



Review article

Chemo-catalytic recycling of PET waste: Progress and prospects for circular economy and valorization

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ABSTRACT

Polyethylene terephthalate (PET), a non-biodegradable single-use plastic, is emerging as a significant environmental issue. The extensive utilization of PET in packaging, especially for disposable products like beverage bottles and food containers, has led to a growing build-up of its waste in landfills, rivers, and oceans, contaminating the ecosystem and eventually infiltrating the global food chain. The limitation of existing physical degradation methods has spurred interest in chemical recycling as a promising alternative for managing PET waste. Traditional, non-catalytic methods for depolymerizing PET are sluggish, energy-intensive, and require high temperatures and/or pressures. However, recent breakthroughs in material chemistry have led to the introduction of innovative strategies that can significantly enhance PET degradation under relatively mild reaction conditions. This review highlights the most recent advances in the development of efficient catalysts such as biomass-waste, mixed metals, zeolites, metal-organic framework, nanomaterials, organocatalysts, and ionic liquids for the processes of glycolysis, methanolysis, and reductive depolymerization. Each section provided a brief overview of the catalyst preparation, functionality, active sites, and reaction mechanism.

1. Introduction

Plastics are a remarkably versatile and indispensable material, often referred to as the “material of a thousand uses” [1]. They possess exceptional capability to be shaped and molded into different forms, making them go-to materials in the apparel and automotive industries and meeting end-user demands in medical and electronics equipment [2]. The phenomenal water and chemical resistance, affordability, simple manufacturing process, impressive strength-to-weight ratio, and convenient handling of the materials have all contributed to their widespread applications [3–5]. In 2012, the global production of plastics amounted to 280 million tons [6] and this figure rose to over 400 million tons a decade later, with approximately 50 % being utilized for single-use applications. If the current consumption rates continue, the estimated plastic production is expected to reach 33 billion tons by 2050 [6]. Each plastics design differs depending on its applications, with each one possessing distinct characteristics that make it ideal for each purpose. Alongside other polymers, including polyethylene (PE), polypropylene (PP), and polystyrene (PS), polyethylene terephthalate (PET)

represents the fourth most manufactured synthetic material [7,8]. PET polyesters, a thermoplastic polymer of ethylene terephthalate monomers with alternating (C₁₀H₈O₄) units, are consistently reliable for a wide range of applications, whether used in the manufacturing of both lightweight and durable packaging materials or in the production of soft and robust textile fiber [9,10]. Global PET production has witnessed a substantial increase, reaching 400 million tons in 2022, over 200 times greater than the 1.5 million tons produced in 1950 [11]. There are three primary methods for synthesizing PET: 1. Esterification of terephthalic acid with ethylene glycol; 2. Transesterification of dimethyl terephthalate with ethylene glycol; and 3. Direct polycondensation of BHET [12].

The remarkable growth in plastic production has been a double-edged sword, as the ‘plastic era’ has brought forth myriad detrimental consequences. The outstanding durability of PET, which makes them so popular for various applications, is one of their major drawbacks [15]. The presence of an aromatic structure makes the post-consumer PET waste highly resistant to hydrolytic and microbial degradation [16]. The slow biodegradation rate, coupled with unregulated usage and

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ineffective waste recycling, has led to the accumulation of PET waste in the environment [17,18]. More than half of the world's PET waste is landfilled, where it is projected to take hundreds of years to degrade [19]. According to a recent report, PET makes up approximately 12 % of global solid waste in volume [20,21]. Based on an analysis conducted by Greenpeace, it has been found that Coca-Cola manufactures a staggering 100 billion PET bottles annually, equivalent to 3400 bottles per second. Unfortunately, only a mere 7 % of these bottles are recycled [22]. Although mechanically recycled PET bottles (rPET) consume 75 % less energy compared to virgin plastic, major brands have not embraced rPET extensively due to its impact on the transparency of the bottles [23].

While plastic pollution is widespread on land, ocean plastics are a prime example of its pervasiveness in the environment. In 2018, the Great Pacific Garbage Patch was estimated to contain approximately 1.8 trillion plastic fragments, with a total mass of 79,000 metric tons, exhibiting continuous annual growth [24]. Approximately 14 million tons of waste plastic have found their way into the oceans every year, with PET materials being the primary contributor to this waste [25–27]. Synthetic PET waste in freshwater and marine ecosystems undergoes physical weathering, generating fragments of plastic waste known as microplastics (1 μm –1 mm) and nanoplastics (< 1 μm), potentially disrupting the food chain and posing a catastrophic threat to human health [28]. In addition, plastic materials may harbor hazardous contaminants, including phthalates, bisphenol A (BPA), polychlorinated biphenyls (PCB's), and nonylphenol ethoxylates (NP) [29]. Bioaccumulation of these harmful chemicals in organisms can have detrimental effects on their growth, reproductive capabilities, and immune systems [28]. Multiple studies have demonstrated the presence of PET debris and microplastics in over 690 species of marine organisms [30]. When ingested, it can lead to internal injuries like perforated gut, ulcerative lesions, and gastric rupture [31]. Additionally, plastic can affect the biochemical responses at a cellular level, causing oxidative stress [32], changes in metabolic parameters [33], reduced enzyme reactivity [34], and cellular necrosis [35]. PET fiber can take a span of up to three decades to decompose within humans and other life forms [36].

Despite their drawbacks, PET provides advantages in our everyday existence, and there are currently no viable alternatives that can be implemented on a large scale right away. Hence, to stop the continuous flow of waste into the environment, there is an urgent need for a plan to build a circular system for PET waste. Traditional mechanical recycling techniques have been chosen for their cost-effectiveness and as a more energy-efficient option for large-scale operations [37]. Mechanical recycling involves a series of physical processes, such as decontamination, shredding, grinding, and melt processing, to convert waste PET into reusable products. Notably, recycling PET requires significantly less energy—up to 88 % less—than producing plastics from new raw materials. However, it encounters challenges such as the production of lower quality PET, which is often only appropriate for making products of low-value or functionality compared to the original materials [38,39]. The repeated reprocessing by physical process has resulted in specification alteration of the material, such as a change in coloration or a decrease in product molecular weight, and thus this approach is commonly stated as 'downgrading' or 'downcycling' process [40,41]. The reduction in molecular weight of recycled plastics is primarily attributed to chain-scission reactions during thermal reprocessing, as well as the presence of acidic impurities and moisture. To mitigate these degradative effects, thorough drying of the plastic waste post-washing, incorporation of chain extenders, and reprocessing under vacuum conditions are recommended [42].

In contrast to mechanical recycling, which depends on physical processes, chemical recycling or advanced recycling focuses on the molecular structure of PET, breaking it down into its chemical building blocks [43]. Chemical recycling aligns with the principles of 'Sustainable Development' by enabling the formation of virgin-quality PET from depolymerized products, reducing dependence on fossil fuels for new

plastic production [44]. This process presents a powerful approach to address the limitations of traditional mechanical systems, effectively 'closing the loop' and establishing a circular economy for this synthetic material [45,46]. Unlike the high viscosity associated with molten polymers obtained in mechanical recycling, monomer-containing solutions derived from chemical recycling exhibit significantly lower viscosity, enabling more efficient separation of impurities. This property facilitates the chemical recycling of complex waste streams containing additives, dyes, blended fabrics, and other contaminants into high-purity recycled products [43]. Chemical depolymerization of PET proceeded via the processes of glycolysis, alcoholysis, pyrolysis, ammonolysis, and hydrolysis. These routes are classified based on the specific chemical reagents used, including glycols, methanol, ammonia, water, or hydroxide (Fig. 1) [47]. Life cycle assessment (LCA) studies have indicated that glycolysis using propylene glycol, aminolysis, and hydrogenolysis exhibit lower global warming potentials, ranging from 4.3 to 5.8 kg CO₂ equivalent per kg of upcycled PET. These processes demonstrate significantly lower environmental burdens compared to the production of virgin PET [48]. Accordingly, there has been significant research conducted in recent years to develop sustainable chemo-catalytic systems for PET recycling [49,50]. Numerous review articles and relevant book chapters that discuss the chemical recycling of PET have existed [51–55]. Despite this great achievement, the majority of review papers published thus far have mostly concentrated on technological advancements related to manufacturing techniques, optimizing reaction yield, achieving energy efficiency, and conducting life cycle assessments. This review has specifically examined the chemo-catalytic recycling of PET waste using highly efficient transesterification catalysts. In the majority of the studies discussed in this review, pretreated PET waste, typically cleaned and shredded, was used as the feedstock for depolymerization. This pretreatment step ensured consistent reaction conditions and enhanced interaction with catalysts or solvents; however, it may limit practical applicability of these methods.

Bibliometric analysis provides a quantitative assessment of scientific publications within a certain field. This technique is crucial for summarizing past research trends and offering academics a structured approach to uncovering innovative methodologies and emerging areas of recent research topics. The method was employed to illuminate the present landscape of PET chemical recycling in plastic waste valorization. Fig. 2 revealed four distinct clusters, each represented by a different color, catalysis (red color), glycolysis (green color), aminolysis (purple color), and alcoholysis (yellow color). The size of each circle mirrors the research intensity within that cluster, while the lines connecting keywords demonstrate the relationships between the subjects. Notably, the glycolysis method has emerged as the most extensively studied area within chemical recycling techniques. Glycolysis presents several notable advantages, including operational simplicity, the ability to proceed under atmospheric pressure and relatively low temperatures, and the use of reagents and products with low volatility and minimal toxicity. Furthermore, the process minimizes environmental impact by avoiding the generation of acidic or alkaline wastewater and facilitates straightforward product recovery through methods such as hot-water extraction, cooling crystallization, and adsorption. Owing to these benefits, particularly the reduced reagent consumption and milder reaction conditions compared to other depolymerization techniques such as methanolysis, glycolysis has emerged as one of the most extensively studied approaches for the chemical recycling of PET waste [56,57]. The dominance of the glycolysis cluster demonstrates that other PET chemical recycling methods are in their nascent periods of research and development. Furthermore, the glycolytic depolymerization of carbonate-based polycarbonate (PC) into bisphenol A (BPA) monomer emerges as a highly effective and promising strategy for the chemical recycling of PC waste [58–60].

In this review, we provide an overview of the literature that discusses current advancements and trends in developing effective and stable

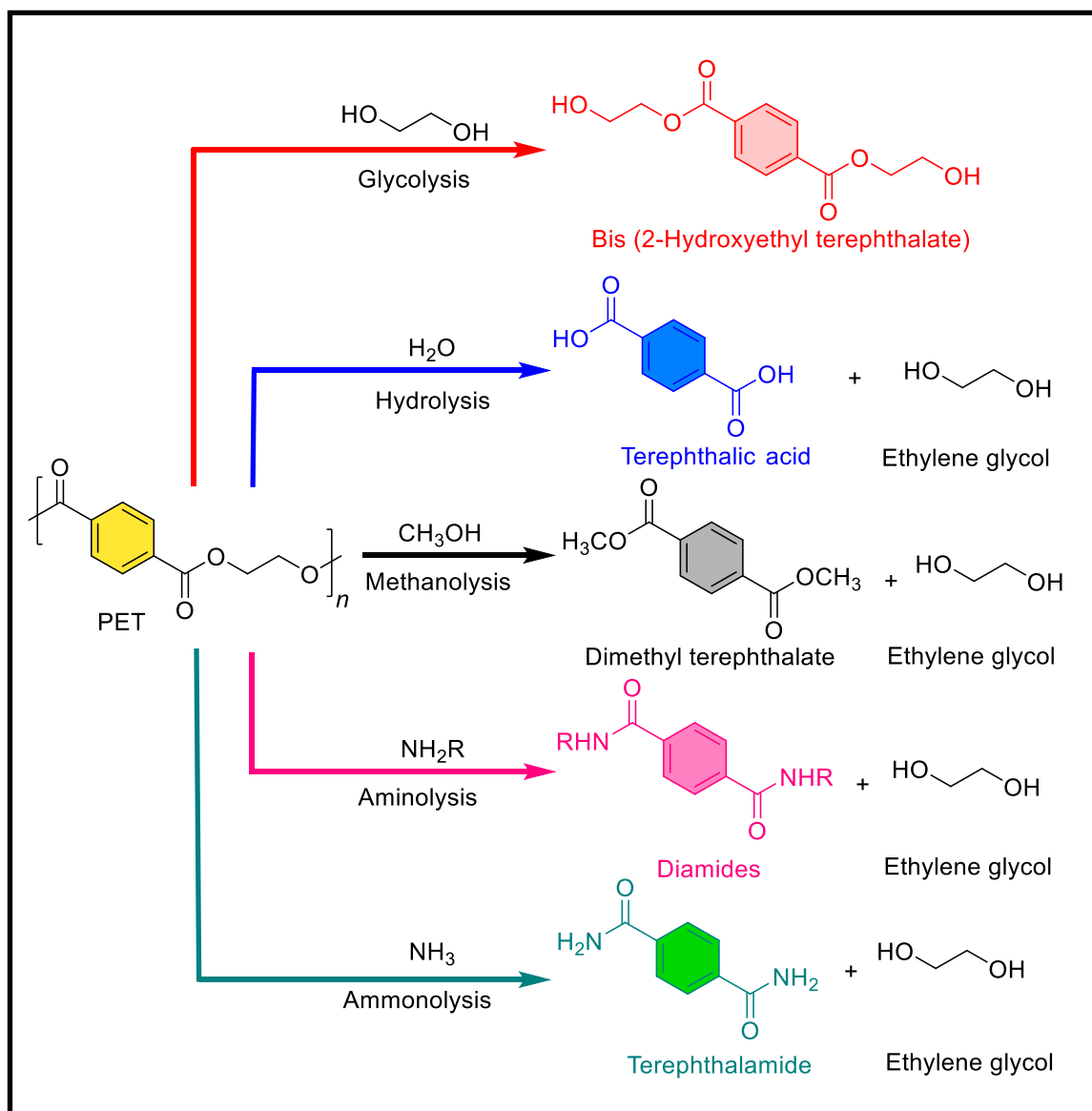


Fig. 1. Chemical recycling processes of PET. Figure adapted from ref. [13,14].

transesterification catalysts for the chemical recycling of PET. We have also emphasized the notable accomplishments, current obstacles, and future strategies for progressing in the area of chemical recycling. However, these processes, including hydrolysis, ammonolysis, and aminolysis, were not reviewed due to their limited commercial applications and lack of extensive study. The novelty of this review can be outlined in the following three key aspects:

i) It presents an overview of recent progress and emerging trends in the development of efficient and stable heterogeneous catalysts for the chemical recycling of PET, addressing the limitations of molecular catalysts, which are often costly and difficult to recover.

ii) Unlike previously published reviews that primarily emphasize polymer degradation methods, this review categorizes catalytic systems based on the nature of their active sites. Each section also incorporates the authors' perspectives and contributions toward designing more robust catalytic materials.

iii) Lastly, the review underscores major breakthroughs, current challenges, and future directions to accelerate advancements in chemo-catalytic PET recycling.

2. Chemical degradation of PET waste

The chemical recycling of PET via glycolysis and methanolysis hinges on catalytic efficiency, which significantly influences the overall process viability in terms of efficiency, scalability, and sustainability. The effectiveness of catalysts—whether including biomass-based materials to advanced systems such as MOFs, zeolites, ionic liquids, mixed metal oxides, and nanomaterials—is influenced by multiple factors, such as:

A. Active site availability and concentration: The availability and nature of active sites significantly influence the efficiency of catalysts used in the glycolysis and methanolysis of PET. For example, utilization of catalysts like ZnO and Fe₃O₄, with mixed metal oxides, can enhance the number of catalytic sites and modify electronic structures, improving their interaction with PET molecules [61].

B. Surface area and porosity: A high surface area and well-defined porosity are indeed beneficial for catalysts, as they enhance contact with PET molecules during glycolysis and methanolysis. Materials like MOFs, with their adjustable porosity, have been actively investigated for PET degradation because of these structural advantages [62].

C. Thermal and Chemical Stability: Catalysts used in the glycolysis

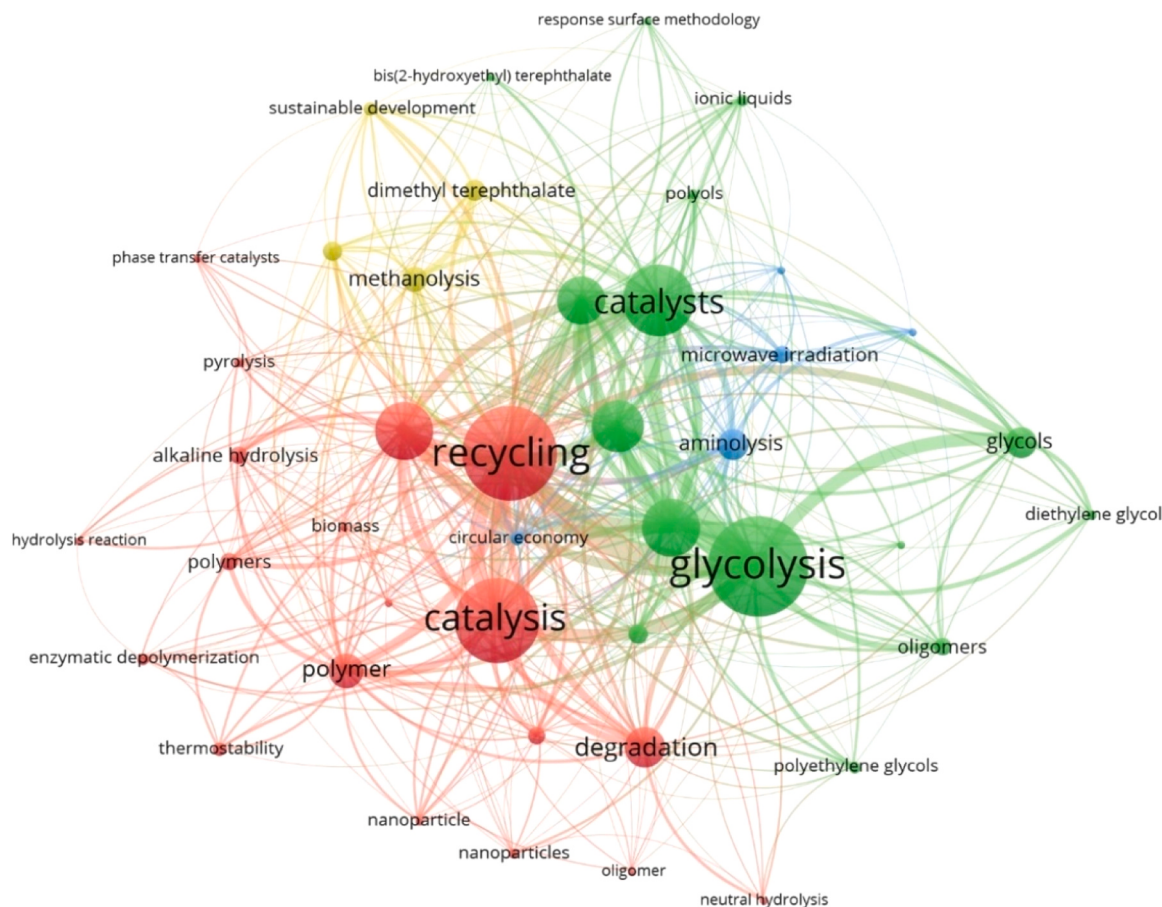


Fig. 2. VOSviewer network visualization of keywords and their relationship to clusters. The bibliometric mapping was created from data indexed in Scopus as of August 14, 2024.

and methanolysis of PET must be able to maintain their structure and function under high temperatures and in the presence of various chemicals. Zeolites are one such example, with Y-type zeolites demonstrating significant yields under specific conditions [63].

D. Acidity or Basicity: The acidity or basicity of a catalyst is indeed a critical factor in determining its catalytic activity in PET depolymerization. For instance, sulfated niobia, which acts as a solid acid catalyst, has been studied for PET depolymerization, and optimizing its calcination temperature and reaction conditions can lead to high yields of bis(2-hydroxyethyl) terephthalate [64].

E. Particle size: The dispersion and particle size of catalysts, especially nanomaterials, significantly influence their performance in PET glycolysis. Smaller nanoparticles generally offer higher surface areas and more active sites, which can lead to enhanced catalytic activity. For example, CeO_2 nanoparticles with a size of 2.7 nm have demonstrated excellent catalytic performance in PET glycolysis, achieving high conversion rates and BHET yields [63].

F. Reusability: The reusability of catalysts is a critical factor determining the economic viability of PET recycling. The ability to recover and reuse catalysts without a significant loss in activity is vital for reducing costs and minimizing waste [65].

2.1. Glycolysis of PET

Among the various chemical recycling routes, glycolysis appears to be the most extensively studied depolymerization techniques [53]. In this process, the breakdown of PET occurs usually at elevated temperatures ranging from 180 to 300 °C. This process necessitates a lengthy reaction duration of 1–8 h in the presence of transesterification catalysts

and glycols as nucleophilic reagents. During the degradation process, the glycol itself diffuses into the PET pellets and caused it to swell up allowing catalyst access to ester bonds, while also promoting catalyst dispersion and potentially enhancing its activity. The desired products afforded by such a process are BHET, low-molecular weight oligomers, and EG [44]. Furthermore, innovative fine-tuning of PET glycolysis through careful modulation of reaction parameters—thermal and pressure settings, catalyst type and concentration, reaction time, and the EG-to-PET ratio—is crucial for achieving economic viability and sustainable process performance. Among the key variables affecting depolymerization, reaction temperature and time hold particular significance. While elevated temperatures may enhance BHET yield, they are associated with increased energy consumption and oligomer formations. Moreover, determining the equilibrium between BHET monomer and dimer is crucial for optimizing product yield.

The glycolyzed BHET monomers can be utilized directly in the current commercial PET production process or employed as a foundational component (via an upcycling process) for the production of polyurethane foams, textile softeners, acrylic coatings, and novel biocompatible synthetic materials [66,67]. The PerPETual factory in India utilizes glycolysis to transform over 2 million post-consumer PET bottles per day into high-quality sustainable filament yarns. These yarns are presently being transported to produce upcycled textiles [16]. Furthermore, recent investigations have shown novel uses for glycolyzed BHET monomers. For instance, Rorrer research groups synthesized glass-fiber-reinforced polymers by combining BHET with bio-origin olefinic acid [68]. A separate study has also reported that the reaction of adipic acid with recovered BHET was able to obtain high-value polyesters [69].

The rate of the non-catalytic glycolysis of PET is quite sluggish, as it demands an activation energy of 32 kcal/mol, which is nearly twice as high as that required for a catalyzed glycolysis process [70]. This non-catalytic procedure generates a mixture of oligo-esters, demanding an intricate separation procedure. In addition, the application of high temperatures (450 °C) and pressures (15.3 MPa) during uncatalyzed glycolysis renders these processes ecologically disadvantageous [71].

Various traditional molecular catalysts, such as metal sulfate [72], metal acetate [73], halides [66], and metal carbonate [74], have been actively employed as transesterification catalysts for the glycolysis of PET. However, metal impurities leaching into the glycolytic product exceed the acceptable limit of 3 mg/L by approximately 7–10 %. Therefore, the remaining metal impurities negatively affect the quality of the subsequent PET product, making it unfit for commercial application [75,76]. Further molecular catalysts, including organometals, ionic liquids, polyoxometalates, and deep eutectic solvents, have also demonstrated their potential usage [77]. However, the task of developing a molecular catalyst that is both economically feasible and capable of efficient separation and recyclability is arduous. This challenge has presented an opportunity for the research community to explore highly stable, capability to function under moderate experimental conditions, and ability to generate high yields of the depolymerized products [78,79].

This section provides a concise overview of the various transesterification catalysts examined for the chemical depolymerization of PET materials.

2.1.1. Biomass waste-derived heterogeneous catalysts

Researchers in the field increasingly embrace catalysts that originate from natural sources, such as biomass, due to their non-toxicity, low corrosiveness, and cost-effectiveness [80,81]. The focus on biomass waste utilization has established a robust foundation for removing hazardous pollutants [82,83], supercapacitor [84], promoting biodiesel production [85,86], facilitating carbon-carbon bond formation [87,88], antibacterial [89], and upcycling plastic waste [90,91]. In addition, the preparation of quantum dots from biomass waste also presents opportunities for diverse applications [92,93].

Biomass-derived activated carbon materials have been widely utilized as catalyst/supports due to their significant porous surface area, adjustable pore structure, and outstanding thermostability [94]. A larger surface area, achieved through smaller particle sizes and high porosity, exposes more active sites for interaction with PET polymer chains, promoting efficient polymer-catalyst interactions and faster reaction speed [95]. However, their synthesis and utilizations can raise environmental concerns, including CO₂ and particulate emissions, reduced reusability from surface contamination, and potential release of polycyclic aromatic hydrocarbons (PAH) if not properly activated.

In 2017, Choi and Kim reported the utilization of carbonized hemp (*Cannabis sativa* L.) as catalyst support and incorporated with vertical MnO₂ nanowires in order to obtain a highly efficient and eco-friendly heterogeneous catalyst for the glycolysis of waste PET [96]. The synthesized composite catalyst, (3D v-MnO₂/HDC), obtained by a hydrothermal method, displayed exceptional catalytic activity due to its large surface area (382.3 m²/g). Under the conditions of 200 °C and a reaction period of 2 h, the catalyst gave an excellent BHET yield (98 %) (see, Table 1, entry 1). However, the recyclability of the catalyst proved challenging. Utilization of calcium oxide (CaO) derived from eggshells and sea food shells as a heterogeneous catalyst for the chemical degradation of post-consumed PET was investigated by Somsok and co-workers [90]. Upon initial investigations of the catalysts, ostrich eggshells calcined at 1000 °C exhibited the highest level of activity. The catalyst was able to obtain a 76.4 % BHET yield within 2 h (see, Table 1, entry 2). The high basicity of ostrich eggshells has a substantial impact on the depolymerization of PET bottles [90].

Recently, there has been an introduction of ash-based solid heterogeneous catalysts derived from biomass combustion for BHET

Table 1

Glycolysis of PET using biomass waste-derived heterogeneous catalysts.

Entry	Catalysts	Reaction condition	BHET yield (%)	Number of cycles reused (yield %)	Ref.
1	3D v-MnO ₂ /HDC	200 °C, 180 min	98	-	[96]
2	Ostrich eggshells	192 °C, 120 min	76.4	-	[90]
3	OPA	190 °C, 90 min	79	5 (62)	[91]
4	BLA	190 °C, 210 min	83	4 (65)	[97]
5	DFPA	190 °C, 90 min	84	5 (71)	[98]

production. In 2021, Vanlaldinpuia research groups reported the synthesis of solid-based heterogeneous catalysts using waste orange peel. The orange peel ash (OPA) catalysts obtained exhibited a typical mesoporous surface area and high basicity due to the presence of oxides and carbonates of calcium (Ca) and potassium (K), as confirmed by different analytical instruments (see Fig. 3). Under the optimized reaction condition, the catalyst was able to obtain as high as 79 % BHET yield (see, Table 1, entry 3). It was assumed that oxides and carbonates of metals serve as an active site during the degradation reaction. These sites deprotonate EG, generating the nucleophile 2-hydroxyethanolate. This nucleophile then attacks the carbonyl carbon in PET's ester bonds to obtain the desired BHET monomer. Furthermore, the catalyst can be reused for up to five catalytic cycles, although each subsequent cycle has a longer reaction time and lower reaction yield. This can be attributed to the leaching of potassium (K) from the catalyst surface [91].

Within the same year, a similar research group employed bamboo leaves, another form of agricultural waste [99], for the preparation of heterogeneous catalysts for PET glycolysis [97]. The bamboo leave ash (BLA) catalyst was obtained by pyrolysis of bamboo leaves at a temperature of 700 °C for 4 h. After the preliminary investigations, the authors reported that a good yield of the product, 83 % yield, was obtained within 3.5 h when the reaction was conducted in the presence of 20 wt% BLA and 16 equiv. EG at 190 °C. The progressive decrease in the number of active sites with each consecutive cycle significantly reduces the reaction yield, reaching a minimum of 65 % yield after the 4th cycle. Under the optimized reaction environments, the glycolysis of red, light yellow, and green colored post-consumed PET bottles gave almost comparable yields to clear PET bottles [97]. Further solidifying their commitment to plastic waste recycling, the same groups recently demonstrated the application of another lignocellulosic biomass waste, dragon fruit peel ash (DFPA), as a solid heterogeneous catalyst. Remarkably, using 4 wt% of the DFPA catalyst and 16 equivalents of EG at 190 °C, yielded an impressive 84 % of the desired BHET monomer in just 1.5 h (see, Table 1, entry 5) [98]. XRF analysis reveals that BLA is predominantly composed of SiO₂ (~78 %), whereas DFPA contains a high proportion of K₂O (~60 %). This compositional difference likely accounts for the superior catalytic performance of DFPA, which demonstrates higher activity within a shorter reaction time and at lower catalyst loading. The elevated K₂O content in DFPA may enhance its basicity and facilitate more effective catalytic activation of the glycolysis process compared to the silica-rich BLA [97,98].

2.1.2. Mixed-metal oxides

The introduction of two metal oxides increases the number of catalytic sites by altering the electronic structure of active metals [100]. This phenomenon intensifies the interaction between the substrate and catalyst, consequently accelerating the reaction rate. In addition, the basicity and structural modification of the mixed metal oxides can be adjusted substantially by altering the aluminium (Al) concentration during the preparation methods [101].

Layered double hydroxides (LDHs), also called anionic clays, were

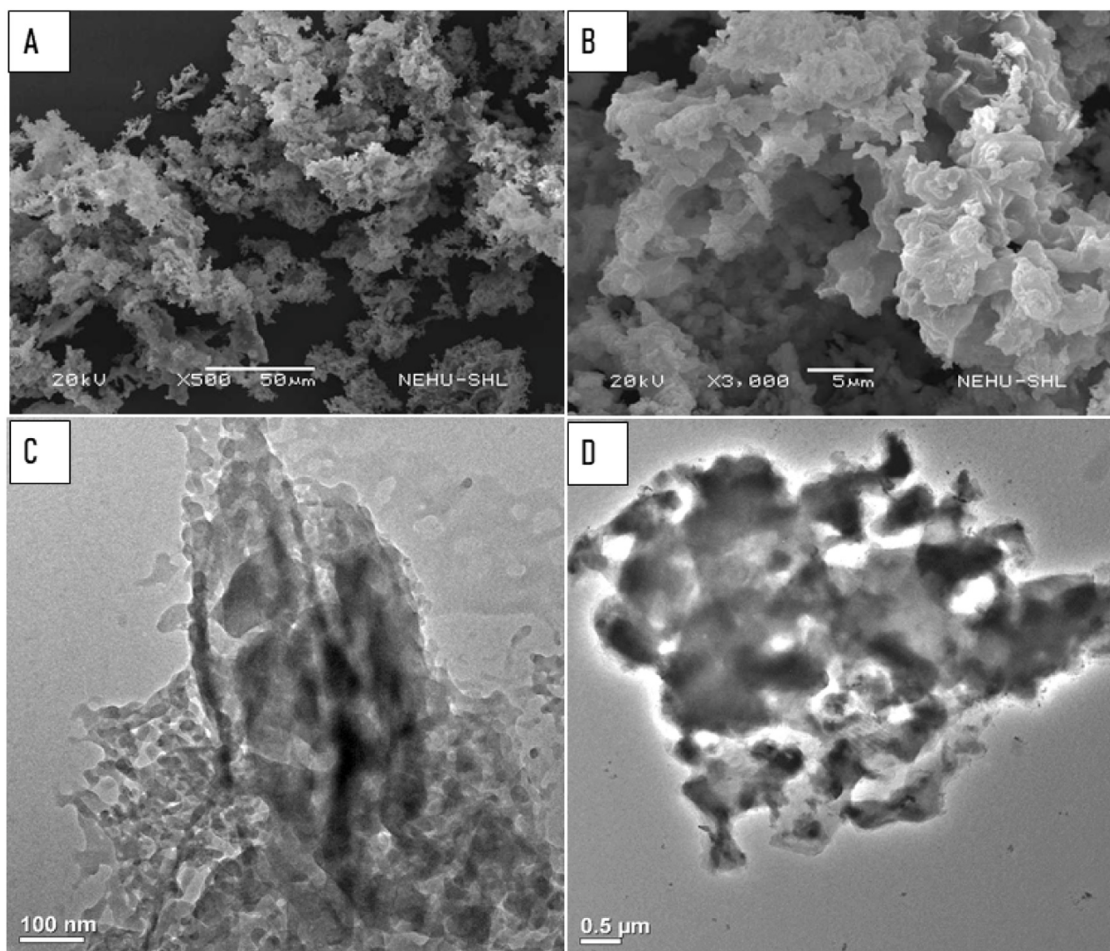


Fig. 3. SEM (A and B) and TEM (C and D) images of OPA [91].

promising candidates for the glycolysis of PET because of their non-toxicity, cost-effectiveness, and eco-friendly preparative steps [109]. The temperature of calcination and ratios of the metal have significantly influenced the performance of such LDHs [110]. The pioneering study conducted by Yang and co-workers in 2012 established the application of calcined Mg-Al hydrotalcites (Mg/Al LDHs) for chemical depolymerization of PET [102]. The coprecipitated hydrotalcites catalysts of different molar ratios of Mg/Al (in a ratio of 2, 3 and 4) were calcined at different temperatures ranging between 300 and 800 °C to obtain the desired Mg/Al LDHs. These calcined hydrotalcite catalysts were labeled as CHT-2, CHT-3, and CHT-4, corresponding to Mg/Al molar ratios of 2, 3, and 4, respectively. The CHT-3 catalyst calcined at 500 °C was found to have the highest basicity, however, increasing the calcination temperature decreases this property. After conducting the initial investigation, it was found that among the catalysts tested, CHT-3 (Mg/Al = 3) exhibited the highest catalytic performance, achieving an 81.3 % yield at 196 °C in 50 min. In comparison, CHT-2 and CHT-4 produced yields of 76.2 % and 65.6 %, respectively, under same experimental conditions as can be seen in Table 2, entries 1–3. Furthermore, CHT-3 demonstrated excellent reusability, maintaining its catalytic activity over three successive reaction cycles without significant loss of efficiency.

The size of the mixed Mg-Al oxides is another determining factor for PET glycolysis. The catalytic efficiency of a granular catalyst, with a diameter of 500 μm, outperforms that of a larger pellet prepared using coprecipitation and wet-mixing technique [111]. At 220 °C, 80 % BHET yield can be obtained in the presence of small granules, which is 40 % higher than with pellet catalyst. However, both catalysts showed similar catalytic efficiency at elevated temperatures. After being reused four

Table 2

Glycolysis of PET using mixed metal oxides catalysts.

Entry	Catalysts	Reaction Condition	BHET yield	Ref.
1	CHT-2	196 °C, 50 min	76.2	[102]
2	CHT-3	196 °C, 50 min	81.3	[102]
3	CHT-4	196 °C, 50 min	65.6	[102]
4	Mg-Al-O@Fe ₃ O ₄	240 °C, 90 min	80	[103]
5	ZnAl-3	196 °C, 85 min	76.4	[104]
6	(Mg-Zn)-Al LDH	190 °C, 180 min	75	[65]
7	S/Zn-Ti-300 °C	180 °C, 180 min	72	[105]
8	(SO ₄ ²⁻ /Co ₃ O ₄)	180 °C, 180 min	72	[106]
9	ZnMn ₂ O ₄	260 °C, 60 min	92.2	[107]
10	CoMn ₂ O ₄	260 °C, 60 min	89	[107]
11	ZnCo ₂ O ₄	260 °C, 60 min	81	[107]
12	CoFe ₂ O ₄ /C10-OAc	195 °C, 150 min	95.4	[108]
13	ZnAl-CO ₃	190 ± 5, 180 min	79.34	[58]
14	ZnAl-CO ₃	190 ± 5, 180 min	79.34	[60]
15	ZnTi-CO ₃	190 ± 5, 180 min	83.74	[59]

times, the pellet catalyst became deactivated as a result of the absorption of glycolyzed BHET and its oligomers on the catalyst surface. Subjecting the pellet catalyst to high temperatures can reactivate it to its virgin state. The glycolyzed BHET monomer was repolymerized with Sb₂O₃ as a catalyst to obtain rPET fiber, which has comparable spinnability and physical characteristics to virgin PET (see Fig. 4).

In 2020, Bahramian reported that dispersing Mg-Al LDHs with 20 wt % titania nanoparticles (NP's) using plasma and thermal routes further enhanced the catalyst's thermal stability and efficacy [112]. In particular, the LDH-FD-DBD catalyst, which underwent treatment with

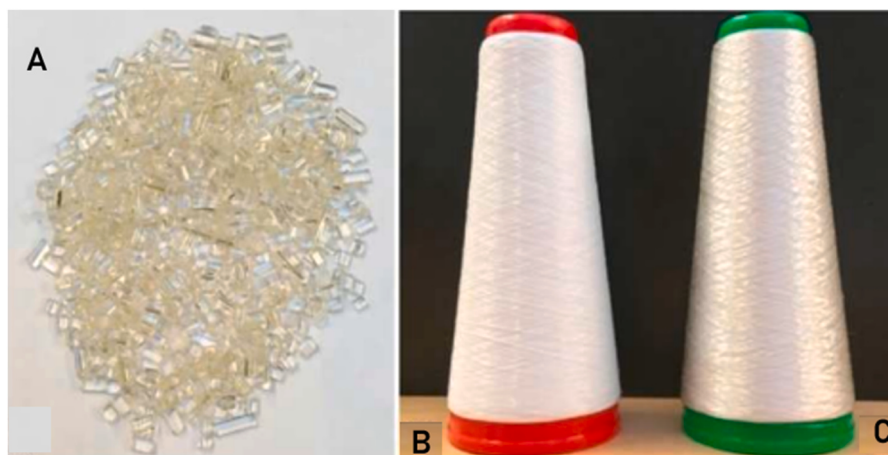


Fig. 4. A. Synthesized r-PET pellets. B. Virgin fibers of PET contain 0.4 wt% TiO₂ and C. r-PET fibers [111].

dielectric-barrier discharge plasma and freeze-drying methods, demonstrated higher activity compared to the catalyst undergoing the calcination process. The high catalytic performance towards PET degradation as well as its excellent reusability can be attributed to the high dispersion rate of the titania NPs on the catalyst surface.

The next year, Guo et al., prepared Mg-Al-O@Fe₃O₄ microparticles by coating nanosized Mg-Al double oxides into Fe₃O₄ microparticles, promoting efficient and sustainable routes for PET depolymerization [103]. The hierarchically structured Mg-Al-O@Fe₃O₄ catalysts possess a high surface area and synergistic metal-metal activations, enabling the formation of BHET monomers in good yield (80 %, see Table 2, entry 4). However, the microparticles experience rapid deactivation after two consecutive cycles as a result of the blockage of active sites by the glycolized PET products. Heat treatment of the catalysts at 1000 °C reactivates the catalysts, which is undeniably an energy-intensive step.

The basicity and surface area of another hydrotalcite, Zn-Al LDHs, were observed to be significantly influenced by its calcination temperature [104]. Although the addition of Al³⁺ diminishes the basic sites of the mixed metal oxides, an increase in the calcination temperature increases their basicity. The Zn-Al hydrotalcite that was calcined at 500 °C (ZnAl-3 catalyst) has the most abundant basic sites and resulted in a greater yield of BHET (76.4 %, see Table 2, entry 5) compared to all other catalysts studied. The acidity and basicity of the mixed oxide catalyst can be further improved by insertion of the Mg²⁺ cation during the preparation process [113]. Eshaq and ElMatwally also reported the preparation of regenerable (Mg-Zn)-Al LDHs catalysts by the coprecipitation method at low supersaturation conditions [65]. The addition of 1 wt% of catalyst to a mixture of 20 g of EG and 2 g of PET at a reaction temperature of 196 °C resulted in the complete depolymerization of post-consumed PET within 3 h, yielding 75 % BHET monomer (see Table 2, entry 6).

PET glycolysis showed significant catalytic efficiency when using sulfate-incorporated metal oxides. Zhu et al., prepared a spectrum of sulfated solid catalysts containing SO₄²⁻/ZnO, SO₄²⁻/TiO₂, and SO₄²⁻/Zn-TiO₂ by precipitation or coprecipitation techniques [105]. The catalysts can be obtained by calcination at temperatures ranging from 200 to 600 °C. After the initial investigation, the surface area and acidic sites of the mixed metal oxides (S/Zn-Ti-200–300 °C) were higher than both the single oxides (S/Zn and S/Ti). Specifically, the S/Zn-Ti-300 °C has demonstrated the best result, obtaining 72 % BHET yield with full PET conversion (see Table 2, entry 7). This can be attributed to the catalyst's mild acidic properties, its amorphous structure, and its high surface area. Later, the same research team demonstrated the preparation of another metal oxide, sulfated cobalt oxide (SO₄²⁻/Co₃O₄) and zinc-modified sulfated oxide (SO₄²⁻/Co-Zn-O), for the same depolymerization reaction [106]. However, there was no substantial change in

the BHET yield under the reported optimized reaction environments (see Table 2, entry 8).

In another study, mixed-oxide spinel of the general formula AB₂O₄ has been investigated for the depolymerization of PET via glycolysis. Imran et al., reported the preparation of ZnMn₂O₄, CoMn₂O₄, and ZnCo₂O₄ catalysts via precipitation and coprecipitation techniques [107]. A preliminary investigation was conducted to examine the influence of various reaction parameters, such as reaction temperature, time, and EG/PET ratios, on the yield of BHET monomer. The mixed metal oxide spinel exhibited superior activity compared to its single metal oxide counterparts due to their higher surface area and greater concentration of the acidic sites. Additionally, the catalytic performance of the spinel catalysts is greatly under the influence of factors such as structure, geometry, and the type of metal cations present. The tetragonal ZnMn₂O₄ spinel demonstrated remarkable activity when compared to the activity of CoMn₂O₄ and ZnCo₂O₄ spinels, obtaining an excellent 92 % BHET yield at 260 °C within 60 min (see Table 2, entries 9–11). A potential explanation for this phenomenon is the larger surface area and abundance of both mild and strong acidic sites. Additional factors, such as the specific spinel geometry and the coordination of tetragonal/octahedral coordination, could potentially influence the observed catalytic activity [107].

However, despite the author's commendable efforts, the reusability experiment of the active tetragonal spinel catalyst was not conducted, and the PET glycolysis relied heavily on a high reaction temperature. Recently, Chen research groups have prepared a series of ionic liquid and employed as surfactants to modify CoFe₂O₄ spinel mixed oxide (see Fig. 5) [108]. Following the initial examination, it was observed that the CoFe₂O₄ spinel catalyst modified with ionic liquid (CoFe₂O₄/C10-OAC) achieved a higher BHET yield of 95.4 % in compared to the unmodified CoFe₂O₄ and pure ionic liquids. Notably, the catalyst can be easily recovered using an external magnet field. The recovered spinel catalyst could be reused for 10 times without any significant loss in its catalytic activity.

Recently, Kumari et. al., reported the synthesis and evaluation of LDHs incorporating various metal ions (M = Zn, Mn, Pb, Cd, Mg, Ni, Co, Ca, Cu) in the form of MAI-CO₃ [58]. These materials exhibit a range of desirable properties, including tunable acidic and basic sites, selective adsorption behavior, anion exchange capacity, adjustable metal composition, biocompatibility, layered morphology, and promising catalytic activity. The catalytic efficiency of different LDH compositions for PET glycolysis followed the trend: Zn > Mn > Li, Pb > Cd > Mg > Ni > Co > Ca > Cu. Under optimized conditions, a BHET yield of 79.34 % was achieved using 0.02 g of ZnAl-CO₃ catalyst after 180 min of reflux, with a crystallization time of 72 h (see Table 2, entry 13). Notably, the glycolysis of colored PET waste yielded the same amount of BHET as

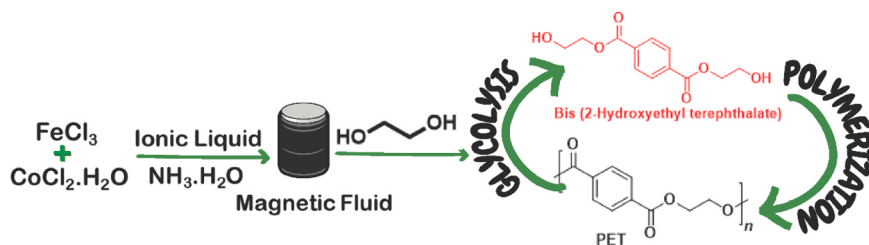


Fig. 5. Glycolysis of PET over modified CoFe_2O_4 magnetic nanoparticle. Adapted from ref. [108] Copyright (2021), with permission from Elsevier.

transparent PET, producing needle-like BHET crystals. Furthermore, recyclability studies demonstrated that the ZnAl-CO_3 catalyst could be reused up to eight times without any loss in catalytic efficiency, maintaining a consistent BHET yield of 79.34 %. In the same year, the research team further reported the synthesis of ZnAl , MgAl , and NiAl LDHs containing various interlayer anions (chloride, nitrate, and carbonate) via the direct co-precipitation method [60]. These LDHs exhibit a layered structure bearing a net positive charge, which is counterbalanced by the intercalated anions, opposite to the structure of conventional cationic clays. Among the synthesized variants, ZnAl-CO_3 LDH demonstrated the highest catalytic activity in the glycolysis of PET, achieving a BHET yield of 79.34 %. (see Table 2, entry 14). These LDH materials are considered promising inorganic catalysts due to their non-toxic nature, cost-effectiveness, and tunable structural features. In a subsequent study, the same research team synthesized ZnM^{IV} - and ZnAlM^{IV} -based LDHs via a one-pot co-precipitation method, followed by calcination to obtain their corresponding mixed metal oxides (MMOs) [59]. These catalysts exhibited notable activity in the glycolytic depolymerization of PET waste. The catalytic performance of the materials followed the order: $\text{ZnTi-LDH} > \text{ZnAlTi-LDH} > \text{ZnTi-MMO} > \text{ZnAlZr-LDH} > \text{ZnAlTi-MMO} > \text{ZnZr-LDH}$. As shown in Table 2, entry 14, ZnTi-CO_3 showed the highest efficiency, achieving a maximum BHET yield of 83.74 %, and remained catalytically stable for up to nine consecutive cycles with only a slight decline in performance. Top of Form Bottom of Form

2.1.3. Porous materials

Zeolites are microporous, aluminosilicate minerals with a remarkable and versatile structure. Despite being renowned for their wide range of applications, including adsorption, catalysis, and ion exchange processes, zeolites have not been thoroughly studied for PET glycolysis. In 2008, Shukla research groups reported the first example of zeolites promoting PET glycolysis [114]. This study explored the potential of two natural zeolites, β -zeolites and Y-zeolites, as transesterification catalysts for depolymerizing waste PET. Both the zeolites positively promoted the reaction under mild conditions. Particularly, Y-zeolites obtained a promising 65 % BHET yield in 8 h at 196 °C reaction temperature, probably due to their favorable structural properties, including a high Si/Al ratio and large mesoporous surface area. While these initial results exhibited the potential of natural zeolites for PET degradation, further research is crucial to gain a deeper understanding of the reaction mechanisms. Efforts should be made to optimize the favorable reaction conditions in order to shorten reaction durations, lower reaction temperatures to enhance energy efficiency, and minimize the formation of byproducts.

Recent research highlights the exciting potential of zeolite-supported heterogeneous catalysts for efficient and effective PET depolymerization [115]. In particular, doping a zeolite support with an optimal amount of zinc oxide (ZnO) has shown remarkable outcomes, outmatching other SBA-15 incorporated metal oxide catalysts. The SBA-15 systems provide high acidic sites, a large surface area, and a significant pore volume. These characteristics endorse excellent dispersion of the active catalyst component, thus enabling reactant adsorption. This catalyst obtained an excellent 91 % BHET yield at 197 °C. Further exploration of natural

bimetallic zeolites presents another promising avenue for the progression of catalytic PET glycolysis.

Other promising PET glycolysis catalysts have been identified, such as nanoclays, clays and microporous structured materials. In a study conducted by Guo et al., Perkalite F100, a synthetic nanoclay composed of Mg–Al layered double hydroxides, was identified as an effective transesterification catalyst for the glycolysis of PET waste [116]. Under optimized conditions (0.5 wt% catalyst loading, 240 °C, 2 h), the reaction yielded over 80 mol% of BHET. The nanoclay's layered nanostructure provides a high specific surface area, which contributes significantly to its catalytic activity. Notably, Perkalite F100 is an environmentally benign material capable of depolymerizing PET with efficiency and selectivity comparable to that of zinc acetate. Vajiravelu et. al., also reported the effective depolymerization of PET using kaolin- and bentonite-supported catalysts containing Lewis acidic species (Zn^{2+} and Al^{3+}) and heteropolyacids (phosphotungstic acid, PWA, and phosphomolybdic acid, PMA) [117]. The catalytic performance of 5 wt% loadings of Zn^{2+} , Al^{3+} , PWA, and PMA on both clay supports was evaluated. Among the two clays, bentonite demonstrated superior performance, particularly in accommodating heteropolyacids, and resulted in higher BHET yields. Complete PET depolymerization was achieved within the temperature range of 180–210 °C, yielding BHET in the range of 78–90 %.

Mesoporous materials, owing to their tunable surface functionalities and well-defined pore formations, facilitate the design of catalysts with improved active site dispersion and pronounced confinement effects. Recently, Yang and Xu's research group reported that a ZnO-supported a mesoporous silica molecular sieve catalyst (5 wt% ZnO/KIT-6), exhibited high catalytic efficiency in the glycolysis of PET, affording a BHET yield of 92.1 % with complete PET conversion at 196 °C over 3 h [118]. Structural characterization revealed that ZnO nanoparticles were uniformly dispersed within the mesoporous channels of KIT-6 and formed strong interactions with surface Si–OH groups, which enhanced the acid strength and increased the density of acid sites, thereby promoting PET depolymerization.

2.1.4. Metal-organic framework catalysts

Metal-organic frameworks (MOF) are an exciting new class of crystalline, non-porous materials attracting significant attention in various research fields [119]. MOF exhibit exceptionally high internal pore volume and external surface area, along with excellent compatibility with organic substrates (such as PET and ethylene glycol), resulting in enhanced catalytic activity for efficient glycolysis [120–122]. However, despite ongoing research focused on developing hybrid MOFs with improved stability, anchoring them onto solid supports for better recyclability, and optimizing post-synthetic modifications, their widespread industrial adoption remains challenging compared to more cost-effective and scalable traditional metal oxide catalysts.

Among the various MOF's reported, zeolite imidazolate frameworks (ZIF-8) stand out, primarily due to their unique combination of zeolite-like structure and impressive thermostability. In 2017, Suo research groups pioneered the utilization of a ZIF-8 catalyst, $[\text{Zn}(\text{min})_2]$ (min = 2-methyl-1H-imidazole), for glycolysis of post-consumed PET (see Fig. 6) [120]. Despite the notable innovations in the exploration of MOFs

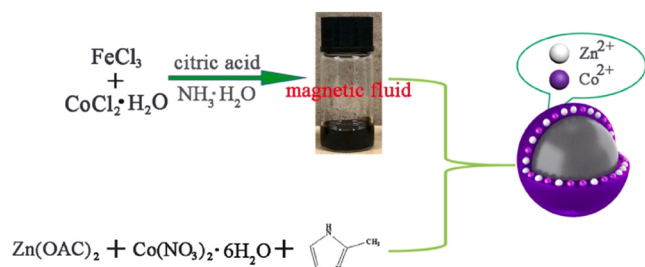


Fig. 6. Synthesis of CoFe₂O₄@ZIF-8/ZIF-67 catalyst [120].

for chemical recycling, the ZIF-8 catalyst obtained a lower BHET yield (68.1 %) compared to traditional metal chloride transesterification catalysts. This difference is likely due to the limited pore size of the ZIF-8 catalyst, which hinders efficient interaction with PET polyesters.

The application of ZIFs as catalyst support presented a promising platform for improving the yield of BHET monomers. For example, Chen research groups reported that bimetallic ZIFs were more reactive than single-metal ZIFs for the promotion of PET glycolysis [123]. In this study, CoFe₂O₄@ZIF-8, CoFe₂O₄@ZIF-67, and CoFe₂O₄@ZIF-8/ZIF-67 composite catalysts were prepared using a one-pot, room temperature method. Among the synthesized MOFs tested, CoFe₂O₄@ZIF-8/ZIF-67 demonstrated superior catalytic activity, obtaining an 85 % BHET yield with complete conversion of the mixed polyesters. The catalytic activity of the catalyst could maintain its efficacy for up to five subsequent cycles, with no significant loss in BHET yield (< 10 %). The authors attributed the performance variances among the prepared catalysts to disparities in their surface area. Based on this observation, further exploration of the activity of ZIF-8 crystal size (ranging from 100 to 1600 nm) can be carried out. This investigation is crucial as crystal size can significantly influence the rate of reaction, selectivity, as well as overall catalytic efficacy.

Immobilizing deep eutectic solvents (DESs) on ZIF supports provides a compelling approach for further improvement of the ZIF catalysts for PET degradation. Chen research group in 2021 explored this strategy by synthesizing DES@ZIF-8 composite catalyst by immobilizing ZIF-8 on DESs comprised of acetamide and various metal salts [124]. After careful optimization of the reaction parameters, such as reaction time, temperature, amounts of EG, and catalyst concentration, they obtained an excellent result: an 82.3 % BHET yield with 100 % PET conversion. During the reaction, the nitrogen atom in acetamide's amino group forms a hydrogen bond with the hydroxyl group of EG, increasing the electronegativity of the hydroxyl oxygen. Concurrently, Zn²⁺ ions from ZnCl₂ coordinate with the carbonyl oxygen in PET, enhancing the positive charge on the carbonyl carbon. This dual activation makes the carbonyl carbon more susceptible to nucleophilic attack by the hydroxyl oxygen of EG, facilitating the formation of BHET (see Fig. 7). Reusability experiments of the catalyst obtained 79.3 % BHET yield after the six cycles.

Metal-azolate framework-6 (MAF-6), a subclass of MOFs, also demonstrated excellent catalytic activity compared to ZIF-based MOFs for PET glycolysis. This improved performance has originated from the larger pore size displayed by the MAF-6s, which enables the diffusion of the BHET dimer intermediate into the porous surface, allowing access to the catalyst's active sites [125]. Wu research groups recently experimented the remarkable activity of MAF-6, obtaining a 92 % PET conversion and an 82 % BHET yield at 180 °C within 4 h. The BHET dimer readily diffuses through the larger pores of MAF-6's, thus leading to a high BHET monomer yield. On the contrary, smaller pores exhibited by the MAF-5 and nonporous MAF-32 demonstrated significantly lower catalytic activity, highlighting the importance of the pore size effect [121].

2.1.5. Nanomaterials

Nano-sized heterogeneous catalysts offer remarkable catalytic

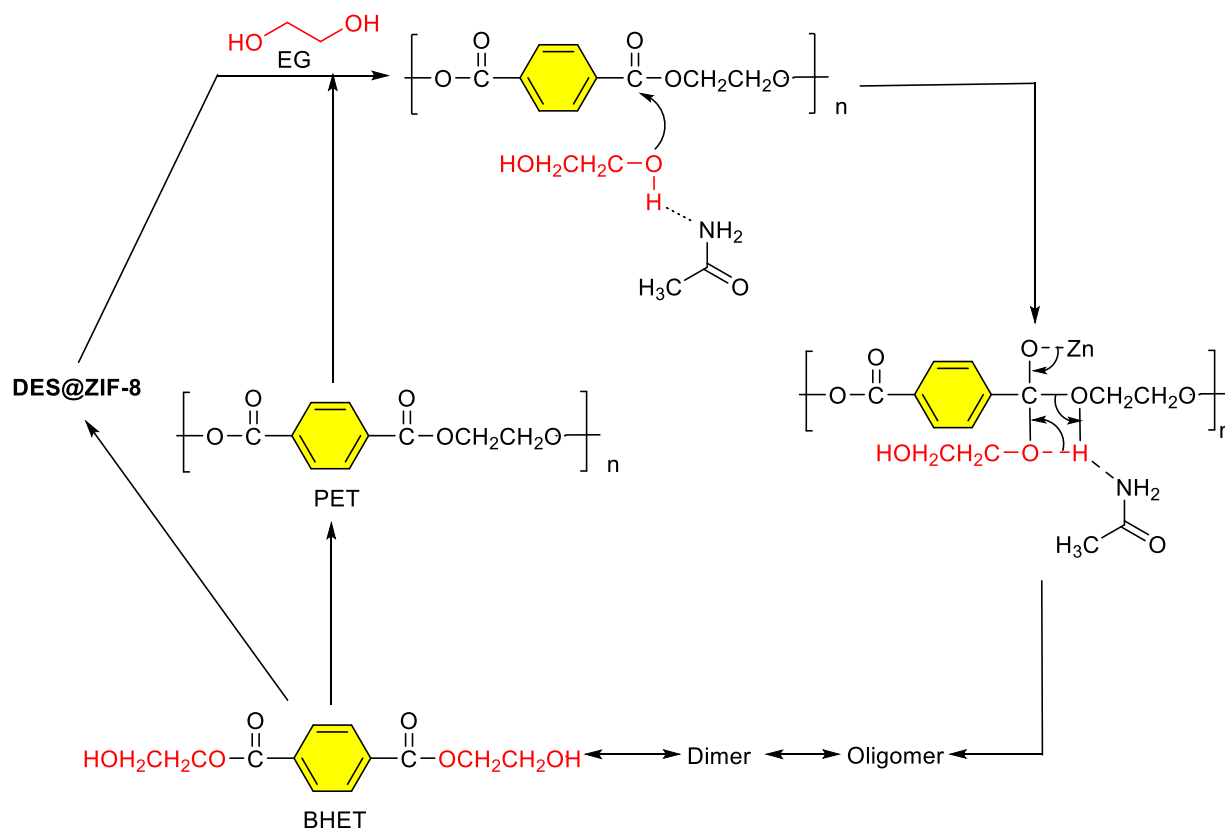


Fig. 7. Mechanism of DES@ZIF-8 promoted PET glycolysis from ref. [124] with permission from Elsevier.

performance due to their high surface area and increased active site density [126]. In addition, the characteristics of the catalysts get altered at the nanoscale, leading to enhance catalytic efficiency for more accessible active sites. For efficient glycolysis of waste PET products, oxide nanoparticles (ONPs) have been effectively employed either alone or as catalyst support for promoting better dispersion of the active metals [127]. The size of the nanocatalysts plays an important role in determining the yield of the BHET monomer. For example: due to their larger specific area and pore size volume, n-ZnO NPs (55 nm) obtained good to excellent BHET yield in contrast to micro-sized m-ZnO (94 nm) (see Table 3, entry 1) [128]. Imran and coworkers demonstrated in 2013 the preparation of ZnO, Mn₃O₄, and Co₃O₄ metal-oxide NPs by a simple precipitation method [107]. In this report, among the tested catalysts, Mn₃O₄ NPs with the smallest particle size (31 nm) gave the best result, obtaining 74 % BHET monomer, further emphasizing the importance of nanoscale design for efficient PET degradation (see Table 3, entries 2–4).

Capping agents such as tannic acid and citrate have been actively involved in controlling the size and agglomeration of the prepared nanoparticles. Renaldi et. al., successfully reported the synthesis of ultrasmall cobalt NPs (approximately 3 nm) using tannic acid as a capping agent and a borohydride reduction method [129]. The prepared catalysts demonstrated efficient glycolysis of PET, enabling a high monomer yield of 77 % at a relatively low reaction temperature of 180 °C (see Table 3, entry 5). A plausible reaction mechanism was outlined in which cobalt ions undergo interaction with the free electron of the PET carboxyl ions, leading to the delocalization of the electron. This increases the susceptibility of the PET's carbonyl group to nucleophilic attack by the EG. The crucial role of tannic acid lies in its ability to increase BHET yield by interacting with EG through its hydroxyl (OH) group. The organic spherical structure composed of tannic acid allows for good dispersion of cobalt nanoparticles, resulting in high BHET selectivity. In addition, the authors acknowledge that other factors such as the type of metal cations and their coordination structure (tetrahedral or octahedral) can facilitate the catalytic activity.

In accordance with the principle of sustainable development, researchers practiced the valorization of electronic waste into efficient transesterification catalysts. The application of recovered zinc (RZnO) and cobalt oxide (RCoO) from used alkaline and lithium-ion batteries as a transesterification catalyst was reported by Fuentes research groups (see Fig. 8) [130]. Initial investigation revealed that the RZnO catalyst exhibited higher catalytic activity, obtaining a 50 % BHET yield within

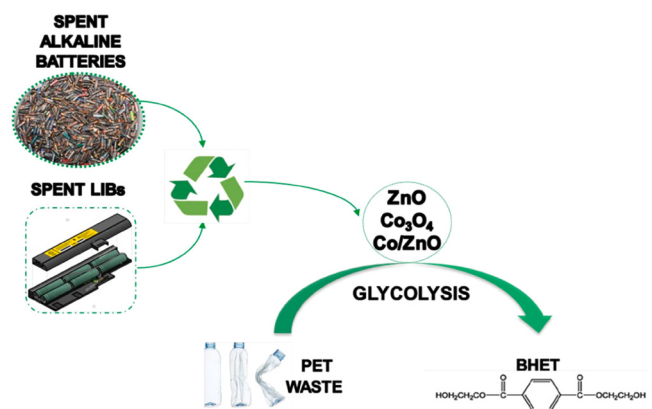


Fig. 8. PET waste glycolysis reaction using recovered ZnO and CoO catalysts. Reproduced from ref. [130].

2 h at 196 °C (see Table 3, entries 6 and 7). This performance can be attributed to the larger specific surface area and smaller particle size of the RZnO compared to the RCoO catalyst. However, further optimization of the reaction is required in order to improve the desired product yield. In another interesting report, magnetically recyclable iron-based catalysts have emerged as attractive candidates due to their large surface area, ease of recoverability, and reusability [53]. Kim et al., demonstrated the first example of magnetically recyclable iron-based nano-catalysts, superparamagnetic γ -Fe₂O₃ NPs, for the promotion of PET glycolysis [131]. After exhaustive preliminary investigation, the authors reported that high reaction temperatures offer efficient PET conversion, while shorter reaction times decrease the potential formation of side-products. Under favorable conditions, a 90 % BHET yield with 100 % PET conversion can be achieved within 1 h at 300 °C (see Table, entry 8). The catalyst's high surface area, excellent crystallinity, and the capability to undergo PET glycolysis through redox processes contributed to its excellent performance. In addition, the nanocatalyst can be reused for up to 10 consecutive runs without a decrease in its catalytic activity.

Layered iron nanosheets have emerged as a promising new class of heterogeneous catalysts for the glycolysis of PET [132]. Researchers have successfully reported the synthesis of ultrathin Fe^{III} and Fe^{II}/Fe^{III} and Cl-Fe^{II}/Fe^{III} LDH nanosheets using the fluid dynamics-induced shear exfoliation technique (see Fig. 9). These nanosheets displayed a thin layered two-dimensional morphology with an ordered hexagonal structure. Remarkably, the synthesized nanosheet catalysts were able to afford 100 % BHET yield with complete conversion of PET at a temperature of 200 °C (see Table 3, entry 9). Notably, ultra-thin Fe^{III} demonstrated superior catalytic performance, demanding a shorter reaction duration and excellent reusability, maintaining 96.5 % yield after the fifth cycle. This remarkable performance can be attributed to the catalyst's ability to promote glycolysis through redox reactions, its large surface area, and the abundance of active sites.

Examples of two-dimensional layered materials such as multiwalled nanotubes (MWCNTs), graphene oxide, hexagonal boron nitride, and titanate nanotubes were proven to be highly efficient for anchoring metal NPs that deliver unique structural characteristics and bifunctional properties to the nano-composite catalyst [144]. The synthesis of stable Fe₂O₃-boosted MWCNTs was reported by ElMetwally research groups for promoting glycolysis of PET (see Table 3, entry 10) [133]. Excellent BHET yield (100 %) could be obtained under the optimized reaction condition. The synergistic effect existing between the magnetite and MWCNTs has greatly improved the degradation of waste PET. In another study, the preparations of titanate nanotubes (TNT) were successfully reported by Lima et al., in 2017 [134]. In comparison with commercially available zinc acetate catalysts, the titanate nanotubes obtained a slightly higher BHET yield (84 %) (see Table, entry 11). Later, by

Table 3
Glycolysis of PET over nanomaterials.

Entry	Catalysts	Reaction Condition	BHET yield (%)	Ref
1	n-ZnO	190 °C, 60 min	90	[128]
2	ZnO	260 °C, 60 min	67	[107]
3	Co ₃ O ₄	260 °C, 60 min	63	[107]
4	Mn ₃ O ₄	260 °C, 60 min	74	[107]
5	Co NPs	180 °C, 180 min	77	[129]
6	RZnO	196 °C, 120 min	50	[130]
7	RCoO	196 °C, 120 min	10	[130]
8	γ -Fe ₂ O ₃	300 °C, 60 min	90	[131]
9	Fe ^{III} nanosheet	200 °C, 30 min	100	[132]
10	Fe ₂ O ₃ -MWCNT	190 °C, 120 min	100	[133]
11	TNT	196 °C, 120 min	84	[134]
12	NaTNT	196 °C, 180 min	80	[135]
13	ZnTNT	196 °C, 180 min	87	[135]
14	Go-Mn ₃ O ₄	300 °C, 80 min	96.7	[136]
15	MnO ₂ /HGO	200 °C, 10 min	100	[137]
16	γ -Fe ₂ O ₃ NGO	195 °C, 180 min	100	[138]
17	Pd/h-BN	100 °C, 30 min	92.1	[139]
18	Fe ₃ O ₄ NPs@h-BNNS	200 °C, 300 min	100	[140]
19	Fe ₃ O ₄ @SiO ₂ @(mim) [FeCl ₄]	180 °C, 1440 min	100	[141]
20	rGO/[TESPMI] ₂ CoCl ₄	190 °C, 180 min	95.2	[142]
21	MnO _x -500	180 °C, 180 min	86	[143]

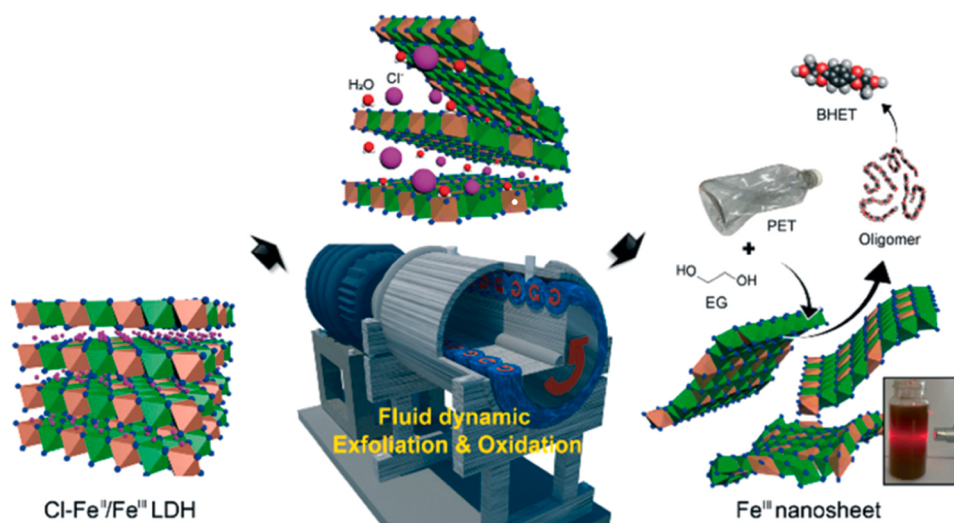


Fig. 9. Fabrication process of layered Fe^{III} nanosheets via fluid dynamics-induced exfoliation (FDSE) method and the catalyzed PET glycolysis reaction. (a) Reproduced from ref. [132] Copyright (2021), with permission from Royal Society of Chemistry.

adopting a facile single-step hydrothermal process, the same research group demonstrated the modification of TNT with sodium (NaTNT) and zinc (ZnTNT) [135]. In this report, the ZnTNT catalyst exhibited higher catalytic activity, achieving an 87 % BHET yield in 3 h at 196 °C (see Table 3, entries 12 and 13).

Graphene oxide (GO) has attracted significant attention as a support material for developing nanocomposite catalysts for the glycolysis of PET. In a pioneering study conducted in 2012, Park research groups described the preparation of $\text{GO-Mn}_3\text{O}_4$ nanocomposite using a facile one-step sonochemical method [136]. As described in see Fig. 10, potassium permanganate (KMnO_4) is reduced to Mn_3O_4 after the oxidation of carbon atoms within the GO support. Although this process obtained an excellent BHET yield of 96.7 %, it demands a relatively high reaction temperature (300 °C) (see Table 3, entry 14). In order to address the high energy demand and further improve BHET yield, Choi et al., developed a nanoporous MnO_2/HGO catalyst [137]. The catalyst was prepared by incorporating an ultra-thin layer of MnO_2 into the GO nanosheet via a solution-based self-limiting reaction. The catalyst was able to afford a 100 % BHET yield with full PET conversion within just 10 min at a lower temperature (see Table 3, entry 15). This remarkable performance was due to the presence of abundant active sites on the catalyst surface, which arise from the interaction between GO and nanoporous MnO_2 .

The incorporation of a nitrogen functional group within the carbon framework of graphene altered the nature of the active catalytic sites and enhanced the electrical conductivity as well as its surface hydrophilicity [145]. Capitalizing on these advantages, Nabid research groups demonstrated the development of a bifunctional superparamagnetic $\gamma\text{-Fe}_2\text{O}_3$ -nitrogen-doped graphene hybrid catalyst, which produced a 100 % BHET yield at 195 °C (see Table 3, entry 16) [138]. This exceptional performance stems from the synergistic effects between the $\gamma\text{-Fe}_2\text{O}_3$ and N-doped graphene material. In addition, the availability of abundant unpaired electrons from the defective sites significantly influenced the catalyst's activity.

In different areas of material sciences, hexagonal boron nitride (h-BN), which is a structural analogue of graphene, has been effectively employed due to its cost-effectiveness, thermostability, excellent chemical inertness, and mechanical strength [146]. The preparation of h-BN nanosheets does not entail combustion in the presence of air and does not absorb light in the visible region; therefore, it is sometimes known as white graphene [147]. The existence of B and N functional groups on the surface of h-BN nanosheets offers remarkable chemical coordination conditions suitable for anchoring metal NPs. This characteristic has prompted researchers to explore h-BN nanosheets as catalysts support for PET chemical recycling [148].

Recently, Kim research groups have explored the application of

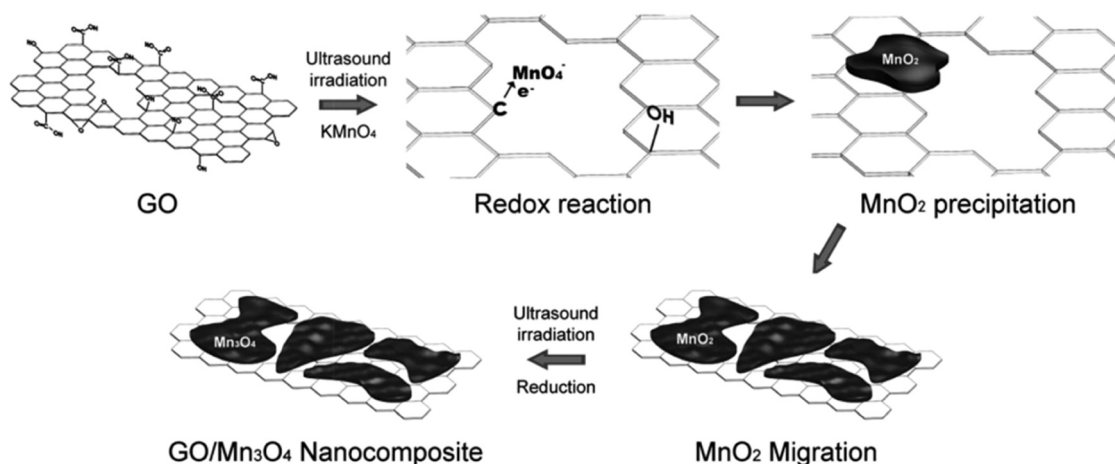


Fig. 10. Sonochemical synthesis of $\text{GO-Mn}_3\text{O}_4$ composite. Reproduced from ref. [136] Copyright (2012), with permission from Royal Society of Chemistry.

metal NPs (Pd, Pt, Ag, and RuO₂)-decorated h-BN catalysts for the glycolysis of PET [139]. This nano-hybrid material was prepared via the fluid-dynamic-induced synthesis process. Among the tested catalysts, Pd-deposited h-BN nano-catalysts gave good to excellent PET conversion and 92.1 % BHET formation when the reaction was conducted at 100 °C for 30 min (see Table 3, entry 17). Although the same Pd/h-BN catalyst was used, the depolymerization reaction conducted on a Taylor-Couette (T-C) flow reactor offers better yield and significantly reduces reaction duration and temperature when compared with the conventional hydrothermal process that occurs above 200 °C for 120 min. Additionally, similar to precious metals, transition metal-doped NPs have also been examined for the conversion of PET to BHET. For instance, Fe₃O₄ nanoparticles were deposited in the hexagonal boron nitride nanosheets (h-BNNS) using a simple hydrothermal process to obtain a Fe₃O₄ NPs@h-BNNS catalyst [140]. In this article, excellent 100 % BHET yield could be obtained when the recycling process was conducted at 200 °C for 5 h (see Table 3, entry 18).

Solid phase ionic liquids (SILs) have received recent scientific attention for the chemical recycling of PET [149,150]. SILs address key challenges associated with conventional ionic liquids (ILs), such as separations, high cost, and recyclability. Immobilizing ILs on magnetic supports provides ease of SILs separation from the reaction mixture using external magnets. In one interesting work, Cano research groups described this approach by modifying the surface of nano-magnetite materials with paramagnetic ILs [141]. This alteration introduced a diverse array of functionalities in the shell, which led to improved thermal inertness and catalytic activity. The authors have deposited the magnetite NPs on silica and incorporated with ionic liquids (ILs) to achieve a paramagnetic ILs coated Fe₃O₄@SiO₂@[mim][FeCl₄] heterogeneous catalyst (where mim = methylimidazolium). The catalysts offered 100 % BHET yield over twelve subsequent runs at a reaction temperature of 180 °C for 24 h (see Table 3, entry 19). The complete recovery of the catalyst can be obtained with an external magnet within 3 min.

In another article, cobalt-based ILs deposited on a graphene support (rGO\TESPMI)₂CoCl₄) were prepared as a transesterification catalyst by Barikani research groups for the glycolysis of PET, (where rGO = reduced graphene oxide and TESPMI = 1-(triethoxysilyl) propyl-3-methylimidazolium) [142]. The reported synergistic effect of graphene and ILs enhanced its catalytic performance, obtaining a 95.2 % BHET yield with full PET conversion (see Table 3, entry 20). ElMetwally and coworkers highlighted the application of bentonite clay as a solid support for immobilizing [Bmim-Fe]([OAc]₃) ionic liquid (where Bmim = 1-butyl-3-methylimidazol-3-ium) [151]. The bentonite clay prevented the interaction between PET and ILs, making a solid-solid phase, which creates EG as the ideal reaction medium for reacting PET with the catalysts. The activation energy required for the depolymerization of PET was found to be 51.6 KJ/mol.

Recently, Swapna and co-workers highlighted the preparation of a

shape-engineered manganese oxide (MnO_x) nanocatalyst having well-defined rod-morphology using a simple hydrothermal synthesis method for efficient PET glycolysis [143]. The nanostructured MnO_x catalyst, particularly when calcined at 500 °C (MnO_x-500), exhibits remarkable catalytic activity, converting used PET bottles into the valuable monomer bis(2-hydroxyethyl) terephthalate monomer under mild conditions. Complete PET conversion was achieved with an 86 % isolated BHET yield (see Table 3, entry 21), surpassing the performance of other metal oxide catalysts like CeO₂, TiO₂, and Nb₂O₅. NH₃-TPD analysis (Fig. 12 C) showed that MnO_x-500 possessed significantly more acid sites (0.44 mmol g⁻¹) compared to MnO_x-300 (0.13 mmol g⁻¹) and MnO_x-400 (0.17 mmol g⁻¹), correlating with its superior catalytic performance. The SEM and TEM image of the MnO_x-500 primarily consists of rod-shaped particles with a length of 200 nm and a width of 80–300 nm (see Fig. 11).

2.1.6. Organocatalysts

It's worth highlighting the recent significant progress made in using metal-free organocatalysts for the chemical recycling of PET. Organocatalysis has proven to be an effective approach for the depolymerization of waste PET. The pioneering work of Hedrick research groups has established the utilization of organocatalyst guanidine-based 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD) for the glycolysis of PET [152]. The TBD functions as a bifunctional catalyst, activating both the oxygen in the carbonyl group of the ester and the alcohol hydrogen through H-bonding interactions (Fig. 12b). This can also proceed through a nucleophilic 'acyl transfer' pathway (Fig. 12a), where the TBD catalyst temporarily forms a covalent bond with the ester. The optimized condition showed that the desired BHET product could be obtained in 78 % yield when the reaction was conducted using 0.7 wt% TBD for 3.5 h at 190 °C. The reusability of the organocatalyst was tested over 10 consecutive runs, consistently forming a BHET yield of over 65 % for each batch. However, the reaction time progressively lengthens with each cycle, likely due to a decrease in catalyst concentration upon multiple recycling stages.

The same research groups explored various nitrogen-based organic catalysts, such as N-methylimidazole (NMI), 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU), 1,5-diazabicyclo[4.3.0]non-5-ene (DBN), 1,4-diazabicyclo[2.2.2]octane (DABCO), 4-(N,N-dimethyl amino)pyridine (DMAP), and N,N-dimethylaniline (DMA), as transesterification catalysts [153]. Among the tested organocatalysts, strong bases such as TBD, DBU, and DBN showed promising results, enabling faster PET degradation with minimal oligomer formations, except for TBD (5.1 wt%). Further investigation revealed that DBU exhibited higher catalytic activity at a lower temperature than TBD. For instance, at a temperature of 190 °C and a catalyst loading of 0.5 %, DBU degrades PET in 220 min, while TBD requires 325 min. The authors describe the difference in catalyst reactivity using diols of varying chain length, ranging from ethylene glycols to 1-octanol. DBU demonstrated exceptional

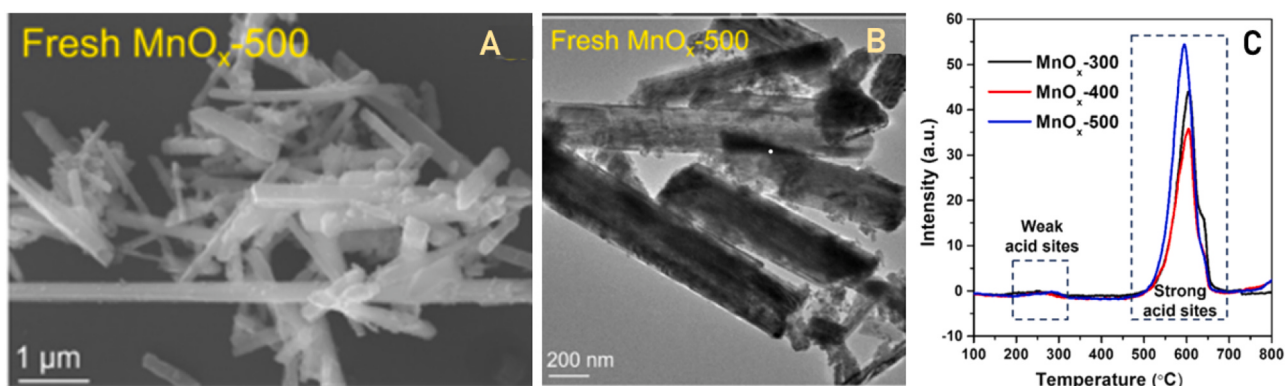


Fig. 11. SEM (A) and TEM (B) images of fresh MnO_x-500 nanocatalyst, and (C) NH₃-TPD of MnO_x-300, MnO_x-400, and MnO_x-500 nanocatalyst [143].

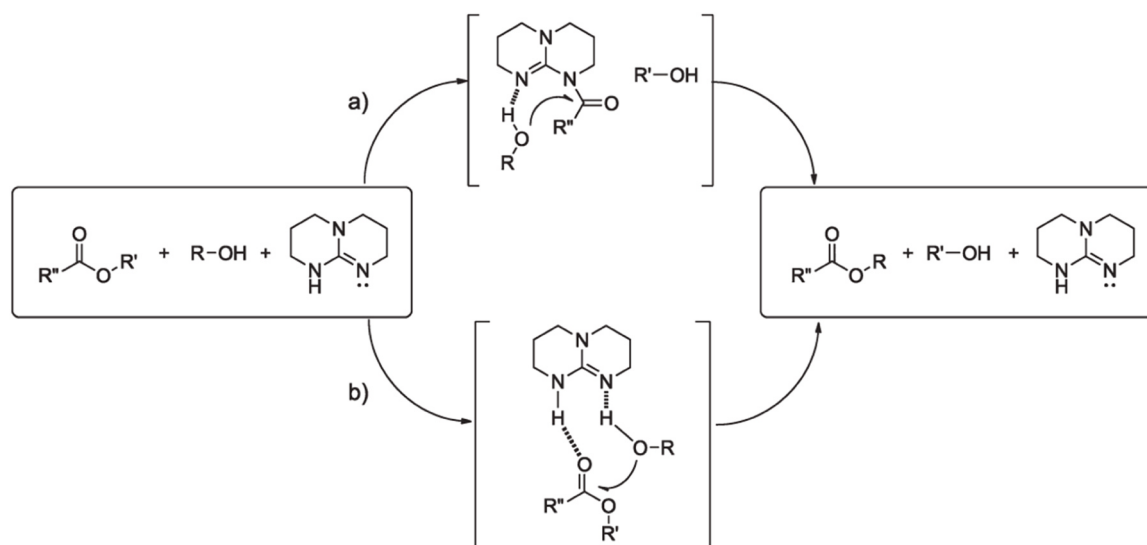


Fig. 12. Two plausible pathways of TBD-catalyzed transesterification: (a) acyl transfer pathway and (b) hydrogen bonding pathway.

performance with short-chain alkanediols under favorable conditions. However, when it comes to PET glycolysis using mono-alcohols and long-chain diols, TBD proved to be the best catalyst.

In another report, the utilization of 1,3-dimethylimidazolium organocatalysts for the chemical recycling of waste PET was described by Holbrey research groups [154]. The synthesized catalyst demonstrates excellent thermostability, exhibiting a remarkable ability to promote PET depolymerization at elevated temperatures. The effect of various reaction parameters has been examined with the goal of enhancing BHET yield. Under the optimized reaction condition, an impressive 60 % BHET monomer was produced in less than 1 h at 180 °C.

Salt-based organocatalysts, which have excellent air and moisture stability, have received considerable recent attention. The application of an equimolar mixture of thermally stable acid:base salt, TBD:MSA (MSA = methane sulfonic acid) catalyst, for the glycolysis of post-consumed PET was discussed [155]. This catalyst exhibits remarkable thermal stability, remaining stable up to 400 °C, a characteristic not commonly observed among organic acids. At a reaction temperature of 180 °C, TBD:MSA effectively degrades waste PET into BHET, obtaining 90 % yield in under 2 h. In addition, the organocatalysts can repolymerized BHET back into PET, with the resulting material showing comparable properties to virgin PET. Notably, the catalyst can be reused for six consecutive runs without any noticeable loss in activity.

The development of highly efficient catalysts that can operate at lower temperatures is imperative. While elevated temperatures may enhance BHET yield, they are associated with increased energy consumption and oligomer formations. Su et al., recently employed readily available cyanamide to depolymerize PET into BHET (100 % yield) at a relatively low temperature (150 °C) [156]. The practical applicability of this glycolysis procedure was showcased via its exceptional performance in the glycolysis of transparent and opaque PET samples as well as polyester foam. The consistent production of high-quality BHET further underscores its potential for large-scale PET recycling applications. A DFT calculation further provided that cyanamide has higher catalytic activity compared to its trimer, melamine. This can be attributed to the formation of a strong H-bond between cyanamide and either PET or EG.

2.1.7. Ionic liquids

The discovery of ionic liquids (ILs), organic salts that are liquid at room temperature, spans a considerable period. In a pioneering study, Walden synthesized the first ionic liquid, ethylammonium nitrate IL, having a melting point of 12 °C in 1914 [157]. These liquids displayed a remarkable set of properties: they are non-volatile, inflammable,

thermally and chemically inert, and exhibit low viscosity, high conductivity, and a wide working temperature range [158–160]. Prized for their unique properties, ILs have found diverse applications across various fields. These include, but are not limited to: electrochemistry, additives, chemical industry, analytical chemistry, advanced materials, and polymer recycling [161].

In the year 2009, Li and Zhang research groups broke new ground by demonstrating the utilization of various ILs for PET waste glycolysis [162]. Their investigation includes ILs such as [bmim]Cl, [bmim]Br, [bmim]H₂PO₄, [bmim]HSO₄, (3-amino-propyl)-tributyl-phosphonium glycine ([3a-C3P(C4)3][Gly]), and (3-amino-propyl)-tributyl-phosphonium alanine ([3a-C3P(C4)3][Ala]) (where bmim = 1-butyl-3-methylimidazolium chloride). Their findings identified [bmim]Cl as the promising candidate for additional examination. Further experiment focused on optimizing effects of different reaction parameters, such as catalyst amount, reaction period, reaction temperature, and water content, for their impact on PET conversion, BHET selectivity, and distribution of the products. The catalysts offered 100 % BHET conversion at a reaction condition of 4.0 g of ILs at 1 atm and 190 °C over an 8-hour reaction period.

Room temperature ILs, recognized for their tunable physicochemical characteristics, have emerged as environmentally friendly solvents and catalysts [163]. In another intriguing study, Jin and co-workers examined highly efficient basic ionic liquids for the degradation of PET waste [164]. Among the tested ILs, [Bmim]OH exhibited superior catalytic activity compared to [Bmim]HCO₃, [Bmim]Cl, and [Bmim]Br. Under the optimized reaction conditions (PET:EG mass ratio of 1:10, 0.1 g (5 wt%) of [Bmim]OH, 190 °C, 2 h), they achieved 100 % PET conversion and a 71.2 % BHET yield, respectively.

ILs have demonstrated the capability to enhance both the solubility of plastic waste and the catalytic activity of depolymerization reactions. Extensive research in the area has explored the application of number of a ionic liquids for degrading plastic waste, particularly for PET and PC materials [165,166]. Among the various ILs studied, those with imidazolium cations have shown particular promise. Table 4 further presents data from other ILs that promoted PET glycolysis reactions:

(where [bmim] = 1-butyl-3-methylimidazolium chloride, [Hmim] = 1-hexyl-3-methylimidazolium, PIL = poly [Bvim]NTf₂, [Ch] = cholinium, [HDBU]m = 1,8-diazabicyclo [5,4,0] undec-7-ene imidazole, OAc = acetate, [Deim] = 1,3 Diethyl Imidazolium)

Table 4
Glycolysis of waste PET using various ionic liquids (ILs).

Sl. No.	Catalysts	Temperature (°C)	Time (h)	BHET yield (%)	Ref.
1.	[bmim]FeCl ₄	150	4	76.4	[167]
2.	[deim][Zn(OAc) ₃]	180	2.5	70.94	[168]
3.	[bmim]ZnCl ₃	190	2	84.9	[169]
4.	[Deim][Zn(OAc) ₃]	175	1.25	80.1	[170]
5.	[bmim]ZnCl ₃	180	5	83.3	[171]
6.	[bmim]OAc	190	3	58.2	[172]
7.	Cu(OAc) ₂ ·[Bmim][OAc] and Zn(OAc) ₂ ·[Bmim][OAc]	190	3	53.95, 45.6	[173]
8.	[bmim] ₂ [CoCl ₄]	175	1.5	81.1	[174]
9.	[Ch][OAc]	180	4	85.2	[175]
10.	Equimolar mixture of [Hmim]ZnCl ₃ and [Hmim]CoCl ₃	190	2	87.1	[176]
11.	PIL-Zn ²⁺	175	2	77.8	[177]
12.	[Ch][Gly]	150	6	51	[178]
13.	[C ₆ TMG]Cl/2ZnCl ₂	195	1.2	92.7	[179]
14.	[EMIm] ₂ TPA	197	1.7	83.6	[180]
15.	[Cho][OAc]	180	-	66.7	[181]
16.	[HDBU]Im	185	1.5	88.9	[182]

2.2. Glycolysis of polyester waste

Among various textile fibers, PET fibers are the most widely used in the industry [183]. These synthetic fibers exhibit extremely low biodegradability, contributing significantly to environmental concerns [184]. Recycling of polyester fibers is more challenging than recycling PET bottles due to the presence of finishing agents, such as antistatic, UV-resistant, and water-repellent compounds, which can trigger side reactions or hinder recycling efficiency. However, glycolysis can still be effectively applied to PET-containing textiles, allowing the depolymerization of polyester into reusable, virgin-grade monomers. For instance, Guo research group developed nanosized Mg–Al double oxides sintered onto magnetic Fe₃O₄ microparticles, forming a hierarchical structure with a high active surface area [185]. This structural feature imparts excellent catalytic performance in the glycolysis of PET, achieving a BHET yield of over 80 mol% using just 0.5 wt% of the Mg–Al–O@Fe₃O₄ catalyst at 240 °C for 90 min. The BHET monomers were repolymerized and spun under the same conditions as virgin PET, producing fibers with mechanical properties, such as tenacity and elongation, comparable to those of virgin PET fibers. Additionally, the catalyst exhibits strong magnetic properties, enabling easy recovery and reuse.

Since a significant portion of textile waste consists of colored polyester fibers dyed with disperse dyes, further complicating recycling efforts. The removal of these dye molecules, either before or after the glycolysis of waste polyester textiles, is a crucial step, as residual dyes can compromise the quality of the final product and may also trigger side reactions or inhibit the depolymerization of PET. The research team led by Ierapetritou, Lobo, and Vlachos developed zinc-based heterogeneous catalysts for the glycolysis of PET waste and successfully applied them to five different colored PET waste samples [186]. Using microwave-assisted heating, PET was depolymerized with a yield exceeding 93 % within just 10 min. The presence of dyes in the colored polyester textiles had no adverse effect on the catalytic reaction. Following the reaction, the dye additives were effectively separated from the BHET monomers by filtration, resulting in high-purity products. In another study, Yu synthesized a novel heterogeneous catalyst, Zn-MCM-41-25, incorporating a Zn-to-Si/Al molar ratio of 25 [187]. This catalyst exhibited excellent performance in the depolymerization of waste PET textiles. Under the optimized reaction conditions, 8 % Zn loading, 5 % catalyst dosage, an EG to PET mass ratio of 6, and a reaction temperature of 200 °C for 1 h and 45 min, complete PET conversion was achieved, along with a BHET yield of 81.4 %. The resulting product was

analyzed using high-performance liquid chromatography, which detected only BHET monomers, dimers, and trimers, with no other by-products observed, confirming that no side reactions occurred between PET and the catalyst, even in the presence of dye molecules.

2.3. Methanolysis of PET

PET alcoholysis is a transesterification reaction that entails the cleavage of the backbone ester linkage of PET to obtain an alkyl terephthalate and EG [188]. This approach is highly favored for the preparation of myriad value-added products with different molecular weights and physicochemical properties. It can be achieved by altering the reaction parameters, such as reaction temperature, pressure, loading of the catalyst, quantity of alcohol, and polymer content [189]. Various alcohols, including butanol, ethanol, benzyl alcohol, 2-ethyl-1-hexanol, trimethylolpropane, etc., have been actively investigated for the process [190]. Recently, there has been a lot of focus on the catalytic methanolysis of PET.

Methanolysis of PET involves the depolymerization of PET in the presence of an excess amount of methanol at relatively high temperatures (180–280 °C) and pressures (20–40 atm), to obtain dimethyl terephthalate (DMT) and ethylene glycol [191]. The incorporation of co-solvents such as p-xylene, dichloromethane (DCM), and dimethyl sulfoxide (DMSO) is common strategy to facilitate a rapid degradation rate even at low temperature by promoting the swelling of PET chips or flakes, and improving polymer accessibility. DMT is an alternative raw material for the production of PET. Its hydrogenated form, 1,4-cyclohexanedimethanol, can be utilized for the production of polyester fiber materials. These fibers are non-toxic and have wide-ranging applications in photography and as anti-fogging agents [192]. In addition, methanolysis exhibits a higher level of tolerance to contaminants. This enables methanolysis to depolymerize low-grade feedstocks, leading to a significant reduction in operational expenses [193]. Similar to the PET glycolysis process, the non-catalytic methanolysis of PET also necessitates a harsh reaction environment and a substantial quantity of reaction solvent. For example: the complete degradation of PET with a 97 % DMT yield could be obtained by subjecting it to supercritical methanol at a high temperature (310 °C) and pressure (51 MPa) [194].

In 1997, Sako and coworkers reported that PET could be fully depolymerized into dimethyl terephthalate, ethylene glycol, and some oligomers within just 30 min in supercritical methanol, in the absence of a catalyst. This depolymerization was achieved at temperatures above 300 °C and pressures around 110 bar. While increasing the pressure beyond the supercritical point did not significantly affect DMT yield, the severity of these conditions renders the process economically unfeasible for large-scale applications and challenging to adapt to continuous flow systems [195]. Microwave-assisted methanolysis is a highly efficient method for achieving good to excellent DMT yields under moderate reaction condition [191]. Studies have shown that the utilization of zinc acetate as a transesterification catalyst produced more than 80 % of the corresponding DMT in just 30 min of microwave irradiation at 160 °C. Numerous metal salts/acetates, DESs, and ILs are extensively examined for this approach [189,196,197].

Pham and Cho research group recently obtained a high DMT yield (93.1 %) in the presence of potassium carbonate (K₂CO₃) and DCM as cosolvent at room temperature [198]. While these molecular catalysts exhibit superior performance, longer reaction times as well as the separation and purification of DMT pose significant challenges that can be addressed by the introduction of atom-efficient heterogeneous transesterification catalysts. However, there are far fewer catalytic systems designed specifically for the methanolysis of PET as compared to the glycolysis process.

Sharma and coworkers demonstrated the first example of PET methanolysis using a heterogeneous catalyst in 2013 [109]. They have employed a two-step method in which PET is initially converted to oligomers by subjecting them to boiling DMSO solvent in the presence of

Mg-Al hydrotalcite catalysts. A transesterification process then follows, where the oligomers are reacted with NaOH in methanol at room temperature to provide DMT and EG. Hydrotalcite, DMSO, and NaOH can be efficiently distilled and recycled in this procedure.

Wang and coworkers have developed a single-step highly efficient method for PET to DMT conversion using ultrasmall ZnO nanoparticles (NPs) as catalyst, eliminating the need for a corrosive alkaline base as a cocatalyst [199]. The catalysts, with a size of 4 nm, provide enhanced reactant-catalyst interaction due to their ultrasmall dimension, resulting in reduced diffusion resistance. Although there was improved catalytic activity, the extraction of ZnO NPs from the reaction mixture proved challenging, leading to a considerable reduction in the degradation of PET after the fifth cycle.

Heterogeneous catalysts obtained from biomass waste have gained significant interest due to their excellent reusability, thermostability, and non-toxic nature. Vanlaldinpuia research groups demonstrated the utilization of bio-waste origin bamboo leaf ash (BLA) as a reusable heterogeneous catalyst for methanolysis of PET. The occurrence of calcium (Ca) and potassium (K) on the catalyst surface has significantly increased the rate of the PET depolymerization reaction, obtaining a 78 % DMT yield at 200 °C within 2 h (Fig. 13). Nevertheless, the performance of the BLA catalyst decreased dramatically throughout the recycling studies due to metal leaching [200]. A year later, a similar research group reported the using Fe₃O₄ deposited on orange peel ash (Fe₃O₄@OPA) as a green, magnetically retrievable transesterification catalyst for PET methanolysis [201]. The catalyst exhibited a significant surface area, a highly mesoporous structure, and exceptional basic strength, resulting in outstanding catalytic performance and achieving a high DMT yield (83 % yield). The authors proposed that the presence of oxides and carbonates of metals serve as Lewis acids, interacting with the carbonyl oxygen in ester groups to increase the polarity and electrophilicity of the carbonyl carbon. This activation facilitates the nucleophilic attack by methanol, leading to the cleavage of ester bonds and yielding the desired product. The catalyst could be reused for 10 consecutive runs, with only a slight decrease in its catalytic activity. The excessive use of methanol to enhance DMT output and minimize by-product formation hinders the scalability of the methanolytic process [202].

Hou research group also reported an efficient catalytic strategy for upcycling waste PET into dimethyl terephthalate (DMT) using environmentally benign Ti_xSi_{1-x}O₂ solid acid catalysts [203]. Characterization revealed that the Lewis acid sites in Ti_xSi_{1-x}O₂ were highly active for the alcoholysis of PET with methanol under mild conditions. Notably, Ti_{0.5}Si_{0.5}O₂ achieved a DMT yield of 98.2 % with complete PET conversion (2.5 wt% loading) within 2 h at 160 °C. Moreover, the catalyst retained its performance over at least five consecutive recycling cycles without significant loss of activity.

To address the challenge of using a large quantity of methanol, Ma research groups developed an innovative single-step procedure involving an in-situ generation of methanol via CO₂ hydrogenation for PET methanolysis (Fig. 14). In the presence of LDHs precursor, the authors have introduced a range of CuFeCr catalysts. Cu₄Fe₁Cr₁ catalyst demonstrated a dual-promotional effect among the examined catalysts

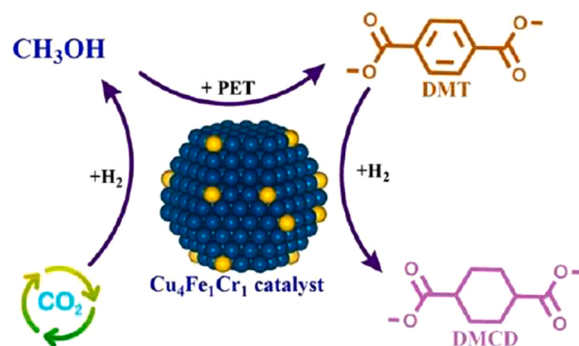


Fig. 14. Synthesis of cyclohexanedicarboxylate (DMCD) from CO₂ and PET using Cu₄Fe₁Cr₁ catalyst. Reproduced from ref.[204] Copyright (2022), with permission from Wiley-VCH.

[204]. It had twice the methanol productivity (11.9 mmol_{g_{cat}}⁻¹) than when PET was not added and facilitated methanolysis of PET. The obtained DMT was subjected to hydrogenation to produce dimethyl cyclohexanedicarboxylate (DMCD) and p-xylene. In addition, the versatility of this method was expanded to include the breakdown of other polyester plastics such as polybutylene terephthalate (PBT) and polyhexamethylene adipate (PHA). The documented synergistic methanolytic approach yields a product that is 2- or 7-times greater than the uncoupled reaction

Furthermore, 1,4-cyclohexanedimethanol (CHDM) is a particularly important monomer used in the production of polyester fibers, unsaturated polymer resins, polyurethane foams, polyester glazing, and hydraulic fluids. CHDM is typically synthesized via the hydrogenation of dimethyl 1,4-cyclohexanedicarboxylate (DMCD), which is derived from PET through methanolysis followed by catalytic hydrogenation. Chang et. al., investigated the hydrogenation of DMCD to CHDM using novel Cu₁/Mg₃Sc₂O₆, Cu₁/Mg₃Al₂O₆, and Cu₁/Mg₃Sc₂O₆-IM catalysts under mild reaction conditions [205]. Among these, Cu₁/Mg₃Sc₂O₆ exhibited superior catalytic performance, achieving 99.3 % DMCD conversion and 97 % selectivity toward CHDM at 250 °C, (weight hourly space velocity) WHSV_{DMCD} = 0.49 h⁻¹, and H₂/DMCD molar ratio of 120 under 2.0 MPa. In comparison, Cu₁/Mg₃Al₂O₆ and Cu₁/Mg₃Sc₂O₆-IM resulted in significantly lower conversions (44.5 % and 17.3 %) and selectivities (36 % and 33 %), respectively. The enhanced CHDM selectivity is attributed to the higher basicity of Cu₁/Mg₃Sc₂O₆, which effectively suppresses the formation of byproducts typically promoted by acidic sites. In another study, Jiang et al. reported the development of a stable Cu-based catalyst (Cu/MgAl₂O₄) with strong metal-support interaction, synthesized via calcination and reduction of a CuMgAl-LDH precursor [206]. The resulting Cu/MgAl₂O₄ catalyst exhibited a high surface area, strong metal-support interaction, and abundant basic sites. The prepared catalyst retained a high surface area of 111.8 m²/g even after calcination at 800 °C, and the dispersion of Cu in the CuMgAl mixed metal oxide reached 42.4 %. The catalyst demonstrated excellent performance in the hydrogenation of PET-derived BHCD to CHDM, achieving a 98 % yield with complete BHCD conversion at 240 °C,

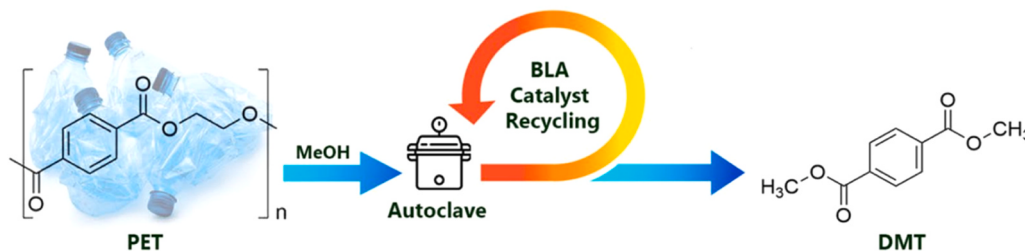


Fig. 13. Methanolysis of PET waste using bamboo leaf ash (BLA) heterogeneous catalyst. (a) Reproduced from ref.[200] Copyright (2022), with permission from Springer.

4 MPa, and a space velocity of $0.525 \text{ g} \cdot \text{g}_{\text{cat}}^{-1} \cdot \text{h}^{-1}$. Recently, Zhao and Hou research group also examined the utilization of a defective sulfonated bifunctional Ru/UiO-66_{def}-SO₃H [207]. The catalyst exhibited excellent catalytic performance not only in the separate methanolysis of PET to DMT and hydrogenation of DMT to DMCD, but also in the one-pot direct conversion of PET to DMCD under mild conditions. Complete PET conversion was achieved with a 97.7 % yield of DMCD at 170 °C and 3 MPa H₂ within 6 h.

The utilization of precious metal catalysts such as Pd or Ru for the chemical degradation of PET significantly limits their industrial applicability due to high cost and limited availability. Recently, Li research groups reported a precious-metal-free tandem catalytic strategy for the direct conversion of PET to 1,4-cyclohexanedimethanol (CHDM) in a CH₃OH/1,4-dioxane mixed solvent system [208] (see Fig. 15). The process proceeds through three consecutive steps without the need for intermediate separation or purification. Initially, PET undergoes methanolysis to form dimethyl terephthalate (DMT), which is subsequently hydrogenated over a base metal NiLa (40 wt%) catalyst to afford dimethyl 1,4-cyclohexanedicarboxylate (DMCD). The final hydrogenolysis of DMCD to CHDM is carried out using a Cu₁Fe₁Al_{0.5} catalyst. This integrated approach resulted in an overall CHDM yield of 90.2 %, demonstrating high efficiency and industrial potential while eliminating the reliance on noble metal catalysts

3. Conversion of PET waste into value added fuels and fine chemicals

Catalytic reductive depolymerization has emerged as a promising platform for the transformation of unwanted waste plastics into value-added products, making it a significant area of recent research interest [209]. Researchers have so far investigated two distinct approaches for this method. In the first method, catalysts are employed to break the carbon-carbon or carbon-hetero bonds of PET, leading to the formation of monocyclic arenes and terephthalic acid (TA). The second method entails the methanolysis of PET into DMT, which is subsequently followed by noble metal-promoted hydrodeoxygenation to obtain valuable compounds and cycloalkanes [210].

The catalytic hydrogenation of PET yielded 1,4-cyclohexanedicarboxylate (PECHD). The presence of an aliphatic ring in PECHD offers improved biodegradability and thermal stability when compared to BHET [211]. The traditional method for the preparation of PECHD, which encompasses hydrogenating dimethyl terephthalate followed by polymerization, has several drawbacks. These limitations involve a harsh reaction environment, a multiple-step synthetic procedure, and the reliance on costly noble metals. In addition, separating the dimethyl 1,4-cyclohexanedicarboxylate intermediate from the crude product is expensive and time consuming. Therefore, it is imperative to develop an efficient, single-step catalytic method for producing PECHD polyester [212].

With this in mind, Tan research groups developed a single-step

selective hydrogenation of PET polyester to PECHD using supported bimetallic Rh_{2.5}Pt_{2.5}/Vulcan XC-72-polyol catalysts [213]. This report explores the use of 1,1,1,3,3,3-hexafluoro-2-propanol as a green, eco-friendly, and economically viable solvent for dissolving PET. At 58 °C and 6.89 MPa of H₂, a full PET conversion with a 98 % PECHD yield could be obtained. The bimetallic Rh-Pt exhibited higher catalytic activity for PET hydrogenation when compared with monometallic Rh catalysts. The remarkable performance can be attributed to the strong aromatic ring adsorption capacity of Rh as well as the increased H₂ spillover facilitated by Pt metal. Furthermore, the authors acknowledge that optimizing the particle size, distribution, and concentration of the active metals are crucial factors for obtaining a high product yield.

In a subsequent study, the catalytic activity of SBA-15 supported Rh-Pt bimetallic (Rh_{2.5}Pt_{2.5}/SBA-15) catalyst was investigated for hydrogenating PET (see Fig. 16) [214]. The study emphasized the synergistic effect of Rh-Pt bimetals in enhancing the efficiency of PET hydrogenation. Notably, under mild reaction conditions, complete hydrogenation of PET was achieved in an aqueous media.

More recently, Cai research groups reported the synthesis of a novel N-doped carbon-supported bimetallic CoMo@NC catalyst via pyrolysis of Mo@ZIF-COZn at 900 °C (see Fig. 17) [215]. During this process, the high temperature pyrolysis of the Mo@ZIF-CoZn precursor converted the ZIF framework into a nitrogen-doped graphitic matrix. Then, the bimetallic components were anchored to the matrix, obtaining Co NPs and Mo nanoclusters. At 260 °C and atmospheric H₂ pressure, the synthesized catalysts fully degrade PET with a 91 % TA yield. The synergistic interaction between the Mo- and Co-sites inside the bimetallic catalyst facilitates efficient hydrogenolysis under mild reaction conditions, achieving a high TA yield. Furthermore, the catalyst exhibits excellent reusability, maintaining stable activity for up to six cycles. In another study, Marks research groups documented the utilization of a carbon-supported single-site C/MoO₂ catalyst for the solvent free hydrogenolysis of PET to terephthalic acid (TA) [216]. This catalyst exhibited high depolymerization efficacy for both commercial and discarded PET. The availability of active molybdenum sites, capable of selectively activating and cleaving the ester groups of PET, accounts for its effectiveness. At a reaction temperature of 260 °C and a hydrogen pressure of 0.1 MPa, the C/MoO₂ catalyst obtained 100 % PET conversion with high a TA yield.

The extension of this approach to the upgradation of lignocellulosic biomass into fine chemicals and high-value fuels offers a compelling opportunity and represents an interesting field of research [217]. This method has recently been utilized for the hydrogenation of PET polyester into high-density aviation fuels and aromatic hydrocarbons in the presence of metal-supported heterogeneous catalysts [218]. Yan research groups exploited a Co/TiO₂ catalyst for the synthesis of arenes from PET through combined depolymerization and catalytic hydrogenation reactions [219]. The catalyst was able to obtain 78.9 % of arenes in 4 h at a reaction temperature of 340 °C. The main drawback of this catalyst was its suboptimal stability, which resulted from the phase shift

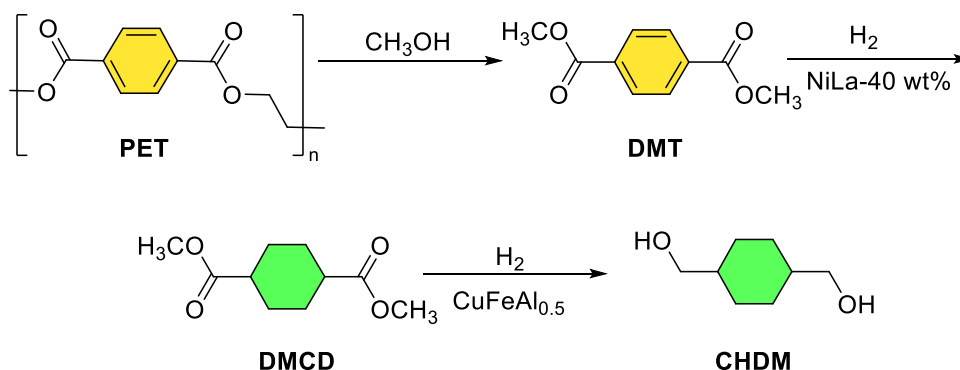


Fig. 15. Synthesis of CHDM from PET [208].

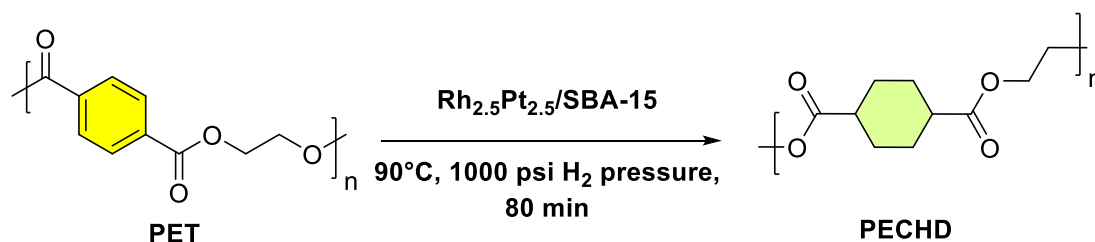


Fig. 16. Catalytic hydrogenation of PET to PECHD using $\text{Rh}_{2.5}\text{Pt}_{2.5}/\text{SBA-15}$ catalyst [214].

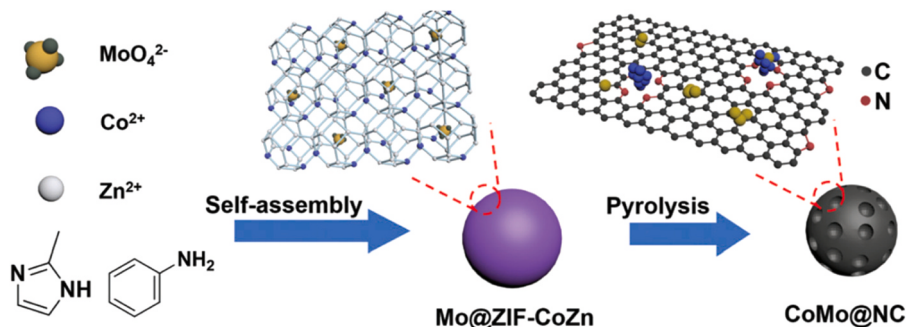


Fig. 17. Schematic illustration of the synthesis of CoMo@NC catalyst.

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of both the support material and the active metal component.

Later, the same research team successfully developed a versatile $\text{Ru}/\text{Nb}_2\text{O}_5$ catalyst that can successfully be employed for the upcycling of waste plastic into monocyclic arenes. This catalyst demonstrates superior catalytic activity in depolymerizing waste PET as well as mixed organic plastic waste into valuable arenes [220]. The reaction mechanism for cleavage of C-C and C-O bonds over $\text{Ru}/\text{Nb}_2\text{O}_5$ catalyst was delineated (see Fig 18). The high oxygen affinity of NbO_x species promoted hydrogenolysis and limited decarboxylation, as a result of the presence of zero-valent Ru species on the $\text{Ru}/\text{Nb}_2\text{O}_5$ catalyst. The Lewis acidic sites, specifically NbO_x species, exhibit a strong affinity for oxygen, enabling selective adsorption and activation of the C-O bond. At the same time, the benzene ring adsorbs onto the NbO_x species and undergoes protonation by the Bronsted acid sites on Nb_2O_5 , activating the $\text{C}_{\text{sp}2}\text{-C}_{\text{sp}3}$ bond. Finally, hydrogen species, dissociated from the Ru nanoparticles, contribute to the precise cleavage of both C-O and C-C bonds within the aromatic plastic structure. The catalyst stability test investigated with mixed plastic waste demonstrated stable yield of arenes even after three runs.

PET's chemical structure is composed of repeating EG units that can serve as a H_2 source during the production of arenes [222]. This

innovative approach was utilized by Wang research groups to develop a H_2 free method for the reductive depolymerization of PET plastic over a $\text{Ru}/\text{Nb}_2\text{O}_5$ catalyst [221]. The method included four main steps: hydrolysis, EG reforming, parallel hydrogenolysis, and decarboxylation reactions. The applicability of the $\text{Ru}/\text{Nb}_2\text{O}_5$ catalyst in depolymerizing different PET materials (including Coca-Cola and polyester film) was investigated. The catalyst obtained BTX (benzene, toluene, and p-xylene) streams with a total 88.7 % and 93.3 % yield, and alkyl aromatic selectivities of 74.5 % and 72.7 %, respectively (Fig. 19). These results established the catalyst's potential for handling common plastic waste streams. In contrast to the earlier study, the conversion of PET promoted by $\text{Ru}/\text{Nb}_2\text{O}_5$ results in the production of 92.4 % arenes under hydrogen-free conditions and at relatively moderate temperatures.

In 2019, Tang research groups introduced a multi-step procedure for synthesizing long hydrocarbon chains from waste PET (Fig. 20) [223]. In this report, the waste PET was initially transformed into DMT through a methanolysis reaction without the influence of any external catalyst. The process was promoted by the solvent (methanol), and resulted in a high yield of DMT (97.3 %) at a temperature of 200 °C. Subsequently, the DMT was isolated from methanol and subjected to hydrogenation to obtain dimethyl cyclohexane-1,4-dicarboxylate (DMCD) under a solvent-free method over Pt/C catalyst. The $\text{Ru-Cu}/\text{SiO}_2$ catalyst successfully converted the DMCD into $\text{C}_7\text{-C}_8$ cycloalkanes and aromatics, achieving an overall yield of 98.4 %.

4. Global strategies and economic insights of PET chemical recycling

Global initiatives, such as the European Union's Plastics Strategy and United Nations Environment Programme resolutions, are driving the adoption of chemical recycling for plastics as a policy to reduce plastic pollution and promote a sustainable economy. The European Union's Plastics Strategy seeks to revolutionize the design, production, use, and recycling of plastics, with ambitious targets of achieving 55 % recycling rate for plastic packaging by 2030. This strategy aimed at reducing marine litter, greenhouse gas emissions, and dependence on imported fossil fuels [224]. Additionally, the United Nations Environment Assembly in March 2022 adopted a landmark resolution to develop an

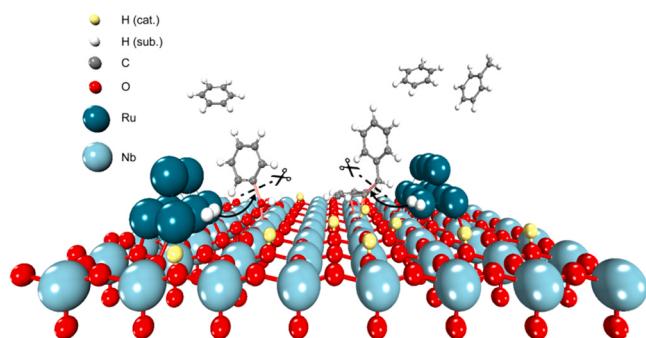


Fig. 18. Proposed reaction mechanism of C-O/C-C cleavage using $\text{Ru}/\text{Nb}_2\text{O}_5$. Reproduced from ref.[220] Copyright (2021), with permission from Wiley-VCH.

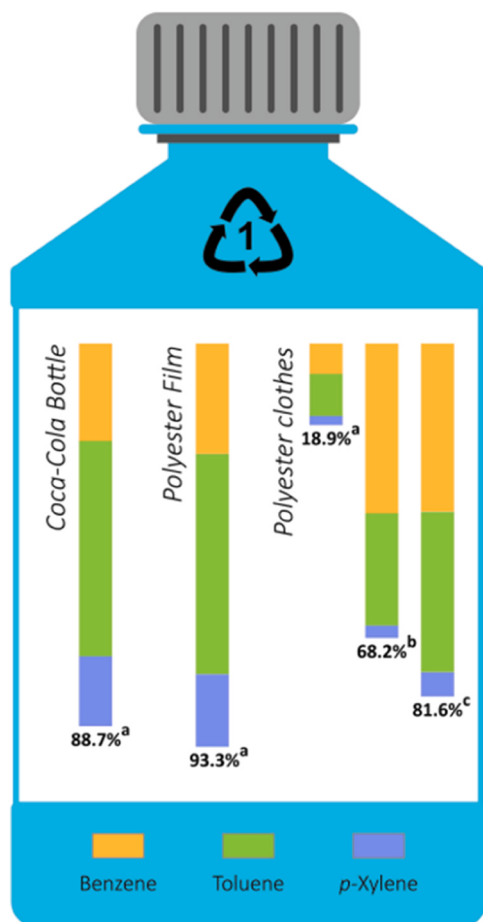


Fig. 19. Results of the conversion of various common PET using Ru/Nb₂O₅ catalyst.

(a) Reproduced from ref.[221] Copyright (2021), with permission from Wiley-VCH.

international, legally binding instrument aimed at ending plastic pollution by 2040. This agreement promotes a comprehensive strategy that addresses the complete lifecycle of plastics, encompassing production, design, and disposal, thereby fostering the adoption of advanced recycling methodologies such as chemical recycling [225]. These policies collectively aim to foster a supportive environment for recycling of plastics by establishing clear targets and frameworks, which in turn stimulates innovation and investment in recycling technologies, ultimately reducing plastic waste and promoting sustainability.

The economic feasibility of large-scale PET chemical recycling hinges on balancing raw material costs, energy consumption, catalyst efficiency, process scalability, and market demand for recycled monomers like BHET, DMT, TPA, and EG. Overcoming major challenges, such as high catalyst expenses, energy-intensive depolymerization, and purification costs, is crucial and can be addressed through biomass-derived or enzyme-based catalysts, process energy optimization, and improved waste sorting [226,227].

The global recycled PET (rPET) market is experiencing rapid growth, driven by increasing environmental concerns and the rising demand for sustainable packaging. It is projected to surpass USD 40 billion by 2033, with a compound annual growth rate (CAGR) of approximately 4 % from 2023 to 2032 [228]. In 2023, a significant portion of the chemical recycling of PET market was concentrated among a few key players, accounted for approximately 86 % of the market share [229]. These major players include SK Chemical, Far Eastern Group, Veolia, SABIC, Indorama Ventures, Mitsubishi Corporation, and JEPLAN, INC. In addition to these companies, the commercialization of PET chemical recycling technologies represents a rapidly evolving and dynamic area of research, with numerous companies and startups successfully showcasing their industrial applicability and economic viability. Ioniqa, Garbo, IBM, and Dupont-Teijin companies are actively pursuing full depolymerization of used PET to BHET via glycolysis process, to create recycle PET granulate [230]. Gr3n's Microwave Assisted Depolymerization (MAD) technology in Spain, a collaborative work between gr3n and Schneider Electric, processed over 40,000 tons of polyester waste annually [231]. Carbios (France) has pioneered enzyme-based PET depolymerization, achieving significant PET breakdown (90 % in 10 h) and partnering with major companies (L'Oréal, Nestlé, and PepsiCo) [230]. Loop Industries (Canada) utilizes a proprietary depolymerization process to produce virgin-quality PET from low-grade waste, collaborating with brands like Evian and Coca-Cola [230]. DePoly (Switzerland) also employs an innovative alkaline hydrolysis process for efficiently breaking down mixed and contaminated PET waste [232]. Furthermore, Revalyu (Germany) has also commercialized glycolysis-based PET depolymerization, converting PET into high-quality rPET for textile and packaging applications [233]. Recently, Eastman Chemical announced the construction of a 250 million USD PET methanolysis facility in Tennessee, with a planned capacity of 100 kilotons per year. The company has been actively involved in PET methanolysis research for several decades, holding patents in the field since the early 1990s [234]. The expansion of chemical recycling of PET is driven by increasing environmental concerns, catalytic efficiency advancements, low-energy depolymerization techniques, and financial incentives like carbon credits and subsidies.

However, when compared to its mechanical counterparts, few initiatives have reached industrial scale likely due to the lack of

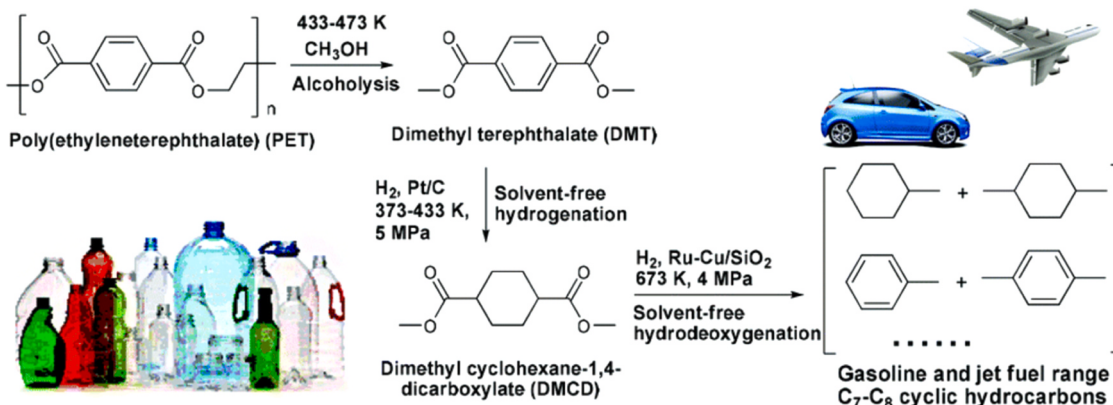


Fig. 20. Reaction pathway for the preparation of gasoline and jet fuel range C₇-C₈ cyclic hydrocarbons from PET waste. Reproduced from ref.[223] Copyright (2019), with permission from Royal Society of Chemistry.

infrastructure for collection, sorting, depolymerization, purification, and storage. As a result, implementation of government regulations supporting circular economy initiatives, alongside corporate sustainability commitments, can significantly enhance the economic feasibility of industrial-scale PET chemical recycling, establishing it as a profitable and sustainable alternative to landfill disposal and mechanical recycling limitations [235,236].

5. Conclusion and future outlook

PET materials offer undeniable benefits to modern society. Their proven versatility and large-scale manufacturing have revolutionized various sectors and greatly improved our quality of life. However, the fact remains that their production far outweighs our capacity to effectively manage them at the end of their life cycle. The exceptional durability and continuous accumulation of waste PET pose significant environmental challenges for mankind. Given our current reliance on plastic infrastructure, demands for a complete and immediate shift to a “plastic-free” society are unfeasible. Instead, the development of innovative chemical recycling and upcycling techniques, with chemists at the forefront, becomes the foreseeable future. The recycling of post-consumer PET material, from fiber for textiles to resin for beverage bottles, addresses the mounting issue of solid waste and offer valuable opportunities for developing virgin raw materials across various sectors, particularly the paint and coatings industry.

The chemical recycling of waste PET over heterogeneous catalysis has compelling advantages over homogeneous catalytic systems. These include ease of separation, lower toxicity, and enhanced stability. To optimize these catalytic transformations for higher reaction rates, a keen understanding of the active sites, their interaction with PET, and the structural properties of the catalysts is essential. In particular, utilization of efficient, cost-effective, and recyclable biomass waste-derived heterogeneous catalysts presents a fascinating alternative to expensive catalysts while also promoting a more sustainable and carbon-neutral future. In addition, while catalyst deactivation upon reuse remains a challenge; immobilization on solid supports like layered double hydroxides (LDHs) emerges as a viable and innovative solution.

Catalytic degradation of waste PET is a widely established process for achieving PET circular economy. This review summarized the various transesterification catalysts employed for the glycolysis and methanolysis of post-consumed PET to its constituent monomers. Glycolysis, as a chemical recycling process, has consistently demonstrated superior cost-effectiveness and economic feasibility. In contrast, methanolysis, a potential degradation method, requires high energy inputs due to the low reactivity of methanol with the PET esters bond. While catalysts with high surface area and balanced acidic/ basic sites demonstrated promising results, it's important to note that most studies were limited to clear, washed PET materials. Additional investigation is crucial to determine their efficacy in processing more complex waste streams, such as mixed plastics, colored, and contaminated PET.

Reductive depolymerization processes enables the production of a range of high-value chemicals. Notably, these chemicals are inaccessible through conventional depolymerization methods, highlighting the unique advantages of this approach. While this methodology holds significant promise, its application to PET waste depolymerization remains limited, with only a handful of examples documented in the literature. The physicochemical properties of the active metal sites, durability of the support matrix, and the cooperative interactions inherent to bimetallic configurations collectively influence the longevity and scalability of the catalytic process. However, challenges such as implementation of cheaper non-metal catalysts, reliance on harsh reaction conditions, limitation in product selectivity, and lengthy reaction times highlighting critical areas for future research and development.

Despite recent academic interest in the chemical recycling of PET waste, its practical implementation remains limited. This is primarily due to the diverse chemical structures of plastics, which define their

unique properties and functions but also affect their susceptibility to chemical depolymerization, thereby complicating the process. Furthermore, the need for appropriate green solvents, not only to effectively dissolve or swell polymers but also to enhance catalytic activity and selectivity, remains critical. The development of efficient catalysts is equally important, particularly those based on inexpensive and non-toxic metals as alternatives to commonly used toxic metals such as Zn, Bi, Sn, and Cr. Additionally, the presence of additives like dyes, plasticizers, and food residues further exacerbates the complexity of the recycling process. Compared to mechanical recycling, current chemical methods are still in the early stages of development in terms of scale and efficiency. In addition, few initiatives have reached industrial scale, likely due to the lack of infrastructure for collection, sorting, depolymerization, purification, and storage.

CRediT authorship contribution statement

Samson Lalmangaihuala: Formal analysis, Software, Writing – original draft; **Monjuly Rongpipi:** Investigation, Writing – review and editing; **Khiangte Vanlaldinpuia:** Conceptualization, Supervision, Validation, Writing – review and editing; **Samuel Lalthazuala Rokhum:** Conceptualization, Supervision, Visualization, Writing – review and 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.

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Data Availability Statement

This is a review article. All the presented data are based on the literature reports.

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