–25], or with polymers [26–29], and demonstrated their utility in extraction processes [30], anode materials for batteries [31], electrochemical supercapacitors [32], bio-based electrocatalytic sensing [33], photocatalysis [34], wastewater treatment [35], energy storage, and other related applications as shown in Figure 2 [36]. In 2021, graphene-based composite materials have been explored across half a dozen review papers, ranging from bio-based graphene nanocomposites [37,38], *via* magnetic GO nanocomposites [39], to graphene nanocomposites for lithium-ion batteries (LIBs) [40], and biomass-generated graphene like carbons for energy devices [41]. None of these reviews focused on waste-derived graphene nanocomposites.

However, the recent publication trend clearly indicates that the incentive towards recycling of waste derivatives into carbonaceous energetic nanomaterials is progressively increasing [42–45]. Various environmental friendly approaches are required to provide value-added solutions for waste recycling to replace non-biodegradable or toxic conventional practices [46,47]. According to the Web of Science search till July of 2022, a total of 62 papers have been published during 2012–2021 regarding graphene nanocomposites using different waste materials followed by an enormous number (58) of new studies in 2021–2022. This shows a significantly increasing and futuristic attention (as presented in Figure 3) of this hot topic among scientific communities.

Based on this, a thorough overview is required to compile all the state-of-the-art publications that reported on waste-derived graphene-based nanomaterials. Therefore, in this narrative review, studies on recent improvement of eco-friendly waste-generated graphene nanocomposites are highlighted. To our surprise, this crucially important topic has not been covered earlier.

The comprehensive search strategy employed to locate all related publications relied on the Web of Science database advanced search option. The selective keywords and the derivatives of "waste," "graphene," "GO," "composite," and "nanocomposite" were used. Peer reviewed research studies in English language were considered for inclusion. Owing to the burgeoning advancement in this field, articles since 2021 are treated separately and they are only involved in statistics without extensive analysis. Instead of providing a critical discussion of the available literature
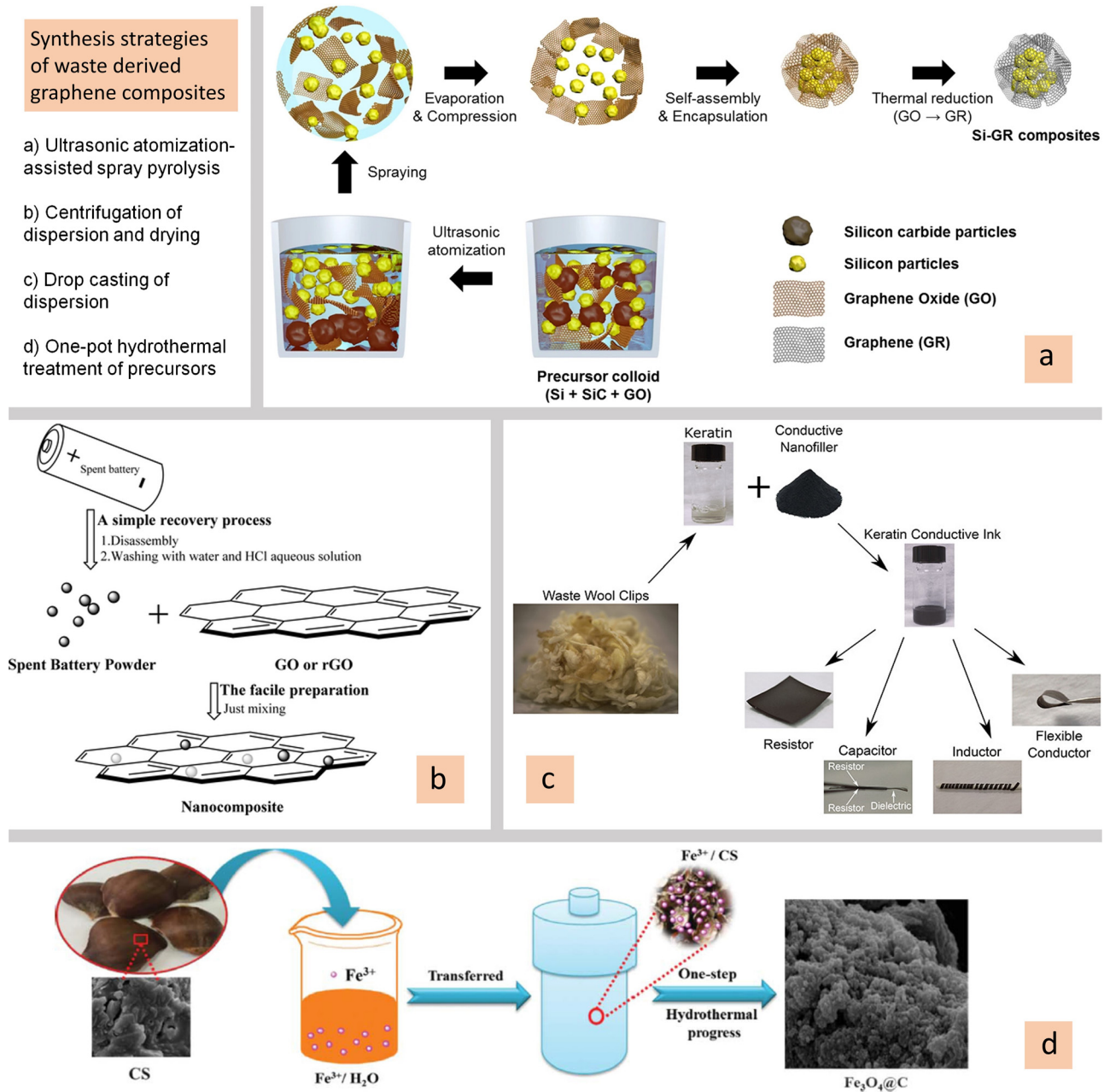
Figure 1: Commonly employed synthesis strategies in the current literature for waste-derived graphene nanocomposite fabrication, showing schematic flowcharts of the synthesis steps. (a) Spray pyrolysis derived silicon sludge waste-generated nanocomposite [219]; (b) physically mixed and dispersed MnO_2/GO and MnO_2/rGO composites from spent battery powder [230]; (c) drop casted waste wool derived keratin/graphene thin films [217]; (d) hydrothermal synthesis of magnetic graphitic carbon nanocomposite using chestnut shell (CS) [224]. All cited display items were reproduced with copyright permission.

(the full list of which is tabulated as Supporting Information), we aimed to collect papers in the form of a narrative review involving the general discussion of waste-derived graphene nanocomposites in order to give a broad perspective of the current state of the art.

Thus, we begin this review with the discussion of the role of waste materials in the environment and their applicability, followed by a brief overview of graphene-based nanocomposites in industry-related applications, highlighting especially those fields that are associated with energy harvesting and storage as displayed in Figure 4. Then, we combine these two areas into one concept, the main focus will be laid on the comprehensive overview of waste-derived energetic graphene composites. By gathering
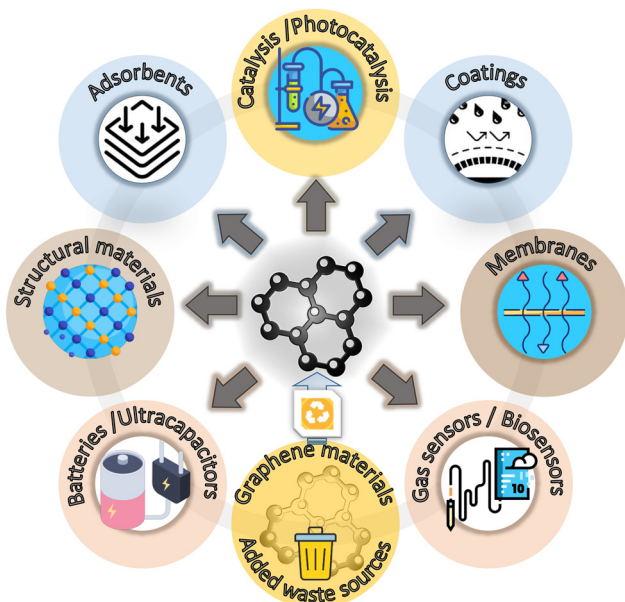
Figure 2: The most notable applications of waste-generated graphene-based nanocomposite materials.

many different examples of their efficient production and use, we foresee that this review may generate further interest in the discovery and use of new waste-associated precursors and novel methodologies towards the safe and efficient conversion of wastes to valuable energy-related products.
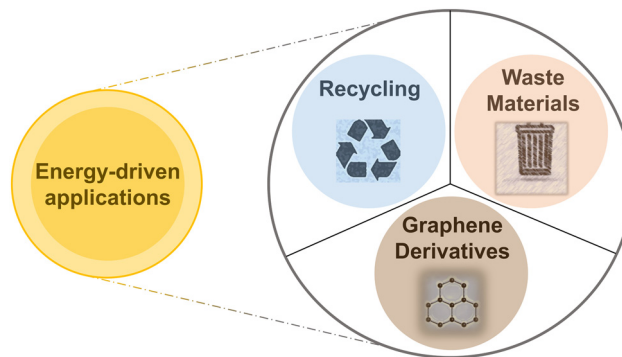
Figure 4: Recycling of waste materials into graphene nanocomposites for energetic applications.

2 Waste materials: their environmental impact and means for possible reuse

Waste materials are generally defined as liquids or solids, which are unwanted, unusable, or at least they lose their basic function after their primary use [48]. The Environmental Quality Act 1974 proclaimed by the Food and Agriculture Organization of the United Nations defines waste as any matter, regardless of solid, liquid, or gaseous form, which are discharged or emitted to the environment resulting in its short-term or long-term change [49,50]. Liquid wastes are, most commonly, residential,
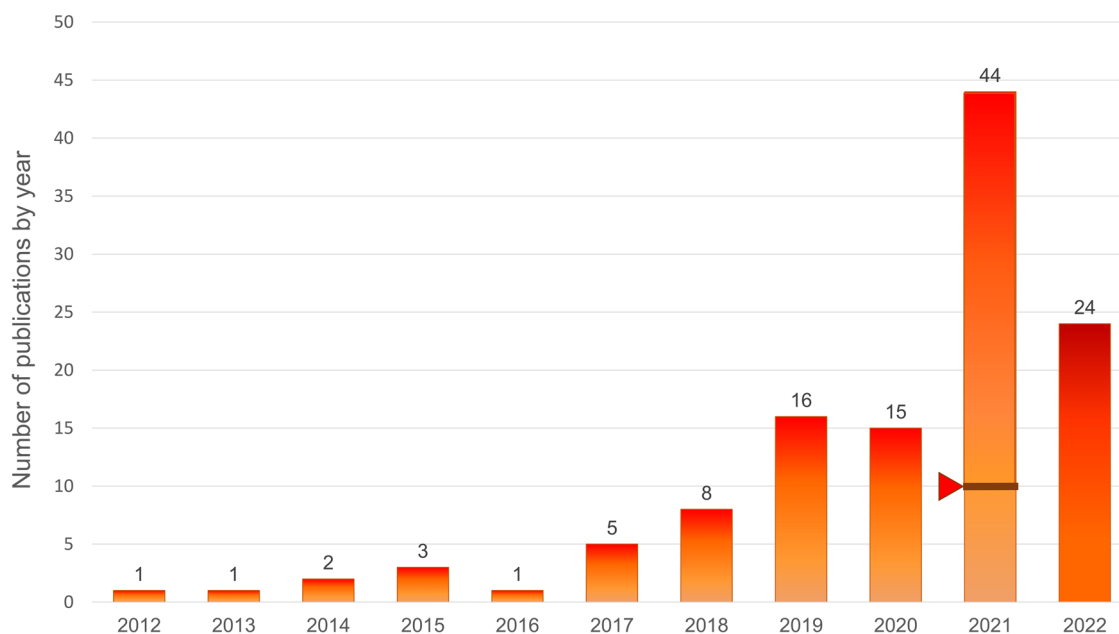
Figure 3: Number of publications related to waste-derived graphene nanocomposites in the past decade.

commercial, or industrial excess wastewaters released after their use such as cleaning, processing, and cooling [51].

Primarily, waste materials originate from commercial, construction, household, industrial, institutional, imported, and public goods or products [52]. Waste materials have also been categorized into solid and municipal waste, hazardous waste, industrial waste, radioactive waste, and agricultural waste [53,54]. These waste materials are disposed irresponsibly to the surrounding deposits, or even illegally to an adjacent abandoned urban or provincial area, which creates an adverse effect to the environment [55]. Therefore, as per current regulations such as Environmental Quality Act 1974 and Environment Quality (Schedule Waste) Regulations 2005, emerging challenges have been observed which are threatening the environmental and waste management [56,57]. Figure 5 illustrates the continent-based annual waste production categories generated by the World Bank Database.

It is noteworthy that the projected values of annual waste production for 2030 and 2050 indicate the largest absolute growth for Sub-Saharan Africa, but the growth rate stays minor in four different regions with already different economic development states (Europe and North America *vs* Middle East and Latin America) [7,58]. This indicates that waste production does not correlate with the current economic development level, but rather with the foreseen rate of development. Hence, the total amount of waste generated is predicted to triple in low-income countries by 2050 [59,60]. In addition, East Asia/Pacific region is expected to generate the world's largest amount of waste *i.e.*, 23%, on the other hand, North Africa and Middle East are estimated to produce around 6%. The highest projection of waste in South and East Asia/Pacific regions require planned strategies to overcome the consequences to the environment, health, and life [61,62].

2.1 Classification of waste materials

One practical way to classify waste materials is based on the level of detrimental effects they have on the environment. Bertram *et al.* [63], classified wastes into four categories: (i) eco-friendly, (ii) biodegradable, (iii) non-biodegradable, and (iv) toxic wastes. They suggested the introduction of color codes to indicate these categories. Herein, gray represents category (i), that is, favorable types of wastes which do not pose any harm to the environment [64]. White and blue color codes symbolize waste materials of category (ii) and (iii), respectively, which degrade by more than 20% of their original mass known as biodegradable, or less than 20% called as non-biodegradable. Finally, toxic and harmful types of waste materials are categorized under brown color code. The color classification index which indicates the environmental impact of waste, and its examples are tabulated in Table 1.
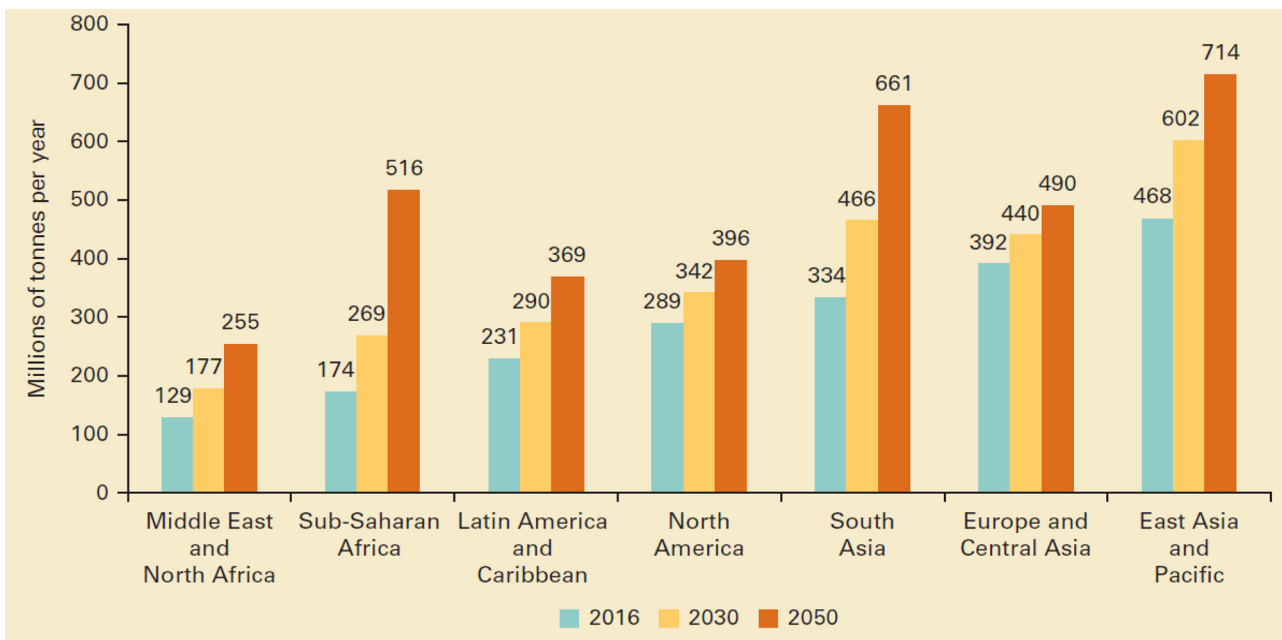
Figure 5: Amount of global waste materials generated in megatons (MT) per year as reported for 2016 and projected for 2030 and 2050 [7].

Table 1: Color classification test procedure conducted to group waste materials, adopted by earlier studies [63,64]

Classification	Color category	Color code based environmental effects	Examples
Eco-friendly waste	Gray	No significant risk to the environment	Food wastes, biomass, *etc.*
Biodegradable waste	White	Biodegradation >20%	Paper wastes, *etc.*
Non-biodegradable waste	Blue	Biodegradation <20%	Plastic, glass, metals, *etc.*
Toxic waste	Brown	Biodegradation <20% and toxicity <10 mg L^{-1}	Batteries, chemical additives, *etc.*

Application of toxic chemicals and additives are widely used in various industries such as oil and gas, electronics, and other energy related sectors [65]. Several authors have reported perilous environmental effects on marine life and humans on using chemical additives such as viscosifiers, lubricants, and surfactants [46,66,67]. Therefore, it is necessary to replace toxic and hazardous chemical additives and use natural and non-toxic materials [68]. In short, the use of eco-friendly and biodegradable materials in the industrial applications reduces the environmental impact and personal safety issues. One of the potential types is biowaste which falls under this category because it is both eco-friendly and biodegradable [69]. Other noteworthy types of waste include hazardous or toxic and non-biodegradable. Thus, biowastes hold great potential as an alternative of expensive and toxic additives in nanomaterials industry [70].

2.2 Recent applicability of waste materials

Accumulation of waste materials is a rising problem that not only threatens the environment but also the public safety. Devadoss *et al.* reported that Malaysia generated 13.9 MT of municipal solid wastes, heavily relying on landfilling for disposal [71]. According to recent surveys [72–74], more than 80% of municipal solid wastes were disposed in the landfills. To reduce the impact of this problem, these waste materials can be recycled and utilized in industries instead of disposing them.

For recycling of dangerous and hazardous wastes, special conditions and measures must be imposed to avoid mass catastrophes [75]. Furthermore, nuclear and radioactive materials must be strictly and completely disregarded from consideration for any types of recycling procedures [76,77]. Besides that, wastes of higher toxicity and complex handling are also unfavorable in any application. This is because waste materials could negatively affect the public health, safety, and the environment [78]. Thus, an increasing demand for the efficient transformation of wastes into benign nanomaterials has emerged to reduce the amount of waste discharged to the environment [79].

Lately, the increase in global concerns towards the sustainability of the Earth has increased public and industrial awareness on the detrimental effects of chemical toxic and non-biodegradable materials [80]. Strong efforts have been exercised to study several non-hazardous waste materials which are applicable in industrial applications such as food, agriculture and construction wastes [81]. Also, recycling of electronic waste has not been extensively adapted though it is a great approach to handle e-waste materials. The most common sources of e-waste include electronic toys, computer chips, circuit boards, batteries, wires, and motherboards [82,83]. Numerous studies have comprehended the recycling of waste materials by differentiating their sources and reutilization, conversion of these waste sources, implementation of disposal treatments, and fabrication into value-added nanomaterials. Finally, energetic applications and energy recovery has been suggested as well [84–86].

3 Graphene nanocomposites for industrial applications

There are versatile graphene materials that have been incorporated into industrially relevant composite matrices [87,26]. The two main categories are (i) chemically derived graphene materials and (ii) graphene particles obtained after physical exfoliation or physical deposition procedures [88–90]. Figure 6 illustrates a classification of commonly used top-down and bottom-up synthetic routes for graphene derivatives [43]. The chemical routes offer, at present, much cheaper and versatile opportunities to obtain the carbonaceous platelets in large quantities, which can serve a whole section of industry with at least a ton-scale/day production rate [91]. Physical exfoliation (scotch tape method and its developments), chemical vapor deposition or laser irradiation-assisted methods [92] cannot currently compete with this scale of production rate. For the chemical routes, the starting material is almost exclusively graphite oxide [93]. Graphite oxide is usually exfoliated in a liquid phase to produce single layers termed as GO, or at least
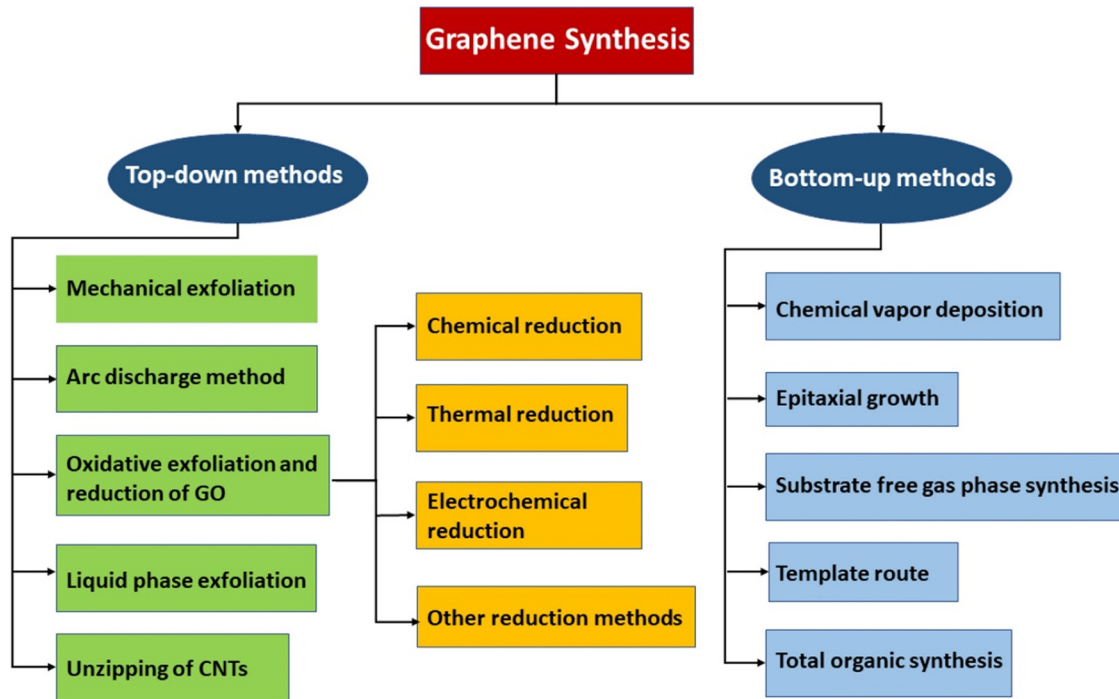
Figure 6: Classification of synthetic routes for graphene-based materials [43].

particles that are dispersed into a few-layered graphene oxide. Graphene-based particles in the final composites are then obtained usually by thermal decomposition of GO (to obtain chemically derived graphene) [94], or by chemical reduction (to obtain reduced graphene oxide, rGO). Other commonly derived forms of graphene include holey graphene, graphene nanoplatelets (GNPs), flower-shaped graphene/GO, graphene nanoribbons (GNRs), and GQDs, *etc.* [95,96]. The origin of high versatility of chemical properties and structural features is the composition of graphene-based composite materials.

Figure 7 shows the distribution diagram of the application fields of waste-generated graphene-based nanocomposites in which the columns are subdivided by the environmental impact of waste source used according to Bertram's classification [63]. The possible ways of reuse are also tabulated in more details in the Supporting Information. First, one finds that energetic nanocomposites are, at least currently, not among the most frequently studied ones. Supercapacitor and battery-related studies rank 4th and 5th and only a couple of papers focus on solar and fuel cell fabrication or heat storage. The most dominant fields are those which efficiently exploit the low electrical or mechanical percolation thresholds provided by the graphene platelets including studies related to structural materials used in construction (29 papers) and conductive materials (7 papers). Second, it is remarkable that most types of wastes are feasible and usable for the construction of composites to be further used in all sorts of applications.

With regard to the remarkable properties of graphene materials, much research has been done by incorporating graphene and its derivatives into nanocomposites to produce novel materials that make use the best properties and characteristics of each material present in these composites [97–99]. The outcomes of these research works have proven that graphene-based nanocomposites significantly improved the performance and functionality of the products, especially in terms of increased electrical conductivity, greater flexibility, thermal resistance, and stability [100,101]. Hence, in Section (3), we discuss the performance of graphene-based nanocomposites in industrial applications. Before this, however, we find it important to make the following remarks on the related terminology:

1) The term "graphene (based) nanocomposite," at least within the context of this review, refers to any composite material which contains highly dispersed (single or few-layered) graphene-like carbon platelets, and it is not restricted to systems in which the major counterpart of the composite (matrix) is graphene-based solid. In all cases, nevertheless, the observable properties of the composites are affected by the graphene content.

2) "Waste-derived" refers to any nanocomposites which include a counterpart obtained from a waste material.
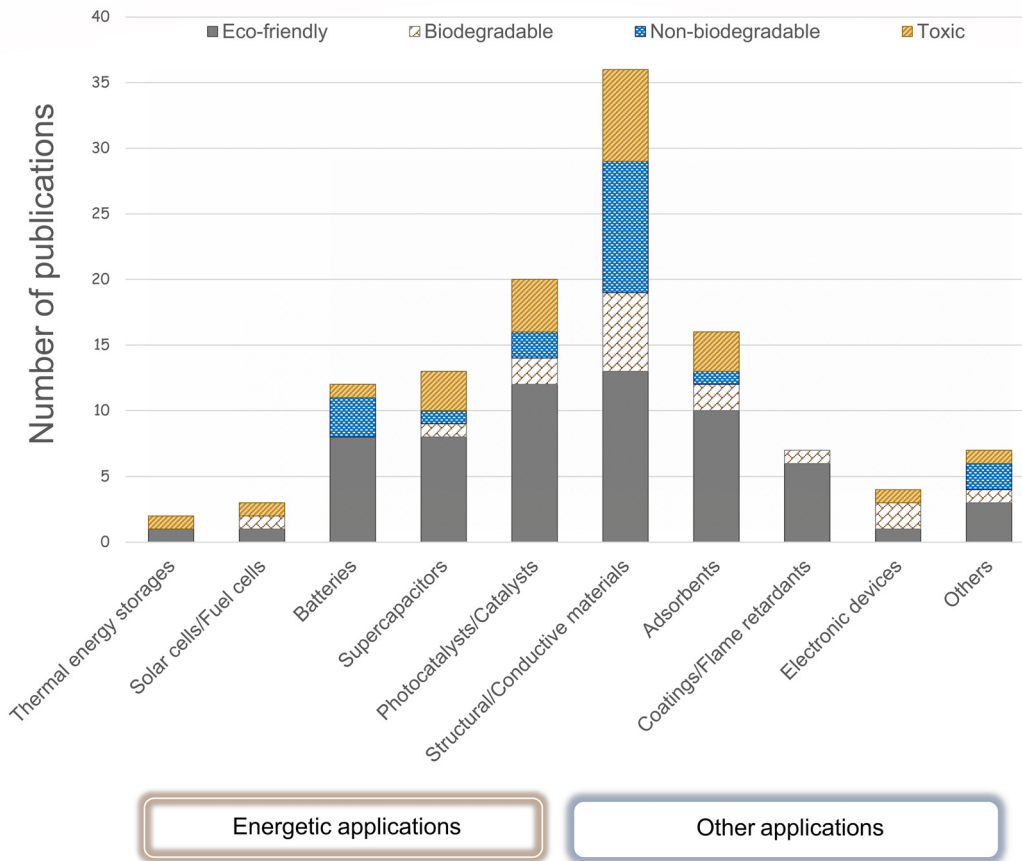
Figure 7: Waste-derived graphene composites in the current literature classified by their respective field of application.

Therefore, wastes may be the source of the carbonaceous phase (GO or derived carbons), which is coupled with a regular nanomaterial (*e.g.*, inorganic oxides) as reported in a few studies [224,237,331]. On the other hand, in an overwhelming majority of currently reported cases, the matrix of the nanocomposite originates from a waste source (*e.g.*, plastic) and graphene platelets (from any sources) are employed as high value-added fillers.

3.1 Energy production and storage

3.1.1 LIBs and sodium-ion batteries (SIBs)

The development of LIBs has been triggered by a surge demand in advanced energy storage due to environmental concerns. LIBs are rechargeable and they have high energy density and long cycle life which make them promising to be utilized as power sources for electric vehicles [102]. SIBs displayed similar electrochemical behavior to LIBs, due to the high availability of sodium

sources with remarkably low cost [103]. SIBs seem as potential candidates to be fully utilized as energy storage systems. The combination of metal oxides and graphene derivatives are promising anode materials and thus are heavily used in market leader products [104]. In particular, power sources based on 1D, 2D, and 3D graphene nanostructures are the best examples of the frontier devices for serving the new wave of medical devices, flexible screens, or wearable electronics [105]. However, the significant difference between the ionic radii and atomic weights of Li^+ and Na^+ are also seen in the transfer properties and phase stability which leads to lower energy density of SIBs. The favorable morphology of composite materials contributes greatly to the increase in overall capacity. This is most evident in the case of composites with metals [106]. Characteristics such as high cyclic stability, large surface area, high capacity, and low electrical resistance [107] are still the primary aspects to consider for energy storage performance, but, for *e.g.*, for flexible batteries, the mechanical and structural properties are also of utmost importance. Graphene-based matrices and coatings are able to enhance electrochemical characteristics

in different ways. First, the various nanostructures (GQDs, rGO, graphene films *etc.*,) may lead to increased active surface area, forming greater surface contact with the electrolyte [23]. These materials are also suitable to make electrodes with lower inactive weight and short diffusion path, which also improve the rate of charging/discharging processes. These coatings and composite matrices have improved the electron transfer processes, which also lead to better power density [108].

3.1.2 Supercapacitors

Supercapacitors (also known as ultracapacitors) are electrical energy storage devices that, unlike conventional solid dielectric-based capacitors, rely on electrochemical double layer capacitance and electrochemical pseudo capacitance which contribute to the total capacitance [109,110]. The important difference between conventional LIBs and supercapacitors is in the quantity of energy stored and the quantity of energy obtained per unit time. Batteries can store lot more energy than supercapacitors, but the latter ones can be charged/discharged much faster, so the development of supercapacitors has recently become a cutting-edge challenge. Especially, the automotive industry and the space agencies are trying to build them into the pioneering hybrid energy storage/ regeneration technologies. The electrodes of supercapacitors have been typically made of porous carbon, and graphene-based nanomaterials could provide a suitable solution to scale up the capacity of these devices as coatings or main components, because they can both act as matrix or filler in composite materials [111]. For instance, GO materials are versatile *en route* products to utilize electrode surface with graphene [112], rGO [113], or graphene-based hybrid nanomaterials [114], to improve electrochemical properties [115]. As the maximum energy storage highly depends on the specific surface area of carbonaceous materials hybridized with metal, metal oxide, and metal sulfide composites, they can be utilized as electrode materials of supercapacitors. Nonetheless, they have excellent electron transport features that enhance conductivity. They may also act as structural materials, because of their small weight and may also play the role of a bonding agent between the components in the composites. However, the direct covalent functionalization of graphene is tedious. Even a recent study shed a light for the applicability of GO to improve the energy storage capacity and power handling capability [116].

3.1.3 Solar cells

A solar cell or photovoltaic cell can be defined as a device that converts the sunlight directly into electricity [117]. In next generation, graphene-based solar cell materials attract serious attention of researchers, due to their good conducting properties, low costs, optical transparency, and lack of toxic environmental effects [118]. Perovskite solar cells (PSCs) are one of the widely used types of solar cells [119]. The power conversion efficiency (PCE) of the solar cells is directly dependent on the electrical conductivity of the matrix of the counter electrode. Graphene and rGO can play a crucial role in the improvement of their performance characteristics. Additionally, the presence of rGO in the composite can possibly prevent the aggregation of fillers or dopants helping to keep the electrically active sites intact during the synthesis process [189]. A few studies revealed that graphene-based nanocomposites could act as hole transport layer [120,121]. In some cases, they even outperformed the polymer-based composites in PSCs, which were proved as the best-known materials for hole transport. They also increased the crystallinity of a perovskite film, leading to reduced leakage current (which causes dropped performance) and decreased the so-called $J–V$ hysteresis, thereby allowing more actual determination of PCE. The optical bandgap of the composites is also a direct function of the degree of oxidation in the GO/rGO composites. In addition, graphene-based matrices have also excellent film forming capabilities, which makes them advantageous for the preparation of solar cells containing brittle and unstable semiconductor films [122]. In dye-sensitized solar cells, carbon-based nanostructures and nanocomposites have been used to improve their electrochemical parameters [123,124].

3.2 Chemical sensors

3.2.1 Biosensors

Biosensors with important features such as low price and rapid, highly sensitive, and greatly selective response have been used extensively for clinical diagnostics, metal ion speciation, and formation of batteries [88,125,126]. With today's technology advancement, the performance of biosensors has been greatly enhanced by graphene-based nanocomposites [127,128]. Graphene-based NPs are capable to act as biomolecule carriers because the selectivity of the device lies on the chemical properties of the immobilized biomolecules [129]. Moreover, their

good film-forming properties makes them suitable to be applied for biosensor fabrication. Graphene materials can be a suitable candidate as a precursor of biosensor membranes or probes. Although GO does not conduct electricity because of the presence of the various oxygen containing functionalities, it can be used as a flat-sheet surface to immobilize biomolecules. Moreover, functionalization by reduction is also able to restore the aromatic π electron system and bonds can be formed with carboxylic groups and N-terminal end on amino-acids or proteins, but epoxide or peroxide groups can also be easily functionalized by acid base or redox reactions. In biosensing, a direct connection leading to electron transfer between the active centrum of an enzyme and the carbon atoms plays key role in the process [130]. Biosensors can be grouped into the enzymatic, non-enzymatic and immunosensor categories [131]. According to the literature, various biomolecules, such as micro-RNA [132,133], nucleic acids [134,135], or biomarkers [136], can be detected with good selectivity by graphene-based biosensors. To enhance the electrochemical behavior, reduction of GO can be easily carried out under mild conditions, but even electrochemically reduced GO is widely used for biosensing.

3.2.2 Gas sensors

Gas sensor is an important device for detecting the variation in the concentration of toxic, or chemically active gases, for example to create safe work environment in industry, where precaution needs to be exercised to avoid air poisoning or to monitor the presence of explosive gases. Selective and sensitive gas detection at room temperature and a wide range of ambient humidity is needed for appropriate environmental monitoring [137]. Graphene-based materials have multiple advances in these devices. First, the presence of graphene in metal–graphene sensors can inhibit the oxidation of the metal component. Moreover, the high surface area to volume ratio is also beneficial and the metal–graphene system is also used to establish kinetic or mass transport selectivity [138]. A large number of studies have reported the applicability of graphene-based nanocomposites incorporated in gas sensors, but carbon nanotubes and nanofibers are also frequently examined [139–141]. Therefore, the most important class of graphene-based nanocomposite type gas sensors is that of electrochemical sensors [88,142]. The gas adsorption on the carbon-modified surface affects the electrochemical properties such as bandgap and charge transfer processes, that may provide a highly selective sensor membrane with remarkable signal/noise ratio [143].

Additionally, varying the structure, morphology, type, and amount of additives or degree of oxidation makes it possible to influence the selectivity. As it was demonstrated for biosensors, GO can also undergo surface functionalization schemes simply, in one step methods. Moreover, graphene nanocomposites make possible to detect wide range of toxic gases for example NO_x [144], CO [145], formaldehyde [146], or CO_2 [147]. Metal oxides or conducting polymers such as polyaniline (PANI) are commonly used fillers for the fabrication of graphene nanocomposites, which are found applicable for gas sensing [137,148,149]. The good film forming ability is also relevant, but the probe also can be formed by 3D (*i.e.*, foam-like) composites [150].

3.3 Corrosion protection

Enormous amount of money is spent on repairing the damage caused by corrosion. Therefore, a wealth of research projects is devoted to produce anti corrosion coatings with excellent properties to overcome the problem [151]. Recently, graphene nanocomposites have become potential candidates to deposit anti-corrosion coatings due to their high chemical stability [152,153]. Besides, graphene matrix is composed of sp^2 hybridized carbon atoms that are linked to each other in a 2D honeycomb lattice structure with high electron-density in its aromatic rings, which makes it impermeable to all molecules [154]. In the past few years, a lot of cheap and simple methods were developed for the mass production of graphene-based anti-corrosion coatings [155]. However, according to some of the latest reports, graphene materials can have a short-term corrosion protection effect but, conversely, corrosion promotion activity of the coating may occur in the long term. It is clearly stated that this phenomenon is related to the percolation threshold phenomena [154]. Above a certain surface coverage of the graphene particles, they even accelerate the corrosion process either due to the kinetic effects (the uncoated patches act as defects on the coating at which the oxidization becomes faster), or because of thermodynamic effects (decrease in the redox potential) [152,156]. It is also associated with the oxidation degree of GO/rGO, which restores the aromatic structure of graphene. In most cases, however, graphene nanocomposite coatings can prevent the formation of defects on the coating. Nonetheless, after the object was oxidized in the end of the process, the graphene coating helped in keeping the object in one piece, preventing the release of the corroded material to the environment [157]. Furthermore, some researchers stated that the rate of corrosion decreases with the increase in the number of graphene layers on the surface, [158,159]

and they did not report the corrosion promoting phenomena, *e.g.*, in the case of plastic/GO coatings. In addition, for monolayer coatings, there is a great importance of the perfect coverage, because structural defects may lead to increased corrosion of the substrate as compared to the bare materials [160]. In summary, the corrosion promotion activity of graphene-based nanocomposites is highly dependent on the quality of the non-carbonaceous component too. Graphene/GO is less suitable to form the matrix in composites, or to be applied only in its pristine state for corrosion protection.

3.4 Membrane processes

Graphene and its derivatives act as a perfect barrier due to high electron density of the aromatic rings that block any atoms or molecules from moving across it. Besides, graphene/GO fillers are the leaders in advanced composite membrane design studies in terms of mechanical properties. In the case of biocomposite membranes, GO loading can also enhance the hydrophilic interactions, dipole interactions, and hydrogen bonding at the same time [161]. In the past decades, water purification became a crucial problem especially for the treatment of municipal sewage or industrial wastewater. Membrane separation is a traditional method to clean the used industrial feed waters and by-product wastewaters [162]. Interestingly, due to their layered structure and special pore system including water permeation channels, pristine GO itself is capable to form membranes with advanced filtering features [163]. However graphene-based nanocomposites are also very competent candidates to beneficially influence the parameters of membranes, but pristine graphene and GO are also suitable to form membranes [164]. Nanocomposite membranes could be applied in a wide range of treatment processes. Among graphene nanocomposites, especially GO has been applied as filler, owing to its ability to keep the structural integrity of the membrane even at high pressures and could be used to form superhydrophilic membranes for increasing the transport rate of the water molecules [165]. For instance, oil-in-water emulsion separation [166] and toxic heavy metal ion removal by adsorption [167] are considerable potential applications of GO composite membranes. It is important to mention that composite membranes can be bestowed by anti-bacterial surface modifiers [168], as GO nanocomposites are extremely capable to immobilize almost any kind of substrates on their surface and the modified carbonaceous lamellae can be easily deposited on the membrane surfaces [169].

It is easily noticeable that polymer/graphene or polymer/GO systems represent the vast majority of carbonaceous composites used for membrane separation applications. The simple reason is that a wide range of polymers (both conducting or non-conducting) can be easily grafted or non-covalently attached to graphene materials in which graphene may act both as the matrix or the filler. Metal-like graphene composite coatings, metal oxide/graphene or GO hybrids, and polymer modified graphene materials and composites containing layered double hydroxides (LDH) and metal organic frameworks were also studied but less frequently [160].

In this part, we summarized the most popular fields of applications of graphene-based nanocomposites, but some of the fields mentioned below are not explained here in detail such as fire retardants or organic catalysts, adsorbent, or photocatalytically active composites. It is due to the minor fraction of literature papers related to waste-derived graphene-based materials in other general applications. Also, the focus of this study is centered towards energy related applications. Nevertheless, we will discuss these from the reusing and valorization point of view.

Without a possibility to give a comprehensive enumeration of all relevant papers in the specific areas, we tabulate representative publications on graphene nanocomposites used in diverse fields of industry, indicating the chemistry of the studied composite counterparts, the objectives of the study, and bullet points of its main findings (Table 2).

4 Recycling of waste materials into graphene nanocomposites

Despite the global COVID-19 pandemic, graphene industry is expected to grow due to increased demand for mobile devices such as smartphones and tablets, as well as opto-electronics and storage devices. While these electronic devices have numerous advantages, they also contribute to a major problem, which is the growing pile of e-waste. According to the World Economic Forum, 50 MT of e-waste are produced each year, but only 20% is recycled, with the remainder often incinerated or dumped in landfills [2–4]. More than just adding to landfill, most e-waste is made of non-biodegradable materials, and it may also contain materials that are potentially hazardous and toxic to our health and the environment if simply discarded. As a result, many problems associated with e-waste management are exacerbated [6,7,15]. To address this issue,

Table 2: Selected publications reporting on the utility of graphene nanocomposites for industrial applications

Applications	Nanocomposite counterparts	Objectives	Performance outcomes	Ref.
LIBs and SIBs	Chitosan/rGO	To enhance the capability rate of LIBs/SIBs	•Support high capacities in Li-ion system ($170\ mA\ hg^{-1}$) and Na-ion system ($80\ mA\ hg^{-1}$)	[170]
	2D GO/1D $LiMn_2O_4$	To improve the cathodic property of LIBs	•Exhibits high specific charge capacity and Coulombic efficiency (~98%) after 100 cycles •Stable structure after 100 cycles	[171]
	GQDs-doped $CuCo_2S_4$	As a supercapacitor electrode and the role of $CuCo_2S_4$ was established to support electrochemical cycling and ion diffusion	•Displayed specific capacitance of $1{,}725\ F\ g^{-1}$ under current density of $0.5\ A\ g^{-1}$ •Cycle life of 10,000 cycles while maintaining 90% of energy storage capability	[172]
	SnS_2/rGO	Carbon particles in nanocomposite helped to prevent SnS_2 agglomeration	•Improved electrochemical performance •Excellent rate capacity •Good cyclic stability	[173]
	V_2O_3/rGO	To investigate Li^+ storage behavior of V_2O_3/rGO	•Achieve high reversible specific capacity of: •$823.4\ mA\ hg^{-1}$ (current density:$0.1\ A\ g^{-1}$) •$407.3\ mA\ hg^{-1}$ (current density: $4.0\ A\ g^{-1}$)	[174]
	3D sandwiched-shape graphene/ $MnCo_2O_4$	As anode material of high-performance LIBs	•Enhanced rate capability •Remarkable cycle performance with high capability	[175]
	Nb_2O_5/rGO	As anode material of LIBs	•Good initial discharge-charge capacity •Capacity retention of $195\ mA\ hg^{-1}$ at $0.2\ A\ g^{-1}$ after 200 cycles and $98\ mA\ hg^{-1}$ at $2\ A\ g^{-1}$ after 1000 cycles	[176]
	$CuSi_2P_3$@Graphene	As anode material	•Great electrochemical performance •High reversible capability over 500 cycles •Long lasting cycling stability of $1{,}394\ mA\ hg^{-1}$ after 1,500 cycles at $2\ A\ g^{-1}$; $1{,}804\ mA\ hg^{-1}$ after 500 cycles at $200\ mA\ g^{-1}$	[177]
	SiOx@Fe_3O_4@FLG	Excellent electrochemical performance	•Outstanding performance of $1{,}550\ mA\ h\ cm^{-3}$ at $0.5\ A\ g^{-1}$ after 500 cycles	[178]
	$Zn_2Ti_3O_8$/rGO	An alternative energy storage device to fill the gap between LIBs and supercapacitors	•Specific capacity of $560\ mA\ hg^{-1}$ at $0.1\ A\ g^{-1}$ and $300\ mA\ hg^{-1}$ at $2.0\ A\ g^{-1}$ with superior rate capability	[179]
	Double shell Co_3Sn_2/ SnO_2@C@Graphene	Superior electrochemical performance as anode material	•Good sodium storage capacity •Reversible charge storage capacity of: •$744\ mA\ hg^{-1}$ after 100 cycles •$605\ mA\ hg^{-1}$ after 300 cycles	[180]
	rGO/Zn/polycarbazole	Supercapacitor device performances and electrochemical measurements were enhanced	•Enhanced the capacitance compared to the single NPs •High power of $442.5\ W\ kg^{-1}$ •Energy storage capabilities ($1.66\ W\ h\ kg^{-1}$	[181]

(Continued)

Table 2: *Continued*

Applications	Nanocomposite counterparts	Objectives	Performance outcomes	Ref.
Supercapacitors	PANI/sulfonated graphene	As negative electrode of the asymmetric supercapacitor	•Extended potential window •85% capacitance retention after 5,000 cycles •Satisfactory energy and power density	[182]
	Poly(3,4 ethylenedioxythiophene) polystyrene sulfonate/GO/glucose	As a novel electrode material for flexible supercapacitors	•Specific capacitance of 16 F g^{-1}	[183]
	N-doped graphene	As an electrode material for supercapacitors	•Higher specific capacitance compared to metal oxides •Better cyclic stability up to 1,000 cycles	[184]
	W$_{18}$O$_{49}$ nanowires-graphene (W$_{18}$O$_{49}$ NWs-rGO//rGO)	As an electrode material for supercapacitors	•Showed pseudocapacitive behavior with: •High specific capacitance •Good rate capability •Good cyclic stability •W$_{18}$O$_{49}$ NWs-rGO/rGO supercapacitor shows energy density of 28.5 W h kg^{-1}	[185]
	Graphene/Ni$_3$V$_2$O$_8$	As electrode material for supercapacitors	•Electrochemical performance better than pristine Ni$_3$V$_2$O$_8$ •Good specific capacitance •Good energy density •Reasonable cyclic stability •Recorded Coulombic efficiency of 83% after 3,000 cycles	[186,187]
	Multivalent VO$_x$/Graphene	As electrode for high-energy-density symmetric supercapacitors	•Obtained high three-electrode specific capacitance of 1,110 F g^{-1} •High energy density of 54 W h kg^{-1} with virtually no capacitance loss after 20,000 cycles •Maintaining 92% energy storage capability after 20,000 cycles	[115]
Solar cells	MoS$_2$/graphene	Fabrication of nanocomposite utilized as a photoanode for dye-sensitized solar cells	•Nanocomposite showed tremendous photo-conversion efficiency of 8.92%	[188]
	MoS$_2$ and N-doped GQDs anchored rGO	Nanocomposites have been fabricated for dye-sensitized solar cells (DSSCs) due to their superior electrochemical properties and high chemical stability	•The DSSCs assembled with fabricated nanocomposites and their photovoltaic power conversion efficiency (η) was •2.01% for •3.92% for MoS$_2$@rGO •3.53% for N-GQDs@MoS$_2$ •4.65% for N-GQDs@MoS$_2$@rGO	[189]
	ZnO/GO	For remediation of MB dye	•33% faster decolorization of 20 mg/L of MB dye	[190]
	TiO$_2$–GO	Improved efficacy of solar cells		[191,192]

(Continued)

Table 2: *Continued*

Applications	Nanocomposite counterparts	Objectives	Performance outcomes	Ref.
Electrochemical biosensors	Copper-carboxylated graphene	To determine electrocatalytic activity for glucose oxidation	• The initial band gap of the TiO_2 showed a significant decrease to 1.81–1.40 eV after incorporation of GO in various amounts	[193]
	CNTs/rGO/BiVO$_4$	Non-enzymatic sensor as it proved that CNTs/rGO/BiVO$_4$ electrode is the best photoelectrochemical electrode for non-enzymatic glucose detection	• Low limit of detection of 7.96 nM • Rapid response time of 2 s and wide linear response range from 0.1 µM to 5.48 mM • High sensitivity of 1,142 µA mM^{-1} cm^{-2} and good selectivity	[194]
	rGO/functionalized-ordered mesoporous carbon	Selective and sensitive electrochemical genobiosensor	• Excellent electrocatalytic activity for providing maximum sensitivity of 501.5 mA cm^{-2} mM^{-1} and good stability with negligible current response	[195]
	Nickle NPs-modified graphene-based GCE (NiNPs/Nafion/graphene/GCE)	Biosensor for sensitive nonenzymatic glucose detection	• Long term stability (21 days) with linear range (0.1–3 µM) • High sensitivity of 0.0188 mA µM^{-1} • Limit of detection: 120 nM • High sensitivity recorded with a linear range of 1–200 µM • Sensitivity of 2,848.6 µA in linear range of 200–10,800 µM: • At concentration <200 µM: • Obtained linear range • Limit of detection: 0.6 µM • Response time: 1–1.5 s • Good stability • Excellent electrical properties	[196]
Gas sensors	rGO–CuFe$_2$O$_4$	High performance H$_2$ gas sensor	• Able to detect H$_2$ at low concentration of ppm at room temperature	[197,198]
	Bi$_2$O$_3$–GO	Ammonia gas sensor imposed superior sensing performance, including quick response and recovery of the Bi$_2$O$_3$–GO nanocomposite sensor	• Bi$_2$O$_3$–GO nanocomposite sensor at 50 ppm NH$_3$ in just 14 s operation duration was ~81.23, which is improved 25-fold and 13-fold compared to pristine GO sensors	[199]
	3D TiO$_2$/graphene-CNT	Highly selective and fast room temperature gas sensing	• Highly selective sensing in VOC mixtures • Good selectivity in the range of 50–500 ppm • Excellent response time in 500 ppm and still satisfying in the range of 100 ppm	[200]
Anti-corrosion coatings	Nickel–graphene	Anti-corrosion for mild steel	• Superior anti-corrosion performance at current density of 9 A m^{-2}	[151]
	GO@CSA–PANI			[201]

Table 2: *Continued*

Applications	Nanocomposite counterparts	Objectives	Performance outcomes	Ref.
		Fabrication of a promising nanomaterial for adsorption and coatings protection	•Good absorption properties of electromagnetic wave •High corrosion resistance •Minimum reflection loss of −48.1 dB and 5.3 GHz with a 2.4 mm matching thickness •Excellent corrosion protection capability (97.43%) after 168 h	[202]
	GO/HAP	As anti-corrosion pigment	•Excellent anti-corrosion performance •The impedance increased by 754.4% due to synergistic effect of the nanocomposite	[203]
	GO–polymer	Nanocomposite membrane for reverse osmosis	•Good water transport properties •An anti-swelling tendency •Stable under severe oxidizing environment	
Membranes	PMMA-*co*-MA-PEG/PC/GNRs	Gas separation membrane	•Due to molecular sieving; •Permselectivity increased by 51.6% •Permeability decreased by 24.7%	[204]
	Functionalized GO/GQDs	Highly efficient reverse osmosis	•Nanocomposite membrane performed remarkably high water permeance up to 5.89 L m⁻² h⁻¹ bar⁻¹ •Rejection ratio of 97.1%	[205]
	Polyvinylidene fluoride functionalized PANI–GO (PVDF–PANI–GO)	Nano-filtration membrane for textile effluents removal	•Nanocomposites membrane with 0.1%w/v GO established a maximum of 98% dye rejection at 0.1 MPa •Recovery ratio reached about 94%	[206]

Note: LIBs, lithium-ion batteries; SIBs, sodium-ion batteries; rGO, reduced graphene oxide; GQDs, graphene quantum dots; FLG, few layered graphene; PANI, polyaniline; GCE, glassy carbon electrode; CSA, camphor sulfonic acid; HAP, hydroxyapatite; PMMA-*co*-MA-PEG/PC/GNR, poly(methyl methacrylate-*co*-methacrylic amide)-polyethylene glycol/polycarbonate and graphene-based nanoribbons.
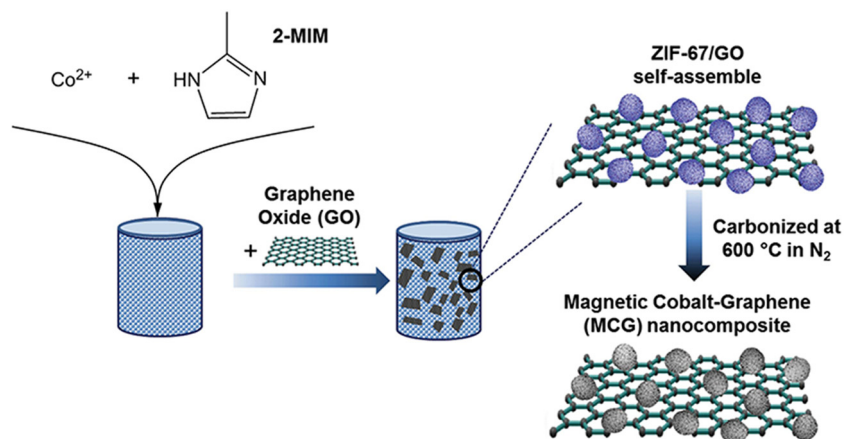
Figure 8: Schematic diagram of synthesis of magnetic cobalt–graphene (MCG) nanocomposite [208].

agricultural waste could be used to create biodegradable materials, which could then be used to make electronic devices. After their lifetime (sometimes rather limited), these obsolete or unserviceable devices (or at least some of their parts) could then be easily degraded upon their disposal. This lays the groundwork for circular economy systems in the electronics industry, increasing their sustainability [61,71,72]. To ensure the understanding of waste-derived nanocomposites fabrication, Figure 8 highlights the GO assembling of cobalt-based metal framework into GO–Co nanocomposite.

4.1 Processing factors of graphene nanocomposites

GO is an oxidized form of graphene that has widespread use as a catalyst support, composite filler, or the precursor of high-surface-area carbon materials [26]. Its benefit as a precursor of processable aqueous suspensions of carbon nanoplatelets is also increasingly realized and exploited, leading to various products of electronics and energy conversion. The presence of oxygenated groups and aromatic domains on the GO structure resulted in excellent organic molecule adsorptive capabilities [39]. However, due to the unavoidable van der Waals interaction, GO layers may agglomerate in aqueous medium, especially at higher salinities or in the solutions of other electrolytes, already at fairly low concentrations [207,208]. As a result, adsorption capacity is reduced, making the recovery process more difficult. One way to prevent the aggregation of GO is to formulate a composite with activated carbon (AC) or other materials conjugated with GO by using waste materials as carbon sources. This will significantly improve the adsorption efficiency due to the

synergism between the porous structures of the two carbon materials [208,209].

A variety of methods for the processing of graphene nanocomposites have been discussed in earlier studies and are not focus of our work. Examples of dedicated publications for nanocomposite formulation are: covalent or electrostatic functionalization by Dreyer *et al.* [18], silanization by Zou *et al.* [99], mechanical mixing by Sun *et al.* [210], functionalization with NPs or *in situ* polymerization by Chen *et al.* [211], utilizing polymers as nanofillers by Ramanathan *et al.* [90], using few layered graphene materials with polymers by Young *et al.* [212], combining multi-layered and monolayered graphene materials with various nanofillers by Shahil *et al.* [213], and utilizing epoxy matrix with graphene nanoplatelets by Monti *et al.* [214].

Most of these studies have discussed a wide series of matrices, fillers, and NPs based on their suitability and application field [125,167]. For the preparation of the composites, melt processing, solution blending, *in situ* polymerization, and sol gel procedures are the most commonly used lab-scale methods relying on van der Waals interactions or the creation of covalent bonds between base material matrices and NPs [90]. Based on the process parameters and the particle size and shape of the component phases, different morphologies can be attained [210]. Platelets, isodimensional or elongated particles are largely used types of NPs. Well dispersed, percolating, and agglomerating are typical arrangements for isodimensional and elongated NPs [53,212,215]. However, micro composite formation, intercalation, and exfoliation offer a broad variety of microstructural arrangements to fabricate nanocomposites using platelet-shaped NPs. Here micro composite refers to inefficient matrix affinity and particles distance [170,172]. Also, intercalated and exfoliated nanocomposites

are noteworthy for their increased interlayer distance and homogeneous filler dispersion in the polymeric matrices, respectively [216]. Figure 9 displays the synthesis mechanism including dispersion, pyrolysis, and oxidization into cobalt oxide/GO nanocomposite.

To fabricate stable waste-derived graphene nanocomposites, a good dispersion of fillers is critical to maintain their properties [46]. Table 3 entails the role of various waste-derivatives into graphene nanocomposites using a variety of matrix, fillers and excellent outcomes for energy related device transformations.

Numerous types of waste materials such as agricultural and other biowastes, steel waste, plastic, paper, and other industrial wastes have shown ability to form stable composites with carbon-based materials by changing their structural morphologies [217,222,227,229]. Recycling of these waste precursors into nanomaterials using several methods have been highlighted repeatedly [215,218]. In order to get the desired morphology of graphene nanocomposites, graphene-based NPs play a vital role which enable interaction with the base matrices [220]. Nevertheless, not all groups of graphene family are successful to get the preferred structures [232,233].

When graphene is used as a filling agent in composites, it usually results in an improvement of their electrical, mechanical, and thermal properties. Consequently, the nanocomposite can perform multiple functions when used in any system. To synthesize GO based nanocomposites, appropriate exfoliation of graphene or GO and their
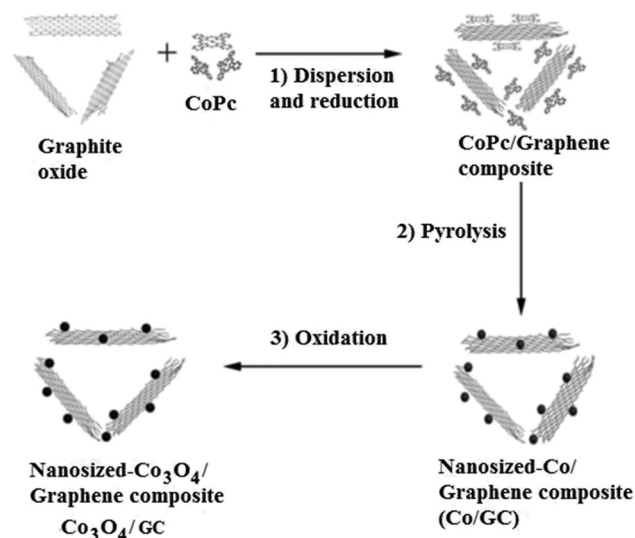
restacking/aggregation are issues to be resolved [235,236]. Conclusively, poor dispersion negatively affects the performance in most kinds of applications. However, greater levels of dispersion can be achieved for GO as compared to graphene due to its oxygenated groups which enables its easy incorporation into water-soluble matrices [238]. Based on recent research works, accommodation of graphene and GO fillers into various types of polymers has been done using *in situ* polymerization, solution intercalation, electrospinning, wet spinning, drop casting, melt intercalation, and freeze-drying procedures [240,241].

A remarkable waste-generated graphene nanocomposite was introduced through keratin wool waste into electronic devices. Energetic nanocomposites were developed by tuning conductivity of nanofillers among GNPs. In order to get homogenous dispersion of GNPs with waste proteins, carbon particles were 100–150 μm thick. By adding 5 wt% of GNPs, the resistance released five folds from 1,010 Ω sq^{-1} to ~105 Ω sq^{-1}, and as low as ≈10 Ω sq^{-1} by adding 30 wt% of GNPs. Moreover, capacitors were fabricated using 1 cm^2 waste keratin with excellent impedance, dielectric properties, and capacitance of 50–550 pF from −85 to 87°. Moreover, this keratin-based graphene nanocomposite was successfully used to develop an insulator with soft and flexible texture with no cracks at ambient temperature [217].

In a recent study, coconut waste has been converted into rGO nanocomposites with high surface area and electrochemical properties for supercapacitors. In its fabrication, ferrocene was used as a catalyst under 300°C opposite to the conventional techniques in which it has been crucial to remove carbon basal plane defects as a strong reducing solvent. The effects of temperature on the oxygen content and band gap were evaluated. It was observed that the increase in the temperature decreased the amount of oxygenated groups in rGO with high amount of defects in rGO lattice structure. Thus, successful efforts were made to recycle defect free rGO nanocomposites from coconut waste [218].

Similarly, an economical and facile approach of coffee waste-derived GO nanocomposites has been introduced for lithium-ion capacitors. Parameters such as particle size, matrix loading, and conductivity for battery electrode and pore size and surface area for capacitor electrode were optimized. It was observed that ball milling method induced a prominent reduction in particle size of 2–5 μm. The incorporation of rGO into the carbon matrix led to significant increase in the overall capacitance of lithium ion capacitor (40–70%) on different applied current rates. This state-of-the-art waste-derived energetic graphene nanocomposite enhanced power and capacitance for up to 80% after

Figure 9: Synthesis of cobalt–graphene nanocomposite (Co/GC) through (1) dispersion of graphite oxide and CoPc using chemical reduction, (2) pyrolysis at the temperature of 800°C, and (3) further oxidation to fabricate nanocomposite at 400°C [208].

Table 3: Summary of waste-generated nanocomposites according to their particle types, sizes, and chemical processing

Type of particle/waste	Matrix	Fabrication process	Filler %	Particle size	Remarks	Ref.
Keratin/waste wool	PDMS	Ink drop casting (Keratin + GNPs)	GNPs 30%	100–150 µm thick films	•Novel transformation of waste wool into electronic devices	[217]
Coconut coir and coconut shell	rGO electrode	Catalytic synthesis with ferrocene	—	—	•Very stable supercapacitors from biomass with high specific capacitance (111.1 F g^{-1} and 60.2 F g^{-1}) •Cyclic stability (99% after 300 cycles)	[218]
Si@rGO/silicon waste sludge	Graphene shell	Ultrasonic-assisted atomization and spray pyrolysis	Si spheres with size ~0.2 µm	0.6–0.8 µm	•One-step synthesis •Electrode in LIBs with high specific capacity (1,626 mA h g^{-1}), stability, and Coulombic efficiency (63.89%)	[219]
Fe$_3$O$_4$/chitosan/graphene/waste batteries	Fe$_3$O$_4$/chitosan	Simple electrochemical method, co-precipitation	Graphene ~54%	—	•Simple and direct electrochemical synthesis method •The form of composite was Fe$_3$O$_4$/chitosan/graphene precipitate •Efficient adsorbent of MB (q_e = 47.35 mg g^{-1}) prepared with co-precipitation method	[220]
Lignin@GO/lignin waste	PVA	Solvent casting method	Alkali lignin – GO hybrid (4:1) nanofiller 4%	—	•Enhanced thermal stability •Synergetic effect on Young's modulus and tensile strength	[221]
BiVO$_4$@cellulose/fruit peels	BiVO$_4$	Direct hydrothermal synthesis	Cellulose – various	24–35 nm	•Good photocatalytic activity with high photodegradation efficiency (87%)	[222]
Graphene@polypropylene/tire waste	Polypropylene homo- and copolymer	Melt mixing	GNPs 1–5 wt%	50 nm sized GNPs in the matrix	•Significantly increased mechanical properties: flexural strength, tensile strength, thermal stability, mechanically beneficial crystal structure	[223]
Fe$_3$O$_4$@C/coconut shell waste	Carbon material similar to GO	Direct hydrothermal synthesis	Fe$_3$O$_4$	Nanoporous material with 5–60 nm pore size distribution	•One-step synthesis method •Magnetic nanocomposite with good magnetic performance •Enhanced surface area •Beneficial adsorption properties (removal efficiency on MB > 90%)	[224]
Polyethylene@graphene/recycled high density polyethylene	Polyethylene	From recycled PE mixing, sonication, and evaporation of the	GNPs (0–8 wt%)	—	•Increased hydrophobicity and enhanced mechanical properties •Slightly increased thermal and mechanical conductivity	[225]

(*Continued*)

Table 3: *Continued*

Type of particle/waste	Matrix	Fabrication process	Filler %	Particle size	Remarks	Ref.
Wood-hydrolysates/mont-morillonite@rGO or GO/forestry waste	Wood-hydrolysates/montmorillonite	solvent, injection molding Hydrothermal treatment of wood chips, mixing and stirring, reduction with hydroiodic acid	GO or rGO (0.3 –2 wt%)	Few microns thick film	•Peak heat release rate reduced with more than 90% compared to that of neat wood hydrolysate •In the case of WH–MMT–rGO good hydrophobicity (88° contact angle) •Superior mechanical properties (~124 MPa maximum tensile strength)	[226]
rGO/MnO$_x$ nanocomposite/ Manganese waste	rGO	Immobilization of manganese compounds on GO, freeze-drying, anneal treatment	MnO$_x$	MnO$_x$ nanospheres with a size of 10–90 nm on rGO from "ultralarge" GO nanosheets 50–90 μm thick film on the electrode	•Coatings of symmetric supercapacitor electrodes •Extremely good capacitance retention after 20,000 cycles (96,6%) •Superior energy storage/conversion performance	[227]
rGO@Au NPs/peanut peel waste	rGO	Mixing and stirring the precursors, free drying	Gold nanoparticles (Au-NPs) 23.29%	Avg. size ~660 nm	•Peanut peel waste catalyzed the formation of the nanocomposite •Efficient removal for MB and MG dyes (~93%) •Good catalytic reduction of 4-aminophenol and 4-nitrophenol	[228]
Graphene–tin oxide nanocomposite/wood and coconut shell charcoal	Wood derived: graphene nanosheets coconut derived: aggregated carbon nanodots and Sn nanorods	One-step hydrothermal treatment	Wood derived: SnO$_2$ nanodots with uniform distribution	Wood derived GTO: SnO$_2$ < 3 nm nanodots on graphene sheets coconut derived GTO: aggregated network of carbon nanodots (5–11 nm) and Sn nanorods (length 20–34 nm)	•Eco-friendly one-step synthesis without hazardous chemicals •One of the highest reported antibacterial activities of its kind (wood derived: 27 ± 1.0 mm, coconut derived: 38 ± 0.7 mm inhibition zone) •Good colloidal stability	[229]
MnO$_2$/GO and MnO$_2$/rGO/spent battery powder	GO or rGO	Mixing, sonicating, stirring	Spent battery powder (MnO$_2$): various mass ratios	20–100 nm	•Excellent cyclic stability (100% after 1,000 cycles) •Significant improvement in the specific capacitance depending on matrix-filler ratio (best on 1:5) 116 F g^{-1} SBP/GO and 150 F g^{-1} SBP/ rGO (two times higher than the original batteries)	[230]

(Continued)

Table 3: *Continued*

Type of particle/waste	Matrix	Fabrication process	Filler %	Particle size	Remarks	Ref.
Twisted graphene coconut husk ash/coconut husk ash	Twisted graphene	Ball milling, washing	Coconut husk ash (100:1,50:1,25:1) CHA: graphene	—	•Simple and mass synthesis •Excellent yield of Biginelli reaction (best 94%) •Solvent free synthesis •Good catalytic stability (80% yield after 6 cycles)	[231]
CNTs-HAP/LLDPE and GO/HAP/LLDPE/waste eggshells	LLDPE	Melt mixing	CNTs–HAP or GO–HAP (1–5 wt%)	HAP particles 50–200 nm	•Increased hardness (best 34%) tensile strength, impact strength, lowering flammability (best 31%) related to the amount of filler	[232]
Tree gum–GO/Tree gum waste	Gum Arabic, gum karaya, gum kondagugu	Solution casting method	GO (0.5, 1.0 wt%)	—	•Utilized as gas barriers with very low oxygen transmission rate (lowest 13.9 cm^3 atm^{-1} day^{-1} m^{-2} •Increasing Young's modulus and tensile strength (68 and 47% best, respectively) •Good thermal stability •Biodegradable	[233]
rGO/TiO$_2$/tea waste	rGO	Hydrothermal method	TiO$_2$ different ratios: 1:4–1:10	—	•Good photocatalytic activity in degradation of wastewater pollutants (methyl orange)	[234]
Lignin–GO–ZnO/palm oil waste	Lignin, GO	Solvothermal method	ZnO (29% Zn(ii) content)	100–200 nm irregular aggregates (ZnO NPs ~100 nm)	•Applicability to remove heavy metal ions from wastewater (Cd(ii) 90%) •Relatively high stability and high thermal stability	[235]
HrGO, SGO, MSG/PET bottle waste	Highly reduced rGO, sulfonated GO, magnetically modified GO	Activation in electric furnace in high T, sulfonating, co-precipitating with Fe$_3$O$_4$	—	—	•Outstanding current density (135.96 mA m^{-2}) •Highest voltage (191 ± 4 mV) •Relatively high mechanical strength •Simple synthesis method using a usual waste (PET) •High amount of adsorbed material for antibiotics (170–236 mg g^{-1}) •Outstanding removal efficiency (99.94% ampicillin, SG)	[236]
CEPG–PANI–Ag/coffee waste	CEPG	Simple synthesis with mixing and stirring in	PANI and Ag	—	•Easy removal of spent adsorbent caused by the magnetic modification	[237]

Table 3: *Continued*

Type of particle/waste	Matrix	Fabrication process	Filler %	Particle size	Remarks	Ref.
		acidic media after extraction		~100 nm avg. pore size with 10–30 nm Ag particles on the surface	• High surface area: 1033.65 m^2 g^{-1}, specific capacity: 1,428 C g^{-1} at 5 mV s^{-1} • Good cyclic stability (94.5% after 5,000 cycles)	[238]
n-Nonadecane–rGO–AC/palm oil waste	rGO/AC	Simple impregnation process	n-Nonadecane (10–90 wt%)	—	• Efficient thermal stability • High phase change latent heat values in the 25–40°C range • High thermal energy storing capability due to encapsulation of n-nonadecane	
ZnO–GO/metallurgic waste	GO	Co-precipitation + annealing	ZnO (other metal ion as contaminants)	—	• Excellent photodegradation efficiency of the azo dye AO$_7$ was found which was significantly higher when waste steel leachate derived zinc precursor was used instead of pure zinc oxide	[239]
CFA–GO–WO$_3$NR/coal fly ash waste	CFA	Hydrothermal	GO–tungsten nanorods	—	• Efficient removal agent of lead ions (max. adsorption capacity 41.51 mg g^{-1}) • Outstanding photocatalytic activity spent adsorbent (original composite + Pb^{2+} in the degradation of acetaminophen 93%	[240]
Polyurethane GO–HAP/PET waste	Polyurethane	*In situ* GO–Hap synthesis + polymerization + SCPL method	GO–HAP (0,1,2,5,10 wt%)	Porous scaffold 0.5–1 µm GO sheets decorated with 10–30 nm HAP platelets of avg. pore size: 236–266 µm	• Remarkable antibacterial activity against Gram-positive and Gram-negative bacteria • Improved mechanical parameters (Young's modulus, tensile strength, flexural strength, hardness, compressive modulus, elongation) • Potential bone tissue engineering applications	[241]

Note: PDMS, polydimethylsiloxane; GNPs, graphene nanoplates; PVA, polyvinylalcohol; rGO, reduced graphene oxide; NPs, nanoparticles; LLDPE, linear low-density polyethylene; CEPG, coffee extracted porous graphene; CFA, coal fly ash waste; SCPL, solvent casting particulate leaching.

3,000 cycles which further heightened to 15,000 cycles [242].

In an interesting study, researchers reported tire waste recycled nanocomposite fabricated using poly (lactic acid) (PLA) and GNPs to accelerate mechanical properties. The energetic nanocomposite was prepared through twin-screw extruder using a die press of 0.5 mm thickness and 25 mm width. It was found that incorporation of GNPs (1, 2, and 3 wt%) in tire waste/PLA blend enhanced the tensile properties. Morphological analysis through scanning electron microscopy (SEM) presented that GNP nanocomposite reduced the formation rate and pore volume. In addition, GNP nanocomposite increased thermal stability and mechanical properties due to heat deformation. Adhesion of PLA with tire waste was not homogenous due to polymeric contents as the surface was heterogeneous and the polar macromolecule of PLA could not interact strongly with the hydrophobic parts of the carbonaceous nanosheets. Thus, functionalization through GNP fillers provided an ideal homogenous dispersion with tire waste (70 wt%) and PLA (30 wt%) nanocomposite. However, the dispersibility of GNPs was limited which paved a way for further research works [243].

Furthermore, sludge waste consisting of Si (40%) has been utilized to convert graphene nanocomposite for LIBs. Recycling of Si from waste was introduced using one-step aerosol method which was also recycled into Si NPs (78%) with SiC mass fraction of 3.2 wt%. Further, ultrasonic atomization spray pyrolysis was deployed to produce Si–graphene nanocomposite as efficient anode material for batteries. As per morphological analysis, the developed Si–graphene nanocomposite turned into a crumpled paper ball with the average size range of 0.6–0.8 μm using different parameters. No variation was found using FESEM technique in the structural analysis. However, XRD showed high peaks of Si and SiC as graphene content was lower compared to Si. By increasing the temperature from 300 to 500°C, the Si–graphene nanocomposite size remained the same (0.66 μm). It was observed that the size distribution of this energetic nanocomposite was homogeneous as it aggregated in the absence of graphene and thus, recovered by aerosol method where the particles were spherical with 0.47 μm diameter. The electrochemical analysis was performed which revealed highly efficient presentation as anode materials in terms of capacity, Coulombic efficiency, and stability for LIBs [219].

The significance of energetic graphene nanocomposites has also been signified by recycling of coffee waste. Coffee waste-derived GO was doped using a polymer (PANI) and silver NPs to fabricate nitrogen doped coffee waste extracted porous graphene (CEPG-PANI-Ag) nanocomposite.

Development of this novel nanocomposite improved electrochemical properties by increasing the electrode surface, porosity, and high surface area of $1033 \, m^2 \, g^{-1}$ for electrons mobility due to combined functionality through nitrogen and oxygenated groups on graphene nanostructure. Morphological analysis through XRD, FESEM, and TEM confirmed the crystalline structure of AgNPs, a reflection at 22.6° diffraction angle, characteristic of graphene, broad peaks of PANI chains, and about ~200 nm size of CEPG-PANI-Ag with homogeneous dispersion. For energetic application, it was demonstrated that developed nanocomposite displayed a very high specific capacity of $1,428 \, C \, g^{-1}$ at $5 \, mV \, s^{-1}$ scan rate and 5,000 cycles stability. This study presented an outstanding example of biowaste-derived energetic graphene nanocomposite for high performance rechargeable batteries [237].

Newly, oil palm waste-derived AC doped with rGO has been developed as a nanocomposite for thermal energy storage. It showed advances of surface area of $680 \, m^2 \, g^{-1}$ and an average pore size of 22 Å. A fruitful intrusion of *n*-nonadecane into the developed nanocomposite was completed and their nitrogen gas adsorption-desorption isotherms were determined. There was no leakage observed during this phase alteration as it was homogeneously dispersed from solid to liquid phase in graphene matrix. This novel, eco-friendly, and thermally stable waste-generated graphene nanocomposite has been introduced for thermal energy storage applications [238].

Likewise, oil palm waste-derived lignin GO (L-GO) was doped with ZnO using 39% carbon and 29% Zn to prepare L-GO-ZnO energetic nanocomposite. In this recent work, biowaste-generated anode material was prepared which was utilized for removal of Cd(II) substrates from wastewater through microbial fuel cells. Self-assembled L-GO was combined with ZnO NPs which enhanced physicochemical properties such as great energy output of $135.96 \, mA \, m^{-2}$ which is eight times greater than typical GO anodes. It further evidenced the removal of Cd(II) from wastewater by using microbial fuel cells *via* waste-generated anodes. It was reported that by changing the morphological characteristics of the designed graphene nanocomposite, 90% of removal efficiency was achieved [235].

In addition, value-added morphological impacts of other remarkable waste-derivatives, such as human hair [244], sisal fibers [245], *Prosopis juliflora* shrubs [246,247], orange peels [248], olive pits [249], green tea waste [250], Indian cake husk [251], peanut hull, bagasse, corn stalk, potato waste, defatted soybean, tobacco rods, fermented rice, prawn shells, and onion husks [252], have been thoroughly evaluated for energetic applications and electronic devices. Interestingly, most of these wastes have been

converted into AC and graphene flakes, rather than graphene nanocomposites.

4.2 Applications of waste-generated graphene nanocomposites

The production of nanomaterials from biomass has been viewed as a bio-based economy and a green solution to local and global pollution issues. Since graphene-based technology is efficient, cost-effective, simple, and environmentally friendly, it has become the solution to today's sustainability issues [41,55]. For example, agriculture waste biomass is a low-cost source for nanomaterial synthesis, and because of its abundance as well as various studies on it, mass production of graphene-derived nanocomposites is possible [234,237,253].

Due to graphene's excellent in-plane properties, new opportunities for tailoring nanocomposites have emerged. Graphene/GO/rGO bio-nanocomposites have a potential to be applied in various fields including adsorption [254], separation of organic and inorganic compounds [255], photocatalytic degradation of organic matters [256], disinfection and antimicrobial compounds [257], hydrogen storage and production [258], and electrochemical biosensors [259]. Therefore, lots of research are now focusing on the strengthening/toughening, electrical conduction, thermal transportation, and photothermal energy conversion of graphene/polymer nanocomposites as well as studying the effect of graphene configuration – layer number, defects, and lateral size, on both intrinsic graphene and graphene/polymer nanocomposites properties [260,261]. A variety of waste sources and their utilization for favorable applications are presented in Figure 10.

Recent studies have shown significant impact of waste-derived graphene nanocomposites among industrial applications. For instance, Rezania *et al.* [263] utilized waste frying oil to synthesize biodiesel using magnetic GO doped mixed metal oxide MGO@MMO nanocomposite. They studied the effect of several experimental parameters such as material amount, methanol-to-oil ratio, temperature, and time. They observed that the optimized parameters were as 94% of yield, 8:1 ratio, and 60°C with the reaction time of 90 min, respectively.

Other recent studies include utilization of steel sludge waste in ZnO–GO nanocomposite for improved catalytic efficiency [239], oil palm biomass derived GO nanocomposites as an effective medium for energy transportation through microbial fuel cells [235] or for thermal energy storage [238]. Coffee waste-derived [242], coconut shell and grapes seed derived [218,264] rGO containing composites find use in supercapacitors, while Bengal gram bean husk derived graphene-like porous carbon nanostructure is applicable for improved adsorption of organic dyes [265]. Additionally, the impact of GO nanocomposite has been evaluated on carbon conversion and methane production using anaerobic digestion of waste activated sludge [266]. Table 4 represents the summary of waste-generated graphene nanocomposites for significant industrial applications and their impacts.

Fabrication of energetic nanocomposites by incorporating nanodimensional phases into bulk matrices can lead to novel structural features and very advantageous properties. Among them, improved thermal stability, mechanical properties, high surface area, electrical stability, and enhanced optoelectronic characteristics are noteworthy [12,13]. It is also mentionable that the value-to-price ratio is the highest in the case of energy storing devices. Supercapacitors and
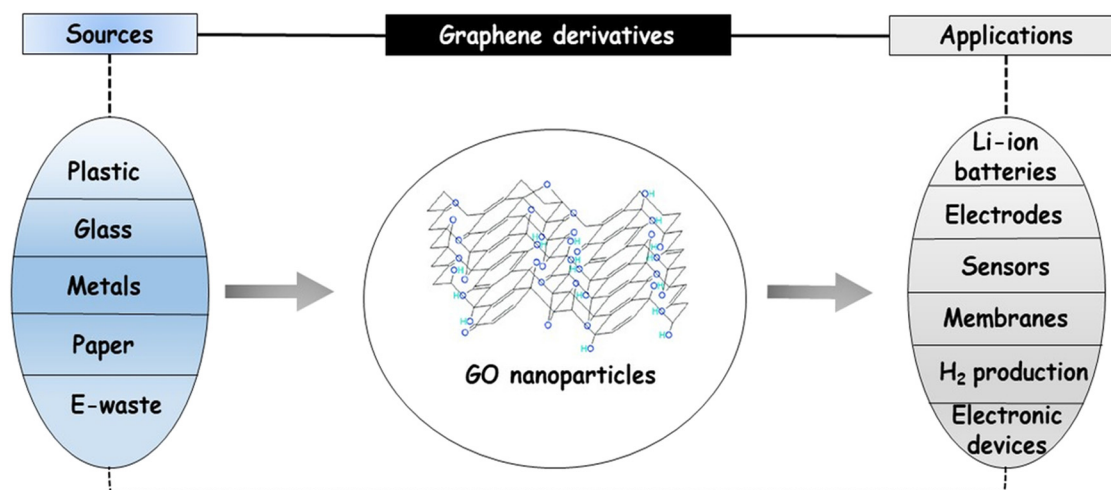
Figure 10: Schematics of waste source incorporation into graphene nanocomposites and their remarkable energetic applications. The structural motif of GO is reproduced with permission from [262].

Table 4: Role of waste sources derived graphene nanocomposites for industrial applications

Waste materials	Nanocomposites	Method of synthesis	Particle size	Graphene concentration	Outcomes	Energetic applications	Ref.
Wool waste	Keratin–graphene nanocomposite	Dispersing	—	5%	Flexible electrodes with remarkable sheet resistance ($\approx 10\ \Omega\ \mathrm{sq}^{-1}$)	Production of sustainable electronic devices	[217]
Sludge waste	Si–GR	One-step aerosol process	532 nm	0.5%	Improved electrochemical performance was evaluated with Si–GR composites	LIBs	[219]
Tire waste	GNPs–PP	Melt mixing	50 nm	1%	Fabrication of tire waste-derived GNPs doped PP nanocomposite	Crystallization and mechanical properties	[223]
Eggshells waste	GO–HAP/LLDPE	*In situ* process	50–200 nm	1, 2, 3, 4 and 5%	HAP-decorated GO filler were used in LLDPE nanocomposite.	Thermo-mechanical properties analysis	[232]
Tree gum waste	GA–GO, GK–GO, KG–GO	Solution casting	200 nm	0.5%	The addition of GO further transformed the brittle gum film into a freestanding film with substantial mechanical strength	Gas barrier properties	[233]
Oil palm waste	L–GO–ZnO	Solvothermal method	—	5%	Removal of Cd (II) from synthetic wastewater by using lignin-based GO–ZnO anodes	Energy transportation *via* microbial fuel cells	[235]
Coffee waste	CEPG–PANI–Ag	Solution polymerization	50 nm	0.5%	Fabrication of novel electrode material using coffee waste-derived GO nanocomposite	Rechargeable batteries	[237]
Tire waste	RW/PLA/GNPs	Twin-screw extruder	—	1, 2, and 3%	PLA combined with recycled tire waste/GNPs nanocomposite	Evaluation of mechanical properties	[243]
Aluminum waste	Graphene–Al_2O_3/Al porous	Ball milling	24–66 nm	4%	Improved electrical conductivity and apparent porosity	Mechanical properties	[267]
Moringa oleifera shell waste	AG@rGONCs	One-pot synthesis	30–50 nm	1%	Green fabrication of reduced graphene-based silver nanocomposites	Sensing of heavy metals	[268]
Cellulose	CMC–GO and CMC–rGO	Solution polymerization	—	0.4%	Graphene-based nanocomposite superabsorbent hydrogels were produced	Improved gel properties using electron beam radiation	[269]
Rice husk waste	GO/OMC	Solution mixing	50 nm	0.5%	Rice husk derived GO-based ordered mesoporous carbon nanocomposites were fabricated	Absorption capacities analysis	[270]
Polystyrene (PS) waste	TiO_2/rGO/g-C_3N_4	Facile calcination	87 nm	2 and 5%	Effective & eco-friendly TiO_2/rGO/g-C_3N_4 nanocomposite-PS floating films were tested	Photocatalytic degradation	[271]
Oil palm waste	GO/cellulose	Simple mixing method	100 μm	0, 1, 2, 3, and 4%	A facile method has been projected to synthesize GO/cellulose aerogel composites that feature thermal stability from waste biomass	Thermal stability analysis	[272]
Wood waste	WH-MMT–GO, WH-MMT-rGO	Hydrothermal synthesis	37.3 nm	0.8%	Nanocomposites were prepared using renewable forestry waste	Fire retardant	[226]
Coconut shell and wood charcoal waste	GTO	Hydrothermal synthesis	20 nm	1%	Facile synthesis of waste-derived graphene–tin oxide	Antibacterial studies	[229]

(Continued)

Table 4: *Continued*

Waste materials	Nanocomposites	Method of synthesis	Particle size	Graphene concentration	Outcomes	Energetic applications	Ref.
Plastic waste	HrGO	Inverse co-precipitation	50 nm	0.5%	Plastic waste-derived highly rGO nanocomposites were produced	Effective adsorption efficiency against antibiotics	[236]
Mushroom and chitosan waste	AgGO	*Ex situ* process	50 nm	4%	Greater activity against *Pseudomonas aeruginosa*	Enhanced microbial study	[273]
Sugarcane straw	BC/rGO	*In situ* fermentation	31,25,42, and 38 nm	1, 2, 3, and 5 wt%	Evaluation of fermentation, mechanical strength, and kinetic studies through nanocomposites	High-value biomedical applications	[274]
Polyurethane (PET bottle)	PU–GO	Polymerization reaction	10–30 nm	1, 2, 5, and 10 wt%	A green synthesis of waste-derived GO nanocomposite	Bone tissue engineering	[241]
Chitosan	SBMGO	Solution mixing	—	0.5%	Schiff base-magnetite–GO nanocomposite was introduced	Removal of heavy metals (Cu,Cr) from wastewater	[275]
E-waste	GOV–Pt	Solvothermal process	50 nm	1 and 2%	Synthesis of GO/vanadium pentoxide (GOV) nanocomposite	Antimicrobial and anti-cancer studies	[276]
Batteries waste	CRGOMC	Solution mixing process	100 nm	1%	Chemically rGO/methyl cellulose nanocomposite was synthesized	Drug delivery	[277]

Note: GA-GO, GO doped with gum Arabic; GK-GO, GO doped with gum karaya; KG-GO, GO doped with kondagogu gum; Si–GR, silicon–graphene composite; AgGO, Silver-based graphene oxide nanocomposite; AG@rGONCs, reduced graphene oxide based silver nanocomposite; CMC–GO and CMC–rGO, carboxymethyl cellulose-graphene oxide/rGO nanocomposite; SBMGO, Schiff base-magnetite–GO nanocomposite; GO-HAP/LLDPE, HAP decorated GO based linear low density polyethylene nanocomposite; HrGO, highly reduced graphene oxide; GO/OMC, graphene oxide based ordered mesoporous carbon nanocomposite; PP, polypropylene; GTO, graphene–tin oxide nanocomposite; WH-MMT-GO/rGO, wood auto-hydrolysates doped with montmorillo-nite/graphene oxide/rGO nanocomposite; GOV-Pt, graphene oxide/vanadium/pentoxide nanocomposite; CRGOMC, chemically reduced graphene oxide/methyl cellulose nanocomposite; RW/PLA/GNPs, recycled waste combined with poly(lactic acid)/graphene nanoplates nanocomposite; L-GO–ZnO, lignin based graphene oxide/zinc oxide nanocomposite.

Table 5: Literature of waste-generated nanomaterials for comparing their performance outcomes

Supercapacitors	Specific capacity (F g^{-1})	Current density (A g^{-1})	Capacity retention	Number of cycles	Energy density Wh kg^{-1}	Power density kW kg^{-1}	Ref.
PANI@sulfonated graphene	752.3	1	85.90%	5,000	23	6.1	[182]
Graphene@Ni$_3$V$_2$O$_8$ Waste-derived	748	0.5	71%	3,000	103.94	46	[186]
rGO/MnOx	34.4	2	96.60%	20,000	4.8	1	[227]
Spent battery powder@rGO	150	1	100%	1,000	—	—	[230]

LIBs	Reversible capacity (mA h g^{-1})	Current density (A g^{-1})	Capacity retention	Number of cycles	Ref.
CuSi$_2$P$_3$@Graphene	2,069	2	—	1,500	[177]
SiOx@Fe$_3$O$_4$@FLG Waste-derived	833.4	0.5	81.80%	500	[178]
Si@GR	1,626	0.2	~75%	50	[219]
CEPG−PANI−Ag	397	5 (mV s^{-1})	94.50%	5,000	[237]

LIBs can be charged and discharged many times providing repeated usage for the composite material. Table 5 contains the energy related materials from cutting edge research articles of composite designing specified in Tables 2 and 4.

Remarkably, the specific capacitance of graphene composite based LIB materials was not higher in magnitude as compared to the waste-derived composites indicating that the composites generated from waste sources can reach similar energy outputs as the state-of-the-art energy storage composite materials. On the other hand, the cyclic stability of waste-generated materials varies over a wide range, which can be an obstacle for mass production in which only the highest performing solids are desired.

There is no significant tendency found regarding the capacitance retention for the battery materials. Based on the above studies, the specific capacitances were already lower in magnitude for waste-generated materials as compared to conventional ones for supercapacitors. On the other hand, one beneficial feature could be that lower power and energy density may lead to much longer life cycle and better capacity retentions which promotes the long-term usage for waste-based energy storage composites.

4.3 Recent studies on graphene nanocomposites for energetic applications (2021 till July 2022)

As direct energy storage materials, supercapacitors became popular among reports published since 2021. Preparing AC matrixes for composites synthesized using pyrolysis were carried out using different fruit waste [278], cigarette filters [279], and canola waste [280]. The resulted AC was mixed with GO. These devices showed a good electric double layer capacitance due to the optimal morphology. One of these studies shed light to the fact that covalent functionalization of AC@GO composites with conductive polymers bestowed pseudocapacitive behavior to these materials [280]. AC derived from animal bone waste was also used in the counter electrode of composite supercapacitor devices [281]. Besides that, one possible way to create the pseudo-capacitance-based composite was to use plastic waste-derived graphene and to grow V_2O_5 nanospheres on the graphene surface *via* hydrothermal method [282]. A similar strategy is that acidic hydrolyzed corn stalk is subjected to hydrothermal treatment to obtain carbon nanospheres and crossing them with GO. Another hydrothermal step provided carbon/rGO hydrogel with appropriate supercapacitor properties [283]. Saccharides can also be used as reducing agents for growing metal NPs on graphene sheets. Industrial molasses was used to fabricate multifunctional composite materials with photocatalytic activity and high gravimetric capacity as supercapacitor [284]. It is important to highlight that the electrochemical performance outcomes of waste-generated nanomaterials (typical examples are shown in Table 5) are competitive as compared to the performance parameters of recently published non-waste-derived supercapacitors or other functional electronic [92,285–291] composites.

According to the studies published in the topic of nanocomposites produced from waste shows that 32 of the 58 research articles were energy related in the last year till July 2022 as presented in Table 6. One can conclude

Table 6: Summary of recent studies containing waste-derived nanomaterials for energetic applications

Waste material	Matrix	Application	Outcomes	Ref.
Expanded polystyrene	Carbon–graphene Xerogel (CGX), flaky graphite (GR) and nickel–zinc ferrite (FeNiZn) composites in PS matrix	Electromagnetic properties	The increase in CGX and GR influenced the increase in the complex permittivity, and that 10 wt% CGX + 50 wt% FeNiZn composite sample demonstrated an absorption peak at 10.5 GHz, thus developing greener and low-weight composite materials to be used in microwave applications	[292]
Waste cotton fiber	rGO NPs and enzyme-treated hemp fiber (HF)	Mechanical, dynamic, and thermogravimetric properties	The cotton/epoxy composite loaded with 0.3 wt% of rGO shows a ~11% increment in tensile strength, ~20% increment in flexural strength, and ~30% increment in impact strength	[293]
Bio-waste	Few layered graphene/SrTiO$_3$/PAN	Photocatalytic water splitting	The developed photocatalytic systems based on FLG derived from bio-wastes, SrTiO$_3$ and PAN are perspective candidates for production of hydrogen under splitting water–alcohol mixtures	[294]
Orange peels, banana peels, and potato starch	GO	Supercapacitor	The superior electrochemical performance is attributed in orange peel derived nano porous carbon/GO due to porous structure	[278]
Biomass	PP as polymer matrix and graphene supported magnetic carbon nanocomposite (MCNC)	Electrical properties	MCNC in PP matrix and formation of network for 15% loading of the filler facilitated 800% higher electromagnetic shielding effectiveness as compared to that of neat PP	[295]
Rock wool waste	rGO	Electrical properties	The obtained rock wool/rGO (RW–rGO) composites showed excellent Electromagnetic interference (EMI) shielding performance (>25 dB) and good hydrophobicity (water contact angle >130°)	[296]
Waste slicing sludge	Boron-doped graphene silicon (BG-3)	Electrochemical performance	BG-3 showed enhanced stability (70% retention after 80 cycles at 500 mA g^{-1}). Moreover, boron-doped graphene silicon/graphite composite (BG-3@G) demonstrated a high capacity of 850 mA h g^{-1} and favorable cycle retention of 76.5% after 180 cycles at 225 mA g^{-1}	[297]
Paddy straw	GQDs	Dielectric and mechanical properties	GQDs-stone nanocomposites exhibited low dielectric constant (ε' ~10) as compared to pristine stone composites (ε' ~ 55)	[298]
Waste batteries	rGO	Photocatalysis	The result shows that visible light with WO$_3$–rGO composite achieved an effective degradation of MB dyes up to 88% in 120 min irradiation	[299]
Loofah sponges	Ni NPs and graphene	EMI shielding	The outstanding performance was achieved that the shielding effectiveness was raised to 57 dB, and the	[300]

(Continued)

Table 6: *Continued*

Waste material	Matrix	Application	Outcomes	Ref.
Polybutadiene (PB) waste	Octadecylamine-functionalized rGO (ODA-rGO)	Energy storage	specific shielding effectiveness was raised to 616.8 dB cm^3 g^{-1}	[301]
Cigarette filter wastes	AC-GO	Supercapacitors	Heat storage capacity as high as 171.5 J g^{-1} Advanced carbon composites delivered specific capacitances of *ca.* 160 F g^{-1} at 0.25 A g^{-1} which correspond to energy densities of *ca.* 65 Wh kg^{-1} at 210 W kg^{-1}	[279]
Waste polystyrene (WPS)	CuO-NPs and rGO	Electrical properties	The electrical conductance of the composite was enhanced from 6.7×10^{-14} to 4×10^{-7} S m^{-1} in contrast to WPS at 2.0×10^6 Hz	[302]
Textile-based waste	Wool/PAN composite fiber and GO	Electrical conductivity	The wool/PAN/rGO knitted fabrics showed higher electrical conductivity ($\sim$1.67 S cm^{-1}) compared to the control PAN/rGO knitted fabrics ($\sim$0.35 S cm^{-1})	[303]
Photovoltaic silicon waste	Si nanowires (SiNWs) and rGO	Electrodes for LIBs	The SiNWs@RGO electrode exhibited an ultrahigh initial Coulombic efficiency (89.5%) and robust cycle stability (2381.7 mA h g^{-1} at 1 A g^{-1} for more than 500 cycles) at high Si content of 76%	[304]
Rice husk	Sb and Si composited with nitrogen-doped graphene (SbSi/NrGO)	Electrodes for LIBs	SbSi/NrGO nanocomposites to be used as high-performance anode materials in high-performance LIBs for advanced applications	[305]
Biomass pulping waste	Lignin nanoparticles (LNPs) and liquid crystal hydroxypropyl cellulose (HPC)	Thin films	The results showed that LNPs provided the HPC-nanocomposite film with acceptable thermal stability	[306]
Waste surgical tools	Gold nanoparticles Au-NPs incorporated with iron/GO (Fe$_2$O$_3$/GO)	Electrodes for LIBs	The results showed that the composite has a stable reversible capacity of about $\sim$980 mA h g^{-1} and was maintained for > 200 cycles	[307]
Biochar	TiO$_2$ and graphene-like biochar (PC)	Photocatalysis	The results showed that the optimal TiO$_2$–PC composite delivered an excellent photocatalytic performance, nearly four times that of TiO$_2$	[308]
Kitchen waste	GO and V$_2$O$_5$	Photocatalytic hydrogen production	Hydrogen production rate of V$_2$O$_5$ increased from 247.52 to 354.15 mol h^{-1} g^{-1}.	[309]
Rice husk	SiO$_2$/rGO and silicon dioxide/nitrogen-doped rGO (SiO$_2$/NrGO)	LIBs	The obtained SiO$_2$/rGO and SiO$_2$/NrGO nanocomposites possessed high capacity and superior cycle performance	[310]
Waste plastic	Vanadium pentoxide and graphene sheets	Supercapacitors	Fabricated cells 1 and 2 showed exceptional specific capacitances of 139.7 and 51.2 F g^{-1} at 5 mV s^{-1} scan rate, respectively	[282]
Waste biomass corn stalks	Carbon microspheres (CSs) and rGO	Supercapacitors	CS/rGO-3 sample-based symmetric supercapacitors showed an excellent specific	[283]

(Continued)

Table 6: *Continued*

Waste material	Matrix	Application	Outcomes	Ref.
			capacitance (281.5 F g⁻¹ at 0.3 A g⁻¹), a good rate performance (81.87%), and an eminent capacitance retention rate (102.2% after 10,000 cycles at 10 A g⁻¹)	
Cooked chicken bone waste (CCBW)	Hausmannite/sulfur rGO (MO/RGO-S) and cobalt-nickel layered double hydroxide (CN)	Supercapacitors	The device produced capacity retention of 85.1% and Coulombic efficiency of 99.7% at 10,000 galvanostatic charge-discharges cycles at 6 A g⁻¹	[281]
Aeschynomene aspera (AA) plant, a sustainable, waste natural carbon source	Nickel oxide (NiO), GO, and rGO	LIBs	The RGO–NiO composite exhibits reversible specific discharge capacity of 847 mA h g⁻¹ at 100 mA g⁻¹ and an excellent rate capability of up to 1,000 mA g⁻¹	[311]
Fly ash	GQDs	Dielectric properties	An increase in the electric dipoles and improved interfacial and chemical bonding of crystalline GQDs with fly ash and epoxy	[312]
Molasses waste	FLG, Au and Ag NPs	Photocatalytic and supercapacitance applications	The Au/FLG (PVDF) and Ag/FLG (PVDF) based electrodes reveal relatively high gravimetric capacitance, 205 and 729 F g⁻¹, respectively	[284]
Canola waste	AC, poly ortho aminophenol (POAP), GO	Supercapacitors	The highest specific capacitance of 1350.5 F g⁻¹ at the current density of 2 A g⁻¹ and retention of 96.2% initial capacitance after 5,000 consecutive charge-discharge cycles verified the superior electrochemical performance	[280]
Waste dry cell batteries	Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), graphite, GO, rGO	Organic solar cells	Highest power conversion efficiency (PCE) of 5.07–6.55%, short-circuit current density (J_{SC}) of 16.61% to 18.42 mA cm⁻², and fill factor (FF) of 42–48%, which was consistent with the enhanced hole mobility of 1.67×10^{-5} to 2.31×10^{-5} cm² V⁻¹ S⁻¹, respectively	[313]
Corn husk	Multilayered graphene/zinc oxide (MLG/ZnO)	Photocatalysis	Enhanced photocatalytic activity of MLG/ZnO nanocomposite in photodegradation of rhodamine B, Congo red, and doxycycline	[314]
Aluminum waste	Al/Al₂O₃, SiC, graphene	Dielectric properties	The microhardness, compressive strength, and elastic moduli increased by about 142.5, 44.8, and 46%	[315,316]

that regarding this trend, composites connected with energy using or energy storage applications of waste materials provides the most sustainable approach of utilizing waste graphene materials as the basis of green graphene composite using technologies.

4.4 New frontiers in waste-derived graphene composites for non-energy related applications

We briefly discuss the best performing non-energy related composite materials reported recently (2021 till July 2022), in terms of their fields of application. Adsorption is one of such fields, for which composite materials were prepared by either hydrothermal or pyrolysis methods. Pyrolysis of waste wood and graphene together afforded graphene@biochar hybrids with high adsorption capacity for Cu^{2+} ions [317]. Sodium silicate originating from rice husk carbonization was mixed with a block-copolymer, GO, and surfactant, subjected to hydrothermal treatment that resulted in highly ordered mesoporous structures with higher adsorption capacity for methylene blue (MB) than non-waste-derived similar materials [318]. Hydrothermally synthesized waste polyacrylonitrile (PAN)/rGO composites demonstrate outstanding oil absorption capacity and good reusability [319].

Waste-derived composites also open new frontiers in catalysis. GO–straw–CuNP composite, in which the plant additive was not completely carbonized, shows optimal porosity, pore size distribution and hydrophilic surface acts as feasible catalyst support in *p*-aminophenol reduction [320]. Sugarcane bagasse was used for the formulation of Cu_2O–Au@rGO composite to analyze sonocatalytic material in decomposition of 4-nitrophenol [321]. Eggshell-transitional metal composites were demonstrated to be appropriate catalysts for carbon nanotube growth [322].

New construction materials still dominate the research on non-energy related waste-derived composites according to the number of publications. Of these, concrete admixture was the most frequently reported topic. One approach was adding waste-derived material combined with GO to the grouting paste [323]. A chitosan waste based nano-sealant was developed [324]. Adding very low amounts of GO to waste concrete powder provides a cement which can then produce a recycled concrete that competes with conventional concretes in terms of flexural and compressive strength [325]. Remarkably, carbonized textile industrial waste combined with GO and colloidal silica has resulted in concrete material with enhanced thermal and mechanical parameters [326]. Polymer wastes can also be utilized with GO for cementitious composite materials. A recent report on upcycling PP waste originating from waste disposable masks revealed that the GO can have a multiple role (dispersing agent and regulator of hydration precursor distribution) in the cement composite production process [327]. Composite waste polymer/GO filler loaded asphalt exhibits comparable performance with those currently used without any filler [328]. Consequently, great quantities of plastic wastes can be utilized in road construction. In summary, polymers, carbons, or waste concrete powders are recently incorporated into cementitious or asphalt matrix in which GO can regulate the structure formation and enhance the mechanical strength, producing high quality materials in a sustainable way.

Finally, bioresource derived wastes are increasingly exploited for a vast range of applications. Rice husk derived plant waste carbons were able to improve the mechanical properties [329] and tribological parameters of aluminum matrices [330]. Related research may attract considerable attention because aluminum composites are largely unexplored. Other recent non-energetic research trends include rubber waste for graphene fabrication [331], eco-friendly routes of graphene synthesis from waste [332], sugar beet bagasse waste for pesticides control [333], blackberry waste extract for graphene nanocomposites [334], or waste grapefruit peel for environmental applications [335]. An increasing emphasis is laid on the use of GO in composite formulation owing to its multifunctionality. GO/cellulose composites with microcrystalline cellulose acid hydrolysis are obtained using the waste acids generated by the Hummers method [336]. Collagen-modified GO finds utility in ultrafiltration [337], fruits and vegetable peel waste in antioxidant analysis of ultra-fast selectively recovered 4-hydroxy benzoic acid [338], walnut shell derived GO/hydroxyapatite nanocomposite in osteogenic differentiation and antimicrobial activity [339], and waste rice husk/PVC/GO nanocomposite as green additives [340]. Recycling of disposable mask affords graphene nanoplatelets [341], sugarcane bagasse agricultural waste can be converted into nanoscale graphene containing photoluminescent and anticorrosive epoxy paints [342], and a green conductive composite of graphene oxide-like char and zero-valent iron was found to be efficient for biogas production [343].

5 Limitations and future recommendations

5.1 Challenges

The use of functional materials incorporated into nanocomposites has undoubtedly increased their performance in various applications. However, there are still many challenges that need to be addressed before the applications of nanocomposites can be fully commercialized as shown in Figure 11.

Several key challenges have been identified and will be discussed further in this section.

- **Toxic effects:** The toxicity of nanocomposites need to be addressed which include health and environmental issues. Both composite counterparts (the carbon and the non-carbonaceous phase) may be toxic not only to human consumers, but to living organisms as well. We especially foresee two main hazards that threat healthy species: metal ions and compounds (typically heavy metals) and organic contaminations (carcinogenic or mutagenic compounds). The former may originate typically from the non-carbon composite counterpart, for example, by the chemical dissolution of semiconductor metal oxide or metal sulfide NPs used in optoelectronic applications (such as cadmium or chromium compounds). However, such metallic contaminations may also originate from the waste-derived graphene-based NPs, in case the waste source is not carefully purified from metallic components. A typical

Figure 11: Key challenges related to waste-generated graphene nanocomposites for future studies.

example is that of rare earth elements, which are commonly found in e-wastes produced from high technology devices, including smart phones, digital cameras, computer hard disks, and fluorescent and light-emitting-diode lights, flat screen televisions, or electronic displays. These electronic devices trashed into e-waste are usually a mixture of the broken metallic functional parts (hard disks, cables, connectors, *etc.*) and the plastic housing. The incomplete disassembly of these electronic devices or when the metallic parts are not removed selectively by magnetic separation will result in the metal contamination of the polymeric waste, which retains its metallic compounds even after the polymer is pyrolyzed or transformed to graphene by other means.

- **Structural modifications and morphology control**: As the key ingredients in producing nanocomposites, size, distribution, shape, surface area, structure, and phase transitions are significant features that should be focused for the analysis of nanostructures which require advanced analytical tools.

- **Green synthesis:** The other, non-metallic type of toxins involve a wide variety of organic molecules, such as aromatic mutagenic or carcinogenic compounds, which may form during the carbonization process of the waste and may slowly dissolve into a water sample or cause skin irritation during physical contact with the nanocomposite assembly. This latter threat may be easily overcome by a proper formulation of the graphene counterpart upon controlled pyrolysis or proper washing and purification.

- **Scale-up and cost-effective production**: Optimization of the aforementioned green techniques are required in order to produce nanomaterials in a large scale at an affordable cost before the applications of nanocomposites can be fully commercialized. Besides, the commercial application of waste-generated nanocomposites beyond the lab-scale necessitates the cost-effective production and consolidation into the end product preserving the stable nanostructures. During the synthesis of nanocomposites, the thermal effects of endothermic and exothermic reactions are remarkable. Therefore, more knowledge on the thermal behavior of the composites would be useful for elaborating the optimized processing conditions. Important factors include burning rate, thermal stability, particle coagulation/sintering effects, and ageing characteristics of the waste-generated nanocomposites. These properties can be tested using standard characterization methods.

- **Environmental impacts and human health**: The handling of the nanocomposites is also an issue. Proper

guideline of handling and storage needs to be implemented in order to prevent any contamination of the nanomaterials. Any contaminations also pose a risk to the human health, because the long-term health effects of many molecular or particulate compounds are unknown. Long-term exposure of waste-derived composites incorporated to devices that are contacted with the skin (*e.g.*, in the form of sensors, wearable electronics) are potentially damaging. However, waste-derived materials do not seem to pose especially larger risks than any other nanocomposite materials already commercialized.

5.2 Future prospects

Nowadays, waste treatment and reuse are one of the most important aspects in industrial production and municipal activities, which generate serious amount of waste. Waste management includes waste collection, transportation, reuse, and landfilling. This study aimed to shed a light on a completely new approach of wastes recycling focusing on bottom-up synthesis of nanocomposites, containing high value-added graphene derived nanomaterials. To promote, this interesting and valuable research direction with globally forward-looking advantages, we would like to present our ideas into a few suggestions.

1) Only a few studies focus on the usage of sulfur–graphene/GO nanocomposites, which have promising applicability in electrical devices such as supercapacitors and batteries [344]. It has to be highlighted that sulfur waste is generated in serious amounts as a petroleum industry by-product and during biogas desulfurization in sewage sludge treatment, which can be perfect sources of waste-derived nanocomposites production.

2) Metal sulfide/GO composites have very similar advantages and they can be produced by one-step methods such as hydrothermal synthesis, which reduces the cost and allows a facile and efficient reuse process. Furthermore, as mentioned before, one-step hydrothermal method was successfully used in the case of metal waste-derived composites.

3) Red mud is a by-product of aluminum industry and it is generated in large quantities. However, very few research studies on its incorporation into graphene derived nanocomposites is known [345,346]. According to its metal composition, it could be a suitable candidate of producing composites with photocatalytic activity, electrical devices (supercapacitors, batteries, gas sensor membranes), and anti-corrosion coatings for anodic protection. Moreover, one-step hydrothermal method is a potential synthesis process to get this new, multifunctional nanocomposite from this unexploited source [347,348].

4) Graphene/LDH nanocomposites can be very efficient adsorbents of heavy metal ions. According to the literature, LDH particles can be prepared by one-step co-precipitation method, which could be formed from the cationic metal ion contamination of wastewaters [349]. In addition, LDH/graphene hybrids can be produced as a result of this very simple synthesis method. As various studies presented, these solids show good adsorption properties for heavy metals [350]. Furthermore, the spent adsorbent can show significant catalytic/photocatalytic activity on the metal centers (Pb, Pd, Hg). It is also important to point out, beside the well-known catalytic combustion of organic pollutants, that Pd centers can also be utilized as selective CO_2 reducing agents.

5) Graphene/cement-based materials proved to be extremely useful for the improvement of mechanical features. To take into account the amount of the globally gathered construction waste, graphene-based nanomaterials can be interesting in the recycling and reuse of construction waste as nanostructured additives. According to the literature, a lot of study has been published about the effects of graphene-based materials on the physicochemical properties of cement-based matrices [351,352], but to the best of our knowledge none of them tried to apply them for waste-derived nanocomposites. To highlight that, the car industry produces a serious amount of limestone as by product, which is a possible raw material for cementitious graphene containing structural materials because the graphene and graphene-based materials can largely affect the pore formation processes.

6) Finally, we suggest that future studies of waste-derived graphene nanocomposites should contain more information on important morphological characteristics, particle size of composite counterparts, or other physical dimensions of the bulk composite structure, such as the layer thickness when the composite is formulated into thin films.

6 Conclusion

Over the years, many great achievements of materials science induced a strong connection between waste management and the development of functional graphene-based materials. To deal with the current extreme climate

and environmental changes, a balance is required to control proper recycling of waste materials, which are excellent carbonaceous sources for nanocomposites. The concept of producing graphene or GO directly from waste is quite appealing but at present a well-established direct conversion methodology similar to the production of waste-derived AC *e.g.*, from bioresources (coconut shell, rice husk, *etc.*) does not exist. In terms of valorization, GO/rGO is currently considered as the high value-added composite counterpart. Accordingly, many types of nanocomposites featuring low amounts of GO or rGO (typically 0.1–2 wt%) are reported to exhibit outstanding performance not only in energy driven applications but also as gas barriers, fire retardants, photocatalysts, or structural materials.

Various composites are synthesized from biomass, eco-friendly waste, or biodegradable sources but they are not valuable for the mitigation of overall negative environmental impacts of wastes. Composites of the abovementioned applications were fabricated from plant wastes but simple and low-cost production methods need to be developed to make their application cost-effective. Cellulose is the most abundant natural source, but many types of non-biodegradable plastic wastes may also be efficiently reutilized by carbonization [353]. The simple recycling methods of industrial metal and metal containing wastes provide the top quality of contemporary waste-derived nanocomposite production [354]. Decoration of graphene with metal NPs for photocatalytic, catalytic, or electrode materials are very effective in terms of valorization.

Regarding the utilization of waste materials for energetic applications, the two critical parameters of the performance of the final composite are usually the filler content and its degree of dispersion, because they vastly influence the percolation threshold of the system [355]. In case the particles represent a long-range connectivity (*e.g.*, continuous electrical pathway), the use of various (non-conductive) matrix materials are possible without a loss of performance. However, for future applications, electric/conductive properties need to be more thoroughly investigated regarding filler-matrix effects.

Biomass waste sources were appropriate for preparing composite materials for all listed applications. However, using eco-friendly waste as precursor does not lower the negative environmental effect of waste, these materials can act as carbon sources and inorganic material sources (*e.g.*, SiO_2 derived from waste rice husk). In terms of environmental effects and cost effectiveness, the energy storage materials derived from industrial metal (*e.g.*, silicon) or metal oxide (manganese oxide waste from

Hummers method or waste batteries) waste means the most noteworthy research direction of the waste-derived graphene composites.

To enhance the functionality, porosity and other properties, numerous carbon-based nanomaterials such as graphene, GO, and rGO-based nanocomposites have been modified through recycling, activation, thermo-chemical exfoliations, and post-treatments. It was observed that this functionalization helped to increase the surface area of these waste-derived graphene nanocomposites for efficient electronic devices and energy storage systems. These nanocomposites also showed great potential for other applications such as drug delivery, gas capturing, biomedical, catalysis, and wastewater treatment. An interesting finding of our literature review is that in almost all cases, the graphene phase was the high added-value component, and only the composite counterpart was derived from waste materials. However, we foresee that nanocomposite materials entirely derived from waste sources will be developed for future valorization.

Additionally, good mechanical properties (as provided by the flexible but strong carbon particles) and high electrical conductivity of waste-derived nanocomposites are effective in sensors and fuel cell applications. Similarly, high surface area, porosity, particle size, and structural chemistry boost the performance of these nanocomposites as electrodes among batteries. In summary, waste-generated energy-driven graphene nanomaterials have the potential to be used as a great alternative of conventional carbon materials (such as carbon blacks or graphite powder) and their composites for the future industrial applications.

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