

Plastic wastes derived carbon materials for green energy and sustainable environmental applications



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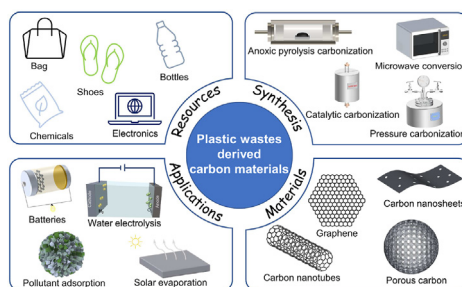
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HIGHLIGHTS

- Strategies for designing carbon materials from plastic wastes are summarized.
- Applications of PWCMS for green energy and sustainable environment are detailed.
- PWCMS' property-performance correlation is analyzed.
- Perspectives in the development of future PWCMS are outlined.

GRAPHICAL ABSTRACT



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ABSTRACT

Plastic waste has become a serious environmental issue and has attracted increasing attention. Various treatment technologies have been developed for the remediation of this waste, including degradation, recycling and upcycling, and transformation to value-added products has been extensively studied. Transitioning plastic waste into carbon-based functional materials is especially attractive because of the practical applications of plastic wastes derived carbon materials (PWCMS) in the field of green energy and in sustainable environmental practices. Herein, recent advances in the preparation and applications of PWCMS are systematically reviewed. Thermal treatment methods for synthesizing carbon-based materials from plastic waste are summarized, including anoxic pyrolysis, catalytic and pressure carbonization techniques, flash Joule heating and microwave conversion. The applications of PWCMS and PWCMS-based composites to green energy storage and production (such as in batteries, supercapacitors and water-splitting systems) and sustainable environmental concepts (pollutant adsorption/degradation, solar evaporation and CO₂ capture) are detailed, with an emphasis on the property-performance correlation. The potential for future development of PWCMS is also examined. This review is meant to provide insights into the advanced applications of PWCMS and to stimulate the future upcycling of plastic waste.

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1. Introduction

Plastics are omnipresent in food packaging, medical devices, construction, transportation, furniture, electrical and electronic devices because these materials provide excellent flexibility, corrosion resistance, high strength-to-weight ratios, good processibility and low fabrication costs [1–3]. However, the rapidly growing production of plastics combined with improper management has generated massive quantities of plastic waste in various environmental spheres [4–6]. By 2100, the mass of plastic waste released on land annually in the European Union is predicted to be 4.73×10^5 to 9.1×10^5 tonnes with approximately 2.5×10^7 to 1.3×10^8 tonnes of plastics released into the ocean [7,8]. There is thus an urgent requirement to reduce plastic waste in the environment. Traditionally, these materials are disposed of either by direct landfilling or incineration, both of which can in turn lead to serious environmental pollution [9]. The alternative approach of converting plastic wastes into value-added substances such as fuels, carbon-based materials and fine chemicals has therefore received increasing attention.

Because plastic waste is rich in carbon, it can serve as a precursor for the synthesis of value-added carbon-based functional materials. Currently, various types of plastic wastes (including polyvinyl chloride (PVC), polystyrene (PS), polyester terephthalate (PET), polyethylene (PE), phenol formaldehyde resin (PFR), low-density polyethylene (LDPE), high-density polyethylene (HDPE), polyacrylonitrile (PAN) and polypropylene (PP)) can be converted into a wide range of carbon materials, such as graphene, carbon dots, carbon nanotubes (CNTs), carbon nanosheets and porous carbon. This can be accomplished via anoxic pyrolysis carbonization, catalytic carbonization, hydrothermal carbonization, flash Joule heating and microwave heating [10,11]. By controlling the synthesis process, it is possible to prepare plastic waste-derived carbon materials (PWCMS) having high degrees of porosity, large specific surface areas (SSAs), rich surface chemistries (including the presence of dopants or functional groups), high conductivity and good stability [12–14]. Accordingly, PWCMS have been widely used in applications related to green energy (such as batteries, supercapacitors and water-splitting systems) or sustainable environmental practices (pollutant adsorption/degradation, solar evaporation and CO₂ capture) [15–20]. PWCMS can provide significant economic and environmental benefits while reducing costs and assisting in the management of plastic waste.

To date, several reviews have summarized the production and applications of PWCMS. Such previous reports have primarily focused on conventional carbonization processes for synthesis [21–25] while some only examine typical carbon materials such as CNTs [13,26,27] or graphene [12]. In addition, the applications of PWCMS in energy storage [3, 9,14], as pollutant adsorbents [3,28] and as CO₂ capture materials (in the form of porous carbon) [29] have been reviewed separately. A thorough review of the synthesis and applications of PWCMS in the fields of green energy and sustainable environmental practices has not yet been published.

The goal of the present review is to provide a comprehensive summary of recent advances in the synthesis and applications of PWCMS. The many thermal treatment methods used to prepare carbon-based materials from plastic wastes are first summarized. Following this, the applications of PWCMS in green energy (batteries, supercapacitors and water splitting) and sustainable environmental technologies (pollutant adsorption/degradation, solar evaporation and CO₂ capture) are analyzed in detail. The property-performance relationship for these materials is also emphasized and perspectives for advanced next-generation functional PWCMS are presented.

2. Synthesis of carbon-based materials from plastic waste

Reforming plastic waste into carbon-based materials generally requires high-temperature treatment/carbonization, which may involve various atmospheres, catalysts, pressure values and heating methods.

Carbonization typically involves pyrolysis followed by an actual carbonization step, although the terms pyrolysis and carbonization are often used interchangeably because these processes can be very similar [20]. Carbon dots, CNTs, carbon sheets, carbon spheres, graphene and irregular porous carbon have all been generated from plastic waste [11, 20,29], and the physicochemical properties of these materials are greatly affected by the synthesis process. Prior to carbonization, pretreatments are required to modify various physicochemical properties of the plastics, with size reduction and stabilization as the two primary objectives. Ball milling is a green and efficient means of reducing the sizes of plastic particles [30,31] and can also ensure the homogeneous distribution of activators such as K₂CO₃ throughout the material. The application of activators generally contributes to porous structures and large SSA. This homogenization improves the subsequent activation process [32]. Stabilization refers to the formation of crosslinked structures via the formation of oxygen-containing functional groups. Thermal oxidation and chemical treatment are two approaches to plastic stabilization [20,33]. Five representative carbonization techniques are introduced in this section, having the general characteristics summarized in Table 1.

2.1. Anoxic pyrolysis carbonization

Anoxic pyrolysis carbonization is a well-known technique for transforming plastic waste into carbon-based materials, typically based on processing in a tube furnace at a relatively high temperature. During this process, the carbonization temperature plays a vital role in modulating the physicochemical properties of the product. A higher carbonization temperature (meaning above 700 °C) provides better pyrolysis with improved porosity and conductivity in addition to fewer oxygen-containing functional groups. The resulting PWCMS may be suitable for electrochemical applications. In contrast, carbon prepared from PS waste at a relatively low temperature of 550 °C contained numerous functional groups and was better suited as an adsorbent for contaminants [37]. Apart from temperature control, another universal strategy for tailoring the properties of PWCMS is post-activation using compounds such as CO₂, KOH or ZnCl₂ to precisely tune porosity. As an example, a KOH-activated tyre pyrolysis char showed significant adsorption of bisphenol A with a high monolayer capacity of 123 mg/g. This performance was superior to that of commercial activated carbon as a result of the large SSA (700 m²/g) and high porosity of the char [38]. In addition, chemical modification (such as via acidic oxidation or doping) can add specific functional groups and heteroatoms to PWCMS [39,40]. The addition of FeCl₃ during the anoxic pyrolysis carbonization of PET waste at 700 °C for 4 h under N₂ was found to generate Fe-doped plastic-char. The presence of Fe greatly promoted the rate of adsorption of 17 α -ethinylestradiol as well as the capacity and the photochemical degradation ability of the plastic-char [41].

2.2. Catalytic carbonization

During catalytic carbonization, plastic waste is pyrolyzed with the assistance of catalysts and transformed into a wide range of carbon-based materials, such as CNTs [42], carbon nanosheets [43], graphene [44] and hierarchically porous carbon (HPC) [45]. In this process, the plastic is first decomposed to generate liquid or gaseous products, after which the desired product is formed on the catalyst surface using the decomposed/degraded material as the feedstock. The catalyst thus plays a dual role by promoting both the initial decomposition and the subsequent formation of the carbon product [46]. A number of earth-abundant catalysts have been developed for the catalytic carbonization of plastic waste, including those based on Ni, Mn, Mo and Fe [42,45–48].

It is worth noting that catalysts may undergo structure reconstruction during the catalytic carbonization process [49,50]. In prior work assessing the catalytic carbonization of PP with Ni, Fe and Fe–Ni catalysts, the PP was first pyrolyzed to form various hydrocarbons that subsequently acted as carbon and hydrogen precursors for the following

Table 1
Summary of thermal treatment methods for synthesizing carbon-based materials from plastic waste.

Methods	Conditions	Typical plastics	Carbon materials	Advantages	Disadvantages
Anoxic pyrolysis carbonization [22]	Carbonization at 500–1000 °C under inert atmosphere or in molten salt, stabilization if needed, activation if needed	PET, PAN, PVC, PE, PS, PFR, polyolefin	Amorphous carbons (activated carbon, mesoporous carbon, carbon fibers)	Simple, low equipment requirements, relatively low cost, applicable for most plastics	Low carbon yields (especially for PS and polyolefin), relatively high energy consumption
Catalytic carbonization [22]	Inert atmosphere, 400–900 °C, catalysts (metal compounds)	PE, PP, PS, PFR, PVC, PVDF ^a , PTFE ^b , PVA ^c , PET	Graphitic carbons (CNTs, graphene, carbon spheres, carbon nanofibers, carbon nanosheets)	Applicable for most plastics, generation of graphitic carbons	Needs catalysts, relatively high energy consumption
Pressure carbonization [34]	Sealed reactors, at 600–850 °C (direct pressure carbonization); Sealed reactors, in the presence of water vapor at 150–300 °C (hydrothermal carbonization)	PP, PE, PVC, PS	Amorphous carbons (activated carbon, carbon dots, carbon spheres)	Sophisticated techniques, high carbon yields	High equipment requirements, relatively high energy consumption
Flash Joule heating [35]	Vacuum reactors, at high current density in a short time (several seconds), conductive substances (carbon black)	PET, PE, PVC, PP, PS	Graphene	Simple, low energy consumption, fast preparation, applicable for most plastics	Difficult management of volatile products, high current involved, needs conductive substances
Microwave conversion [36]	Microwave reactors, microwave irradiation (e.g., 1 kW-h for several minutes), catalysts if needed	PE, PP, PS	CNTs, char	Simple, fast preparation	Relatively high energy consumption, limited carbon yields

^a PVDF: polyvinylidene fluoride.

^b PTFE: polytetrafluoroethylene.

^c PVA: poly(vinyl alcohol).

catalytic decomposition process. During the catalytic breakdown of these carbon-based intermediates, the oxide-based catalysts were simultaneously reduced to active species such as metallic Fe, Ni particles, or Fe–Ni alloys. Interestingly, the bimetallic Fe–Ni catalyst exhibited better performance than the monometallic Ni and Fe catalysts because of the rapid diffusion of carbon intermediates on the surface of the former material. Sequential carbon deposition, diffusion and nucleation proceeded over the catalysts and led to the formation of multiwall CNTs (MWCNTs) (Fig. 1) [48].

2.3. Pressure carbonization

Pressure carbonization refers to carbonization performed at a high pressure in a sealed reactor such as an autoclave [34]. This process is able to synthesize PWCMs with unique structures and in high yields. Based on the pressure source, pressure carbonization may also be referred to as either direct pressure carbonization (autogenic pressure carbonization) or hydrothermal carbonization. The former is conducted under the

pressure generated by gaseous decomposition products or external pressurization, while the latter indicates carbonization occurring under a pressurized atmosphere comprising water/solvent vapor [22].

Direct pressure carbonization. Direct pressure carbonization is performed without liquid addition and involves pressure controlled by programmed temperature variations [51]. The direct pressure carbonization of plastic waste (including PP, PE, PS and PVC) can be accomplished using this technique with or without catalysts [34]. In the absence of a catalyst, carbon spheres are typically formed by carbonizing plastic waste having specific proportions of various components (such as N, O, Cl-containing molecules and hydrocarbons) under relatively high pressures (over 30 MPa) [52,53]. In the presence of a catalyst, PWCMs with different nanostructures, including CNTs and carbon nanosheets, can be obtained. Ni, Fe, Co and Mg-based catalysts are widely used to convert plastic waste into target carbon nanomaterials in conjunction with this technique [34,54,55]. As an example, Ma et al. [56] used MgO as the catalyst for the direct pressure carbonization of a PP/LDPE/HDPE/PS/PET/PVC mixture. Porous carbon sheets were obtained and the

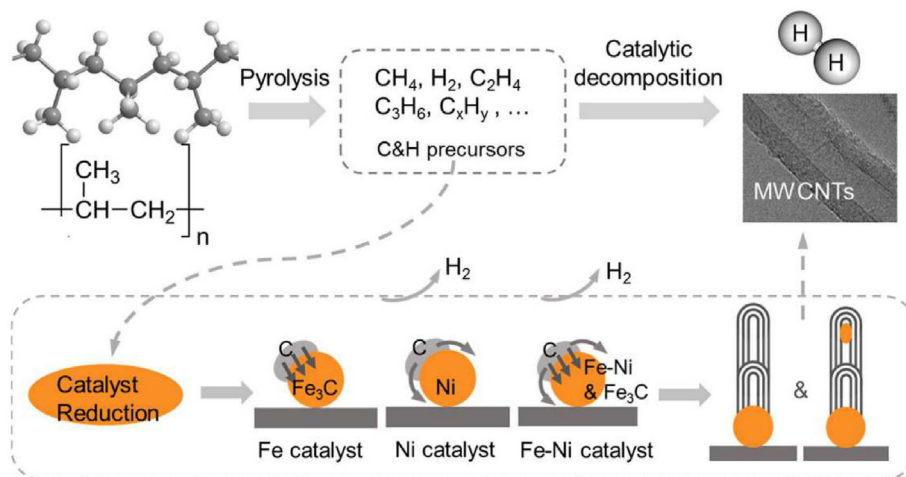


Fig. 1. Diagram of the catalytic carbonization of polypropylene with an Fe/Ni catalyst. Reprinted with permission from Ref. [48] (Copyright 2020 Elsevier).

MgO/plastic mass ratio was found to affect the properties of the product. The maximum surface area ($713 \text{ m}^2/\text{g}$) and pore volume ($5.27 \text{ cm}^3/\text{g}$) were obtained at ratios of 4 and 6, respectively.

Hydrothermal carbonization. In hydrothermal carbonization, waste plastics such as PVC or PP are suspended in aqueous media within an autoclave and subjected to elevated temperatures ($>100 \text{ }^\circ\text{C}$) and pressures ($>0.1 \text{ MPa}$) for a relatively long time span (several to tens of hours) [22]. Both high temperature (over $250 \text{ }^\circ\text{C}$) and low temperature (up to $250 \text{ }^\circ\text{C}$) treatments can be performed. Lower temperatures tend to generate hydrochar while higher temperatures can give graphite, activated carbon, CNTs and other higher value products. The solvent can also effect the physical properties of the product [57]. Hu et al. [58] used H_2O_2 solutions as the reaction solvent to synthesize photoluminescent carbon nanoparticles from waste plastic bags. The nanoparticle size could be precisely regulated by changing the H_2O_2 concentration, with a higher H_2O_2 concentration giving smaller particles.

2.4. Flash Joule heating

Flash Joule heating or carbothermal shock has emerged as an efficient means of converting plastic waste into graphene. In this process, the plastic is heated to a high temperature ($>2700 \text{ K}$) in a relatively short time frame (approximately 100 ms) to obtain a high yield ($>90\%$) of high-quality turbostratic graphene [59]. The experimental equipment required for this process is quite simple, as shown in Fig. 2a, and the treatment involves applying a direct current or alternating current. The conductivity of the plastic waste is often increased by adding carbon black, to ensure suitable Joule heating [59,60]. Tour et al. [35,59–62] fabricated a series of graphene products from plastic waste and other low-cost carbon-based solid wastes (such as biomass and coal). The energy input required to convert plastic waste to graphene was determined to be approximately 23 kJ/g (Fig. 2b) [35], suggesting that this technique would be economically viable on an industrial scale. One issue related to the flash Joule heating technique is that the gaseous products generated by this process have not yet been examined in detail [60].

2.5. Microwave conversion

Similar to flash Joule heating, the microwave conversion/pyrolysis of plastic wastes can be completed in a shorter time than is required when using traditional heating techniques [36]. Microwave treatment often generates multiple products, including carbon-based solids, gaseous products and/or bio-oils [63,64]. The yield of the former is governed by

the reaction temperature, with higher pyrolysis temperatures resulting in decreased char yields as thermal cracking is enhanced [63]. Jie et al. [36] employed low-cost iron-based catalysts/microwave susceptors to initiate the catalytic deconstruction of mixed plastics (Fig. 2c) and reported efficient conversion to CNTs and hydrogen gas ($55.6 \text{ mmol/g}_{\text{plastic}}$). From these data, it is apparent that microwave processing enables the transformation of plastic feedstocks into high-value carbon materials and hydrogen fuel.

Studies have suggested that mixed plastic waste can be co-treated to obtain target carbon materials [36]. However, one should keep in mind that the thermal transformation behavior of plastics is greatly affected by their chemical structures and thermal treatment protocols. Thus, the conversion efficiency of mixed plastic waste can be improved by optimizing the waste composition, by using catalysts (in the case of catalytic thermal processes) and by controlling the thermal treatment conditions (such as the time, temperature and atmosphere) [65].

3. Applications of PWCMS to green energy

Recent studies have found that many PWCMS are redox-active. Implementing electroactive carbon materials obtained from plastic waste in electrochemical energy devices would significantly reduce the cost of such items and could lead to sustainable green energy systems. In this section, the applications of PWCMS and PWCMS-based composites in batteries, supercapacitors and water electrolysis are discussed. The structure-performance relationship is emphasized to highlight the importance of design in different high-performance electroactive materials.

3.1. Batteries

Lithium-ion batteries and sodium-ion batteries are efficient electrochemical energy storage devices and have played a critical role in today's portable electronic devices [66,67]. To meet the performance demands for large-scale energy storage, low-cost electrodes allowing the rapid storage/release of energy and exhibiting high storage capacities with long cycle lifespans are required [68]. Plastic waste has been used to synthesize carbon materials with applications as anodes, cathodes and separators in different battery systems.

Anodes. Non-graphitic carbon materials (that is, amorphous, hard or soft carbon) are suitable as anode materials for alkali-ion batteries and can form energetically stable insertion compounds with lithium/sodium [69]. In this regard, plastic waste has been extensively utilized to develop

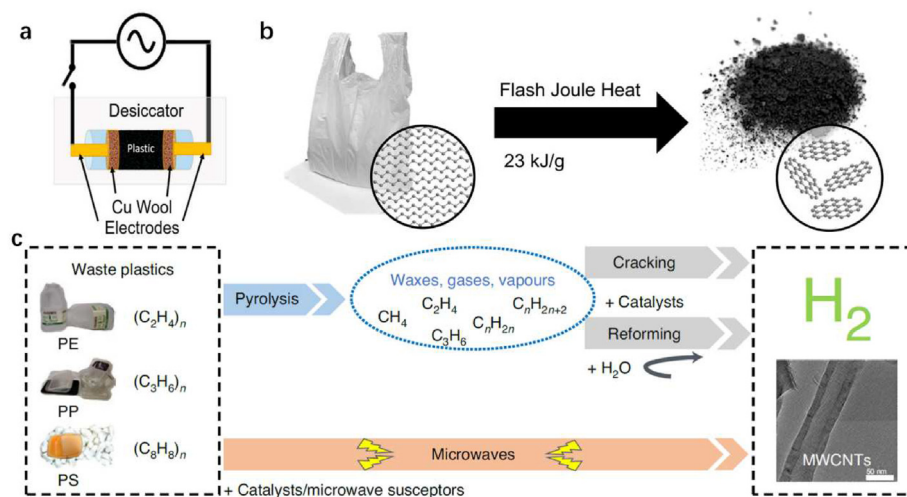


Fig. 2. (a) Diagram of a flash Joule heating set-up (120 V AC circuit). (b) Illustration of the conversion of plastic waste into graphene. Reprinted with permission from Ref. [35] (Copyright 2020 American Chemical Society). (c) Diagram showing the conversion of mixed plastic waste into hydrogen gas and multiwalled carbon nanotubes (scale bar: 50 nm). Reprinted with permission from Ref. [36] (Copyright 2020 Springer Nature).

porous carbon-based anode materials with high reversible storage capacities for alkali ions (such as Na^+ and Li^+). These materials have included activated [69], turbostratic [70,71], soot [72], disordered [73] and hard carbons [74]. Of note, a recent study converted disposable PP masks into non-graphitizable carbon powders via a sulfuric acid treatment followed by pyrolysis (Fig. 3a) [74]. During this process, the stabilization/sulfonation step facilitated crosslinking as well as the formation of polyaromatic hydrocarbons. The hard carbon anode made by this process exhibited an excellent reversible capacity of approximately 340 mAh/g (at a current rate of 0.01 A/g) for Na^+ together with a superior rate capability. Carbon-based composites derived from waste plastic are also promising anode materials [75,76]. In prior work, Wei et al. [75] prepared silicon/carbon nanofiber/carbon composites for use in lithium-ion batteries from waste HDPE and micron-sized silicon particles. The pyrolyzed carbon and carbon nanofibers in this material acted as conductive networks and also limited the volumetric expansion of Si during the Li^+ insertion/deinsertion processes, thus enhancing the anode performance.

Cathodes. PWCMS have also exhibited good performance as cathode materials in different battery systems, including metal-air and metal-sulfur batteries [77,78]. The oxygen reduction reaction (ORR) is a critical process in metal-air batteries and earth-abundant carbon electrocatalysts are extensively used to promote this reaction [79]. The ORR capacity of carbon-based materials can be improved by constructing carbon-metal structures. Cai et al. [77] found that Fe/N-codoped CNTs prepared from waste plastics via pyrolysis at 850 °C showed better ORR performance than Fe-doped CNTs and a commercial Pt/C catalyst. Although the relationship between pyrolysis temperature and carbon structure has been explored, minimal information has been obtained regarding the structure-performance correlation. Compositing carbon with metal materials has been examined, such as in the case of

CaCr_2O_4 /carbon nanoplatelets [78] and NiFe alloy NP/oxidized CNTs composites [80]. The combination of metals and carbon appears to provide a synergistic effect with regard to ORR performance. As an example, in the case of the NiFe alloy NP/oxidized CNTs composite described above, the co-existence of the Fe–Ni alloy, Fe_3C and oxygen-containing groups provided superior electrocatalytic performance while the protective coating of oxidized CNTs on the Fe–Ni alloy enhanced the durability of the material. Carbon materials derived from plastic waste can additionally act as sulfur hosts at the cathode in metal-sulfur batteries. The outward diffusion of lithium polysulfides from the cathode interface can be limited by ensuring chemical affinity and physical blockages based on building hierarchically porous structures and/or heteroatom doping (such as with Ti, O, N or S) [81,82].

Separators. In the development of high-performance metal-sulfur batteries, modifying separators with carbon materials is an efficient approach to limiting the shuttling effect of polysulfides while enhancing redox kinetics and boosting sulfur utilization efficiency [83,84]. Porous carbon materials having large surface areas are suitable for use as separators. In a previous study, negatively charged porous sulfonated carbon synthesized from LDPE was found to inhibit the diffusion of S_n^{2-} as a consequence of repulsive interactions during the discharge process (Fig. 3b) [85]. Similarly, using highly porous carbon materials derived from pyrolyzed plastic waste as the separator can upgrade the performance of lithium-sulfur batteries. Typically, the numerous polar sites (such as metal oxides) and multiple dopants (B or N) on the carbon promote the trapping of lithium polysulfides and improve the redox reaction kinetics [15].

Waste PET has also been used to produce terephthalic acid that was subsequently employed to synthesize disodium terephthalate/single walled CNT composites [86]. An anode made from this material showed good specific discharge capacity (241 mAh/g at 0.1 C) and stable cycling

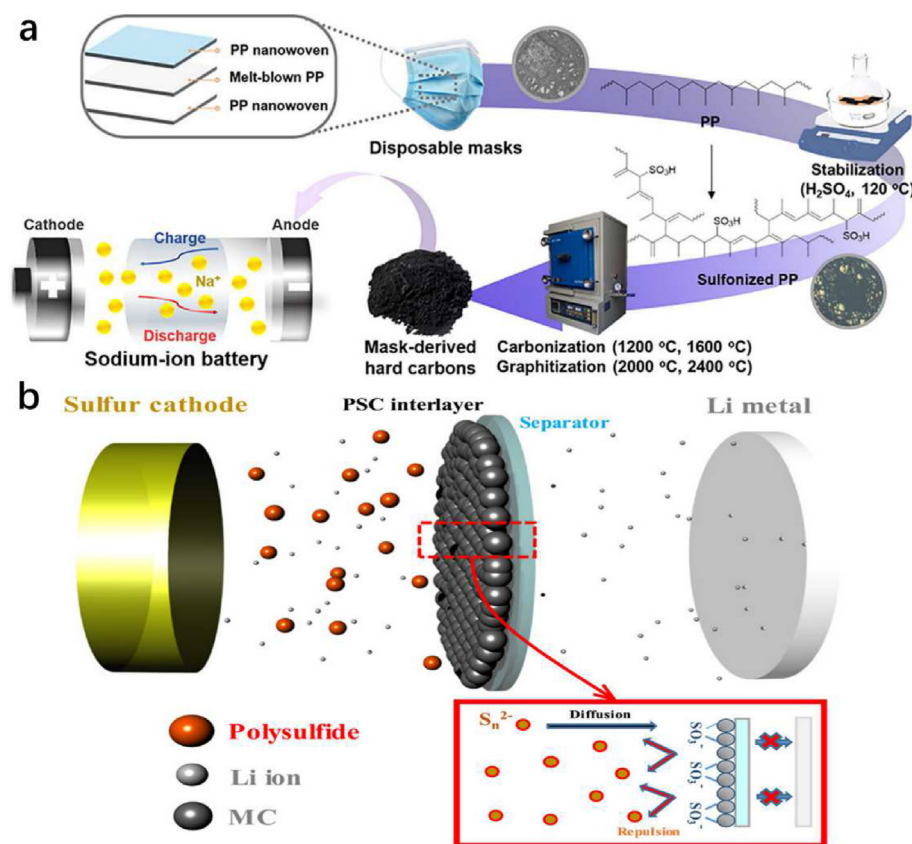


Fig. 3. (a) The synthesis of non-graphitizable carbon powders from polypropylene masks. Reprinted with permission from Ref. [74] (Copyright 2020 Elsevier). (b) Diagram showing the role of a porous sulfonated carbon/separator during the discharge process. Reprinted with permission from Ref. [85] (Copyright 2020 American Chemical Society).

(98.5% coulombic efficiency for 50 cycles) in a Na-ion battery due to the high conductivity and more accessible transmission routes provided by the single walled CNTs. The conversion of plastic waste into small molecules could be used to allow the large-scale production of organic electrode materials for next-generation Na-ion batteries.

3.2. Supercapacitors

Supercapacitors are widely implemented in energy storage devices because they provide high specific power densities, good cyclic stability and long lifespans, rapid charge/discharge and broad working temperature ranges [87]. These devices tend to operate based on one of two charge-storage mechanisms: electrostatic double-layer capacitance and pseudocapacitance. The physicochemical properties of the electrode in the unit (including porosity, chemical composition, SSA and morphology) have a large effect on performance [88]. Previous research has indicated that doping with heteroatoms such as N, O and S can enhance the conductivity, wettability and pseudocapacitance of PWCs, thereby improving the electrochemical performance of such materials [89]. PWCs with distinct structural and compositional features have been used for supercapacitors, including CNTs, carbon nanosheets/flakes, HPC and carbon-based composites.

CNTs. One-dimensional CNTs have been successfully obtained from plastic waste via catalytic carbonization [90,91]. Wen et al. [65] found that a combination of PP, PE and PS could be efficiently upgraded based on carbonization to produce high-quality CNTs in good yields. In addition, CNTs synthesized from waste have outperformed commercial CNTs and carbon black when used in supercapacitors. This improved performance can be attributed to the relatively larger SSA and higher concentrations of oxygen-bearing functional groups of the synthesized CNTs. The multiwall CNTs prepared from waste PP have also exhibited high electrochemical capacity, with a specific capacitance of 59 F/g in 1 N

KOH [90].

Carbon nanosheets. Two-dimensional (2D) carbon nanosheets/flakes have attracted attention owing to their high SSAs, low densities and significant stability [92,93], with graphene being a representative material. Starting from plastic waste, Karakoti et al. [94] synthesized graphene nanosheets comprising only a few layers using montmorillonite clay as a template. Similarly, MgO and Mg(OH)₂ have been employed as templates to convert waste PS into porous carbon flakes [95,96]. As a means of further increasing the SSA of 2D carbon, Wen et al. [92] applied a KOH activation treatment after the catalytic carbonization process. The resulting porous carbon nanosheets featured a high SSA (2236 m²/g) and a large pore volume (3 cm³/g) that synergistically contributed to good supercapacitive capacities (135 F/g in 1 M Na₂SO₄ and 121 F/g in 1 M tetraethylammonium tetrafluoroborate/propylene carbonate).

Hierarchical porous carbon. HPC materials have high SSA values and large pore volumes that promote the exposure of electroactive sites and the penetration/diffusion of electrolytes. The tuning of porosity in these materials has attracted particular interest. Liu et al. found that the co-etching of sp²/sp³ hybridized carbon during the CO₂ activation process promoted HPC formation from waste PET (Fig. 4a) [97]. This material exhibited a large pore volume of 0.51 cm³/g and a high SSA of 2238 m²/g together with a high capacitance of 413 F/g. KOH is another common activator used to provide porosity [16,45,98,99]. Porous structures can be formed as a result of KOH etching, K intercalation and CO₂/CO release (Fig. 4b) [51]. It is worth noting that the KOH concentration determines the generation of the porous structure, the internal crystal structure and the surface chemistry of the carbon product.

Carbon-based composites. Carbon-based hybrids that combine the beneficial properties of various materials are also promising electroactive materials for supercapacitors. Carbon/graphene and carbon/metal oxides originating from plastic waste have been used as electrode materials in supercapacitors [40,95,100,101]. In addition, porous N-doped carbon

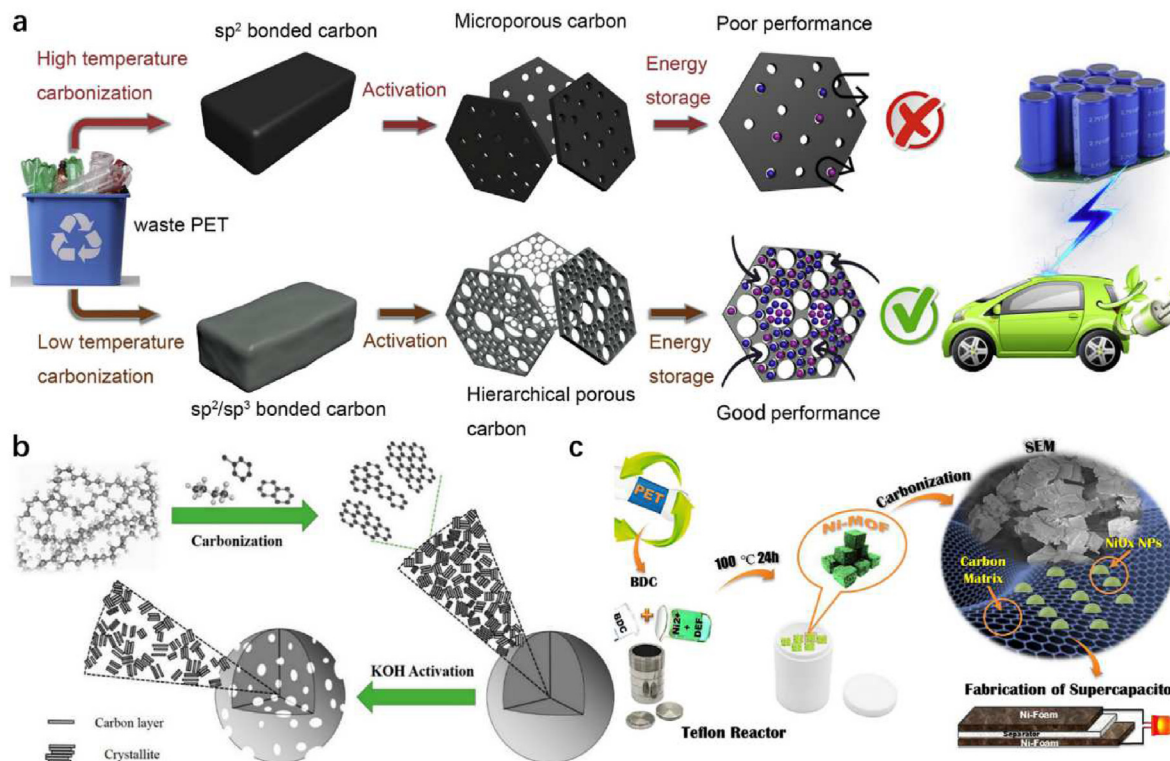


Fig. 4. (a) Synthesis of hierarchical porous carbon from waste polyester terephthalate, using CO₂ as the activation agent. Reprinted with permission from Ref. [97] (Copyright 2020 Elsevier). (b) The autogenic pressure carbonization and KOH activation of waste low-density polyethylene. Reprinted with permission from Ref. [51] (Copyright 2020 American Chemical Society). (c) The synthesis of a NiO_x NP-decorated porous N-doped carbon composite. Reprinted with permission from Ref. [18] (Copyright 2020 Elsevier).

loaded with NiO_x NPs prepared from PET-derived metal-organic frameworks has shown a high specific capacitance of 581.3 F/g (at a scan rate of 5 mV/s in 6 M KOH) with good cycle stability (Fig. 4c) [18]. The high SSA of this material (1523 m²/g) together with its mesoporous structure, high conductivity and excellent electroactivity conferred by the NiO_x all contributed to its exceptional performance.

3.3. Water electrolysis

The development of hydrogen-based energy system is an important aspect of achieving carbon neutrality [102–104]. Currently, water electrolysis is recognized as a practical route to producing high purity green hydrogen [105–107]. The oxygen evolution reaction (OER) and hydrogen evolution reaction (HER) are involved in the water-splitting process and the efficiency of electrolysis is largely determined by the electrocatalytic performance of the associated catalysts. A high-performance catalyst should possess a low overpotential, a small Tafel slope and good stability. To further reduce the fabrication cost of catalysts, much effort has been devoted to designing efficient catalysts from earth-abundant substances, such as minerals and solid wastes [108–116]. PWCMs and PWCMs-based hybrids provide large SSAs, high conductivities, suitable porosities and favorable surface chemistries (such as by incorporating dopants) and so have been applied as HER and OER catalysts.

HER catalysts. Carbon materials are typically used as HER catalysts, although the direct application of PWCMs as HER catalysts is seldomly reported. This can possibly be ascribed to the poor catalytic performance of such materials, such as a current density of 50 mA/cm² at an overpotential of 1 V [117], and so some studies have focused on designing carbon/metal composites to obtain better HER performance. Pandey et al. [17,118–120] made a series of carbon-metal carbide hybrids from waste plastics as potential HER materials. In this prior work, nitrogen present as a dopant in the carbon matrix enhanced the HER activity and the stability of molybdenum carbide/carbon composites [119]. In addition, amorphous surface carbon on the molybdenum carbide showed greater stability than graphitic material in acidic media [17]. Catalysts synthesized at relatively high pyrolysis temperatures also exhibited enhanced HER activity as a consequence of better graphitization of the carbon [118]. Carbon/sulfide composites are also attractive as HER catalysts [121,122]. A carbon-supported MoS₂ hybrid was prepared via a hydrothermal process based on waste plastics (including melamine (CWM) and polyurethane) as carbon precursors [121]. The MoS₂/CWM composite possessed a high carbon content and showed similar HER activity (a low overpotential of 56 mV at 10 mA/cm² and a small Tafel slope of 36.6 mV/dec) to a Pt/C catalyst. A high nitrogen content enhanced the conductivity of the MoS₂ nanosheets and the presence of pyridinic-N provided additional electroactive sites for the HER. Stability is another key factor that governs the application of electrocatalysts [123]. The MoS₂/CWM composite exhibited high stability based on linear sweep voltammetry curves acquired before and after 2000 cycles, which were almost equivalent. This stability was attributed to the strong interactions between the MoS₂ and CWM.

OER catalysts. OER catalysts based on PWCMs have received less attention. Gao et al. [124] proposed an eco-friendly strategy for the preparation of a MnO_x/S-doped graphitized carbon composite. In this material, MnO_x particles were evenly distributed on the carbon sheets and the composite had a mesoporous structure. These structural features contributed to a large SSA and provided numerous active sites and thus good OER activity (including an overpotential of 390 mV at 10 mA/cm² and a small Tafel slope of 67 mV/dec). However, the stability of this material was not examined. Recent studies have focused on the design of bifunctional electrocatalysts for water electrolysis. Ubaidullah et al. [125] designed a N-doped mesoporous carbon functionalized with ZnO from PET waste. As a consequence of the high SSA (939 m²/g) and mesoporosity (pore radius of approximately 25 Å) of this composite, it exhibited good HER activity (an overpotential of 390 mV at 10 mA/cm²

and a Tafel slope of 108 mV/dec) and decent OER activity (an overpotential of 570 mV at 10 mA/cm² and a Tafel slope of 318 mV/dec) in alkaline media. The stability of this catalyst was also suitable although only a short test duration of 300 s was applied. Thus, composites derived from plastic waste show promise as bifunctional catalysts for water splitting but the activity of these materials requires further improvement.

4. Applications of PWCMs to sustainable environmental processes

Environment sustainability is a significant concern in modern society and many techniques based on low-cost materials have been devised as approaches to addressing environmental issues. PWCMs have been applied in diverse sustainable environmental techniques such as pollutant adsorption, pollutant degradation, solar evaporation and CO₂ capture because of their low cost, flexible surface chemistry, large SSAs and complex porous structures.

4.1. Pollutant adsorption

Adsorption is a practical approach to removing pollutants from contaminated water because this method provides high efficiency, cost-effectiveness, facile implementation and adaptability [126–128]. The pollutant removal performance when using this technique is highly dependent on the physicochemical properties of the adsorbent (such as nanostructure, surface functional groups, SSA and wettability) and the nature of the pollutants. Here, applications of PWCMs and PWCMs-based composites to the adsorption of organic pollutants (including dyes and antibiotics) and inorganic contaminants are discussed.

Organic pollutants. PWCMs having distinct nanostructures have been applied as adsorbents for the removal of organic contaminants. Porous carbon [129–131], CNTs [132], carbon nanosheets [133] and graphene [134] are representative high-performance adsorbents. Often, electrostatic attraction, pore filling, hydrogen bonding and π - π interactions are involved in the adsorption of organic pollutants by PWCMs (Fig. 5a) [135]. Thus, efforts have been made to optimize these factors by tuning porosity and composition. As an example, Li et al. [130] synthesized porous carbon through the co-pyrolysis of waste PP and cyanobacteria and this material exhibited a high methylene blue (MB) removal capacity of 667 mg/g. During the pyrolysis process, the PP provided an improved carbon pore structure and numerous functional groups (such as C=O, O-H and N-H) that increased the adsorption performance. KOH activation is another effective means of regulating the porosity of PWCMs [133]. The use of solid KOH as an activator has been found to provide more heterogeneous porosity (macropores: 1.77 cm³/g; mesopores: 0.81 cm³/g; micropores: 0.71 cm³/g) during the fabrication of activated carbon. This technique previously generated a unique pore structure and a high surface area of 1990 m²/g that led to improved pollutant uptake [136].

Coupling PWCMs with other functional nanomaterials (such as metal oxides or alginates) can enhance their adsorption capacity [37,137]. The presence of these additives provides new active metal sites for the adsorption of organic pollutants via chemical bonding [137,138]. Rai et al. [139] found that the Fe-O groups in Fe₃O₄ acted as nucleation site for the enhanced adsorption of cephalixin by magnetic PET-based activated carbon. An additional advantage of magnetic PWCMs-metal oxide composites is that they can be readily separated from the reaction solution, thus facilitating the recycling and reuse of the adsorbent.

Inorganic pollutants. PWCMs have also been used to remove inorganic contaminants, especially heavy metal ions [140,141]. In such cases, oxygen-containing functional groups are the primary adsorption sites [142]. In addition to C=O and O-H groups, Xu et al. [143] showed that the presence of -SO₃H groups on hydrochar obtained from PVC improved the adsorption capacity for Cu(II) and Cr(VI) ions (Fig. 5b). The removal of Cu(II) ions was governed by electrostatic interactions while Cr(VI) anions were likely complexed with phenolic -OH groups and reduced to

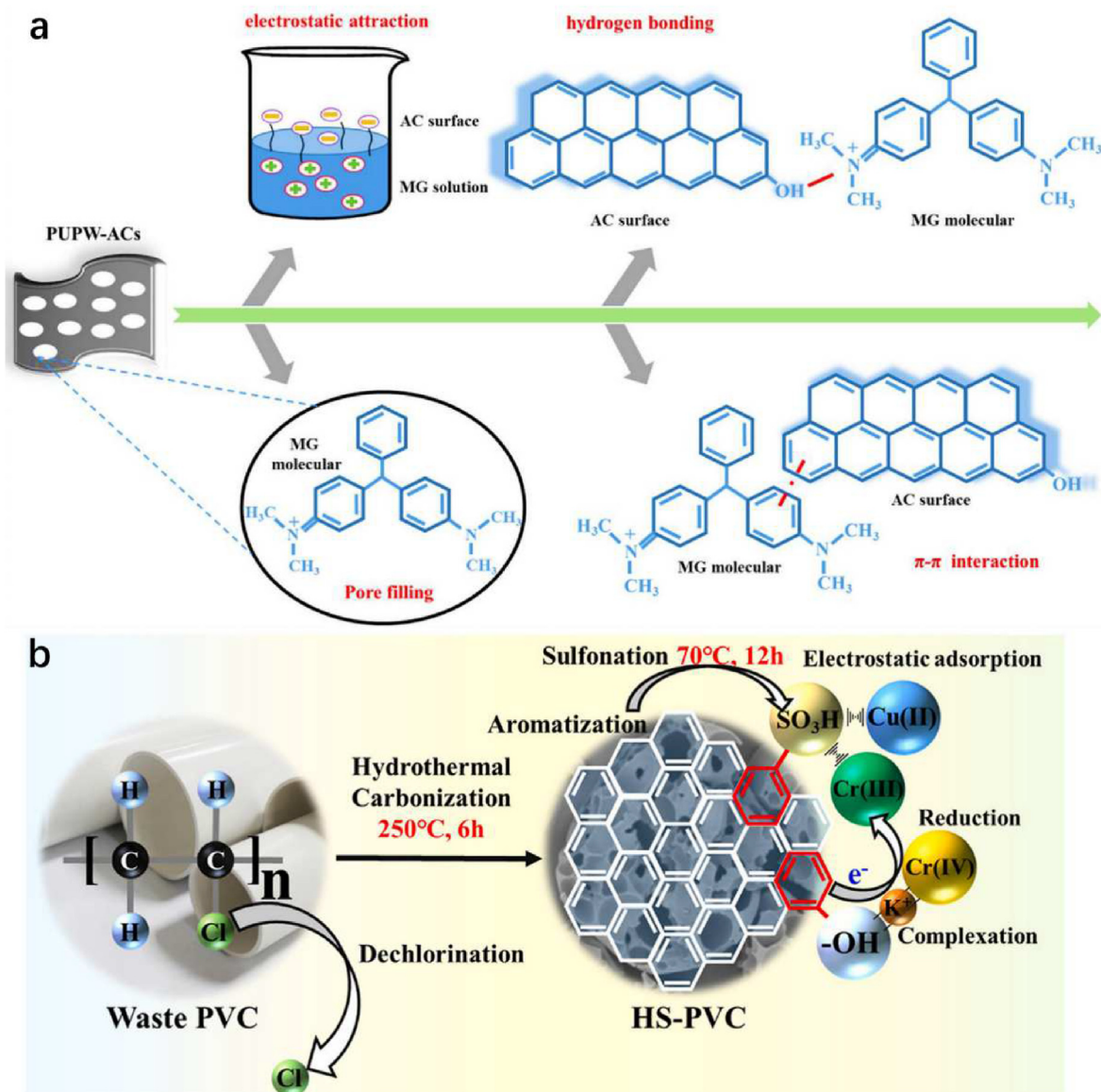


Fig. 5. (a) The adsorption of malachite green dye on activated carbon derived from polyurethane waste. Reprinted with permission from Ref. [135] (Copyright 2020 Elsevier). (b) Diagram summarizing the synthesis of PVC-derived hydrochar and application to the adsorption of Cu(II) and Cr(VI) ions. Reprinted with permission from Ref. [143] (Copyright 2020 Elsevier).

Cr(III) ions by C=C groups on the hydrochar surface. A polypyrrole-modified PET-derived carbon composite was found to efficiently remove NO₃⁻ ions from solution via ion exchange and electrostatic attraction processes [144]. Interestingly, the main binding sites responsible for electrostatic interactions were nitrogen-containing functional groups. Other studies have indicated that the adsorption of inorganic pollutants by PWCMS is mainly governed by surface functional groups, and so generating surface oxygen or nitrogen-containing groups could enhance the adsorption performance toward cations or anions, respectively.

4.2. Pollutant degradation

The degradation of pollutants can mitigate various negative effects on the environment and PWCMS and PWCMS-based composites have been used as catalysts for the oxidation degradation of many pollutants in contaminated water.

Advanced oxidation processes (AOPs). Based on utilizing active radicals (such as hydroxyl (•OH) and sulfate (SO₄^{•-}) radicals) as oxidizing species,

AOPs have been widely used in wastewater treatment because of their high efficiency, low energy consumption, simple operational processes and universal applicability to various pollutants [145]. PWCMS-based catalysts have also been shown to activate persulfate (S₂O₈²⁻) and hydrogen peroxide (H₂O₂) to produce active radicals. In one prior study, sulfur-doped activated carbon derived from waste poly(phenylene sulfide) was applied to S₂O₈²⁻ activation on the basis of both surface-bound radical and non-radical pathways [146]. Composites derived from PWCMS are also capable of activating persulfate to generate SO₄^{•-} and •OH radicals [147,148]. Starting from waste PE and goethite, Kwon et al. [148] developed an iron/carbon composite via co-pyrolysis (Fig. 6a). This material exhibited the efficient degradation of amaranth via reactions with SO₄^{•-} and •OH radicals formed from S₂O₈²⁻ activation. A Cu/carbon hybrid directly fabricated from waste printed circuit boards was also reported to degrade different dyes (including acid orange 7 and orange II) through the action of •OH radicals [149,150], based on an adsorption-degradation process. As depicted in Fig. 6b, small carbon particles effectively adsorbed acid orange 7 and the activation of H₂O₂ by Cu⁰ contributed to the generation of a high concentration of •OH

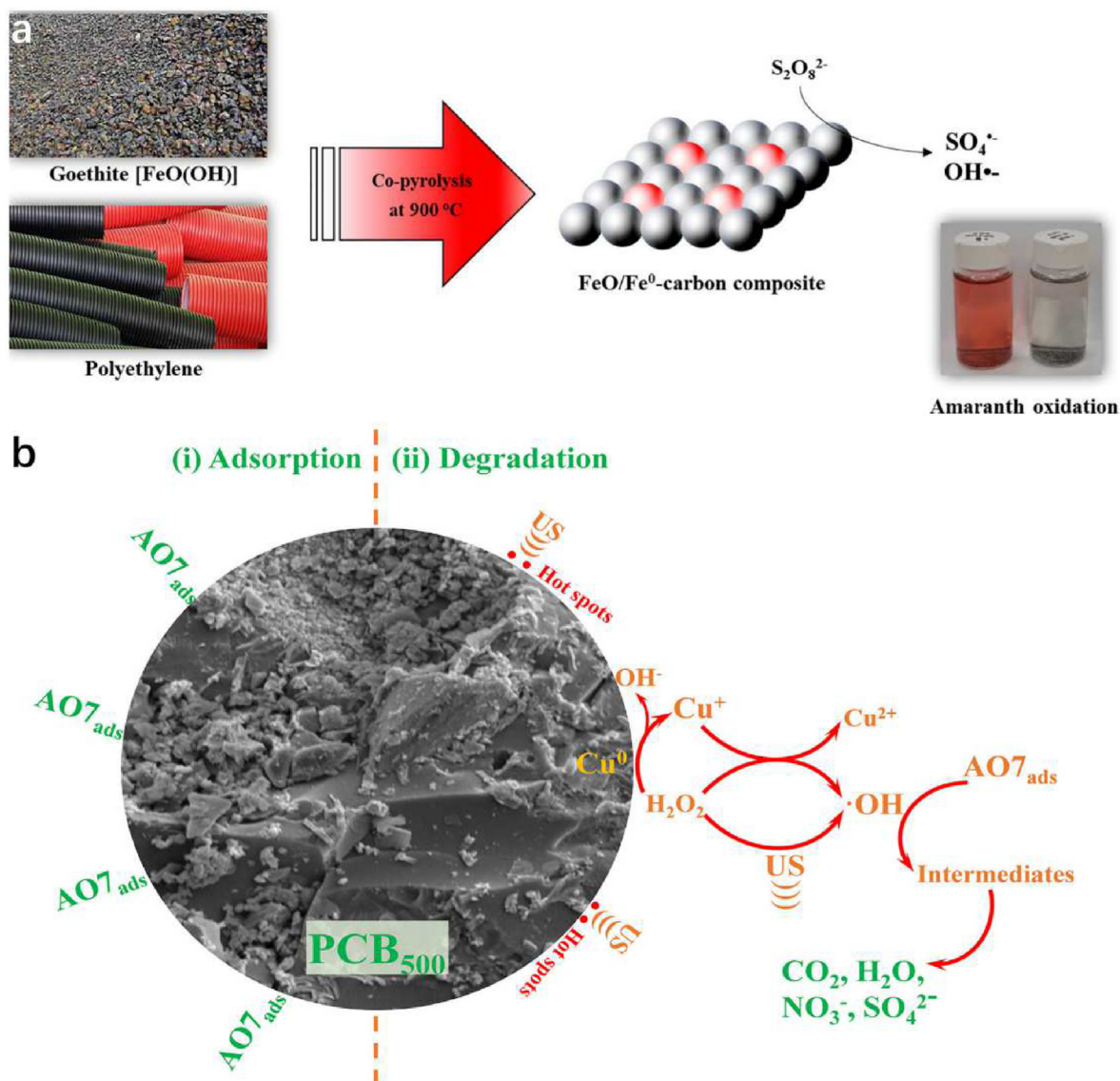


Fig. 6. (a) The synthesis of an iron/carbon composite and its application to amaranth oxidation. Reprinted with permission from Ref. [148] (Copyright 2020 Elsevier). (b) Diagram showing the degradation of acid orange 7 by the Cu/carbon composite. Reprinted with permission from Ref. [150] (Copyright 2020 Wiley-Blackwell).

radicals. The production of •OH radicals could be further enhanced by ultrasonic irradiation [150]. Thus, the Fenton-like system achieved a high degradation efficiency of 93.27% within 90 min.

Photocatalytic degradation. Photocatalysis is an appealing strategy that uses renewable solar energy for contaminant degradation. When photocatalysts are exposed to the appropriate light wavelengths, excited electrons (e^-) and holes (h^+) will be generated [151,152]. The photo-generated h^+ are strong oxidizers and can rapidly degrade pollutants. Several important free radicals (such as •OH) are also produced and can breakdown pollutants into smaller non-toxic or less toxic compounds, including CO₂ and H₂O [153,154]. Several reports have suggested that PWCMS-metal oxide composites can serve as photocatalysts for organic compound degradation [155,156]. The addition of PWCMS can tune the optical properties of the composite and enhance the dispersion of metal oxides. A NiO-MWCNT composite having a bandgap of 3.02 eV was obtained from waste HDPE and was found to degrade MB under visible light. In-depth investigations suggested that the main active radicals were •OH and O₂^{•-}, both of which could react with MB to provide a high decolorization percentage of 92.51% [155].

4.3. Solar evaporation

Solar evaporation is a promising water purification technique intended to convert non-drinkable water into fresh water using solar energy [157]. Solar evaporators with high light absorbance characteristics, rapid water and vapor transport, low heat conduction and minimal fabrication costs are necessary for efficient solar-driven water purification [158]. PWCMS having large SSAs and porous structures may fulfill these criteria. Zhang et al. [159] developed a controlled carbonization technique that generated HPC from waste PET for solar-powered water evaporation (Fig. 7a). During the preparation process, ZnCl₂/NaCl salts were used as porogens to create macropores and mesopores. The product showed a hierarchical porous structure and high SSA together with efficient solar power absorption, rapid water transportation and low thermal conductivity. These properties resulted in a high evaporation rate (1.68 kg/(m²·h) under 1 sun irradiation) and good energy conversion efficiency (97%). Aside from the contribution of the nanostructure of such materials, the surface chemistry also affects the solar evaporation performance. Nitrogen dopants in porous carbon can provide a source of

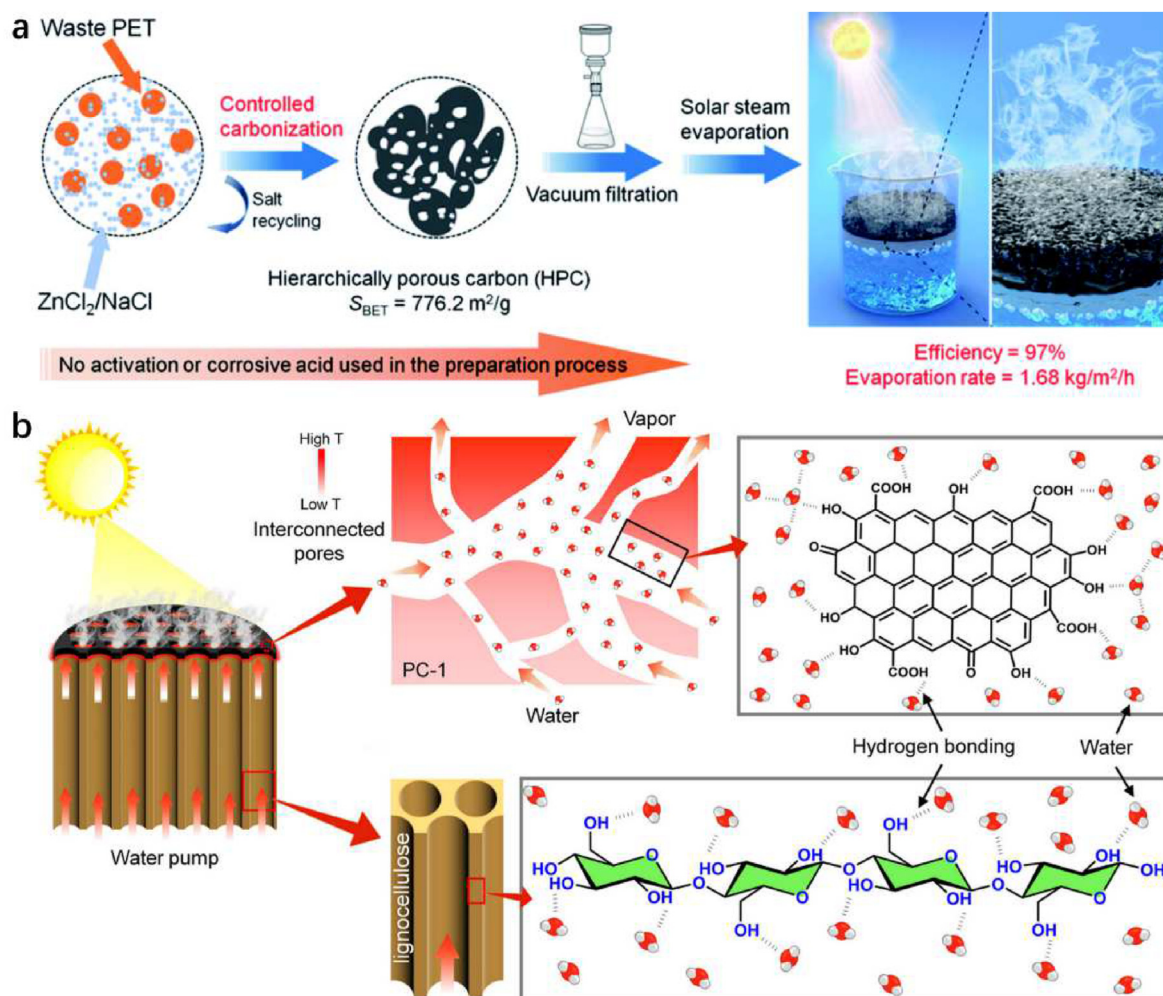


Fig. 7. (a) The controlled carbonization of waste PET using $\text{ZnCl}_2/\text{NaCl}$ molten salts as the catalyst and porogen to fabricate hierarchical porous carbon for solar steam evaporation from seawater. Reprinted with permission from Ref. [159] (Copyright 2020 The Royal Society of Chemistry). (b) Diagram summarizing the solar evaporation mechanism providing interfacial solar steam generation in a bilayer solar evaporator. Reprinted with permission from Ref. [161] (Copyright 2020 Wiley-Blackwell).

electrons that both improves the light absorption properties of the material and reduces the thermal conductivity while lowering heat loss to improve the photothermal conversion efficiency [160]. A high SSA and suitably porous structure also contribute to enhanced CO_2 adsorption capacity.

Another means of improving solar evaporation performance is the design of PWCMs-based hybrids. Starting from PP and NiO, Song et al. [158] synthesized a Ni/carbon nanocomposite consisting of pear-like metallic Ni NPs and cup-stacked CNTs. It is noteworthy that the synergistic effect of the Ni NPs and CS-CNTs in terms of solar absorption facilitated the photothermal conversion process. Taking advantage of the naturally aligned tubular structures in wood, the same group developed a bilayer solar evaporator by coating the wood surface with waste PET-derived porous carbon. This system provided three dimensional microchannels that ensured a continuous water supply and lowered the water evaporation enthalpy by forming hydrogen bonds with water molecules (Fig. 7b). In addition, this porous carbon reduced heat losses by heating the water/air interface, while the many interconnected macro-/meso-/micropores ensured a sufficient water supply for solar vapor generation. The abundant oxygen-containing functional groups on the material also formed numerous hydrogen bonds with water molecules and so promoted the formation of water clusters in the nanopores [161]. These features of the bilayer solar evaporator together contributed to its high performance.

4.4. CO_2 capture

Using PWCMs for CO_2 capture is a promising and sustainable route to simultaneously addressing the urgent plastic waste challenge while limiting global warming. Carbon capture in this manner provides an eco-friendly strategy to close the carbon loop and realize carbon neutrality [19,162]. Considering the large proportion of N_2 in the air, the selective separation of CO_2 from N_2 is required and a representative experimental set-up for CO_2 separation by PWCMs is shown in Fig. 8a [162]. In this process, CO_2 can be selectively captured by the activated carbon, which can be further concentrated for treatment after the desorption process.

The CO_2 capture performance of PWCMs largely depends on the nanostructure/textural properties and surface chemistry of the material. Generally, PWCMs with a large SSA, high porosity and narrow pore size distribution are favorable for CO_2 adsorption [163,164]. These textural features can be obtained by physical (such as by using CO_2) or chemical (including the use of KOH, H_2SO_4 , H_3PO_4 , ZnCl_2 and CH_3COOK) activation [164–168]. Recently, Algozeeb et al. [164] used CH_3COOK as the activator to construct a porous carbon material with pore widths of 0.7–1.4 nm that showed high CO_2 capture efficiency (3.80 ± 0.25 mmol/g at 25 °C). In addition to optimizing textural properties, doping with heteroatoms such as N, O or S can enhance the chemisorption of CO_2 via Lewis acid-base interactions between the functional groups on the absorbent and CO_2 [39]. Oxygen-containing groups (–OH and –COOH),

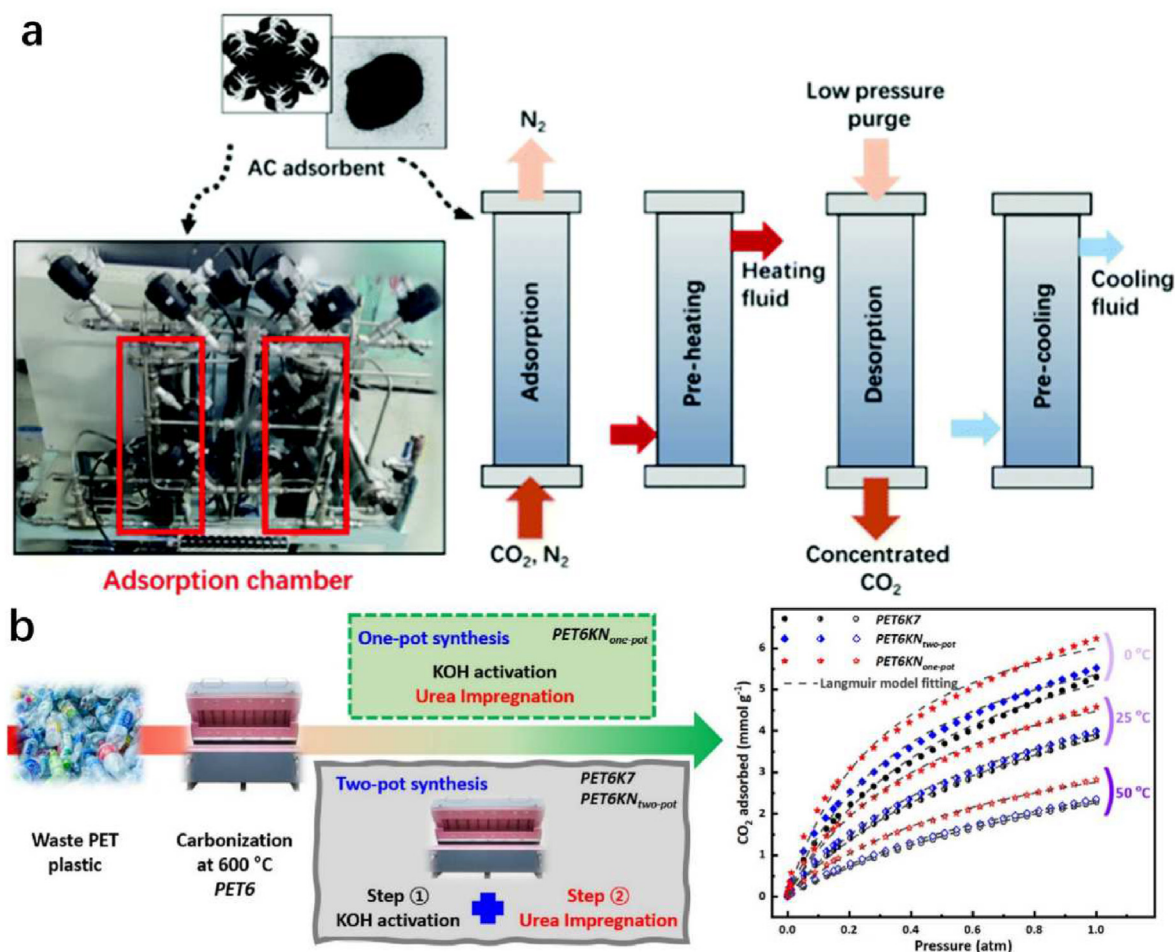


Fig. 8. (a) The selective adsorption of CO₂ by PWCMs (AC: activated carbon). Reprinted with permission from Ref. [162] (Copyright 2020 The Royal Society of Chemistry). (b) Diagram showing the synthesis of N-doped porous carbon materials from waste PET and application to CO₂ adsorption. Reprinted with permission from Ref. [172] (Copyright 2020 Elsevier).

pyrrolic N and oxidized S groups are effective at improving CO₂ adsorption performance [19,169–171]. Using waste PET bottles as the precursor, Yuan et al. [172] manufactured a N-doped microporous carbon based on a one-pot synthesis method (Fig. 8b). Compared with carbon materials synthesized by a two-pot process comprising separate KOH and urea activation steps, this material showed a higher proportion of O and N (22.03% vs. 10.34%) and thus better CO₂ adsorption performance. The authors also found that CO₂ adsorption on the PWCM product was an exothermic process, as the CO₂ uptake amount decreased with increasing temperature.

5. Conclusions and outlook

Converting plastic wastes into carbon-based functional materials represents a step forward toward a circular economy. In this review, the synthesis and applications of PWCMs were examined. It is evident that plastic waste can be transformed into diverse carbon materials via anoxic pyrolysis carbonization, catalytic carbonization, pressure carbonization, flash Joule heating and microwave conversion. Compared with conventional carbonization, flash Joule heating and microwave conversion can produce carbon materials in a shorter time span with high efficiencies. PWCMs have been successfully applied in green energy devices (such as batteries, supercapacitors and water splitting) and in sustainable environmental systems (pollutant adsorption/degradation, solar evaporation and CO₂ capture). The performance of PWCMs can be enhanced by

regulating their nanostructure (especially porosity) and surface chemistry (based on using dopants or forming functional groups). Notwithstanding the outstanding achievements to date, several critical issues remain that require further consideration. These are as below.

- (1) The most widely used conventional carbonization methods generally require long preparation periods and high energy inputs. Alternatively, flash Joule heating and microwave conversion can rapidly produce high-quality carbon materials (including graphene and CNTs) and so deserve more attention. In future, the process optimization and large-scale application of these rapid conversion methods could accelerate the upcycling of plastic wastes.
- (2) Catalysts are important to the conversion of plastics into carbon materials. The rational design of low-cost catalysts for the selective production of PWCMs and value-added chemicals fuels is highly recommended. In addition, monitoring the *in situ* transformation of catalysts during the reaction process is essential to understanding the related catalytic mechanisms.
- (3) Although PWCMs and PWCMs-based composites have shown good performance in a wide range of applications, the mechanisms of adsorption and catalysis are still unclear, often because the structures of most PWCMs are complicated. Therefore, integrated experimental techniques (such as X-ray photoelectron spectroscopy, X-ray absorption spectroscopy and transmission

electron microscopy) and computational methods (including molecular dynamic simulations and density functional theory calculations) are required to resolve the crystal structures of PWCMs. Such data would provide an improved understanding of the related reaction mechanisms on an atomic scale.

- (4) The design of next-generation PWCMs-based functional materials will require several factors to be considered. First, it is important to engineer the porosity of PWCMs and activators are required to tailor the nanostructures of PWCMs-based functional materials. Second, it will be vital to take advantage of the flexible surface chemistry of PWCMs to functionalize the carbon materials with specific functional groups for selected applications. Third, efficient methods of fabricating composites of PWCMs with other functional materials (including metal oxides, single-atom catalysts and 2D transition metal sulfides) to achieve high performance will be required.
- (5) The structural stability and environmental risks of PWCMs should be emphasized. As an example, the breakdown of PWCMs and PWCMs-based composites may lead to metal leaching and nano-sized particles. Once released into soil, air and water, these PWCMs-derived substances could generate secondary pollution. Additionally, PWCMs can react with other species and generate new pollutants in the environment. These issues will be important to the actual applications of PWCMs. Therefore, it is important to assess the stability of PWCMs and PWCMs-based composites in real world scenarios. Another issue is the complete recovery of PWCMs and PWCMs-based composites after their usage.
- (6) For the capture of CO₂ from mixed gases, it appears beneficial to functionalize PWCMs with metals (such as Mg, Al and Ca) to form adsorptive sites that enhance the adsorption selectivity and capacity. In addition, the adsorption mechanisms of CO₂ by PWCMs should be further explored using computational and experimental techniques to elucidate the adsorption processes on the molecular level.
- (7) In addition to the aforementioned energy and environmental applications, it is highly important to use a wide variety of PWCMs in other emerging fields, such as CO₂ reduction, water pollutant electrochemical degradation, sensors and industrial gas purification. The wider application of PWCMs would be expected to mitigate the current plastic waste challenge and reduce the fabrication costs of functional materials.

Author contributions

Zhijie Chen: conceptualization, writing-original draft; **Wenfei Wei:** writing-review & editing; **Bing-Jie Ni:** discussions, writing-review & editing; **Hong Chen:** supervision, writing-review & editing, discussions, project acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.efmat.2022.05.005>.

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