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Complete List of Authors:	Munguía-López, Aurora del Carmen; University of Wisconsin-Madison, Department of Chemical and Biological Engineering Göreke, Dilara; University of Wisconsin-Madison, Department of Chemical and Biological Engineering Sánchez-Rivera, Kevin; University of Wisconsin-Madison, Department of Chemical and Biological Engineering Aguirre-Villegas, Horacio; University of Wisconsin System, Department of Biological Systems Engineering Avraamidou, Styliana; University of Wisconsin-Madison, Department of Chemical and Biological Engineering Huber, George; University of Wisconsin-Madison, Department of Chemical and Biological Engineering Zavala, Victor M.; University of Wisconsin-Madison, Department of Chemical and Biological Engineering

Quantifying the Environmental Benefits of a Solvent-Based Separation Process for Multilayer Plastic Films

Aurora del C. Munguia-Lopez^{¶*}, Dilara Göreke^{¶*}, Kevin L. Sánchez-Rivera[¶], Horacio A. Aguirre-Villegas[‡], Styliani Avraamidou[¶], George W. Huber[¶], and Victor M. Zavala^{¶†}

[¶]Department of Chemical and Biological Engineering, University of Wisconsin-Madison

[‡]Department of Biological Systems Engineering, University of Wisconsin-Madison

Abstract

Food packaging often appears in the form of multilayer (ML) plastic films, which leverage the functional properties of different polymers to achieve specific food protection goals (e.g., oxygen, water, and temperature barriers). These properties are essential to enable long shelf lives, reduce refrigeration usage, mitigate food waste, and increase food accessibility. However, ML film production processes generate large amounts of plastic waste that cannot be mechanically recycled. Recently, we have proposed a process, called solvent-targeted recovery and precipitation (STRAP), that enables the separation and recycling of the constituent polymers of ML films. This technology uses a series of solvent washes that selectively dissolve and precipitate target polymers. Quantifying the environmental benefits of STRAP over virgin resin production is essential for the commercial deployment of this technology. This work uses life cycle assessment (LCA) methods to evaluate these impacts in terms of carbon footprint, energy use, water use, and toxicity. We analyze three STRAP process variants that use anti-solvent and temperature-driven precipitation to treat different ML films. Our analysis reveals that the STRAP-A and STRAP-B process variants can provide environmental benefits over virgin film production. Furthermore, it gives valuable insight into the critical components of ML films (specific polymers) and of the STRAP processes (equipment) that are responsible for the highest impacts. Ultimately, we believe that the proposed analysis framework can lead to the design of more environmentally-friendly ML films and recycling processes.

*Denotes equal contribution.

†Corresponding Author: victor.zavala@wisc.edu

1 Introduction

Advances in the food packaging industry have brought many environmental and societal benefits; for instance, packaging is essential for enabling long shelf-lives, which helps to mitigate food waste and fosters food accessibility (facilitates transport over long distances).¹ ML plastic films are an essential component of packaging applications. These films represent approximately 20% of food packaging in Europe, with projections showing an increase over the following years based on the current demand and trends (e.g., convenience products, single-serve meals, and small packaging).² ML films are built using several layers of different polymers³ that are combined to achieve unique properties (e.g., oxygen, water, and temperature barriers) that help preserve food products and reduce energy use.⁴ Moreover, ML films help reduce packaging costs, as they reduce the amount of plastic material needed to attain these properties (compared to single layer packaging materials).⁵ Films with up to 17 layers of different polymers can be found in applications^{6,7} and more specialized applications might require hundreds of layers.⁸

The production of ML films generates large amounts of waste; this waste is known as post-industrial waste (PIW) and is the result of manufacturing inefficiencies that are difficult to avoid. It is estimated that up to 20% of all ML films produced are discarded as PIW.⁹ Compared to post-consumer waste (PCW), PIW has the advantage that is relatively clean, because it is not contaminated with food or other impurities.¹⁰ Unfortunately, PIW of ML films is not currently recycled¹¹ due to a lack of commercially available technologies. Conventional technologies, such as mechanical recycling, cannot be used to recycle ML films;¹²⁻¹⁴ therefore, ML films end up in landfills or combusted.¹⁵⁻¹⁷ The improper management of ML films can also lead to plastic leakage into the natural environment, with plastic entering the waterways and consequently, the food chain.^{18,19}

Solvent-based separation has been recently proposed as a paradigm that enables the recovery of polymers from ML films. A specific process, known as solvent-targeted recovery and precipitation (STRAP),⁹ uses sequential solvent washes to separate the constituent polymers. The design of these processes has been facilitated via the use of molecular simulations, which enable the identification of selective solvents for targeted polymer layers. Previous work has reported different configurations for the process, called STRAP-A, STRAP-B, and STRAP-C.²⁰ STRAP-A precipitates dissolved polymers via the addition of antisolvents and requires subsequent separation of solvent and antisolvents via distillation (which can be energy intensive). STRAP-B aims to reduce or eliminate the use of antisolvents via using temperature-driven polymer precipitation. Both approaches have been shown to deconstruct an industrial ML film composed of PE, EVOH, and PET (film A1). STRAP-C is a process that combines the two precipitation techniques (antisolvent addition and solvent temperature reduction) and has been designed to separate a more complex ML film (film A2), composed of PE, EVOH, PET, and PEG.

The economic viability of the STRAP processes has been previously studied using detailed techno-economic analysis (TEA).²⁰ This work has found that the minimum selling price of the resins recovered via STRAP is comparable to the average market values of virgin resins. The analysis has also found that the economic benefits increase when precipitation can be achieved via temperature-driven approaches (e.g., STRAP-B and STRAP-C); this is due to the avoidance of distillation, which is expen-

sive from a capital and operation standpoint. Other applications of solvent-based polymer recycling processes, including solid mixtures and electronic waste, have shown economic and environmental benefits when compared to virgin plastics.²¹ However, the environmental impact of the STRAP process has not been reported in the literature; this is essential to foster the adoption of this technology and to identify more environmentally-friendly designs of processes (e.g., solvent selection) and of ML films (e.g., polymer layer selection), as well as to understand the interplay between ML film design and recycling.

Life cycle assessment (LCA) is an established methodology used for quantifying the environmental impacts of consumer products and associated production processes.²² One of the main objectives of LCA is to provide information to industries, policy-makers, and society regarding the environmental impacts and feasibility of emerging processes and technologies. Regarding plastic waste, multiple LCA studies have compared different recycling alternatives such as pyrolysis, mechanical recycling, and energy recovery.^{23,24} Jeswani *et al.*²³ found that pyrolysis and mechanical recycling have a lower contribution to climate change than producing energy products from waste plastics. However, the energy recovery alternative performs better for other impact categories (e.g., acidification, eutrophication, photochemical ozone formation, and human toxicity). An innovative scheme to treat plastics from the waste of electrical and electronic equipment has been studied from an LCA perspective. The environmental benefits of this process were reported under "ideal" and "real" scenarios.²⁵ These studies leverage the LCA methodology to analyze the environmental feasibility of new waste management and production processes. The LCA methodology has also been used as a metric to evaluate the environmental sustainability of some circular economy strategies, including narrowing, slowing, and closing resource flows.²⁶ The goals of a circular economy involve the reduction of waste, use of natural resources, and emission levels. Other key characteristics of circularity are increasing the share of renewable resources by limiting the use of virgin materials and increasing the durability of products through high-quality recycling.²⁷

In this work, we conduct a detailed LCA study of STRAP processes; our goal is to identify if recycling ML films via STRAP can bring benefits over virgin resin products (thus enabling more circular economies). To do so, we evaluate diverse environmental impacts (carbon footprint, energy use, water use, and toxicity) of STRAP and compared them to those of virgin resin production. We analyze STRAP-A, -B, and -C process variants in order to identify the specific components of the STRAP process and of the ML films that are responsible for the highest impacts.

The paper is structured as follows. Section 2 presents the methodology, which includes key assumptions of the study, the description of the STRAP process, as well as details about the LCA approach. Section 3 presents the findings for the different STRAP processes and impact categories. The final section presents conclusions and final remarks of the study.

2 Methodology

The principles of the LCA methodology used in this study are in compliance with the ISO-14040 and 14044-LCA series of standards.^{28,29}

2.1 Scope of the Study

This LCA study aims to quantify the environmental benefits (climate change, energy use, water use, and process toxicity) of the STRAP technology and of the production of ML plastic film from virgin resins. This comparison is essential for assessing the environmental feasibility of the STRAP process and identifying key bottlenecks. The study evaluates the impacts of a potential commercial plant of 3,000 tonnes per year, which has been found to be economically viable (the minimum selling price of the recovered resins is comparable to their market values). We consider the inputs and outputs for three different STRAP processes (STRAP-A, STRAP-B, and STRAP-C) presented in Sánchez-Rivera *et al.*;²⁰ we refer the reader to this study for a detailed description of the process.

The LCA study is performed by taking a product perspective;²³ from this perspective, the STRAP process is seen as an alternative process that produces virgin-grade polymers (raw materials of ML films). Therefore, we compare the environmental impacts of producing ML films from virgin resins (from fossil sources) against producing them using STRAP (which avoids going back to fossil sources). We disaggregate the total impacts into contributions of each polymer layer (e.g., PET, PE, EVOH, PEG), which provides important insight that can enable more sustainable packaging design. Furthermore, we disaggregate the impacts into different components of the STRAP processes, which provides insight that can enable more sustainable process designs.

2.2 System Boundaries

The system boundaries for the material production from virgin resins (from fossil sources) and of STRAP (from ML film PIW) are presented in Figure 1. Following a product perspective, we treat STRAP as another method to produce ML films of virgin-resin quality.^{20,23} In the virgin resins alternative, the system boundaries include the extraction and transportation of raw materials (fossil sources), material processing (production of monomers, polymerization, and ML film extrusion), and consumption of process utilities (energy and water inputs). Any additives such as fillers (fibers), reinforcements, colorants (pigments), or functional additives (stabilizers, flame retardants, lubricants, foaming agents, flame retardants)³⁰ are not considered within the scope of the system boundaries. In the STRAP alternative, the ML film PIW is treated through the recycling technology to isolate the constituent polymer layers. The resins are then extruded to produce an equivalent amount of ML film. Thus, the STRAP system boundaries include the collection and transportation of ML film waste (considering a distance of 1000 km), the material inputs (solvents and utilities), the transportation of the process solvents, and the ML film extrusion to produce the new film from recovered polymers. The film treated in the STRAP process is assumed to be clean PIW; therefore, product use, and treatment (such as screening, sorting, grinding, and cleaning) are not within the scope of the system boundaries. The ability to use PIW is a key benefit compared to PCW.

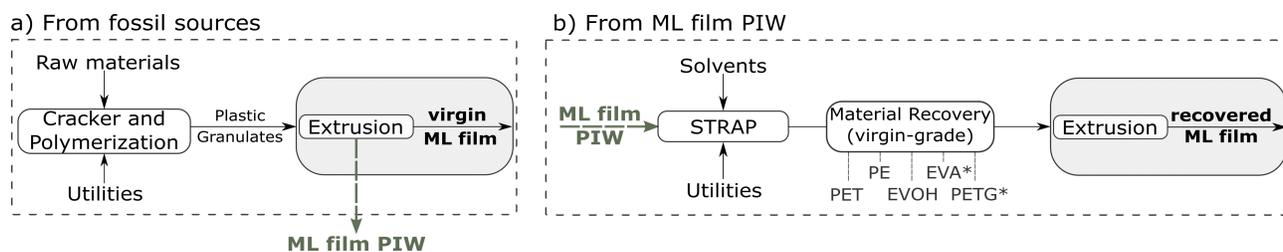


Figure 1: System boundaries for the LCA study. a) Multilayer film production from virgin resins (from fossil sources). b) Multilayer film production via STRAP technology (from ML film PIW).

2.3 Functional Unit

LCA studies can be conducted for systems composed of multiple processes. Defining a functional unit is essential to provide a fair comparison basis, allowing relevant quantitative evaluation and comparison between different strategies serving the same function.³¹ This study defines the functional unit as the production of 1 kg of ML film; therefore, we compare the environmental impacts of producing 1 kg of film from virgin resins with producing 1 kg of film from the recovered resins through the STRAP processes.

2.4 STRAP process description

The STRAP technology aims to recover the polymer components of post-industrial waste of ML films.^{9,20} Particularly, it has been used for a couple of ML films manufactured by Amcor (films A1 and A2). The main building polymers of film A1 are PE, PET, EVOH, and ethylene vinyl acetate (EVA). Film A2 is a more complex film composed of PE, PET, EVOH, and PETG. Three different STRAP processes were previously designed based on the type of film and on the use of different precipitation strategies: precipitation by antisolvent addition or solvent temperature reduction (to 35 °C).

The first variation of the STRAP approach, STRAP-A, was developed for recovering PE (with minor EVA component), PET, and EVOH from ML film A1, following the solvent/antisolvent strategy. In this method, the target polymer is selectively dissolved in a solvent, while the remaining polymers are left insoluble. Then, the polymer-solvent solution is separated from the residual polymers, and an antisolvent is added to precipitate and isolate the targeted polymer.

The STRAP-B process was developed for the same ML film A1. In this process, all the constituent polymers were separated (PE, PET, and EVOH). Additionally, the EVA component from the PE resin was isolated. Regarding the precipitation strategy, instead of adding an antisolvent, the temperature of the polymer-solvent solution (95-110 °C) is lowered to the point that the target polymer becomes insoluble (35 °C). It is worth highlighting that this temperature reduction is done using cooling water. After this, the polymer is precipitated and isolated. The process flow diagrams and a detailed explanation of these processes are reported in the Supporting Information.

The process flow diagram of the STRAP-C approach is presented in Figure 2. This approach is used to recover a more complex material, ML film A2, which consists of PE, PET, EVOH, EVA, and PETG. The process combines a couple of solvent precipitation techniques (antisolvent addition and

solvent temperature reduction), as we describe next.

PETG is the first component to be separated, using a 60% DMF/40% THF (v/v) solution (4 hours, 87 °C), which only solubilizes the targeted resin while leaving the other polymers in the solid phase. These polymers are separated from the solution by mechanical filtration. Then, 1-propanol is added as an antisolvent to facilitate the precipitation of the PETG in the solution. The PETG is then filtered and dried. After this, this recovered polymer is sent to the extruder, and, finally, it is sent to the storage tank in pellet form. The DMF/THF/1-propanol mixture is recycled back into the process after a series of distillation operations. The remaining polymer constituents in the film are recovered following the same steps as STRAP-B. PE and EVA components are selectively dissolved in toluene (4 hours, 110 °C) and separated from EVOH and PET via mechanical filtration. The temperature of the toluene mixture, containing the dissolved polymers PE and EVA, is reduced from 110 °C to 35 °C to enable PE precipitation while the remaining EVA component is recovered by adding acetone which acts as an antisolvent. After filtration and drying, both polymers are sent to the extruder and then to the storage tank. The acetone/toluene mixture is separated by distillation and then recycled back into the process. For the EVOH and PET layers, EVOH is selectively dissolved in a 60% DMSO/40% water (v/v) solution (30 mins, 95 °C) and then cooled down to 35 °C to achieve precipitation. The precipitated EVOH is stored following its filtration, drying, and extrusion processes while the solvent mixture is directly recycled back into the process. After the EVOH separation, the remaining PET component in the dissolution vessel is extruded and stored after filtration and drying. Finally, after the polymers are recovered in pellet form, they are sent to the film extruder to produce virgin-grade ML film. Since the STRAP process recovers the polymers in pellet form, they can be used in several applications. However, in this study, we consider the STRAP process as an alternative method to produce ML films (product perspective); therefore, the film extrusion step needs to be included in the analysis.

The solvent recycling ratios of all solvents and antisolvents are reported in the Supporting Information. Because the recycling rate for all solvents is around 99.90%, the impacts related to the treatment of discarded solvents are assumed to be negligible and excluded from the system boundaries.

A summary of the STRAP processes, including the type of film processed, recovered polymers, solvents, solvent recovery percentage, and precipitation method, is shown in Table 1. The recovered polymers refer to the fraction of the polymers within the film, which is equal to the fraction recovered because the STRAP process has a recovery of nearly 100%.^{9,20} We can see that the same type of film is treated in STRAP-A and STRAP-B. However, the number of recovered polymers and the precipitation methods are different. Also, the solvents and antisolvents used to recover each polymer are similar for all approaches, but there are some essential differences. In STRAP-A, acetone and water are used as antisolvents to recover PE (with a minor EVA component) and EVOH, respectively. In STRAP-B, toluene and a mixture of DMSO and water are used to recover PE and EVOH (using temperature reduction), respectively. In this approach, only a small amount (see Table 2) of antisolvent (acetone) is used to recover the minor component EVA, but the other polymers are precipitated through temperature reduction. On the other hand, in STRAP-C, 1-propanol is the antisolvent that precipitates the PETG of film A2. The amount of antisolvent used is an important process variable

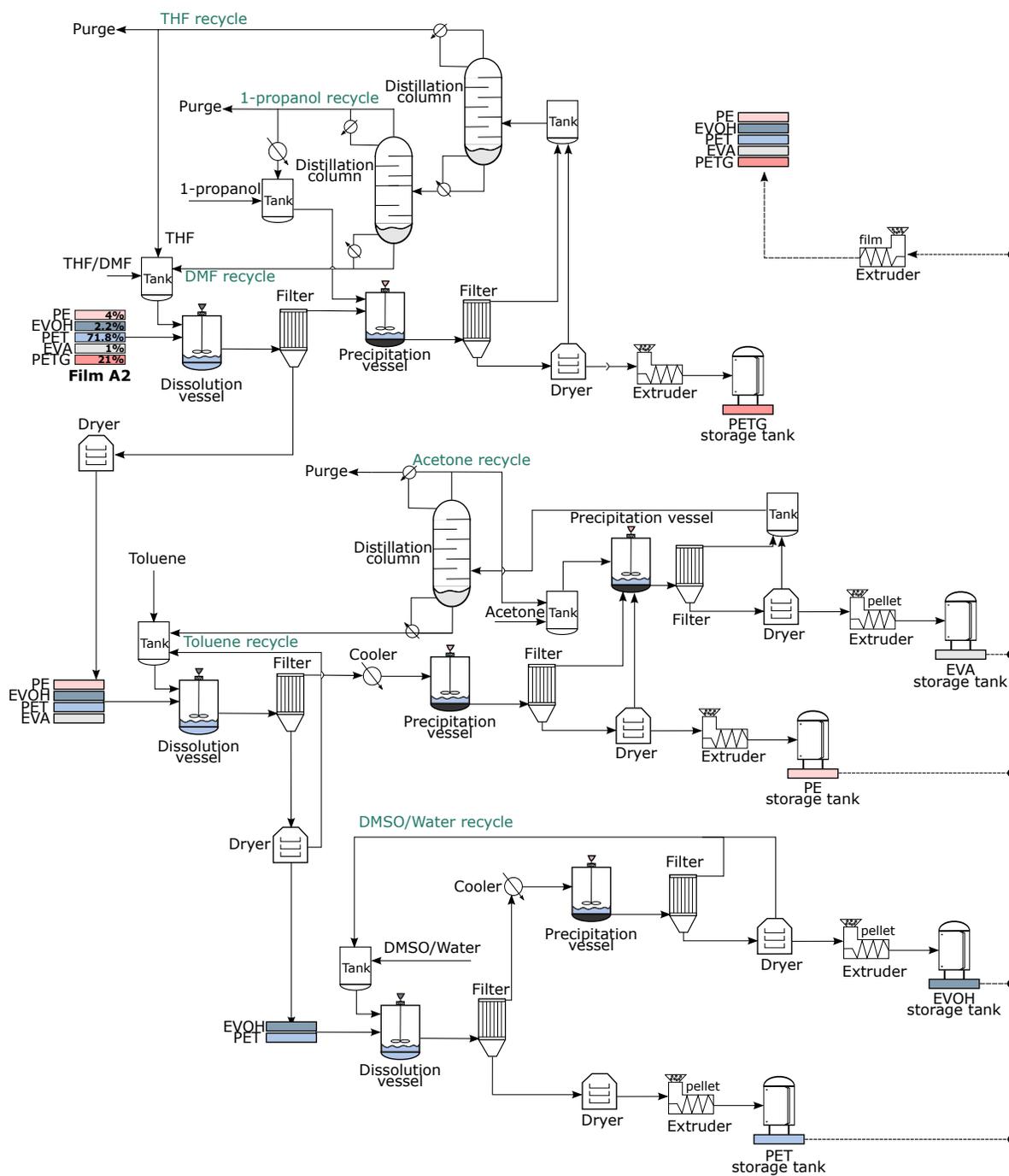


Figure 2: STRAP-C process for recycling ML film A2 composed of PE (4 wt%), EVOH (2.2 wt%), PET (71.8 wt%), EVA (1 wt%), and PETG (21 wt%).

because a distillation unit is required to separate the solvent and antisolvent for their recycling.

Table 1: Summary of STRAP process variants.

Process	ML film	Polymers recovered (wt%)	Solvents and solvent recovery (%)	Precipitation method
STRAP-A	A1	PE* (6.2%)	Toluene, Acetone (99.88%)	Antisolvent addition
		EVOH (3.6%)	DMSO, Water (99.88%)	
		PET (90.2%)	Water (99.88%)	
STRAP-B	A1	PE (5.2%)	Toluene (99.97%)	Temperature reduction
		EVA (1%)	Toluene, Acetone (99.98%)	
		EVOH (3.6%)	DMSO 60%/Water 40% (v/v) (99.99%)	
		PET (90.2%)	Water (99.99%)	
STRAP-C	A2	PETG (21.0%)	DMF 60%/THF 40% (v/v), 1-propanol (99.90%)	Combined
		PE (4%)	Toluene (99.99%)	
		EVA (1%)	Toluene, Acetone (99.99%)	
		EVOH (2.2%)	DMSO 60%/Water 40% (v/v) (99.99%)	
		PET (71.8%)	Water (99.99%)	

* Including 1 wt% of EVA component.

2.5 Inventory Analysis and Impact Assessment

The inventory data used for the LCA analysis is summarized in Table 2. The data includes all the process inputs to recover the resins (solvents and process utilities). These inputs were estimated based on experimental mass balances and on the process simulations of the different STRAP variations. The STRAP process models were developed in Aspen Plus (V11 Aspen Technology).²⁰ The reported data is normalized based on the functional unit (1 kg of ML film). We can see that STRAP-A requires the largest amount of solvents and antisolvents. Utilities used in the process include steam, electricity, and cooling water. Steam is required for distillation and heating operations. Electricity consumption is a result of equipment usages such as dryers, filters, polymer grinders, and extruders. Cooling water is used for distillation and cooling operations (including the temperature reduction process). The water losses due to evaporation are considered in the cooling water requirements. The STRAP-A and STRAP-C approaches require the highest utilities (due to the use of antisolvent-based precipitation).

The factors to estimate the environmental impacts of the processes are obtained from the Ecoinvent and the Environmental Footprint databases. The Ecoinvent 3.6 cut-off by classification database³² is used for the solvent impacts since it has all the necessary factors for the solvents utilized in STRAP processes. Similarly, the impacts related to the production of polymers are modeled based on the Environmental Footprint database.³³ The software openLCA v1.10.3³⁴ is used to perform the LCA calculations with the Environmental Footprint (EF 2.0)³⁵ impact assessment method, providing fair evaluation and comparison for all products within the system boundaries. We observed high regional sensitivity (U.S. and Europe) for some impact categories regarding energy obtained from natural gas (see Supporting Information for more details). Because of this and the lack of data availability, we

Table 2: Annual utility and solvent usage of the STRAP processes for producing 1 kg of ML film.²⁰

Utility/Solvent	Unit	STRAP-A	STRAP-B	STRAP-C
Steam	MJ	82.69	9.51	21.20
Electricity	MJ	1.91	1.58	1.64
Cooling water	m ³	0.07	0.02	0.03
Acetone	ton	14.50	0.79	0.63
Toluene	ton	4.03	0.40	0.31
1-Propanol	ton	-	-	4.05
DMSO	ton	4.03	0.23	0.18
THF	ton	-	-	1.53
DMF	ton	-	-	2.56
Water	ton	135.79	36.67	28.80

present our case study based on European data (materials production, energy carriers, and transportation), which we found to be consistent.

2.5.1 Impact Categories

The impact categories considered in this study are climate change (kg CO₂ eq), energy use (MJ), water use (m³), freshwater ecotoxicity (CTUe), and human toxicity (cancer/non-cancer, CTUh). Climate change impact is an indicator of the global warming potential of the different greenhouse gas emissions (GHGs) of a system and takes into account gases such as carbon dioxide (CO₂), methane (CH₄), and carbon monoxide (CO). Energy use or energy demand are indicators of fossil fuel depletion and are thus key factors for assessing emissions related to fossil-based energy consumption. Water use is a measure of water consumption of the processes weighted by regional water scarcity (depending on the case study). This factor assesses the potential of water deprivation and pollution based on freshwater use and water quality loss at a spatial scale, accounting for water-stressed regions. Process water inputs are multiplied by location-specific water-stress indicators, ranging from 0.1 to 100, to assign higher impacts for water-stressed regions.^{36,37} Regarding toxicity impact factors, we consider freshwater ecotoxicity, a measure of impact on freshwater organisms exposed to toxic chemicals released into the aqueous environment, this measure is expressed as a comparative toxicology unit (CTUe). Also, we consider the human toxicity (cancer/non-cancer), estimated as the increase in human population disease rates based on a unit mass emission of chemicals, this category is expressed as a comparative toxicology unit (CTUh).³⁸

2.5.2 Process LCA

The process LCA is estimated based on the individual contribution of process inputs over the supply chain (raw materials, utilities, transportation, and extrusion) required to manufacture 1 kg of ML film. The impact of the virgin ML film production is estimated as:

$$I_{i, virgin} = \sum_k \underbrace{\left(\underbrace{IF_{i, k}}_{\text{production}} + \underbrace{IF_{i, trans}}_{\text{transportation}} + \underbrace{IF_{i, extr}}_{\text{extrusion}} \right)}_{\text{impact for resin } k} \times f_k \quad (2.1)$$

In Eq. 2.1, $I_{i, virgin}$ refers to the impact assessment for the category indicator i (climate change, energy use, water use, and toxicity factors) for different types of virgin film production. This value is estimated as the sum of the impacts related to the production ($IF_{i, k}$), transportation ($IF_{i, trans}$), and extrusion ($IF_{i, extr}$) multiplied by the material flow rate of each resin f_k . The production impact includes the required raw materials and process utilities. Similarly, the impact i of each STRAP process s for each resin k ($I_{i, s, k}$) is computed as shown in Eq. 2.2. The overall impact i of the STRAP process is estimated by the sum of the impacts for each resin k (Eq. 2.3).

$$I_{i, s, k} = \sum_l \underbrace{IF_{i, l}}_{\text{production}} \times f_{k, l} + \sum_m \underbrace{\left(\underbrace{IF_{i, m}}_{\text{production}} + \underbrace{IF_{i, trans}}_{\text{transportation}} \right)}_{\text{solvent impact}} \times f_{k, m} + \underbrace{IF_{i, extr}}_{\text{extrusion impact}} \times f_k + \underbrace{IF_{i, trans}}_{\text{transport impact}} \times f_k \quad (2.2)$$

$$I_{i, s} = \sum_k I_{i, s, k} \quad (2.3)$$

In Eq. 2.2, the different process impacts (production, transportation, extrusion) are multiplied by the flow rate of each category (utilities, solvent, extrusion, and transport). Specifically, $IF_{i, l}$ denotes the impact factor for process utilities (electricity, steam, cooling water), and it is multiplied by the corresponding flow rates $f_{k, l}$ required for separating resin k . $IF_{i, m}$ and $IF_{i, trans}$ denote the impact factors for the production and transportation of the solvents, and they are multiplied by the solvent flow rates ($f_{k, m}$). The impact of ML film extrusion is estimated by multiplying the extrusion impact ($IF_{i, extr}$) by the flow rate of each resin f_k . Similarly, the last term indicates the estimation of the impact related to the transportation of waste.

3 Results and Discussion

3.1 Climate Change Impacts: Product Perspective

This section compares the climate change impacts of producing an ML film via the STRAP process against ML film production from virgin resins by process utilities. The impacts are expressed per functional unit: the production of 1 kg of an ML film. The STRAP-A and STRAP-B processes are used to treat the ML film A1. Thus, we compare their impacts with the CO₂ eq. emissions related to the production of the same film from virgin resins. Also, we compare the climate change impact of STRAP-C and the CO₂ eq. emissions related to the production of film A2. We present the climate change impact of each method in Figure 3. The impacts related to the production, transportation, and extrusion of producing the virgin ML film are also shown in Figure 3. For the STRAP processes, we present the impacts related to the process utilities (steam, electricity, and cooling water), solvents

(including their transportation), extrusion of the film, and collection and transportation of ML film waste.

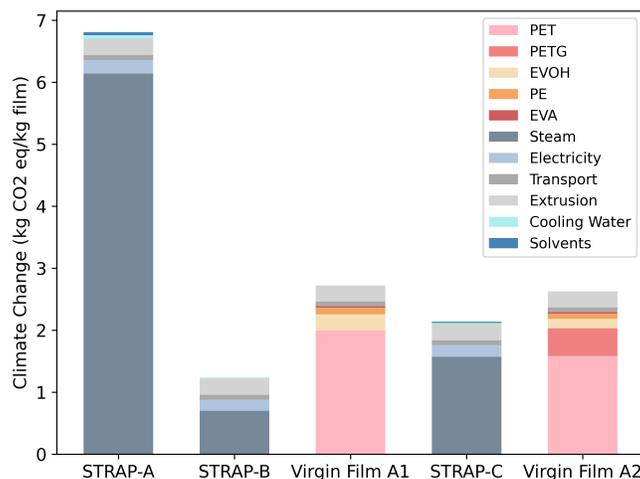


Figure 3: Impact on climate change of the production of 1 kg of A1 and A2 ML films through STRAP technology and through the production of virgin material (from virgin resins).

We can see that the STRAP-B and STRAP-C processes generate less CO₂ eq. emissions per kg of film than the virgin production of films A1 and A2. However, the STRAP-A process has a higher climate change impact, mainly due to the CO₂ eq. emissions associated with the required amount of steam used. This occurs because the precipitation method used in this approach (antisolvent addition) has high steam requirements (due to the use of distillation units). On the other hand, STRAP-B generates 54% fewer emissions than producing film A1 from virgin resins. These avoided impacts highlight the environmental benefits of the STRAP approach and the feasibility of using this process as an alternative for producing virgin-grade ML films. Since both STRAP-A and STRAP-B are used for the ML film A1, we observe that the STRAP-B process is more promising to replace the production of this film from virgin resins.

Regarding climate change impacts of producing the ML film A2, we can see that the STRAP-C process has a lower impact than the material virgin production. We find that STRAP-C generated 18% fewer CO₂ eq. emissions. These environmental benefits show that STRAP-C is a suitable alternative to produce the ML film A2 instead of producing it from virgin resins.

3.1.1 Climate Change Impact and Energy Use per Layer

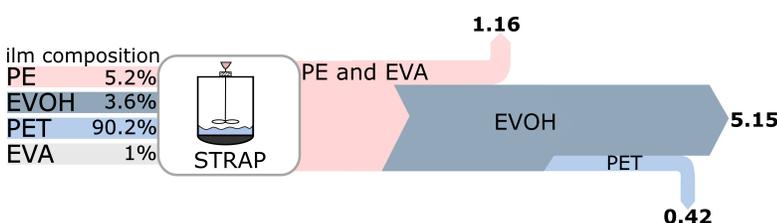
STRAP-A impact and energy use by each constituent polymer of film A1 are presented in Figure 4. The impacts are estimated based on the process inputs and utilities required to separate the corresponding polymer layer and produce (film extrusion) the ML film A1 via the STRAP-A process. We also compare these impacts to the ones related to the material processing to produce the different polymers required for the ML film A1 using virgin manufacturing. Figure 4 (b) shows the energy requirement comparison of the STRAP-A process (outer circle) with virgin film production (inner circle).

As expected, the climate change impacts have the same trend as the energy requirement of the process. For instance, although the EVOH only consists of 3.6 wt% of the film, the EVOH separation step in the STRAP-A approach is the main contributor to energy use and climate change, requiring 76 MJ of energy input with a climate change impact of 5.15 kg CO₂ eq. per kg of film A1. We can see that EVOH production demands less energy in the virgin film production approach. It only requires 6 MJ with a climate change impact of 0.27 kg CO₂ eq. The energy use discrepancy between the virgin manufacturing and recovering of EVOH via STRAP is related to the DMSO/water solvent/antisolvent separation that requires distillation in the STRAP technology.

The other major contributor to the STRAP-A process impacts is the PE separation step (with EVA component) which also requires distillation for toluene/acetone separation. It accounts for 17% of the process energy, requiring 17.56 MJ/kg film A1 and emitting 1.16 kg CO₂ eq./kg film A1. Again, the virgin PE production (with EVA component) requires less energy (4.16 MJ/kg of film A1).

PET recovery via the STRAP process resulted in an energy demand and climate change impact of 7.28 MJ/kg of film A1 and 0.42 kg CO₂ eq./kg of film A1, respectively. For this polymer, its virgin production requires more energy (57.44 MJ/kg of film A1) with a higher climate change impact as well (2.3 kg CO₂ eq./kg of film A1). However, due to the PE and EVOH separation steps, the overall energy requirement of the STRAP-A process is 47.4% higher (100.8 MJ/kg film A1) than the energy required in the manufacturing of virgin ML film (68.4 MJ/kg film A1), with 147.4% higher climate change impact.

a) Climate change impact (kg CO₂ eq./kg film)



b) Energy use (MJ/kg film)

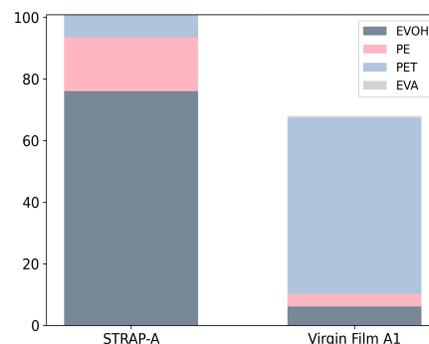


Figure 4: a) Climate change impact disaggregated by polymer layer in the STRAP-A process. b) Energy use comparison of the STRAP-A process with virgin film production by each polymer. The impacts include all the required steps to produce 1 kg of ML film A1 through the different manufacturing alternatives.

STRAP-B energy and climate change impacts per isolated polymer of film A1 are presented in (Fig. 5). The results show that the temperature reduction strategy of this approach reduces the process energy required by 82% compared to STRAP-A. This reduction leads to environmental benefits for the production of the ML film A1 through the STRAP-B process over virgin film production due to lower steam requirements.

In the STRAP-B process, the separation step of the component EVA is included. This step is the main driver for the process energy demand with 9.73 MJ and a climate change impact of 0.66 kg CO₂ eq. per kg of film A1. On the other hand, the virgin production of this polymer requires around ten

times less energy (as shown in Fig. 5 (b)). This difference suggests that avoiding the EVA recovery step can lead to higher environmental benefits. The resins PE and EVA could be recovered together (as in STRAP-A) since PE and EVA copolymer blends are widely used in packaging applications³⁹ and its removal is not necessary for the PE recycling.⁹

The PET layer has an associated energy demand and a climate change impact of 6.91 MJ and 0.41 kg CO₂ eq. per kg of film A1. These values are considerably lower than the virgin PET production impacts (57.44 MJ and 2.3 kg CO₂ eq.). The PE and EVOH separation steps require less energy than virgin film production and have a minor climate change impact. Overall, the STRAP-B process has 57% less climate change impact and 73.2% less energy demand than virgin film production. We can see that the contribution of each polymer in the virgin production method corresponds in general to the composition of the film. For instance, PET is the main component of the film and has the highest energy requirement in virgin film production. However, this behavior does not occur with the STRAP process because the impacts per polymer depend on the process units and inputs required to separate that resin.

a) Climate change impact (kg CO₂ eq./kg film)

b) Energy use (MJ/kg film)

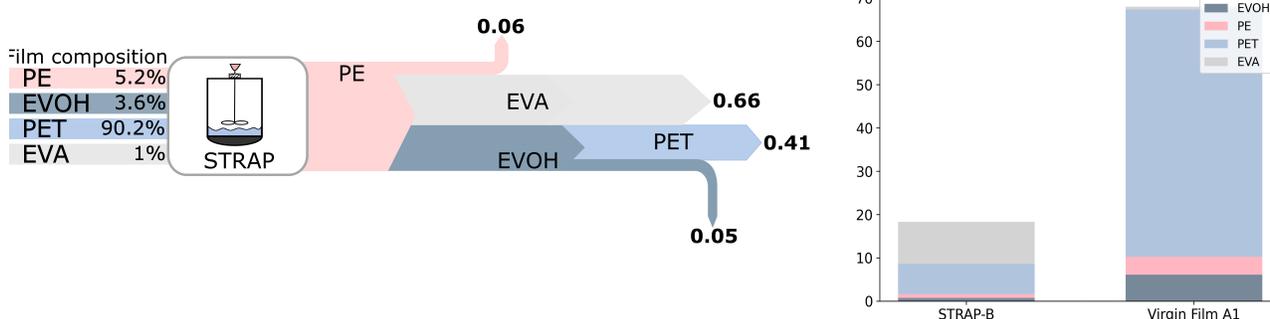


Figure 5: a) Climate change impact disaggregated by polymer layer in the STRAP-B process. b) Energy use comparison of the STRAP-B process with virgin film production by each polymer. The impacts include all the required steps to produce 1 kg of film A1 through the different manufacturing alternatives.

Fig. 6 illustrates the environmental benefits of modifying the STRAP-B process by recovering the polymers PE and EVA together. We can observe that avoiding the emissions generated by the separation of EVA (0.66 kg CO₂ eq. per kg of film A1), the STRAP-B modified process has now 81% less climate change impact than virgin film production.

STRAP-C environmental impacts are presented in Figure 7. As mentioned in the previous discussions, this process was designed for film A2 (which has the additional polymer component PETG). STRAP-C uses a combination of precipitation techniques: the PETG and EVA layers are separated via antisolvent and the remaining layers are recovered via temperature-driven precipitation.

Due to the complexity of film A2, the STRAP-C process requires 74.1% more energy than the STRAP-B approach; this clearly illustrates how the design of the film has a strong influence on overall process impacts. However, we found that STRAP-C has considerably lower energy demand (Figure 7 (b)) and climate change impact (2.07 kg CO₂ eq. per kg film A2) than the virgin film manufacturing alternative (2.63 kg CO₂ eq. per kg film A2). The main contribution to the process energy demand

a) Climate change impact (kg CO₂ eq./kg film)

b) Energy use (MJ/kg film)

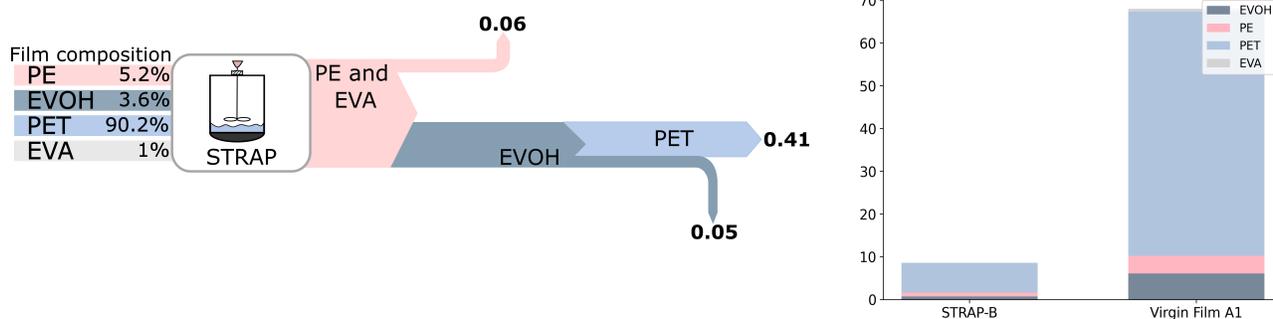


Figure 6: a) Climate change impact disaggregated by polymer layer in the STRAP-B modified process (PE and EVA are recovered together). b) Energy use comparison of the STRAP-B modified process (PE and EVA are recovered together) with virgin film production by each polymer. The impacts include all the required steps to produce 1 kg of film A1 through the different manufacturing alternatives.

and climate change impact comes from the PETG separation, which requires 17.21 MJ/kg of film A2 and generates 1.14 kg CO₂ eq./kg of film A2. The impact due to the EVA isolation represents 24% of the total energy demand (7.65 MJ/kg of film A2). These separation steps require distillation units for solvent/antisolvent separation and recycling which means a higher energy demand. The virgin production of the PETG and EVA components is less energy-intensive because it corresponds to the film composition.

a) Climate change impact (kg CO₂ eq./kg film)

b) Energy use (MJ/kg film)

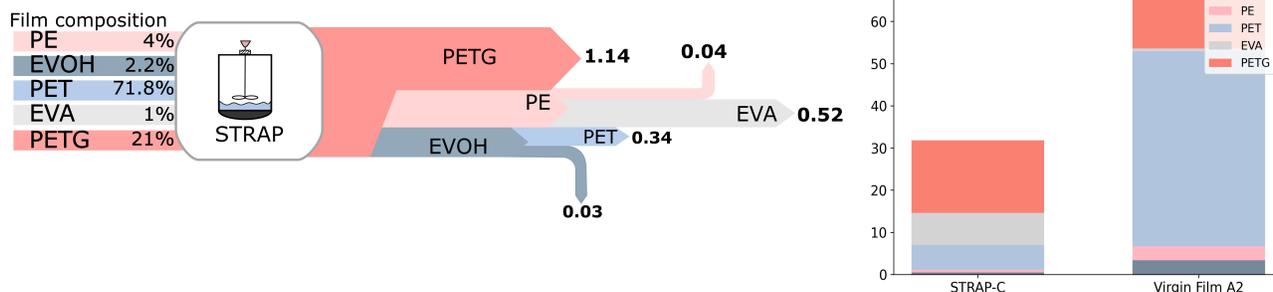


Figure 7: a) Climate change impact disaggregated by polymer layer in the STRAP-C process. b) Energy use comparison of the STRAP-C process with virgin film production by each polymer. The impacts include all the required steps to produce 1 kg of ML film A2 through the different manufacturing alternatives.

The remaining polymers (PET, PE, and EVOH) are separated in the STRAP-C process using the precipitation method of temperature reduction. Thus, their impacts are lower than the virgin film production. The PET layer (71.8 wt%) has an energy and climate change impact per kg of film A2 of 5.84 MJ and 0.34 kg CO₂ eq., respectively, which are considerably lower than the impacts of virgin PET production (45.73 MJ and 1.83 kg CO₂ eq.). The PE separation step has 80% and 60.0% less energy use and climate change impact than virgin PE production (3.22 MJ and 0.10 kg CO₂eq per kg film A2), respectively. Similarly, the EVOH layer in the STRAP-C has 86.5% and 81.25% less energy

demand and climate change impact than the virgin EVOH production (3.56 MJ and 0.16 kg CO₂ eq. per kg film A2).

3.1.2 Climate Change Impact per Processing Step

We also evaluated the climate change impacts associated with each piece of equipment of the STRAP process, in order to identify areas for improvement. The energy requirement is not discussed since the trends of this factor are the same as the climate change impact (as shown in the previous section). Figure 8 shows the GHG emissions of each piece of equipment in the STRAP-A approach. Here, the total impact of isolating each polymer and the impact related to the extrusion of the polymers to produce the recycled film (film extrusion) are presented. The sum of these two impacts (considering the film composition) results in the total impact per polymer.

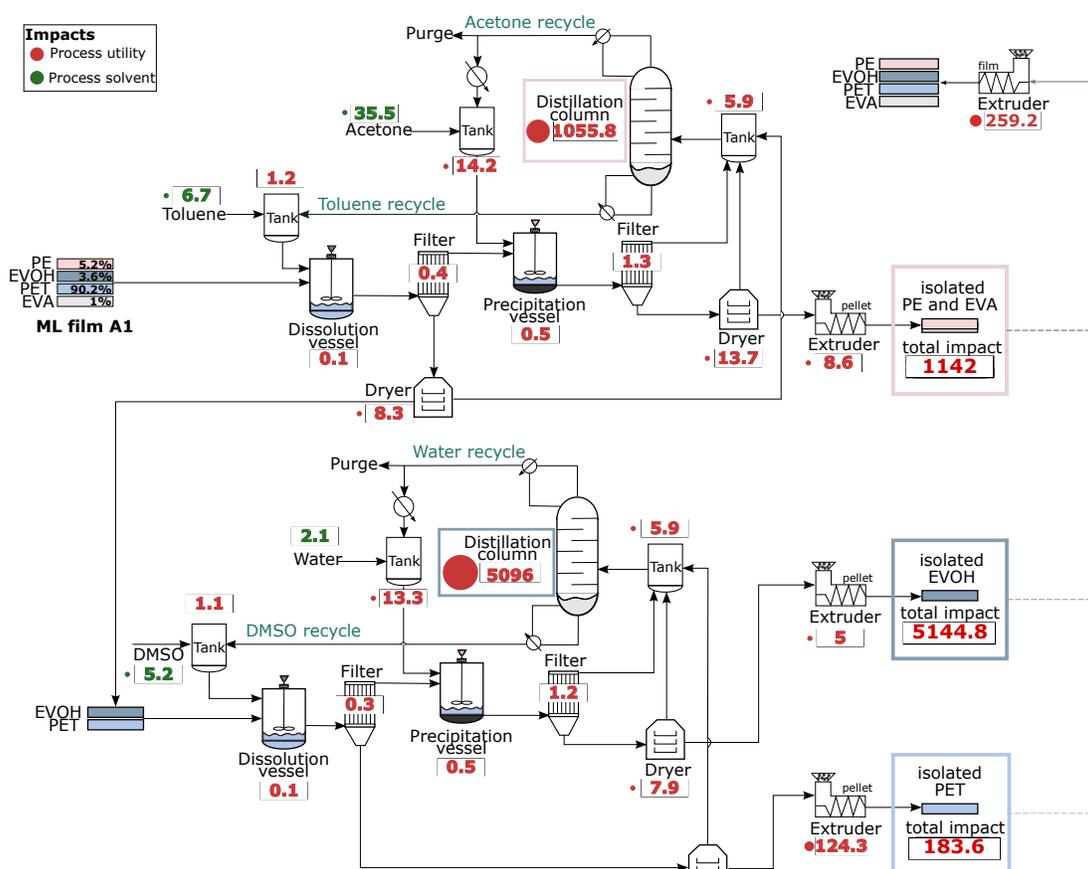


Figure 8: STRAP-A process diagram showing the climate change impact related to process utility (red circles) and process solvent (green circles) per each processing step in g of CO₂ eq. per kg of film A1. The size of the circle represents the amount of generated emissions.

Regarding the equipment climate change impact, we can see that the distillation column, extruders, solvent tanks, and dryers have the highest impacts. The impacts from solvent inputs are related to the production of the fresh solvents fed to the process and estimated by considering the individual solvent impacts multiplied by the make-up solvent used. We can see that these impacts are moderate due to the high solvent recycling rates in the process (around 99.9%). The distillation columns are

required to separate the toluene and acetone, as well as for the DMSO and water separation. Their climate change impact is much higher than the other equipment due to their energy requirement. It is ten times higher than the extruder impact, which is the second-highest contributor to the generated emissions. The high utility demands of the distillation columns make STRAP-A generate a higher amount of GHG than the virgin production method. The high energy demand of distillation columns might be due to factors such as the properties of the solvent/antisolvent solution to be separated and the distillation column inlet conditions (flow rate, temperature, pressure, etc.), as well as the high solvent recycling rate criteria set (99.88%) at the process design stage of STRAP.²⁰ In this regard, improvements in the energy demand of the distillation unit could be achieved by selecting solvents with a low boiling point or reducing the amount of antisolvent used.⁴⁰

Similarly, the climate change impact of each piece of equipment for the STRAP-B approach is presented in Figure 9. In this process, most of the precipitation of the polymers is done by lowering the solvent temperature, which results in considerably fewer GHG emissions for the STRAP-B equipment. However, the solvent/antisolvent precipitation strategy is also used to recover the minor component EVA. Here, a distillation column is required, which represents the highest climate change impact of the process. This result suggests that identifying solvents that allow the precipitation of EVA by temperature reduction or recovering PE and EVA together can be more environmentally benign. The other pieces of equipment that have high environmental impacts are the extruders and dryers.

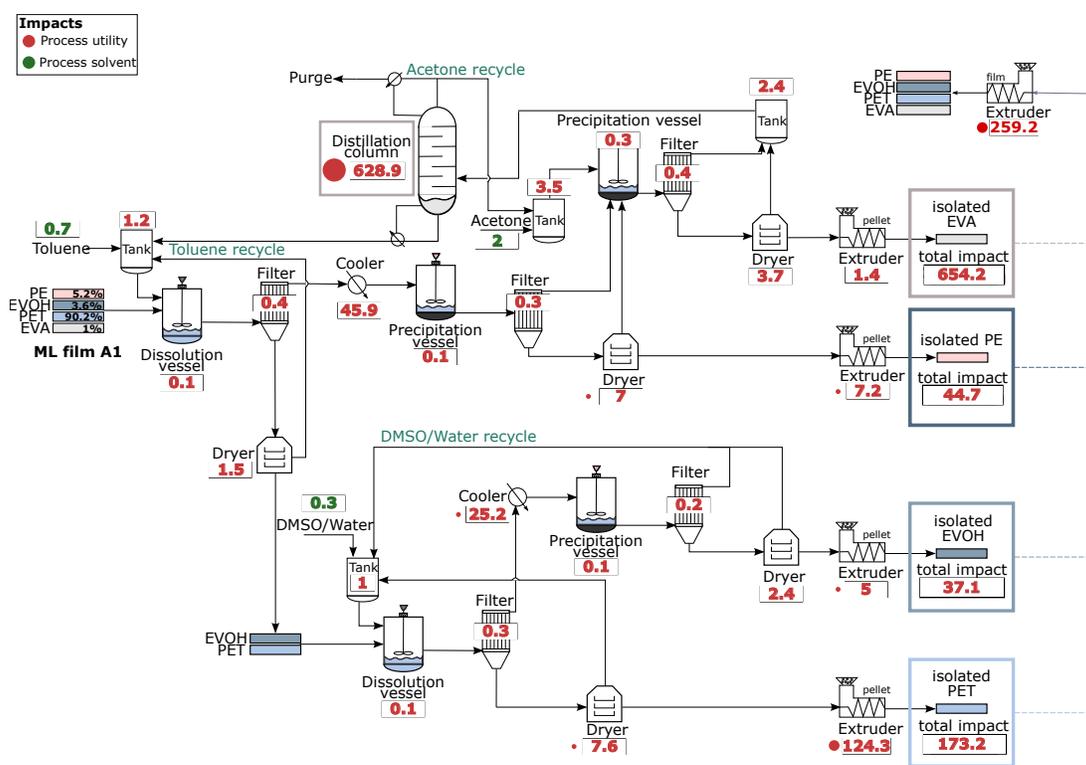


Figure 9: STRAP-B process flow diagram showing the climate change impact related to process utility (red circle) and process solvent (green circle) per each processing step in g of CO₂ eq./kg of film A1. The size of the circle represents the amount of generated emissions

3.2 Water Use: Product Perspective

For the water use analysis, we factor in the direct and indirect water consumption of the processes. We consider the direct water consumption related to cooling water and to the use of water as a solvent. Also, we consider the indirect water consumption associated with the use of water for the extraction of raw materials, production of solvents, and the generation of process utilities (e.g., electricity and steam).

The water use results show that the STRAP-B and STRAP-C approaches require less water than virgin film production (Fig. 10). Manufacturing virgin film A1 results in water usage of 0.70 m³/kg film A1. Of this amount, 90.2% is required for PET production, followed by 7.0% for EVOH production, 1.5% for EVA production, and 1.3% for PE production. Alternatively, recovering the constituent resins of film A1 via the STRAP-A process increases water usage due to the high cooling water requirements by the distillation units. On the other hand, STRAP-B requires 74.5% less water than STRAP-A and 45.7% less water than the virgin production method. For STRAP-C, the water usage is estimated as 0.58 m³/kg of film A2, performing 29.3% better than the virgin film A2 production (0.82 m³/kg of film A2). In all STRAP processes, the main source of water usage is the cooling water (mainly used in distillation operations) followed by water used in heat exchangers. More details on the water use impacts can be found in the Supporting Information.

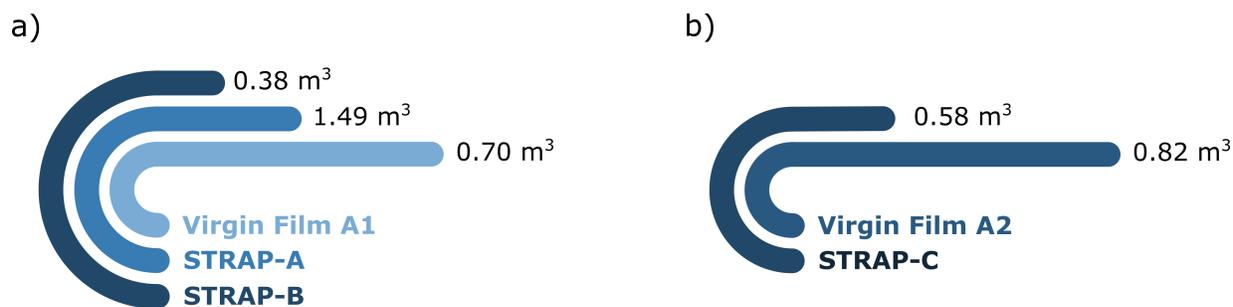


Figure 10: Water usage for a) manufacturing virgin film A1 and STRAP-A and STRAP-B processes, and b) manufacturing virgin film A2 and STRAP-C process.

3.3 Toxicity Impacts: Product Perspective

We now present the environmental impacts related to the toxicity of virgin film production and the STRAP technology evaluating their freshwater ecotoxicity and human toxicity (cancer/non-cancer) impacts. The freshwater ecotoxicity is expressed as a comparative toxicology unit (CTUe) and is a measure of the impact on freshwater organisms exposed to toxic chemicals released into the aqueous environment. Human toxicity (cancer/non-cancer) is expressed as a comparative toxicology unit (CTUh) as well and is estimated as the increase in human population disease rates based on a unit mass emission of chemicals.

Figure 11 (a) shows the freshwater ecotoxicity expressed in the comparative toxicology unit CTUe per kg of film (A1 and A2). For the virgin films, the ecotoxicity values are estimated based on the impacts of the individual polymer production, their transportation, and extrusion. The freshwater

ecotoxicity of the STRAP processes is estimated by considering the utility inputs (steam, electricity, and cooling water) and raw materials (solvents) used in the process, as well as the extrusion to make the new ML film and transportation of ML film waste. The results reveal that the STRAP-A and STRAP-B approaches are 63% and 89% less toxic than the virgin film A1 production. Similarly, STRAP-C is 86% less toxic than virgin film A2 production, suggesting that the STRAP technology is an environmentally safer alternative than virgin ML film production. In the STRAP process, steam is the main source of process ecotoxicity followed by cooling water. We performed a sensitivity analysis for the regional data variability on the toxicity values (see Supporting Information). This analysis reveals that the production method and fuel type used to generate the utilities can affect the process ecotoxicity values. Thus, the ecotoxicity impact can improve by switching to less toxic fuel sources for utility production. Similarly, process ecotoxicity can be reduced by using less toxic solvents. Solvent production impacts reveal that the STRAP-A and STRAP-C have the highest impacts due to the precipitation strategy.

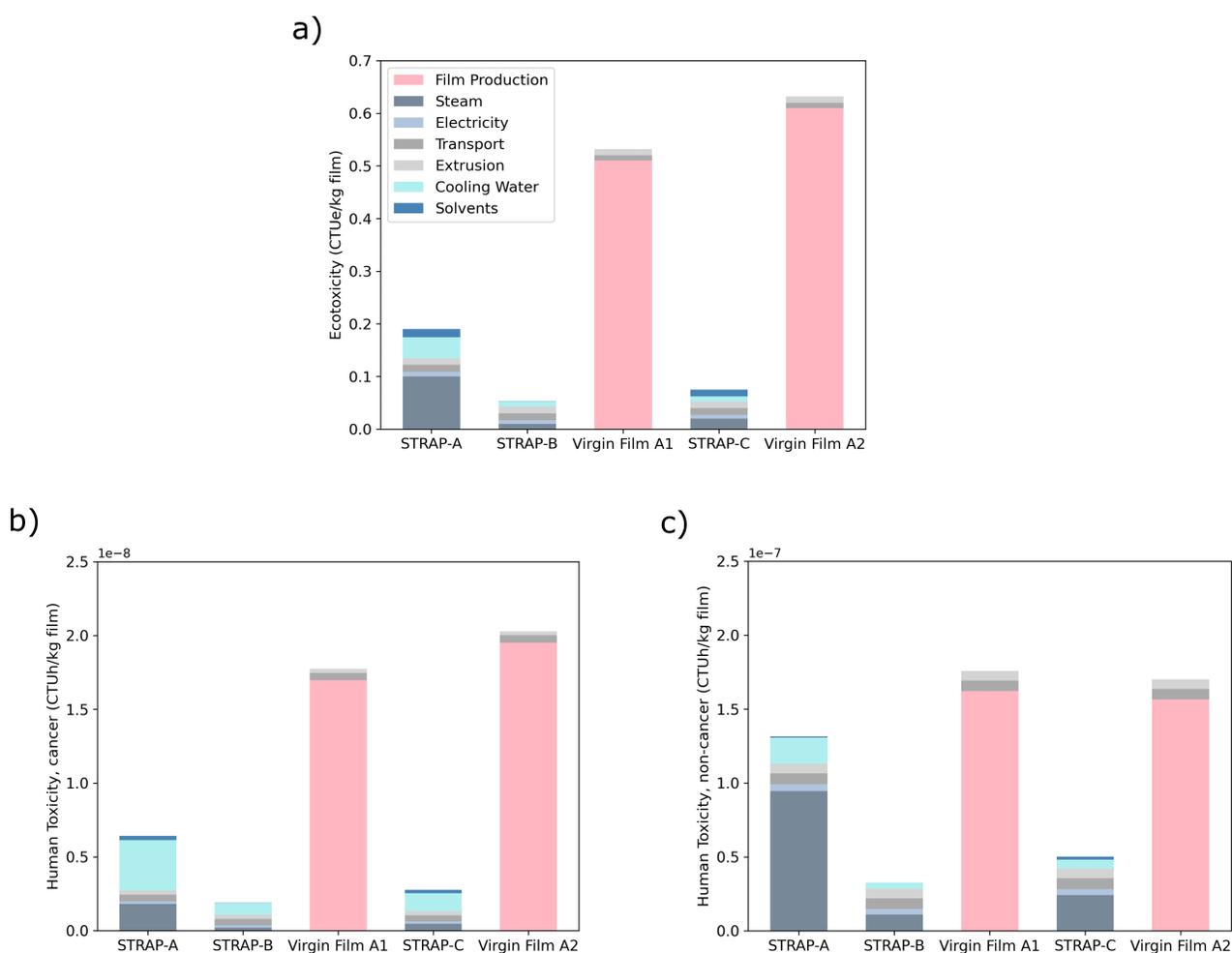


Figure 11: a) Freshwater ecotoxicity impacts (CTUe/kg of film), b) Human toxicity (cancer) impacts (CTUh/kg of film), and c) Human toxicity (non-cancer-impacts) (CTUh/kg of film)

The human toxicity impacts are presented as cancer and non-cancer (see Figure 11 (b) and (c))

and they are expressed using the comparative toxicology unit CTUh. We find that human toxicity results have the same trends as the ecotoxicity impacts. Therefore, the STRAP processes are considerably less toxic to humans than the corresponding film production from fossil-based virgin resins. Human toxicity analysis shows that, on average, STRAP-A is 44% and STRAP-B is 85% less toxic than virgin film A1 production, whereas STRAP-C is 78% less toxic compared to virgin film A2 production. Cancer-related human toxicity impacts show that the main source of toxicity for the STRAP approach is the process utilities (cooling water and steam). The significant contribution of cooling water to human toxicity (cancer) is due to the toxicity impact related to Chromium VI. We can see higher toxicity in STRAP-A and STRAP-C processes because of their higher steam and cooling water requirements. Also, we find that these approaches have higher solvent-related toxicity than STRAP-B. This behavior occurs due to the precipitation technique and the solvents used (mainly DMF and THF).

3.4 Sensitivity Analysis on Process Fuel Sources

We conducted a sensitivity analysis for the climate change impact of the process utilities by changing the energy source for producing steam and electricity. For this analysis, we assume that process energy can come from only one type of energy source. We evaluate different fossil and non-fossil energy sources. The potential of using various non-fossil energy sources to produce steam and electricity has been studied. For instance, Saygin *et al.*⁴¹ reported that the production of steam from biomass can be cost-effective and lead to a significant reduction of emissions. Furthermore, the economic and environmental advantages of meeting energy demands via the deployment of large-scale concentrating solar power systems have been widely studied.⁴²⁻⁴⁴ Similarly, the wind energy development for suitable regions has grown in the last years due to its low costs and emissions.^{45,46} Using nuclear energy and hydropower can also lead to low climate change impacts and are economically feasible alternatives.⁴⁷⁻⁴⁹ Therefore, we compare the impacts of the different energy sources with our base case scenario, which considers European data (due to data availability). For steam, the base case mix includes 75% natural gas, 12.5% hard coal, and 12.5% biomass. Figure 12 shows the impacts of changing the energy source to produce steam using only biomass, natural gas, or coal.

As mentioned in the previous sections, STRAP-A has the highest steam requirements due to the use of distillation units. Therefore, when we change the energy source for steam production, the STRAP-A approach achieves the highest environmental benefits. Producing steam from biomass reduces the climate change impact of STRAP-A by 85%. This reduction makes the STRAP-A process a better alternative than the virgin method. However, switching to natural gas and coal as energy sources results in higher climate change impacts. We can observe the same trends for STRAP-B and STRAP-C processes, in which using biomass leads to environmental benefits, and using fossil sources results in higher impacts. However, even using coal, the emissions of the STRAP-B approach are lower than the virgin alternative. On the other hand, if coal is used to produce the steam required in STRAP-C, then the climate change impact is higher than the virgin method. This behavior occurs due to the steam requirements of each process.

For electricity production, the base case mix includes 27.7% nuclear, 26.6% coal, 21.2% natural

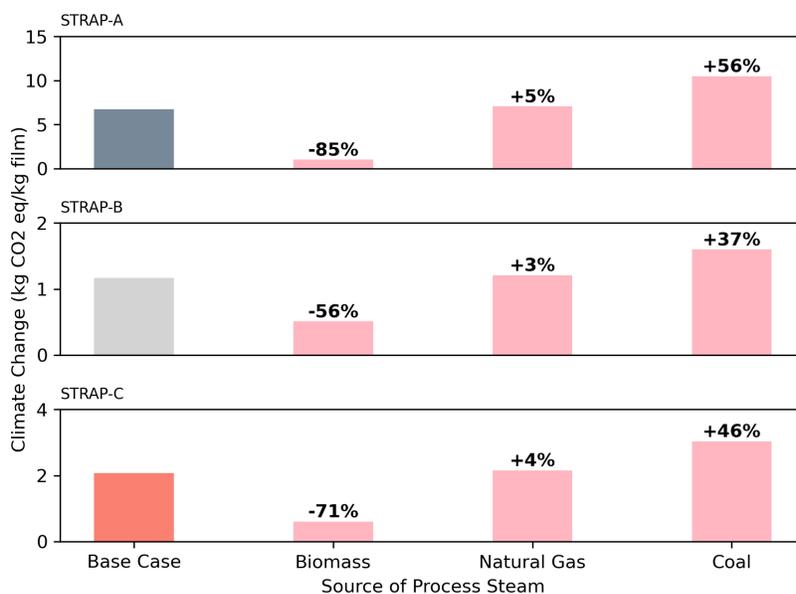


Figure 12: Sensitivity analysis for the climate change impact of the STRAP processes for the energy source to produce steam.

gas, 10.2% hydro, 5.4% wind, 2.2% heavy fuel oil, 2.2% biomass, 1.8% photovoltaic and geothermal, 1.2% biogas, and 1.2% waste. Figure 13 shows the impacts of changing the energy source to produce electricity using only non-fossil energy sources (solar energy, wind power, hydroelectric, and nuclear energy) or fossil energy sources (natural gas and coal).

We can observe that the variations in the climate change impact related to the source of process electricity are lower than the changes related to the source of process steam because the requirements of process steam are higher. Some trends are similar, including lower emissions with non-fossil sources and higher emissions with fossil sources. It is worth highlighting that the emissions reduction with the different non-fossil energy sources is similar. On the other hand, as expected, the increase in emissions from using coal as an energy source is significantly higher than the emissions related to natural gas. However, since the changes with the different sources are small, the impacts of STRAP-A remain higher than the virgin alternative. Similarly, the results of the base case of STRAP-B and STRAP-C do not change since all the energy sources result in a lower climate change impact than the virgin alternatives (even with coal as an energy source).

4 Conclusions and Future Work

This study presented an LCA methodology using a product perspective for assessing the environmental impacts of the STRAP technology. Previous work has shown that the STRAP technology is economically feasible for recovering the constituent polymers of ML film with nearly 100% material efficiency. In this approach, we evaluated the environmental benefits of producing two ML plastic films A1 and A2 (manufactured by Amcor) via the STRAP process. We compared the climate change,

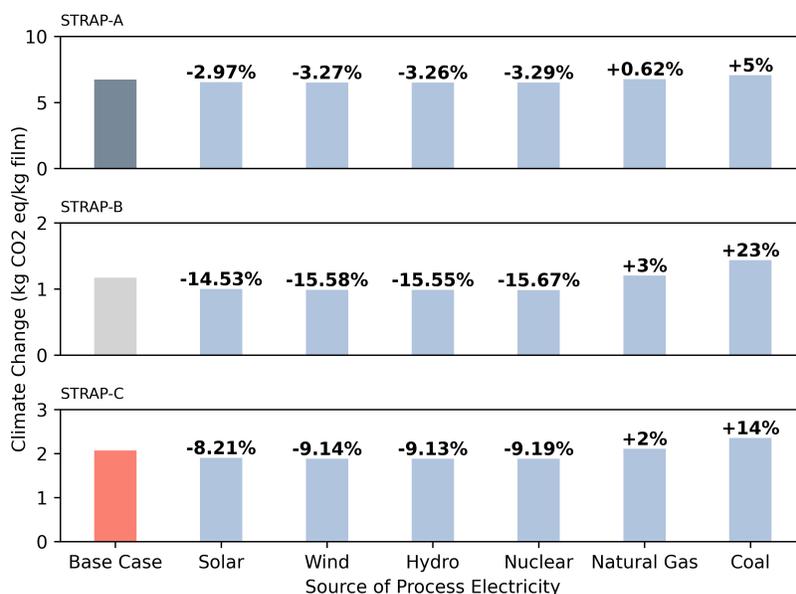


Figure 13: Sensitivity analysis for the climate change impact of the STRAP processes for the energy source to produce electricity.

energy use, water usage, and toxicity (ecotoxicity and human toxicity) impacts of producing 1 kg of ML film via virgin resin production or the STRAP approach. We presented the impact of recovering each layer or polymer (e.g., PET, PE, EVOH) to provide information for a more sustainable packaging design. Furthermore, we estimated the climate change impacts related to each piece of equipment of the STRAP process to identify areas for improvement.

For the ML film A1 (composed of resins PE, EVOH, EVA, and PET), we found that the STRAP-B process performs better than STRAP-A in all the impact factors due to the precipitation method (temperature reduction). Furthermore, the STRAP-B approach provides environmental benefits over virgin film production. Producing ML film via the STRAP-B technology requires 73% less energy and generates 57% fewer GHG emissions. We found that the polymer with the highest impact and lowest composition is EVA which suggests that this polymer should be recovered along with the PE since this is a widely used copolymer blend. The equipment with the highest impact was the distillation columns due to the energy requirements. Similarly, we found environmental benefits for the STRAP-C process over virgin resin production (film A2). This STRAP approach uses 52% less energy and generates 23% fewer CO₂ eq. emissions. Here, PETG is the polymer with the highest impact followed by the impact of the minor component EVA. We found that these impacts are related to the polymer precipitation strategy (antisolvent addition). Therefore, we conclude that temperature-driven precipitation is essential to achieve high environmental benefits.

For the STRAP-B modified process, we illustrated how, by separating two layers together (PE and EVA), the climate change impact can be decreased significantly due to the high energy requirements to separate EVA. For future work, we are interested in developing an optimization framework to identify the optimal process design including which layers to separate. We aim to identify optimal

processes for different types of ML films and provide insights for sustainable packaging. Additionally, we are interested in comparing the STRAP processes with other end-of-life disposal methods, such as pyrolysis, incineration, and landfilling.

In this work, we presented the environmental benefits of using the STRAP process to recycle PIW. Therefore, the environmental burdens of waste sorting and cleaning were not considered. For future work, we are interested in studying the environmental benefits of using solvent-based processes to recycle PCW and evaluating the impacts of sorting and cleaning.

Conflicts of interest

The authors declare no conflict of interest.

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