APPLIED SCIENCES AND ENGINEERING

Closed-loop recyclable plastics from poly(ethyl cyanoacrylate)

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Ethyl cyanoacrylate is a highly reactive monomer that has been used nearly exclusively to make Super Glue and related fast-setting adhesives. Here, we describe transformation of this highly abundant, readily available monomer into a closed-loop recyclable plastic that could supplant currently used (and often unrecycled/unrecyclable) plastics, such as poly(styrene). We report polymerization conditions, plastic-processing methods, and plastic-recycling protocols for poly(ethyl cyanoacrylate) plastics that make the Super Glue monomer a viable starting material for a next generation of closed-loop recyclable plastics. The processes described are scalable, and the plastics can be recycled in a closed-loop process with >90% yields, even when combined with a heterogeneous mixture of other types of plastic.

INTRODUCTION

Most plastics are made from petroleum-derived feedstocks, and the recycling and reuse of these plastics are energy intensive, time intensive, and inefficient, thus leading to both pollution and the accumulation of plastic waste in landfills. To address these drawbacks, we have developed a plastic based on poly(ethyl cyanoacrylate) (PECA), which is prepared from the monomer ethyl cyanoacrylate (more commonly known by the trade names Super Glue and Krazy Glue). This PECA plastic is derived from nonpetroleum starting materials, is scalable and economically competitive with existing petroleum-based products (i.e., the monomer is commercially available in large quantities and is low cost), and has mechanical properties that are commensurate with commercial plastics. Furthermore, recycling is easily performed in a closed-loop process by thermally cracking the polymer and distilling the monomer.

The monomer, ethyl cyanoacrylate, is produced on the ton scale from methanol, chloroacetic acid, and sodium cyanide, which themselves are sourced from hydrogen, nitrogen, methane, carbon monoxide, chlorine, and sodium hydroxide (i.e., none are generated from petroleum) (*1*–*7*). The monomer rapidly polymerizes in biological systems, and the polymer, PECA, has been deemed biocompatible in both in vivo and in vitro studies (*8*, *9*), which is why they are used frequently in biomedical applications (*10*–*12*). Still, the monomer and the polymer have been used only rarely outside of the context of adhesives (*13*–*16*).

To develop poly(ethyl cyanaoacrylate)-based plastics and address the scalability, mechanical properties, and the feasibility of a closed-loop recycling process, we first needed a reliable and reproducible method for preparing the polymer. When Super Glue is used, adventitious water or surface functionalities initiate anionic polymerization of the highly electrophilic ethyl cyanoacrylate monomer (*17*, *18*). Abundant initiation events lead to a broad distribution of short polymers [e.g., polydispersity index (PDI) of 1.56] with number-average molecular weight (M_n) values in the 30-kDa range (fig. S1) (*13*, *19*). Short polymers are acceptable for glues, but they do not lead to plastics with good, reproducible, and tunable mechanical properties. Because polymer length directly affects mechanical properties, and because long polymers are needed to achieve polymer entanglement in plastics, our first task was to develop controlled polymerization conditions for ethyl cyanoacrylate that deliver long polymers with low polydispersity values. Moreover, we sought simple reaction conditions that, ideally, can be translated to industrial scale.

RESULTS

Polymerization of ethyl cyanoacrylate

Initial experiments demonstrated that polymerization reactions must be conducted in poly(ethylene) or poly(propylene) reaction vessels to avoid adhesion of the in situ formed PECA to the container itself (*20*). Many sizes and shapes of such reaction vessels are available and can be generated easily, thus enabling rapid and simultaneous polymerization and shaping of PECA plastics directly in a mold. The fundamental challenge with polymerization of ethyl cyanoacrylate is its high reactivity, which leads to many initiation events, short polymers, and release of heat, the latter being undesirable—from a safety perspective—on an industrial scale. We screened a variety of initiators and reaction conditions, ultimately selecting 25 M monomer in acetone, with initiation occurring at 23°C, open to air, by the addition of substoichiometric quantities (e.g., 5 mole percent) of dimethyl sulfoxide (DMSO) (*13*, *21*). As the polymerization reaction proceeds, the solution becomes a transparent solid, and the reaction continues until the majority of the monomer is consumed. To probe the kinetics of the polymerization process, we followed the course of the reaction using ^{1}H nuclear magnetic resonance (NMR) spectroscopy. For these studies, the reaction was performed under more dilute conditions (12.5 M monomer) than the bulk polymerization reaction so that we could obtain enough data for kinetic analysis before the sample became sufficiently solid to be unreadable by the broadband NMR probe. Under these conditions, a clear change in reaction rate is observed as the first 20% of monomer is converted to polymer (Fig. 1A), and during this period, the solution in the NMR tube transforms from liquid to a viscous liquid to a semisolid material. Polymerization in the semisolid (i.e., beyond approximately 20% conversion of monomer) follows zero-order kinetics

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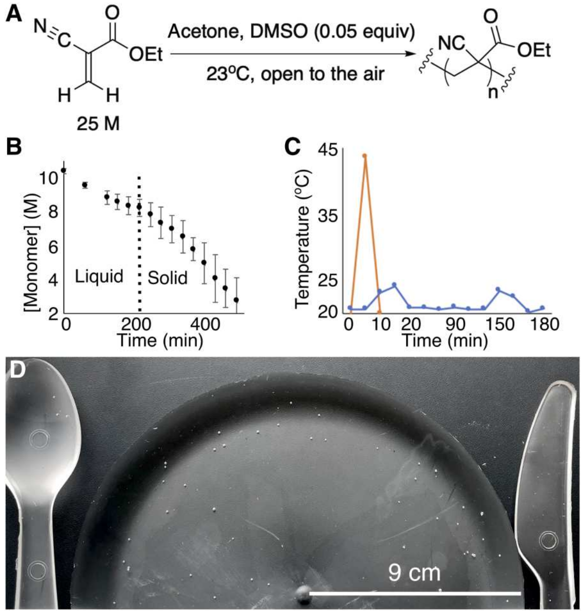


Fig. 1. Direct polymerization and casting of ethyl cyanoacrylate. (**A**) A representative polymerization reaction. (**B**) Monomer consumption was monitored by ^{1}H NMR using 1,3,5-trimethoxybenzene as an internal standard. The data are the average of three separate experiments using 12.5 M monomer (35 mg) in acetone-d_6, with 0.05 equivalents of added DMSO (relative to monomer) at 23°C. The error bars represent the SDs from these averages. The vertical dotted line represents the approximate time point when the viscous liquid transformed into a transparent semisolid in the NMR tube. (**C**) The blue line represents the temperature of a reaction mixture when 30 g of ethyl cyanoacrylate (25 M in acetone) was polymerized by the addition of 0.05 equivalents of DMSO at 23°C. The orange line depicts the temperature for an identical reaction, but where 0.0003 equivalents (0.1 ml) of commercial *N*,*N*-dimethyl-*p*-toluidine was added instead of DMSO. The temperature of the reaction mixture was measured using an infrared thermometer. (**D**) Photograph of a spoon, knife, and plate that were prepared by polymerizing 25 M ethyl cyanoacrylate in acetone at 23°C in shaped poly(propylene) molds by the addition of 0.05 equivalents of DMSO. The shaped PECA products were removed from the molds after 48 hours.

that are consistent with a diffusion-controlled process (rate constant = 0.021 $M\ s^{-1}$). In the absence of added DMSO, ethyl cyanoacrylate does not polymerize, even after a week of stirring in acetone, thus confirming the critical role of DMSO in facilitating the reaction (fig. S2).

Using the 25 M polymerization conditions, we use small quantities of DMSO (e.g., 640 μl for 30 g of monomer) to obtain polymers with M_n values up to 1877 kDa. Polymer entanglement is predicted to begin when PECA is longer than M_n = 30 kDa (*22*); thus, these polymers should afford plastics with substantial polymer entanglement. Likewise, the PDI values are low for the polymers, indicating that most of the polymers will be large enough to become entangled. In one representative example, the addition of 0.05 equivalents of DMSO to 30 g of monomer delivered polymer with an M_n value of 318 ± 43 kDa and an average PDI value of 1.024 ± 0.014.

Heat flow in the polymerization reactions is also low: The temperature of the reaction mixture rose only 3.5°C during the polymerization reaction (Fig. 1B, blue line) for a 30-g batch of monomer. In contrast, *N*,*N*-dimethyl-*p*-toluidine leads to an exothermic and less controllable polymerization reaction (Fig. 1B, orange line); analogous results are obtained with other common polymerization nucleophiles for ethyl cyanoacrylate. Overall, the mild conditions for the substoichiometric DMSO reaction enable straightforward and simultaneous polymerization and molding of shaped objects (Fig. 1C).

By the end of the polymerization reaction, little acetone or DMSO is present, likely due to slow evaporation during the polymerization process. To remove residual solvents, the solid plastic product is conditioned at room temperature, open to air, before performing mechanical testing (table S2). For example, we noted that only approximately 5%, by weight, of acetone and DMSO remained in the polymerized samples after polymerizing a 50-g batch of monomer for 24 hours and that no acetone was detectable by ^{1}H NMR after 48 hours (fig. S5).

Thermal annealing for improved mechanical performance

As cast, the plastics are brittle and weak, with values for ultimate tensile strength of only 5 ± 3 MPa and 8 ± 4% for elongation at break, as indicated by the black data in the stress-strain curve in Fig. 2A. Differential scanning calorimetry (DSC) experiments of the as-cast PECA plastics reveal an exothermic peak at approximately 115°C (Fig. 2B); however, the peak disappears if the sample is cooled in the DSC instrument before thermal decomposition and then heated a second time. Presumably, this exothermic peak corresponds to a reduction of voids between polymer chains that exist in the as-polymerized polymer matrix. The introduction of heat allows for an increase in the uniformity of the material in a process called thermal annealing, which has been used to improve the thermal and mechanical properties of other plastics (*23*). The DSC thermogram of the second heating cycle shows a resolved glass transition temperature (T_g) at approximately 110°C, an endothermic transition at 180°C that we attribute to a reversible depolymerization-repolymerization process, and a sharp onset of degradation (T_o) at approximately 240°C (Fig. 2C) (*24*–*26*). Modulated DSC was used to confirm these transitions, with the T_g corresponding to a step transition in the reversing signal and reversible depolymerization-repolymerization and decomposition appearing in the nonreversing signal (fig. S8).

On the basis of these DSC results, we developed a thermal annealing process for strengthening PECA plastics. The process involves heating the conditioned, as-cast plastic to 130°C, which is above the T_g value but below the onset of thermal decomposition. Figure 2D shows that this annealing treatment resolves the T_g value in bulk samples of PECA in the same way that polymer was annealed in heat-cool-heat experiments in the DSC instrument (Fig. 2C). Figure 2E demonstrates that a heating duration between 20 and 30 min maximizes both the toughness and ductility of the materials, while the blue line in Fig. 2A reveals the impact of 20 min of annealing on the stress-strain curve relative to the as-cast material (black line). On the basis of previous studies on PECA adhesives (*26*, *27*), a slow equilibration between depolymerization and repolymerization likely occurs during this annealing step, although the equilibration reaction does not register on DSC until approximately 180°C. This equilibration process generates shorter polymers after prolonged annealing, as reflected in emerging peaks in gel permeation chromatography (GPC) data compared with the as-cast material (fig. S14). The maximum achievable values for ultimate tensile strength and elongation at break are 22.7 ± 10.8 MPa and

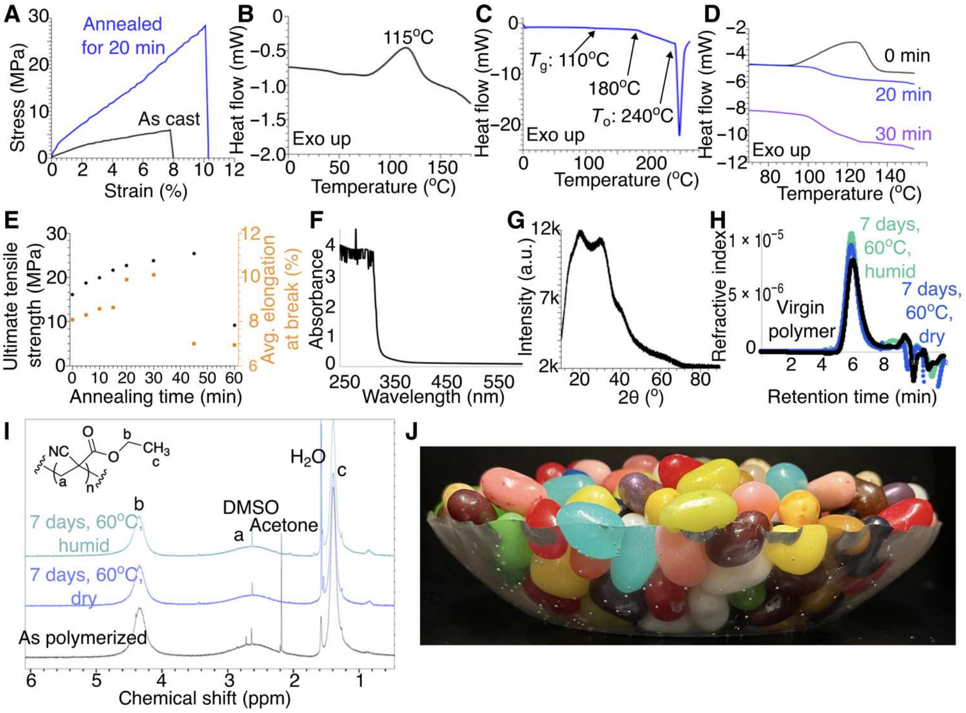


Fig. 2. Thermal, mechanical, and optical properties of PECA plastics. (**A**) Stress-strain curves of PECA plastic as-cast (black) and after 20 min of annealing at 130°C (blue). (**B**) DSC thermogram of the cured plastic upon first heating to 180°C, (C) after cooling, and then reheating the sample until decomposition is reached. (**D**) Overlaid DSC thermograms for samples extracted from bulk PECA plastic during the annealing process (i.e., not annealed directly in the DSC instrument). (**E**) Relationship between ultimate tensile strength and elongation at break values for PECA that was annealed at 130°C for different durations. The data points are the average of a minimum of five samples. (**F**) Ultraviolet-visible spectrum and (**G**) XRD pattern of cured PECA. (**H**) GPC traces and (**I**) ^{1}H NMR spectra of PECA plastic as polymerized and after 7 days in both humid and dry environments at 60°C. (**J**) Decorative fluted bowl made by thermoforming PECA plastic sheets over a ceramic mold.

9.9 ± 5.3%, respectively (Fig. 2E), within a single batch of polymer (M_n = 473 kDa, PDI = 1.076), and 22 ± 1 MPa and 17 ± 14% when averaging the results for five separate batches of polymer. These stress and strain values represent a 4.5-fold increase in strength for the annealed plastic relative to the as-cast material.

Thermoforming PECA plastic

The thermal properties of PECA are useful not only for annealing the plastics but also for thermoforming shaped products, thus enabling a product fabrication method other than direct molding. For example, a 2-mm-thick sheet of as-cast PECA was heated to 130°C until soft and then pressed over a bowl (i.e., mold) to create a bowl with fluted edges, as depicted in Fig. 2J. Water was stored in this bowl at 23°C for 7 days without changes in the physical or mechanical properties of the bowl.

Properties of bulk PECA

The density of the PECA plastic (ρ = 1.10 ± 0.04 g/ml) is between poly(acrylonitrile butadiene styrene) (ABS) and poly(methyl methacrylate) (PMMA), while the hardness (shore D hardness = 70 ± 2) is similar to atactic poly(styrene) (shore D hardness = 80 ± 10) (*28*). The PECA plastics are transparent, absorbing ≥10% of light at wavelengths below 325 nm, but with 90 to 100% transmittance of wavelengths above 400 nm (Fig. 2F). Transparent plastics typically have amorphous polymer morphology, which is the case for these PECA plastics as well, as confirmed by x-ray diffraction (XRD) measurements (Fig. 2G). The stability of these plastics to hot, moist environments is demonstrated by the lack of changes in GPC chromatograms (Fig. 2H) and ^{1}H NMR spectra (Fig. 2I) when PECA plastics are heated at 60°C for 7 days in both dry and humid environments. The annealed PECA plastics display ultimate tensile strengths and percent elongation values that are similar to atactic poly(styrene) (Fig. 3A), while the preannealed samples provide yield compressive strengths at break (51 ± 9 MPa) that are similar to commercial poly(propylene) (Fig. 3B) (*29*).

Solid-state depolymerization of PECA plastics

On an industrial scale, ethyl cyanoacrylate monomer is prepared by thermally cracking ethyl cyanoacrylate oligomers (*24*, *25*) (generated from the condensation of formaldehyde and ethyl cyanoacetate) and distilling the resulting monomers (*30*–*34*). On the basis of this precedent, we reasoned that it might be possible to recycle PECA plastics in the solid state by thermally cracking the long polymers within the plastics. Thus, heating solid PECA plastics to 210°C in the presence of phosphorous pentoxide (P_2O_5) [5% (w/w)], which is used to ensure anhydrous conditions and acidify any adventitious water that may be present in the glassware, resulted in conversion of plastic to monomers. The monomers were collected by distillation either into a Teflon flask or into a glass flask containing P_2O_5 [5% (w/w) with respect to the anticipated quantity of monomer], which is again used to prevent any repolymerization of ethyl cyanoacrylate (Fig. 4, A and B). In both cases, this process provides clean monomer (Fig. 4C) in 93% yield (Fig. 4D), along with a small quantity of black material that remains in the distillation flask. XRD

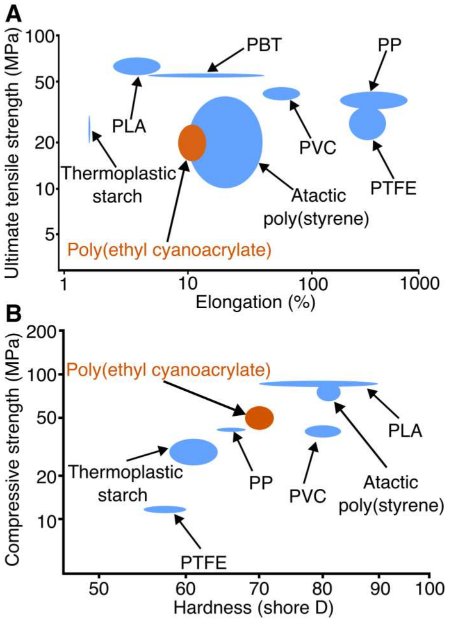


Fig. 3. Ashby plots for PECA plastics. (**A**) Ultimate tensile strength versus percent elongation for PECA plastics that were conditioned for 4 days and preannealed for 20 min. The orange oval represents the average of seven samples, where each sample was measured five times. The size of the oval represents the SD of these averages. Commercial polymers are labeled with standard abbreviations (*29*). (**B**) Compressive strength values versus shore D hardness for PECA plastics that were conditioned for 14 days but not annealed. The orange oval represents the average and SD from the average, as detailed in (A).

measurements and infrared (IR) spectroscopy of this black material reveal the presence of a (0 0 2) plane (Fig. 4E) and nitrogen-carbon double bonds (rather than triple bonds) (Fig. 4F) that is consistent with a nitrogen-substituted graphene precursor, which itself may be useful in other contexts (*35*, *36*). Repolymerization of monomer collected in the Teflon flask results in polymers with target lengths (i.e., M_n = 500 kDa) that provide ultimate tensile strength (24 ± 8 MPa) and percent elongation values (8 ± 2%) that are nearly identical to similar-length polymers prepared from commercial monomer (i.e., M_n = 500 kDa; ultimate tensile strength = 21 ± 4 MPa; percent elongation = 7 ± 2%).

The feasibility of closed-loop recycling of PECA is further improved by successfully cracking and distilling three sequential batches of solid PECA without cleaning the glassware in our apparatus between each batch (table S6). Existing industrial-scale infrastructure for thermally cracking oligomers of ethyl cyanoacrylate should be even more efficient at this type of sequential batch closed-loop processing than our proof-of-concept laboratory equipment. This industrial infrastructure includes a closed-kettle system equipped with a thin-film evaporator that continuously feeds liberated monomer into a double condenser that both condenses and purifies ethyl cyanoacrylate (*37*). Thus, it may be possible to convert this type of existing infrastructure into a recycling facility for PECA.

Depolymerization in mixed-waste streams

A typical unwashed plastic waste stream contains many types of plastics and a variety of contaminants, including paper, aluminum, food residue, adhesives, and any number of other components (*38*). Closed-loop recycling of solid PECA plastic is even possible from this type of crude mixture. In one demonstration, we combined 2 g of solid pieces of PECA plastic with 1.98 g of a shredded mixture of unwashed, unseparated municipal plastic waste. Distilled ethyl cyanoacrylate monomer was collected from this mixture in 75% yield. Since thermal cracking of PECA generates reactive ethyl cyanoacrylate, we attribute the lower yield to the reaction of the monomer with paper and other contaminants in the crude municipal plastic waste before all of the monomer distilled into the collection flask. Consistent with this hypothesis, if the mixture is composed of clean plastics, then monomer recovery increases to 92%. In this latter experiment, 6.8 g of solid PECA plastic was mixed with 4.2 g of ABS, poly(propylene), poly(carbonate), poly(styrene), poly(ethylene terephthalate), low-density poly(ethylene), high-density poly(ethylene), poly(lactic acid), poly(vinyl chloride), and PMMA; the specific proportions are presented in table S5.

DISCUSSION

The high glass transition temperature (T_g = 110°C), tensile strength, percent elongation when stressed, hardness, density, and amorphous morphology of PECA are all similar to atactic poly(styrene). Plastics made from atactic poly(styrene) are challenging to recycle: Mechanical recycling leads to substantial degradation of the polymer chain, and the high temperatures and complex conditions required to induce depolymerization create challenges for closed-loop recycling processes (*39*–*42*). As a result, poly(styrene) plastics are not collected in most curbside recycling programs. Nonetheless, molded and thermoformed poly(styrene) plastics are used routinely for making a variety of products, including yogurt cups; disposable plates, cups, and cutlery; and a variety of other products where rigid, economical plastics are desired. Poly(styrene) currently accounts for 6% of current plastic waste streams, and as such, replacing rigid poly(styrene)-based plastics with PECA plastics would increase the recyclability—particularly the closed-loop recyclability—of at least 6% of plastic waste (*43*). Owing to the excellent materials properties and ease of recycling, PECA may be useful in other contexts other than simply replacing poly(styrene), which would further improve the extent to which a plastic waste stream could be recycled.

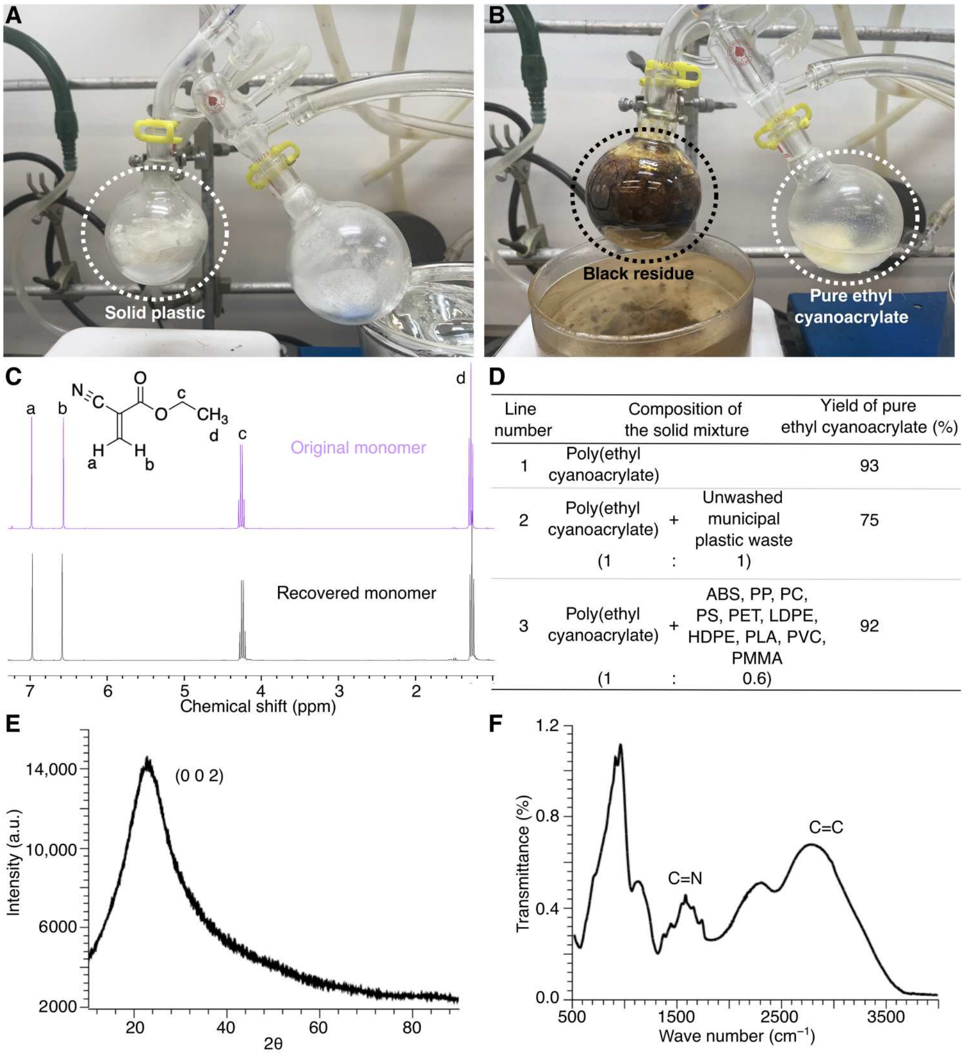
MATERIALS AND METHODS

General considerations

All polymerization reactions were carried out in polyethylene or polypropylene containers to prevent premature polymerization of the ethyl cyanoacrylate monomer and to prevent sticking of the polymer to the walls of the containers. Polymerization reactions were conducted in a laboratory fume hood, where the containers were open to air, at room temperature. ^{1}H NMR spectra were obtained using a Bruker Avance III 300 MHz with a broadband cryoprobe. Chemical shifts (δ) are expressed in parts per million (ppm) scale and are referenced to residual protons of the solvent $CDCl_3$.

Gel permeation chromatography

Molecular weight data were obtained using a Wyatt SEC-MALS system equipped with a Dawn 8^+ multiangle laser light scattering



Line number	Composition of the solid mixture			Yield of pure ethyl cyanoacrylate (%)
1	Poly(ethyl cyanoacrylate)			93
2	Poly(ethyl cyanoacrylate) (1	+ :	Unwashed municipal plastic waste 1)	75
3	Poly(ethyl cyanoacrylate) (1	+ :	ABS, PP, PC, PS, PET, LDPE, HDPE, PLA, PVC, PMMA 0.6)	92

Fig. 4. Transforming solid PECA plastic into liquid ethyl cyanoacrylate monomer. (**A**) Photograph of a distillation apparatus that contains solid PECA (dotted circle); P_2O_5 is in the receiving flask. (**B**) Photograph of the same apparatus as in (A) but after the PECA is cracked and the monomer is distilled. (**C**) ^{1}H NMR spectra showing the purity of the monomer that was collected from the closed-loop recycling process relative to commercial-grade monomer. (**D**) Yields of monomer in the closed-loop recycling process as a function of the composition of the solid mixture. (**E**) XRD spectrum of the black residue in the distillation flask in (B). (**F**) IR spectrum for the same material as in (E).

detector and an Optilab T-rEX refractive index detector and was analyzed using ASTRA software.

Differential scanning calorimetry

Data were recorded using a TA Instruments Q2000 Modulated Differential Scanning Calorimeter using 5- to 10-mg samples. Samples were heated from 0° to 180°C at a rate of 5°C/min with a modulation period of 60 s and a heating amplitude of ±1.6°C.

Mechanical testing

Tensile elongation tests were performed using a Labthink XLW (EC) instrument equipped with a 500-N load cell and serrated grips. All samples were conditioned for 48 hours in an ambient environment before testing. Tests were performed in accordance with ASTM D638-14. Tensile elongation tests were performed using an Instron model 5984 Universal Testing Machine equipped with Bluehill Universal analysis software. All samples were conditioned in an ambient environment, and tests were performed in accordance to ASTM D695-15.

X-ray diffraction

Measurements were obtained using a Rigaku Miniflex 600 at a rate of 10°/min and a step size of 0.02.

Optical characterization

Infrared spectroscopy data were recorded using a Nicollet iS20 Fourier transform IR spectrometer equipped with an attenuated total reflectance diamond plate. A background spectrum was recorded before each sample and was subtracted from the sample data. Samples were measured using 64 scans at a resolution of 4 cm^{-1} with a spacing of 0.482 cm^{-1}. Ultraviolet-visible spectroscopy data were obtained using a Beckman Coulter DU 800 Spectrophotometer from 800 to 200 nm at a rate of 1200 nm/min.

Supplementary Materials

This PDF file includes:
Synthetic Procedures
Figs. S1 to S17
Tables S1 to S6

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