

# A Combined Experimental and Theoretical Study on Textile Recycling: the Dissolution of Elastane

Kim Phan<sup>a</sup>, Sibel Ügdüler<sup>a</sup>, Lies Harinck<sup>a</sup>, Veronique Van Speybroeck<sup>b</sup>, Karen De Clerck<sup>c</sup> and Steven De Meester<sup>a</sup>

<sup>a</sup> Laboratory for Circular Process Engineering (LCPE), Department of Green Chemistry and Technology, Ghent University Campus Kortrijk, Sint-Martens-Latemlaan 2B, B-8500, Kortrijk, Belgium

<sup>b</sup> Center for Molecular Modeling, Ghent University, Technologiepark 46, B-9052, Zwijnaarde, Belgium

<sup>c</sup> Department of Materials, Textiles and Chemical Engineering (MaTCh), Ghent University, Technologiepark 70A, B-9052, Zwijnaarde, Belgium

Keywords: textile, recycling, polymer, valorization, molecular modeling

Presenting author email: [Kim.Phan@Ugent.be](mailto:Kim.Phan@Ugent.be)

## 1. Introduction

The current textile industry operates in a linear way. Clothes are made from large amounts of resources (98 Mt per year) such as oil for the production of synthetic fibers, fertilizers to grow cotton and chemicals to produce dyes and textile auxiliary products. Clothing is also massively underutilised with garments discarded after just seven to ten wears, or even shorter with fast-fashion trends, after which the materials are mostly sent to landfill or to incineration. Despite the relatively high collection rates in some countries, only 13% of the material used for clothing ends up being recycled in lower-value applications (open-loop recycling), while even less than 1% results in a similar quality application (closed-loop recycling). These recycling techniques are either based on mechanical processes or else fibers are chemically broken down. In many of these processes the fiber-recycling processes are scarcely able to separate blends (e.g., cotton/polyester with elastane for wearing comfort). Next to that, the textiles contain dyes and contaminants, leaving a poor color quality of shoddy (Morlet et al., 2017, Payne, 2015). Especially elastane is problematic because this jeopardizes the properties of the main component of the blend upon recycling (e.g., melt processing), leading to an inferior quality of the recycled product. One option is to remove elastane by selective dissolution. The removal of elastane is commonly performed with classical solvents that are often toxic or corrosive (Ramphal et al., 2019). Consequently, a selective solubilization procedure based on greener solvents is required to remove the elastane while leaving the main component of the textile blend intact.

Therefore, new greener solvents should be found for this purpose. Theoretical tools, are frequently applied for predicting the solubility or binding affinity of drugs and additives. Common tools are phase-solubility studies, quantitative-structure property relationships (Das et al., 2020), Hansen solubility parameter calculations via group contribution method (Van Krevelen and Te Nijenhuis, 2009, Hansen, 2007), Flory-Huggins interaction parameter ( $\chi_{12}$ ) (Hansen, 2007), conductor-like screening model for real solvents (COSMO-RS) (Klamt, 1995), COSMO segment activity coefficient model (COSMO-SAC) (Bell et al., 2020), etc. In molecular modeling, solvation energies could be obtained via static calculations using an implicit solvation model or a more computationally expensive explicit solvation model by means of molecular dynamics in combination with the Free Energy Perturbation (FEP) method. These models can provide insights into the solubilization process of textile polymers and predict which (bio)based solvents are most effective. Furthermore, this screening allows a more defined approach for experimental work.

In this work, the solubility of elastane was screened toward their solubility with a large database of solvents by means of Hansen Solubility Parameters and COSMO-RS. The complementary results from these theoretical tools were validated with experimental work. In order to quantify the amount of dissolved textile polymer, a novel thermogravimetric analysis method (TGA-FTIR) was developed to map the solvation ability of various solvents toward elastane.

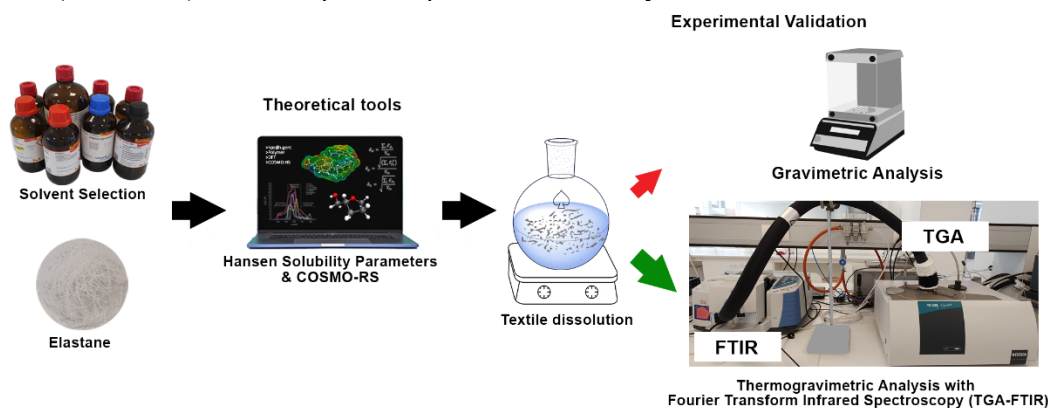


Figure 1. Schematic representation of the selective dissolution procedure.

## 2. Material and Methods

### 2.1. Theoretical Tools

One of the most widely used approach to calculate the solubility of drugs, solvents, polymers, oils and surfactants is the Hansen solubility parameters (HSP) (Hansen, 2004). Briefly, the properties of a compound is described by three types of interactions and thus, three partial solubility parameters: dispersion ( $\delta_d$ ), polar ( $\delta_p$ ) and hydrogen bonding ( $\delta_h$ ). The solubility parameter distance  $R_a$  in the Hansen space gives a qualitative view for the miscibility between solute and solvent. The smaller  $R_a$ , the higher the affinity or miscibility between the polymer and solvent (Hansen, 2007).

$$R_a = \sqrt{4(\delta_{d,A} - \delta_{d,S})^2 + (\delta_{p,A} - \delta_{p,S})^2 + (\delta_{h,A} - \delta_{h,S})^2} \quad (4)$$

where  $\delta_{i,A}$  and  $\delta_{i,S}$  ( $\text{MPa}^{1/2}$ ) are the  $i^{\text{th}}$  component of the polymer and solvent, respectively.

The second screening method is called Conductor-like Screening Model for Realistic Solvent (COSMO-RS). This hybrid solvation model, introduced by Klamt (1995), combines Density Functional Theory (DFT) with Conductor-like Screening Model (COSMO) calculations and is mostly applied for solubility prediction of drugs and polymers (Benazzouz et al., 2014, Walker et al., 2020). The molecular interactions between polymer and solvent are obtained by the statistical averaging of local pair wise interactions of surface segments (i.e., charge densities).

## 2.2. Experimental Section

Elastane filaments were supplied by Utebel and panties with a polyamide/elastane composition of 88/12 were used as a case study. Dimethylacetamide (DMAC), dimethylformamide (DMF), n-methylpyrrolidone (NMP), cyrene (CYR) and tetrahydrofurfuryl alcohol (THFA) (Merck) were the main solvents of interest. A material to liquid ratio of 1:100 was used for the dissolution process and was performed at 100 °C for four hours. Centrifugation was carried out to separate the undissolved elastane from the solution (4000 rpm and 2 hours).

In order to determine the maximal solubilities, a NETZSCH TG 209 F3 Tarsus thermogravimetric analyzer coupled to Nicolet™ iS20 FTIR Spectrometer was applied to perform the analyses. The FTIR allows the detection of the evolved gases derived from the sample decomposed within the TGA. A temperature profile from 35 °C till 750 °C in a  $\text{N}_2$  atmosphere was applied. Calibration curves of elastane (0.1-10 mg), based on the degradation products, and solvents were created for quantification purposes.

## 3. Results and Discussion: Elastane Dissolution

The complementary screening results from HSP and COSMO-RS indicated that aprotic polar solvents are most suitable for solubilizing elastane. The gravimetric approach is a frequently applied method for determining solubilities in various research areas. However, this method comes with large error margins as solvent absorption occurs within the undissolved part of the polymer. Furthermore, additives can also be leached out from the polymer during the dissolution process. As such, one is never certain about the actual amount of polymer dissolved or the amount of solvent entrapped within the fiber. Therefore, the TGA-FTIR technique allows the quantification of the dissolved polymer content by a single measurement. It was observed that the green solvent THFA performed better than CYR and was able to show a competitive elastane solubility to NMP. This dissolution method has been applied to panties as a case study. Figures 1A and 1B illustrate the panty samples before and after treatment with THFA, respectively. Figures 1C-D are SEM images of panties which highlight the elastane removal after the dissolution process.

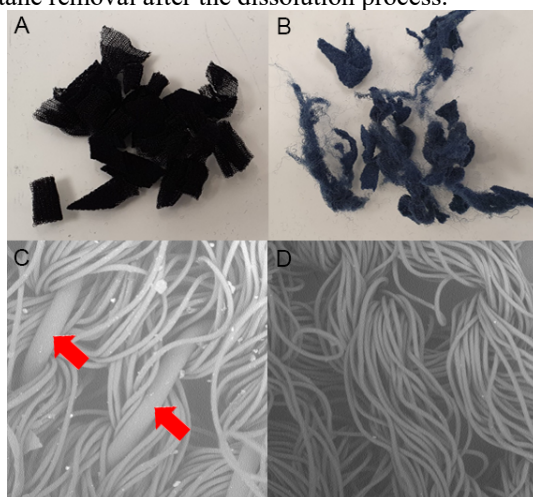


Figure 1. Top: Panty samples (polyamide/elastane: 88/12) before (A) and after (B) treatment with tetrahydrofurfuryl alcohol. Below: SEM images of panties before (C) and after (D) elastane dissolution. The elastane fibers are indicated by arrows.

## 4. Conclusions

In this work, a combined theoretical and experimental study has been performed on the dissolution of elastane. The screening with theoretical tools allows a more defined approach on the type of solvents allow the dissolution of elastane. The results from HSP and COSMO-RS indicate that aprotic polar solvents are most preferred to solubilize elastane. Therefore, cyrene and tetrahydrofurfuryl alcohol were selected as biobased solvents to investigate their performance against classical solvents. Since the gravimetric method does not allow an accurate measure of solubility due to solvent

absorption by the polymer or additive leaching, a TGA-FTIR method has been developed to determine the maximal solubilities of elastane in the selected solvents. This method has been successfully applied on panties as a case study. To the best of the author's knowledge, no work to date has reported TGA-FTIR as quantification method of textile polymers. Further investigations on various textile polymer-solvent combinations are required to ensure the selectivity of this method.

## 5. Acknowledgments

The authors would like to thank Collective Research NETwork (CORNET) and Flanders Innovation & Entrepreneurship (VLAIO) for their financial support. The computational resources (Stevin Supercomputer Infrastructure) and services used in this work were provided by the VSC (Flemish Supercomputer Center), funded by Ghent University, FWO and the Flemish Government – department EWI.

## 6. References

- BELL, I. H., MICKOLEIT, E., HSIEH, C.-M., LIN, S.-T., VRABEC, J., BREITKOPF, C. & JÄGER, A. 2020. A Benchmark Open-Source Implementation of COSMO-SAC. *Journal of Chemical Theory and Computation*, 16, 2635-2646.
- BENAZZOZ, A., MOITY, L., PIERLOT, C., MOLINIER, V. & AUBRY, J.-M. 2014. Hansen approach versus COSMO-RS for predicting the solubility of an organic UV filter in cosmetic solvents. *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 458, 101-109.
- DAS, T., MEHTA, C. H. & NAYAK, U. Y. 2020. Multiple approaches for achieving drug solubility: an in silico perspective. *Drug Discovery Today*, 25, 1206-1212.
- HANSEN, C. M. 2004. 50 Years with solubility parameters—past and future. *Progress in Organic Coatings*, 51, 77-84.
- HANSEN, C. M. 2007. *Hansen solubility parameters: a user's handbook*, CRC press.
- KLAMT, A. 1995. Conductor-like Screening Model for Real Solvents: A New Approach to the Quantitative Calculation of Solvation Phenomena. *The Journal of Physical Chemistry*, 99, 2224-2235.
- MORLET, A., OPSOMER, R., HERRMANN, S., BALMOND, L., GILLET, C. & FUCHS, L. 2017. A New Textiles Economy: Redesigning fashion's future. Ellen MacArthur Foundation.
- PAYNE, A. 2015. 6 - Open- and closed-loop recycling of textile and apparel products. In: MUTHU, S. S. (ed.) *Handbook of Life Cycle Assessment (LCA) of Textiles and Clothing*. Woodhead Publishing.
- RAMPHAL, I., LABINE, A., HAZARD, K., NG, S. & ZHANG, S. 2019. Greener Alternatives to Dimethylformamide Use in Polyurethane Synthetic Leather. 45.
- VAN KREVELEN, D. W. & TE NIJENHUIS, K. 2009. *Properties of polymers: their correlation with chemical structure; their numerical estimation and prediction from additive group contributions*, Elsevier.
- WALKER, T. W., FRELKA, N., SHEN, Z., CHEW, A. K., BANICK, J., GREY, S., KIM, M. S., DUMESIC, J. A., VAN LEHN, R. C. & HUBER, G. W. 2020. Recycling of multilayer plastic packaging materials by solvent-targeted recovery and precipitation. *Science advances*, 6, eaba7599.