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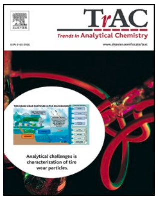

Perspectives on sustainable plastic treatment: A shift from linear to circular economy

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ABSTRACT

The pervasive presence of plastics in the environment, particularly microplastics, has become a significant global challenge, demanding innovative solutions for their management and upcycling. While traditional methods including landfill and incineration face limitations in environmental impact, emerging technologies offer promising pathways for the conversion of plastics into valuable chemicals and fuels, operating under ambient conditions and often utilizing sustainable energy sources. Considering the current research progress in plastic upgrading, it is necessary to summarize the chemical upcycling of plastic waste. To this end, this review provides an overall examination of current and emerging methodologies for plastic treatment, including pyrolysis, hydrogenolysis, photocatalysis, and electrocatalysis. Existing knowledge gaps and future research directions are then proposed. Overall, this review highlights the potential of these novel plastic management approaches in aligning with the principles of a circular economy.

1. Introduction

The escalating prevalence of plastic waste in the environment has emerged as a significant contemporary challenge, raising growing concerns about its potential adverse effects on our ecosystem [1]. However, these concerns have been further exacerbated by the recent outbreak of the COVID-19 pandemic, leading to an increased global demand for plastics in packaging and personal protection [2]. During the pandemic, a staggering amount of over 129 billion face masks and 65 billion gloves were manufactured and discarded each month [3]. Out of the vast array of plastic, nearly 80% end up as waste [4]. In the absence of reasonable treatment, these discarded plastics can contribute to significant pollution of aquatic and terrestrial environments.

Terrestrial ecosystems, regarded as the primary repositories for plastics, are believed to encompass a greater plastic content than marine environments [5]. The degradation of plastic debris by external forces, such as solar radiation or wind, results in the formation of microplastics, characterized by a particle diameter of less than 5 mm [6]. When embedded in soil, the accumulation of these microplastics initiates a series of environmental processes. For example, microplastics have been found to modify soil physicochemical properties, including bulk density, porosity, aggregation, electrical conductivity, water retention capacity, and pH levels [7]. Moreover, microplastics can affect the soil nutrient cycle by altering organic matter composition and nutrient availability [8]. Some studies indicated that microplastics can improve microbial colonization and act as carriers for transporting pollutants and pathogenic bacteria to enhance the spread of antibiotic resistance due to their high specific surface area [9]. These adverse effects on the soil environment indicate the urgency of effective action in waste plastic management.

Among conventional plastic treatment processes, landfilling and mechanical reprocessing stand as the most prevalent and easily scalable methods. Landfilling primarily depends on microbial metabolisms to degrade contaminants [10]. Several reviews have consolidated insights into the intricate biogeochemical transitions transpiring within landfills. This includes a comprehensive five-phase conceptual model encompassing the aerobic phase, anaerobic acid phase, rapid methanogenesis, slow methanogenesis, and the stable phase [11]. Due to the prolonged weathering, the physicochemical properties of plastics can be altered, thereby increasing the release of additives and adsorbed contaminants in plastics, ultimately influencing the stability of nearby ecosystems. In addition, this method is constrained by spatial considerations, and emits

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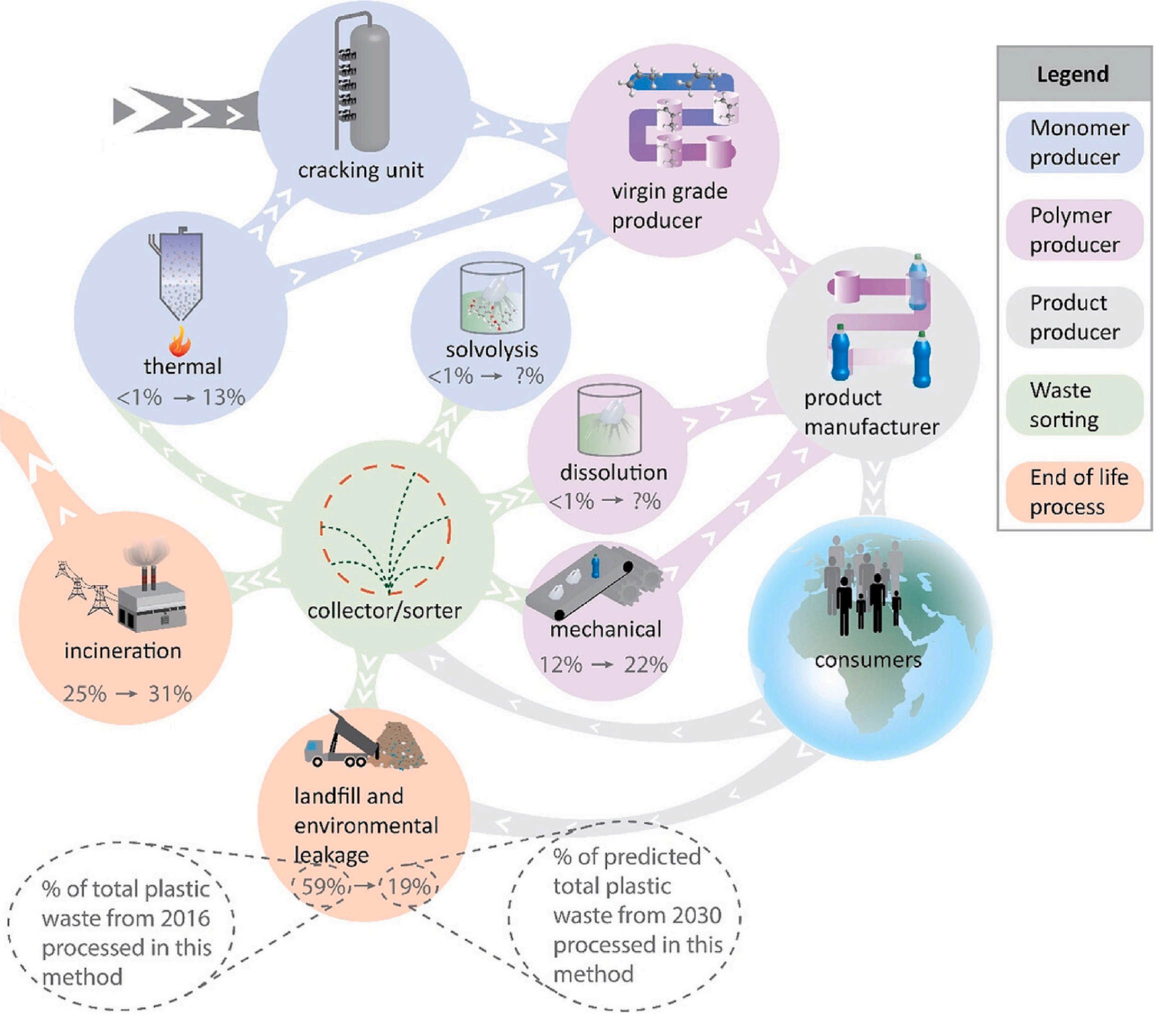

Fig. 1. Illustration of current plastic end-life process with a linear model (bottom left with orange circle) and proposed plastic recycling management with circular economy [40]. Copyright © 2020 The Authors. Published by Wiley-VCH Verlag GmbH & Co. KGaA (CC-BY-NC 4.0). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

undesirable greenhouse gases such as methane and carbon dioxide [12]. Mechanical reprocessing involves the physical processing and reuse of plastic waste. However, the value of recycled plastic products often diminishes due to the thermal and mechanical degradation of polymers, leading to varied use cases [13].

In an ideal circular economy, plastics should originate from renewable or recycled sources [14], a paradigm starkly contrasting the current linear plastic economy of produce, use, and disposal. This linear model, as discussed earlier, disregards numerous environmental concerns, perpetuating the adverse environmental impact of plastic use and disposal. To address this, it is imperative to integrate circular economy principles into current plastic management practices. This integration is pivotal for mitigating plastic pollution and its associated environmental consequences. The central aim of a circular economy is to increase the proportion of plastic undergoing reuse or recycling [15]. To achieve this objective, two plastic recycling strategies could be considered [16]: 1) Depolymerize plastic into monomers or small molecule substances (e.g., CO_2 or H_2) and then transform into new polymer or chemicals; 2) Directly transform plastic into high-value chemicals.

Early research efforts have resulted in the development of pyrolysis and hydrogenolysis techniques, showcasing promising avenues for polymer deconstruction. These methods offer a means to process a variety of plastic waste, generating valuable compounds such as naphtha-containing paraffin, olefins, and aromatics [17,18]. Pyrolysis and hydrogenolysis represent catalytic processes that cleave chemical bonds in organic molecules, yielding molecular fragments. These reactions offer a method to process diverse plastic waste, generating naphtha-containing paraffin, olefins, and aromatics. For example, Nabgan et al. developed bifunctional Ni–Pt nanocatalysts for plastic waste valorization to liquid fuel at 500–700 °C [18]. A hydrogenolysis system was also developed to transform polyethylene (PE) into alkanes using a Pt–SiO_2 catalyst under 300 °C and 1.4 MPa H_2 [17]. Despite not necessitating additional chemical reagents, the elevated reaction temperatures inherent to these techniques can induce side reactions, including monomer dehydration and racemization. Consequently, pyrolysis and hydrogenolysis give rise to intricate hydrocarbon mixtures, posing challenges in separation and thereby limiting their accessibility [19]. Recently, light-driven photocatalysis and electrocatalysis have been recognized as a potentially sustainable method for plastic waste management, utilizing solar energy or sustainable electricity at ambient temperature and pressure. Similar to pyrolysis and hydrogenolysis, both photocatalysis and electrocatalysis offer effective methods for breaking down polymer compounds into monomers. These processes operate under the influence of light irradiation or electric fields, providing a milder alternative even at room temperature and ambient pressure. Notably, $FeCl_3$ and certain acids have been employed as photocatalysts for the depolymerization of polystyrene, demonstrating efficacy at room temperature [20]. The mechanisms showed that the hydrogen atom transfer (HAT) was responsible for the C–H bond break induced by the organic peroxyl radical intermediate [21]. Similar to photocatalysis technology, electrocatalysis represents a sustainable and compelling approach to converting plastics to valuable chemicals because its energy source can come from clean energy. In recent research, Myren et al. developed a two-compartment electrochemical reactor to facilitate the hydrolysis of poly (ethylene terephthalate) (PET) into terephthalate with a yield of 16.9% [22]. In addition, Kim et al. introduced a Zr-doped hematite photoanode that extracts electrons from hydrolyzed PET waste to produce chemical fuels [23]. Considering current progress in plastic upcycling technology, a comprehensive review is needed to further guide future research on the topic.

To facilitate the transition from a linear to a circular economy in waste plastic treatment and avoid the generation of microplastics by conventional linear approach (i.e., landfilling and incineration), this review seeks to provide an overview of the current advancements in the chemical upcycling of plastics. Specifically, it commences with an examination of the urgent need for plastic recycling, followed by a summary of key pyrolysis, hydrogenolysis, photocatalysis, and electrocatalysis techniques. Lastly, this review highlights existing knowledge gaps and provides thoughtful perspectives to guide future progress in plastic upcycling.

2. Advancing waste plastic management towards upgraded recycling

Plastic recycling is paramount in the context of plastic pollution and the circular economy. The prevailing plastic economy largely adheres to a linear model, resulting in substantial waste production (Fig. 1). Plastics, primarily utilized for packaging, often end up in landfills, with reports indicating that from 1950 to 2015, a mere 10% of plastics were recycled, while over 60% were discarded into landfills or the natural environment [24]. Although landfills do sequester embedded greenhouse gases, their linear management approach poses environmental hazards due to the emission of toxic combustion byproducts such as dioxins and furans [25], adding to the environmental burden of this waste disposal method. Furthermore, current plastic production is heavily reliant on petroleum resources, conflicting with societal energy demands. Landfills also discharge microplastics through landfill leachate, potentially endangering soil and underground water, as microplastics act as sinks for hydrophobic organic contaminants [26,27].

Addressing these challenges, the development of a circular economy for plastic treatment emerges as a logical and optimal solution to reduce reliance on finite fossil materials while alleviating environmental risks and managing end-of-life waste plastics through recycling. This strategy has been acknowledged as one of the top-10 emerging technologies in chemistry by IUPAC in 2009 [28]. However, despite its potential advantages, the overall recycling rate remains disconcertingly low [13]. Four main approaches to recycling plastic waste exist, commonly classified as primary, secondary, tertiary, and quaternary recycling [29].

Primary and secondary recycling, categorized under mechanical recycling, encompasses sorting, washing, crushing, melting, and remolding plastics [30]. The distinction between the two lies in the feedstock utilized. Specifically, primary recycling targets the plastic waste with clean and known origin, with the goal of producing products that retain qualities and properties akin to the original plastic [31]. Conversely, post-consumer plastic waste was the feedstock in secondary recycling, necessitating rigorous collection, sorting, and washing to procure clean material for mechanical recycling [32]. Both primary and secondary recycling undergo the crucible of harsh conditions, wherein high temperatures or formidable mechanical forces come into play, catalyzing the inevitable degradation of polymers. A case in point is the pervasive adoption of thermo-mechanical recycling in the reclamation of polyethylene terephthalate (PET) waste, a process entailing the facets of remelting and reprocessing. However, these high temperatures lead to significant thermal degradation, birthing lower-grade plastics characterized by diminished optical, thermal, and mechanical properties. Consequently, their applicability is primarily in domains characterized by less stringent engineering requirements, distinctly distant from the loftier benchmarks set by their original applications [33]. In practice, mechanical recycling is mainly used for handling polyethylene (PE), polypropylene (PP), and PET, while the recovery rates for most other plastics remain negligible in comparison [29].

Another method of recycling involves the conversion of plastic waste into chemical feedstocks, a process referred to as tertiary or chemical recycling [25]. This approach seeks to chemically deconstruct plastic materials, breaking bonds through mechanisms such as glycolysis and pyrolysis, with the goal of depolymerization, rather than mere re-extrusion [16]. Certain polymers, such as PE, present obstacles in depolymerizing into original monomers through reverse synthesis reactions [6,34]. A concept known as “chemical upcycling” has been introduced, showing potential in the development of plastic waste upcycling technology to synthesize chemicals, fuels, or high-value products [35,36]. Chemical recycling not only depolymerizes plastic waste into monomers for new plastic production but also is able to convert plastic waste into value-added chemicals, such as short-chain hydrocarbon fuels or valuable oxygenates to increase the sustainability [37,38].

Quaternary recycling involves the incineration of plastic waste with energy recovery. Discarded plastics are combusted to generate heat for electricity, exhibiting a high heating value akin to crude oil (45 MJ/kg), and ranging between 20 and 40 MJ/kg [39]. However, this method results in net energy loss and substantial greenhouse gas emissions, in addition to toxic gases from materials like polyvinyl chloride (PVC) incineration [40]. This underscores the importance of focusing on mechanical and chemical recycling technologies. In some jurisdictions, including the European Union, energy recovery is not considered a legitimate form of plastic recycling [29].

In summary, the utilization of recycled waste plastic to synthesize valuable chemicals or fuels can effectively mitigate the depletion of fossil fuels, aligning with the green strategy in the circular economy [41]. While primary and secondary recycling are viewed as the most sustainable and cost-effective options, challenges remain, such as the need for uncontaminated waste and the problem of downcycling. Incineration for energy recovery introduces environmental concerns due to emissions and the presence of microplastics in incineration ash. Chemical methods for upcycling plastics into valuable products demonstrate potential for achieving enhanced circular economy objectives.

3. Plastic depolymerization by pyrolysis and hydrogenolysis

The growing concern over the environmental impact of microplastics has led to a shift from the traditional linear model of plastic consumption and disposal to a circular economy framework, emphasizing resource efficiency, waste reduction, and sustainable practices [40]. Within this context, plastic depolymerization has become a key strategy. By breaking down complex polymers into monomers or valuable chemical feedstocks, depolymerization provides opportunities to recycle and repurpose plastics, reducing their negative effects and contributing to sustainability [42–44].

The primary method to increase the value of discarded plastics involves their conversion into higher-value polymers through depolymerization into distinct building blocks, followed by polymerization into new materials [45]. Depolymerization is the reverse of polymerization, and its direction depends on changes in enthalpy and entropy. In exergonic polymerizations, the enthalpy change is negative, indicating that the products have lower energy than the reactants, driving the reaction forward [25]. However, as monomers aggregate to form a polymer, their translational freedom decreases, resulting in a reduction of entropy. Within the context of polymerization, this loss of freedom

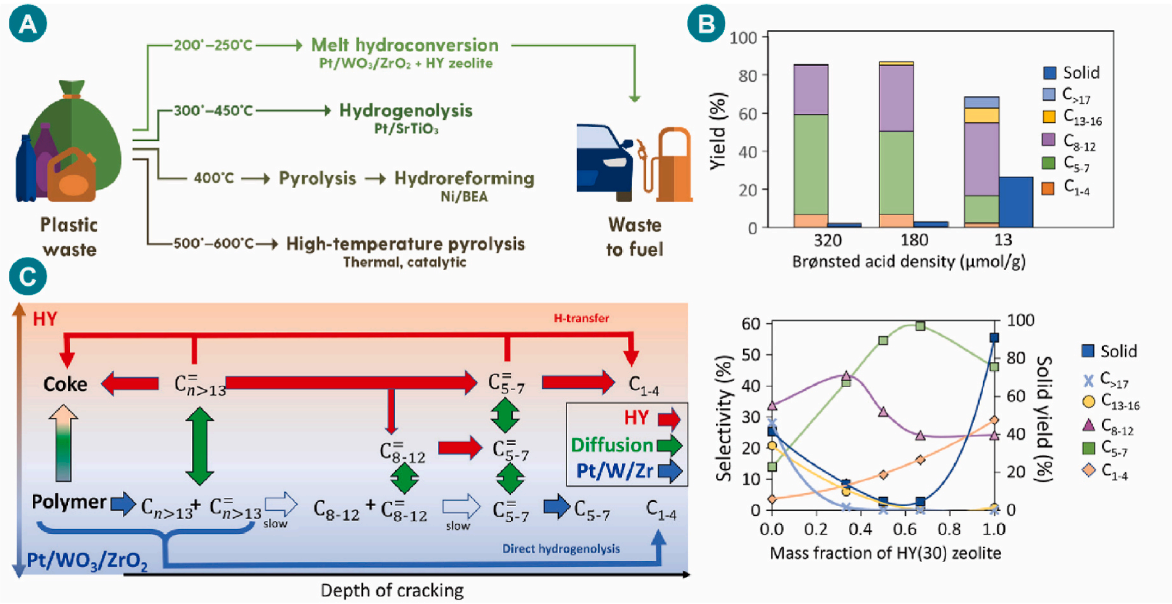

Fig. 2. (A) Illustration of catalytic pyrolysis and hydrolysis of plastic waste to fuels. (B) Depolymerization performance and (C) reaction network of low-density PE over $Pt/WO_3/ZrO_2$ to fuels [55]. Copyright © 2021 The Authors. Published by American Association for the Advancement of Science (CC-BY-NC 4.0).

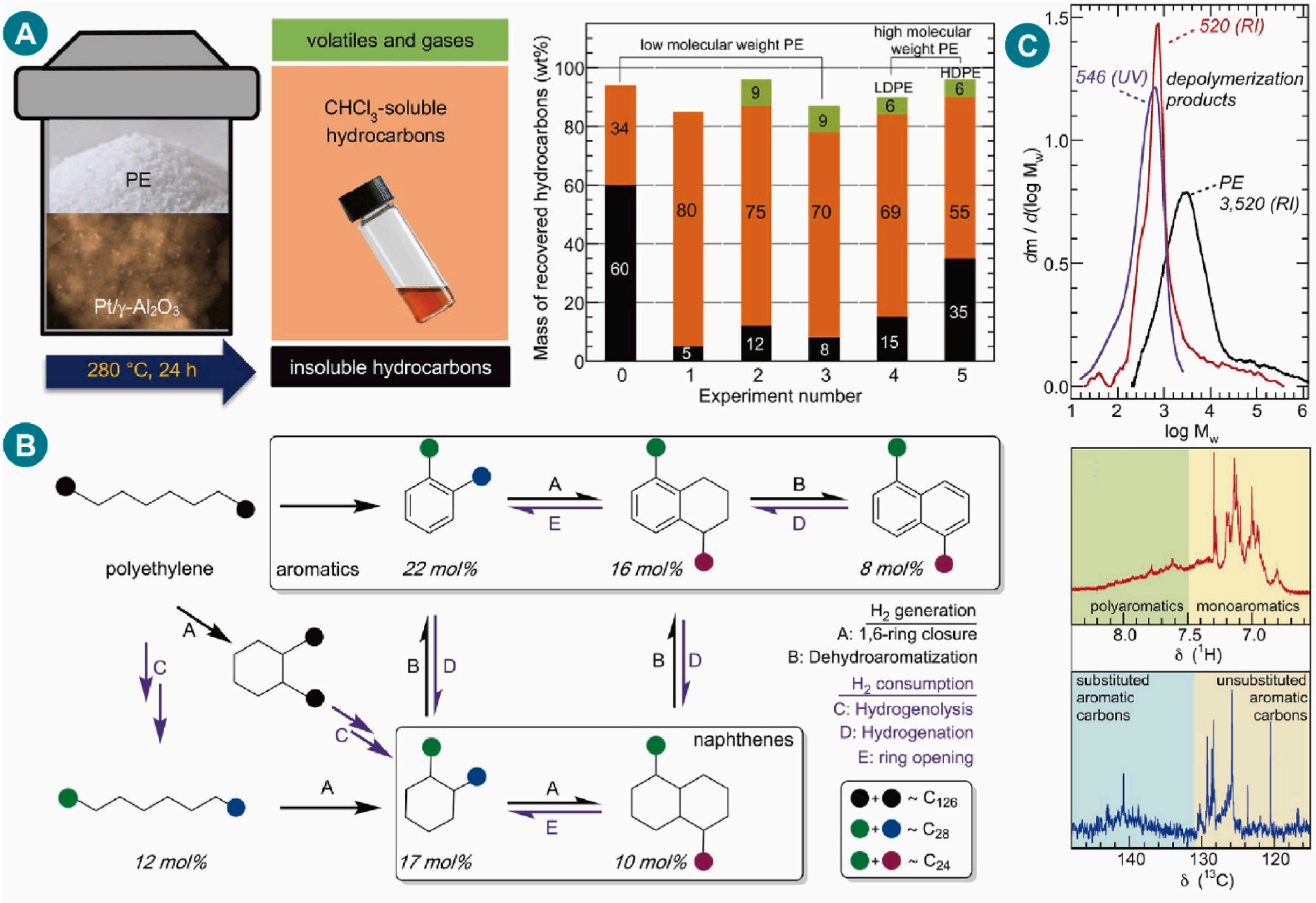

Fig. 3. (A) Illustration of reactor and product fractions in the hydrogenolysis PE process catalyzed by $Pt/\gamma\text{-}Al_2O_3$. (B) The conversion pathway of PE plastic to alkylaromatics and alkylnaphthenes and tandem polyethylene hydrogenolysis/aromatization mechanism. (C) The main liquid hydrocarbon fraction after reaction [56]. Copyright © 2021 The Authors. Published by American Association for the Advancement of Science (CC-BY-NC 4.0).

Table 1
The performance of photocatalysis for plastic waste upcycling.

Plastics	Catalysts	Wavelength (power)	Products	Yield	ROS	Ref.
PS	p-TsOH·H_2O	405 nm (9 w)	Formic acid	72%	singlet oxygen	[66]
Mix plastic	$UO_2(NO_3)_2\cdot 6H_2O$	460 nm (9 w)	Benzoic acid	33%	*UO_2^{2+}	[67]
PLA	ZnO/UiO66-NH_2	300 W Xe lamp	Acetic acid	14.4%	•OH and •O^{2-}	[68]
PET	$CN_X|Ni_2P$	>420 nm (100 mW cm^{-2})	Acetate and formate	–	–	[69]
PS	$FeCl_3$	254 nm	Benzoic acid	10.8%	•Cl	[70]

Note: p-TsOH·H_2O, p-toluenesulfonic acid monohydrate; ROS, reactive oxygen species.

incurs an associated "cost" that must be overcome for the reaction to proceed. For spontaneous exergonic polymerizations, the reduction in enthalpy must exceed the entropic penalty incurred by the loss of monomer translational freedom. In simple terms, the enthalpy change (ΔH) released during the reaction must be sufficiently large to compensate for the decrease in disorder (ΔS) caused by the polymerization process. In this sequence, the reaction temperature, governed by the $\Delta H/\Delta S$ ratio, becomes the key determinant for the feasibility and direction of the polymerization process. The theory of Gibbs free energy explains the existence of a critical temperature where the change in Gibbs free energy is zero, indicating an equilibrium state within the polymerization reaction [46]. In most polymerization processes, both ΔH and ΔS are negative [25]. Therefore, complete reversion to monomer requires elevated temperatures, forming the fundamental principle underlying the thermal strategy of plastic depolymerization.

According to existing literature, a globally distributed group of plastic waste includes PS, PE, polyurethane (PU), PP, PET, and PVC [47,48]. The pyrolysis performance of these plastics, along with their primary products, has been thoroughly summarized in previous reviews [49]. In essence, due to the high dissociation energy of C–C bonds (approximately 300–370 kJ/mol), polyolefins like PE show limited selectivity in thermolysis, primarily producing gases and char [42]. In contrast, PVC predominantly generates HCl due to competing reactions [50]. To refine product distributions and enhance quality, various catalysts are introduced into the pyrolysis processes, such as zeolites, metal catalysts and minerals [49]. For example, recent research indicated that the reaction temperature and reaction time of polymer pyrolysis process have significantly reduced from 490 °C with 120 min–350 °C with 90 min due to the usage of zeolite catalyst [51]. Additionally, catalyst augmentation has shown the ability to enhance the gasoline fraction (C5–C12) within the liquid oil through the cracking of heavier compounds induced by aluminum-substituted mesoporous (Al-SBA-15) catalyst [52].

To prevent undesirable coking and repolymerization reactions, the introduction of high-pressure hydrogen is used, facilitating the cracking of heavy molecules through a process known as hydrogenolysis [53]. In a hydrogen-rich environment, hydrogenolysis operates by cleaving C–C bonds, while hydrogenation simultaneously converts unsaturated molecules produced by hydrogenolysis into saturated products [54]. Similar to catalytic pyrolysis, catalysts play a crucial role in hydrocracking, reducing operational temperature and time, while increasing selectivity towards target products. Some products such as solid waxes and olefins were generated through the pyrolysis of PE within the temperature range of 400–600 °C (Fig. 2) [55]. A recent solvent- and hydrogen-free method, known as tandem hydrogenolysis–aromatization, has been developed for the selective breakdown of PE into valuable long-chain alkylaromatics and alkylcycloalkanes (Fig. 3) [56]. This process, catalyzed by Pt on γ-Al_2O_3 (Pt/γ-Al_2O_3), includes hydrogenolysis,

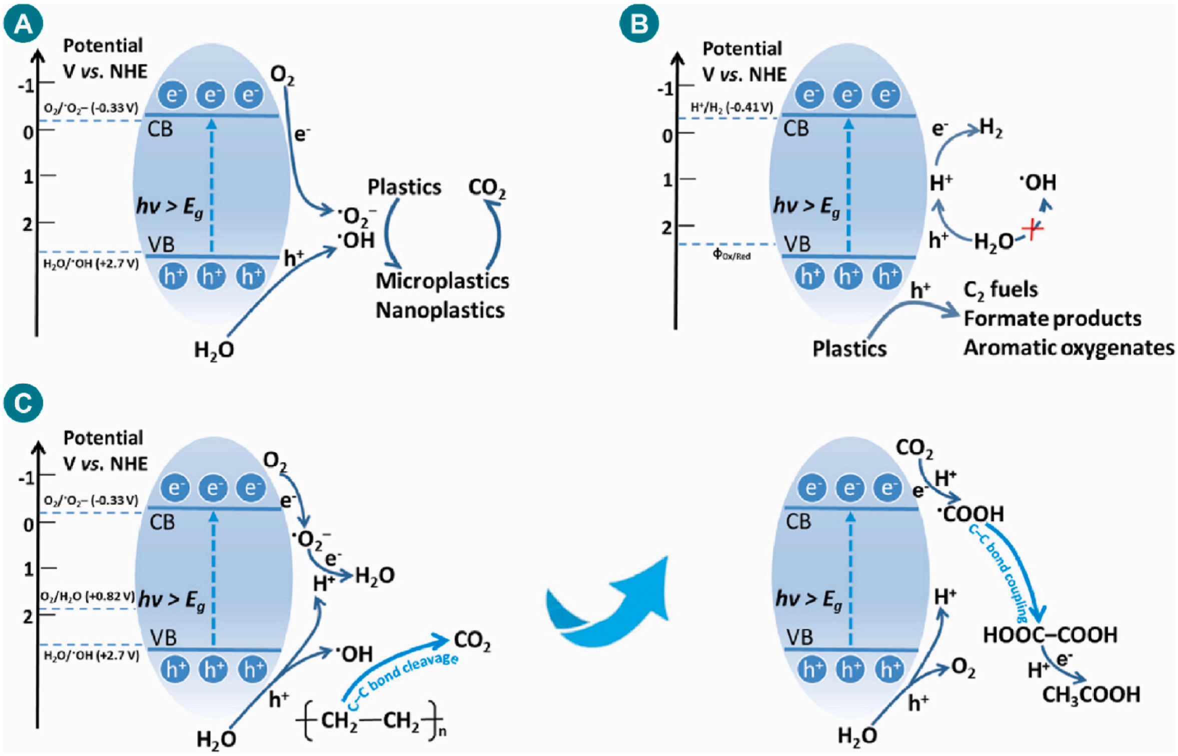

Fig. 4. Photocatalysis mechanisms of (A) plastic degradation with nonselective process, (B) selective fine chemical production and (C) acetic acid production with carbon dioxide as intermediate [61]. Copyright © 2023 American Chemical Society.

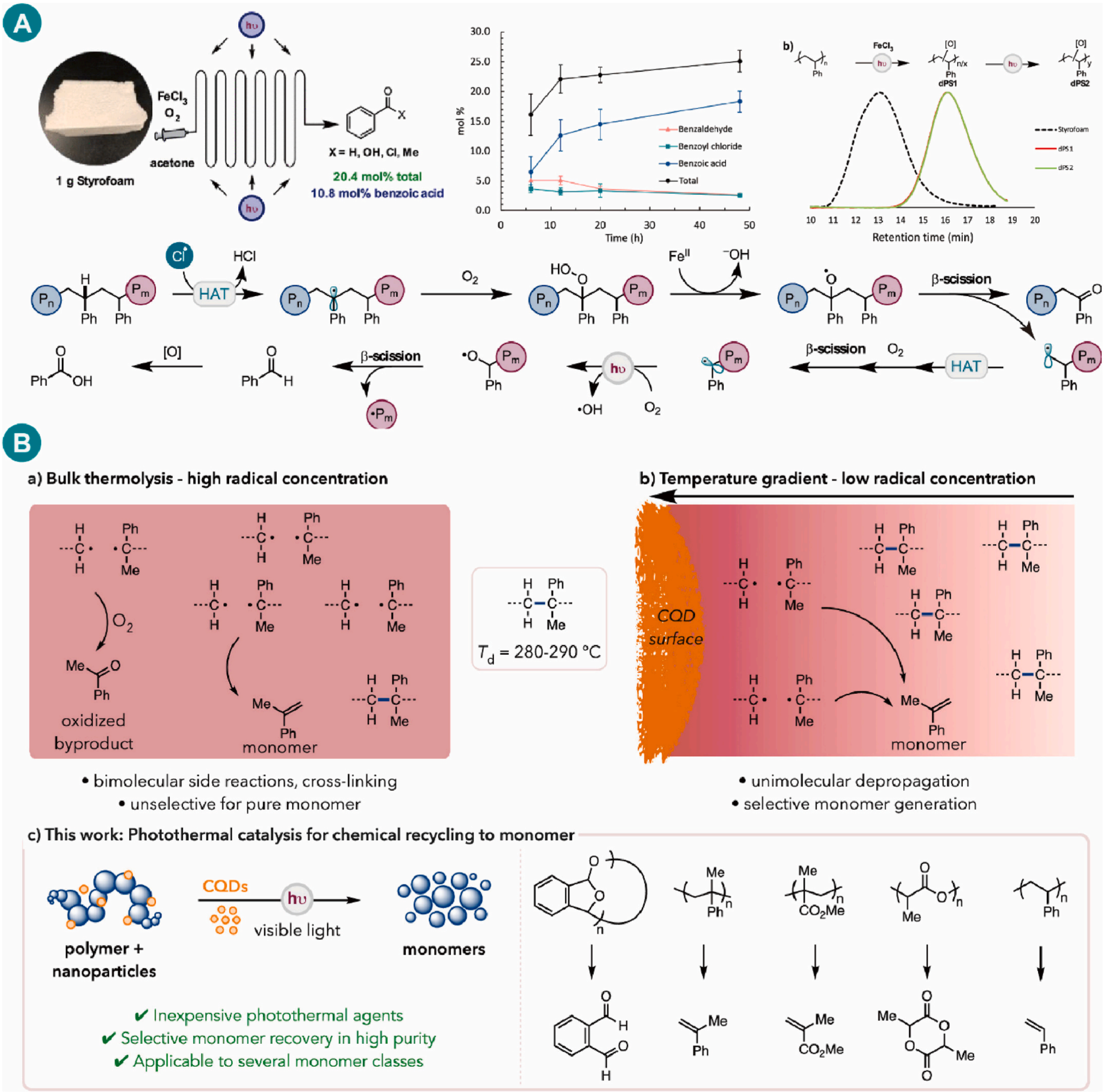

Fig. 5. (A) Mechanisms and performance of $FeCl_3$ induced PS valorization to benzoic acid [70]. (B) Photothermal conversion of various plastics to monomers by using carbon quantum dots [35]. Copyright © 2023 American Chemical Society.

hydrogenation, and ring-opening steps that involve the consumption of H_2 molecules. However, the coexistence of multiple linear and cyclic products with identical carbon numbers presented challenges in subsequent separation efforts [41]. In this context, catalysts featuring active metals on supported structures with open microenvironments or hydrocracking plastic over matched acidic sites and confined metals achieved a high catalytic efficiency and refined target selectivity [57,58].

4. Photocatalysis-induced plastic valorization to chemicals

Upon exposure to ultraviolet radiation or sunlight, plastics undergo a complex process in which their polymer macromolecules directly absorb photons. This absorption induces excited states, subsequently initiating a series of reactions that include chain scission, branching, cross-linking, and oxidation [6]. The inherent degradation time for plastics is notably protracted, extending over centuries. Consequently, the realization of effective photodegradation necessitates the strategic incorporation of suitable catalysts throughout the degradation pathway [4]. Analogous to the photocatalytic degradation of organic pollutants, the treatment of plastic via photocatalysis encompasses three fundamental stages: the absorption of protons by photocatalysts to generate electron-hole pairs, the migration of these pairs, and the ensuing surface reaction [59,60]. Within the context of photocatalytic plastic waste valorization, free radicals, particularly hydroxyl radicals, are instrumental in augmenting degradation efficiency, albeit with reduced selectivity. Typically, the generated hydroxyl radicals and superoxide radicals will

Table 2
The performance of electrochemical systems for plastic waste upcycling.

Catalysts	Cell voltage (V *vs.* RHE)	Current density (mA cm^{-2})	Products	FE (%)	Ref.
Au/$Ni(OH)_2$	1.15	326.2	glycolic acid	96	[82]
Pd/$Ni(OH)_2$	1.15	600	glycolic acid	>85	[83]
OMS-Ni_1-CoP	1.57	10	KDF	96	[84]
$NiCo_2O_4$	1.55	90	Formic acid	90	[85]
$CoNi_{0.25}P$/NF	1.8	500	Formic acid	>80	[80]
Ni_3N/W_5N_4/NF	1.6	120	Formic acid	85	[86]
Pd/NF	0.7	400	carbonate	93	[87]

Note: NF, Ni foam; KDF, potassium diformate.

indiscriminately assail the plastic waste, resulting in the formation of carbon dioxide, nanoplastics, and a composite of organic compounds [61]. Efficient photocatalysts targeting specific redox reactions must exhibit characteristics such as an optimal bandgap, effective separation of photoexcited electron-hole pairs, and suitable band energy levels. Contemporary strategies to enhance photocatalytic efficiency encompass 1) the integration of narrow bandgap materials with their wider bandgap counterparts [62], 2) the creation of heterogeneous structures [63], 3) the construction of photocatalysts with vacancies [64], and 4) the induction of strain [65], all of which contribute to reducing the activation energy required to convert plastics into smaller molecules [59].

Beyond the generation of carbon dioxide, photocatalysis may also yield certain organic acids (Table 1). Fig. 4 delineates the conversion mechanisms from polyethylene (PE) to acetic acid [61]. Initially, hydroxyl radicals are produced from water by separated holes on the photocatalyst surface. These radicals then attack the C–C bonds in PE molecules, forming carbon dioxide. Concurrently, oxygen is reduced into superoxide radicals and water by the electrons in the conduction band. Ultimately, the generated carbon dioxide is reduced into •COOH and further transformed into acetic acid through C–C bond coupling.

Recent studies have also identified the significance of atom transfer processes in polymer photocatalysis depolymerization. Specific research has reported the conversion of PS to benzoic acid with a $FeCl_3$ photocatalyst under visible light (Fig. 5) [35,70]. Fig. 5 shows the detail of this reaction pathway. Carbon-centered radicals are generated within the PS molecules through a HAT process, attributed to the chlorine radical originating from photoinduced ligand-to-metal charge transfer. Subsequently, oxygen participates in the formation of an organic superoxide radical, undergoing β-scission to ultimately transform into benzoic acid [71]. In addition to chlorine radicals, various effective HAT agents have been identified in degrading plastics, including graphitic carbon nitride catalysts, singlet oxygen, fluorenone, and bromine radicals [21,66,72,73].

5. Electrocatalysis for plastic upgrading

Electrocatalysis constitutes a sustainable and robust methodology, finding extensive applications across diverse fields. This includes electrocatalytic processes such as hydrogen evolution, oxygen reduction, carbon dioxide reduction and nitrogen fixation [74,75]. The fundamental principle of electrocatalysis involves the utilization of an external potential to facilitate chemical reactions, thereby allowing meticulous control over reaction pathways and intermediates [76,77]. Specifically, the methodology entails oxidation and reduction reactions at two separate electrodes, interconnected through an external electrical circuit and immersed in an electrolyte medium [36]. This distinctive electrocatalytic approach offers several significant advantages, such as precise modulation of operational processes via electrode potentials, employment of recyclable supporting electrolytes to minimize chemical consumption, utilization of green energy sources to mitigate carbon emissions, and the establishment of compact, modular, and on-demand electrochemical reaction systems with potential for real-world applications [78]. However, a considerable research gap and unexplored territory persist regarding the application of electrocatalysis for the upcycling of plastics into high-value chemicals.

Table 2 shows the performance of current research on electrochemical systems for PET upcycling. Recent investigations have identified PET as a prevalent plastic in electrocatalysis-induced valorization to potassium diformate and terephthalic acid [22,79,80]. Fig. 6 shows the conversion pathway of PET in general electrocatalysis system chemicals [80,81]. The process commences with the initial breakdown of PET under alkaline conditions, yielding terephthalic acid and ethylene glycol. The subsequent electro-driven cleavage of the C–C bond in ethylene glycol leads to formate production. The following step is to transfer formic acid into the PET electrolyte, inducing precipitation and regeneration of purified terephthalic acid through filtration. The reaction sequence concludes with the solidification of potassium diformate via concentration and crystallization.

A two-compartment reactor divided by a glass frit was proposed by Myren et al. to electrochemically depolymerize PET plastics in a 50% methanol solution with 0.1 mol/L NaCl as the electrolyte [22]. The system demonstrated an enhanced hydrolysis of PET into terephthalate, achieving a notable yield of 16.9% at a consistent potential of −2.2 V. High current density and high Faradaic efficiency were identified as essential for scale-up application. In a related development, Zhou et al. employed nickel foam supported by various cobalt and nickel phosphides ($CoNi_{0.25}P$) to achieve efficient electrooxidation of PET into formate, exhibiting a remarkable Faraday efficiency above 80% at 1.8 V [80]. Despite its innovative nature, this study acknowledged certain limitations, including the need for optimization of PET conversion rates and exploration of process scalability [88]. Future research should focus on the development of efficient catalysts, optimization of reaction conditions, and the design of scalable reactors to facilitate the practical application of electrocatalytic plastic upcycling [89].

Additionally, other valuable fuels/chemicals have been obtained by depolymerizing PET through combined electrocatalytic oxidation strategies. For instance, the integration of hydrogen evolution reaction with PET electrooxidation achieved over 95% carbonate selectivity and 95% Faraday efficiency [87]. In this system, ethylene glycol was oxidized to glyoxal and then transformed into glyoxylic acid and oxalic acid sequentially and finally break into carbonate. In another study, electrocatalytic up-cycling of plastics with hydrogen evolution reaction in seawater was achieved using a bifunctional Ni_3N/W_5N_4 catalyst with Janus nanostructures [86]. Glycolic acid was also obtained from PET-derived photoelectrochemical oxidation, as demonstrated by Bhattacharjee et al. in a perovskite-based photoelectrochemical device [90]. This represents a potential example for actual applications in plastic upcycling driven by solar energy [36].

Electrocatalysis has further been applied to inert plastic upcycling, such as PE. A recent nitric acid-assisted strategy was developed to convert PE into succinic, glutaric, and adipic acid, subsequently convertible to alkenes through a non-Kolbe decarboxylation reaction [91–94]. Reisner et al. reported a tandem oxidative depolymerization and electrodecarboxylation reaction to produce hydrocarbons (ethylene and propylene) (Fig. 6) [81]. The PE plastic was initially depolymerized into succinic and glutaric acid. These dicarboxylic acids would then transform into biradicals and finally converted into ethylene and propylene via intramolecular recombination. The primary advantage of this process lies in the conversion of plastic waste into easily separable gaseous products. Nevertheless, further enhancements are warranted, particularly in employing milder conditions in the chemical pretreatment phase and reducing CO_2 emissions in the electrochemical step.

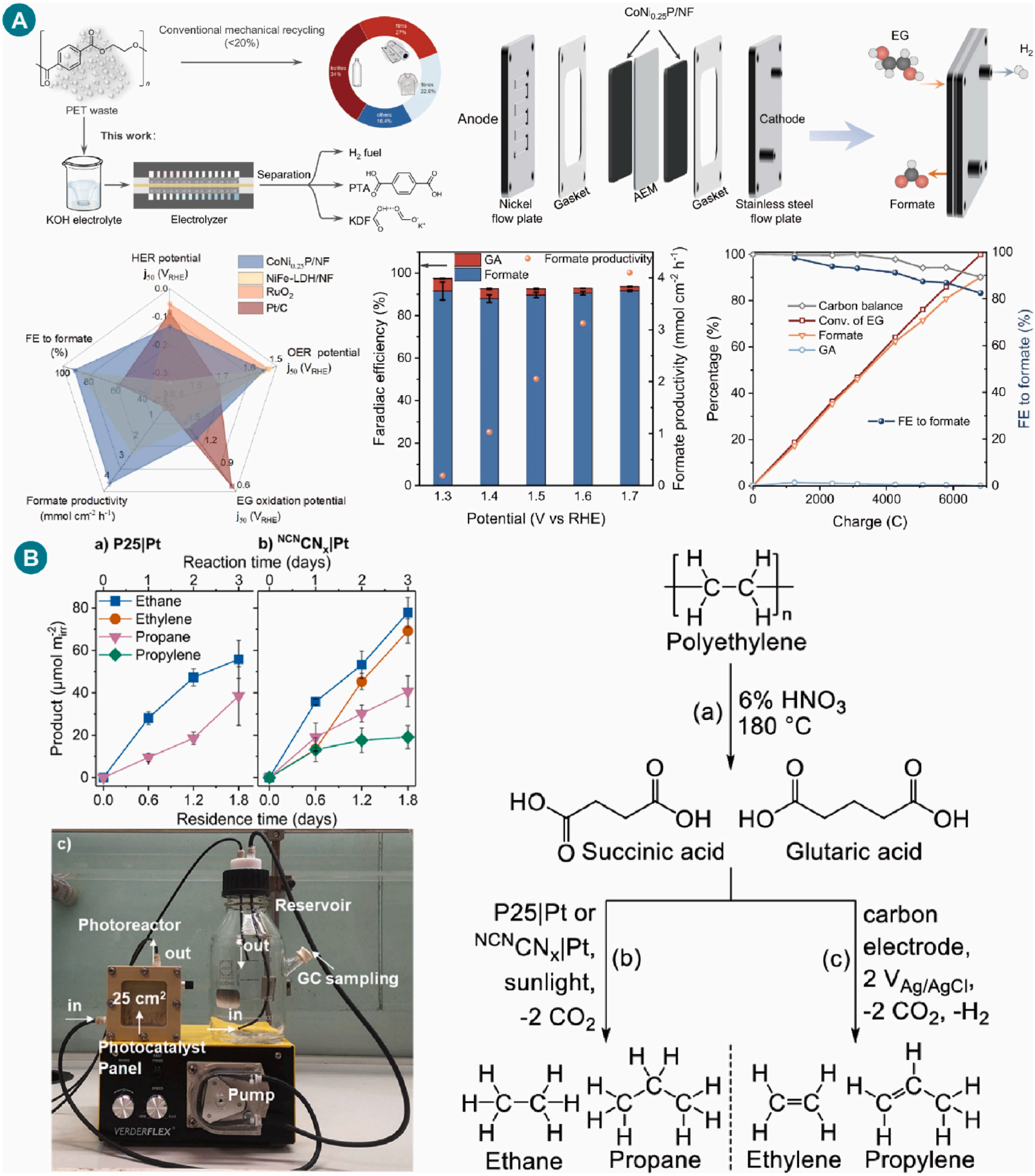

Fig. 6. (A) Conventional pathway for PET recycling with membrane-electrode assembly to produce hydrogen and chemicals [80]. Copyright © 2021, The Authors, Published by Springer Nature (CC-BY-NC 4.0). (B) Illustration of integrated tandem chemical–photo/electrocatalytic processes for PE waste conversion [81]. Copyright © 2021 The Authors. Published by American Chemical Society (CC-BY-NC 4.0).

6. Conclusions and perspectives

The present approach to managing plastic waste, predominantly reliant on landfills and incineration, is deemed unsustainable. These methods contribute to the generation of microplastics through the aging process, integrating them into the environment. Direct treatment of microplastics is considered ineffective due to their minute size, potentially leading to the formation of more toxic nano-plastics. The global plastic waste crisis necessitates the development of innovative and sustainable solutions for managing and valorizing discarded plastics.

Conventional recycling methods, including primary and secondary recycling, result in either secondary pollution or downcycling, limiting recycled plastic to low-quality applications. Therefore, advocating for a proactive approach to recycle plastics into valuable chemicals is crucial for sustainability, replacing current unsustainable practices.

The conventional methods of recycling, including mechanical recycling and incineration, have been complemented by emerging technologies such as chemical recycling, photocatalysis, and electrocatalysis. These novel approaches offer the potential to transform plastic waste into valuable chemicals, fuels, and materials, contributing to a circular economy. Chemical recycling, encompassing pyrolysis, hydrogenolysis, and catalytic processes, has shown promise in depolymerizing plastics into monomers or converting them into higher-value products. Pyrolysis is the most frequently used one, producing liquids (e.g., gasoline) and other products that are very sensitive to the reaction parameters/conditions. However, a low product selectivity is an urgent problem that needs to be solved before wide applications.

Photocatalysis and electrocatalysis represent emerging technologies that leverage renewable energy sources to drive plastic degradation and upcycling. These technologies have experienced an exceptional boost within recent years and advanced to a powerful alternative to traditional thermal recycling technologies. Electrocatalysis offers precise control over reaction pathways and the potential for modular and scalable applications. However, these technologies are still in the nascent stage, and there is plenty of room to improve the selectivity, and scalability. In addition, they always suffer from the relatively low conversion efficiency and the superabundant terminal products of CO_2. Research in this area is still in its infancy, and further exploration is needed to develop efficient catalysts and scalable reactors.

Despite the high transformation efficiency of chemical upcycling from plastic waste to valuable chemicals, urgent challenges remain:

1) Efforts are needed to design catalysts for milder conditions and higher efficiency in pyrolysis and hydrogenolysis, necessitating the development of co-processing technologies to improve product quality.
2) Catalytic technology should expand its application scope, especially to various types of plastics, as current technologies may not be suitable for composite plastic upcycling.
3) Special attention should be given to designing catalytic materials and understanding reaction mechanisms, crucial for enhancing catalytic activity and product selectivity in plastic transformation. The exploitation of *in situ* techniques, though employed in tracing reactive intermediates during mild plastic conversion, requires further refinement.

CRediT authorship contribution statement

Xingdong Shi: Conceptualization, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Zhijie Chen:** Formal analysis, Methodology, Validation, Writing – original draft, Writing – review & editing. **Wei Wei:** Formal analysis, Investigation, Methodology, Writing – original draft, Writing – review & editing. **Bing-Jie Ni:** Conceptualization, Formal analysis, Investigation, Methodology, Project administration, Resources, Supervision, Validation, Writing – original draft, Writing – review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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