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Review

Advancements in the Bio-Degradation of Plastic Waste into Value-Added Chemicals: A Recent Perspective

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ABSTRACT: Plastics are an essential component of modern life, but the plastic waste has caused significant environmental pollution and economic losses. The effective solution to these problems is the biodegradation and high-value conversion of plastic waste. After biodegradation, plastic waste is broken into smaller molecules and eventually transformed into innocuous substances like water, carbon dioxide and biomass. High-value conversion enables plastic waste to be converted into products with higher economic value and environmental friendliness. Based on this, we summarize the biodegradation methods of bioplastics and analyze the shortage of these methods. Subsequently, we summarize the progress of converting the degradation products into value-added chemicals, comprehensively analyze the advantages and disadvantages of these bioconversion process, and propose some strategies to address these disadvantages. Finally, we analyze the significance of establishing a microbial-based conversion process that integrates the degradation and the conversion, and propose some potential strategies.

Keywords: Bioplastics; Bioconversion; Biodegradation

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1. Introduction

Plastics have been applied in the fields of clothing, packaging, pharmaceutical compounds, and building, owing to the performance of light-weight, inexpensive, strong, durable, and flexibility [1,2]. In 2022, the global production of plastics amounted to 400.3 million tons, in which 90% derived from fossil fuels [3]. Given that the majority of plastics were disposable, the accumulation of vast plastic waste has resulted in severe environmental issues. For example, the global life-cycle greenhouse gas emissions from plastics were growing rapidly with an average annual growth rate of 4%. Additionally, the persistence of a substantial amount of plastic waste in the marine environment can cause severe damage to oceanic ecosystems and human health [4,5].

Four main strategies, which were used for plastic waste disposal around the world, included landfill, incineration, regenerative granulation, and pyrolysis [6]. Most plastic waste was degraded and processed through these strategies, but they also had some apparent drawbacks. Landfill carried the risk of soil leaching and compaction, potentially leading to groundwater contamination. The process of plastic waste through waste treatment systems can lead to the accumulation of organic pollutants, which had various adverse effects on human health [7,8]. The improper disposal of plastic waste may result in the loss of its economic value such as traditional plastics recycling process, which often involved energy-intensive treatment steps. In addition, the environmental pollution resulted in additional costs for environmental remediation and treatment [9]. In chemical recycling, pyrolysis and solvent decomposition routes can effectively convert polymers back into monomers and oligomers. However, the low cost of virgin monomers derived from fossil carbon-based feedstocks often rendered chemical recycling commercially unfeasible [10]. Biodegradation and sustainable conversion methods for plastic waste were gaining more attention due to their potential to address the shortcomings of traditional disposal methods. Biodegradation of plastic waste can be achieved through enzymatic degradation or microbial degradation. In comparison to chemical recycling, enzymatic degradation proved to be a

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gentler and more energy-efficient process, with reduced associated environmental pollution. Additionally, the utilization of biological techniques, such as enzyme immobilization, can facilitate the reuse of enzymes and reduce costs. Furthermore, the conversion of waste into high-value compounds enabled the reuse and the recycle of resources, reducing the necessity for virgin resources.

Biodegradation referred to the process in which microorganisms in the environment can convert materials into natural substances [11]. Biodegradable plastics, such as polylactic acid (PLA), poly(butylene succinate) (PBS), poly(butylene adipate terephthalate) (PBAT) and polyhydroxyalkanoates (PHAs) [12], had chemical structures that allowed them to be broken down by microorganisms. Microorganisms mainly broke down these plastics into one-carbon substances such as carbon dioxide and methane [13]. Non-biodegradable synthetic plastics are those that cannot be degraded significantly by microbial cells in the natural environment within a reasonable time. These plastics were usually made from petroleum-based raw materials, including polyethylene terephthalate (PET), polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC), and polystyrene (PS) [14]. Even so, they can be broken down into corresponding monomers or small molecules through enzymatic degradation. Some degradation products had natural value, such as adipic acid (AA) [15]. Some monomers can be converted into value-added compounds [16]. For instance, monomer ethylene glycol (EG) can be converted into glycolic acid (GA) [17]. The economic value of plastic waste was low in its original state, due to the costs associated with its disposal. Conversely, the value was significantly enhanced through the conversion of plastic waste into high-value chemicals. The process of high-value conversion of plastics can be divided into three stages: collection and handling of plastics, degradation and conversion of plastics, and separation of high-value products. Among them, the biodegradation and conversion of plastics was technically feasible. Several molecular manipulation tools can be used to modify microorganisms to enhance their biodegradation efficiency and optimize conversion pathways to meet the needs of industrial production. In addition, in the stage of collection and handling of plastics, the biodegradation process was a relatively low-energy process, with reduced energy costs compared to the physical method of recovery. The value of the target product had a significant influence on the economic viability of the conversion process. For example, the bioconversion of PET to adipic acid was potentially economically beneficial. The bioconversion has been technically achieved in *E. coli* with the conversion rate of 79%. Furthermore, the recycling rate of PET bottles in China has exceeded 94%, which provided an adequate raw material base and reduced the cost of raw materials for the high-value conversion of PET.

So far, many enzymes and microorganisms which can degrade plastics, have been reported. Based on this, this review summarized the current biodegradation and conversion of plastic waste and analyzes the current problems in converting plastics into value-added compounds. As shown in Figure 1, we classified the high-value conversion of plastics into direct conversion of monomers and continuous conversion. Finally, these value-added compounds can be used to manufacture cosmetics, clothes and bioplastic products.

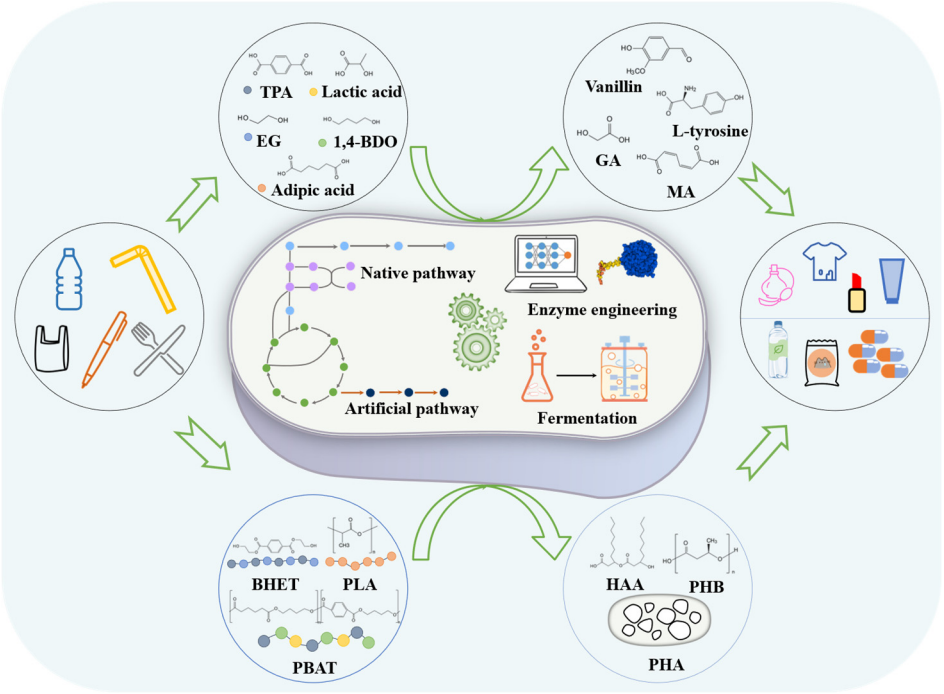


Figure 1. Bioconversion of the degradation products from plastics into value-added chemicals.

2. Biological Depolymerization Methods on Plastic

2.1. Depolymerization of Biodegradable Plastic

Biodegradable plastics, which included PLA, PBS, PBAT and PHAs, were the plastics that can be degraded by microorganisms to generate CO_2, CH_4, and microbial biomass [18]. Some microorganisms that degraded biodegradable plastic were listed in Table1. The biodegradability made these plastics significantly reduced their impact on ecosystems.

2.1.1. PHA

PHAs were biodegradable microbial polyesters that can be produced by microorganisms [19]. The superiority of PHAs compared with other plastics was the complete biodegradability under aerobic and anaerobic conditions [20]. Various microorganisms, which were capable of degrading PHAs, have been discovered in environments such as soil, compost, and ocean (Table 1) [21]. Among these degrading microorganisms, the main bacteria were *Pseudomonas lemoigne* [22], *Streptomyces* sp. IN1 [23], *Comamonas testosteroni* [24], and the main fungi were *Aspergillus fumigatus* [25], *Penicillium citrinum* S2 [26], and *Verticillium leptobactrum* [27]. Generally, the natural environments were conducive to the degradation of PHA in aerobic conditions. Besides, there were still some microorganisms that can degrade PHA under anaerobic conditions, such as *Ilyobacter delafieldii* sp. nov, *Cloacamonales*, *Thermotogales* and *Clostridium* [28–31]. *Ilyobacter delafieldii* sp. nov was the first anaerobic bacterium that can degrade exogenous PHB into 3-hydroxybutyrate. *Clostridium* that isolated from sludge can degrade PHB powder in a pH-controlled bioreactor system. The anaerobic degradation of PHB can be achieved within 3 days.

The enzymatic degradation of PHA can be categorized into intracellular depolymerases and extracellular depolymerases. Intracellular depolymerases broke down PHA chains into small molecules within the PHA-producing strains, and these molecules can be utilized as carbon and energy source for the growth of microorganism [32]. Extracellular depolymerases, which acted on PHA outside of cells, have been identified in various bacteria and fungi. These enzymes can encounter PHA biomaterials and initiate the process of depolymerization [33]. Recently, Zhou et al. investigated that *Schlegelella thermodepolymerans* (*S. thermodepolymerans*) may possess extracellular depolymerases, and *S. thermodepolymerans* showed a faster extracellular PHA degradation speed at 50 °C compared with mesophilic conditions [28,34]. The presence of these enzymes provided the possibility for the degradation of PHA under extreme conditions. However, the complete degradation of PHA to the monomers remained a challenge. For example, the use of base catalysts for the degradation of poly(3-hydroxybutyrate) (P3HB) only produced the non-original monomer transcrotonic acid, which affected the repolymerization of bioplastic P3HB [35].

2.1.2. PLA& PBS

PLA was a biodegradable plastic with a wide range of applications in packaging materials, agriculture and pharmaceuticals. According to PlasticDB (http://plasticdb.org), over 150 strains, 6 uncultured bacteria, and 27 PLA-depolymerases were capable of degrading PLA, blends of PLA, or copolymers of lactic acid. These strains contained bacteria like *Amycolatopsis* 3118 [36], *Bacillus amyloliliquefaciens* [37], *Bacillus smithii* PL21 [38], and as well as fungi *Cryptococcus magnus* [39] and *Penicillium*, etc [40]. The commercial product Proteinase K was a hydrolase which was capable of effectively degrading PLA [41,42]. Additionally, a fungal-like keratinase (CLE1) which was expressed in *Saccharomyces cerevisiae*, produced a crude supernatant that efficiently hydrolyzed different types of PLA materials, releasing up to 9.44 g/L lactic acid from 10 g/L PLA film [43]. PBS was a biodegradable plastic which can be synthesized through polycondensation reaction between 1,4-butanediol (1,4-BDO) and succinic acid. Microorganisms that can degrade PBS, contained *Actinokineaspora riparia*, *Sreptomyces megasporus*, *Micromonospora chalcea*, *Saccharothrix australiensis*, and *Excellospora viridilutea*, etc [44,45]. However, microbial degradation was often slow and required strict environmental condition. Currently, only a few enzymes had the ability of degrading PBS, and their efficiency were generally low. Low-efficiency enzymes prolonged the degradation process, which resulted in extended production cycles and decreased production efficiency. This further increased costs, including energy, labor, and equipment usage costs [46].

2.1.3. PBAT

PBAT, which was synthesized by polymerizing a mixture of AA, 1,4-BDO, and terephthalic acid (TPA) [47], has gained significant attention due to its desirable physical properties. One of its key characteristics was high flexibility

but relatively low strength compared with other bioplastics [48]. Several microorganisms that can degrade PBAT contained *Pseudozyma jejuensis*, *Stenotrophomonas sp. TGJ1*, *Cryptococcus sp. MTCC* 5455, *Bacillus subtilis ATCC* 21332, and *Thermobifida fusca FXJ-1* [49–53]. Biodegradable polyesters were typically broken down by enzymes such as esterases, lipases, proteases and cutinases. Among these hydrolases, lipases and esterases showed low efficiency due to the narrow substrate-binding tunnel, and the lid region that covered the active center. In contrast, cutinases harbored broad substrate-binding cavities and lid-free active site, which appeared to be the most promising PBAT hydrolase [54,55]. For example, the double mutation strategy was applied to modify cutinase that derived from *Thermobifida fusca*, generating a mutant with a more flexible substrate-binding pocket and high decomposition rate [56]. Moreover, cutinase variant TfCut-DM Q132Y can completely degrade PBAT film to TPA in 24 h at 60 ℃, which represented the most efficient and energy-saving choice for enzyme-mediated PBAT decomposition [57]. In addition, biosensors were applied to the high-throughput screening of hydrolases. Shin et al. constructed an AA biosensor based on PcaR transcription factor. The optimized sensor showed high sensitivity (low detection concentration < 10 μM) and dynamic range (~50-fold). In particular, the novel AA sensors can contribute to the discovery and evolution of effective biocatalysts [58]. However, the degradation of PBAT in the actual application process was not ideal. When degrading PBAT mulch that used in agriculture, the degradation process was usually affected by light, temperature, pH and mechanical forces [47]. Therefore, the development of enzymes or bacteria with improved stability and degradation efficiency of PBAT was urgently needed. These stable enzymes or bacteria can maintain activity under different environmental conditions and effectively break down PBAT.

Table 1. Microorganisms that degrade biodegradable plastic.

Plastics	Organisms	Source	Results	References
PHA	*Streptomyces sp. IN1*	Soil	Produced a thermoalkanophilic P(3HB-co-3HV) depolymerase	[23]
	Comamonas testosteroni	Marine	P(3HB) was degraded between pH 9.5 and 10.0	[24]
	Aspergillus fumigatus	Soil	3-hydroxybutyrate was detected as the end product at pH 8.5, 45–60°C	[25]
	Penicillium citrinum S2	Wastewater	β-hydroxybutyrate detected as the major end product of PHB hydrolysis at pH 6.0, 50 °C	[26]
	Verticillium leptobactrum	Soil	Degraded at an erosion rate of 0.03 to 0.64% weight loss per day	[27]
PLA	*Amycolatopsis* 3118	Soil	PLA film disappeared within 2 weeks	[36]
	Bacillus amyloliliquefaciens	Soil	45.5% was degraded at 37 °C after 25 days	[37]
	Bacillus smithii PL21	Compost	Purified enzyme was active against PLA at 60 °C and pH 5	[38]
PBS	*Actinokineaspora riparia*	ICM and IFO	PBS can be degraded by after 30 days	[45]
	Sreptomyces megasporus	ICM and IFO	PBS can be degraded by after 30 days	[45]
	Micromonospora chalcea	ICM and IFO	PBS can be degraded by after 30 days	[45]
	Saccharothrix australiensis	ICM and IFO	PBS can be degraded by after 30 days	[45]
PBAT	*Pseudozyma jejuensis*	Tangerine tree leaves	Weight loss effect exceeding 2%	[49]
	Stenotrophomonas sp. TGJ1	Farmland soil	Degraded 10.14% weight loss of PBAT films in 5 days	[50]
	Cryptococcus sp. MTCC 5455	MTCC	Complete degradation of PBAT films at 25 °C within 216 h	[51]
	Bacillus subtilis ATCC 21332	Soil	2.0 ± 0.1 mg weight loss after 21 days incubation at 30 °C	[52]
	Thermobifida fusca FXJ-1	Compost	Degraded PBAT films to 87.83% in 9 days	[53]

2.2. Depolymerization of Non-biodegradable Plastic

2.2.1. PET

Many methods can be used to degrade PET into monomers TPA and EG, such as hydrolysis, methanolysis, glycolysis, aminolysis, and ammonolysis [59]. Enzymatic hydrolysis offered a milder and more environmentally friendly approach to degrading PET compared with traditional chemical methods. Carboxylic ester hydrolases (EC 3.1.1) such as lipases, esterases, cutinases, and PETases, played a crucial role in the PET hydrolysis [60]. We listed enzymes that degraded PET in Table 2.

Tournier et al. utilized computer-aided enzyme engineering to modify leaf-branch compost cutinase (LCC) derived from leaf-cutinase branch compost metagenome. The LCC-variant (F243I/D238C/S283C/Y127G) has been designed to enhance its depolymerization specific activity, and the introduction of disulfide bonds at the calcium-binding site contributed to thermostability. The LCC-variant catalyzed PET to TPA with a mean productivity of 16.7 $g_{TPA} \cdot l^{-1} \cdot h^{-1}$ [61]. Cutinase 1 (ThcCut1) derived from *Thermobifida cellulosilytica* exhibited higher activity with ester bonds. The ThcCut1 mutant (G63A/F210I/D205C/E254C/Q93G) can degrade 96.2% of the post-consumer PET bottle particles after 96 h [62].

A two-enzyme system of PET hydrolase (PETase) and MHET hydrolase (MHETase) derived from *Ideonella sakaiensis* (*I. sakaiensis*), was employed to depolymerize PET to TPA and EG. The system was biochemically identified and characterized to degrade PET in vitro [63,64]. Recently, Cui et al. improved the robustness of a IsPETase (PETase derived from *I. sakaiensis*) through the GRAPE strategy (greedy accumulated strategy for protein engineering), the IsPETase mutant (DuraPETase) possessed high catalytic efficiency and worked efficiently at 60 °C for more than 24 h [65]. Further, Liu et al. designed DuraPETase through protein engineering. The constructed DuraPETase-4M can degrade 93.3% of amorphous PET powder and 69.8% of PET preformed film after 96 h. These degradation rates were 3.2 and 5.4 times higher than DuraPETase, respectively [66]. Directed evolution was conducted on PETase using a novel high-throughput fluorescence screening assay, which utilized a novel substrate bis(2-hydroxyethyl)2-hydroxyterephthalate (BHET-OH). The generated DepoPETase enabled complete depolymerization of untreated PET wastes at mild temperature [67]. The novel PET hydrolase CaPETase, which combined the advantages of PETase and LCC, exhibited high catalytic activity and high thermal stability at ambient temperature. After rational protein engineering, the variant CaPETase M9 showed a remarkable 41.7-fold increase in activity compared with the wild type at 60 °C [68]. A hydrophobic cell surface display (HCSD) system was constructed as a whole-cell catalyst. The truncated outer membrane hybrid protein FadL exposed PETase and hydrophobic protein HFBII to the surface of *E. coli* and exhibited efficient PET accessibility and degradation, with a higher degradation rate than free PETase [69]. Immobilization of enzymes was also an important method to improve enzyme activity. Immobilization of PETase onto $Co_3(PO_4)_2$ of the nanostructure resulted in an effective increase in enzyme loading, with long reaction times showing that the productivity of TPA was 3.5 times higher than that of the free enzyme [70]. The mining and directed evolution of PET and Phthalate esters (PAEs) hydrolases were slow due to the lack of fast and efficient detection methods. Li et al. established PA and TPA cell sensors by directional evolution transcription factor XylS, and used this sensor to aid the directed evolution of PAE hydrolase GoEst15. The degradation efficiency of dibutyl phthalate and 4-nitrophenylbutyrate in the resulting mutant was increased by 2 and 2.5 times compared with the wild type, respectively [71].

However, PET hydrolases exhibited significant degradation effects primarily on PET with low crystallinity, typically around 10%. If the crystallinity of PET exceeded 20%, high-temperature pretreatment was necessary. This process not only consumed a considerable amount of energy but also increased processing costs [72]. PET was typically composed of a complex structure comprising tightly packed parallel chains in the crystalline regions and disordered chains in the amorphous regions [73]. PET molecules with higher crystallinity were more tightly arranged, with smaller gaps between the molecules, making it more difficult for enzyme molecules to penetrate the structure of the PET molecule. Consequently, the rate of degradation was lower. Additionally, with increased crystallinity, the flexibility and mobility of the polymer chains were constrained, reducing their susceptibility to enzymes. Furthermore, the repetitive units of terephthalate present in PET also limited its mobility and reduced its degradation efficiency [74].

2.2.2. PE, PP, PS, PVC

Plastics with carbon-carbon backbone such as PE, PP, PS, PVC, were difficult to be degraded using microbes, because C-C bonds were resistant to microbial attack and biodegradation. Some Bacterial species and fungi can

depolymerize PE after some forms of pretreatment [75]. Over 20 bacterial genera can degrade PE such as *Pseudomonas*, *Ralstonia*, *Stenotrophomonas*, *Klebsiella* and *Acinetobactor* [76]. Fungi were thought to be more efficient than bacteria for the degradation of PE because they can attach to the hydrophobic surface of the polymers [77]. Fungal laccases and peroxidases, generally used by fungi to degrade lignin, showed high efficiency in degrading PE and PVC. However, the complex biodegradation mechanism of PE was not fully understood yet. Peixoto et al. found a novel role of nitrogen metabolism in the breakdown and oxidation of PE, which was associated with biological production of NOx in three PE-degrading strains of *Comamonas*, *Delftia*, and *Stenotrophomonas* [78]. The biodegradation of PE and PP was challenging due to the lack of unsaturated double-bonds in their structure. Consequently, the abiotic pretreatment before degradation was typically required. Some pretreatment methods, such as ultraviolet radiation (UV), heating, and chemical agent can enhance the degradation efficiency [79,80]. The biodegradation of PS by several microbes such as *Pseudomonas aeruginosa*, *Curvularia*, and *Rhodococcus ruber* etc, has been demonstrated [81–83]. The number of published studies on the degradation of polypropylene by various microorganisms was limited. Among the microorganisms that have been observed to degrade PP were *Pseudomonas* and *Vibrio*, as well as the fungus *Aspergillus niger* [84,85].

Table 2. Enzymes and microorganisms that degrade non-biodegradable plastic.

Plastics	Enzymes/Organisms	Strategies/Conditions	Degradation Efficiency	References
PET	LCC	LCC-mutant (F243I/D238C/S283C/Y127G)	Produced 16.7 g TPA/ L/h	[61]
	ThcCut1	ThcCut1 mutant (G63A/F210I/D205C/E254C/Q93G)	Degraded 96.2% of the post-consumer PET bottle particles after 96 h	[62]
	IsPETase	GRAPE strategy	Worked efficiently at 60 °C	[65]
	DuraPETase	Protein engineering DuraPETase-4M	Degraded 93.3% of amorphous PET powder and 69.8% of PET preformed film after 96 h	[66]
	CaPETase	Variant CaPETase M9	41.7-fold increased in activity compared with the wild type at 60 °C	[68]
	PETase	Immobilization of PETase onto $Co_3(PO_4)_2$ of the nanostructure	Productivity of TPA was 3.5 times higher than that of the free enzyme	[70]
PE	*Pseudomonas sp.*	Incubated at 30 °C after 6 months	Degraded up to 24.22%	[86]
	Stenotrophomonas	Incubated with bacterial isolates for 90 days	PE appeared clearly abraded	[87]
PS	*Pseudomonas aeruginosa*	Incubated at 30 °C after 30 days	10% weight loss	[81]
	Curvularia	Incubated at 25 °C after 9 weeks	Microscopic examination showed adherence	[82]
	Rhodococcus ruber	Incubated at 35 °C after 8 weeks	0.8% weight loss	[83]
PP	*Pseudomonas, Aspergillus niger*	Incubated at 30 °C after 175 days	60% weight loss	[84]
	Bacillus cereus	Incubated at 33 °C after 40 days	12% weight loss	[85]
PVC	*Pseudomonas citronellolis, Bacillus flexus*	Incubated 30 days	Degraded after incubation	[88]

3. Direct Conversion of Monomers into Value-Added Chemicals

The high-value conversion of plastic waste has been widely studied, with particular emphasis on the transformation of plastic monomers. For example, TPA was one of the monomers of PET, which can be converted into value-added compounds such as vanillin. Additionally, some monomers derived from plastic degradation, such as 1,4-BDO and succinic acid, were natural value-added compounds with significant applications.

3.1. EG

EG was widely used as antifreeze agent and chemical intermediate, especially as critical precursor in the production of PET [89,90]. The catabolic pathways of EG to value-added chemicals have been shown in Figure 2. Acetic acid bacteria (AAB), such as *Acetobacter*, *Gluconobacter*, *Gluconacetobacter* and *Komagataeibacter genera*, possessed a unique metabolism that allows them to oxidize several carbohydrates, alcohols and polyols into carboxylic acids, aldehydes or ketones [91]. EG can be converted into GA by several AAB especially *Gluconobacter oxydans* [92]. *Yarrowia lipolytica* (*Y. lipolytica*) may also share an incomplete EG oxidation pathway, because 429.5 mM GA can be obtained in bioreactors from EG [93]. EG was also naturally utilized by other bacteria like *Pseudomonas aeruginosa*. EG can be converted into glycolaldehyde and GA through two functionally redundant periplasmic Quino proteins PedE and PedH [94,95]. Franden et al. clarified the metabolic pathway of EG in *Pseudomonas putida* KT2440 (*P. putida* KT2440). The additional overexpression of the glycolate oxidase (*glcDEF*) operon overcame the toxicity of glycolaldehyde, and the engineered strain can utilize 31 g/L EG to produce medium chain length PHA (mcl-PHA) with a productivity of 0.06 g/g [96].

Wild type *Escherichia coli* (*E. coli*) cannot utilize EG as sole carbon for growth because the propanediol oxidoreductase (FucO) that utilized EG was oxygen sensitive [97]. Lu et al. discovered a FucO mutant (Fuco*) with I7L and L8V, which possessed improved oxygen tolerance and can be used to consume EG [98,99]. Fuco* and glycolaldehyde dehydrogenase (aldA) have been co-expressed in *E. coli* to produce GA. Pandit et al. integrated modeling with experimental investigations and identified oxygen concentration as a crucial metabolic regulator. Finally, a GA titer of 10.4 g/L was attained after 112 h in a fed-batch bioreactor [17]. L-tyrosine was one of the three aromatic amino acids involved in protein biosynthesis [100]. Panda et al. engineered *E. coli* to transform EG into L-tyrosine. The engineered strain produced 2 g/L L-tyrosine from 10 g/L EG through the improvement of EG assimilation and the overcoming of L-tyrosine feedback resistance, and this strain outperformed glucose under the same conditions. Otherwise, a similar quantity of L-tyrosine can be obtained when using EG derived PET waste to replace commercial EG [101]. Frazão et al. extended the range of glycolaldehyde-dependent pathways by demonstrating production of 2,4-dihydroxybutyric acid (DHB). The nonnatural pathway involved several enzymatic steps in the conversion of glycolaldehyde to DHB. The sequential action of the D-threose aldolase, D-threose dehydrogenase, D-threono-1,4-lactonase, D-threonate dehydratase and 2-oxo-4-hydroxybutyrate (OHB) reductase enzyme was employed. Indeed, incorporating an EG oxidation step into the nonnatural pathway enabled the conversion of EG into DHB [102].

Apart from the value-added chemicals mentioned above, EG also can be converted into important platform chemicals and green fuels such as ethanol. Acetyl-CoA was an important metabolic intermediate that can be used to regulate intracellular growth and generate a variety of chemicals such as 3-hydroxypropionic acid, terpenoids and fatty acids [103–105]. The carbon utilization efficiency of glycolaldehyde was found to be low in the native strains, serving a precursor function for producing acetyl-CoA. Recently, some new acetyl-CoA biosynthetic pathways have been created to increase carbon utilization efficiency [106]. A synthetic acetyl-CoA (SACA) pathway was constructed by repurposing glycolaldehyde synthase and acetyl-phosphate synthase, which was verified by ^{13}C-labeled metabolites. Meanwhile, SACA pathway was further verified by cell growth using glycolaldehyde, formaldehyde and methanol as supplemental carbon source. The SACA pathway was proved to be the shortest, ATP-independent, carbon-conserving and oxygen-insensitive pathway for acetyl-CoA biosynthesis [107]. The Ara5P-dependent glycolaldehyde assimilation pathway (GAA) was proposed for the conversion of EG into acetyl-CoA without carbon loss. GAA pathway has been verified in vitro and vivo [108,109]. Acetate and ethanol were important products of the acetyl-CoA metabolic pathway. *I. sakaiensis* was capable of growth on PET as the major carbon and energy source. The consumption of 25 mM EG resulted in the production of 3.3 mM ethanol and 21.8 mM acetate after 17 and 22 days, respectively. [110,111].

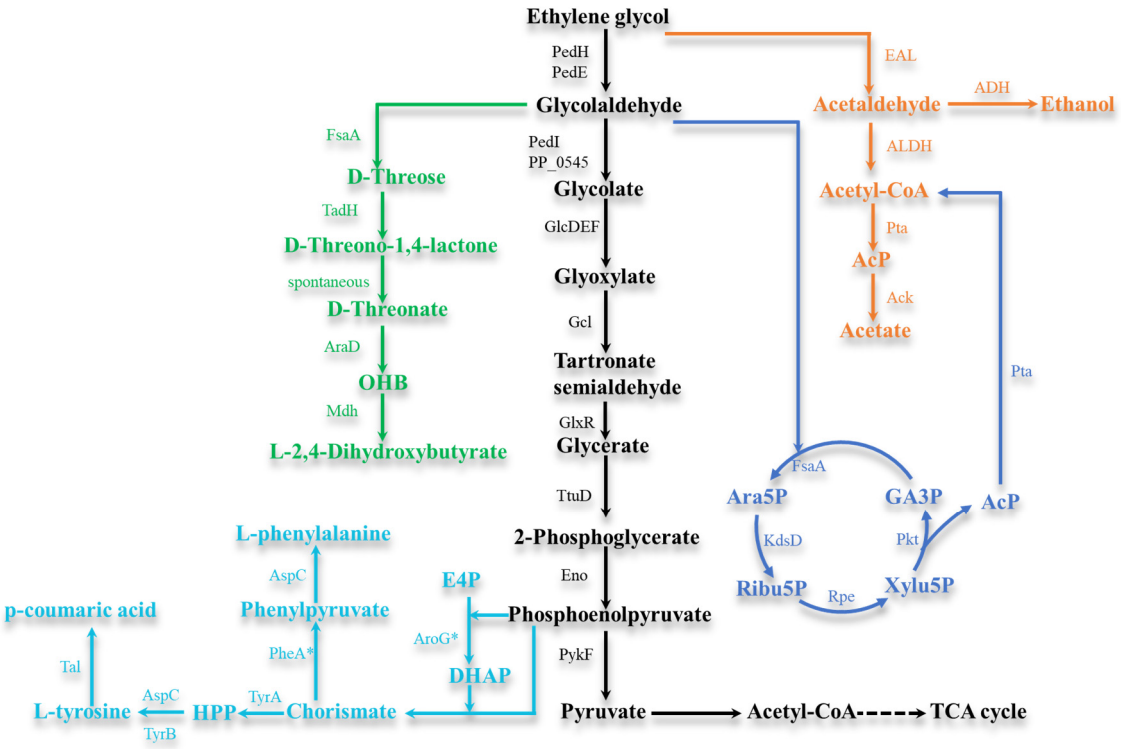


Figure 2. Catabolic pathways of EG to value-added chemicals. OHB:2-oxo-4-hydroxybutyrate; Ara5P: d-arabinose 5-phosphate; Ribu5P: d-ribulose 5-phosphate; Xylu5P: d-xylulose 5-phosphate; GA3P: glyceraldehyde 3-phosphate; E4P: erythrose-4-phosphate; DHAP: 3-deoxy-D-arabino-heptulosonate 7-phosphate; HPP: 4-hydroxyphenylpyruvate. PedE/PedH: PQQ-dependent alcolhol dehydrogenase; PedI: peroxisomal 3-ketoacyl-COA thiolase; GlcDEF: glycolate dehydrogenase; GlxR: tatronate semialdehyde reductase; Gcl: glyoxylate carboligase; TtuD: hydroxypyruvate reductase; Eno: Enolase; pykF: pyruvate kinase; FsaA: D-threose aldolase; TaDH： D-threose dehydrogenase; AraD: D-threonate dehydratase; Mdh: OHB reductase; FsaA: arabinose 5-phosphate aldolase; KdsD: D-arabinose 5-phosphate isomerase; Rpe: D-ribulose 5-phosphate 3-epimerase; Pkt: heterologous phosphoketolase; Pta: phosphate acetyltransferase; AroG*: DAHP synthase D146N; TyrA: chorismate mutase/prephenate dehydrogenase; Tal: tyrosine ammonia lyase; EAL: ethanolamine ammonia lyase; ADH: alcohol dehydrogenase; ALDH: aldehyde dehydrogenase; Ack: acetate kinase. PheA*: Bifunctional chorismate mutase/prephenate dehydratase T326P.

3.2. TPA

Currently, TPA was primarily obtained through the oxidation of p-xylene using either chemical or microbial methods [112,113]. TPA has been widely utilized in various fields, such as polyester fibers, feed additives and pharmaceuticals. Besides, TPA can also be converted into value-added compounds. Several strains, including *Comamonas sp.* E6, *I. sakaiensis*, *P. umsongensis* GO16, and *R. jostii RHA*1.13 [114], have been verified in metabolizing TPA.

TPA can be converted by the engineered microorganisms into five different aromatics or aromatic-derived compounds, including gallic acid, pyrogallol, catechol, muconic acid (MA), and vanillic acid (Figure 3). These five products were produced using protocatechuic acid as the principal intermediate, achieving relatively high molar conversion yields ranging from 32.7% to 92.5% [115]. Vanillin was a widely used additive in the food industry [116]. Sadler et al. developed a novel pathway in engineered *E. coli* for the direct conversion of TPA into vanillin using whole-cell catalysts. The pathway involved several enzymes including terephthalate 1,2-dioxygenase, dihydroxy-3,5-cyclohexadiene-1,4-dicarboxylic acid dehydrogenase, carboxylic acid reductase and catechol O-methyltransferase. After optimizing the reaction conditions, the maximum yield of vanillin reached 119 mg/L with a conversion rate of 79% [117]. In addition, four cleavable vanillin-based polyols were prepared from vanillin, 2,5-furandicarboxylic acid and TPA, followed by the cross-linking with two different isocyanates. The resulting polyurethane samples exhibited reasonable mechanical strength, thermostability, solvent resistance and outstanding degradability [118].

Bayer et al. proposed a new strategy for the conversion of TPA into various diamines. Carboxylic acid reductase (CAR) catalyzed TPA to produce terephthalaldehyde (TAL). TAL was catalyzed through a chemical reductive amination cascade in a one-pot process to produce 1,4-bis-(aminomethyl) benzene and benzylamine [119]. Additionally, para-

xylylenediamine (pXYL), which was used as a building block for polymeric materials, can be obtained of 69 ± 1% yield when combining CAR from *Segniliparus rotundus* (srCAR) and ω-transaminase from *Chromobacterium violaceum* (cvTA) [120]. Valenzuela-Ortega et al. developed the first one-pot bio-upcycling of PET plastic waste into AA in *E. coli*. The rate-limiting enoate reductase (BcER) was interfaced with hydrogen gas and a biocompatible Pd catalyst to synthesize AA. The product conversion is high (79%, 115 mg/L) under ambient conditions in 24 h [15].

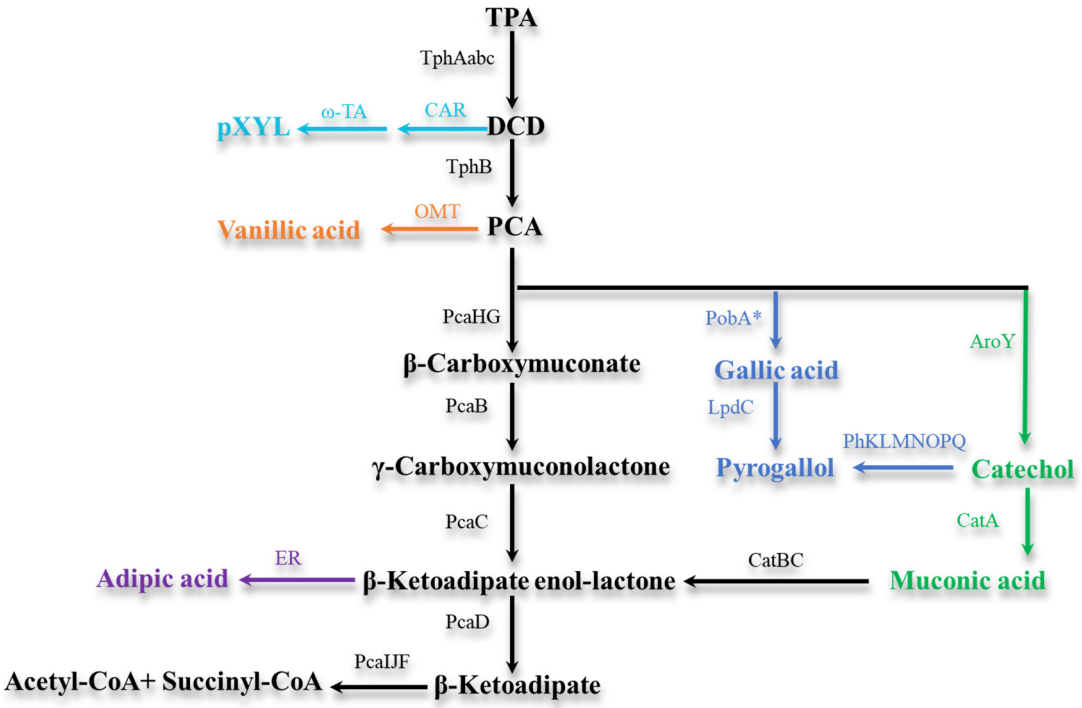


Figure 3. Catabolic pathways of TPA to value-added chemicals. TPA: Terephthalic acid; DCD: 1,6-dihydroxycylohexa-2,4-diene-dicarboxylate. TphAabc: TPA 1,2-dioxygenase; TphB: 1,2-dihydroxy-3,5-cyclohexadiene-1,4-dicarboxylate dehydrogenase; PcaHG: protocatechuate 3,4-dioxygenase; PcaB: 3-carboxy-cis,cis-muconate cycloisomerase; PcaC: γ-carboxy-muconolactone decarboxylase; PcaD: β-ketoadipate enol-lactone; PcaIJF: 3-oxoadipate CoA-transferase; PobA*: p-hydroxybenzoate hydroxylase T294A/Y385F; LpdC: Gallic acid decarboxylase; AroY: PCA decarboxylase; PhKLMNOPQ: phenol hydroxylase; CatA: catechol 1,2-dioxygenase; CatBC: Muconate cycloisomerase 1/Muconolactone Delta-isomerase; ER: Enoate reductase; ω-TA: ω-transaminase; CAR carboxylic acid reductase; OMT:O-methyltransferase.

3.3. AA

AA was a dicarboxylic acid with a wide range of applications, especially used for the production of PBAT and nylon 6-6 fibers, microbial degradation of AA has been characterized in *Acinetobacter baylyi* [121–123]. Fedorchuk et al. reported the biocatalytic conversion of AA into 6-aminocaproic acid (6-ACA) and 1,6-hexamethylenediamine (HMD), the key precursors for nylon synthesis (Figure 4). The cascades of reduction/amination reactions involved carboxylic acid reductases (CARs) and transaminases (TAs). To increase the cascade activity for the transformation of 6-ACA to HMD, three CARs mutant with enhanced activity were designed by protein engineering. For the transformation of AA to HMD (via 6-ACA), the wild type CAR was combined with the variant and two different TAs, resulting in up to 30% conversion to HMD and 70% to 6-ACA [124]. Besides, new CARs were created using accurate protein structure prediction-based virtual screening method, resulting in twice conversion in the enzymatic cascade synthesis of HMD [125]. Additionally, *P. putida* was engineered to produce PHA when utilizing AA as a carbon source. The initial uptake and activation steps for dicarboxylates were conducted, and adaptive laboratory evolution was employed to enable the growth of *P. putida* on AA as the sole carbon source. In a nitrogen-limited mineral medium, the engineered strain produced 25.3 ± 4.2% PHA, representing a yield of 9.2% (g/g of carbon) [126].

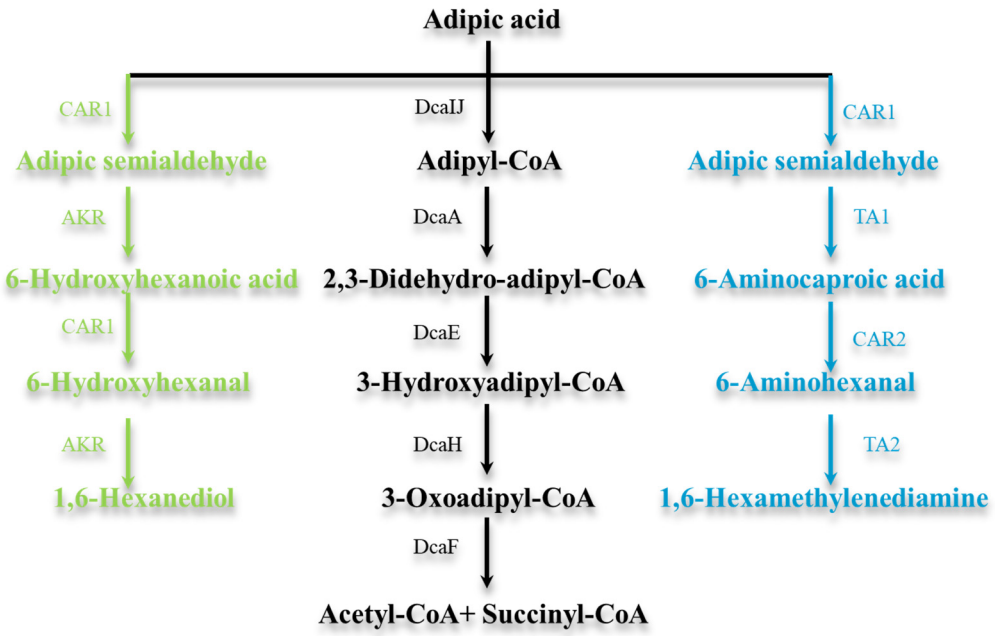


Figure 4. Catabolic pathways of AA to value-added chemicals. DcaIJ: CoA transferase subunits; DcaA: acyl-CoA dehydrogenase; DcaE: enoyl-CoA hydratase; DcaH: hydroxyl-CoA dehydrogenase; DcaF: thiolase; CAR1: carboxylic acid reductase; CAR2: engineered CAR; AKR: aldo-keto reductases; TA1/2: ω-transaminase.

3.4. Lactic acid

Lactic acid was an important building block especially for the production of PLA [127,128]. Three methods were known for the synthesis of polymer PLA (Mw > 10,000), including direct condensation polymerization, azeotropic dehydration condensation, and lactide ring-opening polymerization [129]. Lactic acid was also used in the production of some organic acids. The strain *Megasphaera elsdenii* has been described to consume lactic acid and convert it into n-caproate. In this process, lactic acid was converted to acetyl-CoA, which then entered the reverse β-oxidation cycle. A maximum n-caproate productivity of 6.9 g COD/L-day was observed in 17 days (COD represents chemical oxygen demand), representing the highest sustained lactate to n-caproate conversion rate [130]. Propionate was a valuable platform chemical with versatile applications such as food preservatives. *Clostridium saccharoperbutylacetonicum* has been engineered to produce propionate by implementing the acrylate pathway from *Anaerobiospirillum succiniciproducens* and introducing a D-lactate dehydrogenase enzyme. This modification allowed the bacterium to metabolize sugars to produce propionate as an end product [131].

3.5. Succinic acid and 1,4-BDO

Succinic acid has been recognized as one of the most promising bio-based platform chemicals. In 2017, the market of bio-succinic acid was estimated at 175.7 million US dollars. The market is expected to grow at an annual growth rate of 20%, reaching 900 million US dollars by 2026 [132]. Cui et al. engineered the strictly aerobic yeast *Y. lipolytica* for efficient succinic acid production without pH control, the engineered strain produced 111.9 g/L succinic acid with a yield of 0.79 g/g glucose within 62 h [133]. Succinic acid can be utilized as a precursor for industrially valuable commodity chemicals including 1,4-BDO, 5-Aminolevulinic acid (5-ALA), gamma-butyrolactone, tetrahydrofuran, and as a monomer for manufacturing various polymers [134].

1,4-BDO was involved in the synthesis of PBS and PBAT as a monomer. In 2015, the global market size of 1,4-BDO was valued at 6.19 billion dollars, which is expected to reach 12.6 billion dollars by 2025 [135]. It was known that 1,4-BDO can be produced in *E. coli* by introducing an artificial pathway involving six exogenous genes. In this pathway, succinate and succinyl-CoA were important precursors to synthesize 1,4-BDO [136]. Metabolism of 1,4-BDO has been reported in *P. putida* KT2440 and *Y. lipolytica*. Adaptive Laboratory Evolution enabled strain *P. putida* KT2440 to grow using 1,4-BDO as the sole carbon source.1,4-BDO was oxidized to 4-hydroxybutyrate (4-HBA), which can be further metabolized to support growth by *P. putida* KT2440. [93,137].

4. Continuous Conversion of Plastic into Value-Added Chemicals

The transformation of monomers after plastic degradation partially alleviated the issue of plastic waste. However, the conversion may require an additional carbon source to support growth. Moreover, the separation and purification of plastic monomers from their hydrolysis products are time-consuming and resource-intensive, rendering the entire process unsustainable. Continuous conversion from plastic to value-added compounds demonstrates significant advantages compared with monomers conversion. Continuous conversion eliminated the purification process of intermediates and the need for obtaining purified monomers. Additionally, direct utilization of hydrolysis products also enhanced the efficiency of conversion.

As shown in Figure 5A, Liu et al. proposed a multifunctional *P. putida* KT2440 for converting PET into MA, a precursor for the production of AA or TPA [138]. The engineered strain was constructed to simultaneously secrete PET hydrolase LCC and synthesize MA. The hydrolysis product TPA served as a precursor for MA production, while EG supported cell growth. After optimizing the process, the yield reached 0.50 g MA/g PET [139]. Diao et al. developed a hybrid strategy for converting PET into lycopene (Figure 5D). PET was converted to TPA and EG using alkaline hydrolysis. Then, *Rhodococcus jostii* strain PET (RPET) was able to directly use both TPA and EG as sole carbon sources to produce 1300 μg/L of lycopene, which provided beneficial effects to cellular functions in the human body [140,141].

In addition to using a single strain for the conversion of PET, the hybrid upcycling approach greatly increased the rate of PET degradation and conversion efficiency. Kim et al. developed a one-pot chemo-bioprocess, which integrated PET chemical glycolysis, enzymatic hydrolysis, and bioconversion (Figure 5B). The pivotal strategy involved employing the biocompatible catalyst betaine during glycolysis. Finally, protocatechuic acid and GA were obtained [142]. Besides, *Bacillus subtilis* esterase (Bs2Est) catalyzed TPA to catechol via a whole-cell biotransformation. The catechol solution produced from TPA can be directly used for functional coating on various substrates after simple cell removal from the culture medium without further purification and water-evaporation [143]. Tiso et al. utilized evolved *P. umsongensis* GO16 KS3, which was capable of growth with the PET hydrolysis solution, to produce hydroxyalkanoyloxy alkanoates (HAAs) at a yield of 35 mg/L. HAAs showed surface-active properties and can be polymerized into bio-based poly(amide urethane) (bio-PU) [144,145]. Other than PET, PLA also can be converted into value-added compounds in a one-pot process. PLA can be converted into alanine by a simple ammonia solution treatment using a Ru/TiO2 catalyst. The process had a 77% yield of alanine at 140 °C [146]. Jiao et al. demonstrated a one-pot catalytic process for the direct cracking of PLA into acrylic acid. This process employed an acid catalyst in conjunction with an ionic liquid, Bu4PBr, which facilitated the conversion of PLA to acrylic acid. The acrylic acid was then vaporized directly from the reactor and collected in a collector when PLA was completely converted [147].

The co-culture of microorganisms has emerged as a more promising approach to enhance biosynthetic efficiency. Compared with monoculture, microbial co-culture can reduce the metabolic burden and meet the expression requirements of different genes. Additionally, multiple strains can provide varied cellular environments and confer significant benefits in utilizing mixed substrates. Liu et al. developed a one-step biological process for converting PET oligomers to PHA by the co-cultivation of *E. coli* and *P. putida* KT2440. The system produced PHA utilizing PET as substrate. *E. coli* BL21 (DE3) was used to express and secrete the PET hydrolase (Figure 5C). The genetically modified strain *P. putida* was used to activate the metabolic pathway of TPA and EG while simultaneously overexpressing the PHA synthetic pathway [148]. Liu et al. proposed a co-culture system to directly hydrolyze bis(2-hydroxyethyl) terephthalate (BHET) to generate the PHB in one fermentation step. The system contained a *Y. lipolytica* Po1f harboring PETase from *I. sakaiensis* and a TPA-degrading *Pseudomonas stutzeri* containing the PHB synthesis pathway [149,150]. Bao et al. presented a synthetic microbial consortium that involved two *Pseudomonas putida* strains, specializing in TPA and EG utilization respectively. Compared with a single strain for TPA and EG co-consumption under various substrate conditions, the microbial consortium has reduced catabolic cross-interactions between the pathways for a faster rate of substrate assimilation. The engineered system can be further optimized to design and integrate metabolic pathways that synthesized mcl-PHA and MA [151].

The continuous conversion approach enhanced the efficiency of high-value transformation of plastics. In comparison to monomer conversion, it eliminated the need for monomer separation and purification steps. Moreover, the combination of multiple methods ensured an efficient connection between degradation and conversion, reducing the loss of intermediate products.

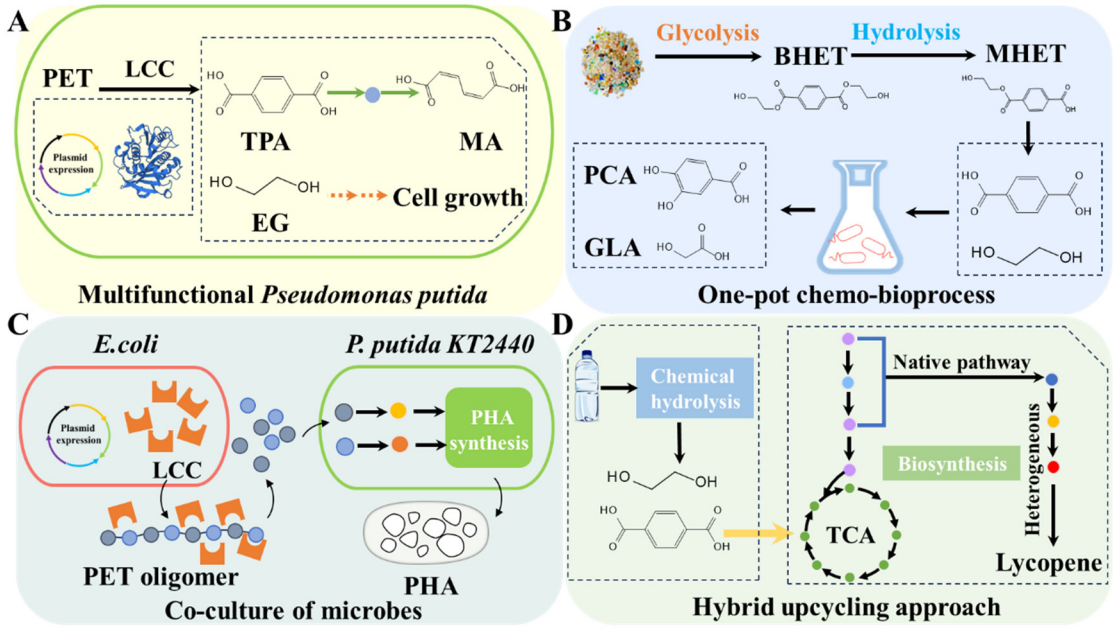


Figure 5. Four methods for continuous conversion of plastic into value-added chemicals.

5. From Plastic to Bioplastic

Bioplastics referred to the plastics of bio-based origin, or the plastics with biodegradable property, they have been applied in packaging, consumer goods, textiles, and biomedical devices. Total production of fossil plastics currently exceeded 380 million tons per year, while global production of 100% bio-based polymers was approximately 2 million tons per year. The global market share of bioplastics was expected to remain low at 2%, mainly driven by upcoming market regulations as well as increased consumer demand for sustainable products. If bioplastics received subsidies and political support, global growth could reach 10–20% [152]. The bioplastics market was expected to grow at an annual rate of 7.1% until 2026 and the revenue from bioplastics was expected to reach $4.4 billion by 2026 [153]. The growing interest of some major organizations and chemical companies in developing and producing sustainable plastics expanded the bioplastics market in the future [154]. As shown in Figure 6, transforming plastic waste into bioplastics through biological methods contributed to a more sustainable method on plastic production and usage. Initially, plastic waste was crushed into polymers through machine processing. The polymers were then biodegraded by enzymes into synthetic monomers, which can be synthesized into bioplastics such as PHA through polymerization. PHA can be reused in food packaging, clothing industry and other fields after processing. This process demonstrated the potential for the reuse of plastic waste.

The conversion of PET into bioplastic PHA was currently being investigated to address plastic pollution and promote sustainable plastic production. *P. umsongensis* GO16 can use TPA as the sole carbon source for the synthesis of both mcl-PHA and short chain length PHA (scl-PHA). Tiso et al. engineered *P. umsongensis* GO16 by adaptive laboratory evolution, which allowed *P. umsongensis* GO16 to use EG as sole carbon source and TPA for PHA synthesis, the PHA concentration peaked at 0.15 g/L [145,155]. Dou et al. proposed a hybrid upcycling approach to converting mixed PET/PLA. They used a biocompatible ionic liquid (IL) to chemically depolymerize plastic and then converted the depolymerized stream via biological upgrading. More than 95% of mixed PET/PLA was depolymerized into the respective monomers, which then served as the sole carbon source for the growth of *Pseudomonas putida*, enabling the conversion of the depolymerized plastics into PHA [156]. Many non-biodegradable plastics (PE, PP, PS) can also be transformed into bioplastics PHA. Plastics were depolymerized and converted into small monomers, which were utilized for the conversion to PHA via fermentation process [157]. *Pseudomonas aeruginosa* PAO-1 accumulated PHA with almost 25% of the cell dry weight as mcl-PHA when supplied with the PE pyrolysis wax in the presence of rhamnolipids. This was the first time that PE was employed as an initial substance for the manufacturing of a biodegradable polymer [158]. Johnston et al. investigated the molecular structure of the PHA that produced via *Cupriavidus necator* H16. *Cupriavidus necator* H16 consumed oxidized PP waste to produce PHA tert-polymer, the ESI-MS/MS analysis confirmed the result [159].

Some research indicated that the products of plastic degradation can be utilized for plastic repolymerization. Li et al. synthesized 554 mg virgin PET directly from the chemical-enzymatic degradation PET solution and completed a closed-loop PET recycling. The entire cycle from degradation to polymerization can be completed within a few days [160]. Youngpreda et al. conducted the re-polymerization of PLA. The protease produced by *Actinomadura keratinilytica* strain T16-1 was used to degrade PLA, and the degradable products lactic acid were re-polymerized by using commercial lipase as a catalyst under a nitrogen atmosphere for 6 h [161]. In addition, circular biological recycling of PHB has been reported. Chemical or enzymatic degradation of PHB can produce crotonic acid (CA), 3-hydoxybutyrate methyl ester (3HMB) and 3-hydroxybutyrate (3HB). *Paraburkholderia sacchari* can utilize 3HB to produce new PHB. Sequential addition of 3HB led to 6.5 g/L total dry weight and 2.5 g/L PHB with 0.26 g/g yield, which was slightly higher than the respective values achieved on glucose [162].

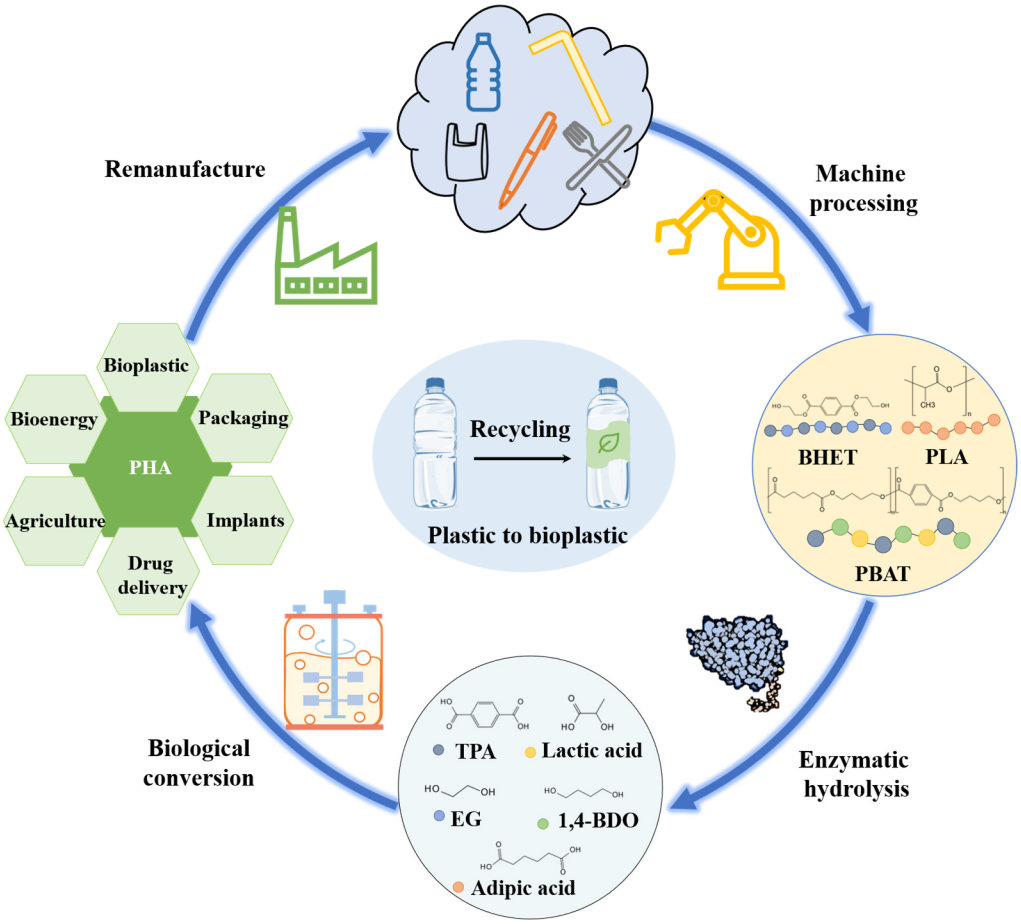


Figure 6. The cycle from plastic to bioplastic.

6. Conclusion and Future Perspectives

The increasing waste and impact caused by plastics have become significant concerns. Converting plastic waste into value-added compounds can greatly alleviate the current accumulation of plastic waste. The design and modification of hydrolase play a crucial role in the degradation of plastics and further conversion process. Currently, the methods for modifying hydrolases involve rational protein design, directed evolution, and random mutagenesis [163]. New artificial intelligence techniques such as machine learning and deep learning are becoming increasingly important in this field. Machine learning can effectively analyze large datasets and hold great promise for accelerating the development of new enzymes. It has been widely applied in enzyme engineering to alleviate the burden of high-throughput experiments [164]. Some machine learning methods have been employed to improve enzyme stability, catalytic activity, and thermostability [165]. Moreover, in the high-value conversion process, the construction of new synthetic pathways also requires machine learning for the exploration of novel enzymes. Some new methods which can convert degradation products have been studied. Electrochemical upcycling has recently emerged as a new alternative, which enables precise control of chemical reactions. Electrocatalytic treatments involve chemical reactions that

facilitate electron transfer through the electrode/electrolyte interface driven by electrical energy [166]. *Tenebrio molitor*'s abundant gut microbiota makes it a promising solution for plastic degradation and the biological retrieval of PHA [167].

The continuous conversion of plastics into value-added compounds avoids the depletion of intermediate degradation products, thereby enhancing conversion efficiency. Currently, the most effective methods involve the combination of chemical and biological approaches or co-cultivation techniques. The combination of chemical and biological approaches involves the modification of degradative enzymes and design of conversion process. Biosensors can be used to optimize the combination of chemical and biological approaches. On the one hand, biosensors have been designed to identify hydrolysis products, thereby screening efficient hydrolase and optimizing the biodegradation [71]. On the other hand, biosensors can be used to control metabolic pathways and reconstruct the conversion circuits [168]. Coupling the sensing elements of biosensors with downstream responsive gene circuits enables the dynamic control of plastic conversion. Besides, the co-culture of microorganisms provides an optimal catalytic environment for continuous conversion of plastics, but the incompatibility between different species may break the balance between biodegradation and bioconversion. Computational modelling can be used to balance degradation and conversion process by designing metabolic network. In addition, several technologies, including genome engineering, macro-proteomics, and macro-metabolomics contribute to the co-culture of microorganisms. In summary, the high value conversion of plastics will be further improved through the development of a range of tools and the combination of technologies.

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Informed Consent Statement

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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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