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## Electrochemical recovery and regeneration of polyethylene terephthalate materials

Chao Chen,<sup>a</sup> Yi Wang,<sup>a</sup> Jing Tang,<sup>ib</sup>\*<sup>b</sup> Qixuan Zhu,<sup>a</sup> Yu Chu,<sup>a</sup> Jiacheng Zhao,<sup>ib</sup><sup>cd</sup>  
Yusuke Yamauchi,<sup>ib</sup>\*<sup>efg</sup> Guiyin Xu\*<sup>a</sup> and Meifang Zhu<sup>ib</sup><sup>a</sup>

Polyethylene terephthalate (PET) is one of the most widely used synthetic polymer materials, which brings great convenience to social life and industrial applications. However, the extensive use of PET and the lack of effective recycling have led to environmental pollution caused by plastic waste. Unlike traditional recycling methods that rely on energy-intensive and corrosive conditions (e.g., high temperature and pressure), electrochemical catalytic technology operates under mild conditions and offers environmental compatibility, and thus has emerged as a promising alternative. The core of this method lies in its catalytic mechanism and selective conversion. This review focuses specifically on the catalytic mechanisms and reaction pathways involved in the electrochemical recycling of PET. It begins by systematically comparing the fundamental differences between electrochemical and conventional methods, with an emphasis on how electrocatalysis enables selective bond cleavage and transformation under ambient conditions. Then, the electrocatalytic mechanism and the detailed reaction process, particularly at the electrode–electrolyte interface, are thoroughly elucidated, including how active species (e.g., hydroxyl radicals or other reactive oxygen species) are generated *in situ* and participate in the stepwise degradation of PET macromolecules. Finally, key electrochemical technologies are systematically summarized and future prospects are discussed, aiming to offer valuable insights for innovating and scaling up industrial-grade PET waste recycling processes based on a deep understanding of the electrocatalytic principles.

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### Wider impact

Although the wide application of polyethylene terephthalate (PET) has brought convenience, its insufficient recovery has led to environmental pollution and other problems. Electrochemical catalysis provides a new approach for the efficient recovery of PET and the regeneration of high value-added products due to its mild, efficient and environmentally friendly characteristics. This review focuses on the progress of electrochemical catalytic recycling of PET materials, including the mechanism of depolymerization under mild conditions, selective synthesis strategies of high value-added products (such as formic acid and glycolic acid), and catalyst design (such as noble metal and transition metal-based catalysts). This field has a broad interdisciplinary interest in facing the global plastic pollution challenge and providing sustainable solutions with low energy consumption and low pollution. Its potential for coupling with renewable energy further highlights the cross-value benefits of environmental and energy science. In the future, this field will develop towards the direction of developing efficient catalytic systems, optimizing process integration and promoting industrial applications. By clarifying the structure–activity relationship between ‘reaction path–product selectivity–material selection’, this review provides theoretical support for the design of new materials and promotes the transformation of materials science to a green and intelligent paradigm oriented by circular economy.

<sup>a</sup> State Key Laboratory for Advanced Fiber Materials, College of Materials Science and Engineering, Donghua University, Shanghai 201620, China. E-mail: xuguiyin@dhu.edu.cn

<sup>b</sup> State Key Laboratory of Petroleum Molecular and Process Engineering, Shanghai Key Laboratory of Green Chemistry and Chemical Processes, School of Chemistry and Molecular Engineering, East China Normal University, Shanghai 200062, China. E-mail: jingtang@chem.ecnu.edu.cn

<sup>c</sup> Xin Feng Ming Group Huzhou Zhongshi Technology Co., Ltd, Huzhou 313021, China

<sup>d</sup> Department of Materials Engineering, Huzhou University, Huzhou 313000, China

<sup>e</sup> Department of Materials Process Engineering, Graduate School of Engineering, Nagoya University, Nagoya 464-8601, Japan. E-mail: y.yamauchi@uq.edu.au

<sup>f</sup> Australian Institute for Bioengineering and Nanotechnology (AIBN) and School of Chemical Engineering, The University of Queensland, Brisbane, Queensland 4000, Australia

<sup>g</sup> Department of Convergent Biotechnology and Advanced Materials Science, Kyung Hee University, 1732 Deogyong-daero, Giheung-gu, Yongin-si, Gyeonggi-do, Seoul 17104, South Korea

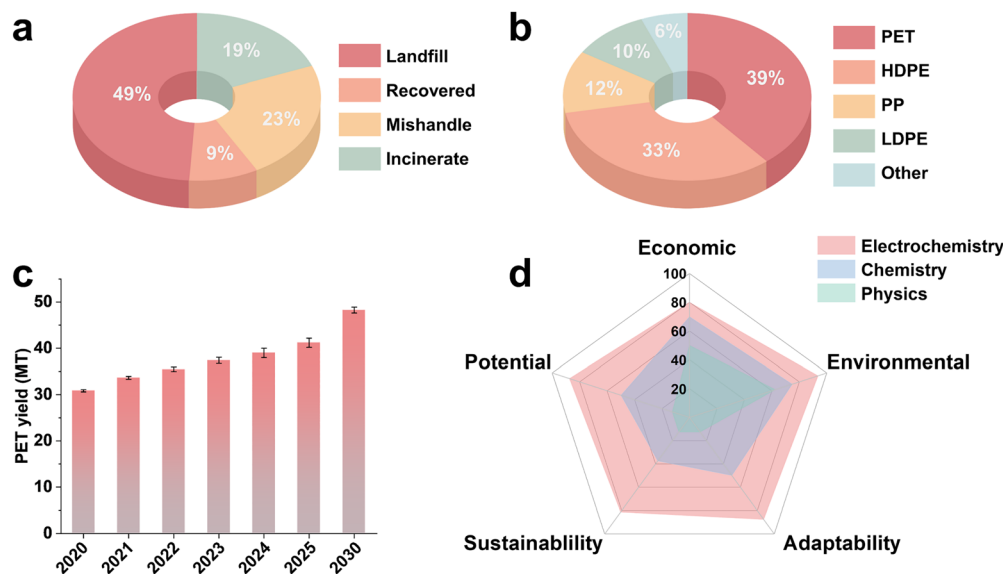
# 1. Introduction

Plastics are widely used in packaging, agriculture, construction, home, and other fields due to their excellent physical properties such as high transparency, good barrier properties, and light weight.<sup>1–3</sup> Among plastics, PET plastic is particularly popular. PET is a thermoplastic polyester with excellent mechanical properties, chemical stability, and thermal stability.<sup>4</sup> In the packaging field, PET materials are often made into various beverage bottles. Its transparent appearance can clearly show the color of the beverage in the bottle and attract the attention of consumers. Moreover, it has a good barrier to gas and water, which can effectively prolong the shelf life of beverages. In agriculture, the PET film can be used to build a greenhouse environment, which allows sufficient sunlight to pass through and creates a warm and suitable growth environment for crops.<sup>5</sup> At the same time, its durability also ensures that the greenhouse can be used stably under various climatic conditions. In the construction industry, PET plastic can be processed into decorative plates, which not only have a beautiful appearance but also have fire resistance, moisture resistance, and other properties, adding a lot of practical value to the building. With the continuous development of science and technology, the PET material is still expanding its application range. In the future, it is expected to show unique advantages in more fields and bring more convenience and change to people's lives.<sup>6,7</sup>

Despite the benefits of PET plastics, the environmental pollution caused by their large-scale use is becoming more and more serious.<sup>9</sup> At present, the limited recycling rate means a large amount of plastic waste is still being landfilled (Fig. 1). More seriously, PET plastics in landfills will continue to release

microplastic particles (particle size  $\leq 5$  mm) during natural degradation.<sup>10,11</sup> These particles can invade the global water system and food chain, which will eventually pose a threat to human health.<sup>12</sup> The traditional physical crushing recovery method has strict requirements on the purity of PET plastics. Mixing other materials or ink-contaminated items will greatly reduce the quality of the recovered materials, resulting in many recyclable materials being forced to degrade or discarded.<sup>13</sup> In addition, incineration will release a large amount of greenhouse gases such as carbon dioxide, further exacerbating climate change.<sup>14,15</sup>

In order to cope with this dilemma, scientists began to explore new PET plastic recycling technologies (Fig. 2a). Among them, the chemical recovery method has gradually emerged.<sup>16–18</sup> It can decompose PET plastics into small molecular monomers, and then re-synthesize high-quality plastics and other products (Fig. 2b). The requirements for the purity of raw materials are relatively low, which greatly improves the recovery efficiency and the quality of recycled materials (Fig. 3a).<sup>19,20</sup> The key path of PET chemical recovery is achieving efficient depolymerization through a controllable bond-breaking-reconstruction mechanism. One strategy is that the transesterification reaction has become one of the core strategies due to its good molecular selectivity. Specifically, the ester bond ( $-\text{COO}-$ ) in the PET molecular chain breaks under the action of specific reagents (such as acid, diol, methanol, *etc.*) and catalysts, and forms new ester bonds or monomers with the depolymerization agent (Fig. 3b).<sup>21</sup> This method is thermodynamically highly feasible ( $\Delta G < 0$ ).<sup>22</sup> Taking acid hydrolysis as an example, PET undergoes a hydrolysis reaction under the catalysis of inorganic acids (such as sulfuric acid or nitric acid).<sup>23</sup> The core mechanism is that the proton ( $\text{H}^+$ ) provided by the acid preferentially attacks



**Fig. 1** Statistics on the development of plastic recycling. (a) Current global plastic processing methods. (b) Renewable plastic usage. (c) Global PET plastic usage from 2020 to 2024 and production projections for 2025 and 2030. (d) Comparison of economic, environmental, applicability, sustainability and potential of electrochemical, physical, and chemical recovery. (Notes: Estimates are based on data from Europe, the United States, and China for the period 2020–2030.<sup>8</sup>)

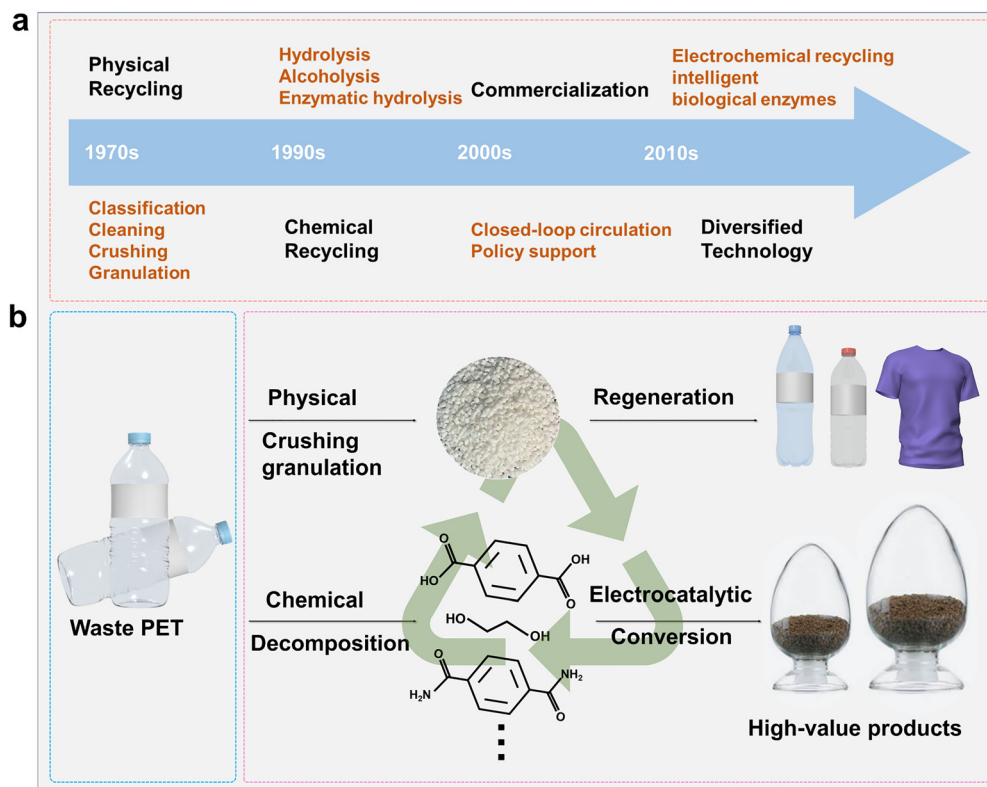
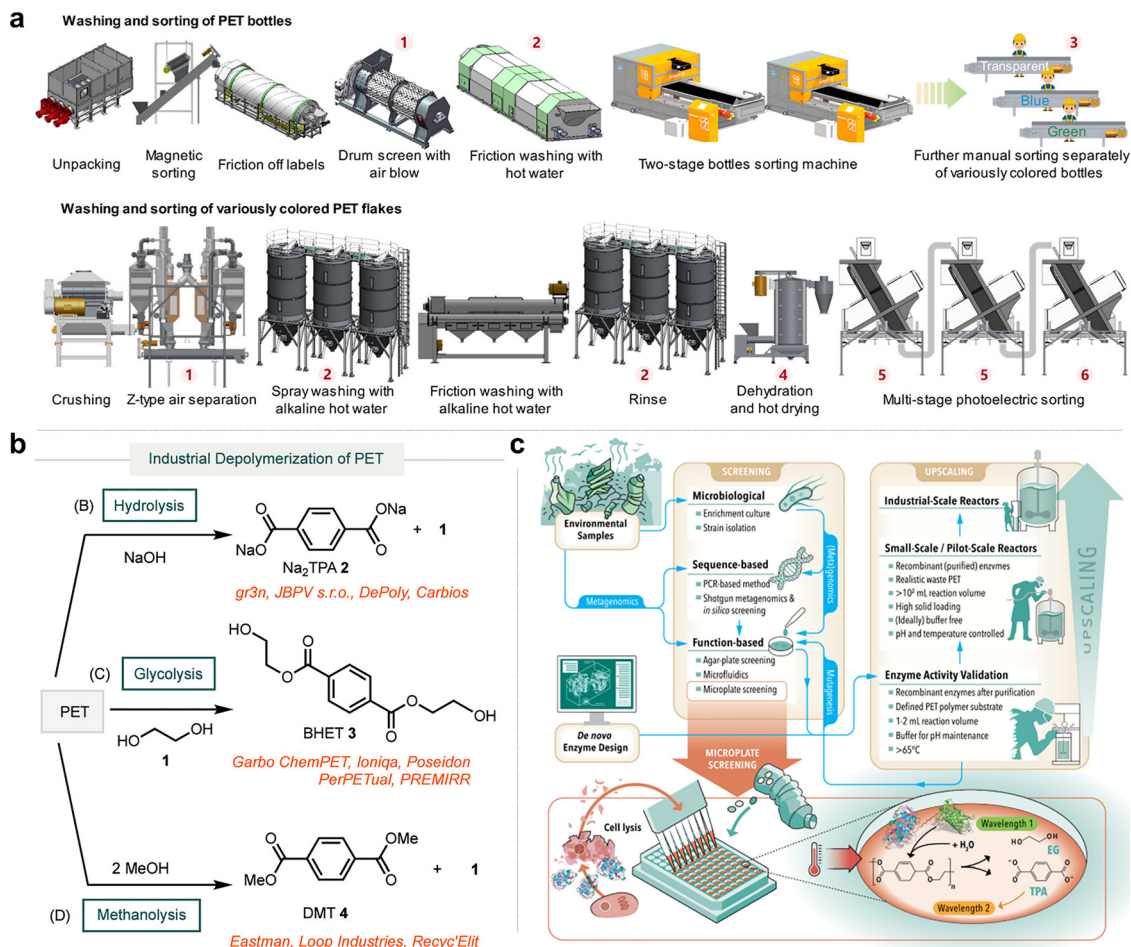


Fig. 2 The development history of PET plastic recycling. (a) The schematic diagram of the development process of PET recycling technology. (b) Physical and chemical methods for PET recycling.

the carbonyl oxygen atom (rather than the hydroxyl oxygen) in the ester bond to form a protonated carbonyl intermediate. This process significantly enhances the electrophilicity of the carbonyl carbon atom, making it vulnerable to the attack of the nucleophilic oxygen atom in the water molecule, eventually leading to the cleavage of the ester bond.<sup>24</sup> After the protonation-induced nucleophilic hydrolysis reaction, the ester bond in the PET chain is broken to form the terephthalic acid (TPA) monomer and ethylene glycol (EG) monomer.<sup>25</sup> It is worth noting that the process does not require high purity of raw materials, and can efficiently convert waste mixed with up to 10% non-PET impurities into TPA with a purity of 99% or more, which fully meets the strict requirements for the synthesis of food-grade recycled PET (rPET).<sup>26</sup>

However, the chemical recovery of the PET process has high energy consumption and high cost. Its depolymerization reaction is complex, resulting in complex product (such as monomers or pyrolysis oil) composition, difficult purification and high cost. At the same time, there are obvious problems such as catalyst deactivation, the challenges of by-product treatment, and disputes surrounding greenhouse gas emissions.<sup>27</sup> In addition, the popularity of the chemical recycling method is low, the relevant industry standards and norms are not perfect, and the awareness and acceptance of chemically recycled plastics in the market also need to be improved, which restricts the large-scale application and promotion of the chemical recycling method to a certain extent. Moreover, as an emerging

plastic recycling technology, the bio-enzyme recycling method is gradually receiving attention, which has high specificity and catalytic activity, and can degrade PET plastics under mild conditions.<sup>28–30</sup> Enzymatic hydrolysis technology relies on the stereospecific catalysis of PET hydrolases (such as cutinase,<sup>31</sup> esterase,<sup>32</sup> etc.). The Ser-His-Asp triplet of the active center of the enzyme selectively breaks the ester bond through a nucleophilic attack mechanism.<sup>33</sup> Compared with the chemical recovery method, the biological enzyme recovery method does not require complex chemical reactions and expensive professional equipment, which greatly reduces the initial investment cost. Moreover, biological enzymes are usually naturally occurring biomolecules, and the risk of environmental pollution during use is minimal, which can effectively avoid the harm caused by chemical reagents (Fig. 3c).<sup>29</sup> However, the biological enzyme recovery method is not perfect. At present, the types of high-efficiency bio-enzymes suitable for PET plastic degradation are still relatively limited. Screening and cultivating high-activity and high-stability enzymes is a complex and time-consuming process.<sup>34</sup> At the same time, the catalytic efficiency of biological enzymes is also greatly affected by environmental factors such as temperature and pH in practical applications. It is necessary to accurately control the reaction conditions to achieve the desired degradation effect. Moreover, the large-scale industrial production technology of biological enzyme recovery is not mature enough, and the transformation from laboratory research to large-scale production faces many technical problems (Table 1).<sup>35</sup>



**Fig. 3** Different methods of recycling PET waste plastics. (a) By combining the physical purification process with photoelectric sorting, PET plastic is recovered from waste beverage containers. Reproduced with permission.<sup>13</sup> Copyright 2024, American Chemical Society. (b) Recycling waste plastics by a chemical method. Reproduced from ref. 36. Published by American Chemical Society under a CC BY-NC-ND 4.0 license. (c) Schematic workflow for discovering, engineering, and finally applying novel PET hydrolases in industrial-scale waste plastic recycling. Reproduced with permission.<sup>28</sup> Copyright 2025. Springer Nature.

**Table 1** Comparison of different traditional recycling methods of waste PET

Recovery method	Principle	Merit	Defect
Physical	It is used to produce low-end plastic products by cleaning, crushing, and melting to form pellets (rPET)	Mature technology; low energy consumption; fast recovery	Multiple recycling leads to performance degradation; degradation cycle, applicable to a single scene
Chemical	Decomposed into monomers (TPA, EG, etc.) by chemical reactions such as hydrolysis, alcoholysis, or ammonolysis, and then repolymerized into virgin PET	The product is pure and can be regenerated into food-grade PET, the method can remove contaminants and impurities; suitable for high-end plastic fibers, etc.	High energy consumption; the cost is higher; there may be by-product pollution
Biological	Enzymes (such as cutinase and esterase) are used to selectively cleave the ester bond of PET to generate TPA and EG	Green environmental protection; high selectivity	Low efficiency; the cost is higher; small-scale processing
Other	Anaerobic pyrolysis of PET to produce oil and gas (such as benzene and ethylene) or carbon black for fuel or chemical raw materials	Easy handling; producing oil and gas can partially replace fossil fuels	High pollution; the composition of the product is complex; carbon emissions are higher

Electrochemical recycling of PET materials provides an effective solution, which can decompose and convert waste PET monomer materials under mild conditions through specific electrochemical

processes.<sup>37</sup> In this process, the electrode material plays a key role, and a suitable electrode can efficiently initiate the breakage of the PET molecular chain. Compared with traditional

recycling methods, electrochemical recycling has significant advantages of low energy consumption and low pollution.<sup>38</sup> It can precisely control the reaction conditions and the formation of products. It can not only decompose PET materials into monomers, but also further convert these monomers into high-value-added chemicals (organic acids, alcohols, olefins, *etc.*), realizing the recycling and value enhancement of plastic waste.<sup>39,40</sup> Moreover, this recycling method can also be combined with renewable energy sources such as solar cells, to provide green electricity, thereby further reducing its impact on the environment.<sup>41,42</sup> With the deepening of research, electrochemical recycling of PET materials is expected to become the mainstream technology to solve the problem of PET waste, and make an important contribution to the recycling and sustainable development of resources.

This review provides a systematic examination of the basic principles of electrochemical PET recycling, going beyond simple advantage comparisons. A deeper understanding of the mechanism of the electrode interface reaction is the key to unlocking selectivity and energy-saving pathways, thus driving this technology towards the circular economy of plastics. Firstly, how the electrocatalytic materials promote the decomposition and conversion of waste PET materials under mild conditions in a specific electrochemical process, and the efficiency of different types of electrocatalytic materials in breaking PET molecular chains are introduced in detail. Compared with the traditional recovery methods, the significant advantages of electrochemical recovery, including low energy consumption and low pollution, are analyzed in depth. The specific mechanism of precise control of reaction conditions and product formation is carefully discussed. At the same time, the specific ways and technical difficulties of decomposing PET materials into monomers and further converting them into high-value-added chemicals are studied. In addition, the feasibility and practical application of the combination of this recovery method and renewable energy are expounded, and the specific effect of this combination on reducing environmental impact is analyzed. Finally, with the deepening of research, the electrochemical recycling of PET materials has become a mainstream technology to solve the problem of PET waste. Moreover, in the later stage, there have been specific directions and potential challenges for electrochemical recycling of PET, making significant contributions to resource recovery and sustainable development. It has also provided comprehensive and in-depth references for research and practice in related fields.

## 2. Electrochemical recovery mechanism

### 2.1. Structural characteristics of PET

The core of electrochemical recycling of PET is to achieve efficient and selective conversion of PET into depolymerization monomers at the electrode interface, and understanding the structural characteristics of PET itself is the basis for analyzing its electrochemical degradation path. PET is primarily synthesized

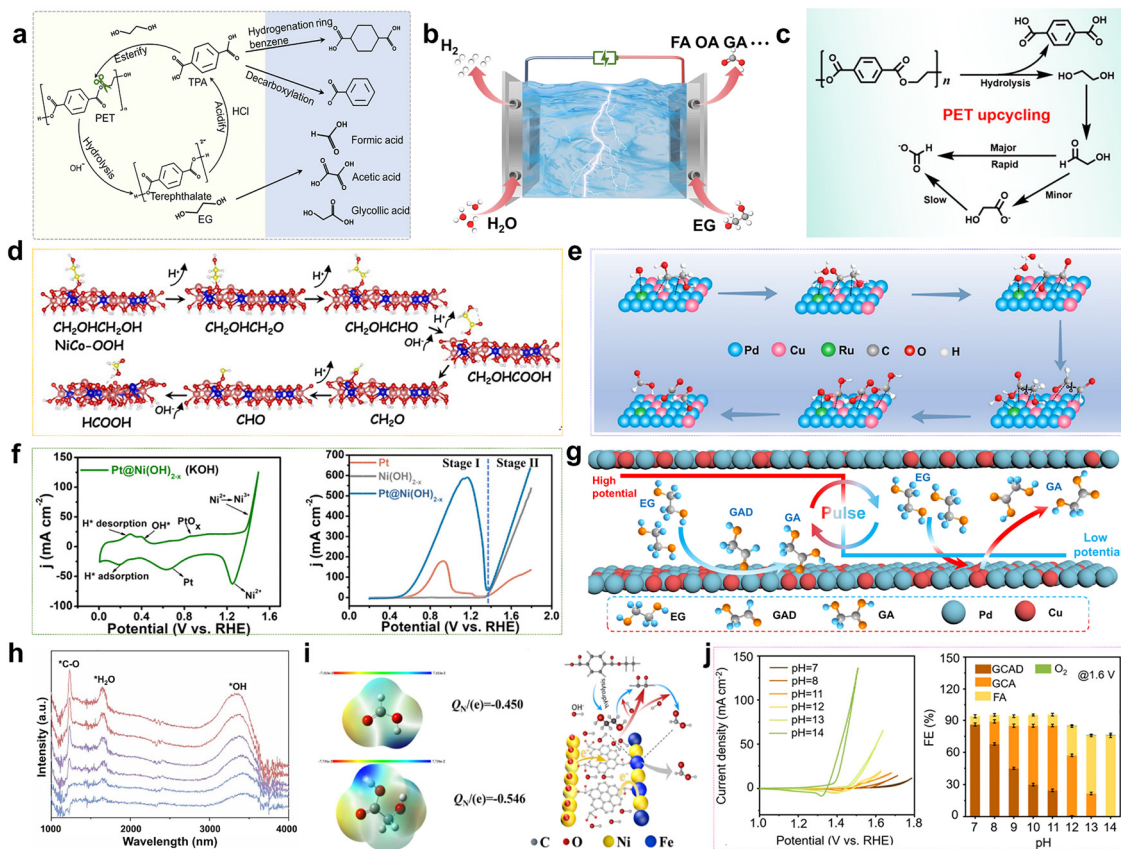
*via* two routes: direct esterification, where terephthalic acid (TPA) reacts directly with ethylene glycol (EG), and transesterification, where dimethyl terephthalate (DMT) first reacts with EG to form bis(2-hydroxyethyl) terephthalate (BHET), which is then polycondensed into PET.<sup>43</sup> In industrial production, the direct esterification of TPA and EG is a more commonly used method. As a polyester material, the repeating unit of PET is composed of the rigid aromatic ring of terephthalic acid and the flexible methylene chain ( $-\text{CH}_2-\text{CH}_2-$ ) of ethylene glycol alternately connected by ester bonds, so that the molecular chain has a highly regular symmetrical structure.<sup>11,44</sup> Due to the synergistic effect of the regular benzene ring plane structure and C–O bond in its molecular chain, it has good mechanical properties, high strength, and hardness. In practical applications, this feature makes it an ideal material for manufacturing beverage bottles, fibers, and other products. The presence of C–O also makes it stable in a weak acid/neutral environment, but it is prone to hydrolysis in an alkaline environment (such as NaOH).<sup>45</sup> This hydrolysis characteristic limits its application under some alkaline conditions, but also provides ideas for its recycling. PET can be decomposed into monomers through alkaline hydrolysis, and then these monomers can be reused to synthesize PET after purification, and realize resource recycling. In today's era of green sustainable development, this recyclable feature makes PET stand out among many materials.

### 2.2. Mechanism of the electrochemical recovery process

Taking alkaline hydrolysis as an example, the process of electrocatalytic regeneration of formate from waste PET plastics mainly includes three steps: (1) PET is hydrolyzed into terephthalate ( $\text{K}_2\text{TPA}$ ) and ethylene glycol monomers in a KOH electrolyte ( $\text{PET} + 2\text{OH}^- \rightarrow \text{TPA}^{2-} + \text{EG} + \text{H}_2\text{O} + \text{e}^-$ ), (2) ethylene glycol in the PET hydrolysate is oxidized at the anode, selectively breaking the C–C bond and generating formate ( $\text{HCOO}^-$ ), while water is reduced at the cathode to produce hydrogen, and (3) terephthalic acid with high purity can be obtained after acidification (Fig. 4a).<sup>46,47</sup> The terephthalic acid precipitate is obtained by filtration, and formate and hydrogen are collected from the electrolyte and gas product, respectively (Fig. 4b). Terephthalic acid can be used as an important chemical raw material to synthesize polyester materials.<sup>48</sup> Formate can be used in many reactions in industrial production, such as leather tanning, printing, dyeing, *etc.*<sup>49</sup> Hydrogen is a clean energy source that can be used for fuel cell power generation or as a reducing agent for chemical production.<sup>50</sup> The entire electrocatalytic waste PET plastic upgrading and recycling process not only effectively recycles waste PET plastic, reducing pollution from plastic waste, but also generates significant economic benefits by producing high-value-added products, providing a sustainable and efficient solution to the problem of plastic waste.

#### 2.2.1. Chemical dissociation and oxidation mechanism.

The electrochemical recycling of PET involves a coupled process of chemical depolymerization and electrochemical oxidation. Firstly, the PET polymer chain is selectively depolymerized through alkaline hydrolysis, breaking the ester bond to obtain purified TPA and EG monomers (Fig. 4c). In an alkaline



**Fig. 4** Electrochemical depolymerization and reaction mechanism. (a) Schematic diagram of PET recycling. (b) Schematic diagram of ion transport and cathode–anode catalytic reaction in an electrocatalytic process. (c) Schematic diagram of electrochemical recovery of regenerated formate from PET. Reproduced with permission.<sup>53</sup> Copyright 2023, American Chemical Society. (d) The mechanism diagram of EG electrochemical conversion to formic acid. Reproduced from ref. 51. Published by Royal Society of Chemistry under a CC BY-NC-ND 4.0 license. (e) Trimetallic mesoporous PdCuRu for efficient formate electrosynthesis from polyethylene terephthalate recycling. Reproduced with permission.<sup>55</sup> Copyright 2025, Springer Nature. (f) CV curve of Pt@Ni(OH)<sub>2-x</sub> in 1 M KOH, and LSV curves of Ni(OH)<sub>2-x</sub>, Pt, and Pt@Ni(OH)<sub>2-x</sub> in 1 M KOH with 0.2 M EG. Reproduced with permission.<sup>53</sup> Copyright 2025, American Chemical Society. (g) Electrocatalytic mechanism of selective electrocatalytic synthesis of GA by PETH. Reproduced with permission.<sup>60</sup> Copyright 2025, Springer Nature. (h) *In situ* ATR-FTIR spectra of Mn/CoOOH of EGOR at different potentials. Reproduced with permission.<sup>64</sup> Copyright 2025, Elsevier. (i) The electrostatic potential calculation of FA and GA, and the schematic diagram of the EG oxidation mechanism on heterostructures. Reproduced with permission.<sup>65</sup> Copyright 2025, Elsevier. (j) The CV curves of the  $\beta$ -NiOOH electrode with 0.1 M EG in the range of pH 7–14 and the FEs of EG oxidation products on  $\beta$ -NiOOH at 1.6 V vs. RHE at different pH values. Reproduced with permission.<sup>66</sup> Copyright 2025, American Chemical Society.

environment, OH<sup>-</sup> with strong nucleophilic ability will attack the ester bond in the PET molecular chain to form an intermediate (the carbonyl carbon (C=O) in the ester bond carries a partial positive charge, making it susceptible to nucleophilic attack), then the intermediate is rapidly reformed, and finally the monomer is formed.<sup>33</sup> Subsequently, the EG monomer undergoes electrochemical oxidation (EGOR) at the anode (Fig. 4d). Crucially, the applied electrode potential determines the reaction path and the final product distribution, which can be adjusted to produce valuable chemicals such as formic acid, glycolic acid, ethanolamine, *etc.*<sup>51</sup>

When the EG molecule is adsorbed on the anode surface, the electron is removed, and the oxidation reaction is initiated under a sufficiently positive potential drive. This reaction proceeds through two possible pathways: C–C cleavage (named C1) and C–C preservation (named C2). The oxidation reaction usually starts from the dehydrogenation of the terminal –CH<sub>2</sub>OH group

to form glycolaldehyde (HOCH<sub>2</sub>–CHO), forming an unstable intermediate. Subsequently, glycolaldehyde may be further oxidized by the adsorbed substance. Glycolaldehyde undergoes adsorbate-induced oxidation (likely nucleophilic attack by hydroxyl species or water) at the –CHO group. This cleaves the C–C bond, producing formate and formaldehyde (which can be further oxidized to CO<sub>2</sub>).<sup>52,53</sup> Glycolaldehyde can be further oxidized, preserving the C–C bond. This typically involves oxidation of the aldehyde group (–CHO) to a carboxylic acid (–COOH), yielding glycolic acid, especially under strong oxidizing conditions or with specific catalysts (Fig. 4e). Moreover, in the ammonia-containing electrolyte (NH<sub>3</sub>/NH<sub>4</sub><sup>+</sup>), ammonia can carry out nucleophilic attack on the carbonyl carbon of the adsorbed glycolaldehyde, instead of direct oxidation, rapid hydrolysis, or *in situ* reduction to form ethanolamine.<sup>54,55</sup> The core mechanism for the electrochemical upgrading of PET lies in the nucleophilic addition of ammonia (NH<sub>3</sub>) or its derivatives to glycolaldehyde

(HOCH<sub>2</sub>-CHO), a key intermediate of ethylene glycol (EG) oxidation, selectively yielding high-value ethanolamine. This pathway, distinct from traditional complete oxidation, demonstrates how electrolyte engineering can steer product selectivity. The process begins with the anodic dehydrogenation of EG adsorbed on the catalyst surface, forming glycolaldehyde.<sup>56</sup> In the ammonia electrolyte (e.g., NH<sub>4</sub><sup>+</sup>/NH<sub>3</sub> buffer), NH<sub>3</sub> acts as a nucleophile, with its lone pair attacking the electrophilic carbonyl carbon of glycolaldehyde. This results in a shift of the  $\pi$ -electron cloud and forms a zwitterionic tetrahedral intermediate.<sup>57</sup> Being highly unstable, this intermediate rapidly undergoes rearrangement. Within the electrochemical environment, it favors reductive amination: the cleavage of the O-H bond is accompanied by C-N bond strengthening, directly eliminating a water molecule to form an iminium cation. Finally, this iminium intermediate is rapidly reduced to the stable product, ethanolamine (HOCH<sub>2</sub>-CH<sub>2</sub>NH<sub>2</sub>), either *via* reduction at the cathode or *via* a surface hydrogen atom transfer mechanism.

The different reaction path shows a dual competitive mechanism (Fig. 4f and g).<sup>53,58</sup> At a lower applied potential (<1.2 V *vs.* RHE), EG electrooxidation can lead to selective glycolate or oxalate formation. However, the reaction kinetics are limited by the limited supply kinetics of the active hydrogen species (H<sup>+</sup>/H), which inevitably leads to competitive hydrogen evolution reaction (HER, the reaction wherein hydrogen molecules are generated by an electrochemical reduction process) resulting in a partial loss of Faraday efficiency.<sup>59</sup> In contrast, a strong oxidation interface is established at a higher applied potential (>1.6 V *vs.* RHE) to drive the oxygen evolution reaction (OER). This process *in situ* produces highly active hydroxyl radicals (\*OH) as OER intermediates.<sup>60</sup> These free radicals promote the non-selective oxidative degradation of EG.<sup>61</sup> The controllable formation of the product is inhibited, and the C-C bond is excessively dissociated to form formate or eventually CO<sub>2</sub>.<sup>62</sup>

Therefore, accurate optimization of key parameters, especially stabilizing the reaction within an optimal potential window, is crucial. The anodic process is carried out by adsorbing intermediates, and the final product is determined by the applied potential and other factors. The complex dynamics of this process involving charge transfer, mass transfer and surface reactivity highlights the challenges and opportunities of optimizing this sustainable recovery method.

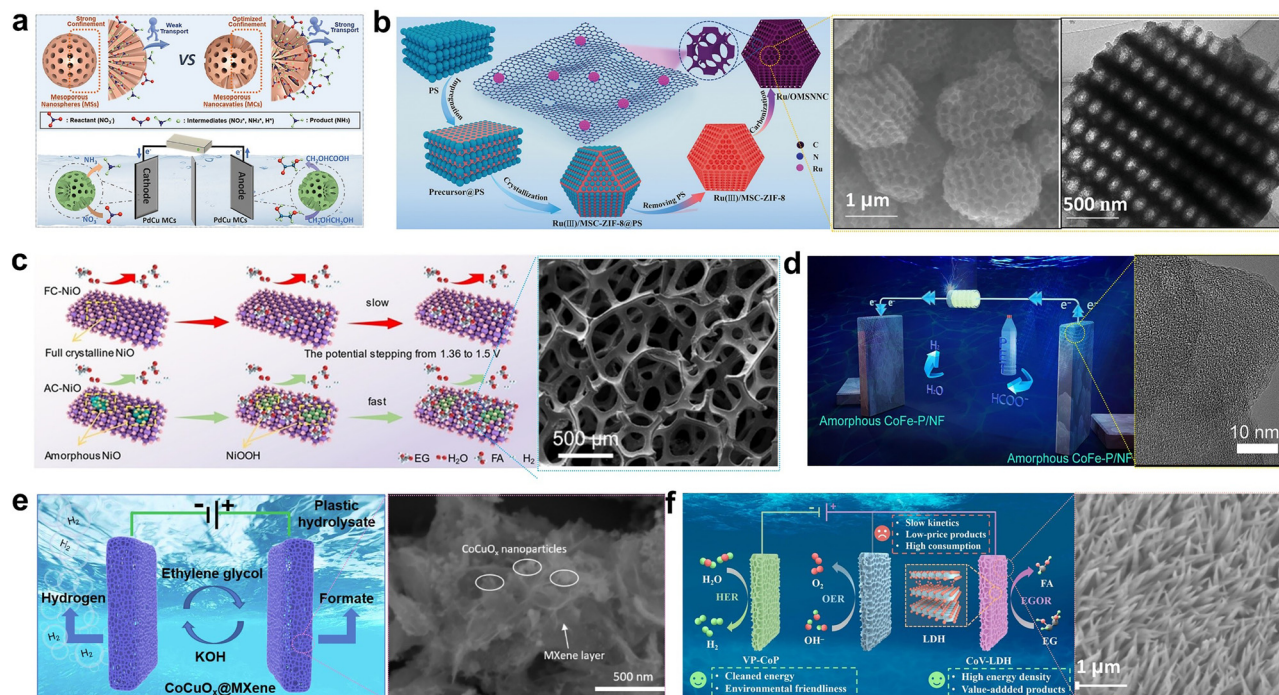
**2.2.2. Interfacial reaction kinetics.** The core of electrochemical recovery of PET is the electrooxidation of ethylene glycol at the electrode-electrolyte interface, which is a complex kinetic process involving adsorption, activation, bond cleavage, and subsequent oxidation pathways, resulting in desired products and various intermediates/by-products.<sup>67</sup> The dissolved EG molecules are first adsorbed on the surface of the electrode catalyst. As the hydrophilic hydroxyl groups of EG interact strongly with the aqueous solution and ions through hydrogen bonds and solvation, under the action of an external electric field, the adsorbed EG molecules show a distinct polarization phenomenon. Specifically, the C-H bonds adjacent to the hydroxyl groups are activated and weakened, making them easy to be deprotonated, and the O-H bonds are polarized to

promote nucleophilic attack or dissociation. This electric field-driven bond weakening directly reduces the activation barrier of the initial oxidative dehydrogenation step (Fig. 4h).<sup>64,68</sup> With the increase of anodic potential, the oxidative cleavage of C-H or O-H bonds initiates the reaction, generating key intermediates and releasing protons and electrons. The specific conditions of intermediates and final products mainly depend on the oxidation mechanism and the sensitivity of these intermediates to further electrochemical oxidation.<sup>69,70</sup> In addition, the competitive oxidation pathway is inherent in electrooxidation kinetics. Ideal partial oxidation and not ideal complete oxidation to produce carbon dioxide occur in parallel. The kinetic competition between selective oxidation and peroxidation pathways determines the selectivity of the product (Fig. 4i).<sup>65,66</sup> Electrolyte composition (ions, pH) plays an important role in interfacial kinetics. Electrolyte ions can be directly involved (e.g., OH<sup>-</sup> acts as a nucleophile in an alkaline medium), regulating the bilayer structure, affecting the local concentration of reactants/intermediates, or interacting with intermediates (e.g., forming stable complexes, especially with metal ions). pH has a great influence on the protonation state of the intermediate product and the thermodynamic benefits of different reaction steps. The mass transfer conditions determine the supply of EG to the surface and the removal of the product, affecting the concentration polarization and the total reaction rate (Fig. 4j).<sup>66,71</sup> As the reaction proceeds, the accumulation of intermediates or by-products on the electrode surface will dynamically change the surface coverage and may change the active sites, thereby affecting the reaction kinetics.

Therefore, the kinetics of PET electrocatalytic depolymerization and by-product formation are governed by potential-driven adsorption and polarization, followed by a kinetically limiting C-O cleavage step. The distribution of products (monomers *vs.* by-products) is determined by the intricate balance of the kinetic rates and selectivity of these parallel and sequential pathways, influenced critically by applied potential, electrode material/catalyst, electrolyte composition (ions, pH), and mass transfer conditions.

### 3. Key technology and challenges of electrochemical recovery

Due to the inertness of PET plastics and their limited solubility in aqueous solutions, they are rarely directly involved in the redox reactions that occur on the electrode surface. Electrolytic recovery of PET is usually carried out using a paired electrolysis process, which consists of two mutually coupled half-reactions. At the anodic side of the electrochemical system, the hydrolyzed monomers undergo an oxidation reaction to produce high-value-added products, with ethanoic acid being the most common product produced from the oxidation of ethylene glycol. On the cathode, the electrons released from the anode are continuously transferred to the cathode and drive the reduction reactions on the cathode side, such as hydrogen evolution reaction (HER), carbon dioxide (CO<sub>2</sub>) reduction, or nitrate reduction (NO<sub>3</sub><sup>-</sup>RR) (Fig. 5a).<sup>56,72</sup> Increasing the yield of



**Fig. 5** Schematics of electrocatalytic upgrading of PET (the virtual wireframes are SEM or TEM images to characterize the morphology). (a) Top: Schematic illustration of enzymatic NO<sub>3</sub><sup>-</sup> to NH<sub>3</sub> electrocatalysis over PdCu MSs and MCs. Bottom: Illustration of the two-electrode coupling system for cathode NO<sub>3</sub><sup>-</sup> to NH<sub>3</sub> and anode PETH to GA electrocatalysis by PdCu MC nanozymes. Reproduced with permission.<sup>77</sup> Copyright 2024, Wiley-VCH. (b) A schematic diagram of the preparation of a Ru/OMSNNC micro-nano reactor for recycling PET plastics. Reproduced with permission.<sup>79</sup> Copyright 2021, Wiley-VCH. (c) Amorphous NiO synthesized by ultra-low temperature oxidation offers fast reconstruction and enhanced active sites for industrial-scale electrocatalytic recovery of PET. Reproduced with permission.<sup>81</sup> Copyright 2024, Wiley-VCH. (d) The self-supporting amorphous cobalt-iron phosphide electrode simultaneously performs electrocatalytic hydrogen evolution and polyethylene terephthalate plastic reforming. Reproduced with permission.<sup>83</sup> Copyright 2024, Elsevier. (e) Electrochemical conversion of plastic hydrolysate to hydrogen and formate by using a CoCuO<sub>x</sub>@MXene catalyst. Reproduced with permission.<sup>84</sup> Copyright 2025, Elsevier. (f) Schematic illustration of hybrid electrolysis for PET plastic upgrading and hydrogen production. Reproduced with permission.<sup>70</sup> Copyright 2025, Elsevier.

the target product is the core objective of the PET electrochemical recovery process. Optimization of key technologies in the electrolysis system can effectively facilitate product generation, including optimization of the electrocatalyst to reduce the reaction activation energy, enhancement of mass transfer efficiency through reactant pretreatment, reaction interface modulation, and so on.

### 3.1. Electrocatalyst design

The electrocatalytic upgrading of PET plastic monomers mainly depends on the anodic oxidation reaction. In the process of anodic oxidation, selecting an appropriate catalyst is very important, as efficient electrocatalysts can significantly reduce the energy requirement for electrolysis reactions.<sup>39</sup> Both precious metal-based and transition metal-based catalysts have been widely employed for the conversion of EG to glycolic acid (GA)<sup>47</sup> and formic acid (FA).<sup>45,73</sup> However, the anodic oxidation reaction also faces challenges of catalyst deactivation during long-term operation. While carbon deposition and poisoning are common causes of performance decay, other critical mechanisms contribute significantly. These include sintering (growth of catalyst nanoparticles leading to surface area loss), phase transformation (change in the active crystal structure

under high anodic potentials), metal leaching (dissolution of active components into the electrolyte), and mechanical erosion or delamination caused by intense oxygen bubble evolution. Usually, these mechanisms work synergistically, accelerating the overall deactivation process.<sup>74</sup> In order to solve these problems, researchers are exploring new catalyst preparation methods and reaction systems. On the one hand, the surface modification and doping of the catalyst can improve its ability to resist carbon deposition and poisoning, and on the other hand, optimizing the reaction conditions, such as adjusting the composition of the electrolyte and the pH value, can improve the reaction environment and enhance the stability of the catalyst.<sup>75,76</sup> In addition, anodic oxidation reaction can be combined with other technologies, such as photocatalysis and biocatalysis, to form a synergistic effect, which can further improve the upgrading efficiency of PET plastic monomers and the diversity of products.<sup>77</sup> Through structural optimization and component modulation of the catalyst matrix, large current density can be achieved at low decomposition voltage, which can effectively improve Faraday efficiency and high selectivity of EG, and guarantee the long-term stable operation of the electrolysis system.

**3.1.1. Structural modulation of electrocatalysts.** The structure of the catalyst is a key factor in determining the efficiency of the electrocatalytic reaction, which significantly affects the

adsorption, electron transfer, and product desorption of the reactants during the electrocatalytic reaction.<sup>78</sup> Construction of nano-defect structures is the most widely used method, which increases the number of active sites involved in the reaction (Fig. 5b).<sup>79</sup> Wu *et al.*<sup>82</sup> constructed a Ru nanocluster-anchored, N-doped, ordered macroporous carbon as an electrocatalytic micro-nanoreactor by pyrolysis of Ru(III) ion-containing ordered microporous ZIF-8 single crystals. Benefiting from the highly interconnected network of macroscopic nanopores, its electrochemical activity is superior to Pt/C. Ouyang *et al.*<sup>80</sup> loaded dendritic nano-silica doped with WO<sub>3</sub> on the surface of the Ni-based catalyst, and the mesoporous and dendritic network structure enhanced the specific surface area and pore volume of the catalyst thereby facilitating the electron transfer from H<sub>x</sub>WO<sub>3-y</sub> to Ni, reducing the activation energy for H<sub>2</sub> conversion and desorption over the catalyst's surface. In addition, the heterojunction structure also showed a contribution to the selectivity of the electrocatalysts. Peng *et al.*<sup>81</sup> grew amorphous nickel oxide *in situ* on a nickel foam (NF) surface. The unique nanolayer structure resulted in a significant increase in the specific surface area and catalytic activity of the electrocatalyst. The rapid transformation of this amorphous NiO to  $\gamma$ -NiOOH at the amorphous interface provides a thermodynamic advantage for the desorption of formate, and its high activity also promotes industrial applications (Fig. 5c). Lu *et al.*<sup>82</sup> loaded RhIn intermetallic compounds on carbon carriers to reduce the theoretical potential of the electrocatalytic reaction, and the loading of RhIn nanoparticles facilitated the optimization of the surface structure of the carbon carriers, which led to the reduction of the onset potential of the electrocatalytic reaction to 0.35 V, showing excellent EG to GA ability.

**3.1.2. Compositional modulation of electrocatalysts.** In the electrochemical recovery of EG, the components and elemental species of the electrocatalysts largely influence their catalytic properties, including the selectivity to the reacting substrates and stability. The elemental species determines whether the oxidation state change of the catalyst can match the potential window of the target reaction. Co and Ni, as the core transition metal catalysts in the electrocatalytic reaction, are widely used in the electrocatalytic reaction due to their high activity, low cost, multivalent flexibility, and tunable electronic structure (Fig. 5d–f).<sup>83,85</sup> The composition of the catalyst directly regulates the electronic structure and geometric structure of its surface active sites. This determines that it is more inclined to catalyze the cleavage of specific chemical bonds in EG molecules and guide the reaction along the desired path.<sup>86</sup> For PET recovery, highly selective oxidation can maximize the yield of the required intermediate product, reduce by-products, and significantly simplify subsequent separation and regeneration processes. Co and Ni are ideal candidate elements for industrial-scale PET recycling due to their low cost.<sup>87,88</sup> Through delicate component design (such as the introduction of an appropriate amount of doping elements or trace precious metals), the activity and selectivity of the unit catalyst can be maximized (reducing the overpotential and improving the current efficiency), while ensuring long life. This is directly converted into

lower energy consumption per unit EG recovery, less catalyst consumption, and higher product yield/purity, which ultimately improves the economic and environmental benefits of the entire PET chemical recovery process.

In summary, the structure and composition of the electrocatalyst are the core factors affecting its performance (selectivity, activity, and stability) in the technical route of electrochemical recovery of PET waste. Selecting core metals with appropriate redox properties (such as Co, Ni) and low cost as the basis, and then introducing other elements (doping or precious metals) for synergy and optimization, can greatly improve the selectivity and stability of the catalyst for EG oxidation. This delicate regulation of catalyst components at the molecular/atomic level is the key to achieving efficient, highly selective, and economical electrochemical cycling of PET, and provides important technical support for the resource utilization of plastic waste.

### 3.2. Effect of electrolytes

The electrochemical recovery of PET essentially involves the catalytic oxidation of its hydrolysis monomer, ethylene glycol (EG).<sup>89</sup> Therefore, the conversion efficiency is critically limited by the EG hydrolysis rate and monomer purity. Various hydrolysis methods are employed to produce EG from PET, including neutral, acid, alkali, and enzymatic hydrolysis. Although acidic hydrolysis makes a certain contribution to the increase of TPA yield, the use of acidic solution will introduce impurities and reduce the purity of the EG solution, which is not conducive to the subsequent catalytic oxidation process.<sup>90,91</sup> Compared with acidic hydrolysis, neutral hydrolysis of PET is an environmentally friendly hydrolysis method.<sup>92,93</sup> In acidic media, the protonation process may promote the formation of key intermediates, while under neutral conditions, the protonation process is more dependent on the regulation of surface adsorption energy. However, neutral hydrolysis is difficult to destroy the mechanical impurities of PET waste, resulting in the reduction of the purity of TPA in the solution, which affects the subsequent purification process.<sup>94</sup> Enzymatic hydrolysis, as an emerging decomposition method, shows great advantages in environmental protection, but its research is still in the laboratory stage, and it is difficult to promote on a large scale.<sup>95,96</sup>

In contrast, alkaline hydrolysis is more suitable for the direct hydrolysis of PET to obtain EG and TPA, and the excess alkaline solution can be used as the electrolyte for the subsequent electrochemical oxidation to recover EG, which avoids the introduction of impurities.<sup>97</sup> Secondly, the alkaline solution can destroy other mechanical impurities in PET waste plastics, which enhances the purity and yield of PTA.<sup>98</sup> However, the conditions of alkaline hydrolysis should also be fully considered, which include the composition of PET waste and the environment of alkaline hydrolysis. On the one hand, PET comes from a wide range of sources, and the composition of PET used in different scenarios has nuances, such as polyester fibers used for clothing and plastic water bottles. This requires that the concentration of the alkaline solution be regulated according to the actual form and specific composition of the reactants. On the other hand, the alkaline hydrolysis reaction is

slow at room temperature and pressure, and the EG monomer preparation is inefficient. In order to accelerate the rapid decomposition of PET, it is necessary to create suitable environmental conditions, such as increasing the reaction temperature and pressure by the hydrothermal method, solar energy and so on (Fig. 7a).<sup>99</sup> In addition, the mass transfer process of the monomer from the bulk solution to the electrode surface also affects the whole process. Improving the mass transfer reaction kinetics of the whole system is very important to improve the conversion efficiency, such as stirring/flow electrolytic cell design, electrode structure design to increase specific surface area/promote convection, concentration control and other strategies.<sup>100,101</sup>

### 3.3. Interface optimization

In the electrolytic recovery of PET, the catalytic oxidation reaction of EG mainly occurs at the electrode/electrolyte interface. The mass transfer of EG from the electrolyte to the electrode surface and the desorption of the oxidized product GA can be accelerated by regulating the interface environment. Effective measures to improve the electrode–electrolyte interface environment include enhancing the wetting rate of the electrode in the electrolyte, optimizing the diffusion behavior of EG in the electrolyte, and enhancing the affinity between EG and the electrode.<sup>102</sup> Endowing the anode surface with super-hydrophilic properties during EG oxidation promotes intimate EG–electrolyte–electrode contact, forming a favorable solid–liquid interface and accelerating anode reaction kinetics. However, high wettability may negatively impact the cathode, where the hydrogen evolution reaction (HER) occurs. Strong electrode/catalyst hydrophilicity can cause hydrogen bubble attachment. Covering bubbles blocks active sites and impedes  $\text{OH}^-$  transfer from the liquid phase to the cathode, inhibiting reaction kinetics and increasing energy consumption.<sup>103</sup> The diffusion behavior of EG in the electrolyte directly affects the mass transfer rate. The concentration gradient is the main driving force for the migration of EG into the diffusion layer at the electrode surface: during the reaction, the EG concentration at the electrode surface decreases due to consumption, while the GA concentration increases due to generation. Reducing the thickness of the diffusion layer significantly accelerates the diffusion of reactants/products, thereby increasing the overall electrolysis efficiency. Enhancing the affinity of EG to the electrode not only improves the interfacial ion transport efficiency, but also promotes the electrode reaction kinetics. For example, increasing the surface roughness of the electrode exposes more reactive sites while enhancing the degree of contact between the electrode and the electrolyte.

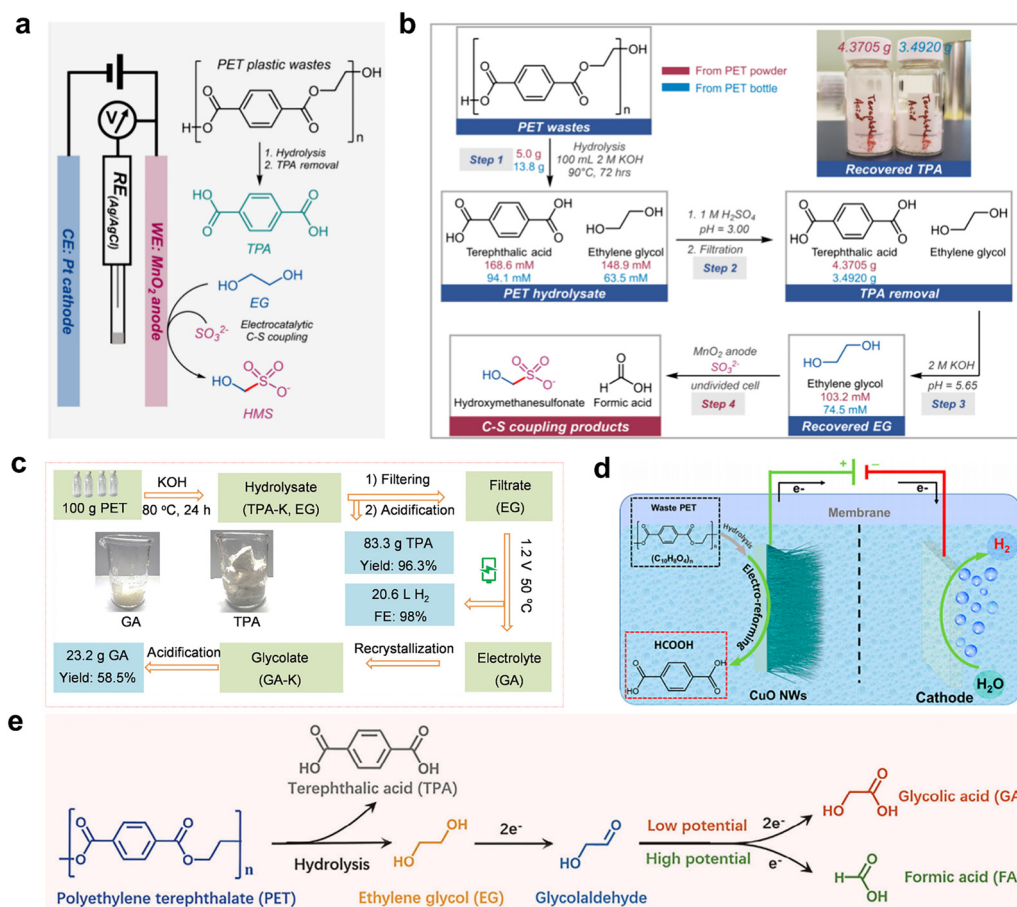
In addition, the key technologies of electrochemical recovery also include electrolytic cell configuration, reactor type, industrial scale, *etc.* The optimization and innovation of these technologies will directly affect the efficiency and cost of recycled PET, which is also a challenge. Among them, the design of electrolytic cell configuration needs to take into account gas separation and current distribution, the selection of reactor type needs to consider thermodynamic stability and material compatibility, and

large-scale production needs to solve problems such as system integration and energy management. In the future, with the development of new materials and intelligent control technology, these key technologies are expected to achieve breakthroughs and promote the large-scale commercial application of electrochemical recovery technology.

## 4. High-value-added products from PET decomposition

The effective electrocatalytic recovery of plastic waste for the preparation of high-value-added products remains an imperfect field. The selectivity of ideal final products (such as specific chemical raw materials, fuels, or fine chemical intermediates) depends heavily on the reaction conditions and is controlled by the complex reaction conditions.<sup>104</sup> Catalyst materials (metal catalysts, carbon-based catalysts, *etc.*), electrolyte selection (solvent composition, supporting electrolyte), electrode potential, reaction temperature, and other parameters greatly affect the reaction path, selectivity, and final product distribution.<sup>56</sup> In the actual production process, the substances produced by the anode and cathode chambers of the electrolytic cell often change with the different reaction parameters, and selectively convert PET into a few high-value target products (such as *p*-xylene, monomers), while minimizing the production of low-value or harmful by-products (such as mixed oligomers, carbon dioxide).

In recent years, the understanding of the formation pathway of carboxylic acid compounds in the process of PET electrocatalytic upgrading has been significantly expanded. The main products of PET alkali hydrolysis are TPA and EG (Fig. 6a and b). TPA is the main organic linker for many crystalline porous structures. The carbon elements in PET plastic waste can be rearranged and combined to form carbon nanotubes or graphene with excellent electrical conductivity, thermal conductivity and mechanical properties. This transformation not only realizes the resource utilization of waste, but also provides high-performance raw materials for electronic devices, energy storage materials, composite materials and other fields.<sup>105</sup> As the core raw material for subsequent electrocatalytic oxidation, ethylene glycol can be converted into high-value carboxylic acids, such as glycolic acid, formic acid and oxalic acid. As the oxidation product of ethylene glycol, carboxylic acid compounds have broad application prospects in industrial fields such as medicine and food additives (Fig. 6c).<sup>106</sup> In addition, controlling the applied potential and pH conditions is beneficial to the formation of oxalic acid by C–C bond cleavage. Further optimization of reaction parameters and development of advanced electrocatalysts are expected to improve the yield and selectivity of other additional products such as carboxylic acids, thereby more effectively increasing the value of PET waste. Moreover, this method realizes the efficient utilization and sustainable development of resources by efficiently converting waste materials into high-value-added products in the electrochemical system, highlighting the importance of electrochemical closed-loop recycling and the upgrading cycle. By optimizing the above



**Fig. 6** Product distribution of electrochemically recycled PET. (a) Scheme of electrocatalytic PET plastic waste to HMS. (b) Commercial PET powder and PET water bottle upcycling pathways; the inset image represents the recovered TPA. 5.0 g of PET powder was hydrolyzed in 2.0 M KOH. Reproduced from ref. 62. Published by American Chemical Society under a CC BY-NC-ND 4.0 license. (c) Schematic illustration of the process of PET electrocatalytic upgrading and product separation. Inset: Photographs of GA and TPA powder. Reproduced with permission.<sup>107</sup> Copyright 2023, Wiley-VCH. (d) Hybrid electrolysis for electrocatalytic upgrading of the PET hydrolysate and hydrogen evolution. Reproduced with permission.<sup>108</sup> Copyright 2022, Physical Chemistry Letters. (e) A scheme illustrating the reaction pathways of PET upcycling. Reproduced with permission.<sup>77</sup> Copyright 2024, Wiley-VCH.

catalytic system, pretreatment and interface process, the electrochemical method can convert PET waste into a variety of high-value-added products, and its distribution is mainly controlled by the reaction path (C1, C2). This section will elaborate on this topic.

#### 4.1. C1 pathway (C–C cleavage)

In the process of electrochemical recovery of PET, ethylene glycol is one of the core products of PET depolymerization, and its oxidation path is crucial to the synthesis of the final product. The main products of the C1 pathway are FA, CO<sub>2</sub>, etc. The C1 oxidation pathway of EG mainly generates single carbon products by cracking the C–C bond, with formate/formate salts (such as HCOONa, HCOONH<sub>4</sub>) being the core target products with economic value (Fig. 6d). The reaction is carried out on the surface of the electrode, and finally the formate was decomposed into a combination of formate ions and cations (such as Na<sup>+</sup> or NH<sub>4</sub><sup>+</sup>) in the electrolyte. In addition, the generated formic acid can theoretically be reused for the synthesis of PET or other polyesters, or used in other chemical processes,

improving the atomic economy and sustainability of the entire recovery route. For industrial applications, the effective separation and purification of the reaction mixture is crucial. Common methods include vacuum distillation after acidification to release liquid formic acid, or solvent extraction. High purity formate crystals, such as sodium formate, can also be directly obtained by evaporation crystallization in an alkaline electrolyte. Purified formic acid/formate can be widely used. It can be directly used as an efficient disinfectant and preservative, and can be deeply processed to form formate (solvent, spice), formamide (solvent), pharmaceutical intermediates, etc. In addition, it can also be used as a clean fuel for direct formic acid fuel cells (DFAFC).<sup>109</sup> Although formic acid (salt) cannot be directly recycled to upgrade PET, it represents a carbon cycle process through its use in other chemical processes, mainly reflected in the conversion of carbon in PET waste into valuable formic acid chemicals, thereby improving the overall atomic economy of the PET life cycle and reducing dependence on fossil raw materials for these specific formic acid applications. Formic acid can be used as a reducing agent or hydrogen carrier in chemical processes, contributing to a wider chemical ecosystem.

Therefore, in the electrochemical recovery of PET, the key to achieve highly selective oxidation of EG to FA (salt) is to regulate its reaction path through the C1 path (Fig. 6e). The core of this pathway is to use Pt or high-performance non-precious metal catalysts to efficiently dehydrogenate EG into acetaldehyde as a key intermediate in an alkaline environment. Subsequently, through the surface activation of the catalyst, acetaldehyde undergoes C–C bond cleavage (the mechanism includes the formation of formyl intermediates or retro-aldol condensation), generating highly oxidized C1 fragments (such as CO, formaldehyde), and finally it is rapidly converted into the target product formate/formic acid.

#### 4.2. C2 pathway (C–C preservation)

The core goal of the C2 pathway is to retain the C–C skeleton of the EG molecule and oxidize it to produce valuable two-carbon products. According to the degree of oxidation, the main products include glycolic acid (4 electron transfer), glyoxylic acid (6 electron transfer) and oxalic acid (10 electron transfer). Among them, glyoxylic acid and oxalic acid have poor stability

and are easy to be further oxidized (decarboxylated) to CO<sub>2</sub> at higher application potentials (Fig. 7d and e).

The effective separation and purification of complex C2 oxidation products (glycolic acid, glyoxylic acid and oxalic acid) from the mixture of reaction products is crucial for industrialization. Among them, glycolic acid is the most stable and valuable product, which can be purified by solvent extraction, ion exchange or crystallization. Glyoxylic acid requires rapid, low-temperature treatment or protective complexation (*e.g.*, with sulfite) to prevent degradation. Purification of oxalic acid depends on acidification-crystallization or precipitation.<sup>107</sup> Purified glycolic acid is a key monomer of high-value-added, biodegradable biomedical polymer polyglycolic acid (PGA), and polylactic-*co*-glycolic acid (PLGA), which can realize the direct recycling of PET waste, and can also be used for cosmetics and cleaning. The application of glycolic acid in bioplastics realizes the high-value upgrading and utilization of PET waste, which complements the C1 cycle and improves the sustainability and economic benefits of PET electrochemical recovery. The purification of glyoxylic acid is the most difficult because of its

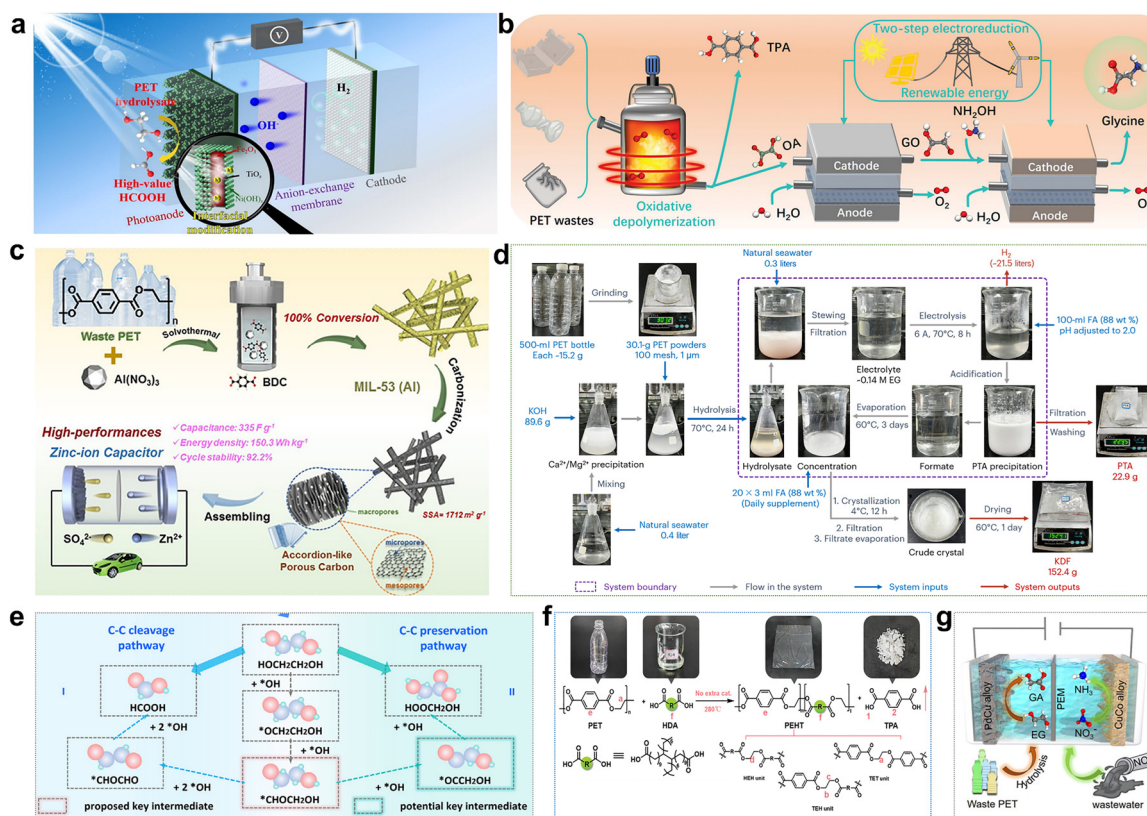


Fig. 7 PET recycling method combined with other technologies. (a) Photoelectrocatalytic preparation of formic acid from PET. Reproduced with permission.<sup>41</sup> Copyright 2023, Elsevier. (b) Schematic illustration of high-value glycine production from PET waste via a spatially decoupled thermochemical-electrochemical system. Reproduced with permission.<sup>110</sup> Copyright 2024, Wiley-VCH. (c) A schematic diagram of MIL-53(Al) and its derived porous carbon for high-performance zinc ion capacitors prepared by directly recycling waste PET bottles. Reproduced with permission.<sup>105</sup> Copyright 2024, Elsevier. (d) A schematic diagram of the technical route for upgrading and recycling of electrochemical seawater PET. Reproduced with permission.<sup>111</sup> Copyright 2024, American Association for the Advancement of Science. (e) Two conversion modes in EGOR: I, C–C cleavage pathway; II, C–C preservation pathway. Reproduced with permission.<sup>11</sup> Copyright 2024, Royal Society of Chemistry. (f) Biodegradable membranes and high purity TPA were obtained by bio-based HDA. Reproduced with permission.<sup>112</sup> Copyright 2024, Wiley-VCH. (g) Paired electrochemical synthesis of glycolic acid and ammonia from polyester and nitrate sewage. Reproduced with permission.<sup>57</sup> Copyright 2025, Cell Press.

extremely active aldehyde group (which can be easily oxidized to oxalic acid or undergo polycondensation). It needs to be separated quickly, operated at low temperature or protected by sulfite complexation. Oxalic acid can be purified by acidification concentration crystallization or the classical calcium salt precipitation method (acidification regeneration after calcium oxalate formation). Crucially, the role of glycolic acid in the synthesis of bioplastics establishes a high-value closed-loop pathway that complements the C1 pathway. This C–C preservation strategy improves the overall sustainability, resource efficiency and economic feasibility of PET electrochemical recovery by directly converting ethylene glycol into valuable functional chemicals such as bio-based polymers, and realizes carbon value in a circular economy.

In summary, while the C2 pathway focuses on preserving the C–C skeleton for high-value chemicals, the C1 pathway is characterized by complete oxidation, emphasizing the need for careful control of catalyst design and reaction parameters to steer selectivity toward desired products.

#### 4.3. Recyclable monomer

Electrochemically recycled monomers (GA) can be converted to other polymers (PGA) for upcycling or closed-loop recycling. EG can polymerize during electrocatalytic oxidation to produce degradable polyesters (such as PGA, PLA), which can be efficiently degraded into CO<sub>2</sub> and H<sub>2</sub>O in the natural environment and by microorganisms (Fig. 7f).<sup>112</sup> Among them, PGA is an important biopolymer, especially in the medical, oil-gas exploitation, and ecology fields because of its high thermal stability ( $T_m = 220\text{--}230\text{ }^\circ\text{C}$ ), mechanical strength (115 MPa), high gas-barrier properties, excellent biocompatibility, and rapid biodegradability. However, there are two core challenges in the electrocatalytic PET preparation of PGA: (I) the low spatiotemporal yield of ethanolic acid from ethylene glycol and (II) the high cost of separation and purification of glycolic acid crystals. In response to these two key technical issues, researchers have recently developed a palladium catalyst loaded with a strong Lewis acid (CoCr<sub>2</sub>O<sub>4</sub>), which promotes the rapid migration of OH<sup>−</sup> in the reaction system through the strong Lewis acid, greatly improving the spatiotemporal yield of ethylene glycol to prepare glycolic acid.<sup>113</sup> A new distillation and alcohol precipitation process has been developed, which not only achieves efficient separation of high-purity glycolic acid crystals, but also completes the recycling of alkali solution/ethylene glycol and the recovery of by-products. Based on these technological innovations, the team independently designed a 500 W level fuel cell

stack system and completed the full process amplification experiment of 20 kg waste PET to PGA (Table 2).

#### 4.4. Cathodal products

While anodic EG oxidation is the core process for PET upcycling, the concurrent cathodic reduction reaction is equally critical. It directly governs the system's energy efficiency, economic feasibility, environmental impact, and overall product value.<sup>114</sup>

Hydrogen (H<sub>2</sub>) produced by the cathode is a non-negligible resource in the whole system. H<sub>2</sub> serves as both a clean energy carrier and a crucial chemical feedstock (e.g., for ammonia synthesis, petroleum refining, hydrogenation reactions). The demand for green hydrogen is substantial in the context of sustainable energy transitions. Especially in alkaline media, HER produces OH<sup>−</sup>, which helps to maintain the alkalinity of the system, which is usually beneficial for the oxidation of anode EG to form formate or stabilize other high-value-added oxidation products, and can inhibit the formation of CO<sub>2</sub>.<sup>48</sup> However, it is necessary to consider the cost and safety of the separation, storage, and utilization of hydrogen. For instance, the separation of hydrogen from the mixed products is usually more complicated than the production of hydrogen from water.

In addition, when the source of PET plastic waste involves nitrate-containing wastewater (e.g., from industrial packaging cleaning), nitrate reduction reaction (NO<sub>3</sub><sup>−</sup>RR) occurs at the cathode, which can convert NO<sub>3</sub><sup>−</sup> into NH<sub>3</sub> or N<sub>2</sub>. Ammonia is widely used in fertilizer manufacturing and chemical production (such as nitric acid, urea).<sup>115,116</sup> The reduction of nitrate in wastewater to ammonia has a dual benefit – water purification and the combined production of basic chemicals – in line with the green environmental development goals. However, it should be noted that NO<sub>3</sub><sup>−</sup>RR may produce nitrite (NO<sub>2</sub><sup>−</sup>), which involves a complex multi-electron transfer mechanism and requires a reasonable catalyst to control the reaction process (Fig. 7g). At the same time, the CO<sub>2</sub> released during PET depolymerization or EG anodic oxidation can also be reused by electrochemical CO<sub>2</sub> reduction reaction (CO<sub>2</sub>RR). As a research hotspot of electrocatalysis, cathode CO<sub>2</sub>RR can convert carbon dioxide into valuable compounds such as methane, ethylene and ethanol, and achieve wider carbon recovery.<sup>117</sup> However, compared with HER and NO<sub>3</sub>RR, CO<sub>2</sub>RR often requires a higher overpotential to achieve good activity and selectivity. The reaction rate and selectivity (target product) are difficult to control, and the product is often more complex. This is primarily due to the inherent thermodynamic stability

Table 2 Summary of partial electrochemical recovery of high-value additional products

Products	Reaction mechanism	Reaction condition	Recovery rate (%)	Value (\$ per ton)	Maturity
TPA	Hydrolysis (ester bond cleavage)	Normal temperature and pressure	92–95	1200–1500	Model of commercialization
EG	Hydrolysis (ester bond cleavage)	Normal temperature and pressure	85–88	800–1000	Quasi-commercialization
GA	Oxidation of EG (partial oxidation)	+0.8 V (vs. SCE)	75–80	4000–6000	Pilot phase
FA	Oxidation byproduct (from EG or others)	+1.0 V (vs. SCE)	85–90	1500–2000	Lab scale
OA	Oxidation of EG (complete oxidation)	+1.2V (vs. SCE)	90–93	3000–4000	Pilot phase
DMT	Methanolysis of PET (transesterification)	60–80 °C, methanol solvent	88–90	1800–2100	Pilot phase

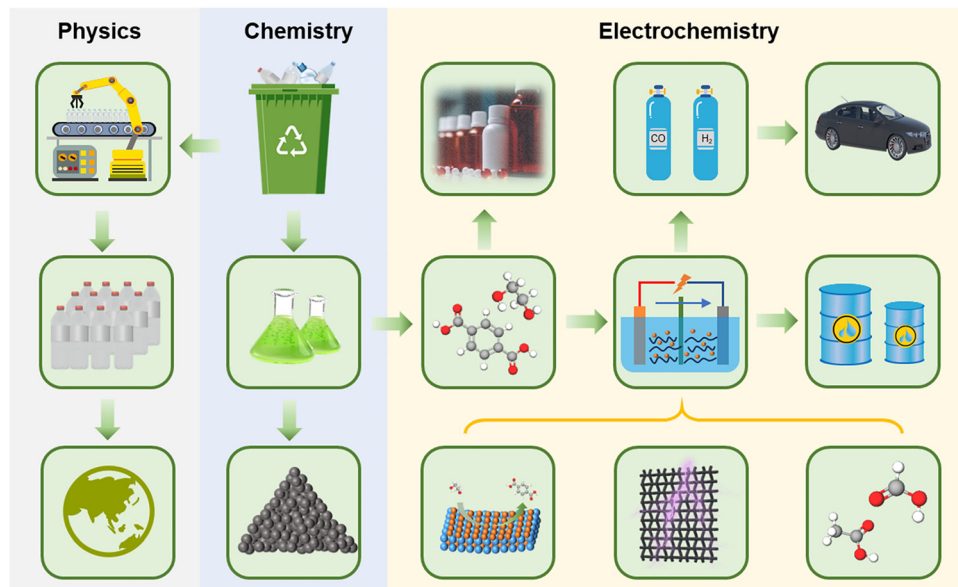


Fig. 8 A schematic diagram of the application of regenerated high-value additives based on different waste PET recycling methods.

of CO<sub>2</sub> molecules and the complex multi-electron transfer processes involved in CO<sub>2</sub> reduction. To address this challenge, researchers have explored various strategies, such as designing advanced electrocatalysts with tailored electronic structures, optimizing reaction conditions, and engineering electrode interfaces (Fig. 8).<sup>118–120</sup>

## 5. Conclusions and prospects

PET plastic products have a profound impact on human production and lifestyle because of their lightweight and corrosion resistance. However, the global annual output is not proportional to the effective recovery rate, forming a serious ecological challenge. Therefore, the establishment of a closed-loop circulation system for PET plastics is crucial to the ecological development of society. Plastic itself is not a pollutant, but a new energy source. The recovery, reprocessing, and reuse of PET plastics through mild electrochemical techniques can minimize their negative impact on the environment. Electrocatalytic recycling technology is a promising strategy for converting plastic waste into a variety of valuable materials. In this paper, the latest progress of electrochemical recycling of waste PET plastics is reviewed, and the reaction mechanism, key catalytic technology, and product distribution of this technology are discussed in detail. At the same time, the research results in this field at home and abroad in recent years are analyzed. The methods and effects used in different studies are compared. The key factors affecting the efficiency and selectivity of electrocatalytic recovery are discussed. The basic principles of electrocatalytic recovery of PET plastics are summarized, including chemical dissociation and chemical reaction interface kinetics in the reaction process. In addition, the challenges faced by the technology in practical applications, such as the stability of electrode materials, the feasibility of large-scale

production, and cost-effectiveness, are highlighted. This technology can efficiently decompose and convert plastics under mild conditions, greatly reducing energy consumption and environmental pollution (Fig. 9). However, this method is still in its infancy, and there are still many challenges. In order to overcome these challenges, we propose to give priority to the following issues in future research work. It is believed that through in-depth research on these aspects, it can provide theoretical support and practical guidance for the further development of electrocatalytic recycling of waste PET plastic technology, and promote it from laboratory research to industrial application, thus opening up a new way to solve the increasingly serious plastic pollution problem.

### 5.1. Development of efficient catalytic systems

For efficient catalytic systems, their stability is one of the key factors that determines whether industrial applications can be achieved. In the actual reaction process, catalytic materials often face harsh reaction conditions such as high temperature, high pressure, strong acid, and alkali, which will lead to changes in the structure of catalytic materials and loss of active components, thereby reducing the activity and selectivity of the catalyst. Therefore, it is of great significance to study the stability of catalytic materials. Traditional materials have obvious limitations in dealing with these harsh conditions. Taking the metal oxide catalyst as an example, its lattice structure is prone to rearrangement at high temperatures, resulting in a decrease in specific surface area and a decrease in active sites. Moreover, in strong acid/alkaline media, metal ions may dissolve into the solution, resulting in irreversible loss of active components. Therefore, the importance of the development of new catalytic materials can be seen. Among them, two-dimensional materials such as metal-organic framework materials (MOFs), graphene, and transition metal chalcogenides show great potential due to their unique structures and

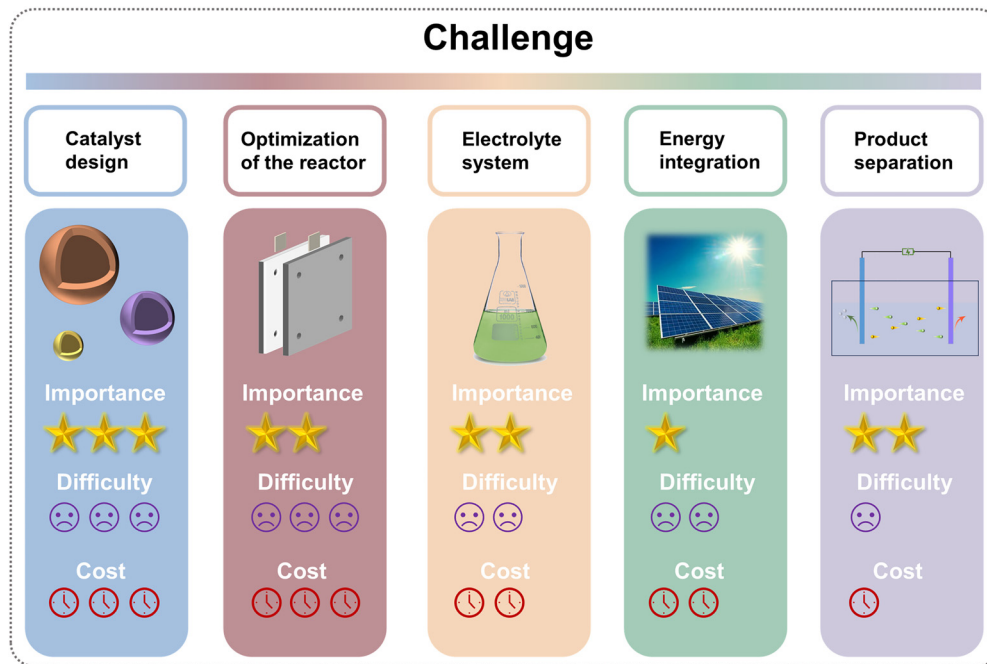


Fig. 9 Future challenges of electrochemical recycling of waste PET.

properties. At the same time, given that nickel (Ni) and cobalt (Co)-based catalysts have shown significant intrinsic activity and cost advantages in the electrochemical recovery of PET reactions, the improvement of their durability has also become a key direction of current research. Through advanced material engineering strategies, such as constructing a core-shell structure, introducing doping elements, forming an alloy, or developing a stable carrier interface, the anti-sintering and anti-dissolution ability of nickel and cobalt active sites under operating conditions can be effectively enhanced, thereby significantly extending the catalyst life. With the continuous deepening of the research on new catalytic materials, we are expected to develop catalytic systems with higher stability and catalytic performance, which will provide strong support for industrial applications in chemical, energy, and other fields. In the future, it is necessary to further explore the preparation methods and catalytic mechanisms of new materials and optimize the performance of materials to meet the needs of different industrial reactions.

### 5.2. Green development and process optimization

With the continuous improvement of environmental awareness and the deepening of the concept of sustainable development, the green development of catalytic reactions has become an important research direction in the field of catalysis. Green catalytic reactions aim to reduce or eliminate the use and production of harmful substances, improve the utilization rate of atoms, and reduce energy consumption. In terms of catalyst design, green and environmentally friendly raw materials should be selected. Traditional catalysts may contain harmful substances such as heavy metals, which will pollute the environment during preparation and use. Therefore, it is of great significance to develop catalysts with non-toxic and harmless metals or non-

metals as active components. Optimizing the reaction solvent is also an important part of achieving green catalytic reactions. Traditional organic solvents often have the disadvantages of volatility and toxicity, which will affect the atmospheric environment and human health. Therefore, it is an effective way to replace traditional organic solvents such as water and supercritical carbon dioxide. In some catalytic reactions, by designing suitable catalysts and reaction systems, the reaction can be carried out efficiently in the aqueous phase, which can not only reduce the use of organic solvents, but also simplify the subsequent separation process. In addition, improving the atomic utilization of catalytic reactions is one of the core objectives of green catalysis. An atomic economy reaction requires the atoms in the reactant molecules to be transformed into the target product as much as possible, reducing the formation of by-products. Through the rational design of the active center and reaction path of the catalyst, the selective catalytic reaction is realized, so that the reaction is carried out in the direction of generating the target product, avoiding unnecessary side reactions and improving the atom utilization rate.

### 5.3. Process integration and industrialization

At present, the electrochemical upgrading of PET plastic waste is mainly in the laboratory stage, and there is still a long way from industrialization. Therefore, the shift to large-scale industrial processes is crucial for the practical implementation of plastic waste electrolysis. The first challenge to be addressed is to ensure the long-term stability of the catalyst and seamless system integration. For the long-term stability of the catalyst, it is necessary to study the deactivation mechanism of the catalyst in the complex electrolytic environment, and improve the ability of the catalyst to resist deactivation factors such as

poisoning and sintering by improving the preparation method of the catalyst and adjusting its composition. In terms of seamless system integration, it is necessary to systematically optimize all aspects of the electrolysis process, such as electrode design, electrolyte selection, reaction device construction, *etc.*, to ensure that all parts work together to improve the overall electrolysis efficiency. At the same time, the establishment of a real-time monitoring system can find and solve the problems in the process of system operation in time. In addition, pilot-scale amplification experiments are needed to gradually verify the feasibility and stability of the process at different scales, and accumulate data and experience for industrial production.

#### 5.4. Policy and industrialization promotion

The government should introduce a series of policies that are conducive to the industrialization of electrochemical upgrading of PET plastic waste. On the one hand, tax incentives and financial subsidies are given to relevant enterprises to reduce the production costs and risks of enterprises, and encourage enterprises to actively participate in the industrialization process in this field. For example, for enterprises that build PET plastic waste electrochemical upgrading plants, a certain proportion of fixed asset investment subsidies are given; for enterprises that produce upgraded products that meet the standards, corporate income tax for a certain period of time is reduced. On the other hand, strict plastic waste management regulations should be formulated to improve the recycling rate of plastic waste and provide sufficient raw material supply for the electrochemical upgrading of PET plastic waste. At the same time, a sound market access and supervision mechanism should be established to ensure the quality and safety of upgraded products and promote the healthy development of the market. In addition, the government can also strengthen the connection between universities, scientific research institutions, and enterprises by organizing industry-university-research cooperation projects, accelerate the transformation and application of scientific research results, and promote the rapid development of PET plastic waste electrochemical upgrading industry.

### Author contributions

Chao Chen: investigation, formal analysis, data curation, visualization, writing – original draft; Yi Wang: investigation, data curation; Qixuan Zhu: investigation, data curation; Yu Chu: investigation, data curation; Jiacheng Zhao: investigation; Jing Tang: writing – review & editing, supervision; Yusuke Yamauchi: methodology, formal analysis, conceptualization; Guiyin Xu: writing – review & editing, methodology, funding acquisition, formal analysis, conceptualization. Meifang Zhu: supervision, methodology, conceptualization.

### Conflicts of interest

The authors declare no conflict of interest.

### Data availability

Data availability is not applicable to this article as no new data were created or analyzed in this study.

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