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A roll-to-roll chitosan finishing strategy for elastane recovery†

Eleanor C. Grosvenor,[‡] Malachi Cohen,[‡] Caterina Czibula,^{a,b} Emma M. Sellin,^a Kayla T. Ghezzi,^c Natalie C. Fisher,^a Jana B. Schaubeder,^b Sara E. Branovsky,^a Gabrielle N. Wood,^{a,d} Jeffrey J. Richards^c and Cécile A. C. Chazot^{*a}

Elastane fibers, renowned for their balanced strength, elasticity, and comfort, are a prevalent component in blended fabrics. However, their strong adhesion within core-spun yarns and resistance to chemical dissolution pose significant challenges for separation and recycling. The lack of a universal single-solvent strategy across blend types limits the scalability of selective dissolution recycling. Here, we propose an alternative approach using a dissolvable chitosan (CS) finishing layer applied to elastane fibers, which can be selectively removed at end-of-life to enable separation from sheath fibers. We implemented a continuous dip-coating process and demonstrated its feasibility at pilot scale using a roll-to-roll setup. By tuning solution viscosity, we achieved uniform, conformal coatings on neat elastane. A 4 wt% CS solution in 0.5 N HCl yielded a 5–10 μm-thick coating that forms strong non-covalent interactions with the elastane core without compromising the elastic modulus or energy dissipation under cyclic strain. The CS layer can be redissolved under mild acidic conditions, preserving the chemical integrity of the recovered elastane. This proof-of-concept highlights CS dip-coating as a promising finishing strategy for scalable elastane recovery from diverse fiber blends via selective dissolution.

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Introduction

Clothing is an important means of self-expression with nuanced social, cultural, and even political implications. Rapid trend cycles and population growth drive the demand for new styles, which led textile manufacturers to produce 124 million tons of garments in 2024. Future garment production is projected to reach 160 million tons by 2030.^{1,2} This growth generates substantial waste, with roughly 18 million tons of material discarded by the garment industry alone in 2020.^{3,4} Synthetic fibers comprise 57% of this waste,¹ making them a major contributor. Due to the difficulty in separating synthetic fibers in scalable recycling streams, over 70% of clothing fibers end up in landfills or incinerated, with less than 1% recycled into new garments.^{5,6}

Elastane, also known as spandex or LYCRA® fiber, is ubiquitous in the textile industry. Elastane fibers contain at least 85% polyurethane–polyurea, a linear block copolymer with alternating rigid and flexible segments.^{7,8} The flexible segments allow elastane to be reversibly stretched by up to 600%,^{9,10} and the rigid segments participate in hydrogen bonding, providing strength to the fiber.⁹ The balance between elasticity and tenacity (fiber tensile strength) of elastane yarns can be modulated simply by changing the denier (mass in grams per 9000 meters of fiber).¹¹ As a result, stretch fabrics containing even small amounts of elastane (~1–5%) are comfortable without sacrificing tear strength,^{11–13} underscoring its popularity in sportswear and casual wear. For most applications, elastane is blended with other materials through core spinning, in which roving of polyamide, polyester, or nylon is twisted around the elastane core fiber.^{14,15} This core-spun structure, along with elastane's strong adhesion to sheath fibers¹⁶ and resistance to most organic solvents, complicates the separation of these blended synthetic yarns and the recovery of individual materials at the end of their life cycle.¹⁷ Standard mechanical recycling is ineffective for core-spun elastane, as it clogs machinery and hinders the recycling of sheath fibers.^{18–20}

Neat elastane is incompatible with most other standard recycling methods.^{18,19,21} Depolymerization, which breaks covalent bonds to recover monomers, is the most promising method for

^aDepartment of Materials Science and Engineering, Northwestern University, Evanston, IL, USA. E-mail: cchazot@northwestern.edu

^bInstitute of Bioproducts and Paper Technology, Graz University of Technology, Graz, Austria

^cDepartment of Chemical Engineering, Northwestern University, Evanston, IL, USA

^dDepartment of Chemical Engineering, Howard University, Washington, DC, USA

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‡ These authors contributed equally to the work.





Fig. 1 (a) Illustration of the elastane fiber with the dip-coated chitosan (CS) layer as interface between the elastane and the sheath fibers to improve recycling via interface layer redissolution. (b) Hansen solubility plot of relevant textile polymer fibers and CS. (c) Roll-to-roll (R2R) pulley system applied in this work. The elastane fiber is guided into the CS bath, and then a fan is used to induce drying of the coated fiber. (d) Several SEM images illustrating the elastane fiber, which is a bundle of 10 single filaments (plies), successfully coated with CS.

Influence of bath concentration on CS content and interaction

With the continuous R2R process established, we investigated the impact of CS concentration and bath rheology on coating thickness (Fig. 2).^{51,52} CS-HCl solutions exhibit distinct rheological regimes depending on polymer concentration (also denoted as "bath conc."), each characterized by varying degrees of shear thinning (Fig. 2a). In the first regime (bath conc. < 0.75 wt%), the solutions are Newtonian. As concentration increases, the solution transitions from the dilute to semi-dilute regime. This transition corresponds to a logarithmic increase in zero-shear viscosity η_0 (Fig. 2b) and stronger shear-thinning. We found that only baths with CS concentrations in the semi-dilute regime (3–7 wt%, our processing window) resulted in the formation of a CS coating on elastane.

The formation of a CS coating was further confirmed by TGA (Fig. 2c and d). The presence of the CS coating is qualitatively supported by the characteristic shape of the mass loss curves (Fig. 2c). Increasing the CS bath concentration leads to a decrease in the onset decomposition temperature. Additionally, the residual mass at 645 °C provides further evidence of increased coating thickness with higher bath concentrations (Fig. 2d). Pure CS had a residual mass of around 40%, while neat elastane had a residual mass below 3% (Fig. 2c). As CS bath concentration increases from 3 to 6 wt%, the average residual mass of the coated fibers triples, denoting a corresponding increase in coating thickness (Fig. 2d). Bath concentrations below 3 wt% resulted in no significant residual mass increase compared to neat elastane, likely due to their low

capillary number and the resulting absence of CS coating.⁵³ We also observed a plateau in residual mass for CS bath concentrations above 6 wt%, attributed to the formation of a non-uniform beaded coating due to significant non-Newtonian flow effects or air entrainment.^{53,54} These residual mass results also demonstrate that CS coating can enhance flame retardancy of elastane fibers, a useful property for performance textiles.

FTIR spectroscopy was used to provide further evidence of successful CS coating and probe chemical interaction between CS and elastane (Fig. 2e, f and Fig. S4†). In the virgin elastane spectrum, there is a sharp peak at 1102 cm⁻¹ followed by a lower-intensity shoulder (between 900 and 1030 cm⁻¹) corresponding to the C–O (ether) bond.⁵⁵ In pure CS, a strong, broad absorption peak between 800 and 1220 cm⁻¹ is present and characteristic of C–O–C in the CS saccharide repeat unit.⁵⁶ We noted an increase in absorption intensity within this characteristic CS region as the bath concentration increased, particularly along the lower wavenumber edge of the investigated range (below 1000 cm⁻¹) (Fig. 2e). Integrating the relevant broad aggregate peak (between 900 cm⁻¹ and 1170 cm⁻¹) and normalizing by a vibrational signature only present in the elastane signal (from 1685 to 1760 cm⁻¹, comprising the amide I and ester vibrational modes) further confirms the increase in CS coating thickness with bath concentration in the range of 3 to 5 wt% (Fig. 2f and Fig. S4†). Fibers coated with 6 and 7 wt% CS solutions showed a lower characteristic CS signal, likely due to coating non-uniformity. Fibers with a thin CS coating (4–5 wt% bath concentration) featured an amplified intensity





Fig. 2 (a) Flow curves for 25 concentrations between 0 and 7 wt% CS in aqueous 0.5 N HCl. (b) Associated zero-shear viscosity which highlights the dilute regime, semi-dilute regime, and processing window for the bath concentrations. (c) TGA curves for CS, virgin elastane fiber, and coated elastane fibers (2–7 wt% bath concentrations). (d) Average residual mass of fibers in TGA experiment at 645 °C. (e) Overlaid FTIR spectra for 3 wt%, 4 wt%, 5 wt%, 7 wt% coated fibers, virgin elastane, and CS in two regions of interest: 1300–1135 cm^{-1} , and 1170–900 cm^{-1} . These regions encompass the CH_2 (CS) bend at 1258 cm^{-1} (i), the C–O–C (CS) stretch at 1150 cm^{-1} (ii), and the broad absorption peak for C–O–C (CS) below 1000 cm^{-1} (iii). (f) Ratio of the calculated area of the C–O–C peak in the 1170–900 cm^{-1} region to the reference peak in the 1685 to 1760 cm^{-1} region for 3 wt%, 4 wt%, 5 wt%, 6 wt%, and 7 wt% coated fibers.

for vibrational modes characteristic of the pyranose ring of CS (CH_2 bending at 1258 cm^{-1} and asymmetric stretching of the C–O–C bridge at 1150 cm^{-1}) (Fig. 2e).^{57,58} This suggests significant molecular interaction between CS and elastane, in turn affecting the vibrational degrees of freedom of the saccharide units and their amplitude.⁵⁷ Fibers coated in high-viscosity 6 and 7 wt% solutions do not show the same vibrational mode modulation due to coating heterogeneities and potential decrease of CS-elastane intermolecular interaction at large coating thicknesses.

Mechanical properties of the coated fibers

We then evaluated the effect of the dip-coating process and CS coating on elastane's copolymer structure and mechanical performance. Preservation of crystallinity after dip-coating treatment was confirmed by WAXS, comparing the spectra for virgin elastane, 4 wt%, and 7 wt% coated fibers (Fig. S5†). This suggests that exposure to HCl did not affect the elastane copolymer morphology and crystalline domains. Using a standardized cyclic tensile testing procedure, we assessed the impact of CS coating thickness on reversible stretchability under uniaxial periodic strain (Fig. 3 and Fig. S3†).

Elastane fibers show a large hysteresis under cyclic load, both in the presence and absence of CS coating.^{59,60} The CS

coating appears to significantly affect the shape of the first load cycle, with sudden stress drops at large CS bath concentrations (Fig. 3a). Evaluating the elastic modulus for the initial slope of the first tensile load resulted in average values which are between 5 and 15 MPa, with a slight decrease with the addition of a CS coating (Fig. 3b). However, these values were calculated by neglecting variations of cross-sectional area and CS coating thickness between fibers. These variations are likely the source of the slight differences observed in elastic modulus.

Hysteresis is an indication of the energy dissipated in the fiber during one loading-unloading cycle. In elastane fibers, the dissipated energy of the first cycle is about three times larger than in subsequent cycles (Fig. 3c). Comparing the energy dissipated across the CS bath concentration range, the values are very similar for the 3–4 wt% coated fibers, which exhibited slightly higher values in every cycle, while the 5–6 wt% coated fibers have lower or unchanged values compared to the uncoated fibers. Only the 7 wt% coated fibers showed stronger deviations, with higher dissipation values than the uncoated fibers in cycles 1–4. Moreover, in cycle 1, several instant drops in stress are observed at higher strain values in 7 wt% coated fibers (Fig. 3a). Macroscopic observations of individual ply failure at stress drops suggest that





Fig. 3 (a) Stress–strain diagrams from cyclic tensile testing of the CS-coated elastane fibers at different CS concentrations (0 wt%, 3 wt%, 4 wt%, 5 wt%, 6 wt%, and 7 wt%) performed for 5 hysteresis cycles. (b) Results of the elastic modulus determined from the first cycle for the different CS concentrations. (c) Dissipated energy obtained for each cycle for the different CS concentrations by calculating area under the hysteresis curves.

immersion in a high-viscosity 7 wt% bath solution results in decreased ply interaction and fiber weakening during the coating process.

Although CS and elastane have good chemical compatibility for adhesion, there seems to be a mechanical mismatch between the two materials. CS exhibits wide-ranging mechanical properties (*e.g.*, tensile strength values range from a few MPa to 100 MPa), which are tunable to a certain degree by pro-

cessing and additives.⁶¹ In direct comparison to elastane, which exhibits typical values of 1 MPa for tensile strength and up to 10 MPa for elastic modulus, CS is significantly stiffer and more brittle.⁶² In most cases, CS-based films and fibers have an elongation at break below 50%, whereas elastane reaches stretchability values of up to 600% due to its thermoplastic elastomeric nature.^{61,63} Therefore, it is surprising that coating elastane with CS does not lead to a significant increase in elastic modulus or decrease in dissipated energy. Observed coating lift-off, fracture, and ply separation under mechanical stress suggest that the strong mechanical property mismatch dominated the observed tensile properties, resulting in decreased macroscale adhesion despite strong intermolecular hydrogen bonding.

However, a quantified description of the mechanical behavior of neat and coated elastane is complicated by variations in the fibers' structural arrangement. The elastane fibers featured ten individual plies solely interacting through weak physical or electrostatic interactions. Therefore, stretching the whole elastane fiber assembly can induce reorientation and shifting of the plies into different packing geometries. We observed this rearrangement through optical microscopy of microtome-sectioned longitudinal slices taken at multiple positions along several centimeters of a continuous elastane fiber (Fig. S6†). This geometric reorganization of the plies likely occurs during the R2R dip-coating process of fibers under tension, particularly in high viscosity solutions (*e.g.*, 6–7 wt% bath conc.). Overall, mechanical property characterization indicates that the dip-coating process has minimal effect on elastane's stretchability. However, high bath viscosity can significantly influence the stretched state and cross-sectional area of coated elastane fibers, affecting failure modes. The impact of dip-coating on ply configuration was further evaluated through direct morphological analysis of CS-coated fibers (Fig. 4). Low magnification POM observation of the CS-coated fibers revealed that the 4 wt% coated fibers have an intact thin CS coating, while the 5–7 wt% coated fibers showed varied coating thickness, along with CS fracture and delamination along the fiber length (Fig. 4a). For a bath concentration of 7 wt%, regions where the coating is still intact showed significant compression of the elastane core in comparison to uncoated sections. Heterogeneities in the CS coating along the fiber direction are also accompanied by changes in coating surface morphology, as measured by AFM (Fig. 4b). Whereas the uncoated fiber exhibited the anisotropic linear pattern typical of the surface of spun synthetic fibers, the surface morphology evolves with increasing coating thickness. At 3 wt% and 5 wt%, the surface appeared homogeneously and conformally coated, as evidenced by the presence of the linear pattern from the underlying elastane fiber. However, fibers finished in 7 wt% solutions exhibit lateral cracks of several μm in length within the CS coating. In summary, elastane fibers finished using bath concentrations of 4–5 wt% featured the most uniform CS coating with no significant impact on the stretchability, failure mechanism, and ply morphology compared to neat elastane.





Fig. 4 (a) POM images of 4 wt%, 6 wt%, and 7 wt% bath concentrations demonstrating adherence of the CS coating to the fiber. (b) Representative $10 \times 10 \mu\text{m}^2$ AFM topography images (z-scale: 400 nm) for 0 wt%, 3 wt%, 5 wt%, and 7 wt% bath concentrations.

Selective dissolution of CS coating

We tested the chemical resistance of the 4 and 5 wt% coated fibers by selective dissolution of CS in aqueous 0.5 N HCl (Fig. 5). Elastane fibers were successfully recovered from the HCl bath without visible damage or ply separation. FTIR analysis confirmed that the coating dissolution did not induce significant changes in the chemical structure of the elastane (Fig. 5a). Additionally, partial dissolution of the coating was evident from a reduction in the C–O–C stretching signal,

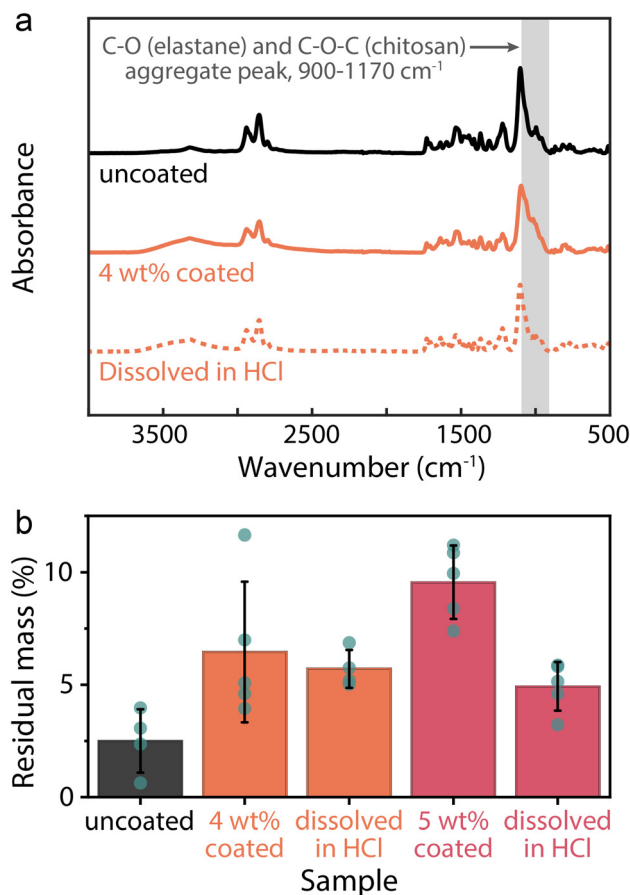


Fig. 5 (a) Representative FTIR spectra of virgin elastane, 4 wt% coated fiber, and 4 wt% fiber immersed in aqueous HCl. The region of interest is highlighted. (b) Residual mass from TGA curves, calculated for virgin elastane and 4–5 wt% coated fibers before and after immersion in HCl. Small circular data points represent individual trials.

characteristic of the CS layer, following immersion. These FTIR spectra exhibited no notable differences in the region of interest between the 4 wt% and 5 wt% CS coatings (Fig. S7†). Interestingly, TGA conducted on fibers treated with HCl for CS removal only showed a decrease in residual mass after coating dissolution for the 5 wt% coated fibers, while the 4 wt% coated fibers showed no change (Fig. 5b). Combined with the FTIR analysis in Fig. 2e and f, we expect that the chemical interaction between the CS coating and elastane fiber is stronger for thinner coatings (*i.e.*, bath solutions of lower concentration) due to enhanced adhesion *via* hydrogen bonding interactions. After immersion in HCl, most of the 4 wt% coating stays intact, while the outer layer of the 5 wt% coating is expected to be redissolved. Furthermore, both the 4 wt% and 5 wt% coated fibers exhibited higher residual mass than the neat fibers, likely due to CS residue firmly adhering to the elastane surface. Thus, once a certain coating thickness is reached with the 4 wt% bath, additional CS appears to adhere less effectively. Overall, we showed that selective coating dissolution can enable the recovery of elastane fibers with minimal residual CS adhered to the surface. The balance



between coating thickness and adhesion can be tailored to enhance recyclability *via* selective dissolution or to improve chemical resistance for further textile processing, such as dyeing.

Conclusions

We have designed and demonstrated the conformal coating of elastane fibers with CS, an abundant natural polymer that can be selectively dissolved to facilitate neat elastane recovery. We implemented the method in a pilot-scale roll-to-roll dip-coating apparatus and studied the impact of bath concentration and viscosity on coating morphology. CS bath concentrations of 4–5 wt% resulted in a uniform coating with the best adhesion to the elastane core. We demonstrated that the CS coating did not impair elastane's stretchability and that it enhanced the thermal stability of the resulting fibers. Lastly, we demonstrated how adjusting the thickness and spatially resolved intermolecular interactions can tailor the chemical resistance of the CS finishing layer. This enables selective coating dissolution under acidic conditions, allowing elastane fibers to be recovered without detectable mechanical or chemical degradation.

Further improvement in adhesion could be achieved by decreasing the mechanical mismatch between CS and elastane or enhancing hydrophobicity. Indeed, preliminary testing of the water resistance of the 4–5 wt% coatings and subsequent analysis by FTIR (Fig. S7†) suggests that immersion in de-ionized water for as little as 30 seconds may result in some coating loss (Fig. S8†). This redissolution behavior at neutral pH could pose challenges for textile durability during regular washing. This issue could be addressed by incorporating more hydrophobic biopolymers into the coating or by refining processing conditions during the finishing steps. For example, the current coating strategy does not include an acid neutralization step, which may improve the water resistance of the CS layer. Surface pretreatments commonly applied to cotton and polyesters (*e.g.*, silanization and UV/ozone plasma) could also be adapted to elastane.^{64,65} Additionally, tuning fiber velocity to increase the capillary number during coating may enhance adhesion.⁵³

Beyond addressing these practical considerations, this work serves as a proof-of-concept for scalable elastane finishing with improved recovery upon selective dissolution. It also offers critical insights into the balance of CS-elastane molecular interactions and mechanical property mismatches leading to uniform coating formation and property enhancement. The design principles and fundamental knowledge generated here can be leveraged to explore other relevant biopolymers, such as cellulose ethers and acetates, as dissolvable interface layers for textile recycling. This advancement broadens the applicability of the demonstrated fiber finishing and selective coating dissolution methods, paving the way for innovative recycling solutions for a wide range of blended fibers and textiles.

Author contributions

C. A. C. C., E. C. G. and M. C. conceptualized the research. C. A. C. C. supervised the research. G. W. conducted preliminary work leading to the results presented here. M. C. built the R2R apparatus. M. C., E. C. G. and N. F. conducted fiber coating/redissolution experiments. E. S. and M. C. conducted SEM imaging. M. C. and E. C. G. conducted FTIR measurements. M. C., C. C., J. S. and E. C. G. conducted mechanical testing and analysis. S. B. and C. A. C. C. conducted TGA. K. T. G. conducted rheometry under the supervision of J. J. R., E. C. G., C. C. and N. F. conducted POM imaging. E. C. G. and M. C. wrote the first draft of the manuscript. C. A. C. C. edited the manuscript and oversaw its writing. All authors commented on the manuscript.

Conflicts of interest

There are no conflicts to declare.

Data availability

The FTIR, TGA, and stress/strain data supporting this study are available at: E. C. Grosvenor, M. Cohen, & C. A. C. Chazot (2025). A roll-to-roll chitosan finishing strategy for elastane recovery [Data set]. Northwestern University. <https://doi.org/10.5281/zenodo.15863582>.

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