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Article

Synthesis and Chemical Recovery of Castor Oil-Based Poly(ester amides) with PE-Like Performance

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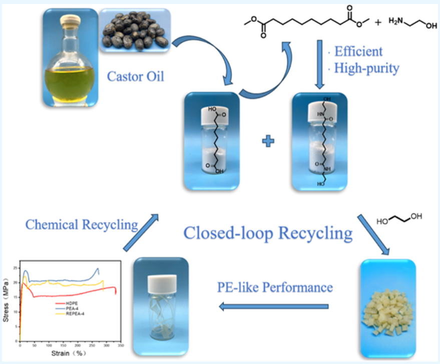
ABSTRACT: Biobased closed-loop recyclable olefin-like materials are environmentally friendly and have promising application prospects. However, such materials are scarce, and especially the olefin-like materials developed so far are limited to polyester materials. Therefore, developing nonpolyester biobased olefin-like materials is of significant interest. In this study, a castor oil-based poly(ester amide) (PEA) with polyethylene-like properties and closed-loop recyclability was successfully prepared. The obtained PEA materials exhibit thermal and mechanical properties comparable to polyethylene and surpass those of previously reported linear poly(ester amide)s. Notably, PEA can be completely hydrolyzed to its precursors, N^1,N^{10}-bis(3-hydroxypropyl) decanediamide (DSE) and sebacic acid (SA), with yields of 73.1−75.8 and 92.8−96.2%, respectively. Further experiments demonstrated that repolymerization of recovered DSE and SA produced REPEA materials with mechanical properties equivalent to the original polymer. This study achieves closed-loop recycling for biobased linear nonpolyester materials, which offers a novel approach to design olefin-like sustainable materials.



INTRODUCTION

Petrochemical-derived plastics have become an indispensable part of modern daily life, with polyolefins being among the most widely consumed materials.[1−3] Polyethylene, in particular, is extensively used in plastic bags, straws, packaging, films, and other applications. However, polyethylene's inability to degrade naturally poses significant environmental challenges.[4,5] Disposal often involves incineration, leading to substantial carbon dioxide emissions, exacerbating global warming, and creating severe societal impacts. Furthermore, the over-exploitation of fossil fuels has strained resources, leaving their supply insufficient to meet the growing demand for fuels and polymer materials.[6] Addressing the dual challenges of resource scarcity and environmental degradation has become a critical priority for scientists and governments. Among potential solutions, the recycling of polyolefin materials is particularly desirable.[7,8]

Normally, polyethylene recycling is divided into two categories: physical and chemical.[9−11] Physical recycling involves reprocessing polyethylene plastic waste into products that are commonly used via methods such as melting, extrusion, injection molding, and others. However, this process often results in significant performance degradation of the material and involves high costs, limiting its scalability.[12−14] Chemical recycling, on the other hand, remains a challenge due to polyethylene's robust C−C bond structure, which requires depolymerization at high temperatures (600 °C), with the yield less than 10%. Therefore, it is necessary to develop a cost-effective and energy-efficient method for depolymerizing polyethylene and recovering polyolefin materials.[15,16]

A notable attempt to address polyethylene's degradation involves inserting photodegradable ketone bonds into high-density polyethylene chains. This approach allows for controlled degradation by adjusting the density of ketone bonds.[5] However, the process requires complex catalysts, increasing production costs, and does not address the recycling of degraded materials. In addition, this study solves the problem that high-density polyethylene is difficult to degrade, but without considering the recycling process after degradation, it cannot solve the problem of lack of oil resources, which means that it is particularly important to find sustainable substitutes for polyolefin materials.

Häußler et al. designed the linear diacid and diol monomers from oleic acid through olefin metathesis and other reactions.[3] Then the high molecular weight polyester (PE-18,18) was prepared by melting polycondensation of octanedioic acid and octadecanediol, and polycarbonate (PC-18) was prepared by reacting octadecanediol with CO_2. These materials exhibit crystalline structures and crystallinity properties comparable to high-density polyethylene (HDPE). Thus, the prepared polyester and polycarbonate have similar mechanical performance to those of HDPE. On this basis, the alcoholysis reaction

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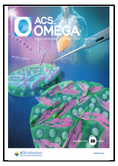

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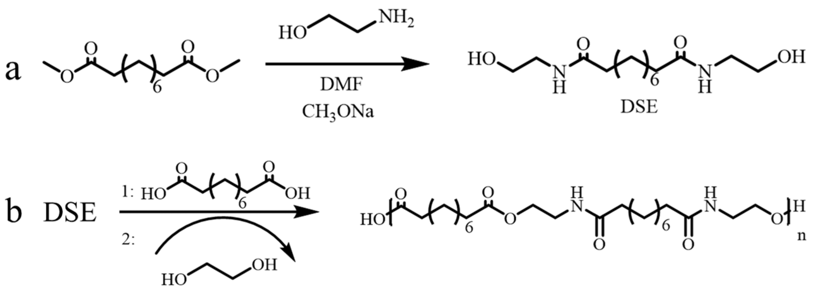
Scheme 1. Synthesis Route of the (a) Amide Diol Monomer and (b) Poly(ester amide)



of the material is conducted at 150 °C in alkaline methanol or ethanol as the solvent. It was found that the material was depolymerized into diol monomer. Remarkably, the performance of repolymerized RPE-18,18 closely matched the original PE-18,18, demonstrating the feasibility of closed-loop recycling for olefin-like materials. This innovation significantly reduces dependence on traditional polyethylene and highlights the potential of polyolefin-like materials.[17,18]

It is demonstrated that linear long-chain polyesters show promise as polyolefin alternatives, but their reliance on costly long-chain diacid and diol precursors derived from vegetable oils via olefin metathesis limited the industrial scalability.[3] Additionally, the properties of linear polyester materials are closely related to the chain length of their reaction monomers, which results in the large-scale production of such PE-like polyester materials being highly dependent on expensive monomers derived from vegetable oils.[14,15] It is still a great challenge to produce polyethylene-like (PE-like) polyesters on a large scale at a lower cost.[19]

As we all know, the introduction of amide bonds into the polyester backbone makes poly(ester amide) (PEA), which could improve the thermodynamic properties of the material.[20,21] PEA not only improves the thermodynamic properties but also retains degradability because of the introduction of amide bonds.[22] It is hypothesized that incorporating amide bonds into a long-chain polyester backbone could yield PE-like materials with desirable properties.[23]

Researchers are most concerned about the degradability of PEA materials.[24−28] Previous studies mainly focused on the degradation of this type of materials under different pH conditions,[29] enzymatic[24] or in vivo degradation,[25] and there are problems such as slow degradation and incomplete degradation in these studies. To facilitate monomer recovery, the degradation of ester bonds while preserving amide bonds is essential. The degradation and recovery of polyamide materials such as polycaprolactam (PA6)[26] and lactam 5-azabicyclo[2.2.1]octan-6-one (5/6-LM)[27] have been reported in the literature, and the degradation temperatures are as high as 300 and 260 °C. The reaction temperature of 160 °C was used for the complete closed-loop recovery of PEA material, in which both amide and ester bonds are effectively broken, allowing for efficient recovery of monomers.[28]

As illustrated in Scheme 1, this study introduces an amide bond into the polyester backbone through the amidation reaction. Aliphatic diester reacts with amino alcohol to form amide diol monomers, which are subsequently polymerized with diacid to prepare linear biobased poly(ester amide). The properties of the polymer are controlled by adjusting the density of the amide bond on the polymer chain. Furthermore, the polymer can be depolymerized to amide diol and diacid monomer by alkaline hydrolysis. These recovered monomers can be repolymerized to PEA in a certain proportion. This enables the closed-loop utilization of PE-like materials. The biobased diacid/diester and sebacic acid/dimethyl sebacic acid are derived from castor oil through alkali hydrolysis, while the amino alcohol is ethanolamine. In general, the low cost of preparing these polyester amides makes them suitable for widespread application, offering a sustainable and economically viable alternative to traditional polyolefins.

■ EXPERIMENTAL SECTION

Materials. Dimethyl sebacate (98%, Aladdin), ethanolamine (99%, Aladdin), *N*,*N*-dimethylformamide (DMF, 99%, Macklin), sodium methoxide (5 mol/L in methanol, TCI), sebacic acid (SA, 99%, Aladdin), ethylene glycol (EG, 99%, Aladdin), tetrabutyl titanate (TBT, 98%, Aladdin), and potassium hydroxide (KOH, 99%, Macklin), sulfuric acid (99%, Xilong Scientific), deionized water (it's made by laboratory instruments).

Synthesis of Diol Monomer Containing Amide Bond (DSE). Specifically, in a 100 mL round-bottomed flask, 23.03 g (0.1 mol) of dimethyl sebacate, 12.82 g (0.21 mol) of ethanolamine, and 20 mL of DMF were added successively. After purged by N_2 for 30 min, 0.2 mL (0.001 mol) of sodium methanol was added to the system and sealed. The round-bottomed flask was placed in an oil bath at 65 °C and stirred for 12 h. At the end of the reaction, the liquid was recrystallized three times with 50 mL of deionized water to obtain 21.34 g of white crystals, with a 67% yield.[30]

Synthesis of PEA Prepolymer. 20.22 g (0.1 mol) of SA and 30.44 g (0.095 mol) of DSE were added to a 100 mL three-necked flask. The system was placed in an oil bath equipped with a magnetic stirrer set at 100 rpm; the temperature was heated to 180 °C. The reaction was purged with nitrogen for 3 h. After the mixture was cooled, a milky white solid was obtained.

Synthesis of PEA. The prepared PEA prepolymer, 1.55 g (0.025 mol) of glycol, and 0.08 g of TBT were added to the high-pressure reactor. The reactor purged with nitrogen was heated to 180 °C and stirred at 100 rpm for 3 h. The vacuum in the reaction system was lowered to a level of 100 Pa, followed by a gradual increase in the temperature to 230 °C. The reaction was terminated when the stirring current of the reactor exceeded 1 A.

Chemical Recycling of PEA. 20 g of PEA-4 and 140 mL of water containing 10 wt % KOH were added to a high-pressure reactor equipped with a magnetic stirrer. Exhaust the air in the reactor was ejected by nitrogen. Then the system was closed, and the reactor was heated to 100 °C and stirred at 100 rpm for 3 h. The product was a slightly yellow liquid, and the

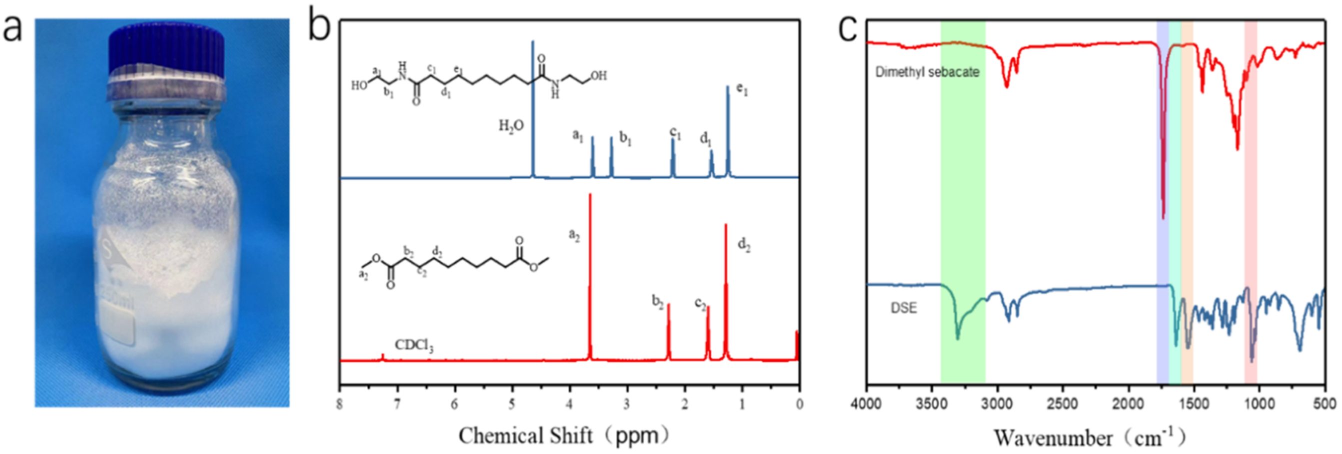


Figure 1. (a) Photo of DSE. (b) ^{1}H NMR spectra of monomers (DSE). (c) FT-IR spectra of DSE and dimethyl sebacate.

pH value of the product was adjusted to neutral by diluting sulfuric acid at a concentration of 10%. The white solids of sebacic acid collected after filtration were washed three times with deionized water, dried, and weighed. The filtrate was recrystallized in water three times. The solid products DSE after recrystallization were collected and weighed after drying.

Repolymerization of PEA. REPEA was prepared from recycled sebacic acid and DSE in a certain proportion according to the above method, and the above experimental operations were repeated.

Characteristic. The ^{1}H nuclear magnetic resonance (^{1}H NMR) spectra of the monomers were recorded with an AVANCEIII Bruker NMR spectrometer (Bruker, Switzerland) with D_2O as the deuterated solvent, operating at 600 MHz, respectively. The samples were dissolved at a concentration of 5 mg in 0.5 mL, and the chemical shift of the solvent was 4.79 ppm.

Differential scanning calorimetry (DSC) measurements were performed with a TA 250 instrument (TA Instruments) under a nitrogen atmosphere. PEAs were first heated to 150 °C at a heating rate of 50 °C/min and then cooled to −50 °C at a rate of 50 °C/min; after an isothermal treatment for 3 min, a second heating cycle was executed at a rate of 10 °C/min.

Thermogravimetric analysis (TGA) was carried out with a TG 209F3 instrument (NETZSCH Instruments, Germany); the mass was set at 100 °C for 10 min and then heated to 700 °C at a rate of 10 °C/min under a nitrogen atmosphere.

Fourier transform infrared spectroscopy (FT-IR) spectra were recorded with a Micro-FT-IR Cary660 spectrometer (Agilent) in the wavenumber range of 400−4000 cm^{-1} with a resolution of 4 cm^{-1}, and the signal was averaged over 32 scans at room temperature.

A tensile test was carried out on a universal mechanical testing machine (Suns Instruments, China), in which the samples were obtained by a vacuum laminator at 10 MPa and 150 °C for 8 min. The transparent yellow film with a thickness of 0.2 mm was cut into a dumbbell shape, and then a 100 N load cell was used at a speed of 10 mm min^{-1} at room temperature. The mechanical data were obtained by a mean value of five specimens of each sample.

The melt index of the material was measured on the melt flow rate meter (HDS-1800, China), the polymer is prepared into regular particles by the granulator, it is put into the barrel of the instrument that has been preheated at 190 °C for 30 min, and then put on 2.16 kg weights to squeeze the piston rod down. After uniform discharge, samples are collected every 15 s; 3−5 samples are collected, and after cooling, they are placed on the balance, respectively, weighing their mass and taking the average value. After this, the average value is input into the test instrument to calculate the result.

■ RESULTS AND DISCUSSION

Preparation of Amide Diol Monomer (DSE). According to previous literature, long-chain aliphatic diol monomers can be purified through recrystallization with water.[3] As shown in Scheme 1a and Figure 1a, DSE produced from the reaction of dimethyl sebacate with ethanolamine forms a white, crystalline powder. Typically, after three cycles of recrystallization, the yield of DSE exceeds 60%. The post-treatment operation is straightforward, requiring only pure water for recrystallization followed by drying. Additionally, this method enables the production of more than 500 g of monomer per reaction.

Figure 1b demonstrates the successful preparation of DSE, confirmed by comparing its ^{1}H NMR data for dimethyl sebacate. The peak at 3.60 ppm belongs to the proton of the methylene group adjacent to the hydroxyl, while the peak at 3.28 ppm is attributed to the methylene proton next to the amide. These two characteristic peaks indicate the occurrence of the transesterification reaction between the amino group of ethanolamine and the ester group of dimethyl sebacate, forming hydroxyl and amide bonds. In addition, the peaks at 2.20, 1.53, and 1.25 ppm correspond to the methylene proton within the sebacate chain. These data strongly confirm the high efficiency of the amidation reaction between the amino and ester groups.

Figure 1c shows the infrared spectra of the DSE monomer and its precursor, dimethyl sebacate. In the spectrum of dimethyl sebacate, a pronounced peak of ~1740 cm^{-1} C═O ester is very obvious, and it completely disappears in the white powder obtained after the reaction is finished and purified; this phenomenon indicates that the ester bond in the raw material reacts with the amino group, and the carbonyl peak in the ester bond is converted to the carbonyl group in the amide group. The presence of ethanolamine in the reaction system is evidenced in the DSE spectrum by distinct peaks at ~1635 cm^{-1} (amide I, C═O stretching) and ~1541 cm^{-1} (amide II, combination of C−N stretching and CO−NH bending vibrations). The spectra of DSE also indicate the appearance of a strong H-bond peak at ~3300 cm^{-1} (N−H H-bonded) with a shoulder at ~3400 cm^{-1} (O−H H-bonded). The above

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data strongly proved the successful preparation of the amide diol monomer. The DSC curve of the amide diol is shown in Figure S1, the melting point of the monomer is 133.6 °C, so in the subsequent polymerization, we started to stir when the temperature of the reactor reached 150 °C.

Synthesis and Characterization of PEA. To prepare poly(ester amide) materials with excellent mechanical properties, different feeding ratios are selected for the reaction; the specific ratios are shown in Table 1, and the polycondensation reaction can be seen in Scheme 1b. From the table, we can see that the input of sebacic acid is 0.1 mol, and then the total input of DSE and ethylene glycol is 0.11 mol. Glycol is incorporated to modify the ratio of amide bonds, and its high reactivity contributes to the enhanced mechanical properties of the resulting polymer. It is known from the literature that in the production process of polyester, the addition of diol is often excessive, which ensures that the polymer has a high viscosity and molecular weight.[31] Subsequent experiments found that the prepared PEA could not be dissolved in DMF, THF, CH_3Cl, even HFIP, and other solvents, so the tests of the polymers such as ^1H NMR and GPC could not be carried out normally. So, we use the melt flow rate of the test material to indirectly prove that the material has a relatively high molecular weight. As shown in Figure S3, melt indices of PEAs, commercially available polylactic acid (PLA), and high-density polyethylene (HDPE) were measured at 190 °C and loaded at 2.16 kg. The melt indices of PEAs were higher than those of HDPE but lower than those of PLA, indicating that the material had high viscosity and high molecular weight similar to those of commercially available products.

Table 1. PEA Polymer-Specific Feed Ratio

	DSE (mol)	SA (mol)	EG (mol)
PEA-1	0.07	0.1	0.04
PEA-2	0.08	0.1	0.03
PEA-3	0.09	0.1	0.02
PEA-4	0.095	0.1	0.015

The infrared spectrum of the polymer is shown in Figure 2a. All PEAs show the characteristic IR peaks at ∼1732 cm^{-1} C═O ester, ∼ 1635 cm^{-1} amide I, C═O, and ∼1541 cm^{-1} amide II, C−N + CO−NH bend. The IR spectra of PEAs also indicate the appearance of a strong H-bond peak at ∼3300 cm^{-1} (N−H H-bonded) with a shoulder at ∼3400 cm^{-1}(O−H H-bonded). These details indicate the successful synthesis of PEA at different feed ratios.[7,32−35]

As shown in Figure 2b, both the melting point (T_m) and the glass-transition temperature (T_g) of PEA increased with the increase of amide bond content, which was consistent with the behavior of amide bonds in polymer; the introduction of amide bond has a great influence on the crystallization of polymer and the ability of molecular chain movement. The introduction of the amide bond can improve the crystallinity of the material and thus increase the melting point of the material. However, the high crystallinity in the material will hinder the movement of the molecular chain segment so that the glass-transition temperature of the material will also be increased to a certain extent. Among them, the melting point of PEA-4 reached 108 °C, which exceeded the boiling point of water at atmospheric pressure (100 °C), which greatly expanded the application range of the material. As shown in Figure 2c,d, the degradation temperature of each proportion of PEA is about 300 °C, which

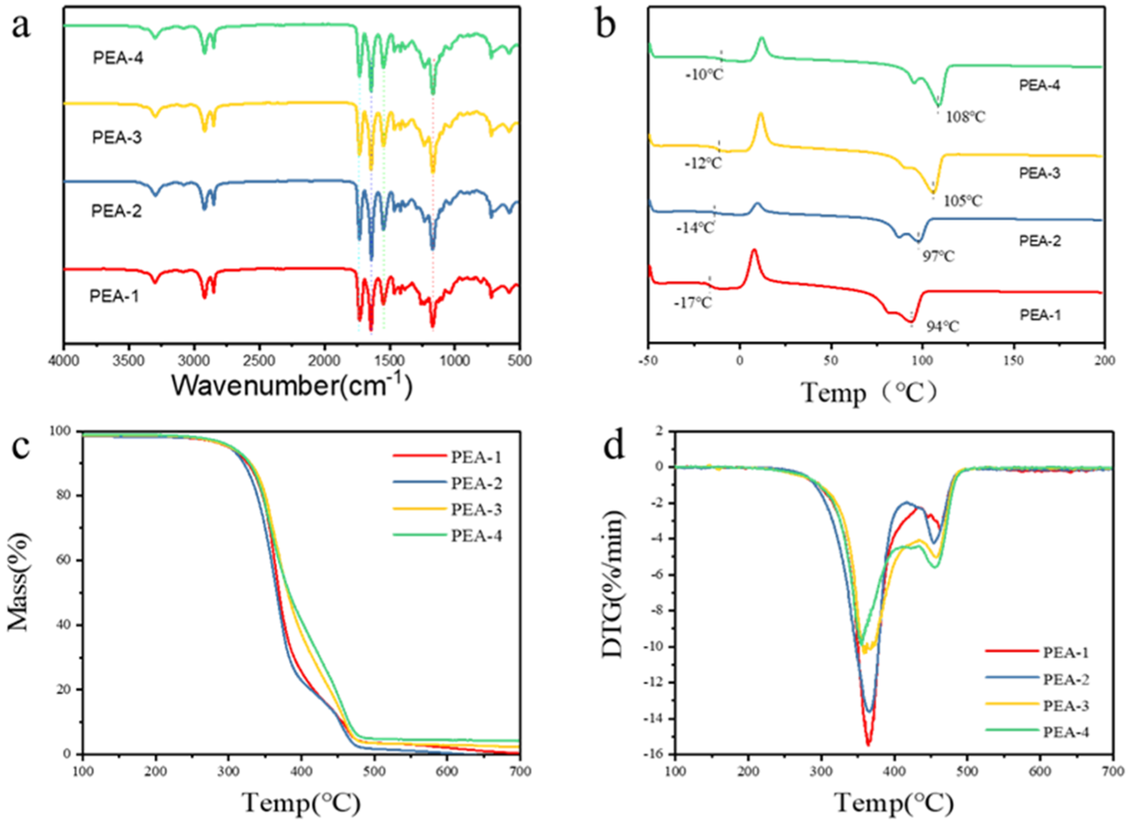

Figure 2. (a) FT-IR spectra, (b) DSC, (c) TGA, and (d) DTG curves of poly(ester amide)s.

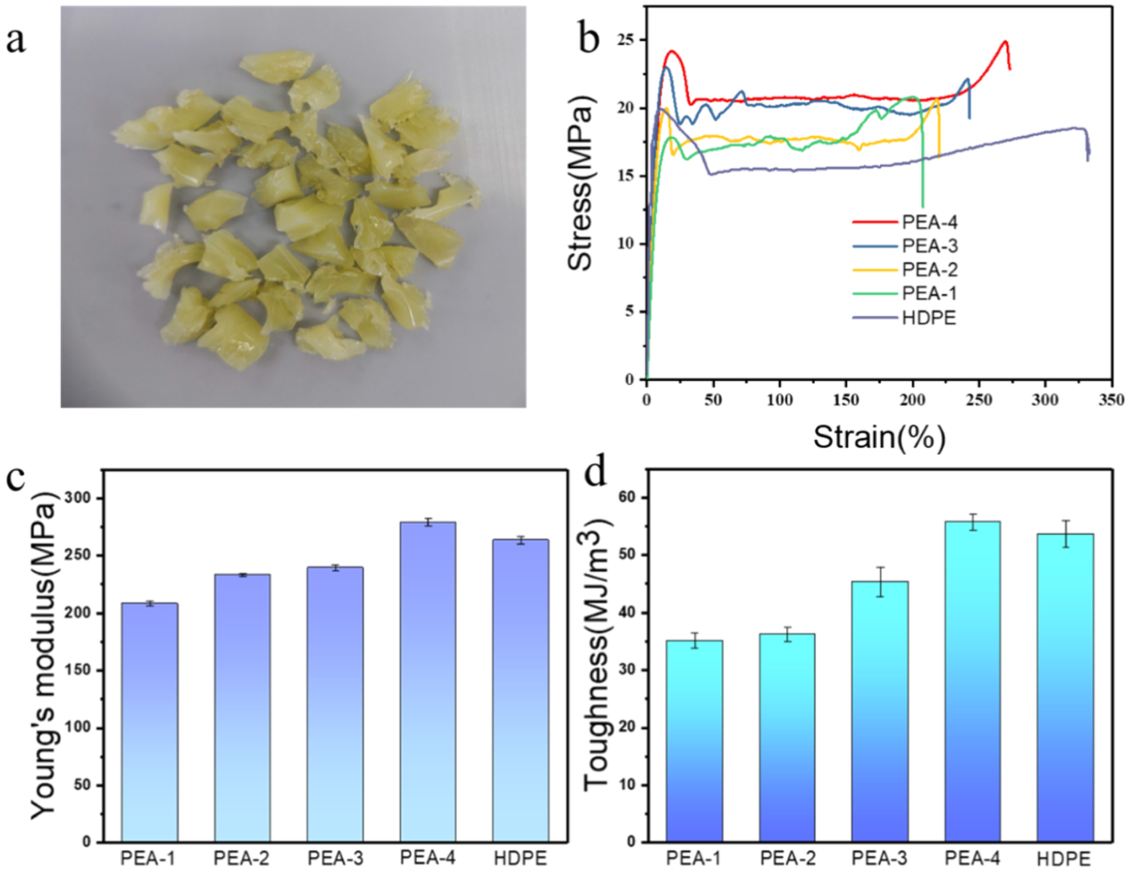

Figure 3. (a) Photo of PEA-4, (b) strain−stress curves, (c) a bar chart of Young's modulus, and (d) the tensile toughness of PEAs and HDPE.

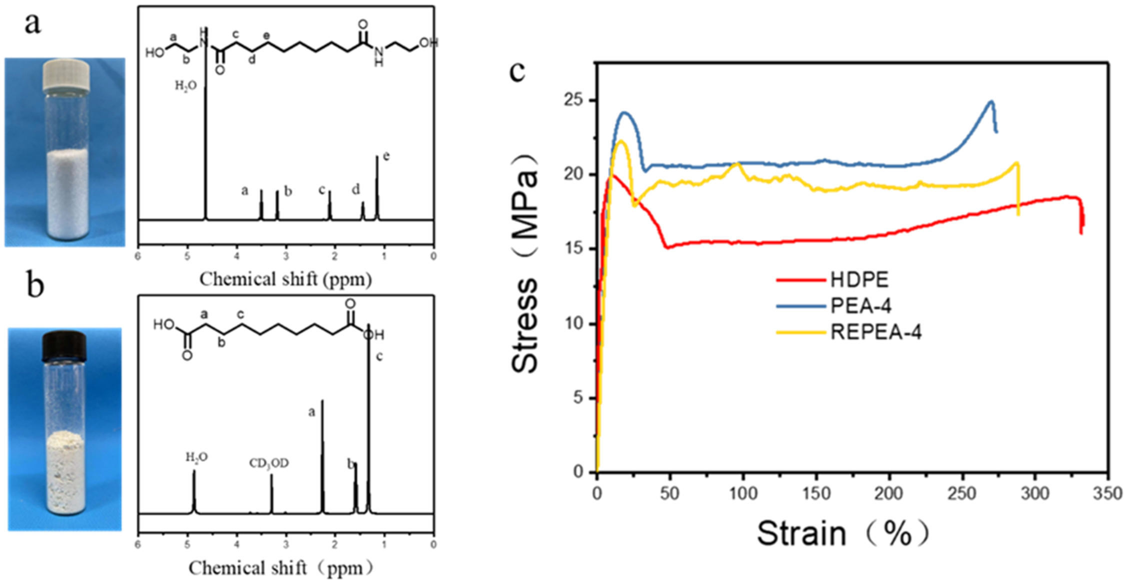

Figure 4. ^{1}H NMR spectra of DSE (a) and SA (b) obtained by chemical recovery and (c) strain−stress curves of PEA by repolymerization.

has a high processing temperature range. As can be seen from Figure 2d, the degradation of the polymer is divided into two stages, first the degradation of the ester group and then the degradation of the amide bond, which is in line with the description in the literature.[6]

To evaluate the mechanical properties of PEA-1, PEA-2, PEA-3, and PEA-4, polymer films were subjected to tensile tests. All polymer films showed good mechanical properties with a tensile strength in the range of 20−25 MPa, a toughness of 35.1−55.7 MJ/m^3, and elongation at break of 210−280%. With the increase of the DSE content and the increase of amide bond density, the mechanical properties of the polymer are gradually improved. It can be seen that the fracture stress and strain of PEA-4 reach a maximum of 25 MPa and 280%, followed by PEA-3, PEA-2, and PEA-1. We found that the mechanical behavior of the polymer is very similar to that of the widely used high-density polyethylene (HDPE), and the collected results are provided in Figure 3b. The specific values are shown in Figure S2 and Table S1; the polymer is expected to be an alternative to HDPE.

It can be seen from Figure 3c that with the increase of the density of amide bonds in the polymer system, the Young's modulus of the material also increases, and the hardness of the material increases. This result is also consistent with the initial idea that the amide bond can improve the strength of the polyester material. More importantly, the modulus of PEAs is very close to that of HDPE. Figure 3d shows the tensile toughness of the PEA material, which also increased with the increase of amide bond density. These data indicate that the amide bond plays a role in regulating the properties of polyester materials. Among them, PEA-4 possesses higher tensile toughness than that of HDPE. It is concluded that PEA-4 shows better mechanical properties than that of HDPE, which indicates a very promising biobased and recyclable materials.

Hydrolysis and Repolymerization of PEA. Since PEA contains a large number of ester groups, according to the literature,[3] hydrolysis experiments we performed on the polymer and two white solid powders were obtained by separation. The two powders were analyzed by ^{1}H NMR. As shown in Figure 4a,b, the ^{1}H NMR spectrum of white powder obtained by recrystallization is consistent with the ^{1}H NMR spectrum of DSE. The ^{1}H NMR spectrum of the white powder filtered after acidification matches that of sebacic acid. Details are shown in Figures S3 and S4. It was proved that the products obtained by PEA hydrolysis were sebacic acid and DSE with yields of 92.8−96.2 and 73.1−75.8% (Table S2), respectively. Owing to the low price of ethylene glycol, the high cost of recovery, and the complexity of the process, ethylene glycol is not recovered after polymer hydrolysis. In the polymerization stage of recycled raw materials, fresh glycol is added to react with recovered DSE and sebacic acid. DSC test of REPEA-4 shows that its T_m and T_g values are basically consistent with those of PEA-4 (Figure S5), and the FT-IR curve of REPEA-4 is shown in Figure S6. Tensile tests of the material obtained after secondary polymerization are shown in Figure 4c. The properties are not significantly different from those of the first prepared polymer, indicating that the material can achieve a complete closed-loop recovery process and has a great advantage over the nondegradable petroleum-based material HDPE in application.

CONCLUSIONS

In summary, this study has developed a novel method for the polymerization of high-performance poly(ester amide)s. And the mechanical properties of the polymers are similar to high-density polyethylene (HDPE), which is superior to all reported linear poly(ester amide)s. In addition, the material also poses the property of complete alkaline hydrolysis, which can be completely depolymerized into monomers and repolymerized, forming a closed-loop recycling process. This study provides a new idea for replacing polyethylene materials, solving the shortage of fossil resources, and protecting the environment.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.4c04503.

DSC second heating curves of DSE; Stress−strain curve of PEA-1−4; Column chart of melting index of PEA PLA and HDPE; Summary of PEA thermodynamic properties; NMR spectra of DSE after hydrolysis; NMR spectra of SA after hydrolysis; Repeated monomer yield from chemical recovery of PEA-4; DSC curve of REPEA-4; and FT-IR curve of REPEA-4. (PDF)

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Notes

The authors declare no competing financial interest.

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