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Renewable Polymers

Closed-Loop Recycling of Poly(Imine-Carbonate) Derived from Plastic Waste and Bio-based Resources

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Dedicated to Professor Klaus Müllen and Professor Ivan Gutman on the occasions of their 75th birthdays

Abstract: Closed-loop recycling of polymers represents the key technology to convert plastic waste in a sustainable fashion. Efficient chemical recycling and upcycling strategies are thus highly sought-after to establish a circular plastic economy. Here, we present the selective chemical depolymerization of polycarbonate by employing a vanillin derivative as bio-based feedstock. The resulting di-vanillin carbonate monomer was used in combination with various amines to construct a library of reprocessable poly(imine-carbonate)s, which show tailor-made thermal and mechanical properties. These novel poly(imine-carbonate)s exhibit excellent recyclability under acidic and energy-efficient conditions. This allows the recovery of monomers in high yields and purity for immediate reuse, even when mixed with various commodity plastics. This work provides exciting new insights in the design of bio-based circular polymers produced by upcycling of plastic waste with minimal environmental impact.

Introduction

Recycling of polymeric materials is a major scientific challenge of modern society as the ever-increasing production of plastic goods and their predominant linear use cause severe environmental burden.[1,2] Since global recycling rates remain low,[3] most of the plastic waste is either incinerated or disposed of in landfills. Thus, creating a circular economy for plastics is an auspicious strategy to counteract the current take-make-waste consumption model.[3–9] Closed-loop recycling also provides ample opportunities to conserve the vast volume of chemical resources that are imbedded in synthetic macromolecules. To this end, chemical recycling which encompasses the controlled cleaving of chemical bonds in polymers to recover the monomeric species in high yields represents a sustainable method to produce fresh polymers without loss of quality and resources.[10–16] This strategy additionally allows for the generation of value-added monomers from traditional polymers and thus provides access to upcycled polymeric materials with superior properties.[5,7,17]

Generally, polymers decorated with functional groups, such as esters and carbonates, are intrinsically more suitable for selective depolymerizations.[1] Chemical recycling of Bisphenol A polycarbonate (BPA-PC), a commodity polymer with widespread applications, is of particular interest.[18] In stark contrast to conventional plastic waste management methods, it enables the controlled capture of Bisphenol A which poses a health concern to humans when exposed to the environment.[19] Considering the high value of carbonyl compounds, polycarbonate waste also serves as a green source of carbonyl functionalities, thus rendering it an ideal candidate for plastic upcycling. Recently, Sardon and co-workers demonstrated the organocatalyzed depolymerization of BPA-PC into various heterocycles,[20] diols,[21] and bis-imidazole[22] as value-added feedstocks. In addition, upcycling of polycarbonates paves the way for the manufacture of novel polymers, such as poly(aryl ether sulfone)s,[23] bicyclic diol based polycarbonate,[24] epoxy resins,[25,26] polyurethanes,[27] and non-isocyanate polyureas.[28]

Despite these excellent strategies to obtain innovative materials by upcycling of polycarbonates, the recyclability of the newly formed polymers, to our surprise, has never been investigated. Yet, implementing recyclability as a core design criterium for upcycled polymers would significantly increase their impact on sustainability. The installation of reversible covalent bonds into chemically crosslinked polymers is an effective method to develop circular polymeric materials.[29–32] These materials are well-known as covalent adaptable networks (CANs) or vitrimers, taking advantage of both thermoset properties (e.g., high mechanical performance) and thermoplastic behavior (e.g., ductility at elevated temperatures, reprocessability).[33–37] Due to the dynamic character of these polymeric networks, CANs are considered sustainable alternatives to non-recyclable polymers. The introduction of reversible chemical linkages

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facilitates in certain cases the chemical depolymerization of thermosets back into original monomers.[12, 16, 38, 39]

Herein, we combine for the first time upcycling of polycarbonates and the benefits of reversible covalent bonds to create new high-performance materials that are reprocessable as well as chemically recyclable and illustrate their cradle-to-cradle life cycle (Figure 1). To elevate the sustainability of our approach, we perform the selective depolymerization of BPA-PC using a bio-based vanillin derivative instead of petrochemical reagents. Thereby, we recover the monomer BPA, which can immediately be reused in making fresh BPA-PC, and obtain di-vanillin ethoxy carbonate (DVEC) as a new building block. Through condensation reaction with various primary amines, we prepared either linear or crosslinked poly(imine-carbonate)s, P(ImC)s, which show full recyclability under acidic conditions and malleability at elevated temperatures due to the dynamic nature of the imine bonds. Moreover, we demonstrate the selective separation of P(ImC)s from mixed plastic waste streams. Our approach to generate new circular polymeric materials through bio-based upcycling of polycarbonates contributes to the design of innovative and sustainable plastic waste treatment methodologies.

Results and Discussion

Upcycling of BPA-PC into Poly(imine-carbonate)s

First, we developed a sustainable and green protocol for the selective depolymerization of polycarbonate under solvent-free conditions with the hydroxyethylated derivative of vanillin (HEV), a widely available bio-based compound. To facilitate the transesterification reaction, 1,5,7-triazabicyclo-[4.4.0]dec-5-ene (TBD) was employed as an organocatalyst,[40, 41] which is known for its high efficiency for BPA-PC depolymerization.[42] ^{1}H NMR spectroscopy analysis showed quantitative conversion (>98%) of BPA-PC pellets into BPA and bis(2-(4-formyl-2-methoxyphenoxy)-ethyl) carbonate (DVEC) after a reaction time of 2 h at 160°C (see Supporting Information for detailed synthesis descriptions and characterization of obtained compounds). Lowering the temperature to 140 and 120°C, while keeping it above the melting point of HEV ($T_m = 95°C$), resulted in longer reaction times (Figure S1). After the reaction, BPA and DVEC were isolated by flash column chromatography with yields of 71% and 67%, respectively. While BPA can be reused to produce fresh BPA-PC and thereby establish a separate closed-loop recycling scheme, we focused on the newly produced monomer DVEC to synthesize and study various poly(imine-carbonate)s. To examine structure–property relationships, we used three diamines, namely 4,7,10-trioxa-1,13-tridecanediamine (OPA), Jeffamine-D230 (D230), and 4,4′-methylenedianiline (MDA), to create linear P(ImC)s, and tris(2-aminoethyl)amine (TREN) to prepare a crosslinked polymer network (Scheme 1). To further explore the design and property space of poly(imine-carbonate)s, we prepared crosslinked polymer networks which include either flexible or rigid diamines in addition to TREN as the crosslinking unit (molar ratio of diamine/TREN = 3/2 and 2/2.6, respectively. See Scheme 1 and Table S1). The condensation reactions were performed in DMF at 80°C and the final polymer samples were obtained after solvent casting, drying of polymers, and hot-pressing at 120°C. The polymers were characterized by ^{1}H NMR, FT-IR, and SEC (Figures S2–S6, Table S2). ^{1}H NMR and FT-IR spectra
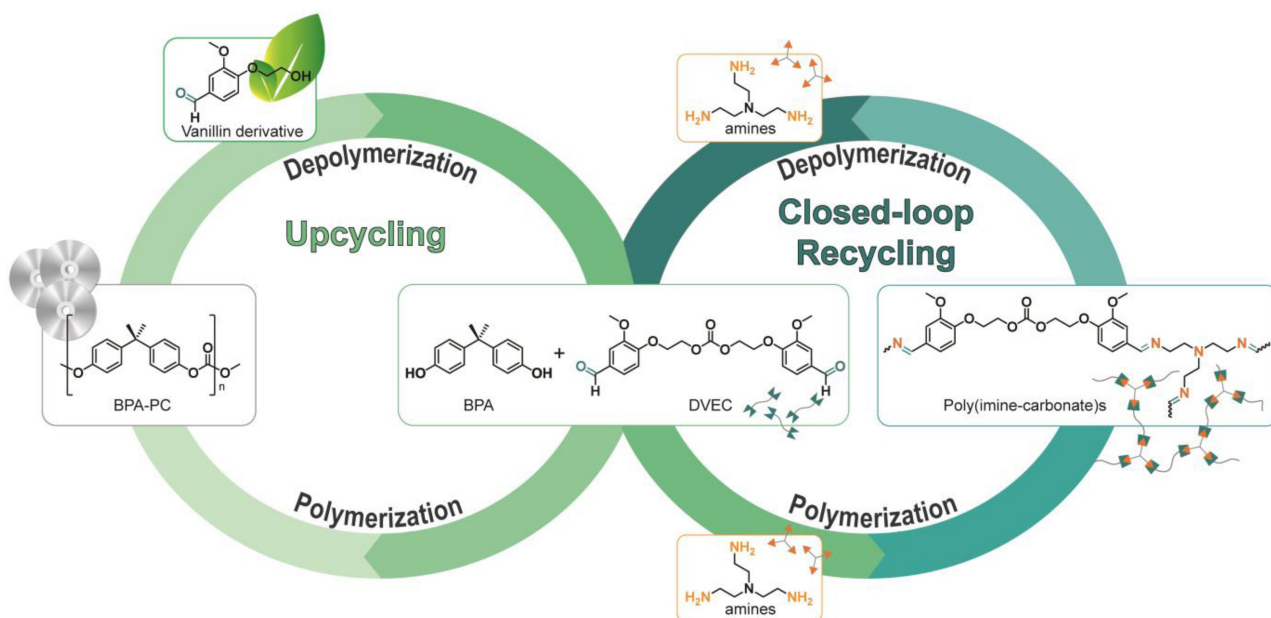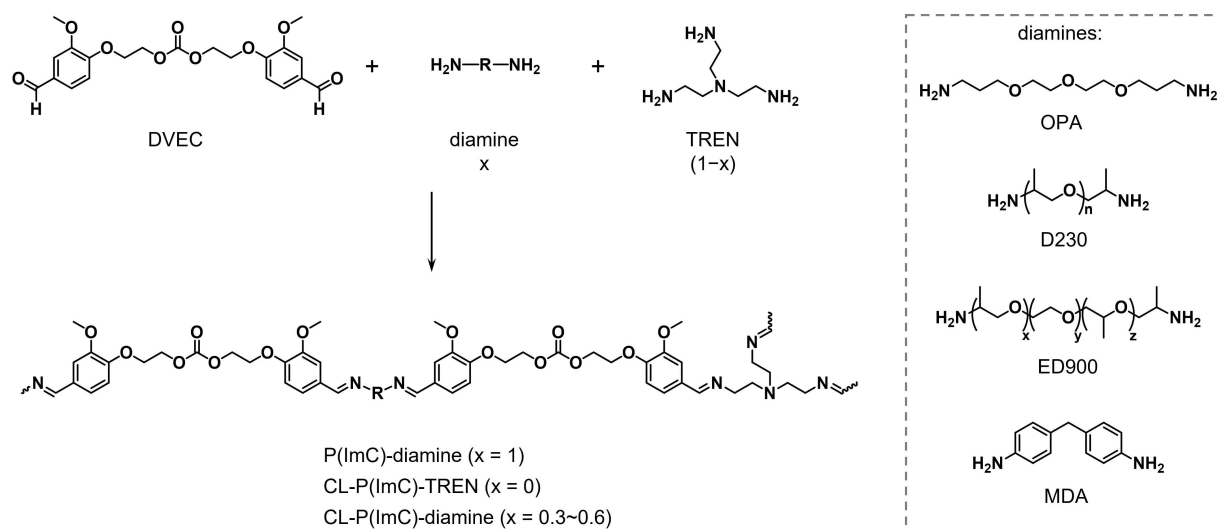

Figure 1. Upcycling of BPA-PC with a bio-based vanillin derivative into novel dialdehyde monomer DVEC which can be polymerized by using primary amines into poly(imine-carbonate)s. P(ImC)s show full recyclability under acidic conditions, thereby enabling a closed-loop recycling scheme.

Scheme 1. Preparation of linear and crosslinked poly(imine-carbonate)s with a series of diamines (OPA, D230, ED900, and MDA) and TREN as the crosslinking unit.

suggest the formation of imine bonds and consumption of aldehyde groups after polymerization. In ^{1}H NMR spectra of soluble linear P(ImC)s the newly formed imine signals manifest at around 8.2 ppm, while the monomer aldehyde signals at 9.9 ppm vanished. In FT-IR spectra a new peak at around 1640 cm^{-1} emerged and the signal at 1681 cm^{-1} disappeared, which correspond to C=N stretch vibration and C=O stretch absorption band of aromatic aldehydes, respectively.

Thermal and Mechanical Properties of P(ImC)s

The thermal properties of linear and crosslinked P(ImC)s were investigated using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). In TGA measurements, all polymers showed a high initial decomposition temperature ($T_{d5\%}$) in the range of 270 to 330°C (see Figure S7 and Table S3), which is comparable with other polyimine vitrimers.[43,44] The highest values for residual weight at the end of the TGA measurements were obtained with CL-P(ImC)-TREN and P(ImC)-MDA due to the high crosslink density and content of aromatic units within the polymeric structure, respectively. By DSC measurements, we found that through the careful selection of amines a broad range of material properties ($T_g = -13.1$–88.0°C, Figure 2A) can be covered, ranging from elastomer-like (low T_g) to rigid materials (high T_g). Furthermore, the obtained results indicate that the incorporation of more rigid aromatic diamines (i.e., MDA) into the polymer backbone increases the T_g, while more flexible diamines, such as ED900, D230 and OPA, lower the transition temperature. The viscoelastic properties of the crosslinked P(ImC)s were investigated by dynamic mechanical analysis (DMA, Figure 2B) and the values for T_g obtained from tan δ are in good agreement with DSC measurements. The mechanical properties of CL-P(ImC)s were investigated by tensile tests

(Figure 2C, Table S4). It was observed that the type of diamine in the network affects the mechanical properties. CL-P(ImC)-MDA showed the highest strength of 84.5 MPa and lowest elongation at break with 4.3%, while the introduction of flexible chains in CL-P(ImC)-OPA facilitates chain movement and led to reduced strength of 47.7 MPa and increased elongation at break with 35.3%. These results indicate that the amines' functionality and chain segment rigidity determine not only T_g, but also the strength of polymeric networks. This observation is in line with other polyimine structures,[43] while CL-P(ImC)-MDA exhibits a comparably high strength for imine-containing polymers.

Reprocessability of CL-P(ImC)s

The malleability at elevated temperatures is a crucial characteristic of vitrimers or CANs in terms of reshaping and reprocessing.[45,46] Since the polymeric network of CL-P(ImC)s is constructed by dynamic covalent imine bonds, topological thermal rearrangement is expected. To investigate the dynamic exchange behavior of the crosslinked polymers, a stress relaxation experiment was carried out using compression-molded CL-P(ImC)s. The time dependence of normalized relaxation modulus $G_{(t)}/G_{0.1}$ at different temperatures of CL-P(ImC)s is shown in Figures 3A–C. The relaxation time was recorded as τ^* where $G_{(t)}/G_{0.1}$ reached $1/e$. All three CL-P(ImC)s exhibit full stress relaxation and showed temperature dependence typical for CAN materials.[47–50] By comparing the different amine building blocks, a significant relaxation time difference was observed at 120°C with $\tau^* = 1.0$ s for CL-P(ImC)-OPA, 3.7 s for CL-P(ImC)-TREN, and 18.4 s for CL-P(ImC)-MDA. The activation energy (E_a) of the imine bond exchange reaction was further calculated using the Arrhenius equation and ranged from 78–115 kJ mol^{-1} (Figure 3D). These results show the slowest bond exchange rate for CL-P(ImC)-MDA
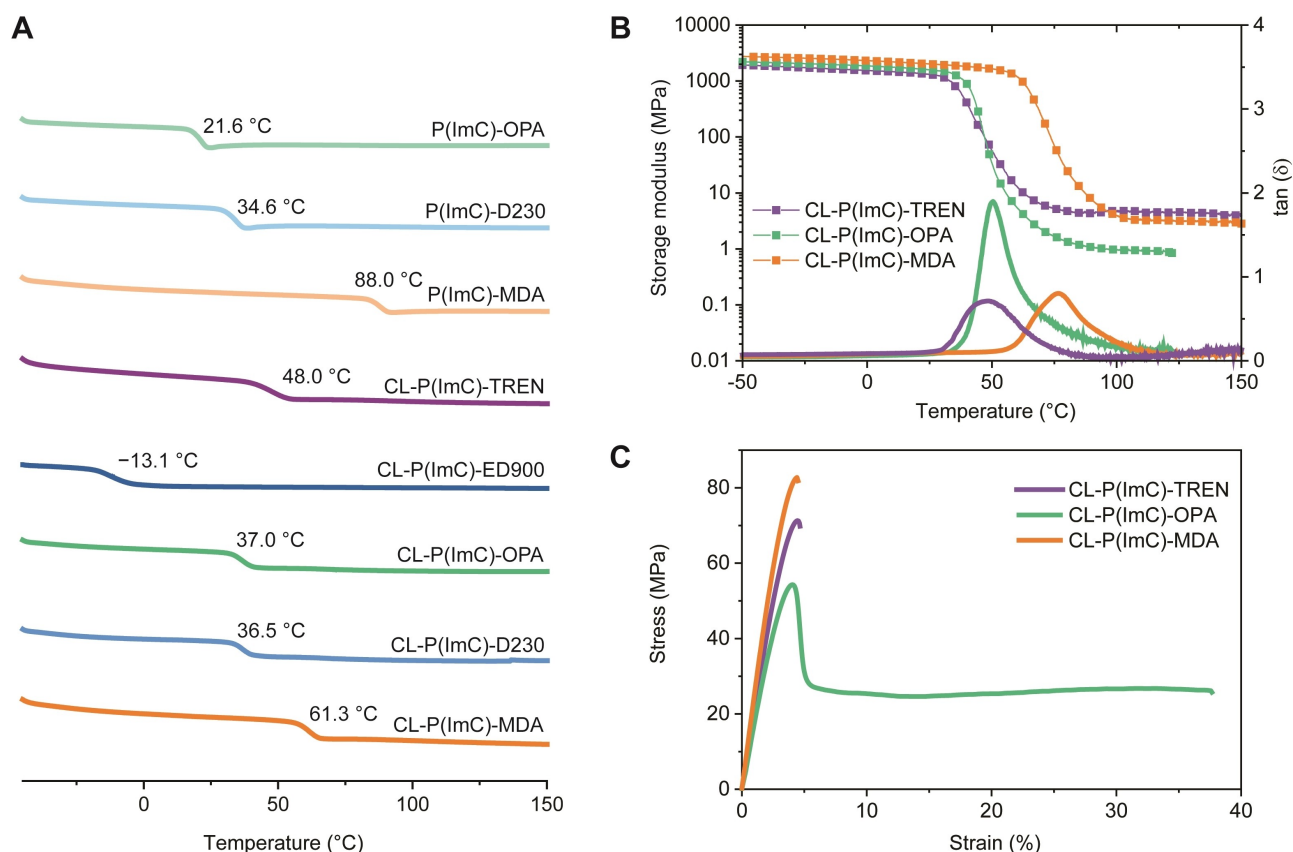
Figure 2. Thermal and mechanical characterization. A) DSC measurements of linear and crosslinked P(ImC)s showing the second cycle (T_g values are given for each polymer). B) DMA measurements of crosslinked CL-P(ImC)s showing storage moduli and tan δ. C) Stress-strain curves of crosslinked CL-P(ImC)s.

and the fastest rate for CL-P(ImC)-OPA and thus are in good agreement with the measured relaxation times.

To investigate the reprocessability of poly(imine-carbonate)s, a CL-P(ImC)-TREN polymer film was cut into small pieces and then reprocessed by hot-pressing at 120 °C and 10 MPa for 1 h (Figure 4A). After compression molding, the newly formed polymer film was subjected to a tensile test. The stress-strain curves of both original and reprocessed CL-P(ImC)-TREN were almost identical (Figure 4B). The reprocessed film showed slightly higher stress and decreased elongation at break. This slight alteration can be explained by an increase of crosslinking density.[43] Overall, the polymeric imine network displays excellent reprocessability and no loss in mechanical strength.

Closed-Loop Recycling via Chemical Depolymerization

Interested in creating closed-loop recycling, we studied and optimized the chemical depolymerization of CL-P(ImC)-TREN to selectively recover the monomeric building blocks, DVEC and TREN (Figures 5A and B). As imine bonds can conveniently be cleaved under acidic conditions,[29,51–53] CL-P(ImC)-TREN was treated with a solution of 0.1 M HCl and THF (v:v = 9:1) at room temperature for 2 h under continuous stirring. Within a few minutes, the bulk material

started to break, the mixture became turbid, and the formed DVEC precipitated as a fine powder, while TREN HCl remained dissolved in the aqueous mixture. After completed depolymerization, DVEC was filtered, washed with deionized water and EtOH, and dried in the oven at 60 °C (recovery yield 92 %). In addition, TREN was recovered in 93 % yield from the acidic supernate by neutralizing the HCl salt via an ion-exchange resin (Amberlyst A26) and removing the solvent. The high purity and chemical integrity of the recovered monomers was confirmed by ¹H NMR spectroscopy (Figures 5C and S8), thus rendering them ideal for producing fresh polymer samples and closing the recycling loop. In addition, the chemical recycling and separation of recycled materials from the heterogeneity and wide range of compositions in plastic waste streams are key points toward a sustainable economy.[12,13,39,54] Thus, the depolymerization of CL-P(ImC)-TREN and monomer recovery from mixed plastic waste were examined (Figure 5D). Pieces from various plastic products, such as PP (straw, green), HDPE (solvent bottle cap, red), PET (plastic bottle, transparent), and PC (laboratory safety goggles, transparent), were mixed with CL-P(ImC)-TREN. The polymer mixture was exposed to the same acidic depolymerization conditions. Due to the orthogonal reactivity and the chemical stability of the other plastic products under the used conditions, CL-P(ImC)-TREN was selectively depoly-
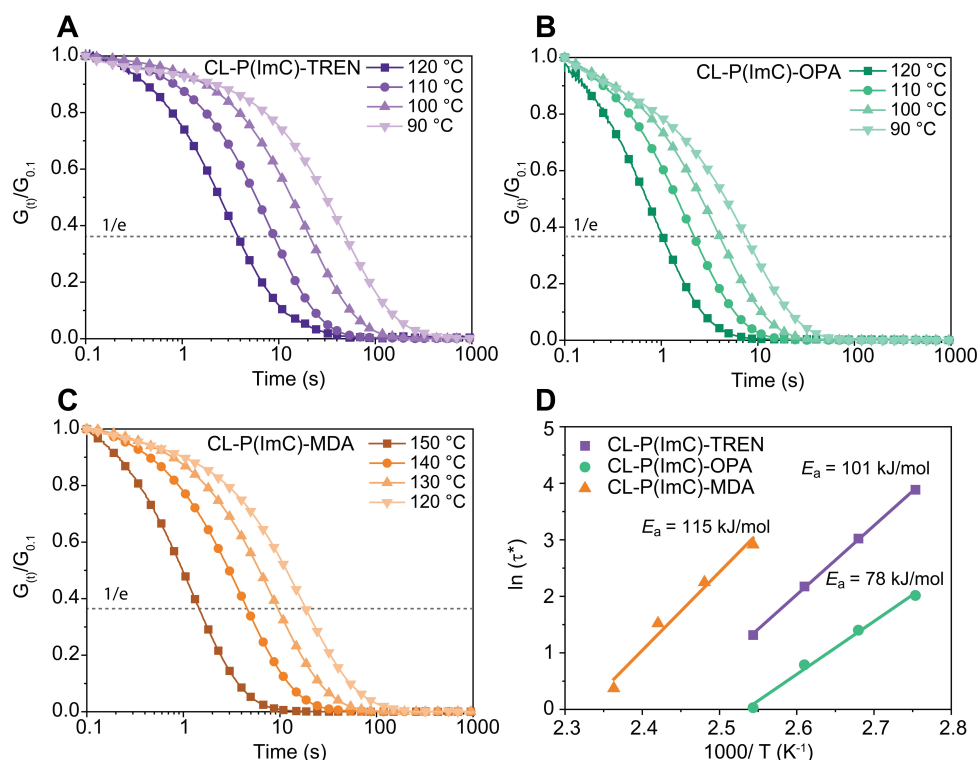
Figure 3. Normalized stress-relaxation at different temperatures of A) CL-P(ImC)-TREN, B) CL-P(ImC)-OPA, and C) CL-ImC-MDA. D) Fitted curves between 1000/*T* and characteristic relaxation time according to Arrhenius law.
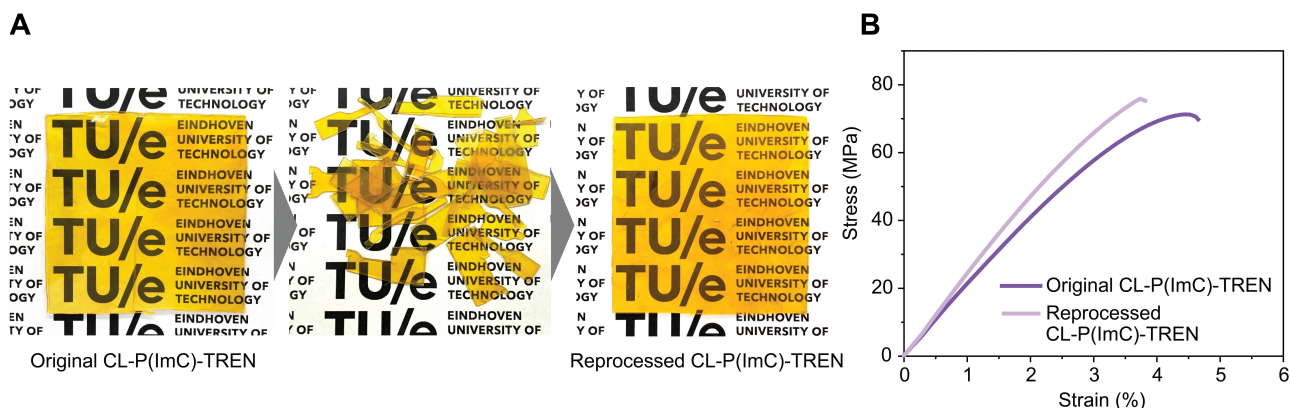

Figure 4. A) Cutting a polymer film of CL-P(ImC)-TREN into small pieces followed by reprocessing via hot-pressing at 120 °C and 10 MPa for 1 h. B) Stress-strain curves of original and reprocessed CL-P(ImC)-TREN.

merized. As a result, the original monomers, DVEC and TREN, could be easily separated from the mixed plastic waste, while the other plastic products remained stable in their original state (Table S5).

Conclusion

We demonstrated the upcycling of BPA-PC waste into the bio-based dialdehyde-containing monomer (DVEC) which was successfully used for the preparation of a library of polyimine vitrimers. Our modular approach, as illustrated by

using a series of diamines and/or triamine (TREN) as the starting materials, enabled us to cover a broad range of thermal and mechanical properties. Finally, the reprocessing as well as the efficient and selective closed-loop recycling of crosslinked poly(imine-carbonate) were performed. Notably, our polymeric system showed convenient separation from mixed plastic waste streams. Our environmentally friendly approach to create next-generation polymeric materials through depolymerization of common polycarbonate contributes to the design of innovative and sustainable plastic waste treatment methodologies. Moreover, these fully recyclable materials have the potential to replace non-recyclable
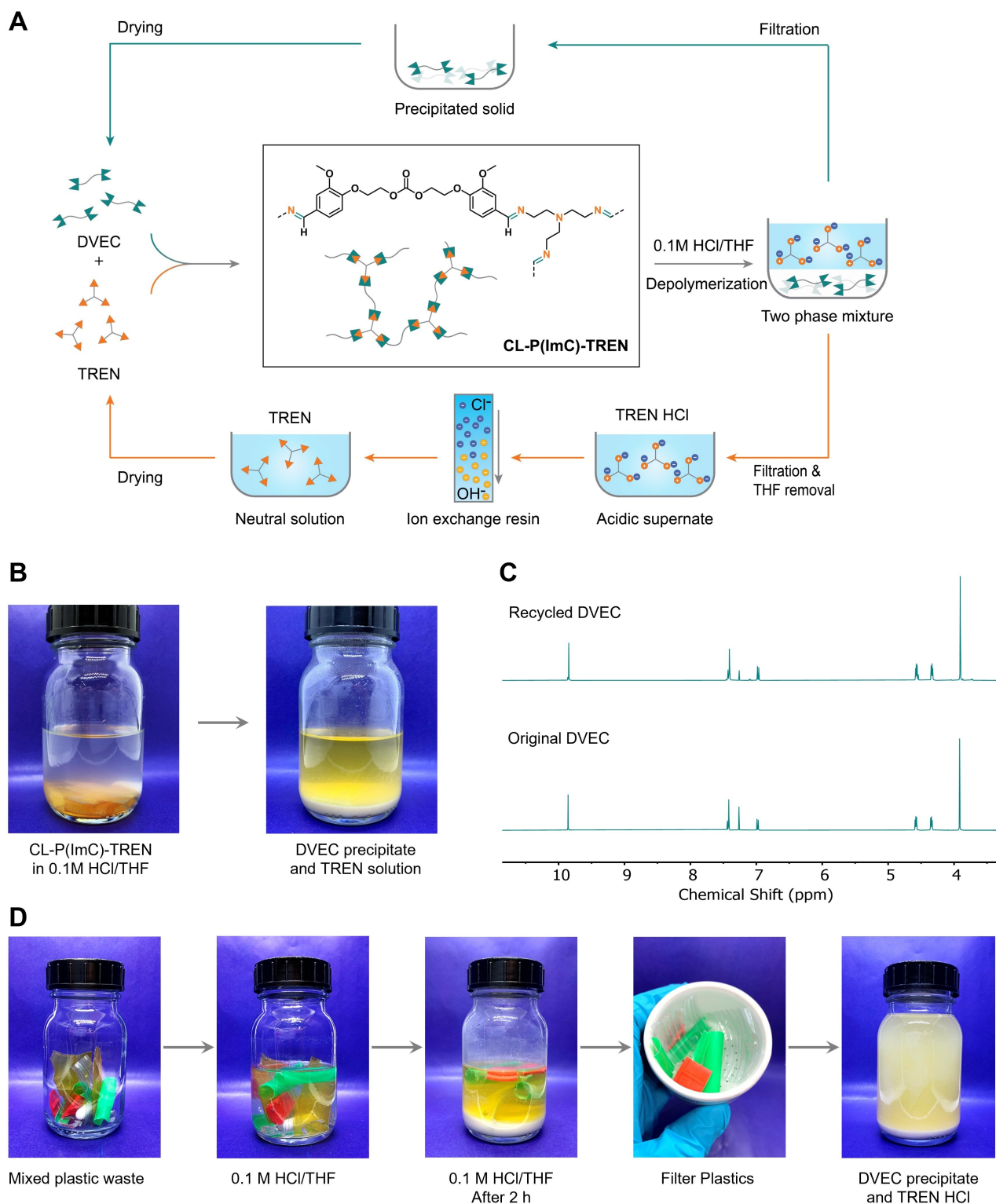
Figure 5. A) Schematic representation of the closed-loop recycling scheme demonstrated with crosslinked CL-P(ImC)-TREN. First, CL-P(ImC)-TREN was synthesized through condensation between DVEC and TREN. The chemical depolymerization was achieved under acidic conditions, followed by separation and purification steps to recover the pure monomers which can be used to prepare fresh polymers without loss of performance. B) Photographs showing the chemical depolymerization of CL-P(ImC)-TREN under acidic condition with a solution of 0.1 M HCl and THF (v:v=9:1) at room temperature for 2 h under continuous stirring. Both monomers were purified and isolated. C) ^{1}H NMR spectra of original DVEC (bottom) and recycled DVEC (top) in CDCl$_3$ (400 MHz, 25 °C). D) Photographs showing the selective chemical depolymerization process of CL-P(ImC)-TREN (yellow) from a plastic waste mixture containing pieces of PP (green), HDPE (red), PET (transparent), and PC (transparent). After applying acidic conditions for 2 h, the obtained monomers DVEC and TREN HCl were separated from the plastic mixture through filtration, purified, and isolated.

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thermosets in various application areas. Because our method merges for the first time vital aspects of sustainability and circularity, that is use of bio-based resources, upcycling of commodity plastics, and closed-loop recycling of new polymers, we envision that this novel concept will lead to advancements regarding plastic pollution. Since this energy-efficient strategy is also applicable to other carbonyl containing plastics (e.g., polyesters), exciting new paths for establishing a circular economy become accessible.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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