Monomer recycling of polyethylene terephthalate, polycarbonate and polyethers : scalable processes to achieve high carbon circularity

Reference:

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Monomer recycling of polyethylene terephthalate, polycarbonate and polyethers: scalable processes to achieve high carbon circularity

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List of abbreviations

[Bmim][Ac]	1-butyl-3-methylimidazolium acetate
[Bmim][OH]	1-butyl-3-methylimidazolium hydroxide
[Ch][Ac]	Choline acetate
[Ch][For]	Choline formate
ABS	Acrylonitrile butadiene styrene
BHET	Bis-hydroxyethyl terephthalate
BPA	Bisphenol-A
CAGR	Compound annual growth rate
CaO NPs	Calcium oxide nanoparticles
CeO_2	Cerium oxide
DABCO	1,4-diazabicyclo[2.2.2]octane
DBSA	dodecylbenzene sulfonic acid
DBU	1,8-diazabicyclo (5.4.0)undec-7-ene
DMAP	4-(dimethylamino)pyridine
DMT	Dimethyl terephthalate
EARTO	European Association of Research and Technology Organizations
EG	Ethylene glycol
$FeCl_3$	Iron chloride
FRC	Fiber reinforced composites
GO	Graphene oxide
IL	Ionic liquid
IPP	Isopropylene phenol
K_3PO_4	Potassium phosphate
LCC	Leaf-branch compost cutinase
LDO	Layered double oxides
MeOH	Methanol
MMT	Million metric tons per year
Mn_3O_4	Manganese oxide
Na2CO3	Sodium carbonate
NaOH	Sodium hydroxide
NMP	N-methyl-2-pyrrolidone
OP	Oxygenated plastic
PA	Polyamide
PC	Polycarbonate
PE	Polyethylene
PEF	Polyethylene furanoate
PEG	Polyethylene glycol
PET	Polyethylene terephtalate
PG	Propylene glycol
PHA	Poly(hydroxyalkanoate)
PHA	polyhydroxyalkanoate

PLA	Poly lactic acid
PP	Polypropylene
PPG	poly(propylene glycol)
PS	Polystyrene
PTMG	poly(tetramethylene glycol)
PU	Polyurethane
PVC	Polyvinyl chloride
SiO_2 NP	Silicon dioxide nanoparticle
TA	Terephthalic acid
TBACl	Tetrabutyl ammonium chloride
TBD	Triazabicyclodecene
THF	Tetrahydrofuran
TMDS	1,1,3,3 Tetramethyldisiloxane
TRL	Technology readiness levels
VA	Vinyl acetate
$Zn(OTf)_2$	Zinc trifluoromethanesulfonate
ZnO	Zinc oxide

Abstract:

This review presents a comprehensive description of the current pathways used in the chemical recycling of oxygenated plastics, with a specific focus on poly(ethylene terephthalate) (PET), poly(bisphenol-A carbonate) (PC), and polyethers including anhydride-cured epoxies. For PC and PET, the emphasis is on processes that achieve high depolymerization efficiencies as well as monomer selectivity and the potential to simplify downstream processing for the recovery of pure monomers. In the case of epoxies, this work focuses on depolymerization processes that produce curable molecules, as studies on epoxy depolymerization are scarce. To assess scalability, different depolymerization pathways are compared for each polymer based on the process conditions and monomer yields. The review concludes with the discussion on potentials and challenges of the distinct depolymerization pathways that have been developed for oxygenated plastics, such as hydrolysis, alcoholysis, and reductive depolymerization.

Keywords: *oxygenated plastics, chemical recycling, monomer recycling, depolymerization, circular economy*

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1. Introduction:

With rising plastic demand, the consumption of oil for plastic production is predicted to increase threefold by 2050 and to reach 15–20% of the total oil production [1]. This will further exacerbate the demand for already depleting fossil fuel sources. Furthermore, the ecological disruptions caused by the excessive use and improper disposal of plastics are evident. A substantial amount of untreated plastic waste can be found in oceans and on continents [2, 3]. Based on estimations, nearly 30% of all plastics produced between 1950 and 2015 are still in use, while only 7% have been recycled, and 8% have been incinerated (Fig. 1). Unfortunately, a significant proportion (55%) accumulates in landfills and oceans (Fig. 1) [2]. If this trend continues to 2050, enormous amount of plastics will enter the oceans and even exceed the quantity of fish in oceans [2, 4].

Worldwide 390.7 million tons of plastics were produced in 2021 and is projected to grow at a compound annual growth rate of nearly 5% (Fig. 1) [5-8]. Around 10% of global plastic production consists of oxygenated plastics (OPs), which contain C-O bond in their polymer backbone. Among various OPs, poly(ethylene terephthalate) (PET) dominates the market with a production volume of more than 60 million tons/year. Whereas approximately 4.92 million tons of polycarbonates and 3.6 million tons of epoxy resins are produced annually [6, 7, 9, 10]. Mitigating the plastic problem and reducing dependency on fossil resources for plastic production is a point of debate in policy-making and scientific communities. Plastics derived from renewable sources have been proposed as one of the solutions [11-13]. In the context of oxygenated plastics, researchers have investigated bio-based alternatives as potential substitutes for PET. Typically, investigated alternatives are poly(ethylene furanoate) (PEF), poly(

lactic acid) (PLA) and polyhydroxyalkanoates (PHAs) [5, 14]. However, these oxygenated bio-based alternatives are still in early stages of development, representing only 0.5% of global plastic production and not capable of replacing plastics derived from fossil sources [5]. A significant portion (55%) of these bioplastics is not biodegradable [15]. Additionally, they do not provide a solution for managing or treating current plastic waste. Implementing efficient plastic recycling processes is crucial to manage plastic pollution. This applies to the waste generated during the production process (post-industrial waste), that can be recycled more easily with minimal cleaning (primary recycling), and to post-consumer waste. Post-consumer waste requires a more elaborate sorting and cleaning process based on the composition of the waste streams (secondary recycling) [16].

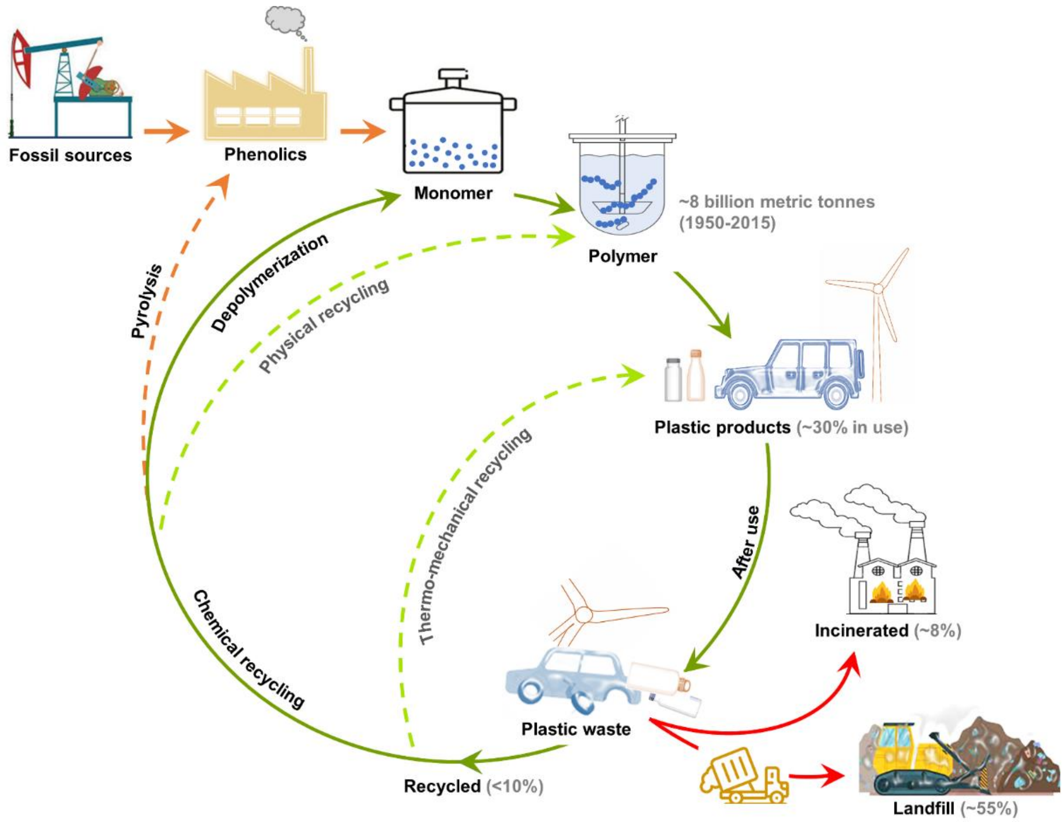


Fig. 1. Global plastic production, use, disposal and recycling between 1950 to 2015. Data taken from ref. [2, 3].

The traditional approach to recycle plastic waste is based on thermomechanical processing. After sorting and cleaning, the end-of-life materials undergo grinding and pelletization [16]. Although the process is simple and relatively inexpensive, partial degradation of the polymer is inevitable. The presence of residual additives can deteriorate the mechanical properties and limit the application as well as economic value of the recycled material [17-20]. Therefore, thermomechanical recycling often leads to downcycling instead of true recycling. To eliminate these drawbacks of thermomechanical recycling, recently dissolution-precipitation is considered as an alternative and physical recycling approach to recover intact polymers from a mixed plastic waste stream (Fig. 1) [21-23]. With further development and utilization of computational solvent selection tools, overall process costs can be reduced further and prices of recovered polymers can match those of virgin polymers [24, 25].

Although physical recycling methods can be very effective in treating relatively pure materials, difficulties arise while handling waste stream impurities, complex blends, and thermosets. Polyolefins, for example, account for about 70% of the global polymer consumption (Fig. 2) and are mostly mechanically recycled if the waste stream allows. Otherwise, polyolefin waste is pyrolyzed (chemical recycling) to lubricants (C_{22}–C_{50}) and alternative fuel (C_5–C_{21}) with high calorific value [26-29]. Currently, depolymerization of polyolefins remains a high energy-consuming process due to the high depolymerization energy required to break C-C polymeric backbone (Fig. 2b). With advancements in catalytic research, one can foresee improvements in the depolymerization processes of waste polyolefins into various valuable chemicals [28-30]. However, chemical recycling technologies that can selectively deconstruct polymers into their constituent monomers/building blocks can simplify the downstream purification steps and make the process economically more attractive. This is desirable to achieve circularity in plastic production. Monomers recovered after these depolymerization processes can be reused indefinitely for plastic production, and thereby reducing dependency on fossil sources.

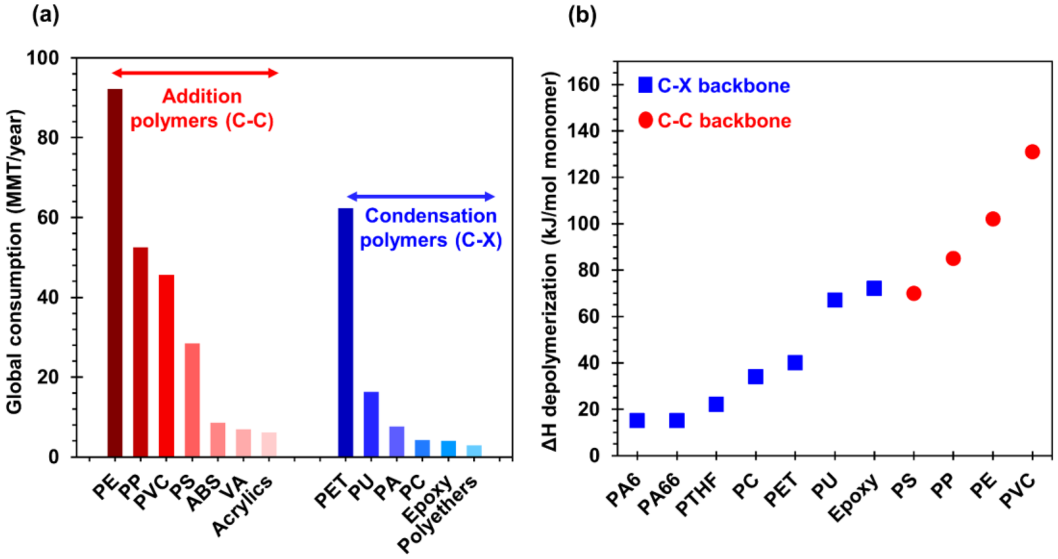


Fig. 2. (a) Consumption of addition (C-C) and condensation polymers (C-X) in million metric tons/year (MMT/year). Data taken from ref. [6, 7]. Plastics with consumption >3.0 MMT/year are presented in the figure. (b) The heat of depolymerization (ΔH) of condensation and addition polymers. Data taken from ref. [31, 32]. Polymers presented in the figures are: polyethylene (PE), polypropylene (PP), poly(vinyl chloride) (PVC), polystyrene (PS), vinyl acetate (VA), acrylonitrile butadiene styrene (ABS), poly(ethylene terephthalate) (PET), polyamide (PA), polycarbonate (PC), and polyurethane (PU).

Selective cleavage of the desired bonds in the polymer chain is necessary for efficient recovery of constituent monomers. The lower depolymerization energy of condensation polymers (C-X) allows selective cleaving the C-O or C-N bonds while keeping the C-C bonds intact

(Fig. 2b) [31, 32]. However, when the complexities in the heteroatom containing polymeric network increase, the energy required for depolymerization also starts to increase. For example, crosslinking in epoxies and polyurethanes is mainly responsible for the higher ΔH of depolymerization (Fig. 2b). When it comes to consumption of condensation polymers, consumption of C-O backbone containing polymers (OPs) are the most popular (Fig. 2a). Unlike polyolefin, the pyrolysis of OPs is not economically viable operation because it primarily yields gases (CO and CO_2), char and a small amount of liquid products with low calorific value [27, 33-35]. Therefore, OPs are transformed to (i) new building blocks that can be utilized for the preparation of valuable materials (open-loop recycling or upcycling) [36-42] or (ii) monomers via selective C-O cleavage to recover the constituent monomers [7, 42-47]. Depolymerization to the monomer is also known as closed-loop recycling because it allows the reintegration of the recovered monomers to create the same polymeric materials [44-47]. This is more promising than open-loop recycling to ensure circularity in OP production and address plastic pollution. Apart from reducing plastic pollution, development in depolymerization technologies for OPs can contribute to multiple Sustainable Development Goal (SDG) identified by United Nations [48].

Several competitive approaches have been investigated for the efficient depolymerization of OPs [45, 47, 49-51], and they can be broadly classified as hydrolysis, alcoholysis, and aminolysis. Recently, reductive pathways were also investigated to improve monomer selectivity and efficiency. Several recent reviews summarize and discuss latest advancements in the field of plastic recycling [21, 28-30, 40, 43, 52]. This enhances the overall understanding of processes and technologies available in this field. However, discussions on the scalability of existing and emerging technologies are rare. To address this gap, this review will focus on efficient depolymerization processes for OPs (PET, PC, and polyethers) that have been selected based on their efficiency and monomer selectivity. Typically, the scalability of novel processes depends on their operating conditions, depolymerization efficiency, and monomer selectivity. These factors have a significant impact on the cost of downstream processing. To identify the most efficient and potentially scalable technologies, comparison is made for each polymer based on their operating conditions, products, and current Technology Readiness Level (TRL).

2. Depolymerization of PET

PET is a semicrystalline polymer synthesized via transesterification of ethylene glycol (EG) with terephthalic acid (TA) or dimethyl terephthalate (DMT). The unique mechanical and optical properties of PET make it a widely used polymer in textiles, packaging and composites. This makes PET as the most commonly used OP and gives it a high economic significance [53]. When it comes to recycling, PET is primarily recycled via mechanical recycling, which decrease the quality of recycled material. Therefore, recycled materials are often blended with virgin PET to meet the quality requirements. PET materials are also converted into fibers for use in textiles [54, 55]. However, presence of other fibers as a blend of PET and complex composition of textiles makes the mechanical recycling quite challenging.

In recent years, extensive research has been conducted on the chemical recycling of PET via depolymerization into valuable building blocks (Fig. 3) [56, 57]. Processes such as methanolysis, hydrolysis, and glycolysis are used to depolymerize PET to monomeric building blocks that can be used to resynthesize the virgin PET (Fig. 3). On the other hand, ammonolysis and aminolysis produce molecules that are unsuitable for PET synthesis. Products of ammonolysis and aminolysis can be upcycled to prepare other materials. Therefore, these upcycling processes are not discussed in this review.

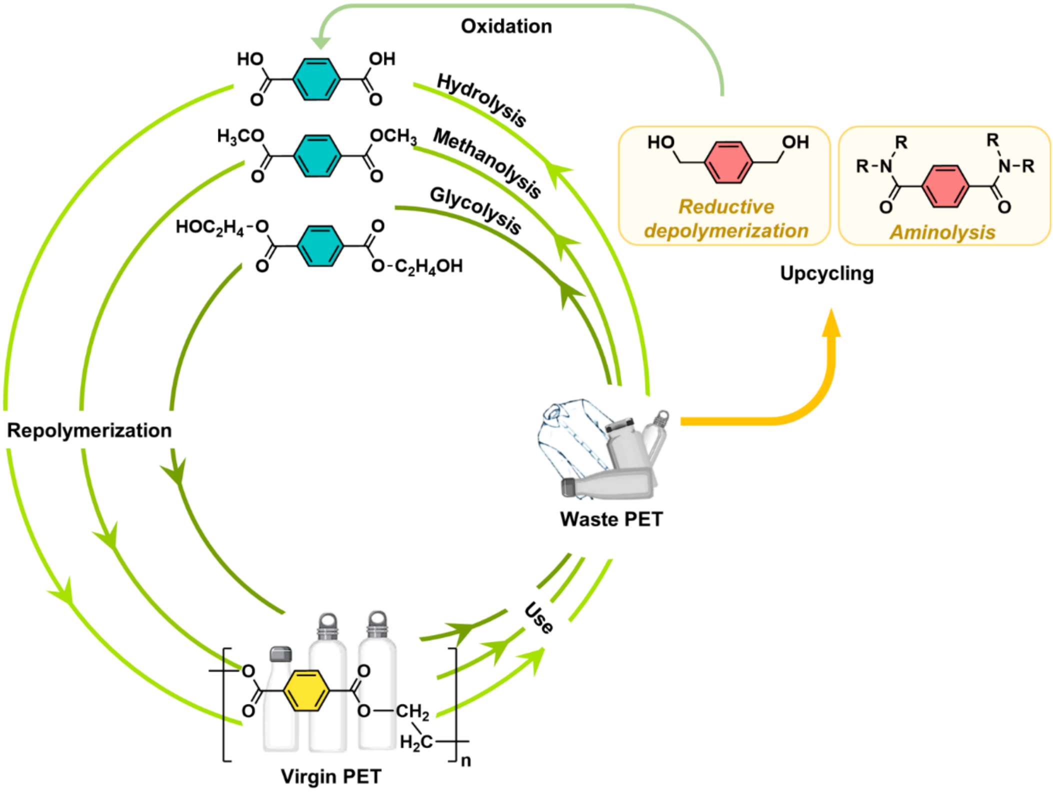


Fig. 3. Overview of PET depolymerization into monomers and other valuable building blocks.

2.1 Depolymerization of PET by hydrolysis.

Hydrolyzing the ester bonds in PET is a well-established and simple depolymerization process for the recovery of TA and EG with high yields (>80%). In general, hydrolysis can be performed under neutral [58, 59], acidic [60, 61], and alkaline conditions (Fig. 4a) [62-64]. The limited polymer-water contact combined with the semi-crystalline nature of PET requires high temperatures and pressures to fully hydrolyze ester bonds [65]. Neutral hydrolysis is an eco-friendly technique performed in water or steam at elevated temperatures and pressures (≥200 °C, 1–4 MPa, Fig. 4a) [66-68]. These processes require long reaction time to achieve complete depolymerization of PET. Although different catalysts have been investigated to accelerate the process, alkali-metal catalysts are the most commonly used catalysts [69, 70]. However, it was observed that temperature is the dominant factor affects the conversion and increasing the temperature above the melting point of PET accelerate the depolymerization rate significantly [71]. This problem of harsh reaction condition can be addressed by employing acidic or alkaline hydrolysis by activating the carbonyl group in PET in the presence of an acidic or basic catalyst.

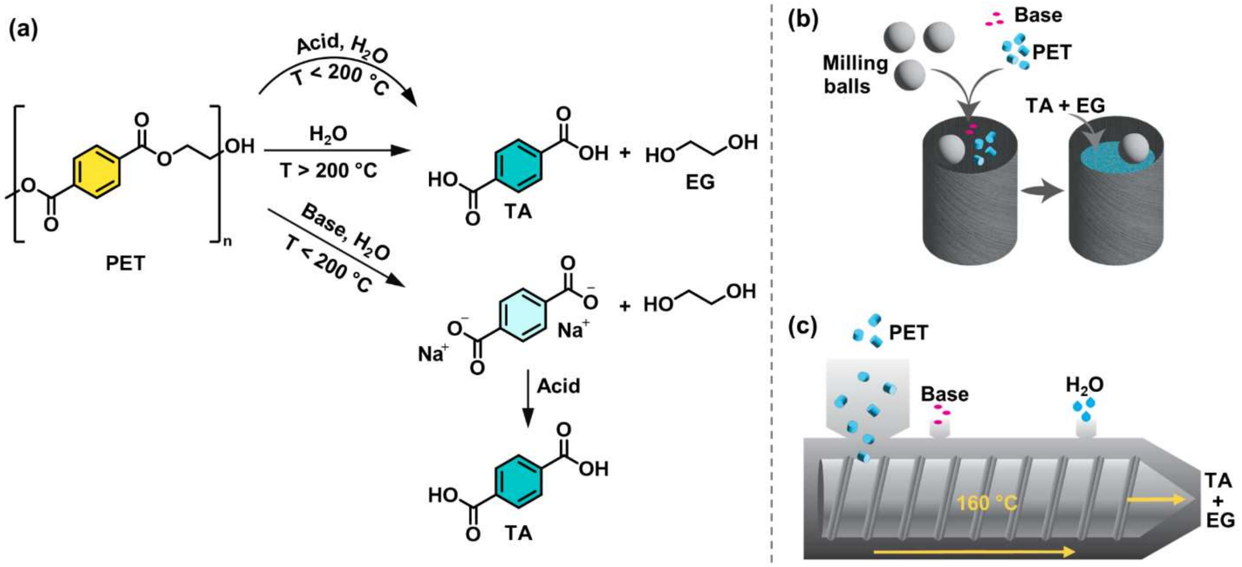


Fig. 4. (a) Hydrolysis of PET under acidic, neutral, or basic conditions. (b) Schematic representation showing stress-induced PET depolymerization by ball milling and (c) continuous depolymerization in the extruder. The figure is adapted from ref. [72].

2.1.1 Acid hydrolysis

Acid hydrolysis is mainly performed in highly concentrated acid solutions (e.g., H_2SO_4 and HNO_3) to prevent the need of elevated reaction temperatures and pressures (Fig. 4a). Examples of TA recovery from PET by low temperature (<100 °C) acid hydrolysis use concentrated H_2SO_4 or HNO_3 solutions [73, 74]. However, these technologies require more than 80 wt% of acid to complete the hydrolysis of PET. A high concentration of acid is highly corrosive and leads to the generation of a large quantity of salt containing wastewater. As a result, the purification of EG and the recycling of the acid pose a challenge [73], which increases the overall cost of the process.

In the past decade, only limited advancements have been made in acid hydrolysis of PET. In one of the studies, Yoshioka et al. demonstrated that reducing the acid content to below 50 wt% (10 M) requires a relatively higher temperature (150 °C) [60]. From a process and environmental standpoint, this method does not offer any significant advantages over previously reported acid hydrolysis methods. Additionally, a recent report from Yang et al. suggests that TA can also act as a catalyst in aqueous media for the hydrolysis of PET [75]. Although such processes require higher reaction temperatures and longer reaction times, acid removal steps is eliminated from the process and thereby reducing wastewater generation.

2.1.2 Alkaline hydrolysis

Similar to acids hydrolysis, strong bases like NaOH and KOH (4–20 wt%) were used for PET hydrolysis (Fig. 4a). However, the process feasibility, efficiency, and environmental friendliness remain a challenge [76]. Phase transfer catalysts, such as ammonium salts, have been used to promote water-to-polymer contact and increase the hydrolysis efficiency [77, 78]. However, intermediate terephthalate salts formed at the end of the process require additional process steps involving large quantities of acidic water to recover TA (Fig. 4a) [79]. To improve polymer-to-water contact during PET hydrolysis and to make the process economically viable, stress-induced depolymerization and microwave heating were investigated [80, 81]. It has been reported that applying mechanical stress to the reaction mixture during alkaline hydrolysis of PET can achieve process intensification [72, 80, 81]. In the first process, the ball milling of PET and

NaOH mixtures quantitatively depolymerized PET into TA over 95% selectivity (Fig. 4b). In the second case, a mixture of PET/NaOH/water (1:0.44:1.4 by mass) was passed through a twin-screw extruder at 160 °C. High mechanical stress in the presence of NaOH resulted in hydrolysis of PET within 1 minute and with ~97% disodium terephthalate yield (Fig. 4c) [72]. The process is reported to achieve a continuous throughput of 20 $kg{\cdot}h^{-1}$ [72]. Such processes address issues related to conventional alkaline hydrolysis of PET and opens the possibility to move towards a continuous process. It is worth mentioning here that an additional acid treatment step is still necessary to recover TA from disodium terephthalate [72].

2.2 Depolymerization of PET by glycolysis

Glycolysis is the oldest and most frequently used industrial depolymerization technique for PET [82]. During glycolysis, glycols diffuse through the polymer chains and swell the polymer matrix, followed by oligomerization via transesterification. Further glycolysis leads to the formation of monomeric polyhydric alcohols [83, 84]. Glycols, such as EG and propylene glycol (PG), can be used for glycolysis [76, 85-89]. However, the use of EG is well-established because it forms bis-hydroxyethyl terephthalate (BHET), which is the principal building block of PET. It also eliminates the first transesterification step required to form BHET from TA and EG [90, 91]. During PET glycolysis, catalysts are often used to facilitate depolymerization. Currently, researchers are developing innovative methods and optimizing existing processes. These include solvent-assisted glycolysis, microwave heating, and supercritical processing [92-94]. In recent reports, these methods have been combined with an active catalyst to achieve a faster reaction with improved selectivity for BHET, thus making the process more viable. Catalysts developed for PET glycolysis include homogeneous catalysts such as metal salts [95, 96], ionic liquids [89, 97-99], and heterogeneous catalysts like ZnO [100, 101].

2.2.1 Homogeneously catalyzed glycolysis

a. Metal-salt-catalyzed glycolysis: Acetates of Zn, Mn, Co, and Pd are commonly employed as homogeneous catalysts for PET glycolysis due to their high catalytic activity and cost-effectiveness. In this case, simultaneous activation of carbonyl groups in PET and EG takes place by the metal center and anion of the salt respectively (Fig. 5a). The activities of these metal acetates follows the order: Zn > Mn > Co > Pb. Despite the high glycolytic potential of zinc acetate, the poor miscibility of PET in EG slows down depolymerization kinetics. To overcome this limitation, a dissolution-depolymerization strategy was adopted utilizing co-solvents such as DMSO or xylene [95, 96]. The solubilization of PET facilitated the access to ester bonds and resulting in rapid depolymerization with BHET yield over 80% [95, 96]. Complete removal of these high-boiling-point solvents from the recovered BHET can be challenging and carries the risk of BHET contamination. Moreover, soluble heavy metal catalysts (such as Zn, Pd) require an additional separation process to prevent any potential environmental or health hazards [102]. In this regard, mild alkali salts were found to be quite effective in minimizing the negative health impacts of heavy metal salts without compromising their efficacy [103, 104]. Despite of high BHET yield and low cost, additional purification steps in downstream processing remain a major drawback of homogeneously catalyzed processes.

b. Ionic liquid (IL)-catalyzed glycolysis: ILs are salts with low melting temperatures (≤100 °C) that have gained attention due to their excellent catalytic performance. ILs are promising candidates for the glycolysis because of their non-volatility, good solvation properties for both inorganic and organic compounds and ease of tuning their properties [105, 106]. In the case of ILs-catalyzed glycolysis, the cationic component of the IL activates the C=O group of the ester, and the hydroxyl group of EG attacks the ester group of PET (Fig. 5a) [107]. ILs with stronger basicity are known to exhibit higher catalytic activity compared to ILs with weaker basicity [108]. For example, 1-butyl-3-methylimidazolium hydroxide ([Bmim][OH]) displayed higher activity than [Bmim][HCO_3], [Bmim][Cl], and [Bmim][Br]. During the PET glycolysis, 5 wt% [Bmim][OH] at 190 °C completely depolymerize PET with ~71% BHET yield after 2 h. [108] Considering the high catalytic activity of basic ILs, Lewis base ILs such as 1-butyl-3-methylimidazolium acetate ([Bmim][Ac]) were also investigated for the glycolysis of PET [89]. Although complete depolymerization of PET was achieved within three hours, selectivity for BHET was only 58%, which was significantly lower than the BHET yield obtained using the basic IL [Bmim][OH] [89]. Overall, the BHET selectivity of IL-catalyzed glycolysis was slightly lower than that of the other processes described in this review (<80%). Despite their toxicity, imidazolium-based ILs have been extensively studied for glycolysis. This is mainly because of the ease of tuning the physical and chemical properties of ILs, which enables the design of a diverse library of ILs with different catalytic activities. To address the issue of toxicity associated with imidazolium-based ILs, Gonçalves et al. investigated the potential of choline-based ILs that are less toxic alternatives [109]. Accordingly, choline formate ([Ch][For]) and choline acetate ([Ch][OAc]) were used for PET glycolysis, and efficient depolymerization of PET with over 80% BHET yield was achieved [110].

2.2.2 Heterogeneously catalyzed glycolysis

Heterogeneous catalysts have been widely used to address the issue of catalyst separation from monomers and solvents (Fig. 5a). High surface area and porous structure of these catalysts offer high activity for depolymerization. Moreover, heterogeneous catalysts are also highly stable in nature and easy to recover, which makes them easy to recycle. One of the earliest reports in 2008 described the use of β- and γ-zeolites as transesterification catalysts for PET depolymerization. Complete depolymerization was observed with a ~65% BHET yield after eight hours of reaction at 196 °C [101]. Metal oxide-doped silica nanoparticles (NPs) were also found to enhance PET glycolysis [84]. In this case, ZnO-doped SiO_2 NPs were more effective than CeO_2-doped SiO_2 NPs. Imran et al. reported that manganese oxide (Mn_3O_4)-doped SiO_2 NPs were active to catalyze the glycolysis of PET, and >90% BHET yield was obtained after 80 min of reaction [111]. In a separate study, Park et al. showed that nearly 96% BHET yield was obtained using Mn_3O_4-doped graphene oxide (GO-Mn_3O_4) as the catalyst [112]. When unsupported Mn_3O_4 NPs was used for glycolysis, only 82% BHET yield was obtained under similar reaction conditions. This confirms the positive effect of a high active surface area and the stabilizing action of the support [112].

Another promising study explored the transesterification ability of aluminum-magnesium-layered double hydroxide, known as Perkalite F100 [113]. Interestingly, ~82% BHET yield was obtained at 0.1 wt% catalyst loading, highlighting the superior transesterification ability of Perkalite F100 compared to zinc acetate and sodium bicarbonate [113]. Magnetic particle-doped catalysts have also been explored to enhance catalyst recovery and facilitate recycling. For example, substituting ZnO with the spinel of Fe_3O_4 led to an improvement in the glycolysis of PET and facilitated the retrieval of the catalyst via magnetic separation [114]. In this case, complete depolymerization with ~80% BHET selectivity was observed within two hour [114].

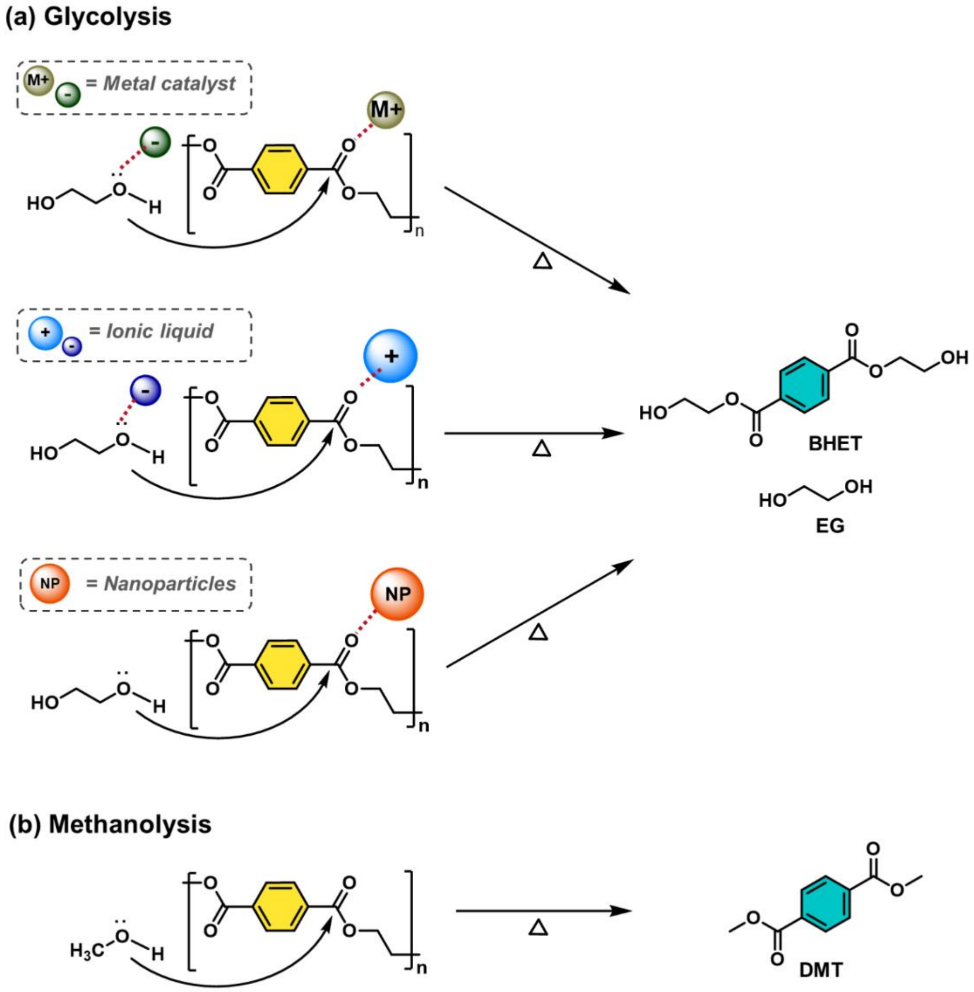


Fig. 5. (a) General scheme illustrating the glycolysis PET via activation of the carbonyl group by metal cation of soluble metal catalysts, the cations of ionic liquid, and nanoparticles/heterogeneous catalysts. (b) Reaction scheme showing depolymerization of PET to dimethyl terephthalate (DMT) through methanolysis.

2.3 Depolymerization of PET by methanolysis

Methanolysis is another method to depolymerize PET, and its mechanism is similar to that of glycolysis (Fig. 5b). The lone pair of electrons on the oxygen atom in methanol attacks the ester bond of PET and leading to its cleavage, which subsequently depolymerizes PET to form DMT and

EG. DMT is not the direct monomer of PET, but it can be purified relatively easily and can be used as a feedstock for PET production [91]. In the last half of the 20th century, numerous patents related to methanolysis were filed [115-117]. Usually, high temperatures (180–280 °C) and pressures (20–40 bar) was used for successful methanolysis [79]. This was also the case for the Eastman-Kodak process, in which pure DMT was produced [118]. More recently, a few researchers have renewed their focus on methanolysis and developed processes that operate under milder conditions. For example, methanolysis developed by Loop Industries can be carried at ambient temperatures using a basic or alkali metal catalysts [119]. Additionally, Pham et al. have developed an ambient temperature methanolysis process by combining K_2CO_3 as the catalyst and an aprotic polar solvent to achieve more than 90% DMT yield after 24 h [120].

2.4 Depolymerization of PET by biocatalysts

Progress in biotechnology for the selective hydrolysis of ester bonds has opened up the possibility of depolymerizing PET using biocatalysts. However, the semicrystalline nature of PET hinders enzymatic depolymerization and leading to slow reaction with moderate yields. In an effort to enhance bio-catalyzed depolymerizations, Kim et al. combined the glycolysis of PET with enzymatic hydrolysis in the presence of compatible biocatalysts [121]. In this case, betaine was used as the catalyst for the glycolysis in the first step, followed by enzymatic hydrolysis of PET into TA and EG. For enzymatic hydrolysis, IsPETase and IsMHETase were used (Fig. 6). Leaf-branch compost cutinase (LCC) has also been identified as an enzyme for PET depolymerization. However, in this case heating was required to slightly soften PET to achieve a noticeable reaction rate. Reaction temperatures above 65 °C unfortunately denature the enzyme. To overcome this limitation, LCC was modified to improve its thermal stability and it led to more than 90% depolymerization after 10 h of incubation at 72 °C [122].

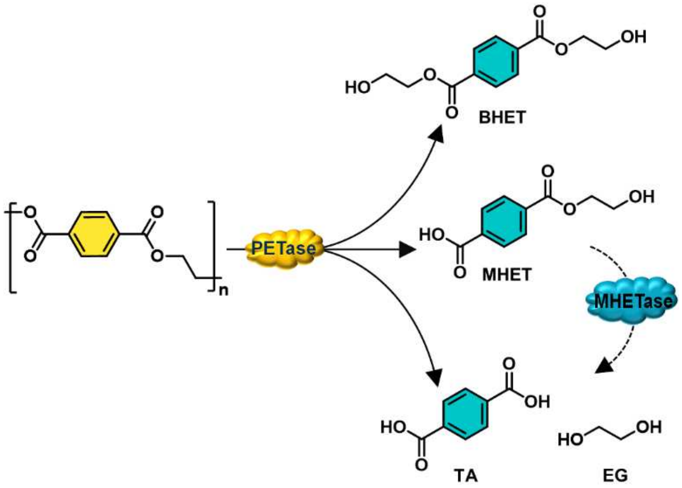


Fig. 6. Schematic overview of PETase and MHETase as biocatalysts for depolymerizing PET into BHET, MHET, TPA, and EG. The figure is adapted from ref. [123].

2.5 Depolymerization of PET by reduction

Reductive depolymerization of PET is a new and alternative method for converting PET-based plastics into valuable building blocks or recovering TA (Fig. 7a). To date, only a limited number of studies have been reported in the literature, demonstrating the use of homogeneous and heterogeneous catalysts for the reductive depolymerization of PET. Borane catalysts [124] as well as pincer complexes of ruthenium (Ru) [125, 126], iridium(Ir) [127], and iron (Fe) have been employed as catalysts in the presence of molecular H_2 or silanes as reducing agents to depolymerize PET (Fig. 7b). Feghali et al. demonstrated that commercially available Tris(pentafluorophenyl)borane ($B(C_6F_5)_3$) and [Ph_3C^+, $B(C_6F_5)_4^-$] can act as catalysts for the hydrosilylation of PET to form disilyl ethers (path 1, Fig. 7a) [124]. The use of air-stable polymethylhydrosiloxane and tetramethyldisiloxane as reductants simplified the process and complete depolymerization was achieved with high yield (91%) [124]. Later, disilyl ether obtained as the depolymerization product can be hydrolyzed to 1,4-phenylenedimethanol (PD). In contrast, Krall et al. used ruthenium pincer complexes at 120 °C and elevated pressures (40 bars) for the depolymerization of PET to produce PD and EG (Path 2, Fig. 7a) [126]. PD obtained after both depolymerization pathways (path 1, 2, Fig. 7a) can be directly used as a building block for the production of various other polymers [128, 129] or oxidized to TA for the synthesis of PET [130, 131]. Both Pathways suffer from long reaction times, multistep processes, and the loss of expensive catalysts, making these reductive pathways unattractive for scale-up.

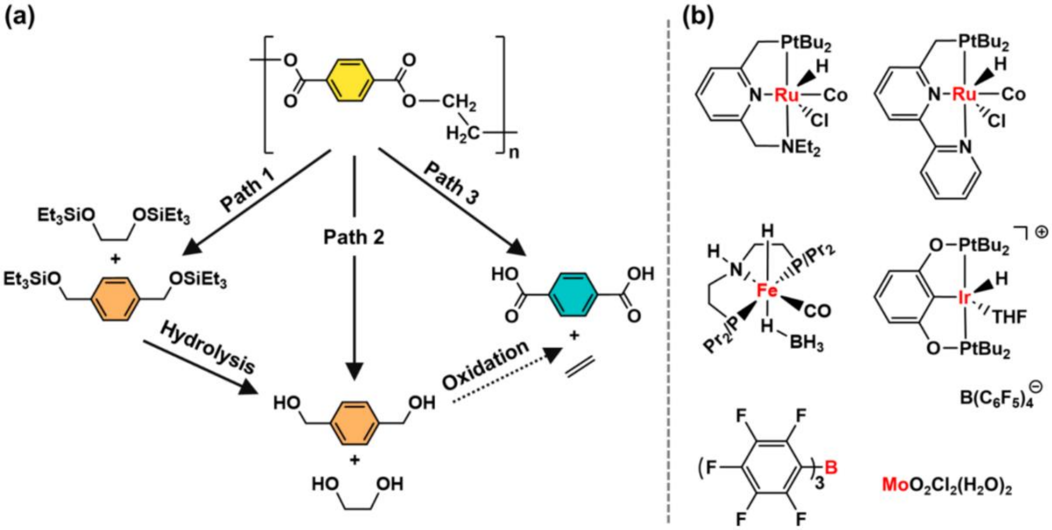


Fig. 7. (a) Reductive depolymerization pathways and intermediate products formed during PET depolymerization. (b) Typical catalysts used for reductive depolymerization of OPs.

Alternatively, Marks et al. reported a direct hydrogenolysis pathway of PET to TA using a carbon-supported molybdenum-dioxo catalyst (MoO_2/C) (path 3, Fig. 7a) [132]. The high thermal stability of heterogeneous catalysts allows the depolymerization of PET at melting temperatures without solvents. Depolymerization proceeded in the presence of H_2 at 1 atm, and after 24 h of reaction 90% yield of TA was obtained. The heterogeneous nature and stability of the active sites in MoO_2/C facilitated recycling without decreasing the TA yield. The abundance of MoO_2, high stability, absence of a solvent and high yield of the process make it an attractive option for scale-up. However, the long reaction time and high depolymerization temperature are two major drawbacks of this process [132]. Developing more efficient non-noble metal-based heterogeneous catalysts having ability to decrease the reaction time and temperature can make the reductive depolymerization process more attractive at an industrial scale.

2.6 Comparison of different PET depolymerization processes

Among the various OPs depolymerization processes, considerable progress has been made in case of PET depolymerization, and several technologies are currently being utilized in the industry with a TRL 7–9. In this report, the TRL classification suggestions of the European Association of Research and Technology Organizations (EARTO) were used to compare the different PET depolymerization processes reported in the literature [133]. Although different routes are available for PET depolymerization, the ones that allow a smooth PET synthesis using established routes are relatively more developed and therefore TA and BHET routes are mainly used for commercial PET production. For example, most mature PET depolymerization processes are hydrolysis [51, 134-136] and glycolysis [44, 46, 137] that produce TA and BHET respectively. The recovered TA and BHET can be used directly for PET synthesis. As a result, the TRL of hydrolysis and glycolysis are in the range of 7–9. Comparison of different PET depolymerization methods was done by showing the lower and upper limit of the required reaction conditions (Fig. 8a) and by calculating the normalized output of each process using Eq. 1. Output minima and maxima were determined and plotted against the temperature of the particular process (Fig. 8b). As can be seen from the spider diagrams (Fig. 8a), glycolysis requires milder temperatures and lower catalyst loadings compared to hydrolysis to reach similar conversions and selectivity. Additionally, hydrolysis of PET tends to produce more hazardous wastewater than other processes described. Therefore, glycolysis of PET is the preferred technology for scale-up when comparing to hydrolysis. Reducing the wastewater generation or reusing it would make hydrolysis more economically appealing to industries [134].

$$\text{Normalized output/h} = (\text{polymer loading }\% \times \text{conversion} \times \text{selectivity})/\text{time (h)} \qquad (1)$$

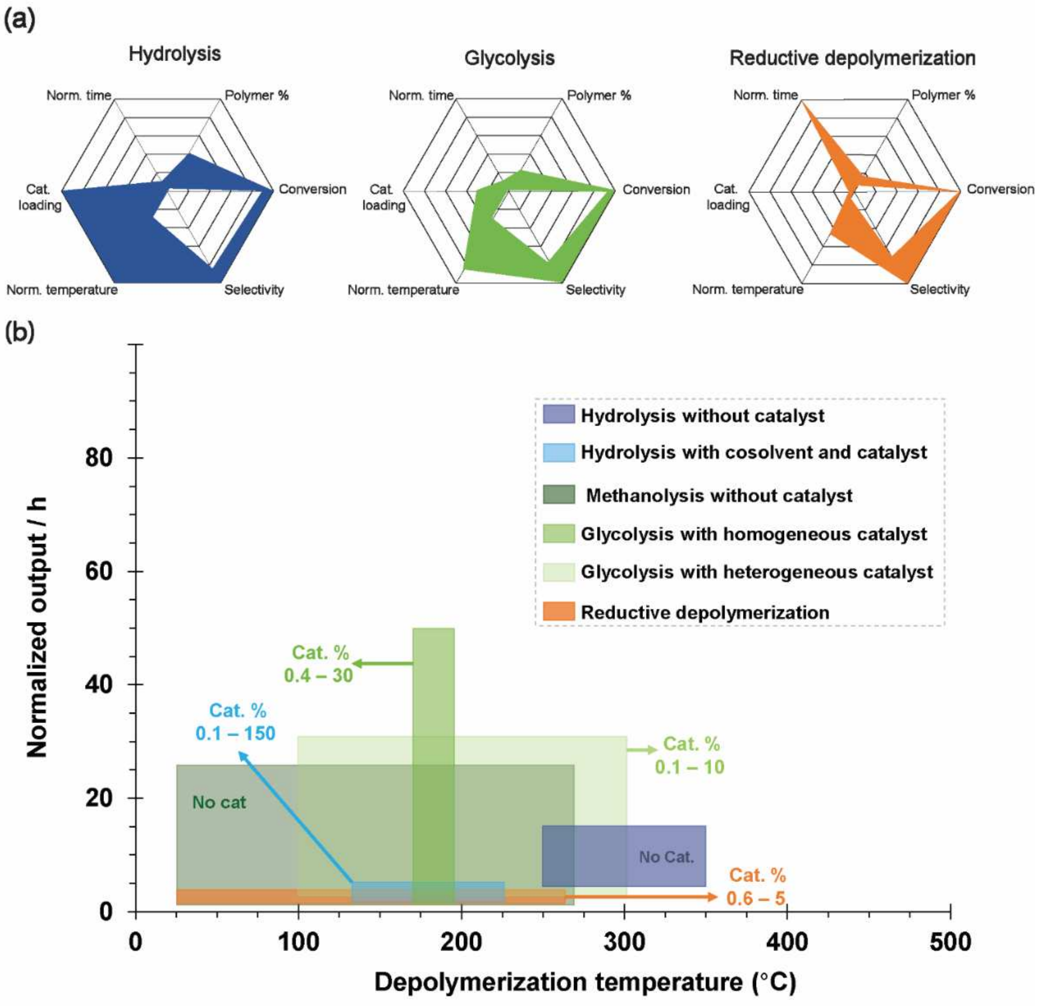


Fig. 8. (a) Spider diagrams illustrates the impact of depolymerization parameters on conversion and monomer selectivity in a specific process. The shaded area represents the upper and lower limits for time, catalyst and polymer loading, temperature, conversion and selectivity towards the monomer for each processes. (b) Comparison of different PET depolymerization methods based on the calculated normalized output. Reports with high depolymerization efficiencies and monomer yields (~90%) were considered for this comparison. The catalyst loading % was calculated based on the weight to PET (cat.) used during the depolymerization and is given as inserts at their respective locations. All data is listed in Table 1 and the normalized output was calculated using Eq. 1.

Although less popular, methanolysis is also used for large-scale depolymerization of PET (TRL 7–9). However, this method is limited to companies that operate DMT-based PET production plants [45, 47]. Compared to all other processes, reductive depolymerization of PET is relatively new and is mainly at the lab-scale development stage (TRL 3–4). The output of reductive depolymerization is currently extremely low and relatively long times are required to obtain reasonable conversions (Fig. 8). Additionally, an important factor here is the recovery and reuse of the catalyst. The use of noble-metal-based or homogeneous catalysts makes the process unattractive for industrial use. Heterogeneous catalysts, for example, will facilitate the separation and hence ease the downstream processing of the process making the process already more interesting for industrial application. Therefore, efficient and cost-effective catalysts that can be easily recycled and operated under low H_2 pressure must be developed for the future implementation on pilot and industrial scales.

Table 1. Different process conditions of PET depolymerization and output of the reaction.

No	Solvent	Polymer loading (%)	Catalyst	Cat. %	Time (h)	Temp. (°C)	Conv. (%)	Yield (%)	Ref.
Hydrolysis of PET									
1	H_2O	9	$[(CH_3)_3N(C_{16}H_{33})]_3PW_{12}O_{40}$	7	2	145	100	93	[78]
2	H_2O	16	H_2SO_4	600	5	135	90	97	[138]
3	H_2O	11	TA (R)	0.1	3	220	100	96	[75]
4	H_2O /EG	6	$NaHCO_3$	90	1.16	180	98	94	[64]
5	H_2O /EG	10	NaOH	100	0.75	100	-	93	[139]
6	H_2O	41	NaOH	44		160	100	98	[72]
7	DEG	10.7	NaOH/GDS	55	0.42	160	100	90	[140]
8	THF/EG	3	KOH	150	1	100	100	98	[141]
9	MeOH	5	KOH		0.016	120^a	100	100	[142]
10	H_2O	10	Sea water	0.35	2	205	87	96	[59]
11	H_2O	11	None	-	6	250	100	99	[58]
13	H_2O	5	None	-	1	250	92	86	[143]
14	H_2O	20	None	-	1	350	91	85	[143]
15	H_2O	10	None	0	0.5	300	-	90	[144]
Glycolysis of PET									
16	EG	9	Cyanamide	5	2.5	190	100	95	[145]
17	EG	19	Orange peel ash	10	1.5	190	100	79	[146]
18	EG	20	TBD : MSA	30	2	180	100	91	[147]
19	EG	20	[Ch][OAc]	5	4	180	100	85.2	[110]
20	EG	20	1,3- DMU/$Zn(OAc)_2$	5	0.3	190	100	82	[148]
21	EG	20	Urea/$ZnCl_2$	10	0.5	170	100	83	[149]
22	EG	16	Acetamide/$ZnCl_2$	0.4	1.6	195	100	85.2	[150]
23	EG	2	[Bmim]Cl/$ZnCl_2$	1.25	2	190	100	84.9	[99]
24	EG	8	$ZnMn_2O_4$	1		260	100	92.2	[151]
25	EG	8	Mn_3O_4/SNPs	1	1.3	300	-	90	[111]
26	EG	21	GO–Mn_3O_4	1	1.3	300	-	96	[112]
27	EG	5	MnO_2/GO	0.01	0.17	200	100	100	[152]
28	EG	21	γ-Fe_2O_3	5	1	300	95	90	[153]
29	EG	8	Fe_3O_4/MWCNT	5	2	190	100	100	[154]
30	EG	7	γ-Fe_2O_3/N_2-doped graphene	10	3	195	100	100	[155]
31	EG	3	Fe_3O_4/h-BNNS	0.6	5	200	100	100	[156]
32	EG	22	Fe_2O_3 on MoS_2	10	3	225	97	90	[157]
33	EG	3.5	Co NPs	1.5	3	180	96	77	[158]
34	EG	15	Pd/hBN	10	0.5	100	98	92	[159]
Methanolysis									
35	MeOH	27	K_2CO_3	2	14	25	100	93	[120]
36	MeOH	14	None	0	0.5	270	100	95	[160]

Reductive depolymerization of PET									
35	1,4 dioxane	17.5	[Ru(triphos)tmm]/HNTf$_2$	1.2	16	140	99	87	[161]
36	Anisole/THF	7	Milstein catalyst	5	48	160	99	-	[126]
37	Benzene	17	LaN*3/HBpin	0.6	24	100	-	71	[162]
38	DCM/TMDS	7	$B(C_6F_5)_3/Et_2SiH$	5	3	25	100	77	[124]

[a] microwave heating, MeOH = Methanol; TA = therephtalic acid; GDS = glycidoxy propyl trimethoxy silane/diethanol amine; TBD = triazabicyclodecene ; MSA = methanesulfonic acid; Ch = choline; 1,3-DMU = 1,3-dimethylurea; bmim = 1-methyl-3-butylimidazolium; MWCNT = multiwalled carbon nanotubes; h-BNNS = hexagonal-boron nitride nanosheets; hBN = hexagonal boron nitride; triphos = (1,1,1-tri(diphenylphosphinomethyl)ethane; tmm = trimethylenemethane; HNTf$_2$ = bis(trifluoromethanesulfonyl)imide; HBpin = pinacolborane; (R) = recycled catalyst.

3. Depolymerization of polycarbonate

Commercial poly(bisphenol A carbonate) is mainly referred as polycarbonate (PC). The building blocks of PC provide excellent transparency, thermal stability, impact resistance, and mechanical properties. While most engineering polymers become brittle and stiff below their T_g, PC retains its ductile behavior [163]. These properties make PC a highly desirable polymer for application in electronic equipment, engineering plastics, automobiles, optical devices, and sports equipment (Fig. 9a). However, BPA used in most PC synthesis is an endocrine-disrupting chemical [164, 165]. In attempts to reduce or replace BPA, several BPA analogues (BPB, BPS, and BPF, Fig. 9b) are being used and marketed as BPA-free plastics. Unfortunately, most of these analogues have comparable toxicity to BPA [166, 167] and triggered the search for safer alternatives. In one such development, bio-sourced isosorbide was investigated for polycarbonate synthesis (Fig. 9b) and these novel isosorbide based PCs display excellent thermal, mechanical, and optical properties This indicates their potential as alternatives to the existing BPA based PCs. With the commercial availability of polymerization-grade bio-sourced isosorbide

[168], bio-based polycarbonates like DURABIO® from Mitsubishi [169] and PLANEXT® from Teijin [170] are now available for consumers. Despite possessing all the desired properties, bio-based polycarbonates require time to replace conventional PCs. With growing awareness of plastic circularity and attempts to minimize the risk of BPA exposure, PC recycling is gaining attention.

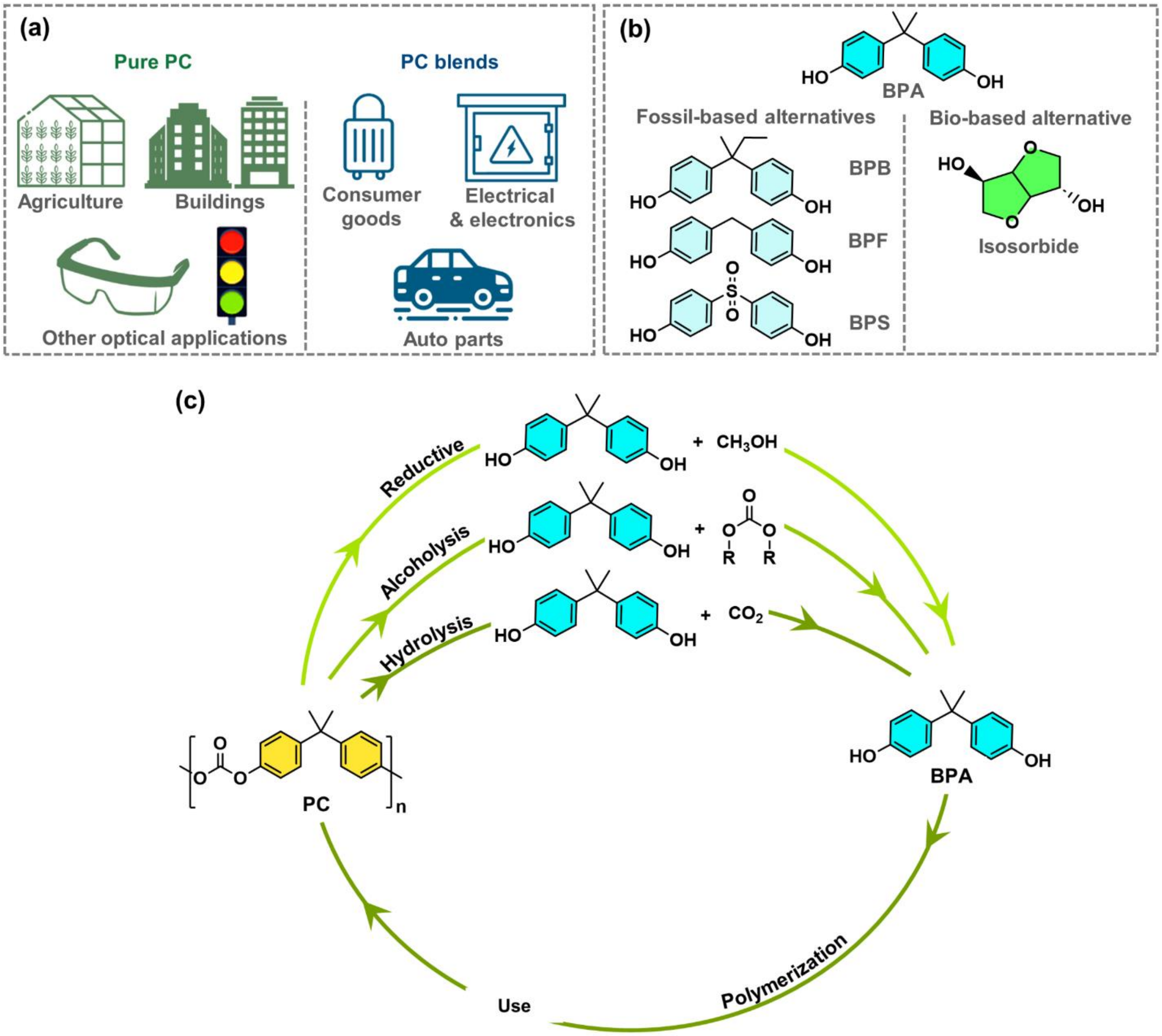


Fig. 9. (a) Examples of PC applications in pure form and as blends. Pure PC is preferred in some of the applications because of its transparency and high impact resistance. (b) BPA and its fossil and bio-based alternatives, such as bio-isosorbide, for PC preparation. (c) Different chemical recycling pathways reported for the recovery of BPA from PC wastes.

Except few applications, PC is mainly blended with other polymers to produce durable components that are exposed to various weathering conditions. This leads to a significant variation in the different PC waste feedstock (Fig. 9a). Additionally, the potential release of BPA during thermomechanical recycling increases the risk of environmental pollution and human exposure. These factors make the mechanical recycling of PC very challenging. Consequently, most PC-based waste ends up in landfills, and the slow release of BPA resulting from the degradation in landfills pollutes surrounding land and water. The risk of BPA pollution and lack of mechanical recycling of PC require urgent attention. Therefore, our discussion primarily focuses on the depolymerization processes of PC, which offer the possibility of recovering BPA (Fig. 9c).

3.1 Depolymerization of PC by hydrolysis

Nucleophilic attack by water molecules on ester bonds offers the possibility to depolymerize PC to BPA (Fig. 10a). Compared to semicrystalline PET, the amorphous nature of PC facilitates hydrolysis under milder conditions. However, the rigidity and hydrophobic nature of the PC molecules severely limit the diffusion of the solvent into the polymer, thereby restricting the contact between the polymer and water. In an attempt to overcome these constraints, early studies were carried out at high temperatures, typically above the melting temperature of PC (T_m=243 °C), to facilitate the contact between water and PC to enhance the hydrolytic cleavage of carbonate linkages [171-173]. Unfortunately, harsh reaction conditions often led to partial degradation of BPA and lowering the BPA selectivity (Fig. 10a). The combination of harsh reaction conditions and low BPA selectivity renders these processes very unattractive. To achieve hydrolysis under milder conditions and reduce the risk of BPA degradation, different catalysts were investigated [174-179]. Additionally, a lower reaction temperature with prolonged reaction time was also investigated to improve BPA yield [171]. Unfortunately, these strategies led to lower BPA yields.

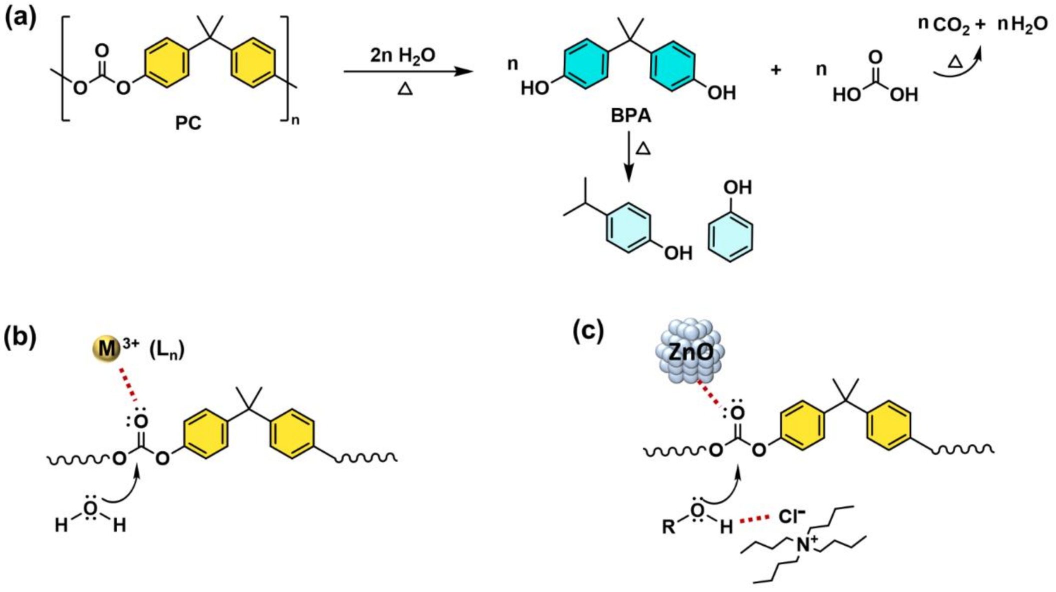


Fig. 10. (a) Scheme showing the hydrolysis of PC to BPA and the degradation of BPA to phenol and isopropylene phenol (IPP) [180, 181]. (b) Activation of carbonyl groups in PC by metal triflates for hydrolysis. M = lanthanum, scandium, and ytterbium, L_n = $(O_3SCF_3)_3$ [178, 182]. (c) Synergy between TBACl and ZnO as a bifunctional acid-base catalyst for the activation of the carbonate linkage in PC [183]. Copyright 2017. Adapted with permission from Elsevier Science Ltd.

3.1.1 Homogeneously catalyzed hydrolysis

a. Alkali catalyzed hydrolysis: In an effort to improve the BPA yield, Lewis bases were used during the hydrolysis of PC. Complete depolymerization of PC was possible within 3 h at a low temperature (130 °C) using 1000 mM aqueous solution of Na_2CO_3 [174]. Unfortunately, it was difficult to prevent BPA degradation even at low reaction temperatures, particularly under high Na_2CO_3 loading. As a result, only 56% BPA yield was obtained [174]. Using a stronger base like NaOH (0.6 $mol·kg^{-1}$) under a semicontinuous reaction condition (160 °C, 2 h), a significant improvement in BPA yield was obtained (94%) [175]. High water (3000 g/g of PC) and NaOH consumption were the major drawbacks of this process (Table 2) [175].

The hydrolytic depolymerization of PC under homogeneous conditions was also investigated by adding a solvent to the hydrolysis reaction mixture. The dissolution of PC in the co-solvent improved the accessibility of the carbonate linkages for hydrolysis. Consequently, PC was hydrolyzed under moderate conditions, resulting in a high BPA selectivity. For example, Liu et al. observed that the use of 1,4-dioxane accelerated the NaOH-catalyzed hydrolysis of PC at 100 °C and achieved nearly 94% BPA yield after 8 h of reaction [184]. As demonstrated by the authors, such process can operate with high polymer loading (up to 20 wt%) (Table 2, Entry 3). Similarly, a glycerin-water mixture was used as the solvent for NaOH-catalyzed hydrolysis of PC at 50% loading and 150 °C microwave heating (Table 2) [185]. Heat transfer limitation was mitigated by microwave heating and a high depolymerization rate was achieved. At high glycerin composition (80%) rapid depolymerization was observed and complete depolymerization of PC was achieved within two minutes [185]. Lower glycerin concentration required a longer reaction time (0.4 h) for PC depolymerization. Interestingly, BPA selectivity in both reaction conditions remained high (~100%) [185].

b. IL catalyzed hydrolysis: In line with the homogeneous hydrolysis route, Song et al. investigated the use of ILs as solvents for PC hydrolysis. In addition to promoting polymer dissolution, ILs also act as catalysts. As observed in the case of PET, basicity of ILs has a strong influence on the hydrolysis of PC [176, 177]. For example, hydrolysis in [Bmim][Ac], which is more basic than [Bmim][Cl], requires a lower temperature (140 °C) than [Bmim][Cl] (165 °C) (Table 2). In both ILs, complete depolymerization was achieved after 3 h of reaction with high BPA yield (≥95%) [176, 177]. It was also observed that a higher water content in the reaction medium than the optimized nH_2O/nPC ratio (5–10%) slow down the depolymerization significantly. This was attributed to the poor swelling of PC and dilution of the catalytic activity of ILs in the presence of water above the optimized limit of 5–10%. Similarly, when a stronger Lewis base IL, such as 1,8-diazabicyclo (5.4.0)undec-7-ene (DBU)-based ILs was used, complete PC hydrolysis was possible within 3 h at a very high polymer loading (75 wt%) [186]. Very high BPA selectivity (98.5%) was also obtained after the reaction [186]. Low reaction temperature in case of IL-catalyzed hydrolysis prevents any unwanted side reaction and results high BPA selectivity. The DBU-IL studied for the hydrolysis was reused more than five times without sacrificing their activity and selectivity. Despite the distinct advantages of IL-catalyzed hydrolysis in terms of PC loading (35–75%), BPA yield, and reusability, the use of ILs for large-scale PC depolymerization is still limited due to their high cost and potential toxicity [187, 188].

c. Metal-catalyzed hydrolysis: The hydrolysis of PC through the activation of carbonate linkages with the assistance of a rare-earth metal center was investigated (Fig. 10b) [178]. Metal triflates ($M(O_3SCF_3)_3$) of Lanthanum (La), Scandium (Sc), and Ytterbium (Yb) were used to catalyze the hydrolysis of PC in the presence of THF as a co-solvent. Metal triflates enhanced the PC hydrolysis rate by activating the carbonyl oxygen in PC through the metal center (M^{3+}) and making it susceptible to nucleophilic attack by water molecule (Fig. 10b) [178]. Although all the investigated metal triflates were effective for the hydrolysis of PC (Table 2), complete depolymerization with 97% BPA selectivity was observed only in the case of $La(O_3SCF_3)_3$. Despite of selective depoymerization ability, this process require high catalyst loading, long reaction time (6 h) and no information is available on the reusability of metal triflates, which make the process unsuitable for scale-up (Table 2) [178].

3.1.2 Heterogeneously catalyzed hydrolysis

To simplify the BPA and catalyst recovery, researchers have investigated the use of heterogeneous catalysts for PC hydrolysis. Many catalysts were reported in the literature including Al_2O_3, MnO_2, monoclinic ZrO_2, CeO_2 NPs, CaO-stabilized ZrO_2, and ZrO_2 nanocrystal. Nonetheless, CeO_2 NPs were found to be efficient even with a very low loading (0.03%, Table 2) [179]. The CeO_2 NPs completely hydrolyzed PC with ~90% BPA selectivity. The remaining depolymerized fraction consisted of phenol and IPP formed due to the decomposition of BPA at high reaction temperature (200 °C). Unfortunately, no further investigations have been conducted to improve the BPA selectivity.

Emami et al. used a magnetic Lewis acid catalyst based on nano-silica for the hydrolysis of PC in a mixture of diethylene glycol and water. Diethylene glycol in the reaction mixture enabled the hydrolysis at high PC loading (~37%) and achieved complete depolymerization with 100% BPA selectivity after 1.5 h [189]. Moderate hydrolysis temperature (160 °C) and short reaction time (1.5 h) prevented the degradation of BPA and achieved high BPA selectivity. Moreover, the magnetic nature of the catalyst facilitated its recovery and reuse [189]. In another example, naturally occurring magnesium-rich chlorite clay (clinochlore) was used to catalyze the hydrolysis of PC. THF was used to homogenize the reaction mixture and to promote contact between the heterogeneous catalyst and PC molecules. Although complete hydrolysis of PC with 98% BPA selectivity can be achieved, the high clinochlore loading (70%) coupled with the high reaction temperature (200 °C) are the major drawbacks [190]. Since clinochlore is a naturally sourced material, batch-to-batch variation can also influence the catalytic performance and inhibit their acceptance for large-scale processes [190]. On the other hand, Iannone et al. used the combination of tetrabutylammonium chloride (TBACl) and ZnO NPs for the hydrolysis of PC in the presence of THF [183]. In this catalytic system, ZnO NPs enhanced the electrophilicity of the carbonyl in PC, and TBACl anions acted as H-bonding acceptors as shown in Fig. 10c. The synergistic effect of ZnO NPs and TBACl afforded efficient hydrolysis of PC with 99% BPA yield even at 100 °C. Hybrid catalysts also displayed excellent reusability. However, long reaction time (7 h) is the major drawback of the process and may hinder its upscaling [183].

3.2 Depolymerization of PC by alcoholysis

Alcoholysis is an alternative method for depolymerizing PC to BPA via a multistep process: (i) random transesterification of PC chains to oligomers mediated by alcohol, (ii) subsequent transesterification to form soluble oligomers, and (iii) homogeneous transesterification of low molecular weight products to yield BPA and corresponding carbonates (Fig. 11). Unlike hydrolysis, CO_2 is not released during alcoholysis. Instead, the carbonyl group of PC forms the corresponding carbonate. Alcoholysis of PC requires harsh reaction conditions ($T>200$ °C) due to poor solubility of PC in alcohols and weak nucleophilicity of alcohols. Increasing the molecular weight of the alcohol can also increase the steric hindrance and requires more energy for the transesterification of PC. Therefore, methanol that offers smallest molecular structure and higher nucleophilicity is preferred compared to other alcohols and processes using methanol for depolymerization of PC referred as methanolysis.

In one of the early reports, Kim et al. carried out methanolysis of PC at temperature between 160 and 220 °C. Complete depolymerization with ~90% BPA yield was obtained after 15 minutes of reaction at 220 °C [191]. Increasing the methanol content in the reaction mixture had a positive impact on the BPA yield. However, elevated reaction temperatures (220 °C) led to the degradation of BPA and ultimately decreasing the BPA selectivity [191]. To improve BPA selectivity, Ikenaga et al. used microwave heating (190 °C) [192]. Although a marginal increase in BPA selectivity (94%) was observed at this temperature, long reaction time (3 h) was required to depolymerize PC (Table 2) [192]. Moreover, these processes operate at a very low polymer loading (6–10%), which negatively affect the BPA output of the reaction [191, 192]. In an attempt to depolymerize PC under milder conditions, improve the BPA selectivity and output, catalyzed methanolysis in the presence of co-solvents were investigated.

3.2.1 Homogeneously catalyzed alcoholysis

a. Alkali metal catalyzed alcoholysis: The inclusion of an alkali hydroxide in the reaction mixture found to expedite the PC methanolysis by simultaneously activating the carbonyl function of PC and methanol thus promoting the transesterification (Fig. 11a). To improve BPA selectivity, Arias et al. used microwave heating for alkali catalyzed (KOH) methanolysis (120 °C). KOH in combination with uniform heating achieved successfully depolymerized PC within 2 minutes with 98% BPA yield (Table 2) [193]. The process also displayed similar effectiveness for the depolymerization of PC-PET blend. Although the process has advantages in terms of BPA selectivity and short reaction time, low polymer loading (6 wt%) is a major drawback [193]. If the polymer loading of this ultrafast depolymerization process can be improved further, it will be attractive for large-scale adaptation [193]. In another study, He et al. used toluene as a co-solvent during NaOH-catalyzed methanolysis of PC [194]. Presence of co-solvent allowed to operate the methanolysis under high polymer loading (38 wt%) and moderate temperature (60 °C). The homogeneous nature of the reaction mixture obtained complete depolymerization within 15 minutes with nearly 95% BPA yield (Table 2) [194]. These observations are quite promising and make the process industrially attractive.

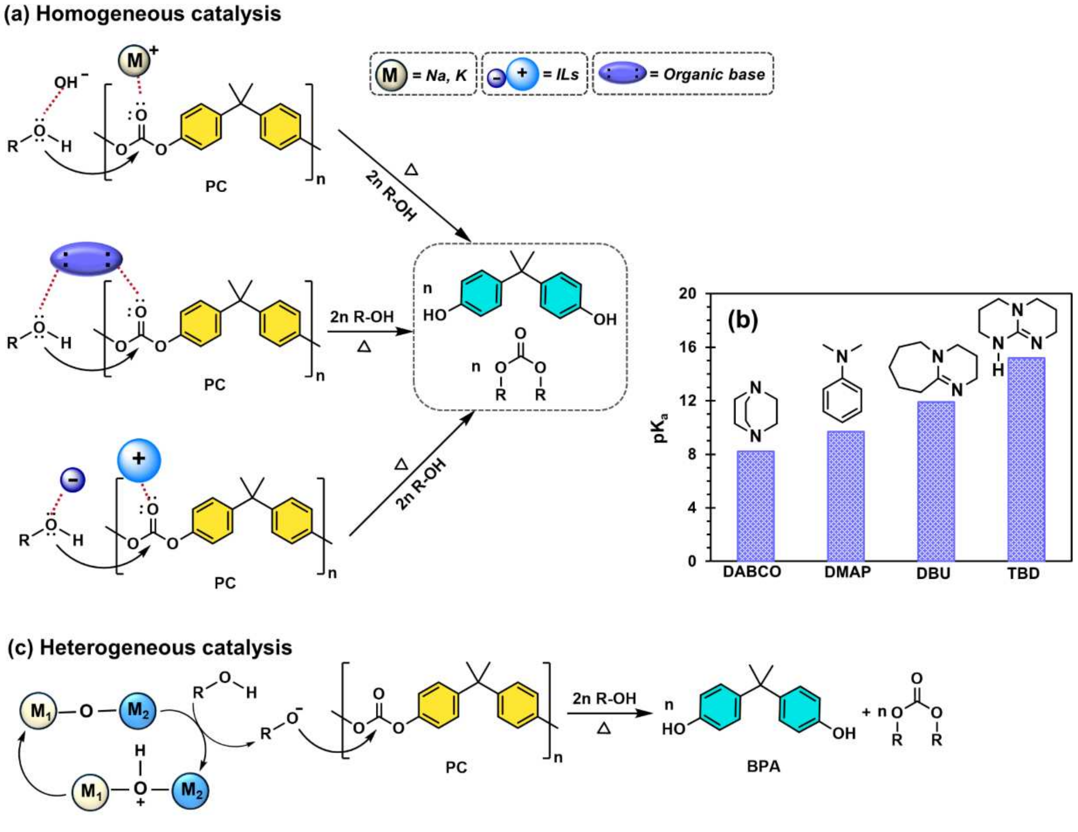


Fig. 11. (a) Simultaneous activation of the carbonyl group of PC and the nucleophile using homogeneous catalysts such as alkali metal hydroxides, organic bases, and ILs for methanolysis.

(b) pKa values of various organic bases in water. Data taken from ref. [195-197]. (c) Activation of methanol by alkaline earth metal oxides during methanolysis of PC (R = CH_3).

b. Organic base-catalyzed alcoholysis: Like alkali metal hydroxides, organic bases were also found to promote methanolysis (Fig. 11). Organic bases like DBU, 1,4-diazabicyclo[2.2.2]octane (DABCO), and 4-(dimethylamino)pyridine (DMAP) were investigated for methanolysis of PC at a moderate reaction temperature (≤100 °C) [198]. Poor thermal stability of these catalysts is the main reason for selecting a moderate reaction temperature. As a result, high catalyst loading (up to 10 wt%) is required to maintain an appreciable depolymerization rate. It was also observed that basicity of the catalyst has a positive influence on the methanolysis rate (Fig. 11b), and catalyst loading could be significantly decreased using a catalyst with strong basicity. For example, catalysts with lower basicity (e.g., DABCO and DMAP) require longer depolymerization time (Table 2), whereas catalysts such as DBU can significantly decrease the reaction time. All the catalysts achieved complete depolymerization with ≥96% BPA yield [198]. When Alberti et al. used microwave heating (180 °C) for DMAP catalyzed methanolysis, only 25% of DMAP was required as compared to conventional heating and methanolysis was complete within 5-10 min with 99% BPA selectivity (Table 2) [198, 199]. In this case, uniform heating and short reaction time allowed the methanolysis under high PC loading (14–28 wt%) without sacrificing BPA selectivity [199]. As mentioned earlier, the use of THF as a co-solvent can decrease the depolymerization temperature from 180 to 140 °C. However, the dilution of the reaction medium led to a visible decrease in the depolymerization rate, although the BPA selectivity remained high (99%) [199].

Methanolysis was possible at a lower temperature (75 °C) when methyl-THF and dimethyl carbonate (DMC) were used as co-solvents in the presence of stronger bases, such as 1,5,7-triazabicyclo [4.4.0]-dec-5-ene (TBD, 1%) [200]. The excellent BPA yield (>96%) obtained under moderate reaction conditions (75 °C, 1 mol% TBD) is the main advantage of this process. Even though organic catalysts can reduce the depolymerization temperatures (from 120 °C to 70 °C) and improve the BPA selectivity, there is a lack of literature on recovery and reuse of the catalysts [198]. Therefore, a more systematic investigation is required to assess the scalability and economic viability of the organic-base catalyzed alcoholysis process on a large scale [200].

c. IL-catalyzed alcoholysis: As discussed in the context of PET depolymerization, the dual roles of ILs have been exploited for the methanolysis of PC. As Ils act as solvents and catalysts, it is challenging to classify their roles compared to other catalytic systems. The typical IL-mediated transesterification of PC is shown in Fig. 11a. As observed in the case of IL-catalyzed hydrolysis, the basicity of the IL also strongly influences the methanolysis of PC. With the increase in the basicity of Ils ([Bmim][Cl]) > [Bmim][Ac] > [HDBU][Lac], the temperature and catalyst requirements for methanolysis decreased (Table 2) [201-203]. Regardless of the polymer loading, the selectivity of BPA remained high (≥96%). In the case of [HDBU][Lac], the reaction temperature significantly affected the IL loading and the depolymerization rate. The depolymerization was completed within 1 h in the presence of 0.7% [HDBU][Lac] at 120 °C. [203] Additionally, the depolymerization of PC was investigated using Ils with Lewis acid and base characteristics [204, 205]. IL-mediated methanolysis typically operate at high polymer loading (25–60 wt%) and moderate reaction temperature. Moreover, ILs achieve high depolymerization efficiency and they are easy to reuse. Despite their impressive performance in PC methanolysis, it is important to note that ILs are expensive and potentially toxic [109]. This raises concerns regarding their suitability for large-scale depolymerization.

3.2.2 Heterogeneously catalyzed alcoholysis

Apart from soluble catalysts, heterogeneous alkali metal oxides like CaO have also been studied for methanolysis. These catalysts offer ease of recovery, reusability, and low cost. An early example of heterogeneously catalyzed PC methanolysis involved the use of SBA-15 supported CaO and CaO-CeO_2 nanoparticles (NPs) in the presence of THF as a co-solvent. The nano-channels of SBA-15 facilitate the uniform distribution and accessibility of active CaO NPs that function as weak basic sites for the activation of the carbonyl linkages in PC. Due to the weak basic nature, a high catalyst loading (15–30 wt%) is required for depolymerization. Moderate reaction temperature minimizes the degradation of BPA and maintains high BPA selectivity (95%) even after 3 h long reaction time (Table 2) [206, 207]. In addition, both CaO-SBA-15 and CaO-CeO_2-SBA-15 catalysts displayed excellent reusability [206, 207]. In another study, Zhao et al. prepared uniformly distributed CaO NPs in a three-dimensional monocellular silica foam (MCF) to catalyze methanolysis of PC in presence of THF as a co-solvent [208]. Although the methanolysis conditions were nearly identical to the CaO-SBA-15 and CaO-CeO_2-SBA-15 catalyzed reactions (Table 2), only half the amount of CaO-MCF was required for the complete depolymerization (Table 2). This can be attributed to the improved accessibility of CaO NPs in the three-dimensional matrix of MCF [208]. However, a slight decrease in BPA selectivity was observed after the third reuse cycle [208].

Similarly, Liu et al. also synthesized calcium-doped γ-alumina catalyst with a large aspect ratio and featuring a worm-hole framework (Ca-Al_2O_3) [209]. In contrast to other CaO-based catalysts discussed earlier, only 3 wt% loading of Ca-Al_2O_3 was used for this THF-assisted methanolysis (Table 2) at 130 °C. Low catalyst loading shows the active nature of the catalyst and the methanolysis was completed within 3 h with 97% BPA yield [209]. Despite the easy recovery and recyclability of such CaO-based catalysts, there can be an economic drawback due to the need for additional solvent recovery steps.

Eliminating the use of co-solvents during methanolysis can simplify this process. Keeping this in view, heterogeneous catalysts with strong basic characteristics were investigated for the depolymerization of PC. In one such example, Huang et al. reported the use of Mg/Al-layered double oxides (LDOs) for PC depolymerization [210]. The strong basic nature of Mg/Al-LDOs arises from the surface OH^- and O_2^- groups linked to Mg and Al atoms, which can readily deprotonate methanol to generate alkoxide ions, favoring the transesterification of PC (Fig. 11c). Complete depolymerization was achieved after 1 h with 98% BPA yield at a moderate reaction temperature of 110 °C and a catalyst loading of only 3 wt% (Table 2). Most importantly, the absence of a co-solvent, high polymer loading (~60 wt%), easy catalyst reusability make the process highly attractive for scale-up [210].

3.2.3 Depolymerization of PC by alcoholysis using other alcohols

Alcohols such as ethanol, ethylene glycol, and propylene glycol have also been investigated for the depolymerization of PC. The products obtained from depolymerization using these alcohols are shown in Fig. 12. An early example of PC glycolysis was reported by Kim et al., achieving a BPA yield over 95% after a 1.25 h reaction at 220 °C [211]. Rosi et al. and Lin et al.

investigated base-catalyzed glycolysis of PC in the presence of various bases (NaOH, KOH, Na_2CO_3) and metal oxides (ZnO, CaO) [212, 213]. A high reaction temperature (180–200 °C) was favorable for achieving faster depolymerization rates (Table 2). However, a mixture of products was obtained under different reaction conditions, indicating poor selectivity of the glycolysis process [212, 213]. Working on glycolysis of PC, Oku et al. reported that the selectivity can be tuned towards product D_1 (100%, Fig. 13a) by carrying out the glycolysis in the presence of a low NaOH loading and an excess amount of ethylene carbonate (Product C_1, Fig. 13a) [214]. A high reaction temperature (180 °C) achieved complete depolymerization after 10 minutes, resulting in a 100% yield of product D_1 (Fig. 13a) [214]. The depolymerization product is not suitable for virgin PC synthesis, but it can serve as a building block for other polymeric materials. Later, Nikje et al. reported that it is possible to tune the glycolysis of PC toward high BPA yield (92%) by using high EG and NaOH loading under microwave heating (180 °C) [215]. The combination of microwave heating and high NaOH loading resulted a fast glycolysis and complete depolymerization of PC was achieved within eight minutes [215]. Recently, Do et al. found that co-solvent (methyl-THF) assisted glycolysis of PC can be carried out at a very low temperature (30 °C) using TBD as the catalyst [200]. High BPA yield (98%) and low reaction temperature are two main advantages of the process [200]. However, very long reaction time (12 h) and the use of co-solvents are major drawbacks of such a process.

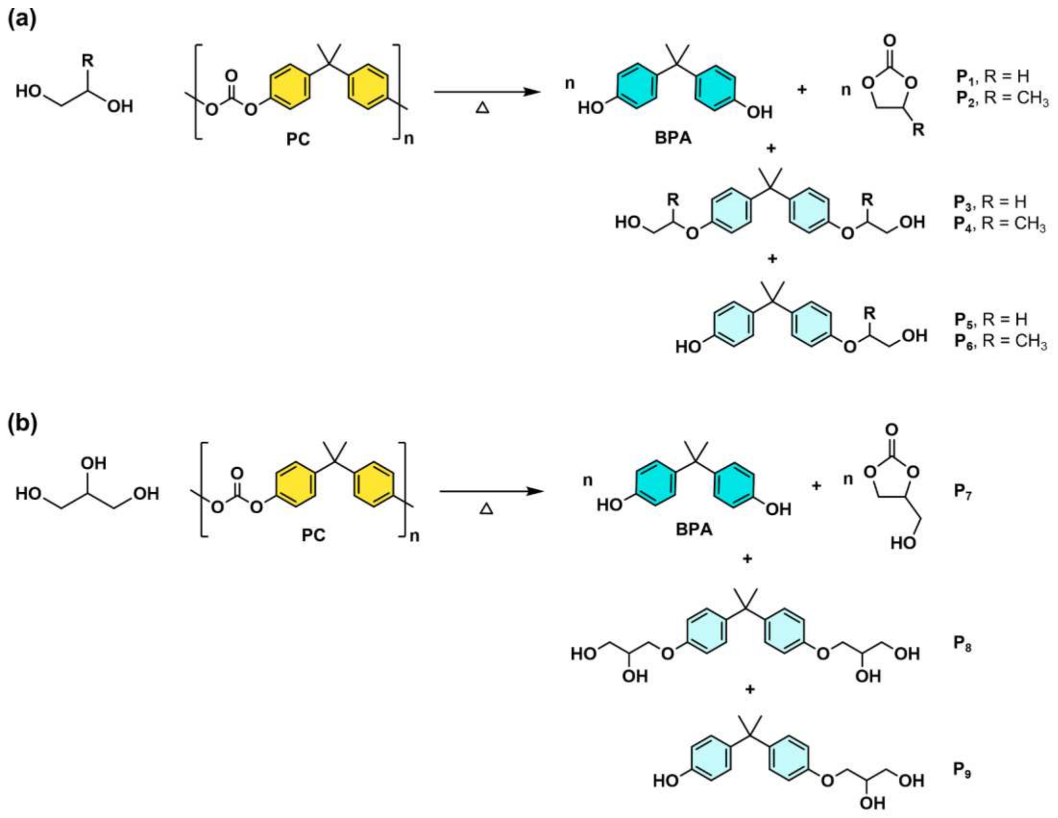
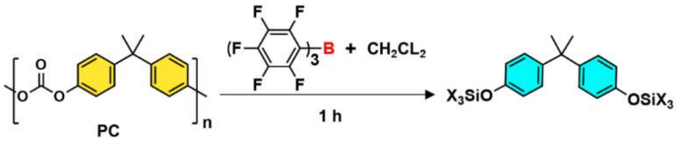


Fig. 12. Alcoholysis of PC using alcohols other than methanol.

When propylene glycol (PG) was used for the alcoholysis of PC in the presence of Na_2CO_3 and ZnO, Lin et al. and Rosi et al. observed a mixture of products [212, 213]. The inclusion of THF and urea in the reaction mixture led to the formation of product D_2 with a 95% yield [213]. However, Do et al. found that by using TBD as a catalyst and a co-solvent (Methyl THF), PC depolymerization can occur at 30 °C to achieve ~93% BPA yield after a 12 h reaction [200]. Similarly, glycerol was also investigated as a transesterification reagent to depolymerize PC into BPA (Fig. 12b). Hidaka et al. observed that glycerol can depolymerize PC to BPA with ~100% yield after 25 minutes of reaction in the presence of KOH and dioxane at 100 °C [216]. In a DBU-catalyzed alcoholysis of PC in presence of glycerol and THF obtained 97% BPA yield [217]. Homogeneous reaction mixture and presence of a base allowed to carry out glycolysis at a mild temperature of 60 °C [217]. It is important to note that higher reaction temperatures or the use of a co-solvent is necessary to achieve appreciable depolymerization rate in presence of these high molecular-weight alcohols.



	BPA-Si Yield
Et_3SiH (4.2 mol. eq.)	*82%*
TMDS (2.2 mol. eq.)	*98%*

Fig. 13. Reaction scheme showing the reductive depolymerization of PC using silanes in the presence of borane catalysts ($B(C_6F_5)_3$) and the subsequent formation of disilylated-BPA [124]. Disilylated-BPA formed after depolymerization can be hydrolyzed to BPA.

3.3 Depolymerization of PC by reduction

Reductive depolymerization of PC was also investigated as an alternative to hydrolysis and alcoholysis. The most efficient reductive depolymerizations reported in the literature focus on homogeneous catalysts utilizing silane as a reducing agent. For example, Feghali et al. used an $Et_3SiH/B(C_6F_5)_3$ catalytic system for the room-temperature depolymerization of PC with 82% disilylated BPA (BPA-Si) yield (Fig. 13). Subsequently, the BPA-Si was hydrolyzed to BPA (Table 2) [124]. Use of TMDS as the silane had a positive impact on depolymerization, and the yield of BPA-Si improved significantly (98%) under similar reaction conditions [124]. Likewise, Monsigny et al. used an Iridium pincer complex as a catalyst for PC depolymerization (Fig. 7b) [127]. The iridium catalyst was found to be weaker compared to the $B(C_6F_5)_3$ catalyst [124], and only 88% BPA-Si yield was obtained after 6 h of reaction at 65 °C [127]. Hydrogenative depolymerization of PC with a high BPA yield is also possible using Ru- and Fe-pincer catalysts [161, 218-220]. However, reductive depolymerizations of PC using homogeneous catalysts require long reaction times, rendering these processes economically unviable at the current stage of development (Table 2).

In addition to the use of homogeneous catalysts, heterogeneous catalysts have also been investigated for reductive depolymerization of PC. However, the main objective of these researches was to synthesize jet fuel-grade cycloalkanes from PC waste through the hydrodeoxygenation pathway [221, 222]. In two separate reports, Wang et al. used Rh/C and RANEY® Ni in the presence of an ultra-stable acidic zeolite as a co-catalyst for efficient hydrodeoxygenation of PC [221, 222]. During the preparation of this review, no report on the reductive depolymerization of PC using heterogeneous catalysts was found.

3.4 Comparison of different PC depolymerization processes.

Fig. 14 shows the comparison of different depolymerization pathways reported for PC. Fig 14a showing the broad comparison between processes shows that conversion and BPA selcectivity of all processes are high (nearly 100%). However, the polymer loading in the reaction mixture was very different in among processes. For example, methanolysis can be carried out under high polymer loading (>50 wt%), whereas lowest polymer loading was used during reductive depolymerization. Similar reaction time distribution was observed for both hydrolysis and methanolysis (Table 2, Fig. 14a), except reductive depolymerization where very long reaction time was required for complete depolymerization. In terms of catalyst loading, reductive processes use highly active homogeneous metal catalysts, which allows them to operate under lowest catalyst loading. Overall, similar amount of catalyst loading is required for hydrolysis and alcoholysis. In both cases, very high catalyst loading was required when heterogeneous catalysts and ILs were used. However, major difference between the processes was observed when output of various processes were compared (Fig. 14b).

Uncatalyzed methanolysis is carried out under high temperature and low polymer loading, which lead to uncontrolled reactions with poor BPA yield (Fig. 14b). Overall, catalyzed methanolysis is more efficient than any other PC depolymerization methods. A significant reduction in reaction temperature and improvement in output can be achieved by using a catalyst during methanolysis (Fig. 14b). Presence of co-solvents in during catalyzed methanolysis can decrease the reaction temperature further. However, no improvement in output was observed following such strategy. Rather, use of a co-solvent require an extra separation step after depolymerization. In case of hydrolysis, synergy of both catalyst and co-solvent was required to achieve appreciable BPA output during depolymerization. Except one report (entry 4, table 2), overall BPA output of hydrolysis is significantly lower compared to that of the catalyzed methanolysis (Fig. 14b). Since such a high BPA output value was observed only once, it was not used for preparation of Fig. 14b Because it is only one case, Moreover, hydrolysis requires a slightly higher temperature than catalyzed methanolysis (Fig. 14a, b). Like PET, reductive depolymerization of PC also suffers from a long reaction time, low polymer loading, and the use of soluble catalysts (Fig. 14a). As a result, the output of reductive processes was the lowest (Fig. 14b).

During the preparation of the review, it was observed that most of the studies focused on pure PC, while a significant portion of PC is actually used as a blend with other polymers. Moreover, the long lifespan of PC-based components and the lack of detailed information in the PC value chain are major obstacles to upscaling PC depolymerization processes. To the best of our knowledge, no large-scale setup has been found for PC depolymerization during the preparation of this review, despite numerous reports on lab-scale PC depolymerization processes (TRL 3–4). Large-scale PC depolymerization is possible by addressing bottlenecks related to the identification, collection, and selective sorting of PC waste.

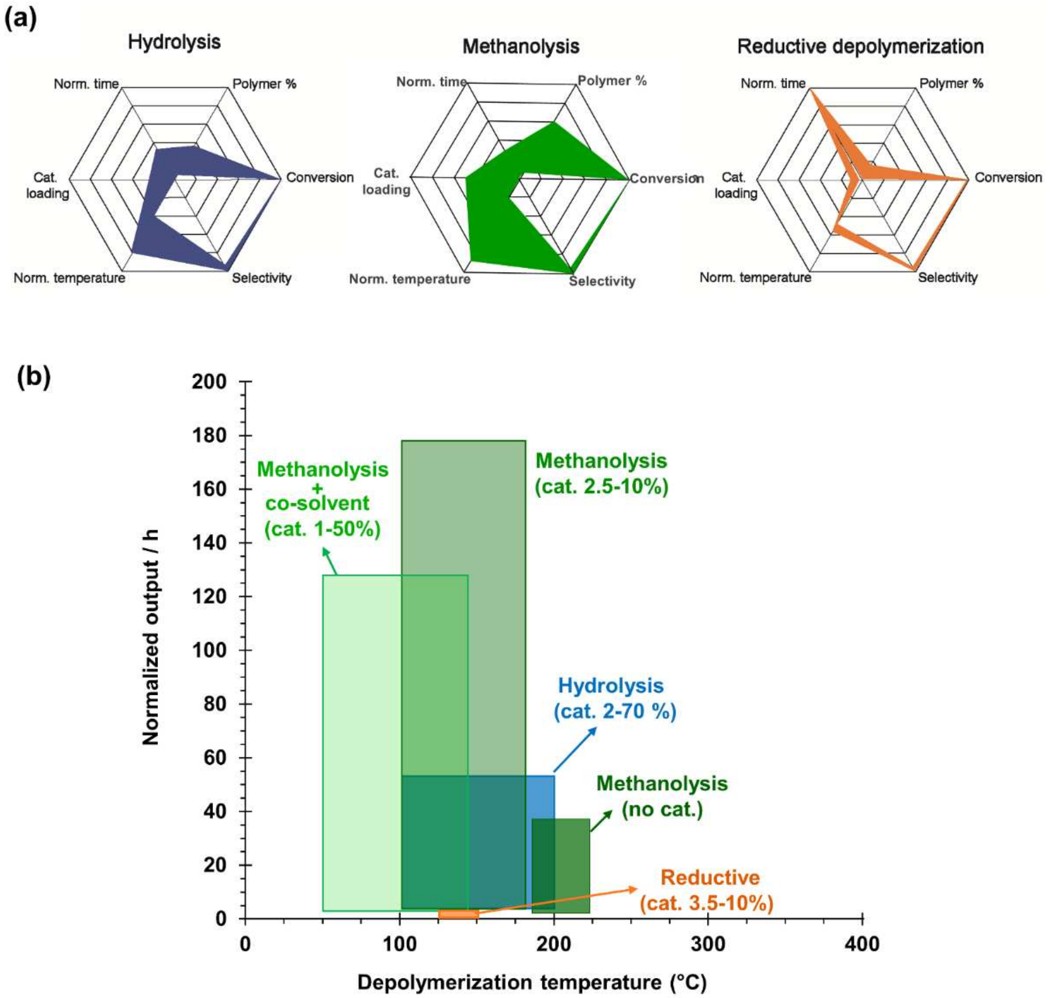


Fig. 14. (a) Spider diagram illustrating various PC depolymerization reaction parameters in a specific process and their impact on conversion and BPA selectivity. The shaded area represents the upper and lower limits for time, catalyst and polymer loading, temperature, conversion and selectivity towards the monomer for each processes. (b) Comparison of different depolymerization methods based on BPA output. Reports with high BPA yield (>95%) were

considered for this comparison. “cat. ” refers to catalysts loading (%) on the weight of PC used for depolymerization. In case of ionic liquid (IL) based processes, IL loading ≤50% is considered as catalyst and IL loading >50% is considered as a solvent. Data used for preparation of the figure are listed in Table 2 and normalized output was calculated using Eq. 1 and figure was plotted as described in Sec. 2.6.

Table 2: Depolymerization of polycarbonate (PC)

No	Solvent	Polymer loading (%)	Catalyst	Cat. %	Time (h)	Temp. (°C)	Conv. (%)	BPA Yield (%)	Ref.
Hydrolysis of PC									
1[a]	NH_3 sol. (0.6 mol/kg)	0.04	none	0	0.83	160	100	94	[175]
2[a]	H_2O	-	none	0	0.83	160	100	94	[175]
3	Dioxane:H_2O (4:1)	20	NaOH	10	8	100	100	95	[184]
4	Glycerin:H_20 (4:1)	50	NaOH	2	0.03	150[b]	100	100	[185]
5	Glycerin:H_20 (1:4)	50	NaOH	2	0.4	150[b]	100	100	[185]
6	H_20	31	[Bmim][Cl] (R)	150	3	165	100	95	[177]
7	H_20	35	[Bmim][Ac] (R)	150	3	140	100	96	[176]
8	H_20	75	[HDBU][LAc] (R)	10	3	140	100	98.5	[186]
9	THF:H_20	5.5	TBAC + ZnO NPs (R)	1.6+5.5	7	100	100	99	[183]
10	THF: H_2O	8.5	$La(O_3SCF_3)_3$	23.3	6	160	100	97	[178]
11	H_20	5	CeO_2 NPs	0.03	3	200	100	90	[179]
12	DEG:H_2O (2.33:1)	37.5	$Fe_3O_4/SiO_2/NH_2$ NPs	5.3	1.3	160	100	100	[189]
13	THF:H_2O	14	Chlorite (R)	70	6	200	99	98	[190]
Alcoholysis of PC									
14	MeOH	10	none	0	0.25	220	100	90	[191]
15	MeOH	6	none	0	3	190	100	94	[192]
16	MeOH	6	KOH		0.03	120	100	98	[193]
17	Toluene:MeOH	33.5	NaOH	2.5	0.25	60	100	96	[194]
18	MeOH	18	DBU	10	0.5	100	100	99	[198]
19	MeOH	18	DMAP	10	2.0	100	100	97	[198]
20	MeOH	18	DABCO	10	3.25	100	100	96	[198]

21	MeOH	14.4	DMAP	2.5	0.08	180^{b}	100	99	[199]
22	MeOH	28.8	DMAP	2.5	0.16	180^{b}	100	99	[199]
23	THF:MeOH (2:1)	10	DMAP	2.5	0.7	140^{b}	100	99	[199]
24	MeTHF:MeOH	10	TBD	1	12	50	100	99	[200]
25	DMC:MeOH	10	TBD	1	6	50	100	99	[200]
26	DMC:MeOH	10	TBD	1	2	75	100	99	[200]
27	[Bmim][Cl]:MeOH (1:1)	33	[Bmim][Cl] (R)	100	2.5	105	100	95.5	[201]
28	[Bmim][Cl]:MeOH (1:3)	25	[Bmim][Cl] (R)	50	2.5	105	100	95.5	[201]
29	[Bmim][Ac]:MeOH	36	[Bmim][Ac] (R)	100	2.5	90	100	95.6	[202]
30	[Bmim][Ac]:MeOH	44	[Bmim][Ac] (R)	50	2.5	90	100	95.6	[202]
31	[Bmim][Ac]:MeOH	33	[Bmim][Ac] (R)	50	2.5	90	100	95.6	[202]
32	MeOH	57	[HDBU] [LAc] (R)	12	2.5	90	100	98	[203]
33	MeOH	61	[HDBU] [LAc] (R)	0.7	1	120	100	99	[203]
34	MeOH	54	[Bmim]Cl·2FeCl$_3$	10	3	120	100	98	[204]
35	MeOH	49	[HDBU][Suc] (R)	27	2	70	100	96	[205]
36	MeOH: TBAC	4.5	TBAC + ZnO (R)	1.6+5.5	7	100	100	97	[183]
39	THF:MeOH	25	10%CaO/SBA-15 (R)	33	3	130	100	97	[206]
40	THF:MeOH	26	15%CaO/Ce-SBA-15 (R)	30	3	130	100	95	[207]
41	THF:MeOH	24	12% CaO/MCF	13	2.5	125	100	96.5	[208]
42	THF: MeOH	28	Ca-Al$_2$O$_3$	3	3	130	100	96.5	[209]
43	MeOH	60	Mg$_3$Al-LDO	3	1	110	100	98.3	[210]
44	Benzyl alcohol	6	None	0	3	250	100	100	[192]
45	1-2 Propanediol	4.5	TBAC + ZnO (R)	1.6+5.5	7	100	98	100	[183]
46	EG	73	Mg (OAc)$_2$	0.06	1	180	99	90	[223]
47	EG (Zn (OAc)$_2$)	73	Zn (OAc)$_2$	0.07	1	180	99	96	[223]
48	DMSO: Ethanolamine	18	None	0	2	80	100	96	[224]
49	Ethanolamine	80	None	0	47	120	100	98	[224]
50	Aniline: tetrabutylammonium chloride	3.5	TBAC+ZnO (R)	1.6+5.5	7	100	99	96	[183]
Reductive depolymerization of PC									
51	1,4 dioxane	7.5	[Ru(triphos)tmm]	4.1	16	140	99	99	[161]
52	THF	16	Milstein Catalyst II	9	24	140	99	99	[218]
53	THF	16	Milstein Catalyst II + KOtBu	10	24	120	99	99	[219]
54	THF	4	Fe Pincer catalyst	3.5	24	120	99	99	[220]

^{a} semi-continuous, ^{b}microwave assisted

TBAC = Tetrabutylammonium chloride; Me-THF = Methyl tetrahydrofuran; MeOH = Methanol; Mg$_3$Al-LDO = Mg$_3$Al-Layered double oxide

EG- ethylene glycol; DEG= Diethyleneglycol; DMSO- dimethyl sulfoxide.

4. Polyether depolymerization

Polymers containing ether linkages (C-O-C) in their backbone are commonly known as polyethers [225]. Polyethers can be aliphatic or aromatic, with properties suitable for various applications. In general, polyethers can be classified as linear or cross-linked. Poly(ethylene oxide) (PEO), poly(propylene oxide) (PPO), and polytetrahydrofuran are examples of linear polyethers, whereas anhydride-cured epoxies are classified as crosslinked polyethers. Linear aliphatic polyethers are flexible with low crystallinity and are widely used in pharmaceuticals, cosmetics, lubricants, and surfactants. Low production volume (Fig. 2a), and easy degradation of linear polyethers prevent their accumulation in the environment [226]. This is possibly the reason for lack of interest to depolymerize polyethers. However, increasing awareness of circularity, efforts are being directed towards the depolymerization of polyethers.

On the other hand, epoxy resins are the main example of crosslinked polyether, and they are known for their exceptional physical properties, structural stability, solvent resistance, and low thermal shrinkage [227-229]. These properties make epoxies suitable for matrices in fiber-reinforced composites (FRC) designed for load-bearing structural materials such as windmill blades, automobile components, and various other applications. Crosslinks in epoxy resins, which are desirable in high-performance, pose a significant obstacle for recycling [230-233]. As a result, epoxy-based materials are either disposed of in landfills or incinerated, leading to negative environmental impacts. Considering the high-value fibers in FRCs (carbon fiber or glass fiber), various depolymerization methods have been investigated to recover the fibers. However, the recovery and reuse of organic matrices/epoxies has largely been neglected. This not only leads to polymer waste but also raises environmental concerns, as the remaining epoxy is not biodegradable and typically contains toxic molecules, particularly BPA [164, 165]. Incorporation of reprocessable covalent networks offers a possibility to enhance their lifespan of thermosets or achieve recyclability [234-236]. However, such technologies are at their early stage of the development and suffer from poor thermo-mechanical properties. Therefore, in the following section, we summarize the efforts made over the past decades to depolymerize linear polyethers and epoxies.

4.1 Depolymerization of linear polyethers

Depolymerization of polyethers is accomplished by activating the ether units with the assistance of transition-metal catalysts in acid chlorides and polyethers were depolymerized into monomeric chloroesters. Later these molecules can be used as building blocks for the preparation of other polymers (Fig. 15a) [237-241]. Enthaler et al. investigated the depolymerization of various polyethers including poly(ethylene glycol) (PEG), polytetrahydrofuran (PTF), and poly(propylene oxide) (PPO) using an anhydrous $ZnCl_2$ as the catalyst in the presence of acyl chloride. All depolymerization reactions resulted more than 80% monomer yield after the reaction. It was also observed that $FeCl_2 \cdot 4H_2O$ can also catalyze the depolymerization PTF and PEG (Fig. 15a) [238]. $FeCl_2 \cdot 4H_2O$ displayed superior catalytic activity compared to anhydrous $ZnCl_2$ and achieved complete depolymerization with ~90% monomer yield after 24 h of reaction at 100 °C. However, the separation of these homogeneous catalysts

(ZnCl$_2$ and FeCl$_2$·4H$_2$O) after the reaction is very challenging. This led to the investigation of heterogeneously catalyzed depolymerization of polyethers. In one such example, proton–exchanged montmorillonite was used to depolymerize PEG, poly(propylene glycol) (PPG), and poly(tetramethylene glycol) (PTMG) in the presence of acyl chloride [242]. At optimized conditions (100–130 °C, 24–48 h), ~80% of monomer yield was obtained. When benzoic anhydride was used instead of acyl chlorides, depolymerization of PEG was possible under milder conditions (100 °C, 12 h) with >85% yield [243]. XRD analysis revealed that depolymerization occurs at the proton-exchanged montmorillonite as shown in Fig. 15b [242, 243].

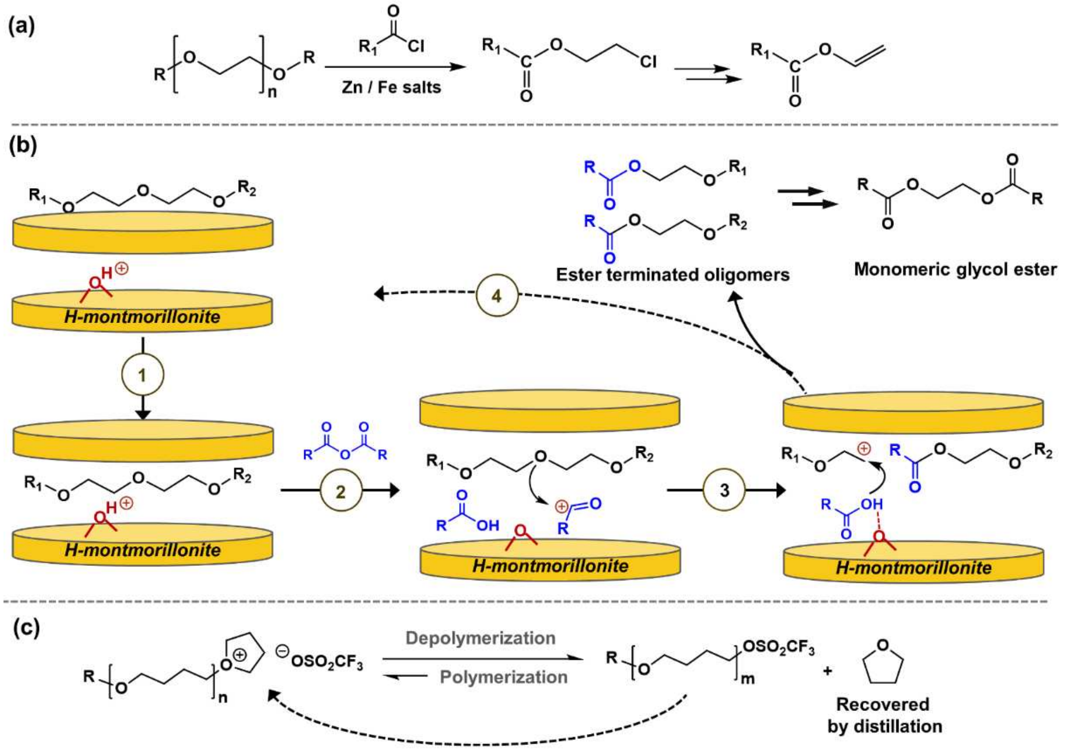


Fig. 15. (a) Polyethers depolymerization of by acid chloride and a transition metal catalyst [237-241]. (b) C-O bond cleavage in presence of proton-exchanged montmorillonite (H-

montmorillonite). The figure is adapted from ref. [243]. (c) Depolymerization of polytetrahydrofuran via a ring-closing using a triflate catalyst.

To eliminate the requirement of acid chlorides in the depolymerization process, the ring-closing depolymerization of polytetrahydrofuran was studied in presence of $FeCl_3$ and $Zn(OTf)_2$ (Fig. 15c) [239, 240]. THF formed during the depolymerization was removed continuously to drive the equilibrium in favor further depolymerization. Although $FeCl_3$ achieved ~90% THF yield after 20 minutes of reaction, a rapid decrease in activity was observed during subsequent recycling [239]. On the other hand, $Zn(OTf)_2$ was found to be a more robust catalyst and can maintain 84% of its activity even after the 9th cycle [240].

4.2 Depolymerization of Epoxies

Crosslinked network in epoxies that provides it necessary solvent resistance is a major hindrance during the depolymerization of epoxies by restricting the diffusion of solvent and catalysts. Therefore, the depolymerization of epoxies has mainly been carried out at elevated temperature using solvents, and either in the absence or presence of soluble catalysts [244-248]. In such situation, heterogeneous catalysts can suffer from severe diffusion limitation and slowdown the depolymerization reaction significantly. Therefore, use of heterogeneous catalysts is avoided during epoxy depolymerization reactions. Epoxies available in the market consist of different types of resins and crosslinkers, making it challenging to develop a standardized depolymerization strategy. Depolymerization in an epoxy network is dependent on the type of crosslinker used for its preparation. For example, anhydride-cured epoxies are mainly depolymerized via alcoholysis [245, 247, 249, 250] or aminolysis [251, 252] of ether bonds whereas amine-cured epoxies are depolymerized via the activation of N-atoms using a weakly coordinating Lewis acid catalyst [253, 254]. Considering that this review is focused on C-O bond depolymerization, only anhydride-cured epoxies will be discussed in the following sections. More details on the depolymerization of amine-cured epoxies can be found in recent reports [253-257].

4.2.1 Depolymerization of epoxies by hydrolysis

Early reports on depolymerization of anhydride-cured epoxies are mainly focused on the use of supercritical water (373 °C and 22 MPa) and methanol (240 °C and 8 MPa) [244, 246-248]. High depolymerization temperatures often degrade the main building blocks, rendering the depolymerized products unusable [244, 246-248]. To prevent degradation of the building blocks and maintain their functionality, Liu et al. used phosphotungstic acid, tungstosilicic acid, and phosphomolybdic acid as catalysts for hydrolysis [258]. Environmentally benign phosphotungstic acid (HPW) was found to be the most efficient among the tested catalyst. Moreover, HPW selectively cleaved the ester bonds in the network and obtained a cleaner product (Fig. 16a). Under the optimized conditions, 10 wt% HPW completely hydrolyzed the epoxy within 5 h at a reaction temperature of 190 °C. The depolymerized product was insoluble in water, which facilitated its separation from the catalyst solution by simple filtration. The catalyst solution was reused six times without any noticeable changes in the depolymerization efficiency. The oligomeric products recovered after depolymerization contain –OH and –COOH groups and offer a possibility to reuse it for the preparation of thermoset material [258].

In an innovative approach, Zhang et al. used amphiphilic dodecylbenzene sulfonic acid (DBSA) as a catalyst for hydrolysis of epoxy at a relatively lower temperature (170 °C, Fig. 16b) [259]. Amphiphilic nature of DBSA allowed its accumulation at the water–epoxy interface and initiate an acid-catalyzed hydrolysis from the surface of the epoxy rather than the bulk (Fig. 16b). Lower depolymerization temperatures minimized any undesired degradation of constituent molecules, and ~54 wt% of BPA was recovered after the reaction [259]. In a recent report, Zhang et al. used a solvent-assisted approach to carry out the acid-catalyzed hydrolysis of the epoxy in a swollen state [260]. They used γ-Valerolactone as the solvent to swell the epoxy and p-toluenesulfonic acid as the catalyst [260]. Complete depolymerization of the epoxy was achieved with recovery of nearly 87% of the constituent building blocks comprising of copolymers of styrene, methacrylic acid, and BPA-diglycidyl ether. The catalytic system was stable and was reused four times without any loss in activity. Subsequently, depolymerized building blocks could be reused for the preparation of thermoset [260]. A high reaction temperature (210 °C) and long time (12 h) are two major drawbacks of this process.

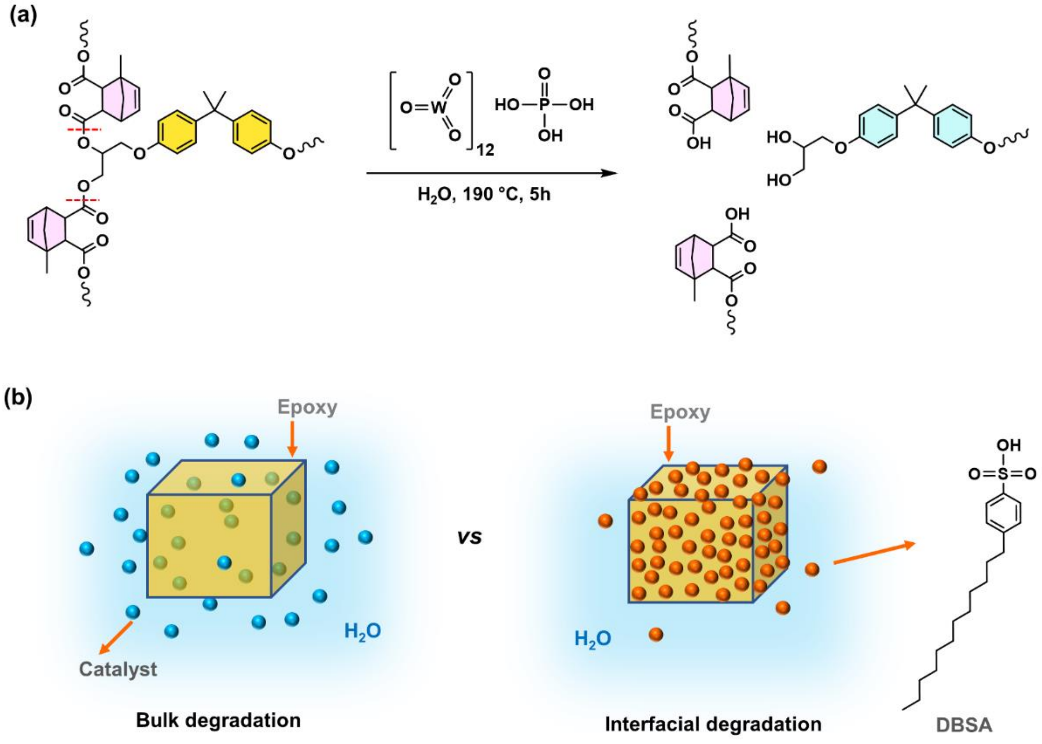


Fig. 16. (a) Hydrolysis of anhydride-cured epoxy in the presence of the HPW catalyst [258]. (b) Schematic representation of bulk and interfacial degradation of the epoxy. DBSA was used as the catalyst for interfacial depolymerization. The figure is adapted from ref. [259].

4.2.2 Depolymerization of epoxies by alcoholysis

Supercritical methanol can also lead to the depolymerization of epoxy resins under milder conditions compared to supercritical water (Table 3, entries 7, 8) [245, 261, 262]. While epoxy supercritical hydrolysis results in extensively degraded products, supercritical methanolysis obtain depolymerized products that can be re-cured [245, 262]. It is important to note that epoxy depolymerization primarily targets the recovery of high-value fibers from composites, and harsh depolymerization conditions can have a detrimental effect on the properties of the recovered fibers. This led to the use of catalysts in order to reduce the depolymerization temperature and time [263-266].

Zhao et al. followed an alcoholysis-hydrolysis approach using K_3PO_4 as the transesterification catalyst [263]. With the aid of K_3PO_4, it was possible to completely depolymerize epoxy (9 wt%) at a moderate temperature (110 °C, Table 3) [263]. Initial alcoholysis of the epoxy formed oligomers, which were then hydrolyzed into smaller molecules with carboxyl groups. After purification, the depolymerized products with carboxyl groups can be used as curing agents. K_3PO_4 is insoluble in alcohols at room temperature and can be easily removed from the reaction mixture [263]. KOH was found to have a positive impact on the transesterification of epoxy. However, the solubility of KOH in alcohols complicates the purification step [263]. In this study, ethanol was investigated for the alcoholysis of epoxy in the presence of K_3PO_4. Being a weaker nucleophile, ethanol requires a slightly higher depolymerization temperature (Table 3) [263]. In contrast to the alcoholysis-hydrolysis approach, Yang et al. followed a hydrolysis-alcoholysis approach at 180 °C using the PEG200-NaOH catalytic system [264]. In this mixed catalytic system, PEG200 forms a crown-like structure at the end of the polymeric chain in the presence of Na^+, and enhancing the reactivity of the base (Fig. 17a) [264]. As shown in Fig. 17a, initial depolymerization proceeds via hydrolysis, followed by alcoholysis with the help of nucleophilic PEG200 (Fig. 17a) [264]. As a result, epoxy was completely depolymerized after 50 minutes.

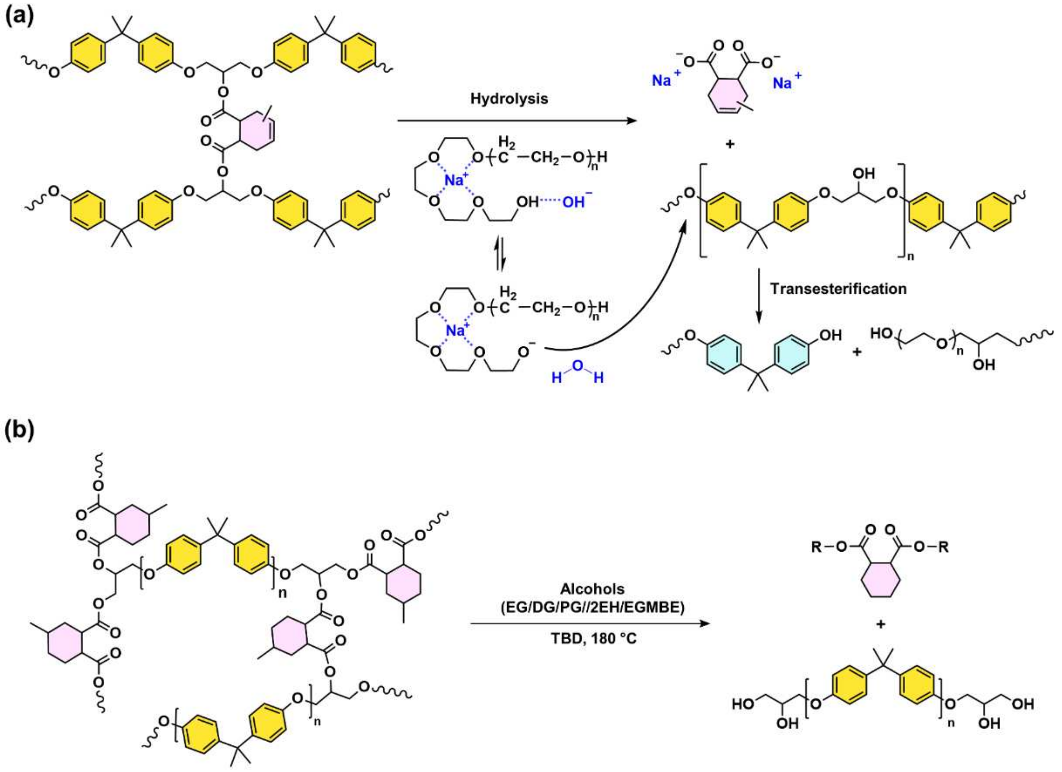


Fig. 17. (a) Plausible depolymerization mechanism of anhydride-cured epoxy using the PEG200-NaOH catalyst system. The figure is adapted from ref. [264]. (b) Depolymerization of epoxy via

alcoholysis in the presence of TBD and different alcohols (EG, PG-Propylene glycol, 2EH-2-Ethylhexanol and EGMBE- Ethylene glycol mono butyl ether).

In a base-catalyzed alcoholysis of epoxy, Kuang et al. used TBD as the catalyst [266] in presence of various alcohols such as EG, diethylene glycol, propylene glycol, 2-ethyl hexanol, and ethylene glycol monobutyl ether. Among different alcohols, ethylene glycol monobutyl ether was found to be the most efficient one and epoxy was completely depolymerized within 2 h at 180 °C (Table 3). The depolymerized product was a mixture of diesters and oligomeric tetra-alcohol (Fig. 17b) [266]. It was observed that the swelling of epoxy in different alcohols determines the thickness of the surface layer and ultimately affects the depolymerization [266]. Kuang et al. used a swollen epoxy system for faster depolymerization [265]. N-methyl-2-pyrrolidone (NMP) was used as the solvent during the organic base (TBD)-catalyzed glycolysis of epoxy resins [265]. The swelling of the epoxy in NMP facilitated the diffusion of EG and TBD throughout the matrix. Complete depolymerization of the epoxy was achieved within 1.5 h at 170 °C (Table 3), [265] which is significantly faster than glycolysis under similar conditions in the absence of NMP [266]. Moreover, high concentration of –OH in EG (35.8 mol/L) also favored the transesterification. Mild reaction conditions preserved the functionality in the depolymerized molecules for future reuse [265]. Alternatively, Pérez et al. investigated the IL-catalyzed glycolysis of epoxy by using ILs with different alkyl chain lengths and anions [267]. As mentioned in the PET and PC sections, ILs also play a dual role as both solvent and catalyst in the glycolysis of epoxy [267]. Butylimidazolium (acetate) (BMIm[Ac]) and EG mixtures (20:80) were found to depolymerize epoxy within 1.5 h at

150 °C (Table 3). Increasing the alkyl chains length of IL, slow depolymerization was observed, which can be attributed to poor diffusion of long chain ILs (Table 3) [267].

4.2.3 Depolymerization of epoxy by aminolysis

A recent report suggests that ester linkages in the anhydride-cured epoxies can be cleaved with help of long-chain amines to achieve complete depolymerization (Fig. 18) [251, 252, 268, 269]. It was also observed that aminolytic depolymerization proceed at a mild reaction temperature in the absence of any catalyst and the depolymerized products can be recycled for various applications. Lin et al. used 1-hexylamine at 125 °C to depolymerize epoxy into phenoxy resin (Fig. 18) [251, 252]. Based on the crosslinkers used in the epoxy, N^1, N^3-dihexylisophthalamide, and dihydroxy urea were also recovered after depolymerization [251, 252]. Subsequently, the recovered phenoxy resin was reused for the preparation of adhesives and paints. However, Lin et al. observed that the nature of crosslinkers can significantly influence the efficiency of depolymerization [251]. Zhao et al. followed a similar depolymerization pathway using diethylenetriamine for aminolysis. Complete depolymerization (99%) was achieved after 50 minutes of reaction at 130 °C [269]. Based on the amines used during depolymerization, oligomers can be obtained with either –OH or $-NH_2$ groups (Fig. 18). In separate reports, Li et al. [270] and Zhang et al. [271] used ethanolamine (EA) for the aminolytic depolymerization of epoxy. Li et al. observed a complete depolymerization within 1 h at 160 °C using KOH as the catalyst [270]. Whereas, under almost similar reaction conditions and in the absence of KOH, Zhang et al. observed a longer reaction time (4 h) [271]. The use of EA for depolymerization afforded bifunctional oligomers (Fig. 18), which can be recycled for epoxy and polyurethane synthesis after appropriate modification [270, 271].

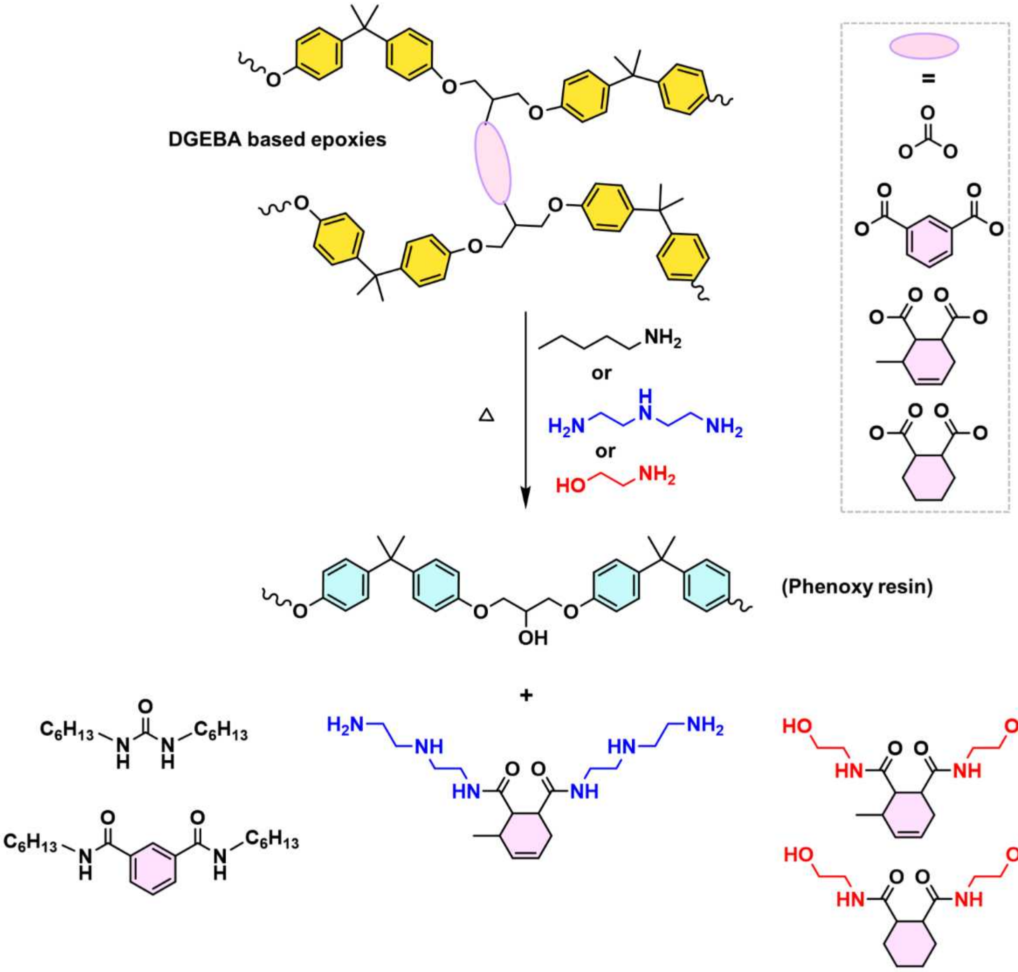


Fig. 18. Generalized reaction scheme showing aminolysis of anhydride-cured epoxy by various amines, resulting in the formation of phenoxy resin as the main depolymerized product, along with additional products based on the type of crosslinkers used for epoxy preparation and the amine used during depolymerization [252]. The figure is adapted from ref. [251, 252, 269-271].

4.3 Comparison of different epoxy depolymerization methods

Only a few reports are available on polyethers depolymerization processes at laboratory scale (TRL 3–4). The lack of sufficient reports makes it difficult to draw any conclusions regarding different linear polyether depolymerization processes. Therefore, this review compares different epoxy depolymerization processes that demonstrate the reusability of depolymerized matrices (epoxies). For a better understanding, the normalized outputs of the different processes were compared against reaction temperatures and catalyst loading (Fig. 19). As shown in fig. 19, hydrolysis without catalyst requires a high operating temperature and most of the reactions were carried out at a low epoxy loading, which led to very low output. High reaction temperature (>400 °C) also renders the depolymerized products unusable. The use of catalysts during hydrolysis allowed improved the depolymerization output by decreasing the reaction time or by improving the initial epoxy loading in the reaction. In case of base catalyzed hydrolysis, operating temperatures were high (Fig. 19, Table 3) and depolymerized products were not curable. On the other hand, acid catalyzed hydrolysis was possible at low temperature (<200 °C) and maintained the recurability of depolymerized products (Table 3).

As shown in Fig. 19, alcoholysis in the absence of a catalyst were carried out within 250–300 °C and addition of a catalyst in the reaction mixture significantly decreased the reaction temperature. Lower reaction temperature in alcoholysis prevented unwanted degradation of depolymerized products and they were curable. Despite of several advantages, output of catalyzed alcoholysis was lowest among different processes (Fig. 19) and catalyst loading was found to be high. In contrast, aminolysis of epoxy required moderate temperatures (125–160 °C) with higher output compared to alcoholysis process. Moreover, aminolysis allows to tune the end group functionality of the depolymerized products. Among these methods, aminolysis is relatively new and can be performed effectively without catalyst. Further optimization is required to improve the efficiency of aminolysis. To the best of our knowledge, different processes discussed in this review are mainly carried out on a laboratory scale (TRL 3–4).

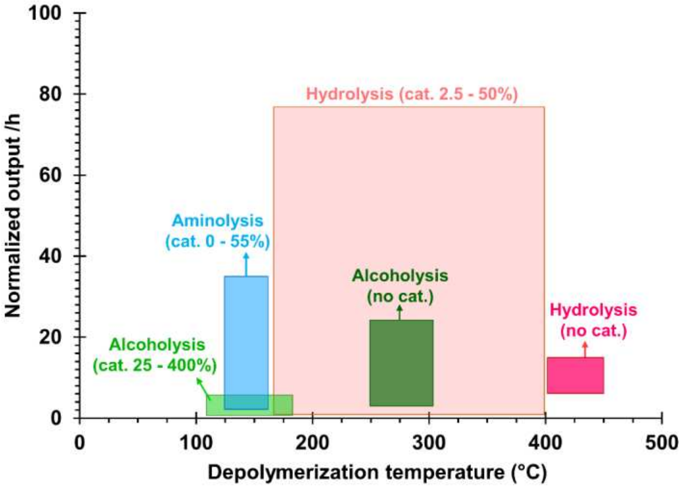


Fig. 19. Comparison of different anhydride-cured epoxy depolymerization processes based on depolymerization efficiency (output) and overall operating conditions (temperature and catalyst loading). "cat." refers to the catalysts loading % calculated based on the weight of epoxy. "no Cat." refers to the absence of catalyst in the reaction mixture. All data was taken from Table 3 and normalized output was calculated by using Eq 2.

$$\text{Normalized output/h} = (\text{polymer loading}\ \% \times \text{conversion})/\text{time (h)} \qquad (2)$$

Table 3: Depolymerization of epoxy

No	Solvent	Polymer loading (%)	Catalyst	Cat. (%)	Time (h)	Temp. (^oC)	Conv. (%)	Curability	Ref.
1	H_20	3.5	None	None	0,5	440	97	no	[244]
2	H_20	-	K_2CO_3	2.5	0.75	400	70.9	no	[246]
3	H_20	-	K_2CO_3	5	4	350	71	no	[247]
4	H_20	20	KOH	5	0.25	400	96	no	[248]
5	H_20	10	HPW	50	5	190	100	yes	[258]
6	H_20	8	DBSA	9	10	170	99	BPA[a]	[259]
7	MeOH	-	None	None	1.5	270	100	yes	[245]
8	MeOH	-	None	None	1	270	100	yes	[262]
9	MeOH	9	K_3PO_4	25	3	110	96.2	yes	[263]
10	EtOH	9	K_3PO_4	25	3	120	95	yes	[263]
11	EtOH	9	KOH	25	3	120	100	yes	[263]
12	PEG200	4	NaOH	0.8	0.8	180	99	no	[264]
13	EG/NMP	5	TBD	87.5	1,5	170	95	yes	[265]
14	EGMBE	5	TBD	94	2	180	100	yes	[266]
15	EG/BMIm[Ac]	5	BMIm[Ac]	400	2.5	150	100	yes	[267]
16	EG/BMIm[prop]	5	BMIm[prop]	400	3.5	150	100	yes	[267]
17	Hexylamine (HA)	4.5	None	None	2	125	100	yes	[251]
18	HA	4.1	None	None	6	125	100	yes	[252]
19	Diethylenetriamine	29	None	None	0.83	130	98	yes	[269]
20	Ethanolamine	4	KOH	54	1	160	100	yes	[270]
21	Ethanolamine	25	None	None	4	160	100	yes	[271]

[a]BPA was obtained as the depolymerization product.

During preparation of this review, it was observed that depolymerization of OPs are mainly achieved via hydrolysis, alcoholysis, aminolysis and reductive pathways. Hydrolysis is the simplest way to depolymerize OPs to their constituent monomers. Water being a weak nucleophile, hydrolysis require relatively higher temperature compared to other processes. Comparing individual polymers, it was observed that hydrolysis of PET produces TA and EG. Whereas, hydrolytic depolymerization of PC produces BPA and constituent CO_2 in the polymer chain is lost.

In case of epoxy, no such depolymerization related mass loss is observed. When it comes to alcoholysis, glycolysis is the most widely used pathway for PET depolymerization and BHET obtained after depolymerization can be used directly for PET synthesis. Methanolysis is another efficient pathway to depolymerize PET to DMT. Due to commercial reasons, methanolysis is a less preferred process these days. On the other hand, methanolysis is the most widely used method to depolymerize PC to BPA and DMC. Interestingly, only few reports are available on alternative alcoholysis methods to depolymerize PC. When it comes to alcoholysis of epoxies, all kind of alcoholysis display similar depolymerization efficiency and afforded re-curable products. It is well known that increasing the nucleophilicity of reagents can depolymerize OPs at moderate reaction conditions. Based on this concept, nucleophilic amines were investigated for depolymerization of OPs. However, PET aminolysis products are not suitable to resynthesize PET. In contrary, aminolysis of PC produce BPA, which can be utilized to resynthesize PC. Aminolysis of PC is quite new and output of such process are low compared to hydrolysis and alcoholysis. Interestingly, aminolysis of epoxy is quite relatively easy affording re-curable products and depolymerization can proceed at lower reaction temperature (100–160 °C). Use of reductive pathway to depolymerize OPs is new and output of these processes are quite low for both PET and PC. Additionally, reductive pathways need a post-depolymerization step to recover original monomer, which ultimately decrease the output and increase the overall cost of the process. Among all processes, reductive depolymerization pathway of both OPs (PET and PC) are quite new and need further optimization to improve the output.

5. Conclusions and perspectives

In this review, we discussed and compared recently reported efficient depolymerization technologies for PET, PC, and polyethers. It was observed that depolymerization technologies of PET through hydrolysis, glycolysis, and methanolysis are mature (TRL 7–9). On the contrary, the last few decades witnessed a decreasing number of reports on methanolysis of PET, which can be attributed to the transition of commercial PET synthesis from the DMT route to the TA and BHET routes. When it comes to depolymerization of PC, methanolysis shows potential for scale-up. Although base-catalyzed methanolysis is quite efficient, downstream purification steps to recover catalyst and monomer is a challenge due to the homogeneous nature of catalysts. In this context, further investigation is required to improve the depolymerization efficiency of heterogeneous catalysts, that can also simplify the downstream operations and improve the economic viability of methanolysis. Despite numerous reports on efficient depolymerization processes, no large-scale PC depolymerization facility was found during preparation of this review. Reasons for the absence of large scale depolymerization facility could be: (i) PC being predominantly used as blends with various polymers, which makes the sorting and collection of PC waste quite challenging. (ii) PC waste source are scattered which also makes it difficult to ensure a continuous supply of PC waste, a crucial requirement for large-scale operations. Conversely, epoxy depolymerization primarily serves as a method to recover high-value reinforcement fibers in the composites rather than recycling of epoxy. The concept of depolymerizing epoxy and reusing the depolymerized product for the preparation of new material is novel and is a step in the right direction. Aminolysis and alcoholysis of epoxies are quite promising for anhydride cured epoxy resins and allows to recover recurable depolymerized products. Milder operating conditions and high depolymerization efficiency of these two processes make them potential candidates for scale-up.

Despite substantial progress in lab-scale depolymerization of OPs, large-scale operations based on these technologies are limited. We believe few changes in the approach are necessary to expedite scale up. These approaches are: First, future research activities need to demonstrate the technology by using OP blends containing additives that are encountered in real waste streams. At present, most studies in literature deal with pure and relatively clean OPs, which are far from the real scenario. As a result, transforming an efficient lab-scale depolymerization process into an industrial-scale process is not obvious, as it needs to address factors such as decrease in depolymerization efficiency due to varying feedstock quality, and losses occur during downstream processing. Second, the slow implementation of chemical recycling facilities is attributed to lengthy procedures for obtaining permits, particularly for waste storage and processing. This issue can be resolved by implementing favorable policies to streamline the process. Third, approaches needs to be considered to ensure an uninterrupted supply of feedstock for economically viable operation. For example, the complex and scattered waste stream of PC makes it logistically difficult to operate large-scale BPA recovery plants from PC waste. This bottleneck can be resolved by developing effective and efficient sorting techniques in parallel with depolymerization technology development. Fourth, techno-economic assessments of different processes need to be conducted to gain sufficient insights on the economic feasibility and challenges associated with the process. This will help determine the technical targets that need to be achieved. Finally, it is important to implement concepts of eco-design into OP products to reach sustainable plastic circularity. This involves replacing the multiplastics products with high-quality mono-plastics that contains fewer additives. This is essential to facilitate downstream processing and decrease the overall cost of depolymerization processes.

Research activities and technological progress in the field of plastic depolymerization contribute directly to Sustainable Development Goal (SDG) 9 through innovation for new industrial infrastructure towards sustainable growth [48]. Such industrial infrastructure scattered around the globe will generate new employment opportunities for the local population and contribute to economic growth (SDG 8). Growing research activities in this field align with SDG 12 by raising awareness in society and encouraging sustainable production and consumption behavior. Progress in the field of chemical recycling will decrease fossil fuel consumption and greenhouse gas emissions, thereby which fulfilling the objectives of SDG 13 in the long term. OP depolymerization will significantly decrease the amount of plastic in landfills as well as water bodies, addressing SDG 14 by preventing further pollution of water bodies by macro and microplastics. This has an indirect and positive impact on providing clean drinking water to a larger population (SDG 6). Decreasing the amount of plastics reaching landfill prevents land deterioration and meets the objectives of SDG 15. Overall, plastic depolymerization promotes SDG 17 by stimulating partnerships at different levels, technology transfer, and infrastructure development for a sustainable growth.

Conflicts of interest

There are no conflicts to declare.

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