

Closed-Loop Recyclable Poly(imine-acetal)s with Dual-Cleavable Bonds for Primary Building Block Recovery

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Closed-Loop Recyclable Poly(imine-acetal)s with Dual-Cleavable Bonds for Primary Building Block Recovery

Keita Saito,^[a] Tankut Türel,^[a] Fabian Eisenreich,^[a] and Željko Tomović^{*,[a]}

Chemical recycling offers a promising solution for the end-of-life treatment of synthetic polymers. However, the efficient recovery of well-defined recycled building blocks continues to be a major challenge, especially for crosslinked thermosets. Here, we developed vanillin-based polymer networks functionalized with dual-cleavable imine and acetal bonds that facilitate chemical recycling to primary building blocks and their convenient separation at the molecular level. A library of crosslinked poly(imine-acetal)s was synthesized by combining the in-bulk synthesized liquid di-vanillin acetal monomer (DVA) with commercially available liquid di- and triamines under solvent-free conditions. These thermosets showed tailor-made thermal and mechanical properties along

with outstanding chemical recyclability. Under aqueous acidic conditions, poly(imine-acetal)s selectively and completely disintegrate into small molecules. During the polymer design stage, these compounds were carefully selected to enable facile separation without tedious techniques. As a result, the primary building blocks were isolated in high yields and purity and immediately reused to produce fresh polymers with identical thermomechanical properties. Since our “design for recycling” concept aims at obtaining the primary building blocks rather than monomers after depolymerization, a plethora of possibilities are unlocked to utilize these chemical resources, including closed-loop recycling as portrayed.

Introduction

Traditional thermosetting materials are irreplaceable in industry and our daily lives due to their excellent mechanical and thermal properties.^[1] Nevertheless, they cannot be recycled due to their permanently and covalently crosslinked architectures. As a result, most of them end up as plastic waste, either being incinerated or disposed of in landfills after consumption.^[2–5] To address this issue, depolymerization of crosslinked materials into original monomers by controlled cleavage of the chemical bonds is an efficient strategy.^[6–13] This process enables the generation of fresh identical polymers without the loss of their material performance, thereby closing the recycling loop.^[6,7,9,10,14,15] Hence, chemical recycling to monomers represents a powerful approach, offering crucial opportunities to conserve the chemical resources, and thus provides a sustainable solution for end-of-life plastic products.

Significant efforts have been devoted to the development of chemically recyclable polymer networks, specifically aiming to achieve closed-loop recycling. This objective is

accomplished by employing reversible chemical bonds, such as carbonate,^[9] diketoenamine,^[6,12] boronic ester,^[16] disulfide,^[17] imine,^[7] and acetal linkages,^[18] into the polymer backbone. In extension to this, the incorporation of two types of reversible bonds into macromolecular structures, taking advantage of each character, is an effective strategy to create desirable, recyclable polymers with tunable properties.^[17,19–23] For instance, Lehn and co-workers combined imine moieties and polyester units to prepare a “green dynamer” which exhibited doubly degradable features, based on chemical and biological degradability.^[20]

In addition to implementing recyclability as a core feature of polymer design, the use of bio-based reagents as building blocks enhances the sustainability even further.^[24,25] Among the renewable chemicals, vanillin and its derivatives are of particular interest as building blocks due to their rigid aromatic chemical structure and functional groups.^[26,27] The aldehyde group of vanillin, for instance, provides versatile possibilities through the formation of imine bonds with amines, which are well-known motifs to render polymeric structures cleavable by hydrolysis under acidic conditions.^[25,28–33] Recently, our group developed vanillin-based polyimine thermosets through the upcycling of plastic waste, which exhibited tailor-made material performance and chemical recyclability to monomers.^[7] In addition, introducing imine linkages has also been applied to produce polyurethane^[19,34–36] and epoxy networks.^[37–43]

However, once the hydroxy group of vanillin reacted with other species, such as phosphate, isocyanate, or epoxy derivatives, it is virtually impossible to cleave the newly formed bonds under mild conditions.^[19,28,44] Considering the low yields of vanillin production from lignin,^[45,46] its recovery from synthetic polymers represents a critical advantage. Yet,

[a] K. Saito, Dr. T. Türel, Dr. F. Eisenreich, Prof. Dr. Ž. Tomović
 Polymer Performance Materials Group, Department of Chemical Engineering and Chemistry and Institute for Complex Molecular Systems
 Eindhoven University of Technology
 5600 MB Eindhoven (The Netherlands)
 E-mail: z.tomovic@tue.nl

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research conducted on vanillin-based polymers has not prioritized the development of selective depolymerization processes that yield pure vanillin as a primary building block. It is crucial to conceptually differentiate recycling strategies that aim to obtain monomers from those that target primary building blocks. While the former method is ideal for keeping the chemical resources in a closed loop, the recycled monomers are typically tied to one specific polymer type. In contrast, chemical recycling to primary building blocks unlocks extensive design space to create, for instance, other polymer families or industrially relevant chemicals. While this chemical recycling concept may be beneficial, it is largely unexplored.

In this work, we combine both imine and acetal bonds to construct vanillin-based polymer networks, which facilitate chemical recycling to primary building blocks due to these dual-cleavable bonds (Figure 1). Through condensation reaction between the in-bulk synthesized and liquid di-vanillin acetal monomer (DVA) and selected commercial liquid di- and triamines, we prepared several crosslinked poly(imine-acetal)s, P(ImA)s, with tailor-made thermal and mechanical properties. Notably, due to simultaneous hydrolysis of imine and acetal bonds, the polymers selectively disintegrate into primary building blocks, including pure vanillin, diethylene glycol, respective amine, and acetaldehyde. These molecules can easily be separated in high yield and purity and immediately reused for producing fresh and identical polymers in a manner of closed-loop recycling. Moreover, we demonstrated the selective depolymerization and separation of P(ImA) from a plastic waste mixture. This novel concept of recyclable-by-design thermosets for primary building block

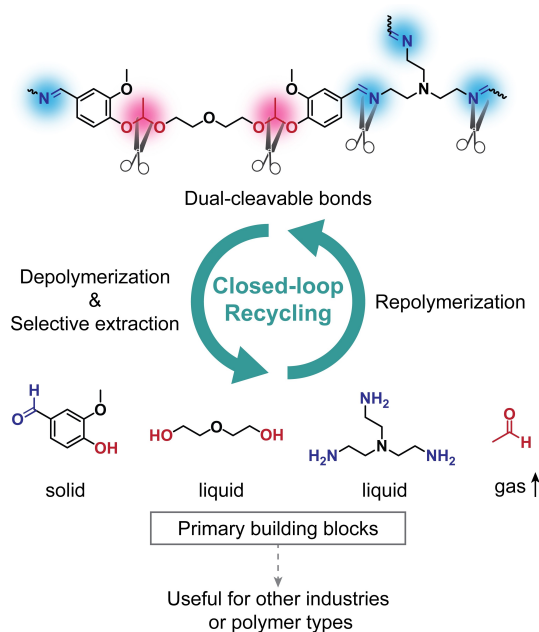


Figure 1. Design of a polymeric network containing dual-cleavable bonds. The simultaneous hydrolysis of imine and acetal bonds facilitates the depolymerization back into primary building blocks.

recovery provides an important advancement for the creation of sustainable materials that consider the end-of-life states.

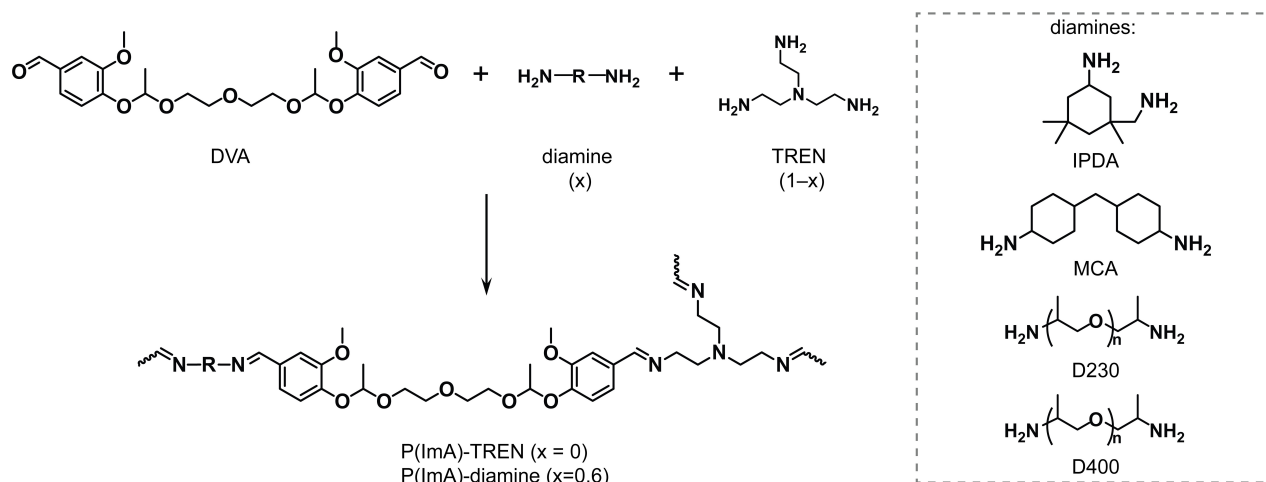
Results and Discussion

Synthesis and Characterization of Poly(imine-acetal)s

The synthesis of DVA monomer was performed using catalyst-free, click-type addition reaction between vanillin and di-vinyl ether.^[47] The characterization of DVA was performed using ¹H NMR, ¹³C NMR, and FTIR spectroscopy as well as MALDI-TOF spectrometry (Figure S1–S2). Appearance of acetal protons at 5.61 ppm and disappearance of vinyl ether groups at 6.50 ppm showed that the conversion was successful and complete. Tris(2-aminoethyl)amine (TREN) was utilized as a crosslinker to prepare the crosslinked P(ImA)s. To further examine structure-property relationships, a variety of diamines, such as isophorone diamine (IPDA), 4,4'-methylenebis(cyclohexylamine) (MCA), Jeffamine D230 (D230), and Jeffamine D400 (D400), were incorporated in addition to TREN in molar ratios of diamine/TREN = 3/2 (Scheme 1 and Table S1). The condensation reactions between aldehyde and amine were carried out in bulk conditions and the final polymer films were obtained after curing in a vacuum oven at 80 °C and hot-pressing at 120 °C and 50 kN for 30 min. The obtained P(ImA)s were analyzed by FTIR spectroscopy which showed that the characteristic signal of C=O stretching vibration belonging to the aromatic aldehyde at 1682 cm⁻¹ vanished. On the contrary, a new signal for C=N stretching was observed at 1643 cm⁻¹, suggesting the successful polymerization through Schiff base formation (Figure S3). Furthermore, the hydrolytic and solvent resistance of P(ImA)s were confirmed under selected conditions (Figure S4–S8 and Table S2–S3).

Thermal and Mechanical Properties of Poly(imine-acetal)s

The thermal properties of P(ImA)s were investigated using thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) (Figure 2A, Figure S9, and Table S4–S5). All polymers showed good thermal stability with an initial decomposition temperature ($T_{d5\%}$) ranging from 227.6 to 267.0 °C. P(ImA)-TREN displayed the highest value for residual weight, which is attributed to its high crosslinking density. DSC results demonstrated that the glass transition temperatures (T_g) were in the range between 0.5 to 41.8 °C. The incorporation of cyclic diamines, such as IPDA and MCA, into the polymer backbone increased the T_g values, while the utilization of flexible diamines (i.e., D230 and D400) caused the opposite effect. These results indicate the pivotal role of diamines in modulating the thermal material properties, spanning from rubbery to glassy, to cater to the diverse requirements of polymer applications. Thermomechanical characterization of P(ImA)s was performed by dynamic mechanical analysis (DMA) (Figure 2B, Table S5). The T_g values



Scheme 1. Preparation of crosslinked poly(imine-acetal)s with a series of diamines (IPDA, MCA, D230, and D400) and triamine (TREN).

obtained from the maxima of the $\tan \delta$ curves showed the same trend as those obtained by DSC measurements. The mechanical performance of P(ImA)s was analyzed by tensile tests (Figure 2C, Figure S10, Table S6). The incorporation of rigid, cyclic diamines into polymer networks led to more rigid materials with superior mechanical properties. P(ImA)-IPDA showed the highest tensile strength of 41.8 MPa and lowest elongation at break with 13.3%. In contrast, networks with flexible diamines, P(ImA)-D230 and P(ImA)-D400, exhibited elastomer-like behavior with lower tensile strength of 8.9 and 1.6 MPa and higher elongation at break values of 267 and 244%, respectively. These results were supported by the thermal performance characterization which indicated the differences between glassy and rubbery state at serving temperatures.

Since imine and acetal groups are known to be interchangeable bonds,^[18,30,47–49] P(ImA) networks may exhibit thermal rearrangement at elevated temperature and pressure. Thus, we investigated the reprocessability of P(ImA)-TREN network, which was selected as the representative material (Figure S11–S13). To investigate the reprocessability of P(ImA)-TREN, small pieces were cut from the original polymer film and then reprocessed by hot-pressing at 120 °C and 50 kN for 30 min to produce a fresh homogeneous film (Figure S12A). The reprocessed P(ImA)-TREN showed comparable tensile stress to the original film (Figure S12B).

Chemical Recycling of Poly(imine-acetal)

The presence of dual-cleavable bonds in the polymer network facilitates the chemical depolymerization back into defined small molecules (Figure 3A). During the molecular design stage, careful attention was devoted to ensuring simple separation of primary building blocks for this chemically recyclable polymer. As a result, the process of depolymerization yields distinct solid, liquid, and gaseous components. In addition, the compounds bear various func-

tional groups, including hydroxy and amino groups, which further facilitates their separation and isolation. To demonstrate on-demand chemical recycling, P(ImA)-TREN was treated with 0.1 M HCl solution under continuous stirring at 50 °C. Within 30 min, the polymer network completely depolymerized and a homogeneous solution was obtained (Figure 3B).

Due to the simultaneous cleavage of imine and acetal bonds under acidic conditions, small and defined molecules including vanillin, diethylene glycol (DEG), acetaldehyde, and TREN-HCl were formed. The separation of these compounds was straightforward and accomplished by using the characteristics of their chemical structures (i.e., solubility and salt formation) without using tedious methods, such as column chromatography (Figure S14). First, vanillin was recovered by extracting the aqueous mixture with ethyl acetate. After removal of water, acetone was added to the residual mixture to selectively dissolve DEG, while TREN-HCl remained undissolved as a solid. After filtration, DEG was recovered upon evaporation of acetone. Neutral TREN was eventually obtained after treating it with an ion-exchange resin and further purified by vacuum distillation. Acetaldehyde was released as a gas, which may be recovered using distillation method in an industrial scale.^[50] The recycling yields of vanillin, DEG, and TREN were gratifyingly high with values of 97, 91, and 75%, respectively. In addition, their purity and chemical integrity were confirmed by ¹H NMR spectroscopy (Figure 3C), which facilitate the production of fresh polymers and closure of the recycling loop. In addition, the effective 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. Thus, we performed selective depolymerization of P(ImA)-TREN from a plastic waste stream as a proof of concept (Figure 3D). P(ImA)-TREN was mixed with plastic pieces from various products, such as PET (plastic bottle, transparent), HDPE (solvent bottle cap, red), PP (straw, green), and PC (laboratory safety goggles, transparent). The plastic mixture

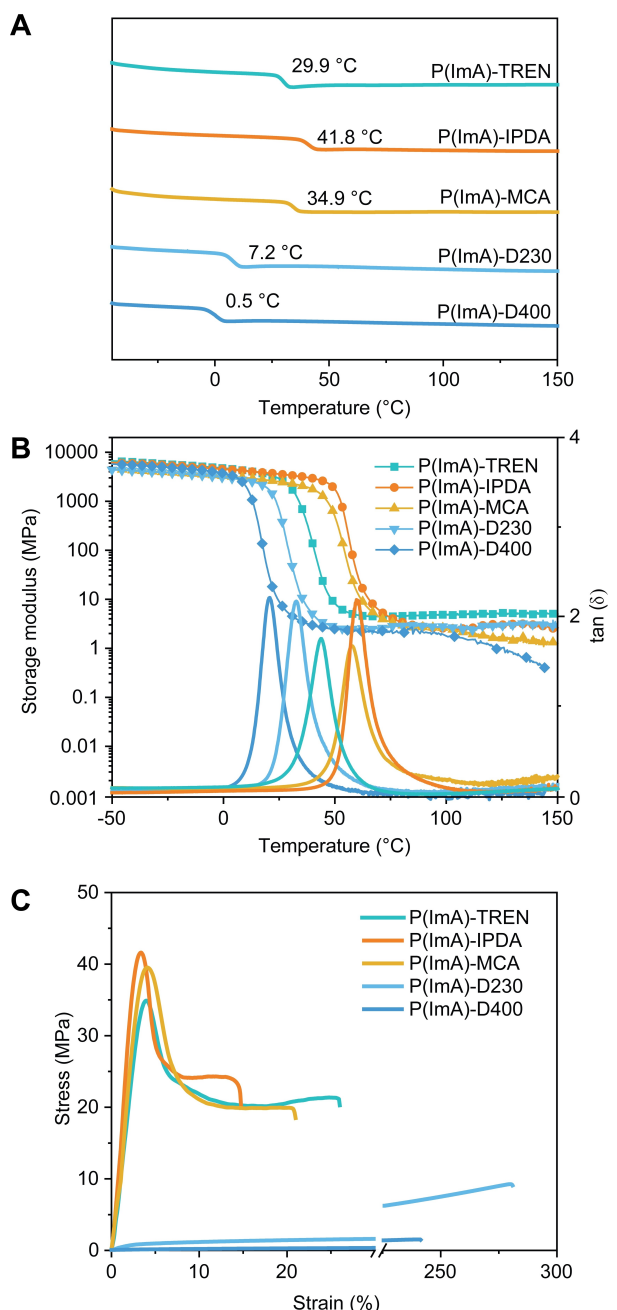


Figure 2. Thermal and mechanical properties of prepared poly(imine-acetal)s. A) DSC measurements showing glass transitions. B) DMA curves showing storage moduli (MPa) and $\tan(\delta)$ values. C) Stress-strain curves of P(ImA)s (the extended graph is recorded in Figure S10).

was treated with the same acidic depolymerization condition. Due to the orthogonal reactivity and the chemical stability of the other plastic products under the used conditions, selective depolymerization of P(ImA)-TREN was achieved. Consequently, well-defined molecules including vanillin, DEG, and TREN, could be easily separated from the mixed plastic waste, while the other plastic products remained stable in their original state (Table S7). This unique advantage highlights the effectiveness of chemical recycling, which

cannot be achieved through mechanical or thermal recycling methods.

Moreover, recycled vanillin was utilized to produce a new batch of DVA monomer, which showed identical signals in ^1H NMR spectra as compared to the original one (Figure S15). Then, a new generation of P(ImA)-TREN was produced using recycled DVA and TREN. According to FTIR, DSC, DMA, and tensile test experiments, the recycled P(ImA)-TREN exhibited nearly identical properties as the original thermoset (Figure 4A–D). For instance, T_g values of the pristine and recycled P(ImA)-TREN were measured to be 29.9 and 29.1 °C by DSC as well as 43.3 and 44.9 °C by DMA, respectively. Tensile tests showed similar mechanical properties with a tensile strength of 37.6 MPa (36.1 MPa for original material) and an elongation at break with 27.0% (25.8% for original material). This closed-loop recycling process allows for the sustainable use P(ImA)-TREN, as it can be depolymerized and repolymerized repeatedly without the loss of their chemical integrity and material performance, which contributes to reducing plastic waste and environmental impact.

Conclusions

In conclusion, we demonstrated the successful synthesis of vanillin-based polymer networks that incorporate both imine and acetal bonds. The liquid di-vanillin acetal monomer (DVA) was used in solvent-free imine condensation reactions with commercially available liquid diamines and TREN as crosslinker to produce a library of poly(imine-acetal) networks. These thermosets exhibited tailor-made thermal and mechanical properties, ranging from rubbery to glassy materials, enabling the ability to cater to the diverse requirements of polymer applications. Based on the on-demand cleavable bonds, these thermoset materials could be readily subjected to chemical recycling. Notably, due to simultaneous hydrolysis of imine and acetal bonds, primary building blocks, including native vanillin, were obtained and easily separated in high yields and purities. The recovered compounds were reused to produce fresh polymers with identical thermomechanical features, thereby providing an effective closed-loop recycling scheme. In this study, the critical role of the recycle isolation is emphasized on par with the recycling process itself, as both are integral to achieving peak efficiency. We envision that our concept of carefully selecting solid, liquid, and gaseous primary building blocks and connecting them with cleavable linkages provides a valuable blueprint for streamlining the recycle recovery and enhancing the design space of circular polymers.

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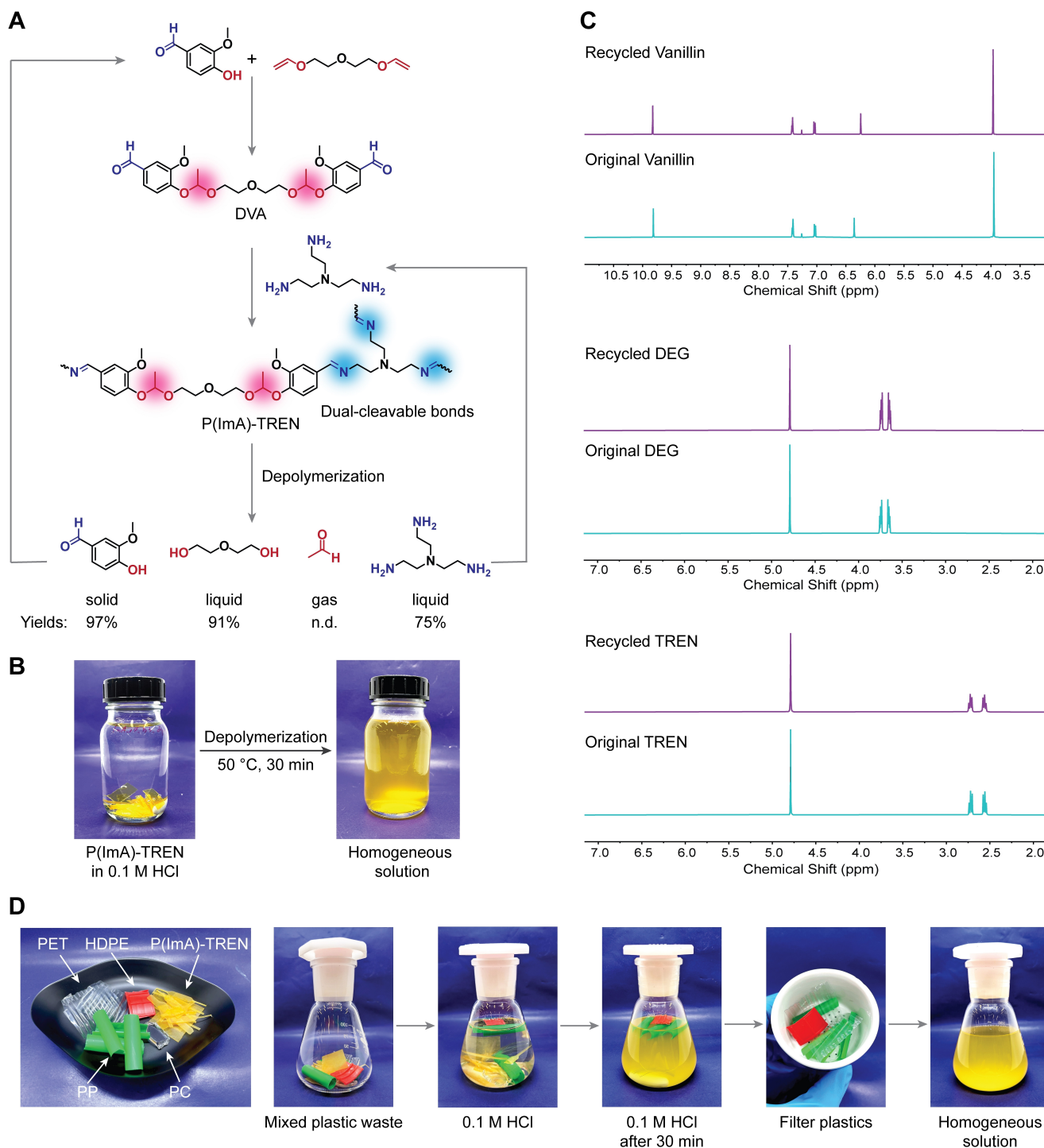


Figure 3. Chemical recycling of poly(imine-acetal). A) Schematic illustration of the closed-loop recycling strategy of P(ImA)-TREN, including the synthesis of the crosslinked polymer through condensation between DVA and TREN, followed by on-demand chemical depolymerization under acidic condition. The pure building blocks vanillin, DEG, and TREN were recovered after simple purification steps. The retrieved vanillin can be used to produce new DVA, which can then enter the scheme as a monomer to prepare a new generation of P(ImA)-TREN through condensation with recycled TREN. B) Photographs showing P(ImA)-TREN in a 0.1 M HCl solution before and after complete depolymerization by continuous stirring at 50 °C for 30 min. C) ¹H NMR spectra of original vanillin and recycled vanillin in CDCl₃ (400 MHz, 25 °C) (top). ¹H NMR spectra of original DEG and recycled DEG in D₂O (400 MHz, 25 °C) (middle). ¹H NMR spectra of original TREN and recycled TREN in D₂O (400 MHz, 25 °C) (bottom). D) Photographs showing the selective chemical depolymerization process of P(ImA)-TREN (yellow) from a plastic waste mixture containing pieces of PET (transparent), HDPE (red), PP (green), and PC (transparent).

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Conflict of Interests

The authors declare no conflict of interest.

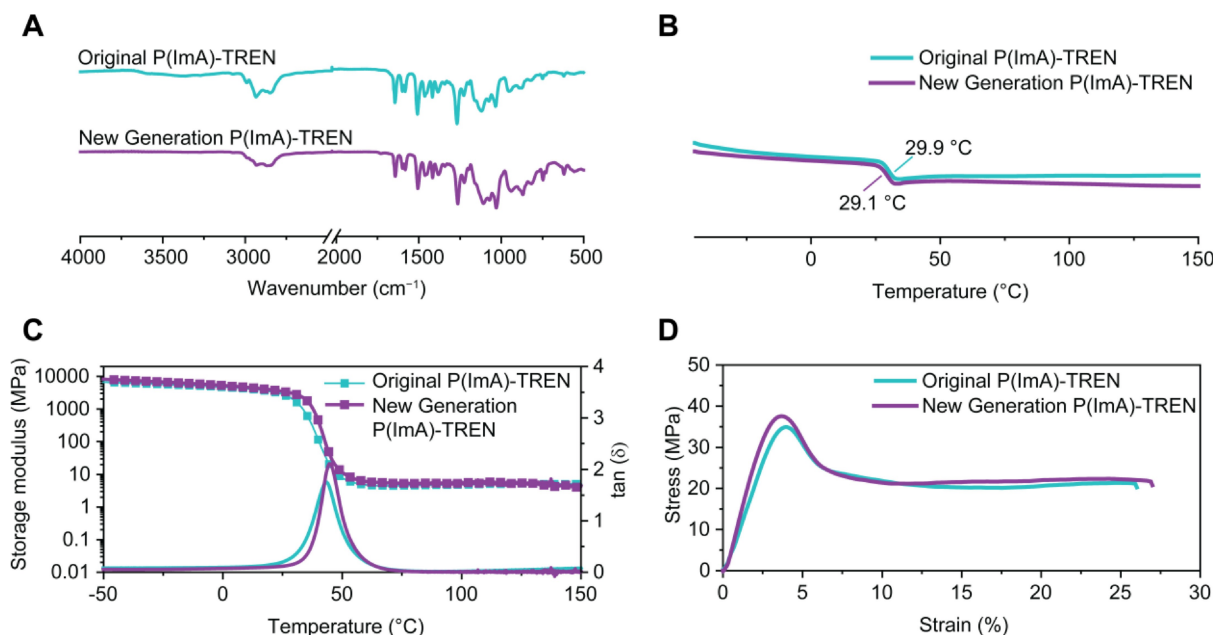


Figure 4. Characterization of regenerated P(ImA)-TREN through the chemical recycling. A) FTIR spectra, B) DMA curves, C) DSC curves, and D) stress-strain curves of original P(ImA)-TREN and regenerated P(ImA)-TREN.

Data Availability Statement

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

Keywords: Design for recycling · Chemical recycling · Closed-loop recycling · Dual cleavable bonds · Bio-based monomer

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