doi.org/10.1002/cnma.202400409

CHEMNANOMAT
Review

www.chemnanomat.org

Upcycling Plastic Waste into Valuable Carbon Nanomaterials

Ling Chi[a] and Muhammad Omar Shaikh*[a]

The global issue of plastic waste accumulation is now widely acknowledged as a significant environmental challenge that affects all aspects of life, economies, and natural ecosystems worldwide. Hence, it is crucial to develop sustainable solutions to traditional disposal methods. One promising solution involves upcycling plastic waste into valuable carbon nanomaterials such as carbon nanotubes, graphene, and carbon nanofibers, among others. This critical review provides an overview of the problems associated with plastics, including their various types and properties, as well as their significant impact on the environment and the methods currently employed for waste management. Furthermore, it delves into recent advancements in upcycling plastic waste into carbon nanomaterials through four state-of-the-art methods with the potential for scaling up and enabling industrial applications: thermal decomposition, flash joule heating (FJH), chemical vapor decomposition (CVD), and stepwise conversion. For each method, highly influential and seminal papers were selected, and their research approaches and observed results were thoroughly analysed. This upcycling approach transforms plastic waste into valuable resources, promoting a waste-to-value concept that reduces environmental impact and supports the circular economy. By creating new materials from discarded plastics, it addresses waste management challenges while generating economic value.

1. Introduction

1.1. Problems of Plastic Waste

The global production of solid waste encompasses a wide range of materials, such as plastics, woody biomass, food waste, tires, animal manure, and various combinations.[1] Among these waste types, plastic waste (PW) has gained significant and growing attention due to its extensive production scale and environmental repercussions.[2] Initially, the term "plastic" referred to flexible and easily molded materials. However, its definition has evolved over time to encompass a diverse array of materials known as polymers.[3] Nonrenewable crude oil is the primary raw material for manufacturing plastic products. While the first synthetic polymer was invented by James Wesley Hyatt in 1869, the demand for plastic for military requirements skyrocketed during World War II. In multiple applications, plastics have replaced materials such as wood, metal, and glass. They have been used in a variety of products ranging from packaging to medical equipment and electronic devices owing to their advantageous properties such as affordability, lightweight, water resistance, malleability, and durability.[4,5]

The versatility of plastics has led to continuous growth in their production over the years. The production of plastic has seen a notable rise, surging from 1.5 million metric tons in 1950–390.7 million metric tons in 2021.[6] According to projections, our environment and landfills could significantly accumulate approximately 12 billion tons of PW by 2050.[7] In addition, the utilization of non-renewable resources for plastic production is expected to rise from 6% in 2014–20% by 2050, primarily driven by the substantial increase in plastic consumption.[8] The COVID-19 pandemic has exacerbated the predicament with a significant surge in the utilization of protective equipment, single-use utensils, and plastic-wrapped food, potentially causing a new environmental catastrophe.[9] The continuous production of plastics generates vast quantities of PW. The Organization for Economic Co-operation and Development (OECD) countries are responsible for 14% of total plastic leakage, including 11% from macroplastics and 35% from microplastics.[10] Around two-thirds of PW originates from plastics with a lifespan of fewer than five years, of which packaging accounts for 40%, consumables 12%, and clothes and textiles 11%.[10] By 2015, the total amount of PW produced had reached approximately 6300 Mt, as depicted in Figure 1. Out of this, only about 9% went through the process of recycling, 12% was burnt and the rest (79%) was deposited into landfills or released into the environment.[11]

Plastics make a substantial contribution to waste and pollution around the world. The accumulation of significant volumes of waste in landfills and oceans primarily results from the high durability of plastics. The primary focus in the design of most synthetic polymers is to achieve attributes of longevity and high performance, rather than prioritizing easy recyclability or biodegradability. As a consequence of this emphasis on durability and performance, plastics exhibit a high level of resistance to degradation, leading to significant waste accumulation in landfills and oceans.[12,13] The production of plastic also contributes to greenhouse gas emissions, and it is projected that by 2050, these emissions will reach 2.8 gigatons per year. In the United States, plastic manufacturing alone accounts for

---

[a] L. Chi, M. Omar Shaikh
Sustainability Science and Management, Tunghai University, Taichung 407224, Taiwan
Tel: +886 4 23590121 ext.39204
E-mail: omar@thu.edu.tw

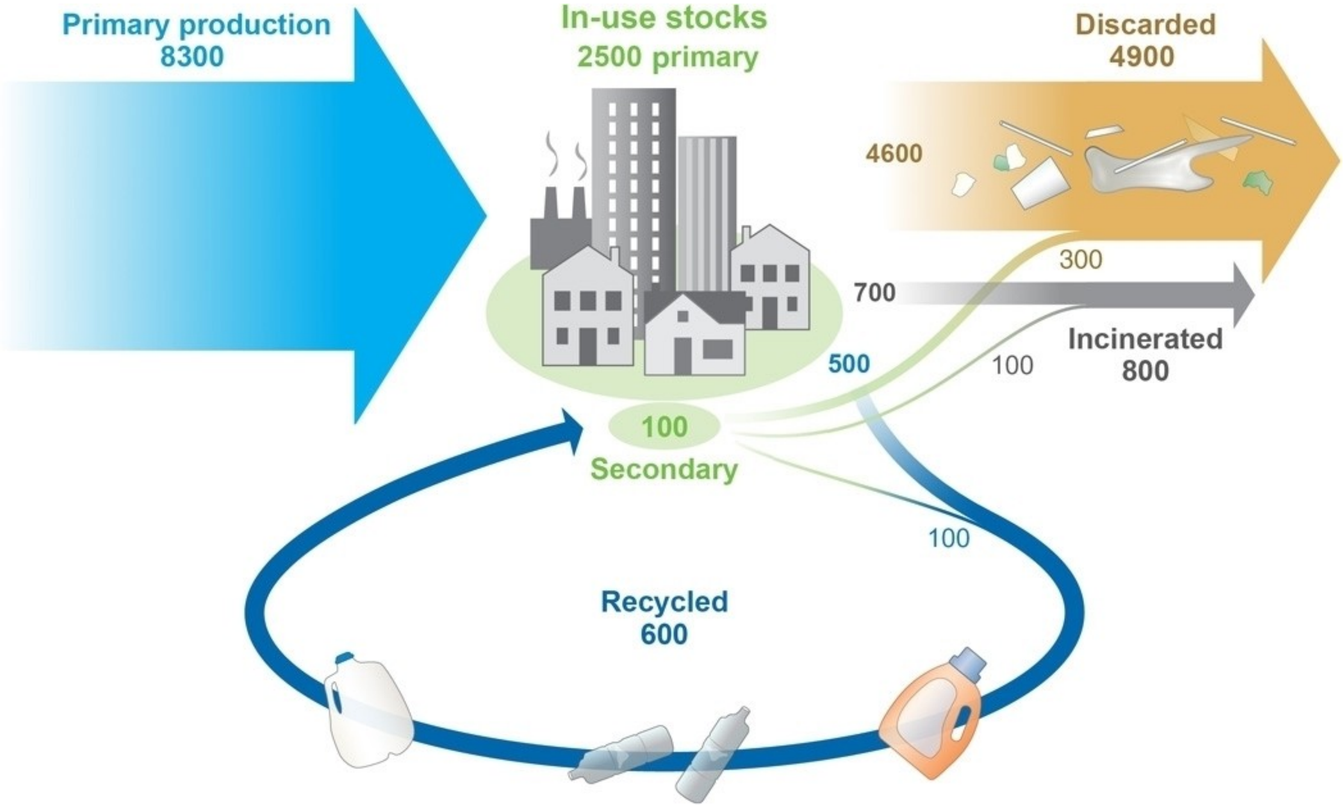


Figure 1. The global production, consumption, and outcomes of polymer resins, synthetic fibers, and additives between 1950 and 2015, were measured in millions of metric tons. Reprinted with permission from Ref..[11]

approximately 114 million tons of CO2 emissions annually.[14] This substantial amount of CO2 emissions underscores the environmental impact of plastic manufacturing in the US. Additionally, when using biomass, PW can produce significant side effects in terrestrial acidification and nutrient eutrophication of water resources.[15] The correlation between plastics and climate is apparent in their dispersal and impacts, with climate change expected to increase the dispersion of plastics in the natural environment through heightened occurrences of extreme weather events and flooding.[16]

Furthermore, plastic has detrimental effects on various environmental factors, including soil temperature and habitat suitability, disease prevalence in coral reefs, the rate of melting of polar ice, and human health.[17–20] These impacts are often considered “poorly reversible” due to the challenges in mitigating plastic emissions, contributing to modifications in habitats within both terrestrial and aquatic ecosystems, and posing a threat to keystone species.[21] Moreover, substantial quantities of plastic have been detected within the digestive systems of wildlife, spanning from minuscule Antarctic krill to immense sperm whales.[22,23] The entanglement of animals in

Lynn Chi holds a Bachelor’s degree in Sustainability Science and Management from Tunghai University. Her undergraduate research was centred on the development of sustainable cellulose-based supercapacitors utilizing nitrogen-doped graphene derived from recycled polyethylene terephthalate (PET) bottles. Currently, her research interests lie in the application of green chemistry principles and waste-to-value strategies, with a particular focus on integrating comprehensive characterization techniques and electrochemical measurements to advance energy storage technologies.

Muhammad Omar Shaikh is a Professor of Sustainability Science and Management at Tunghai University, Taiwan, and the director of Advanced NanoTech Laboratory (ANT Labs). He holds a BEng and MEng in Materials Science and Engineering from Imperial College London and a PhD in Mechatronic Science and Engineering from Southern Taiwan University of Science and Technology. Over the past decade, his research has centred on advanced nanotechnologies for healthcare and energy. His work includes innovations in biosensors for point-of-care diagnostics, tactile sensors for robotics, gas sensors, wearable energy harvesters, and technologies for solar fuels and CO_2 conversion and utilization.

ropes and fishing lines also presents a severe threat to numerous species, particularly rare ones like Australian sea lions.[24] Reports indicate that the ingestion and interaction with plastic can result in reduced body size, nutritional deficiencies, altered blood chemistry, and increased contamination levels.[25–28]

1.2. Types of Plastics

Plastics are macromolecules formed through polymerization, a process that chemically connects individual units of identical or different molecules to create long-chain structures.[29] These macromolecules can be shaped and modeled by applying pressure, heat, or other external forces. As a result of this polymerization process, plastics acquire properties that are distinct from those of the initial molecules.[30,31] Polymers are further classified into rubbers or elastomers, plastics, and fibers based on their specific properties and characteristics.[32,33] The two primary categories of plastics are thermoplastics and thermosets.

Thermoplastics exhibit a basic molecular structure comprising chemically autonomous macromolecules. Being subjected to heat, they can undergo softening or melting, allowing for shaping, forming, welding, and subsequent solidification upon cooling. These types of plastics can endure numerous cycles of heating and cooling without substantial damage, facilitating reprocessing and recycling procedures. The recycling process typically involves collecting, sorting, and cleaning used plastics before shredding, melting, and extruding them into pellets or flakes for manufacturing new products. Various additives or fillers can be incorporated to enhance certain properties like thermal or chemical stability, as well as ultraviolet resistance. Comprising approximately 80% or more of the overall plastic consumption, thermoplastics are exemplified by commodity types like polyethylene (PE), polyethylene terephthalate (PET), polypropylene (PP), polystyrene (PS), and polyvinyl chloride (PVC).[34]

Thermosetting plastics also consist of independent macromolecules. However, they possess a three-dimensional (3D) structure achieved through chemical cross-linking, either during processing or after. The cross-links formed between the chains of thermosetting plastics restrict their mobility and relative displacement, resulting in certain advantages and disadvantages. One advantage is their resistance to heat degradation as they cannot be melted. However, a significant drawback is that the cross-linking process is time-consuming, leading to longer production cycles often requiring heating.[33] Recycling thermosetting polymers is more challenging compared to recycling thermoplastics due to their irreversible chemical cross-linking, which makes them less amenable to melting and reprocessing. Thermosetting plastics account for approximately 12–20% of total plastic consumption. Examples of thermosetting plastics include urea-formaldehyde, phenol-formaldehyde, melamine-formaldehyde, and thermosetting polyester.[35]

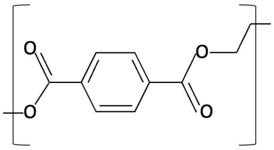
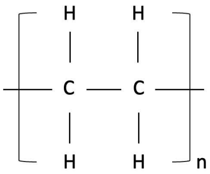
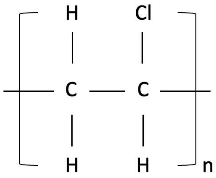
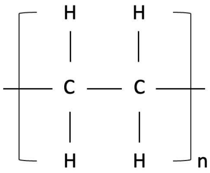
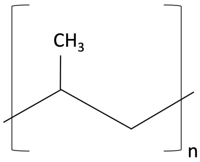
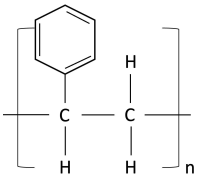
Thermoplastics constitute the most widely utilized plastics in our daily lives. Generally, PW comprises a blend of diverse polymers, such as PET, PE, PS, PP, and PVC.[36] Table 1 provides information on these polymers. To aid in identification, the Society of the Plastics Industry has assigned different recycling codes to each of them. PET, which is frequently used for containers, labels, and films, is assigned recycling code 1.[37] PE can be categorized as either high-density polyethylene (HDPE) or low-density polyethylene (LDPE), depending on the polymerization method, the structure of the polymer chains, and molecular weight.[38] Resin code 2, HDPE, is represented by the chemical formula (C2H4)n, and is commonly found in milk bottles and oil containers.[39] Resin code 4 designates LDPE, characterized by a more branched molecular structure. This type of plastic is predominantly employed for the fabrication of plastic bags and wire insulation.[40] PVC, commonly found in water pipes, electrical wiring covers, and construction, is assigned recycling code 3.[41] Resin codes 5 and 6 signify PP and PS, respectively. PP is employed in the production of automotive bumpers, luggage, and medical containers, while PS is utilized for household appliances and food containers.[42] Under the resin code 7, a range of polymers is encompassed, such as polybutylene terephthalate, polylactic acid (PLA), multi-layered mixed polymers, polycarbonate (PC), nylon, acrylonitrile butadiene styrene, and acrylic.[43] PLA, recognized for its biodegradability and commendable mechanical attributes, finds application in diverse sectors including bone engineering scaffolds and drinking straws.[44] Belonging to the code 7 group, PC stands out as a notable plastic. Its inherent robustness and remarkable ability to endure high temperatures underscore its prominence as a leading material for engineering applications.[45]

1.3. Plastic Waste-to-Value

In order to tackle the problem of PW and reduce environmental pollution, PW is usually incinerated or disposed of in landfills. However, while incineration can significantly reduce the volume of PW by up to 90–99%,[46] it also generates harmful by-products such as furnas, hydrocarbons, polycyclic aromatic, and monoxide.[47] Additionally, the residual ash generated by incineration contains microplastics which pose a serious threat to the ecosystem. Consequently, these concerns drive the rapid growth of PW recycling research.

There are three primary approaches to plastic recycling: mechanical, biological, and chemical. Each approach involves a distinct process for breaking down PW and transforming it into new products. Mechanical recycling involves sorting, washing, drying, shredding, and re-granulating plastics to obtain secondary plastics. Currently, this method is the primary means of disposing of mass PW. Nonetheless, the secondary plastics generated through this method exhibit limited mechanical strength and unpredictable rheological characteristics, rendering them unsuitable for application in fields such as packaging and medicine.[48] The recycling procedure also involves considerable expenses in terms of labor and electricity, encompassing tasks like sorting, shredding, cleaning, and melting. Another approach to PW recycling is biological recycling, which involves cultivating microorganisms such as fungi and bacteria or

Table 1. The molecular structures and applications of commonly used plastics.

Plastic types	Resin identification code	Molecular structures	Density (g/cm^3)	Melting point (°C)	Final products
PET	♳		1.38	≥250	Plastic bottles, food containers, jars, cups, food trays
HDPE	♴	 $\left[-\overset{\displaystyle H}{\underset{\displaystyle H}{C}}-\overset{\displaystyle H}{\underset{\displaystyle H}{C}}-\right]_n$	0.941–0.96	130	Plastic bottles, chemical containers, pipes, toys
PVC	♵	 $\left[-\overset{\displaystyle H}{\underset{\displaystyle H}{C}}-\overset{\displaystyle Cl}{\underset{\displaystyle H}{C}}-\right]_n$	1.38	100–260	Construction, water, pipes, electrical wiring covers, windows
LDPE	♶	 $\left[-\overset{\displaystyle H}{\underset{\displaystyle H}{C}}-\overset{\displaystyle H}{\underset{\displaystyle H}{C}}-\right]_n$	0.91–0.925	115	Plastic bags, wire insulation, plastic film
PP	♷		0.855	130–171	Reusable water bottles, medical containers, luggage
PS	♸		0.96–1.04	240	Packaging foam, household appliances, IT equipment
Others	♹				Eyeglasses, electronics, fiberglass, CDs/DVDs

utilizing enzyme extraction.[49] This method can only break plastics apart into oligomers or monomers with hydrolysable ester or amide bonds in the main chain.[50] Ultimately, chemical recycling employs either heat or chemical reactions to disintegrate PW into its fundamental building blocks or monomers. These resulting components can subsequently be utilized in the production of fresh products. Chemical recycling has garnered significant attention compared to mechanical and biological methods. This heightened interest stems from its unique ability to revert post-consumer waste (PW) back to its original monomers. This process enables the repolymerization of materials into virgin-grade substances without compromising the material's properties or its economic value.

Moreover, the integration of chemical, biological, and hybrid catalytic techniques, as illustrated in Figure 2, is actively being researched. These methods aim to break down, recycle, and upcycle plastics, playing a pivotal role in advancing plastic recovery efforts and tackling the global PW issue.[51] For PW that proves too expensive or impractical to recycle via traditional mechanical or chemical methods, the emerging field of upcycling shows great promise as illustrated in Figure 2B. This innovative approach harnesses chemical or engineering strategies to reposition plastic waste at the forefront of a new value

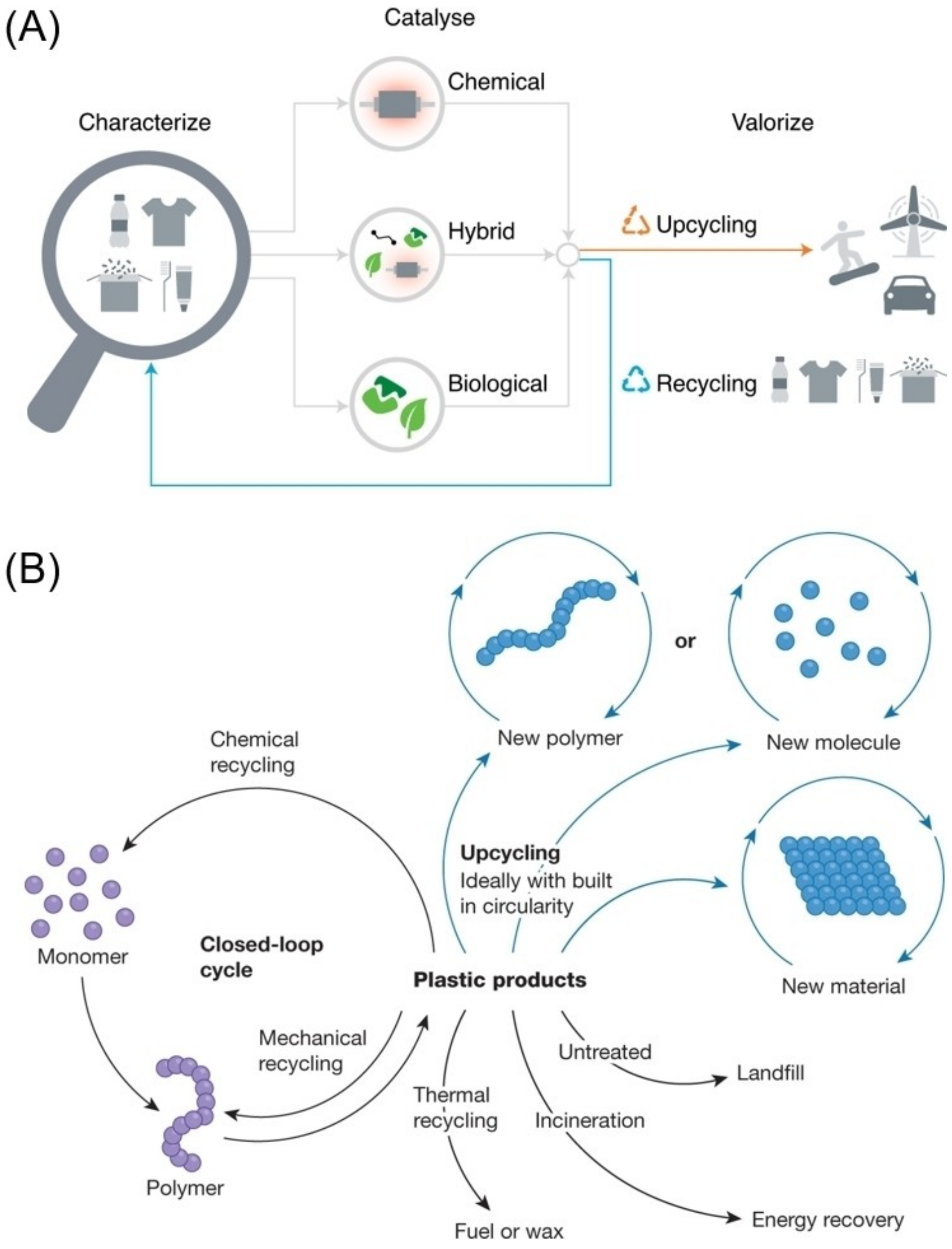


Figure 2. (A) Harnessing chemical, biological and hybrid processes to facilitate recycling and upcycling of plastic waste. Reprinted with permission from Ref..[51] (B) Upcycling represents a complementary approach that works in tandem with chemical and mechanical recycling methods, ultimately resulting in the production of entirely new polymers or materials. Reprinted with permission from Ref.[52]

chain. Within this framework, PW transforms into a readily available and cost-effective starting material for synthesizing new materials or molecules. Addressing the challenge of turning PW into materials that possess increased economic value remains a significant hurdle. This task involves intricate chemical, economic, and environmental considerations that are interconnected. Novel concepts are emerging that aim to tap into high-value markets within the circular economy for plastics and find inventive solutions for repurposing plastic waste into materials with elevated worth and utility.

One promising solution to tackle this issue involves upcycling PW into valuable carbon-based nanomaterials. This approach not only helps combat plastic pollution but also offers the additional advantage of reducing carbon emissions. Specifically, carbon-based nanomaterials or nanocarbons, including fullerenes, nanotubes, graphene, activated carbon nanoparticles, and nanodiamonds, possess unique properties that make them highly desirable. These properties include high mechanical flexibility, exceptionally large surface areas, low toxicity, biocompatibility, and the ability to modify their mechanical, electrical, thermal, optical, and chemical characteristics. Consequently, these nanocarbons find wide-ranging practical applications in various fields,[52–62] and some examples include:

- Water and Air Purification: Nanocarbons are effective water and air purification filtration agents. Because of their expansive surface area and adept adsorption capacities, they possess the ability to eliminate impurities, pollutants, and even microorganisms, thereby contributing to purer and safer water and air conditions.

- Composites: Nanocarbons are employed to enhance the properties of composites in industries such as aerospace, automotive, construction, and electronics. By incorporating nanocarbons into the composite materials, they can improve strength, durability, and conductivity.
- Conductive Coatings and Paints: Nanocarbons are utilized to add conductivity, corrosion resistance, and strength to coatings used in various applications, including electronics and aerospace. These coatings provide enhanced performance, protection, and longevity to the surfaces they are applied to.
- Chemical and Biosensors: Nanocarbons play a pivotal role in the advancement of robust sensors used in chemical and biosensing applications. These sensors enable the detection and analysis of substances in environmental monitoring, healthcare, and food safety, contributing to improved diagnostics and safety measures.
- Bioimaging and Drug Delivery: Nanocarbons are employed as imaging agents and targeted drug carriers in bioimaging and drug delivery systems. Their unique properties allow for better visualization of biological structures and precise delivery of therapeutic agents, enhancing diagnostic accuracy and therapeutic outcomes.
- Catalysts: Nanocarbons are used as catalysts to accelerate chemical reactions in various fields, including energy production, chemical synthesis, and environmental remediation. Their high surface area and unique surface chemistry make them efficient catalysts for promoting desired chemical transformations.
- Energy Harvesting and Storage: Nanocarbons find applications in energy storage and harvesting devices such as batteries, supercapacitors, solar cells, and fuel cells. Their remarkable surface area and exceptional conductivity facilitate enhanced energy storage capabilities, quicker charging rates, and higher energy conversion efficiency, contributing to the evolution of more efficient and sustainable energy systems.

The ongoing research and development of carbon-based nanomaterials from plastics has the potential to reshape technology, address global challenges, and advance sustainability efforts. By effectively converting PW into valuable resources, this approach presents a promising resolution to the environmental consequences of PW while unlocking new possibilities for innovation and circular economic growth. Embracing upcycling as a key strategy in waste management can pave the way for a more sustainable future.

2. Conversion of Plastic Waste to Carbon Materials

The utilization of PW as a raw material to produce value-added items and materials has emerged as a prospective tactic to augment the attractiveness of plastic recycling.[63–68] Given that plastic contains a substantial amount of carbon, transforming waste plastic into valuable carbon-based materials presents an ideal approach for waste management and carbon material production.[69–71] Consequently, a range of methodologies has been devised to produce high-quality carbon materials, including graphite, graphene, fullerene, activated carbon, and carbon nanotubes (CNTs), which find diverse applications across multiple fields.[72–76] This section will delve into the primary methods employed for converting PW into valuable carbon materials, namely thermal decomposition, flash joule heating (FJH), chemical vapor decomposition (CVD), and stepwise conversion. Each method will be discussed in detail in the subsequent subsections.

2.1. Thermal Decomposition

Thermal decomposition is a chemical process that occurs when a compound is exposed to high temperatures, leading to its breakdown into simpler chemical species. According to the American Society for Testing and Materials (ASTM), thermal decomposition involves significant chemical changes resulting from high temperatures.[77] In contrast, thermal degradation refers to the loss of physical, mechanical, or electrical properties that can happen when a material, product, or assembly is exposed to elevated temperatures. During thermal decomposition, solid materials generate combustible fuel vapours that can burn above the material.[78] This process can continue as long as the burning gases provide sufficient heat to the material, enabling the ongoing production of fuel vapours or volatiles. Thus, it can become a continuous feedback loop if the material keeps burning, ensuring the quality of the products obtained.

Among the various thermal decomposition processes for utilizing PW, pyrolysis is a promising solution. Pyrolysis is a process that thermally decomposes PW in an oxygenless environment to produce valuable chemicals in the form of pyrolytic gases and oils, as well as heat energy. One advantage of pyrolysis is its ability to handle various types of PW, including PS, PP, PE, HDPE, LDPE, and mixed waste plastics. Pyrolysis stands out from other recycling methods in that it doesn't require the raw materials to be isolated on the basis of their polymer composition, making it more convenient and cost-effective. Pyrolysis can be performed using different types of reactors and heating sources, such as electrical heaters or burners. The choice of reactor and operating conditions can impact the quality and yield of the pyrolysis products. For instance, with increased temperatures and residence times, the yield of pyrolytic oils can be improved while reducing the formation of char and gas. Conversely, lower temperatures and shorter residence times can favour the production of pyrolytic gases. However, a fundamental challenge in the thermal decomposition of waste plastics is the low thermal conductivity, which makes it challenging to uniformly distribute heat to the treated materials. This often leads to localized overheating of the reacting material and excessive precipitation of elemental carbon, which strongly adheres to the heating surfaces of the reactor. Table 2 presents various carbonization processes and their conditions for synthesizing carbon materials.

Thermal decomposition serves as an efficient and affordable technique to produce innovative carbon nanomaterials, including 3D sponge nitrogen-doped graphene (NG), from discarded PET bottles combined with urea at various temperatures through a straightforward and environmentally friendly one-step process, as depicted in Figure 2.[81] In this procedure, the initial PET waste was appropriately sized and screened before being mixed with urea at different ratios. Subsequently, the blend was introduced into a stainless-steel autoclave positioned within an electric furnace, and then subjected to a heating for duration of five hours, either at 600 °C or 800 °C. Following an overnight cooling period, the resultant dark products were accumulated and crushed, culminating in the production of NG with nitrogen atoms integrated into the graphitic lattice. To facilitate a comparison of capacitive performance, researchers also fabricated samples of pure graphene (G) specimens along with NG samples having varied ratios of urea to PET. The utilization of X-ray photoelectron spectroscopy (XPS) for analyzing the thermal decomposition process, with varying quantities of urea employed, revealed a reduction in the nitrogen content within the resultant samples as the quantity of urea was diminished. Cyclic voltammetry and impedance spectroscopy assessments disclosed that the fixation of nitrogen, impacting the structure and morphology of the synthesized graphene, amplified the transmission of charge and the diffusion of ions. Transmission electron microscopy (TEM) images displayed transparent, large, few-layered graphene exhibiting a laminar morphology, with embedded nitrogen atoms within the multi-layered and polycrystalline structure. High-resolution TEM images confirmed the presence of corrugated microstructures, while selected area electron diffraction patterns indicated the existence of wrinkled features. The urea and PET thermal decomposition process resulted in gas release, leading to the formation of a highly porous structure, as confirmed by morphological characterization. The resulting 3D porous framework facilitated ion transfer pathways and significantly enhanced the material's surface area, making it well-suited for electrochemical energy storage applications. Ultimately, this economically viable and eco-friendly approach facilitated the large-scale production of nitrogen-doped graphene (NG) from discarded plastic materials, thereby offering prospects for the upcycling of PW.

The production of carbon nanomaterials (CNMs) and hydrogen-rich gases as byproducts was also successfully achieved through the processes of pyrolysis and catalysis using different types of PW, as illustrated in Figure 3.[82] Two sets of catalysts were synthesized, employing Co, Fe, and Ni as active metals and MgO as the support material. The first set of catalysts was prepared using conventional impregnation (Co–Fe/MgO, Co–Ni/MgO, and Fe–Ni/MgO), while the second set utilized coprecipitation (Co–Fe-Mg, Co–Ni-Mg, and Fe–Ni-Mg). The experimental setup included a two-stage reactor: the first stage involved plastic pyrolysis and the generation of hydrocarbon volatiles, while the second stage focused on catalytic reforming. The performance of each bimetallic catalyst was evaluated by analyzing the resulting products, which included CNMs and high-value gases. Furthermore, a comprehensive comparison was conducted among products derived from four types of plastic: LDPE plastic bags, PP plastic bottles, PS plastic lids, and PET mineral water bottles. The comparison primarily considered factors such as yield, carbon purity, and adsorption capacity. The findings indicated that the Fe–Ni-Mg catalyst, prepared using the precipitation method with increased pH, exhibited the most effective conversion of plastic. Among the different plastic types, LDPE bags demonstrated a relatively high yield of hydrogen at 35.27 mmol/gplastic, while PS lids exhibited a relatively high yield of carbon nanomaterials at 38.26 wt %. The carbon nanomaterials derived from the plastics were employed as adsorbents for wastewater treatment and demonstrated a significant adsorption capacity (approximately 180 mg/gCNM) for metal cations such as Fe, Ag, and Ni (Figure 4).

2.2. Flash Joule Heating

Flash Joule heating (FJH) is an innovative method for synthesizing superior carbon materials, specifically aimed at generating high-quality carbon nanomaterials. By employing FJH, it becomes possible to convert waste plastic substances into substantial quantities of superior-grade turbostratic graphene by directing a discharge of direct current through the precursor materials using sizeable capacitors. Two variants of FJH are utilized for the production of turbostratic flash graphene (FG): direct current flash Joule heating (DC-FJH) and alternating current flash Joule heating (AC-FJH). This state-of-the-art method utilizes electrical energy to initiate fast joule heating (FJH) in PW, resulting in a rapid elevation of the carbon source's temperature within a brief period (Figure 5).

The conversion of carbon-based initial materials into substantial amounts of turbostratic graphene through the FJH process has been effectively accomplished by passing direct current discharge via large capacitors. However, challenges remain in the production of alternative carbon allotropes, such as nanodiamonds and concentric carbon materials. Likewise, there are still challenges in achieving the covalent functionalization of various carbon allotropes through the FJH process. In a previous study, researchers utilized a solvent-free FJH method to create three different fluorinated carbon allotropes: fluorinated nanodiamonds (FND), fluorinated flash graphene (FFG), and fluorinated concentric carbon (FCC), as shown in Figure 6.[83] This was accomplished by rapidly heating organic fluorine compounds and fluoride precursors for a short duration. The spectroscopic analysis confirmed changes in the electronic states and detected various short and long-range structural arrangements within the distinct fluorinated carbon allotropes. The correlation between the duration of the heating process and the resulting phases and compositions was demonstrated, with longer heating durations leading to the graphitization of FND into FCC. This study provides evidence that FJH is an effective, rapid, and cost-effective method for synthesizing fluorinated carbon allotropes in under a second

Table 2. Summary of carbonization processes for synthesizing carbon materials from PW.[79,80]

Carbonization processes		Conditions	Typical polymers	Carbon materials	Advantages and Disadvantages
Anoxic pyrolysis carbonization	No stabilization	• Carbonization temperature: 500–1000° °C • Atmosphere: inert or in molten salt • Activation	PET, PF resin	Amorphous carbons (activated carbon, mesoporous carbon, carbon fibers)	Advantages: • Simple setup process • Versatile for different plastics • Produces diverse carbon materials Disadvantages: • Energy intensive process • Low yields • Inconsistent quality
	Oxidated stabilization	• Carbonization temperature: 500–1000° °C • Oxidation temperature: 200–350° °C • Atmosphere: inert Activation	PAN, LDPE, PVC		
	Chemical stabilization	• Sulfonation or Friedel–Crafts reaction • Activation	PE, PS		
Catalytic carbonization		• Carbonization temperature: 400–900° °C • Atmosphere: inert, in the presence of metal compound catalysts	PE, PP, PS, PFR, PVC, PVDF, PTFE, PVA, PET	Graphitic carbons (carbon nanotubes, carbon nanofibers, carbon nanosheets, graphene, carbon spheres, carbon foam)	Advantages: • Produces high-quality carbon • Energy efficient operation (Catalytic carbonization) • Higher production yields (Catalytic pressure carbonization) Disadvantages: • Requires expensive catalysts • Complex process (Catalytic carbonization) • Safety risks (Catalytic pressure carbonization)
Catalytic pressure carbonization		• Carbonization temperature: 600–850° °C • Synthesis: in sealed reactor and in the presence of metal compound catalysts	PE, PP, PS		
Pressure carbonization	Pressurized atmosphere	• Carbonization temperature: 600–850° °C • Synthesis: in sealed reactor	PE, PP, PS, PVC	Amorphous carbons (activated carbon, carbon dots, carbon spheres)	Advantages: • Produces high-quality carbon • High yields • Efficient conversion Disadvantages: • Energy intensive process • Safety risks due to pressure • High operation costs
	Hydrothermal carbonization	• Carbonization temperature: 150–300° °C • Synthesis: in sealed reactor and in the presence of water vapor	PVC		

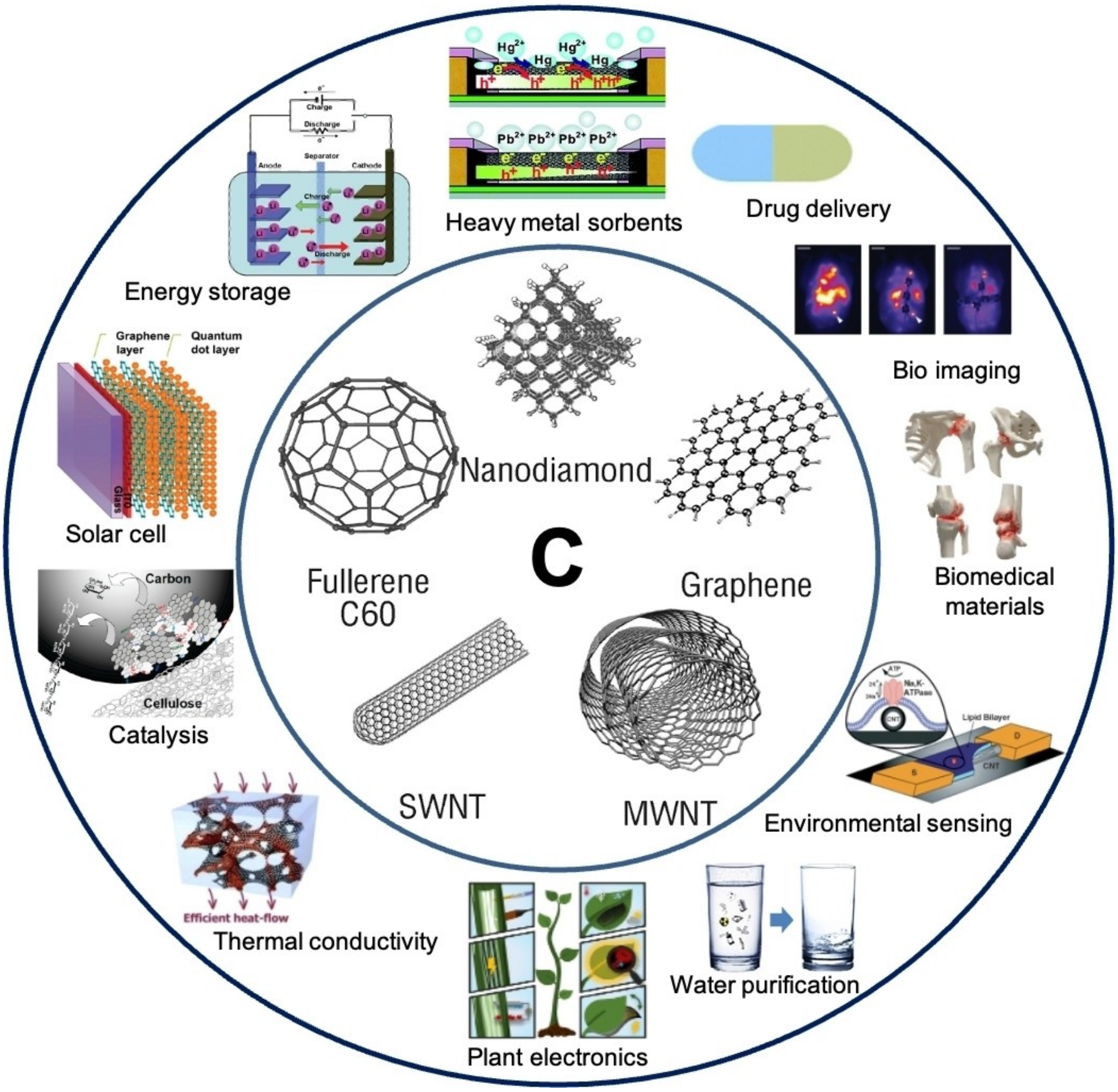


Figure 3. Diverse applications of Nanocarbons. Reprinted with permission from Ref..[52–62]

while allowing for customization of their properties and structures to meet diverse application requirements.

Another research study applied FJH as a technique for the conversion of PW into flash graphene (FG), as shown in Figure 7.[84] The process produced FG, carbon oligomers, hydrogen, and light hydrocarbons. In the pursuit of premium-grade graphene, a sequence involving alternating current (AC) and direct current (DC) flashes was employed. The sample underwent AC-FJH with an AC voltage of 120 V and frequency of 60 Hz for about 8 seconds in a vacuum desiccator for outgassing. Subsequently, DC-FJH was carried out on the specimens utilizing a capacitor bank comprising 10 capacitors having capacities of 450 V and 60 mF. These capacitors were charged to 110 V and then discharged for a duration of 500 ms, resulting in the attainment of enhanced quality of FG. The FJH method has been demonstrated to be effective in treating PW mixtures without a catalyst, rendering it appropriate for the management of PW in landfills. The required energy was approximately 23 kJ/g, equivalent to around $125 in electricity per ton of PW, suggesting economic viability for expansion. TEM imaging revealed that the FG has a turbostratic structure of 27 nm average size and 3.45 Å interlayer spacing, facilitating dispersion in liquids and composites. Raman spectroscopy showed FG with an I2D/IG peak ratio of up to 6 and a low-intensity D band. Fourier transform infrared spectroscopy characterized the carbon oligomers obtained, which exhibited resembles chemical structures to the initial PW. Gas-phase analysis indicated significant hydrogen and light hydrocarbon formation. Considering the naturally occurring and low-toxicity nature of graphene, this method represents an eco-beneficial approach to recycling PW.

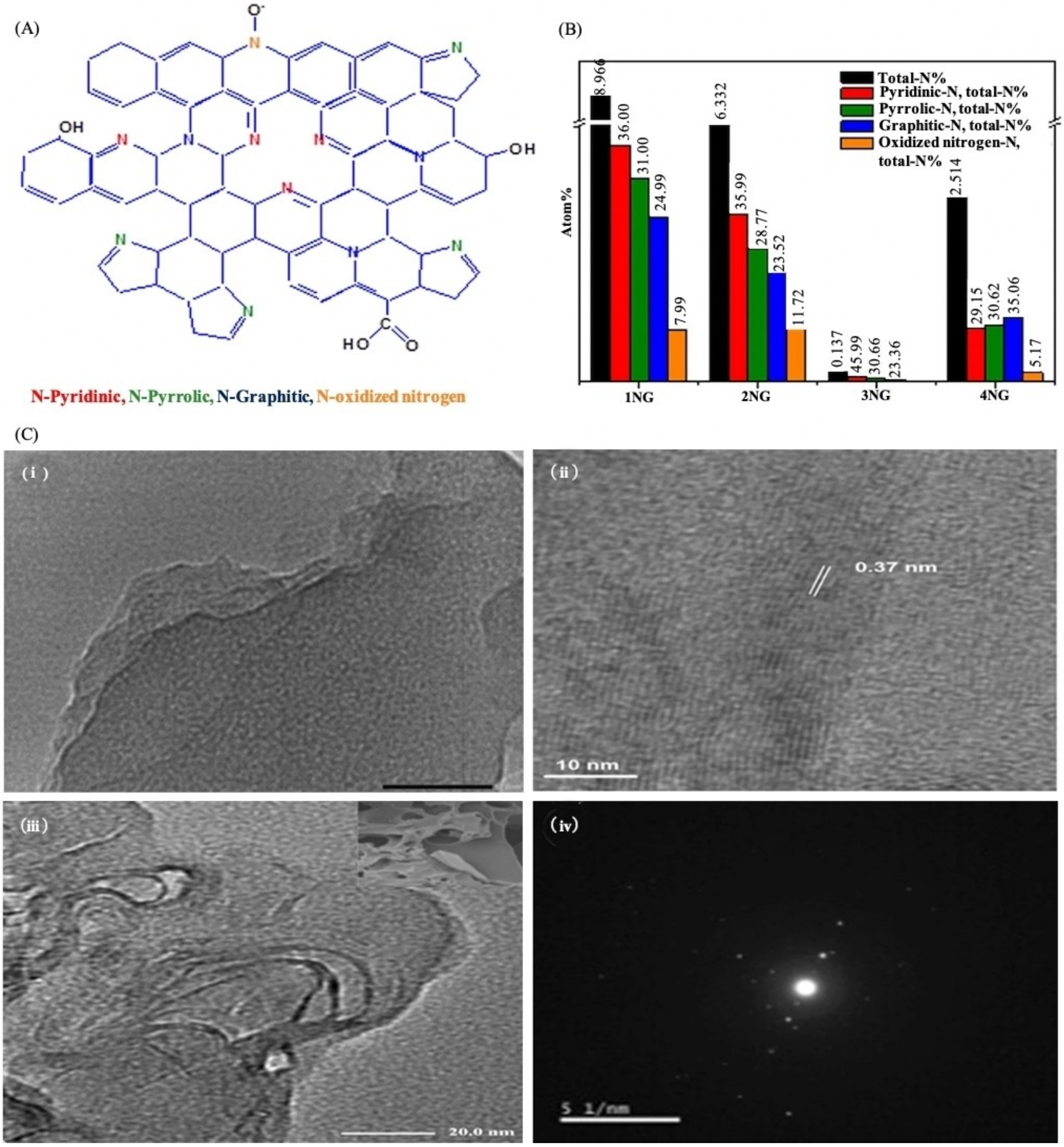


Figure 4. Development of high-performance supercapacitor based on a novel green synthesis protocol for 3D NG. (A) Schematic of the structure of NG. (B) The atomic percentage of nitrogen in each NG sample and its components. (C) (i) TEM image and (ii) HR-TEM image of G, (iii) corresponding TEM image with inset SEM image and (iv) selected area diffraction pattern (SAED) of NG. Reprinted with permission from Ref..[81]

The yields of FG are directly influenced by the characteristics of the materials involved. AC-FJH proves to be effective in producing FG from various thermoplastics such as PET, HDPE, PVC, LDPE, PP, and PS. The output of AC-FG derived from various thermoplastics shows a correlation with the thermal resistance of the starting material. In other words, plastics with higher thermal stability yield a higher FG output while generating fewer volatile oligomers. This relationship between thermal stability and FG yield presents an opportunity to utilize minimal electricity to convert PW into higher-value materials.[85] Embracing this approach holds the potential for substantial advancement in attaining plastic neutrality and curbing greenhouse gas emissions across the complete life cycle, ranging from initial production to the subsequent upcycling process.

2.3. Chemical Vapor Deposition (CVD)

Chemical vapor deposition (CVD) is an advanced deposition method that enables the production of high-quality solid

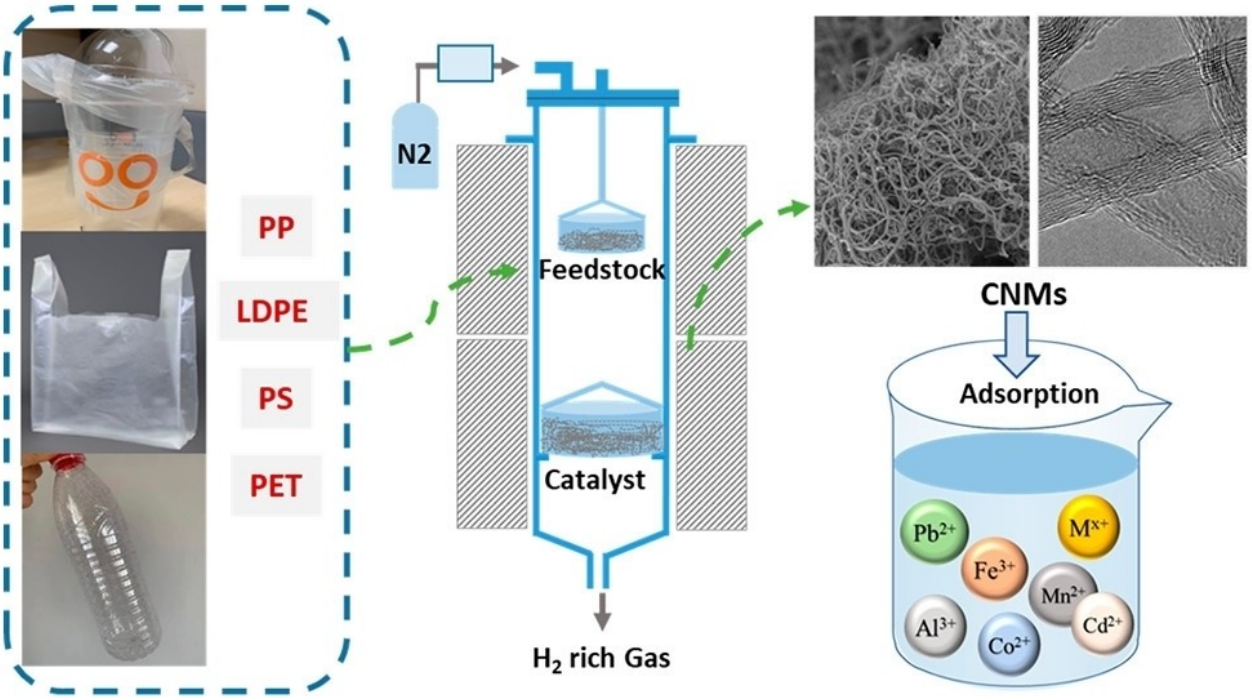


Figure 5. Converting plastic packaging waste into carbon nanomaterials (CNMs) using catalyst materials. Reprinted with permission from Ref..[82]

materials with exceptional performance characteristics, particularly under vacuum conditions. This technique relies on the occurrence of chemical reactions involving organometallic or halide compounds, which lead to the formation of the desired deposited material. One fascinating application of CVD involves the conversion of PW into CNTs, a process that generally comprises two primary stages. The process starts with the conversion of PW into volatile vapors in an oxygenated environment under ambient temperature, followed by transformation into CNTs at high temperature and pressure using CVD in the presence of a catalyst. Ongoing research and technological advancements in CVD are focused on optimizing process parameters, catalyst selection, and scaling up production to enable efficient and sustainable utilization of PW in the production of CNTs, graphene, and other advanced materials. The benefits of CVD include its high growth rate, substantial product yield, and the capability to produce epitaxial thin films. However, the technique is intricate and requires elevated temperatures, which can be considered a disadvantage.

The recycling of PW to treasure was implemented using a cost-effective, facile, and reliable CVD system for industrial-scale upcycling of various types of PW, including PET, PE, PMMA, PP, PS, and PVC, into high-quality graphene foil (GF), as illustrated in Figure 8.[86] A polycrystalline nickel foil was used as a catalyst and inserted into a quartz tube, then tempered at 1050 °C for 30 min in an Ar/H2 atmosphere to remove metal surface oxides. After annealing, the carbon source is melted and gradually injected into the CVD reactor. Then, GF was obtained by removing the nickel foil by etching it with ferric chloride/hydrochloric acid, followed by cleaning it with deionized water. The resulting GF exhibits a remarkable electrical conductivity of $3824\ \mathrm{S\,cm^{-1}}$, surpassing conventional free-standing graphene films processed at ultrahigh temperatures of 2200–2500 °C. Furthermore, GFs can be used as flexible components such as stand-alone electrodes in foldable Li-ion batteries, and exhibit stable electrochemical performance. Additionally, it demonstrated rapid and extremely low-voltage reactivity, allowing it to be used as a flexible electrothermal heater capable of reaching 322.6 °C with a 5 V input voltage.[87,88] The efficient upcycling of PW into valuable GF opens up new possibilities for graphene utilization across diverse fields.

Another study presented an innovative upcycling approach to synthesize high-quality monocrystalline graphene on polycrystalline Cu foil using discarded PW as a carbon source. The research utilized an ambient pressure chemical vapor deposition (AP-CVD) process, as depicted in Figure 9.[89] Copper foil functioned as the substrate, while the waste substance was used as the carbon source for graphene production. To minimize the number of nucleation sites, the Cu substrate underwent annealing in an H2 atmosphere at 1020 °C for 30 minutes before introducing the feedstock. Subsequently, individual graphene crystals were deposited onto a SiO2/Si substrate using a PMMA supporting layer which was eventually dissolved using acetone. The injection rate of decomposed polymer components from the pyrolysis of waste plastics had a dramatic impact on single graphene crystals outgrowth. Through meticulous regulation of the pyrolysis rate, the research accomplished the successful synthesis of sizable hexagonal and round-shaped single-crystal graphene structures. Images captured through optical microscopy showcased hexagonal graphene crystals positioned on the Cu substrate. These crystals exhibited a propensity to amalgamate, giving rise

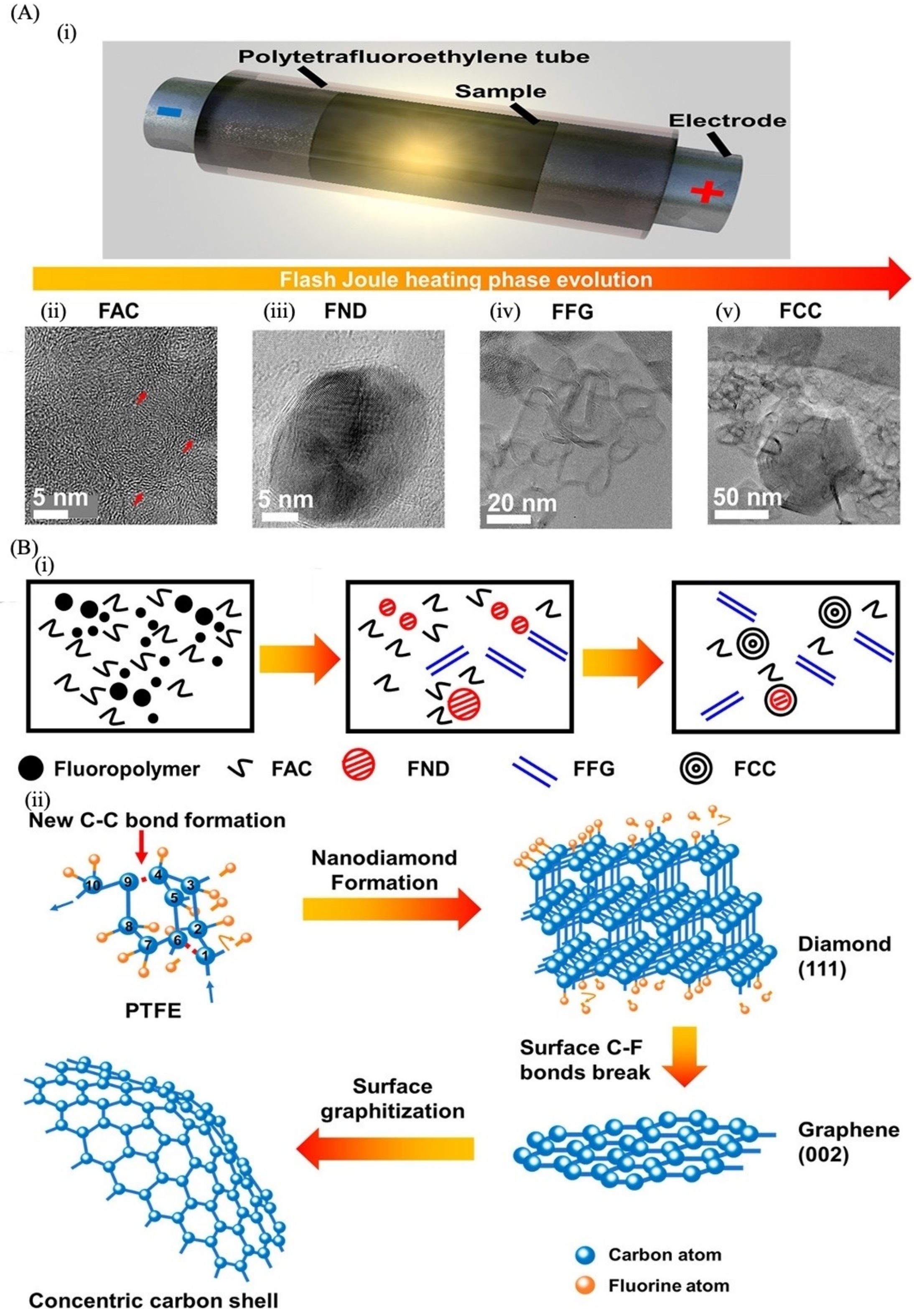


Figure 6. Ultrafast and controllable phase evolution by FJH. (A) Phase evolution of fluorinated nanocarbons. (i) Schematic of FJH setup. (ii–v) HR-TEM images of nanocarbon at different flash stages. (ii) FAC (iii) FNDs (iv) FFG (v) FCC products. (B) Mechanism for the evolution of fluorinated nanocarbons phases. (i) Schematic of the flash products at different stages of the FJH process (addition of energy input). (ii) Schematic of the formation of FND from PTFE, followed by subsequent conversion of the FND into polyhedral FCC. Reprinted with permission from Ref..[83]

to more extensive clusters or domains, thereby facilitating the creation of larger graphene structures. The SEM images showed the formation of a second layer of graphene on top of the first, indicating the formation of bilayer graphene. The high quality

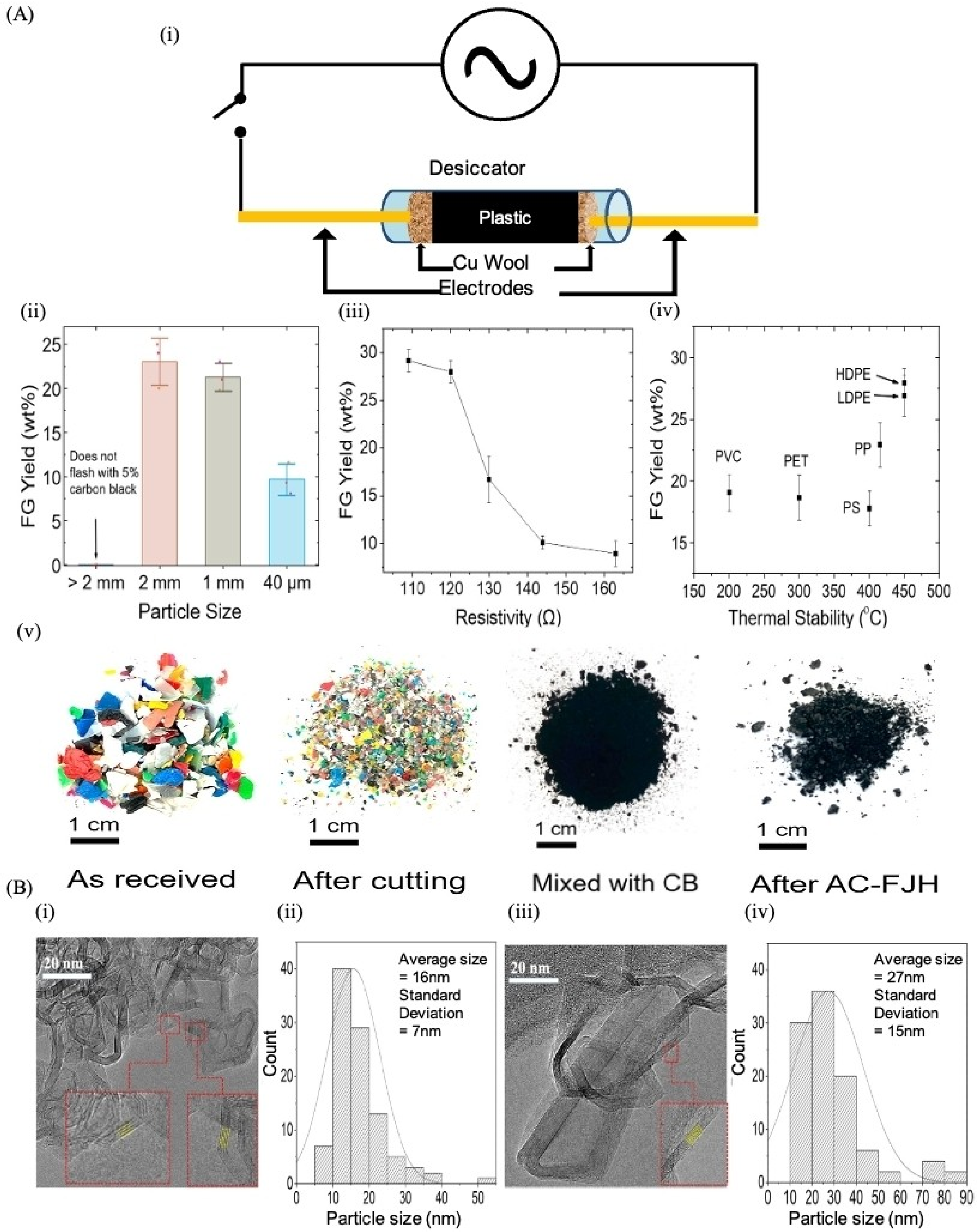


Figure 7. Conversion of PW into FG. (A) (i) Schematic of the AC-FG synthesis and (ii) AC-FG yield from HDPE. (iii) Impact of the initial resistivity of the HDPE/CB blend on the yield of AC-FG. (iv) Typical AC-FG yields obtained from various plastics when the initial resistance is set to 120 Ω. (v) Sequential images showing the progression from postconsumer plastic, as received from a recycler, to subsequent stages and final transformation into FG using AC-FJH. (B) (i) TEM image of AC-FG from HDPE and (ii) particle count. (iii) TEM image of AC/DC-FG from HDPE and (iv) particle count. Reprinted with permission from Ref..[84]

of the graphene crystals, regardless of their hexagonal or round shape, was verified by Raman studies. The results of this study highlight the considerable potential of using PW as a direct feedstock for the growth of high-quality graphene, thereby converting it into a high-value product.

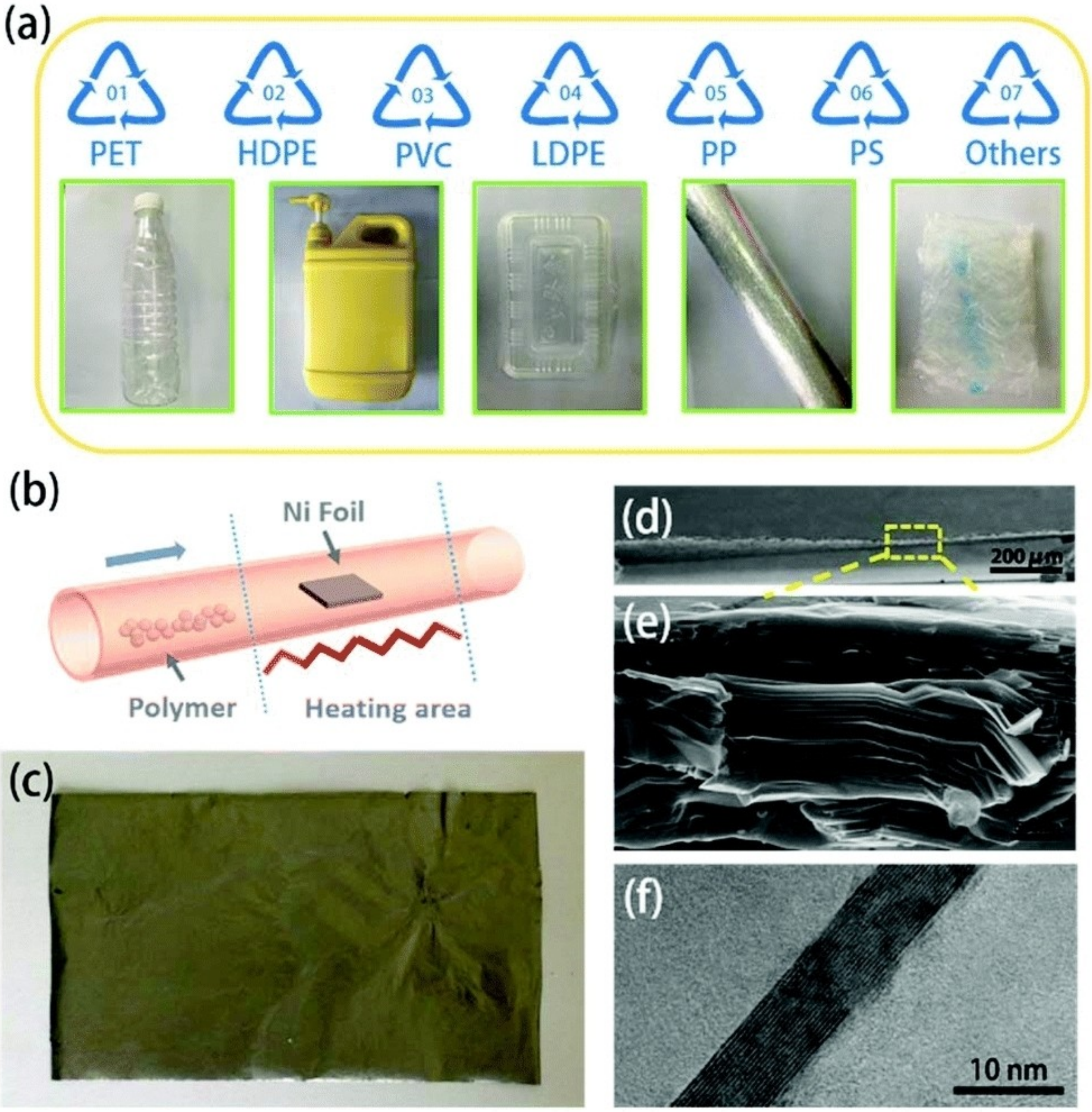


Figure 8. Trash to treasure: converting PW into a useful graphene foil (GF). (a) Raw waste plastic materials used in the experiment. (b) Schematic of the fabrication process of the GF. (c) Image of GF. (d and e) SEM images of the GF edge at different magnifications. (f) TEM image of the few-layer graphene in the structure of GF. Reprinted with permission from Ref..[86]

In summary, CVD has emerged as a highly effective and mature synthesis method for transforming PW into carbon materials. The resulting products exhibit exceptional properties, including high electrical conductivity, energy capacity, and mechanical flexibility. By precisely controlling the rate of injection of degraded polymer constituents into the CVD process, specific characteristics of the products can be fine-tuned. For instance, parameters such as pore size and graphene layer thickness can be adjusted to meet desired specifications. The ability to tailor the properties of these materials during the CVD process enhances their versatility and opens up new possibilities for their utilization in diverse fields.

2.4. Stepwise Conversion

The stepwise conversion method is a versatile and effective technique that converts PW into valuable carbon materials through a series of stages. Originally developed to cater to the unique requirements of industrial applications, this method offers several advantages over other approaches. By following a sequential series of reactions, starting with the pyrolysis of plastics, the stepwise conversion process ensures a controlled and efficient transformation of carbon materials. In the first step of the process, the plastics undergo pyrolysis, and generate gaseous products, primarily hydrocarbons, as well as solid residues. In the second step, the gaseous products are directed downstream to encounter carefully selected catalysts. These catalysts play a crucial role in facilitating the subsequent

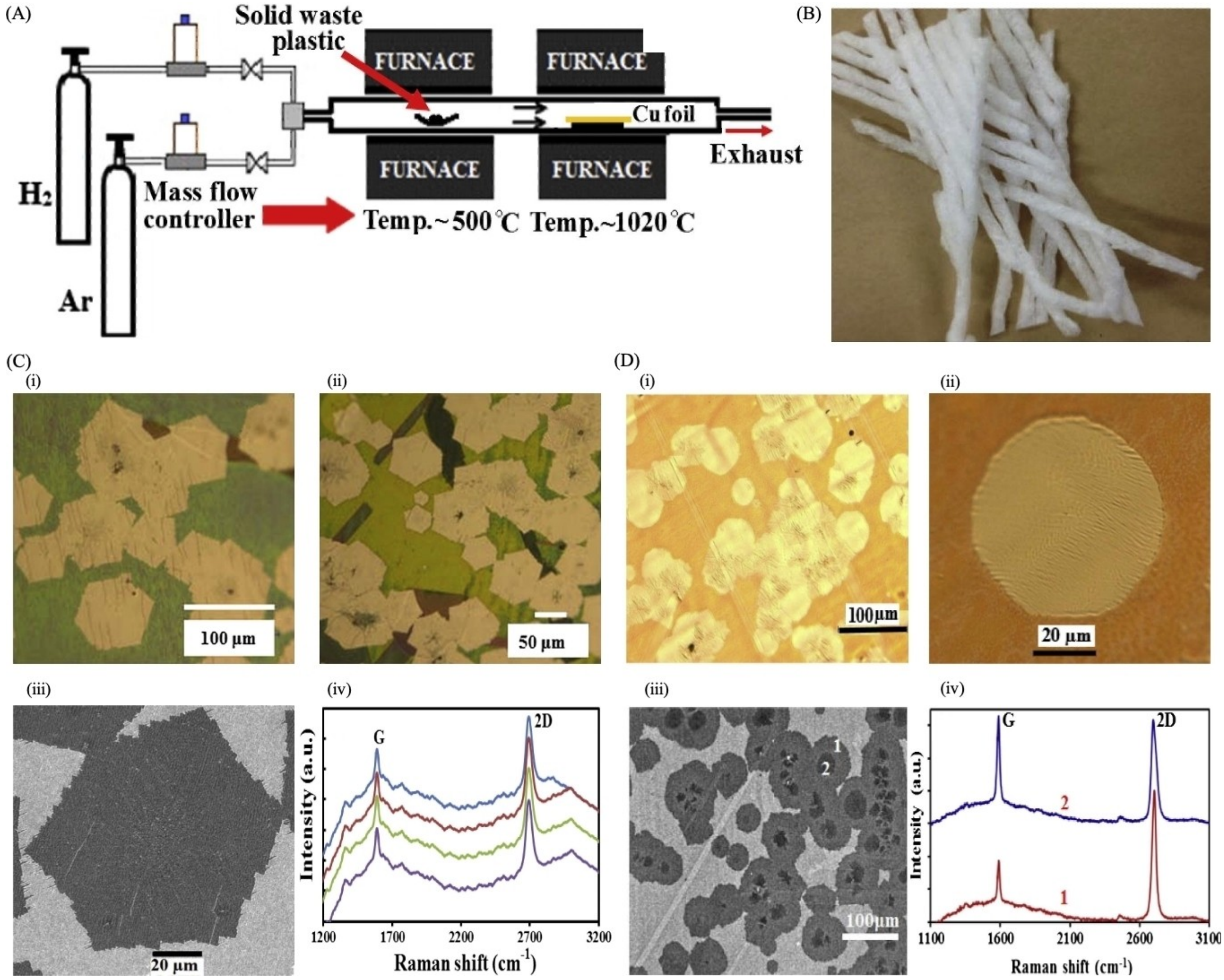


Figure 9. Synthesis of high-quality graphene from solid waste plastic by CVD. (A) Schematic of the CVD process. (B) PW is used as carbon feedstock. (C) Hexagonal graphene crystals (i, ii) Optical microscopy (iii) SEM image and (iv) Raman spectra of the graphene crystal. (D) Round-shaped graphene crystals (i, ii) Optical microscopy (iii) SEM image and (iv) Raman spectra of few-layer graphene crystals. Reprinted with permission from Ref..[89]

reactions and are chosen based on their ability to promote the desired carbon conversion. Under the influence of the catalysts, the gaseous hydrocarbons undergo further transformations, forming CNTs and other highly versatile carbon-based materials.

In a previous study, CNTs were produced successfully by synthesising them from easily available waste polyethylene employing a stepwise pyrolysis-combustion technique, as shown in Figure 10.[90] The process involved pyrolyzing polyethylene, which was in the form of strips, pellets, or ground particles, to produce gaseous by-products. Subsequently, these vaporized products were combined with a gas containing oxygen and ignited, resulting in the formation of a flame rich in fuel. The effluent produced by the combustion process resulting from this flame was used to synthesize multi-walled carbon nanotubes (MWCNTs), yielding approximately 10% by mass in relation to the initial carbon in the fuel, prior to purification procedures. Plastics were subjected to pyrolysis, yielding light hydrocarbons (LHCs) predominantly in the form of a gaseous stream comprising LHCs and hydrogen (H2). This stream was subsequently employed directly as raw material for the synthesis of CNTs. Alternatively, it was blended with gases containing oxygen and subjected to combustion, generating CNT feedstock with distinct compositions. The production of feedstock gases and the synthesis of CNT took place in distinct reactors, allowing independent control over their operational parameters. Within an upscaled industrial framework, the potential exists to collect and harness the energy discharged during the exothermic combustion of PW. This could potentially compensate for the energy needed for the pyrolysis of solid plastics and improve the overall energy efficiency of the process while reducing the cost associated with CNT production.

In a separate study, a two-step process was employed to transform PP into MWCNTs and hydrogen-rich gas, as schematically illustrated in Figure 11.[91] The proposed method comprised two distinct stages: the preliminary catalytic pyrolysis of

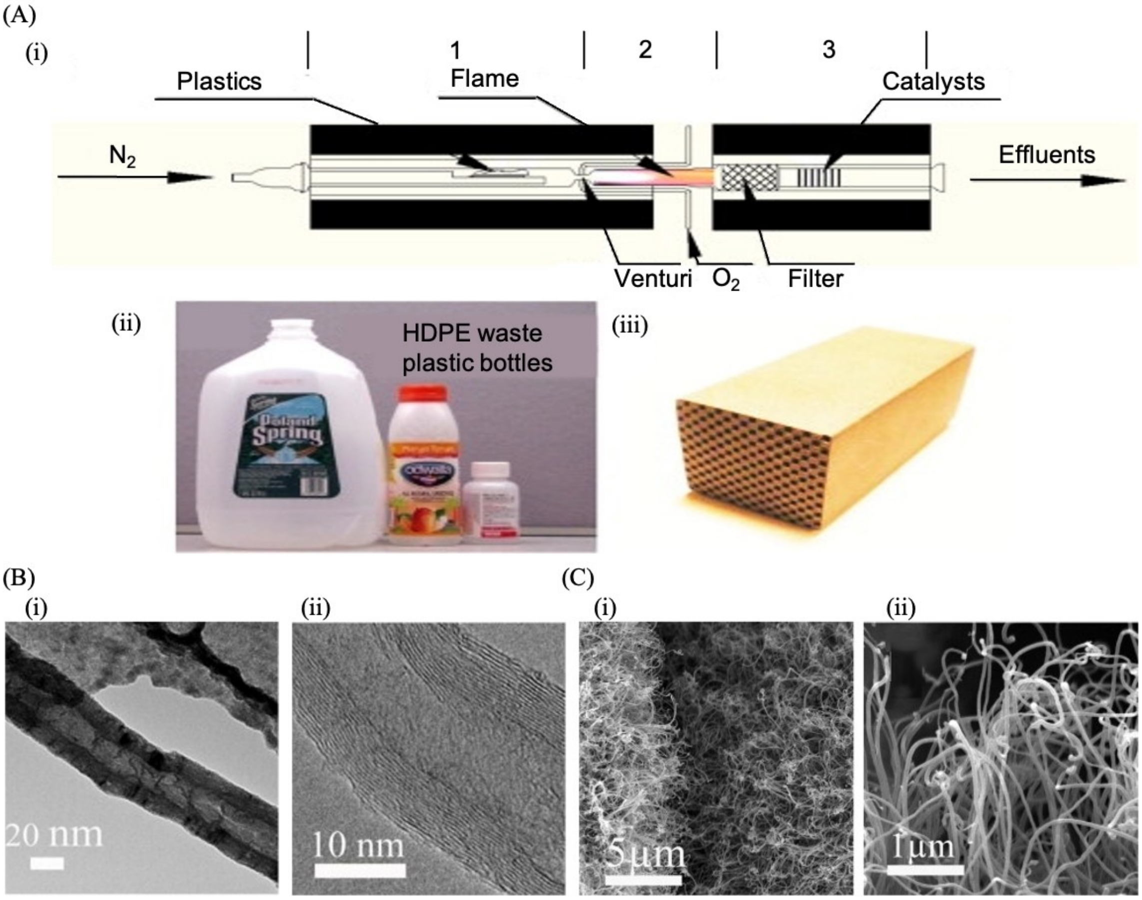


Figure 10. Synthesis of CNTs by sequential pyrolysis and combustion of polyethylene. (A) (i) Schematic of three-stage step-wise CNTs synthesis system (zone 1: pyrolysis, 2: combustion, and 3: synthesis stage). (ii) HDPE waste plastic bottles are used as carbon feedstock. (iii) Ceramic filter. (B)TEM images of CNTs. (i) HRTEM image (ii) CNTs with few-layer graphene. (C) SEM images of CNTs generated by the combustion of HDPE PW at two different magnifications. (i) 5 μm. (ii) 1 μm. Reprinted with permission from Ref..[90]

PP using HZSM-5 zeolite, succeeded by subsequent catalytic decomposition of the resultant pyrolysis gases to yield MWCNTs and hydrogen via CVD nickel catalysts within a moving-bed reactor. SEM and TEM imaging showed that the carbon products generated in the fluidized bed reactor were present as MWCNTs. XRD and TGA analysis showed that higher decomposition temperatures led to the production of CNTs with a greater degree of crystallinity. The effects of pyrolysis temperature (550–750 °C) and decomposition temperature (500–800 °C) on the efficacy of the two-stage reaction system were thoroughly examined. The quantity of MWCNTs and the concentration of hydrogen increased as the decomposition temperature rose, reaching a peak at 700 °C. The level of graphitization and thermal stability of the MWCNTs synthesized at higher decomposition temperatures exceeded those synthesized at lower decomposition temperatures. The pyrolysis temperature influenced the amount and composition of the pyrolysis gas, as well as the yield of MWCNTs. This research presents a promising method for converting PW into carbon nanomaterials and hydrogen.

Based on the findings of the aforementioned studies, it is evident that the quantity and composition of pyrolysis gas and the overall yield of products using the stepwise conversion method are significantly determined by the pyrolysis temperature. Moreover, the choice of catalysts significantly influences the characteristics of the resulting products, leading to variations in their shapes, such as straight, tortuous, or coiled configurations. It is worth noting that while pyrolysis and decomposition processes generally necessitate high temperatures and energy input, their energy efficiency can be enhanced by employing a heat exchanger to recirculate the released heat back into the pyrolyzer, thereby facilitating the gasification of the incoming precursor. By implementing this heat recovery mechanism, the energy efficiency of the pyrolysis and decomposition processes can be enhanced, resulting in a more sustainable and economically viable operation.

3. Conclusion and Future Outlook

Plastic is ubiquitous in modern society due to its versatility and cost-effectiveness. However, the uncontrolled proliferation of PW poses a severe environmental and health crisis. Transforming PW into high-value carbon materials represents a compelling and sustainable solution to this burgeoning issue. This review meticulously explores the current state of PW

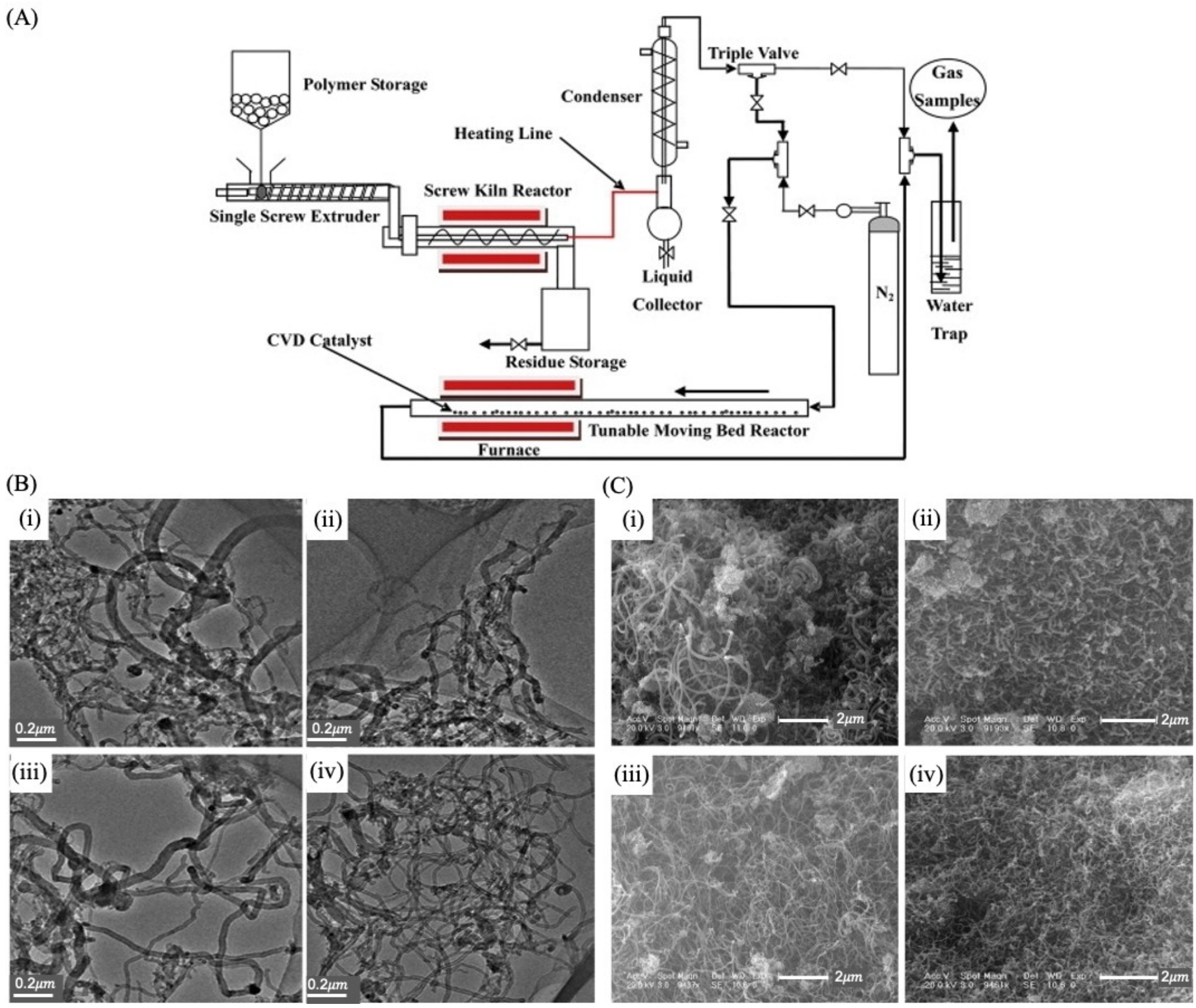


Figure 11. Catalytic pyrolysis of polypropylene to synthesize MWCNTs and hydrogen through a two-stage process. (A) Schematic of the two-stage step-wise reaction system. (B) TEM images of the MWCNTs were obtained at different decomposition temperatures: (i) 500, (ii) 600, (iii) 700, and (iv) 800 °C. (C) SEM images of the MWCNTs obtained at different decomposition temperatures: (i) 500, (ii) 600, (iii) 700, and (iv) 800 °C. Reprinted with permission from Ref..[91]

upcycling, focusing on four primary methodologies: thermal decomposition, flash joule heating (FJH), chemical vapor deposition (CVD), and stepwise conversion.

Thermal decomposition, particularly through pyrolysis, remains the most established and cost-effective method for generating carbon nanomaterials from plastic waste. Pyrolysis offers high yields and economic advantages, producing CNTs at costs ranging from $50–$200 per gram.[92,93] This cost is markedly lower compared to the $1,000–$10,000 per gram associated with CVD methods.[94,95] This significant cost disparity underscores the economic viability of pyrolysis for large-scale applications.

FJH is recognized for its rapid production of high-quality carbon nanomaterials, with costs ranging approximately from $200–$500 per gram. This makes FJH a cost-effective alternative to CVD, yielding carbon nanomaterials with superior purity and fewer defects.[96,97] In contrast, stepwise conversion, while less prevalent, involves a multi-stage process that is generally more expensive than pyrolysis but less costly than CVD, with costs typically between $500 and $1,000 per gram.[98]

Despite these advancements, the current infrastructure for PW management remains inadequate, leading to inefficiencies in waste processing. A transformative shift in plastic upcycling technologies and the development of value-added products are crucial for enhancing economic feasibility. Moreover, robust policies aimed at reducing plastic production, promoting reuse, and improving recycling efforts are essential to addressing this issue.

Achieving plastic neutrality and advancing environmental sustainability necessitate a coordinated effort across scientific research, engineering innovation, and policy-making. An integrated, cross-disciplinary approach is critical to overcoming the complexities of PW management and progressing towards a circular plastic economy. The urgency of this challenge underscores the need for innovative, collaborative solutions to effectively address the global plastic waste crisis.

Acknowledgements

The authors would like to thank the Ministry of Science and Technology, Taiwan, for financially supporting this research under Contract No. 111-2813-C-029-042-E.

Conflict of Interests

The authors declare that they have no conflicts of interest.

Keywords: Upcycling · Waste-to-value · Plastic waste · Carbon nanomaterials · Circular economy

[1] X. Zhao, M. Korey, K. Li, K. Copenhaver, H. Tekinalp, S. Celik, S. Ozcan, *Chem. Eng. J.* **2022**, *428*, 131928.
[2] G. Celik, R. M. Kennedy, R. A. Hackler, M. Ferrandon, A. Tennakoon, S. Patnaik, M. Delferro, *ACS Cent. Sci.* **2019**, *5*, 1795–1803.
[3] J. Huang, A. Veksha, W. P. Chan, A. Giannis, G. Lisak, *Renewable Sustainable Energy Rev.* **2022**, *154*, 111866.
[4] A. Dey, C. V. Dhumal, P. Sengupta, A. Kumar, N. K. Pramanik, T. Alam, *J. Food Sci. Technol.* **2021**, *58*, 3251–3269.
[5] K. J. Groh, T. Backhaus, B. Carney-Almroth, B. Geueke, P. A. Inostroza, A. Lennquist, J. Muncke, *Sci. Total Environ.* **2019**, *651*, 3253–3268.
[6] *Plastic production worldwide 2021* Statista **2023**, June 12, Statista https://www.statista.com/statistics/282732/global-production-of-plastics-since-1950/.<
[7] R. Geyer, J. R. Jambeck, K. L. Law, *Sci. Adv.* **2017**, *3*, e1700782.
[8] I. Agenda, In *The World Economic Forum: Geneva, Switzerland* **2016**, *36*.
[9] K. R. Vanapalli, H. B. Sharma, V. P. Ranjan, B. Samal, J. Bhattacharya, B. K. Dubey, S. Goel, *Sci. Total Environ.* **2021**, *750*, 141514.
[10] OECD Global Plastics Outlook Database, Plastic pollution is growing relentlessly as waste management and recycling fall short, says OECD. https://www.oecd.org/environment/plastic-pollution-is-growing-relentlessly-as-waste-management-and-recycling-fall-short.htm.
[11] R. Geyer, J. R. Jambeck, K. L. Law, *Sci. Adv.* **2017**, *3*, e1700782.
[12] A. Sangroniz, J. B. Zhu, X. Tang, A. Etxeberria, E. Y. X. Chen, H. Sardon, *Nat. Commun.* **2019**, *10*, 1–7.
[13] X. Jiao, K. Zheng, Q. Chen, X. Li, Y. Li, W. Shao, Y. Xie, *Angew. Chem. Int. Ed.* **2020**, *59*, 15497–15501.
[14] K. L. Law, N. Starr, T. R. Siegler, J. R. Jambeck, N. J. Mallos, G. H. Leonard, *Sci. Adv.* **2020**, *6*, eabd0288.
[15] R. Meys, A. Kätelhön, M. Bachmann, B. Winter, C. Zibunas, S. Suh, A. Bardow, *Science* **2021**, *374*, 71–76.
[16] H. V. Ford, N. H. Jones, A. J. Davies, B. J. Godley, J. R. Jambeck, I. E. Napper, H. J. Koldewey, *Sci. Total Environ.* **2022**, *806*, 150392.
[17] J. L. Lavers, J. Rivers-Auty, A. L. Bond, *J. Hazard. Mater.* **2021**, *416*, 126140.
[18] J. B. Lamb, B. L. Willis, E. A. Fiorenza, C. S. Couch, R. Howard, D. N. Rader, C. D. Harvell, *Science* **2018**, *359*, 460–462.
[19] Y. Zhang, T. Gao, S. Kang, H. Shi, L. Mai, D. Allen, S. Allen, *Earth Sci. Rev.* **2022**, *226*, 103924.
[20] J. M. Donkers, E. M. Höppener, I. Grigoriev, L. Will, B. N. Melgert, B. van der Zaan, I. M. Kooter, *Microplast. Nanoplast.* **2022**, *2*, 1–18.
[21] M. MacLeod, H. P. H. Arp, M. B. Tekman, A. Jahnke, *Science* **2021**, *373*, 61–65.
[22] A. L. Dawson, S. Kawaguchi, C. K. King, K. A. Townsend, R. King, W. M. Huston, S. M. Bengtson Nash, *Nat. Commun.* **2018**, *9*, 1–8.
[23] B. Unger, E. L. B. Rebolledo, R. Deaville, A. Gröne, L. L. IJsseldijk, M. F. Leopold, H. Herr, *Mar. Pollut. Bull.* **2016**, *112*, 134–141.
[24] B. Page, J. McKenzie, R. McIntosh, A. Baylis, A. Morrissey, N. Calvert, S. D. Goldsworthy, *Mar. Pollut. Bull.* **2004**, *49*, 33–42.
[25] J. L. Lavers, A. L. Bond, I. Hutton, *Environ. Pollut.* **2014**, *187*, 124–129.
[26] P. S. Puskic, J. L. Lavers, L. R. Adams, M. Grünenwald, I. Hutton, A. L. Bond, *Conserv. Physiol.* **2019**, *7*, coz017.
[27] J. L. Lavers, I. Hutton, A. L. Bond, *Environ. Sci. Technol.* **2019**, *53*, 9224–9231.
[28] J. L. Lavers, A. L. Bond, *Mar. Pollut. Bull.* **2016**, *110*, 493–500.
[29] S. Amin, M. Amin, *Rev. Adv. Mater. Sci.* **2011**, *29*, 15–30.
[30] M. C. S. Ribeiro, A. Fiúza, A. Ferreira, M. D. L. Dinis, A. C. Meira Castro, J. P. Meixedo, M. R. Alvim, *Recycling* **2016**, *1*, 178–193.
[31] C. S. Brazel, S. L. Rosen, *Fundamental Principles of Polymeric Materials* John Wiley & Sons, **2012**, 456–460.
[32] Y. Yang, R. Boom, B. Irion, D. J. van Heerden, P. Kuiper, H. de Wit, *Chem. Eng. Process.: Process Intensif.* **2012**, *51*, 53–68.
[33] M. E. Grigore, *Recycling* **2017**, *2*, 24.
[34] M. Biron, *Thermoplastics and Thermoplastic Composites*, William Andrew **2018**.
[35] J. Nicholson, *Royal Society of Chemistry* **2017**.
[36] Y. Chen, A. K. Awasthi, F. Wei, Q. Tan, J. Li, *Sci. Total Environ.* **2021**, *752*, 141772.
[37] Z. Liu, Q. Qin, Z. Hu, L. Yan, U. I. Ieong, Y. Xu, *Environ. Pollut.* **2020**, *265*, 114926.
[38] J. H. Arndt, R. Brüll, T. Macko, F. Perez, J. Panitzky, *Ind. Eng. Chem. Res.* **2021**, *60*, 14968–14976.
[39] Q. Zhang, M. U. Khan, X. Lin, W. Yi, H. Lei, *J. Cleaner Prod.* **2020**, *262*, 121251.
[40] J. M. Restrepo-Flórez, A. Bassi, M. R. Thompson, *Int. Biodeterior. Biodegrad.* **2014**, *88*, 83–90.
[41] S. Kumagai, J. Lu, Y. Fukushima, H. Ohno, T. Kameda, T. Yoshioka, *Resour. Conserv. Recycl.* **2018**, *133*, 354–361.
[42] L. Yang, J. Gao, Y. Liu, G. Zhuang, X. Peng, W. M. Wu, X. Zhuang, *Chemosphere* **2021**, *262*, 127818.
[43] S. Nanda, F. Berruti, *Environ. Chem. Lett.* **2021**, *19*, 123–148.
[44] K. Shi, Z. Bai, T. Su, Z. Wang, *Int. J. Biol. Macromol.* **2019**, *126*, 436–442.
[45] D. K. Ojha, R. Vinu, In *Waste Biorefinery* Elsevier **2018**, 349–391.
[46] A. Demetrious, E. Crossin, *J. Mater. Cycles Waste Manage.* **2019**, *21*, 850–860.
[47] Z. Yang, F. Lü, H. Zhang, W. Wang, L. Shao, J. Ye, P. He, *J. Hazard. Mater.* **2021**, *401*, 123429.
[48] S. M. Al-Salem, M. H. Behbehani, A. Al-Hazza'a, J. C. Arnold, S. M. Alston, A. A. Al-Rowaih, H. Karam, *J. Mater. Cycles Waste Manage.* **2019**, *21*, 1106–1122.
[49] R. Koshti, L. Mehta, N. Samarth, *J. Polym. Environ.* **2018**, *26*, 3520–3529.
[50] R. Wei, T. Tiso, J. Bertling, K. O'Connor, L. M. Blank, U. T. Bornscheuer, *Nat. Catal.* **2020**, *3*, 867–871.
[51] L. D. Ellis, N. A. Rorrer, K. P. Sullivan, M. Otto, J. E. McGeehan, Y. Román-Leshkov, G. T. Beckham, *Nat. Catal.* **2021**, *4*, 539–556.
[52] J. J. Klemeš, Y. V. Fan, P. Jiang, *Energy Sources Part A* **2021**, *43*, 1549–1565.
[53] M. S. Mauter, M. Elimelech, *Environ. Sci. Technol.* **2008**, *42*, 5843–5859.
[54] K. D. Patel, R. K. Singh, H. W. Kim, *Mater. Horiz.* **2019**, *6*, 434–469.
[55] L. Dai, D. W. Chang, J. B. Baek, W. Lu, *Small* **2012**, *8*, 1130–1166.
[56] A. K. Wanekaya, *Analyst* **2011**, *136*, 4383–4391.
[57] S. C. J. Huang, A. B. Artyukhin, N. Misra, J. A. Martinez, P. A. Stroeve, C. P. Grigoropoulos, A. Noy, *Nano Lett.* **2010**, *10*, 1812–1816.
[58] M. Nasrollahzadeh, M. Sajjadi, S. Iravani, R. S. Varma, *Chemosphere* **2021**, *263*, 128005.
[59] G. Dufil, I. Bernacka-Wojcik, A. Armada-Moreira, E. Stavrinidou, *Chem. Rev.* **2021**, *122*, 4847–4883.
[60] M. Islam, A. D. Lantada, D. Mager, J. G. Korvink, *Adv. Healthcare Mater.* **2022**, *11*, 2101834.
[61] X. Tong, N. Li, M. Zeng, Q. Wang, *Renewable Sustainable Energy Rev.* **2019**, *108*, 398–422.
[62] S. Suganuma, K. Nakajima, M. Kitano, D. Yamaguchi, H. Kato, S. Hayashi, M. Hara, *J. Am. Chem. Soc.* **2008**, *130*, 12787–12793.
[63] D. Ghosh, A. F. Bagley, Y. J. Na, M. J. Birrer, S. N. Bhatia, A. M. Belcher, *Proc. Natl. Acad. Sci.* **2014**, *111*, 13948–13953.
[64] H. Liu, L. Liu, Z. Fan, J. Liu, H. Wang, X. Wen, G. Hu, J. Gong, *Chem. Eng. J.* **2024**, *485*, 149690.
[65] Q. Liu, H. Wang, X. Wen, G. Hu, H. Liu, Z. Gong, S. Bi, Q. Wei, R. Niu, J. Gong, *Sustainable Mater. Technol.* **2024**, *41*, e01022.
[66] J. Liu, Z. Fan, H. Liu, L. Liu, X. Wen, H. Wang, R. Niu, H. Wang, J. Cheng, J. Gong, *J. Environ. Chem. Eng.* **2024**, *12*, 112363.
[67] J. Choi, I. Yang, S. S. Kim, S. Y. Cho, S. Lee, *Macromol. Rapid Commun.* **2022**, *43*, 2100467.
[68] X. Chen, Y. Wang, L. Zhang, *ChemSusChem* **2021**, *14*, 4137–4151.

[69] O. Vieira, R. S. Ribeiro, J. L. D. de Tuesta, H. T. Gomes, A. M. Silva, *Chem. Eng. J.* **2022**, *428*, 131399.
[70] Q. Li, K. Yao, G. Zhang, J. Gong, E. Mijowska, K. Kierzek, T. Tang, *Part. Part. Syst. Charact.* **2015**, *32*, 874–879.
[71] X. Liu, Y. Wen, X. Chen, T. Tang, E. Mijowska, *Sci. Total Environ.* **2020**, *723*, 138055.
[72] Z. Jiang, R. Song, W. Bi, J. Lu, T. Tang, *Carbon* **2007**, *45*, 449–458.
[73] M. F. De Volder, S. H. Tawfick, R. H. Baughman, A. J. Hart, *Science* **2013**, *339*, 535–539.
[74] L. Dai, D. W. Chang, J. B. Baek, W. Lu, *Small* **2012**, *8*, 1130–1166.
[75] A. Kumar, K. Sharma, A. R. Dixit, *J. Mater. Sci.* **2020**, *55*, 2682–2724.
[76] M. Bassyouni, A. E. Mansi, A. Elgabry, B. A. Ibrahim, O. A. Kassem, R. Alhebeshy, *Appl. Phys. A* **2020**, *126*, 1–33.
[77] ASTM E176 *Standard Terminology of Fire Standards*, West Conshohocken, PA American Society for Testing and Materials, **2006**.
[78] C. L. Beyler, M. M. Hirschler, *SFPE Handbook of Fire Protection Engineering* **2002**, *2(7)*, 111–131.
[79] S. Chen, Z. Liu, S. Jiang, H. Hou, *Sci. Total Environ.* **2020**, *710*, 136250.
[80] Z. Chen, W. Wei, B. J. Ni, H. Chen, *Environ. Funct. Mater.* **2022**, *1*(1), 34–48.
[81] N. A. Elessawy, J. El Nady, W. Wazeer, A. B. Kashyout, *Sci. Rep.* **2019**, *9*, 1–10.
[82] D. Yao, H. Li, B. C. Mohan, A. K. Prabhakar, Y. Dai, C. H. Wang, *ACS Sustainable Chem. Eng.* **2022**, *10*, 1125–1136.
[83] W. Chen, J. T. Li, Z. Wang, W. A. Algozeeb, D. X. Luong, C. Kittrell, J. M. Tour, *ACS Nano* **2021**, *15*, 11158–11167.
[84] W. A. Algozeeb, P. E. Savas, D. X. Luong, W. Chen, C. Kittrell, M. Bhat, J. M. Tour, *ACS Nano* **2020**, *14*, 15595–15604.
[85] J. Zheng, S. Suh, *Nat. Clim. Change* **2019**, *9*, 374–378.
[86] L. Cui, X. Wang, N. Chen, B. Ji, L. Qu, *Nanoscale* **2017**, *9*, 9089–9094.
[87] G. Xin, H. Sun, T. Hu, H. R. Fard, X. Sun, N. Koratkar, J. Lian, *Adv. Mater.* **2014**, *26*, 4521–4526.
[88] Y. Chen, K. Fu, S. Zhu, W. Luo, Y. Wang, Y. Li, L. Hu, *Nano Lett.* **2016**, *16*, 3616–3623.
[89] S. Sharma, G. Kalita, R. Hirano, S. M. Shinde, R. Papon, H. Ohtani, M. Tanemura, *Carbon* **2014**, *72*, 66–73.
[90] C. Zhuo, B. Hall, H. Richter, Y. Levendis, *Carbon* **2010**, *48*, 4024–4034.
[91] J. Liu, Z. Jiang, H. Yu, T. Tang, *Polym. Degrad. Stab.* **2011**, *96*, 1711–1719.
[92] K. S. Kim, H. H. Lee, M. H. Lee, *J. Mater. Chem. A* **2019**, *7*, 3571–3580.
[93] A. B. Smith, J. M. Davis, L. J. Reynolds, *Waste Manage.* **2019**, *85*, 230–238.
[94] R. D. Johnson, M. E. Anderson, H. T. Wang, *Carbon* **2018**, *127*, 497–507.
[95] T. L. Clark, R. H. Peterson, M. J. Sutherland, *Nanotechnol. Rev.* **2019**, *8*, 89–101.
[96] J. Zhang, A. C. Pope, C. C. Schlesinger, R. J. P. Williams, *Nat. Nanotechnol.* **2019**, *14*, 623–628.
[97] J. Smith, et al., *Mater. Sci. Eng. Rep.* **2024**, *150*, 89–105.
[98] M. K. Gupta, L. J. Cohen, P. S. Verma, *Mater. Sci. Eng.* **2018**, *560*, 75–83.

Manuscript received: July 2, 2024
Revised manuscript received: July 31, 2024
Accepted manuscript online: August 16, 2024
Version of record online: October 17, 2024