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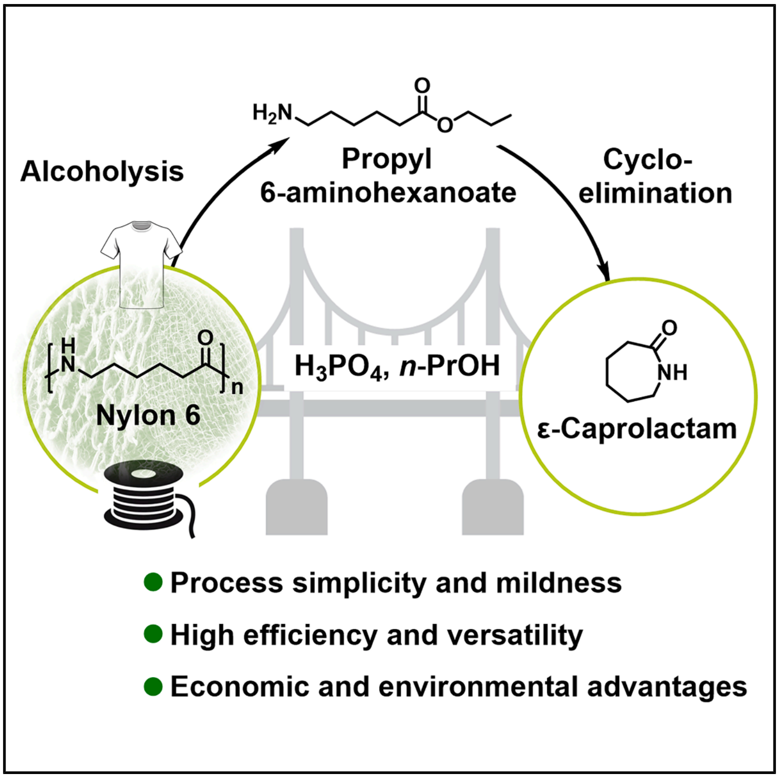
Alcoholysis of nylon 6 waste to ε-caprolactam promoted by phosphoric acid

Graphical abstract



Highlights

- Acid-catalyzed alcoholysis of nylon 6 enables selective production of ε-caprolactam
- Kinetic studies reveal that alcoholysis of nylon 6 is the rate-determining step
- Depolymerization of commercial nylon 6 feedstocks shows broad compatibility
- The process offers both economic benefits and environmental advantages

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In brief

Materials made from nylon 6 are widely used but difficult to recycle back into their original monomer, ε-caprolactam. This work introduces a simple acid-catalyzed alcoholysis method that enables efficient and selective depolymerization of nylon 6 into ε-caprolactam. Additionally, this process shows excellent compatibility with a range of real-world nylon 6 materials, including post-consumer fishing net waste, thread, 3D-printing filament, fabric, and carpet, offering a practical pathway toward closed-loop recycling with both environmental and economic benefits.

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Alcoholysis of nylon 6 waste to ε-caprolactam promoted by phosphoric acid

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THE BIGGER PICTURE Unlike traditional mechanical recycling approaches that often result in downcycled materials with inferior properties, the chemical depolymerization of thermoplastics to their constituent monomers represents a critical step toward achieving a circular economy for plastics recycling. Polyamide 6 (nylon 6), a semicrystalline thermoplastic, is widely used in the textile, automotive, electronics, packaging, fishing, and medical industries. However, its conventional production relies heavily on fossil resources, resulting in substantial greenhouse gas emissions. Nevertheless, its inherent chemical resistance and the lack of efficient recycling technologies often result in its accumulation in landfills or marine environments, thereby contributing to the global plastic pollution crisis. In this work, we report an acid-catalyzed alcoholysis strategy capable of selectively depolymerizing both model and post-consumer nylon 6 substrates to ε-caprolactam in high yield. This process exhibits notable techno-economic and environmental advantages over primary production pathways, thereby representing a promising approach toward the scalable implementation of closed-loop recycling for polyamide-based materials.

SUMMARY

Nylon 6 is widely used in high-performance materials. However, the inherent structural rigidity of nylon 6 inhibits catalytic depolymerization to its monomer, ε-caprolactam. Here, we demonstrate an acid-catalyzed alcoholysis process that depolymerizes nylon 6 to ε-caprolactam, achieving yields up to 74% using *n*-propanol as solvent and phosphoric acid as catalyst at 220°C for 2 h. We further applied the process to commercial nylon-containing products, demonstrating ε-caprolactam yields of 67% to 76%. Process modeling and techno-economic analysis estimated a minimum selling price of recycled nylon 6 at \$1.79/kg, 30% lower than the 5-year average market price of virgin nylon 6. These estimates depend on assumed costs and process performance and may vary with future conditions. Life cycle assessment indicated that nylon 6 produced via alcoholysis can reduce greenhouse gas emissions by up to 63% compared with primary production. Overall, this study demonstrates a facile, cost-effective, and environmentally beneficial process for nylon 6 chemical recycling.

INTRODUCTION

Nylon 6 is a semi-crystalline polymer synthesized via the ring-opening polymerization of ε-caprolactam.[1–3] Owing to its high strength, durability, and elasticity, nylon 6 has found numerous applications in the textile, automotive, electronics, packaging, fishing, and medical industries.[1–3] As a result, the annual global consumption of nylon 6 has reached 5.5 million metric tons

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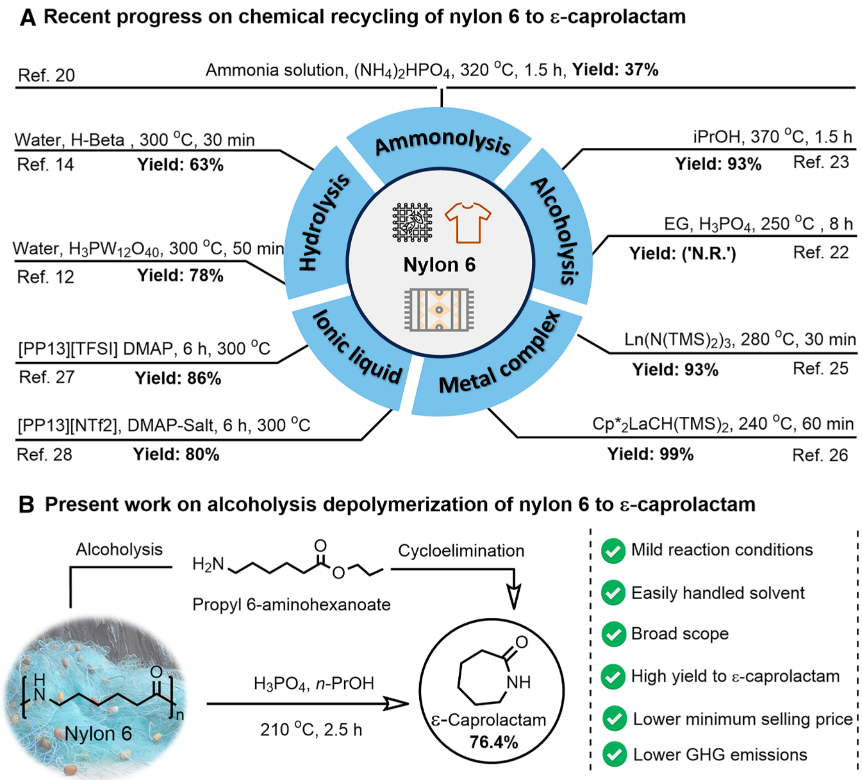

Figure 1. Overview of reported methodologies to produce ε-caprolactam from nylon 6

(A and B) (A) from the literature and (B) in this work. Chemical abbreviations and their full names can be found in the supplemental information.

(MMTs) as of 2020, less than 2% of which is produced from recycled monomer.[4] Meanwhile, the supply chain emissions for manufacturing ε-caprolactam in the US are 2.3 kg $CO_2e/kg_{\varepsilon\text{-caprolactam}}$ and require 60 $MJ/kg_{\varepsilon\text{-caprolactam}}$,[5] while nylon 6 is associated with 4 kg $CO_2e/kg_{nylon\text{-}6}$ of emissions and consumes 117 $MJ/kg_{nylon\text{-}6}$.[6] Moreover, due to its chemical resistance and a lack of effective recycling methodologies, nylon 6 waste often ends up in landfills or the ocean, contributing to the global plastic pollution crisis.[7]

Given that mechanical recycling results in compromised nylon 6 properties, such as reduced molar mass, tensile strength, and thermal stability, which are critical for high-performance applications, there is a need to develop viable methods for the chemical recycling of nylon 6 to its monomer, ε-caprolactam.[8–11] To date, numerous efforts have pursued chemical recycling of nylon 6 via hydrolysis,[12–16] pyrolysis,[17–19] ammonolysis,[20,21] alcoholysis,[22,23] hydrogenolysis,[24] organometallic catalysis,[25,26] ionic liquid-mediated depolymerization,[27,28] and base-mediated depolymerization.[29] Many of these methods are capable of selectively depolymerizing nylon 6 to ε-caprolactam, which can be subsequently reused to produce pristine nylon 6, enabling closed-loop recyclability (Figure 1A).

The selective transformation of nylon 6 to ε-caprolactam typically proceeds via one of two reaction mechanisms: (1) chain-end back-biting, where activation of both the terminal amide and opposite free amine group by an organometallic complex allows for the selective generation of ε-caprolactam from a nylon 6 end group during each catalytic cycle[25,26]; or (2) random scission, where amide bonds in nylon 6 are coordinated and activated by an acid catalyst, thereby making it susceptible to nucleophilic attack by water, alcohol, or ammonia. This latter process initially yields either 6-aminocaproic acid (via hydrolysis), an alkyl 6-aminohexanoate (via alcoholysis), or 6-aminohexamide (via ammonolysis) as an intermediate, all of which can subsequently undergo a cycloelimination reaction to produce ε-caprolactam.[22,23] The random scission mechanism can alternatively proceed in the presence of a strong base catalyst, which facilitates amide deprotonation, thereby generating a nucleophilic nitrogen anion. This anion can then attack the carbonyl group of the adjacent amide, driving intramolecular cyclization and resulting in the formation of ε-caprolactam.[29]

Here, inspired by previous findings where the combination of an acid catalyst and a nucleophile can synergistically promote amide bond cleavage of anilide and nylon 6,6,[30,31] we demonstrate a catalytic method for the selective depolymerization of nylon 6 to ε-caprolactam at yields up to 74% via phosphoric acid-catalyzed alcoholysis in *n*-propanol (*n*-PrOH) at 220°C for 2 h (Figure 1B). We further demonstrate the applicability of this method toward commercial nylon 6, including post-consumer fishing net waste, thread, 3D printing filament, fabric, and carpet, all of which were converted to ε-caprolactam at yields ranging from 67% to 76%. Techno-economic analysis (TEA) and life cycle assessment (LCA) estimate a 30% decrease in minimum selling price (MSP) for recycled nylon 6 (r-nylon 6) compared with primary manufacturing and a 63% decrease in resulting greenhouse gas (GHG) emissions.

RESULTS AND DISCUSSION

Establishing optimized reaction conditions for nylon 6 depolymerization

We initially hypothesized that an acid catalyst could serve as an active center for both alcoholysis and cycloelimination by coordinating with carbonyl oxygens, thereby enhancing their susceptibility to nucleophilic attack by alcohols. To this end, a range of different acid catalysts, including homogeneous Lewis and Brønsted acids, were screened in *n*-PrOH for nylon 6 depolymerization at 220°C (Figure 2B; Table S1). Uncatalyzed alcoholysis of nylon 6 at these conditions was ineffective, consistent with previous reports.[22] However, in the presence of Brønsted acids

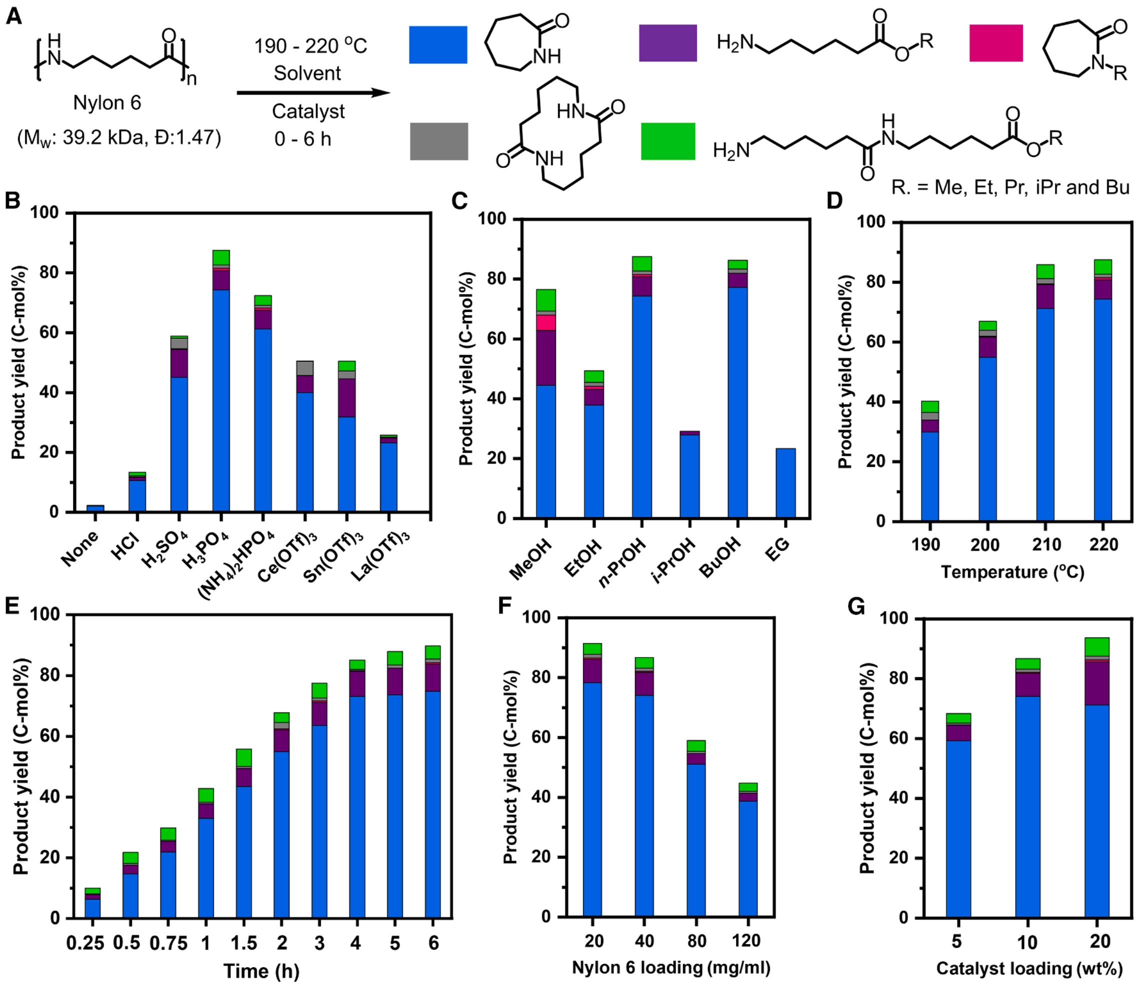

Figure 2. Influence of reaction conditions on nylon 6 depolymerization

Unless otherwise specified, the following reaction conditions were used: 200 mg nylon 6 pellet (Sigma-Aldrich), 0.2 mmol H_3PO_4, 220°C, 5 mL *n*-PrOH, 2 h.
(A) Nylon 6 depolymerization scheme.
(B) Catalyst screening results: HCl: 0.6 mmol, H_2SO_4: 0.3 mmol, and others: 0.2 mmol.
(C) Solvent screening results.
(D) Temperature screening results.
(E) Reaction time effects (200°C).
(F) Nylon 6 loading screening results (210°C, 2.5 h).
(G) Catalyst loading with respect to nylon 6 screening results (210°C, 2.5 h). Tabulated data are provided in supplemental information.

(HCl, H_2SO_4, and H_3PO_4), ε-caprolactam yield improved to 10.6%, 45.1%, and 74.4%, respectively, implying the presence of an acid catalyzes alcoholysis of nylon 6 while also potentially aiding subsequent cycloelimination of alcohol. Interestingly, the various acids demonstrated substantial differences in nylon 6 alcoholysis promotion, suggesting potential differences with respect to either acid solubilities or dissociation constants in *n*-PrOH. Aside from ε-caprolactam, alcoholysis also produces propyl 6-aminohexanoate and other dimeric intermediates, namely 1,8-diazacyclotetradecane-2,9-dione and propyl 6-(6-aminohexanamido)hexanoate (Figure S1). The use of a phosphate salt ($(NH_4)_2HPO_4$) likewise resulted in ε-caprolactam generation at a yield of 61%, confirming NH_4^+ ions, which can also act as Brønsted acids, are capable of coordinating carbonyl groups and thereby promoting nucleophilic attack by *n*-PrOH. By contrast, homogeneous Lewis acid catalysts such as $Sn(OTf)_3$, $Ce(OTf)_4$, and $La(OTf)_3$ afforded lower ε-caprolactam yields ranging from 20% to 40%. The superior performance of the H_3PO_4 system is primarily attributed to its stronger local Brønsted activation of the amide and to solvent and interfacial effects that enhance proton-mediated pathways relative to Lewis acid coordination.

Based on the promising resulting ε-caprolactam yields when combining H_3PO_4 and *n*-PrOH, we further examined the influence of solvents on reaction performance (Figure 2C; Table S2). Volatile alcohols were examined owing to their low cost and ease of separation. Methanol (MeOH) led to a moderate yield of 45%, higher than that achieved using ethanol (EtOH) (35%). These results were inferior compared with those for larger protic alcohols, where *n*-PrOH and *n*-butanol (*n*-BuOH) achieved yields of 74% and 77%, respectively. This is in line with previous observations where, under supercritical reaction conditions, higher yields of ε-caprolactam were typically obtained in those larger alcohols from the depolymerization of nylon 6 via alcoholysis.[22] Conversely, isopropanol (*i*-PrOH) resulted in a decreased yield of 28%, which was slightly higher than that obtained with ethylene glycol (EG) (23%). These differences in reactivity could be related to differences in solvent-substrate interactions (e.g., hydrogen bond donating or accepting propensity),[30] which could potentially enhance/reduce the electrophilicity of carbonyl groups or the nucleophilicity of utilized alcohols, thereby modifying the efficacy of a given system for the cleavage of amide bonds in nylon 6.[30]

Moving forward with *n*-PrOH as a viable solvent, depolymerization temperatures of 190°C, 200°C, 210°C, and 220°C were investigated (Figure 2D; Table S3). ε-Caprolactam yields generally increased with increasing reaction temperature, from ∼30% at 190°C to 74% at 220°C. The influence of reaction time was also investigated at 200°C (Figure 2E; Table S4). The production of ε-caprolactam was favored at longer reaction times, reaching a yield of 75% after 6 h. Interestingly, this increase was coupled with an increase from 2% at 0.25 h to 9% at 6 h in the production of propyl 6-aminohexanoate.

Next, owing to the effect that reactor sizing can have on process economics (*vide infra*), substrate loading was examined. Increased nylon 6 loadings resulted in decreased depolymerization extents (Figures S2 and S3) and yields of ε-caprolactam, from 78% at a loading of 20 mg/mL to 39% at 120 mg/mL (Figure 2F; Table S5). In turn, given the effect that the catalyst/substrate ratio appears to exhibit on process performance, the influence of catalyst loading on nylon 6 alcoholysis was also investigated. Unsurprisingly, increasing the catalyst loading from 5 to 10 wt % H_3PO_4 enhanced the ε-caprolactam yield from 59% to 74%. However, further increasing the concentration to 20 wt % H_3PO_4 resulted in a decreased ε-caprolactam yield of 71%, instead promoting the production of propyl 6-aminohexanoate (Figure 2G; Table S6).

Mechanistic insights into the depolymerization of nylon 6

To elucidate the reaction mechanism and kinetics for the depolymerization of nylon 6, a series of probe reactions on model substrates was conducted. It was initially postulated that the reaction occurs predominantly via a propyl 6-aminohexanoate intermediate (Figure 3A), as the compound was observed in many of the depolymerization reactions shown in Figure 2. The intermediate nature of propyl 6-aminohexanoate was verified using a purified standard, which underwent rapid, near-complete cycloelimination of *n*-PrOH (90% conversion) to ε-caprolactam in 15 min at similar conditions to those in Figure 3B. Further increasing the reaction time led to no significant variation in the reagent and product concentrations, suggesting the presence of a reversible step. This hypothesis was confirmed by subjecting pure ε-caprolactam to similar conditions, which led to the formation of propyl 6-aminohexanoate (Figure 3C), indicating an equilibrium between the two species.

Based on the observed product distributions and model compound results, an acid-catalyzed reaction mechanism involving consecutive alcoholysis and cycloelimination of *n*-PrOH is proposed (Figure 3E). In stage 1, the amide bond of nylon 6 is initially coordinated and activated by a Brønsted acid, making it susceptible to nucleophilic attack by *n*-PrOH to produce propyl 6-aminohexanoate. This can then undergo cycloelimination of *n*-PrOH during stage 2 to yield ε-caprolactam, with *n*-PrOH being regenerated in the process.

To estimate relative reaction rates for the system, a simple first-order, pseudo-homogeneous kinetic model was developed utilizing the network outlined in Figure 3A. For ease of calculation, and owing to the large excess of *n*-PrOH in the system, we assumed the alcohol concentration in the liquid phase to be constant and thereby lumped the effects of *n*-PrOH concentration into the apparent kinetic rate constants ($k_n' = k_n C_{n-PrOH}$). The ratio [$K_2^{eq} = k_{2f}/k_{2r}' = 8.0$] was assumed to be constant as defined by the equilibrium concentrations shown in Figure 3C. Finally, the system was further simplified by assuming that the equilibrium between nylon 6 and its depolymerized intermediates is negligible under the conditions studied.

Using this simplified system, experimental concentrations of ε-caprolactam and propyl 6-aminohexanoate were fit to a first-order power law model by optimization of the first-order kinetic constants: k_1' and k_{2f} (Figure 3D). A sum-squared-error cost function was utilized by assuming an initial molar nylon concentration equivalent to the molar concentration of monomer subunits within the nylon polymer, and the kinetic constant k_1' was determined to be 0.423 h^{-1}. However, k_{2f} demonstrated substantial initial guess sensitivity. As a result, a sensitivity analysis was implemented by fixing k_1', determining the parameters k_{2f} and k_{2r}' to be >20 h^{-1} and >2.5 h^{-1}, respectively. These results reinforce experimental observations suggesting the release of monomers from nylon 6 via alcoholysis proceeds slower than cycloelimination. The full calculation details are provided in the supplemental information.

Depolymerization of commercial nylon feedstocks to ε-caprolactam

Following our reaction optimization and mechanistic investigation, we explored depolymerization of commercial nylon 6 plastics, including commercial fishing nets (green and white), 3D printing filament, film, t-shirt fabric, carpet, and thread, all of which were chopped into pieces of ∼1 mm in length before reactions were conducted (Table S8 for materials characterization).[8,32] Nylon 6 powder and film both resulted in yields of ∼70% to ε-caprolactam under optimized reaction conditions (Figure 4; Table S7). Meanwhile, the depolymerization of fishing nets (green, white, or a mixture of both), a significant contributor to ocean plastic waste pollution,[33] achieved the highest yield of 76%. Apart from demonstrating that our process works on commercial polymers, this result also suggests that this chemistry is resilient to dyes and other additives found in commercial nylon 6 materials.

We also applied the system to carpet fibers, which are a major waste stream in landfills.[34,35] At optimized reaction conditions,

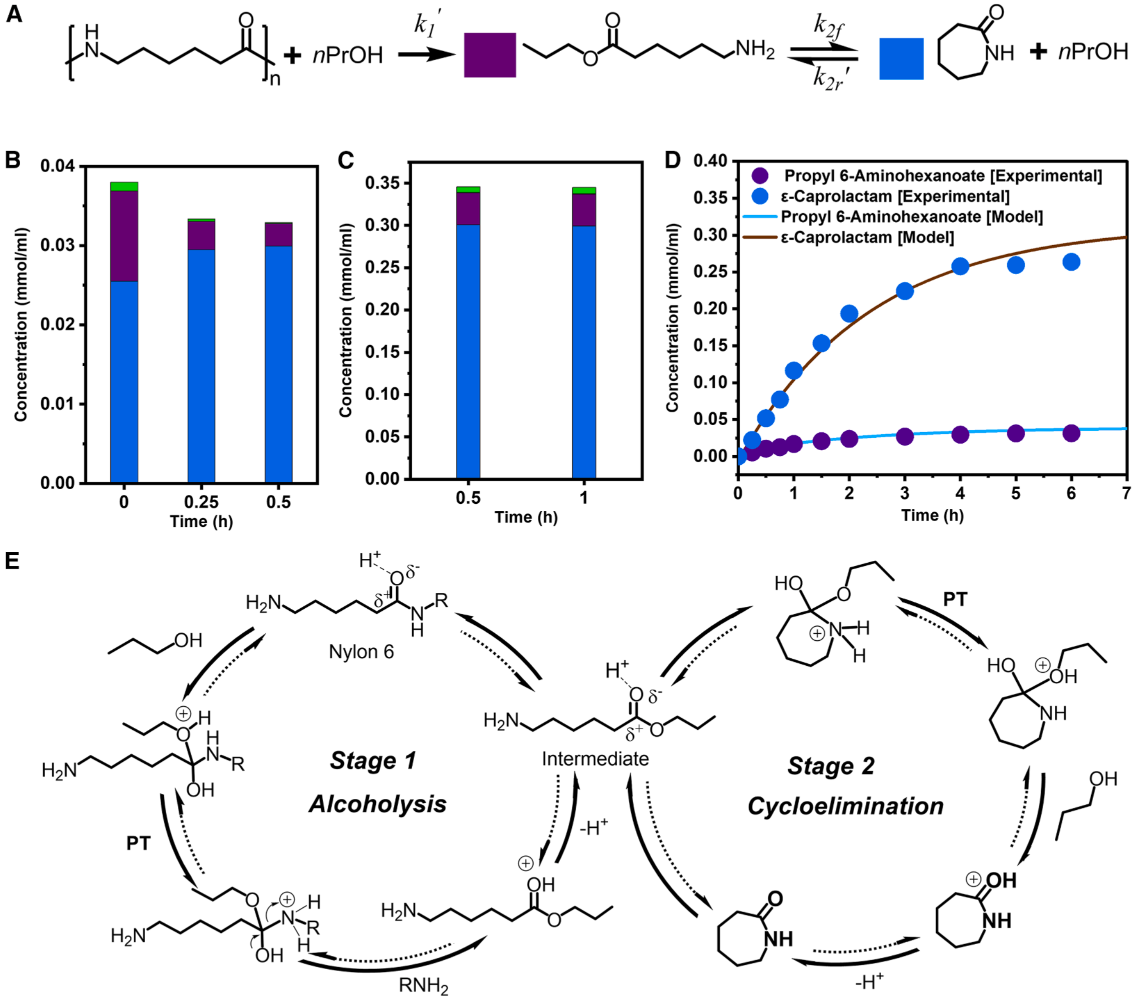

Figure 3. Elucidation of the reaction pathway by means of model experiments and kinetic analysis

(A) Initial reaction scheme used for the kinetic modeling of nylon 6 depolymerization.

(B) Concentration of propyl 6-aminohexanoate as a function of reaction time during ring closure. Reaction conditions: 0.293 mmol propyl 6-aminohexanoate, 0.033 mmol H_3PO_4, 180°C, 5 mL *n*-PrOH, 0–0.5 h.

(C) Concentration of ε-caprolactam as a function of time during ring opening. Reaction conditions: 1.77 mmol ε-caprolactam, 0.2 mmol H_3PO_4, 200°C, 5 mL *n*-PrOH, 0.5–1 h.

(D) Product concentrations (experimental and modeled) as a function of reaction time. Reaction conditions: 200 mg nylon 6, 0.2 mmol H_3PO_4, 200°C, 5 mL *n*-PrOH, 0–6 h.

(E) Proposed reaction mechanism for nylon 6 depolymerization via acid-catalyzed alcoholysis. The code for kinetic modeling is provided as a separate file, named Data S1: MATLAB scripts used for kinetic modeling. PT: proton transfer.

the carpet was converted to ε-caprolactam at a yield of 73%. Nylon thread, which is predominantly used in textiles, generated a yield of 75%. Nylon T-shirt fabric yielded 69%, while 3D printing filament containing 25 wt % glass fibers afforded a yield of 70%.

To further assess the impurity tolerance of this reaction, both washed and unwashed fishing nets recovered from the ocean were tested. Washed fishing nets resulted in an ε-caprolactam yield of 70%, whereas a yield of 68% was achieved for the unwashed nets. This indicates that impurities present in the unwashed nets do not compromise the deconstruction efficiency of the process. Finally, subjecting a multilayer film containing 25 wt % of nylon 6 and a variety of other materials to our process resulted in a yield of 69% of ε-caprolactam, further demonstrating the validity of our methodology toward mixed plastic waste.

Process modeling, TEA, and LCA of a conceptual nylon 6 depolymerization process

Based on the promising results obtained for ε-caprolactam yields, we conducted process modeling, TEA, and LCA to evaluate the economics and environmental impacts of nylon 6 depolymerization via alcoholysis. Process modeling was conducted with Aspen Plus v14.1, and the missing property data for the

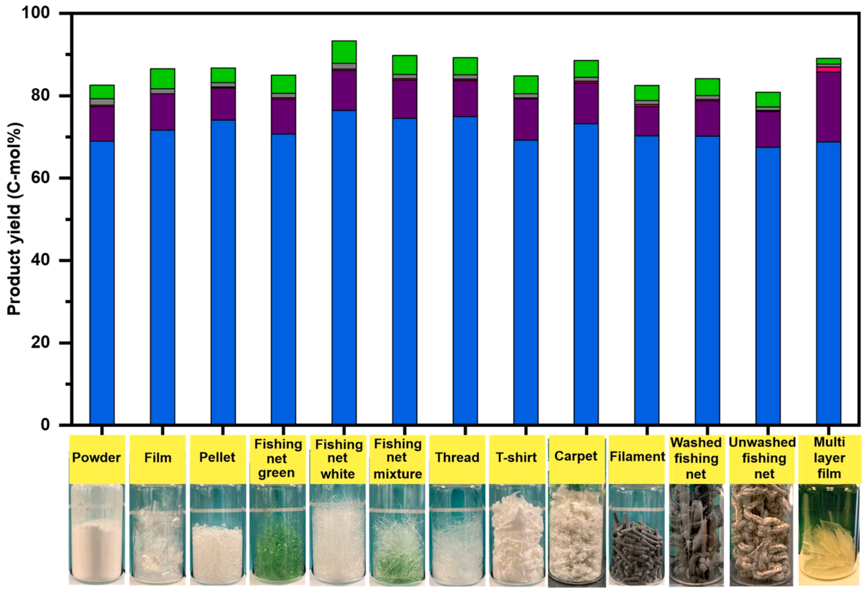

Figure 4. Depolymerization of commercial nylon 6 feedstocks to ε-caprolactam

Reaction conditions: 200 mg nylon 6 substrate, 0.2 mmol H_3PO_4, 210°C, 5 mL *n*-PrOH, 2.5 h. The composition of each nylon 6 material is described in Table S8. All yields were determined based on the actual content of nylon 6 and were corrected to account for the presence of impurities. Tabulated data are provided in the supplemental information.

oligomeric compounds were estimated using Aspen's property estimation tool. Sensitivity analysis and Monte Carlo simulations confirmed that these estimations did not significantly influence the overall outcomes of the TEA and LCA. Further details on the property estimation methods and thermodynamic models used are provided in the supplemental information. For TEA, we used a discounted cash flow analysis approach to estimate the MSP with a 2022 US$ cost basis. LCA was conducted with Brightway 2.9.7, using ecoinvent v3.9.1 for background data. GHG impacts were assessed using IPCC 2021,[36] while all other impact categories were evaluated using the ReCiPe midpoint hierarchical method.[37] A complete description of the process modeling, TEA, and LCA, including design assumptions, historical prices of chemicals and utilities, details regarding total capital investment (TCI) and annual operational expenditure (OPEX) calculations, and detailed flowsheets, is provided in the supplemental information.

A simplified process flow diagram for a conceptual nylon 6 recycling facility is shown in Figure 5. In the base case scenario, the plant capacity was modeled at 100 metric tons per day (MTPD), or 34,000 metric tons per year of nylon 6 waste, which represents 5% of 2022 nylon 6 consumption in North America.[38,39] For process modeling, fishing nets were selected as the feedstock. Owing to their mono-material design, nets offer high feedstock quality compared with other nylon 6 waste streams, but they also incur higher recovery costs due to low bulk density and dispersed collection points.[40] The feedstock is modeled as 96 wt % nylon 6 and 4 wt % impurities (modeled as 1.2 wt % sand and 2.4 wt % high-density polyethylene [HDPE])[41,42] sourced at \$0.80/kg, which includes transportation costs (\$0.65/kg) and collection and processing of the fishing nets (\$0.15/kg). LCA data for the collection and pretreatment of post-use nylon 6 were not available. Thus, custom inventories were created by modifying known inventories for other post-consumer recycled plastics[43] to include estimated transportation distance for post-use nylon fishing nets and removal of irrelevant entries such as sorting at a materials recovery facility (Table S17).

In the process, nylon 6 is shredded and washed to remove contaminants like sand, salt, and polyolefins.[44] Following pretreatment, nylon 6 is extruded and fed to the depolymerization reactor at 210°C and 320 psi for 2.5 h. In the presence of H_3PO_4 catalyst and *n*-PrOH solvent, nylon 6 undergoes depolymerization, yielding 77% ε-caprolactam and 23% oligomers. Following depolymerization, *n*-PrOH is recovered via distillation and recycled to the reactor, with near-complete *n*-PrOH recovery. The ε-caprolactam-rich bottom product of the *n*-PrOH recovery column is fed to a wiped-film evaporator. The resulting vapor stream is sent to the water recovery column, and the oligomer-rich liquid stream is directed to an oligomer depolymerization reactor, where 85% of the oligomers undergo depolymerization to ε-caprolactam.[45] The ε-caprolactam is removed continuously from the top of the oligomer depolymerization reactor by stripping with superheated steam (steam to ε-caprolactam feed ratio of 5:1)[46] and then recovered by distillation.

After two distillation steps, the recovered ε-caprolactam stream is 97 wt % pure and is introduced to a column reactor for polymerization. The r-nylon 6 polymer from the column reactor is extruded into pellets and contacted with hot water in a counter-current extraction column to remove remaining ε-caprolactam and oligomers. The purified r-nylon 6 product is subjected to solid-liquid separation using a disc centrifuge and fed into a rotary dryer for moisture removal, resulting in a final composition of 99 wt % r-nylon 6 and 1 wt % water.

Throughout the process, water and residual ε-caprolactam are recycled where possible. For example, unreacted ε-caprolactam from polymerization is recovered by distillation and recycled. When recovery is not possible, such as with wash water from pretreatment and unreacted oligomers from the depolymerization reactor, the waste is neutralized with calcium hydroxide and sent to wastewater treatment.

The TCI for the base case scenario was estimated at \$105.6 million (MM) with a total installed cost of \$55 MM (Figure 6A; Table S13). The caprolactam polymerization and caprolactam and water recovery sections were the most capital intensive, driven by the costs of the column reactor and distillation

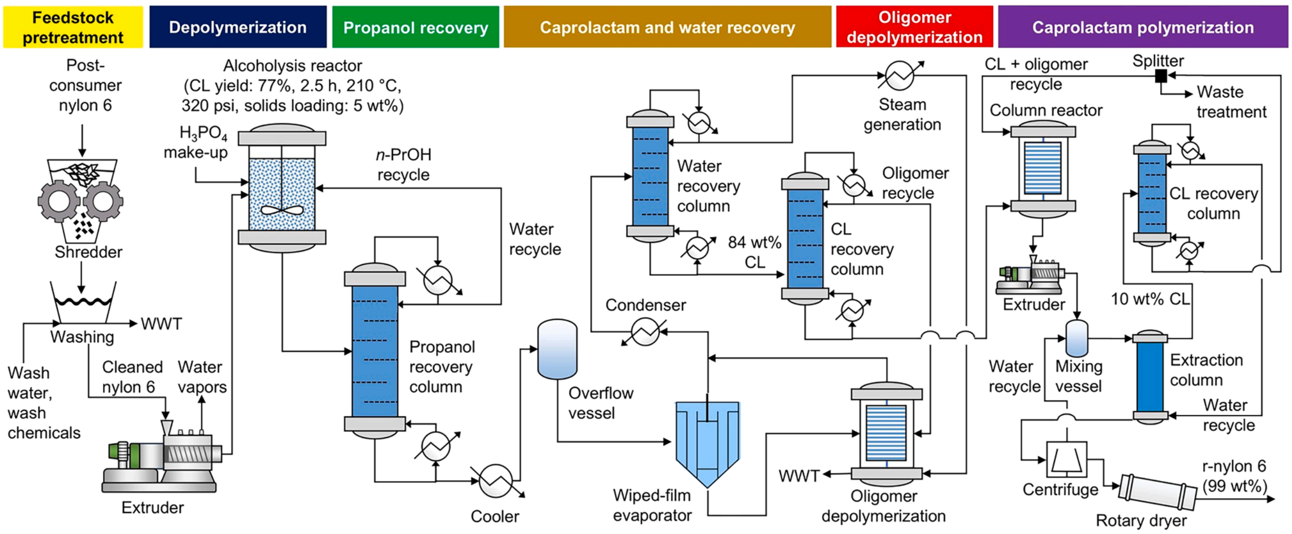

Figure 5. Simplified process flow diagram (PFD) for depolymerization of nylon 6 to ε-caprolactam via acid-catalyzed alcoholysis
Not all equipment is shown here. Detailed PFDs are shown in Figures S4–S5. ε-caprolactam is abbreviated as CL in the PFD. WWT is wastewater treatment.

columns, accounting for $14.5 MM and $12.6 MM, respectively. The total annual OPEX was estimated to be $43.1 MM (Figure 6B; Table S14), including a feedstock cost of $27.3 MM (63%, purchased at $0.80/kg). The estimated TCI and OPEX for alcoholysis align with figures reported for comparable polymer recycling processes.[47–49] Based on the TCI and OPEX estimates, a discounted cash flow rate of return analysis was performed to estimate the MSP of r-nylon 6 at $1.79/kg (Figure 6C; Table S15), with the two largest contributions coming from the feedstock (48% of overall MSP) and the capital charge (24% of overall MSP). Assuming that there is existing capacity for polymerization on-site, as would be the case if the depolymerization plant was co-located with an existing nylon 6 production facility, the MSP would drop to $1.59/kg without a newly installed caprolactam polymerization section (Table S15).

r-Nylon 6 produced via alcoholysis is predicted to exhibit lower environmental impacts than virgin nylon 6 (v-nylon 6) across 5 out of 14 categories, including 63%, 65%, and 75% reductions in GHG emissions, fossil resource use, and water use, respectively (Figure S9). These environmental benefits arise from circumventing the energy- and reagent-intensive synthesis of ε-caprolactam in the primary production process. Specifically, the alcoholysis route avoids the use of benzene feedstock; on-purpose hydrogen for benzene hydrogenation; ammonia-derived nitric oxide and hydroxylamine chemistry; concentrated sulfuric acid (oleum) used in the Beckmann rearrangement; toluene for solvent extraction and purification; and the soluble cobalt, palladium, or platinum-based catalysts that drive the oxidation and hydrogenation trains required in primary production.[5,50]

Conversely, life cycle impacts due to ecotoxicity, land use, and mineral resources, among several others, are estimated to be higher, partly due to the electricity and process heat demand (Figure S9; Table S19). Many of the environmental impacts in the nylon 6 alcoholysis process can be linked to the feedstock (9%–89% contribution to total alcoholysis impacts), mainly due to transportation, H_3PO_4 catalyst (2%–61%), process heat (6%–41%), cooling water (0.5%–25%), and electricity (1%–48%). The other process consumables, including *n*-PrOH, nitrogen, process water, and calcium hydroxide, exhibit minimal environmental impacts (Figure S9). Additionally, the calculated E-factor of 0.65 for the proposed plant, defined as the mass ratio of waste to desired product, falls within the expected range for bulk chemicals, which typically lies between <1 and 5.[51]

A second cradle-to-grave system boundary was considered to evaluate the effect of recycling nylon-6 multiple times via alcoholysis, using a system expansion approach.[52] The recycling process achieves a mass yield of 92.7%. Due to data limitations, an optimistic scenario was assumed, with a 100% recovery rate (i.e., no losses during collection or sorting) and fully recycled monomer content in the final product. Under these assumptions, the model predicts 13.7 potential product lifetimes. However, lower collection rates significantly reduce the attainable number of lifetimes. For example, a 30% recovery rate yields just 1.4 effective lifetimes (Figure S14). For more details regarding the multiple lifetimes methodology, refer to supplemental information.

Univariate sensitivity analysis was performed to investigate the impact of key process drivers on the MSP and environmental performance of alcoholysis, as shown in Figures 6D and 6E (data in Tables S16 and S20). Even under pessimistic scenarios, r-nylon 6 remains less expensive and generally more environmentally favorable than v-nylon 6. Feedstock cost had the biggest influence on the MSP, with a 20% variation changing the MSP by ±12%. However, r-nylon 6 remains competitive with historical v-nylon 6 prices ($1.63–$2.91/kg) even with feedstock prices as high as $1.50/kg (Figure S6). While increases in solids loading, catalyst loading, plant capacity, and feedstock contamination had moderate effects on MSP, environmental performance was primarily driven by solids and catalyst loading,

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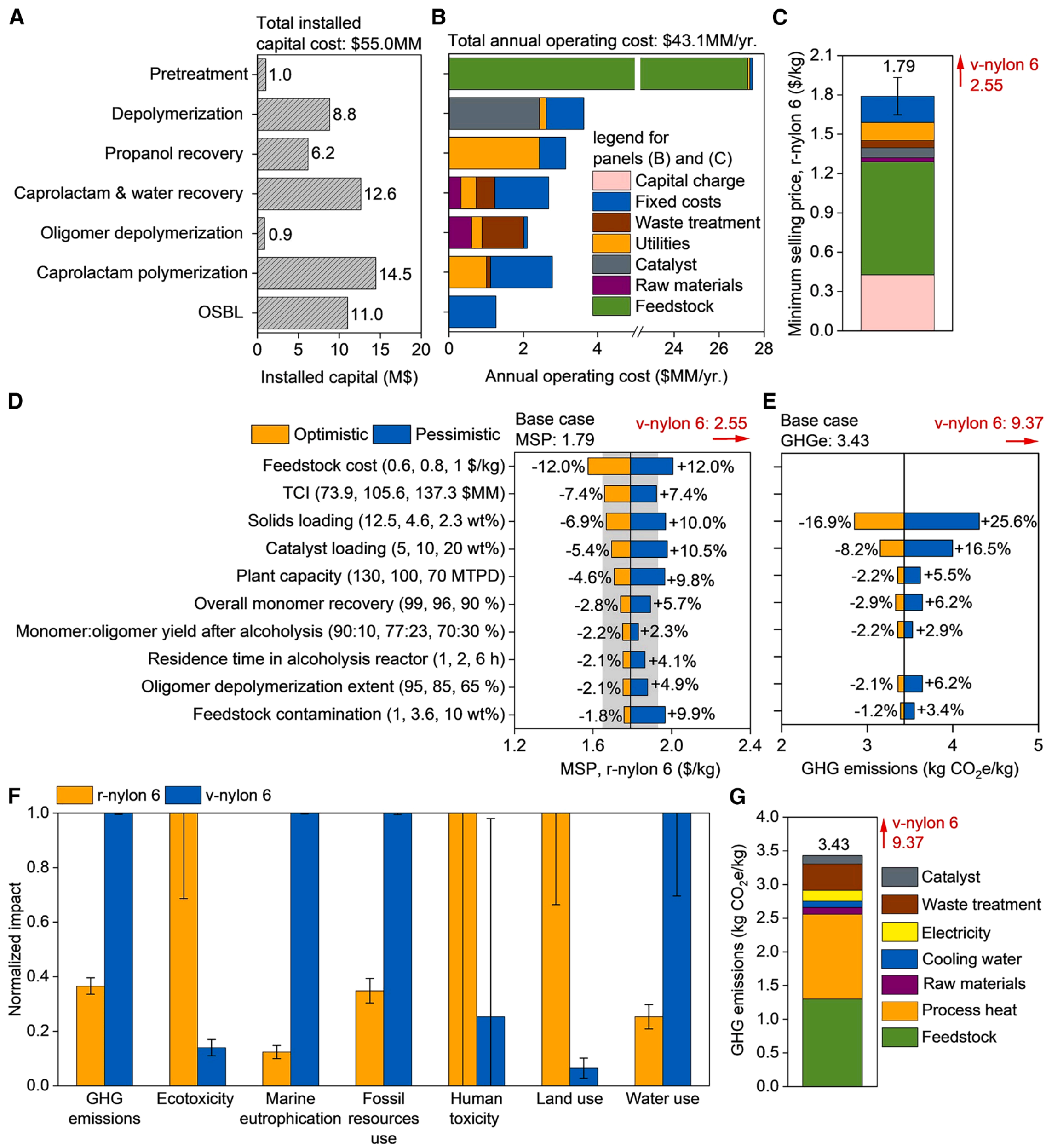

Figure 6. TEA and LCA results for the conceptual nylon-6 alcoholysis process

(A) Total installed capital cost breakdown by process area, including outside battery limit (OSBL).

(B) Annual operating cost breakdown by process area.

(C) Minimum selling price (MSP) breakdown for nylon 6 depolymerization via alcoholysis for plant capacity of 100 metric tons per day (MTPD). The error bars show the normalized standard deviation determined through Monte Carlo analysis with 1,000 runs centered on the mean. The market price of virgin nylon 6 (v-nylon 6) is listed for reference.

(D) Univariate sensitivity analysis results for the MSP of recycled nylon 6 (r-nylon 6). The shaded area in gray represents the uncertainty range in the base case MSP, estimated by Monte Carlo analysis with 1,000 runs centered on the mean. The 5-year average market price of v-nylon 6 is shown for reference.

(E) Univariate sensitivity analysis results for greenhouse gas (GHG) emissions of r-nylon 6 across various process parameters. GHG emissions for v-nylon 6 are listed for reference.

(F) Cradle-to-gate LCA results for selected impact categories for the base case and v-nylon 6 production. The error bars show the normalized standard deviation determined through Monte Carlo analysis with 1,000 runs centered on the mean. For the rest of the impact categories, refer to Table S19.

(G) The GHG impact for the proposed design broken down by component contribution. GHG emissions for v-nylon 6 are listed for reference. All data shown in Figure 6 are available in the supplemental information: Table S13 for (A), Table S14 for (B), Table S15 for (C), Table S16 for (D), Table S20 for (E), and Table S19 for (F) and (G).

with the GHG emissions increasing by nearly 26% for lower solids loading (2.3 wt %) (Figure 6E) and human toxicity increasing by nearly 38% for higher catalyst loading (20 wt %) (Figure S11). Other parameters, such as monomer recovery, alcoholysis reactor residence time, monomer-to-oligomer yield after alcoholysis, and oligomer depolymerization extent, exhibited relatively minor impacts on cost and GHG emissions (Figures 6D and 6E). Variations in reaction temperature had no impact on MSP due to energy integration across the process, and solvent (*n*-PrOH) recovery was excluded from the analysis as near-complete removal is required prior to the downstream polymerization step. Real-world fishing nets can contain insoluble pigments such as TiO_2; however, the base case in Figure 5 assumed a pigment-free feedstock to establish a clear TEA baseline. An additional scenario with fishing nets containing 2 wt % TiO_2 was modeled by incorporating a pressure leaf filtration step post alcoholysis and prior to solvent recovery. This resulted in only a minor increase in capital and operating costs, with a 1.5% increase in the MSP of r-nylon 6.

Additionally, a multivariate sensitivity analysis was carried out to understand how monomer yield after alcoholysis and solids loading affect the MSP of r-nylon 6. The results show that increasing both yield and solids loading leads to a lower MSP, with solids loading having a particularly strong impact. At low yield (50%) and low solids loading (2.5 wt %), the MSP is ~\$1.88/kg, whereas at yields above 90% and solids loadings above 15 wt %, MSP drops to ~\$1.69/kg. These insights highlight the importance of optimizing yield and solids loading to further improve process viability (Figure S8).

Based on this analysis, we propose several potential process improvements to further improve process feasibility. A key consideration is the feedstock purity. While a high-purity feedstock (high nylon 6 content and low contaminant content) can improve the alcoholysis process efficiency, the associated cost premium contradicts the low-cost feedstock requirement from the MSP sensitivity analysis. Establishing a robust supply chain is thus essential. Implementing renewable energy sources for heating and electricity across the entire plant can further decrease net GHG emissions by 39% (Figure S13). While the influence of changes in the monomer-to-oligomer yield ratio on the MSP is relatively minor, achieving monomer yields of 80% or higher is desirable to reduce the need for energy-intensive oligomer separation. By increasing solids loading to 12.5 wt % and beyond, solvent requirements can be minimized, thereby reducing costs and associated environmental burdens.

When considering the scale-up potential of new technologies, it is important to avoid burden shifting. Therefore, in addition to examining the costs and environmental impacts of nylon 6 depolymerization, the environmental risk implications of this technology were investigated using the framework developed by Uekert et al.[53] The analysis revealed that the alcoholysis process does not involve any substances currently classified as toxic by the Environmental Protection Agency's (EPA) Toxic Release Inventory (accessed July 2025).[54] However, the use of H_3PO_4 catalysts should be monitored, as it can be corrosive at high concentrations.[55] The upstream processes associated with the collection, cleaning, and sorting of plastic waste utilize toxic and corrosive chemicals like sulfuric acid and sodium hydroxide.[54] While ferric chloride and sodium hydroxide are also used in the cleaning step, their concentrations remain well below toxic levels.[56] Further, it is important to note that the manufacturing of nylon 6 textiles may have considerable supply chain concerns, given that the production of textiles has been reported to involve child labor in various countries, including Bangladesh, Cambodia, Ghana, North Korea, Pakistan, Vietnam, Ethiopia, and China.[57,58] Increasing the recycling of post-consumer nylon-6 could reduce reliance on virgin polymer production, thereby mitigating associated labor risks. Consequently, advancements in nylon recycling technologies can substantially contribute to more sustainable and socially responsible supply chains.

Conclusions

The work presented herein demonstrates a simple acid-catalyzed alcoholysis method for the chemical recycling of nylon 6, enabling selective recovery of ε-caprolactam at excellent yields from both virgin and post-consumer nylon 6 substrates. In addition, the process offers both economic benefits, including a 30% reduction in recycled nylon MSP, and environmental advantages, including a 63% decrease in resulting GHG emissions with respect to primary production of nylon 6. The economic analysis is limited by assumptions regarding input costs, scale, and process performance. These factors may change with future market developments and technological advancements, and thus, the reported economic indicators should be considered as indicative. Overall, our process addresses a variety of challenges associated with the recycling of nylon 6 and provides a viable path to a circular nylon economy.

METHODS

Detailed methods regarding the experimental and modeling procedures can be found in the supplemental information.

RESOURCE AVAILABILITY

Lead contact

Further information and requests for resources and reagents should be directed to the lead contact, Yuriy Román-Leshkov (yroman@mit.edu).

Materials availability

All the reagents required to generate the materials described in this study are all commercially available or can be prepared as indicated in the supplemental information. All materials generated in this study are available from the lead contact upon reasonable request.

Data and code availability

All data supporting the findings of this study are included within the article and its supplemental information and are also available from the authors upon request.

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AUTHOR CONTRIBUTIONS

Conceptualization, X. Wu, R.P.B.A., G.T.B., Y.R.-L., and S.S.S.; methodology, X. Wu, R.P.B.A., X. Wang, and M.S.W.; investigation, X. Wu, R.P.B.A., X. Wang, M.S.W., A.E.B., G.D., J.M., C.L., E.V.R., S.J.H., J.S.D., T.U., and Y.S.-H.; visualization, X. Wu, R.P.B.A., and X. Wang; writing – original draft, X. Wu and R.P.B.A.; writing – review and editing, X. Wu, R.P.B.A., X. Wang, A.E.B., G.D., M.S.W., G.T.B., J.S.D., T.U., and E.V.R.; funding acquisition, G.T.B. and Y.R.-L.; resources, G.T.B. and Y.R.-L.; supervision, G.T.B. and Y.R.-L.

DECLARATION OF INTERESTS

X. Wu., S.S.S., G.T.B., and Y.R.-L. are co-inventors on a patent application that covers the technology to depolymerize nylon 6 waste.

SUPPLEMENTAL INFORMATION

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