

Mini Review

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Recent advance in poly(bisphenol a carbonate) chemical recycling: catalysts, reaction strategies, and its applications

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Abstract

The chemical upcycling of waste plastics into high-value-added products such as monomers, fuels, or fine chemicals represents a promising strategy for mitigating the adverse effects of massive end-of-life plastics. Poly(bisphenol A carbonate) (BPA-PC) stands out as a notable engineering plastic due to its exceptional overall performance; however, its durability and potential environmental toxicity make its recycling imperative. Although a lot of reviews about plastic degradation have been done before our review, the progress for plastic degradation needs to be constantly updated and summarized due to the rapid development of this field. Meanwhile, BPA-PC, as an important notable engineering plastic, previous reviews only focused on its depolymerization into monomers and missed their further conversion into final chemicals. which In this concise review, we summarize recent developments in the chemical upcycling of BPA-PC to valuable chemicals, emphasizing the role of various catalysts and reagents. Some of the most utilized chemical upcycling strategies such as alcoholysis, aminolysis and



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upcycling of BPA-PC in “polymer-to-polymer” format to reproduce new polymers are elucidated in detail. Finally, we provide insights into the future prospects of chemical upcycling for waste BPA-PC.

Keywords: Poly(bisphenol A carbonate), chemical upcycling, alcoholysis, aminolysis, high-value-added fine chemicals

INTRODUCTION

The annual global production of plastics has surpassed 460 million tons, yet only about 9% of waste plastics are recycled. Plastics have profoundly advanced human society, driving significant developments across various sectors^[1-3]. However, their accumulation, due to their stable and non-biodegradable nature, has led to serious environmental problems, including soil pollution, marine pollution, and climate change^[4-6]. Waste plastics are considered one of the major crises threatening the sustainable development. Consequently, the transition from the traditional “linear plastic economic model” to the promising “circular plastic economic model” has garnered extensive attention^[7]. Currently, plastic recycling methods are primarily categorized into mechanical recycling, energy recovery (incineration), and chemical recycling^[8,9]. Despite the widespread application of mechanical recycling and incineration for large-scale plastic waste treatment, these methods have critical drawbacks. Mechanical recycling results in materials with lower molecular weight and reduced thermal and mechanical performance compared to the virgin materials^[10,11], while incineration releases hazardous gases and microplastics^[12,13]. In contrast, chemical recycling of waste plastics into initial monomers or high-value chemicals^[14,15] with high resource utilization, economic benefits, environmental friendliness and quality controllability will be regarded as a significant driving force for the circular plastic economic model.

As one of the fastest-growing engineering plastics, poly(bisphenol A carbonate) (BPA-PC) boasts excellent optical properties, heat resistance, impact resistance, toughness and flame retardancy. These attributes make BPA-PC highly versatile, finding applications in electronics, electrical appliances, sheet containers, automotive industry, medical devices and protective equipment^[16]. It is projected that by 2024, the global demand for BPA-PC will reach 7.72 million tons^[17]. However, the extraordinary durability of BPA-PC leads to its persistent accumulation in the environment, gradually releasing harmful bisphenol A (BPA)^[18-20]. Therefore, the efficient chemical recycling of BPA-PC is imperative. The primary methods for chemical recycling of BPA-PC include pyrolysis, hydrolysis, reduction, alcoholysis, and aminolysis^[21-26]. Pyrolysis, characterized by random and non-selective bond breaking, results in complex products and requires high reaction temperatures with significant energy consumption, thereby increasing costs and reducing practicability^[27-29]. Additionally, carbonic acid, a by-product of BPA-PC pyrolysis, decomposes into carbon dioxide and water, complicating its reuse^[30,31]. Although high conversion rates can be achieved in BPA-PC reduction under suitable conditions, the requirement for high-pressure hydrogen necessitates equipment with substantial pressure-bearing capacity, limiting scalability^[32,33]. In contrast, employing alcohols or amines as nucleophilic reagents for depolymerization, such as in alcoholysis and aminolysis, presents a more promising alternative. These processes capture the carbonyl portion of polymers, generating high-value carbonyl products such as organic carbonates, carbamates and ureas^[34]. Thus, they offer greater practical and economic value and have garnered significant attention in recent years.

In this comprehensive review, a systematic summary for chemical upcycling of BPA-PC to BPA and high-value-added carbonate derivatives with various depolymerization reagents was presented. According to the depolymerization reagents and products structures, the main categorization was as follows: (1) Alcoholysis (methanolysis, ethanolysis and phenolysis glycolysis); (2) Aminolysis of BPA-PC and depolymerization of BPA-PC by other nucleophilic reagents; (3) Reuse of BPA-PC depolymerization monomers to synthesis of

new polymers. Moreover, we provide insights into the future potential and prospects of chemical upcycling for waste BPA-PC.

ALCOHOLYSIS OF BPA-PC

Methanolysis, ethanolysis and phenolysis of BPA-PC to linear carbonates

As shown in Figure 1, alcoholysis of BPA-PC with different structures of alcohols could obtain unified product BPA and corresponding carbonates. Dimethyl carbonate (DMC), diethyl carbonate (DEC), and diphenyl carbonate (DPC) could be produced with methanol, ethanol, and phenol as nucleophilic reagents for the chemical recycling of BPA-PC, respectively. This section is classified and summarized according to the types of catalysts.

Alkali metal catalysis

As early as 1998, Hu *et al.* initially investigated the alkali-catalyzed methanolysis of BPA-PC, which found that only 7% yield of BPA was obtained catalyzed by the NaOH (8.5 mol% relative to carbonate units of BPA-PC) in methanol at 60 °C after 330 min. However, the depolymerization rate was significantly accelerated when toluene or dioxane was added as a co-solvent. For example, in a mixed solvent of MeOH (1 mL) and toluene (1 mL), BPA-PC was completely depolymerized by a similar treatment for 70 min, and the products BPA (96%) and DMC (100%) were obtained in high yields. The reaction time could be further shortened by increasing the amount of the co-solvent. Since BPA-PC is virtually insoluble in methanol, a plausible reason for the accelerated action of co-solvents is that they can dissolve or solubilize the surface of solid BPA-PC and promote contact between reaction reagents, thereby increasing the rate of methanolysis^[35]. Subsequently, Piñero *et al.* developed a greener process for alkali-catalyzed methanolysis of BPA-PC under supercritical or near-critical conditions in a semi-continuous laboratory plant^[36]. The relationship between operating conditions such as reaction temperature, pressure, alcohol/co-solvent ratio, and catalyst concentration with product yield, selectivity, and reaction rate were systematically investigated. The best yields of BPA and DMC were achieved at the conditions of 120–140 °C, 10 MPa, and 1.5–2 kg/m³ NaOH/pure methanol, while the reaction selectivity and yield of the products were reduced with less toxic water used as the co-solvent. Chiu *et al.* used a batch autoclave reactor to explore the methanolysis of BPA-PC. It was found that under high temperature and subcritical conditions, BPA-PC could also be effectively depolymerized without adding a catalyst^[37]. Under the optimal reaction conditions, when the molar ratio of MeOH/BPA-PC was 37.5 at 240 °C, it only took 5 min for complete methanolysis of BPA-PC, with the yields for BPA and DMC being 90.7% and 95.2%, respectively. Furthermore, Kim *et al.* revealed that increasing the MeOH/BPA-PC molar ratio to 71 in the temperature range of 160–220 °C without catalysts and toxic solvents resulted in higher BPA and DMC yields, but this further increased the energy and cost requirements. When the ethylene glycol (EG)/MeOH co-solvent system is used, the depolymerization yield does not grow^[38].

The monomer BPA and DPC would also be obtained with phenol as a depolymerization reagent catalyzed by alkali metal halides, such as potassium fluoride. The product yields could reach as high as 90% and the turnover frequency (TOF) value was as high as 1,392 h⁻¹. Moreover, the mixture of BPA, DPC and phenol was verified to be the starting point of the polymerization to produce new BPA-PC, thus completing the closed cycle^[39].

Ionic liquid catalysis

Despite the above progress, strong alkali-base catalysts or high temperature and pressure are inevitable. It is desirable to explore more mild and efficient catalytic systems. As the pioneer of ionic liquid catalytic depolymerization, Liu *et al.* developed a new process for BPA-PC methanolysis using ionic liquids as

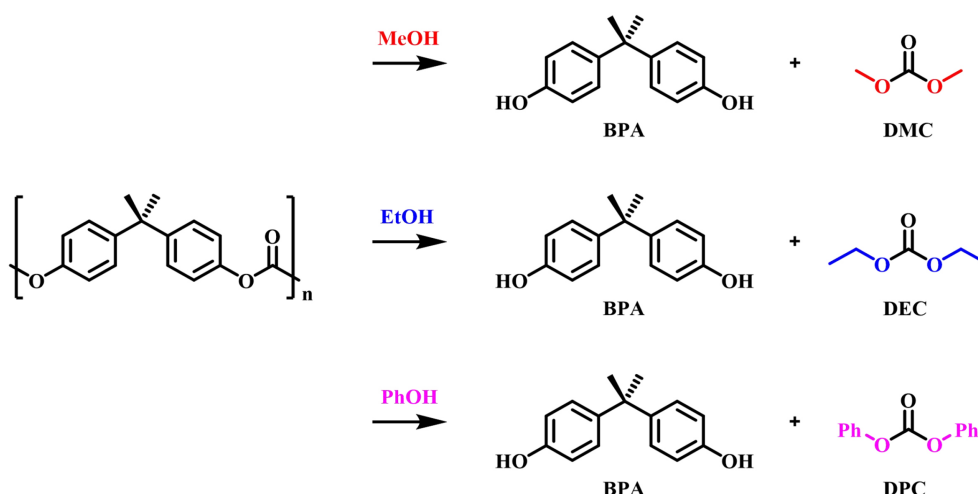


Figure 1. Methanolysis, ethanolysis and phenolysis of BPA-PC. BPA-PC: Poly(bisphenol A carbonate).

catalysts^[40,41]. Under the specified conditions, the yields of BPA and DMC exceeded 95% after 2.5 h using the ionic liquid 1-butyl-3-methylimidazolium chloride ([Bmim][Cl]) or 1-butyl-3-methylimidazolium acetate ([Bmim][Ac]) as a catalyst. Moreover, the ionic liquid could be reused 6-8 times with almost no loss of reactivity. This work avoided the use of toxic transition metal ions and organic solvents, providing a green and promising choice for the practical recycling of BPA-PC wastes.

Organic base catalysis

Organic catalysts are considered as metal-free eco-friendly catalysts. 1,8-Diazabicyclic [5.4.0] undecan-7-ene (DBU), as an organic catalyst, was first employed for the alcoholysis of BPA-PC^[42]. Other organocatalysts, such as 1,4-diazobicyclic [2.2.2] octane (DABCO) and 4-(dimethylamino) pyridine (DMAP), were less active than the amidine base DBU. The study was extended to EtOH and EtOH/MeOH mixtures. The ethanolysis reaction yielded BPA and DEC in quantitative yields, whereas in MeOH/EtOH solutions, mixed products of BPA and carbonates [DMC, DEC, methyl ethyl carbonate (MEC)] were obtained. Besides, Alberti and Enthaler studied the methanolysis of BPA-PC by microwave-assisted heating in the presence of catalytic amounts of DMAP^[43]. Excellent activities with a TOF of up to $1,164 \text{ h}^{-1}$ were observed at 180°C .

Zinc catalysis

In recent years, eco-friendly zinc-based catalysts were found to have high activity for alcoholysis as well. Yang *et al.* have successfully achieved the selective depolymerization of BPA-PC and other commercial plastics under mild conditions by utilizing the bis(trimethylsilyl) amino zinc ($[\text{Zn}(\text{HMDS})_2]$) as multifunctional catalyst and methanol as the depolymerization reagent^[44]. Payne *et al.* also prepared a series of Zn^{II} -complex catalysts based on half-salan ligands, which were successfully applied for the rapid methanol depolymerization (12-18 min) of BPA-PC even at room temperature and low catalyst loading conditions^[45].

Compared with various catalysts mentioned above, Alkali and Zn-based catalysts have excellent reactivity, organic catalysts avoid the use of metals, and ionic liquid catalytic depolymerization processes provide a green and sustainable pathway for the practical recycling of BPA-PC wastes.

Glycolysis of BPA-PC to functionalized cyclic carbonates

Oku *et al.* investigated the reuse of waste BPA-PC as a chemical reagent using EG as a depolymerization reagent catalyzed by NaOH (0.1 equiv.) at 180 °C, yielding monohydroxyethyl ether of BPA (MHE-BPA, 42%), bishydroxyethyl ether of BPA (BHE-BPA, 11%), and BPA (42%) [Figure 2]^[46]. They concluded that ethylidene carbonate (EC) is an intermediate in the reaction which would react with the simultaneously formed BPA to obtain the final products. It was also found that quantitative conversion of BPA-PC to BHE-BPA was achieved within 20 min by adding additional EC (1.6 mol equiv.) to the same reaction system. Subsequently, Kim *et al.* further explored the non-catalyzed glycolysis of BPA-PC^[47]. Unexpectedly, only BPA was found in the glycolysis product, which implied that EC did not react with BPA under non-catalytic conditions, but rapidly broken down into linear carbonates to produce CO₂. Through the optimization of the reaction conditions, they found that BPA could achieve 95.6% of the maximum yield for only 85 min with weight ratio of EG/BPA-PC = 4, at 220 °C. To achieve efficient solvent-free glycolysis of BPA-PC, Nifant'ev *et al.* systematically studied the catalytic effects of acetate salts such as Li, Na, K, Mg, Ca and Zn on glycolysis of BPA-PC^[48]. In the presence of Mg(OAc)₂ (0.5 mol%, reaction for 15 min) or Zn(OAc)₂ (0.1 mol%, reaction for 60 min) at 180 °C, BPA and EC were obtained with high yields of 90%, 98% and 96%, 98%, respectively. This method combined BPA-PC recycling to BPA with recycling to cyclic carbonate using a simple, low-cost, non-toxic metal catalyst, and is a promising chemical recycling strategy for waste polycarbonate.

Functionalized cyclic carbonates are important intermediates in the manufacture of fine chemicals; propylene carbonate and vinyl carbonate, as representative cyclic carbonates, are widely used as polar non-protonic solvents and electrolytes in batteries^[49,50]. Therefore, increasing attention has been paid to the conversion of BPA-PC to BPA and functionalized cyclic carbonates, since the carbonyl portion of BPA-PC can be used as an equivalent reagent instead of toxic CO and phosgene conforming to the goals of “green chemistry” and “atom economy”.

The pioneering work was done by Hata *et al.* in 2003; they treated BPA-PC with nucleophiles such as alkanedithiols, mercaptoethanol, aminoethanethiol, and aminoethanol in the presence of catalytic amounts of NaOH to obtain BPA and corresponding cyclic heterocarbonates [1,3-dithiopentyl-2-one (DTO) and 1,3-dithiane-2-one (DTA)] with high yields as shown in Figure 3^[34]. It was proved that BPA-PC can be used as a phosgene equivalent for industrial purposes. In 2009, they continued to study the alcoholysis of BPA-PC by biomass-based polyols such as glycerol (Gly) and glucose (Glu) [Figure 4]. In addition to obtaining BPA, cyclic carbonates hydroxymethyldioxolane (HMDO) and dicarbonate (Glu-DC) were produced in 97% and 40% yields, respectively^[51].

In addition, researchers have successively developed a variety of efficient catalytic systems. For example, in 2017, Iannone *et al.* developed a bifunctional acid/base catalyst based on the composition of nanostructured zinc oxide and tetrabutylammonium chloride (ZnO-NPs/NBu₄Cl), which enabled efficient chemical recovery of BPA-PC in 7 h at 100 °C^[52]. Diverse nucleophilic reagents, including water, alcohols, amines, polyols, aminols and polyamines, were universally applicable, and a group of high-value compounds, such as functionalized carbonates and ureas were obtained [Figure 5]. The catalyst was proved to be easy to recover and could be recycled many times without obvious decay of reactivity. However, a strong excess of the nucleophilic reagent was essential. In 2018, Quaranta *et al.* further studied the glycolysis reaction catalyzed by DBU based on their previous work^[53]. The high-yield BPA monomer and the corresponding cyclic carbonate were smoothly obtained by depolymerizing BPA-PC waste with 1,2-propylene glycol or Gly. Do *et al.* also compared a series of transesterification catalysts for the alcoholysis of BPA-PC, and they found that 1,5,7-triazabicyclo[4.4.0]-dec-5-ene (TBD) exhibited the best performance, which led to the successful synthesis of a series of high-value-added organic carbonates at only 30 °C [Figure 6]^[54].

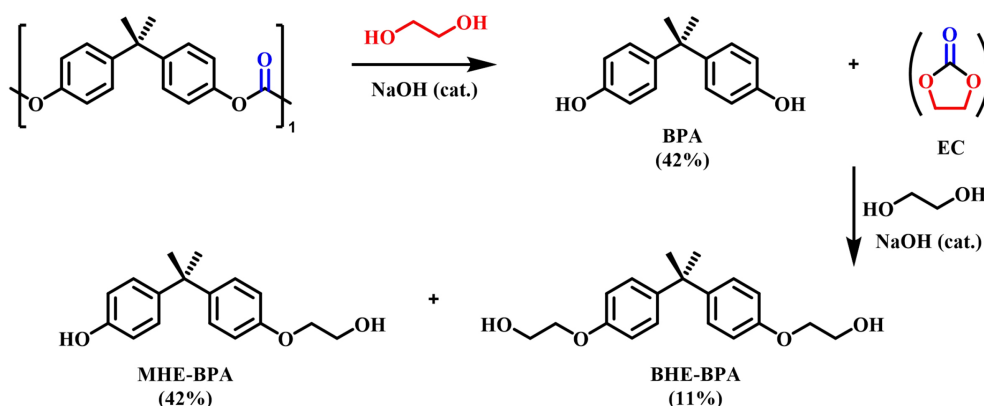


Figure 2. Glycolysis of BPA-PC (isolated yield). BPA-PC: Poly(bisphenol A carbonate).

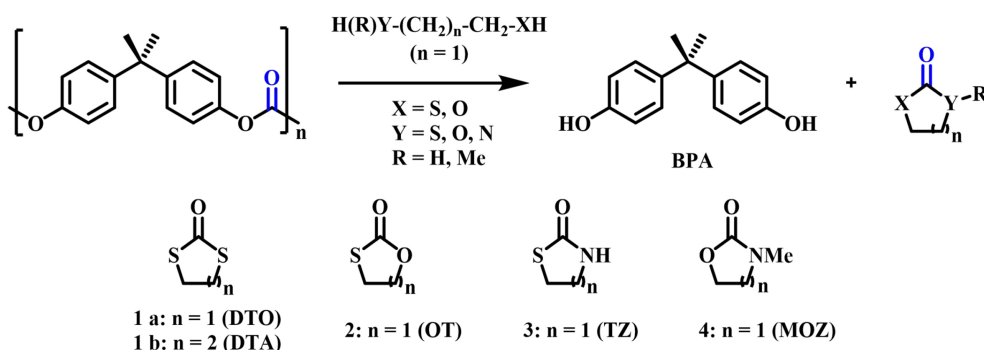


Figure 3. Depolymerization of BPA-PC by alkanedithiols, mercaptoethanol, aminoethanethiol, and aminoethanol. BPA-PC: Poly(bisphenol A carbonate).

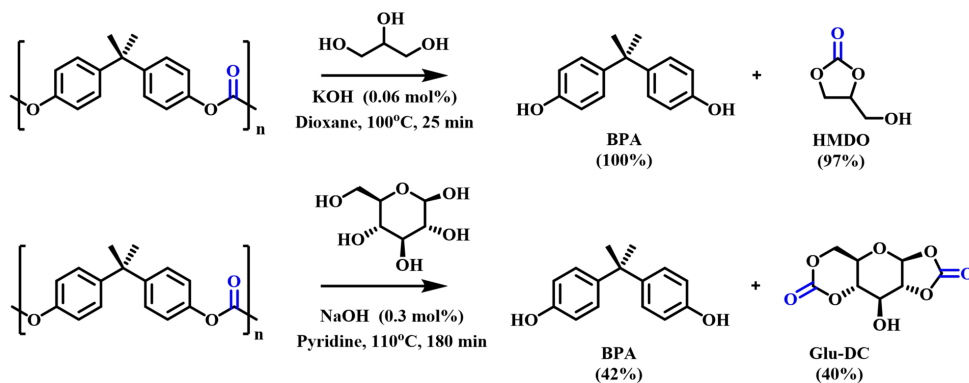


Figure 4. Alcoholysis of BPA-PC by glycerol (Gly) and glucose (Glu) (isolated yield). BPA-PC: Poly(bisphenol A carbonate).

Jehanno *et al.* had new progress in 2020 using protic ionic salt TBD: methanesulfonic acid (MSA) as a catalyst to obtain five-membered heterocyclic monomers and six-membered cyclic carbonates with good to excellent yields without solvents [Figure 7]^[55]. In 2021, they successfully subjected this catalyst to the selective and sequential depolymerization of mixed plastics such as BPA-PC and polyethylene terephthalate (PET) under solvent-free conditions and high temperatures (100–160 °C)^[56]. In 2023, Olazabal *et al.* further achieved the complete depolymerization of BPA-PC using 1-methylimidazole as a solvent and imidazole as

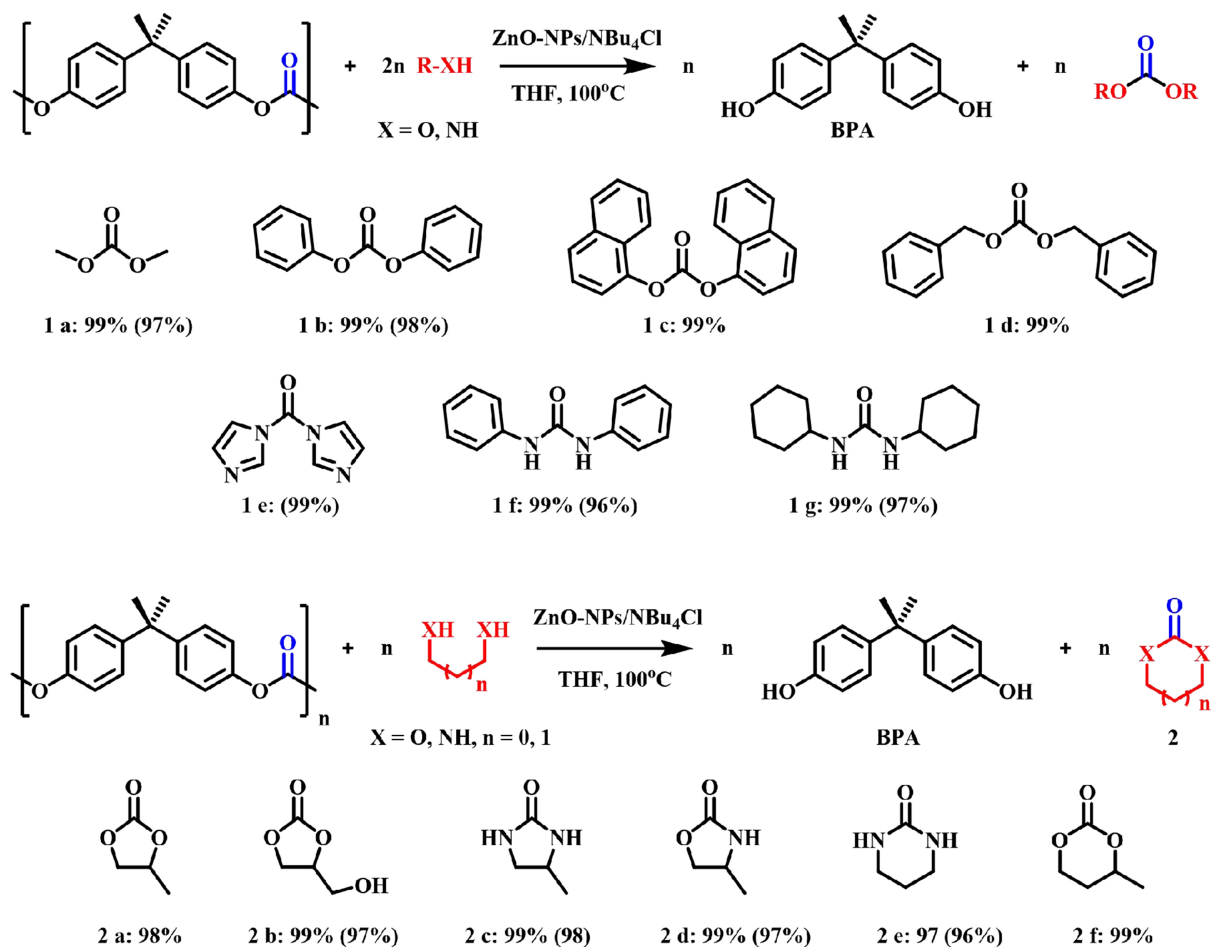


Figure 5. ZnO-NPs/NBu₄Cl catalyzed depolymerization of BPA-PC (GLC yield; in brackets, isolated yields). BPA-PC: Poly(bisphenol A carbonate).

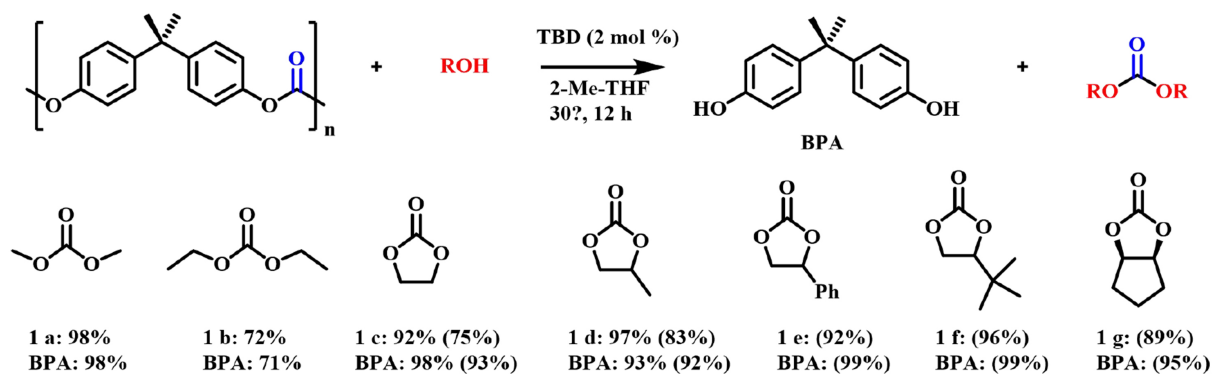


Figure 6. TBD-catalyzed alcoholysis of BPA-PC. (NMR yields; numbers in parenthesis are isolation yields). TBD: 1,5,7-triazabicyclo[4.4.0]-dec-5-ene; BPA-PC: poly(bisphenol A carbonate).

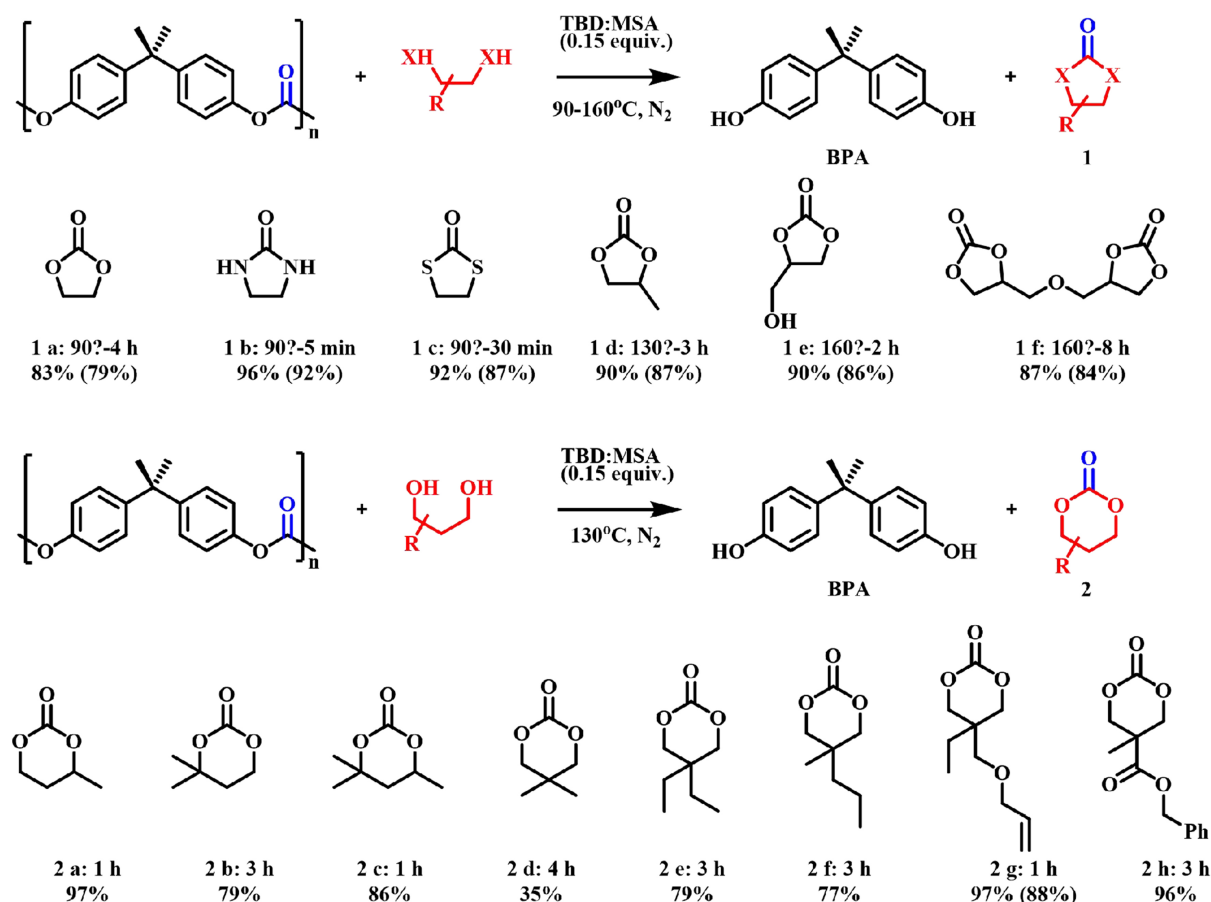


Figure 7. TBD: MSA-catalyzed depolymerization of BPA-PC (NMR yields; numbers in parenthesis are isolation yields). TBD: 1,5,7-triazabicyclo[4.4.0]-dec-5-ene; MSA: methanesulfonic acid; BPA-PC: poly(bisphenol A carbonate).

a catalyst at 50 °C for 3 h, obtaining high trimethylene carbonate (TMC) yields (81%) [Figure 8]^[57]. This method was also applied to other nucleophilic reagents, resulting in the synthesis of a number of different cyclic carbonyl-containing intermediates.

AMINOLYSIS OF BPA-PC AND DEPOLYMERIZATION OF BPA-PC BY OTHER NUCLEOPHILIC REAGENTS

In addition to alcohols, other nucleophiles such as amines have also been studied for the depolymerization of BPA-PC. Hata *et al.* initially investigated the aminolysis of BPA-PC with dialkylamines in 2002^[58]. However, the aminolysis process using monoamines such as dimethylamine and diethylamine was not successful, giving BPA monomers and other oligomeric carbamate intermediates low yields. When diamine was used instead of monoamines, the reaction proceeded smoothly to give BPA and cyclic urea derivatives such as 1,3-dimethyl-2-imidazolidinone (DMI), and the six-membered cyclic urea 2,6-dimethyl-2,6-diazacyclohexanone (DMDACH) in almost quantitative yields as shown in Figure 9. The aminolysis of carbamates with monoamines in the second step proceeds much more slowly than the cleavage of the carbonate group in the first step. However, with diamines, the intramolecular cyclization in the second step provides a kinetic advantage, thereby promoting the conversion to urea. In 2015, Singh *et al.* reported an unprecedented green method to react BPA-PC with primary amines to generate urea derivatives^[59]. The carbonate moiety in BPA-PC was successfully extracted at 80 °C to prepare useful urea derivatives in

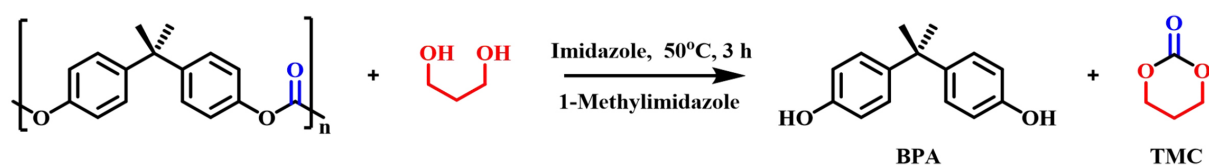


Figure 8. Imidazole-catalyzed alcoholysis of BPA-PC. BPA-PC: Poly(bisphenol A carbonate).

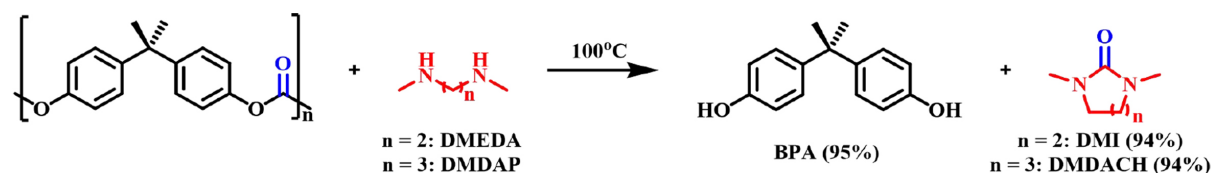


Figure 9. Depolymerization of BPA-PC by DMEDA, DMDAP, (NMR yields). BPA-PC: Poly(bisphenol A carbonate); DMEDA: N,N'-dimethylethylenediamine; DMDAP: N,N'-dimethyl-1,3-propanediamine.

moderate yields, without the use of any catalysts or toxic organic solvents [Figure 10].

Recently, Wang *et al.* proposed an “amino-alcoholysis” strategy for upcycling BPA-PC plastic waste to BPA monomers and high-value-added chiral 2-oxazolidinone chemicals via ZnX_2 -catalyzed depolymerization of chiral amino alcohols under mild conditions [Figure 11]^[60]. In addition, high yields and selectivity were achieved in the sequential depolymerization of BPA-PC/PET mixed plastics. Using dimethyl sulfoxide (DMSO) as a strong polar solvent, the reaction can proceed smoothly without the necessity for a catalyst^[61]. More importantly, BPA-PC could be efficiently depolymerized under solvent-free conditions with only one equivalent of depolymerizing agent. This “amino-alcoholysis” strategy was proved to be suitable for different types of commercial BPA-PC, and the feasibility of the method for practical application was further verified by scaling up the reaction. The recovered high-purity BPA was repolymerized to prepare new polymers, thus realizing closed-loop recycling. In addition, oxazolidinone compounds with different structures were obtained, demonstrating the generalizability of the method, which is important for plastic recycling and economic sustainability.

Jung *et al.* used hydroxamic acid as a nucleophilic reagent for TBD-catalyzed depolymerization of BPA-PC to obtain a series of dioxazolone^[62]. The resulting dioxazolones could be subjected to one-pot C-H amidation to generate amides, which are important synthetic building blocks. As shown in Figure 12, amides 3a-3e were obtained in yields in the range of 69%-79%, while furanoic acid-derived dioxazolone showed low yields in transforming into amide 3f and BPA. This protocol promoted a green and sustainable process using BPA-PC as a carbonyl source instead of the toxic phosgene or 1,1'-carbonyldiimidazole (CDI) reagent in dioxazolone synthesis.

The chemical depolymerization of BPA-PC using α -hydroxyketone as a nucleophilic reagent was performed by Onida *et al.* in 2023^[63]. The reaction was catalyzed by TBD in 2-Me-THF at room temperature to give higher value-added vinylidene carbonate in moderate to high yields (30%-97%, 13 instances) [Figure 13]. The success of TBD was attributed to its ability to activate both α -hydroxyketone and the carbonyl group of BPA-PC via hydrogen bonding. The practicality of the method was demonstrated by further scale-up experiments.

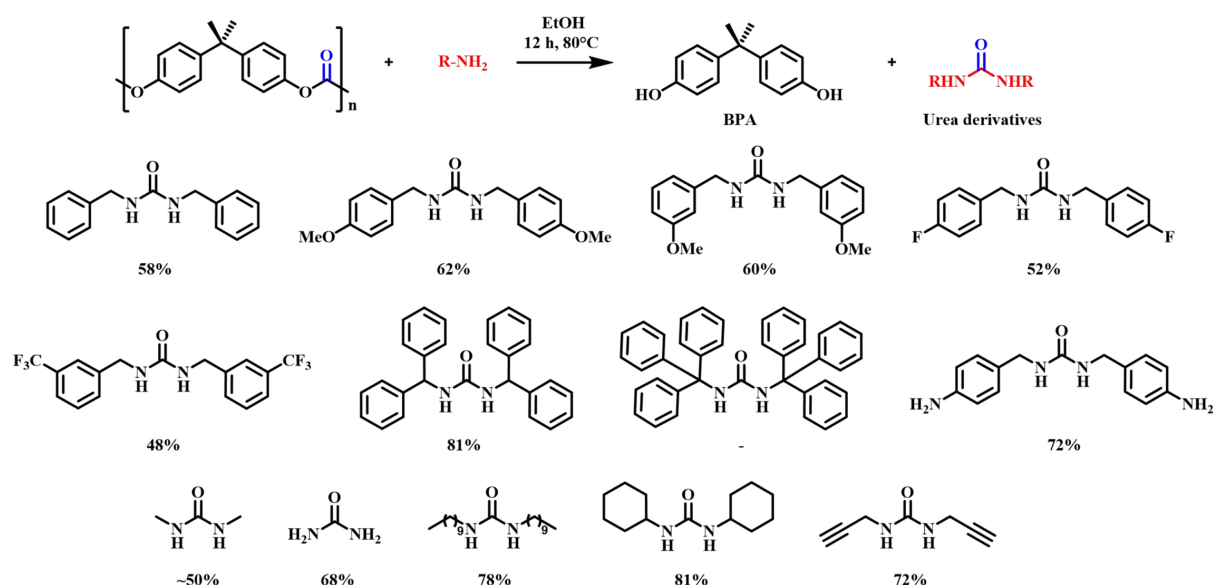


Figure 10. Depolymerization of BPA-PC by primary amines (isolated yield). BPA-PC: Poly(bisphenol A carbonate).

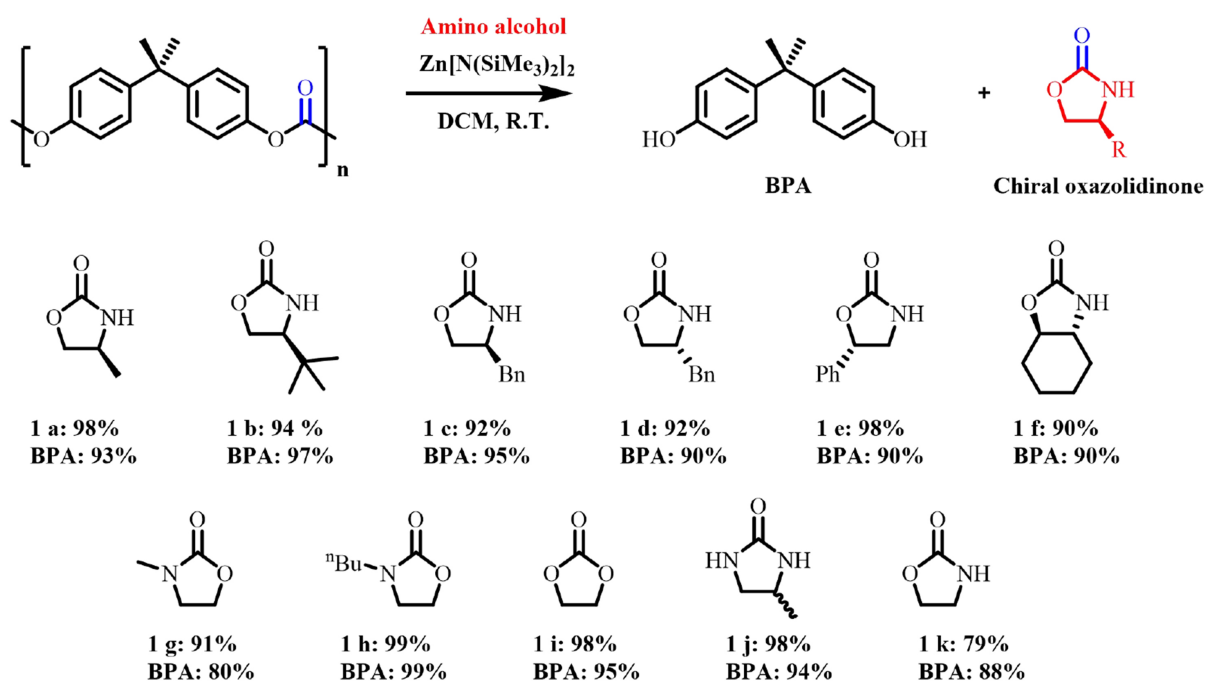


Figure 11. Depolymerization of BPA-PC by amino alcohols (NMR yields). BPA-PC: Poly(bisphenol A carbonate).

REUSE OF BPA-PC DEPOLYMERIZATION MONOMERS TO PRODUCE NEW POLYMERS

The efficient depolymerization of BPA-PC plastics through alcoholysis, aminolysis and other methods has resulted in a variety of multifunctional monomer products which could be utilized as raw materials for the synthesis of new polymer materials. In this regard, the use of hydroxyethyl ether products of BPA as rigid copolymerization monomers has attracted attention. Oku *et al.* have shown that the addition of additional EC is the key to the formation of bis(hydroxyethyl) ether (BHEEB) during glycolysis of BPA-PC in the

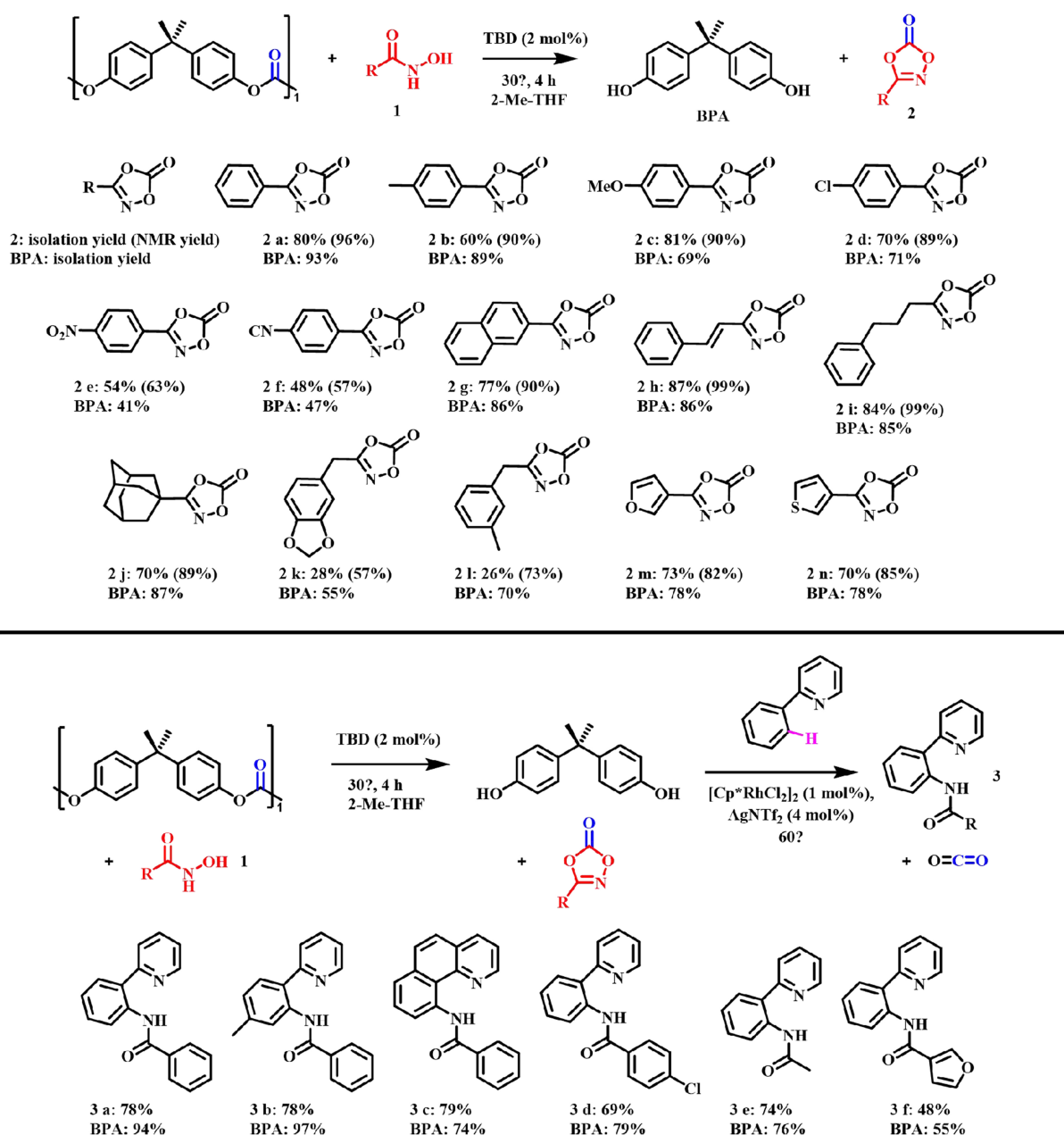


Figure 12. Depolymerization of BPA-PC by hydroxamic acid and one-pot C-H amidation. (isolation yields; numbers in parenthesis are NMR yields). BPA-PC: Poly(bisphenol A carbonate).

presence of NaOH. Despite the fact that a number of inexpensive synthetic routes for EC have been developed, EC is not yet a commercial product, which restricts the industrial application of the method^[46]. Instead, Lin *et al.* further developed a one-pot atmospheric pressure alkoxylation process based on the studies of Oku *et al.*^[46,64]. On the basis of the completion of the first step in depolymerization, catalytic ZnO and cheap urea were added to the reaction mixture as carbonylation agents, and cyclic carbonates were generated in situ from EG or propylene glycol, which could be used to complete the ethoxylation or propoxylation process. Finally, bishydroxyethyl ether of BPA (BHE-BPA) or bishydroxypropyl ether of BPA (BHP-BPA) was generated as the final product. In comparison, this method offers higher selectivity and yields for the BPA-based diol (more than 90%). BHE and BHP products have been successfully applied as

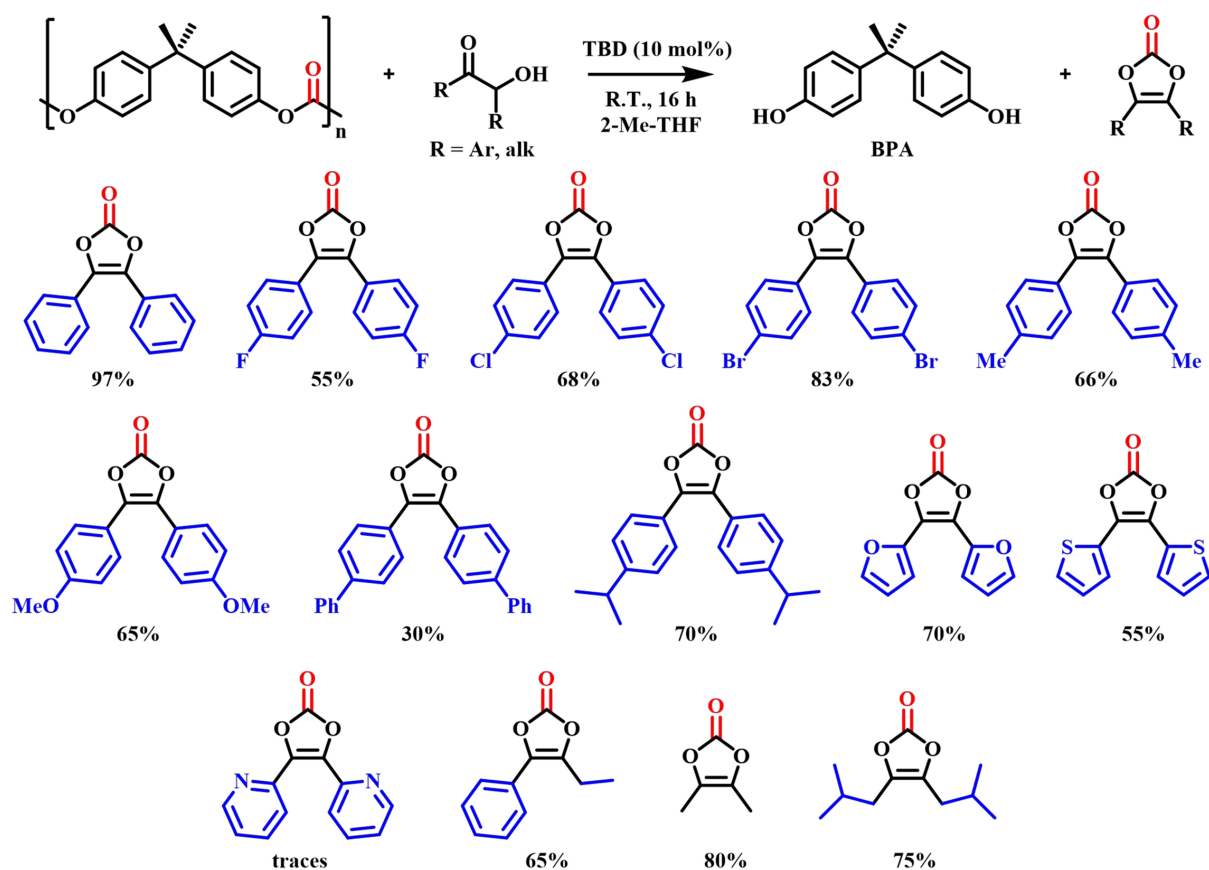


Figure 13. Depolymerization of BPA-PC by α -hydroxyketone (isolated yields). BPA-PC: Poly(bisphenol A carbonate).

important raw materials in the preparation of high molecular weight polyurethane (PU) elastomers and plastics with excellent mechanical properties. In addition, Berti *et al.* have shown that it is possible to insert BPA groups into the polyester chain by reacting with BHEEB, resulting in terephthalate polyesters with higher mechanical properties and thermal stability^[65].

In 2018, Pang *et al.* successfully achieved the synthesis of a series of BPA-PC homopolymers and copolymers using the chemical recycling of BPA-PC and a novel bicyclic diol octahydro-2,5-pentalenediol (OPD) deriving from naturally-occurring citric acid [Figure 14]^[66]. It was found that BHEEB not only compensated for the low reactivity of OPD, but also improved the brittleness of OPD-based polycarbonates without affecting other properties.

Gioia *et al.* further optimized the synthesis process of BHEEB [Figure 15], with an environmentally friendly catalyst K_2CO_3 ^[67]. Subsequently, purified BHEEB was polymerized with succinic acid (SA) and isosorbide (IS) which were derived from renewable sources. In the presence of a tin-based catalyst (MTBO), low molecular weight polyesters were obtained with properties suitable for coatings applications [Figure 15]. Furthermore, a second route was developed for recycling BPA-PC [Figure 15]. BPA-PC reacted with vinyl carbonate via selective decarboxylation to produce polyether-carbonate, followed by partial depolymerization using diols to further reduce its relative molecular weight while increasing its hydroxyl group number. The oligomer is also suitable for powder coating applications.

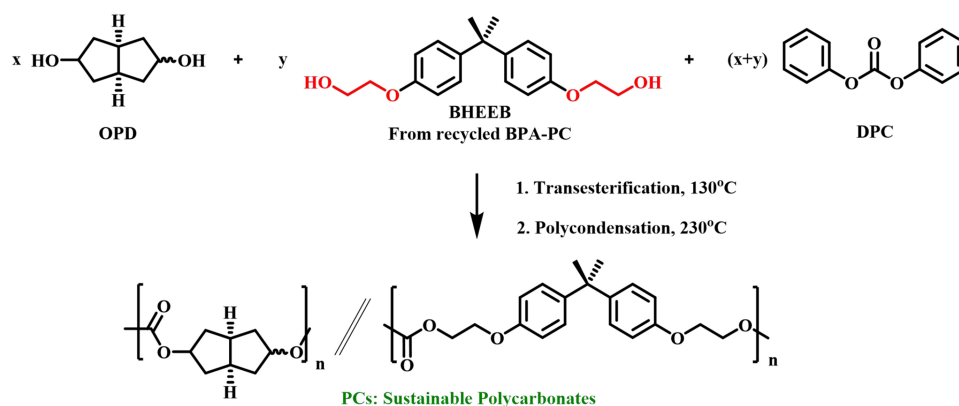


Figure 14. Conversion of BPA-PC to sustainable polycarbonate. BPA-PC: Poly(bisphenol A carbonate).

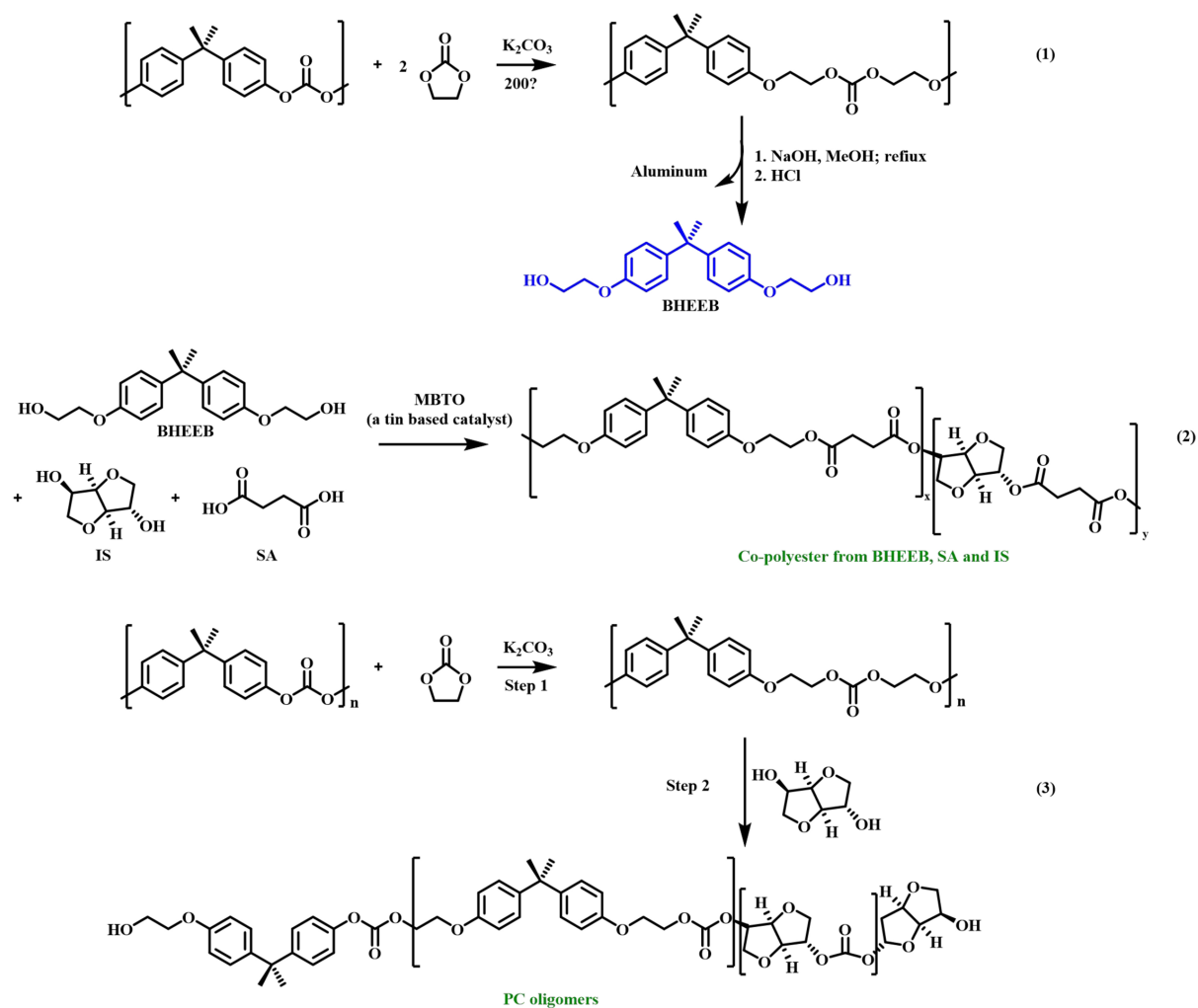


Figure 15. Conversion of BPA-PC into novel polymers for powder coating applications. BPA-PC: Poly(bisphenol A carbonate).

Saito *et al.* used diols with different chain lengths to depolymerize BPA-PC into new diols containing carbonates, such as bis(3-hydroxypropyl) carbonate (3C), bis(4-hydroxybutyl) carbonate (4C) and bis(5-

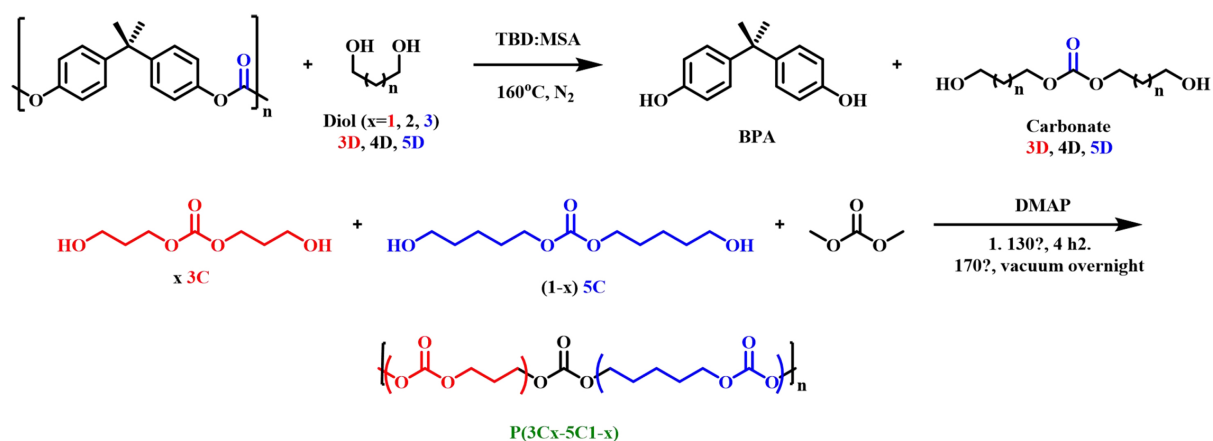


Figure 16. Conversion of BPA-PC to polymer electrolytes for batteries. BPA-PC: Poly(bisphenol A carbonate).

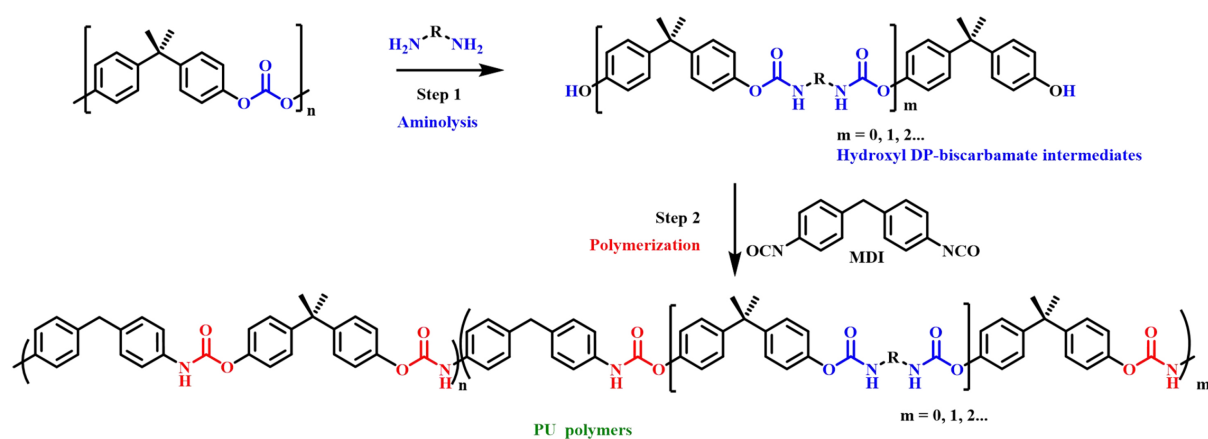


Figure 17. Conversion of BPA-PC into multifunctional intermediates to produce various PU polymers. BPA-PC: Poly(bisphenol A carbonate); PU: polyurethane.

hydroxypentyl) carbonate (5C); they could be further applied to obtain a group of value-added aliphatic polycarbonates through organic catalyzed polycondensation [Figure 16]^[68]. These new polymers showed promising ionic conductivity values that could serve as sustainable polymer electrolytes for solid-state batteries. Wu *et al.* investigated the conversion of BPA-PC into multifunctional intermediates by depolymerization of BPA-PC with aliphatic diamines in the absence of catalysts under mild conditions [Figure 17]^[69]. The first step of aminolysis offered hydroxyl-terminated short-chain extenders or prepolymers. Subsequently, a variety of PU polymers were synthesized in a one-pot by adding reagents such as diisocyanates. The process achieved 100% atom economy efficiency. Quaranta *et al.* reacted BPA-PC with a long-chain diamine to provide the monomer BPA (83%-95%, isolated yield) and successfully converted the waste BPA-PC to polyurea [Figure 18]. This is a more environmentally friendly method of synthesizing polyurea without the use of isocyanates^[70]. The above successful examples demonstrated that BPA-PC can be used as a feedstock for the synthesis of new functional polymers with different properties.

Other studies of recycling BPA-PC to synthesize bio-based polycarbonates have also been reported. For example, in 2021, De Leo *et al.* achieved quantitative conversion of BPA-PC into BPA and DPC using zinc acetate and ionic liquids as catalysts and phenol as a nucleophilic reagent. The latter was subjected to a melt transesterification reaction with curcumin (CM) from natural polyphenols or its tetrahydrogenated

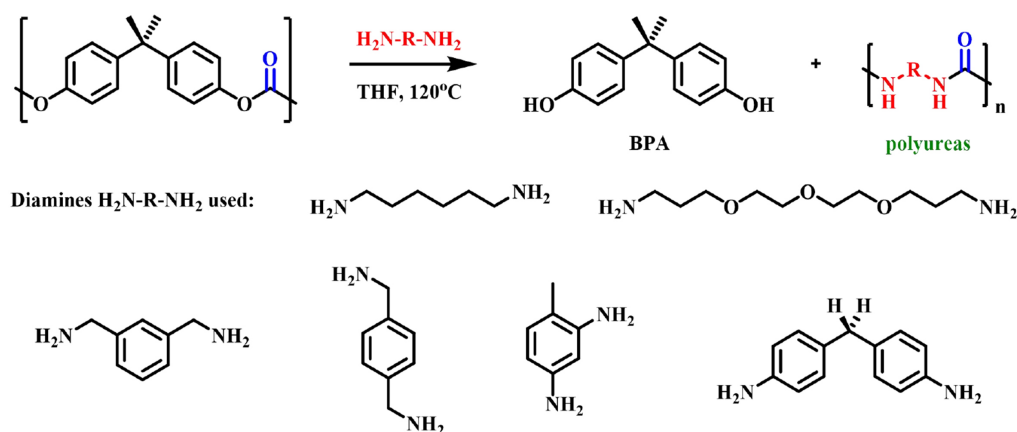


Figure 18. Reuse of BPA-PC as polyureas. BPA-PC: Poly(bisphenol A carbonate).

colorless product (THCM) to obtain the corresponding bio-based polycarbonates CM-PC and THCM-PC, respectively [Figure 19]^[71]. An innovative study was carried out by Saito *et al.* in 2022. BPA-PC was upcycled with biobased vanillin derivatives into novel dialdehyde monomers (DVEC), which could be transformed into poly(imide-carbonate) [P(ImC)] by polymerization with primary amines as shown in Figure 20^[72]. P(ImC) showed full recyclability under acidic conditions, thus enabling closed-loop recycling. It is noteworthy that the P(ImC) prepared using different diamines and triamines [tris(2-aminoethyl)amine (TREN)] as starting materials has readily tunable thermal and mechanical properties, which have broader application prospects.

CONCLUSION AND OUTLOOK

As summarized in this review, chemical recovery strategies for alcoholysis, aminolysis or other nucleophiles attack on BPA-PC have been widely studied. In order to achieve complete conversion of the polymer, researchers have systematically explored reaction conditions such as solvents, temperatures, pressures, concentrations of depolymerization reagents, types and loading of catalysts. However, in the early research, these processes generally have some drawbacks such as the use of high toxicity and environmental organic solvents or transition metal catalysts, harsh reaction conditions and large energy consumption. Therefore, increasing green and sustainable BPA-PC depolymerization processes have been developed in recent years via efficient and low-cost non-toxic metal catalysts, organic catalysts and ionic liquid catalysts, and green solvent systems. In addition, milder and greener catalyst-free or solvent-free reaction conditions have also been reported. Based on the development of BPA-PC upcycling technologies, the regeneration of new polymer materials from monomer products obtained through the BPA-PC upcycling was also achieved, indicating its potential practical values.

Although methanolysis can effectively yield BPA and DMC, the low boiling point of DMC makes it prone to mixing with residual reaction by-products, necessitating the use of multi-stage distillation or solvent extraction for separation. This complex separation process may limit the scalability of industrial applications. Compared with methanolysis, DEC obtained via ethanolysis has a higher boiling point, making separation relatively easier. However, due to potential azeotrope formation between DEC and by-products, precise distillation equipment is still required for effective separation. Given that aminolysis yields various amino derivatives, traditional separation methods may require multiple extractions or column chromatography techniques. To enhance separation efficiency, exploring continuous flow synthesis systems or membrane separation technology could reduce cross-contamination of products.

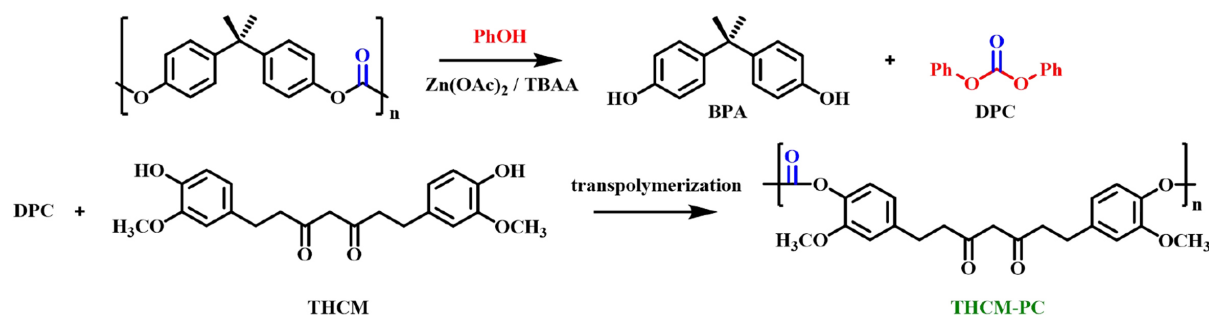


Figure 19. Conversion of BPA-PC to bio-based polycarbonate THCM-PC. BPA-PC: Poly(bisphenol A carbonate).

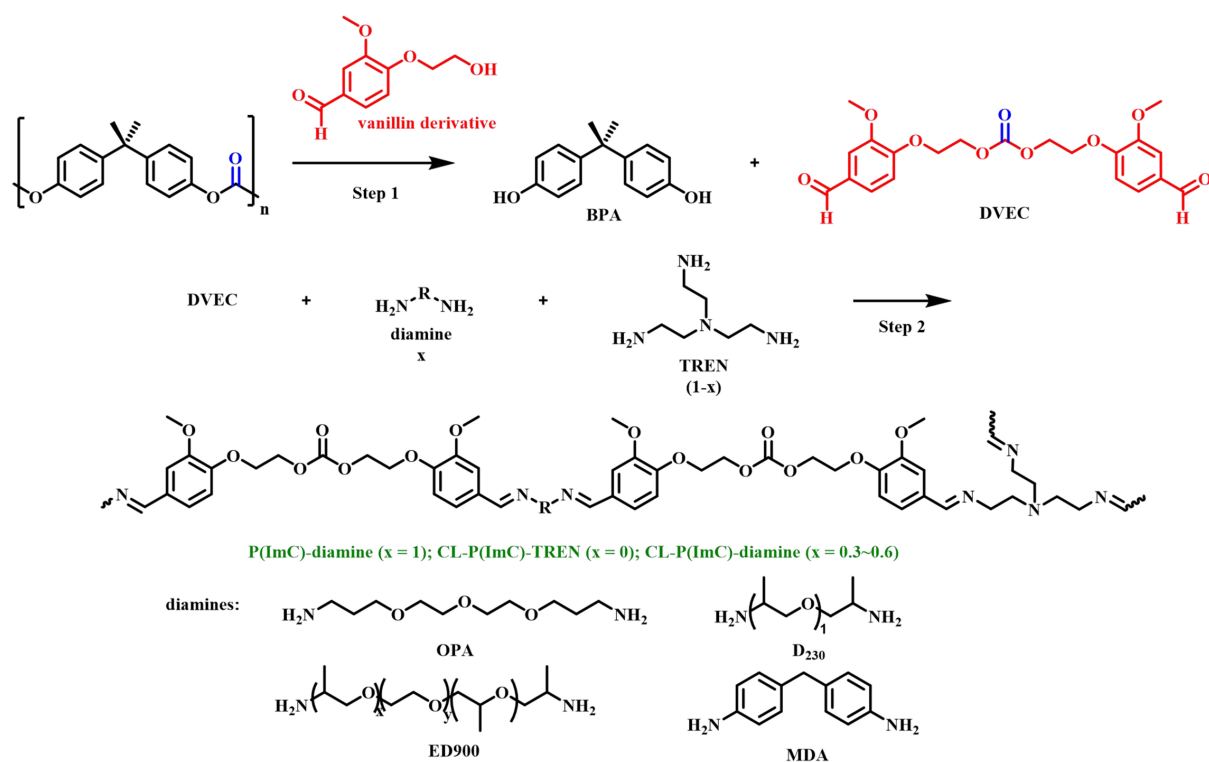


Figure 20. Conversion of BPA-PC to bio-based polycarbonate P(ImC). BPA-PC: Poly(bisphenol A carbonate).

In the future, efficient and low energy consumption depolymerization technology is an important trend, which needs more efficient catalysts and sustainable depolymerization processes. Except for the homogeneous catalysis depolymerization of BPA-PC, heterogeneous catalysis processes have the advantage of allowing the catalyst to be reused several times, but they also exhibit low reactive activity. Meanwhile, the sustainable processes are also imperative demands for the industrialization of BPA-PC depolymerization. Moreover, although chemical catalysts play a crucial role in depolymerization reactions, the challenges of product separation and purification must be addressed to achieve large-scale industrial implementation of these methods. Future research should focus more on these aspects to develop cost-effective and sustainable recycling processes.

DECLARATIONS

Authors' contributions

Proposed the topic of this review: Song T, Xu G

Prepared the manuscript: Li S, Zhu G, Yang R

Revised the manuscript: Hou H, Song T, Wang Q

Availability of data and materials

Not applicable.

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Conflicts of interest

All authors declared that there are no conflicts of interest.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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