MOLECULAR RECYCLING OF PLASTICS FACILITATED BY SOLVENTS

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Abstract
Only a small fraction of the plastics produced is being recycled, with the great majority landfilled or released into the environment. Mechanical recycling is currently used to recycle plastic, however, this method is efficient only for homogeneous and non-contaminated feedstock, and for easily identifiable objects such as bottles made of PET or HDPE. Notably, mechanical recycling cannot easily handle plastic films which constitute about 40% of all plastic packaging used. Polyolefins in the plastic waste stream can be processed via pyrolysis, the most common among chemical recycling processes. Pyrolysis, however, decomposes the polymers, resulting in undesirable greenhouse gas (GHG) emissions. Further, pyrolysis is not viewed as constituting recycling when its product, pyrolysis oil, is not converted into new polymers. Plastics recycling research in our team utilizes physical, solvent-based processes that do not break down the polymer chains. This constitutes true plastic-to-plastic recycling. Such molecular recycling processes leave the polymer chains intact, thus maintaining their embodied energy and emitting relatively little GHG. Examples on molecular recycling are highlighted on the (i) separation of polyolefin mixtures through dissolution/precipitation in solvents that switch between a form that dissolves polyolefins and another that does not, hence facilitating the cycling and reuse of the solvent, and (ii) recovery of polyethylene from multilayer films via solvent-assisted delamination and separation, which retain the majority component, polyethylene, in the solid form, hence reducing greatly solvent amounts. The recovered polyolefins, following appropriate processing, can replace primary materials without loss of properties or performance and, hence, meet demand by customers and corporations to incorporate recycled plastics into products.

Keywords: flexible films, advanced recycling, chemical recycling, LCA, circular economy, sustainability

1. Introduction and Motivation
Plastics are useful because their properties are modular and amenable to first principles design. We are approaching half a billion tons of plastic annual production worldwide, which is one trillion (!) pounds, and further growth is expected.[1] Durability, low density, color, and low cost make plastics desirable and long-lasting, but disposable. Thus, plastic waste is increasing, and less than 10% is recycled in the US, with most plastic being landfilled.[2] Opportunities abound for recovering plastic, but technical and economic challenges limit plastics recycling.[3]
Packaging consumes a large fraction (close to half) of the plastics used world-wide. Plastic film production devotes an estimated 25% of production volume to manufacturing multilayer films.[4, 5] Multilayer films are beneficial, even essential, in several applications, notably in food and pharmaceutics.[6] Multilayer packaging achieves superior protection and durability through the integration of the unique functionalities of the constituent polymers (Figure 1). The major component of multilayer packaging is a polyolefin, on the basis of its low cost, easy processing, excellent moisture barrier properties, and chemical inertness, however, most multilayer packages consist of three or more layers of various polymers, often with an added barrier layer or coating.[6] As the expectations and specifications for plastic packaging functions become ever more demanding, the complexity of packaging is increasing. Up until now, the great majority of polymer-based multilayer packaging ends up in landfills. This leaves a lot of value to be recovered.

Plastics offer unique, often irreplaceable, properties and will continue to be used,[6] but plastic waste threatens the environment.[7] Even if sorted, multilayer plastics and flexible films cannot be reprocessed and are being landfilled.[4, 8] While pyrolysis can recover value from such plastics, this involves high energy demand and high greenhouse gas (GHG) emissions.[9] These basic premises underscore our research on the solvent-assisted molecular recycling of plastics with a focus on polyolefins and multilayer films.

Upcycling of sorted plastic is achieved through dissolution of select types of plastic in environmentally responsible solvents to recover desirable materials (i.e., polyolefins), separate them from additives or impurities, and render them suitable for reuse. New routes are developed for the recycling of polymer-based multilayer packaging by addressing the delamination of multilayer packaging into the individual constituent films, and the fractionation of the different types of plastic. The innovations advanced here will enable the recovery from post-consumer multilayer packaging waste of useful plastics such as polyolefins, for which great demand exists for incorporation into new products with certain minimum recycled plastic content. Delamination is energy-efficient and low-GHG emitting, as it leaves the majority of the polymer content of multilayer films in its original, solid state, with no polymer degradation involved.

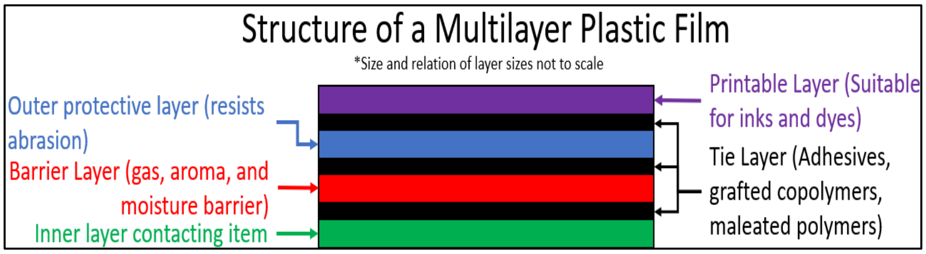

Figure 1: Schematic of a multilayer film.

2. Review of Related Work

Plastic film waste buildup has become a global problem. Attempting to reduce use or substitute plastics are good steps to reduce overall film waste volume, but are insufficient.[10] Recycling of plastic film waste is needed. The following processes can be used to this end: mechanical recycling, pyrolysis, dissolution-precipitation, and delamination. Life cycle assessment can be used to evaluate and compare the environmental impacts of the various processes.

2.1. Mechanical Recycling

Mechanical recycling is the prevalent method used for plastic waste (Figure 2), yet the great majority of current mechanical recycling processes do not handle plastic films. Material Recovery Facilities (MRF) avoid films due to the extra costs in cleaning and handling, and the low-quality of the recycled polymer obtained from plastic film waste[11].

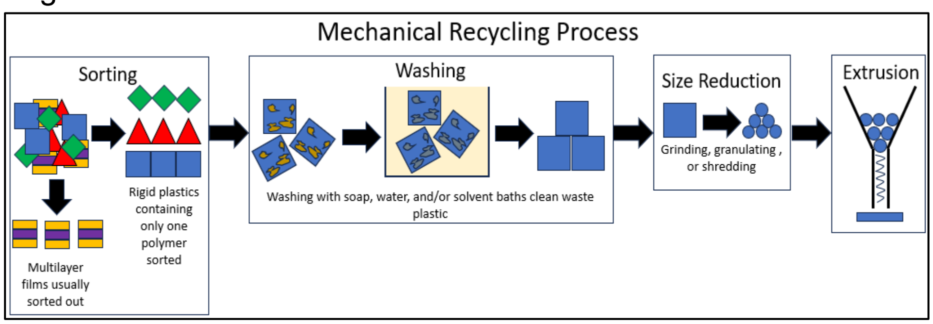

Figure 2: Schematic of mechanical recycling.

Despite these issues, there have been positive developments in the mechanical recycling of plastic films. For example, Waste Management disclosed a pilot scheme for recovering plastic film waste in Illinois as of 2022.[12] AMP Robotics is utilizing AI-powered sorting to sort films.[13] Improvements to sorting technology are valuable to MRFs due to challenges posed by multilayer film waste.[14, 15] EREMA is a company that produces extruders capable of handling both post-industrial and post-consumer plastic film waste for mechanical recycling.[16] Should advancements in sortation and mechanical recycling continue, plastic films could eventually be a viable feedstock for MRFs.

2.2. Pyrolysis

When mechanical recycling is not an option, chemical recycling can be an alternative. The most common method of chemical recycling, pyrolysis, involves thermal decomposition of long-chain polymers at high temperatures and no oxygen (Figure 3).[17] Concerns for pyrolysis include its energy consumption and products.[17] The various hydrocarbons produced from pyrolysis need further processing to be turned in a desired product.[17] Pyrolysis of waste plastic requires large feedstock volumes to be profitable, and the profitability is very sensitive to the (volatile) price of petroleum.[18]

Pyrolysis does have benefits, as it can handle mixed plastic waste, including contaminated feedstocks.[19] TotalEnergies has pyrolysis plants in Spain, France, and the US that can use flexible plastics as feedstock.[20, 21] According to TotalEnergies, products from its pyrolysis plants are intended to produce high quality materials with a portion to be used for food-grade resins.[20] Pyrolysis offers a complement to mechanical recycling, however GHG and high energy consumption attenuate the appeal of pyrolysis of plastic film waste.

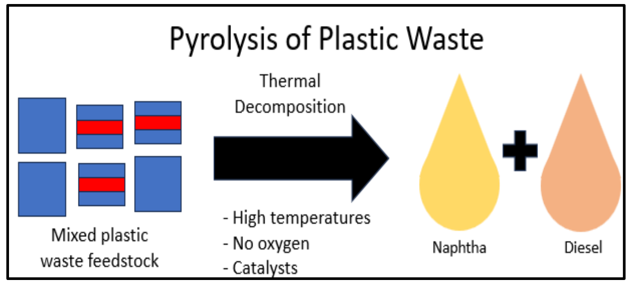

Figure 3: Pyrolysis process and products.

2.3. Dissolution-Precipitation Recycling of Plastic Films

Dissolution-precipitation provides a means of recycling plastic film waste without the high energy consumption and emissions of pyrolysis. Dissolution-precipitation recycling works by dissolving each of the polymer layers (one at a time) creating a solvent slurry (Figure 4).[22] Following filtration of undissolved solids, dissolved material is collected via precipitation.[22] Precipitation is generally performed using an antisolvent, cooling, or a combination of the two.[8] Solvent selection should account for risks inherent to specific solvents, as hazardous solvents like toluene are often used based on compatible solvent-solute affinity.[8] The precipitated polymer is almost the same as the starting material, is its molecular weight distribution may be affected.
An application of dissolution-precipitation to the case of plastic film is the so-called solvent targeted recovery and precipitation (STRAP) process that works by taking a characterized and identified multilayer plastic film and uses specific solvents and antisolvents to dissolve all polymers present sequentially, layer by layer.[23]

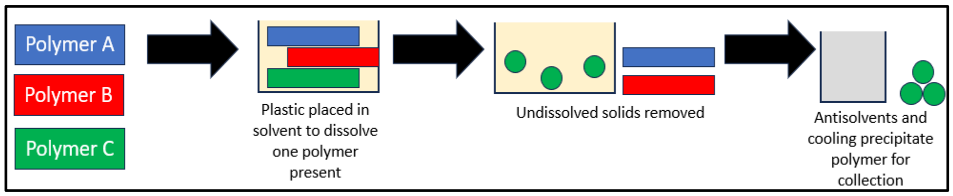

Figure 4: Schematic of dissolution-precipitation process.

2.4. Life Cycle Assessment (LCA)

LCA is used to evaluate environmental impacts of a product or process, from raw material extraction to disposal[24], as well as to identify opportunities for improvement and inform decision-making, by allowing comparisons between current and prospective approaches.[25, 26] LCA is valuable in competitive markets, as environmentally friendly products and services are increasingly valued by consumers.[27, 28] LCA can also identify areas for improvement, including end-of-life management, materials replacement, energy reduction opportunities.[29, 30]
For polymer-based multilayer packaging (PMP), LCA has been used to identify opportunities to reduce material use[31], improve recycling rates[32], and/or apply renewable energy sources in manufacturing.[33] In the case of dissolution-precipitation, a comprehensive LCA of various PMP waste management (WM) practices is required to ensure a thorough evaluation of environmental impacts, and facilitate informed and sustainable choices. An LCA can be defined in different boundaries (Figure 5) that include or exclude certain process steps, depending on the goal and scope of the analysis.

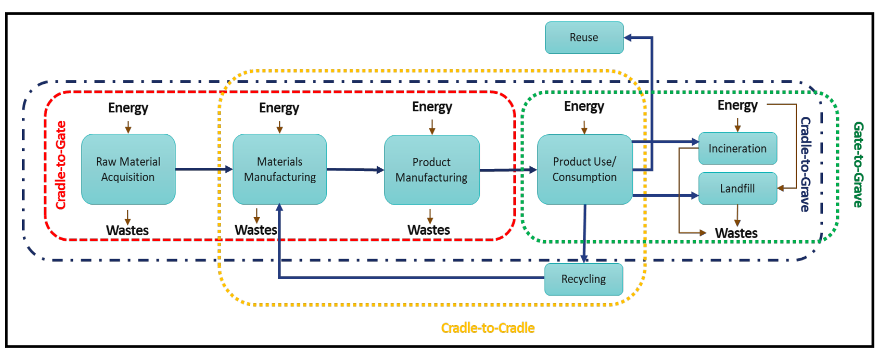

Figure 5: Various boundaries that can be defined in an LCA study.

3. Technology Approach

3.1. Polyolefin Dissolution in Plastics Recycling

In order to obtain insights on the underlying mechanism of polyolefin dissolution, and assess the impact of temperature and other operating conditions in this process, in this project we designed an experimental setup to obtain over time:

(i) mass of dissolved polyolefin; and (ii) crystallinity of undissolved polyolefin. To obtain the time evolution of polymer crystallinity, we used temperature-controlled FT-IR spectroscopy (Figure 6), and to obtain the polymer dissolution rate we measured the polymer mass loss at specific time intervals. In addition to the experiments, a comprehensive mathematical model to describe the swelling and dissolution of polyolefins has been developed that captures the transport phenomena and thermodynamics governing this process.[34]

The experimental data are used to validate the model and to obtain values for the two fitted parameters, rate of disentanglement (r_{dis}) and decrystallization constant (K_{dec}), which have physical interpretation regarding the mechanism of polyolefin dissolution (Figure 7), The data shown here are obtained in xylene, a well-known polyolefin solvent, but the dissolution mechanism should apply to other solvents. Further, the validated model is employed to perform a parametric analysis by simulating different possible processing conditions in order to assess the impact of different parameters. The model indicates that the dissolution process of semicrystalline polymers can be controlled by either r_{dis} or K_{dec}. When decrystallization is important, the dissolution rate increases more with a reduction in crystallinity compared to reduction in the film thickness. However, when the dissolution is controlled by polymer chain disentanglement, a reduction in the thickness could accelerate dissolution. Very few articles have published experimental data on polyolefins dissolution, dating back to the 1970s and pertaining only to the dissolution of PP.[35, 36] Further, no published articles address modeling of polyolefin dissolution kinetics.

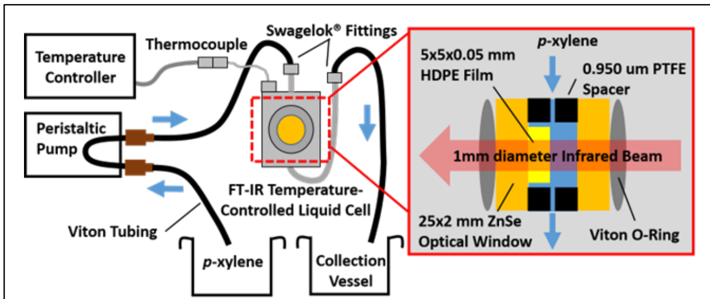

Figure 6. Schematic of the FT-IR temperature-controlled liquid cell used to study polyolefin dissolution kinetics in this study.

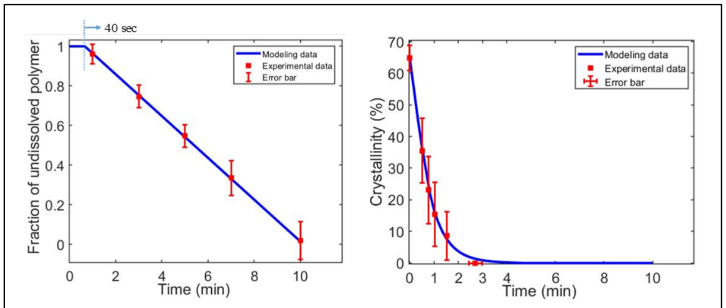

Figure 7: Fitting experimental results (dots) with modeling predictions (line): (a) fraction of undissolved 0.1mm thick HDPE film in *p*-xylene; (b) crystallinity over time at T=104 °C.

3.2. Delamination of Plastic Films

The flexible films used in packaging often involve more than one types of plastic (Figure 1) and more than just plastic. The addition to plastic film of paper or metal like aluminum leads to waste that is difficult to recycle due to being heterogenous.[37] Delamination, or separation of layers, focuses on sequestering one type of polymer. This can be done by targeting adhesives to separate layers, or through attacking the layer that the plastic is adhered to, such as aluminum.[37] Once the multilayer film is separated via delamination, the remaining strips can then be extracted as monolayer film strips (Figure 8). Compared to the dissolution-precipitation process, delamination is less energy and emission intensive as it does not dissolve the majority of plastic. One of the drawbacks of delamination is solvent selection, as the adhesive or tie layer can make selecting a proper solvent difficult.[38] Delamination is beneficial for dealing with flexible plastic as it recovers solid plastic layers without the polymers degrading[39]. Pretreatment of plastic waste would benefit the delamination.[40]

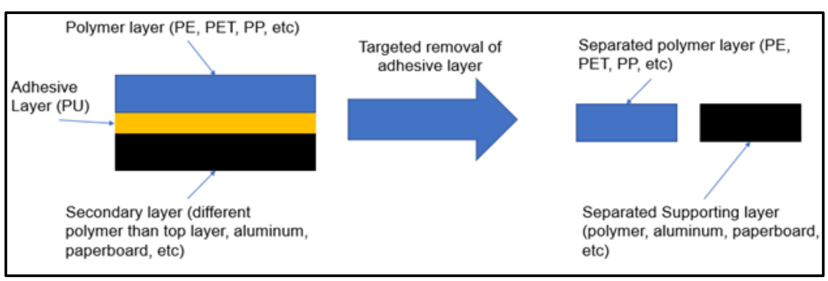

Figure 8: Schematic of delamination process.

Delamination relies on targeting for recovery a specific layer of the plastic film waste. Removal of an adhesive layer would promote separation and allow for recovery of the different layers present in the flexible plastic waste.[41] Carboxylic acids are reported as good delaminating agents for PET/PE (polyethylene terephthalate / polyethylene) films, but the environmental impact that they have may present a hindrance on a large scale.[39]

In our work we have achieved solvent-based delamination in four different multilayer plastic film compositions in under an hour. The four film compositions are: LLDPE/Nylon 6/LLDPE, LLDPE/EVOH/LLDPE, HDPE/EVOH/HDPE, and LLDPE/PET. Delamination is induced by dissolving a minority by mass binding layer (Figure 9) or by partial dissolution and swelling of a targeted polymer layer. Solvents used include carboxylic acids and phenols. The delamination process is performed at a relatively mild temperature (80°C) and results in complete recovery of the polyethylene present initially in the film. Recovered PE purity ranges from 98% to 100% based on solvent selection, temperature, and the time that film spends in the solvent. The delaminated films are recovered through density separation in the delaminating solvent, with all PE being skimmed off the surface of the solvent. The remaining solvent is collected and recycled in a closed-loop process.

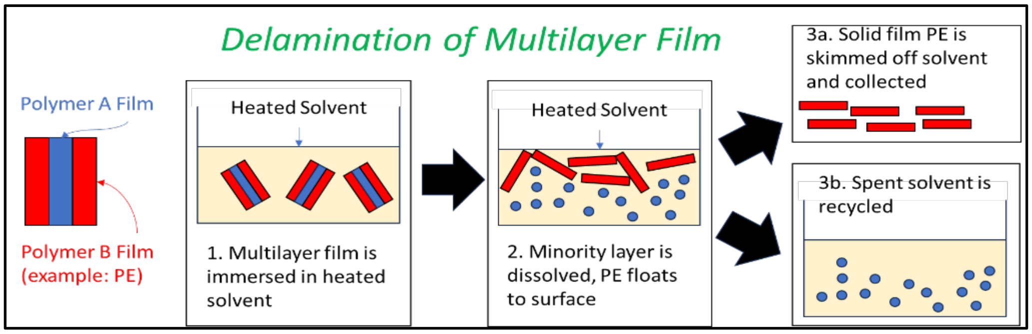

Figure 9: Delamination process via dissolution of minority by mass layer.

3.3. Life Cycle Assessment (LCA) of Plastic Film Recycling

Most published LCA studies of polymer-based multilayer packaging have been conducted in Europe[42-46], and rather limited research on this topic in the US, with a focus on their entire life cycle.[47, 48] PMP waste in the US is typically landfilled or incinerated[49]. Available studies on PMP recycling primarily focus on packaging having PET as the major component [50, 51]. However, polyethylene (PE) and its variants account for the largest share of plastic waste, particularly in plastic packaging [52, 53]. This research addresses the relative sustainability of PMP waste by conducting LCA of the solvent delamination recycling method developed in this project, alongside incineration and landfilling PMP waste management.

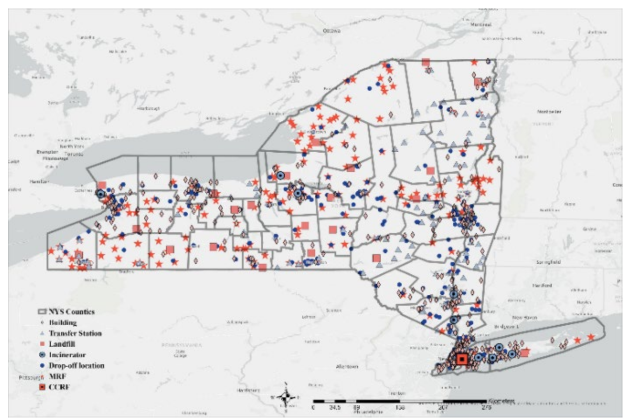

Figure 10: Distribution of waste management facilities in NYS, including the proposed recycling facility (CCRF) and randomly selected buildings and retail return drop off sites considered in this study.

The Cradle-to-Grave boundary (Figure 5) was selected for virgin PMPs. However, a Cradle-to-Cradle approach emphasizes a closed-loop system associated with the circular economy which is considered for the recycling method developed in this study. In this approach, waste generated after PMP use is considered a resource that can be reused as raw material in the next cycle of production. This circular approach minimizes waste and reduces the need for resource extraction, leading to sustainable products. This research considers explicitly post-customer transportation which has been neglected in previous packaging life cycle studies. This work quantifies the environmental impact of post-customer transport of PMP in NYS, taking into account current and future waste management possibilities. Actual waste management facilities, including transfer stations, landfills, incinerators, MRFs, and retail return drop off sites were mapped, and travel distances calculated for a statistically representative subset of NYS buildings. Further, a proposed central chemical recycling facility was considered (Figure 10). Compared to landfilling, total transportation GHG emissions are 64%, 264%, and 158% higher for incineration, MRF with chemical recycling, and retail return with chemical recycling, respectively. This emphasizes the value of well-dispersed and frequent landfill locations, relative to the few-and-far-between chemical recycling and incineration sites – a possible hindrance to advancing film recycling in the US.[54]

A comparison between the results obtained from LCA of the determined landfilling and incineration scenarios, in the Cradle-to-Grave boundary, showed that the manufacturing phase has the highest impact compared to the post-consumer phase in both scenarios. The assessment revealed that almost half of the total fossil-based CO_2 emitted into the air from PMP production is associated with the production of ethylene from petroleum and natural gas.

4. Discussion

4.1. Perspective on Dissolution-Precipitation Molecular Separation of Plastics

Dissolution-precipitation can be environmentally friendly for plastics recycling. Vollmer et al.[55] found that switching from incineration to dissolution/precipitation recycling can reduce CO_2 emissions by up to 75 %. Samori et al.[56] present LCA findings for formic acid treatment, pyrolysis, and SHS (switchable hydrophilicity solvents) that highlight favorable results for dissolution-precipitation employing the SHS process that utilizes CO_2 to switch between a solvent form that dissolves polyolefins and a form that does not, and thus allows polymer dissolution and precipitation to be accomplished in the same solvent with no need for antisolvent and the associated solvent separation steps and expenses. This study emphasized the significance of integrating green chemistry principles at the initial design phase.[56]
This project develops an efficient dissolution-precipitation recycling process targeted to polyolefins such as LDPE, HDPE, and PP. We investigate solvents like amine-based SHS, non-amine-based SHS, and non-SHS and address challenges related to incomplete polymer precipitation. Successful implementation could elevate recycling rates, contribute to a circular plastic economy, and pave the way for recycling diverse polymer types while mitigating environmental and health risks associated with hazardous additives.

4.2. Perspective and Future Directions on the Delamination of Plastic Films

Our work has successfully induced delamination in under an hour for four different film compositions. The recovered solid films are of high purity and recover all polyethylene initially present in the films.
Further work towards the precipitation of dissolved materials form solvent, the recycling of solvent for reuse in the delamination process, and a better understanding of the energy and materials cost of the process are required. Precipitating out dissolved materials allows for the recovery of all materials present and not just the desired polyethylene. Additionally, spent solvent should be reused for as many process cycles as possible, and the efficiency of recycled solvent in the delamination process should be documented.

4.3. Perspective and Future Directions on LCA of Plastic Film Recycling

This study highlights key phases in the life cycle of polymer-based multilayer packaging, assessing the environmental impact of all steps of the life cycle for both virgin and recycled PMP. Identifying the most and least impactful stages aids decision-makers in choosing sustainable options. Our results reveal significant environmental impacts in the production of virgin PMPs, emphasizing the potential benefits of recycling approaches, through reducing the environmental impact of manufacturing virgin PMPs and the need for landfilling and incineration.
To bridge the gap between innovation and industrial implementation, future research should focus on optimizing recycling processes for efficiency and economic viability at industrial scale. This entails refining technology, addressing scalability, and maximizing resource utilization. Additionally, efforts should enhance the economic viability of recycling technologies by streamlining production processes and reducing operational expenses.

5. Conclusions and Recommendations

Polyolefins comprise the largest volume of commodity plastics, but they can be difficult to recycle due to the molecular-level incorporation of other plastics, additives, colorants, and/or fillers.
The solvent-based polyolefin separation methodology that we pursue is environmentally responsible on the basis of the solvents selected and the greatly reduced CO_2 emissions (no polymer chains break down), and can be readily implemented in industrial practice and extended to the separation of other types of plastics.
The delamination of multilayer films developed here leaves intact, in the solid state, polyethylene which is the majority component of the film and main target for recovery and recycling. This solvent-based multilayer film delamination process is environmentally responsible on the basis of low energy usage (relatively mild temperatures and low solvent usage as the majority of the polymer remains solid) and low GHG emissions.
The different types of polyolefins to be recovered from the solvent separation process can meet the demand by both customers and corporations to incorporate recycled plastics into products while maintaining desirable specifications.
Collaboration with industry stakeholders, policymakers, and waste management systems is crucial for practical application of any recycling technology, including the multilayer film delamination approach developed in this project. Adapting methods to meet industry standards and recognizing regulatory landscapes are vital for successful implementation. A multidisciplinary approach, integrating technological innovation and economic feasibility, is essential to transition research from the lab to industrial practices, making a tangible impact on plastic recycling.

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Prof. Alexandridis' research utilizes molecular interactions and supramolecular assemblies to develop processes that are environment friendly and energy efficient, and products with desired properties and function. He is leading projects on chemical recycling, recycling of multilayer films, and recycling of textiles. He has authored over 200 articles and 6 US patents (Google Scholar h-index 83 and 25,700 citations). [www.cbe.buffalo.edu/alexandridis]

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Marina Tsianou

Prof. Tsianou leads research that involves the design, development, and characterization of molecularly-engineered materials with desirable functionalities. She contributes to plastic recycling with expertise in nanostructured polymers in films and on surfaces, polymer dissolution, nano- and meso-scale organization and characterization.

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