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Applications of Waste Polyethylene Terephthalate (PET) Based Nanostructured Materials: A Review

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Abstract

While polyethylene terephthalate (PET) has enjoyed widespread use, a large volume of plastic waste has also been produced as a result, which is detrimental to the environment. Traditional treatment of plastic waste, such as landfilling and incinerating waste, causes environmental pollution and poses risks to public health. Recycling PET waste into useful chemicals or upcycling the waste into high value-added materials can be remedies. This review first provides a brief introduction of the synthesis, structure, properties, and applications of virgin PET. Then the conversion process of waste PET into high value-added materials for different applications are introduced. The conversion mechanisms (including degradation, recycling and upcycling) are detailed. The advanced applications of these upgraded materials in energy storage devices (supercapacitors, lithium-ion batteries, and microbial fuel cells), and for water treatment (to remove dyes, heavy metals, and antibiotics), environmental remediation (for air filtration, CO_2 adsorption, and oil removal) and catalysis (to produce H_2, photoreduce CO_2, and remove toxic chemicals) are discussed at length. In general, this review details the exploration of advanced technologies for the transformation of waste PET into nanostructured materials for various applications, and provides insights into the role of high value-added waste products in sustainability and economic development.

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Keywords

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1. Introduction

Plastic is an indispensable part of everyday life and is used in almost every field. However, most conventional plastics are prepared from petroleum-based chemicals and non-biodegradable, which leads to serious environmental pollution after their disposal. It is estimated that the amount of plastic waste generated worldwide was around 353 million metric tons in 2019, and most were directly discharged into the natural environment. If current policies continue, the amount of plastic waste is anticipated to triple by 2060. (Geyer et al., 2017) Landfilling and incinerating are the two most conventional techniques for treatment of plastic waste. (Zhang, F. et al., 2021) However, landfilling these nonbiodegradable plastics takes up increasingly more land space due to their extremely slow rate of degradation, which may result in soil and water pollution and even cause land deterioration. Plastic incineration requires a large amount of energy and releases harmful or toxic substances into the air, such as carbon dioxide (CO_2), sulfur oxides, and various hazardous particulate substances. (Damayanti et al., 2022) In recent decades, degradation, recycling and upcycling have become the mainstream solutions for plastic waste treatment (Abdelbasir, Sabah M. et al., 2020; Choi et al., 2022; Sadeghi et al., 2021; Singh et al., 2021). Degradation comprises biodegradation, photodegradation, thermodegradation, mechanochemical degradation and other degradation methods, where the plastic waste is converted into water and CO_2 and eventually released into the environment (Thachnatharen et al., 2021; Zhang, F. et al., 2021). Recycling mainly includes physical, resource, and energy recycling. Physical recycling usually refers to sorting and separating, crushing, cleaning, and regranulation of waste. Resource recycling involves chemolysis and thermolysis including gasification, pyrolysis, and hydrogenolysis. Energy recycling mainly refers to incineration. (Wu et al., 2022) Upcycling is to transform waste plastics into carbonaceous materials with diverse nanotextures and morphologies, such as carbon nanotubes (CNTs), carbon dots, graphene, 3D carbon architectures, porous carbon, and carbon-based composites, which can be achieved via catalytic, anoxic pyrolysis, and hydrothermal carbonization, and microwave and flash Joule heating. (Chen et al., 2022)

Polyethylene terephthalate (PET) is one of the most widely used polymers globally because it is high in strength, lightweight, and has good thermal stability and a low production cost. The mass production of PET products however generates a massive amount of waste, which causes serious environmental problems if the waste is discharged into the environment. In order to prevent such environmental problems and strive for environmental sustainability, physical recycling has attracted universal attention owing to its simple operations and low cost. (Soong et al., 2022) To ensure

the performance of reprocessed products, physical recycling requires high quality PET waste, such as waste that is a single-type of plastic, uncontaminated and clean. Some waste plastics need to be melted and re-granulated, which may result in deterioration of the properties of the reprocessed products due to the reduction in molecular weight resulting from chain-scission. (Awaja and Pavel, 2005) Resource recycling is another popular strategy to recycle waste PET, in which the PET macromolecular chains are broken down to acquire useful chemicals. The obtained chemicals can be further used to synthesize with high-value materials for various applications. For example, terephthalate (BDC) is one of the most useful chemicals made from depolymerized PET, which is widely used to produce metal-organic frameworks (MOFs) with a crystalline porous structure. (Shanmugam et al., 2022; Ubaidullah et al., 2021) MIL-53 (Cr) and MIL-101 (Cr) are two types of PET-based MOFs with a large surface area and have been applied for the adsorption of CO_2, N_2, and H_2 gases (Lo et al., 2016). MOF-5 can be embedded into mesoporous carbon to develop structures with a large surface area and excellent active sites. These structures not only can be applied as electrodes but also as next-generation energy storage devices. (Ubaidullah et al., 2021) Besides, waste PET is rich in carbon, which can be utilized for the synthesis of high value-added carbon materials with a large specific surface area, high degree of porosity, high conductivity, and excellent surface functions (due to the presence of functional or group dopants). (Chen et al., 2022) These carbonaceous materials can be widely applied in sustainable environmental strategies including for pollutant adsorption (Chan and Zinchenko, 2022; Ubaidullah et al., 2020) and CO_2 capture, and for green energy purposes, such as supercapacitors and water splitting technologies (Al-Enizi et al., 2020a; Al-Enizi et al., 2020b).

Currently, a number of review articles have summarized the transformation techniques of waste plastics into valuable carbonaceous materials (Chen et al., 2022; Choi et al., 2022; Jiang et al., 2023) and MOFs (El-Sayed and Yuan, 2020; Lagae-Capelle et al., 2020; Shanmugam et al., 2022). However, descriptions of the different applications of upgraded materials are still insufficient. The purpose of this review is to provide a comprehensive summary of the recent technologies that transform waste PET into high-value materials and their advanced applications in various fields. Specifically, the recycling and upcycling techniques for waste PET are introduced. Following this, the fabrication process of various waste PET-based MOFs and carbonaceous materials and their advanced applications for energy storage, water treatment, environmental remediation and as catalysts are summarized in detail. Based on the current findings, insights into the transformation technologies of waste-to-value nanostructured materials and their potential applications are provided.

2. PET

PET is one of the most commercially important thermoplastic polymers in the past few

decades. Superior properties such as high strength and modulus and high heat deformation temperature make PET wide spreads in almost all fields. This section gives a brief introduction of the production, structure, properties, and applications of virgin PET. The most advanced management strategies of waste PET were also introduced.

2.1 Virgin PET

PET is produced from ethylene glycol (EG) with either terephthalic acid (TPA) or dimethyl terephthalate (DMT). (De Vos et al., 2021) The production process of PET generally consists of three steps: (1) polymerization of bis(hydroxyethyl) terephthalate (BHET), (2) pre-polymerization, and (3) polycondensation. (Awaja and Pavel, 2005) In the first step, the polymerization of BHET involves two types of reactions. One type is an esterification reaction where EG is reacted with TPA and another type is a transesterification reaction where EG reacts with DMT. The transesterification method is more preferred in commercial production of PET because of the improved catalytic action and easier purification process. The second step is related to the pre-polymerization process, where BHET and some oligomers are polymerized to a degree of polymerization (DP) of up to 30. The DP of the product is further improved to 100 in the third step. Solid state polymerization (SSP) is required to produce PET products with a higher molecular weight and a DP of up to 150. (Awaja and Pavel, 2005; Mandal and Dey, 2019) Finally, based on the final product requirements and applications, PET pellets can be processed through various techniques, such as fiber spinning, extrusion, foaming, and blow and injection molding. (Ji, 2013)

The chemical structure of PET consists of numerous monomer units that contain BDC and ethylene. The aromatic ring in the repeating units imparts PET with high strength and modulus. (Mandal and Dey, 2019) PET can be colorless and semi-crystalline or transparent amorphous, which is determined by the crystalline state of the PET. The crystalline state is influenced by the cooling rate of the PET. A slow cooling process can produce a semi-crystalline polymer, while rapid cooling leads to an amorphous nature. Besides, additives and heat treatment of the polymer melt can also influence the crystallinity of PET. (Sinha et al., 2010) As a result, amorphous PET has a transparent appearance and high impact resistance, while semi-crystalline PET has a cloudy appearance but higher strength and can endure more severe outdoor environments (Mandal and Dey, 2019). PET has a glass transition temperature (Tg) between 65 °C and 140 °C, and melting temperature (Tm) around 260 °C. (Sinha et al., 2010) PET has superior thermal properties; excellent resistance to oils, aliphatic hydrocarbons, alcohols, and diluted acids; and moderate resistance to aromatic hydrocarbons, diluted alkalis, and halogenated hydrocarbons. (Ben Zair et al., 2021) In addition, PET has good electrical properties and excellent moisture and gas (oxygen, CO_2) barrier properties.

PET products are widely used in various fields as they have superior physical, mechanical, and chemical properties. One of the most important applications is

packaging. Owing to its excellent mechanical performance and gas barrier properties, PET is usually used to produce bottles for water and carbonated drinks, food packaging films, food trays, plastic bags and cosmetic jars. (Nisticò, 2020) PET is also widely used in the textile industry in different forms. Uniaxial stretching with applied tension yields high-strength fibers, which can be used to produce strong, light, and tear-resistant products, such as staple fibers, filaments, and technical yarns, while the fabric is used extensively in apparel and household products, from pants and shirts to curtains and bedding. Polyester fabrics are particularly popular for children's wear due to its toughness, and garments owing to its anti-wrinkle and anti-shrinkage abilities. (Wang et al., 2019) PET is also used in the electrical and electronics industry and usually used to manufacture devices that are subjected to electrical encapsulation, liquid crystal displays, solar junction boxes, and photovoltaic parts. Other applications of PET include automotive parts, disposable vehicle interiors, surgical supplies, and insulation tape. (Awaja and Pavel, 2005)

2.2 Recycling and upcycling of waste PET

The extensive use of PET products has caused difficulties in their disposal and management. A large number of strategies have been proposed to manage these non-biodegradable plastic wastes. (Zhou et al., 2022) To ensure that they are disposed in an environmentally friendly manner, the most promising way is to recycle these wastes and transform them into high value materials. There are three strategies to recycle PET, including physical and chemical recycling and energy recovery. (Awaja and Pavel, 2005; Lu et al., 2022) Physical recycling can be further classified into primary and mechanical recycling. The former mainly targets single-type of plastic, and uncontaminated pre-consumer waste, which can be used as a second-grade material to ensure product quality. (Ravi and Vadukumpully, 2016) The latter usually include processes of sorting and separating, crushing, cleaning and regranulation of plastics. The drawback of this method is the deterioration of product performance due to the degradation of the polymer. Chemical recycling, or resource recycling, involves the depolymerization of the PET molecular chains. (Nikles and Farahat, 2005) Useful chemicals are obtained in the forms of monomers, oligomers, petroleum liquids and gases through various depolymerization methods, such as hydrolysis, glycolysis, methanolysis, and aminolysis. (Al-Sabagh et al., 2016) When the physical and chemical recycling methods are challenging, or, economically or environmentally not viable, energy recycling can be applied to recover thermal energy. Waste heat can be used for generating electricity, and residual heat from waste steam can be utilized to heat industrial or residential buildings. (Sinha et al., 2010)

The resource-recycled chemicals could be further employed to synthesize useful materials. For example, BDC is one of the most useful chemicals made from depolymerized PET, which is widely used to produce MOFs with a crystalline porous structure. MOFs have been the subject of research attention due to their wide range of applications in addressing environmental and energy problems, such as H_2 production,

CO_2 adsorption, water treatment, catalysis, sensing and energy storage. (Gangaraju et al., 2023) The structure of MOFs consists of metal ions/clusters and organic linkers, both of which can be recycled from various wastes. (Masoomi et al., 2019) MOFs are rationally designed frameworks with adjustable porosity, tunable pore sizes, and tailored structures for different applications. In particular, organic linkers play a vital role in controlling reactions and changing the pore size in MOFs. The synthesis of MOFs from waste PET can be realized by using one-pot synthesis or a two-step approach. The former consists of two stages, i.e., hydrolysis of PET and on-site synthesis of MOFs. The latter involves the degradation of PET into BDC and the purification of BDC first, and then adding the BDC into metal salts to develop MOFs. (El-Sayed and Yuan, 2020)

Although recycling is an environmentally friendly and sustainable approach to treating waste PET, the conventional recycling strategy still has some limitations. (Abdelbasir, Sabah M et al., 2020) For example, separating and cleaning waste can be laborious, and the recycled products have compromised strength, flexibility and durability. The oligomer or liquid oil produced from resource recycling has limited applications. In this case, upcycling waste PET into carbonaceous functional materials is considered as a novel technology to manage waste plastics. (Jiang et al., 2023) Waste plastic can be transformed into a wide range of carbon materials, such as CNTs, carbon dots, porous carbon and graphene. (Gong et al., 2019; Mukherjee et al., 2019) The steps for transforming PET into carbon-based materials include a pre-treatment, carbonization, and a post-treatment. Carbonization is the most crucial step in transforming waste plastics into carbonaceous materials, which can be achieved through various ways, including catalytic, hydrothermal and anoxic pyrolysis carbonization, and microwave and flash Joule heating. (Shanmugam et al., 2022) Carbon-based nanostructured materials derived from waste PET have a relatively wide range of applications, such as for energy storage devices, as adsorbents for water treatment or harmful or toxic gases. (Chen et al., 2022; Mukherjee et al., 2019)

2.3 Impacts, feasibility and regulations

The environmental impacts of all proposed recycling and upcycling strategies are supposed to be minimized, which could be evaluated by means of life cycle assessment (LCA) or circular economy indicators. The LCA is a methodology to evaluate the environmental impacts through the consumptions of materials and energy, disposal of wastes and emissions in the environment. (Korley et al., 2021) One of the cases is the carbon footprint calculation of the re-polymerization of recycled PET into fiberglass/plastic composites. (Jehanno et al., 2022) According to the performed analysis, the upcycling processes is potential to save CO_2 by avoiding incineration and production of virgin polymers, with each kg of waste PET avoiding 3.92 kg of CO_2-equivalent emission. Besides, the upcycled polymer could be recycled again, which further increase the benefit to the environment. The carbon footprint calculation was conducted on three different recycling or upcycling scenarios, and the results

demonstrate that polymer recycling and upcycling have feasible environmental benefit, which can reduce the emissions of CO_2, the major contributor to greenhouse effect. Other studies have also proved that the recycling strategy can achieve better environmental benefit compared to landfill or incineration. (AlMa'adeed et al., 2012; Wäger and Hischier, 2015)

High cost is one of the most common reasons that impede the industrialization process of new technologies. As for the synthesis of MOFs, the expensive precursors have limited the economic feasibility to achieve a sustainable and scalable production. (El-Sayed and Yuan, 2020) However, several published studies have proved that the cost could be reduced and meet the industrialization requirements. For example, the price of 1 kg of MIL-53(Al) fabricated from recycled aluminum foil and PET bottles was only one-third of that of Basolite A100 provided by Sigma-Aldrich. (Panda et al., 2020) On one hand, the optimization of reaction conditions could achieve a PET conversion rate of almost 92-100%, thus realizing a high BDC yield and high MOF yield from useless inexpensive waste. (El-Sayed and Yuan, 2020) On the other hand, the recycling of plastic waste require energy. Bataineh has investigated the LCA of recycling waste PET and revealed that the total required energy to recycle PET sheet are only 14–17% of the production of virgin PET sheet. (Bataineh, 2020) Besides, solar electricity was also used in some regions to reduce the cost of recycling waste PET bottles. (Choudhary et al., 2019) All efforts have ensured the economic feasibility of the recycling and upcycling process of waste PET, offering an integrated pattern to realize economic benefit and achieve a sustainable environment.

Plethora of nanomaterials have been generated and used to advance technologies in numerous industrial applications in the past few decades. The increasing utilization of nanomaterials has raised concerns about potential risk, and require adequate risk assessments methods and regulations. (Abdelbasir, Sabah M et al., 2020; Scheringer, 2008) Different countries and regions have formulated corresponding laws and policies on the production, use and disposal of nanomaterials. (Miernicki et al., 2019) For example, in the European Union (EU), different legal acts contain specific provisions for nanomaterials regulations, such as for food, cosmetics, devices, and biocides. (Jehanno et al., 2022) With the advancement of new nanomaterials technology, it is necessary to update or formulate new laws and regulations to supervise the production of nanomaterials.

3. Applications

Recent studies have explored the transformation technologies of waste PET into high value-added materials. The reasonable utilization of these nanostructured materials can help achieve both environmental sustainability and economic benefits. In this section, the applications of PET-based nanostructured materials in energy storage devices, water treatment, environmental remediation and catalysis, as well as their conversion process are discussed.

3.1 Energy devices

Energy devices is one of the most important applications of PET-based nanostructured materials. (Fan et al., 2023) Energy storage devices function mainly based on the transport of electrons and the interaction of ions within the material (Nayak et al., 2022), which requires the materials to have high porosity, a large surface area, excellent electrical conductivity, and electrochemical stability. (Jiang et al., 2023) Waste PET derived nanostructured materials can be utilized to fabricate various energy storage devices owing to their diverse morphologies, nanostructure, and physicochemical properties. (Al-Enizi et al., 2020a; Dubey et al., 2023; Liu et al., 2020; Pandey et al., 2021; Shah et al., 2022; Ubaidullah et al., 2021) One example is the fabrication of waste PET-based hierarchical porous carbons, which are used as supercapacitor electrodes. (Zhang, H. et al., 2021) Potassium hydroxide (KOH)-activated carbon (AC-K) yields a high specific surface area of 2683 m^2/g and demonstrates an excellent specific capacitance of 325 F/g at a current density of 0.5 A/g. AC-K electrodes could retain about 92% of the initial specific capacitance after 5000 charge and discharge cycles. Another example of waste PET-based supercapacitor electrodes is the nickel oxide (NiOx)@ nitrogenous porous carbon (NPC) nanocomposite (NiOx nanoparticle modified nitrogen (N_2)-doped porous carbon). (Al-Enizi et al., 2020b) The NiOx@NPC nanocomposite is prepared from the carbonization of an Ni-MOF that is synthesized from BDC through the one-pot method. The modification of NPC can enhance electrical conductivity, electrical stability, and charge relocation operation, thus enhancing the performance of the supercapacitor. With a high specific surface area of 1523 m^2/g, the NiOx@NPC composite electrode has a superior capacitance of 581 F/g in a three-electrode setup, and excellent capacitance of 291 F/g in a two-electrode system. In addition, the NiOx@NPC composite shows cyclic stability after 5000 segments of cyclic voltammetry (CV) at 50 mV/s. Alhokbany et al. (Alhokbany et al., 2020) reported a bimetallic $NiCo_2O_4$@ N_2-doped carbon nanocomposite that was used in a high-performance supercapacitor. Waste PET were first depolymerized to produce BDC, then the BDC was further prepared to create bimetallic Ni/cobalt (Co) metal organic frameworks (Ni/Co-MOFs). The $NiCo_2O_4$@ N_2-doped carbon nanocomposite was synthesized from the carbonization of Ni/Co-MOFs and finally used to fabricate the supercapacitor. (Alhokbany et al., 2020) It is reported that the $NiCo_2O_4$@ N_2-doped carbon nanocomposite yields a high mesoporosity and large surface area of about 15 nm and 813 m^2/g, respectively. The nanocomposite also exhibits a superior electrochemical performance with excellent specific capacitance of about 913 F/g at 1 A/g, and about 890 F/g at 5 mV/s in a 6 m KOH electrolyte. Besides, the $NiCo_2O_4$@ N_2-doped carbon nanocomposite has good cyclic stability with 99% capacity retention after 5 cycles.

Waste PET-based nanostructured materials are also used in lithium-ion batteries (LIBs) (Kim et al., 2020; Półrolniczak et al., 2020) The heat treatment of waste PET with tin(II) chloride ($SnCl_2$) in a molten KCl-LiCl eutectic system yields SnO_2-embeded

terephthalic acid (SnO_2@terephthalic acid), which can be used as an anode for LIBs. The SnO_2@terephthalic acid sample prepared at 500 ℃ (PDN-500) shows a higher electrical conductivity of 447.3 S/m, a lithium-ion storage performance of 498 mAh/g after 500 cycles, and a Li-ion diffusion coefficient of 8.51 $\times 10^{-10}$ cm^2/s after 300 cycles. Li-ion storage contributes to over 42% of the cycling performance and capacitive storage (Kamali and Li, 2023). Dense carbon has also been shown to be a promising anode material for LIBs. Ehi-Eromosele et al. (Ehi-Eromosele et al., 2022) reported the use of waste PET derived activated carbon for the anode of LIBs through ionothermal synthesis. In particular, the use of choline chloride deep eutectic salts (CU-DES) can be used to synthesize activated carbon materials with high levels of ordering and a high degree of N_2 doping. The prepared activated carbon materials have a good electrochemical performance with an initial gravimetric capacity of about 460 mAh/g and a Coulombic efficiency of about 53%. The materials also demonstrated good cycling stability with a capacity retention of 98% after the 100th cycle. The $Co_{0.8}$Ni-MOF derived from waste PET bottles is considered to be a promising anode material for LIBs, as shown in **Figure 1**. (Wang et al., 2022) This $Co_{0.8}$Ni-MOF shows initial discharge and charge capacities of 2496 and 1729 mAh/g, respectively. Even after 200 cycles, the $Co_{0.8}$Ni-MOF electrode can still retain a high Coulombic efficiency of over 99%.

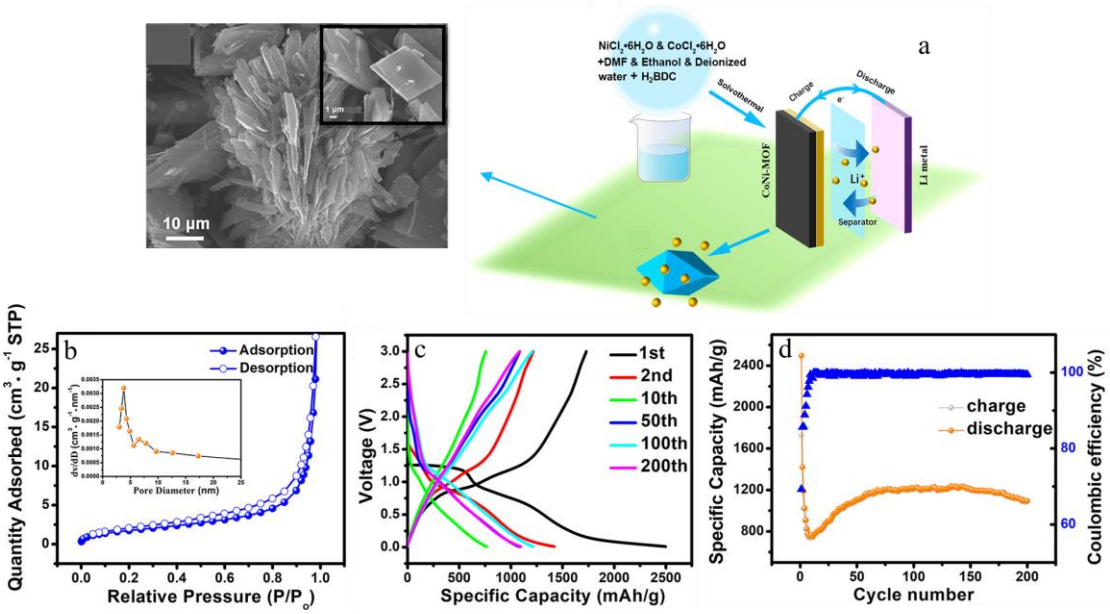

Figure 1. (a) Preparation process of $Co_{0.8}$Ni-MOF, (b) N_2 adsorption−desorption isotherm of $Co_{0.8}$Ni-MOF (inset shows plotted Barrett-Joyner-Halenda (BJH) pore size distribution), (c) Galvanostatic charge−discharge profiles with current density of 500 mA/g, and (d) cycling performance and Coulombic efficiency with current density of 500 mA/g. (Wang et al., 2022)

Microbial fuel cells (MFCs) have received attention as a sustainable technology for electricity generation from wastewater. However, their high ohmic resistance and low power output, and the high cost of electrode materials and membranes that are used in MFCs have limited their practical use and application. A number of strategies have been

proposed to build electrodes that would produce MFCs with higher output power density and electrical energy. (He et al., 2007; Jang et al., 2004; Song et al., 2009) For instance, terephthalic acid monomer (t) derived from the depolymerization of waste PET has been used as an organic linker to form Fe-t-MOFs. (Kaur et al., 2021) After the modification of polyaniline (PANI), the resultant Fe-t-MOF/PANI nanocomposite is coated on a conductive disk and functions as an electrode for MFCs. The resultant MFC has a high-power density of 680 mW/m^2, open circuit potential value of 0.67 V and a satisfactory limiting current density of 3500 mA/m^2, as shown in **Figure 2**. The prepared Fe-t-MOF/PANI electrode is biocompatible, stable, and low in cost, so it is a promising material that can be used to replace more costly carbon-based electrodes in fuel cells.

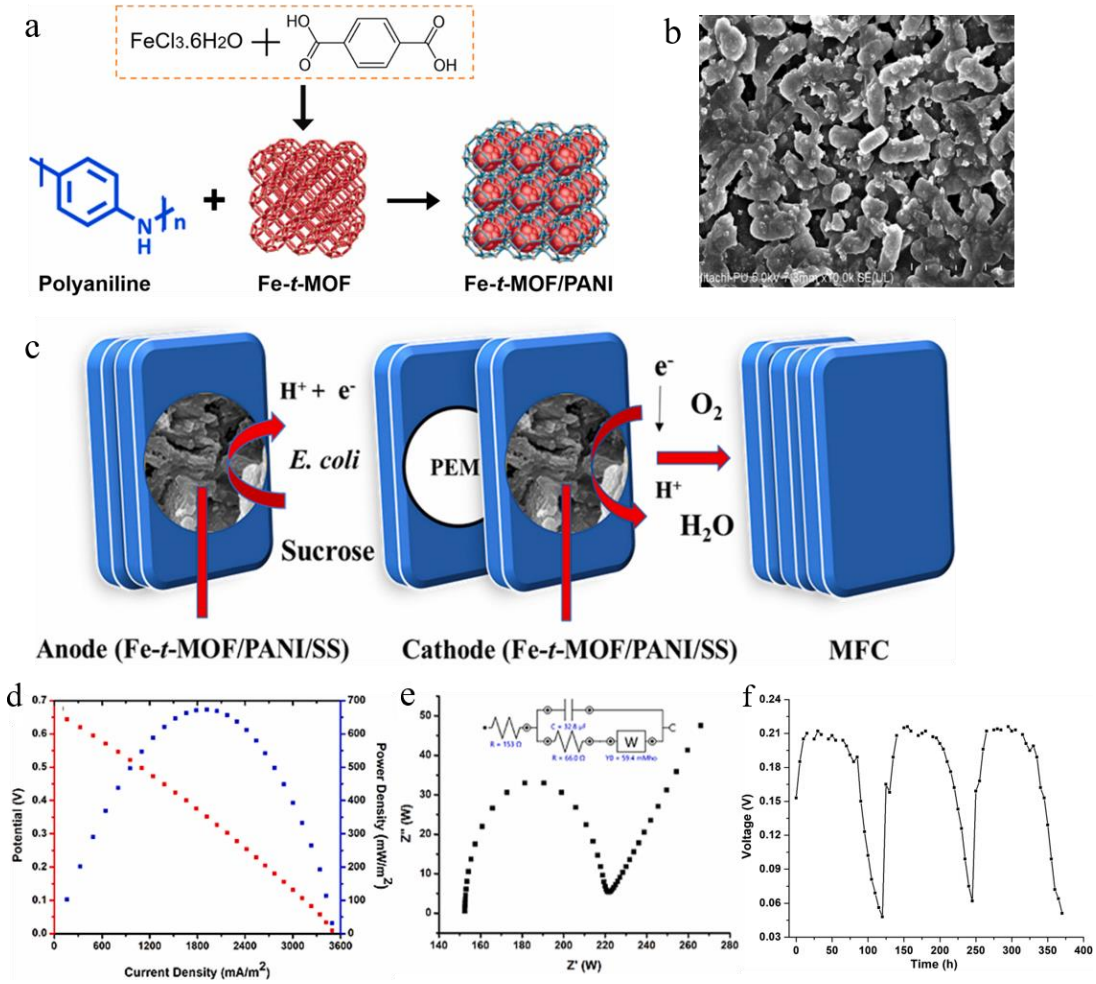

Figure 2. (a) Structure and synthesis process of Fe-t-MOF/PANI nanocomposite, (b) biofilm growth on Fe-t-MOF/PANI nanocomposite electrode, (c) schematic diagram of the working principle of an MFC, (d) open-circuit potential, (e) plotted power density, and (f) voltage-time analysis of stability of prepared electrode using 1000 ohm. (Kaur et al., 2021)

3.2 Water treatment

Water pollution is detrimental to ecosystems and contamination of water is a threat to

public health. Industrial waste discharge and human activities are the main causes of water pollution. In order to reduce the impact of water pollution on sustainability, the treatment of waste water is paramount. Wastewater contains suspended and dissolved matter from agricultural, industrial, commercial or domestic sources including dyes, plant nutrients, heavy metals, pesticides, antibiotics, and pathogenic microorganisms. (Kurniawan et al., 2012) Commonly used wastewater treatment strategies include chemical, physical, physico-chemical, and biological methods. Chemical procedures used to remove contaminants safely include the use of ozone, ultraviolet (UV) light and chlorine to kill viruses, bacteria and other pathogens, while pre-chlorination is used to control biological growth. Physical strategies include sedimentation, filtration and degasification. (Hassani et al., 2022) Sedimentation removes the suspended solids in floc (aggregated solid matter), filtration removes solid particles while degasification removes dissolved gases. The physico-chemical methods include membrane filtration, ion exchange, electrochemical treatment, and adsorption with activated carbon . Finally, biological methods eliminate suspended and dissolved organic chemicals through biodegradation. (Liquete et al., 2016)

Dyes in water pollutants can be toxic, carcinogenic and mutagenic. The most common method to remove dyes from waste water is adsorption. Activated carbon is a commonly used adsorbent to absorb impurities. Debina et al. (Debina et al., 2020) reported the synthesis of activated carbons from waste PET and the pseudo-stem of bananas through pyrolysis. The prepared adsorbent has a large specific surface area of about 424 m^2/g and shows a maximum retention of 94.7% with a pH value of 4. Pyrolysis can also transform waste PET bottles into graphene, which is an excellent adsorption material. Therefore, pyrolysis is a feasible technique to produce materials that adsorb dyes, such as acid blue 25 (AB 25) and methylene blue (MB). (El Essawy et al., 2017) Activated graphene (AG) and nano-ferromagnetic activated graphene （NFMAG) can be further developed though the modification of synthesized graphene (SG). Compared to SG, AG and NFMAG show higher maximum monolayer adsorption capacity and better adsorption capacity. (Mensah et al., 2022)

Magnetic materials that contain Fe, Ni, Co and their compounds are important functional materials that can be used for water treatment. A PET-derived magnetic adsorbent has been synthesized by the co-precipitation of PET oligomer (terephthalamide) through ammonolysis with Fe_3O_4 nanoparticles. (Chan and Zinchenko, 2022) Chan and Zinchenko (Chan and Zinchenko, 2022) used a magnetic adsorbent for the effective and rapid removal of an anionic dye called Congo Red (CR) which shows excellent adsorption capacity of CR of around 780 mg/g. Wei and Kamali (Wei and Kamali, 2021) constructed a magnetic nanoparticle embedded mesoporous carbon (Co_3Fe_7/$CoFe_2O_4$@carbon) composite to adsorb organic dyes from aqueous solutions. The Fe_3Co_7 nanoparticles were synthesized through the ball-milling of CoO and Fe_2O_3 in the presence of hexane, while $CoFe_2O_4$ was generated when the ball-milled Fe_2O_3/CoO was heated in molten salt. Waste PET was heated together with NaCl

and Fe_2O_3/CoO at 900 ℃ for 60 min to create a mesoporous carbon nanostructure (MCN) decorated with Co_3Fe_7/$CoFe_2O_4$ nanoparticles, see **Figure 3**. The mesoporous carbon nanostructured absorbent shows an exceptional monolayer adsorption capacity for MB and methyl orange (MO) dyes of 278 mg/g and 238 mg/g, respectively. With a high saturation magnetization (Ms) value of 17.9 emu/g, the magnetic adsorbent can be easily isolated from its mixture with water by using an external magnetic field. In addition, the recycled product still retains a high adsorption capacity of more than 95% of its original capacity after six regeneration cycles. These features make Co_3Fe_7/$CoFe_2O_4$@carbon a stable, effective and affordable adsorbent for practical uses.

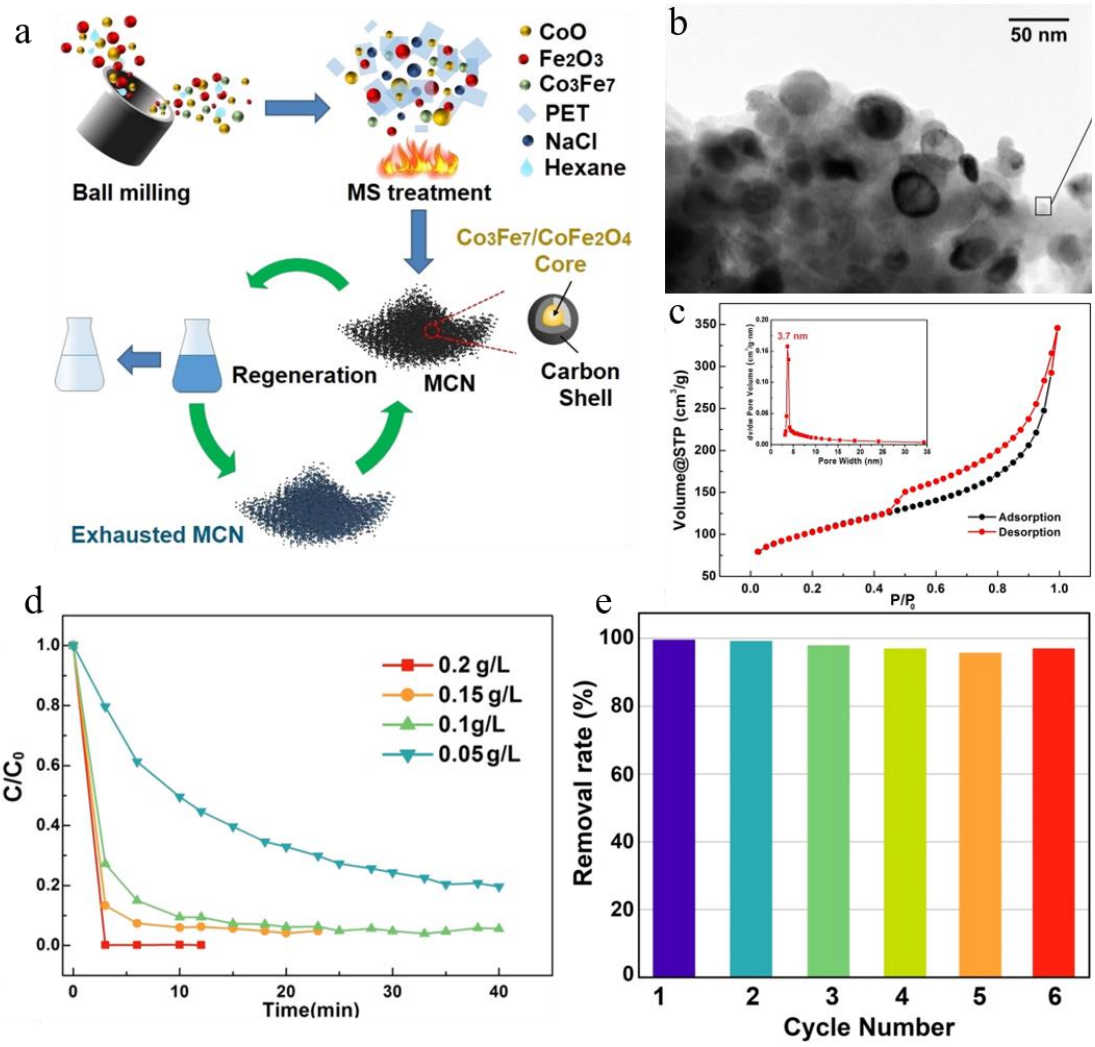

Figure 3. (a) Schematic illustration of method to fabricate MCN, (b) transmission electron microscope image of MCN morphology with presence of nanoparticles less than 40 nm in diameter embedded in carbon substrate, (c) N_2 adsorption-desorption isotherms and pore size distribution (inset), (d) plotted efficiency of MCN in removing MB with different dosages of carbon at dye concentration of 100 mg/L, and (e) plotted efficiency of thermal process used for regeneration of exhausted MCN, expressed as dye removal rate (%) vs number of adsorption cycles. (Wei and Kamali, 2021)

Heavy metals, such as titanium (Ti), chromium (Cr), manganese (Mn), arsenic (As), and lead (Pb), are well-known toxic pollutants in water. Exposure to these heavy metals for a long period of time and their accumulation in the body will cause severe damage to human health. They also damage the aquatic biota. Heavy metal ions can be removed from aqueous solutions by techniques such as physical adsorption, electrostatic interactions, complexation, and precipitation (Inyang et al., 2016). Wang et al. (Wang et al., 2012) reported the use of a waste PET-based chelating fiber as an adsorbent to remove Ni^{2+} and Cu^{2+} from waste water. The chelating fiber greatly absorbs Ni^{2+} and Cu^{2+} in an alkaline medium, and its adsorption capacity could still be maintained at 93% after five cycles. Roy et al. (Roy et al., 2021) fabricated a flexible and structurally durable 3D nanofiber-based aerogel by using recycled waste PET bottles as the raw materials for removing various heavy metals from polluted water. This aerogel has an efficient adsorption capacity between 94.5 and 98.3% for Zn(II), Hg(II), and Pb(II) under optimum experimental conditions. Kalimuthu et al. (Kalimuthu et al., 2022) synthesized various waste PET-based MOFs including Zr-MOF, Fe-MOF, and La-MOF to evaluate their removal capacity for arsenate. The adsorption process can be explained by surface complexation, ligand exchange and electrostatic interactions mechanisms. Kalimuthu et al. (Kalimuthu et al., 2022) reported that Zr-MOF, Fe-MOF, and La-MOF show a high maximum adsorption capacity of 85.72, 70.02, and 114.28 mg/g at a pH of 7, respectively, and more than 90% of the adsorption capacity can still be retained after four cycles. Ghosh and Das (Ghosh and Das, 2021) fabricated Sn(II)-MOF by utilizing waste PET derived BDC as the organic linker**.** The adsorbent showed substantial capacity for PO_4^{3-} and AsO_4^{3-} adsorption with values of 126.58 mg/g and 90.90 mg/g, respectively. Besides, the fabricated Sn(II)-MOF showed superior anti-interference ability when co-existing anions are present, which demonstrate the impressive effectiveness of removing about 99% of arsenate ions (AsO_4^{3-}) from aqueous samples obtained from lake and tap water. In addition, Sn(II)-MOF can be further transformed to SnO_2 nanoparticles, which shows an excellent adsorption capacity of 52.63 mg/g for manganese(2+) (Mn^{2+}).

Antibiotics are widely used as drugs for treating infectious diseases. However, they have a negative impact on the ecosystem when their concentration exceeds the normal concentration in the ecological environment. (De Smit et al., 2017) For example, antibiotics in natural water environments have toxic effects on certain organisms, thereby affecting their normal growth or reproduction. (Jery et al., 2023) In some cases, the overuse of antibiotics can induce the development of drug-resistant bacteria or resistant genes. Common methods for the removal of antibiotics from water include photolysis (Ge et al., 2019), electrochemical degradation (Wang and Zhuan, 2020), biodegradation (Zhi et al., 2019), UV radiation (Rizzo et al., 2013), adsorption (Li et al., 2018), and membrane filtration (Sharma et al., 2017). Waste PET can be used to fabricate magnetic carbon/iron composites to adsorb antibiotics from water. (Wang et al., 2023) Rai and Singh (Rai and Singh, 2018) reported that waste PET bottles can be converted into activated carbon (PETAC) through pyrolysis, then the ferromagnetic

iron oxides are intercalated onto the PETAC matrix to fabricate magnetic PETAC (M-PETAC) to adsorb cephalexin from water. The M-PETAC has a strong saturation magnetization of 35.4 emu/g and an adsorption capacity of 71.42 mg /g for cephalexin. The magnetic PET carbon composite has a desorption efficiency of 42.11%, which points to the recycling potential of the material. Jung et al. (Jung et al., 2020) fabricated a Fe-MOF derived magnetic porous carbon (α-Fe/Fe_3C) composite, which was used as an adsorbent for tetracycline hydrochloride (TCH). The prepared α-Fe/Fe_3C composite has a mesoporous texture with a surface area of 194.11 m^2/g and a pore volume of 0.285 cm^3/g, and adsorption capacity of 158.9 mg/g at pH values of 6. Compared with the initial adsorption capacity, the adsorption capacity of α-Fe/Fe_3C composites for TCH is reduced by less than 10%. Elessawy et al. (Elessawy et al., 2020) reported a novel technique of the synthesis of functionalized magnetic fullerene nanocomposites (FMFNs) for the removal of ciprofloxacin antibiotic. The FMFN was synthesized via a one-step catalytic thermal decomposition of waste PET, see **Figure 4**. The prepared FMFN has a large surface area of 336.84 m^2/g with micropores and mesopores volumes. The superparamagnetic property of FMFN can prevent the aggregation of adsorbent, thus enhancing the adsorption process. As shown in Figure 4(d), the left tube that contains E. coli culture shows + ve bacterial growth, and the middle tube that contains FMFN adsorbed with Ciprofloxacin showed no growth of bacteria. The right-side tube is an E. coli culture and filtrate water, which show no bacterial growth, which means that the FMFN has adsorbed all of the Ciprofloxacin.

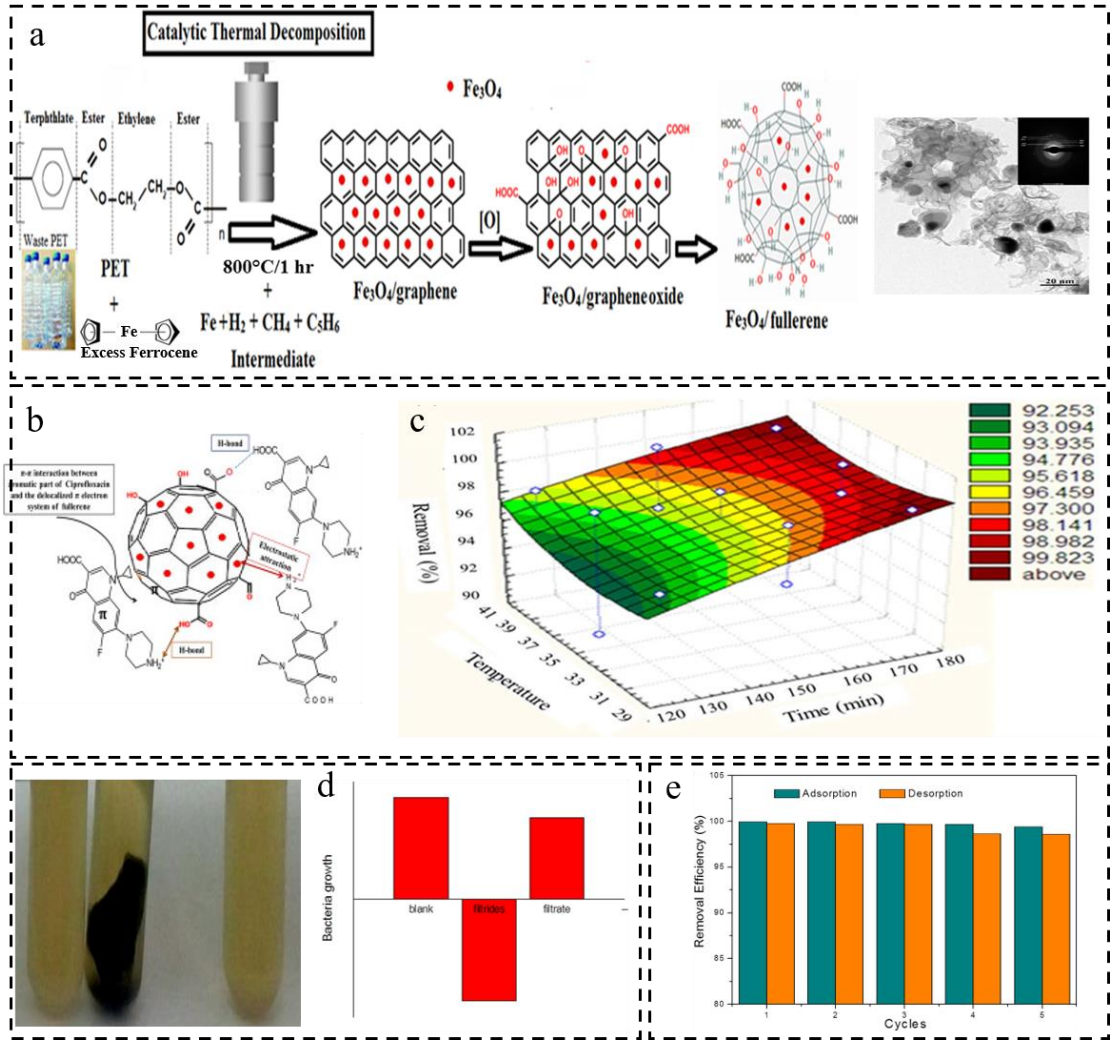

Figure 4. Schematic illustration for the one-step formation of FMFNs, (b) different types of interactions involved in the adsorption of ciprofloxacin on FMFN, (c) 3D plot of ciprofloxacin removal (%) versus process variables onto prepared FMFN, (d) confirmation test of the efficiency of adsorption process on E. coli growth, and (e) adsorption-desorption cycles of ciprofloxacin on FMFNs. (Elessawy et al., 2020)

3.3 Environmental remediation

Industry development has brought about a series of environmental problems. Performing environmental remediation to remove contaminants or pollutants from environmental media such as air, soil, groundwater, or ocean, is one of the important ways of achieving sustainable development. In recent decades, air pollution is deemed a global problem that may endanger all human health. (Dunlap, 2014) Especially, there has been an increase in air pollution with increased particulate matter ($PM_{0.1–10}$) concentration. (Anderson et al., 2012) Filtration has been widely applied to remove particulate matter from the atmosphere due to its high collection efficiency, low cost and simplicity of use. Bonfim et al. (Bonfim et al., 2021) used waste PET bottles as a filter media by using electrospinning for air filtration. The filtration capacity of the fibrous membrane was determined by examining the penetration ability for sodium

chloride (NaCl) aerosol particles with a diameter of 9 to 300 nm. The prepared fibers obtained a diameter of 0.65 μm to 3.25 μm. The filtration results showed a collection efficiency of 99% for nanoparticles, which indicates that the filter media could be applied in air filtration. Strain et al. (Strain et al., 2014) reported a tough fibrous membrane for smoke filtration. The fibrous membrane was produced from waste PET bottles by using electrospinning. The diameter of the fiber was controlled within a range of 0.4 μm to 4.3 μm. The air filtration was determined by passing cigarette smoke through the fibrous membrane. The results revealed that fiber with an average diameter of 1.0 μm shows the highest tensile strength and toughness of 62.5 MPa and 65.8 MJ/m^3, respectively, and at the same time, exhibits the optimal combination of mechanical property and filtration capacity (32g/g). Ashish et al. (Kakoria et al., 2021) fabricated two types of nanofiber mats from waste PET via supersonic solution blowing (SSB) and electrospinning (ES) for filtration of $PM_{0.1-2}$, as shown in **Figure 5(a)**. Compared to ES mats, the SSB mats sandwiched between two cotton fabric exhibited better particle filtration efficiency (PFE). The SSB-16 mats demonstrated the PFE for PM_2 of 99.7% with a quality factor (QF) of 0.03 Pa^{-1} and pressure drop (ΔP) of 11.66 Pa/cm^2. The SSB-16 mask retained over 99% of its PM_2 removal capacity after 10 washing and drying cycles.

CO_2 is considered a major contributor to global warming and should be responsible for melting glaciers and rising sea levels. In addition to reducing carbon emissions, capturing atmospheric CO_2 and converting it into valuable products is a promising route to achieve sustainability. (Lo et al., 2016; Teo et al., 2022) Using waste PET bottles as raw material to develop activated carbon for CO_2 adsorption offers a promising strategy to solve two environmental problems simultaneously. (Li et al., 2023; Shen, 2022) Wang et al. (Wang et al., 2020) explored the possibility of using negative emissions during the carbon capture process with waste PET as the CO_2 adsorbent. They conducted sensitivity analyses on CO_2 capture, heat energy needed for CO_2 desorption and bottle recycling to show that the transformation of waste PET to CO_2 adsorbent can realize negative CO_2 emissions, as an environmentally friendly strategy to close the carbon loop. Kaur et al. (Kaur et al., 2019) developed a waste PET derived adsorbent for CO_2 capture. Waste PET was carbonized at different temperatures, then chemically activated by using different KOH: PET mass ratios. The optimum sample, PET-3-700 (with a KOH: PET mass ratio of 3 and an activation temperature of 700 °C), provides a large surface area of 1690 m^2/g and a micropore volume of 0.78 cm^3/g, which offers a maximum CO_2 uptake capacity of 2.31 mmol/g under 100% CO_2 flow at an adsorption temperature of 30 °C. Besides, the prepared carbon adsorbent can be regenerated through four cycles of adsorption-desorption. PET plastics have also been converted to microporous carbon through N_2 doping and one-pot synthesis, see **Figures 5 (b-d)**. (Yuan et al., 2020) The prepared microporous carbon has a high CO_2 adsorption capacity of 4.58 mmol/g at 25 °C and 6.23 mmol/g at 0 °C (1 atm). In particular, the N_2 doped microporous carbon shows a high CO_2 selectivity over N_2, which means that it is practical to treat flue gases (90% N_2 and 10% CO_2) at different temperatures.

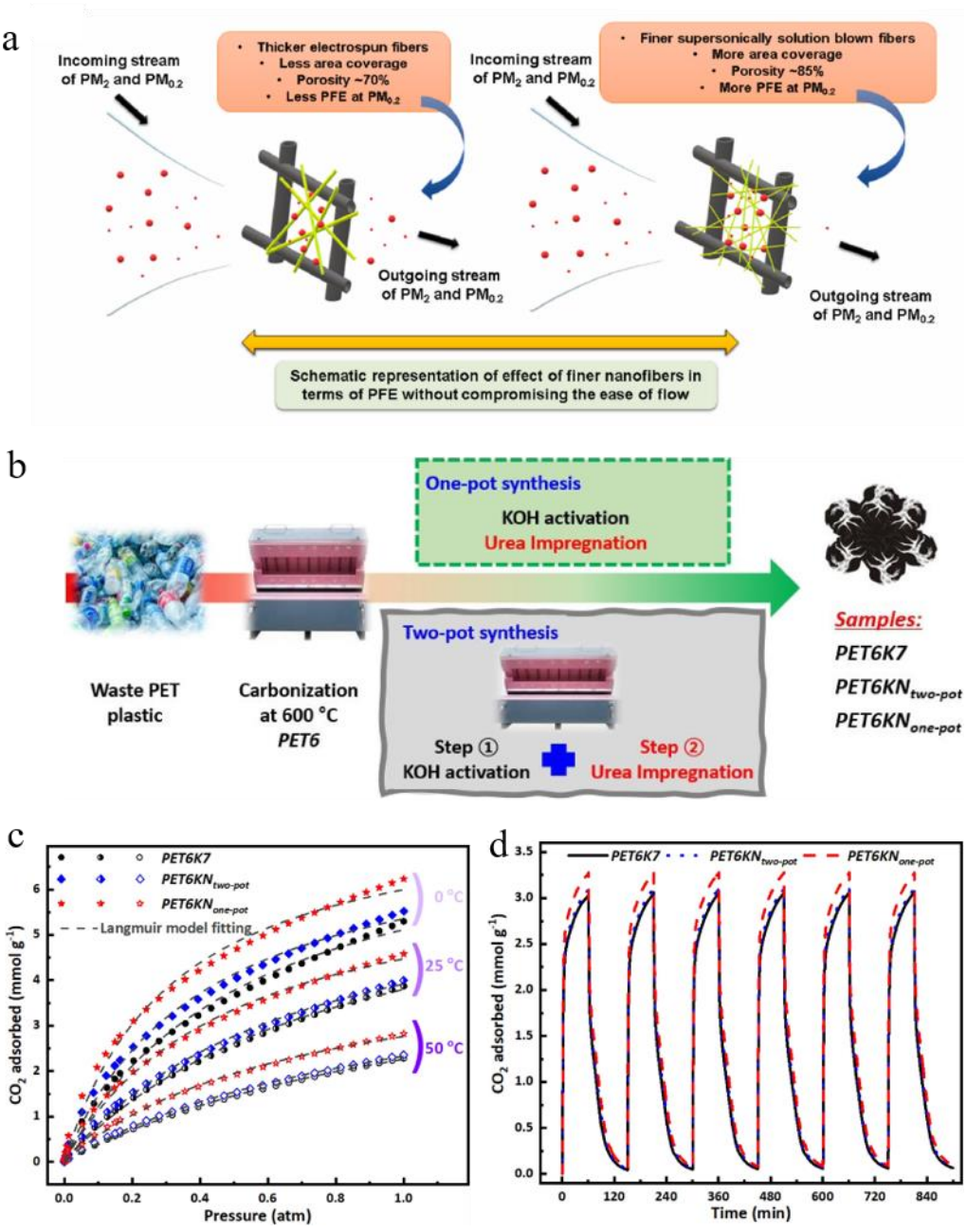

Figure 5. (a) Schematic illustration of effect of enhanced count of fibers in SSB-16 mats compared to ES mats, (Kakoria et al., 2021) (b) Synthesis scheme of N-doped microporous carbons derived from waste PET, (c) CO_2 adsorption isotherms at 0, 25, and 50 °C for PET6K7 (circle), $PET6KN_{two\text{-}pot}$ (diamond), and $PET6KN_{one\text{-}pot}$ (star). The dashed fitted lines are obtained from a Langmuir model, and (d) cyclic tests of CO_2 adsorption-desorption at 30 °C and 1 atm. (Yuan et al., 2020)

Oil pollution also causes environmental problems, such as air, soil, and ocean pollution, and damages the ecological system (Choi and Cloud, 1992). Adsorption is also considered to be an efficient strategy to remove oil spills. It was reported that waste PET materials can be used to clean up various pollutants in soils, such as oil and some heavy metals, thus providing a solution to remove spilled automotive oil in soils.

(Adebajo et al., 2003; Ahmad et al., 2014) Janqamsari et al. (Janqamsari et al., 2021) synthesized an efficient oil adsorbent (a polyvinyl alcohol (PVA)-recycled (rPET) nanocomposite) by using recycled PET fibers and PVA. Carbon nanotubes (CNTs) were further embedded on the PVA-rPET nanocomposite to determine their influence on porosity, density and oil absorption capacity. The prepared PVA-rPET adsorbents have low density and high porosity of 0.043-0.097 g/cm^3 and 92.93-96.87%, respectively. The PVA-rPET sample has a maximum absorption capacity of kerosene oil close to 12 g/g, which is enhanced to 15.1 g/g with the addition of 2wt% CNTs onto the aerogel structure. The optimum aerogel adsorbent has a large surface area of about 272 m^2/g and a four-cycle reusability, demonstrating the potential applications for absorbing oil from water, the atmosphere and soil. Waste PET derived nanofibrous membranes can also be applied to remove oil from water in natural environments. Topuz et al. (Topuz et al., 2022) dissolved waste PET in organic solvent systems and produced a PET membrane with the electrospinning method. The morphology of the nanofibers was controlled by adjusting the concentration of the polymer solution. The PET nanofibrous membranes have high adsorption capacity of 11.1 ± 1, 19.6 ± 1.8, 22.9 ± 2, and 19.3 ± 1.6 g/g for gasoline, diesel, crude oil, and pump oil, respectively. The PET nanofibrous membranes also have high flexibility and Young's modulus according to a dynamic mechanical analysis.

3.4 Catalysis

Catalysis is another important application of waste PET based nanostructured materials. (Chan and Zinchenko, 2023; Semyonov et al., 2022) Hydrogen (H_2) is a prospective green energy resource because it has zero carbon emission and high energy density, and electrochemical water splitting is the most practical and effective approach to produce H_2. (Chen et al., 2023) Ubaidullah et al. [19] reported the use of an N_2 doped mesoporous carbon functionalized zinc oxide nanocomposite (ZnO@NMC) for electro-catalysis in hydrogen evolution reactions (HERs) and oxygen evolution reactions (OERs). The ZnO@NMC nanocomposite was fabricated from MOF-5, which was built from waste PET-based BDC via a low temperature solvothermal approach. The prepared ZnO@NMC nanocomposite has a high meso-porosity with a pore radius of about 25 Å, and a large specific surface area of 939 m^2/g, which offer many active sites and ease of charge relocation for electrocatalytic HERs and OERs. The study showed that the prepared ZnO@NMC nanocomposite has enhanced electrocatalytic activity for both HER and OER in a 0.5 M KOH solution, with over-potential (η_{10}) and a Tafel slope of 0.39 mV and ~108 mV/ dec for HER, and 0.57 V and ~318 mV/dec for OER. (Ubaidullah et al., 2020) Veksha et al. (Veksha et al., 2020) converted flexible PET packaging waste into multi-walled CNTs (MWCNTs) for electrocatalytic applications. During OER, the electrodes from the MWCNTs outperform conventional Pt-based and commercial MWCNT electrodes.

Upcycling waste PET and electroplated sludge (EPS) into MOFs can both reduce pollution and encourage a sustainable economy. Song et al. (Song et al., 2021)

synthesized an Ni-MOF nanocrystal by using waste PET and EPS, to develop a nanocatalyst that would facilitate the photoreduction of CO_2 to generate a CO and H_2 reaction. The Ni-MOFs are successfully synthesized even in the presence of coexisting ions such as Fe^{3+} and Cu^{2+}. The Ni-MOF nanocatalyst has a superior photocatalytic performance for the photoreduction of CO_2 to CO, which offers a CO evolution rate of around 10 mmol/Hg. (Song et al., 2021) Lo et al. (Lo et al., 2016) used waste PET bottle material as the starting precursor to construct five BDC-based nanoporous trivalent MOFs including MIL-47, MIL-53(Ga), MIL-53(Al), MIL-53(Cr), and MIL-101(Cr). The gas adsorption tests for MIL-53(Cr) and MIL-101(Cr) showed their superior CO_2 and H_2 uptake capability. Besides, the prepared MIL-47 and MIL-101(Cr) can transform adsorbed CO_2 to corresponding cyclic carbonates, thus indicating their good catalytic performance.

Waste PET derived nanoporous carbon (NPC) can be used as a carrier to prepare NPC supported Pt catalysts (Pt/NPCs) to catalyze the reductive alkylation of p-aminodiphenylamine with methyl isoamyl ketone. (Wenlong et al., 2019) The catalytic efficiency and reusability of the Pt/NPCs could be enhanced by increasing the pore size of the NPCs when the pore size is smaller than 14 nm. The conversion rate of p-aminodiphenylamine can still be maintained at 98% after 10 cycles. Gangaraju et al. (Gangaraju et al., 2022) reported the fabrication of Ni-based MOFs on CNTs (Ni-MOF/CNTs) and their application for arsenic removal and electrocatalytic oxidation of urea. The Ni-based CNTs were synthesized through the pyrolysis of nickelocene under microwave heating, then waste PET derived BDC, sodium nitrate ($NaNO_3$) and the Ni-based CNT were heated at 150 ℃ for 6 hours. The nano-sized Ni-MOF particles were decorated on the defect-rich CNTs which finally formed the Ni-MOF/CNTs (**Figure 6**). When used for arsenic removal, the Ni-MOF@CNT showed high removal ability in a wide range of pH from 4.5 to 8.5. At the same time, the fabricated Ni-MOF@CNT electrodes showed enhanced current response, lower onset potential and charge-transfer resistance when utilized as electrocatalysts in reactions of urea oxidation.

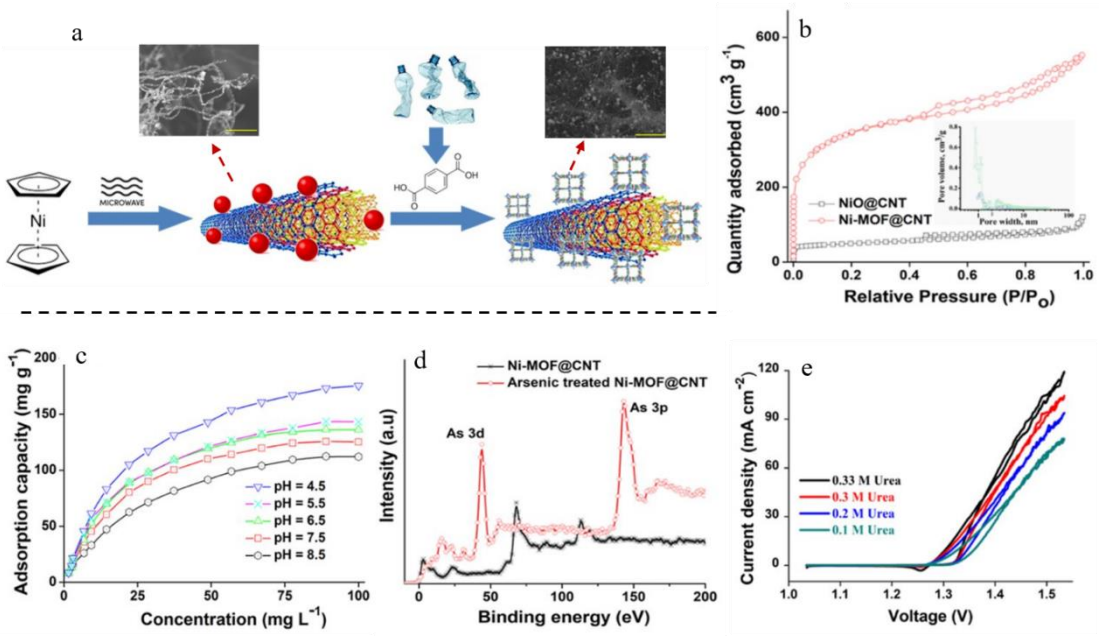

Figure 6. (a) Schematic of synthesis of Ni-MOF@CNT from nickelocene and waste PET, (b) Brunauer-Emmett-Teller (BET) surface area of NiO@CNT and Ni-MOF@CNT, (c) adsorption isotherms of arsenic on Ni-MOF@CNT nanostructures at various pH values, (d) low binding energy X-ray photoelectron spectroscopy peaks that show adsorption of arsenic peaks on Ni-MOF@CNT, and (e) urea electro-oxidation CV curves of Ni-MOF@CNT with increasing urea concentration. (Gangaraju et al., 2022)

3.5 Other applications

Waste PET-based nanostructured materials can also be used in many other fields. In particular, microbial contamination has once again substantially threatened public health during the rampage of the COVID-19 pandemic globally. (May, 2021) Researchers are now focusing on preventing and controlling viruses and targeting microbial organisms to prevent the next global pandemic. Recycling waste PET to fabricate antimicrobial nanostructured materials has attracted much research attention. For example, Vázquez et al. (Vázquez et al., 2021) developed antibacterial and antifungal zinc oxide nanoparticle-doped PET nanofibrous membranes through electrospinning. The prepared nanofibrous membranes are effect against Escherichia coli and penicillium. Grumezescu et al. (Grumezescu et al., 2019) fabricated antimicrobial electrospun PET nanofibrous membranes decorated with Ag nanoparticles. The prepared membranes showed an antibacterial effect on gram-positive and gram-negative bacteria. The in vitro and in vivo biocompatibility tests indicated the promising biomedical applications of these membranes, such as for wound dressings implant coating, etc. Soltanolzakerin-Sorkhabi et al. (Soltanolzakerin-Sorkhabi et al., 2023) also developed Ag decorated PET nanofibrous membranes, which have good antibacterial and antibiofilm activity against the Pseudomonas aeruginosa, Escherichia coli, and Staphylococcus aureus strains. Their excellent antimicrobial performance has enhanced their popularity for various applications, such as water

purification and air filters.

Koh et al. (Koh et al., 2018) developed a series of PET-based aerogels for acoustic and thermal insulation applications. Acoustic and thermal insulated aerogels were fabricated from waste PET fibers, PVA and a crosslinker glutaraldehyde (GA) by using freeze-drying. The prepared aerogels have low densities that range from 0.007 to 0.026 g/cm^3, a porous network structure with a porosity of 98.3 to 99.5%, and high flexibility with a low compressive Young's modulus that ranges from 1.16 to 2.87 kPa. These waste PET-based aerogels have excellent acoustic absorption capacity, as well as good insulation properties with a low thermal conductivity that ranges from 0.035 to 0.038 W/m.K. Photochromism can be used for the anticounterfeiting of commercial commodities. El-Newehy et al. doped (El-Newehy et al., 2022) highly phosphorescent rare-earth aluminate nano-sized (REAN) particles in waste PET solutions to fabricate REAN@PET nanofibrous films for anticounterfeiting purposes. The diameter of the nanofiber ranges from 180 to 220 nm, and the nanofiber shows enhanced superhydrophobicity with a larger REAN ratio. The REAN@PET nanofibrous films show rapid and reversible photochromism when exposed under UV light. Besides, the recycled PET plastics are also widely used in construction and buildings (Behzadian and Shahrajabian, 2019; Ge et al., 2014; Nematzadeh et al., 2020), packaging (Ciobanu et al., 2023; Sánchez et al., 2014) and textiles (Albini et al., 2019; Harmsen et al., 2021; Park and Kim, 2014).

4 Conclusion and future directions

In this literature review, the conversion process of waste PET into high value-added materials for a variety of different applications has been summarized. In particular, PET derived MOFs and carbonaceous materials can be widely applied for energy storage (in supercapacitors, LIBs, and MFCs), water treatment (to remove dyes, heavy metals, and antibiotics), environmental remediation (for air filtration, CO_2 adsorption, and oil removal) and catalysis (to produce H_2, photoreduce CO_2, and remove toxic chemicals). The performance of these high value-added materials can be improved by tuning their nanostructure (especially porosity) and surface properties (by doping or forming functional groups). The transformation of waste PET into high value-added nanostructured materials contributes to both environmental sustainability and the economy. Although much work has been done to transform and apply these upgraded nanomaterials to date, there are still several challenges that need to be addressed in future work, as follows.

(1) The efficiency of transformation processes should be increased to promote upcycling practices and large-scale applications. For example, conventional carbonization techniques are energy and time consuming, and the fabrication of carbonaceous electrodes require complex instruments and synthetic processes.

(2) The performance of the final products contributes to the replacement of traditional materials with novel waste PET-based functional materials. In addition to the

innovativeness of these materials, more research is needed to improve the performance of these materials to meet the different requirements of practical applications.

(3) The stability of the synthesized functional materials is of great importance for environmental protection. The breakdown of these materials may leach heavy metal or other toxic components, which will cause environmental pollution.

In conclusion, converting waste plastics into functional nanostructured materials for different applications has been successful in lab experiments and industrial trials. However, more needs to be done in future studies to explore efficient and reliable conversion processes to fabricate low-cost, high-performance, and stable materials that can be mass-produced for a variety of different applications.

Conflicts of interest

The authors declare no conflict of interest.

Acknowledgments

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1178