ARTICLE IN PRESS

Nature Communications

https://doi.org/10.1038/s41467-026-68936-w

Article in Press

Cracking the code of multi-layer films to promote circularity in single-use plastic packaging

Received: 4 September 2025

Accepted: 21 January 2026

Ethan C. Quinn, Levi J. Hamernik, Jeffrey N. Law, Ryan W. Clarke, Maya Milrod, Shivani Kozarekar, Rebecca M. Mick, Margaret J. Sobkowicz, Linda J. Broadbelt, Brandon C. Knott & Katrina M. Knauer

Cite this article as: Quinn, E.C., Hamernik, L.J., Law, J.N. *et al.* Cracking the code of multi-layer films to promote circularity in single-use plastic packaging. *Nat Commun* (2026). https://doi.org/10.1038/s41467-026-68936-w

We are providing an unedited version of this manuscript to give early access to its findings. Before final publication, the manuscript will undergo further editing. Please note there may be errors present which affect the content, and all legal disclaimers apply.

If this paper is publishing under a Transparent Peer Review model then Peer Review reports will publish with the final article.

ARTICLE IN PRESS

© The Author(s) 2026. **Open Access** This article is licensed under a Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International License, which permits any non-commercial use, sharing, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if you modified the licensed material. You do not have permission under this licence to share adapted material derived from this article or parts of it. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit http://creativecommons.org/licenses/by-nc-nd/4.0/.

Cracking the Code of Multi-Layer Films to Promote Circularity in Single-Use Plastic Packaging

Ethan C. Quinn,[1,2,3,‡] Levi J. Hamernik,[2,3,‡] Jeffrey N. Law,[2,‡] Ryan W. Clarke,[4] Maya Milrod,[5] Shivani Kozarekar,[6] Rebecca M. Mick,[7] Margaret J. Sobkowicz,[8] Linda J. Broadbelt,[6] Brandon C. Knott,[2,]* Katrina M. Knauer[2,3,]*

[1]Department of Chemistry, Colorado State University, Fort Collins, CO 80523, United States.
[2]Renewable Resources and Enabling Sciences Center, National Laboratory of the Rockies, Golden, CO 80401, United States.
[3]BOTTLE Consortium, Golden, CO 80401, United States.
[4]Hexion Inc., 180 East Broad St., Columbus, OH 43215, United States.
[5]Department of Chemical and Biological Engineering, McCormick School of Engineering and Applied Science, Northwestern University, Evanston, IL 60208, United States.
[6]Department of Chemistry, Northwestern University, Evanston, IL 60208, United States.
[7]Association of Plastic Recyclers, 2050 M Street NW, Washington, DC 20036, United States.
[8]Department of Plastics Engineering, University of Massachusetts Lowell, Lowell, MA 01854, United States.
[‡]These authors contributed equally.
* corresponding authors:
brandon.knott@nrel.gov
katrina.knauer@nrel.gov

Abstract

Multi-layer film packaging (MLF) revolutionized food preservation by combining diverse material layers to optimize barrier properties, mechanical strength, and shelf-life. These materials are essential for transporting perishables across various climates and allow for access to fresh goods in “food deserts”, but they pose significant recycling challenges due to their structural complexity. This perspective examines key structure-property relationships governing barrier performance and highlights innovations in material design. We explore how machine learning can predict performance metrics and propose recyclable alternatives, integrating data-driven approaches with material science insights. By challenging the status quo of MLF design, we advocate for circularity in food packaging, inspiring innovation at the intersection of sustainability, material science, and artificial intelligence.

Introduction

Multi-layer films (MLFs) have become a cornerstone form factor for a diverse range of consumer products. In 2018, the world produced more than 100 million tonnes of MLFs, anticipated to reach 140 million tonnes by 2025[1]. Food preservation in particular has been revolutionized by MLFs, extending shelf life and substantially reducing global food waste[2,3]. By combining materials with complementary strengths, the multi-layer structure unites individual advantages into a balanced, high-performance film. Such films, often composites of polymers and metalized species, integrate barrier layers for low oxygen and moisture permeation, structural layers for mechanical robustness, and tie layers that serve as the adhesive during lamination. The complex engineering that has yielded such technological significance is ultimately becoming overshadowed by incompatibility with recycling pathways and damaging environmental persistence at end-of-life, both of which demand urgent attention.

A staggering proportion of MLFs, often designed for single-use applications, escape proper waste management systems and accumulate in the environment, contributing to the growing crisis of plastic pollution[4]. Flexible packaging like MLFs, characterized by low bulk density and light weight, is particularly problematic. While advantageous for reducing material use and transportation costs, the low density of these materials makes them easily transported by wind and water, resulting in widespread distribution in marine and terrestrial ecosystems[5]. Estimates suggest that millions of tons of plastic waste enter the environment annually, a significant portion originating from single-use MLFs[6]. This is particularly problematic in developing countries where a majority of daily-use products are sold in single-use MLFs[7].

A key obstacle in mitigating the environmental impact of MLFs lies in the incompatibility with existing recycling systems. Conventional mechanical recycling infrastructure is largely optimized for rigid plastics, such as bottles and containers, which are easier to collect, sort, and process[8]. In contrast, MLFs are difficult to collect and transport due to their low bulk density. Furthermore, the multi-material nature of many MLFs impedes sortation[4,9]. To achieve the performance attributes necessary for packaging applications, MLFs generally feature several different polymers laminated together, including polyethylene (PE), polyvinylidene dichloride (PVDC), polyethylene terephthalate (PET), polyamides (PAs), and ethylene vinyl alcohol (EVOH). These polymers are frequently incompatible when melt blended by mechanical recycling, and the physical separation of discrete layers remains functionally impractical. Chemical processing technologies like selective dissolution and precipitation have demonstrated the effective separation and recovery of discrete polymer components of MLFs[1]. However, these processes frequently yield polymers with diminished thermal properties and remain economically and energetically intensive, therefore limiting adoption at scale[10,11].

Among the many challenges associated with MLF recycling, barrier layers, most notably PVDC and EVOH, remain the most pressing. Halogenated polymers such as PVDC are particularly problematic in recycling processes because they can generate harmful and damaging byproducts at high temperatures, while EVOH can render otherwise recyclable polyolefin mono-layer films unrecyclable. When EVOH is melt-blended with a non-polar polyolefin such as PE during mechanical recycling, the two polymers are inherently immiscible and phase-separate. This phase separation leads to weak interfacial adhesion and, consequently, poor mechanical properties in the recycled material unless an appropriate compatibilizer is introduced to promote interfacial bonding between the two phases. Progress has been made in creating new polymeric materials with comparable barrier properties to incumbent technologies while offering avenues for recyclability and reduced environmental persistence[12,13,14,15,16,17,18,19,20]. Researchers have explored synthetic strategies to tune the density, crystallinity, hydrophobicity, and chemistry of more “sustainable” polymers in pursuit of replacing PVDC and EVOH barrier layers. Designing polymers that deliver layer-specific performance while maintaining the same chemistry (i.e. mono-material design) is also a major area of interest[21]. Yet, there are often trade-offs between recyclability and performance, trade-offs that must not only be carefully managed but strategically overcome if truly circular, high-performance packaging is to be realized.

Computer-aided synthesis planning (CASP), machine learning (ML), and artificial intelligence (AI), offer transformative potential in tackling the challenge of redesigning MLF packaging. These tools can accelerate the discovery of polymers with enhanced barrier properties, mechanical strength, and recyclability and/or biodegradability/compostability built into the design. By leveraging existing datasets of polymer structures and properties, AI can predict optimal formulations and guide material design, reducing reliance on iterative experimentation[22]. Moreover, AI-driven insights can optimize recycling processes, from improving sortation efficiency to enhancing the economic feasibility of chemical recycling methods[23]. Furthermore, emerging ML databases can also narrow down design scopes, for example, to identify alternatives to particularly problematic polymers like PVDC and even focus on biologically derived building blocks allowing for the utilization of biogenic carbon in the production of future packaging materials.

The utilization of MLFs exemplifies the complex trade-offs between functionality and sustainability. While their role in modern food systems is indispensable, the environmental impacts associated with their disposal is unsustainable. Addressing these challenges requires a multifaceted approach, integrating advances in material science, recycling technologies, and waste collection frameworks. By leveraging emerging ML tools like PolyID[24] and fostering a culture of innovation and collaboration, industry can pave the way for a more sustainable future. This perspective explores opportunities to advance MLF sustainability, emphasizing the need for systemic solutions to ensure their environmental and economic viability. We examine the key structure-property relationships that dictate barrier performance in MLF design, analyze the current and future landscape of MLF packaging and recycling, and highlight how PolyID, and other ML tools, can predict and accelerate recyclable-by-design alternatives (**Figure 1**).

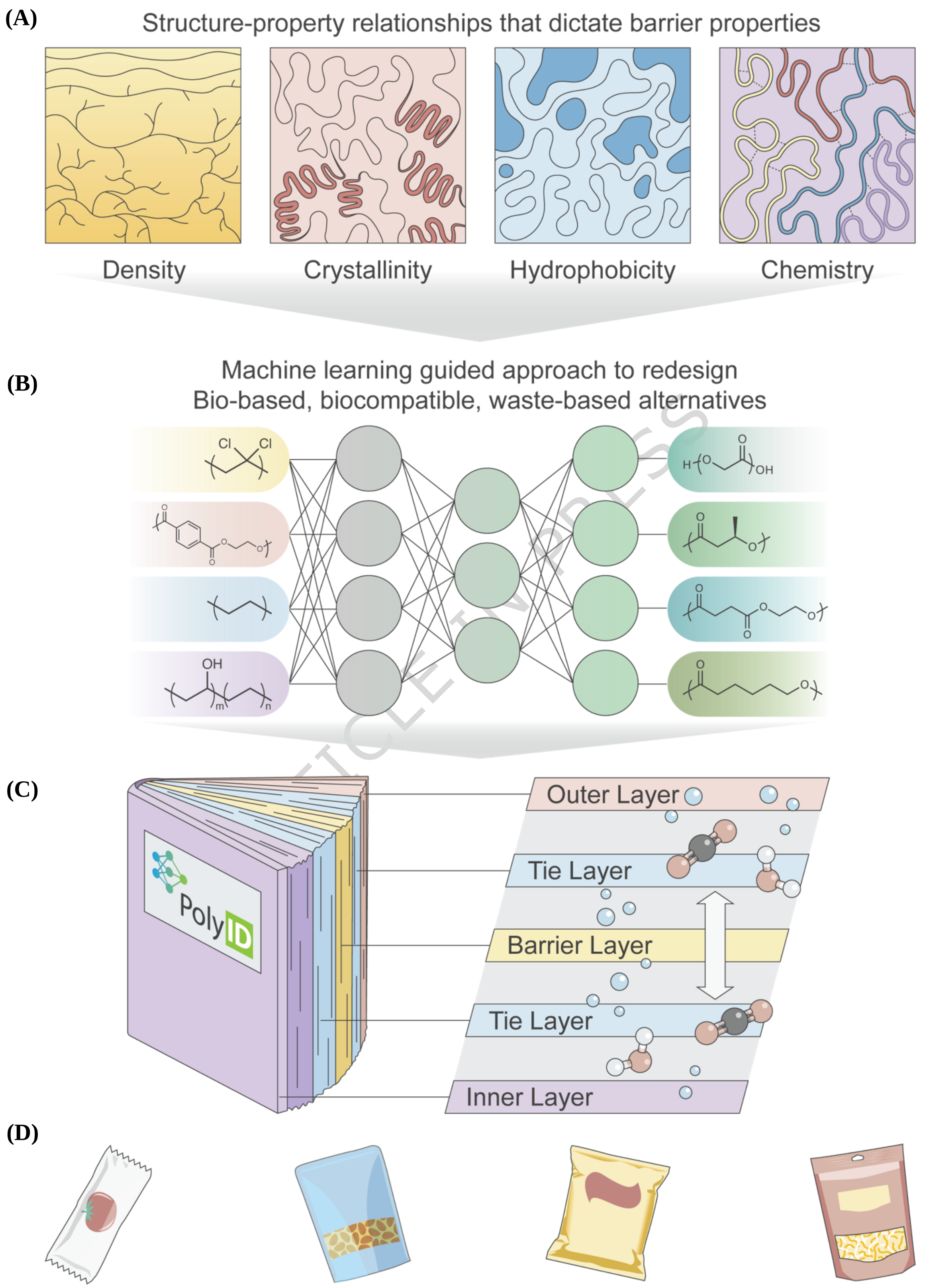

Figure 1. A proposed ML-guided approach to designing recyclable and/or compostable multi-layer films (MLFs) that match the food preservation metrics and target performance of today's conventional materials.

(A) Key structure-property relationships of current MLF polymers are studied; (B) Properties of known polymers are fed into an ML model (PolyID) to predict polymers (in this case study we targeted polyesters) with similar properties; (C) Using the output from PolyID, a new portfolio of polyesters for MLF applications is generated; (D) Examples of the types of MLF packaging targeted in this perspective.

Legacy Multi-Layer Films Packaging

Flexible packaging, like typical MLFs, represent a significant advancement in materials engineering, aimed at addressing the diverse and often competing physical requirements of polymers used in packaging applications. Packaging materials have long needed to balance mechanical integrity, barrier properties, thermal stability, shelf-life, and ease of processing. Flexible packaging materials date back to the early 20th century, with the introduction of cellophane[26], but it's barrier performance was limited, making it insufficient for applications requiring long-term preservation[27]. To address these shortcomings, nitrocellulose lacquer coatings were applied to cellophane, significantly improving its moisture resistance and marking one of the first examples of layered film packaging designed to enhance functionality allowing them to be used in household products[28,29].

Synthetic polymers drastically increased the efficacy of packaging technologies. PE, polypropylene (PP), and PET are widely used in flexible packaging due to their mechanical strength, heat sealability, clarity, and moisture resistance[27,30]. Biaxial orientation[31] of PP (BOPP) and PET (BOPET) greatly improve the tensile strength, puncture resistance, and gas and moisture permeability, enabling thinner films and reduced material usage without sacrificing performance. PVDC, commercialized under the trade name Saran by Dow Chemical[32], is the gold standard for barrier films, achieving extraordinarily low permeability to both oxygen (oxygen permeation coefficient, $PO_2 < 0.005$ Barrer) and water vapor (water vapor permeation coefficient, $PH_2O < 50$ Barrer), enabling prolonged shelf lives and improved food preservation compared to prior flexible packaging[33]. The superior gas and water vapor barrier properties of PVDC arise from a combination of several structural features that limit permeation through the material. The polar nature of the chlorinated backbone results in strong polymer-polymer and polymer-permeant interactions that provide hydrophobicity and minimizes gas solubility within the polymer. Moreover, the crystalline morphology and high density (~1.7 g/cm^3) cooperatively result in low fractional free volume, effectively impeding permeant diffusion[34,35].

Despite the barrier benefits of PVDC, challenges associated with narrow processing windows (difference between melting and degradation temperature), high cost of manufacturing, and hazards posed by its halogenated nature, prompted the development of alternative barrier polymers[36,37,38]. EVOH copolymers are a promising class of extrudable high barrier polymers with exceptionally low oxygen permeation that surpassed even PVDC, owing to the high polymer-permeant interaction of the polar -OH functionality and O_2 gas[39]. However, EVOH exhibits increased sensitivity to moisture due to its hydrophilic nature, resulting in high water vapor absorption, plasticization, and increased oxygen permeability over time. For example, the oxygen permeability of EVOH is approximately 300 times higher when measured at 100% relative humidity compared to 0% relative humidity[34], highlighting the challenges associated with designing materials that possess both good oxygen and water vapor permeability across diverse environmental conditions[40].

Even with the substantial advancements in polymer chemistry and processing, currently there is no single polymer that can deliver all the necessary properties for the evolving demands of packaging applications and growing food scarcity. Thus, MLF packaging, where different polymer layers are combined through extrusion and adhesive lamination to create composite films that strategically balance multiple performance properties in a single film, is needed[41].

Recently, MLF packaging has evolved into highly engineered, complex systems, often comprising ten or more discrete layers[2,25]. These films integrate combinations of distinct barrier and support polymers in conjunction with sealant layers, adhesion-promoting tie layers, organic and inorganic processing additives, and even metalized films. Each layer is optimized to deliver specific performance attributes, such as puncture resistance, aroma retention, light obstruction, and hermetic sealing. **Figure 2** displays the key properties and performance metrics for the most common polymer materials used in MLF layers.

Despite their functional benefits, MLFs present significant challenges for researchers and sustainability efforts. While similar polymers are used across MLFs, the proprietary nature of their formulations often hinder in-depth analysis. Furthermore, the heterogeneous composition of MLFs complicates recycling and end-of-life management. Conventional recycling systems are typically unable to separate and process the multiple, often incompatible, polymers used in these films, frequently resulting in landfilling, incineration, or loss to the environment.

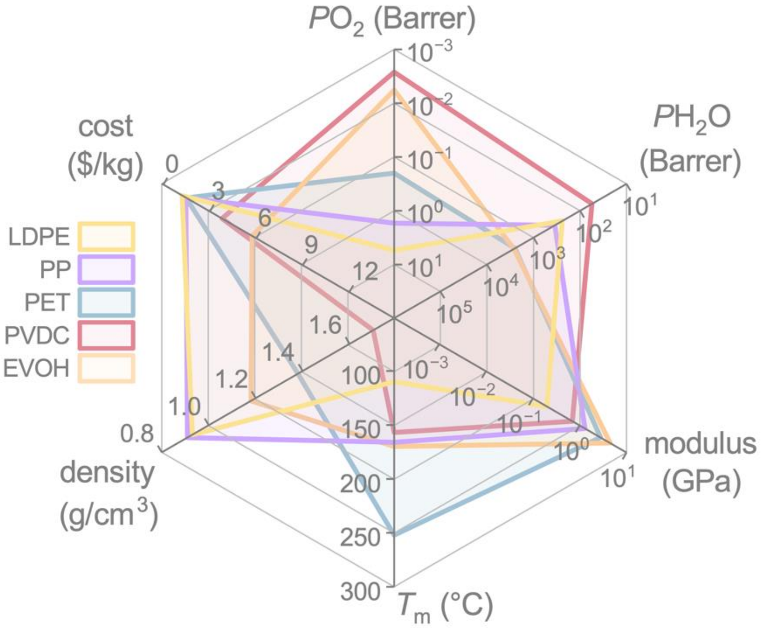

Figure 2. Property comparison of conventional polymers used in MLFs. Summary of reported oxygen barrier (presented as oxygen permeability coefficient, PO_2), water vapor barrier (presented as water vapor permeability coefficient, PH_2O), melting temperature (T_m), modulus, density, and average cost (based on 2024 market reports) for incumbent polymer materials commonly used in multi-layer film layers. Barrier measurements and modulus are presented on logarithmic scales. Cost, density, and barrier measurements are presented on inverted scales. Barrer is equivalent to $10^{-10}((cm_{STP}^3*cm)/(cm^2*s*cmHg))$. LDPE: low-density polyethylene, PP: polypropylene, PET: polyethylene terephthalate, PVDC: polyvinylidene dichloride, EVOH: ethylene vinyl alcohol.

Current End-of-Life Approaches for Multilayer Films

Efforts to address the end-of-life challenges of MLFs have led to the exploration of several recycling strategies, including mechanical recycling, pyrolysis, and dissolution[42]. Each of these approaches offers distinct advantages and limitations when applied to existing MLF packaging. Our primary focus in this section is on the opportunities and challenges associated with specific recycling technologies. We first include a brief discussion of some advancements in waste management of MLFs.

Many recent advances in the management of plastic waste (e.g. sorting, marking, or identification) utilize AI and ML coupled with imaging and spectroscopy approaches for rapid, nondestructive characterization[43], including in detection of microplastics[44]. MLFs have two main challenges in identification: weak signal reflection due to thin profiles, and transparency, where the film and underlying materials both reflect into the sensor. Koinig et. al., demonstrated successfully a method to classify mono- from multi-layer films inline using near-infrared spectroscopy in transflectance mode[45]. However, automated separation of different types of multi-layer films remains an open challenge.

Once materials have been collected and sorted, the next major hurdle is recycling. Mechanical recycling is the most established method and involves the collection, sorting, cleaning, and remelting (via extrusion) of plastic waste into recycled materials. While this process works well for single-material streams, it is impeded by MLFs composed of incompatible polymers (e.g. PE and PET) or structures containing barrier layers like aluminum, PVDC, and EVOH. The incompatibility between layers can result in poor melt quality, discoloration, and degraded mechanical properties in the recycled material. Efforts have been made to compatibilize immiscible polymers found in MLFs, thereby mitigating property degradation. The most common is an addition of a reactive polymer that can graft to both phases, reinforcing interfaces to prevent stress concentration[46,47,48,49]. However, these additives simultaneously reduce aesthetics, and the process is not universal for the range of compositions that may be found in mixed MLF waste[50].

Pyrolysis thermally decomposes plastic waste in the absence of oxygen, breaking it down into oils, gases, waxes, and char[51]. This process can recover valuable hydrocarbons and generate virgin-like naphtha feedstocks for new polymer production. However, pyrolysis typically requires high energy input and elevated operating temperatures, which can lead to significant emissions and generate byproducts that require additional treatment. While often marketed as a solution for mixed plastic waste, pyrolysis is largely limited to polyolefins such as PE, PP, or polystyrene (PS); polymers containing heteroatoms such as PET, EVOH, and PAs (i.e. nylons) are generally incompatible, depending on the level at which they are present. Halogenated polymers like PVDC also pose challenges, producing corrosive and toxic chlorinated byproducts that necessitate complex gas-cleaning systems.

Solvent-targeted recovery and precipitation (STRAP) is an emerging approach that selectively dissolves and separates individual polymer layers using tailored solvents[1]. STRAP offers the potential to recover high-purity polymers from MLFs, enabling true material circularity and preserving the quality of the separated components. Unlike pyrolysis, STRAP operates at lower temperatures and avoids bond breaking, instead focusing on the physical separation of intact polymer chains. However, the process depends on the careful selection of solvents, which can be costly, and may raise concerns regarding solvent recovery, toxicity, and energy use for solvent evaporation and recycling. Furthermore, highly complex multilayer structures, especially those containing insoluble crosslinked or highly engineered tie layers, can still pose challenges for efficient separation.

Collectively, these end-of-life strategies (summarized in **Figure 3**) represent important steps towards MLF packaging waste. However, each faces significant technical and economic challenges when applied to today's highly engineered multi-layer structures. Advancing packaging design through design-for-recyclability and the development of mono-material MLFs will be critical to improving compatibility with recycling systems and enabling a more circular packaging economy. A key research focus is the identification of non-halogenated barrier layers capable of replacing PVDC while maintaining essential oxygen and moisture barrier performance. These emerging technologies offer the potential to fundamentally reshape the future of MLF packaging by balancing high performance with environmental sustainability.

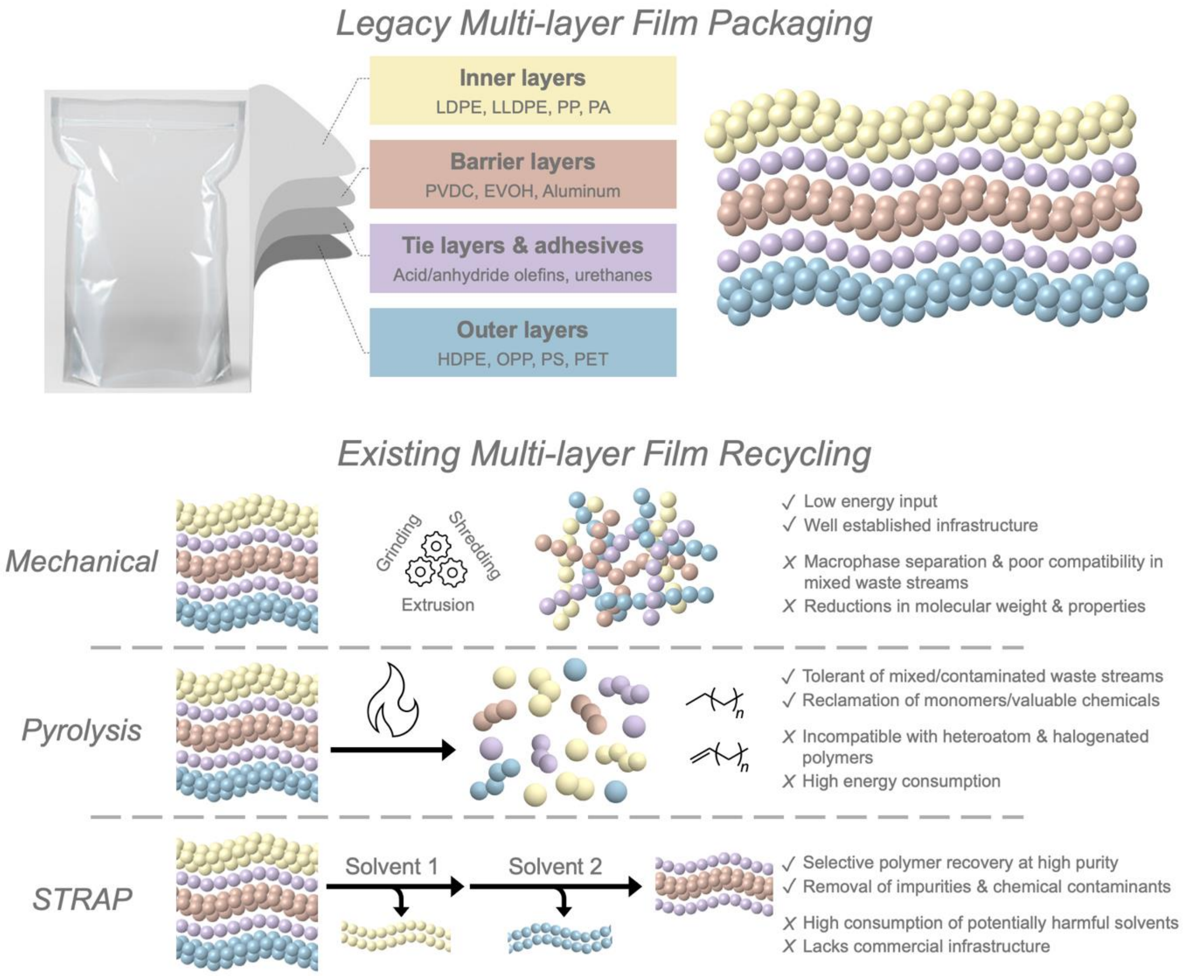

Figure 3. **Generalized legacy MLF structure c and current end-of-life recycling strategies.** Legacy MLF packaging configurations and existing MLF recycling strategies including mechanical recycling, pyrolysis, and STRAP. LDPE: low-density polyethylene, LLDPE: linear low-density polyethylene, PP: polypropylene, PA: polyamide, PVDC: polyvinylidene dichloride, EVOH: ethylene vinyl alcohol, HDPE: high-density polyethylene, OPP: oriented polypropylene, PS: polystyrene, PET: polyethylene terephthalate.

Future Multi-Layer Film Packaging

As researchers advance robust recycling platforms capable of handling complex, heterogeneous plastic waste streams, parallel efforts are focusing on the development of more sustainable and recyclable packaging designs. When considering redesigning MLFs, the primary challenge is meeting the exceptional barrier performance provided by aluminum, PVDC, and EVOH (respectively and in combination). Here, we will highlight efforts made towards designs for mechanical recycling, chemical recycling, and composting end-of-life systems.

For mechanical recycling, efforts have focused on lowering material variability to a single polymer or compatible polymers. A theoretical basis for redesign choices is that selected polymers show superior compatibility with other polyolefins[52]. In previous work, recyclability has been improved using a mono-material approach primarily through polyolefins[53]. One notable example is an all-PE film manufactured via biaxial orientation (BOPE), which can achieve higher crystallinity and reduced polymer chain mobility. This results in enhanced mechanical properties at thinner gauges and offers moderate improvements in oxygen barrier performance[53]. Another promising design-for-recycling strategy involves the incorporation of specialized tie or adhesive layers that can be selectively activated by chemical or thermal triggers to release at end-of-life, enabling separation and recovery of the individual layers for mechanical recycling[54].

Solvolysis-based chemical recycling targets depolymerization in polymer backbones containing heteroatoms (e.g. ester bonds in polyester) and can produce near quantitative yields of parent monomers at relatively mild conditions when compared to pyrolysis. The high carbon efficiency of solvolysis has prompted discussions on whether a transition to an “all-polyester” approach to redesigning complex plastic materials could improve recycling rates and carbon circularity[57]. However, it remains challenging to develop heteroatom-backbone polymers that offer strong barrier performance against both water vapor and gases. As a result, achieving the desired barrier performance often requires combining multiple polymers within a single structure, an approach that inherently increases MLF complexity and hinders recyclability.

Currently, there is no single polymer that has all the properties necessary for typical food packaging applications, necessitating multi-layered architectures. As we consider redesign, recent life cycle assessments (LCAs) and planetary boundary analyses have made it clear that improving recycling rates alone will not bring the plastics supply chain within a sustainable operating space[55]. To meet climate and sustainability goals, we must simultaneously increase recycling rates and shift towards materials derived from biogenic carbon. Select polyolefin producers are investing in bioethanol to produce bio-PE. However, the recycling challenges remain. Commercially available bio-based polyesters such as polylactic acid (PLA) and polyhydroxyalkanoates (PHAs) are currently marketed as compostable and/or biodegradable but are typically rejected from mechanical recycling streams. However, recently reported tecno-economic assessment (TEA) and LCA studies on mixed polyester recycling reveal the economic and environmental benefits of transitioning plastics to an all-polyester based platform coupled with chemical recycling[56,57]. These studies suggest that multiple polyesters can be combined in a multi-layer structure while still being recyclable by solvolysis-based platforms.

Polyesters are historically not known for their barrier properties. Thus, redesign efforts in polyester chemistry for packaging materials have focused on improving the barrier performance against moisture and gases[12,15,16,58]. In the pursuit of new high-barrier polyesters, it is important to understand the chemical and physical factors that govern gas permeation. In bulk polymer films, several key characteristics including morphology, chain mobility, polarity, and intermolecular interactions contribute to mass transport through the material[59,60]. Permeation of non-interacting gases (i.e. O_2, CO_2, N_2, etc.) occurs through a combination of solution-diffusion mechanisms, where the permeant initially dissolves into the polymer, before diffusing to the opposing interface[61]. Polar and apolar functional groups can attract or repel permeants, which alter the solubility of the gas within the polymer. The intermolecular forces that determine polymer-polymer and polymer-permeant interactions can be captured in the cohesive energy density, which has proven to be an effective method for correlating barrier performance with polymer structure[59].

The diffusion of gases through films is primarily governed by the polymer's fractional free volume[62]. The free volume, or the interstitial space between polymer chains, is influenced by physical parameters such as glass transition, crystallinity, and processing-induced chain alignment. In semi-crystalline polymers, crystallites function as impermeable domains, forcing permeants into tortuous pathways through amorphous regions that imped diffusion[60]. In contrast, fully amorphous polymers typically exhibit high gas permeability due to larger free volume, making them less suitable as barrier layers in MLFs.

Unlike non-interacting gases like O_2 and CO_2, water vapor permeation is comparatively more complex due to its small kinetic diameter and strong hydrogen-bonding capacity[34]. Hydrophilic polymers with high hydrogen-bonding capabilities result in high rates of water vapor sorption, resulting in high permeability. Permeation is further complicated as polymers can often be swelled and plasticized upon the solubilization of water molecules, resulting in increased free volume, and non-linear diffusion kinetics upon exposure to varying humidity[40]. Interactions between water molecules and the polymer substrate can therefore change polymer physical characteristics and alter the permeation of other gases, a phenomenon observed in current EVOH barrier materials[39].

Following these guidelines, it is intuitive that polymers with high self-association (high cohesive energy density), low free volume, and high crystallinity will possess the best barrier resistance to permeation[59]. The polar nature and moderate crystallinity of current commercial polyesters such as PET, PLA, or polybutylene succinate (PBS) results in reduced oxygen permeation compared to polyolefins, but increased water vapor permeation due to reduced hydrophobicity[63]. Surprisingly, polyglycolic acid (PGA), the simplest linear aliphatic polyester, exhibits exceptional water and oxygen barrier properties ($PO_2 < 0.005$ Barrer, $PH_2O < 150$ Barrer) that rival PVDC, despite being a non-halogenated biodegradable polymer[64,65]. The barrier performance of PGA stems from to its high crystallinity and high concentration of permeant-interacting ester groups; however, the high crystallinity of PGA yields a highly brittle polymer and an elevated melting temperature that poses challenges for MLF processing and application[66,67].

It is therefore critical to identify new, redesigned polyesters that achieve balanced barrier properties, mechanical performance, and manufacturability to realize an all-polyester MLF packaging platform. A recent example from Chen and coworkers disclosed a series of spiro-valerolactone-based polyesters that yielded reduced water vapor permeation compared to low-density polyethylene (LDPE; $PH_2O < 200$ Barrer) and oxygen permeation lower than PET ($PO_2 < 0.05$ Barrer)[58]. These materials also had tunable ductility and excellent adhesive properties, making them promising candidates for mono-material design. Other advancements in sustainable barrier materials have typically employed sugar-based polymers like polysaccharides, nanocellulose, cellulose, and chitin and also include the use of dynamic chemistries while still enabling compostable packaging material[68,69,70,71,72].

As our modeling efforts are primarily based around polyesters this perspective looks at developing new polyester-based chemistry with ML. As mentioned, there are a multitude of other methods to create sustainable packaging that would benefit from the usage of ML. The computational methods discussed (vide infra) can, and should, be extended to modeling these other material types and we encourage the use of this platform to explore varying chemistries.

Computationally-Driven Discovery of Polymers

A comprehensive pursuit of MLFs with improved end-of-life outcomes must include consideration of a wide range of polymer building blocks, particularly those derivable from natural and biological sources. Given the vast design space for bio-derivable building blocks, high-fidelity computational screening procedures are essential to narrow the possibilities to the most promising formulations for synthesis and characterization. AI is a broad field of computer science focused on creating systems that can perform tasks typically requiring human intelligence, such as reasoning, problem solving, and decision-making. ML is a subfield of AI that uses data and algorithms to learn patterns and relationships, enabling systems to improve their performance on tasks over time without being explicitly programmed. In the context of materials and specifically polymers discovery, AI/ML can be coupled with databases to enable property predictions and even de novo design of materials. New approaches to map molecular features to polymer performance are being pursued via the development and application of increasingly advanced high-throughput ML tools for polymer property prediction[73,74,75,76,77,78,79]. A very comprehensive introduction to data-driven approaches for polymer design was reported by McDonald *et. al.*[80], and Tran *et. al.*[81].

ML approaches broadly fall into two categories: inverse design and forward design. Inverse design first specifies desired properties and then predicts chemistries likely to have those properties, e.g. via generative ML models. Using generative models thus broadens the discovery space beyond existing materials and chemical motifs and has produced some promising results for a variety of properties and applications[82,83,84,85,86,87]. On the other hand, these approaches can also generate polymeric materials wherein the building blocks are difficult to synthesize and/or polymerize. In general, molecular simulations have been deployed as a part of inverse design approaches for data generation and model validation, but experimental synthesis and characterization to validate polymer chemistries and properties has been limited. Conversely, forward design approaches start from a list of potential molecules (monomers and/or their polymer formulations) and aim to identify which are the most promising for a target application based on predicted polymer properties. Examples of forward design approaches include RadonPy[88], PolyGNN[89], and PolyID[24]. PolyID is a multioutput, message passing neural network (MPNN) designed to facilitate quantitative structure–property relationship (QSPR) analysis for polymers[24]. PolyID takes as input a representation of a polymer chain by performing *in silico* polymerization, with degree of polymerization 18, of monomers specified using the simplified molecular-input line-entry system

(SMILES) strings to encode chemical structures. This allows for specification of comonomer ratio (if applicable) and random regio-orientations. Atom and bond feature vectors are built from the molecular structure using basic descriptors from the RDKit Python package. These vectors are then updated during message passing, where vectors of neighboring atoms and bonds are concatenated and passed through two feed forward layers, reducing the vector size to the original length. Finally, the bond vectors are averaged, and passed through separate feed forward layers, one for each target property. The PolyID MPNN, originally trained on ~1,800 experimental data points mined from literature and existing polymer databases, predicts polymer performance attributes, including thermal (glass transition and melting temperatures), density, mechanical (elastic modulus, tensile strength, and elongation at break), and barrier properties (permeability of oxygen, nitrogen, carbon dioxide, and water vapor). Typical accuracies, measured using the mean absolute error (MAE) from 10-fold cross validation, fall around 20 °C for T_g, 25-30 °C for T_m, 0.05 g/cm^3 for density, $10^{0.4}$ (2.5) Barrer for permeabilities, and approximately 300 MPa for elastic modulus.

Certain features that are essential to the design of circular MLFs are particularly ill-captured by current computational approaches, including water vapor permeability and end-of-life outcomes. Regarding the former, there is currently no publicly available database of water vapor permeability for common polymers. In addition, the interactions of water vapor with polymer films tend to differ widely from that of other gases due to the formation of hydrogen bonds, often leading to high solubility and subsequent swelling in polymers[34]. Temperature, relative humidity, polymer density, crystallinity, and measurement approach can also have a large effect on the measured and reported permeability. These factors and the resulting scarcity of accessible and standardized data can make modeling films with ML challenging and resulting predictions unreliable. Augmentation of experimental datasets with molecular modeling outputs for training ML models has been demonstrated[88], including for gas barrier properties[90] but not yet for water vapor permeability. As far as we are aware, currently only PolyID includes water vapor permeability as a prediction target. In the original PolyID publication, just 27 data points were included for water vapor permeability to train PolyID, all of which came from non-public databases. We have recently expanded that number to 55 by including more publicly available literature data and in-house experimental measurements (reducing the mean absolute error from $10^{2.6}$ to $10^{0.6}$ Barrer for $P\mathrm{H_2O}$ predictions).

The ability to design polymers with targeted properties for MLFs primarily depends on 1) the pool of candidate materials, 2) the ability to make accurate predictions, and 3) the synthesizability of the monomers and polymers. As discussed above, inverse design approaches can expand the possibilities for molecular generation beyond known materials. A more direct approach to expand the candidate pool is to apply chemical transformations to source molecules to create new monomers, somewhat analogically to the experimental chemist's approach. Applying this set of chemical reactions to the initial pool would expand the material discovery space while leaving a direct recipe for creating desired monomers. DORAnet (previously Pickaxe)[91], an open-source python package, chemically transforms an initial set of molecules according to well-defined reaction rules and is well-suited for such a purpose. DORAnet has two main types of reactions: 1) *synthetic*, which transforms molecules according to a 1,224 set of industrially utilized chemical reactions, and 2) *enzymatic*, which employs 22,803 reactions accessible by biological enzymes. DORAnet also has built-in tools to filter out reactions or molecules that do not meet certain criteria, including filters based on monomer attributes such as halogen moieties or high molecular weight, reaction thermodynamics (e.g. using the tool eQuilibrator), and monomer properties critical for polymerization such as boiling point or solubility. Integrating a monomer generation scheme (e.g. DORAnet) with a polymer property prediction tool (e.g. PolyID) allows for strategic expansion of the initial materials discovery space, feeding realistic possibilities to ML models to target the most promising candidate materials for synthesis.

To demonstrate the possibilities of such an integrated computational workflow for polymer discovery for MLF applications, we apply DORAnet and PolyID to a large pool of bio-derivable and purchasable monomers from KEGG and Sigma data sources, respectively. In this example, we target homopolyesters (ring opened structures) and diol + diacid polyesters, as they have an increased likelihood of biodegradability and recyclability via hydrolysis[92]. KEGG contains 290 diols, 115 diacids, and 146 multi-functional monomers which containing both an alcohol and an acid group while Sigma contains 537, 166, and 285 of the same groups, respectively, with 84 diols and 40 diacids overlapping between the two. These monomers can be combined into 82,612 polyesters. We sought to restrict the polyester combinations to those for which PolyID is most likely to make accurate predictions. Thus, we applied a domain of validity filter (DoV) of 10, wherein polymers that contain 10 or more chemical substructures not seen in training are removed, reducing the number of polyesters to 21,779. To cast as wide a net as possible, we also applied all DORAnet synthetic and enzymatic reactions to the molecules in KEGG and Sigma, resulting in 14,703 diols and 9,851 diacids with a molecular weight less than 250 g/mol, which can be combined to form over 144 million polyesters. Targeting polycondensation reactions, we exclude diols with a boiling point over 250 °C as excess diol needs to be distilled off during polymerization, with diacid as a limiting reagent. Thus, we applied a recently developed MPNN-based tool, Chemperium[93], to predict the boiling point of each diol, and filtered those over 250 °C, leaving 1,791 diols. This reduced the number of polyester combinations to 17.6 million. A DoV filter of 10 brought the total to 2.3 million polyesters. A graphical representation of this example ML workflow to design an all-polyester MLF is presented in **Figure 4**.

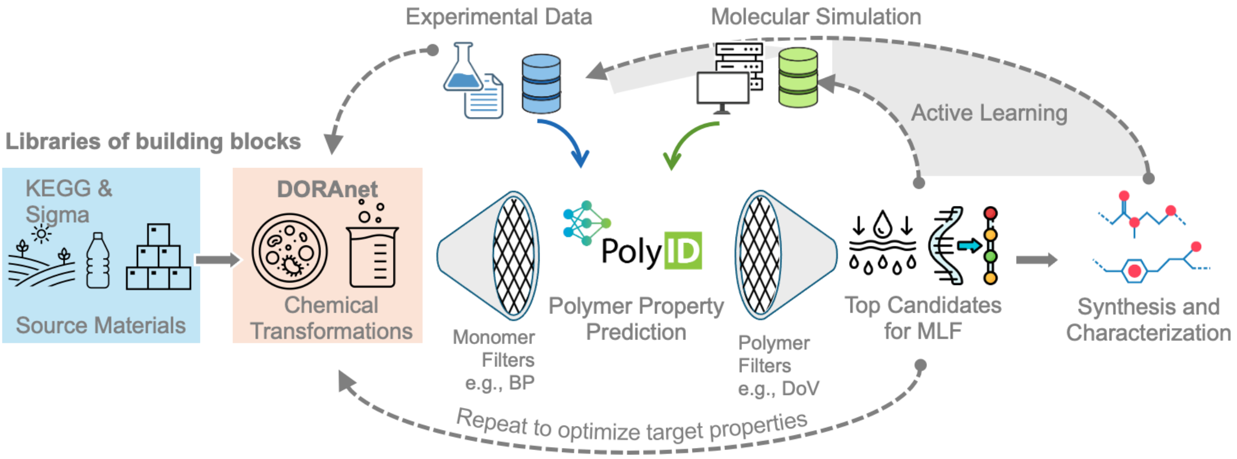

Figure 4. Integrated PolyID-DORAnet workflow for polymer discovery and property prediction. DORAnet applies *in situ* chemical reactions to potential source materials to naturally expand the monomer discover space. PolyID, trained on experimental and/or molecular simulation data, is then used to identify candidate polymer formulations with desired properties. While this work does not perform molecular simulations, active learning, or repeated DORAnet-PolyID loops to optimize target properties, we include those steps in the figure to illustrate a holistic approach to continually improve the model predictions and target candidates. BP: boiling point, DoV: domain of validity, MLF: multi-layer film.

The PolyID pipeline, including *in silico* polymerization and prediction of ten properties, was applied to each of the candidate polyesters from before and after applying DORAnet. We focus on the results with DORAnet-generated monomers (**Figure 5**), with pre-DORAnet results presented in the supplementary information (**Figure S1**). **Figure 5** shows an overview of the predicted properties, highlighting relationships between barrier, density, and elastic modulus (with an expanded dataset in **Figure S2**). In the context of polyesters for MLF applications, each layer requires different property requirements (as shown by the incumbent materials in **Figure 2**), including layer-specific melting temperature for heat sealing, adhesive characteristics, and various mechanical properties. Our preliminary focus is on the most important properties for MLFs for food packaging, namely oxygen and water vapor permeability, assuming application-relevant thermal and mechanical properties. We note that barrier properties can also vary greatly depending on the use temperature, relative humidity, and polymer crystallinity, e.g. enhanced barrier performance is often noted at temperatures below the polymer's T_g, which are not directly modeled by PolyID. The ability to integrate crystallinity into PolyID, either by including it as an input to the model, updating the model architecture (see below), or as a prediction target based on processing conditions, would likely greatly improve prediction accuracy, although significantly expanded datasets are needed.

Navigating the large, multidimensional space of the property predictions can be challenging, and thus we created interactive versions of **Figure 5** to enable real-time exploration of the property space. Using these interactive plots, we highlight a few examples in **Figure 5** of polyester systems with under-explored structural features that may be well suited for MLFs, including those with metabolites, naphthalenes, phosphates, as well as a polyester with a strong monomer candidate for bio-production at scale, 2-pyrone-4,6-dicarboxylic acid (PDC)[94]. **Figure S3** presents all the predicted properties for select polyesters from PolyID.

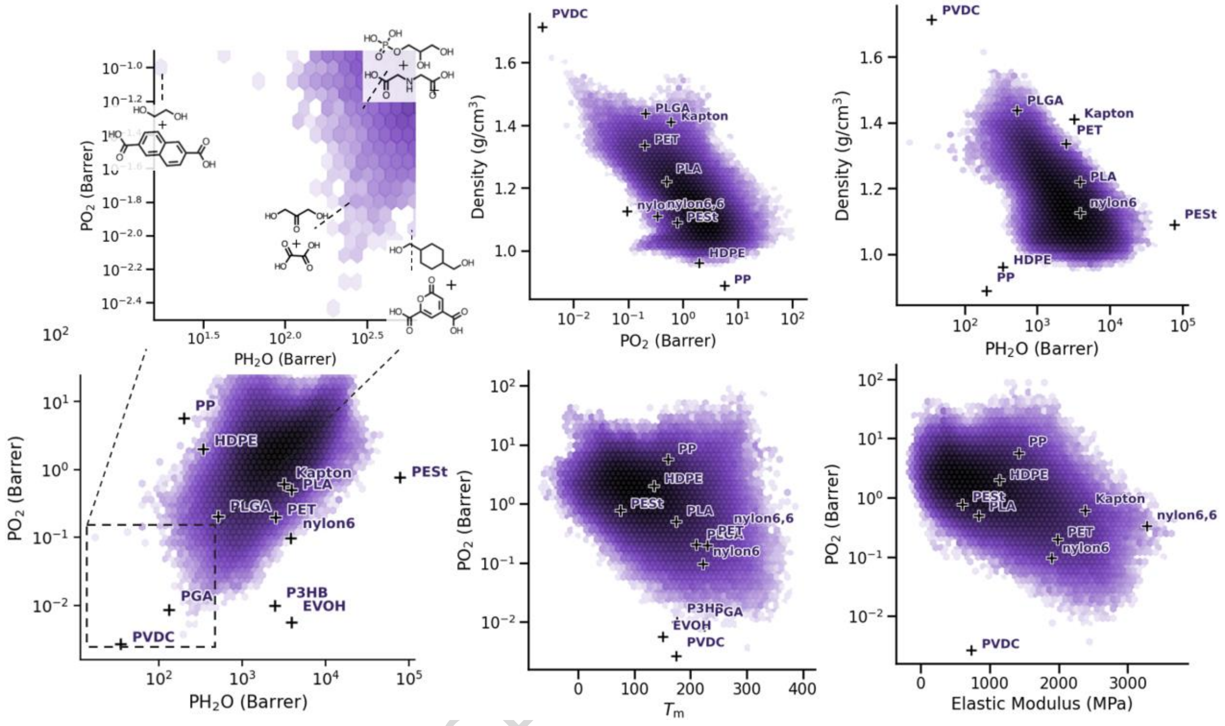

Figure 5. PolyID-generated predictions of polymer properties for polyesters produced from monomers derived from KEGG and Sigma databases and one round of DORAnet transformations. See **Figure S2** for more predicted property comparisons. Highlighted monomers are examples of under-explored chemistries for MLFs with relatively strong predicted barrier properties. PVDC: polyvinylidene dichloride, PLGA: poly(lactic-co-glycolic acid), PET: polyethylene terephthalate, PLA: polylactic acid, HDPE: high-density polyethylene, PP: polypropylene, P3HB: poly(3-hydroxybutyrate), EVOH: ethylene vinyl alcohol, PESt: poly(ethylene stearate).

In **Figure 6** we highlight the key properties of select commercially available bio-based polyesters as well as a new polyester predicted by PolyID (PolyID P1). While several of the polyesters are predicted to achieve relatively robust oxygen or water vapor barrier performance, none of the predictions reach the combined barrier performance of PVDC. It may be difficult for polyesters, and other heteroatom containing polymers, to reach the barrier performance of the heavily halogenated carbon backbone of PVDC that produces a tightly packed, highly crystalline, and hydrophobic polymer film. While PolyID does not explicitly include crystallinity information in the model training, optimizing these polyesters for increased crystallinity could lead to improved barrier performance. Another important consideration is compliance with food packaging regulations. PolyID, like most ML tools for polymer design, does not predict toxicity or health and safety concerns for specific molecules. Therefore, these analyses should also be performed on predicted molecules using tools such as the Environmental Protection Agency's (EPA) Toxicity

Estimation Software Tool (TEST) before down-selecting target polymers for food-contact packaging[95].

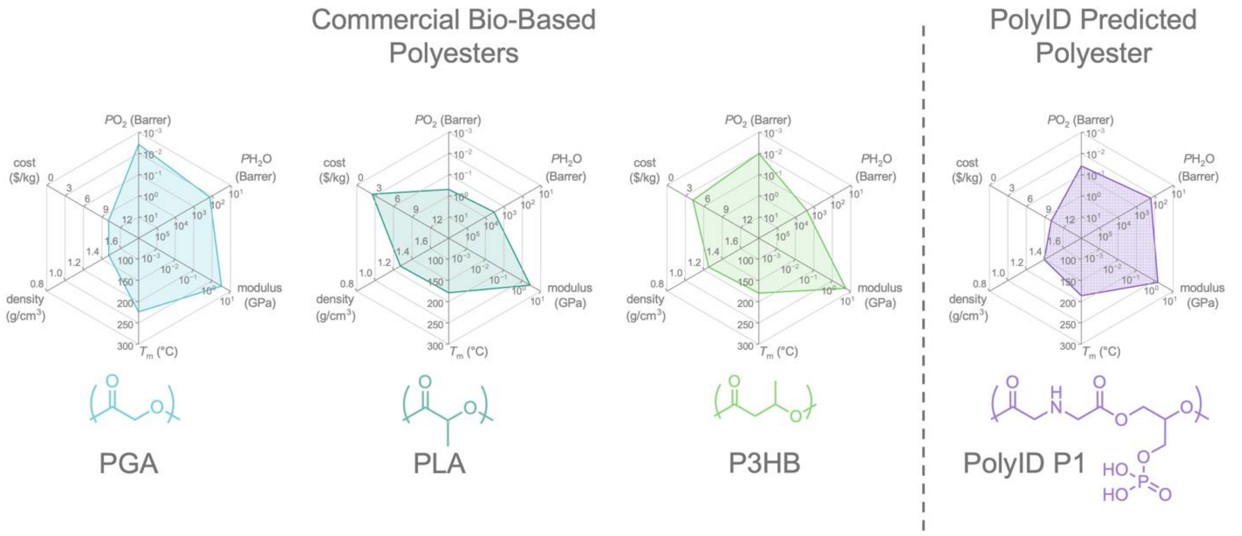

Figure 6. Property comparison of emerging biopolymers and predicted polyester structure for potential use in future MLFs. Summary of oxygen barrier (presented as oxygen permeability coefficient, PO_2), water vapor barrier (presented as water vapor permeability coefficient, PH_2O), melting temperature (T_m), modulus, density, and average cost (based on 2024 market reports for polyglycolide (PGA), polylactic acid (PLA), and poly(3-hydroxybutyrate) (P3HB) for existing and a new polyester predicted by PolyID (PolyID P1). Barrier measurements and modulus are presented on logarithmic scales. Cost, density, and barrier measurements are presented on inverted scales.

Machine Learning-Guided Polymer Processing

While ML and AI-based tools can effectively predict polymers with target properties, polymer processing itself plays a critical role in determining the final film performance. MLFs are typically produced using one of three primary methods: co-extrusion, lamination, or coating. The co-extrusion process (illustrated in **Figure 7**) involves melting two or more polymeric materials via extrusion, joining them together while in the molten state, and then cooling the resulting multilayer structure[96]. Common co-extrusion techniques include flat die (cast) extrusion, blown film extrusion, and sheet extrusion. In lamination, pre-formed layers, often produced by extrusion or coating, are bonded using either adhesives or heat[97]. These laminates may include a variety of materials such as polymeric films, metallic foils, and paper, each selected for specific barrier or mechanical properties. Lamination methods include adhesive lamination, extrusion lamination, hot melt lamination, and wax lamination[2]. The coating method, while like extrusion lamination, differs in that it does not involve a secondary substrate or web. Instead, a functional coating is directly applied onto a base film, resulting in a two-layer structure. Common coating techniques include aqueous dispersion, solvent-based, vacuum, and hot melt coating.

Barrier properties, mechanical strength, aesthetic appeal, and cost-effectiveness of the final MLF all depend on the type of processing applied. The method is selected depending on the desired specific properties and appearance of the packaging structure. Predicting the processability of polymeric systems and how processing parameters influence material properties is an emerging and impactful application for ML and AI. Researchers can contribute by updating databases on polymer properties to include viscoelastic behavior, the effects of processing conditions on crystallinity, and critical parameters for producing high-barrier films, such as residence time, processing temperature, crystallinity, and orientation. Sharing this data openly can strengthen ML predictions for more processable materials.

Another challenge when attempting to prototype and validate new polymer systems in applications such as MLFs if that polymer processing trials typically require tens of kilograms of material, much of which is discarded as waste during process optimization. This presents another key opportunity for ML and AI integration. Embedding in-line characterization tools within polymer processing workflows and feeding real-time data back into ML algorithms could enable better prediction of optimal processing windows for new polymers, reducing experimental iteration and material waste (**Figure 7**). However, in-line characterization tools remain limited. Today, they can reliably measure only a few properties such as melt flow, which provides insight into polymer degradation and thermal stability during processing, and infrared signatures, which indicate potential chemical changes. As a result, most critical film characterization still occurs manually after processing using separate, stand-alone analytical tools. While this approach remains time limited, it is still possible to integrate these characterization tools to ML. For example, Python codes could be designed to automatically collect data from key polymer characterization tools such as differential scanning calorimetry (DSC), dynamic mechanical analysis (DMA), rheometers, and thermogravimetric analyzers (TGAs) to feed back into ML algorithms[98].

Within the materials community, autonomous experimentation platforms, referred to as self-driving laboratories, are being developed to perform experiments with little to no intervention from human scientists by leveraging active learning algorithms. These automated and autonomous experiments promise to help scientists discover materials with optimized properties more quickly, map phase spaces more accurately, and use less material in the pursuit of these goals. Several in-line analysis tools are already available, including spectroscopic, rheologic, and process state measurements[99]. Furthermore, automated systems can naturally integrate databases and materials ML platforms as the metadata for each sample is likely already digitized as part of the preparation process. One example of this approach was outlined by Wang *et al.* where they presented an AI driven automated material laboratory (Polybot) designed to autonomously explore processing pathways for achieving high-conductivity, low-defect electronic polymers films[100]. Polybot demonstrates a successful autonomous experimental campaign and designed recipes for scaled-up fabrication of transparent conductive thin films with target conductivity. A similar Polybot approach could be applied to processing trials for new MLF materials, adapting in-line measurement tools to align with polymer properties specific to MLFs. The challenge is not merely to build smarter tools, but to create smarter systems that amplify human insight rather than replace it and that push the frontiers of knowledge forward while remaining firmly grounded in the values that define scientific progress.

Several barriers must be addressed before research laboratories can fully leverage automated experimental platforms. Bringing all relevant tools and instrumentation online and making them controllable through software is often nontrivial, particularly for older equipment that lacks modern interfaces. In addition, robust safety protocols must be developed to protect both valuable instruments and personnel operating near robotic systems. Because robotics, sensors, and supporting infrastructure require substantial financial investment, automation platforms must be sufficiently flexible to accommodate variation in workflows, sample types, and operating conditions. Large language models (LLMs) such as ChatGPT can help lower these integration barriers by translating well-structured documentation of instrument capabilities and control interfaces into executable scripts, command sequences, and workflow logic, thereby streamlining the process of bringing heterogeneous laboratory instruments under unified automated control[101].

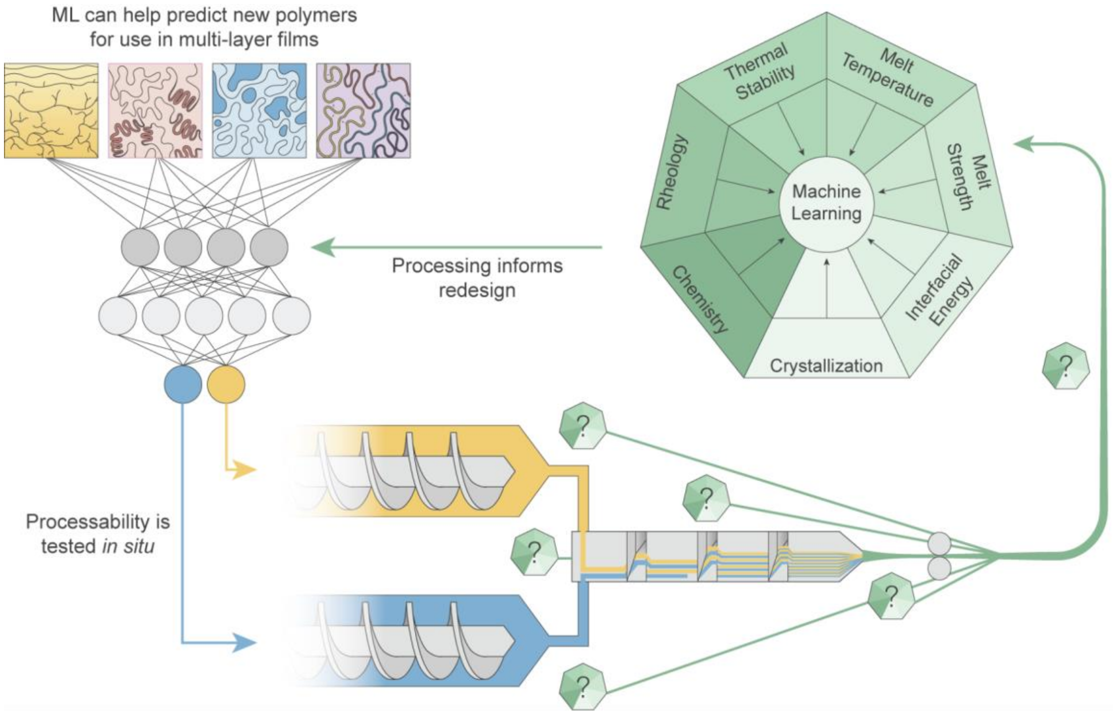

Figure 7. Self-driving laboratories. Proposed schematic for combining in-line characterization to multi-layer film processing coupled with ML to better inform redesign efforts, reduce iteration cycles, and accelerate scale-up/commercialization.

Conclusion and Future Outlook

The redesign of MLFs encompasses a multi-objective optimization problem for polymer performance (barrier, thermal, and mechanical), processability requirements, as well as environmental and economic viability. Going forward, there is significant opportunity for computational tools to guide the development of next-generation MLFs, but several challenges must be addressed. The relative dearth of publicly available data that links polymer structure to performance attributes, particularly gas and vapor barrier properties, is a significant hindrance to model prediction accuracy. In addition, the lack of standardization for reporting permeability and transmission rates (e.g. units, normalization by film thickness, polymer processing conditions, and explicitly reported experimental conditions such as relative humidity and temperature) adds another layer of complexity. The success of modern deep learning tools for protein structure prediction[102] and protein design[103,104] have been enabled by the existence and curation of the open-source Protein Data Bank[105]. Polymers certainly present unique challenges, yet the development of a central repository for polymer properties, within a polymer-appropriate data model (e.g. the Community Resource for Innovation in Polymer Technology, CRIPT[106]), could similarly facilitate innovation in the application of AI/ML tools to solve polymer design challenges. Another promising avenue could be high-throughput data generation via molecular modeling approaches through density functional theory (DFT), molecular dynamics (MD), and COSMO-RS, producing data that can then be utilized to train predictive neural networks[90,107].

Alternative approaches to getting more out of less data can also be done at the ML stage with techniques such as transfer learning wherein abundant data collected on more easily measured/estimated properties can pre-train ML models to be further refined with less abundant, experimentally-measured properties[108].

Current ML models for polymers primarily focus on the atomistic-level and struggle to capture and design for higher-order effects at the polymer chain level such as crystallization, molecular weight, level of entanglement, viscosity, and in the case of thermosets such as covalent adaptable networks, features such as cross-link density and distance between cross-links. Challenges in designing for these topological features are due partially to the scarcity of high-fidelity data for these properties, and partially to the limitations of current model architectures. Hierarchical[109] or topological neural networks[110] for example, that incorporate interactions across multiple scales, would likely have more success in capturing and designing for effects from atomistic- and chain-level differences. In addition, correlation-based ML models (as utilized in this work) inherently interpolate within the domain of the training data. To extrapolate to new chemistries and designs that fall outside the known distribution, ML approaches that enforce physics[111] and/or analogical reasoning[112] are needed.

Rational design of MLFs is also hindered by a limited understanding about basic mechanisms by which small molecules (e.g. O_2, H_2O, CO_2, N_2) traverse polymer films. Beyond general trends with respect to crystallinity, density, and other basic physical properties, little is known about the molecular level permeation mechanisms of these small molecules. Both simulation and experimentation are needed to investigate these mechanisms. MD simulations can provide detailed, quantitative insights at spatiotemporal scales not readily available experimentally and are well-suited to describe the complex polymer-fluid and polymer-gas interactions critical for characterizing barrier properties. Advanced X-ray and neutron scattering experiments can complement atomistic simulations to probe specific structure-property relationships that govern barrier performance in polymer systems. Also lacking are robust models to predict the processing conditions that would yield optimized morphologies to achieve barrier and other properties. These would require *in silico* predictions of material rheology and crystallization kinetics to feed to computational fluid dynamics simulations of melt processing behavior. The elucidation of detailed gas permeation mechanisms through polymer films via integration of molecular modeling, ML, and advanced material characterization via scattering could spur the development of design principles for packaging materials.

Methods to directly predict biodegradability or recyclability of a polymer are challenging due to the lack of amount and standardization of data, although some approaches are being developed[22]. Lin and Zhang demonstrated the potential of ML to predict aerobic biodegradation using a dataset of 74 polymers (R^2=0.66)[113]. Kern et al. performed a large-scale search for sustainable, chemically recyclable ring-opening polymerization (ROP) polymers using ML to optimize for specific properties[114]. The development of an ML model to accurately predict ceiling temperature (T_c) as a thermodynamic measure of the conditions favoring recyclability would be particularly valuable towards narrowing the search space for circular MLFs. We are currently developing ML models that can predict enthalpy and entropy of polymerization, which determine T_c, for systems that undergo chain-growth polymerization. For the candidates in this study that are formed via polycondensation, the degree of polymerization for step-growth polymers based on equilibrium conversion can be used to guide selection of polymerization conditions, albeit not a proper T_c, to define the potential for recyclability at a given set of conditions. Kinetic modeling as demonstrated by Coile *et al*. can be used to quantitatively assess this metric as a function of diverse backbone compositions[115]. As soon as these environmental and processing factors can be modeled and predicted, they can be included in a multi-objective design space[116] to ensure candidates meet these stringent requirements.

Despite significant advancements, the path to achieving circularity in MLFs remains fraught with challenges. A systemic shift in how packaging is designed, produced, used, and disposed of is essential. Collaboration across stakeholders including manufacturers, policymakers, researchers, and consumers will be critical to driving this transformation. At the same time, public awareness campaigns and readjusted consumer expectations can foster demand for sustainable alternatives. Regardless, innovations in recycling, polymer design, polymer processing, and AI/ML will continue to drive progress in the space of waste management and reduction.

Data Availability

The data that supports the findings presented in this perspective are available in the Supplementary Information. Polymer water vapor permeability data used to train the PolyID model are available at doi.org/10.5281/zenodo.18262440. All data are available from the corresponding author upon request.

Code Availability

The code to run and train the PolyID model and to run DORAnet are available at github.com/NREL/polyid and github.com/wsprague-nu/doranet, respectively. An updated web-based interface that serves the models and makes predictions is available at https://polyid.nrel.gov

References

1. Walker TW, *et al.* Recycling of multilayer plastic packaging materials by solvent-targeted recovery and precipitation. *Sci. Adv.* **6**, eaba7599 (2020).
2. Anukiruthika T, Sethupathy P, Wilson A, Kashampur K, Moses JA, Anandharamakrishnan C. Multilayer packaging: Advances in preparation techniques and emerging food applications. *CRFSFS* **19**, 1156-1186 (2020).
3. Coles R, Kirwan MJ. *Food and beverage packaging technology*. John Wiley & Sons (2011).
4. Horodytska O, Valdés FJ, Fullana A. Plastic flexible films waste management–A state of art review. *Waste Manag.* **77**, 413-425 (2018).
5. *Plastics and the Environment*. John Wiley & Sons (2003).
6. Schnurr REJ*, et al.* Reducing marine pollution from single-use plastics (SUPs): A review. *Mar. Pollut. Bull.* **137**, 157-171 (2018).
7. Tun TZ*, et al.* Polymer types and additive concentrations in single-use plastic products collected from Indonesia, Japan, Myanmar, and Thailand. *Sci. Total Environ.* **889**, 163983 (2023).
8. Hopewell J, Dvorak R, Kosior E. Plastics recycling: challenges and opportunities. *Philos. Trans. R. Soc. B or Phil. Trans. R. Soc. B-Biol. Sci.* **364**, 2115-2126 (2009).
9. Schyns ZOG, Shaver MP. Mechanical Recycling of Packaging Plastics: A Review. *Macromol. Rapid Commun.* **42**, 2000415 (2021).
10. Cecon VS, Curtzwiler GW, Vorst KL. A Study on Recycled Polymers Recovered from Multilayer Plastic Packaging Films by Solvent-Targeted Recovery and Precipitation (STRAP). *Macromol. Mater. Eng.* **307**, 2200346 (2022).
11. Tamizhdurai P*, et al.* A state-of-the-art review of multilayer packaging recycling: Challenges, alternatives, and outlook. *J. Clean. Prod.* **447**, 141403 (2024).
12. Hu Z, Bernsten SN, Shi C, Sangroniz A, Chen EYX, Miyake GM. Terpenoid-based high-performance polyester with tacticity-independent crystallinity and chemical circularity. *Chem* **10**, 3040-3054 (2024).
13. Quinn EC*, et al.* Installing Controlled Stereo-Defects Yields Semicrystalline and Biodegradable Poly(3-Hydroxybutyrate) with High Toughness and Optical Clarity. *JACS* **145**, 5795-5802 (2023).
14. Sangroniz A*, et al.* Improving the barrier properties of a biodegradable polyester for packaging applications. *Eur. Polym. J.* **115**, 76-85 (2019).
15. Sangroniz A, Zhu J-B, Etxeberria A, Chen EY-X, Sardon H. Modulating the Crystallinity of a Circular Plastic towards Packaging Material with Outstanding Barrier Properties. *Macromol. Rapid Commun.* **43**, 2200008 (2022).
16. Sangroniz A, Zhu J-B, Tang X, Etxeberria A, Chen EYX, Sardon H. Packaging materials with desired mechanical and barrier properties and full chemical recyclability. *Nat. Commun.* **10**, 3559 (2019).

17. Trinh BM, Chang BP, Mekonnen TH. The barrier properties of sustainable multiphase and multicomponent packaging materials: A review. *Prog. Mater. Sci.* **133**, 101071 (2023).
18. Choi K, Hong SH. Chemically recyclable oxygen-protective polymers developed by ring-opening metathesis homopolymerization of cyclohexene derivatives. *Chem* **9**, 2637-2654 (2023).
19. Jang Y-J, Nguyen S, Hillmyer MA. Chemically Recyclable Linear and Branched Polyethylenes Synthesized from Stoichiometrically Self-Balanced Telechelic Polyethylenes. *JACS* **146**, 4771-4782 (2024).
20. Anwar MA, Suprihatin, Sasongko NA, Najib M, Pranoto B. Challenges and prospects of multilayer plastic waste management in several countries: A systematic literature review. *CSCEE* **10**, 100911 (2024).
21. Quinn EC, Knauer KM, Beckham GT, Chen EYX. Mono-material product design with bio-based, circular, and biodegradable polymers. *One Earth* **6**, 582-586 (2023).
22. Chen T, *et al.* Machine intelligence-accelerated discovery of all-natural plastic substitutes. *Nat. Nanotechnol.* **19**, 782-791 (2024).
23. Fang Y, Wen Y, Dai L, Wang C-H, You S, Li W. Artificial intelligence in plastic recycling and conversion: A review. *Resour. conserv. recycl.* **215**, 108090 (2025).
24. Wilson AN, *et al.* PolyID: Artificial Intelligence for Discovering Performance-Advantaged and Sustainable Polymers. *Macromolecules* **56**, 8547-8557 (2023).
25. Risch SJ. Food Packaging History and Innovations. *J. Agric. Food Chem.* **57**, 8089-8092 (2009).
26. Morris BA. Flexible packaging past, present and future: Reflections on a century of technology advancement. *J. Plast. Film Sheet* **40**, 151-170 (2024).
27. Wagner Jr JR. *Multilayer flexible packaging*. William Andrew (2016).
28. Wing HJ. Water Impedance of Nitro-cellulose Films. *J. Ind. Eng. Chem.* **28**, 786-788 (1936).
29. Paunonen SI. Strength and barrier enhancements of cellophane and cellulose derivative films: a review. (2013).
30. Morris BA. *The science and technology of flexible packaging: multilayer films from resin and process to end use*. William Andrew (2022).
31. Demeuse MT. *Biaxial stretching of film: Principles and applications*. Elsevier (2011).
32. Wessling RA, Gibbs DS, DeLassus PT, Obi BE, Howell BA. Vinylidene Chloride Monomer and Polymers. In: *Kirk-Othmer Encyclopedia of Chemical Technology*).
33. Morris BA. 4 - Commonly Used Resins and Substrates in Flexible Packaging. In: *The Science and Technology of Flexible Packaging* (ed Morris BA). William Andrew Publishing (2017).
34. Piringer OG, Baner AL. *Plastic packaging materials for food: barrier function, mass transport, quality assurance, and legislation*. John Wiley & Sons (2008).
35. DeLassus P. Barrier Polymers. In: *Kirk-Othmer Encyclopedia of Chemical Technology*).
36. Mokwena KK, Tang J. Ethylene Vinyl Alcohol: A Review of Barrier Properties for Packaging Shelf Stable Foods. *Crit. Rev. Food Sci. Nutr.* **52**, 640-650 (2012).
37. Dunn T. *Manufacturing flexible packaging: materials, machinery, and techniques*. William Andrew (2014).
38. Brody AL. Packaging of Foods. In: *Encyclopedia of Food Microbiology (Second Edition)* (eds Batt CA, Tortorello ML). Academic Press (2014).

39. Maes C, Luyten W, Herremans G, Peeters R, Carleer R, Buntinx M. Recent Updates on the Barrier Properties of Ethylene Vinyl Alcohol Copolymer (EVOH): A Review. *Polym. Rev.* **58**, 209-246 (2018).
40. Morris BA. *The Science and Technology of Flexible Packaging: Multilayer Films from Resin and Process to End Use*. William Andrew (2016).
41. Mount E. Chapter 6 - Coextrusion equipment for multilayer flat films and sheets. In: *Multilayer Flexible Packaging* (ed Wagner JR). William Andrew Publishing (2010).
42. Soares CTdM, Ek M, Östmark E, Gällstedt M, Karlsson S. Recycling of multi-material multilayer plastic packaging: Current trends and future scenarios. *Resour. conserv. recycl.* **176**, 105905 (2022).
43. Sutliff BP, Goyal S, Martin TB, Beaucage PA, Audus DJ, Orski SV. Correlating Near-Infrared Spectra to Bulk Properties in Polyolefins. *Macromolecules* **57**, 2329-2338 (2024).
44. Sunil M, *et al.* Machine learning assisted Raman spectroscopy: A viable approach for the detection of microplastics. *JWPE* **60**, 105150 (2024).
45. Koinig G, Kuhn N, Fink T, Grath E, Tischberger-Aldrian A. Inline classification of polymer films using Machine learning methods. *Waste Manag.* **174**, 290-299 (2024).
46. Barbosa FD, Staffa LH, Costa LC. Recycling of PE/PA/EVOH Multilayer Flexible Packaging Films via Reactive Compatibilization. *J. Appl. Polym. Sci.* **n/a**, e57332.
47. Pracella M, Chionna D, Ishak R, Galeski A. Recycling of PET and Polyolefin Based Packaging Materials by Reactive Blending. *Polym. Plast. Technol. Eng.* **43**, 1711-1722 (2004).
48. Samios CK, Kalfoglou NK. Compatibilization of poly(ethylene-co-vinyl alcohol) (EVOH) and EVOH/HDPE blends with ionomers. Structure and properties. *Polymer* **39**, 3863-3870 (1998).
49. Zhan K, *et al.* Impact of thermomechanical reprocessing on multilayer plastic packaging blend. *Polym. Degrad. Stab.* **222**, 110710 (2024).
50. Maile K. Plastics Recyclers Europe announces new findings for PE film recycling. In: *Recycling Today* (2019).
51. Anuar Sharuddin SD, Abnisa F, Wan Daud WMA, Aroua MK. A review on pyrolysis of plastic wastes. *Energy Convers. Manag.* **115**, 308-326 (2016).
52. Bauer A-S, Tacker M, Uysal-Unalan I, Cruz RMS, Varzakas T, Krauter V. Recyclability and Redesign Challenges in Multilayer Flexible Food Packaging—A Review. *Foods* **10**, 2702 (2021).
53. Carullo D, *et al.* Testing a coated PE-based mono-material for food packaging applications: an in-depth performance comparison with conventional multi-layer configurations. *Food Packaging and Shelf Life* **39**, 101143 (2023).
54. Adam HB, Yousfi M, Maazouz A, Lamnawar K. Recycling of Multilayer Polymeric Barrier Films: an Overview of Recent Pioneering Works and Main Challenges. *Macromol. Mater. Eng.* **310**, 2400414 (2025).
55. Meys R, *et al.* Achieving net-zero greenhouse gas emission plastics by a circular carbon economy. *Science* **374**, 71-76 (2021).
56. DesVeaux JS, *et al.* Mixed polyester recycling can enable a circular plastic economy with environmental benefits. *One Earth* **7**, 2204-2222 (2024).
57. Curley JB, *et al.* Closed-loop recycling of mixed polyesters via catalytic methanolysis and monomer separations. *Nat. Chem. Eng.* **2**, 568-580 (2025).

58. Han C-T, *et al.* Circular Polymer Designed by Regulating Entropy: Spiro-Valerolactone-Based Polyesters with High Gas Barriers and Adhesion Strength. *JACS* **147**, 4511-4519 (2025).
59. Lagaron JM, Catalá R, Gavara R. Structural characteristics defining high barrier properties in polymeric materials. *MS&T* **20**, 1-7 (2004).
60. Siracusa V. Food Packaging Permeability Behaviour: A Report. *Int. J. Polym. Sci.* **2012**, 302029 (2012).
61. Salame M, Steingiser S. Barrier Polymers. *Polym. Plast. Technol. Eng.* **8**, 155-175 (1977).
62. Klopffer MH, Flaconneche B. Transport Properties of Gases in Polymers: Bibliographic Review. *OGST* **56**, 223-244 (2001).
63. Wu F, Misra M, Mohanty AK. Challenges and new opportunities on barrier performance of biodegradable polymers for sustainable packaging. *Prog. Polym. Sci.* **117**, 101395 (2021).
64. Murcia Valderrama MA, van Putten R-J, Gruter G-JM. PLGA Barrier Materials from CO2. The influence of Lactide Co-monomer on Glycolic Acid Polyesters. *ACS Appl. Polym. Mater.* **2**, 2706-2718 (2020).
65. Altay E, Jang Y-J, Kua XQ, Hillmyer MA. Synthesis, Microstructure, and Properties of High-Molar-Mass Polyglycolide Copolymers with Isolated Methyl Defects. *Biomacromolecules* **22**, 2532-2543 (2021).
66. Jem KJ, Tan B. The development and challenges of poly (lactic acid) and poly (glycolic acid). *Adv. Ind. Eng. Polym. Res.* **3**, 60-70 (2020).
67. Samantaray PK, *et al.* Poly(glycolic acid) (PGA): a versatile building block expanding high performance and sustainable bioplastic applications. *Green Chem.* **22**, 4055-4081 (2020).
68. Basu S, Plucinski A, Catchmark JM. Sustainable barrier materials based on polysaccharide polyelectrolyte complexes. *Green Chem.* **19**, 4080-4092 (2017).
69. Nair SS, Zhu JY, Deng Y, Ragauskas AJ. High performance green barriers based on nanocellulose. *Sustain. Chem. Process.* **2**, 23 (2014).
70. Su Z, *et al.* Robust, high-barrier, and fully recyclable cellulose-based plastic replacement enabled by a dynamic imine polymer. *J. Mater. Chem. A* **8**, 14082-14090 (2020).
71. Yu Z, Ji Y, Bourg V, Bilgen M, Meredith JC. Chitin- and cellulose-based sustainable barrier materials: a review. *Emerg. Mater.* **3**, 919-936 (2020).
72. Zeng J, *et al.* Development of high-barrier composite films for sustainable reduction of non-biodegradable materials in food packaging application. *Carbohydr. Polym.* **330**, 121824 (2024).
73. Bradford G, *et al.* Chemistry-Informed Machine Learning for Polymer Electrolyte Discovery. *ACS Cent. Sci.* **9**, 206-216 (2023).
74. Doan Tran H, *et al.* Machine-learning predictions of polymer properties with Polymer Genome. *J. Appl. Phys.* **128**, (2020).
75. Kim C, Batra R, Chen L, Tran H, Ramprasad R. Polymer design using genetic algorithm and machine learning. *Comput. Mater. Sci.* **186**, 110067 (2021).
76. Martin TB, Audus DJ. Emerging Trends in Machine Learning: A Polymer Perspective. *ACS Polym. Au* **3**, 239-258 (2023).
77. Mysona JA, Nealey PF, de Pablo JJ. Machine Learning Models and Dimensionality Reduction for Prediction of Polymer Properties. *Macromolecules* **57**, 1988-1997 (2024).
78. Qiu H, Sun Z-Y. On-demand reverse design of polymers with PolyTAO. *Npj Comput. Mater.* **10**, 273 (2024).

79. Andraju N, Curtzwiler GW, Ji Y, Kozliak E, Ranganathan P. Machine-Learning-Based Predictions of Polymer and Postconsumer Recycled Polymer Properties: A Comprehensive Review. *ACS Appl. Mater. Interfaces.* **14**, 42771-42790 (2022).
80. McDonald SM, Augustine EK, Lanners Q, Rudin C, Catherine Brinson L, Becker ML. Applied machine learning as a driver for polymeric biomaterials design. *Nat. Commun.* **14**, 4838 (2023).
81. Tran H, *et al.* Design of functional and sustainable polymers assisted by artificial intelligence. *Nat. Rev. Mater.* **9**, 866-886 (2024).
82. Khajeh A, Lei X, Ye W, Yang Z, Schweigert D, Kwon H-K. A self-improvable polymer discovery framework based on conditional generative model. *arXiv preprint arXiv:2312.04013*, (2023).
83. Yang Z, Ye W, Lei X, Schweigert D, Kwon H-K, Khajeh A. De novo design of polymer electrolytes using GPT-based and diffusion-based generative models. *Npj Comput. Mater.* **10**, 296 (2024).
84. Kim S, Schroeder CM, Jackson NE. Open Macromolecular Genome: Generative Design of Synthetically Accessible Polymers. *ACS Polym. Au* **3**, 318-330 (2023).
85. Liao V, Jayaraman A. Inverse Design of Block Polymer Materials with Desired Nanoscale Structure and Macroscale Properties. *JACS Au* **5**, 2810-2824 (2025).
86. Vogel G, Weber JM. Inverse design of copolymers including stoichiometry and chain architecture. *Chem. Sci.* **16**, 1161-1178 (2025).
87. Zhou T, Wu Z, Chilukoti HK, Müller-Plathe F. Sequence-Engineering Polyethylene–Polypropylene Copolymers with High Thermal Conductivity Using a Molecular-Dynamics-Based Genetic Algorithm. *J. Chem. Theory Comput.* **17**, 3772-3782 (2021).
88. Hayashi Y, Shiomi J, Morikawa J, Yoshida R. RadonPy: automated physical property calculation using all-atom classical molecular dynamics simulations for polymer informatics. *Npj Comput. Mater.* **8**, 222 (2022).
89. Gurnani R, Kuenneth C, Toland A, Ramprasad R. Polymer Informatics at Scale with Multitask Graph Neural Networks. *Chem. Mater.* **35**, 1560-1567 (2023).
90. Phan BK, Shen K-H, Gurnani R, Tran H, Lively R, Ramprasad R. Gas permeability, diffusivity, and solubility in polymers: Simulation-experiment data fusion and multi-task machine learning. *Npj Comput. Mater.* **10**, 186 (2024).
91. Shebek KM, Strutz J, Broadbelt LJ, Tyo KEJ. Pickaxe: a Python library for the prediction of novel metabolic reactions. *BMC Bioinformatics* **24**, 106 (2023).
92. Wang Y, van Putten R-J, Tietema A, Parsons JR, Gruter G-JM. Polyester biodegradability: importance and potential for optimisation. *Green Chem.* **26**, 3698-3716 (2024).
93. Dobbelaere MR, Lengyel I, Stevens CV, Van Geem KM. Geometric deep learning for molecular property predictions with chemical accuracy across chemical space. *J. Cheminform.* **16**, 99 (2024).
94. Zhou D, *et al.* Multi-step biosynthesis of the biodegradable polyester monomer 2-pyrone-4,6-dicarboxylic acid from glucose. *Biotechnol. Biofuels Bioprod.* **16**, 92 (2023).
95. EPA US. *User's Guide for T.E.S.T. (version 5.1) (Toxicity Estimation Software Tool): A Program to Estimate Toxicity from Molecular Structure* (2020).
96. Messin T, *et al.* Structure and Barrier Properties of Multinanolayered Biodegradable PLA/PBSA Films: Confinement Effect via Forced Assembly Coextrusion. *ACS Appl. Mater. Interfaces.* **9**, 29101-29112 (2017).

97. Dziadowiec D, Matykiewicz D, Szostak M, Andrzejewski J. Overview of the Cast Polyolefin Film Extrusion Technology for Multi-Layer Packaging Applications. *Materials* **16**, 1071 (2023).
98. van den Hurk RS, Pirok BWJ, Bos TS. The Role of Artificial Intelligence and Machine Learning in Polymer Characterization: Emerging Trends and Perspectives. *Chromatographia* **88**, 357-363 (2025).
99. Abeykoon C. Sensing technologies for process monitoring in polymer extrusion: A comprehensive review on past, present and future aspects. *Measurement: Sensors* **22**, 100381 (2022).
100. Wang C, *et al.* Autonomous platform for solution processing of electronic polymers. *Nat. Commun.* **16**, 1498 (2025).
101. Xie Y, He K, Castellanos-Gomez A. Toward Full Autonomous Laboratory Instrumentation Control with Large Language Models. *Small Struct.* **6**, 2500173 (2025).
102. Abramson J, *et al.* Accurate structure prediction of biomolecular interactions with AlphaFold 3. *Nature* **630**, 493-500 (2024).
103. Dauparas J, *et al.* Robust deep learning–based protein sequence design using ProteinMPNN. *Science* **378**, 49-56 (2022).
104. Watson JL, *et al.* De novo design of protein structure and function with RFdiffusion. *Nature* **620**, 1089-1100 (2023).
105. Berman HM, *et al.* The Protein Data Bank. *Nucleic Acids Res.* **28**, 235-242 (2000).
106. Walsh DJ, *et al.* Community Resource for Innovation in Polymer Technology (CRIPT): A Scalable Polymer Material Data Structure. *ACS Cent. Sci.* **9**, 330-338 (2023).
107. Gormley AJ, Webb MA. Machine learning in combinatorial polymer chemistry. *Nat. Rev. Mater.* **6**, 642-644 (2021).
108. Liu G, Jiang M. Transfer learning with diffusion model for polymer property prediction. In: *Workshop on "Machine Learning for Materials" ICLR* (2023).
109. Ahn J, Irianti GP, Choe Y, Hur S-M. Enhancing deep learning predictive models with HAPPY (Hierarchically Abstracted rePeat unit of PolYmers) representation. *Npj Comput. Mater.* **10**, 110 (2024).
110. Shen C, Zhang Y, Han F, Xia K. Molecular topological deep learning for polymer property prediction. *arXiv preprint arXiv:2410.04765*, (2024).
111. Jain A, Gurnani R, Rajan A, Qi HJ, Ramprasad R. A physics-enforced neural network to predict polymer melt viscosity. *Npj Comput. Mater.* **11**, 42 (2025).
112. Segal N, Netanyahu A, Greenman KP, Agrawal P, Gómez-Bombarelli R. Known Unknowns: Out-of-Distribution Property Prediction in Materials and Molecules. *Npj Comput. Mater.* **11**, 345 (2025).
113. Lin C, Zhang H. Polymer Biodegradation in Aquatic Environments: A Machine Learning Model Informed by Meta-Analysis of Structure-Biodegradation Relationships. *Environ. Sci. Technol.* **59**, 1253-1263 (2025).
114. Kern J, Su Y-L, Gutekunst W, Ramprasad R. An informatics framework for the design of sustainable, chemically recyclable, synthetically accessible, and durable polymers. *Npj Comput. Mater.* **11**, 182 (2025).
115. Coile MW, Harmon RE, Wang G, SriBala G, Broadbelt LJ. Kinetic Monte Carlo Tool for Kinetic Modeling of Linear Step-Growth Polymerization: Insight into Recycling of Polyurethanes. *Macromol. Theory Simul.* **31**, 2100058 (2022).

116. S. V SS, *et al.* Multi-objective goal-directed optimization of de novo stable organic radicals for aqueous redox flow batteries. *Nat. Mach. Intell.* **4**, 720-730 (2022).

Acknowledgements

Funding was provided by the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, Advanced Materials and Manufacturing Technologies Office (AMMTO), and Bioenergy Technologies Office (BETO). This work was performed as part of the Bio-Optimized Technologies to keep Thermoplastics out of Landfills and the Environment (BOTTLE) Consortium and was supported by AMMTO and BETO under contract DEAC36-08GO28308 with the National Renewable Energy Laboratory (NREL), operated by Alliance for Sustainable Energy, LLC. The views expressed in the article do not necessarily represent the views of the DOE or the U.S. Government. The U.S. Government retains and the publisher, by accepting the article for publication, acknowledges that the U.S. Government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this work, or allow others to do so, for U.S. Government purposes. SK and MM also gratefully acknowledge the support of RePLACE (Redesigning Polymers to Leverage A Circular Economy), funded by the Office of Science of the U.S. Department of Energy via award no. DR-SC0022290. We also thank Scivetica with their help with graphics.

Competing Interests Statement

The authors have no competing interests to declare.

Author Contributions Statement

E.C.Q, L.J.H., J.N.L., B.C.K., and K.M.K. conceived of the idea; E.C.Q, L.J.H., and J.N.L wrote the original manuscript; E.C.Q, L.J.H., J.N.L., B.C.K., and K.M.K. created the figures; R.W.C., M.M., S.K., R.M.M., and M.J.S., wrote and edited on select sections of the manuscript; L.J.B. and B.C.K build the model and neural network for PolyID; L.J.B., B.C.K., and K.M.K secured funding; all authors edited the manuscript.

Figure Legends/Captions (for main text figures)

Figure 1. A proposed ML-guided approach to designing recyclable and/or compostable multi-layer films (MLFs) that match the food preservation metrics and target performance of today's conventional materials. (A) Key structure-property relationships of current MLF polymers are studied; (B) Properties of known polymers are fed into an ML model (PolyID) to predict polymers (in this case study we targeted polyesters) with similar properties; (C) Using the output from PolyID, a new portfolio of polyesters for MLF applications is generated; (D) Examples of the types of MLF packaging targeted in this perspective.

Figure 2. Property comparison of conventional polymers used in MLFs. Summary of reported oxygen barrier (presented as oxygen permeability coefficient, PO_2), water vapor barrier (presented as water vapor permeability coefficient, PH_2O), melting temperature (T_m), modulus, density, and average cost (based on 2024 market reports) for incumbent polymer materials commonly used in multi-layer film layers. Barrier measurements and modulus are presented on logarithmic scales. Cost, density, and barrier measurements are presented on inverted scales. Barrer is

equivalent to $10^{-10}((cm_{STP}^3 * cm)/(cm^2 * s * cmHg))$. LDPE: low-density polyethylene, PP: polypropylene, PET: polyethylene terephthalate, PVDC: polyvinylidene dichloride, EVOH: ethylene vinyl alcohol.

Figure 3. **Generalized legacy MLF structure c and current end-of-life recycling strategies.** Legacy MLF packaging configurations and existing MLF recycling strategies including mechanical recycling, pyrolysis, and STRAP. LDPE: low-density polyethylene, LLDPE: linear low-density polyethylene, PP: polypropylene, PA: polyamide, PVDC: polyvinylidene dichloride, EVOH: ethylene vinyl alcohol, HDPE: high-density polyethylene, OPP: oriented polypropylene, PS: polystyrene, PET: polyethylene terephthalate.

Figure 4. Integrated PolyID-DORAnet workflow for polymer discovery and property prediction. DORAnet applies *in situ* chemical reactions to potential source materials to naturally expand the monomer discover space. PolyID, trained on experimental and/or molecular simulation data, is then used to identify candidate polymer formulations with desired properties. While this work does not perform molecular simulations, active learning, or repeated DORAnet-PolyID loops to optimize target properties, we include those steps in the figure to illustrate a holistic approach to continually improve the model predictions and target candidates. BP: boiling point, DoV: domain of validity, MLF: multi-layer film.

Figure 5. PolyID-generated predictions of polymer properties for polyesters produced from monomers derived from KEGG and Sigma databases and one round of DORAnet transformations. See **Figure S2** for more predicted property comparisons. Highlighted monomers are examples of under-explored chemistries for MLFs with relatively strong predicted barrier properties. PVDC: polyvinylidene dichloride, PLGA: poly(lactic-co-glycolic acid), PET: polyethylene terephthalate, PLA: polylactic acid, HDPE: high-density polyethylene, PP: polypropylene, P3HB: poly(3-hydroxybutyrate), EVOH: ethylene vinyl alcohol, PESt: poly(ethylene stearate).

Figure 6. Property comparison of emerging biopolymers and predicted polyester structure for potential use in future MLFs. Summary of oxygen barrier (presented as oxygen permeability coefficient, PO_2), water vapor barrier (presented as water vapor permeability coefficient, PH_2O), melting temperature (T_m), modulus, density, and average cost (based on 2024 market reports for polyglycolide (PGA), polylactic acid (PLA), and poly(3-hydroxybutyrate) (P3HB) for existing and a new polyester predicted by PolyID (PolyID P1). Barrier measurements and modulus are presented on logarithmic scales. Cost, density, and barrier measurements are presented on inverted scales.

Figure 7. Self-driving laboratories. Proposed schematic for combining in-line characterization to multi-layer film processing coupled with ML to better inform redesign efforts, reduce iteration cycles, and accelerate scale-up/commercialization.

Editorial Summary

Multi-layer film packaging revolutionized food preservation by combining diverse material layers to optimize barrier properties, mechanical strength, and shelf-life but they pose significant recycling challenges due to their structural complexity. This perspective examines key structure-property relationships governing barrier performance and highlights innovations in material design.

Peer review information: *Nature Communications* thanks Mehran Ghasemlou, Dina Maniar, and the other, anonymous, reviewers for their contribution to the peer review of this work.