ACS NANO

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REVIEW

Catalytic Upgrading of Plastic Wastes into High-Value Carbon Nanomaterials: Synthesis and Applications

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Cite This: *ACS Nano* 2025, 19, 12734–12761

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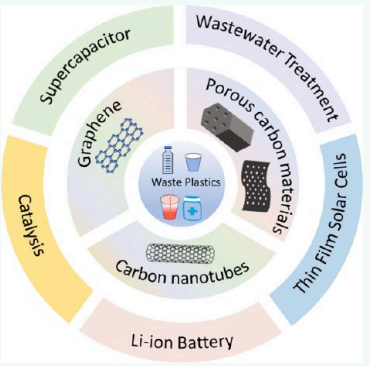
ABSTRACT: The surge in waste plastics has placed a serious burden on the global ecosystem. Traditional recycling methods are insufficient to handle the growing volume of plastic waste, highlighting the urgent demand for innovative recycling technologies. Transforming plastics into high-value carbon nanomaterials is a simple and efficient resource recovery strategy, especially effective for handling mixed or hard-to-separate plastic waste. This method not only simplifies the sorting of discarded plastics but also offers significant advantages in recovery efficiency and processing convenience. This review systematically summarized various technologies for converting plastics into carbon nanomaterials, focusing on the catalytic mechanisms of different conversion methods. We also analyzed how various catalysts, catalytic temperatures, and metal−support interactions affect the yield and quality of carbon nanomaterials. Additionally, the potential applications of carbon nanomaterials in environmental remediation, energy storage, and catalysis are also evaluated. The ongoing challenges and future research directions in this field are critically discussed, which will ultimately facilitate more effective resource recovery from plastics and contribute to the realization of a circular economy. We believe that this review will inspire more creativity in designing such win−win reaction systems to realize a "waste treat waste" concept.



KEYWORDS: *plastic wastes, chemical upgrading, catalytic conversion, carbon nanomaterials, synthesis strategies, catalytic mechanism, applications, challenges and perspectives*

1. INTRODUCTION

With the ongoing development of society, plastics have become widely used in industries such as food packaging, water supply, and healthcare, owing to their low cost and excellent corrosion resistance.[1−7] Plastics Europe reports that global plastic production increased from 1.7 million tons in 1950 to 367 million tons by 2021.[8] However, the chemical stability of plastics leads to slow degradation, resulting in the rapid accumulation of plastic waste in the environment. By 2050, it is estimated that around 20 million tons of plastic waste may remain in landfills or natural environments.[8] These plastics can pollute soil and water, disrupt ecosystems, spread disease and pose serious threats to marine life and human health.[9] Current plastic waste disposal methods mainly rely on incineration and landfilling, which waste resources and pose significant risks to ecosystems.[10] Therefore, improving the recycling efficiency and resource recovery of plastic waste has become a pressing issue.

Plastic recycling generally involves two main methods: mechanical and chemical recycling (Figure 1). In mechanical recycling, plastics can degrade with repeated use, which reduces their mechanical properties and reuse value. In contrast, chemical recycling technology is used to add value to waste by breaking chemical bonds in the polymer chain and breaking down waste plastics into smaller molecules, such as gases, liquid fuels, or single molecules. This method is not only capable of handling complex mixtures of plastics, but also removes impurities and produces high-quality recycled materials.[11−14] Waste plastics are often a mixture of different types, requiring presorting before chemical recycling. This increases both the complexity and cost of the process. To tackle this challenge, chemical conversion of waste plastics into carbon nanomaterials has emerged as a more appealing solution. This method not only simplifies the sorting of

Received: February 24, 2025
Revised: March 20, 2025
Accepted: March 20, 2025
Published: March 29, 2025

https://doi.org/10.1021/acsnano.5c03391
ACS Nano 2025, 19, 12734−12761

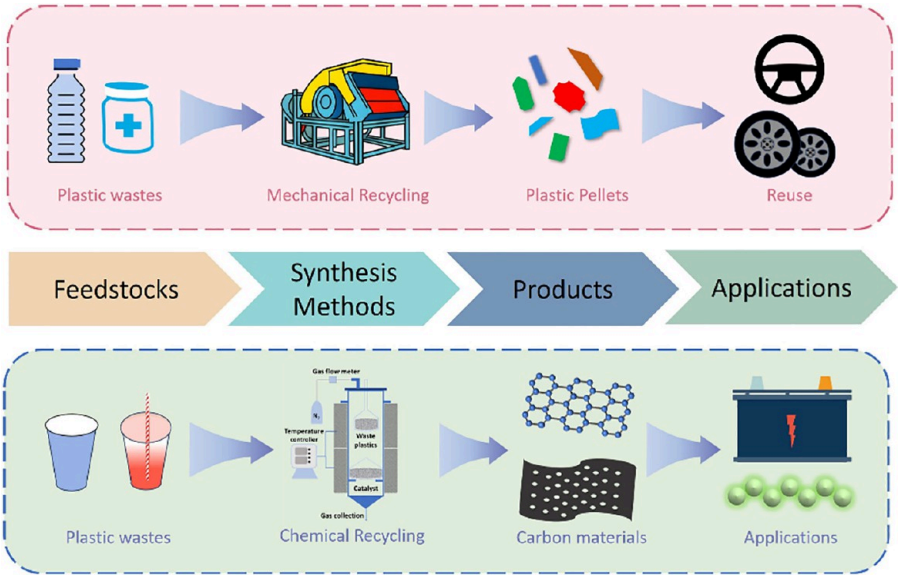


Figure 1. Recycling and upgrading of plastic wastes and potential applications of subsequent products.

mixed plastics but also provides better adaptability.[15] Carbon nanomaterials, with high porosity, large surface area, abundant functional groups, and good conductivity, are widely used in applications such as batteries, supercapacitors, water electrolysis, pollutant removal, solar evaporation, and CO_2 capture.[16,17] Therefore, developing carbon nanomaterials not only provides an efficient plastic wastes recycling pathway but also offers solutions for several important applications.[18,19]

The existing literature review discusses the conversion of plastic waste into carbon nanomaterials, such as activated carbon (AC) and carbon nanotubes (CNTs), and analyzes the effects of different treatments and production process parameters on the properties of carbon nanomaterials. However, there are some limitations: on the one hand, the application of emerging technologies such as flash joule heating (FJH) technology and microwave-assisted catalytic technology is rarely included in this field; on the other hand, the elaboration on the application of carbon nanomaterials is not comprehensive enough, and there is no mention of the use of life cycle analysis (LCA) and techno-economic analysis (TEA) methods to study the carbonization process.[20−22] Herein, this review provides an overview of recent advancements in the chemical conversion of plastics into carbon nanomaterials. It highlights several widely used conversion methods, including hydrothermal carbonization, high-temperature pyrolysis, templating, rapid carbonization, and pressurized carbonization. We also discuss the key factors influencing the synthesis of carbon nanomaterials in detail. The applications of carbon nanomaterials in environmental remediation, energy storage, and catalysis are evaluated. The challenges and future directions for the plastics-to-carbon pathway are outlined. We hope this article inspires further innovation in greener plastics-to-carbon processes and promotes the efficient recycling of plastic resources.

2. A BRIEF PRIMER ON PLASTICS AND COMMON CHEMICAL CONVERSION METHODS

Plastics can be classified into three categories based on their usage: general-purpose plastics, including polyethylene (PE), polypropylene (PP), poly(vinyl chloride) (PVC), polystyrene (PS), and polyethylene terephthalate (PET), are widely used due to their high production volume, versatile applications, excellent molding properties, and low cost.[23] These plastics are primarily used in industrial products and consumer goods. Engineering plastics, such as polyamide, polycarbonate, and polyformaldehyde, can endure external forces, exhibit strong mechanical properties, resist temperature extremes, and maintain dimensional stability, making them suitable for structural applications.[24−26] Special plastics, including fluoroplastics and silicones, have unique properties and are mainly used in special fields such as aviation and aerospace. General-purpose plastics, which has the highest production volume among the three types, significantly contribute to environmental pollution. Therefore, the traditional chemical recycling methods for general-purpose plastics are introduced.

PE is odorless, nontoxic, and waxy and exhibits excellent low-temperature resistance. Additionally, PE is chemically stable and resistant to most acids and alkalis and is widely used in manufacturing. During the degradation of PE waste, its structure—composed of repeating ethylene monomer units—forms relatively weak C−C bonds, making them prone to hydrogen attack and cleavage, leading to hydrogenolysis reactions. Therefore, hydrogenolysis has become a commonly used method in the chemical recycling of PE. PE involves a catalyst that cleaves the C−C bonds in the molecular chain, generating shorter carbon-chain molecules. However, excessive methane production can pose safety risks and is less valuable as a fuel compared to liquid or solid hydrocarbons, which are undesirable reaction products. Wang and co-workers investigated the mechanism of methane formation during hydrogenolysis, identifying two main pathways (Figure 2a). The first pathway involves the direct cleavage of terminal C−C bonds, where two surface alkyl groups are hydrogenated and desorbed, forming methane without further reactions. The second pathway occurs when at least one alkyl group remains on the catalyst. This group undergoes further dehydrogenation and C−C cleavage, resulting in successive molecular cracking that generates multiple methane molecules. The process continues until the surface alkyl groups are fully hydrogenated and desorbed.[27] After elucidating the mechanism of methane formation, Sun and co-workers synthesized alloyed RuPt/ZrO_2 catalysts by incorporating Pt into a monometallic Ru/ZrO_2 catalyst. Experimental characterization demonstrated that the RuPt alloy exhibits exceptional C−H activation ability, accelerating the hydrogenolysis rate by promoting initial C−H activation while preventing terminal C−C cleavage and methane production. This approach is highly effective for enhancing the hydrogenolysis of PE into liquid fuels.[28] Additionally, the hydrogenolysis of PE faces several challenges. Catalysts significantly influence reaction efficiency and product selectivity, while harsh

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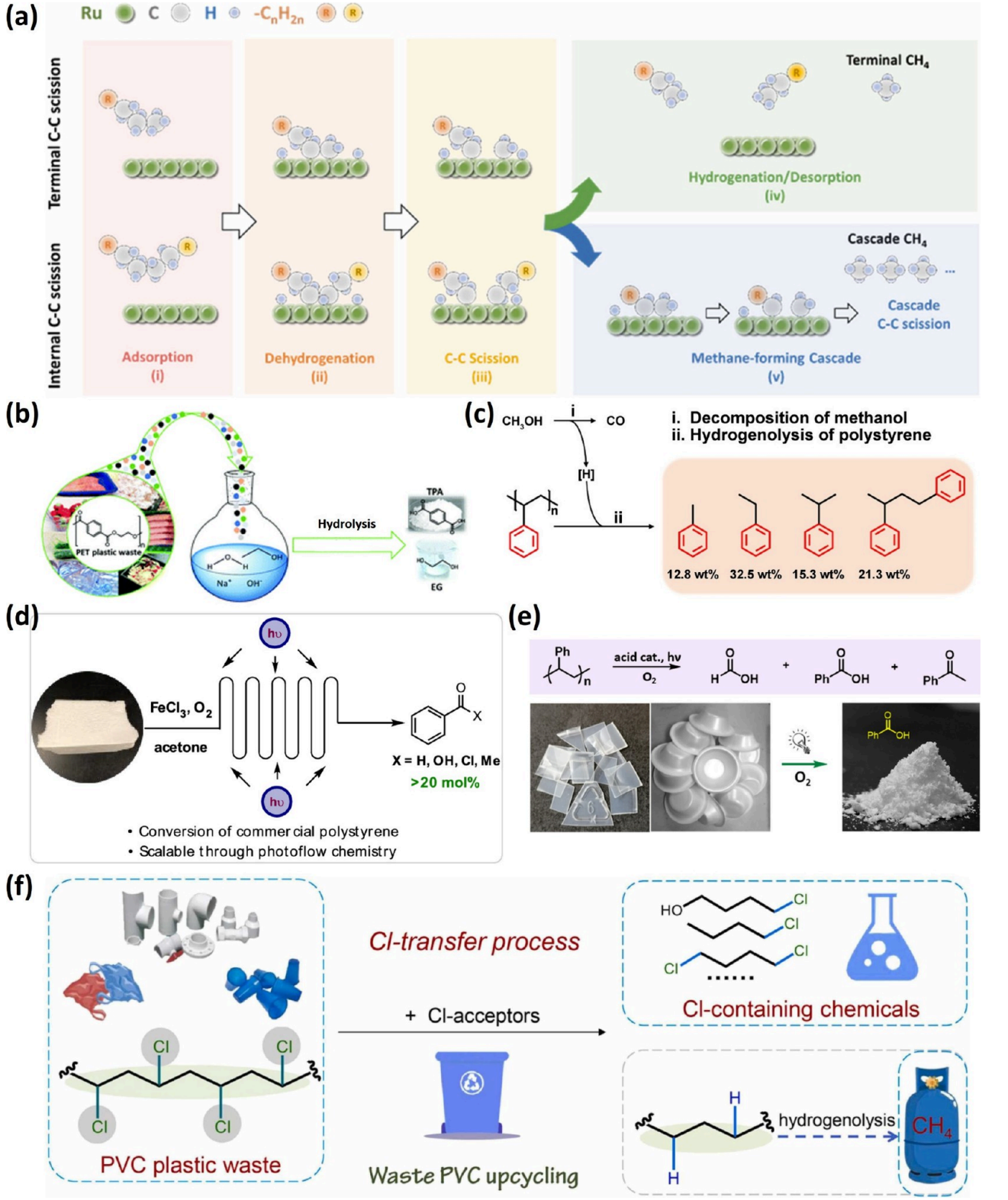

Figure 2. (a) Mechanism diagram of hydrogenolysis of LDPE to produce methane. Reproduced with permission from ref 27. Copyright 2022 Elsevier. (b) Alkaline hydrolysis of PET. Reprinted with permission under a Creative Commons CC BY license from ref 30. Copyright 2020 Royal Society of Chemistry. (c) Reaction scheme of PS-MAD on Ru/SiO_2. Reproduced with permission from ref 34. Copyright 2024 Wiley-VCH. (d) Photocatalytic decomposition of commercial PS. Reproduced with permission from ref 36. Copyright 2022 American Chemical Society. (e) Photoacid-catalyzed degradation of PS. Reproduced with permission from ref 37. Copyright 2022 American Chemical Society. (f) Recycling program for waste PVC. Reproduced with permission from ref 39. Copyright 2023 Elsevier.

reaction conditions require further optimization to improve the process.

PET is widely used in the manufacture of beverage bottles, food packaging, fibers, and a variety of other plastic products because of its

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ACS Nano 2025, 19, 12734−12761

Table 1. Common Conversion Methods for Common Plastics

feedstock	method	reaction conditions	yield and conversion	advantages	challenges	ref
PE	hydrolysis	240 °C, 3 MPa, catalyst $RuPt/ZrO_2$	C20: 93.9% in 1 h	good catalytic performance, high product selectivity	catalyst stability and economics, imperfect reaction mechanism	27, 28
			LDPE: 77.9% in 8 h			
PET	alkaline hydrolysis	80 °C, 20 min, 5 wt % NaOH, $EtOH:H_2O$ = 60:40, 250 or 500 rpm	conversion: 95%	mild conditions, high conversion and yield	high alkali concentration, complex postprocessing	30
			yield: 95%			
	acid hydrolysis	200 °C, 2 h, PET:water = 1:10 (mass ratio)	77% (nitric acid, 200 °C), 85% (HY zeolite, 270 °C)	lower environmental impact, renewable catalysts	catalyst stability, more byproduct	31
PS	PS-MAD	280 °C, 6 h, catalyst Ru/SiO_2	conversion: 93.2 wt %	high conversion, high product recovery	reaction conditions unclear, poor versatility	34
			yield: 84.3 wt %			
	photocatalysis	20 h, catalyst $FeCl_3$, solvent acetone, oxygen-enriched environment	conversion: close to 100%	high efficiency, high product selectivity, mild reaction conditions	restricted reaction conditions, more byproducts	36
			yield: 23 mol %.			
	photoacid catalysis	acid catalysts, 1 bar O_2, violet-blue light at 405 nm, benzene:acetonitrile = 1:1, 15 h	formic acid: 72%	mild reaction conditions, high selectivity	complex postprocessing, poor versatility	37
			benzoic acid: 40%			
			acetic acid: 2%			
PVC	Cl-transfer strategy	180 °C, 2 MPa, 10−20 h, catalyst Ru/Al_2O_3	Cl utilization efficiency: 56.3% (THF as chlorine acceptor)	high conversion efficiency, environmental friendliness	lower selectivity, catalyst stability, cost issues	39
			yield: 56.3%			
			solids conversion: 91.8%			
	PVC−PET copolymerization	230 °C, 8 h, solvent Bu_4PCl, catalysts $ZnCl_2$ and Bu_4PCl	conversion: 100%	efficient chlorine use, lower temperature, low cost.	catalyst stability, difficulty in reprocessing DHPVC	40
			yield: TPA, 98%; EDC, 90%			

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ACS Nano 2025, 19, 12734−12761

transparency, mechanical strength, and chemical resistance.[29] As a chemically stable polymer, PET contains ester bonds in its molecular structure, which can be broken under appropriate conditions. The long polymer chains are depolymerized into terephthalic acid (TPA), ethylene glycol, and other chemicals. Ügdüler and co-workers conducted the hydrolysis of PET in an alkaline solution at atmospheric pressure (Figure 2b). They optimized reaction conditions by adjusting the temperature (50 and 80 °C), sodium hydroxide concentration (5, 10, and 15 wt %), ethanol-to-water volume ratio (20, 60, and 100 vol %), and stirring rate (250 and 500 rpm). The optimal hydrolysis conditions were determined to be 80 °C. A 95% yield was achieved within 20 min under optimal conditions: 80 °C, a sodium hydroxide concentration of 5 wt %, an ethanol-to-water ratio of 60:40 ($EtOH/H_2O$), and a stirring rate of 250 or 500 rpm. A two-step alkaline hydrolysis was performed on postconsumer PET samples with varying particle sizes. The study revealed that sample thickness and crystallinity significantly affected the hydrolysis rate. Notably, the best results were achieved when the PET particle size was less than 500 μm.[30] While alkaline solutions are commonly used for PET hydrolysis, Pereira and colleagues explored the effects of various acid catalysts on the hydrolysis of PET and the recovery of TPA. They employed diverse acid catalysts, including zeolites, inorganic acids, ionic liquids, carboxylic acids, metal salts, and carbon dioxide (CO_2), to perform acid hydrolysis. The study revealed that TPA yield was strongly influenced by the pH of certain catalyst solutions, particularly aliphatic carboxylic acids, nitric acid, and CO_2. However, TPA yields were also influenced by solubility limitations, oxidation, and anionic effects (for metal salts) when using metal salts, ionic liquids, sulfuric acid, or aromatic carboxylic acids as catalysts. Reacting at 200 °C for 2 h, TPA yields reached up to 80% using carboxylic acids and metal salts, offering a promising approach for PET chemical recycling. However, the acidic hydrolysis of PET still faces several challenges, primarily including the high hydrolysis temperature, oxidation of TPA, and the formation of byproducts. Therefore, further research is urgently needed to optimize reaction conditions and improve recycling efficiency.[31] PET hydrolysis has become a widely used method for degradation due to its high recycling efficiency, minimal byproduct formation, and milder reaction conditions. However, challenges remain, such as reducing the concentration of alkaline treatment and enhancing catalyst stability. These issues require further research and optimization.

PS is a polymer synthesized from styrene monomer by free radical polymerization. It is a colorless, transparent thermoplastic with a glass transition temperature above 100 °C and is commonly used in the production of disposable containers and foam lunchboxes. The popularity of COVID-19 has led to a proliferation of single-use plastic products and a dramatic increase in PS waste. However, less than 1% of PS is currently recycled due to difficulties in cleaning and separation.[32,33] In view of this situation, numerous chemical recycling methods have been investigated for PS. Zeng and colleagues developed a methanol-assisted PS depolymerization (PS-MAD) process (Figure 2c). Reacting at 280 °C for 6 h, a high yield of liquid product was obtained, accounting for 93.2 wt % of the original PS, with a productivity of 118.1 $mmol_{carbon}{\cdot}g_{cat.}{}^{-1}{\cdot}h^{-1}$. The main constituents were valuable alkylbenzenes (monocyclic aromatic hydrocarbons and diphenyl alkanes), which together accounted for 84.3% of the liquid product.[34] However, the process requires high temperatures and significant energy consumption, making the depolymerization of PS under mild conditions an urgent issue to address. Photocatalytic recovery is a commonly used chemical method for PS, gaining widespread attention due to its mild conditions and low cost.[35] Oh and Stache employed a catalytically controlled photo-oxidative degradation method, in which $FeCl_3$ undergoes homolytic cleavage under white light irradiation, forming a chlorine radical that extracts an electron-rich hydrogen atom from the polymer backbone.[36] In an oxygen-enriched environment, high-molecular-weight PS (>90 $kg{\cdot}mol^{-1}$) was degraded to below 1 $kg{\cdot}mol^{-1}$, producing up to 23 mol % benzoyl products. This method was subsequently applied to commercial PS, resulting in similar yields (Figure 2d). Huang and colleagues developed a new method for photoacid-catalyzed degradation of PS waste (Figure 2e).[37] Using *p*-toluenesulfonic acid monohydrate ($pTsOH{\cdot}H_2O$) as a catalyst, heavy oxygen ($^1O_2$) was generated by irradiation at 405 nm, leading to the oxidative cleavage of PS to benzoic acid, formic acid and acetophenone. The high degradation yields obtained from applying this process to everyday plastic products open up new avenues for the management and recycling of PS waste. High temperatures and high currents in PS degradation often result in the formation of additional byproducts, which makes photocatalytic technology a preferred method for PS degradation. Although photocatalysis has achieved some success in PS recycling, future research should focus on enhancing product selectivity and reducing the degradation cycle time.

PVC is a thermoplastic synthesized by polymerizing vinyl chloride monomers, commonly used in construction materials, medical equipment, and household goods. PVC is classified into two types: hard PVC, typically used for pipes and door/window frames, and soft PVC, which is made flexible by adding plasticizers, making it suitable for products such as wire insulation and artificial leather. PVC degradation is challenging, and traditional recycling methods can release hazardous substances, such as dioxins, due to the high chlorine content (up to 56%) in PVC.[38] Therefore, in PVC degradation, researchers have attempted to utilize Cl to promote the reaction rather than release Cl from PVC. Feng and colleagues proposed a novel chlorine transfer strategy for the conversion of waste PVC by Cl transfer using Ru/Al_2O_3 as a catalyst. Chlorine acceptors (tetrahydrofuran, dibutyl ether, and butanol) open the ring under the action of the Al_2O_3 Lewis acid site to generate carbon cations and alkoxide anions, and at the same time, the Ru site activates the C−Cl bond of the PVC; the carbon cations then accept the chlorine intermediates of PVC to form chlorine polymers; subsequently, the C−O bond of the intermediates is broken by hydrogenolysis under the synergistic action of the Al_2O_3 acid site and Ru metal site; the C−O bond of the intermediates is broken by hydrolysis. Then the carbon cation accepts the chlorine in PVC to form a polymer intermediate; subsequently, the C−O bond of the intermediate is broken by hydrolysis under the synergistic action of Al_2O_3 acid site and Ru metal site; finally, chlorobactene is desorbed from the surface of the catalyst to form a PE-like polymer (Figure 2f). The Cl-transfer system can also up-cycle several common PVC-containing wastes and has proven effective in scale-up trials. This work offers a catalytic pathway to integrate waste PVC plastics into the organochloride supply chain within the framework of a circular economy.[39] Cao and co-workers proposed a strategy for the coprocessing of PET and PVC. In this scheme, the first step is the dichlorination of PVC, where Bu_4PCl is used as a catalyst and solvent, and at a reaction temperature of 230 °C, the PVC is dechlorinated to produce hydrogen chloride (HCl) and a black solid dechlorinated poly(vinyl chloride) (DHPVC), which is retained in the chlorine-containing liquid. The next step is the depolymerization of PET, which is initiated by the attack of the HCl produced by the PVC dichlorination reaction on the C−O bond of the PET backbone. Following the nucleophilic substitution reaction of chloride ions, TPA, and 1,2-dichloroethane (EDC) are produced. In addition, there is a synergistic interaction between the chloride ion and the Lewis acid catalyst, and this synergy contributes to the efficiency of the reaction as well as the selectivity of the products. Under the reaction condition of 230 °C, the product yield could reach about 95%, showing excellent results in practical applications.[40] Regarding the treatment of PVC, most of the current research focuses on exploiting the potential value of chlorine in PVC with the aim of converting chlorine, which is detrimental to polymer conversion and prone to catalyst poisoning, into usable products, providing new ideas for sustainable plastic waste management. However, the current catalytic conditions are still harsh and this issue needs to be further explored. If the efficient conversion can be achieved under milder conditions, it will be more helpful for its industrialization.

As shown in Table 1, chemical recycling requires different methods for different types of plastics. However, real world plastic waste is typically a complex mixture of plastics, which makes effective degradation and recycling with a single method challenging.

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ACS Nano 2025, 19, 12734−12761

Converting plastic waste into carbon nanomaterials through a one-pot process is a promising approach. This method avoids the need for sorting and pretreatment of waste plastics, thus reducing treatment costs and achieving high-value conversion of waste plastics. It also effectively reduces carbon emissions, ensures product stability and prevents secondary pollution.

3. PLASTIC-DERIVED CARBON NANOMATERIALS

To recycle waste plastics, various methods have been developed to convert them into high-value chemicals. Among these, carbon nanomaterials have gained attention for their excellent properties. However, different synthesis methods and influencing factors lead to variations in morphology and structure, resulting in diverse types of carbon nanomaterials. As shown in Figure 3, graphene, porous carbon, and carbon

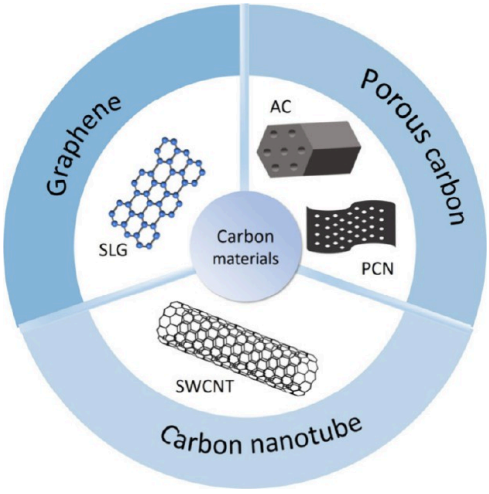

Figure 3. Three common carbon nanomaterials derived from the carbonization of plastic wastes.

nanotubes are three common carbon nanomaterials derived from the carbonization of plastics. They exhibit significant differences in structural dimensions and performance characteristics. Graphene is a two-dimensional honeycomb crystal structure with excellent electrical conductivity and mechanical strength due to the thickness of only one atomic layer. Carbon nanotubes combine excellent electron transport and axial mechanical properties with their one-dimensional tubular structure and high aspect ratio. Porous carbon has a high specific surface area (up to 3000 $m^2 \cdot g^{-1}$) and adsorption capacity due to its three-dimensional interconnected pore network structure with a pore size distribution covering micro-, meso-, and microporous systems, and its surface chemistry can be modulated by doping. These structural differences also determine their potential for different applications in en1uergy storage catalysis and composite materials.

Graphene is distinguished among nanomaterials by its exceptional physical properties. Its carrier mobility can reach 15000 $cm^2 \cdot V^{-1} \cdot s^{-1}$ at room temperature, significantly exceeding that of traditional semiconductor materials. The theoretical thermal conductivity of single-layer graphene is up to 5300 $W \cdot m^{-1} \cdot K^{-1}$, making it one of the best materials for this property. Furthermore, graphene exhibits remarkable strength, with a theoretical Young's modulus of 1.0 TPa, placing it among the strongest known materials (Figure 4a).[41] These excellent properties have enabled widespread applications of graphene materials in fields such as energy storage, electronics, sensors, and biomedicine.[42−44]

Single-layer graphene has a thickness equivalent to that of a carbon atom, approximately 0.34 nm.[45] Chemical vapor deposition (CVD) is currently the primary method for producing single-layer graphene. This technique uses waste PET as a raw material to deposit graphene on a metallic substrate.[46] Graphene produced by this method is highly valuable; recent data indicate that commercial graphene prices range from approximately $67000−200000 per ton. This process offers an effective means for the high-value utilization of plastic waste. This process offers an effective means for the high-value utilization of plastic waste. Like single-layer graphene, bilayer and multilayer graphene also exhibit high electron mobility, mechanical strength, flexibility, and chemical stability. However, bilayer graphene typically displays a Bernal stacking order, enhancing its semiconductor-like properties.[47] Bilayer graphene typically exhibits a Bernal stacking order, making it more akin to semiconductors, which holds great promise.[48] Bilayer and multilayer graphene can be synthesized through solid-state CVD. Using nickel foils as the substrate, multilayer graphene foils with micrometer thickness are produced at 1050 °C from six types of plastic waste (PMMA, PS, PP, PVC, PE, and PET). Flash graphene (FG) is a type of graphene produced through the FJH technique. In this progress, a carbon source is rapidly heated to 3000 K, leading to fast graphitization.[49] Current techniques can synthesize holey and wrinkled flash graphene (HWFG) from mixed plastic waste in seconds. The specific surface area can reach 650−874 $m^2 \cdot g^{-1}$, which is significantly higher than that of conventional graphene materials. In terms of microstructure, HWFG has two- and three-dimensional porous structures rich in micropores and mesopores (Figure 4b,c). In terms of electrical properties, HWFG exhibits good electrical conductivity and its electrical conductivity is excellent.[50] At the level of thermal properties, HWFG has good thermal stability and is able to retain 90% of its mass at a high temperature of 750 °C. In addition, HWFG has a high defect concentration. Together, these properties ensure that HWFG is well suited for metal-free hydrogen evolution reaction (HER) electrocatalysts, lithium metal battery cathodes and CO_2 gas adsorption materials.[51,52] Moreover, the simplicity of this innovative process significantly lowers graphene production costs, enhancing its feasibility for industrial applications.

Plastic-derived graphene materials have the potential to disrupt the conventional reliance on fossil fuels for the synthesis of graphene, thereby transforming waste into a valuable resource. From a structural and functional standpoint, this material has the capacity to satisfy the requirements of numerous disciplines, exhibiting distinctive advantages in the domains of environmental and energy-related applications. However, significant challenges persist in terms of industrialization, including the development of a robust preparation process, effective cost management, and ensuring consistent performance. Nevertheless, with current technological advances, there is a strong possibility of achieving this goal (i.e., the industrial application of plastics-derived graphene materials) in the future. Indeed, it is even expected that the conversion process from plastics to graphene materials can be completed under low energy consumption conditions, thus fully achieving the purpose of resource recycling.

The porous carbon materials are produced by carbonization of plastics and are similar to conventional porous carbon

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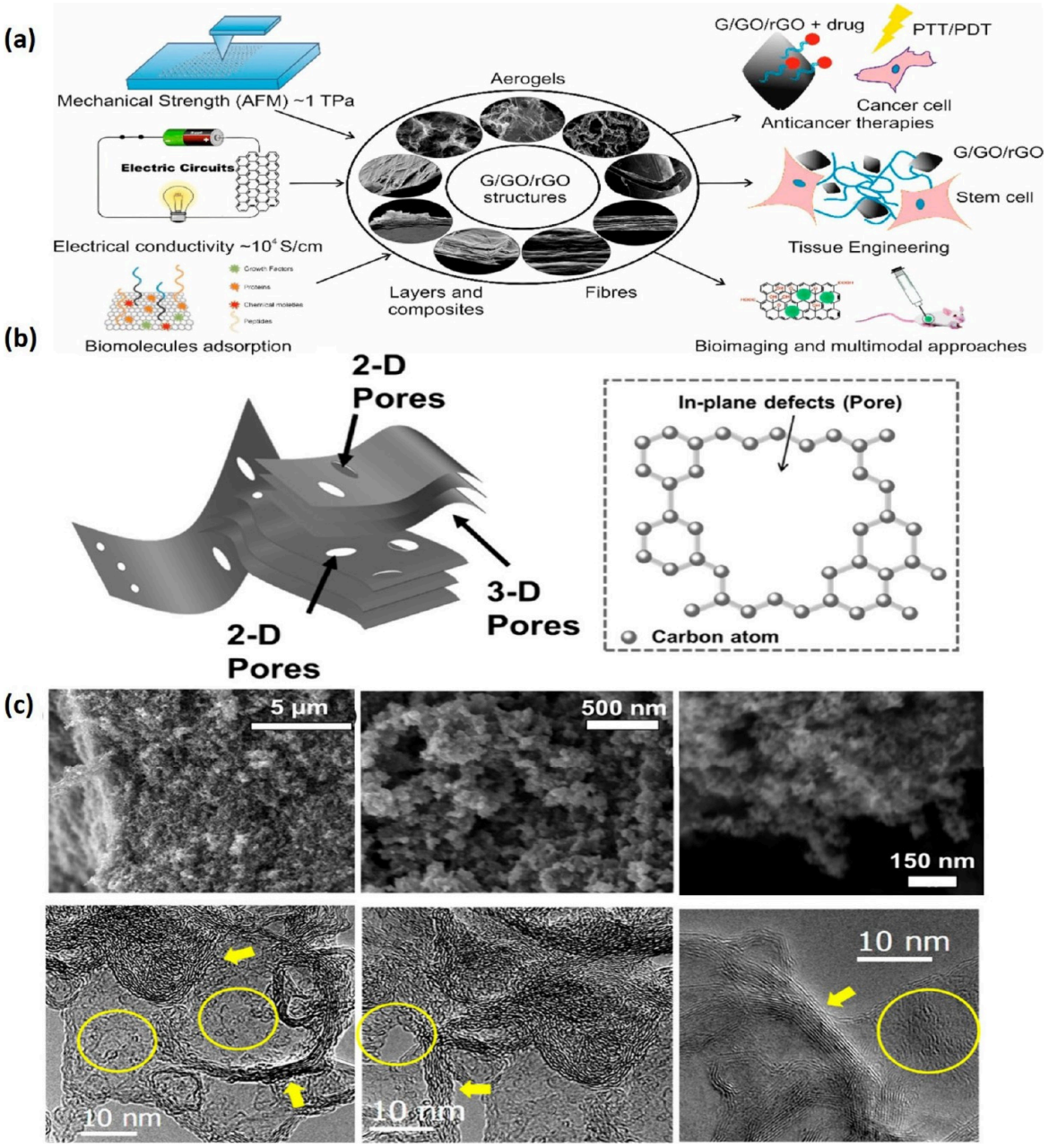

Figure 4. (a) Demonstration illustrating the diverse structures of graphene and their exceptional properties and potential applications in biomedicine. Reproduced with permission from ref 44. Copyright 2018 MDPI. (b) Two- and three-dimensional porous structures in HWFGs. (c) Electron microscopy image of HWFG. The top row of images are SEM images showing three-dimensional macropores; the bottom row of images are TEM images showing two-dimensional meso- and micropores. Parts b and c were reproduced with permission from ref 52. Copyright 2022 American Chemical Society.

materials in that they have a microporous structure, with pore sizes between 0.7 and 1.4 nm, and are rich in micropores and mesopores, presenting both two-dimensional and three-dimensional porous structures. The specific surface area of the material is large, for example, some materials can reach 930.062 $m^2 \cdot g^{-1}$ and even up to 2507 $m^2 \cdot g^{-1}$. Under different conditions, the material shows excellent adsorption capacity for CO_2, with adsorption amounts up to 17.0 ± 1.1 wt % and 6.90 $mmol \cdot g^{-1}$, respectively, and the material shows good stability in multiple adsorption−desorption cycles. The material exhibits good thermal stability, retaining 90% of its mass at 750 °C.[52−55] It also has good electrical conductivity and thermal stability. At the same time, the conductivity is good and the electrical conductivity is excellent, although the defect concentration is high. These properties give this material great potential for energy storage (Figure 5a),[56,57] catalysis (Figure 5b),[58] adsorption and separation. Activated carbon, one of the most common porous carbon materials, is produced through two main steps: carbonization of the raw material to create a carbon surface, followed by activation through chemical oxidation or heat treatment to enhance surface quality (Figure 5c).[59] This process makes activated carbon one of the most widely used adsorbents.

As a result, the porous carbon material derived from plastic carbonization retains its original structural properties while effectively reducing costs, making it an ideal alternative to

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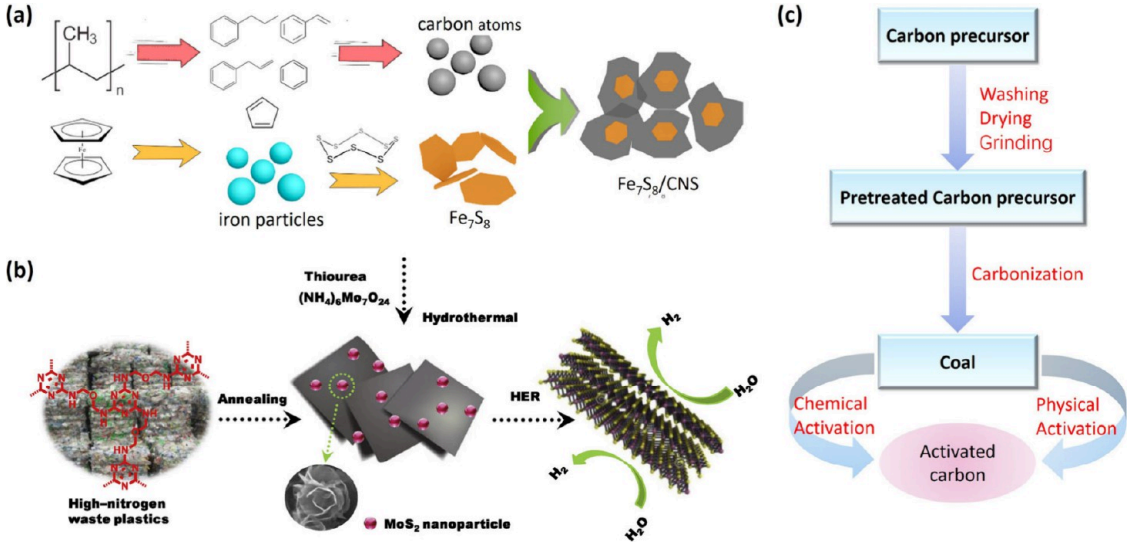

Figure 5. (a) Roadmap for the generation of Fe_7S_8/CNS composites from waste PP. Reprinted with permission under a Creative Commons CC BY license from ref 57. Copyright 2021 Elsevier. (b) Scheme of conductive nitrogen-doped carbon carriers for the conversion of high-nitrogen-containing waste plastics into HER catalysts at elevated temperatures. Reproduced with permission from ref 58. Copyright 2020 Elsevier. (c) General synthesis route of activated carbon. Reprinted with permission under a Creative Commons CC BY license from ref 59. Copyright 2020 MDPI.

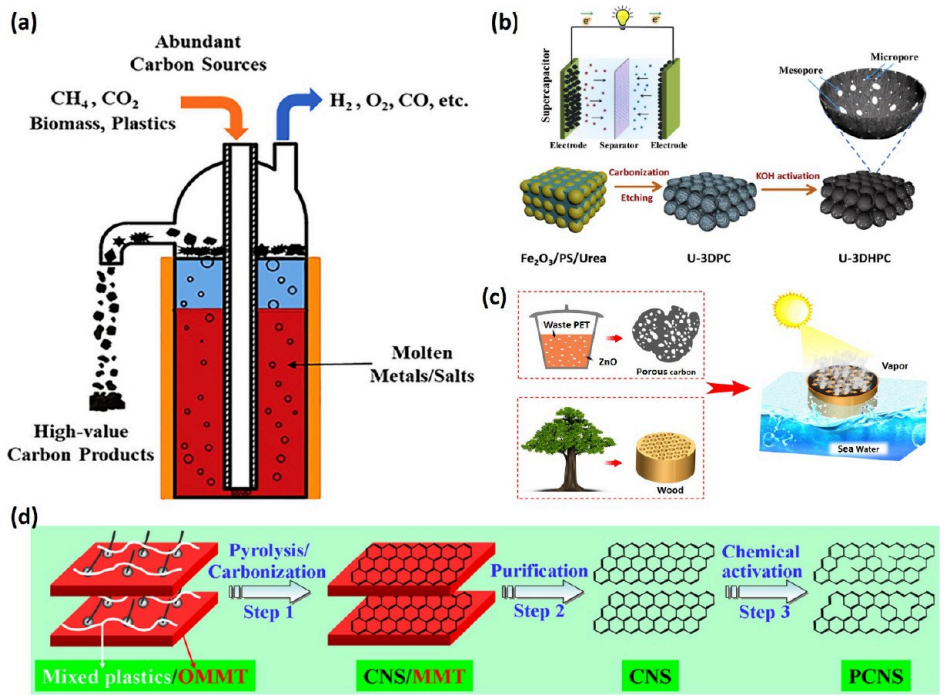

Figure 6. (a) Typical molten pyrolysis system. Reproduced with permission from ref 65. Copyright 2023 Wiley-VCH. (b) Schematic diagram of the synthesis process for the preparation of 3D multistage porous carbon. Reproduced with permission from ref 71. Copyright 2020 Elsevier. (c) Production of porous carbon from PET for solar evaporator applications. Reprinted with permission under a Creative Commons CC BY license from ref 73. Copyright 2022 Wiley-VCH. (d) Process of synthesizing PCNS from mixed plastics. Reproduced with permission from ref 74. Copyright 2014 American Chemical Society.

functional carbon materials. Of particular note, the material exhibits excellent performance in terms of CO_2 adsorption and electrical conductivity. However, further optimization in terms of defect control and structural homogeneity is required to meet the demands of higher end applications.

CNTs are nanoscale tubular materials composed of carbon atoms, characterized by their high strength, lightweight nature, excellent conductivity, and chemical stability. These properties make CNTs highly promising for a wide range of applications in energy storage, nanotechnology, and materials science.[60,61] The production methods for CNTs are not yet fully optimized, with three main techniques in use: laser ablation, arc discharge, and CVD.[62] The CVD method is increasingly popular due to its cost-effectiveness, simplicity, and high synthesis rates of CNTs, with process temperatures ranging from 600 to 1200 °C and efficiencies over 90%. The CVD method has also been

https://doi.org/10.1021/acsnano.5c03391
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applied to produce carbon nanotubes from plastic waste, showing increasing yields and improved properties.[63] Carbon nanotubes generated from the conversion of waste plastics using FJH technology can be synthesized into morphologically controllable one-dimensional structures. The diameter and morphology of the carbon nanotubes can be controlled by adjusting the concentration and type of metal salt catalysts such as iron, nickel and cobalt. The mechanical properties of the carbon nanotubes are superior to those of current commercial carbon nanotubes, and when used in nanocomposites, they can significantly improve their mechanical properties, especially in terms of tensile strength and toughness, reinforcing the fact that they have good reinforcing ability and good bonding properties with the matrix material, and can effectively improve the overall material properties.[64]

Waste plastic-derived carbon nanotube materials have electrical and thermal conductivity close to that of ideal graphene, a property that makes them suitable for flexible electronic devices or thermal management materials. In addition, the functional groups (e.g., −OH, −COOH) on the surface of the material can be modified to enhance compatibility with specific substrates, thus expanding their applications in energy storage or catalytic supports. Overall, these materials show great potential in terms of resource recycling, performance enhancement and cost control. However, key issues such as process stability, large-scale production and environmental risk control still need to be addressed in practical applications.

4. SYNTHESIS STRATEGIES FOR CONVERTING WASTE PLASTICS TO CARBON NANOMATERIALS

Converting waste plastics into carbon nanomaterials provides notable environmental and economic advantages. This process reduces waste volume, mitigates environmental burdens from landfilling and incineration, and converts nondegradable plastics into sustainable carbon nanomaterials, thereby supporting the carbon cycle. High-value carbon products, including activated carbon and carbon nanotubes, are widely used in energy storage, water treatment, and catalysis, improving resource efficiency. These carbon nanomaterials also act as stable catalyst supports, enhancing catalytic performance and stability. Key processes, including pyrolysis, chemical activation, and microwave processing, provide technological support for advancing the circular economy.

4.1. High-Temperature Pyrolysis. High-temperature pyrolysis offers significant advantages in plastic recycling. This method can process various common plastic wastes and eliminates the need for complex sorting and pretreatment. Additionally, pyrolysis effectively reduces plastic waste volume, thereby mitigating its environmental impact. It also generates carbon nanotubes, which have significant potential applications in energy storage, catalysis, and materials science. The molten salt pyrolysis technology commonly used today was developed from the high temperature pyrolysis technology, which uses molten salt as the pyrolysis reaction medium, and is an effective method of converting plastics into carbon nanomaterials (Figure 6a). Two types of molten media are primarily used: molten metals (e.g., Ni, Cu, Sn, Fe, and Ga) and molten salts (e.g., KCl, NaCl, $ZnCl_2$, KBr, and NaBr), with each presenting unique advantages and challenges.[65] Frank Riedewald and colleagues developed a molten metal pyrolysis reactor that efficiently decomposes plastic waste. Direct contact between waste plastics and molten metal enables complete pyrolysis within 15 min, significantly faster than traditional indirect reactors. This method produces hydrocarbons, amorphous carbon, and recyclable catalysts, facilitating efficient material recovery and reuse.[66] Molten salts offer excellent heat transfer properties and lower the pyrolysis temperatures (420−480 °C), improving energy efficiency and reducing cost. Kong and colleagues cracked a mixture of PP and anhydride-modified PP in a high-pressure reactor to synthesize helical carbon nanotubes with diameters ranging from 20 to 60 nm. At 400 °C, amorphous carbon was identified as the primary product.[67] Along with carbon nanomaterials, the process yields high-value chemicals such as light oils, paraffins, and aromatics.

Molten salt pyrolysis is a cost-effective and energy-efficient alternative to conventional high-temperature pyrolysis, but it can also release harmful gases, so effective emission control measures are required.[68−70] High-temperature pyrolysis technology is promising for plastics recycling, but it is important to balance economic feasibility and environmental impact in practical applications. Future research should focus on optimizing process conditions, improving material recovery rates and developing sustainable emission control technologies to increase the viability of these processes.

4.2. Template-Assisted Method. The template method for carbonizing plastics regulates the carbonation process using specific template materials to produce carbon nanomaterials with defined morphologies and structures. The success of this method depends on the selection of the template, as its characteristics directly influence the final morphology and performance of the carbon nanomaterials. Depending on the template's nature, this method can be classified into hard and soft template methods.

The hard template method typically employs structurally stable inorganic materials (such as SiO_2, Al_2O_3) as template, allowing for the preparation of carbon nanomaterials with highly ordered pore structures that can be easily removed. Ma and colleagues used Fe_2O_3 particles as catalysts and templates to carbonize PS waste into novel three-dimensional (3D) hierarchical porous carbon (Figure 6b). They further applied KOH activation to the macroscopic pore walls to generate micropores and mesopores, resulting in carbon nanomaterials with high specific capacitance and good multiplicity performance, which were applied to supercapacitors.[71] Wang and colleagues synthesized nitrogen-doped hierarchical porous carbons using polyacrylonitrile as a precursor, with nano silica (nano-SiO_2) and $ZnCl_2$ serving as pore-forming agents in a one-step pyrolysis process. This carbon material shows great potential for capacitive deionization applications, effectively removing salt and heavy metal ions.[72] In addition, Liu and colleagues employed an active template method to carbonize plastics. They utilized metal additives in PVC as hard templates to prepare porous carbon materials with a high specific surface area (1164 $m^2 \cdot g^{-1}$) and well-controlled micropore, mesopore, and macropore structures via KOH activation and acid washing. This material not only enhances light absorption ability but also promotes the formation of water molecule clusters, thereby reducing the enthalpy of water evaporation. It was applied in solar evaporators to achieve efficient water transport and steam release (Figure 6c).[73]

The soft template method relies on self-assembled structures of surfactants and other flexible molecules as templates, allowing for diverse and easily regulated template structures. For instance, Gong and colleagues prepared carbon nanosheets

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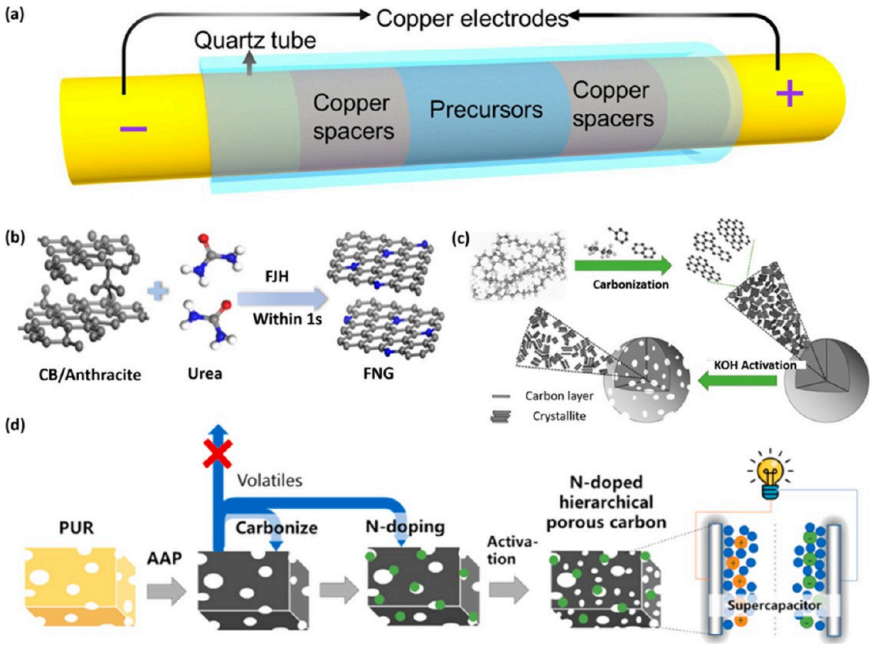

Figure 7. (a) Typical FJH schematics. Reproduced with permission from ref 78. Copyright 2022 American Chemical Society. (b) Synthesis of nitrogen-doped graphene by the FJH method. Reprinted with permission under a Creative Commons CC BY license from ref 78. Copyright 2022 American Chemical Society. (c) Mechanism diagram of autogenous pressure carbonization and KOH activation of LDPE. Reproduced with permission from ref 79. Copyright 2019 American Chemical Society. (d) Schematic diagram of NHPC synthesis. Reproduced with permission from ref 80. Copyright 2022 Elsevier.

(CNSs) through catalytic carbonization of organically modified montmorillonite, using PP, PE, PS, PET, and PVC as raw materials (Figure 6d). The resulting KOH-activated porous CNS (PCNS) exhibited a high specific surface area (1734 $m^2 \cdot g^{-1}$), a large pore volume (2.441 $cm^3 \cdot g^{-1}$), and high purity (over 99.5%), demonstrating excellent performance in CO_2 absorption and hydrogen storage.[74] In addition, Lian and colleagues enhanced the thermal stability of PE during heat treatment by introducing magnesium carbonate pentahydrate. This approach provided a template for MgO while contributing to the carbonation process. The resulting carbon material improved the porosity and nitrogen content after ammonia activation, significantly enhancing its electrochemical performance.[75]

Template carbonized plastic technology can convert plastic carbonization into carbon nanomaterials with high mechanical properties, improving strength, hardness and wear resistance, and the carbon layer formed by carbonization helps to improve flame retardancy, which has a broad application prospect. However, the technology is a complex process that requires precise control of multiple parameters and high equipment and process requirements, which increases the cost and technical difficulty. In addition, due to the complexity of plastic types and compositions, carbonization is prone to quality instability, which affects product consistency and performance. There is therefore a need to further develop new catalysts to increase yield, reduce cost and better control the microstructure of the products to achieve more efficient use of resources.

4.3. Rapid Carbonation. FJH technology is a highly efficient method that can rapidly heat materials to extremely high temperatures (Figure 7a is a typical FJH device). It is both environmentally friendly and cost-effective for treating plastic waste.[64] Luong and colleagues converted raw materials into "flash-heated graphene" (FG) by heating carbon source material to 3000 K in less than one second. This process yields FG with over 90% efficiency, which can be applied in sustainable 3D printing and sensor technology.[76] Advincula and colleagues optimized the FJH process to successfully convert rubber waste into flash-heated graphene, which exhibits good dispersion and has the potential to enhance cement-based materials.[77] Zhu and colleagues developed a one-pot, solvent- and catalyst-free FJH method to synthesize nitrogen-doped graphene (FNG), amorphous carbon black and urea precursors are rapidly transformed into high-quality FNG in less than one second using short electrical pulses and intense blackbody radiation flashes (Figure 7b). The resulting FNG features a highly graphitized, swirled structure with a surface-normalized capacitance of 152.8 $\mu F \cdot cm^{-2}$ at 1 $A \cdot g^{-1}$, a capacitance retention of 86.1% at 128 $A \cdot g^{-1}$, and a breakdown relaxation time of 30.2 ms. These properties make FNG a promising candidate for high-performance supercapacitors.[78]

The technology is highly efficient and can improve productivity. In addition, the FJH method has lower energy requirements (86−92% reduction) and global warming potential (92−94% reduction) compared to conventional synthesis methods.[64] However, it requires specialized equipment and is associated with high equipment costs and difficult process control. Regarding equipment improvement, with the development of technology, the equipment cost is expected to decrease while the performance and stability will improve, thus laying the foundation for wide-scale application. For process optimization, an in-depth study of the heating mechanism and carbonization process will be conducted to optimize the parameters, thereby improving the carbonization effect and surface quality.

4.4. Pressurized Carbonation. Pressurized carbonization of plastics is a process that converts plastics into carbon nanomaterials through high temperature and pressure. This technology is commonly employed to convert waste plastics into valuable carbon nanomaterials or to produce high-

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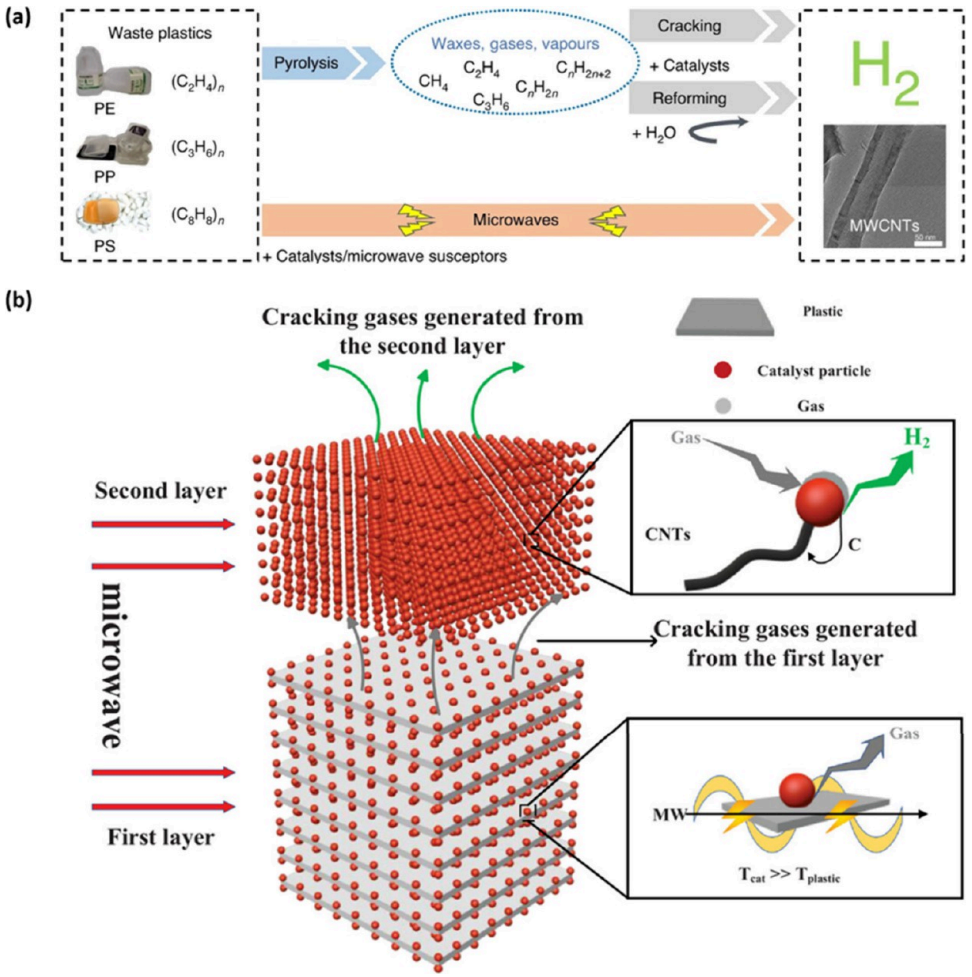

Figure 8. (a) Schematic diagram of the two-stage pyrolysis gasification process and one-stage microwave catalytic process. Reproduced with permission from ref 81. Copyright 2020 Springer Nature. (b) Schematic diagram of the reaction process of the DLMP method. Reproduced with permission from ref 82. Copyright 2024 Wiley-VCH.

performance carbon/carbon composites. Pressurized carbonization enhances the mechanical and chemical stability of materials. Zhang and colleagues used low-density polyethylene (LDPE) as a precursor and decomposed it into short-chain aliphatic hydrocarbons under high-temperature conditions. Confining these small-molecule products in a closed reaction vessel leads to an increase in pressure inside the vessel, which drives the aromatization polycondensation reaction, resulting in the formation of a carbon layer. To keep the surface energy at a minimum, spheres are formed spontaneously. This is followed by activation and etching with potassium hydroxide (KOH), which embeds metal K and releases CO/CO_2, resulting in the production of high-porosity carbon (HPC). The resulting HPC exhibits a micron-sized spherical carbon morphology with graded pores, a high specific surface area of up to 3059 $m^2 \cdot g^{-1}$ and abundant surface functional groups. As an electrode material for supercapacitors, HPC exhibits excellent electrochemical properties: a specific capacitance of up to 355 $F \cdot g^{-1}$, an energy density of 9.81 $Wh \cdot kg^{-1}$ and a power density of 450 W in 6 M KOH electrolyte at a current density of 0.2 $A \cdot g^{-1}$, and has excellent cycling stability (Figure 7c).[79] Zhou and colleagues converted polyurethane (PUR) foams into nitrogen-doped hierarchical porous carbon (NHPC) using PUR foams as substrates, also by autogenous atmosphere pyrolysis (AAP)-KOH activation (Figure 7d). The method achieved a carbon yield of 55.0%, which is more than 17 times higher than that of conventional PUR pyrolysis, and the KOH activation resulted in a significant increase in the specific surface area of the carbon material to 2057 $m^2 \cdot g^{-1}$ and the introduction of a hierarchical porous structure with O-containing functional groups into the carbon material, which was used to enhance the performance of the NHPC for supercapacitors. Electrochemical measurements showed that the NHPC exhibited a high specific capacitance of 342 $F \cdot g^{-1}$ (133 $F \cdot cm^{-3}$), low resistance and excellent cycling stability at 0.5 $A \cdot g^{-1}$ current. The energy and power densities of the supercapacitor were improved to 11.3 $Wh \cdot kg^{-1}$ and 250 $W \cdot kg^{-1}$ respectively.[80]

Pressurized carbonization technology improves the mechanical properties and chemical stability of carbon nanomaterials but is energy-intensive. To enhance its economic feasibility, future research should focus on economic-technical analyses.

4.5. Microwave-Assisted Catalytic Pyrolysis. Microwave-assisted catalytic pyrolysis is a chemical process that combines microwave radiation with a catalyst. This method takes advantage of the efficient heating properties of microwave radiation to accelerate catalytic reactions and improve their efficiency. Microwave energy heats the reaction system quickly and uniformly, thereby enhancing catalytic efficiency. Jie and colleagues proposed a one-step microwave decom-

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ACS Nano 2025, 19, 12734−12761

position process for high-density polyethylene (HDPE). The plastic was mechanically shredded and mixed with $FeAlO_x$ catalyst particles (microwave receptors) in a 1:1 weight ratio. During the reaction, microwave electromagnetic energy is absorbed directly at the microwave absorption sites of the catalyst particles. As a result, heat is rapidly generated at these sites, without first heating the surrounding HDPE sheet. This method decomposes plastics quickly, reducing side reactions typical of CP, and achieving good control of hydrogen production and high yields. Additionally, a high carbon yield of 1,560 mg per gram of catalyst (relative to each gram of plastic) can be achieved through continuous addition of plastic, with the composition containing over 92% multiwalled carbon nanotubes (MWCNTs; Figure 8a).[81]

Wang and colleagues developed a simple double-layer microwave-assisted pyrolysis (DLMP) method for PE, also using $FeAlO_x$ catalysts. They covered the catalyst and PE blends with a new catalyst layer, enabling efficient conversion of PE into hydrogen and multiwalled carbon nanotubes. They achieved a high hydrogen yield of 66.4 $mmol \cdot g^{-1}$ PE, approximately 93% of the theoretical upper limit for hydrogen yield in the PE pyrolysis process. The high hydrogen yield is attributed to further pyrolysis of the oil fraction generated in the first layer, producing a gas product in the second layer, which reduces the oil fraction in the gas product. However, this also results in a reduction in the amount of multiwalled carbon nanotubes produced in the second layer (Figure 8b). Thus, DLMP is effective in producing higher yields of H_2, but the diameter distribution of the produced carbon nanotubes is not uniform, requiring further modification before use.[82] Jiang and colleagues employed microwave pyrolysis to aid the carbonation of postconsumer PET plastic using microwave absorbers, cobalt nitrate and α-cellulose, converting PET into a porous carbon catalyst material activated with peroxomonosulfate (PMS) for carbamazepine (CBZ) degradation. The synthesized carbon material exhibits remarkable porous structural properties, with active components uniformly anchored in the material. After PMS activation, the material effectively accelerated CBZ degradation, realizing the 'waste for waste' concept and offering a new approach to antibiotic degradation in water.[83]

The techno-economic evaluation performed by Li and coworkers showed that microwave processing (MP) technology has significant advantages in terms of hydrogen yield and product distribution compared to conventional pyrolysis (CP). In particular, the hydrogen utilization efficiency can be as high as 97.65% when using an iron-based catalyst. In addition, with an internal rate of return of 39% and a payback period of 2.5 years, MP technology is significantly more profitable than CP technology. In particular, with the use of catalysts, MP technology can achieve a return of US$577 per ton of plastic.[84]

It can be seen that microwave pyrolysis for plastic carbonization has considerable potential for plastic waste treatment due to its low energy consumption and high efficiency. However, several factors influence its application, including microwave power, pyrolysis temperature, catalyst type, and microwave absorbers. Future research should prioritize optimizing these key parameters and balancing their effects to improve the technology's overall efficiency and stability.

As shown in Table 2, the current methods for converting plastics into carbon nanomaterials are mainly pyrolysis or catalytic conversion, which convert waste plastics into high-

Table 2. Summary of Carbonation Methods

method	reaction temperature (°C)	product	advantages	challenges	ref
high-temperature pyrolysis	400–1050	graphene carbon nanotubes	simple pretreatment, simple technology, diversified products	more byproducts, high energy consumption, stable product quality	65–70
Template-assisted method	200–950	graphene carbon nanotubes, porous carbon materials	uniform and controllable product morphology, higher quality of carbon materials	harsh reaction conditions and difficult template removal	71–75
rapid carbonation	400–1000	graphene, carbon nanotubes	high efficiency and possibility to prepare carbon materials with special properties	requires precise control of reaction conditions and is costly	76–78
pressurized carbonation	700–1050	graphene carbon nanotubes, porous carbon materials	improvement of properties such as hardness, strength and wear resistance of materials, improvement of heat resistance of materials	complex processes, high technology costs	79, 80
microwave-assisted catalytic pyrolysis	50–600	solid carbon, carbon nanomaterials	high thermal efficiency, fast reaction speed, high product selectivity	low technology maturity and need to develop efficient microwave absorption catalysts	81–84

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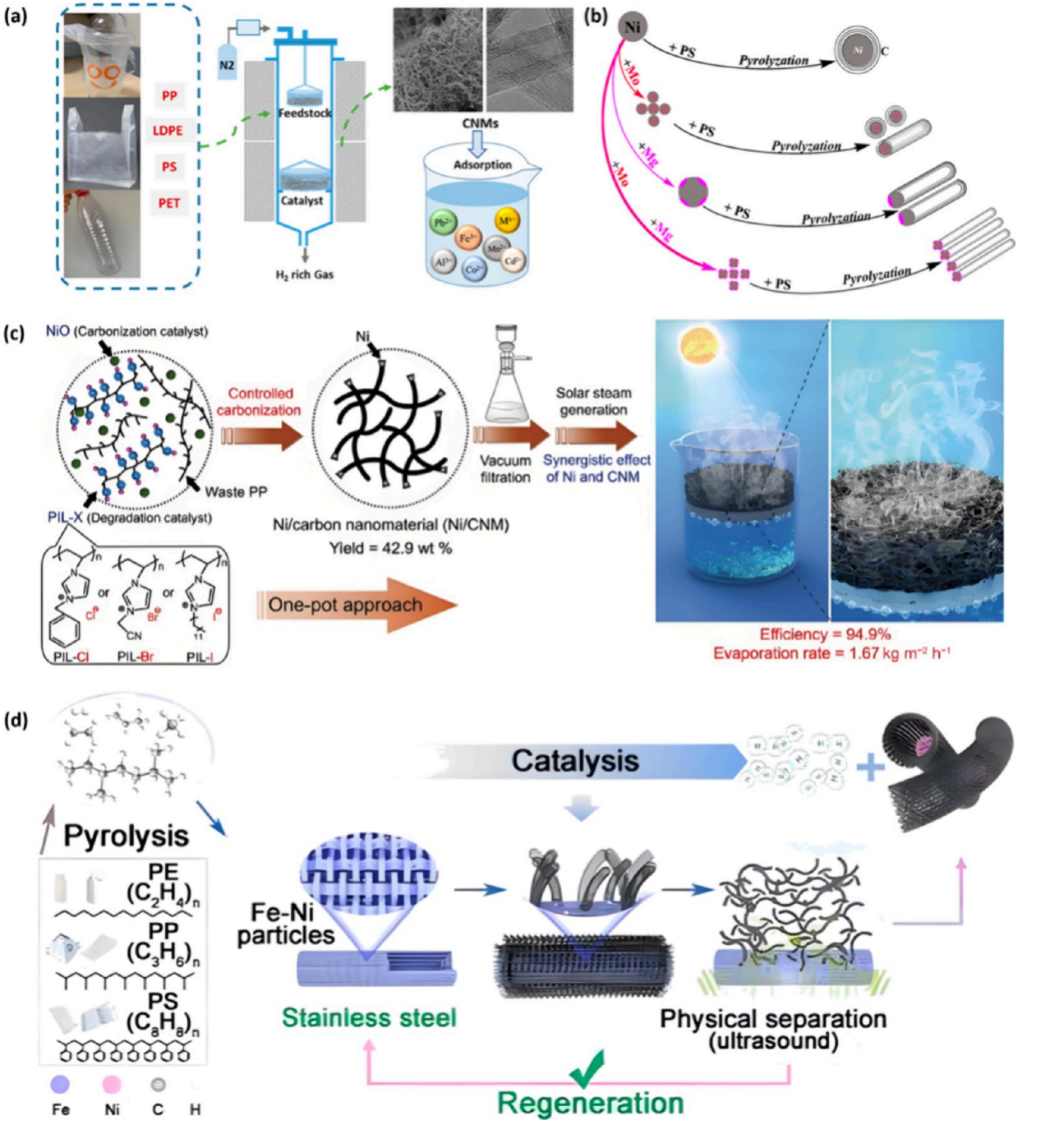

Figure 9. (a) Pyrolysis catalysis of waste plastics to produce carbon nanomaterials with hydrogen-rich gas. Reproduced with permission from ref 87. Copyright 2022 American Chemical Society. (b) Schematic illustration of the effects of molybdenum and nickel on PS carbonization. Reproduced with permission from ref 90. Copyright 2017 American Chemical Society. (c) Schematic diagram of Ni/CNM carbonized by PP Ni/CNM for solar steam power. Reproduced with permission from ref 93. Copyright 2020 Science China Press. (d) Catalytic pyrolysis of plastics into MWCNTs and H_2 over monolithic multilayer stainless steel mesh catalysts. Reproduced with permission from ref 94. Copyright 2023 National Academy of Sciences.

value carbon nanomaterials such as graphene and carbon nanotubes. These methods have great potential for resource recovery and pollution reduction compared to traditional treatment methods. However, their practical application is limited by three major challenges: first, high energy requirements and high operating costs; second, the risk of secondary pollution, in particular emissions of volatile organic compounds (VOCs); and third, technical limitations in treating plastics with different levels of contaminants. Further optimization of reaction conditions, development of efficient catalysts and improvement of the stability of product properties are needed to achieve large-scale industrialization.

5. KEY FACTORS FOR PLASTICS CONVERSION INTO CARBON NANOMATERIALS

Converting plastics to carbon nanomaterials involves various factors that influence the properties of the final product, including catalyst selection, reaction conditions, and the

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characteristics of the plastic feedstock. The activity and selectivity of the catalyst directly determine the type of carbon material produced. Additionally, reaction conditions—such as temperature, pressure, and reaction time—are key parameters that regulate both yield and quality. Variations in raw materials, including polymer types and molecular weights, also significantly impact product properties. Thus, a thorough understanding and precise control of these variables are critical for improving the performance and yield of carbon nanomaterials.

5.1. Catalyst Structure Regulation. Catalysts are crucial in the plastic carbonization process, the size effect of the catalyst, preparation methods, and the metal−support interactions can influence the yield and quality of carbon nanomaterials.

5.1.1. Metal Components. Metal catalysts play a crucial role in accelerating pyrolysis and carbonization reactions, leading to higher yields and improved product quality.[85] They can also control reaction pathways, enhancing product selectivity and boosting the conductivity and mechanical strength of the resulting carbon nanomaterials. Transition metals, such as iron, cobalt, and nickel, are particularly favored for their high catalytic activity and low cost, making them ideal candidates for these reactions.[22]

Yao and colleagues prepared catalysts with varying Ni−Fe molar ratios using the impregnation method to pyrolysis real waste plastics, producing hydrogen and carbon nanotubes. Experimental results indicated that catalysts with higher Fe content produced more hydrogen and deposited carbon due to their superior cracking ability and lower interaction between active sites and supports. Ni enhances the thermal stability and graphitization of carbon. Hydrogen concentration and yield peaked at 73.93 vol % and 84.72 $mg{\cdot}g^{-1}$ of plastic, respectively, when the Ni molar ratio was 1:3.[86] They also investigated the effect of six different catalysts on the yields of hydrogen and carbon nanomaterials, and the experimental results showed that among the six different catalysts (Figure 9a), Co−Fe−Mg and Ni−Fe−Mg exhibited higher carbon yield and lower liquid yield, Fe−Ni−Mg performed best, with a carbon yield of 30.25 wt % and a hydrogen yield of 31.52 $mg{\cdot}g^{-1}$ plastic. The bimetallic catalyst activities ranked in descending order: Fe−Ni, Co−Fe, and Co−Ni, indicating that iron atoms play a crucial role in carbon material synthesis.[87]

Ni-based catalysts are highly effective in converting waste plastics and hydrocarbons into hydrogen by efficiently breaking C−C bond.[88] Studies have explored adding different metals to Ni-based catalysts, with Ni−Mn−Al catalysts emerging as the most promising for coproducing hydrogen and carbon nanotubes. Li and colleagues demonstrated that Ni plays a pivotal role in catalyzing the conversion of PS to CNTs. Ni not only serves as the main active component of the catalyst but also is key in the formation mechanism of CNTs.[89] The addition of Mo and Mg to Ni-based metallic materials has distinct effects: Mo facilitates the reduction of catalyst particles by forming the $NiMoO_4$ phase, while Mg, despite not altering catalyst particle size, enhances carbon nanotube growth through $NiMgO_2$ formation, effectively controlling carbon solubility (Figure 9b).[90] Wang and colleagues, found that in the catalytic pyrolysis of waste plastics for carbon nanotube production, a significant amount of carbon was generated on the catalyst, predominantly as multiwalled carbon nanotubes. The Ni catalyst produced the highest yield of filamentary carbon (~93 wt %). However, when Mg-based catalysts were employed, the strong metal-carrier interaction inhibited carbon nanotube growth, resulting in shorter and irregular nanotubes.[91]

The metal molar ratio can control the yield of carbon nanomaterials. Wu and colleagues. found that Ni−Mn−Al catalysts with a 4:4:4 molar ratio produced higher carbon yields than those with a 4:2:4 ratio. This is attributed to the higher Mn content, which increases reducible metal oxides and enhances catalytic efficiency.[92] Song and colleagues employed poly(ionic liquid) (PIL) and nickel oxide (NiO) as composite catalysts to convert waste PP into Ni/CNM via a controlled carbonization process (Figure 9c). The morphology and structural characteristics of Ni/CNM were precisely regulated by adjusting the amount of PIL added; Ni/CNM comprised cuprous carbon nanotubes (CS-CNTs) and pear-shaped nickel metal nanoparticles. The synergistic effect of Ni and CS-CNT in solar light absorption endows Ni/CNM with excellent photothermal conversion performance. Furthermore, Ni/CNM exhibits a high specific surface area and abundant micropores, mesopores, and macropores, creating a three-dimensional (3D) porous network for efficient water supply and vapor channeling.[93] Furthermore, Liu and colleagues employed a monolithic multilayer stainless steel mesh catalyst for the catalytic pyrolysis of plastics to produce MWCNTs and H_2 (Figure 9d). The experimental results demonstrated recovery efficiencies of 86% for carbon and 70% for hydrogen. This achievement results from a unique redox dynamic process combined with careful modifications to the catalyst's surface microstructure, which effectively enhance the full exposure of the active sites. This series of sophisticated design and modulation directs the formation pathway toward MWCNTs while effectively limiting the production of complex macromolecular hydrocarbon byproducts. Notably, after 10 cycles, the decline in carbon recovery efficiency was only 5%, strongly demonstrating the stability of the selected catalyst system during long-term operation. Furthermore, this catalyst, owing to its excellent vapor−solid−solid conversion mechanism, demonstrates the capability to efficiently convert various aromatic hydrocarbons into high-quality multiwalled carbon nanotubes, and exhibits high compatibility and flexibility in processing different types of waste plastics.[94]

The impact of metal catalyst components on the plastic carbonization process exhibits multidimensional complexity. In active component systems, Fe-based catalysts modulate catalytic efficiency through particle size and crystallinity optimization, while Ni-based variants demonstrate superior hydrocarbon dissociation capabilities. Co-based systems exhibit exceptional structural stability under reaction conditions, whereas Mn- and Zn-based additives serve as effective promoters for olefin selectivity enhancement. Bimetallic catalysts reveal synergistic mechanisms, where precise metal ratio adjustments dictate catalytic performance. Collectively, these compositional and structural parameters govern both activity and selectivity in plastic carbonization. Future investigations should prioritize mechanistic elucidation of component interactions through advanced characterization techniques, coupled with computational modeling for rational catalyst design.

5.1.2. Particle Size of the Catalyst. The size effect of catalysts significantly influences the diameter of carbon nanotubes. Research by Kukovitsky and co-workers show that, typically, smaller metal particles lead to the growth of carbon nanotubes with smaller diameters, while larger metal

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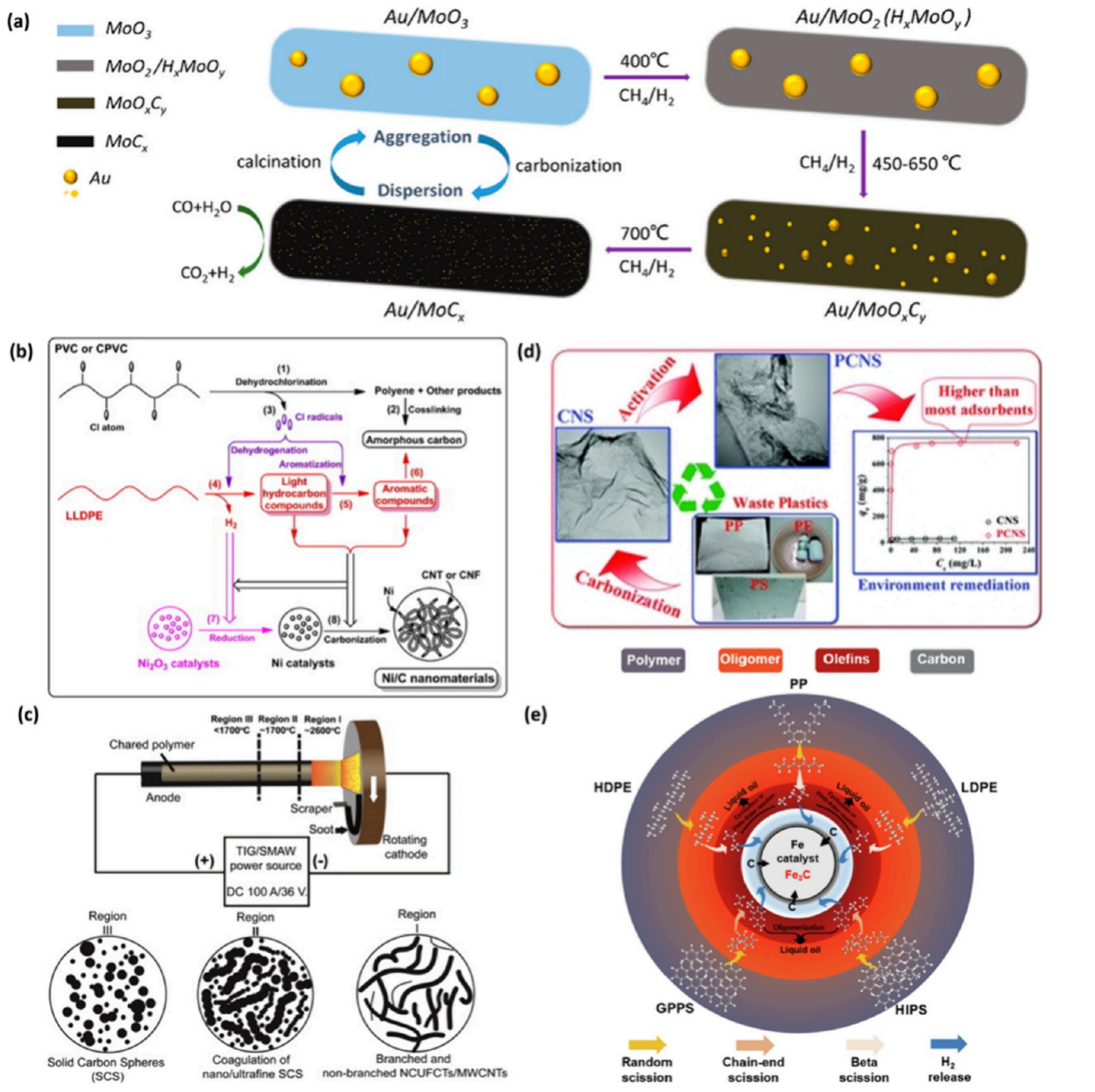

Figure 10. (a) Schematic of the SMSI effect between metal carbide catalysts. Reproduced with permission from ref 97. Copyright 2018 American Chemical Society. (b) One-pot catalytic carbonization of LLDPE with Ni_2O_3 and PVC resin to produce magnetic Ni/C nanomaterials. Reproduced with permission from ref 101. Copyright 2013 Elsevier. (c) Representation of conversion to NCUFCTs, MWCNTs, and SCSs in PET waste by using arc-discharge technology. Reproduced with permission from ref 103. Copyright 2014 Elsevier. (d) Carbonizing real mixed plastics and activating with KOH to produce PCNS. Reproduced with permission from ref 106. Copyright 2015 Royal Society of Chemistry. (e) Catalytic conversion of various types of plastics over Fe/Al_2O_3 catalysts. Reproduced with permission from ref 107. Copyright 2021 Elsevier.

particles result in larger diameter nanotubes. This occurs because metal particles act as catalysts, with their surfaces controlling the deposition rate and pattern of carbon atoms, thus influencing the nanotube diameter.[95] Furthermore, the size of metal particles may impact the catalyst's activity and selectivity, influencing the growth mechanism of carbon nanotubes.

Liu and colleagues developed nickel- and iron-based catalysts with different particle sizes for the thermal catalytic conversion of waste plastics into carbon nanotubes and hydrogen gas. Their results showed that iron-based catalysts with large particle sizes (about 80 nm), achieved the highest hydrogen production (about 25.60 mmol $H_2 \cdot g^{-1}$ plastic) and the highest carbon production (29 wt %), as well as the highest proportion of graphite carbon; In addition, whether Fe- or Ni-based catalysts, larger metal particles produced more hydrogen production than smaller particles.[96]

Catalyst particle size exerts multiple effects on the plastic carbonization process, encompassing catalytic efficiency, reaction selectivity, and stability. Smaller particle size enhances catalytic efficiency as it offers a larger surface area, providing more reaction sites and reducing activation energy. It also

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ACS Nano 2025, 19, 12734−12761

influences reaction selectivity by altering product distribution and controlling reaction pathways. Larger particle size exhibits better abrasion resistance, while smaller particles may have weaker antipoisoning capabilities. In practical applications, the appropriate particle size of the catalyst should be selected based on the specific reaction system and requirements. Although some understanding has been gained, future research could also explore the synergistic mechanisms between particle size and other factors, precisely prepare catalysts with specific particle sizes, and study their microscopic properties and reaction behaviors.

5.1.3. Strong Metal–Support Interactions. Metal supports can influence both the reaction efficiency and the properties of the final products. As shown in Figure 10a, strong metal–support interactions (SMSI) can modify the electronic structure and geometric configure ration of metal active sites, thereby affecting the activity and selectivity of the carbonization reaction.[97] Acomb and colleagues synthesized nickel, iron, cobalt, and copper catalysts via impregnation for producing carbon nanotubes and hydrogen from LDPE. Carbon nanotubes were successfully generated on nickel, iron, and cobalt catalysts, but were scarcely observed on copper. Iron and nickel catalysts yielded the highest hydrogen and carbon nanotube production due to optimal metal–support interactions, which were neither too strong (as in cobalt) nor too weak (as in copper). This demonstrates that metal–support interactions are a key factor in carbon nanotube production.[98]

Yao and colleagues investigated the effect of catalyst composition and support material. Among the monometallic catalysts, Fe/γ-Al_2O_3 achieved the highest hydrogen yield (22.9 mmol $H_2 \cdot g^{-1}$ plastic) and carbon nanotube yield (195 $mg \cdot g^{-1}$ plastic), followed by Fe/α-Al_2O_3, Ni/γ-Al_2O_3 and Ni/α-Al_2O_3. Bimetallic Ni–Fe catalysts exhibited higher catalytic activity in terms of hydrogen yield than the monometallic Ni or Fe catalysts, due to optimal interactions between the metal and the support.[66]

They also selected four different porous materials, namely, ZSM-5, MCM-41, NKF-5, and H-Beta as catalyst supports and Ni–Fe bimetal as the active component. During the pyrolysis of waste plastics, the Ni–Fe/MCM-41 catalyst exhibited the highest catalytic activity, yielding carbon material with a mass fraction of 55.60%. This indicates that the metal active sites of the catalyst were more fully exposed during the reaction process, which in turn enhanced its activity. This is attributed to the good metal dispersion and moderate metal carrier interactions of the Ni–Fe/MCM-41 catalyst, which can effectively promote the formation of carbon nanotubes, and the formed carbon nanotubes have high purity and graphitization degree. It can be seen that regulating the metal–support interaction can optimize the performance of the catalysts, and the appropriate metal ratio and carrier type can improve the activity and selectivity of the catalysts, thus increasing the yield and quality of the carbon nanotubes.[99]

Metal–carrier interactions affect polymer carbonization in a number of ways. They can increase the reactivity and influence product selectivity by altering reaction pathways, increase the dispersion and stability of metal particles to extend catalyst life, and induce charge transfer and modulate electronic structure to influence catalytic performance, reactant adsorption, and activation. However, there are some drawbacks to such catalysts; too strong metal-carrier interactions can limit the accessibility of active sites and increase the complexity and cost of preparation. Further studies of the interactions between different metals and novel supports and their modes are needed to develop better catalyst systems.

5.1.4. Preparation Temperature of the Catalyst. The calcination temperature significantly influences catalysts in several ways, including phase transformation, particle size, metal–support interactions, catalytic activity, and product distribution. An optimal calcination temperature enhances the interactions between the metal and support, thereby improving the activity and selectivity of catalyst. Moreover, variations in calcination temperature affect the sintering behavior of catalyst particles, impacting their surface area and activity. Furthermore, calcination temperature influences the properties of reaction products, such as the diameter and uniformity of carbon nanotubes. Therefore, appropriately adjusting the calcination temperature is crucial for optimizing catalyst performance.

Acomb and co-workers recognized that different calcination temperatures affect the structure of the catalyst and thus the yield of carbon nanotubes with hydrogen. They therefore calcined Ni-based catalysts at 500 and 750 °C respectively and used them to produce carbon nanotubes and hydrogen from LDPE feedstock. It was found that the catalyst calcined at 750 °C was able to produce more hydrogen during the reaction and the resulting carbon nanotubes were of higher purity. This is mainly due to the weaker metal–support interactions between the catalysts generated at lower calcination temperatures, which resulted in more severe sintering of the catalysts and affected the yield of carbon nanotubes.[91] Chai and colleagues investigated the effect of catalyst calcination temperature on the diameter uniformity of CNTs synthesized via methane decomposition. The CoO-MoO/Al_2O_3 catalysts used were not prehydrogen reduced. The results demonstrated that calcination temperature significantly influenced CNT diameter uniformity. CNT diameters from CoO-MoO/Al_2O_3 catalysts calcined at 300, 450, 600, and 700 °C were 13.4 ± 8.4, 12.6 ± 5.1, 10.7 ± 3.2, and 9.0 ± 1.4 nm, respectively. This suggests that increasing calcination temperature leads to smaller diameters, but at 750 °C the catalyst became inactive in methane decomposition.[100] Kong and colleagues examined the impact of different calcination temperatures on the performance of the catalyst, and the experimental results showed that the reaction started at 400 °C, when the main product was amorphous carbon. The yield of CNTs continued to increase with increasing temperature, and when the temperature reached 700 °C, more than 80% of the products were CNTs, both straight and helical.[70] The calcination temperature of the catalyst affects its activity, which subsequently influences the morphology, structure, and yield of carbon nanomaterials. Thus, precise control of the calcination temperature is essential for optimizing carbon material production.

Appropriate calcination temperature can promote the formation and exposure of catalyst active sites to increase activity, while too high a temperature can led to sintering of active components and deactivation of active components due to a reduction in specific surface area; it can also improve structural stability and increase the ability to resist sintering, carbon build-up and toxicity, while too high or too low a temperature can reduce stability. By precisely controlling the calcination temperature, it is expected that catalysts with better performance and stability can be developed, further improving the efficiency and reducing the cost of plastics carbonation.

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5.2. Catalytic Reaction Conditions. *5.2.1. Pyrolysis Temperature.* In the catalytic pyrolysis of plastics, reaction temperatures significantly influence the yield of CNTs. Gong and colleagues converted linear low-density polyethylene (LLDPE) into magnetic Ni/C nanomaterials through the combined catalysis of Ni_2O_3 and PVC resin (Figure 10b). The study found that increasing the pyrolysis temperature enhanced the yield of magnetic Ni/C nanomaterials, likely due to higher temperatures promoting nickel catalyst activity, which favors the growth of CNTs or carbon nanofibers.[101] Bajad and colleagues successfully performed batch synthesis of carbon nanotubes using waste plastics as the hydrocarbon source and Ni/Mo/MgO as the catalysts. The production process revealed that increasing the cracking temperature from 450 to 700 °C reduced the oil yield from 20 g per 30 g of PE to 1.2 g per 30 g of PE, while the yield of CNTs increased from 0.23 g per 30 g of PE to 6.033 g per 30 g of PE.[102] Yang and colleagues investigated the effect of catalytic reaction temperature on CNTs and hydrogen yields in a waste plastic gasification system. Experiments were conducted at temperatures of 600, 680, and 750 °C using H−Ni/Al_2O_3 as the catalyst. Results indicated that hydrogen concentration increased from 34.53 vol % to 36.13 vol % with rising reaction temperature, and higher temperatures promoted CNT formation by enhancing methane dry reforming, hydrocarbon dry reforming, and direct decomposition. Although higher reaction temperature enhanced the yield of hydrogen and CNT, the highest quality CNT was obtained at 680 °C. At this temperature, the CNT structure is smoother and more uniform.[87]

Berkmans and colleagues synthesized carbon nanomaterials from PET waste using the arc discharge method, where varying temperatures in different regions of the anode affected the morphology of the carbon nanomaterials during the arc discharge process. As illustrated in Figure 10c, in the lower temperature region (~1700 °C), ultrafine and nanosized solid carbon spheres (SCSs) were formed, with average diameters of 221 and 100 nm. In contrast, in the higher temperature region (~2600 °C), these carbon spheres transformed into long "Y"-shaped branched and unbranched nanochanneled ultrafine carbon tubes (NCUFCTs) and MWCNTs, with average diameters of 364 and 95 nm.[103]

Veksha and colleagues found that at 500 °C, the noncondensable cracking gases of mixed plastics were converted into a mixture of carbon nanocages and multiwalled carbon nanotubes, exhibiting a higher degree of graphitization than the carbon nanomaterials produced from PP and LDPE, which predominantly consisted of multiwalled carbon nanotubes. In contrast, at 800 °C, the synthesis of carbon nanomaterials occurred through the formation of intermediates (such as methane, ethylene, and condensable oils), which reacted on nickel-based catalysts to produce multiwalled carbon nanotubes. This indicates that, at higher synthesis temperatures, carbon nanomaterials of consistent quality can be obtained, regardless of the heterogeneity of the plastic feedstock.[104]

5.2.2. Steam Injection Volume. Acomb and colleagues investigated the effect of controlling steam input during pyrolysis gasification affects the quality of carbon nanotubes. They found that increasing the rate of steam injection enhanced the hydrogen yield from plastic waste via steam reforming. Varying the steam injection rate revealed that increasing steam amounts raised the yield of carbon nanotubes. This occurs because steam acts as a weak oxidizing agent that reacts with amorphous carbon, which can deactivate the catalyst, promoting the growth of longer and purer CNTs. However, once a critical concentration is reached, the yield decreases drastically.[105] Therefore, regulating the steam injection rate can effectively control the yields of hydrogen and carbon nanotubes during the pyrolysis gasification of waste plastics.

Variations in reaction conditions can affect the carbonization process of plastics, which in turn affects the quality, yield and structure of the resulting carbon nanomaterials. Among the reaction conditions, temperature variation plays a key role. Different temperatures will change the thermal movement of the plastic molecules, which will affect the rate and extent of the carbonization reaction and ultimately be reflected in the quality, yield and structure of the carbon nanomaterials. Differences in the oxidizing or reducing nature of the reaction atmosphere can also affect the direction of the carbonization reaction. For this reason, the reaction conditions must be carefully controlled to produce carbon nanomaterials with specific structures.

5.3. Plastic Feedstock. Different plastic raw materials differ in type, composition, structure, and carbonization yield, and these differences affect plastic carbonization in many ways. For example, high-molecular-weight polymers such as polyimide have high thermal stability and are less susceptible to carbonization, while low-molecular-weight polymers such as PP tend to carbonize at high temperatures due to the structure of their molecular chains. Plastics contain a high proportion of carbon, a characteristic that makes the conversion of waste plastics into high value-added carbon nanomaterials important in the recycling of waste plastics and the production of functional carbon nanomaterials. Different plastic raw materials can produce carbon nanomaterials with different structures during the carbonization process. In addition, the carbonization yields of different raw materials vary. Taking waste PP as an example, its carbonization yield will change significantly under certain conditions. In conclusion, in the research and application of plastic carbonization, it is necessary to select the appropriate carbonization conditions and methods according to the characteristics of plastic raw materials.

In 2015, Gong and colleagues investigated the conversion of real-world waste plastics (PP, PE, and PS) into porous carbon nanosheets (PCNSs) for the adsorption of organic dyes from wastewater (Figure 10d). The PCNSs exhibited excellent properties due to their high specific surface area, large pore volume, and various adsorption mechanisms, including pore-filling, hydrogen bonding, π-π interactions, and electrostatic interactions.[106] In selecting precursors for carbon material production, PP, PE, and PS are considered suitable due to their high carbon content. However, Aboul-Enein and colleagues found that PS produces aromatic hydrocarbons during pyrolysis, making it less effective as a precursor for carbon synthesis.[66] Veksha and colleagues examined how different plastic feedstocks influence the yield of carbon nanomaterials using catalytic chemical vapor deposition (CCVD) to convert noncondensable gases from plastic pyrolysis at 500 or 800 °C. At 500 °C, the hybrid plastic MP, consisting of carbon nanocages and multiwalled carbon nanotubes, showed a higher degree of graphitization compared to carbon nanomaterials obtained from PP and LDPE alone. In addition, the carbon yield was highest from PP, followed by LDPE, with MP yielding the least carbon.[104]

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ACS Nano 2025, 19, 12734−12761

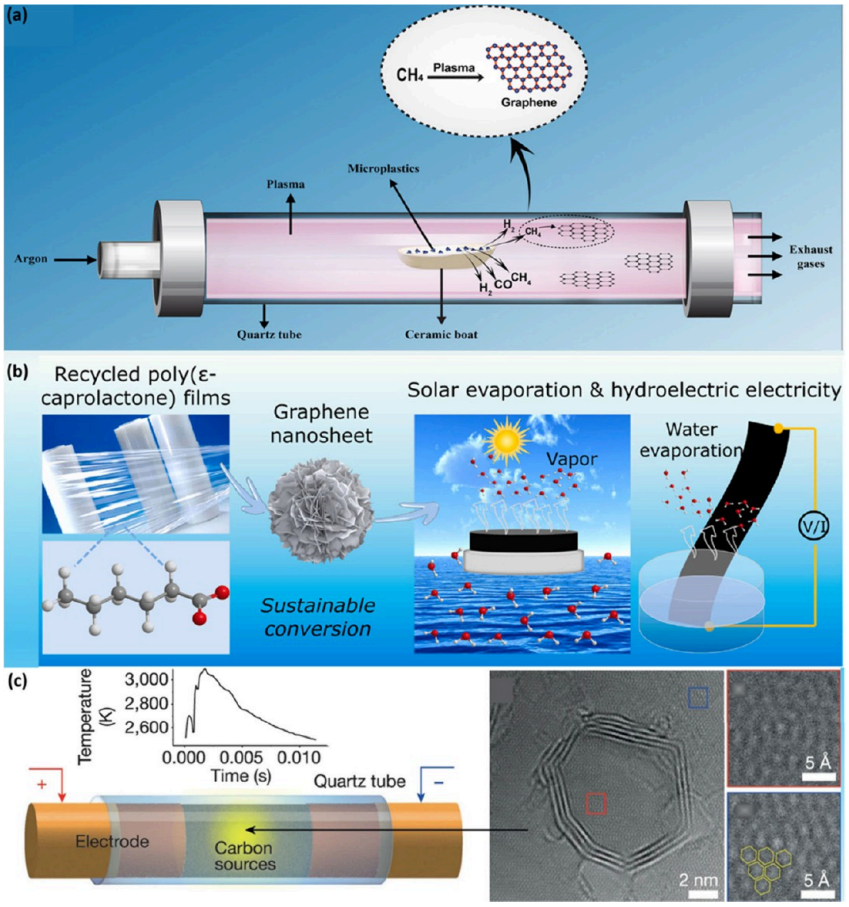

Figure 11. (a) APMP system for the synthesis of graphene using PE microplastics. Reprinted with permission under a Creative Commons CC BY license from ref 109. Copyright 2024 Wiley-VCH. (b) Design of GNS-*x* evaporator made from PCL waste for solar powered interfacial evaporation and hydroelectric power generation. Reproduced with permission from ref 111. Copyright 2024 Elsevier. (c) Schematic diagram of the FJH process and a plot of temperature rise versus time in the flash evaporation process. Reprinted with permission under a Creative Commons CC BY license from ref 76. Copyright 2020 Springer Nature.

Cai and colleagues investigated the pyrolysis-catalytic conversion of five types of waste plastics: PP, HDPE, LDPE, high-impact polystyrene (HIPS), and general-purpose polystyrene (GPPS) using Fe/Al_2O_3 catalysts (Figure 10e). Their results showed that HIPS and GPPS had higher solid carbon deposition yields (49.4 and 48.7 wt %), over 10 wt % more than PP, HDPE, and LDPE. This is due to the formation of small molecules like styrene and benzene, which support CNT growth during HIPS and GPPS pyrolysis. Thus, PS-based plastics are more suitable for producing solid carbon nanomaterials, particularly those with more amorphous carbon. In contrast, PP, HDPE, and LDPE yielded carbon deposits with over 75% graphitic carbon, making them more appropriate for CNT production. Additionally, ethylene from PE enhanced carbon deposition, resulting in more uniform distribution and less amorphous carbon compared to PP.[107]

Yao and colleagues investigated the effect of different types of different plastic raw materials affect the production of carbon nanomaterials (CNMs). They found that PS plastics yielded the highest CNM, likely due to their high carbon content, low hydrogen content, and greater liquid product generation. LDPE produced carbon and gas yields as PP, likely because their chemical structures and compositions are similar. PET, with its high oxygen content (around 33 wt %), had low carbon yields as oxygen bonds with carbon during pyrolysis, generating large amounts of CO_2, making PET unsuitable for CNM production. When investigating mixed plastics, they found that even with PET present, the final product yields were not significantly affected. This suggests that mixed plastics can be used directly for carbon material production in industrial applications without the need for sorting.[85]

Catalysts, reaction conditions and the nature of the feedstock are critical influences in the carbonization of plastics. At the catalyst level, the type of catalyst, the choice of metal and the synergies between the support materials have a significant effect on many properties, while deactivation mechanisms such as carbon build-up and sintering must also be considered. In the realm of reaction conditions, temperature has a significant effect on reaction rates and product distribution, pressure affects reaction pathways and equipment costs, reaction time requires a balance between conversion and selectivity, and atmosphere can affect reaction pathways and product properties. In terms of feedstock, the type of plastic determines thermal stability and decomposition behaviors, impurities can poison the catalyst or alter the reaction pathway, requiring pretreatment, and molecular structure affects the decomposition mechanism and product distribution. These factors are interrelated and affect each other, and future research should focus on dissecting the multiscale process correlation and revealing the dynamic laws through in situ characterization techniques, thus promoting the industrial application of plastic carbonation.

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ACS Nano 2025, 19, 12734−12761

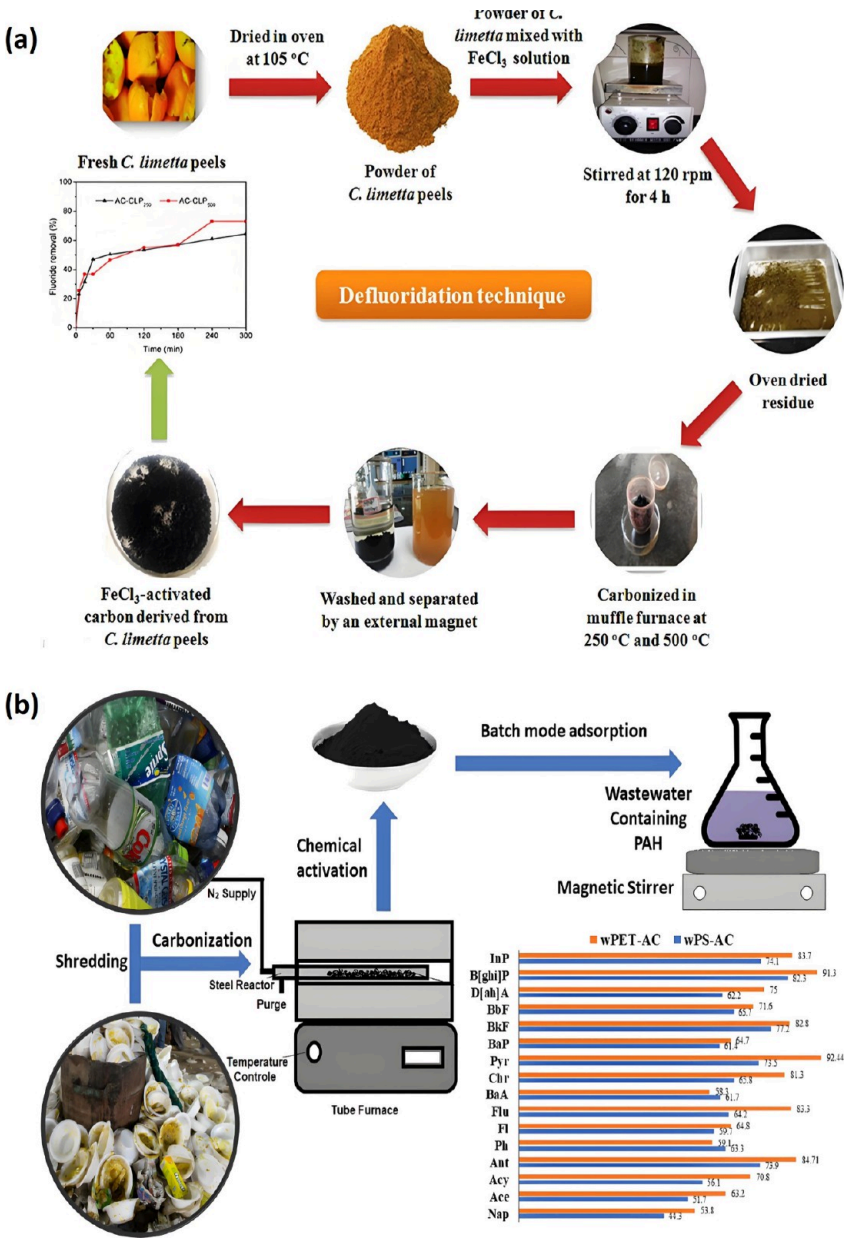

Figure 12. (a) Citrus peel waste AC-CLP. Reproduced with permission from ref 120. Copyright 2020 Elsevier. (b) Chemical modification of PET and PS for production of activated carbon. Reproduced with permission from ref 122. Copyright 2021 IWA Publishing.

6. APPLICATIONS OF CARBON NANOMATERIALS

Converting waste plastics into high-value carbon nanomaterials is an effective approach for producing high-value products at low cost, demonstrating strong economic feasibility. Additionally, carbon nanomaterials derived from waste plastics typically exhibit excellent electrical conductivity and thermal stability, making them ideal for high-performance applications such as supercapacitors and conductive materials.

6.1. Application of Graphene. Graphene is a thin-layered carbon material with excellent thermal conductivity, mechanical strength, current density, electron mobility, and large surface area, which can be applied to environmental remediation, materials science and other fields.[108] The graphene material obtained from the conversion of waste plastics can be applied to the adsorption of perfluorooctanoic acid (PFOA) in water with excellent performance; it can be used as a substrate material for photovoltaic evaporator or recycled as an additive for PUR foam, which can effectively improve the performance of the material.

Zafar and colleagues proposed an atmospheric pressure microwave plasma (APMP) synthesis technique (Figure 10a).[109] Compared with traditional graphene synthesis methods, the APMP method of this technology breaks through the inherent limitations of multistep reactions, improves the production efficiency, and also circumvents the problems of impurity of intermediates that may occur in multistep reactions as well as the difficulty of precisely controlling the reaction conditions. In this technology, a plasma breaks down microplastics into gases such as methane, ethylene, ethane, CO_2, hydrogen, and carbon monoxide. Directed deposition of the carbon source onto the quartz tube wall was achieved by controlling the plasma power density (2.5−3.8 kW·cm^{-3}) and residence time (50−200 ms). The adsorption capacity of the synthesized graphene for PFOA was about 30% for samples

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ACS Nano 2025, 19, 12734−12761

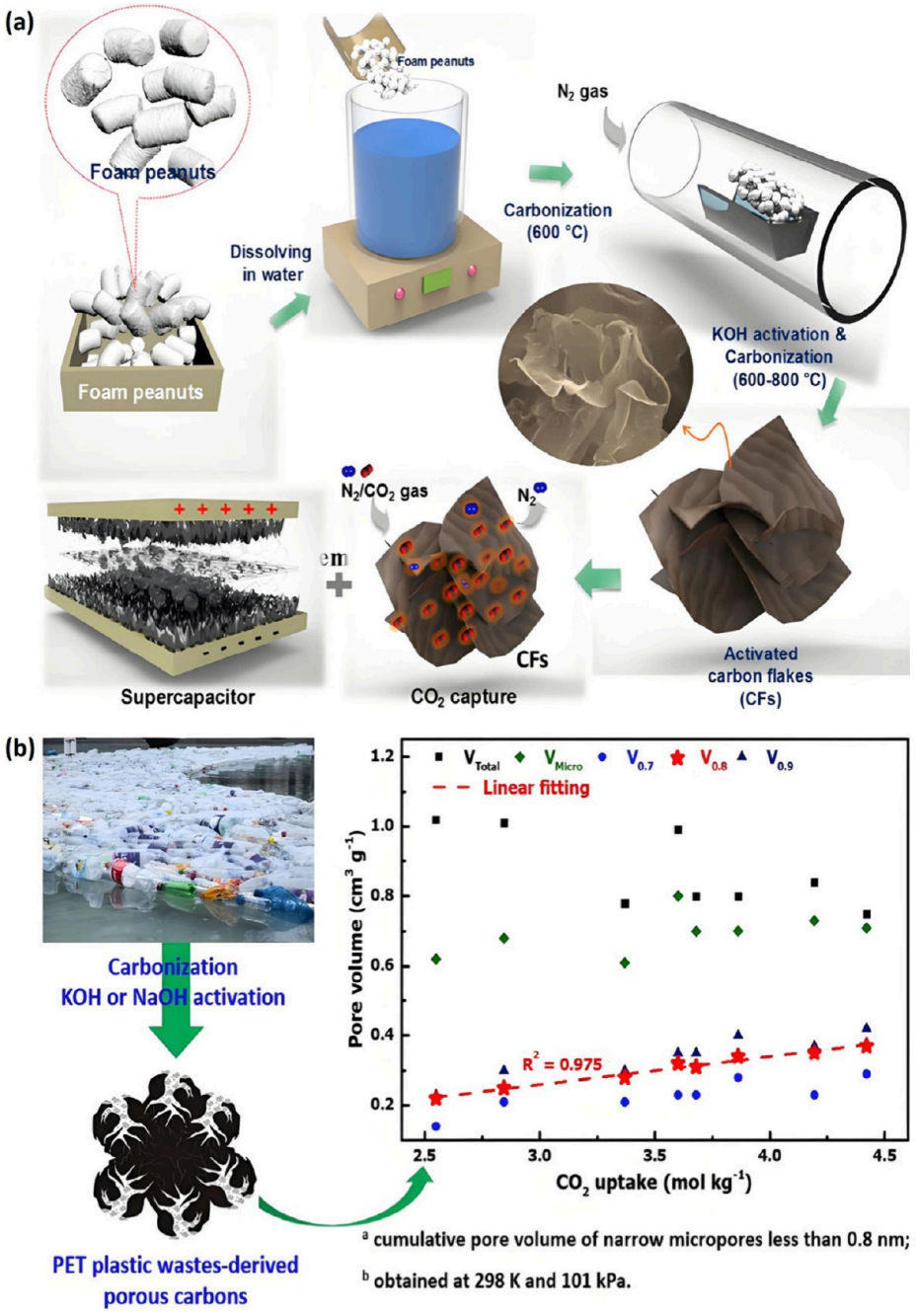

Figure 13. (a) Synthesis of foamed carbon nanosheets for CO_2 capture and supercapacitors. Reproduced with permission from ref 123. Copyright 2019 American Chemical Society. (b) PET as a precursor for the synthesis of microporous carbon for CO_2 capture. Reproduced with permission from ref 124. Copyright 2020 Elsevier.

ultrasonicated for 15 min and 32% for 30 min, which is higher than the 3.3% reported for graphene oxide samples adsorbed using an orbital oscillator.[110] The FeG composite, which was further formed by combining with iron oxides, showed more than 90% adsorption of PFOA. The advantage of this composite is that it combines the properties of the mineral and carbon phases and can provide multiple binding sites to enhance the adsorption of PFOA.

Hu and colleagues converted waste poly(ε-caprolactone) (PCL) into graphene using a salt-assisted carbonation strategy. The process involved milling PCL with NaOH at 400 rpm for 2 h, where PCL underwent solid-state alkaline hydrolysis and depolymerized into Na-CL. The Na-CL was then pyrolyzed in a nitrogen atmosphere at various temperatures for 1 h, followed by aqueous scrubbing, washing, and drying. The graphene has a 7−8 layers structure with curved edges, structural discontinuities, and is rich in oxygen-containing groups. A graphene-based hybrid evaporator was then prepared using the impregnation method (Figure 11b). It exhibited a high sunlight absorption rate of 98%, excellent photothermal conversion performance, strong water transport capacity, low water evaporation enthalpy, and a thermal conductivity of 0.06 $W \cdot m^{-1} \cdot K^{-1}$. The water evaporation rate reached 2.92 $kg \cdot m^{-2} \cdot h^{-1}$, surpassing many advanced photothermal materials. This achievement not only promotes the upcycling of waste plastics,

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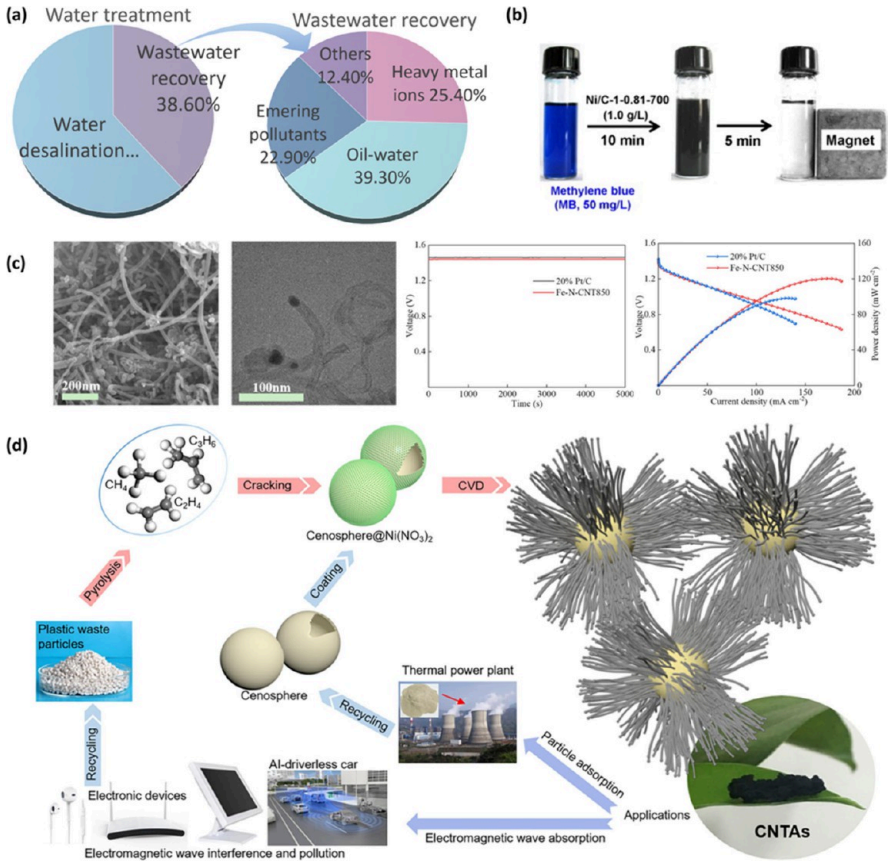

Figure 14. (a) CNT composite membranes for water treatment. Reprinted with permission under a Creative Commons CC BY license from ref 125. Copyright 2017 MDPI. (b) Photographs of the adsorption behavior and magnetic separation phenomena of magnetic Ni/C-1-0.81-700 nanomaterials. Reproduced with permission from ref 101. Copyright 2013 Elsevier. (c) Plastic waste-derived Fe−N−CNTs for zinc−air cells. Reprinted with permission under a Creative Commons CC BY license from ref 127. Copyright 2020 Wiley-VCH. (d) Flowchart for the preparation and applications of CNTAs. Reproduced with permission from ref 129. Copyright 2024 Elsevier.

but also provides a new platform for the manufacture of fresh water and power cogeneration equipment.[111]

When cars are scrapped, most precious metals and electronic components can be recycled, but plastics weighing up to 350 kg often end up in landfill.[112] In 2020, Tour and his team reported a method for producing graphene from mixed waste plastics (Figure 11c).[76] The method involves heating a waste plastic resistor to extremely high temperatures through a rapid discharge process, turning it into highly carbonized plastic. An electric current is then applied, converting it into graphene by breaking and rearranging the C−C bonds. The graphene can be used as an additive in PUR foams for automotive acoustic insulation and vibration management. It improves the foam's performance by 34% in tensile strength and 25% in low-frequency noise absorption, facilitating efficient recycling of plastics.

Obviously, graphene materials derived from waste plastics can be effectively used in the field of environmental remediation and material modification. In addition, due to its excellent conductivity, graphene can be used as the anode of lithium-ion batteries, the substrate of photocatalysts, and has great potential for application in supercapacitors, flexible electrodes, and biosensors.[113−119] However, the applicability of graphene from plastic waste needs to be further explored.

6.2. Application of Porous Carbon Materials. Porous carbon materials are widely used for their high specific surface area and well-developed pore structure. These materials possess excellent adsorption properties, making them particularly suitable for water treatment and CO_2 capture. Carbon materials have been produced from various waste sources, and porous carbon materials derived from the carbonization of plastics can also be used to remove contaminants such as fluoride ions and polycyclic aromatic hydrocarbons (PAHs) from water and to capture CO_2.

Siddique and colleagues used iron-impregnated activated carbons (AC-CLPs) derived from waste lemon peels to remove fluoride ions from water (Figure 12a).[120] The adsorbents were prepared by activating selected biomass with $FeCl_3$, followed by carbonization at two different temperatures, 250 and 500 °C. The optimal adsorption conditions for both carbon materials, determined through experimental investigation, were pH = 6.6, an adsorbent dosage of 1.0 $g{\cdot}L^{-1}$, and an adsorption time of 240 min. According to the Langmuir isotherm, the adsorption of fluoride ions in wastewater was 4.926 and 9.709 $mg{\cdot}g^{-1}$ for AC-CLP250 and AC-CLP500. Pallarés and co-workers physically activated barley straw using CO_2 and steam to convert it into activated carbon (Figure 12b), which exhibited good microporosity and a surface area comparable to that of commercial activated carbon.[121] This surface area was comparable to that of commercial activated carbon, indicating its potential as a low-cost precursor for activated carbon production. Ilyas prepared AC by carbonizing waste PET and PS, which showed greater effectiveness in adsorbing PAHs from aqueous solutions. The optimal

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ACS Nano 2025, 19, 12734−12761

adsorption conditions, determined experimentally, included an initial PAHs concentration of 40 ppm, a contact time of 2 h, pH values of 3, 5, and 7, a temperature of 50 °C, and an adsorbent dosage of 0.8 g.[122] Compared to commercial adsorbents, this type of AC exhibits significant PAH adsorption capacity, offering an economical and environmentally friendly solution for industrial wastewater treatment. It suggests that this AC holds promise for PAH removal and recovery applications.

Hiremath and colleagues synthesized two-dimensional (2D) activated carbon nanosheets (CFs) from foam materials through simple carbonization and KOH activation for CO_2 adsorption. The resulting carbon exhibited a large specific surface area, excellent cycling stability, and superior energy storage properties, making it ideal for CO_2 capture and supercapacitors (Figure 13a).[123] Yuan and colleagues utilized discarded PET plastic bottles as carbon precursors to prepare microporous carbon via carbonization and chemical activation (Figure 13b). The samples demonstrated high CO_2 adsorption, good CO_2 selectivity, easy regeneration, outstanding cycling stability, and fast CO_2 adsorption−desorption kinetics. These characteristics are critical for practical CO_2 capture applications. DFT calculations confirmed that narrow micropores and functional groups play a key role in achieving high adsorption capacity and selectivity. This material can be applied to carbon capture and storage while addressing environmental issues caused by PET waste.[124] It can be seen that the porous carbon materials prepared from waste plastics also show excellent performance in water treatment and CO_2 capture, which opens up a new way of recycling waste plastics and helps to practice the concept of waste to waste.

6.3. Application of Carbon Nanotubes. Carbon nanotubes are nanomaterials with special structure and excellent performance, which have many advantages over other carbon nanomaterials: mechanical properties, high tensile strength (50−200 GPa, 100 times that of steel), low density (1/66 that of steel) and high flexibility; electrical properties, high conductivity (10,000 times that of copper), low energy loss, and unique electrical properties (metallic or semiconducting); thermal properties, high thermal conductivity (more than 3000 $W{\cdot}m^{-1}{\cdot}K^{-1}$); and high specific surface area, good chemical stability, and hydrogen storage properties. These excellent properties of carbon nanotubes absolutely determine its unique advantages in environmental remediation, electronic components, and materials applications.

The high specific surface area and unique electronic structure of carbon nanotubes (CNTs) highlight their potential in catalytic reactions and environmental purification. Numerous studies have explored the application of CNT composite membranes in water treatment, including desalination, oil−water separation, and the removal of heavy metal ions and emerging pollutants (Figure 14a).[125] Deokar and colleagues synthesized carbon nanotubes (CNTs) from waste PE bottles for the removal of the herbicide diuron in aqueous solution. The study results showed that the CNTs efficiently adsorbed diuron, with an adsorption capacity of approximately 40.37 $mg{\cdot}g^{-1}$ at 303 K, making them a low-cost and highly efficient adsorbent for water treatment.[126] Gong et al. prepared magnetic nickel/carbon (Ni/C) nanomaterials through the catalytic carbonization of LLDPE using a one-pot method. This process involved cocatalysis with Ni_2O_3 and PVC resin. They controlled the yield and morphology of the magnetic Ni/C nanomaterials by adjusting the amount of PVC resin in the LLDPE/5Ni_2O_3 mixture. The metallic nickel endows the Ni/C nanomaterials with magnetic properties, facilitating their reuse in dye adsorption applications (Figure 14b).[101]

CNTs also have a wide range of applications in electronic materials. Cai and co-workers proposed a method to prepare CNTs on a Fe−Al_2O_3 system using pyrolysis of plastic waste (Figure 14c).[127] Specifically, carbon nanotubes were first prepared by pyrolyzing plastic waste on Fe−Al_2O_3, followed by coheating the Fe-CNTs with melamine in an inert atmosphere. Higher doping temperatures help increase the extent of graphite formation and can induce the conversion of nitrogen into a more stable form. The electrocatalysts obtained by pyrolysis at 850 °C showed excellent performance compared to commercial Pt/C catalysts. The electrocatalysts obtained by pyrolysis at 850 °C showed excellent performance when Fe−N−CNTs was used. When zinc−air cells were assembled using Fe−N−CNT850 and Pt/C catalysts, respectively, the Fe−N−CNT850-based zinc−air cells operated stably for over 5000 s at a high open-circuit potential of 1.44 V, which was only 0.02 V lower than that of the Pt/C-based zinc−air cells (1.46 V). Additionally, the Fe−N−CNT850-based zinc−air battery has a higher peak power density of 120.44 $mW{\cdot}cm^{-2}$ compared to 98.53 $mW{\cdot}cm^{-2}$ for the Pt/C-based battery.

Abbas and colleagues investigated the synthesis of MWCNTs in a CCVD reactor, using noncondensable gases from the pyrolysis of flexible plastic packaging waste.[128] The MWCNTs were then treated with nitric acid (HNO_3), and two functionalized samples, labeled FMWCNT-3 and FMWCNT-6, were prepared based on differences in boiling time. The longest discharge time was observed for FMWCNT-3 at a current density of 1 $A{\cdot}g^{-1}$, indicating its highest specific capacitance. Under different current densities, the plastic waste-derived MWCNTs exhibited superior capacitance performance compared to commercial C-MWCNTs. Among these, the specific capacitance of FMWCNT-3 was significantly enhanced after functionalization, while the capacitance of FMWCNT-6 rapidly decreased with increasing current density. Although FMWCNT-3 has the smallest specific surface area, its oxygen and nitrogen functional groups enhance its electrochemical performance, suggesting that the influence of functional groups outweighs surface area as a factor. These results suggest that multiwalled carbon nanotubes synthesized from flexible plastic packaging waste could be a superior alternative to commercial multiwalled carbon nanotubes.

Cai and co-workers constructed carbon-based electromagnetic wave (EMW) absorbing materials using carbon nanotubes derived from plastic packaging waste in discarded electronic devices, primarily consisting of PE and PP.[129] During preparation, due to the inherent sensitivity of carbon nanotubes to processing parameters, they successfully created a three-dimensional network-structured carbon nanotube aerogel (CNTA) with radial nanotube arrays by modulating their microstructure (Figure 14d). CNTAs exhibit excellent performance in EMW absorption. In particular, CNTA-2, with a minimum reflection loss (RL) of −59.5 dB at a thickness of 2.7 mm, has an effective absorption bandwidth covering 8.48 GHz. In addition, CNTA-2 exhibits excellent thermal insulating properties, with a thermal conductivity of 33.5 $mW{\cdot}m^{-1}{\cdot}K^{-1}$, which is able to effectively CNTA-2 also exhibits excellent hydrophobicity, with a water contact angle of more than 150°, indicating that it can effectively resist water penetration. This property allows the CNTAs to excel in self-cleaning applications and maintain cleanliness in environments

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ACS Nano 2025, 19, 12734−12761

Figure 15. Challenges and future research directions for plastic carbonization.

with different pH values. These multifunctional properties make CNTAs very promising for applications in modern technology and in the protection of the environment.

CNTs converted from waste plastics have already shown great advantages in water treatment, electronic components and materials applications.[130−135] However, there is still much room for research in this field to ensure its effectiveness, stability and safety in practical applications, and to achieve further breakthroughs in various aspects such as resource recycling, environmental protection and the development of high-performance materials.

7. CHALLENGES AND PERSPECTIVES

This review focuses on the catalytic conversion of waste plastics into carbon nanomaterials and the applications of different carbon nanomaterials. Some key factors affecting the quality and yield of synthesized carbon nanomaterials are discussed. Finally, the applications of the three derived carbon nanomaterials, graphene, porous carbon and carbon nanotubes, in various fields are discussed. While various technologies exist for converting plastics into carbon nanomaterials, the efficiency and yield of synthesizing carbon from waste plastics have significant potential for improvement. Future research should focus on developing novel catalysts and optimizing reaction conditions to produce higher-quality carbon nanomaterials. These innovations could significantly enhance the conversion process and improve the performance of the final products. Additionally, we summarize the current challenges of this technology and offer insights into future research directions, as shown in Figure 15. These efforts aim to push waste plastic conversion technology to new heights to address the global challenge of plastic waste.

(1) **A deep understanding of the plastic carbonization process can enhance both the yield and quality of carbon nanomaterials.** Precise control of temperature, pressure, reaction time, and catalysts during the carbonization process optimizes the reaction rate and steers the process toward favorable conditions. Understanding the plastic carbonization process also allows for the adjustment of reaction conditions, enabling the formation of various carbon morphologies, such as porous and nanostructured materials. These morphologies directly influence the performance of carbon nanomaterials. Porous structures increase the specific surface area, making them ideal for applications like adsorption, while nanostructures enhance electrical and mechanical properties, improving efficiency in applications such as electronics and environmental remediation. Therefore, optimizing the plastic carbonization process increases yield and significantly improves the performance of carbon nanomaterials, offering more efficient solutions for practical applications.

(2) **Development of novel high-efficiency catalysts and green processes.** Novel catalysts can significantly lower the reaction temperature in plastic-to-carbon conversion, reducing energy consumption and improving economic feasibility. High-efficiency catalysts should focus on controlling the morphology and types of carbon nanomaterials to improve their industrial applicability. Several innovative catalysts have been tested, such as silica-supported zirconium monohydride,[136] bimetallic,[87] core−shell,[137] and transition metal salt catalysts.[64] These catalysts have shown strong performance in plastic-to-carbon conversion. However, reducing catalyst production costs remains a key challenge in achieving more sustainable and environmentally friendly production methods. The diversification of green technologies will be a crucial trend in future developments. In addition to microwave plasma and 3D printing, emerging techniques like electrochemical catalysis will play a key role in plastic-to-carbon conversion.[138−141] These technologies improve conversion efficiency while operating under milder conditions, further reducing energy consumption and emissions. Process integration will be essential for the development of green technologies. For example, catalyst design can be integrated with reactor optimization and exhaust gas (CO_2, CO, CH_4, C_xH_y, HCl, NO_x, etc.) recovery systems to create a unified green process. This integrated approach will offer sustainable solutions for industrial applications.

(3) **TEA and LCA of plastic to carbon conversion.** TEA of plastic to carbon conversion mainly includes the pyrolysis conditions of waste plastics (temperature or electricity) as well as the quality and yield of the product obtained. TEA of the plastic carbonization process allows the correlation between process parameters and production efficiency to be studied in depth. Thus, the optimal combination of process parameters can be clarified to achieve the goal of improving production efficiency and reducing production costs. In addition, the cost

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analysis and market demand analysis of plastic carbonization products can help formulate a reasonable pricing strategy to enhance the competitiveness of the products in the market. Therefore, optimizing the reaction conditions, minimizing the energy input and increasing the yield and quality of the carbon material obtained is essential to increase the net market return. LCA is an essential method for evaluating the environmental impacts, resource consumption, and economic benefits of converting plastics into carbon nanomaterials. This analysis covers the entire process, from production and use of plastics to their recycling and final conversion into carbon nanomaterials, helping assess the sustainability and circular economy potential of plastic waste management. For example, Ügdüler performed a LCA to evaluate pure monomer extraction through alkaline hydrolysis of PET trays and its associated carbon footprint. The study showed that this recycling method significantly reduces emissions compared to both producing the same amount of virgin monomer (with a carbon footprint of 3.9 kg of CO_2-eq) and PET incineration with energy recovery. In addition, the carbon material preparation process inevitably produces pollutant gases (VOCs), and a good post-treatment of exhaust gases is of great benefit to the environment. These findings highlight more environmentally friendly processing conditions and provide guidance for optimizing hydrolysis recycling.[30] Moreover, using milder reaction conditions can further reduce energy consumption and economic costs. Overall, LCA not only improves the environmental sustainability of the conversion process but also reduces economic costs, offering critical support for the sustainable management of plastic waste.

(4) **Scale-up experiments to industrial applications.** Although a lot of research has been done in the field of plastic waste treatment, most of the current degradation technologies are still limited to laboratory scale. There is a huge gap between laboratory and industrial production, as the laboratory environment is ideal and parameters can be precisely controlled, while the industrial production environment is complex and involves factors such as efficiency, cost, safety and stability. Therefore, to apply degradation technologies to industrial production, it is necessary to conduct large-scale experiments, gradually evaluate different technologies, and select technologies suitable for industrial production based on industrial production requirements. After selecting the appropriate technology, we can promote the industrial application of plastic carbonization technology, which is of far-reaching significance for the treatment of plastic waste, and can add value to plastic waste by converting it into high-value products, which will help to recycle plastic waste and solve the related environmental and resource problems. In conclusion, although plastic carbonation technology demonstrates significant potential in laboratory settings, its industrial-scale implementation encounters challenges such as reactor design, process optimization, and system stability. Scaling up production necessitates continuous research and refinement to ensure economic viability and consistent system performance. These efforts will facilitate the transition of plastic carbonation from laboratory research to industrial applications, enabling efficient and cost-effective utilization of plastic waste while advancing a green economy and sustainable development. We believe that this review will inspire more creativity in designing such win−win reaction systems to realize a "waste treat waste" concept.

ASSOCIATED CONTENT

Data Availability Statement

No primary research results, software, or code has been included, and no new data were generated or analyzed as part of this review.

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (22178258 and 22308254) and Talent Program Fund of Tianjin University (0701321039 and 0903074107). This work was also supported by a grant from the Key Laboratory of Recycling and Eco-Treatment of Waste Biomass of Zhejiang Province (No. 2024HZYB02).

VOCABULARY

carbonation = process by which plastics undergo complex chemical transformations under certain conditions and their organic components are gradually converted into carbonaceous materials

life cycle analysis (LCA) = methodology for assessing the environmental, resource, and health impacts of plastics from raw material acquisition, manufacturing, use, and disposal to inform plastics management and decision making

chemical upgrading = process of chemically breaking down waste plastics into monomers, oligomers, or other valuable

https://doi.org/10.1021/acsnano.5c03391
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chemicals and then using these breakdown products to resynthesize new plastics or other chemical products
plastics = polymer-based synthetic materials that can be formed, processed, and molded through polymerization reactions, have specific physical and chemical properties, and are widely used in industry and daily life
techno-economic analysis (TEA) = analytical method that provides a basis for industry decision making by comprehensively evaluating the technical and economic aspects of plastics from production to disposal, including environmental and political impacts

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