Dissertation

Mechanical recycling and design of multi-layer packaging: Constraints, opportunities and challenges

carried out for the purpose of obtaining the degree of Doctor technicae (Dr. techn.), submitted at TU Wien, Faculty of Mechanical and Industrial Engineering, by

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Danksagung

Ein freundliches Wort kostet nichts, und dennoch ist es das Schönste aller Geschenke.
-
Daphne du Maurier

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Abstract

The use of polymers in food packaging allows economically efficient manufacturing and transport of lightweight low-cost versatile products able to protect sensitive food content while meeting the demands of the customers. A single polymer can often not meet all the application requirements such as good sealability, low-temperature resistance, barrier against gas and moisture as well as high mechanical stability. As different polymers are usually not compatible with each other, the mechanical recycling of such single-use systems has proven to be rather challenging and no industrial solution has been developed to date to address this issue.

In recent years, the emphasis in mechanical recycling has primarily been on "design from recycling" approaches, which have sometimes led to the production of lower-quality products due to material down-cycling compared to the performance of virgin polymers. To improve recyclability, it is essential to also consider "design for recycling" aspects during the product development process. This includes careful material selection and combination, as well as optimizing reprocessing parameters and exploring the potential for system compatibilization in a cost-effective manner.

The thesis proposes a comprehensive approach to assess the primary constraints and opportunities for the recycling of multi-layer packaging, involving analysis of package structure, mechanical recycling methods, and secondary material tests on a diverse range of post-consumer multi-layer packaging. This study included approximately 240 different packaging products from food and cosmetic packaging to cover the Austrian market for short-shelf-life consumer goods. Current multi-layer packaging is not recyclable without the development of effective separation and sorting techniques. Even separated waste fractions or individual layers of multi-layer packaging demonstrated significant variation in physical properties compared to virgin materials.

The use of different packaging materials, driven by functional and marketing requirements, resulted in structurally different types of packaging for the same product, leading to heterogeneity in the recycling stream. Contamination with minor material components, such as different polymers or other materials like aluminum or paper, further complicated the recycling process and affected the physical properties and aesthetics of the resulting recyclates. Thus, addressing the challenges posed by multi-layer packaging will require innovative solutions to enhance recyclability and reduce contamination in order to achieve more sustainable packaging systems.

The possible concern that improving the recyclability of multi-layer films leads to a decrease in packaging efficiency is likely baseless. Packaging analysis has shown that commercially available structures without PET and polyolefins already exist in various product areas. Examples such as PP-based packaging for sliced ham demonstrated that recyclable multi-layer structures can closely resemble the property profile of virgin polymers, and could even undergo successful functional closed-loop recycling. However, compatibilization of mixed polymer waste remains a challenge due to varying polymer ratios and unpredictable material properties. Bridging the gap between design for recycling and design from recycling may be achieved through holistic design choices, careful material selection, and managing profit margins, process adaptations, and product appearance. Harmonization of products and processes will be essential for achieving circularity in multi-layer packaging and making them a valuable source of material in the future.

Kurzfassung

Der Einsatz von Polymeren in Lebensmittelverpackungen ermöglicht die wirtschaftliche Herstellung und den Transport leichter, kostengünstiger und vielseitiger Produkte, die empfindliche Lebensmittel schützen und gleichzeitig den Anforderungen der Kunden gerecht werden. Ein einzelnes Polymer kann oft nicht alle Anwendungsanforderungen erfüllen, wie z. B. gute Siegelfähigkeit, Temperaturbeständigkeit, Barriere gegen Gas und Feuchtigkeit sowie hohe mechanische Stabilität. Da verschiedene Polymere in der Regel nicht miteinander kompatibel sind, hat sich das mechanische Rezyklieren solcher Einwegprodukte als herausfordernd erwiesen, und bis heute wurde keine industrielle Lösung für dieses Problem entwickelt.

In den letzten Jahren lag der Schwerpunkt des mechanischen Rezyklierens vor allem auf dem "Design from Recycling"-Ansatz, der mitunter dazu führte, dass Produkte von geringerer Qualität hergestellt wurden, weil das Material im Vergleich zur Leistung von Neuware schlechter abschneidet. Um die Rezyklierfähigkeit zu verbessern, ist es wichtig, bei der Produktentwicklung auch Aspekte des "Design for Recycling" zu berücksichtigen. Dazu gehören eine sorgfältige Materialauswahl und -zusammensetzung sowie die Optimierung der Wiederaufbereitungsparameter und die Miteinbeziehung des Potenzials kosteneffizienter Kompatibilisierungsmöglichkeiten.

Die Arbeit bietet einen umfassenden Überblick zur Bewertung von Hindernissen und Möglichkeiten für das Rezyklieren von Mehrschichtverpackungen, einschließlich der Analyse von Verpackungsstrukturen, Durchführung mechanischer Rezyklierversuche und Testung der entstandenen Rezyklate. Die Studie umfasste etwa 240 verschiedene Verpackungen von Lebensmitteln und Kosmetika (Abbildung des österreichischen Marktes). Die aktuellen Mehrschichtverpackungen sind ohne effektivere Trenn- und Sortiertechniken nicht rezyklierbar, da selbst sortenreine Fraktionen erhebliche Unterschiede zu den Eigenschaften von Neuware aufwiesen.

Der Einsatz unterschiedlicher Verpackungsmaterialien, bedingt durch funktionale und marketingbezogene Anforderungen, führte zu unterschiedlichen Verpackungen für dasselbe Produkt, was die Heterogenität der Rezyklatströme begünstigte. Verunreinigungen mit geringen Mengen von Fremdpolymeren oder anderen Materialien (Aluminium, Papier) erschwerten den Rezyklierprozess zusätzlich und beeinflussten die physikalischen Eigenschaften und das Aussehen der resultierenden Rezyklate. Daher erfordert die Bewältigung der Herausforderungen von Mehrschichtverpackungen innovative Lösungen, um die Rezyklierfähigkeit zu verbessern und Verunreinigungen zu reduzieren, um nachhaltigere Verpackungssysteme zu generieren.

Die mögliche Sorge, dass die Verbesserung der Rezyklierfähigkeit von Mehrschichtfolien zu einer Verringerung der Verpackungseffizienz führt, ist überwiegend unbegründet. Verpackungsanalysen haben gezeigt, dass bereits kommerziell verfügbare Strukturen ohne Kombination von PET mit Polyolefinen in verschiedenen Produktbereichen existieren. Beispiele wie PP-basierte Verpackungen für Aufschnitt haben gezeigt, dass rezyklierbare Mehrschichtstrukturen dem Eigenschaftsprofil von neuwertigen Polymeren nahe kommen können und sogar die technische Umsetzbarkeit eines geschlossenen Kreislaufs konnte demonstriert werden. Die Kompatibilisierung von gemischten Polymerabfällen bleibt jedoch aufgrund unterschiedlicher Polymerverhältnisse und unvorhersehbarer Materialeigenschaften eine Herausforderung. Die Verbindung zwischen "Design for Recycling" und "Design from Recycling" kann durch ganzheitliche Gestaltung, sorgfältige Materialauswahl sowie die Anpassung der Erwartungen an Gewinnpannen, Produktionsprozesse und Produktoptik geschaffen werden. Die Harmonisierung von Produkten und Prozessen wird entscheidend sein, um die Kreislauffähigkeit von Mehrschichtverpackungen zu erreichen und sie zu einer wertvollen Materialquelle für die Zukunft zu machen.

Published Material

Research Paper

Publication 1:
Seier, M., Archodoulaki, V.-M., Koch, T., Duscher, B., & Gahleitner, M. (2022). Polyethylene terephthalate based multilayer food packaging: Deterioration effects during mechanical recycling. Food Packaging and Shelf Life, 33, Article 100890. https://doi.org/10.1016/j.fpsl.2022.100890

Publication 2:
Seier, M., Archodoulaki, V.-M., Koch, T., Duscher, B., & Gahleitner, M. (2023). Prospects for Recyclable Multilayer Packaging: A Case Study. Polymers, 15(13), 2966. https://doi.org/10.3390/polym15132966

Publication 3:
Seier, M., Roitner, J., Archodoulaki, V.-M. & Jones, M.P. (2023). Design from recycling: Overcoming barriers in regranulate use in a circular economy. Resources, Conservation and Recycling, 196, 107052. ttps://doi.org/10.1016/j.resconrec.2023.107052.

Publication 4 (not included in the thesis):
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Seier, M., & Archodoulaki, V.-M. (2021). Herausforderungen im mechanischen Recycling von Einweg-Kunststoffen und Möglichkeiten neuer Konzepte. In Internationale wissenschaftliche Tagung Polymertec 2021 (p. 44).

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Patent Applications

EP 22177222
„Mechanically recyclable modified atmosphere packaging system based on PP” (filing date 03.06.2022)

EP 23163969
"All-PP modified atmosphere packaging system suitable for mechanical recycling" (filing date 24.03.2023)

List of Figures

List of Tables

List of Abbreviations

CP	Cheese Packaging
DIN	Deutsche Industrie Norm
DSC	Differential Scanning Calorimetry
EFSA	European Food Safety Authority
EN	European Norm
EPS	Expanded Polystyrene
EVA	Ethylene-Vinyl-Acetate
EVOH	Ethylene-Vinyl-Alcohol
FDA	Food and Drug Administration
FT-(N)IR	Fourier Transform (Near) Infrared
HIPS	High-Impact Polystyrene
IML	In-Mold Labeling
ISO	International Standard Organization
LVE	Linear Viscoelastic Range
MAP	Modified Atmosphere Packaging
MCR	Modular Compact Rheometer
MWD	Molecular Weight Distribution
MW	Molecular Weight
NIAS	Non-Intentionally Added Substances
PA	Polyamide
PC	Post Consumer
PE	Polyethylene
PE-HD	Polyethylene high density
PE-LD	Polyethylene low density
PE-LLD	Polyethylene linear low density
PET	Polyethylene terephthalate
PLA	Polylactic Acid
PP	Polypropylene
PS	Polystyrene
PU	Polyurethane
SEM	Scanning Electron Microscopy
SER	Sentmanant Extensional Rheometer
SP	Sausage Packaging
SPI	Society of the Plastic Industry
UV	Ultra Violet
A	Cross-section area
a_{tN}	Tensile impact toughness
$\dot{\gamma}$	Shear
CoF	Coefficient of Friction
c_p	Heat capacity
δ	Loss angle
$tan(\delta)$	Loss factor
ε	Strain
$\dot{\varepsilon}$	Strain rate
ε_b	Elongation at break
E_t	Elastic modulus
E_p	Puncture energy
F	Force

G	Gibbs free energy
G'	Storage modulus
G''	Loss modulus
η	Viscosity
η^*	Complex viscosity
η_0	Zero shear viscosity
$\eta(t)$	Highest extensional viscosity
$\eta_0(t)$	Extensional viscosity of LVE curve
H	Enthalpy
l	Displacement
m	Mass
MFR	Melt Flow Rate
ν	Heating/cooling rate
OTR	Oxygen Transmission Rate
ρ	Density
$\dot{Q}$	Heat flow
σ	Stress
S	Entropy
SHC	Strain Hardening Coefficient
T	Temperature
T_c	Crystallization temperature
T_g	Glass transition temperature
T_m	Melt temperature
$WVTR$	Water Vapor Transmission Rate
φ	Volume fraction
ω	Angular frequency

Contents

1 Introduction

Over the past few years, there has been a significant increase in the use of plastics due to their diverse properties and application possibilities. Polypropylene, polyethylene, polyethylene terephthalate, and polystyrene are among the most important polymers used globally [1], finding their way into various industries such as automotive, medical, building, and construction. Despite their versatility, the packaging industry remains the largest consumer of these polymers [2], with products having a short lifespan, also known as single-use items. As a result, the recycling of products with a short lifespan has become a pressing issue, leading to the ban of single-use bags, straws, and cutlery, among other polymer products. The challenge is particularly acute for packaging made of multi-layer films or containing several types of polymers, as they are difficult to recycle, and no viable industrial solutions have been developed to address this challenge.

In order to evaluate the recycling potential of multi-material packaging, it is necessary to gain profound information about the polymers used in multi-layer films. There are no standards that specify a uniform structure for laminated films, meaning that there is a wide variety of product variants on the market. However, manufacturers are usually unwilling to disclose comprehensive company-internal data. Although the literature provides good knowledge about the structure and content of various packaging products, there are also strong deviations found concerning that topic.

This thesis assesses to investigate the primary constraints and opportunities for the recycling of multi-layer packaging using a comprehensive approach that involves analyzing the package structure (layer materials and thicknesses), mechanical recycling methods (cold wash, grinding, extrusion), and secondary material tests (morphology, mechanics, rheology, thermal properties) on a diverse range of post-consumer packaging. A total number of approximately 240 different packaging products from food and cosmetic packaging are included in the study to widely cover the Austrian market for short-shelf-life consumer goods.

By conducting a structural analysis, it should be possible to quantify the individual polymers in the packaging categories by mass percentages, allowing for the estimation of waste streams generated. Besides the consideration of material combinations, whose formation during the recycling process is inevitable (inseparable multi-layers, polymers with similar densities), different mixtures are also specifically tested in order to be able to define maximum values for specific contaminants.

The study also aims to provide for a comparison of recycled materials versus virgin polymer resins to understand the degradation processes through the individual mechanical recycling steps. Detailed consideration of various contaminants and their origins (inks, food residues, different adhesives, fillers), as well as their effects on the recyclate, are only partially content of the work, as it would have exceeded the scope.

Ideally, the results of the work will enable a comprehensive linkage between design for and design from recycling for a wide variety of packaging products. The transition from linear to a circular economy for primary packaging products is currently still limited by safety concerns due to a lack of sufficiently efficient purification methods Still, the proper design for recyclability is a first step towards circularity.

1.1 Plastics in packaging: A little history

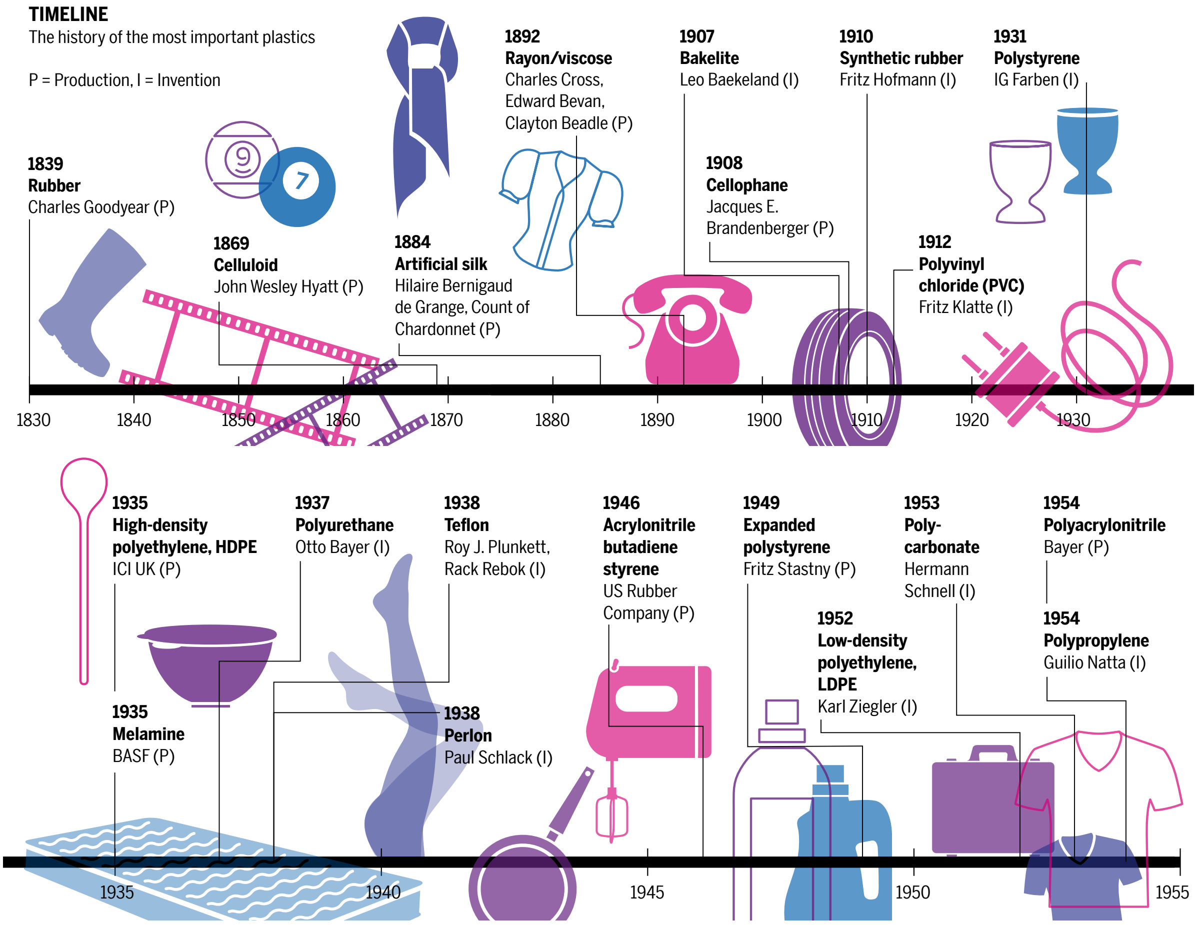


Figure 1: Invention history of the most important (packaging) polymers from 1830-1955. [1]

The earliest attempts to wrap foods in polymeric materials were made with cellophane films around 1900. At that time, a significant development was seen in the field, with new materials and technologies transforming not only the way goods were packaged but also their transportation. Cellophane was first used for packaging cigarettes in the 1920s, and by the 1930s, it had become a popular packaging material for food products. [3,4]

In the 1930s, new types of polymers, such as polyethylene (PE) and polystyrene (PS), were developed, which opened up new possibilities for packaging. PE, a lightweight, flexible plastic, was first produced in the 1930s and was used for packaging applications such as bags and wraps. [5] Films made from PE quickly replaced waxed papers as bread storage bags and also wax coatings used for heat-sealable paperboard. [6, 7] PS, a hard, brittle plastic, was first produced in 1931 and was used for packaging applications such as containers and cups. [8] However, the primary use of polymers for packaging applications started after World War II, in which PE had been produced in very high quantities. The demand for new packaging overgrew as a result of the need to transport goods efficiently and safely for the war effort. [3] This led to the development of new materials such as nylon, which was used for parachute fabric and eventually found its way into other applications such as food packaging. [9]

In the 1950s, the development of high-density polyethylene (PE-HD) and polypropylene (PP) further expanded the possibilities for polymer packaging. These materials were stronger, more durable, and more heat-resistant than earlier plastics, which made them ideal for a range of applications, from food packaging to automotive parts. [10] These new materials also paved the way for the development of more sophisticated packaging technologies, such as blow molding and injection molding, which made it possible to create a wide range of shapes and sizes for packaging containers. [11] The manufacturing of bi-axially oriented PP was discovered in a blown film process in the 1960s, which was a critical factor in the general replacement of cellophane due to cost efficiency and environmentally friendly production (figure 1). [4]

One of the most significant developments in polymer packaging between 1950-1970 was the introduction of polyethylene terephthalate (PET) in the 1960s. This new material was transparent, lightweight, and strong, which made it ideal for use in a wide range of packaging applications. However, in the beginning, PET was mainly used for the manufacturing of bottles to carry carbonated drinks but became more popular for pouches soon due to its high thermal stability, which allowed for reheating food in boiling water. [12, 13]

Soon, another major development was the introduction of vacuum packaging, which helped to extend the shelf life of many food products. Vacuum packaging involves removing air from a package to create a vacuum, which helps to reduce the growth of bacteria and other microorganisms. This made it possible to preserve foods for longer periods of time, which helped to reduce food waste and spoilage. [14, 15] The concept was later adapted and involved altering of the composition of air surrounding a food product to extend its shelf life. This can be achieved by removing oxygen from the package, replacing it with another gas such as nitrogen or carbon dioxide, or by controlling the level of oxygen and other gases inside the package, which is today widely known as modified atmosphere packaging (MAP). [16, 17] From that point, many attempts have been made to improve food packaging structures and especially permeability properties. Primarily, different coatings (metal, wax), polyamide (PA) films, blends including paraffin wax, or organic fillers, were used as an approach to improve barrier function. [18–21] The use of ethylene-vinyl-acetate (EVA) and ethylene-vinyl-alcohol (EVOH) copolymers, which were discovered to have an excellent barrier against oxygen, started around 1975 and massively improved packaging technologies. [22–24]

At the same time, co-extrusion, for the production of multi-layer films, was introduced, which most likely was a key factor for the success of especially these resins. Due to their high popularity and functionality, the combination of PET and EVOH became more and more present in packaging applications. [25] Next to the increasing use of multi-layer packaging, the introduction of expanded polystyrene (EPS) and polyurethane (PU) foams, which offered improved cushioning and insulation properties for a range of packaging applications, were distinctive for the time between 1970-1990. [26]

The rapidly growing range of polymer packaging materials and their widespread application also raised increasing awareness of the environmental impact of packaging. [27, 28] One of the most critical steps in that context was the first establishment of recycling codes for polymers in 1988 by the Society of the Plastics Industry (SPI), a trade association representing the plastics industry in the United States. These codes were created to help consumers identify the different types of plastic materials and facilitate the recycling process. [29]

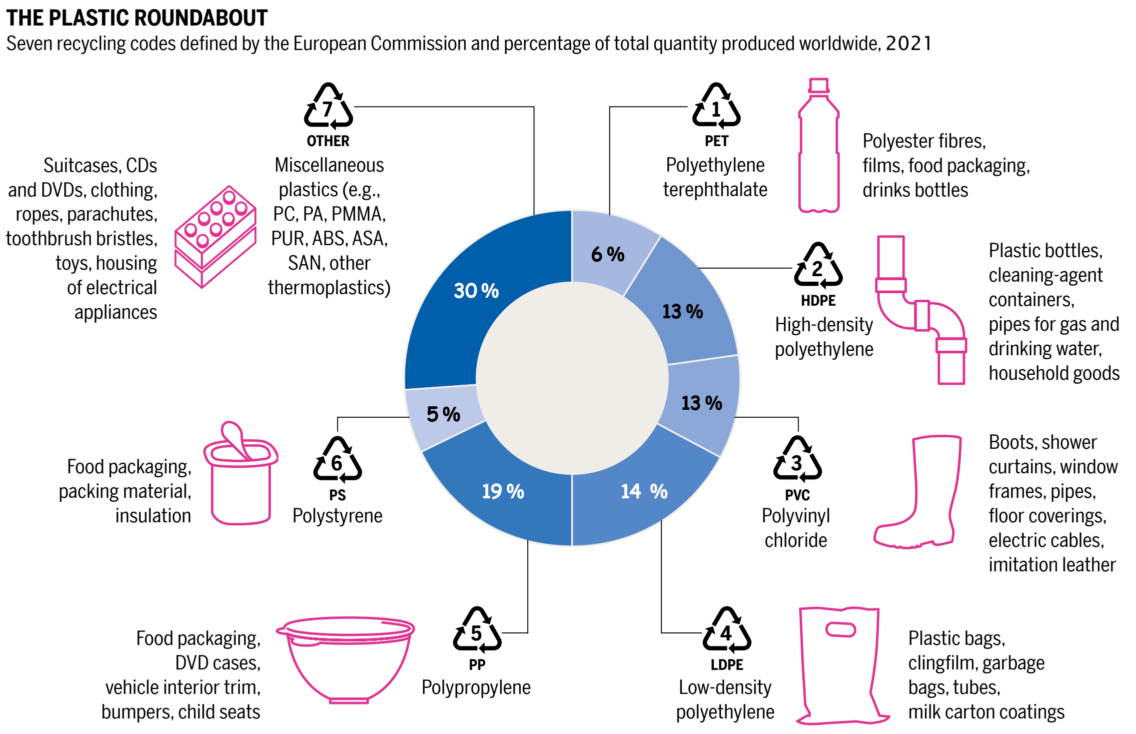


Figure 2: Polymer recycling codes, resin application and individual percentage of globally produced quantities in 2021 [1, 2].

Figure 2 depicts the seven recycling codes for the most frequently used polymers, some application examples, and their annual production quantities in 2021. The applications demonstrate that most of these polymers are used today in packing either as bottles (PET, PE-HD), cups (PS, PP), bags (PE-LD), or combined multi-component structures (other). [1]

During the recent 20-30 years, only minor material selection changes were made in packaging compared to the period from 1930-1990. The time was rather shaped by technological innovations and process improvements. This includes the development of bio-based and degradable materials and coatings such as polylactic acid (PLA). [30–32] Unfortunately, most of these materials are still too expensive to allow economically attractive application, do not meet all the technical requirements needed for the substitution of commercial polymers, or their compostation requirements are not industrially applicable yet. [33] Also, the potential use of nano-materials for the food market is often discussed and could bring many advantages, such as improvement of the barrier properties, thermal stability, strength, and durability. Nevertheless, future developments in this sector will strongly depend on the outcome of ongoing discussions and studies regarding the environmental impact and possible toxicity of nano-materials. [34]

Next to that, 3D printing technologies started to offer new possibilities for customized and on-demand packaging solutions in small quantities. [35, 36] Advanced active and smart packaging systems, which incorporate antimicrobial agents, oxygen scavengers, and other additives to improve the shelf life and safety of food products, became increasingly popular as well and will be further improved in the future. [37]

1.2 Primary packaging: Materials, Manufacturing, Requirements

Primary packaging refers to the layer of packaging that directly encloses a product, often coming into direct contact with it. It is the first layer of packaging that a consumer will see and interact with, and its primary function is to protect the product from damage, contamination, or deterioration during transport, storage, and use. [38]

The materials used for primary packaging can vary depending on the product and its intended use. For example, food and pharmaceutical products may require different types of primary packaging materials to ensure safety, freshness, and efficacy. Common materials for primary packaging include plastics, glass, metal, and paper-based materials, frequently in combination. [39]

Next to the necessary functional requirements, the packaging design can also impact the product's usability and consumer experience by shaping the consumer's perception of the product, making it a critical consideration for manufacturers and marketers alike. Unfortunately, this enormous marketing potential promotes the mixture of different materials to realize a specific look, utilization of large prints, and complex surface finishing methods which negatively influence the recyclability and hence the product sustainability. [40, 41]

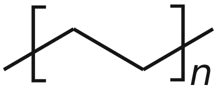
1.2.1 Packaging materials

Polyethylene (PE)

PE (table 2) is a widely used thermoplastic polymer that is known for its versatility and low cost. One of the key advantages of PE is its resistance to chemicals, moisture, and impact. PE can be found in different types, including high-density polyethylene (PE-HD), low-density polyethylene (PE-LD), and linear low-density polyethylene (PE-LLD). It is produced by the polymerization of ethylene gas, a simple organic compound derived from natural gas or petroleum. The polymerization process involves the addition of ethylene monomer units to form a long-chain molecule with repeating ethylene units. This process can be carried out by using a variety of methods, which including free radical polymerization, Ziegler-Natta polymerization, and metallocene catalysis. [42]

PE packaging offers many advantages, such as heat seal-ability due to its low melting point and excellent barrier against water vapor. PE-HD is stiffer compared to PE-LD and PE-LLD and therefore used for rigid blow-molding containers and bottles for beverages (ketchup) and cosmetic products (shampoo). Contrarily, PE-LD and PE-LLD are commonly used for the manufacturing of flexible films by film blowing or casting processes. PE is also used in multi-layer packaging structures, where it can be combined with other materials such as EVOH, PA, or aluminum foil to provide superior barrier properties. Blending of PE-LD and PE-LLD resins prior to manufacturing is also a common practice to enhance process stability (e.g., bubble stability in film blowing). While PE-HD results in opaque products, films from PE-LLD and PE-LD are highly transparent. [3,43]

Table 2: Properties and structure of PE [44,45]

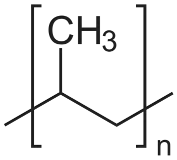
Properties		**Structure**
Melt temperature T_m	105-135 °C	
Desitiy ρ	0.91-0,96 g/cm^3	
Elastic modulus E_t	200-1400 MPa	
Water vapor transmission rate $WVTR$	0.6-1 $g/m^2 \cdot d$	
Oxygen transmission rate OTR	50-200 $cm^3 \cdot mm/m^2 \cdot d \cdot atm$	

Polypropylene (PP)

PP (table 3) is a thermoplastic polymer that is widely used in a variety of applications due to its unique combination of properties, including high stiffness, low density, and excellent chemical resistance. Comparable to PE, PP is produced by the polymerization of propylene monomer, which results in different PP types. PP homopolymer is the most common type and is produced from a single monomer, propylene. PP copolymers are both made by the copolymerization of propylene and a small amount of ethylene which gives the material some PE-like properties such as increased toughness. [46,47]

PP homopolymer, which has a high degree of stiffness, can be used in rigid packaging such as containers, caps, and closures. One advantage of PP homopolymer in packaging is its relatively high melting point which makes it suitable for use in microwaveable food trays or pouches. PP is also a highly customizable material, which allows for the production of packaging with specific properties, such as opacity or clarity, depending on the requirements of the product being packaged. Comparable to PE, PP copolymers can be manufactured into films by casting or blowing processes. Cast PP films are commonly used for food packaging, as they offer excellent clarity and are easy to print on. Blown PP films are more durable and have better puncture resistance, making them suitable for industrial and heavy-duty packaging applications. PP is offered in a wide viscosity range which dictates suitable processing methods. While highly viscous types are used for the manufacturing of films or thermoforming of cups, low-viscosity types are suitable for injection molding. [3,4]

Table 3: Properties and structure of PP [44,45]

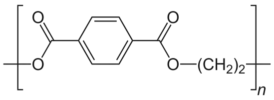
Properties		**Structure**
Melt temperature T_m	145-170 °C	
Desitiy ρ	0.90-0.91 g/cm^3	
Elastic modulus E_t	900-2000 MPa	
Water vapor transmission rate $WVTR$	0.7-0.8 $g/m^2 \cdot d$	
Oxygen transmission rate OTR	50-100 $cm^3 \cdot mm/m^2 \cdot d \cdot atm$	

Polyethylene terephthalate (PET)

PET (table 4), a thermoplastic polymer, is mainly used in the production of a variety of products, including bottles, packaging films, and fibers. One of the main material advantages is its high strength and durability and excellent clarity. Depending on the processing conditions (slow or rapid cooling), crystals within the polymer melt may or may not form, resulting in a semi-crystalline or amorph material. PET is produced through the polymerization of terephthalic acid or its dimethyl ester with ethylene glycol. The polymerization process results in the formation of long chains of PET molecules that are linked together through ester bonds. Drying of the material prior to processing is necessary as the absorbance of too much moisture leads to scission of hydrogen bonds and, subsequently deterioration. [4]

PET offers a moderate barrier against oxygen and moisture, which makes the material attractive for a wide range of different packaging applications for beverages, juices, and other liquid foods, as well as snacks and other dry foods. PET bottles and jars also offer good resistance to impact and temperature changes, making them suitable for hot and cold-filled products. Even though PET is often used for rigid bottles or containers, PET films offer excellent transparency, flexibility and have good dimensional stability. The films can be produced as mono- or multi-layer structures together with other polymer films such as PE, EVOH, or PA to offer tailored storage possibilities. [3]

Table 4: Properties and structure of PET [44,45]

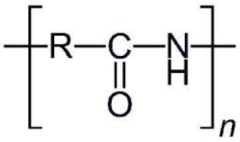
Properties		**Structure**
Melt temperature T_m	255 °C	
Desitiy ρ	1.37 g/cm^3	
Elastic modulus E_t	3100 MPa	
Water vapor transmission rate $WVTR$	5 $g/m^2 \cdot d$	
Oxygen transmission rate OTR	1-5 $cm^3 \cdot mm/m^2 \cdot d \cdot atm$	

Polyamide (PA)

PA (table 5) is a thermoplastic polymer that is commonly used in a wide range of applications due to its excellent mechanical properties, chemical resistance, and thermal stability. It is also known as nylon, which is one of the most popular PA materials. PA is made by reacting a diamine with a dicarboxylic acid or a lactam monomer. This polymerization process produces a long-chain molecule with repeating amide linkages, which gives PA its unique properties. There are many different PA types on the market (PA 6; PA 6,6; PA 12; PA 6,12 etc.), which can be distinguished by their nomenclature. The basic reaction is based on one (A-B) or two different (A-A/B-B) functional groups and leads to linear poly-condensates. The number indicates the number of C atoms in the respective monomer unit. In the case of PA 66, therefore, there are two different monomers (AA and BB), each containing 6 C atoms. [29]

PA is predominantly used for flexible packaging and is known for its excellent barrier properties, strength, and durability. The material provides high gas, flavor, and aroma barrier to maintain the taste and keep products fresh. It can be used as a mono-layer if very good puncture resistance is required (e.g., sausage casing with metal clamps) or together with other polymers such as PET in multi-layer structures. PA is commonly used as PA 6 in meat and cheese packaging, as well as in snack food packaging. [48]

Table 5: Properties and structure of PA 6 [44, 45]

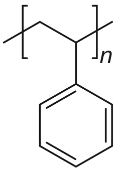
Properties		**Structure**
Melt temperature T_m	220 °C	 $\left[R-\underset{\overset{\|}{O}}{C}-\underset{H}{N} \right]_n$
Desitiy ρ	1.13 g/cm^3	
Elastic modulus E_t	1400 MPa	
Water vapor transmission rate $WVTR$	10-20 $g/m^2 \cdot d$	
Oxygen transmission rate OTR	1.4 $cm^3 \cdot mm/m^2 \cdot d \cdot atm$	

Polystyrene (PS)

PS (table 6) is an amorphous thermoplastic polymer. One of the main advantages of PS is its lightweight and rigid nature, which makes it ideal for use in packaging materials. It is also an excellent insulator and is commonly used in the construction industry for insulation purposes. The polymerization process for PS can be carried out using two different methods, bulk polymerization, and suspension polymerization. In bulk polymerization, the styrene monomers are heated and polymerized in a single reaction vessel, resulting in the formation of a solid mass of PS. This method is typically used for the production of high-impact PS (HIPS), which is a rubber-modified version of PS with improved toughness. [49]

PS is used in both rigid and flexible packaging and is known for its excellent clarity, insulation properties, and cost-effectiveness. The high degree of transparency makes it an ideal choice for packaging products that need to be visually appealing, such as cosmetics, toys, and electronics. To safeguard consumer products from damage, PS in foam form is often used to absorb transportation and handling vibrations. CD and DVD cases, foam packing peanuts for shipping, meat/poultry/egg trays, and food packaging are among the typical products made from PS. HIPS, which has a naturally white coloring, is next to PP, the most popular material for the manufacturing of yogurt cups. [3, 29]

Table 6: Properties and structure of PS [44, 45]

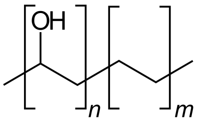
Properties		**Structure**
Softening temperature T	95-100 °C	
Desitiy ρ	1.05 g/cm^3	
Elastic modulus E_t	3200-3250 MPa	
Water vapor transmission rate $WVTR$	12 $g/m^2 \cdot d$	
Oxygen transmission rate OTR	100-150 $cm^3 \cdot mm/m^2 \cdot d \cdot atm$	

Ethylene-vinyl-alcohol (EVOH)

EVOH (table 7), a thermoplastic polymer, is typically used as a component in multi-layer packaging structures, where it is combined with other materials such as PE, PP, or PET. The polymer is highly impermeable to gases such as oxygen and carbon dioxide, which makes it an effective barrier material in food packaging. It is synthesized through the copolymerization of ethylene and vinyl alcohol monomers. EVOH can be produced in different grades or compositions depending on the specific application. The ethylene content in the copolymer can vary from 15% to 50% by weight, with higher ethylene content providing better flexibility and lower oxygen barrier properties, while higher vinyl alcohol content provides better oxygen barrier properties and stiffness. [48]

EVOH offers excellent oxygen barrier properties, which makes it very attractive for use in vacuum packaging or MAPs. The material is commonly used in the production of flexible packaging for foods such as meat, cheese, and bakery products. Despite the excellent barrier properties against oxygen, the material is sensitive to moisture, which often necessitates the combination of EVOH in 3-layer composites with polyolefins such as PE. [3]

Table 7: Properties and structure of EVOH [44, 50]

Properties		**Structure**
Melt temperature T_m	165-195 °C	
Desitiy ρ	1.12-1.24 g/cm^3	
Elastic modulus E_t	1900-3500 MPa	
Water vapor transmission rate $WVTR$	8-12 $g/m^2 \cdot d$	
Oxygen transmission rate OTR	0.04-0.4 $cm^3 \cdot mm/m^2 \cdot d \cdot atm$	

1.2.2 Manufacturing processes

Polymer packaging is used extensively in many industries, including food and beverage, pharmaceutical, and consumer goods. The manufacturing process for polymer packaging typically involves several production steps, including resin selection, molding or extrusion, and finishing.

Polymer selection is a crucial step in the manufacturing process because different polymers have different properties that make them suitable for different types of packaging and manufacturing processes (table 8). Proper processes are mainly dictated by the material viscosity, which is commonly expressed as melt flow rate (MFR). [51]

Table 8: Manufacturing processes and product applications depending on polymer viscosity [51, 52]

Manufacturing process	Viscosity range	Product output
Film blowing	very high-high	bags, pouches, lids, labels
Blow molding, thermoforming, foaming	high-moderate	bottles, cups, containers, foam
Film casting	moderate-low	bags, pouches, lids, labels
Injection molding	low-very low	bottles, bottle caps, cups

Extrusion

Following the material selection, the next processing step for resins is commonly extrusion. The property profile of the raw resins can be further influenced by a broad range of additives and fillers. These are usually added prior to or during the extrusion process. The main objectives are stabilization (heat stabilizer, UV stabilizer), the coloring of the material (pigments), facilitation of processing (anti-slip, anti-static) and improvement of the service properties (titan dioxide, calcium stearate). [51, 53]

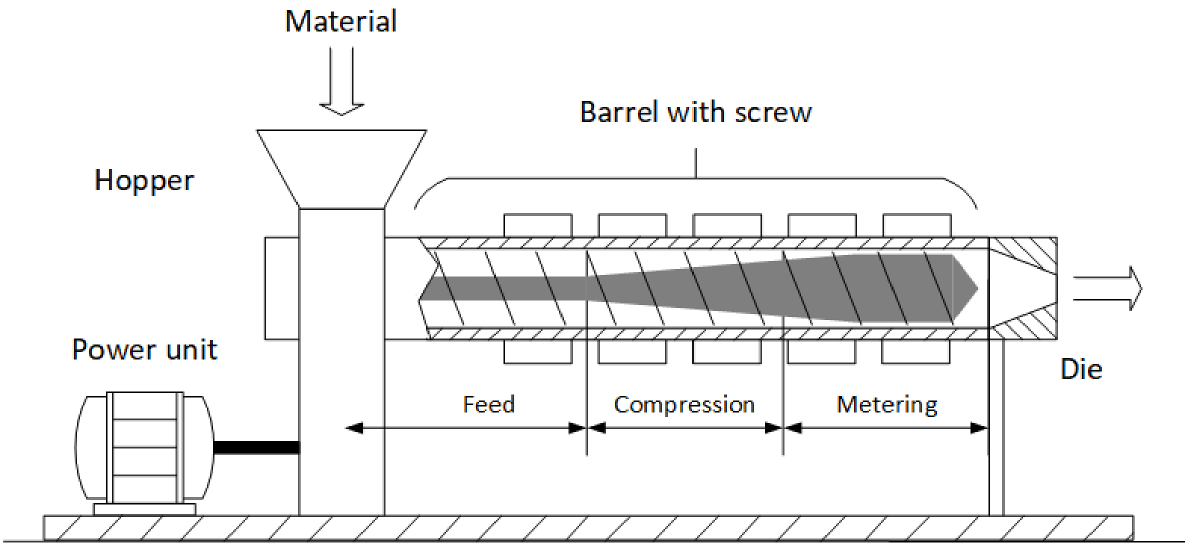


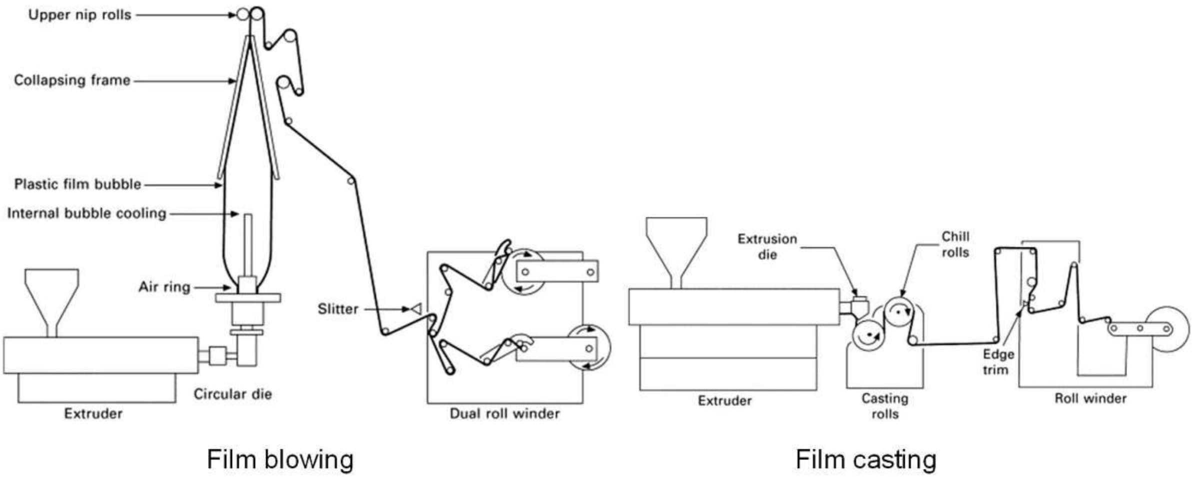
Figure 3: Schematic illustration of an extruder-own illustration based on [54]

Extruders come in a variety of types and sizes and are used to melt and mix polymer materials and force them through a die to create a desired shape or profile. The basic components of an extruder include a hopper, barrel, screw, and die (figure 3). The hopper is where the raw materials are loaded into the extruder, while the barrel is the heated chamber where the materials are melted and mixed. The screw is used to convey and mix the materials as they move through the barrel, and the die is used to shape the molten polymer as it exits the extruder. [55]

Extruders can be classified based on their screw design, which can be either single-screw or twin-screw. Single-screw extruders are the most common type and are used for a wide range of applications. They have a single screw that rotates within the barrel and conveys the materials through the extruder. Twin-screw extruders, on the other hand, have two screws that rotate concurrent or counter-concurrent in the barrel to homogenize the material. [51]

Film blowing and casting

After extrusion, there are two options to process the extrudate into films (figure 4), film blowing (tubular process) or casting (flat process).



Film blowing

Film casting

Figure 4: Schematic illustration of a film blowing (left) and a cast film (right) process [56]

Film blowing is commonly carried out by forming a bubble vertically upwards. The molten polymer is extruded from the plasticating extruder and passed through an annular die to form a tube, which is then sealed at the nip rollers. Subsequently, air is introduced into the tube, causing it to inflate into a bubble that increases in diameter and orients the film in the transverse or cross direction. The degree of orientation is determined by the blow-up ratio, which is the ratio of the bubble's diameter to the die's diameter and is limited by the polymer's melt strength. A higher melt strength allows for a higher blow ratio. To cool the polymer bubble below its melting temperature, chilled air is blown onto the film's outer surface. The point where the melt solidifies and crystallization occurs is known as the frost line. It is essential to control the cooling rate carefully to manage the transition from liquid to solid state. A slower cooling rate results in larger crystals, leading to reduced transparency and gloss in the film. The flattened bubble is then transported by nip rolls to the in-line slitters and roll winders at the line's base, resulting in lay-flat tubing. The resulting film is ideally characterized by high tensile strength in the machine direction, a smooth surface, and good tear resistance. [55–57]

At the beginning of the casting process, the molten polymer is conveyed by an extruder to a slot die with an adjustable narrow opening. This opening controls both the flow rate and the initial thickness of the emerging film. To prevent polymer surge through the die, which could lead to uneven film thickness and inconsistent performance, a reservoir is utilized. The extrudate is then deposited onto large, chilled metal rolls to produce either a sheet or a film. If the film or sheet is not stretched, there is no orientation of the material, and its width, thickness, stiffness, and barrier properties remain unchanged as determined by the slot die. For sheet applications, the plastic melt is often extruded onto a temperature-controlled three-tier calender stack instead of a chill roll, which smooths out the sheet's surface and can provide a textured finish if desired. The calendered sheet is subsequently cooled by passing through several chill rolls or a quench tank before being wound up for shipment or further conversion, such as thermoforming. [51, 56]

Multi-layer films

The manufacturing of multi-layer films can either be realized by a direct coextrusion process or subsequent lamination of the individually produced layers to produce a structure commonly comprising 3-9 different polymer layers (figure 5). Since most polymer melts cannot be easily combined with each other due to structural differences, the use of adhesives as an intermediate tie layer is necessary. They play an essential role in ensuring that the individual layers bond together seamlessly to create a strong and durable film. Coextrusion adhesives can come in various forms, including hot melt adhesives, solvent-based adhesives, and water-based adhesives. They can be applied to one or both surfaces of the polymers to be bonded, depending on the specific application requirements. [58]

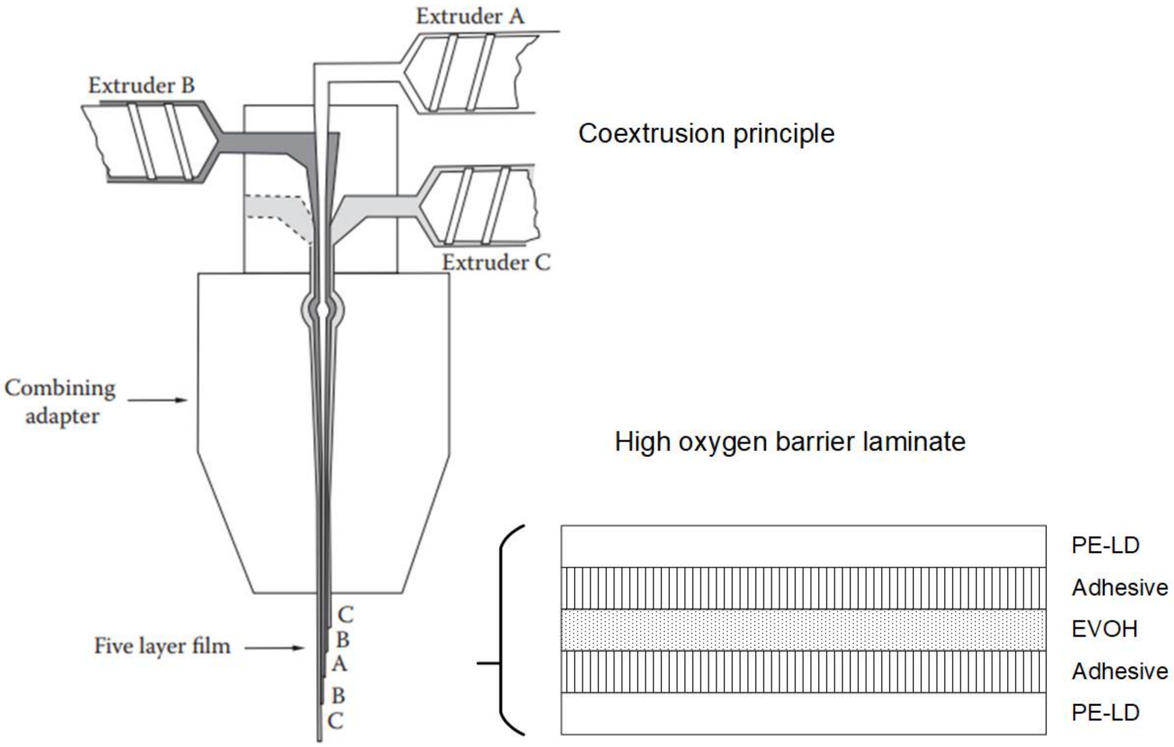


Figure 5: Manufacturing of multi-layer films - own illustration based on [55, 56]

Direct coextrusion can be realized within blown-film production lines as well as cast-film production lines. Accordingly, the polymers are combined in the molten state, thus several extruders are used simultaneously, with a separate extruder being used for each polymer type. In cast extrusion, each extrudate is directed to the so-called feedblock, which combines all supplied polymer streams into a single stream before the mass is pumped through the slot die. The slot die is referred to as a single-manifold die when only one stream flows through it. Through the combination with a feedblock films with multiple polymers can be produced. There are also other designs, such as the multi-manifold die, through which numerous streams can flow. With or without a pre-installed feedblock, a wide variety of multilayer films can be produced. In film-blowing processes, coextrusion is commonly realized with multi-manifold dies. Since the extrudates have a shorter flow path to the die exit, the film layers subsequently exhibit less distortion at the interfaces. [4, 59]

Lamination makes it possible not only to bond polymer layers together but also to incorporate layers of aluminum, paper, or cardboard, which is not feasible in the coextrusion process. In lamination, a distinction is made between extrusion lamination and adhesive lamination. In extrusion lamination, the extrudate is applied to a carrier material, which is often referred to as a substrate. The substrate, such as polymer film or paper board, is continuously unwound from a roll and brought into contact with the extrudate. If necessary, the carrier material is pre-treated (flame, ozone, or plasma treatment) to ensure sufficient adhesion of the materials at the interfaces. Adhesion lamination describes a method for bonding different layers to each other in a roll-to-roll process. The layers to be bonded have already been produced and wound onto rolls in a previous step. In adhesion lamination, these rolls are unwound, and one or both layers are coated with an adhesive at the contact surface and bonded together. Subsequently, the adhesives are usually cured with heat, moisture, UV radiation, or electron radiation. Adhesion lamination is generally divided into dry lamination, wet lamination, and UV curing processes. [48, 56]

Thermoforming

The thermoforming process starts by heating a sheet of uniform thickness, which can be a mono-material, a coextrusion, or a laminate. The sheet is then shaped by drawing it over or into a mold to create a rigid or semi-rigid shape. Once the desired shape is formed, the excess material is trimmed off, leaving a rim around the finished product. However, as the depth of the object being formed increases, there is a higher risk that the material may thin out or even break. This is a significant disadvantage of the thermoforming process, especially for complex shapes that require greater depth. Furthermore, a vacuum can be employed to facilitate the process, particularly when the overall wall thickness is crucial. [51]

Various techniques can be used to shape the product. The traditional approach involves draping the heated sheet over a cavity or plug mold, drawing a vacuum, and molding the sheet to fit the shape of the mold. This method is suitable for shallow thermoformed packaging components with simple designs. However, if the depth exceeds the diameter, plug-assist vacuum forming is the better option. In conventional vacuum thermoforming, there is a risk that the sheet will not conform well to the contour of the mold, particularly in the bottom edges, as the sheet is formed and the wall thins. Plug assist helps to alleviate some of these issues by serving as a heat sink and displacing the material more evenly, thus reducing the thinning of the wall section (figure 6). [56, 60]

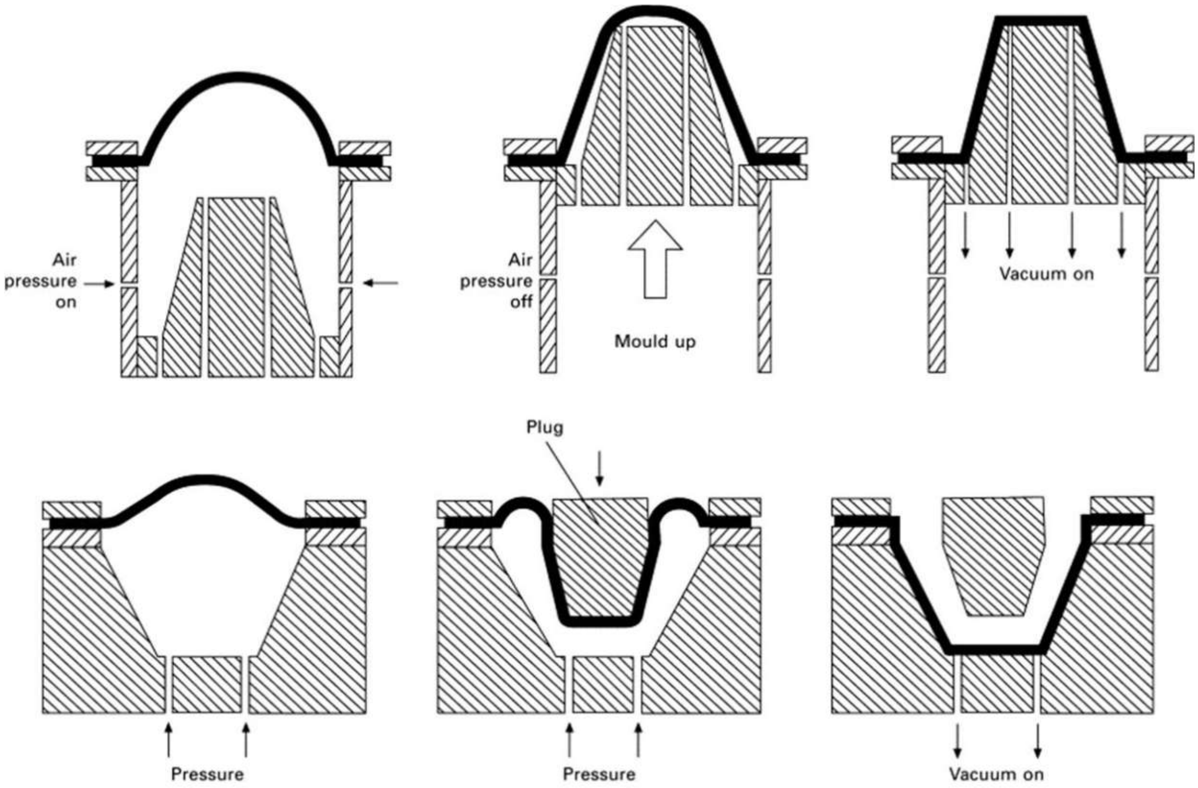


Figure 6: Vacuum assisted thermoforming process with and without a plug [56]

To prevent deformation, the molding must be cooled before being removed from the mold. This step is particularly critical when thermoforming polypropylene, which has a high shrinkage rate. Without sufficient cooling, the material can continue to shrink for several days after being removed from the mold. Cooling is achieved by directing temperature-controlled water through cooling channels integrated into the tooling. After the article has been formed and cooled, it must be cut from the sheet. The cutting process results in a rim around the article that can be utilized for heat sealing. [56]

Blow molding

Blow molding (figure 7) is commonly used for the production of plastic bottle packaging (e.g., in the cosmetics industry). In this specific molding process, the plastic mass is first extruded through a ring nozzle in a tubular shape. Two halves of a blowing mold then enclose the tube and squeeze the drainage area at the bottom, resulting in a seam at the bottom of the product. A sizing mandrel is inserted into the top opening of the product, and the polymer is pressed against the walls of the two halves of the mold by an increase in pressure. The material then cools down and solidifies in the desired shape. After production, the two mold halves open, and the hollow body is released. Additional post-processing may be required, such as removing excess material from the top and bottom or checking the hollow body for leaks. [61]

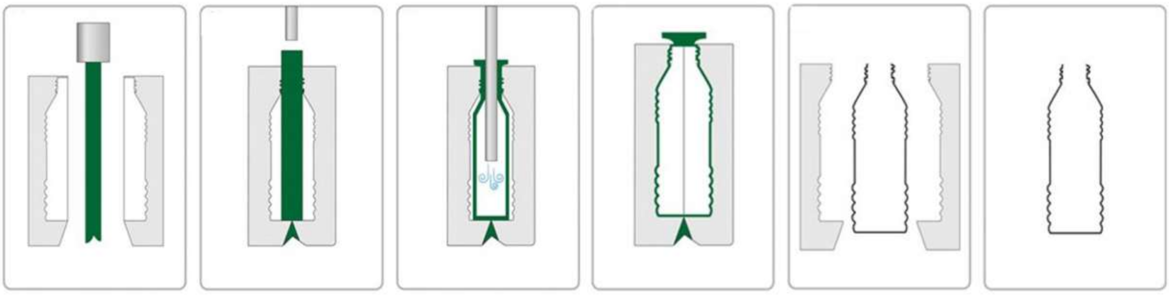


Figure 7: Bottle blow molding process [61]

Foaming

Polymer foaming processes involve the use of a blowing agent, which is added to the polymer in its molten state. The blowing agent can be a physical or chemical agent, and it generates gas bubbles within the polymer matrix, and the gas bubbles expand and form a cellular structure, creating the foam. The foam structure is then set by cooling the material, and it is stabilized by the solidified polymer matrix. Foaming processes can be carried out using various techniques, such as extrusion foaming. Polymer foaming can also be tailored to produce different foam structures, including closed-cell, open-cell, and semi-closed-cell foams. Closed-cell foams have dense cell walls, which prevent gas and liquid from passing through them. Open-cell foams have interconnected cells, allowing gas and liquid to flow through them. Semi-closed-cell foams have partially closed cells, which provide a balance between the properties of closed and open-cell foams. [62]

Injection molding

The injection molding process (figure 8) is capable of producing highly accurate thermoplastic parts from polymer granules. After polymer melting in an extruder, the liquid polymer is then injected into a mold through an injection point, which is known as a gate. The amount of polymer required to fill the mold is precisely controlled by the reciprocating screw or ram piston in the injection molding machine. The mold is made up of two or more steel parts. One of these parts includes a cavity that corresponds to the desired shape, finish and thickness of the molded part. The other part has a corresponding profile that fits into the cavity. To keep the mold closed until the part has cooled and can be ejected, the two parts are clamped together with a force that is strong enough. Multi-cavity molds are utilized to increase production output and reduce costs. The number of cavities that can be used depends on the size of the cavity and the locking force of the injection molding machine. [56, 63]

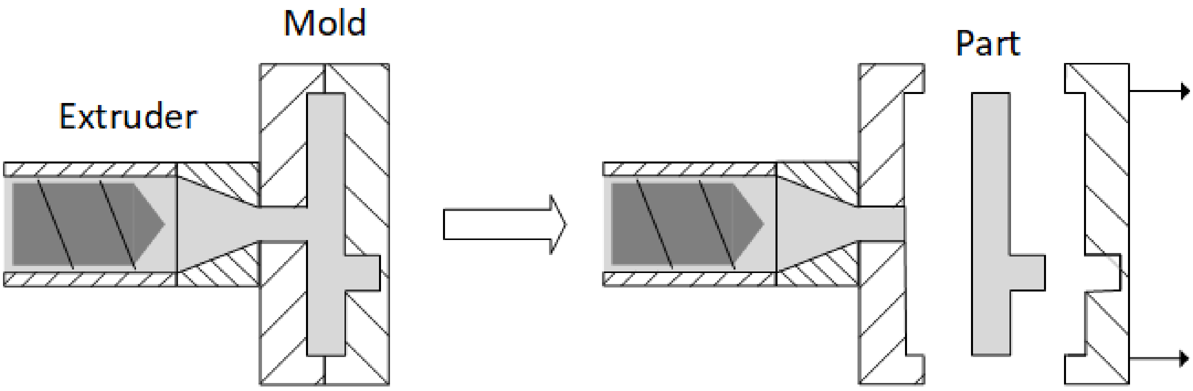


Figure 8: Injection molding process - own illustration based on [64]

Even though the processes are similar and lead to similar products, thermoforming, and injection molding have fundamental differences. Thermoforming requires more material than injection molding due to wall thinning, resulting in waste during cutting. However, tooling costs for injection molding are commonly higher due to accuracy and pressure requirements. Multi-layer film part production for thermoforming is simple, while it is still under development for injection molding but, thermoformed moldings lack the dimensional accuracy of injection molded items. [56]

Decoration and surface finishing

Polymer packaging offers a variety of decoration options to enhance its visual appeal and brand recognition (figure 9). These options include printing, labeling, embossing, hot stamping, and holographic effects. Printing can be done using different techniques such as flexography, gravure, digital, or screen printing. Labels can be applied using pressure-sensitive, heat-shrinkable, or in-mold techniques. Embossing and hot stamping add a tactile effect to the packaging, while holographic effects provide a striking visual impact. The choice of decoration depends on the packaging material, design, and intended use, as well as the desired level of branding and shelf appeal, which are widely driven by marketing aspects. [3]

Printing on packaging material can be realized through direct or indirect methods, as well as stencil printing. The basic principle involves transferring ink from an engraved plate to the substrate through contact, with indirect printing using an intermediate rubber blanket. Stencil printing involves passing ink through a stencil onto the substrate. For polymers such as PE, PET and PP high quality printing is only possible after surface treatment. Due to their non-polar surfaces, adhesion between the ink and the polymer is not possible. Chemical, electrical, flame, and solvent treatments are some of the techniques used to oxidize the surface of these plastics for better ink adhesion. Further, printing techniques can be divided into direct and reverse printing. Reverse printing is a printing technique where the ink is applied to the inner side of the material and does not come into contact with human skin, while in direct printing processes, the packaging surfaces are directly printed. [48, 65]

Figure 9: Decoration options for yogurt cups - own illustration

Labeling serves as a communication tool for packaging, providing consumers with information on nutritional content, net weight, product usage, and other relevant details. By incorporating unique branding, labeling can act as a subtle marketing tool, and by incorporating bar codes, it can also aid in identification during check-out. Although printing technologies are very common for many packaging products, some require labeling. Product labeling also allows for the combination of different material types, such as paper labels on polymer bottles. The most basic form of labels is the glued-on type, which is made up of sheet material (usually paper) that has been printed and trimmed to the appropriate size. The label is secured to the package using an adhesive, which is either applied during the labeling process or activated with moisture just before application during the manufacturing process. This labeling method is commonly utilized for high-volume products like beer, soft drinks, wines, and canned foods where a quick application is necessary. [55,66]

In-mold labeling (IML) is a decoration method used for blow-molded bottles, injection-molded or thermoformed containers, and cups. Polymer IMLs are commonly made from PE and are compatible with various polymer containers. What sets IMLs apart from traditional glue-on labels is the application of a heat-seal coating on the back of the IML stock during manufacturing. Labels made from film offer superior heat, moisture, and chemical resistance compared to those made from paper. During the IML process, a label is placed in the open mold and held in position using vacuum ports, electrostatic attraction, or other suitable methods. The mold closes, and molten plastic resin is injected into the mold, taking on the shape of the object. The hot plastic surrounds the label, incorporating it as a fundamental part of the molded object. [67]

Shrink sleeves are a type of label made from a thermoplastic polymer film that is printed and then shrunk to fit the shape of the container through the application of heat. These labels provide complete 360-degree coverage of the container, allowing for more extensive branding and design opportunities compared to "traditional" labels. [68]

Additional surface optics or film protection can be realized through various coating possibilities. This technique makes polymer packaging suitable for applications that are commonly dominated by glass or aluminium. To achieve the necessary optical clarity, scratch and abrasion resistance, and weather exposure (depending on the application), polymer parts require a coating or multiple coatings. Metalizing enables the utilization of polymers in place of metal components and also enhances decorative possibilities. Coating systems are categorized as either thermoplastic or thermoset. Thermoplastic systems undergo only a physical change during the curing process, with the film hardening over time due to the loss of solvent. Conversely, thermoset systems undergo a chemical reaction during curing, which is triggered by thermal energy, radiation, or oxidation. Polymer coatings can be made from a wide range of materials depending on the specific application requirements, including polyurethanes, epoxies, silicones, and acrylics. [67]

1.2.3 Functional requirements for packaging

Functional requirements for packaging are the specific characteristics and features that a packaging solution must possess in order to effectively protect, preserve, and transport the product it contains. These requirements are determined by the product being packaged and the conditions it will encounter throughout its journey from manufacturing to distribution and ultimately to the end user. Those functional performance parameters have to ensure consistency throughout production and must undergo strict quality control. [48]

Controlling the thickness of the film is one of the critical factors in ensuring consistent heat sealing, printing, and mechanical strength. However, measuring the thickness of the film is a challenging task, as individual points will vary significantly. In addition to controlling the film thickness, it is crucial to measure the WVTR accurately and at a predetermined temperature and humidity. Similarly, gas barrier measurements, including oxygen, carbon dioxide, and nitrogen, need to be taken at a known temperature and humidity. Moreover, not all polymers have a grease barrier, which is essential for packaging high-fat content products like butter and dry pet foods. The type of fat also plays an essential role in determining the effectiveness of the barrier. [3, 4]

The coefficient of friction (CoF) is another critical attribute of film that impacts packaging functionality. The lower the CoF, the more slippery a surface is, and it is essential to have a constant CoF within the film to ensure smooth movement over packaging machinery. Heat sealability is another important attribute for most packaging applications. The ideal multilayer film for heat sealing should have a significantly higher sealing temperature on the outer part of the film than the inner, with the inner part of the film having a wide sealing range. In some cases, a cold-seal adhesive is used, such as when packaging a chocolate bar. [56, 68]

Film stiffness is another critical attribute that impacts packaging functionality, particularly when making bags, sachets, and pouches that need to stand up without sagging. The thicker the material, the greater the stiffness, but this also increases the cost. Therefore, for many applications, stiffness at the lowest thickness is a preferred attribute. Adequate puncture resistance is another essential packaging property, especially when packaging items with sharp edges. Low puncture resistance can result in holes in the film, significantly reducing the effectiveness of any barrier that has been engineered into it. Ensuring packaging functionality requires careful consideration of several factors, and a thorough understanding of these attributes is critical to achieving the desired packaging outcome. [56]

1.3 Recycling perspectives

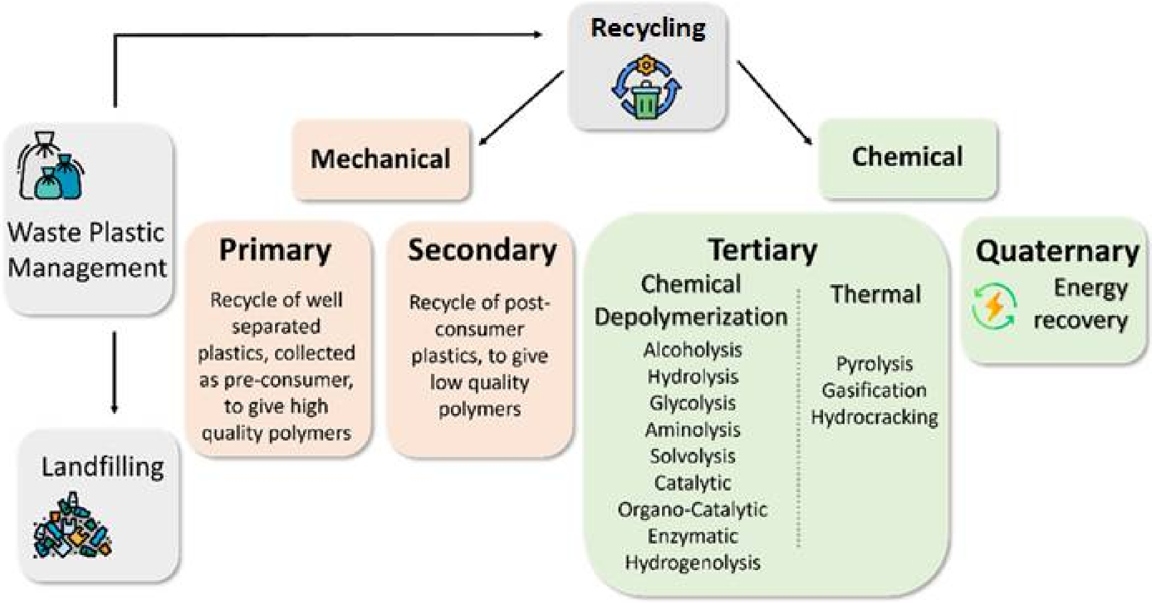


Figure 10: Polymer recycling techniques [69]

The disposal of polymers and especially short shelf life polymer products such as packaging poses a significant challenge as they are commonly non-biodegradable and can persist in the environment for centuries. There are several perspectives on recycling of polymers to prevent landfilling, which include mechanical, chemical, thermal, and biological recycling (figure 10). [69]

There are two primary recycling methods: "closed-loop recycling" and "open-loop recycling". Closed-loop recycling involves recycling plastic without significantly changing its inherent properties, allowing it to be used in the same application as the original material, such as bottle-to-bottle recycling. On the other hand, open-loop recycling involves changing the inherent properties of the recycled plastic, making it unsuitable for the original application but usable for other plastic product manufacturing, such as bottle-to-fiber recycling. The appropriateness of each approach depends on various factors, including the level of contamination in the plastic waste, the properties of the polymer, and the specific approval requirements for the application in question. [70]

Within the open- or closed-loop scenarios, several recycling technologies exist. Mechanical recycling, widely known as primary or secondary recycling, involves the physical processing of waste polymers to produce new products. It is the preferred recycling method due to its simplicity, cost-effectiveness, ability to produce high-quality recycled products, reliability, and lower environmental impact compared to other disposal methods. Chemical recycling involves the use of chemical processes to break down polymers into their constituent monomers, which can be used to produce new polymers. This approach has the potential to recycle a wider range of polymers, including complex structures such as multi-layer films. Biological recycling, also known as biodegradation, involves the use of microorganisms to break down polymers into their constituent components. However, this approach is still in its early stages of development and might gain more importance in the future. The thermal recycling process involves the conversion of waste polymers into heat or electricity, which can be used to power industrial processes or feed into the electricity grid. Thermal recycling converts polymer waste into energy. The process typically involves the combustion or gasification of waste polymers, which breaks them down into their constituent components, including carbon dioxide and water. [71–73]

1.3.1 Degradation mechanisms

One aspect that makes polymer recycling challenging in comparison to other materials, such as metals, is that their structure naturally undergoes degradation during their processing and service life. Polymer degradation is the process by which polymers break down into their constituent components due to environmental factors such as heat, light, moisture, and exposure to chemicals. There are several mechanisms of polymer degradation, including thermal degradation, photo-degradation, hydrolysis, oxidation, and biodegradation. [74, 75]

Thermal degradation is a process of polymer degradation that occurs due to exposure to high temperatures (e.g., extrusion). It involves the breaking of chemical bonds within the polymer, leading to the formation of smaller molecules such as monomers, oligomers, and gases. The process of thermal degradation can be accelerated by the presence of oxygen, which promotes the formation of free radicals that initiate the degradation process. [76]

Photo-degradation describes the degradation due to exposure to light, particularly UV radiation. UV radiation can break down the chemical bonds within the polymer, leading to the formation of free radicals that can initiate the degradation process. This can result in changes in the physical properties of the polymer, such as brittleness, discoloration, and loss of mechanical strength. [77]

Biodegradation is the degradation of polymers by microorganisms such as bacteria and fungi. Biodegradation involves the breaking of chemical bonds within the polymer by the action of enzymes produced by microorganisms. Biodegradation is a natural process that can occur in the environment, but the rate of biodegradation depends on various factors, such as the type of polymer, the environmental conditions, and the presence of microorganisms capable of degrading the polymer. [78]

Polymer degradation caused by exposure to water or other aqueous environments is known as hydrolysis. This process involves the breakdown of chemical bonds within the polymer by a hydrolysis reaction, which results in the formation of smaller molecules such as monomers and oligomers. The presence of acids, bases, or enzymes can accelerate hydrolysis by promoting the reaction. [74]

Oxidation describes the degradation of polymers due to exposure to oxygen, which can lead to the formation of free radicals that can initiate the degradation process. The oxidation process can be accelerated by exposure to heat or light and typically results in changes in the physical properties of the polymer, such as loss of mechanical strength or changes in color. [76]

Polymer degradation mechanisms are complex and depend on various environmental factors such as heat, light, moisture, exposure to chemicals, and the presence of microorganisms. Due to their various chemical structures, polymers commonly demonstrate opposed degradation behavior under the same environmental influence factors. If exposed to thermo-mechanical stress, PP tends to chain scission (loss in viscosity), while PE typically undergoes branching reactions (increase in viscosity) which has to be considered during reprocessing. Understanding these mechanisms is essential for the development of viable recycling techniques to produce homogeneous secondary materials of high quality. [79]

1.3.2 Mechanical recycling process

Mechanical recycling is a process that involves a combination of various processing steps, including collection, identification, sorting, grinding, washing, agglomerating, and compounding (figure 11). One major advantage of mechanical recycling is that it can be implemented in a decentralized manner. The recycling plants are simple and cost-effective and require relatively low energy and resource inputs compared to chemical recycling plants. While optimization of the individual processing steps has the potential to partially improve the properties of the resulting material (polymer recyclate), such as smell, purity, and color, the quality of the recyclates widely depends on the quality and purity of the input stream (plastic waste). [70, 80]

Another critical aspect that makes polymer recycling very challenging is the fact that different types of polymers are not compatible with each other. When different polymers are mixed together during recycling, they will likely form blends with different physical and chemical properties compared to the original materials. Incompatibility between polymers can arise due to differences in their chemical structures, molecular weights, and processing conditions. For example, some polymers may be hydrophobic (repel water), while others are hydrophilic (attract water). When these two types of polymers are mixed, they can separate into distinct phases, which weakens the resulting material. The formation of such blends can especially not be avoided with multi-layer packaging, as the individual layers cannot be easily separated during the recycling process. This problem highlights the importance of waste stream purification during the mechanical recycling process. [81, 82]

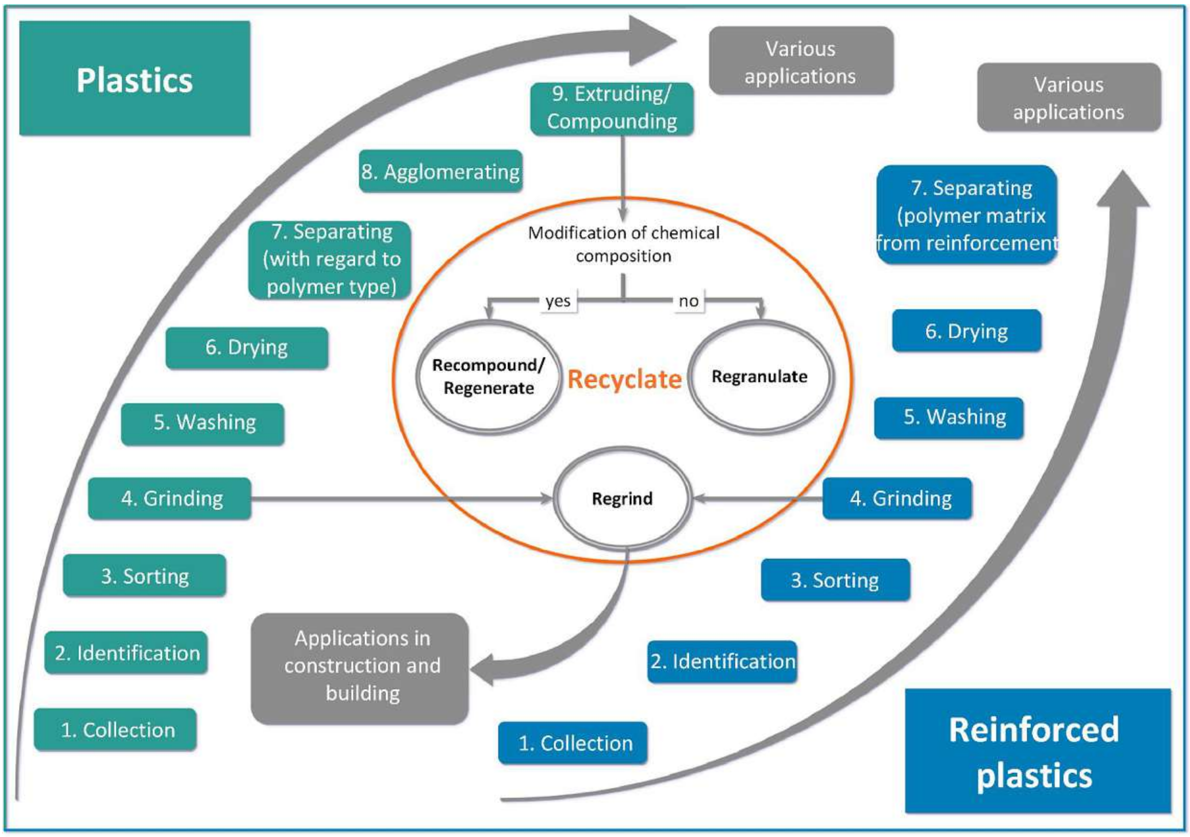


Figure 11: Steps and output of the mechanical recycling process [70]

Collection

There are no universally applicable guidelines for the collection of plastic waste. Some countries have stringent guidelines regarding the separation and collection of waste (e.g., Japan), while others are very liberal. Ultimately, the differences can even extend to locally implemented collection points (municipal level in Austria). Collection systems can be generally divided into two categories, single-stream recycling, and dual-stream or multi-stream recycling. With single-stream recycling, household waste is disposed of in a single waste container, whereas with dual-stream or multi-stream recycling, households separate their waste into at least two types of waste. [83, 84]

It is common to separate at least paper, plastics, aluminum, organic waste, and residual waste. In Austria, from 2023 onwards, plastics and aluminum will be collected uniformly together in blue-yellow bins across the whole country. Therefore, the self-responsibility of companies and households plays a significant role in disposing of waste as sorted, clean, and dry as possible. Public or private waste disposal companies transport the collected household waste to a waste sorting facility. [85]

Identification, sorting and grinding

The quality of the delivered waste is strongly dependent on the type of collection system (single-stream or multi-stream), but operators of sorting plants are often also waste collectors and can thus influence the type of collection. The first step in the recycling plant (tearing of collection bags) serves to break down the waste into its components, loosen the collected material, and prepare it for transport on conveyor belts. Single-shaft shredders, which are operated at low speed, have established themselves as collection bag openers. [85]

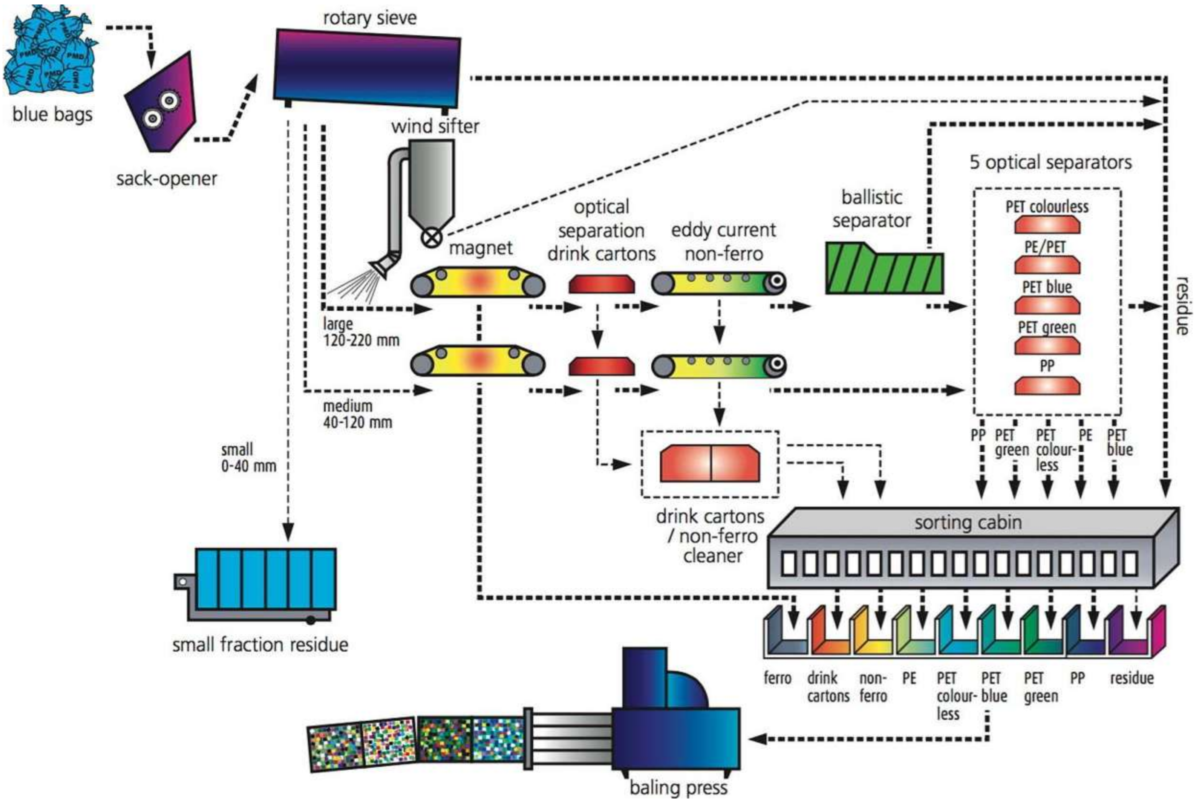


Figure 12: Identification and sorting technologies for municipal waste [86]

Following the opening process, the first sorting process step, also called classification, takes place. The waste material is conveyed into a rotary sieve and using sieving technology, mass flows with different particle sizes (<40 mm, 40-120 mm, 120-220 mm) are generated. The material stream with the smallest components (<40 mm) is rejected and not used for further processing. The material flow with the largest elements (120-220 mm), as well as the main mass flow (40-120 mm), are fed to an air classifier system. [86]

The air classifier system uses a combination of airflow and mechanical vibration to separate the different types of polymers. The airflow within the classifier creates a series of air streams that move the polymer particles through the machine. The particles are sorted based on their size and shape, with larger and heavier particles settling first and smaller and lighter particles moving further through the machine. The separated materials are collected in different compartments or bins. [87]

Air classification is commonly followed by magnetic sorting and eddy current sorting (figure 12). The former is used to separate ferromagnetic metals (iron-carbon alloys) from the material stream. This is achieved using an electromagnet or permanent magnet that creates a magnetic field with which the magnetizable components are attracted and separated. Eddy current sorting is used to sort out non-ferrous metals, especially aluminum (beverage cans, food cans, coffee capsules, and inner coating of liquid cartons). This separation principle requires that the iron metals have already been removed from the waste stream, otherwise the ferromagnetic particles would stick to the conveyor belt, which is placed above the magnet. [88]

The remaining polymer-based waste stream then undergoes ballistic separation, which is used to divide the stream into rigid (bottles, cups, containers) and flat (films) components which are usually incinerated. The rigid polymer products are further separated by optical methods such as FT-NIR (Fourier Transform Near Infrared), optical color recognition sensors, and manual sorting by trained operators. The separated fractions are either sold for further recycling and purification or shredded and processed into regrind. [70, 89]

Regrind recyclates are frequently utilized in building and construction applications as a substitute for sand aggregates and fillers. Both polymers and fiber-reinforced polymer composite regrinds are incorporated into self-compacting concrete and mortar. This method has great potential for lightweight constructions. The utilization of recycled HIPS and PE-LD as sand replacement in concrete has shown a decrease in workability, density, and compressive strength with an increase in the amount of recycled polymers. [90–92]

Washing and drying

Washing technologies are an essential aspect of the advanced polymer recycling process, and they are used to remove contaminants and impurities from waste polymers. Hot washing involves the use of hot water or steam to clean the waste polymers. The temperature used is typically between 80-100°C. The hot water or steam is used to dissolve and remove contaminants such as adhesives, labels, and inks. The waste polymers are placed in a washing tank, and the hot water or steam is circulated through the tank to remove the impurities. The wastewater from the hot wash process is then treated to remove any remaining contaminants before being discharged. [93]

Cold washing, on the other hand, uses cold water or solvents to clean the waste polymers. The cold water or solvent is used to dissolve and remove contaminants such as dirt, dust, and grease. Both hot and cold washing methods have their advantages and disadvantages. Hot washing is more effective in removing contaminants such as adhesives, labels, and inks, but it requires more energy and can be more expensive. Cold washing is less effective in eliminating specific types of contaminants, but it is more energy-efficient and can be less expensive. Subsequent drying completes the washing process. As washing of polymer waste has less impact on the mechanical material properties but influences smell and color, the recyclate application will mostly justify the degree of purification. [94–96]

Further processing and compounding

Depending on to what degree the plastic waste has already been separated (PET/polyolefin, polyolefins alone, all mixed), it may be necessary to add some further sorting steps for division into individual polymer types. For instance, using NIR scanners, which can detect individual polymer particles over a large area, or float-sink (flotation) processes for density separation. [70]

Flotation, which utilizes water as a flotation agent, is the primary technique for sorting shredded flakes. This cost-effective method separates polymers with densities below $1 g/cm^3$, such as unfilled PP and PE, by causing them to float while the other common polymers like PS, PET, and PA will sink. It is possible to use denser media than water for flotation to achieve further separation of the sink fraction. However, some polymers found in post-consumer waste have a density range rather than a single-density value. Moreover, these density ranges often overlap (figure 13), making it difficult to achieve complete separation of these polymers into mono-streams. [86, 90]

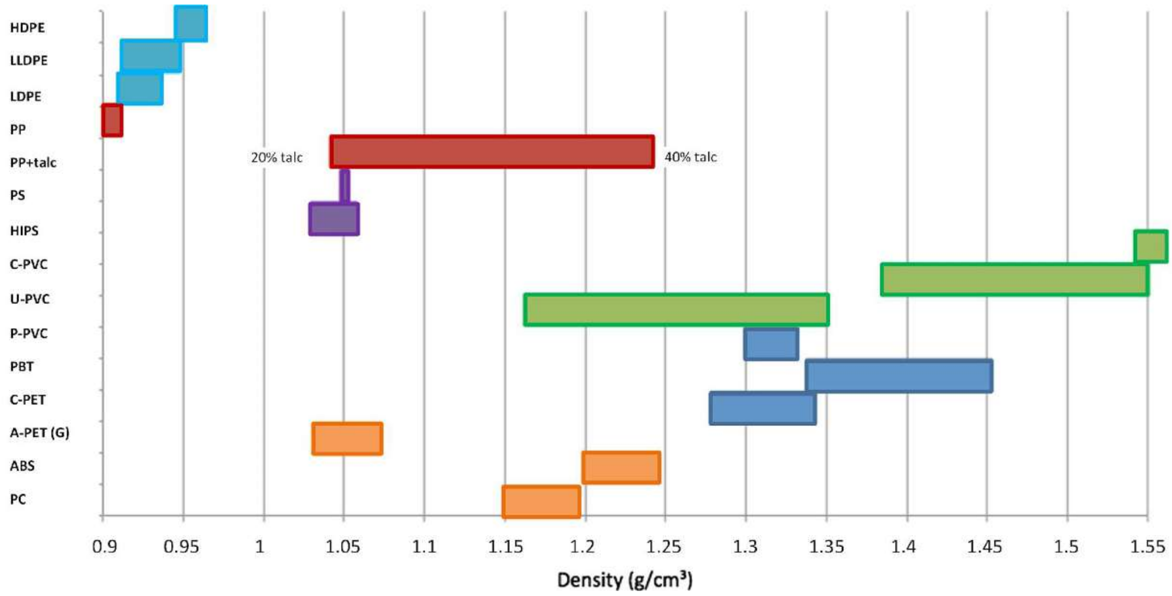


Figure 13: Density ranges for common waste polymers [86]

Regranulate or regenerate is then produced via extrusion. During the extrusion of the recycled materials, melt filtration is a useful technique to remove remaining non-melting contaminations from the melt, as these would inevitably reduce the quality and properties of the recyclate. Typically removed fractions include small wood, paper, aged rubber particles, and higher-melting polymers (e.g., PET in PP processed at 220 °C). Melt filters come in different mesh sizes. A smaller mesh size takes out more contaminations, and is more complex in production but it will also lead to improved process stability and polymer quality. [70, 86]

Without further modification of the composition through chemicals, the recyclate is called regranulate. Chemical treatment can be used to successfully prevent or reduce down-cycling. Specifically, in cases of unintentional mixing of two or more polymers, such as in multi-layer packaging, compatibilizers can increase the compatibility of the polymers with each other. Commonly reactive and non-reactive compatibilizers are available. During the extrusion of PET, reactive copolymers of styrene, glycidyl methacrylate, and butyl acrylate are utilized to extend the polymer chain, resulting in an almost 100% increase in tensile strength. Peroxide-induced long chain branching offers the possibility to increase the melt strength of PP, which makes it suitable for film-blowing or foaming processes. Other modification options include stabilizers (prevention of degradation), lubricants (reduction of friction and shear stress), anti-statics (prevention of dust accumulation), and impact modifiers (increase of toughness and impact resistance). Those modified recyclates are classified as recompounds or regenerates. [70, 97–99]

Ideally, the concept of closed- or open-loop recycling involves the preservation of a polymer material's inherent properties not only after the initial recycling process but also after multiple recycling processes. However, with mechanical recycling, it's only feasible to maintain the plastic's properties through a few recycling loops. This constraint is due to the breakdown of the polymers' molecular structure caused by high-temperature and high-pressure extrusion processing, resulting in shear but also reinforced by contaminants that lead to the formation of unintentional material mixtures. Nevertheless, to keep the cycle alive as long as possible, the generation of purified mono-material fractions during the recycling process plays an essential role. [70, 100]

1.3.3 Challenges, solution approaches and design for/from recycling aspects

Design for recycling and design from recycling are two critical aspects of polymer product design that are closely connected. Design for recycling involves designing products with the end-of-life in mind, ensuring that they can be easily recycled and reused. This includes using materials that are easily recyclable, designing products that can be easily disassembled, and avoiding the use of non-recyclable components. By designing products for recycling, the material value can be preserved, reducing waste and the need for virgin materials. On the other hand, design from recycling involves using recycled materials as the primary source for creating new products. This approach requires a thorough understanding of the properties and limitations of recycled materials and involves designing products that can be manufactured using recycled materials, while still meeting performance and quality requirements. The link between design for and design from recycling can help to reduce the environmental impact of the product by reducing the need for virgin materials while also reducing waste and energy consumption. However, it is essential to consider all the issues involved in the mechanical recycling process, as well as those that may arise in connection with different manufacturing processes. [101–104]

Separation limitations

The sorting performance of NIR devices is restricted by various factors. These include high belt speeds and short measuring times, as well as packaging surfaces that are black, dirty, wet, or fully printed and overlapping, and labels that cover over 30% of the packaging and involve different polymers. In addition, NIR sorters have limitations in various sorting tasks, such as distinguishing between food and non-food packaging, sorting brand-specific materials for extended producer responsibility, or identifying certain plastic types, such as polyethylene terephthalate amorphous/glycol-modified (PET-A/G) and polypropylene homopolymer/-copolymer (PP-H/C), and certain multilayer packaging. [105]

The polymer density can be altered in various ways, such as by adding fillers or by expanding the polymers to create foamed materials. These modifications can result in deviations from the typical density values. In addition, multi-layer composite materials, or multi-layers, can also cause changes in density. By incorporating different materials with varying densities into a single layer, the overall density of the multi-layer material can also be altered. These modifications might impact the sorting performance of certain separation technologies that rely on density measurements, such as sink-float separation. [86, 105]

Non-intentionally added substances (NIAS)

Stakeholders are concerned about the potential migration of harmful substances from recycled packaging to the product. This concern is not limited to the food packaging sector but also extends to other industries, such as hygienic packaging. Non-intentionally added substances (NIAS), which are undesired chemicals, have been identified as potential source of contamination. NIAS can arise from the degradation of polymeric materials and their additives, as well as from contaminants that migrate from external sources. Oligomers of the polymer material, degradation products from antioxidant additives, and odorants such as limonene are among the most prevalent NIAS. Additionally, substances that were intentionally added (IAS) during the manufacturing process (catalysts, additives, inks, fillers) might later demonstrate a certain risk for consumers in recyclates. [106–108]

(Im)miscibility of polymers

Polymer blends are inevitably formed within the mechanical recycling of a mixed polymer waste stream, as it is impossible to entirely separate the different types of polymers. A polymer blend contains at least one other polymeric contaminant > 2 wt.%. The miscibility of polymer blends is determined by their thermodynamics. [86]

The simplest explanation of polymer miscibility is given by the Gibbs free energy of mixing (equation 1).

$$\Delta G_{mix} = \Delta H_{mix} - T\Delta S_{mix} \leqslant 0 \tag{1}$$

$$(\partial^2 G/\partial\varphi) > 0 \tag{2}$$

The Gibbs free energy can be expressed as the sum of two components (A, B), the enthalpy (ΔH) of mixing, and the entropy of mixing (ΔS). This relationship is dependent on the absolute temperature, denoted by T. The thermodynamic equilibrium determines whether mixtures of different polymers are (partially) miscible or immiscible. A blend that is completely miscible will form only if the Gibbs free energy is negative and if the criterion in equation 2 is met in addition. In this equation, φ is the volume fraction of component B. The compatibility of two polymers is generally increased as their chemical similarity increases. [109]

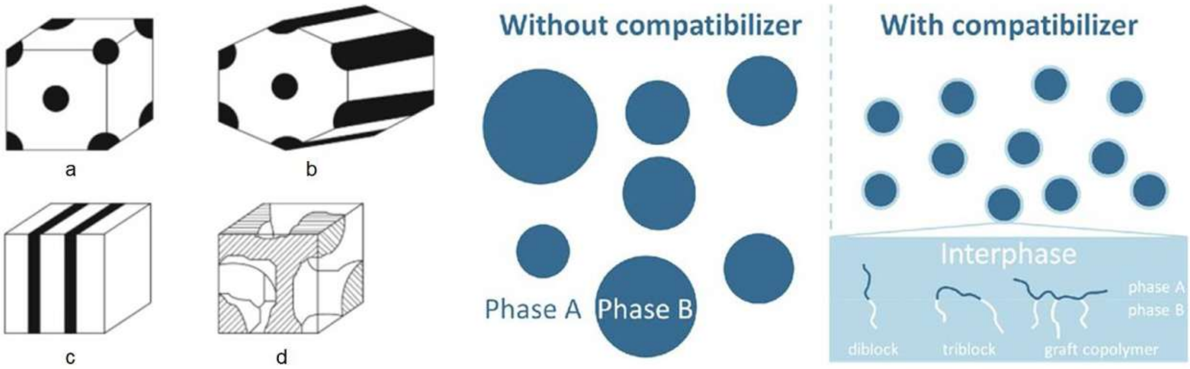


Figure 14: Immiscible polymer blend morphologies (a: droplets, b: cylinder, c: laminar, d: co-continuous) and the examplary effect of compatibilizers [110,111]

Homogeneous blends, also known as completely miscible blends, exhibit a one-phase morphology. In contrast, immiscible blends can display a variety of morphologies, such as spherical droplets, cylinders, fibers, sheets, or co-continuous phases. To realize improved compatibility, macromolecules with different structures can be employed, including diblock, triblock, or graft copolymers. The process of compatibilization typically involves the creation of an interphase that enhances interfacial adhesion and leads to a finer morphology with a smaller average diameter of the dispersed particles (figure 14). [86,111]

Profound knowledge about polymer miscibility and avoidance of the combination of certain polymer compositions in multi-layers such as polyester and polyolefins has the potential to significantly increase product sustainability. [112]

Lack of standardization

Standardization is essential for the sustainable introduction of a global supply chain for polymer recyclates. However, there are currently few international standards [113] for polymer recycling due to differences in economic, political, and legal frameworks. The supply chain for polymer recyclates is mainly based on agreements and cooperation between recyclate suppliers and manufacturers of polymer products. [114–116]

To develop a functioning market for polymer recyclates, material quality, clear responsibilities for recyclate properties, and supply guarantees are prerequisites. Traceability of recycled materials, standardized sampling and characterization methods, and application- and product-specific requirements must be considered. Material properties need to be provided in technical and safety data sheets. Corresponding standards could guarantee the implementation of these requirements and serve as a valuable communication tool for the supply chain. [70, 114, 117]

Performance versus cost

Mechanically recycled plastics are currently the most commonly used recyclates, but using them for industrial applications is disadvantageous due to their lower quality and occasionally higher cost compared to virgin plastics. The most stringent quality requirements are for food-grade recyclates, which must be certified by FDA (Food and Drug Administration) or EFSA (European Food Safety Authority) through a challenge test to ensure they do not pose a risk to human health. However, using mechanically recycled plastics in applications like medicine is currently not possible due to low traceability of plastic waste and lack of high-quality recycling technologies. Improving the quality of mechanically recycled plastics is possible but comes at a higher production cost due to additional processing steps and the use of additives. [70, 118]

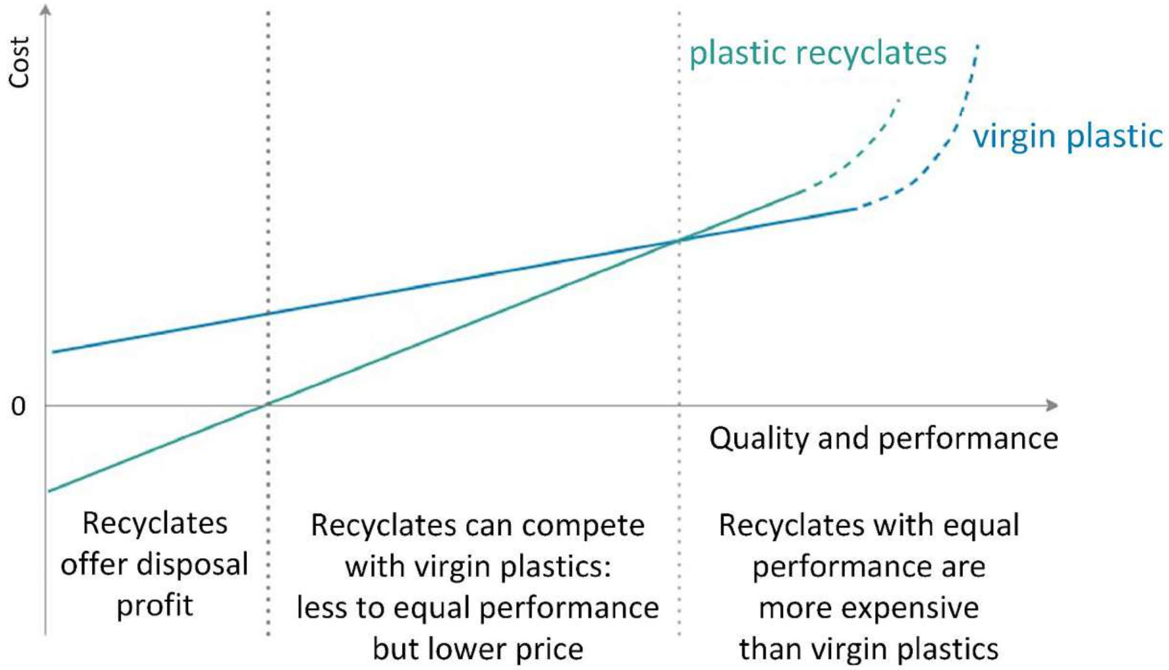


Figure 15: Performance/cost comparison of virgin polymer and recycled polymer waste [70]

Figure 15 depicts a simplified comparison between the quality and cost of mechanically recycled and virgin polymers in three regions. The second region represents a price-performance area where recyclates can compete with virgin polymers in terms of quality but not cost due to the need for additional processing steps and the low cost of virgin polymers. The third region represents high-quality polymers requiring special approval (e.g., food grade), resulting from complex recycling processes where ecological criteria are driving the recycling efforts. The introduction of design for recycling strategies and better pre-sorting of waste streams could simplify the challenge and reduce the processing cost in the second and third regions. [70]

Comprehension of waste management by consumers

Improper disposal of plastic waste is a significant problem, as shown by a study in Germany where 30% of lightweight packaging waste was sent to the wrong recovery route. This is not due to a lack of willpower, as most consumers have a positive attitude towards waste separation education, but rather a lack of understanding. Additionally, packaging distributors are increasingly making consumers responsible for separating packaging, such as requiring the removal of not only the aluminum lid but also the paper band on yogurt cups. The issue of food and other residues remaining in the packaging results in unpleasant odors in recycled material. This is caused by the consumers not fully emptying the packaging, as well as packaging designs that inhibit effective emptying. [105, 119, 120]

Both design for and from recycling strategies require close collaborations between distinct professional fields such as (material) engineers, product designers, and marketing specialists. If a harmonization of those collaborations can be realized, ideally supported by legal guidelines and standards, material circularity of polymer products can be accomplished. [86]

2 Methodology

2.1 Characterization of packaging

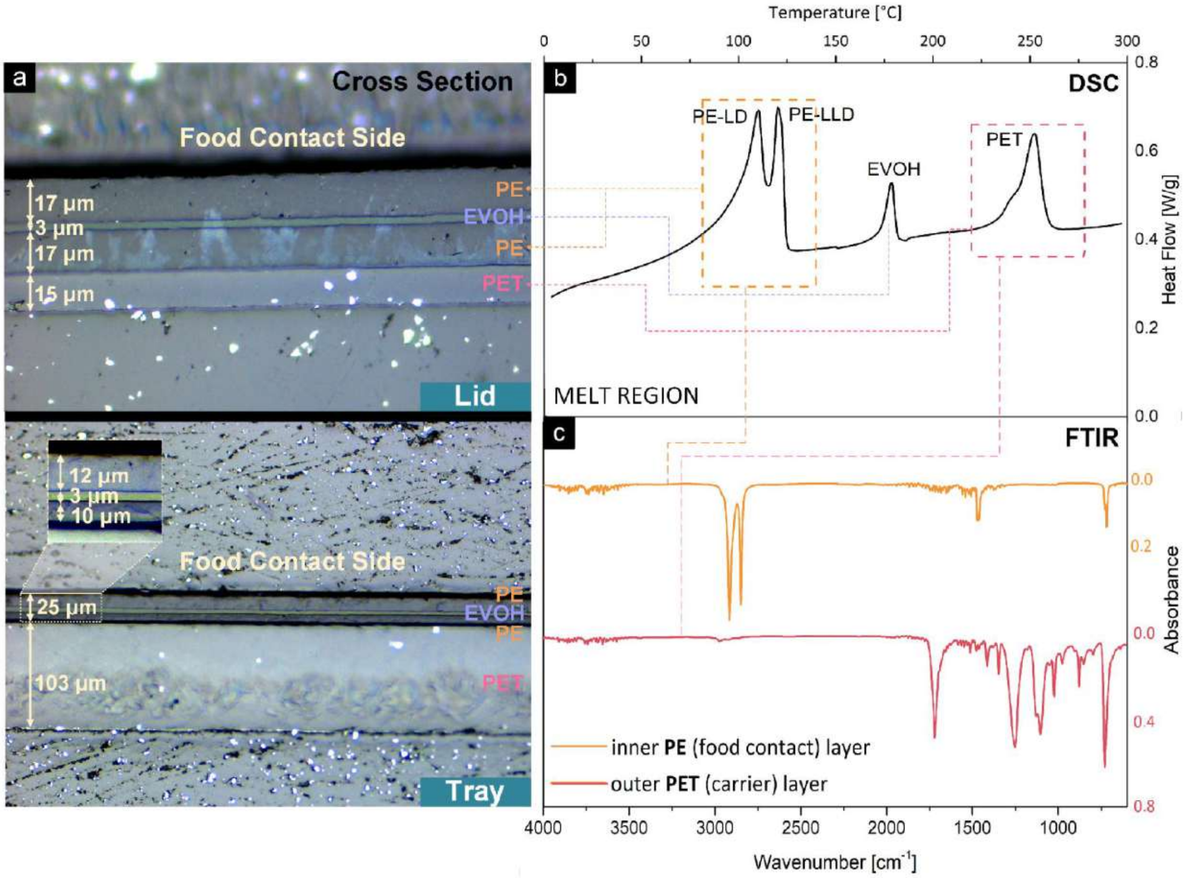


Figure 16: Packaging characterization trough (a) light micrographs of film cross-sections, (b) DSC and (c) FT-IR of inner and outer layer exemplary shown for a MAP lid and tray [121]

The analysis of about 240 different packaging products, which were collected from waste bins in eastern Austria between 2018-2023, was realized with a combination of Differential-Scanning-Calorimetry (DSC), Fourier-Transform Infrared spectroscopy (FT-IR) and light microscopy (figure 16). FT-IR revealed the outer layers, while characterization of all thermoplastic materials within the structure was possible by DSC (analysis of T_m). Additionally, the number of layers and their thicknesses were analyzed using light micrographs. For mono-layer packaging, DSC or FT-IR alone is suitable for the material characterization. The percentual mass proportions of the individual polymers were calculated using volumetric mass density. [122]

2.1.1 DSC

During DSC analysis, the heat flows of a material sample and a reference (air) are measured as a function of temperature. The resulting difference between the sample and reference can be used to determine parameters such as glass transition temperature(T_g), crystallization temperature(T_c), melting temperature (T_m), and also the degree of crystallization. In the so-called heat flow principle, the sample and reference are located in the same oven and subjected to the same temperature program (figure 17). [123]

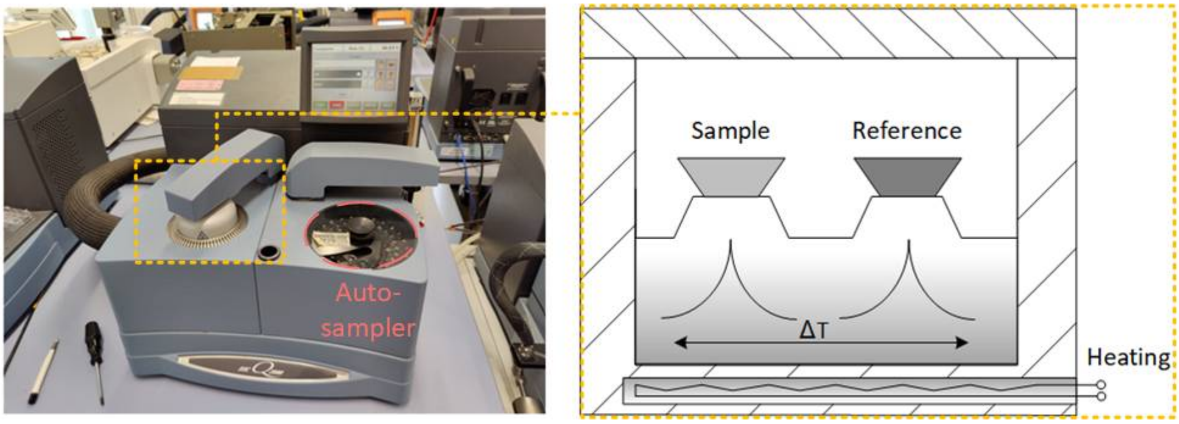


Figure 17: DSC testing device (left) and DSC measurement principle (right) - own illustration based on [124]

The temperature of the sample and reference is continuously measured. Due to temperature reactions of the material, such as crystallization or recrystallization, glass transition or melting, a temperature difference ΔT is created. This difference subsequently results in a non-constant heat flow. Heat flow $\dot{Q}$ and enthalpy change ΔH are determined by incorporating a constant heating or cooling rate ν, the sample mass m, and the specific heat capacity c_p according to the equations 3 and 4. [125]

$$\dot{Q} = m \cdot \nu \cdot c_p \tag{3}$$

$$\Delta H = \int c_p \cdot dT \tag{4}$$

After positioning the sample with a weight of 2-40 mg and reference in the oven, they are usually subjected to a temperature program consisting of heating, cooling, and heating again. Heating twice is necessary because any impurities or processing influences may become visible in the first heating cycle, which will only disappear after complete melting and crystallization. [123]

The measurements were carried out using a Q2000 instrument (TA Instruments) according to DIN EN ISO 11357 [126]. Standard aluminum pans were used for sample preparation. Between 5 and 6 mg of each material were weighed and measured between 0-300 °C with heating and cooling rates of 10K/min.

2.1.2 FT-IR

FT-IR spectroscopy is a non-destructive method for the characterization of materials. The technology is based on the principle that molecules of a substance are set into vibration by electromagnetic waves in the infrared range. A part of the incoming radiation passes through the sample unhindered, while the rest is absorbed or reflected. [127]

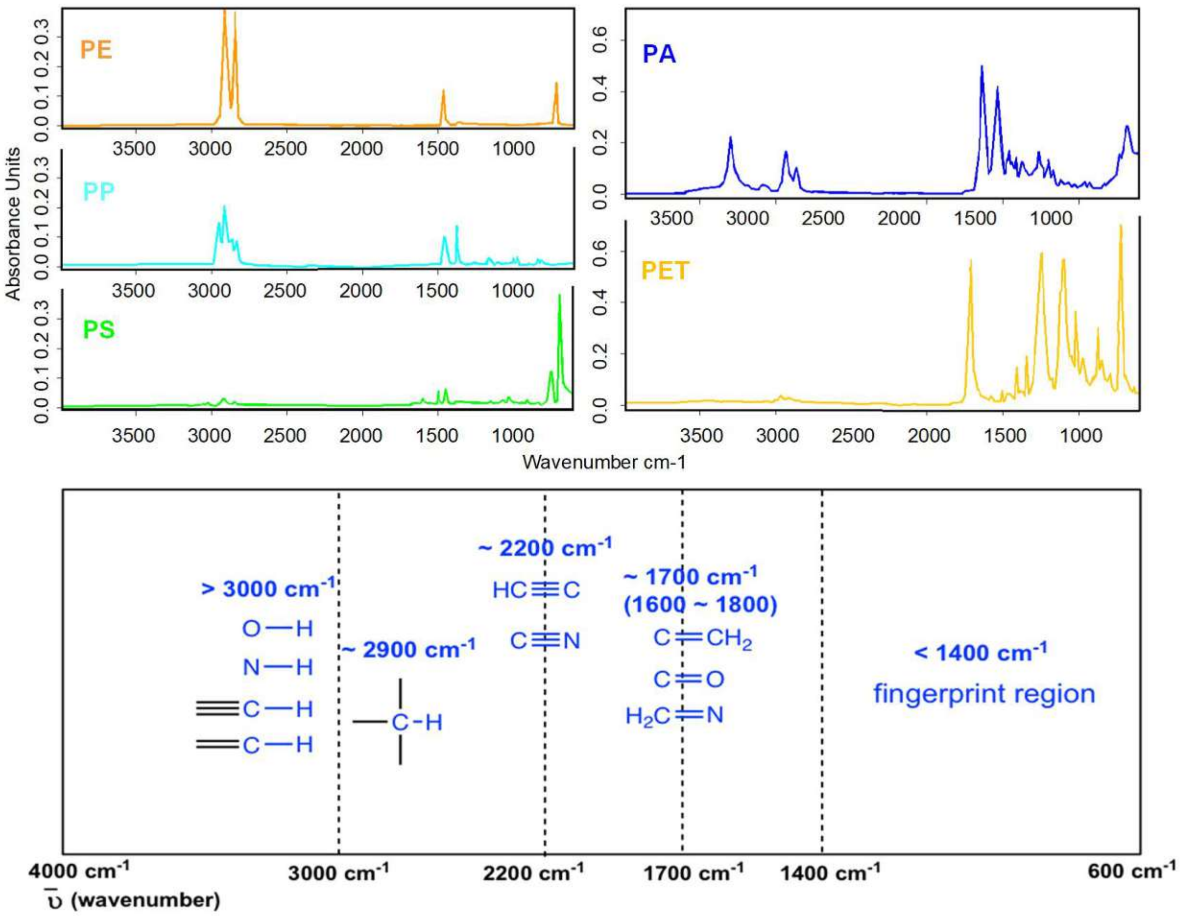


Figure 18: FT-IR sample spectra for PE, PP, PS, PA, PET and characteristic IR regions of important chemical groups - own illustration based on [128]

The principle of the measuring device is based on a Michelson interferometer. A beam of light coming from the source is split into two beams. One of the beams is directed towards a fixed mirror, while the other is directed towards a linearly movable mirror. The two beams are detected and transformed into a spectrum by means of Fourier transformation. [129]

Peaks in the spectrum can be identified by their position and intensity. The position of a peak corresponds to the frequency at which molecular vibrations occur, and the intensity indicates the strength of the vibration. Peaks can be assigned to functional groups in the sample based on their characteristic frequencies. The sum of the characteristic frequencies can then be clearly assigned to different polymers for proper identification (figure 18). [130]

Inner and outer layers of post-consumer packaging were investigated using a Bruker Tensor 27 spectrometer equipped with an attenuated total reflection (ATR) diamond (DuraSample IR II) with single reflection. A total number of 16 scans with a resolution of 4 cm^{-1} were conducted between $600 - 4000$ cm^{-1}.

2.1.3 Light microscopy

Figure 19: Light microscopy preparation procedure

For microscopy analysis (figure 19), packaging samples measuring 20x12 mm were cut, cleaned with ethanol, and placed in a sample holder. The samples were embedded with a two-component epoxy resin (Araldite AY103 + REN HY956). To prevent the packaging samples from floating, only a small amount of resin was used for the first filling (filling height 3-5 mm). Samples were then kept in a vacuum chamber for 15 min to reduce air bubbles and subsequently stored on a flat surface for 1-2 hours until the second filling (full height). Prior to automatic grinding and polishing (9) with a Struers TegraForce-31 polisher, the curing time was at least 12 h.

Table 9: Grinding and polishing program for light microscopy film preparation

Processing step	**Force [N]**	**Time [min]**	**Speed [rpm]**	**Grain size**	**Lubricant**	**Suspension**
1	15	1	300	SiC paper #320	water	-
2	10	1	300	SiC paper #500	water	-
3	10	2	150	SiC paper #1000	water	-
4	10	2	150	SiC paper #2000	water	-
5	10	2	300	SiC paper #4000	water	-
6	10	5	150	MD-Dur 3 μm	DP-Lub. blue	DP-Susp. 3μm
7	10	3	150	MD-Chem	DP-Lub. red	OP-U

Thicknesses of the individual layers were measured with a Zeiss Axio Imager M2m light microscope and analyzed with the corresponding Axio Vision software. For a sufficiently good representation, a 20- to 50-fold magnification was necessary. Polymer mass content was estimated for every sample using volumetric mass density relation.

2.2 Reprocessing and evaluation

Prior to processing, post-consumer packaging waste was de-labeled, cleaned (manual cold wash), and shredded (universal cutting mill Fritsch Pulverisette 19 with 4 mm sieve insert) in line with industrial processing steps. Before extrusion PET containing material was additionally dried for at least 4 h at 80 °C. Due to the manual selection of the packaging and detailed structural examinations, a high degree of purity of the individual recyclate fractions could be realized if desired.

2.2.1 Extrusion

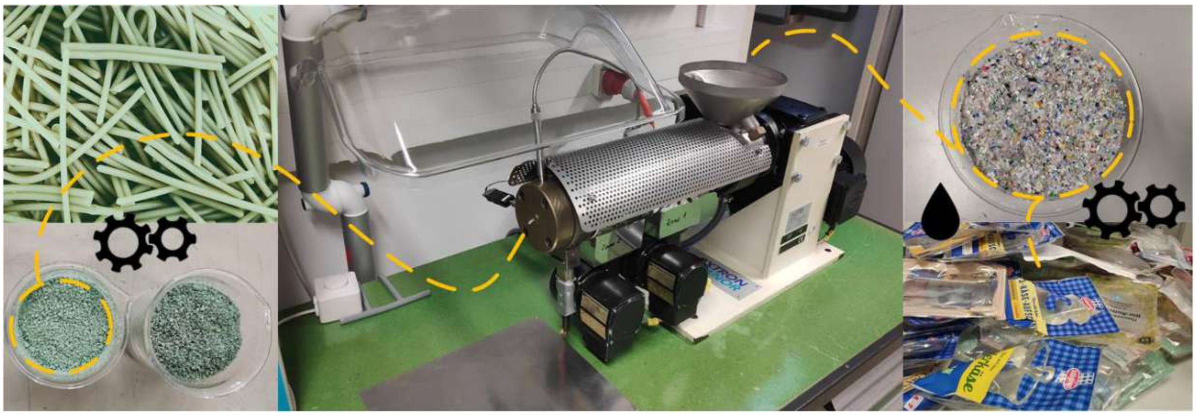


Figure 20: Preparation and extrusion of post-consumer waste

Virgin materials, polymer blends, and post-consumer recyclates were processed in an Extron EX-18-26-1.5 single screw extruder (figure 20) before test specimens production and further material testing. The extruder had a screw diameter of 18 mm, a length/diameter ratio of 25:1 and 3 individual heating zones. Screw speed was set to 70 rpm, and the temperatures were selected according to the processed material:

- **Zone 1** (Feed): 165 °C (polyolefins), 260 °C (PET containing material)
- **Zone 2** (Compounding): 220-240 °C (polyolefins), 275 °C (PET containing material)
- **Zone 3** (Die): 200-240 °C (polyolefins), 275 °C (PET containing material)

Compared to industrial extruders, lab devices are typically much smaller and less powerful. The throughput rates are lower, and the control units are less complex, not allowing for precise adjustment of specific parameters such as extrusion pressure, temperatures, and operation speed (table 10). However, both extruder types work on the same basic principle, and since industrial devices have more options regarding purification (degassing zones and melt filtration), the recyclate quality in the laboratory can be seen as a kind of worst-case scenario concerning the processing. [131]

Table 10: Differences between industrial recycling extruders and lab equippment [131,132]

Feature	Indsutrial	Lab
Throughput	up to 6 t/h	approximately 1.5 kg/h
Screw	double screw (diameter 80-280 mm)	single screw (diameter 18 mm)
Heating	5 or more zones	3 zones
Output	pellets	strands
Filter	melt filter	-
Dosing	dosing of additives during extrusion	-
Degassing	degassing zone for improved purity	-

2.2.2 Aging (simulation)

The reprocessing of post-industrial spare material has always been a common practice since polymer processing started. Production residues or waste, such as sprues from injection molded parts or edge trim, are shredded on-site and mixed with other virgin material. Consequently, the resulting products contain material that has been damaged to a certain degree. After all, even with the utmost care, degradation of the material due to mechanical stress and heat during processing cannot be avoided. For this reason, the effect of multiple processing steps on the material properties is of great interest not only for post-industrial waste but especially for post-consumer waste, since the longer material can be kept in the cycle, the more resources are saved. [133]

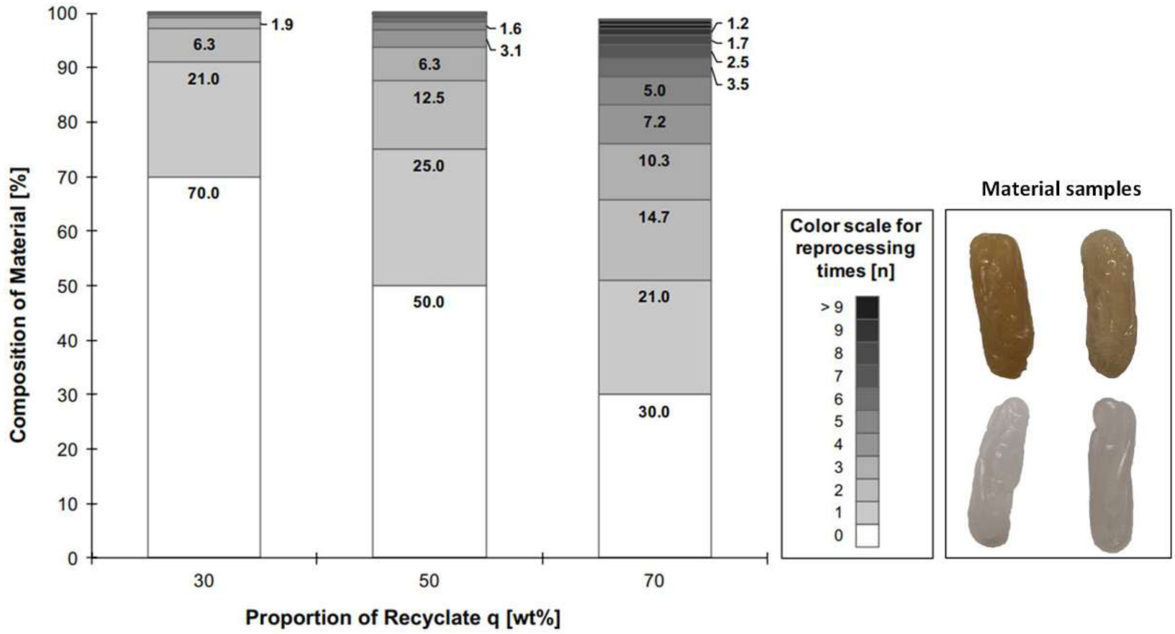


Figure 21: Composition of recyclate/virgin blend after n steps for 30%, 50% and 70% recyclate proportion [134]

If a proportion q of shredded residues is added to the raw material, the resulting product will contain twice the processed and damaged material. The resulting residues also contain this portion q and are again mixed with raw material in a certain ratio. If this process is repeated, the following equation 5 can be used for specification [134]:

$$\sum_{i=1}^{n} q^{n-1}(1-q) = 1 \tag{5}$$

When using small proportions of recyclate, the recyclate material typically contains minimal amounts of highly degraded material that has undergone multiple processing cycles. Figure 21 demonstrates the material composition for various mixing ratios of recycled and virgin material. The first column, representing a 30% recycled and 70% virgin material mix, shows that the regrind material contains less than 0.8% of material that has been reprocessed five times or more.

The majority of the material is virgin, with only small fractions having been processed once, twice, or three times. As the proportions of material less than 1% have negligible influence on material properties, the properties are determined primarily by fractions that have undergone four or fewer processing cycles. Thus, it can be concluded that the material properties of blends with small amounts of recyclate will not drop below a certain level. [134]

However, regrind material with high proportions of recyclate contains significant amounts of highly degraded material, as shown in the right column of figure 21, where 70% of the regrind is recycled, and 30% is virgin material. This regrind material contains 5.0% material that has been reprocessed five times or more, along with smaller amounts of material that has undergone fewer processing cycles. Even after nine processing cycles, the material still contains 1.2% of the original material. Although this mix contains significant portions of highly degraded material, it can still be useful for specific applications. [134]

The simulation of multiple recycling cycles was realized by repeated shredding and extrusion according to the parameters from section 2.2.1 but without phases of product manufacturing and use in-between. Therefore, only the degradation from reprocessing but not during the service life is considered.

2.3 Film blowing

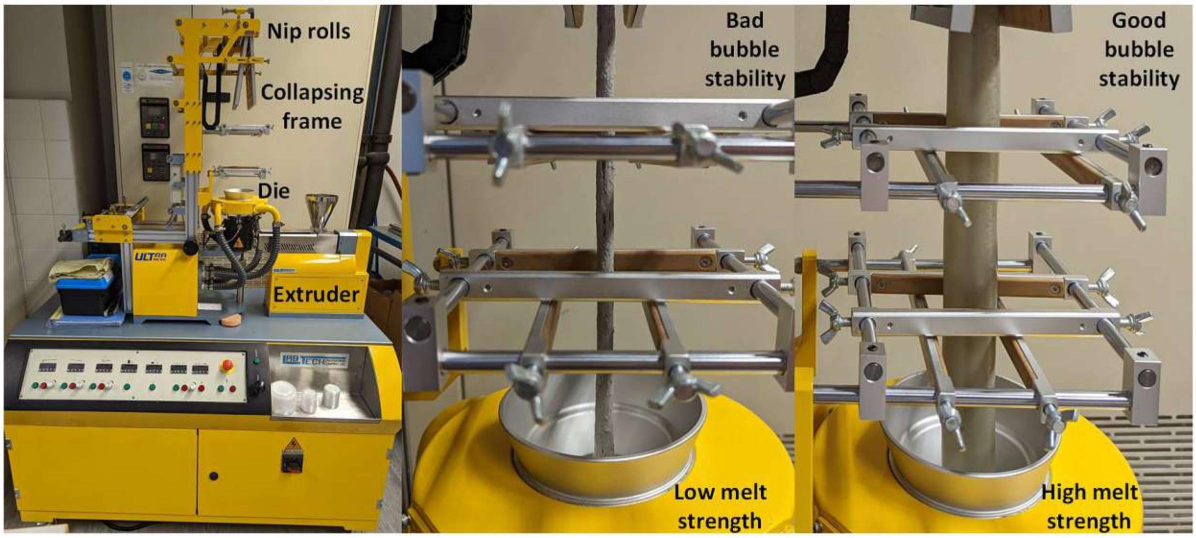


Figure 22: Micro film blowing line and sample for good/bad bubble stability of recyclate

Since many packaging products have films as their base, it is of great interest to (at least) functionally realize the manufacturing of recyclates into products of the same economic value. For that purpose polyolefin-based materials that demonstrated good melt strength were processed into 50 μm thick films (figure 22) utilizing an Ultra Micro blown-film line (LabTech Engineering) using the following parameters:

- **Extrusion temperature**: 200 °C
- **Die temperature**: 180 °C
- **Airflow cooling fan**: 1700 rpm
- **Pull-off speed rollers**: 1.1 m/min

2.4 Test specimens

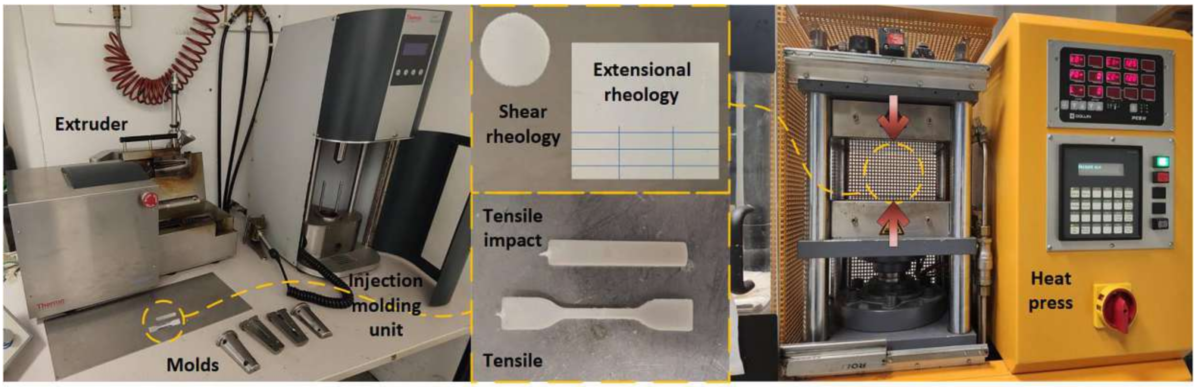


Figure 23: Manufacturing of mechanical (tensile, tensile impact) and rheological (shear, extensional) test specimens trough injection- and compression molding

For material characterization, mechanical end rheological test specimens were produced (figure 23). The test specimens for tensile testing and tensile impact testing were manufactured by injection molding using a Haake Mini Jet II after previous extrusion in a Haake Mini Lab twin-screw extruder with the following parameters:

- **Screw speed**: 100 rpm
- **Melt temperature**: 220-240 °C (polyolefins), 275 °C (PET containing material)
- **Mold temperature**: 50 °C ± 10%
- **Injection pressure**: 350 bar
- **Injection time**: 10 s

Specimens for dynamic shear and extensional rheology were produced by compression molding with a Collin P 200 P heat press at a pressure of 100 bar and in line with extrusion temperatures (220-240 °C (polyolefins), 275 °C (PET containing material)). Discs with a diameter of 25 mm and a thickness of 1.2 mm and squares of 0.8 mm thickness, and a side length of 60 mm were generated using punched aluminum frames sandwiched between steel plates and separated by Teflon sheets.

2.5 Rheology

To describe the deformation and flow behavior of materials, the relative displacement of material elements with respect to each other is examined in rheological investigations. Through the application of external forces, permanent deformations can be induced. This process, which is referred to as flow, can be observed for both solids as well as for liquids. The internal resistance of a fluid to external stress is described as material viscosity. The measuring methods can be classified according to the type of load (shear, strain, bending) or according to the rheometer type used (e.g., rotational viscometer or capillary rheometer). [135,136]

2.5.1 MFR

MFR testing is a widely used technique for characterizing the rheological properties of polymers, especially for industrial purposes, serving as an easy and fast quality measurement tool. The test was performed to ISO 1133 [137] and involves melting a small amount of polymer (3-5 g) and extruding it through a capillary tube under a constant load (2.16 kg) and temperature (220-240 °C (polyolefins), 275 °C (PET containing material)). Rate of extrusion is measured and reported as the melt flow rate in grams per 10 minutes (g/10 min). [137]

The MFR of a polymer is influenced by various factors, including its molecular weight, molecular weight distribution, and chemical composition. Polymers with higher molecular weights typically have lower MFR values, as they require more energy to flow. Conversely, polymers with lower molecular weights have higher MFR values as they flow more easily. Various additives, even in small quantities, can also influence material flowability and wall adhesion properties and hence the MFR. [135]

2.5.2 Shear rheology

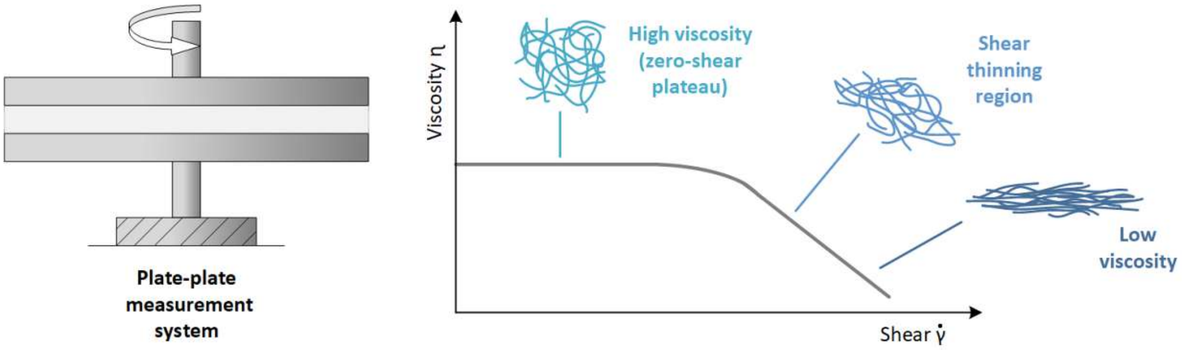


Figure 24: Shear rheology measurement principle and polymer melt behavior - own illustration based on [138]

In shear rheological tests, the specimens are subjected to shear stress. Compact rheometers that can either be operated in rotatory or oscillatory mode equipped with a plate-plate system are suitable for that purpose. The material sample is placed between two parallel plates, one of which is rotated and the other is anchored in a stationary fix (figure 24).The variation of parameters such as gap height, load intensity, shear rate, or temperature results in a large number of measurement program possibilities. For this thesis, frequency sweeps and steady shear measurements were performed with the parameters given in table 11 using an Anton Paar MCR 302 compact rheometer equipped with a PP25-SN39840 measurement tool and a nitrogen-purged heating hood and heating plate. [139]

Table 11: Machine parameters for shear rheology measurements

	Frequency sweep	**Steady shear test**
Number of measurement points	25	1000
Temperature [°C]	220-240/275	180
Angular frequency [rad/s]	628-0.1	-
Shear rate [1/s]	-	0.1 and 0.001

Frequency sweeps allow for the determination of storage (G' [Pa]) and loss modulus (G" [Pa]) as a function of a variable angular frequency (ω [rad/s]) and the temperature, which remains constant. With G' and G", the complex viscosity (η^* $[Pa \cdot s]$) can further be determined via the Cox-Merz relation (equation 6). [140]

$$|\eta^*| \equiv \frac{|G^*|}{\omega} = \frac{|G''|}{\omega}\left[1 + (\frac{G'}{G''})^2\right]^{0.5} \tag{6}$$

The complex viscosity is indirectly proportional to the MFR but describes the melt flow under dynamic shear contrary to constant shear. G' represents the elastic response of a material or its ability to store energy when subjected to a deformation. A material with a high storage modulus will resist deformation and be more elastic, while a material with a low storage modulus will be more fluid-like and easier to deform. G" represents the viscous response of a material or its ability to dissipate energy when subjected to deformation. The cross-over point of G' and G" indicates the transition from the predominance of elastic to viscous behavior of the polymer melt. A shift in the cross-over point is associated with changes in molecular weight (MW) and molecular weight distribution (MWD) of the polymer melts. A cross-over at lower frequencies is related to the presence of longer or branched molecules with longer relaxation times, while a vertical shift to lower G values is associated with a broadening of the MWD. Using the ratio of loss modulus to storage modulus, loss angle δ and loss factor $tan(\delta)$ can be calculated as a function of ω. $tan(\delta)$ can give information about deviating relaxation times which is directly correlated to the macromolecular structure. [141–143]

Steady shear measurements were used to determine the base curves (linear viscoelastic range, LVE) for extensional rheology. Changes in viscosity were measured over time with constant temperature and pre-defined shear rates.

2.5.3 Extensional rheology

Figure 25: Extensional rheology measurement principle and polymer melt behavior

In contrast to shear rheology, the clamped test specimen is subjected to a tensile force in extensional rheology measurements, which subsequently leads to an elongation of the material (figure 25). Since the specimens are subjected to a different load in this test method, the resulting viscosity is also different and referred to as extensional viscosity. Extensional viscosity consistently demonstrates higher values compared to shear viscosity, and for conversion, a factor of 3 can be used, according to Trouton. [144]

The LVE curve represents the behavior of a polymer melt with linear chain structures and poor strain hardening properties. If the polymer melt demonstrates high melt strength, the extensional viscosity values exceed those of the LVE curve. Contrary, low melt strength leads to an alignment of the viscosity curves. [145]

To provide a more quantitative description of the strain hardening phenomenon and, consequently, the melt strength, strain hardening coefficients (SHC) can be calculated according to equation 7. $\eta(t)$ refers to the highest extensional viscosity for the corresponding strain rate, while $\eta_0(t)$ denotes the extensional viscosity of the LVE curve. [146, 147]

$$SHC = \frac{\eta(t)}{\eta_0(t)} \tag{7}$$

For extensional rheology measurements, the compact rheometer was equipped with a CTD 450 heating chamber (nitrogen purged) and an SER-HPV 1 (Sentmanat Extensional Rheometer) measurement tool. Test specimens (8x20 mm) were strained at three different rates (5 s^{-1}, 1 s^{-1} and 0.1 s^{-1}) at a temperature of 180 °C in line with the film-blowing procedure.

2.6 Mechanics

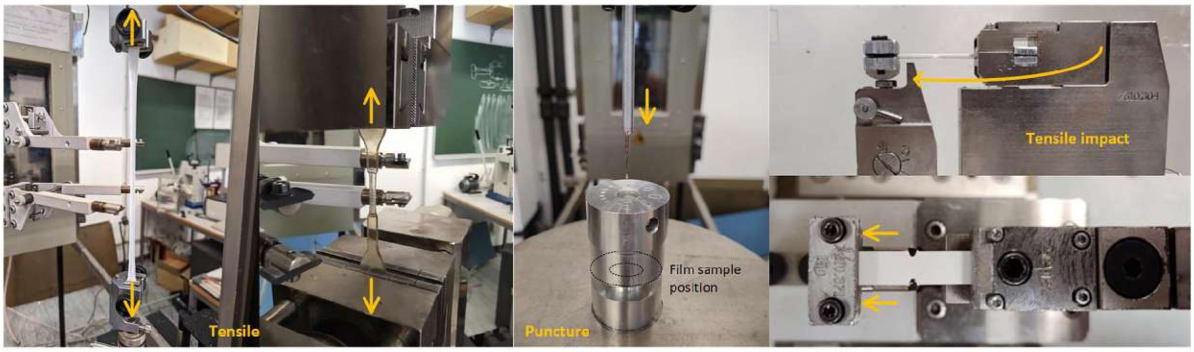


Figure 26: Tensile, puncture and tensile impact testing procedures

2.6.1 Injection molded specimens

In the conventional tensile test, bone-shaped test specimens are strained at a constant cross-head speed in an axial direction, and the test is completed as soon as the specimen breaks (figure 26). During the test, a corresponding time-dependent reaction force F is generated within the specimen due to the external load. Measurement of that force combined with the specimen cross-section (A) allows the determination of the component stress σ [MPa] (quotient of F and A). The material also undergoes a time-dependent change in length which can be expressed as strain ε [%] and is associated with material ductility. Using Hook's law, the modulus of elasticity (E_t $[MPa]$) can be calculated from σ and ε, which is a measure of the material stiffness. Tensile testing was in accordance with DIN EN ISO 527-2-5A [148] and performed on a universal testing machine (ZwickRoell Z050) at a cross-head speed of 10 mm/min. The testing device was equipped with a 1 kN load cell and an extensometer. [149]

For tensile impact testing, an external load is also applied in an axial direction of the test specimen. According to DIN EN ISO 8256 [150], the tests can be described as tensile tests with relatively high deformation speed (figure 26). The measurement results (tensile impact toughness a_{tN} $[kJ/m^2]$) represent values that can be associated with material toughness. a_{tN} indicates the amount of energy that is consumed for the rupture of the test specimen by a falling hammer, based on the specimen cross-section. Prior to testing, the specimens were notched on both sides using a Ceast Vis-Notch device. Subsequently, they were tested with an Instron 9050 pendulum (equipped with a 2 J hammer and a 15 g cross-head mass). [151]

2.6.2 Film samples

To get an idea of the packaging film and lab blown film performance under load, tensile tests were carried out according to DIN EN ISO 727-3 [152] with strips measuring 100x10 mm (clamping length 70 mm) at a cross-head speed of 50 mm/min (figure 26). Whenever possible, film samples were measured in longitudinal and transverse directions due to potential differences in film orientation.

In addition to the parameters from tensile testing, the puncture resistance was evaluated as an essential factor associated with packaging stability. Punctual damage to the film structure could likely lead to an unintentional exchange of gas and, subsequently product spoilage. Testing was performed according to DIN EN 14477 [153] (ZwickRoell Z050) with round film samples having a diameter of 20 mm at a test speed of 10 mm/min. The test involves measurement of the force F [N] and displacement l [mm], which is required to penetrate a film sample by a needle with a rounded tip (figure 26). Puncture energy (E_p $[mJ]$) as a single parameter can be calculated through the derivation of the resulting force-displacement curve. As thinner structures are more likely to suffer from external damage, the focus was laid on the testing of film samples with thicknesses < 150 μm, which excluded most rigid packaging structures. Samples were always perforated according to packaging functionality from outside to inside. [4,153]

2.7 Morphology

Scanning electron microscopy is based on the principle that a finely focused electron beam ($<$ 1 nm) is passed over the surface of the object to be imaged. Secondary and back-scattered electrons are generated on this surface. Through the interaction of electrons and the material surface, a signal is generated. This signal corresponds to the number of emitted secondary or back-cattered electrons. Subsequently, imaging is carried out with the aid of a Braun tube, in which the bundled beams are deflected or modulated accordingly. To exclude interactions with the environment, the electron beam and the sample must be in a vacuum. Sample images from the millimeter to the nanometer range can be realized, with the images produced having a very high depth of field. For insulating samples such as polymers, it is necessary to apply a conductive layer of noble metals or carbons. This can be realized by vapor deposition or sputtering. [154]

For morphology characterization, the fractured surfaces of the test specimens from the impact tensile test were examined with an FEI Philips XL30 scanning electron microscope. Prior to imaging, the samples were coated with gold (Agar Sputter Coater B7340). Weibull distributions of the particle sizes were generated by measuring the areas of 100-200 individual particles in order to calculate the average diameters.

2.8 Material overview

2.8.1 Virgin and industrial regranulates

Virgin polymer types for the reference (blends) were selected according to their field of application (food grade, film type) and processed without any further treatment. Table 12 gives an overview of the processed resin types. MFR values are obtained from data sheets (except from PET) and therefore measured under different conditions.

Table 12: Virgin polymers [155–164]

Material	Type	Purpose	MFR [g/10 min]
Braskem PP H7058-25R	PP homopolymer	thin wall injection molding	25 (230 °C, 2.16 kg)
Borealis PP HC600TF	PP homopolymer	thermoformed packaging	2.8 (230 °C, 2.16 kg)
Borealis PP BC918CF	PP block copolymer	flexible packaging	3.0 (230 °C, 2.16 kg)
Borealis PP BA110CF	PP block copolymer	unoriented blown film	0.85 (230 °C, 2.16 kg)
Ineos PE LL6608AF	PE linear low density	blend blown film application	0.9 (190 °C, 2.16 kg)
Dow PE Agility EC7000	PE low density	sealant in multi-layers films	3.9 (190 °C, 2.16 kg)
Dow PE 310E	PE low density	blown film	0.75 (190 °C, 2.16 kg)
Borealis PE MB6561	PE high density	injection molding	1.5 (190 °C, 2.16 kg)
Invista Polyclear 1101	PET	various packaging applications	45 (275 °C, 2.16 kg)
Soarnol ET3803RB	EVOH 38 mol%	gas barrier in multi-layer films	4.0 (210 °C, 2.16 kg)

Industrial regranulates were used for the comparison of industrial and lab recycling processes. The different types are displayed in table 13.

Table 13: Industrial regranulates [165–167]

Material	Type	Characzeristic	MFR [g/10 min]
Rissland PE-LD regranulate	PE low density	white color	3.2 (230 °C, 2.16 kg)
Duales System 70002 PE regranulate	PE high density	black color	1.0 (230 °C, 2.16 kg)
MTM Plastics Dipolen S regranulate	PE and PP mix	grey color	5.1 (230 °C, 2.16 kg)
Total RPPC03GR regranulate	PP mix	grey color	3.1 (230 °C, 2.16 kg)

2.8.2 Post Consumer

Post-consumer packaging products from different categories which were processed into recyclates are provided in tables 14 - 18.

Table 14: Post-consumer PP-based MAPs

Modified atmosphere packaging (PP-based)		
Collection period	04/2021 - 07/2022	
Characteristic	packaging from sliced ham; contains only polyolefins; distinction between lids and trays; multiple processing	

Table 15: Post-consumer PET-based MAPs

Modified atmosphere packaging (PET-based)		
Collection period	06/2018 - 12/2020	
Characteristic	packaging from sliced sausage and cheese products; distinction between lids and trays	

Table 16: Post-consumer frozen food packaging

Frozen food packaging		
Collection period	02/2022 - 08/2022	
Characteristic	packaging from various frozen food products; distinction between polyolefin and polyolefin/PET blends	

Table 17: Post-consumer cosmetic packaging

Cosmetic packaging		
Collection period	05/2021 - 11/2021	
Characteristic	packaging from various cosmetic products; distinction between polyolefin and polyolefin/PET blends	

Table 18: Post-consumer snack and ready-made meals packaging

Snack and ready-made meals packaging		
Collection period	06/2020 - 08/2022	
Characteristic	packaging from various snacks and ready-made meals; distinction between polyolefin and polyolefin/PET blends	

2.8.3 Additives

Table 19: Processing additives [168–170]

Additive	Characteristic
Lubobit Lucofin 1492M HG	modified ethylene butyl acrylate copolymer grafted with maleic anhydride, increased polarity and adhesion properties, reactive
Ampacet ReVive 311E	commercial compatibilizer for multi-layer films
Kraton G1643M	non-reactive styrene- ethylene/butylene copolymer, 20% styrene content

Among the used additives were different chemicals based on reactive and non-reactive interaction principles (table 19).

3 Summary of the publications

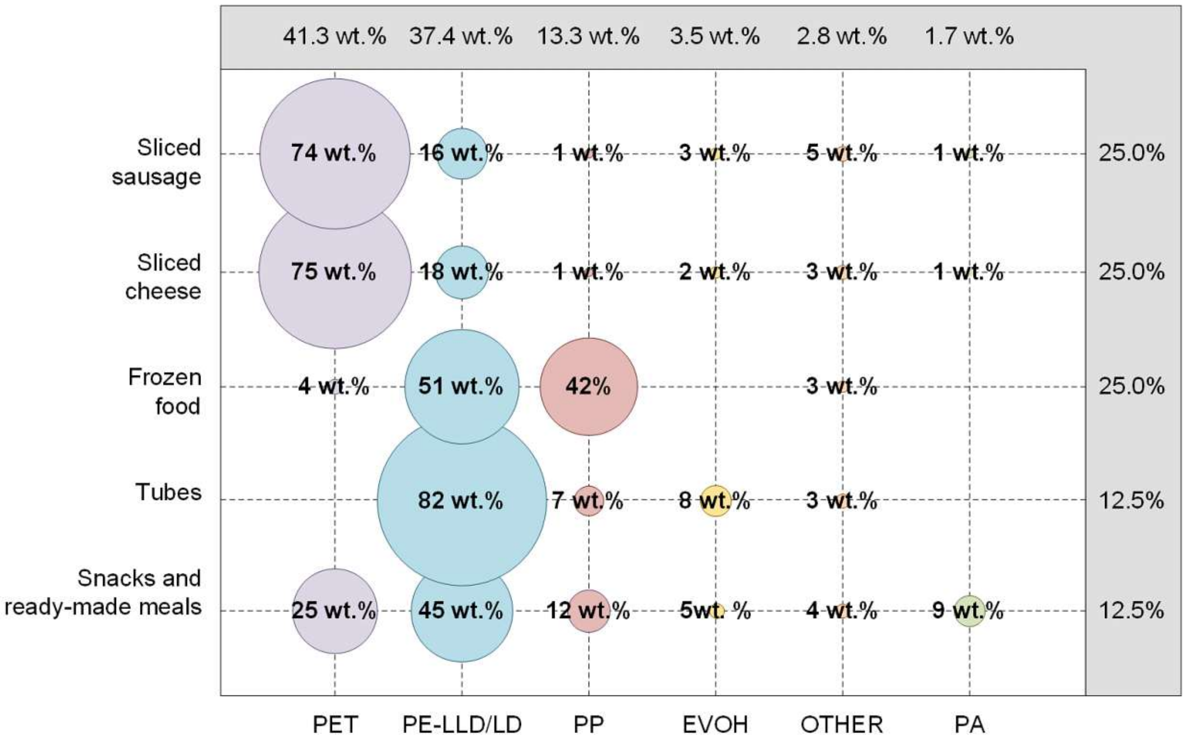


Figure 27: Waste stream composition of post-consumer packaging derived from structure analysis and separated by product packaging category (sliced sausage, sliced cheese, frozen food, cosmetic tubes, snacks and ready-made meals) and polymer type (PET, PE-LLD/LD blend, PP, EVOH, OTHER minor components and PA).

Figure 27 gives an overview of the waste stream composition of all (approximately 240) investigated packaging products which were divided into packaging types (sliced sausage, sliced cheese, frozen food, cosmetic tubes, snacks, and ready-made meals) and polymeric constituents. PET and PE were the major constituents of the overall packaging waste stream, with proportions of 41.3 wt.% and 37.4 wt.%. Packaging made from sliced sausage and cheese has comparable multi-layer structures, resulting in consistent waste streams. However, when other types of packaging are combined, the compositions become more varied and less predictable. This presents a significant challenge for mechanical recycling, as unpredictable mixtures and highly incompatible material combinations such as PET and PE result in low-quality secondary materials (brittleness, bad smell, grey color), which are often difficult to process and unsuitable for secondary packaging applications.

Based on the packaging analysis, reference model blends from virgin polymer materials were designed, and their properties were investigated alongside the recycled packaging products. Deteriorating factors were isolated, possible compatibilization approaches were tested for their economic viability and utility, and the applicability of the recyclates in secondary products was analyzed. Based on those findings, design for and from recycling principles were identified.

3.1 Modified atmosphere packaging (Publication 1+2)

Modified atmosphere packaging (MAP) is commonly known as a two-component system consisting of a sealed lid and tray with multiple layers of materials that encase food items within a modified gas atmosphere. The effectiveness of the packaging depends on its ability to sustain the modified atmosphere for an extended period, which is achieved by preventing gas diffusion through the materials and protecting the contents from external damage.

In order to consider a separate recycling scenario for the packaging parts (lid, tray), those structures were investigated independently from each other for sliced sausage (SP) and sliced cheese (CP) packaging. SP and CP trays which represented ~ 80 wt.% of the total packaging mass, had comparable compositions comprising PET (SP: 84 wt.%, CP: 83 wt.%), blends of PE-LD and PE-LLD (SP: 8 wt.%, CP: 11 wt.%), adhesives (SP: 4 wt.%, CP: 2 wt.%), EVOH (SP: 2 wt.%, CP: 1 wt.%) and other minor constituents such as paper and PA. Contrary, lids contained much higher PE contents (SP: 50 wt.%, CP: 46 wt.%) accompanied by PET (SP: 33 wt.%, CP: 43 wt.%), EVOH (SP: 7 wt.%, CP: 5 wt.%), adhesives (SP: 6 wt.%, CP: 4 wt.%) and other minor constituents. While lids and trays, as individual system parts, demonstrate uniform waste streams, their combination as a whole packaging system leads to a greater variation in PET/PE ratio which are the main constituents (trays: ~ 84/10 wt.%, lids: ~ 38/48 wt.%, overall: 71/19 wt.%) and hence poorer recyclability.

PE was always present as a blend comprising PE-LD and PE-LLD in different ratios (common industrial practice to improve bubble stability during film blowing), resulting in either PE-LD or PE-LLD rich blends in the packaging barrier structures (PE-EVOH-PE) of lids and trays. Based on those findings, PE-LD and PE-LD rich blends representing the packaging parts (lid, tray, lid+tray) comprising PE, PET, and EVOH were processed from virgin material and compared to corresponding recycled post-consumer packaging and virgin PET (main constituent of MAP waste stream).

Table 20: E_t [MPa], ε_b [%], MFR [g/10 min] and a_{tN} $[kJ/m^2]$ of individual system parts (lid, tray) and the entire packaging system (lid+tray). Extruded virgin material blends are divided into PE-LD- and PE-LLD-rich compositions and compared to reprocessed post-consumer MAPs and virgin PET (main constituent of the MAP waste stream).

	$MFR\ [g/10min]$	$E_t\ [MPa]$	ε_b [%]	$a_{tN}\ [kJ/m^2]$
Lid blend PE-LD rich	15 ± 1.5	1110 ± 103	50 ± 4	36 ± 3
Lid blend PE-LLD rich	5 ± 0.4	990 ± 82	78 ± 7	42 ± 4
Lid post-consumer	28 ± 1.9	890 ± 86	3 ± 0.4	10 ± 1
Tray blend PE-LD rich	31 ± 2.7	2096 ± 203	25 ± 2	47 ± 5
Tray blend PE-LLD rich	21 ± 1.9	2113 ± 208	152 ± 15	56 ± 5
Tray post-consumer	82 ± 8.0	2286 ± 219	2 ± 0.2	6 ± 0.5
Lid + Tray PE-LD rich	28 ± 2.6	1831 ± 185	23 ± 2	45 ± 5
Lid + Tray PE-LLD rich	17 ± 1.5	1734 ± 172	88 ± 8	48 ± 5
Lid + Tray post-consumer	65 ± 6.3	1830 ± 172	2 ± 0.2	5 ± 0.4
Virgin PET reference	23 ± 1.9	2328 ± 228	324 ± 31	58 ± 5

Table 20 summarizes mechanical properties (E_t, ε_b, a_{tN}) and MFR values of the investigated packaging components and their reference blends. The mechanical properties and MFR of PE-LLD rich tray blends, containing the least PE-based contamination, most closely resembled those of virgin PET with deviations in MFR, E_t and $a_{tN} < 10\%$. However, ε_b was drastically reduced from 324% (virgin PET) to 152% (tray blend PE-LLD rich). The further increases in PE content of lid and lid+tray blends promoted deterioration effects and the highest percentage deviation from the PET reference for E_t (PE-LD rich: 52%, PE-LLD rich: 58%), MFR (PE-LD rich: 35%, PE-LLD rich: 78%), a_{tN} (PE-LD rich: 38%, PE-LLD rich: 28%) and ε_b (PE-LD rich: 85%, PE-LLD rich: 76%) in lid blends. For the post-consumer recyclate blends, even greater deterioration effects were observed with the highest MFR (lid post-consumer: 28 g/10 min, tray post-consumer: 82 g/10 min, lid+tray post-consumer: 65 g/10 min) and lowest ε_b and a_{tN} values $\leq 10\ \%/kJ/m^2$.

These various material properties are undoubtedly a result of the poor compatibility of PET and PE. To evaluate temperature- and compatibility-induced deterioration effects, the multi-layer structures were further divided into quarternary (PE-LD and PE-LLD rich lid+tray blends representing the whole packaging), tertiary (PE-LD and PE-LLD rich blends combined with EVOH representing the barrier structure) and binary (PE-LD and PE-LLD rich blends without the influence of EVOH) blends.

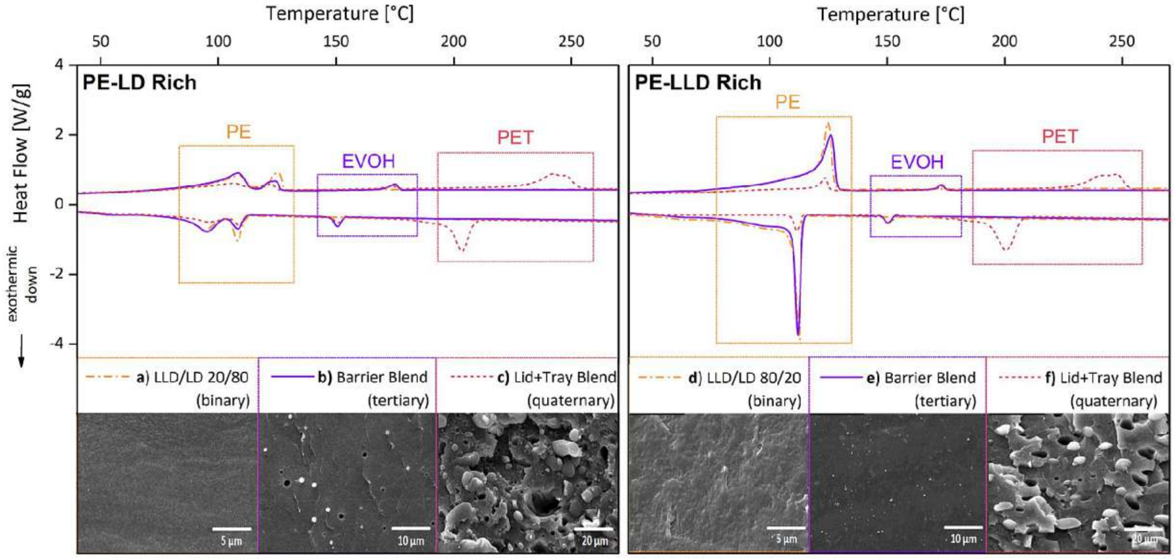


Figure 28: Component miscibility of PE-LD- (left) and PE-LLD-rich (right) blends demonstrated using DSC thermograms and scanning electron micrographs of (a, d) binary PE systems (LLD/LD 20/80, LLD/LD 80/20, reference), (b, e) tertiary barrier blends representing the barrier layer in MAPs and (c, f) quaternary lid+tray blends representative of the total MAP waste stream. [121]

The DSC thermograms (figure 28) of different types of PE in binary blends (LLD/LD 20/80) showed that they exhibit distinct melting and crystallization peaks. Even after blending with EVOH and PET, the peak separation between PE-LD and PE-LLD is still visible. However, samples rich in PE-LLD demonstrate only a single peak in the PE melt region, indicating better miscibility. Those findings were supported by SEM micrographs as PE-LLD rich tertiary blends (figure 28 e) exhibited a visibly finer morphology and smaller dispersed EVOH particles ($< 1\,\mu$m) compared to the PE-LD rich tertiary blends (figure 28 b). The binary system showed no phase separation and hence no conclusive evidence of a homogeneous mixture. Contrary, the quarternary blends demonstrated clear phase separation with dispersed particles $> 20\,\mu$m.

To prevent thermal degradation, ideal polymer reprocessing environments utilize temperatures only slightly higher than the polymer's melt region, whereas, in the case of MAP reprocessing, the presence of PET in the system requires temperatures exceeding 250 °C, despite the other polymer components having much lower melting points. At increased extrusion temperatures of 275 °C representative for combined polyolefine-polyester recycling, PE-LD- (LLD/LD 20/80) and PE-LLD- (LLD/LD 80/20) rich binary and tertiary barrier blends exhibited decreased MFR (table 21) and increased complex viscosity values compared to an extrusion temperature of 220 °C (representative for pure polyolefine recycling). Temperature-dependent mechanical properties, such as E_t, a_{tN} and ε_b, varied less than rheological properties (deviations $< 15\%$), with the exception of the ε_b values, which were more sensitive to changes in processing temperature.

Table 21: Impact of extrusion temperature T on MFR, E_t, a_{tN}, and ε_b of PE-LD- (LLD/LD 20/80) and PE-LLD-rich (LLD/LD 80/20) binary and tertiary (barrier) blends containing EVOH.

Sample		MFR [g/10 min]	E_t [MPa]	a_{tN} [kJ/m^2]	ε_b [%]
		T= 220 °C			
PE-LD	**LLD/LD 20/80**	3.9 ± 0.4	200 ± 3	215 ± 11	126 ± 13
rich	**Barrier blend**	3.5 ± 0.3	227 ± 13	230 ± 9	122 ± 15
PE-LLD	**LLD/LD 80/20**	1.5 ± 0.1	325 ± 12	229 ± 21	98 ± 9
rich	**Barrier blend**	1.9 ± 0.2	380 ± 29	278 ± 27	104 ± 13
		T= 275 °C			
PE-LD	**LLD/LD 20/80**	3.2 ± 0.3	192 ± 10	243 ± 12	139 ± 17
rich	**Barrier blend**	3.0 ± 0.3	219 ± 14	227 ± 7	135 ± 6
PE-LLD	**LLD/LD 80/20**	0.8 ± 0.1	317 ± 5	255 ± 18	143 ± 19
rich	**Barrier blend**	1.0 ± 0.1	332 ± 7	301 ± 6	152 ± 8

Incompatibility of PET and PE, along with temperature-induced degradation at processing temperatures > 250 °C, makes the recyclate of MAPs comprising PET and PE unsuitable for any viable secondary application, even if only comparably small amounts of contaminants (trays) are present. It was further investigated whether compatibilizers can sufficiently improve the properties of the recyclate. Since the separation of lid and tray would be too complex and costly anyway with current industrial sorting techniques, the whole MAP system (lid+tray) was considered for these investigations. Figure 29 demonstrates the impact of three different compatibilizers in quantities of 3 wt.% and 7 wt.% on the properties of post-consumer lid+tray recyclates compared to uncompatibilized recyclate.

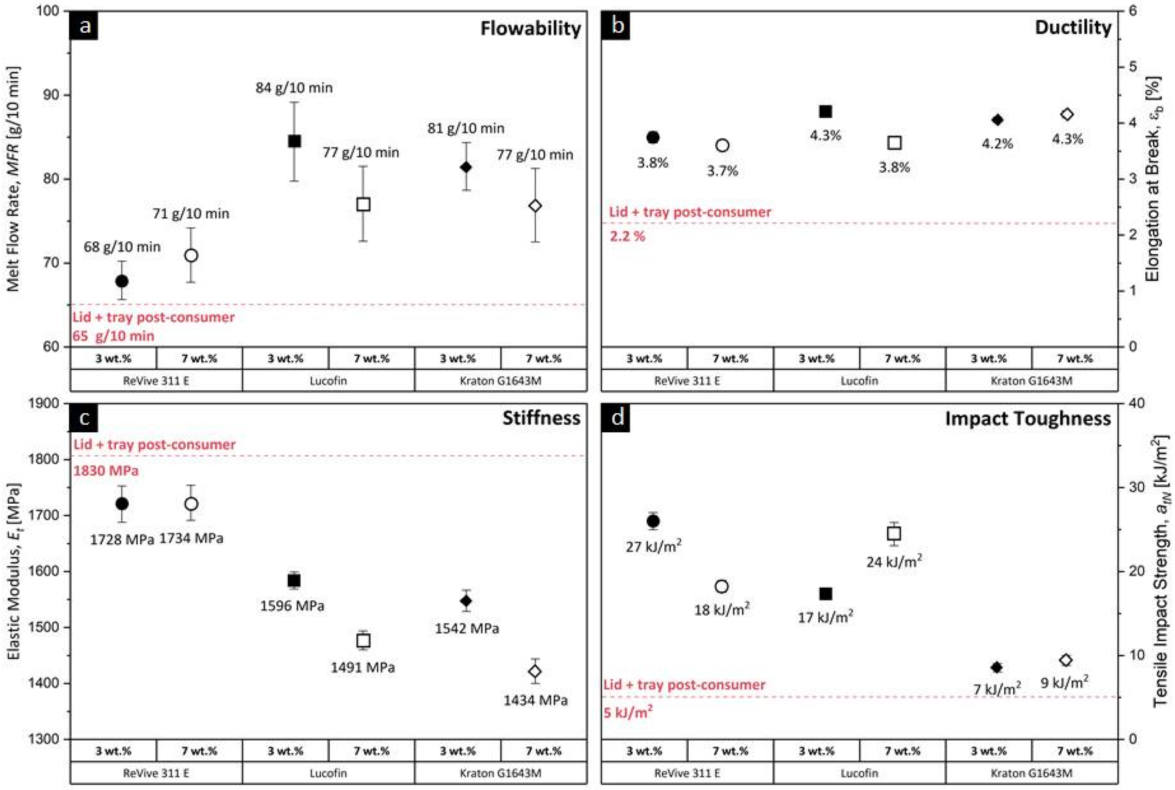


Figure 29: (a) MFR [g/10 min], (b) ε_b [%], (c) E_t [MPa] (d) a_{tN} $[kJ/m^2]$ of compatibilized (3 wt.% compatibilizer solid markers, 7 wt.% compatibilizer hollow markers) post-consumer lid+tray recyclates compared to post-consumer lid+tray recyclates without compatibilizer (dashed line).

The used compatibilizers were selected based on preliminary tests on virgin PE-LD and PE-LLD rich lid+tray blends [171]. Ductility and toughness could be greatly improved by the addition of compatibilizers (increase of the impact strength from 5 kJ/m^2 to 27 kJ/m^2 for 3 wt.% ReVive 311 E). However, the E_t (between 1434 MPa for 7 wt.% Kraton G1643M and 1734 MPa for 7 wt.% ReVive 311 E compared to 1830 MPa for the uncompatibilized recyclate) and MFR (between 68 g/10 min for 3 wt.% ReVive 311 E and 84 g/10 min for 3 wt.% Lucofin compared to 65 g/10 min for the uncompatibilized recyclate) values were consistently worsened. Additionally, a higher content of compatibilizer did not necessarily lead to improved material properties, but could also have the opposite effect (ReVive 311 E). On the one hand, these experiments demonstrated that the selection of the appropriate compatibilizer for a recycling waste stream is very complex and not necessarily always effective, and on the other hand, that the improvement is still far too low in the case of MAP recycling to be used in any secondary application.

Without the rapid development of suitable delamination methods for the recycling process, the only effective improvement for the recyclability of MAPs seems to lie in a change in their structure and, therefore, product design. Although the combination of PET with PE accounted for 90-95% of the market share for MAPs, it was not the only available product structure. Figure 30 depicts MAP structures other than the common PET-PE multi-layers. The used barrier structure consisted of three layers (PE-EVOH-PE) comparable to common MAPs but was combined with layers of paper, PA, or PP. Paper-polymer composites can have a positive environmental impact if they are easily separable and disposed separately. In the case of the paper tray, however, this is not possible since the paper layers are coated with PE and do not consist of paper alone (PE proportion $>$ 20 wt,%). The use of PA instead of PET offers some advantages, such as an improved odor barrier, but the combined recycling of PE and PA poses comparable challenges as the common MAPs. However, the packaging structure comprising PP, EVOH, and only a small proportion of PE ($<$ 5 wt.%) in the trays seemed very promising to demonstrate improved recyclability compared to PET-PE packaging.

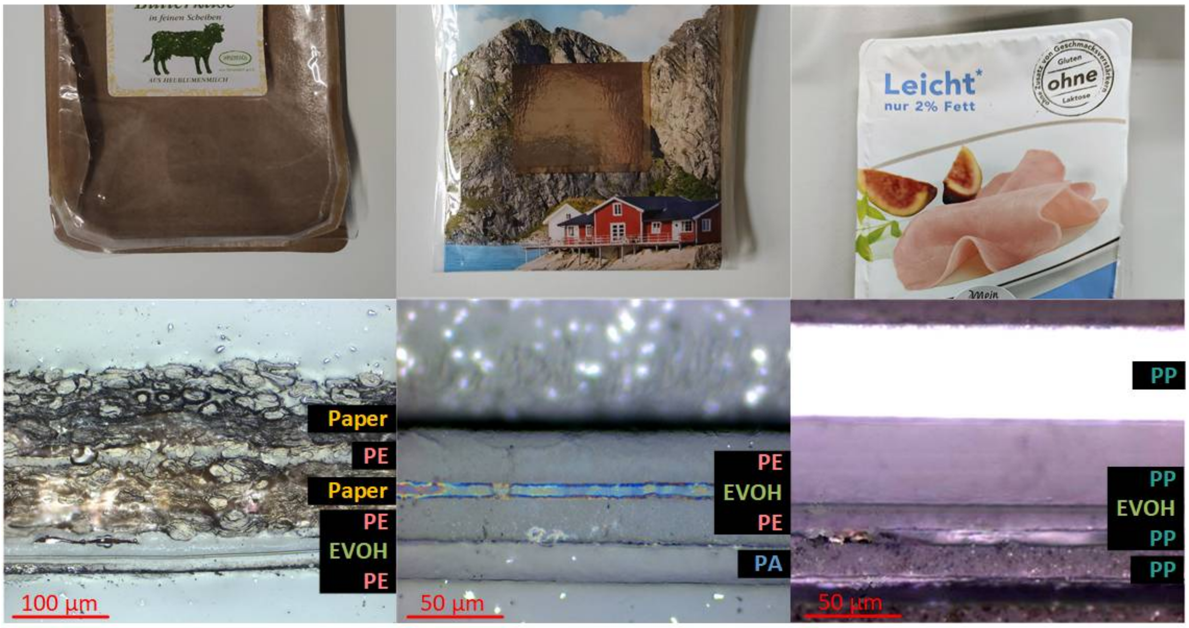


Figure 30: Packaging structure samples comprising a common barrier structure (PE-EVOH-PE) combined with paper (left), PA (middle) and PP (right).

Therefore, these PP-comprising MAPs were further investigated and demonstrated excellent recyclability. After one recycling cycle of post-consumer packaging, the material properties closely resembled those of virgin PP types used for film applications (MFR: 3.7 g/10 min compared to 3.3 g/10 min for virgin PP copolymer, E_t: 1245 MPa compared to 1282 MPA, ε_b: 832% compared to 884%, a_{tN}: 64 kJ/m^2 compared to 136 kJ/m^2). Tensile impact toughness, which was the only property that demonstrated considerably lower values, is highly dependent on the ethylene content, which was lower in MAPs as they also contained PP homopolymer. Due to the excellent material properties after one recycling step, the recyclate was subsequently extruded and shredded ten times, and the properties (MFR, η_0 E_t, a_{tN}, and ε_b) were determined after 3, 5, 7 and 10 reprocessing steps (table 22).

Table 22: MFR, η_0 E_t, a_{tN}, and ε_b of ten times reprocessed post-consumer PP based MAPs (lid+tray

.

Processing	MFR [g/10 min]	η_0 $[Pas]$	E_t [MPa]	ε_b [%]	a_{tN} $[kJ/m^2]$
1x	3.7 ± 0.3	4580 ± 421	1245 ± 118	832 ± 78	64 ± 6
3x	4.9 ± 0.5	3497 ± 321	1064 ± 101	944 ± 87	66 ± 6
5x	6.4 ± 0.6	2974 ± 299	1062 ± 104	917 ± 89	68 ± 7
7x	8.1 ± 0.8	2497 ± 221	1024 ± 98	875 ± 82	78 ± 7
10x	11.3 ± 1.2	1973 ± 187	944 ± 91	871 ± 88	69 ± 7

E_t was decreased by 25%, with the most significant drop between the first and the third reprocessing step. A slight increase of ε_b from 832% for the first processing step up to 871% for the tenth step was observed, again with the most significant increase between the first and third step. The tensile impact strength remained relatively constant during the ten reprocessing steps, with values ranging between 64 kJ/m^2 and 69 kJ/m^2. Over the ten processing steps, the MFR was gradually increased from 3.7 g/10 min to 11.3 g/10 min while conversely, the zero-shear viscosity was decreased from 4580 Pas to 1973 Pas. This material degradation is typically associated with the re-extrusion of PP [172] but also depends on branching or cross-linking reactions of the ethylene-containing polymer chains of PP copolymers.

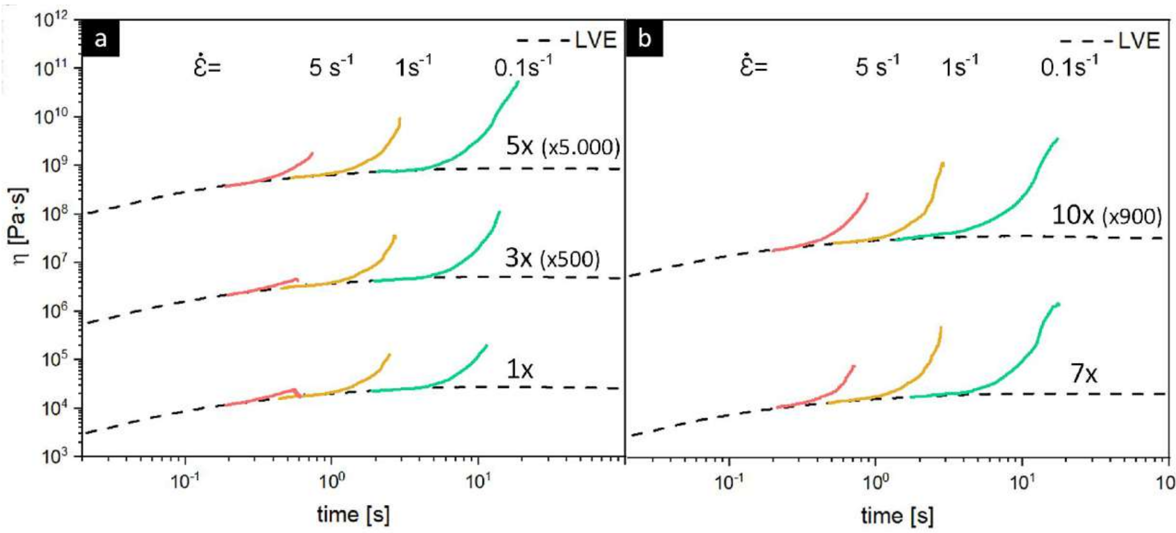


Figure 31: Extensional rheology curves of 10 times recycled (a, b) PP-based MAPs (lid+tray). [173]

Figure 31 represents the extensional rheology curves of the ten times reprocessed (1x, 3x, 5x, 7x, 10x) PP-based MAPs. High-quality film production and process stability require the use of high melt strength polymers, to which PP homopolymers usually do not belong as a result of their linear chain structure. Ethylene present in copolymers is responsible for the strain hardening of the melt, which was observed in all analyzed post-consumer PP MAP blends, particularly at lower strain rates of 0.1 s^{-1} and 1 s^{-1}. The following film-blowing experiments of the recyclates turned out successfully, and also the mechanical film properties were widely maintained over the ten processing steps (E_p varied between 4.4 mJ and 4.6 mJ, E_t decreased from 882 MPa to 842 MPa in the longitudinal direction and from 632 MPa to 500 MPa in the transverse direction). Despite not having a flawless surface, the manufactured films' mechanical properties were not significantly affected. Better optics would have required a higher degree of purification that would have consumed significant resources, such as energy, chemicals, and water, prior to processing.

Nevertheless, not only the recyclability has to be considered but also the fundamental packaging function, which involves sufficient barrier properties and mechanical stability of the packaging. For that purpose, barrier and mechanical properties of different multi-layer film systems were analyzed and compared to each other.

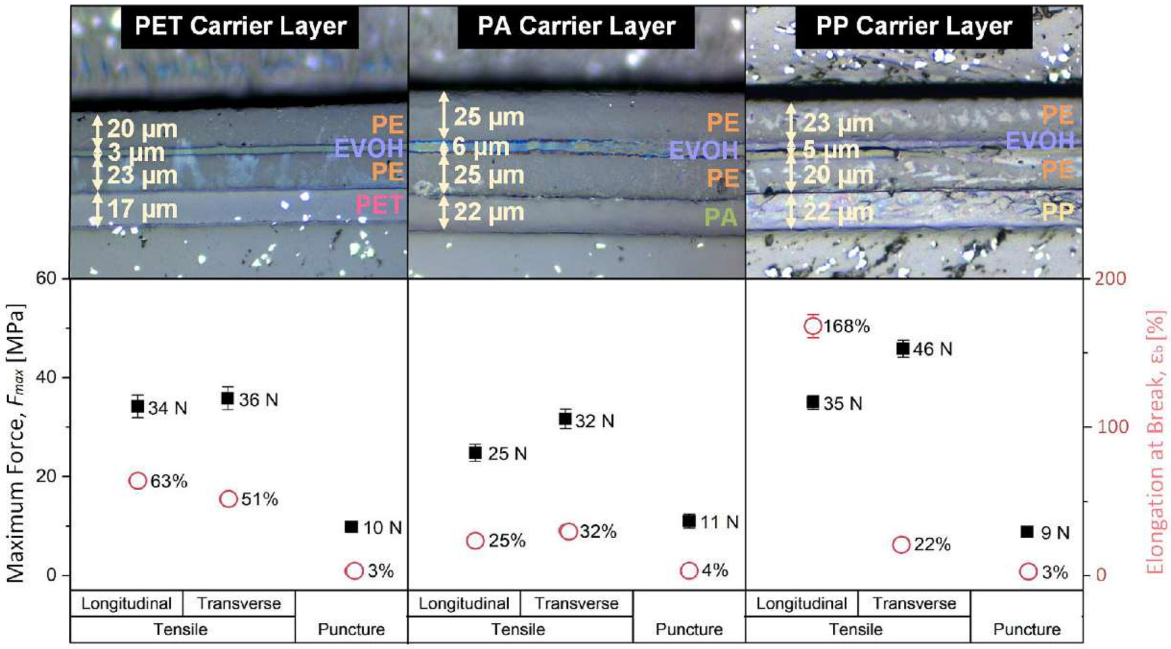


Figure 32: Impact of the carrier layer material (packaging stability) on F_{max} and ε_b (tensile and puncture resistance testing) of multilayer film samples. [121]

Figure 32 compares the impact of different carrier layer materials (PET, PA, PP) laminated to similar barrier layers on the mechanical properties of commercially available MAP lids. PET, PA, and PP all demonstrated an F_{max} of 10 ± 1 N for puncture resistance, although PET was the stiffest among these materials. Improving the puncture resistance of PA and PP can be realized by using mono- or bi-axial film orientation. The ε_b values of the films show significant differences in the longitudinal and transverse measurement directions of the PP carrier layer (168% compared to 22%), which are attributed to the effects of monoaxial film orientation. In common MAP samples, the barrier function of blocking oxygen and moisture can be attributed to a three-layer PE-EVOH-PE composition.

A single PET layer with a thickness of 255 μm (typical carrier layer) provided a WVTR of 2.02 $g/(m^2 \cdot day)$ and an OTR of 13.94 $cm^3/(m^2 \cdot day \cdot bar)$. A combination of 225 μm PET and 35 μm PE resulted in an increased OTR of 18.62 $cm^3/(m^2 \cdot day \cdot bar)$ while the WVTR decreased to 1.36 $g/(m^2 \cdot day)$ as PE is less permeable to moisture than PET alone but more readily transmits oxygen. The presence of only a thin EVOH layer (154 μm PET + 15 μm PE + 5 μm EVOH + 17 μm PE) radically reduced the OTR to 2.16 $cm^3/(m^2 \cdot day \cdot bar)$ while the WVTR remained almost constant at 1.23 $g/(m^2 \cdot day)$.

The combined findings clearly demonstrate that improvement in the recyclability of MAPs could be realized through a combination of polyolefins with EVOH, which is also meeting the functional packaging requirements.

3.2 Other multi-layer packaging (unpublished)

Of all the polymer-based packages analyzed (figure 27), more than 90% contained 2 or more layers. Cosmetic tubes was the only product sector that did not contain any PET but 82 wt.% PE-LLD/LD, 7 wt.% PP, 8 wt.% EVOH, and 3 wt.% OTHER. Packaging from frozen foods only contained a minor portion of PET (4 wt.%) together with 51 wt.% PE-LLD/LD, 42 wt.% PP and 3 wt.% OTHER components. Packaging from snacks and ready-made meals was similar to MAPs but comprised a smaller PET proportion of 25 wt.% next to 45 wt.% PE-LLD/LD, 12 wt.% PP, 5 wt.% EVOH, 4 wt.% OTHER and 9 wt.% PA (highest PA proportion of all packaging sectors). While cosmetic tubes and packaging for frozen foods demonstrated rather homogeneous waste streams, especially not containing high amounts of PET, the whole mixture of all multi-layer packaging resulted in a quite heterogeneous composition which is thus hard to recycle.

Table 23: Composition and layer thicknesses of packaging structures storing similar products but comprising different polymer layers.

Product	Category	PE (LD/LLD)	PET	PP	EVOH	PA
Mixed berries	Frozen	20/8/41 μm	-	-	-	-
Strawberries	Frozen	14/30/14 μm	-	21 μm	-	-
Broccoli	Frozen	17/34/17 μm	-	-	-	-
Peas	Frozen	10/30 μm	15 μm	-	-	-
Grated Mozzarella	Snacks and ready-made	112 μm	-	-	-	-
Grated Pizza cheese	Snacks and ready-made	74 μm	-	-	-	26 μm
Shower gel	Cosmetic tubes	500 μm	-	-	-	-
Shower gel	Cosmetic tubes	272/195 μm	-	-	40 μm	-
Shower gel	Cosmetic tubes	-	-	513 μm	-	-

One of the main reasons leading to heterogeneous recycling streams from multi-layer packaging is the use of various different material combinations, which mainly, but not entirely, supports packaging functionality and product protection. The choice to design packaging for the same or similar products differently is mainly driven by marketing related rather than practical factors. Moreover, dynamic economic factors such as resin, transportation, and energy costs or material availability influence short-term production decisions, which contributes to the generation of more heterogeneous waste streams.

Table 23 compares the various structures (polymer types, layer thicknesses) from packaging storing similar products (frozen vegetables and fruit, grated cheese, shower gel) from different categories (frozen food, snacks and ready-made meals, cosmetic tubes). The shelf life was comparable for all products from the individual categories. Except for the shower gel tube containing only PP copolymer, all packaging samples contained at least 1-3 layers of PE. Even if the packaging is entirely composed of PE, it is not necessarily a mono-material packaging, as can be seen in the examples of frozen food packaging, which contain three individual layers of PE (mixed berries, broccoli). These layers comprise blends of PE-LD and PE-LLD, which are not necessarily the same resin types and could be mixed in a different ratio for each layer. Therefore, in the worst case (from a recycling perspective) a packaging made from PE only could still contain six or more different PE resin types. The other frozen food packaging samples contained additional layers of PET (peas) and PP (strawberries). Overall packaging thicknesses were similar, ranging between 55-68 μm. Grated mozzarella was packed in 112 μm of PE, while grated pizza cheese packaging comprised a combination of PE (74 μm) and PA (26 μm) which again resulted in similar overall thicknesses. Shower gel was either stored in tubes comprising a single PE layer (500 μm), a three-layered PE-EVOH-PE structure (507 μm) or a single layer of PP copolymer (513 μm).

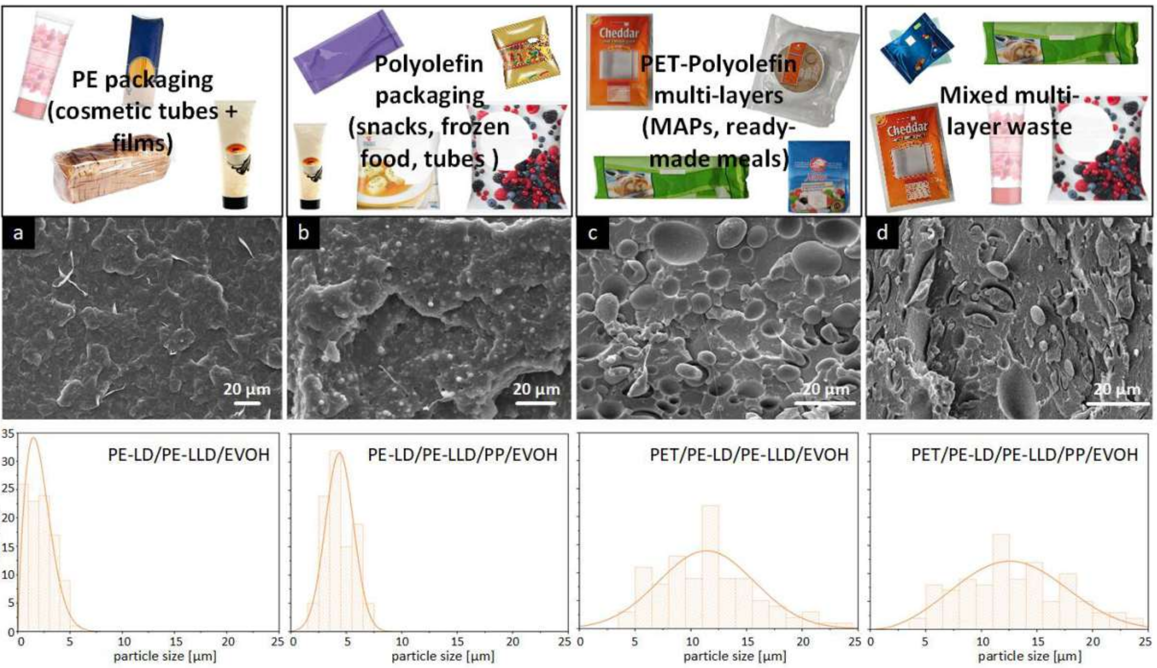
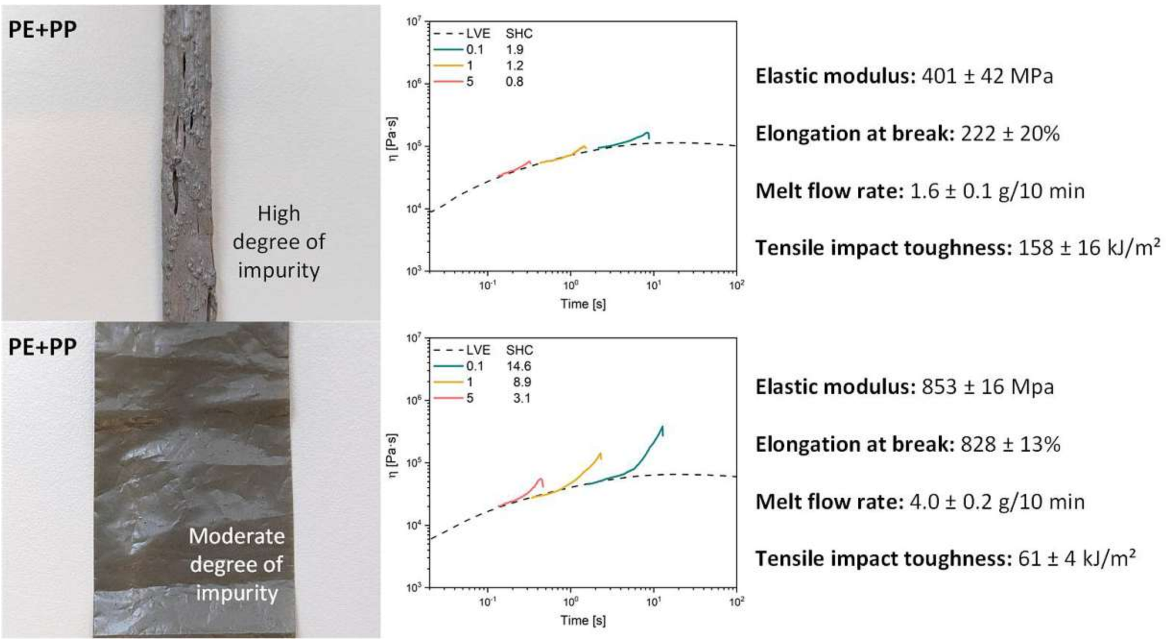


Figure 33: Recyclate morphology of common post-consumer packaging blends comprising (a) PE-LD/PE-LLD/EVOH, (b) PE-LD/PE-LLD/PP/EVOH, (c) PET/PE-LD/PE-LLD/EVOH, (d) PET/PE-LD/PE-LLD/PP/EVOH demonstrated using scanning electron micrographs and particle size distribution curves. - adapted

Despite great similarities, as the structural difference often is only given by one layer, this slight deviation of product design disturbs the recycling process and secondary material homogeneity leading to a great loss in quality. Figure 33 displays recyclate morphologies with their corresponding particle size distribution curves of possible post-consumer blend scenarios from multi-layer packaging. Blends from pure PE packaging (most cosmetic tubes and PE films), containing only EVOH in minor quantities (5 wt.%) as a second component, exhibited finer morphologies and only small (< 5 μm) homogeneously distributed particles compared to blends containing more, structurally different polymers (mixed multi-layer waste). The size and distribution of dispersed particles depend on polarity differences and the viscosity ratio of the blend partners, which led to more convenient conditions to form a homogeneous structure for the PE and polyolefin-based (polyolefin packaging from frozen food, tubes and snacks) blends. These blends demonstrated very narrow particle size distributions with maximum particle sizes of 8 μm. Contrary, the addition of PET (packaging from MAPs and ready-made meals, mixed multi-layer waste) resulted in broad particle size distributions and larger particle sizes with diameters up to 25 μm, indicating lower quality and material brittleness in comparison to the polyolefin-based blends.

Nevertheless, even if similar structures are found, not containing combinations of polyolefins and PET, this is no guarantee for good recyclability because not only polymer-polymer contaminations are problematic, but also those caused by fillers, additives, adhesives, and decorations, which is why their use should be carefully considered.



Elastic modulus: 401 ± 42 MPa

Elongation at break: 222 ± 20%

Melt flow rate: 1.6 ± 0.1 g/10 min

Tensile impact toughness: 158 ± 16 kJ/m²

Elastic modulus: 853 ± 16 Mpa

Elongation at break: 828 ± 13%

Melt flow rate: 4.0 ± 0.2 g/10 min

Tensile impact toughness: 61 ± 4 kJ/m²

Figure 34: Film-blowing samples (left), extensional rheology curves (middle) and physical properties (right) of PE-PP post-consumer packaging blends containing a high and moderate degree of impurities.

Figure 34 compares film-blowing samples, extensional rheology curves, and physical properties of post-consumer packaging PE-PP blends containing a high and moderate degree of impurities. The blends with high degrees of impurities resulted from frozen food packaging, which commonly contained thick coating layers ($\sim$ 6-8 μm), large prints covering the whole packaging film and one layer of a filler-containing polymer such as titanium dioxide or calcium stearate. These blends contained about 75 wt.% of PE and 25 wt.% of PP, which is reflected in the mechanical properties (E_t : 401 MPa, ε_b : 222%, a_{tN} : 158 kJ/m^2) which were comparable to those of virgin PE. Although PE is usually better intended for film-blowing applications, the blends demonstrated hardly any strain hardening behavior with SHC values between 0.8-1.9. As predicted by the rheology experiments, the blends could not be processed into films due to insufficient bubble stability caused by visibly large (1-3 mm) particles in the polymer melt.

Contrary, PE-PP blends from snack packaging contained $\sim$ 30 wt.% PE and 70 wt.% PP but had a lower filler content, fewer prints, and thinner coatings. The mechanical properties were closer to those of virgin PP ($E_t : 853$ MPa, $\varepsilon_b : 828\%$, $a_{tN} : 61$ kJ/m^2) and extensional rheology measurements demonstrated better strain hardening (SHC: 3.1-14.6) compared to the blends from frozen food packaging. Consequently, the blends could be manufactured into homogeneous films demonstrating only small specks ($<$ 1mm).

In packaging design, there is a preference for using lightweight structures and materials to conserve resources and reduce manufacturing costs. However, it is essential to consider that specific contaminants, such as inks from prints, attached labels, and sealing and adhesives, can easily make up to 12 wt.% of the entire packaging and therefore influence not only the recycling process itself but also the appearance and quality of the resulting recyclate.

3.3 The design from recycling dilemma (Publication 3)

During the recycling process of polymers, the high temperatures and shear forces that occur in re-extrusion cause an increase in cross-linking and branching (primary mechanism for PE) as well as chain scission (primary mechanism for PP), leading to changes in molecular weight and weight distribution. This alteration affects the MFR and melt strength, resulting in unfavorable changes in machine parameters such as injection speed and pressure in injection molding to prevent flash which leads to a waste of material. In more severe cases, it can limit polymer processing options and ultimately restrict the range of products and applications for which the polymer mixture can be used.

The changes in MFR and resulting impact on processing options and associated applications make repurposing of recycled polymers a desirable option. This involves utilizing the material property changes in the recycled polymer and using it for a different application for which it is now more suitable rather than its original intended purpose. Repurposing to an application with a longer service life than the original one would also offer a chance to slow down the reintroduction of the material into the recycling loop and subsequently decrease polymer degradation. Unfortunately, the composition of recycling streams poses the greatest challenge that would argue against this approach.

About 90% of recycled materials consist of post-consumer waste, mostly packaging, which is made up of a few selected polymers preferred for this purpose such as PE-LD, PE-HD, PP, and PET. Post-consumer waste is typically dirty and mechanically deformed, making it significantly different from the desired recycled output material. In contrast, post-industrial waste is usually very clean and more easily recycled, even for use in the same application (closed-loop). Repurposing post-consumer input material is a challenging task since it mainly consists of a limited range of polymers designed to meet specific property profiles for a single application. Thus, it is quite challenging to utilize these materials for a different application with different functional requirements. PE-LD and PE-LLD for example, possess mechanical properties that are best suited for film production, making it challenging to use them for other applications beyond their original purpose, such as packaging. Currently closed-loop systems in packaging remain limited to PET-to-PET bottle recycling and PE-HD milk containers in certain countries.

Closed-loop recycling is undoubtedly the desired option for waste treatment, but color and odor problems in recycled polyolefins coupled with legal requirements associated with food packaging and reductions in their mechanical properties typically make this impossible. The migration of volatile organic compounds into the polymer matrix limits the usability of the recyclate for products that require light colors or in which the migrated substances could demonstrate a risk for human health. To address unpleasant odors, washing procedures present a possible purification option. However, these washing procedures commonly do not further improve the mechanical properties of the material, making their inclusion less viable from an economic and ecological perspective.

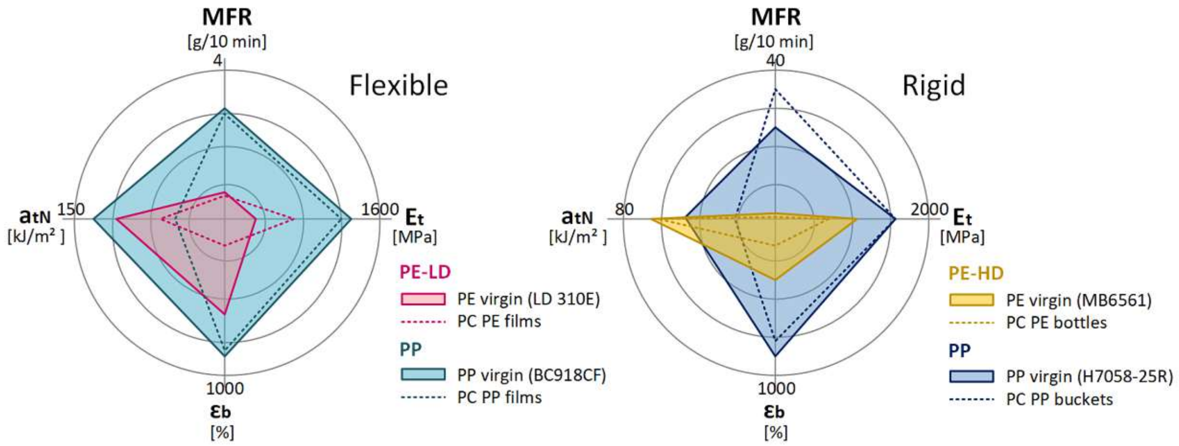
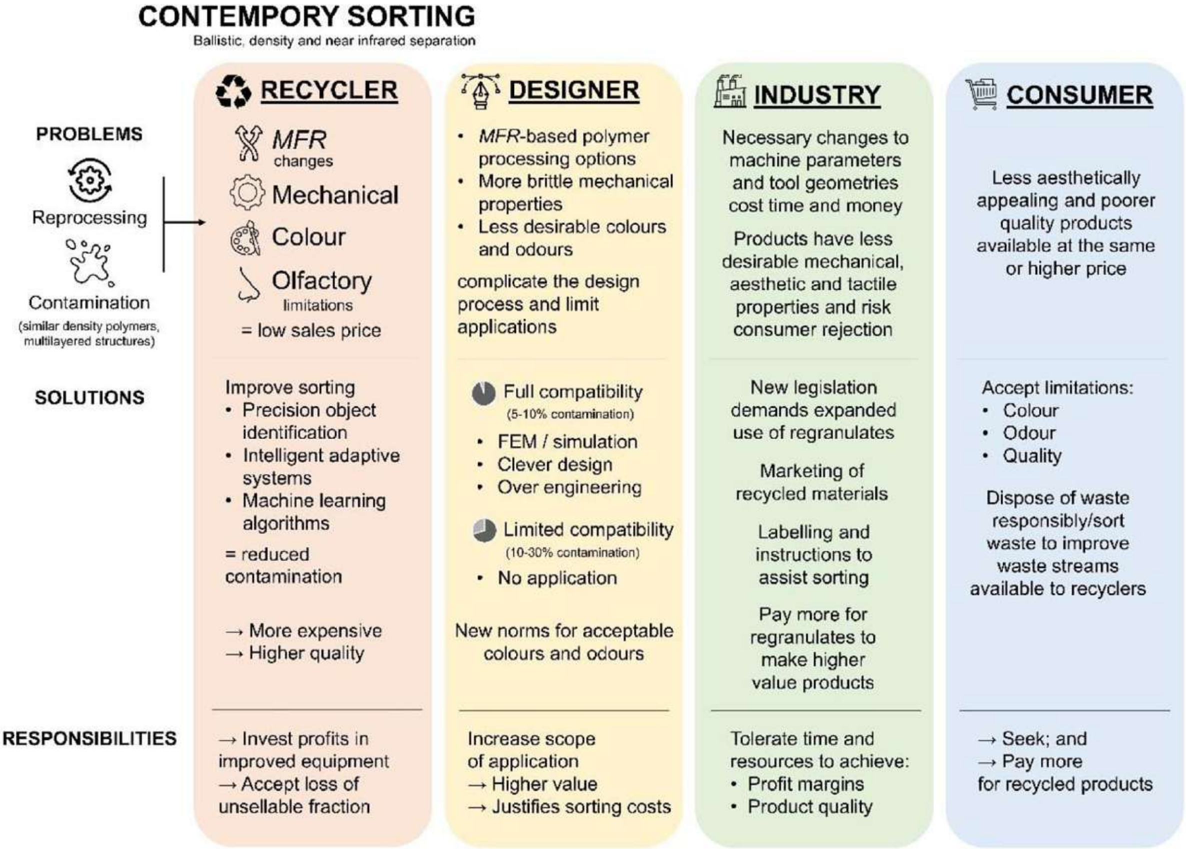


Figure 35: Property profiles of virgin (PE-LD, PE-HD, PP copolymer, PP homopolymer) and PC (post-consumer) (flexible, rigid) material samples - adapted from [174]

Figure 35 demonstrates that even recyclate properties from single material waste streams deviate from those of virgin resins for the same application which makes the design of secondary product applications challenging. Next to processing and application difficulties, consumer perception is one of the greatest issues hindering the use of recyclates. Or perhaps companies are hesitant to use recycled materials in their products due to concerns about reduced material properties, less desirable colors, or odors, fearing that consumers may reject them. However, this fear may be unfounded, as environmentally friendly and recycled products currently have an exceptionally positive image in society.

Designers play an essential role in shaping the image and function of products, which ultimately determines their success. While marketing can influence consumer perceptions, the look and performance of a product are the most important factors. Designers can also make a product seem "cool" by creating innovative designs that challenge traditional norms and set new standards for acceptable color variations, odors, and material properties. As we move towards a more sustainable future, designers must be willing to embrace the imperfections of recycled materials and showcase their environmental benefits. Design from recycling and design for recycling are closely related. When products are designed to be easily sorted and recycled, it benefits the design from recycling stage by producing higher quality recycled materials with fewer impurities and better mechanical properties. This makes the design process easier. The diversification of the material stream is also important. If designers want to work with specific polymers during the design from recycling stage, they must introduce those polymers into circulation during the design stage.



CONTEMPORY SORTING
Ballistic, density and near infrared separation

	RECYCLER	**DESIGNER**	**INDUSTRY**	**CONSUMER**
PROBLEMS Reprocessing Contamination (similar density polymers, multilayered structures)	*MFR* changes Mechanical Colour Olfactory limitations = low sales price	• *MFR*-based polymer processing options • More brittle mechanical properties • Less desirable colours and odours complicate the design process and limit applications	Necessary changes to machine parameters and tool geometries cost time and money Products have less desirable mechanical, aesthetic and tactile properties and risk consumer rejection	Less aesthetically appealing and poorer quality products available at the same or higher price
SOLUTIONS	Improve sorting • Precision object identification • Intelligent adaptive systems • Machine learning algorithms = reduced contamination → More expensive → Higher quality	Full compatibility (5-10% contamination) • FEM / simulation • Clever design • Over engineering Limited compatibility (10-30% contamination) • No application New norms for acceptable colours and odours	New legislation demands expanded use of regranulates Marketing of recycled materials Labelling and instructions to assist sorting Pay more for regranulates to make higher value products	Accept limitations: • Colour • Odour • Quality Dispose of waste responsibly/sort waste to improve waste streams available to recyclers
RESPONSIBILITIES	→ Invest profits in improved equipment → Accept loss of unsellable fraction	Increase scope of application → Higher value → Justifies sorting costs	Tolerate time and resources to achieve: • Profit margins • Product quality	→ Seek; and → Pay more for recycled products

Figure 36: Interlinked and co-dependent problems, solutions and responsibilities of recyclers, designers, manufacturers (industry) and consumers currently hindering design from recycling practices [174].

However, contamination is currently the main issue in recycling streams, limiting the usability of recycled materials for design from recycling. Effective use of recyclates for secondary applications relies on mixtures containing less than 5-10% of impurities. Considering the common practice of blending PE-LD with PE-LLD for improved process stability, inseparable multi-layer films (including adhesives), the frequent use of fillers, the necessity of EVOH as an oxygen barrier for sensitive products in addition to various decorations (e.g., labels or prints) and potential migration products from food residues, this degree of purity is challenging to realise. Improved sorting and recycling practices are essential in achieving better quality recycled polymer mixtures for designers. Current sorting technology is limited and more sophisticated technologies are under development, but they require greater market demand for recycled polymers and higher product values to compensate for additional costs. Designers are key drivers in this process but are hesitant due to the inferior quality of recycled materials compared to virgin polymers (figure 36). Future policy and legislation should focus on establishing and adopting new protocols and standards to reduce contamination levels to less than 5% considering product re-designs, limitation of material combinations and improvement in sorting accuracy.

4 Conclusion and Outlook

Multi-layer packaging as it is currently designed has no chance to be recyclable without the rapid development of novel viable separation and sorting techniques. The broad variety of packaging materials used in multi-layers, inevitably leads to the formation of incompatible polymer blends during the recycling process. This results in unpredictable changes to the property profile, with variations greater than 10% observed for MFR, E_t, ε_b, and a_{tN} when compared to the individual polymers. Even the analyzed single- waste fractions or separated MAP lids and trays, which individually resulted in similar waste compositions predominantly demonstrated a variation in physical properties, of at least 5%, compared to virgin references. It is possible that four or more different PE-LD/PE-LLD types are present in an individual multi-layer packaging film. In many multi-layers there are two or more PE layers, which do not necessarily have to contain the same PE types. However, the fact that PE is predominantly found as a blend of PE-LD and PE-LLD with varying ratios in multi-layer packaging makes it difficult to predict the properties of the recyclate, since the materials differ not only in their chain structure but also in their mechanical and melt properties. Such property variations were even observed solely for the mixture of PE-LD and PE-LD, which is a common practice to improve processability or bubble stability of PE films during film blowing.

In the scope of this thesis, around 240 different packages from the categories sliced cheese and sausage, frozen food, cosmetic tubes and snacks and ready-made meals were analyzed, from which more than 90% contained two or more layers and subsequently different material combinations. Unfortunately, the use of different materials is not only justified by functional requirements, but also a result of design and marketing specifications (shape, print, surface texture, transparency). This results in the availability of various, structurally different, types of packaging for the same product. Especially concerning that, as soon as PET is combined with polyolefins, which is the case for all investigated packaging categories except cosmetic tubes, a sharp ductile to brittle transition is observed ($\varepsilon_b < 5\%$), the necessity of such a broad packaging variety should be critically questioned. Besides the inhomogeneous morphology (strong phase separation with particles > 20 µm) of incompatible PET/polyolefin blends and their subsequent loss in mechanical strength, further deterioration of the polyolefin proportion (chain scission and branching reactions) is promoted by the required (> 250 °C) extrusion temperatures during the mechanical recycling process.

If two or more different polymers are blended, they often have different molecular structures, polarities, and other characteristics that can lead to poor mixing and phase separation in the final blend. Compatibilizers commonly work by having segments that can interact with all or at least two polymers in the blend, promoting adhesion and reducing interfacial tension. The specific interactions that are required for bonding are also influenced by the polymer ratios. If the polymer ratios change, the overall composition of the blend changes, affecting the types and strengths of intermolecular interactions. Polymer ratios within recyclates might greatly vary and therefore significantly complicate the selection and dosing of an appropriate compatibilizer.

Of course, this work does not consider food technical aspects in any way, which is why it is difficult to make a statement about the exact necessary design of food packaging. Especially fresh products may have very different requirements for packaging (e.g., marginal differences between Gouda and Cheddar cheese), but the need for different packaging types for cosmetic tubes may most likely depend more on coincidence or marketing aspects, such as conserving the smell of heavily scented products. Next to heterogeneous design of polymer based multi-layer packaging, that are hardly suitable for recycling, popular hybrid structures, combining polymers with other materials, introduce contaminants like aluminium (coatings) or paper (labels and compounds) into the waste stream. These contaminants do not only alter the physical properties of the resulting recyclates but also lead to color changes (mostly gray or dark) and bad smell. This clearly further limits the applicability of the resulting secondary material.

The potential concern that improving the recyclability of multi-layer films can only work at the expense of a decrease in packaging efficiency, such as a shorter shelf-life, is probably baseless. The packaging analysis demonstrated that structures which do not combine PET and polyolefins are already commercially available in various product areas but unfortunately not exclusively. PP-based 'Berger' MAPs were an excellent example for recyclable multi-layer structures as the reprocessed individual lids and trays and their combination closely resembled the property profile of virgin PP copolymer. Further, ten-times reprocessed recyclates could be successfully manufactured into films demonstrating the functional possibility of closed-loop recycling. More of these design-for recycling approaches in product composition are urgently needed, as efficient compatibilization of mixed polymer waste is hardly possible.

While some multi-layer packaging is almost impossible to replace, others are just being invented for marketing reasons. For example, it will be almost impossible to store coffee without the oxygen and aroma blocking properties of EVOH or aluminium layers. Alternatively, containers made of other metals or ceramics could be used, which would in turn have an unfavorable impact on the overall product life cycle assessment due to the higher weight and or manufacturing effort (e.g., energy consumtion during processing). On the other hand, paper-polymer composites are often sold as particularly sustainable because they save on plastic consumption. This may be true, as long as the materials can be easily separated and are actually separated. The paper-polymer trays, commonly used to replace the PET-PE MAP trays, comprise several PE-coated paper layers and an additional EVOH barrier which are laminated together. The result is a polymer content of >20 wt.% in these trays which makes them neither suitable for disposal in a paper waste stream nor in a polymer waste stream due to the high amount of contaminants for each case. Such packaging designs are literally green washing and do not benefit an increase in recycling rates.

Currently, it is clearly impossible to meet the narrow property range of product demands, with the majority of recyclates, for which the individual materials of the multi-layers were originally designed for. However, the small proportion of polyolefin-only packaging (e.g., Berger ham) indicates the recycling potential that can be realized within the investigated waste fractions if the packaging is appropriately designed for recycling. The missing link between design for and design from recycling can likely be achieved through holistic design choices, careful material selection based on legal guidelines, and adaption of profit margins, processes, and product appearance. Harmonization of products and processes will be essential for the circularity of multi-layers making them a valuable source of material in the future.

This thesis presented a holistic method for evaluating some of the main limitations and possibilities concerning the design and mechanical recycling of multi-layer packaging. The following future perspective presents possible avenues for further research to advance sustainable packaging practices.

One possible direction for future work is to continue exploring more efficient purification methods on a lab scale, such as melt filtration and degassing, to improve the quality and purity of recycled materials. These methods can help to remove residues, increase the yield of recycled materials, and make the mechanical recycling process more viable.

Recycling simulations of alternative designs on an industrial scale will also be important to ensure the feasibility and sustainability of new designs. These simulations can be carried out in cooperation with industry partners who are willing to process such packaging materials, and will allow for a detailed analysis of the potential barriers to implementation. As a specific example, film-to-film recycling of PP-based Berger ham on an industrial scale, similar to what was done on a laboratory scale, would be interesting. Due to strict food regulations, the use of recycled film for direct food contact would currently not be realistic, but the use as secondary packaging or middle layer between virgin materials is conceivable.

Future work could also focus on the specific evaluation of the impact from additives, fillers, and decoration on the physical properties of the recyclates. This will also involve analysing the potential environmental impact of these materials, and identifying ways to reduce their negative effects.

Finally, it would be important to consider some essential points during the development of new packaging products. Packaging functionality and recyclability need to be the focus to make packaging more sustainable. Whenever possible, the use of mono-layers or at least polyolefin-only or PET-only solutions is highly recommended to prevent poor quality recyclate mixtures. If improved barrier is necessary, preferably EVOH or alternatively aluminium layers need to be used as sparingly as possible but should in any case not exceed 3-5 wt.% of the whole packaging. For the labels, polymers made of the same material as the packaging itself should be preferred over paper to prevent contamination and the used adhesives should be removable with current purifying practices (NaOH in combination with hot or cold wash).

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Appendices

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Polyethylene terephthalate based multilayer food packaging: Deterioration effects during mechanical recycling

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ABSTRACT

Modified atmosphere packaging (MAP) comprises a multilayer structure of polymers and adhesives offering excellent protection to food products. While this widely used structure improves shelf life, its recyclability is restricted by polymer incompatibilities and difficulty separating individual layers during sorting, which limits the viability of this prominent waste stream for 2030 European Union (EU) reuse and recycling targets. This study assessed the recycling potential of MAPs by system part and functional layer based on composition, component miscibility and temperature and incompatibility induced changes in physical and mechanical properties. Lids and trays alone exhibited similar compositions, but when combined their PET (polyethylene terephthalate)-PE (polyethylene) ratios varied considerably resulting in embrittlement and reduced toughness indicating limited compatibility and recyclability. Although PET-PE immiscibility resulted in phase separation causing inhomogeneity induced loss in ductility and toughness, barrier structures exhibited better phase adhesion and more homogeneous morphologies. Nonetheless, the PET dictated processing temperature, greatly exceeding that of PE, promoted cross-linking effects. The barrier and carrier layers of current MAPs must be delaminated within waste sorting processes to be recyclable. More economic and ecological recycling of MAPs to meet EU targets will require designs with more compatible polymer components.

1. Introduction

A major application of polymers is the preservation of food products with converter demands of 20.3 tons (of 50.7 tons overall market demand) for the packaging sector in the European Union (PlasticsEurope, 2021). Modified atmosphere packaging (MAP), widely described as a sealed two component lid/tray system comprising multiple joined material layers that enclose foods with modified gas atmosphere, extends the shelf life of sensitive foods by up to several weeks or even months compared to products exposed to unmodified atmosphere (Barukčić et al. 2020; Kargwal, Garg, Singh, Garg, & Kumar, 2020). Packaging functionality depends on the maintenance of the modified atmosphere for as long as possible, hindering gas diffusion through the material and damage from external forces (puncture, tearing). This is realized through a layered composition of structurally distinct polymer films (Chen et al., 2020; Rossaint, Klausmann, Herbert, & Kreyenschmidt, 2014).

While this layered composition provides improved product protection during service life it does make the recycling of MAPs in line with product sustainability criteria very challenging. MAP packaging typically comprises low density polyethylene (PE-LD), linear low density polyethylene (PE-LLD), polyamide (PA), polypropylene (PP), ethylene vinyl alcohol (EVOH) and polyethylene terephthalate (PET). These constituents are not compatible with each other due to differences in chemical structure. Additionally, individual layers cannot be separated in an economically viable manner using current industrial sorting techniques as such separation requires chemical dissolution of the adhesives (Walker et al., 2020). MAPs are commonly rejected for mechanical recycling, the preferred choice for closed loop recycling (primary recycling), as reprocessing without specific additives to improve compatibility leads to considerable deterioration of material properties (secondary recycling or downcycling). MAPs are subsequently primarily used for energy recovery (quaternary recycling) (Schyns & Shaver, 2021).

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New government regulations will dictate the use of secondary materials in new products and recycling quotas of 55% for the packaging sector by 2030, clearly demonstrating the need for new design and recycling strategies (European Commission Directive, 2018). Recent studies on the mechanical recyclability of multilayer films have focussed on the compatibilization of PET/PA-PE/PP in different ratios (Akshaya, Palaniappan, Sowmya, Rasana, & Jayanarayanan, 2020; Czarnecka-Komorowska, Nowak-Grzebyta, Gawdzińska, Mysiukiewicz, & Tomasik, 2021; Nomura et al., 2020). These films often contain considerable quantities of additives (> 5 wt%) with improvements in the material limited to targeted mechanical properties such as toughness, rather than homogenization of the mixture, which is important for the production of recyclate with consistent material composition and properties (Maris et al. 2018). The use of these additives has an enormous environmental impact on the sustainability of mechanical recycling through consumption of energy during material transport and synthesis, dependence on non-renewable petroleum resources and groundwater contamination with hazardous substances, such as phosphorus (Gu, Guo, Zhang, Summers, & Hall, 2017). Studies focussing on polymer compatibility widely investigate binary blend systems with and without compatibilizers but do not consider all MAP constituents in their relevant ratios as they exist in commercial packaging. They therefore do not provide an accurate representation of the material mixture that recyclers receive for reprocessing. The impact of EVOH on PE, PP or PET has also only been studied in the context of barrier properties rather than blend homogeneity and recyclability (Ge, Fortuna, Lei, & Lu, 2016; Horodytska, Valdés, & Fullana, 2018).

This study evaluated the mechanical recycling potential of MAPs. Polymer blends based on a preliminary structural study of 60 post consumer MAPs, representative of the Austrian product market, were produced and processed in line with common industrial recycling practices and their mechanical, rheological and morphological material properties analysed. Physical properties such as viscosity or impact toughness of blends, representing system parts (lid, tray) were compared to a PET reference, which constituted the largest polymeric mass fraction in the MAPs, to evaluate possible advantages in lid and tray separation during mechanical recycling. Mechanical (tensile, puncture) and barrier (water vapor, oxygen) testing was also completed on the functional layers (carrier, barrier) of the lid and tray films. The impact of variation in processing temperature and component miscibility on blends representing the functional layers was finally assessed to isolate the key factors contributing to specific deterioration effects, such as changes in structural and physical properties.

2. Materials and methods

2.1. Materials

60 individual post-consumer MAPs (30 sausage/cheese packaging) were collected from household bins in eastern Austria to investigate the polymeric constituents of typical MAP waste streams. Virgin food grade PET (Invista Polyclear refresh 1101), PE-LLD (Ineos LL6208AF, MFR = 0.9 g/10 min), PE-LD (Dow Agility EC7000, MFR = 3.9 g/10 min) and EVOH (Soarnol ET3803RB) with 38 mol%) were used for the production of reference blends representative of the waste stream. PET represented the carrier layers while various ratios of PE-LLD, PE-LD and EVOH were used to represent the barrier layers. All materials were intended for the manufacturing of packaging films and used as received.

2.2. Production of reference blends and test specimens for rheology and mechanical property evaluation

Polymer reference blends (Table 1) were designed to represent the system parts (lid, tray) based on the mean polymer mass fractions determined during post-consumer MAP polymeric constituent analysis. Further analysis of lid and tray mechanical and barrier properties prompted a categorical division into functional layers (carrier, barrier). An Extron single screw extruder (EX-18–26–1.5, Extron Engineering Oy, Toijala, Finland) with three individual heating zones, a screw diameter of 18 mm and a length/diameter ratio of 25:1 was used to melt blend manually pre-mixed virgin polymer resins. The screw speed was 70 rpm and the processing temperature was varied to simulate pure polyolefin ($T_{Polyolefin}$: 220 °C) and combined polyolefin/polyester ($T_{Polyester}$: 275 °C) waste streams.

Extruded polymer strands were shredded using a universal cutting mill (Pulverisette19, Fritsch, Idar-Oberstein, Germany) equipped with a 4 mm sieve insert prior to further processing. Tensile and tensile impact test specimens were produced using a Haake Mini Lab II twin screw extruder coupled with a Haake Mini Jet II injection molding unit (Thermo Fisher Scientific, Waltham, MA, USA). Extrusion temperatures were selected based on compounding temperatures (220/275 °C) with a screw speed of 100 rpm, a mold temperature of 40 °C and pressure of 350 bar (10 s injection time). Dynamic shear rheology specimens were produced using compression molding (Collin P 200 P, Maitenbeth, Germany) at extrusion temperatures of 220/275 °C and a pressure of 100 bar. Punched aluminum frames sandwiched between steel plates and separated by Teflon® sheets were used to generate discs 25 mm in diameter and 1.2 mm thick.

2.3. Analysis of the polymeric constituents of the post-consumer MAPs

Inner (food contact) and outer (carrier) MAP layers were investigated using a Brucker Tensor 27 Fourier-transform infrared (FT-IR) spectrometer (Billerica, MA, USA) equipped with an attenuated total reflection (ATR) diamond (DuraSample IR II) with single reflection. 16 scans were conducted between 600 and 4000 cm^{-1} with a resolution of 4 cm^{-1}. This is in line with industrially applied sorting techniques (Kumagai, Suyama, Sato, Amano, & Ogawa, 2002).

Table 1
Composition of PE-LD or PE-LLD rich reference blends representing disassembled packaging structures comprising virgin PET, PE-LD, PE-LLD and EVOH based on the analysis of the post-consumer MAPs. Extrusion temperature variation simulated reprocessing within a pure polyolefin ($T_{Polyolefin}$ = 220 °C) or a combined polyolefin/polyester ($T_{Polyester}$ = 275 °C) waste stream.

Composition				Blend model		Extrusion	
PE-LD	PE-LLD	EVOH	PET	MAP component	Representation	$T_{Polyolefin}$	$T_{Polyester}$
17.6 wt%	4.4 wt%	3.0 wt%	75.0 wt%	‘Lid + Tray Blend’	Overall MAP composition	–	275 °C
4.4 wt%	17.6 wt%	3.0 wt%	75.0 wt%			–	275 °C
8.0 wt%	2.0 wt%	3.0 wt%	87.0 wt%	‘Tray Blend’	Composition of trays	–	275 °C
2.0 wt%	8.0 wt%	3.0 wt%	87.0 wt%			–	275 °C
34.2 wt%	19.8 wt%	3.0 wt%	43.0 wt%	‘Lid Blend’	Composition of lids	–	275 °C
19.8 wt%	34.2 wt%	3.0 wt%	43.0 wt%			–	275 °C
78.5 wt%	18.5 wt%	3.0 wt%	–	‘Barrier Blend’	Barrier structures (with influence of EVOH)	220 °C	275 °C
18.5 wt%	78.5 wt%	3.0 wt%	–			220 °C	275 °C
80.0 wt%	20.0 wt%	–	–	‘LLD/LD 20/80′	Barrier structures (without influence of EVOH)	220 °C	275 °C
20.0 wt%	80.0 wt%	–	–	‘LLD/LD 80/20′		220 °C	275 °C

Differential scanning calorimetry (DSC) of the films was completed with a TA-Instruments TA Q2000 instrument (Newcastle, DE, USA) and used to supplement the FT-IR results, which cannot detect the presence of polymers located within the middle layers of the films. Unique peaks were identified for each meltable component present in the system using changes in heat flow attributable to the melting of spherulites within the crystalline polymer region. This also allows distinctions between different types of the same polymer, such as PE-LD and PE-LLD (Grellmann & Seidler, 2013). Samples with a mass of 5–6 mg were deposited into aluminum pans, heated from 0 °C to 300 °C, cooled to 0 °C and reheated to 300 °C at a rate of 10 K/min under a nitrogen atmosphere.

Mass content was estimated using volumetric mass density. The layer thicknesses of film cross sections were measured using a Zeiss Axio Imager M2m light microscope (Oberkochen, Germany). 20 × 12 mm samples were cut from the lids and trays, cleaned with ethanol, placed in a sample holder and embedded with a two-component epoxy resin (Araldite AY103 + REN HY956). The cured samples were ground and polished using a Struers TegraForce-31 polisher (Copenhagen, Denmark).

2.4. MAP barrier property analysis

Water vapor transmission rate (WVTR) tests were performed from the inside of each sample out and oxygen transmission rate (OTR) in the opposite direction. WVTR testing was completed at 23 °C with a relative humidity of 85% (PERMATRAN-W 3/34 G, Mocon, Minneapolis, MN, USA) in accordance with ISO 15106–2 (International Organization for Standardization, 2005). OTR was assessed in accordance with ISO 15105–2 (International Organization for Standardization, 2003) at 23 °C and a relative humidity of 0% (GDP-C, Brugger Feinmechanik GmbH, Munich, Germany).

2.5. Rheology of the reference blends

Characterization of the polymer melts was completed using dynamic shear rheology measurements and melt flow rate (*MFR*) at constant shear. Dynamic shear rheology was achieved using frequency sweeps on an MCR 302 rheometer (Anton Paar, Graz, Austria) equipped with a plate-plate system (1 mm gap size) and a heating hood purged with nitrogen. Temperature was constant (220/275 °C) during the experiments while deformation was raised logarithmically from 1% to 2% at a frequency ranging from 628 rad/s to 0.01 rad/s. MFR measurements were performed in accordance with ISO 1133–1 (International Organization for Standardization, 2011) under a load of 2.16 kg and temperatures of 220/275 °C on a MeltFloW basic device (Karg Industrietechnik, Krailling, Germany). Shear rate values (γ) were calculated based on the *MFR* using Eq. (1) (Rudin & Chang, 1978).

$$\gamma \left[1 \Big/ \mathrm{s}\right] = 1.83 \times \frac{MFR\ [\mathrm{g}/10\ \mathrm{min}]}{\rho_m\ [\mathrm{g}/\mathrm{cm}^3]} \qquad (1)$$

2.6. Mechanical properties of the reference blends and MAP films

Tensile impact strength test specimens measuring 60 × 10 × 1 mm were notched on both sides with a Vis-Notch (Ceast, Darmstadt, Germany) device and tested using an Instron 9050 (2 J hammer and 15 g crosshead mass; Ceast, Darmstadt, Germany) pendulum in accordance with ISO 8256/1 A (International Organization for Standardization, 2004). Tensile testing was performed to ISO 527–2-A5 (International Organization for Standardization, 2012) at a speed of 10 mm/min using a universal testing machine (ZwickRoell Z050, Ulm, Germany) equipped with a 1 kN load cell and an extensometer.

Mechanical film properties were characterized through tensile testing in accordance with ISO 527–3 (International Organization for Standardization, 2018) using 100 × 10 mm test strips (70 mm clamping length) and puncture resistance testing in accordance with DIN EN 14477 (DIN EN 14477, 2004) using round samples at a crosshead speed of 10 mm/min.

2.7. Morphological characterization of the reference blends

The fractured surfaces of the tensile impact test specimens were investigated using scanning electron microscopy (SEM) with an FEI Philips XL30 microscope (Hillsboro, OR, USA). Samples were coated with gold (Agar Sputter Coater B7340, Essex, UK) prior to imaging.

3. Results and discussion

3.1. Polymeric constituents and uniformity of the post-consumer MAP waste stream

Combined DSC, FT-IR and light microscopy analysis of MAPs revealed a three-layer co-extrusion PE-EVOH-PE structure in most samples laminated to a thick (> 100 μm for trays) or thin (< 30 μm for lids) PET layer (Fig. 1). PE was present either as a PE-LD or PE-LLD rich blend as demonstrated through melting peak separation and peak height variation in the DSC thermograms.

Sausage- (SP) and cheese- (CP) MAPs contained 71 wt% PET, 19 wt %. PE (PE-LD and PE-LLD), 3 wt% EVOH, 3 wt% adhesives and 4 wt% other minor constituents, such as paper board, polypropylene and polyamide (Fig. 2).

SP trays contained 84 wt% PET, 8 wt% PE, 2 wt% EVOH and 4 wt% adhesives in addition to other minor constituents. The same constituents were identified in CP trays which contained 83 wt% PET, 11 wt% PE, 1 wt% EVOH, 2 wt% adhesives and 3 wt% other minor constituents. Waste streams generated from trays, which represent ~80% of the packaging mass, were subsequently nearly identical.

Lids exhibited more than four times the PE content of trays (50 wt% compared to 8 wt% for SP and 46 wt% compared to 11 wt% for CP) and approximately half the PET content (33 wt% compared to 84 wt% for SP and 43 wt% compared to 83 wt% for CP). The mass fractions of EVOH (7 wt% for SP compared to 5 wt% for CP), adhesives (6 wt% for SP compared to 4 wt% for CP) and other components (4 wt% for SP compared to 2 wt% for CP) were slightly higher in SP compared to CP. The PE (50 wt% for SP compared to 46 wt% for CP) and PET (33 wt% for SP compared to 43 wt% for CP) contents in SP and CP nevertheless make the lid waste streams similar in composition.

SP and CP as individual system parts (lid, tray) have similar multilayer structures with the potential to form a uniform waste stream (based on polymer types and layer thicknesses), which is important for mechanical recyclability as the probability of contamination from compositional variations in the waste stream is reduced. However, when combined to form a single packaging system the variation in the ratio of the main polymeric constituents, which is ~48/38 wt% PE/PET for lids and ~10/84 wt% for trays results in a more heterogeneous and hence less easily recycled waste stream.

3.2. Recycling potential of MAPs by system part (lid, tray) and functional layer (carrier/barrier)

Design for recycling strategies aim to establish a balance in essential product function, which for MAPs is the protection of food against external forces and gas migration, and maximized recyclability, commonly acknowledged to be characterized by deviations in physical properties, such as toughness, ductility or melt strength during recycling compared to virgin materials. Consideration of the connection between function claims involving barrier properties for MAPs and potential recyclability is especially important not to exclude closed loop recycling in advance. (Demets et al. 2021).

A direct comparison of the stiffness (E_t), ductility (ε_b), flowability (as described using *MFR*) and impact toughness (a_{tN}) of the investigated blends (lid, tray, lid+tray), representative of the various MAP system

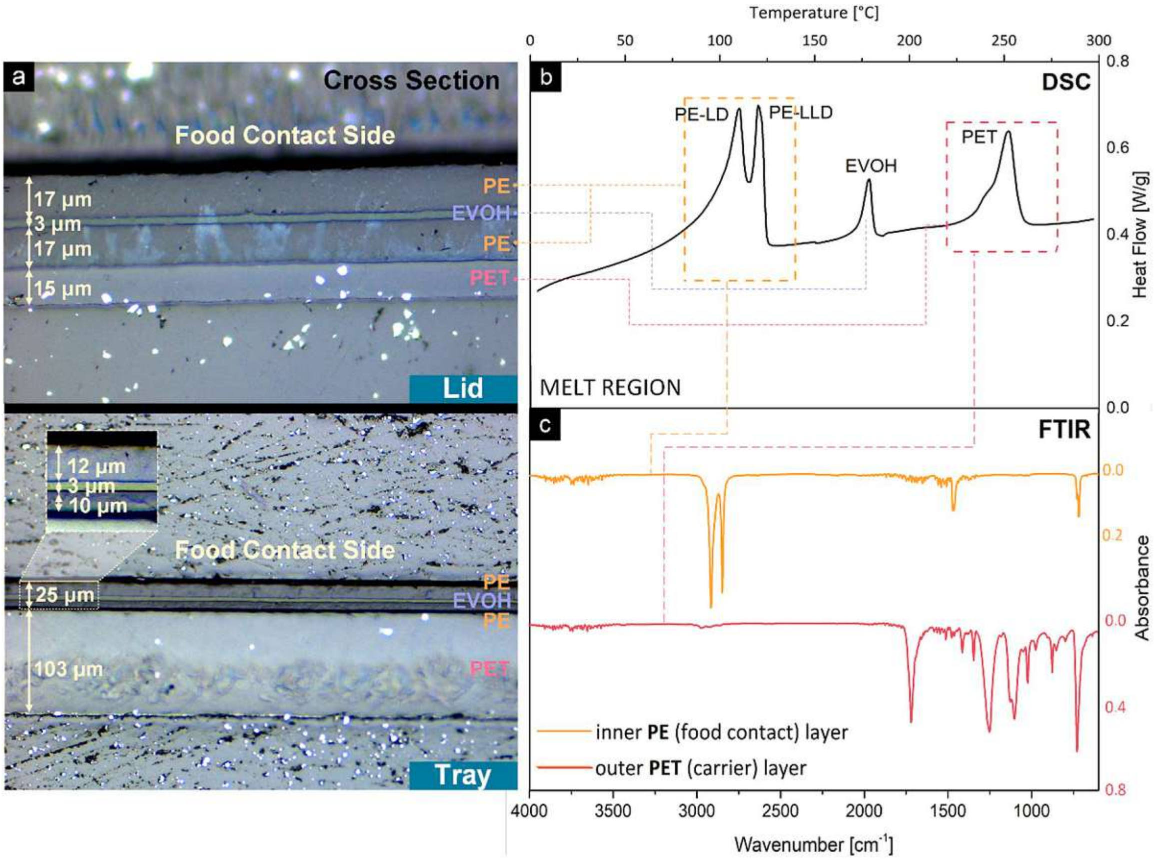

Fig. 1. Representative samples of the most common MAP structures characterized by (a) light microscopy depicting cross sections, (b) DSC demonstrating distinctions between the polymers based on melting temperature and (c) FT-IR spectroscopy identifying inner (food contact) and outer (carrier) layers.

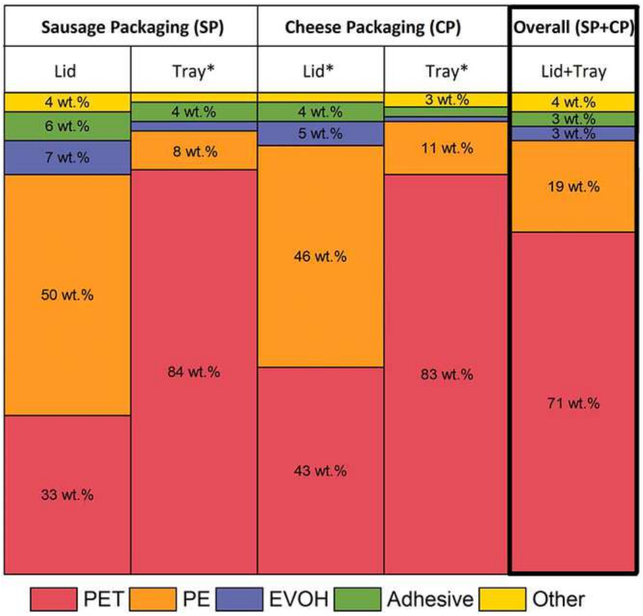

Fig. 2. MAP waste stream composition (PET, PE, EVOH, adhesive and 'Other' minor constituents) derived from structure analysis separated by product type (SP/CP) and system component (lid/tray). *Components ≤ 2 wt% could not be labelled (SP trays and CP lids: 2 wt% paper board, PA and PP; SP trays: 2 wt% EVOH; CP trays: 1 wt% EVOH and 2 wt% adhesives).

parts, is shown in Fig. 3. The mechanical properties and *MFR* of PE-LLD rich tray blends, containing the least PE based contamination, most closely resembled those of PET with deviations in *MFR*, E_t and a_{tN} < 10% (*MFR*: 23 g/10 min compared to 21 g/10 min, E_t: 2328 MPa compared to 2113 MPa, a_{tN}: 58 kJ/m^2 compared to 56 kJ/m^2). However, the ε_b decreased from 324% (virgin PET) to 152% (PE-LLD rich tray blend), most likely due to the immiscibility of PET and PE (Delva, Deceur, van Damme, & Ragaert, 2017). Further increases in the PE content of lid and lid+tray blends promoted deterioration effects and the highest percentage deviation from the PET reference for E_t (PE-LD rich: 52%, PE-LLD rich: 58%), MFR (PE-LD rich: 35%, PE-LLD rich: 78%), a_{tN} (PE-LD rich: 38%, PE-LLD rich: 28%) and ε_b (PE-LD rich: 85%, PE-LLD rich: 76%) in lid blends. An extreme ductile to brittle transition was observed for PE-LD rich blends with ε_b values ≤ 50%. This clearly demonstrated that the mechanical properties of most MAPs deviated considerably from the reference properties of PET irrespective of whether they had a higher (tray) or lower (lid) ratio of incompatible PET-PE constituents. This effect would be exacerbated by further impurities, such as acrylic or polyurethane based adhesives (Aznar et al. 2011), food residues and inks common in real-life waste streams.

While most carrier layers comprised PET, rare exceptions included PA and PP. PET, PA and PP all demonstrated an F_{max} of 10 ± 1 N for puncture resistance, although PET was the stiffest amongst these materials. The puncture resistance of PA and PP can be improved through mono- or biaxial film orientation. The effects of monoaxial film orientation are visible in the ε_b values of the films, which differ notably in the longitudinal and transverse measurement directions of the PP carrier layer (168% compared to 22%). These orientation effects improve the mechanical stability of PP and PA and could, especially for PP, provide more efficient reprocessing opportunities (superior to PET) when combined with a PE-based barrier layer due to better compatibility between

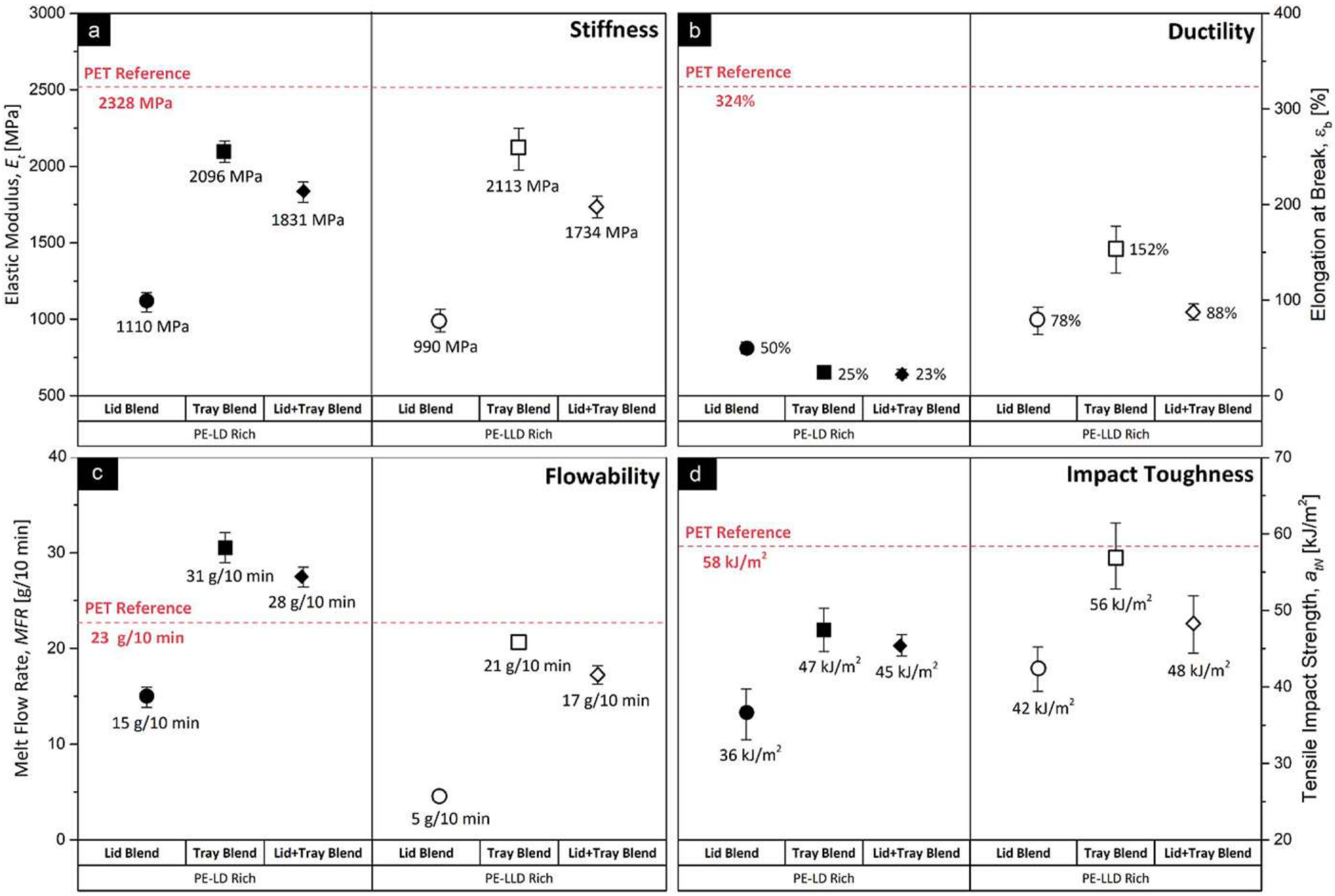

Fig. 3. (a) E_t [MPa], (b) ε_b [%], *MFR* [g/10 min] for 2.16 kg at 275 °C and (d) a_{tN} [kJ/m^2] of individual system parts (lid, tray) and the entire packaging system (lid+tray). Blends are divided into PE-LD- (solid markers) and PE-LLD-rich (hollow markers) compositions and compared to virgin PET (main constituent of the MAP waste stream, dashed line).

the polyolefins and the use of similar processing temperatures (Schyns & Shaver, 2021) (Fig. 4).

The three-layer PE-EVOH-PE composition found in most samples is responsible for the MAP barrier function, blocking oxygen and moisture transmission (Rux, Luca, & Mahajan, 2019; Tajeddin, Ahmadi, Sohrab, & Chenarbon, 2018). PET carrier layers measuring 255 µm (typical carrier layer) provide a WVTR of 2.02 g/(m^2·day) and an OTR of 13.94 cm^3/(m^2·day·bar). A combination of 225 µm PET and 35 µm PE resulted in an increased OTR of 18.62 cm^3/(m^2·day·bar) while the WVTR decreased to 1.36 g/(m^2·day) as PE is less permeable to moisture than PET alone but more readily transmits oxygen due to differences in the polarities of PET and PE (McKeen, 2011). The presence of a thin EVOH layer (154 µm PET + 15 µm PE + 5 µm EVOH + 17 µm PE) radically reduced the OTR to 2.16 cm^3/(m^2.day.bar) while the WVTR remained almost constant at 1.23 g/(m^2·day).

3.3. Component miscibility

Immiscible blends of two or more polymer components do not satisfy thermodynamic phase stability conditions and will consequently undergo phase separation driven by the degree of structural difference between the blend partners, which can lead to embrittlement and decrease recyclate quality (Dorigato, 2021).

DSC thermograms indicated that various PE types in PE-LD rich binary blends (LLD/LD 20/80) exhibit different melting- (PE-LD: 107 °C, PE-LLD: 122 °C) and crystallization (PE-LD: 96 °C, PE-LLD: 108 °C) peaks (Fig. 5). Peak separation between PE-LD and PE-LLD constituents is still visible if EVOH (melting: 167 °C, crystallization: 145 °C) is blended into the binary system and PET (melting: 248 °C, crystallization: 200 °C) added as a fourth component. Conversely, PE-LLD rich samples exhibit a single peak in the PE melt region at 123 °C which can be attributed to PE-LLD in the blend composition and indicates better component miscibility (Morris, 2016). Literature suggests that PE miscibility may be influenced by chain length (long chain branches hinder complete mixtures) rather than by differences in molecular weight or viscosity, which are often suggested to be key factors in general polymer blend miscibility (Zhao & Choi, 2006).

Tertiary blends representing the barrier layer (Fig. 5b, e) exhibited a finer morphology and smaller dispersed EVOH particles (< 1 µm) than the PE-LD rich system suggesting better miscibility in PE-LLD rich systems as supported by SEM micrographs. Binary systems (Fig. 5a, d) showed no particle dispersion and hence no conclusive evidence of a homogeneous mixture. White surfaces visible in LLD/LD 80/20 blends could indicate slightly more ductile fracture behavior (Sundararaj, Macosko, Rolando, & Chan, 1992). Quaternary blends representing the overall MAP waste stream (Fig. 5c, f) exhibited clear phase separation (dispersed particles >20 µm), which indicated a highly incompatible and brittle (no void formation or fibrillation) mixture.

3.4. Temperature effects during mechanical recycling

Ideal polymer reprocessing environments utilize temperatures only slightly higher than those of the polymer's melt region to prevent deterioration induced by thermal degradation (Pielichowski & Njuguna, 2005). MAP reprocessing temperatures must be > 250 °C due to the presence of PET in these systems despite the other polymer components

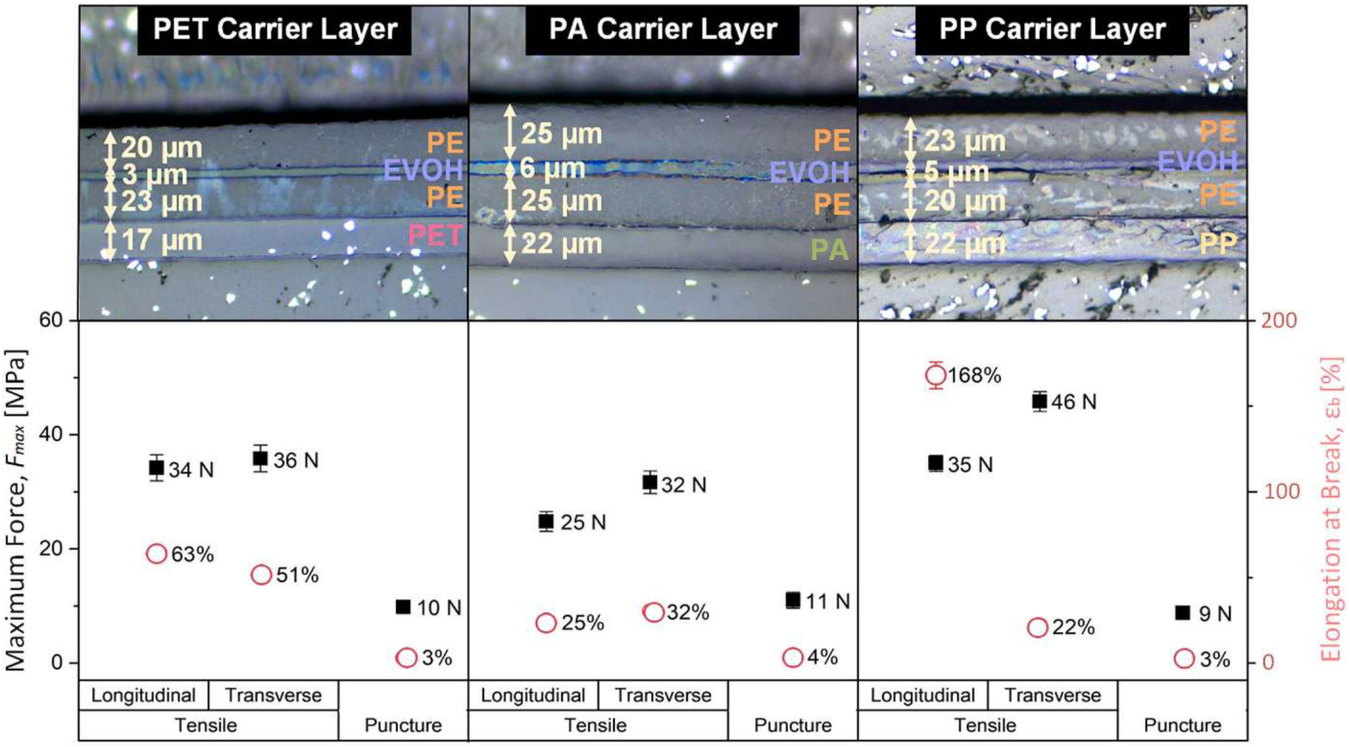

Fig. 4. Impact of the carrier layer material (packaging stability) on F_{max} and ε_b (tensile and puncture resistance testing) of multilayer film samples. Film structures are illustrated using light micrographs (upper row) depicting barrier layers (PE-EVOH-PE) of similar thicknesses but containing PET (left), PA (middle) and PP (right) carrier layers.

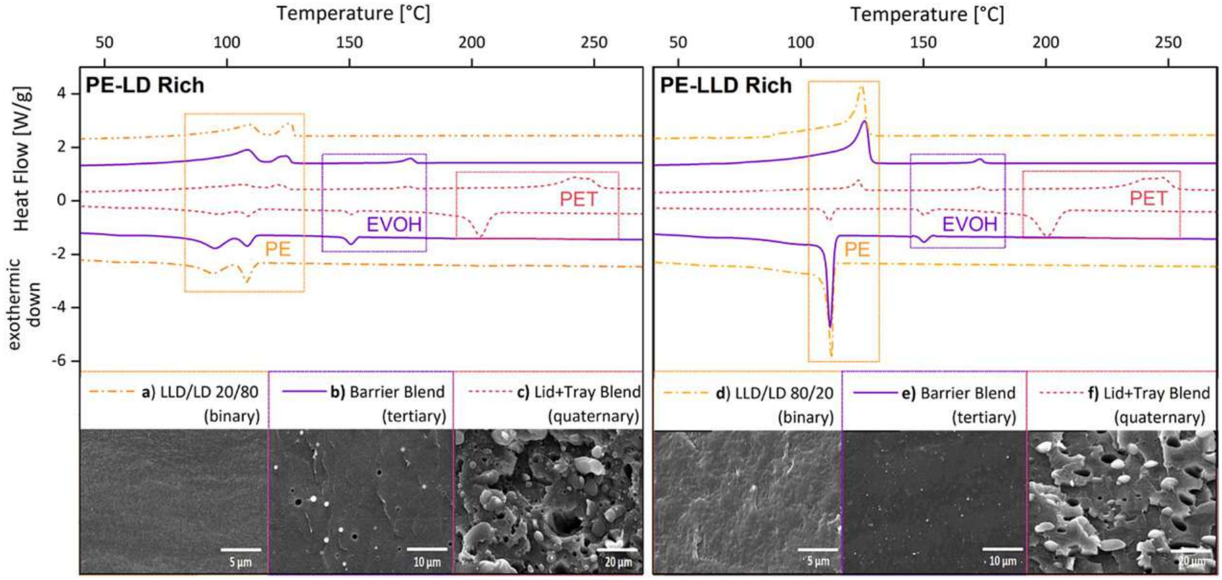

Fig. 5. Component miscibility of PE-LD- (left) and PE-LLD-rich (right) blends demonstrated using DSC thermograms and scanning electron micrographs of (a, d) binary PE systems (LLD/LD 20/80, LLD/LD 80/20, reference), (b, e) tertiary barrier blends representing the barrier layer in MAPs and (c, f) quaternary lid+tray blends representative of the total MAP waste stream.

having much lower melting points (EVOH: ~165 °C, PE-LLD: ~120 °C, PE-LD: ~110 °C).

MFR and dynamic shear rheology measurements for PE-LD- (LLD/LD 20/80) and PE-LLD- (LLD/LD 80/20) rich binary and tertiary barrier blends representative of pure polyolefin and combined polyolefin-polyester waste streams exhibited decreased *MFR* and increased complex viscosity values at extrusion temperatures of 275 °C (Fig. 6). *MFR* values for PE-LD rich blends reduced by 20% as the temperature was raised from 220 °C to 275 °C (3.9 g/10 min compared to 3.2 g/10 min for LLD/LD 20/80 and 3.5 g/10 min compared to 3.0 g/10 min for barrier blends) and approximately halved for PE-LLD rich blends (1.5 g/10 min compared to 0.8 g/10 min for LLD/LD 80/20 and 1.9 g/10 min compared to 1.0 g/10 min for barrier blends). This behavior can be attributed to temperature induced cross-linking effects in PE frequently reported at processing temperatures > 250 °C (Mendes, Cunha, & Bernardo, 2011).

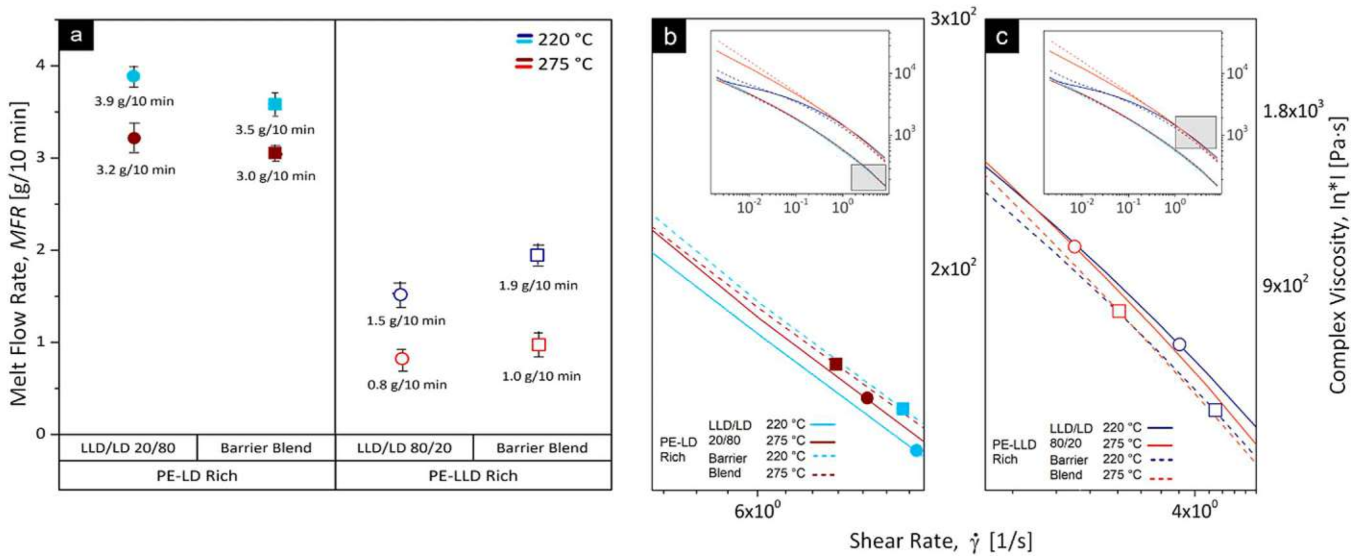

Fig. 6. (a) MFR [g/10 min] at constant shear for a 2.16 kg load at 220 °C and (b, c) complex viscosity curves measured at 220 °C for PE-LD- (LLD/LD 20/80), PE-LLD-rich (LLD/LD 80/20) and tertiary (barrier) blends containing EVOH following extrusion at 220 °C or 275 °C. Correlation between the two measurements as detailed in (b, c) is represented using colored markers on the complex viscosity curves.

Temperature dependent mechanical properties, such as E_t, a_{tN} and ε_b, varied less than rheological properties (deviations < 15%), with the exception of the ε_b values of PE-LLD rich blends (Table 2). ε_b increased ~45% (98 ± 9% compared to 143 ± 19% for LLD/LD 80/20 and 104 ± 13% compared to 152 ± 8% for barrier blends) at an extrusion temperature of 220 °C compared to 275 °C. This indicated that ε_b is more sensitive to changes in processing temperature than E_t and a_{tN}, a phenomenon also noted in other studies (Yin *et al.* 2015).

4. Conclusion

Deterioration effects associated with mechanical recycling processes were studied to estimate the recycling potential of MAPs using blends representative of individual system parts (lid, tray) and functional layers (carrier, barrier). The composition of the blends was based on a study of 60 commercial MAPs. Changes in physical properties, such as viscosity and impact toughness in the solid and molten state, resulting from pure polyolefin and polyolefin-polyester recycling temperatures were investigated in addition to correlations between blend constituents representing individual system parts or layers, blend homogeneity, changes in polymer chain structure and miscibility of individual components. MAP constituent analysis revealed very similar compositions for lids and trays individually, comprising a PET layer laminated to a coextruded three layer structure of EVOH sandwiched between two PE layers (with various ratios of PE-LD and PE-LLD) but considerable variation in the PET-PE ratio of lids compared to trays, which limits the uniformity of the packaging waste stream as a single entity. Virgin blends representing the compositions of lids and trays exhibited limited compatibility resulting in embrittlement and reduced toughness. The recyclate is subsequently not suitable for products of similar economic value to packaging films, which limit the economic viability of mechanical lid/tray recycling of MAPs. Design for recycling strategies must consider essential packaging function (mechanical stability and barrier function against gas diffusion) which are commonly achieved using PET and PE-EVOH-PE layers. While PET and PE were immiscible with strong phase separation (particles > 20 μm) visible in their morphology leading to material inhomogeneity and loss of toughness and ductility, the tertiary blend representing the barrier structure only exhibited dispersed particles < 1 μm in size making it more homogeneous. Further deterioration during mechanical recycling was promoted by the > 250 °C extrusion temperature required to melt PET, which exceeded the melting point of PE (PE-LD: 110 °C, PE-LLD: 120 °C) by more than 100 °C. This high processing temperature resulted in structural changes such as branching and crosslinking in the PE-based barrier layers.

These results demonstrate that MAPs in their current form require delamination of barrier and carrier layers and sorting into PE and PET waste streams to be recyclable in primary recycling systems. Next to sorting techniques and degradation issues, organic and inorganic contamination resulting from flawed sorting practices, traces of not economically efficient removeable inks, adhesives, labels and food residues or widely used additives/fillers, such as calcium stearate, contribute to the decrease of homogeneity and loss of quality during mechanical polymer recycling. Consideration of various contaminants and their origins as well as their effects on the recyclate would have exceeded the scope of this work. Nevertheless, new MAP designs or the substitution of incompatible components in existing designs are required to make primary recycling of MAPs an economically and ecologically viable process that contributes towards meeting the recycling targets of the future.

Table 2
Impact of extrusion temperature $T_{extrusion}$ on tensile impact strength a_{tN} [kJ/m²], elastic modulus, E_t [MPa] and elongation at break, ε_b [%] of PE-LD- (LLD/LD 20/80) and PE-LLD-rich (LLD/LD 80/20) binary and tertiary (barrier) blends containing EVOH.

Sample		$T_{extrusion}$ = 220 °C			$T_{extrusion}$ = 275 °C		
		a_{tN} [kJ/m²]	E_t [MPa]	ε_b [%]	a_{tN} [kJ/m²]	E_t [MPa]	ε_b [%]
PE-LD Rich	LLD/LD 20/80	215 ± 11	200 ± 3	126 ± 13	243 ± 12	192 ± 10	139 ± 17
	Barrier Blend	230 ± 9	227 ± 13	122 ± 15	227 ± 7	219 ± 14	135 ± 6
PE-LLD Rich	LLD/LD 80/20	229 ± 21	325 ± 12	98 ± 9	255 ± 18	317 ± 5	143 ± 19
	Barrier Blend	278 ± 27	380 ± 29	104 ± 13	301 ± 6	332 ± 7	152 ± 8

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

The data presented in this study are available on request from the corresponding author.

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Prospects for Recyclable Multilayer Packaging: A Case Study

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Abstract: Food preservation is an essential application for polymers, particularly in packaging. Complex multilayer films, such as those used for modified atmosphere packaging (MAP), extend the shelf life of sensitive foods. These mostly contain various polymers to achieve the necessary combination of mechanic, optic, and barrier properties that limit their recyclability. As the European Union's Circular Economy Action Plan calls for sustainable products and business models, including waste prevention policies and recycling quotas, with plastic packaging being a high priority, solutions towards more sustainable multilayer packaging are urgently needed. This study evaluated and compared the recycling potential of functionally equivalent PET (polyethylene terephthalate) and PP (polypropylene) post-consumer MAP through structure analysis and recycling simulation. The structure analysis revealed that both types of MAP contained functional (stability) and barrier layers (oxygen and moisture). The recycling simulation showed that the PP-based packaging was recyclable 10 times, maintaining its mechanical properties and functionality. At the same time, the PET-based MAP resulted in a highly brittle material that was unsuitable for reprocessing into similar economic value products. The secondary material from the PP-based MAP was successfully manufactured into films, demonstrating the functional possibility of closed-loop recycling. The transition from a linear to a circular economy for MAP is currently still limited by safety concerns due to a lack of sufficient and efficient purification methods, but the proper design of multilayers for recyclability is a first step towards circularity.

Keywords: multilayer packaging; recycling; modified atmosphere; polyolefins; waste management

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1. Introduction

Food preservation is one of the most common applications for polymers. Packaging accounts for approximately 44% of the global annual converter demand of 390.7 million tons [1]. Complex multilayer films, such as those used for modified atmosphere packaging (MAP), have a share of about 30% within this sector [2]. MAP is a sealed multilayer material system (consisting of a lid and a tray) that extends the shelf life of sensitive foods by creating a modified gas atmosphere. The longevity of the modified atmosphere is essential for its effectiveness. Therefore, the selected material must prevent gas diffusion and damage from external forces before content use [3]. A low water vapor transmission rate (WVTR) and oxygen transmission rate (OTR) are specifically required for this, typically requiring the combination of nonpolar and polar polymer elements.

Consequently, MAP may contain layers of up to nine or more different polymers, which are typically polyethylene (PE), polypropylene (PP), polyamide (PA), ethylene vinyl alcohol (EVOH), and polyethylene terephthalate (PET). The manufacturing process of multilayer films involves several steps, starting with the extrusion of virgin polymer resins. After extrusion, the polymer melt can be processed into films by blowing (tubular process) or casting (flat process). In order to form a multilayer structure, individual film layers can be combined in a molten (co-extrusion) or solid-state (lamination, with or without adhesives). Since most polymer melts cannot be easily combined due to structural differences, the use of adhesives (tie layers) and primers is necessary for the co-extrusion process. For surface finishing and decoration, MAP is commonly coated (glossy surface), printed (direct or reverse), or labeled (adhesive-attached paper or polymer labels) [4–7].

Those tightly assembled packaging structures, however, complicate the recycling process. Current mechanical recycling technologies are limited by their reliance on single-variety separation, making them unsuitable for the viable reprocessing of conventional MAP [8]. According to various studies on polymer streams [9,10] and recycling systems, this is one of the limiting factors for the necessary massive increase in plastic recycling.

While pure polyolefin [11] (PE and PP) and PET [12] streams can be recycled rather well, most MAP is, even if collected, currently ending up in the 'reject' streams of recycling units and incinerated for energy recovery. This is in contradiction with the European Union's Circular Economy Action Plan (CEAP), adopted in March 2020 as a part of the "Green Deal" to make the EU (European Union) economy sustainable and reach the UN (United Nations) Sustainable Development Goals by 2030 [13,14]. The CEAP aims to make sustainable products and business models the norm, including waste prevention policies such as using recyclates in new products and recycling quotas of 55% in the packaging sector [15]. Plastic packaging is a high priority for circularity implementation and will be a focus for future actions and legislation.

Avoidance or substituting polymers with other materials is often considered a first-solution approach for reducing polymer waste. However, removing or replacing functional plastics from food packaging is only possible by sacrificing the numerous advantages and conveniences they offer [16]. Despite the broad scientific research effort in developing biobased materials for food packaging, their application is not yet viable due to both the lack of proper industrial recycling capacities [17,18] and because of a shortfall in performance. Using polymer substitutes, such as paper, aluminum, or glass, may quickly lead to higher transportation and manufacturing costs, not benefiting the carbon footprint or sustainability life cycle analysis [19–21].

However, there have been some promising recent research in certain packaging areas lately. Korte et al. [22] demonstrated that groundwood pulp and sugar cane trays demonstrated better preserving characteristics for tomatoes when compared to their other references, including rPET (recycling PET). Chen et al. [23] found that the recyclability of beverage-paper-based composites could be improved by extending producer responsibility, benefiting the recycling rate. Tsironi et al. [12] discussed the current trends regarding the future sustainability of PET bottles, and Sid et al. [24] identified bio-sourced polymers as an emerging alternative to conventional food packaging. The urgent need for a more comprehensive evaluation, as well as comparison tools, to assess the sustainability of packaging products and materials is also proposed in the literature [21,25].

Nevertheless, although polymer multilayer packaging is currently predominantly incinerated, it is still widely considered the most environmentally beneficial solution when compared to substitutes [26–28]. One key reason is the aforementioned balance between the mechanical performance required for logistics and handling, the thermal stability required for filling and/or thermal treatment, the optics required for customer appeal, and the barrier properties (WVTR and OTR) required for content preservation. In this respect, Table 1 presents an overview of these properties for one of the most frequently cited biobased and biodegradable polymers, poly(lactic acid) (PLA), as well as a number of conventional versions. PET films demonstrate high-temperature stability (melt temperature (T_m): 256 °C) and moderate barrier against oxygen and moisture (OTR: 20.0 $cm^3/m^2 \cdot d \cdot bar$, WVTR: 4.0 $g/m^2 \cdot d$). PP has moderate temperature resistance (T_m: 165 °C) and a poor oxygen barrier (OTR: 800.0 $cm^3/m^2 \cdot d \cdot bar$), though it has an excellent moisture barrier (WVTR: 0.6 $g/m^2 \cdot d$), while PLA has the lowest temperature resistance (T_m: 146 °C) and a poor blocking function against oxygen and moisture (OTR: 280.0 $cm^3/m^2 \cdot d \cdot bar$, WVTR: 14.0 $g/m^2 \cdot d$). Only the multilayer combinations (PET/EVOH/PE, PP/EVOH/PE) result in the optimum balance of temperature resistance, stability, and barrier properties necessary for MAP films. One can easily see that the target property combination is further out of reach for PLA than for other polymers. Among other factors, this results from the very low crystallization speed [29] of this polymer, which makes the glass transition temperature (T_g) the more relevant stability parameter. While PLA may be suitable for certain packaging applications, such as fresh produce that requires short shelf life and limited gas barrier requirements, its gas permeability, moisture barrier, and temperature sensitivity make it less suitable for modified atmosphere packaging.

Table 1. Comparison of elastic modulus (E_t), glass transition temperature (T_g, main load-bearing layer), melt temperature (T_m, main load-bearing layer), oxygen transmission rate (OTR), and water vapor transmission rate (WVTR) for conventional packaging films compared to PLA (poly(lactic acid)). * Normalized to 100 µm in thickness [30–32].

Film Sample	E_t [MPa]	T_g [°C]	T_m [°C]	OTR [$cm^3/m^2 \cdot d \cdot bar$]	WVTR [$g/m^2 \cdot d$]
PET	3021	70	256	20.0 *	4.0 *
PP	1530	0	165	800.0 *	0.6 *
PLA	3600	55	146	280.0 *	14.0 *
PET/PE	2851	70	256	18.6	1.3
PET/EVOH/PE	3395	70	256	1.5	1.1
PP/EVOH/PE	2368	0	165	0.4	0.2

This clearly indicates the need for recyclable polymer-based multilayer packaging, for which a critical analysis of the existing packaging recyclability (and the derivation of new structures from these results) will play a decisive role [33]. Recently, the research interest in the field of flexible packaging recycling has rapidly grown, highlighting the importance of advancing packaging sustainability [34–39]. The work is mainly focused on either the "design for" recycling (invention of novel packaging films) [40–44] or "design from" recycling (recycling possibilities [45] and recyclate application) [46–49] aspects but does not connect the topics for a holistic overview.

This study was performed to evaluate the mechanical recycling potential of commercial PET- (majority) and PP-based (minority) post-consumer MAP, which is representative of the Austrian product market [50], and to connect the "design for" (packaging structure) and "design from" (assessment of recyclate properties and possible secondary applications) recycling aspects by following the scheme presented in Figure 1. It should be noted here that comparisons to other European countries, like Belgium [10], confirm the dominance of PP and PET in packaging waste streams (next to PE), adding to the logic of this choice.

The exact packaging structure (polymeric constituents, number of layers, and their thicknesses) of the individual components (lid or tray) was investigated to examine recycling (in combination with the results from secondary material testing) in terms of which packaging design structures are more or less suitable for mechanical recycling. Blends of the post-consumer MAP were processed following industrial procedures (cold-wash, grinding, and extrusion) [51] prior to the analysis of their mechanical, rheological, and morphological material properties, which were compared to each other. PP-based MAP (lid + tray) was further reprocessed up to 10 times (material samples were collected after 1, 3, 5, 7, and 10 reprocessing steps), tested mechanically (elastic modulus, elongation at break, and tensile impact toughness) and rheologically tested (melt flow rate, zero-shear viscosity, and extensional rheology) and was subsequently processed into new films using a micro blown-film line to simulate film-to-film closed-loop recycling. In order to highlight the functional possibility of the film-to-film recycling (possible secondary application) of PP-based MAP, the tensile modulus and puncture resistance of the blown films was tested and compared to virgin PP lids, which had similar thicknesses.

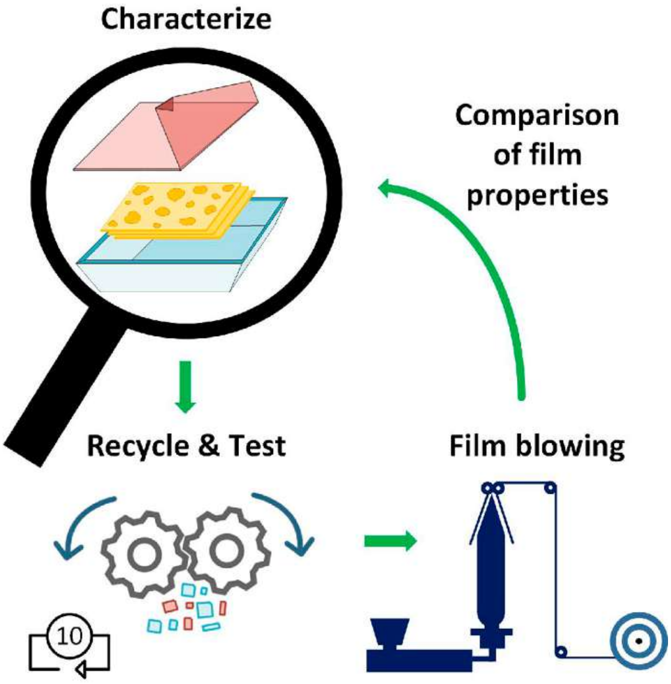


Figure 1. Research scheme.

2. Materials and Methods

2.1. Materials and Test Specimen

PP ('Berger' ham)- and PET ('Grana Padano' cheese)-based post-consumer MAP was collected from household bins in eastern Austria to assess the recycling potential of the two different packaging products. Individual multilayer structures were analyzed and mechanically reprocessed (comparable to industrial practices), and properties, such as melt flow rate and stiffness, were tested and compared for material quality evaluation [51]. The recyclability of the system components was investigated by separate (lid or tray) and combined (lid + tray) reprocessing, according to Table 2. Currently, there is no uniform standard procedure for the mechanical recycling of polymer waste, but there are some existing guidelines, specifications, and pieces of research on possible certification systems for waste polymers, which provide guidance for processing and testing [52,53].

Table 2. Composition of investigated post-consumer polypropylene (PP)- and polyethylene terephthalate (PET)-based blends from modified atmosphere packaging (MAP) representing the disassembled (lid or tray) and combined packaging (lid + tray) structure processed at T_{PP} = 220 °C (PP-based packaging) or T_{PET} = 275 °C (PET-based packaging).

Composition (Estimated from Analysis)				**Blend Model**	**Extrusion**		
EVOH	**PE-LD/LLD**	**PET**	**PP**	**Representation**	**T_{PET}**	**T_{PP}**	**Processing Steps**
4 wt.%	23 wt.%	73 wt.%	-	'PET Lid + Tray'	275 °C	-	1×
2 wt.%	13 wt.%	85 wt.%	-	'PET Tray'	275 °C	-	1×
11 wt.%	66 wt.%	23 wt.%	-	'PET Lid'	275 °C	-	1×
4 wt.%	4 wt.%	-	92 wt.%	'PP Lid + Tray'	-	220 °C	10×
3 wt.%	5 wt.%	-	92 wt.%	'PP Tray'	-	220 °C	1×
6 wt.%	-	-	94 wt.%	'PP Lid'	-	220 °C	1×

Prior to the mechanical recycling process, the MAP was sorted, cold-washed, and cut manually. A singled screw Extron extruder (EX-18-26-1.5, Extron Engineering Oy, Toijala, Finland) with a screw diameter of 18 mm and a length/diameter ratio of 25:1, equipped with three individual heating zones, was used for melt blending. Extrusion temperature was varied according to the packaging components based on the typical processing temperatures for the respective polymers (T_{PP}: 220 °C for PP and T_{PET}: 275 °C for PET-based packaging), and the screw speed was set to 70 rpm (this was found to give good homogeneity in earlier studies). The material strands from extrusion were shredded with a universal cutting mill (Pulverisette19, Fritsch, Idar-Oberstein, Germany) that had a 4 mm sieve inserted. For 'PP Lid + Tray', the process of extrusion and grinding was repeated up to 10 times (1×–10×, material samples were collected after 1, 3, 5, 7, and 10 reprocessing steps) to demonstrate the stability resp. degradation behavior of the PP-based MAP. The same reprocessing procedure could not be carried out for the PET-based packaging due to inferior material quality and liquid-like behavior after only one processing step.

2.2. Test Specimen

Test specimens for tensile and tensile impact testing, according to ISO 527-2 [54] and ISO 8256 [55], were produced by injection molding using a Haake Mini Lab II twin-screw extruder coupled with a Haake Mini Jet II injection molding unit (Thermo Fisher Scientific, Waltham, MA, USA). Extrusion temperature was set according to compounding temperature (T_{PP}: 220 °C for PP and T_{PET}: 275 °C for PET-based packaging); screw speed was 100 rpm, and mold temperature for injection was 40 °C (pressure: 350 bar, injection time: 10 s).

Specimens for dynamic shear and extensional rheology were produced by compression molding (Collin P 200 P, Maitenbeth, Germany) at a pressure of 100 bar and in line with extrusion temperatures of 220/275 °C. Discs with a diameter of 25 mm and a thickness of 1.2 mm and squares of 0.8 mm in thickness and a side length of 60 mm were generated using punched aluminum frames sandwiched between steel plates and were separated by Teflon® sheets.

To demonstrate the possibility of film-to-film recycling, the recyclates were converted into 50 µm-thick films using an Ultra Micro blown-film line (LabTech Engineering, Samut Prakan, Thailand). Extrusion temperature was set to 200 °C; the die temperature was 180 °C; the fan for airflow cooling was set to 1700 rpm, and the pull-off speed of both roller sets was 1.1 m/min.

2.3. Multilayer Structure Analysis of Post-consumer MAP

The characterization of multilayer packaging structures was based on the user guide for the identification of polymers in multilayer films used in food contact materials released by the European Commission [56]. The inner and outer layers of post-consumer packaging were analyzed using a Bruker Tensor 27 Fourier-transform infrared (FT-IR) spectrometer (Billerica, MA, USA) with an attenuated total reflection (ATR) diamond (DuraSample IR II) and single reflection. A total of 16 scans with a 4 cm^{-1} resolution between 600–4000 cm^{-1} were conducted to identify the polymer spectra.

Differential scanning calorimetry (DSC) was used to detect all polymers within the outer layers included in the multilayer film structure by monitoring changes in heat flow attributed to physical changes such as melting [57]. Packaging samples were analyzed using a TA-Instruments TA Q2000 device (Newcastle, DE, USA) in a nitrogen atmosphere, with a sample mass of 5–6 mg and a temperature range of 0–300 °C, increasing at a rate of 10 K/min.

The thickness of each layer was determined using a Zeiss Axio Imager M2m light microscope (Oberkochen, Germany). Sections of 20 × 12 mm were cut from the post-consumer packaging and embedded with a two-component epoxy resin (Araldite AY103 + REN HY956) before being ground and polished with a Struers TegraForce-31 polisher (Copenhagen, Denmark). The percentual polymer mass proportion given in Table 1 was calculated using volumetric mass density (ρ_{PET}: 1.34 g/cm^3, ρ_{EVOH}: 1.16 g/cm^3, ρ_{PE}: 0.93 g/cm^3, and ρ_{PP}: 0.9 g/cm^3) [58]. The combined mass proportions of the 'Lid + Tray' fractions resulted from the fact that the tray represents about 80 wt.% of the total packaging mass and the lid about 20 wt.% (detailed wt.% calculation can be found in the supplementary material).

2.4. Mechanical Properties of Recyclate Blends and Films

Specimens of 60 × 10 × 1 mm for the tensile impact strength test were notched on both sides using a Vis-Notch device (Instron, Darmstadt, Germany) and were tested with an Instron 9050 pendulum (equipped with 2 J hammer and 15 g crosshead mass; Instron, Darmstadt, Germany), according to ISO 8256/1A [55], to obtain the tensile impact strength a_{tN}. Tensile testing was carried out in accordance with ISO 527-2-5A [54] and was performed on a universal testing machine (ZwickRoell Z050, Ulm, Germany) at a speed of 10 mm/min, equipped with a 1 kN load cell and an extensometer.

Mechanical testing of blown film properties was carried out in the tensile mode, according to ISO 527-3 [59], using strips measuring 100 × 10 mm (70 mm clamping length) and by puncture-resistance testing, performed according to DIN EN 14477 [60] on round samples at a crosshead speed of 10 mm/min.

2.5. Rheological Properties of Recyclate Blends

Dynamic shear rheology was performed via frequency sweeps on an MCR 302 rheometer (Anton Paar, Graz, Austria) equipped with a plate–plate system (1 mm gap size) and a nitrogen-purged heating hood. The temperature was held constant at 220 °C during the experiments, while deformation was raised logarithmically from 1% to 2% at a frequency range between 628 rad/s and 0.01 rad/s.

For extensional rheology measurements, the testing device was equipped with a CTD 450 heating chamber (nitrogen purged) and an SER-HPV 1 Sentmanat Extensional Rheometer (Xpansion instruments, Tallmadge, OH, USA). Test specimens were strained at three different rates ($5\ s^{-1}$, $1\ s^{-1}$, and $0.1\ s^{-1}$) at a temperature of 180 °C, in line with the film-blowing procedure. Base-line curves (LVE) were measured using a plate–plate system, using shear rates of $0.001\ s^{-1}$ and $0.1\ s^{-1}$.

Melt flow rate (MFR) measurements were performed according to ISO 1133 method A [61] under a load of 2.16 kg at 220/275 °C by using a manual testing device (MeltFloW basic, Karg Industrietechnik, Krailling, Germany).

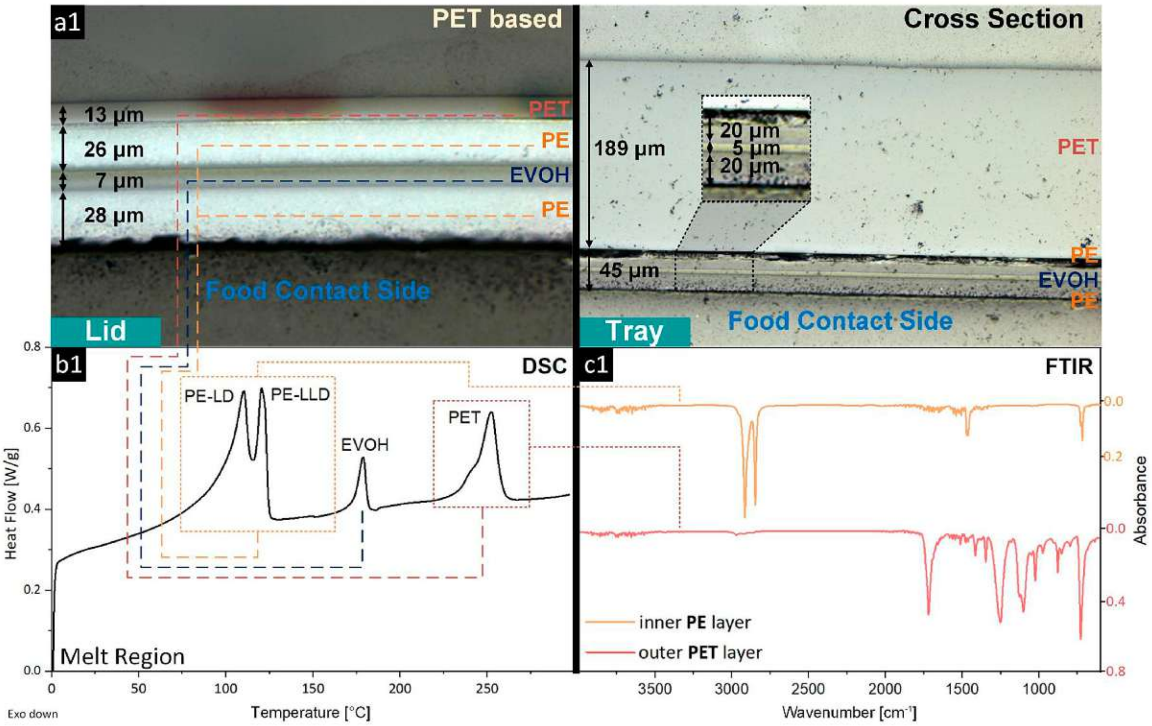
2.6. Morphological Characterization of the Recyclate Blends

The morphology of the recycling blends that were recycled once was characterized using scanning electron microscopy (SEM) for the fractured tensile impact test specimen surfaces with an FEI Philips XL30 microscope (Hillsboro, OR, USA). Prior to imaging, the test samples were coated with gold (Agar Sputter Coater B7340, Essex, UK). Average particle size was estimated from an optical assessment of the fracture surfaces, considering the fact that this gives only a rough indication.

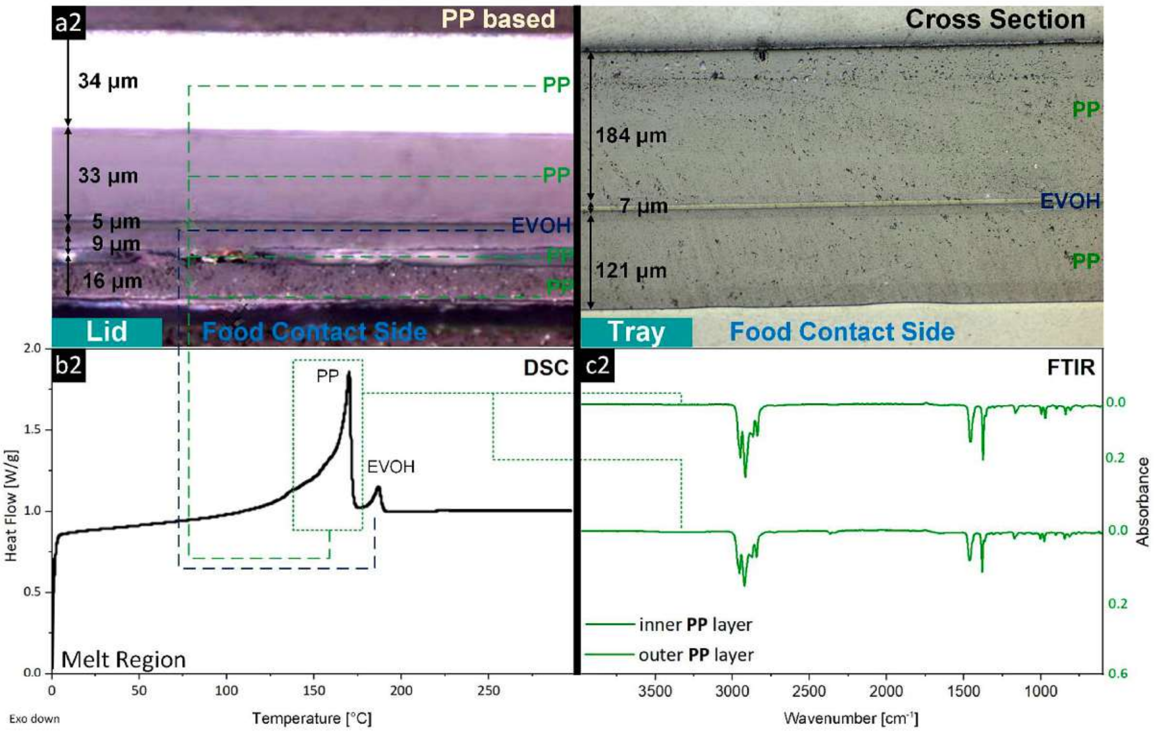
3. Results and Discussion

3.1. Multilayer Structure Analysis of Post-consumer PET- and PP-Based MAP Lids and Trays

The multilayer analysis of the post-consumer MAP was realized through a combination of DSC (melt peak identification of all polymers in the multilayer film), FT-IR (identification of inner and outer multilayer film layers), and light microscopy (thickness of individual layers for mass content estimation through volumetric mass density), which is schematically presented in Figure 2. The analysis revealed that the PP-based lids and trays were thicker (lid: 97 μm; tray: 312 μm) when compared to the PET-based components (lid: 74 μm; tray: 234 μm). However, when having the same geometry, the PP packaging remains lighter due to its lower density of 0.9 g/cm^3 when compared to the 1.34 g/cm^3 of PET, which results in lower logistic costs.



(A) PET based packaging



(B) PP based packaging

Figure 2. Packaging structure of PET (**a1**–**c1**)- and PP (**a2**–**c2**)-based MAP characterized by (**a**) light microscopy of film cross-sections, depicting the individual layers, (**b**) DSC (measurement of packaging film), revealing the melting peaks of the components, and (**c**) FT-IR spectroscopy (measurement of packaging film) for the identification of the inner (food contact) and outer layers. FT-IR and DSC curves of trays are provided in the Supplementary Materials (Figures S1–S4).

The lids and trays of the PET-based MAP consisted of four layers, which can be divided into the barrier (food contact side) and carrier layers according to their functionality. The barrier layer for the lids and trays was similar, comprising one layer of oxygen-blocking EVOH (lid: 7 μm; tray: 5 μm) sandwiched between two layers of moisture-blocking PE (lid: 26/28 μm; tray: 20 μm) [7]. The DSC revealed that PE was present as a blend of PE-LD and PE-LLD (distinct melt peaks at 108 °C and 122 °C). A 13 μm-thin (lid) and 189 μm-thick (tray) PET layer were used to provide sufficient mechanical stability for the packaging against external damage such as punctures, tearing, or buckling [50]. The trays only contained removable paper labels but were otherwise transparent, while the lids were reverse printed. This technique prevents the ink from coming into direct contact with human skin or food products, as it is situated between the first and second film layers [62]. However, this makes the partial or complete removal of the color during the recycling process very difficult or even impossible, subsequently leading to the undesirable greyish coloring of the secondary material.

The PP-based MAP lids presented a five-layer structure with four PP layers (34/33/9/16 μm) and one layer of EVOH (5 μm), providing a barrier against oxygen. The PP was present as a mixture of PP copolymer (high toughness) and PP homopolymer (increased stiffness), estimated by the broad melting peak of PP between 145–160 °C, providing a sufficient moisture barrier and mechanical stability regarding the packaging. [7]. The trays only consisted of three layers: two thick layers of PP (184/121 μm) sandwiching one thin layer of EVOH (7 μm). The trays also contained a small portion of PE, which was observed in the DSC analysis, most likely functioning as a tie layer material for the co-extrusion of PP and EVOH [63]. Contrary to the PET-based lids, the PP-based lids were printed directly, allowing for the more efficient removal of the ink during the recycling process using flotation de-inking processes [64].

The lids and trays of the PET-based MAP consisted of similar polymers, but these polymers were present in different ratios (Table 1). The PET trays contained about 85 wt.% PET and 13 wt.% PE, while the lids contained only 23 wt.% PET and 66 wt.% PE. Polymer–polymer contamination plays an important role in the mechanical recycling process, as the mixture of highly incompatible polymers, such as PE and PET, leads to secondary materials of inferior quality when compared to virgin polymers [65]. The recyclate quality of the PET trays could be improved by using compatibilization agents [66] due to the lower PE contamination (13 wt.%) when compared to the lids (66 wt.%). However, when the PET lids and trays are combined to form a single packaging system, the recyclability and effectiveness of the compatibilization agents are limited (higher amounts of compatibilization agent are required) by increasing PE contamination (23 wt.%) when compared to the PET trays alone [67]. On the contrary, the PP-based MAP lids and trays contained >90 wt.% PP, resulting in reasonably homogeneous and, hence, more easily recyclable waste streams for both the individual components (lid or tray) and the entire packaging (lid + tray).

3.2. Comparison of the Property Profile of Reprocessed PET- and PP-Based MAP

Extrusion within the mechanical recycling process causes the inevitable formation of polymer blends from the inseparable layers. The more incompatible the individual components are to each other, the more unpredictable the resulting properties of the recyclate, subsequently making the design of and finding a suitable application for those materials very challenging [68] or requiring the use of a compatibilization agent.

Figure 3 compares the elastic modulus (E_t), elongation at break (ε_b), melt flow rate (MFR), and tensile impact strength (a_{tN}) of recycled PET and PP-based post-consumer MAP, divided into individual system components (lid or tray) and entire packaging (lid + tray). In the material testing, the examined packaging types achieved very contrasting results. While the MFR for PP-based MAP varied between 2.7 g/10 min for the lids and 4.5 g/10 min for the trays, that of PET-based MAP was between 27.9 g/10 min for the lids due to the high PE content (66 wt.%), and 82.3 g/10 min for the trays. This leads to entirely different processing options regarding the secondary material, with MFR being the determining factor. PP-based MAP with an MFR of < 5 g/10 min offers the chance of being reprocessed into products of the same or similar economic value, such as films, while the high MFR of PET-based structures limits their applicability regarding injection molding [69], which is not the application the packaging film was originally designed for.

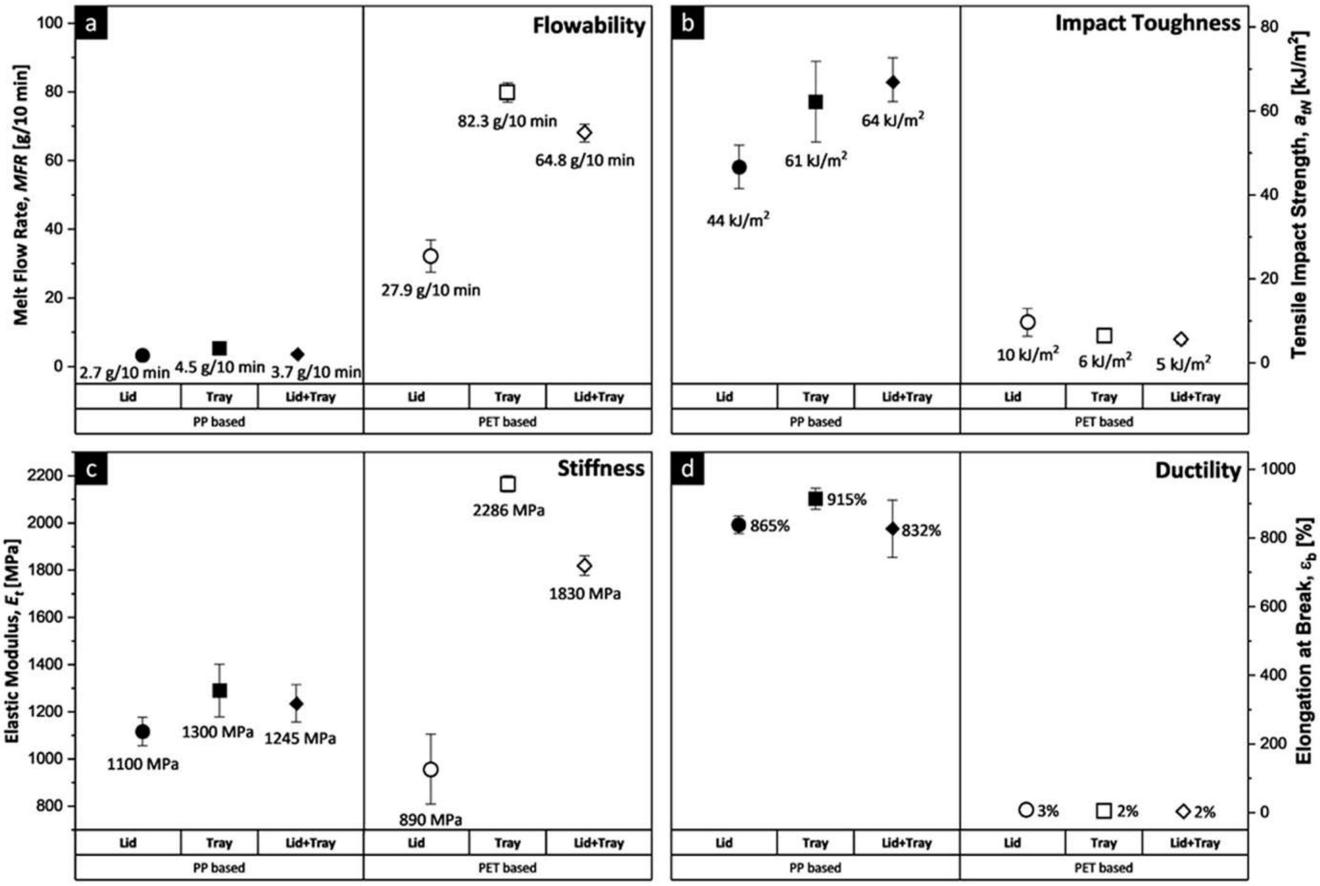


Figure 3. (**a**) Melt flow rate MFR [g/10 min], measured under a load of 2.16 kg at 220 °C (PP-based) and 275 °C (PET-based), (**b**) tensile impact strength a_{tN} [kJ/m^2], (**c**) elastic modulus E_t [MPa], and (**d**) elongation at break ε_b [%] of one-time recycled PP (solid markers)- and PET (hollow markers)-based post-consumer MAP divided into individual components (lid or tray) and entire packaging (lid + tray).

The tensile impact toughness of the PET-based MAP blends was extremely low, demonstrating values below 15 kJ/m^2. Due to the relatively high fraction of PE (66 wt.%), which, in virgin form, demonstrates a_{tN} values of >150 kJ/m^2, the PET lid had an a_{tN} of 10 kJ/m^2, while this was halved for the trays. However, toughness is an essential characteristic of high-value polymer products such as packaging. Tough packaging ensures that the product is safe from damage or breakage during transportation, storage, and handling. It provides a layer of cushioning and absorbs shocks and vibrations that may occur during handling and transport [7]. The PP lids had a four times higher a_{tN} of 44 kJ/m^2, and the PP trays presented an even higher value of 61 kJ/m^2, which most likely resulted in a toughening effect from the presence of 5 wt.% PE within the trays, making the material a suitable source for secondary packaging applications.

The E_t values of polypropylene are strongly dependent on whether a homopolymer (up to 2000 MPa) or a copolymer is used and, in the case of the latter, also on how high the polyethylene content is, which decreases the modulus (between 800 MPa and 1400 MPa) [58]. The PP trays exhibited the highest E_t value of 1300 MPa, while the values for the PP lid + tray (1245 MPa) and PP lid (1100 Mpa) were only slightly lower. Virgin

PET is stiffer compared to PP, presenting E_t values of about 3000 Mpa. For PET-based MAP recycling blends, the E_t ranged from 890 MPa for the lids (lowest PET content of 23 wt.%) to 2286 MPa for the trays (highest PET content of 85 wt.%).

Due to the mixture of the highly incompatible phases of PET and PE in PET-based MAP, the test specimen demonstrated brittle fracture behavior with ε_b values below 5%. The PP-based MAP blends were very ductile and exhibited ε_b values between 832% (lid + tray) and 915% (tray). The combined property profile (MFR, E_t, $\varepsilon_{b,}$ and a_{tN}) clearly indicated that the recyclate from the PP-based MAP demonstrated higher recycling potential (low MFR, combined with moderate stiffness and high ductility and toughness) when compared to the recyclate from the PET-based MAP (moderate to high stiffness but high MFR and low ductility and toughness). In order to be considered economically recyclable, a material needs to retain its chemical and mechanical characteristics while being easily sortable by recycling companies. Since multilayers cannot simply be disassembled for recycling, the packaging design must ensure that they can be efficiently transformed and sorted at a reasonable cost without compromising their performance.

Samples of the recyclates, together with the scanning electron micrographs of the PET- and PP-based blends, are presented in Figure 4. For both the PET- and PP-based fractions, an apparent influence in color from the inks was observed even though different printing methods (reverse and direct) were used (no de-inking processes were part of the study). However, the directly printed (PP-based) blends resulted in lighter shades of beige, especially when compared to the dark brown PET-based trays. Besides the inks from printing, other contaminants, such as adhesives, sealants, or food residues, which migrate into the polymer matrix, contribute to the grey coloring and bad smell of recyclates [70]. Even though the recyclates from the PP-based MAP demonstrated a considerably more attractive appearance when compared to the PET-based MAP, coloring is not necessarily an indication of better or worse material performance. Hot- or cold-wash processes are frequently used in industrial waste treatment procedures to improve color and odor, but these are only ecologically viable if the secondary application demands low smell and inclusion content, as the mechanical parameters are hardly affected [71].

However, the morphology of a polymer blend, specifically its size, shape, and distribution of the dispersed phase, has a great influence on its performance. It affects properties such as mechanical strength or thermal stability [72]. The blends from the post-consumer PP-based MAP, which contained structurally similar blend partners, exhibited very fine morphologies and only relatively small particles (average < 5 µm), which were homogeneously distributed. No notable difference in particle size, particle size distribution, and surface condition between the morphologies of the PP lid, PP tray, and PP lid + tray blends was found. In contrast, the blends derived from the PET-based MAP demonstrated clear phase separation at the particle-matrix boundary and more coarse particles with average diameters of >20 µm, indicating the combination of highly incompatible phases (PET and PE) and a brittle structure confirmed by a ε_b of < 5% obtained from tensile testing. The blends from the PET trays and PET lids + trays, which contained predominantly PET (tray: 85 wt.%, lid + tray: 73 wt.%), had similar morphologies, comprising dispersed PE particles within a PET matrix (major component), while conversely, the PET lids containing 66 wt.% PE exhibited dispersed PET particles within a PE matrix (major component). Accordingly, the morphologies of the PET- and PP-based MAP recyclates are in good agreement with the mechanical and rheological properties.

3.3. Multiple Processing of the PP-Based MAP

3.3.1. Mechanical Properties

In order to accomplish sustainable material circularity, it is essential to design products that can withstand not only single but multiple recycling procedures. In this case, not just the degree of organic and inorganic contaminants and compatibility of system components must be considered, but also how well that material (combination) can resist thermomechanical damage [73].

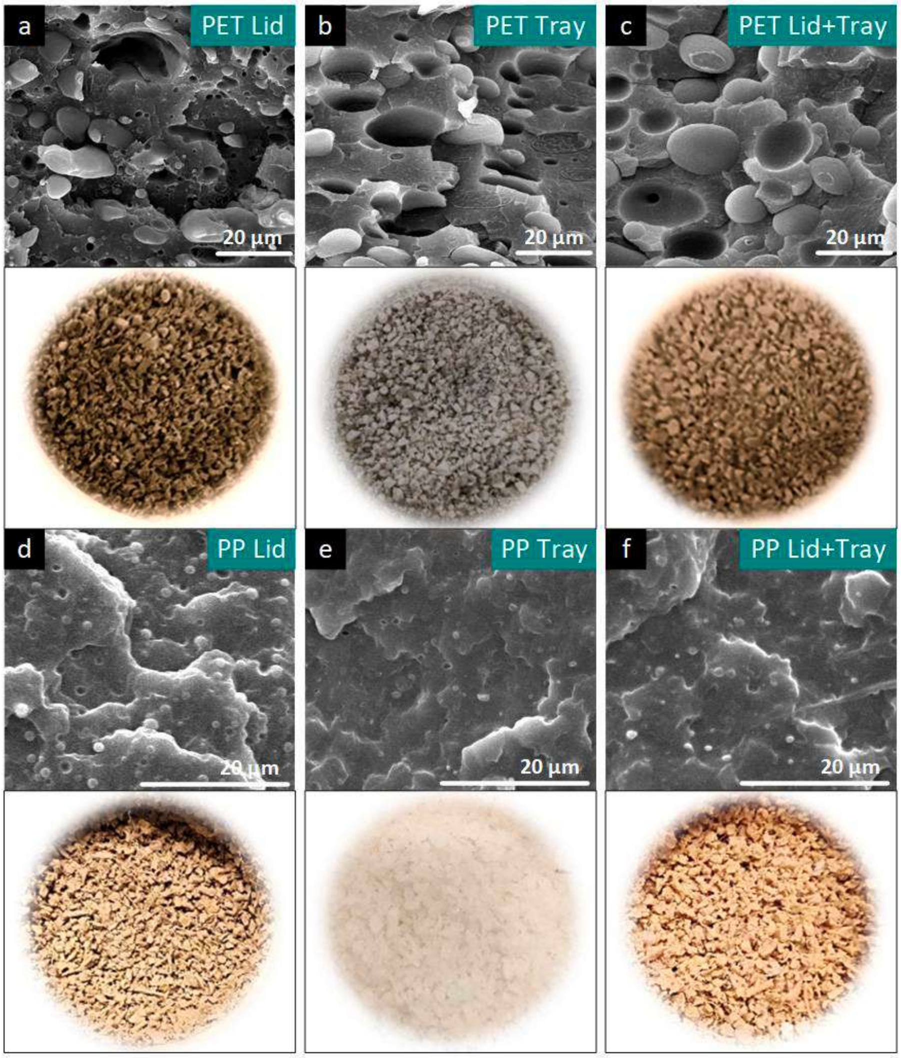


Figure 4. Samples of one-time recycled secondary materials and their morphologies, obtained from scanning electron microscopy of recycled (**a**–**c**) PET- and (**d**–**f**) PP-based post-consumer MAP divided into lids (**a**,**d**), trays (**b**,**e**), and lids + trays (**c**,**f**).

Due to poor mechanical properties and a very high MFR, which led to liquid-like behavior, the repeated reprocessing of the PET-based MAP was not possible. In contrast to this, Figure 5 presents the E_t, ε_b, and a_{tN} after 1, 3, 5, 7, and 10 recycles of the post-consumer PP-based MAP (lids + trays). Within the 10 reprocessing steps, the E_t decreased by 25% (1245 MPa for the first recycling step, compared to 944 MPa for the tenth step), with the most significant drop occurring between the first and the third step (decreasing from 1245 MPa to 1064 MPa). This stiffness reduction is commonly seen in the degradation of PP and is normally explained as a result of reduced crystallinity and skin-layer orientation [74,75]. ε_b slightly increased from 832% for the first processing step up to 871% for the tenth step, which is in line with slightly reduced crystallinity. Again, the greatest change in values was observed between the first and the third step, with an increase of 13% from 832% to 944%. Tensile impact strength held relatively constant within the standard deviation during the 10 reprocessing steps, varying between 64 kJ/m^2 (first step) and 69 kJ/m^2 (tenth step). Even though there was a moderate drop in stiffness with increasing reprocessing steps, no embrittlement of the material was observed over 10 recycling cycles, with ductility and toughness remaining largely constant, indicating the good processing stability of the recyclates. However, these changes in mechanical properties could likely be reduced with the addition of processing stabilizers [76].

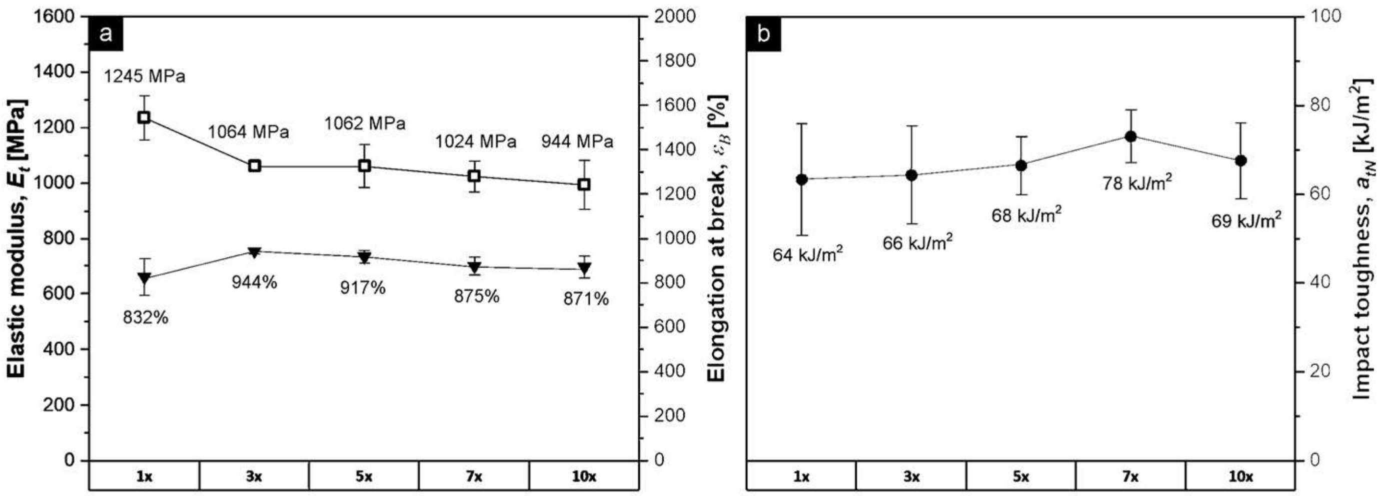


Figure 5. (**a**) E_t (hollow markers) and ε_b (solid markers) from tensile testing and (**b**) a_{tN} from tensile impact testing (injection molded specimen) of 10 times recycled PP-based MAP (lid + tray). The x-axis marks the number of reprocessing cycles.

3.3.2. Rheology

The high temperatures and shear forces associated with the mechanical recycling process of polymers result in increased chain scission related to changes in molecular weight, especially in the case of PP. The resulting change in molecular weight affects steady shear MFR, indirectly proportional dynamic shear viscosity values, and, subsequently, melt strength, which limits the reprocessing options [48]. The change in MFR and zero shear viscosity corresponds to an approximate reduction in average molecular weight from 400 to 300 kg/mol, which is a range where the effect on mechanical properties is only limited [77].

During the 10 reprocessing steps of the PP-based post-consumer MAP (Figure 6), the MFR was gradually increased from 3.7 g/10 min (first step) to 11.3 g/10 min (tenth step) while, conversely, the zero-shear viscosity was decreased from 4580 Pas (first step) to 1973 Pas (tenth step). This behavior is typically reported for the re-extrusion of PP and is associated with material degradation and the β-scission of polymer chains. However, the percentual change of the measured values depends on the PP type, as homopolymers may demonstrate an MFR increase of 400% within five recycling cycles, while an increase of 100–150% in MFR is reported for PP copolymers [78,79]. This deviant degradation behavior may be explained by the branching or crosslinking reactions of the ethylene-containing polymer chains. Nevertheless, the major PP homopolymer portion will predominantly dictate the degradation process and, thus, the trend of MFR and viscosity during reprocessing [78]. In order to support those findings, crossover points (crossover modulus G_c and crossover frequency ω_c) of storage (G') and loss modulus (G'') from the dynamic shear rheology measurements are presented in Table 3.

The crossover point indicates the transition from predominantly elastic to viscous behavior regarding the polymer melt. A shift in the crossover point can be associated with changes in molecular weight (M_W) and molecular weight distribution (MMD) [80–82]. The location of the crossover point at higher frequencies indicates the presence of shorter or broken polymer chains (shorter relaxation times), while a shift to higher frequencies indicates longer chains or branching reactions. Conversely, a shift to higher G_c values is related to a broadening of the MMD and vice versa for a shift to lower values [83]. For the 10 times recycled PP-based lid + tray blends, a clear shift in ω_c to higher values (45 rad/s to 82 rad/s) and, subsequently, a loss in M_w indicates chain scission through thermo-mechanical degradation during the individual reprocessing steps, which was observed. However, the highest MFR after 10 processing cycles of 11 g/10 min is still low enough for the production of films from the recyclate [84].

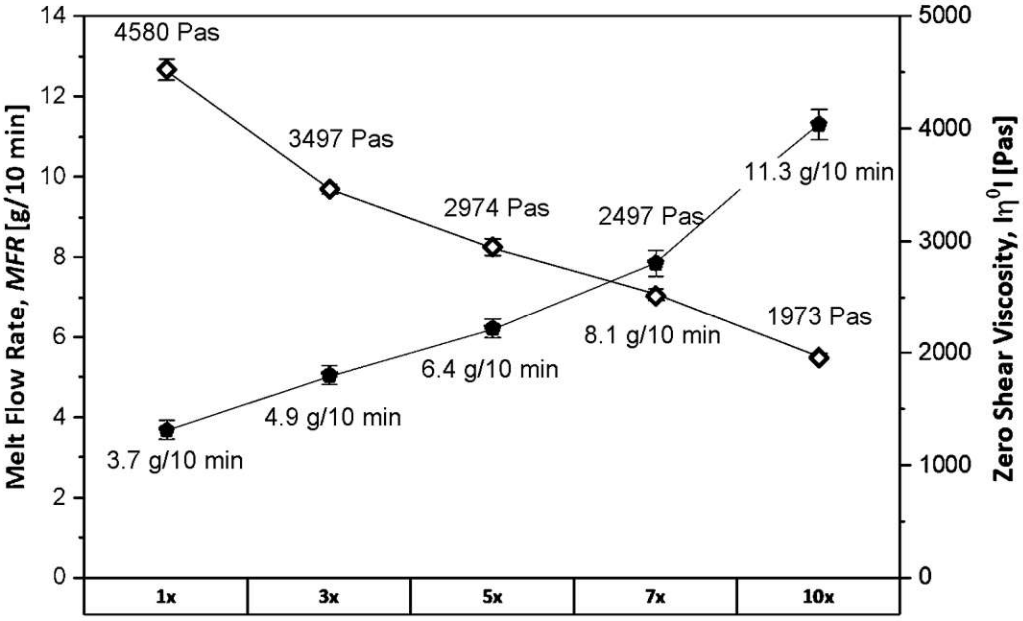


Figure 6. MFR (solid markers) and zero shear viscosity (hollow markers) of 10 times recycled PP-based MAP (lid + tray) measured under constant (2.16 kg load) and dynamic shear stress at 220 °C. The x-axis marks the number of reprocessing cycles.

Table 3. Results of crossover point ($G' = G''$) expressed as crossover frequency ω_c and crossover modulus G_c from dynamic shear rheology measurements at 220 °C of 10 times recycled PP-based MAP (lid + tray). Allowing the estimation of higher (↑)/lower (↓) molecular weight (M_W) and molecular weight distribution (MMD).

Sample	ω_c [rad/s]	G_c [Pa]	M_W/MMD
PP lid + tray 1×	45	31,230	
PP lid + tray 3×	59	31,522	M_w ↓, MMD ↑
PP lid + tray 5×	64	31,610	M_w ↓, MMD ↑
PP lid + tray 7×	73	31,789	M_w ↓, MMD ↑
PP lid + tray 10×	82	31,816	M_w ↓, MMD ↑

The stress resistance to the uniaxial extension of polymer melts is closely related to chain length and the degree of branching, which is visualized in Figure 7, which demonstrates the extensional rheology curves of the 10 times reprocessed PP-based MAP [85]. Good process stability and the output of high-quality films can only be realized with high melt strength polymers due to their relatively high extensional shear forces, to which PP homopolymers usually do not belong as a result of their linear chain structure [86]. Only the presence of ethylene in the copolymers results in the strain hardening of the melt, which was observed for all analyzed post-consumer PP MAP blends, especially at the lower strain rates of 0.1 and 1 s^{-1}. While all blends exhibited pronounced strain hardening for all processing steps at shear rates of 0.1 and 1 s^{-1}, for a strain of 5 s^{-1}, the strain hardening effect gradually increased from the fifth up to the tenth reprocessing step, indicating branching reactions within the ethylene phase [87]. The extensional rheology measurement has shown that the melt strength of the recyclates is sufficient for application in processes that require high extensional viscosities, such as film blowing (possible design from recycling application).

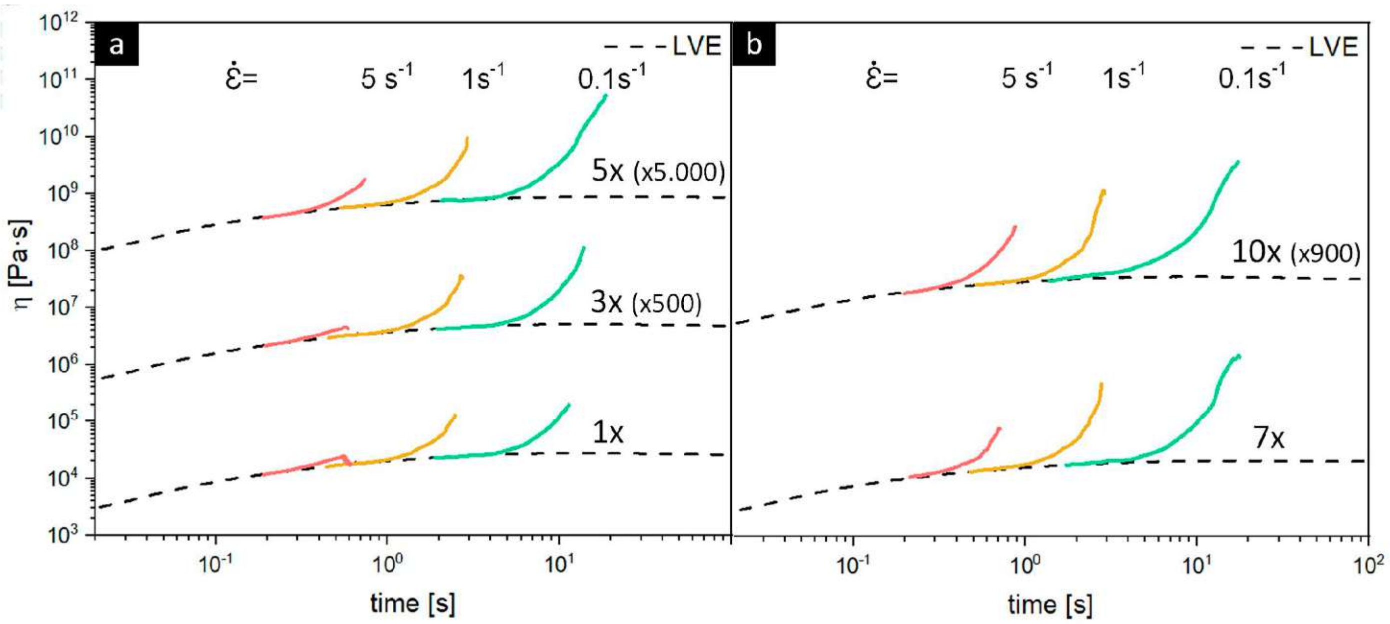


Figure 7. Vertically shifted extensional rheology curves of 10 times recycled (**a**,**b**) PP-based MAP (lid + tray) measured at 180 °C with strain rates of 5 s^{-1} (red lines), 1 s^{-1} (yellow lines), and 0.1 s^{-1} (green lines).

3.4. Film Blowing of Reprocessed PP-Based MAP

In order to support the assumptions from the extensional rheology measurements and to demonstrate the application possibilities of the recyclates, the reprocessed PP-based MAP was manufactured into films, and its appearance and mechanical properties (elastic modulus and puncture energy) were compared to the lid of the original PP-based packaging. Figure 8 depicts blown-film samples with a thickness of 50 µm from 1, 3, 5, 7, and 10 times reprocessed PP-based MAP (Figure 8b–f), accompanied by the elastic modulus E_t (longitudinal and transverse film direction) and puncture energy E_p from the mechanical film testing, as compared to the virgin PP packaging lid (Figure 8a) with a thickness of 100 µm. All recycling blends could be successfully manufactured into films, which confirmed the results from the extensional rheology measurements. The films were opaque and presented a light beige coloring comparable to the color of the recyclate granules and included small (<0.5 mm) spots formed from contaminants or branched gels.

E_p remained rather constant within the 10 reprocessing steps, demonstrating a mean average value of 4.5 mJ. This was 3.3 mJ lower when compared to the virgin MAP lid, but as the parameter is increased linearly via film thickness (virgin lid: 100 µm; recycled film: 50 µm), puncture resistance widely remained the same during the recycling procedure. E_t, which is not affected by film thickness but processing parameters, such as blow-up ratio or film orientation, was about 35% lower in both measurement directions after the first recycling step (longitudinal: 882 Mpa; transverse: 632 MPa) when compared to virgin material (longitudinal: 1384 MPa, transverse: 990 MPa). From the first to the tenth processing step, E_t decreased only slightly in the longitudinal measurement direction from 882 MPa to 842 MPa and from 632 MPa to 500 MPa in the transverse measurement direction, which is in agreement with the approximated molecular weight loss [77]. The mechanical properties of the recycling films were comparable to the original PP-based packaging film properties, which demonstrates the possibility of film-to-film recycling and, therefore, the application of the recyclate in secondary products similar to the purpose they were originally designed for. Currently, food-grade to food-grade recycling is limited by safety concerns regarding the migration or formation of harmful substances during the recycling process, but nonetheless, the recyclates could be used as a middle layer in a multilayer structure to reduce virgin material use [88].

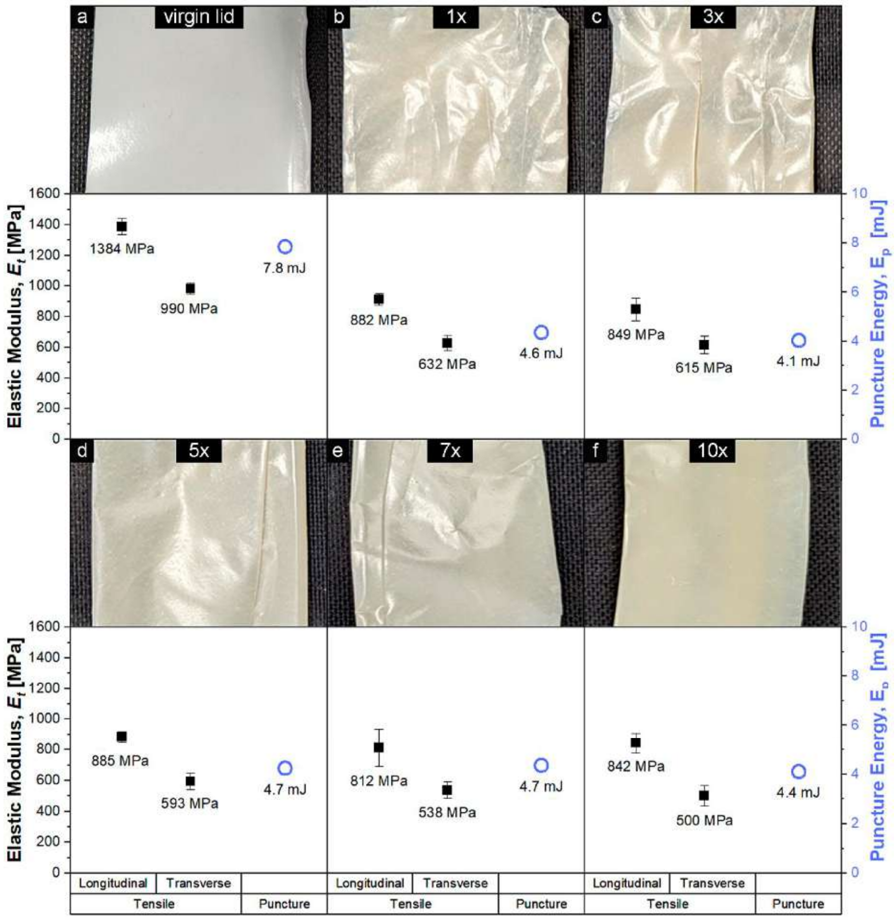


Figure 8. E_t from tensile and puncture energy and E_p from puncture-resistance testing (blown film) of 10 times recycled PP-based MAP (lid + tray) with a thickness of 50 µm (**b**–**f**), compared to the virgin packaging lid with a thickness of 100 µm (**a**).

4. Conclusions

To date, most MAP is, even if collected, currently ending up in the 'reject' streams of recycling units and is incinerated for energy recovery. The leap from a linear to a circular economy for MAP can only succeed if the packaging can be recycled into products of similar economic value (film-to-film recycling). Therefore, repeated film-to-film recycling and the maintenance of film properties over more than one reprocessing step offer great potential to approach circularity and establish multilayers as a valuable source of material in the future.

The prospects for the mechanical recycling of comparable (purpose and shelf-life) commercial post-consumer PET- and PP-based MAP were evaluated via a structural comparison (analyzed using FT-IR, DSC, and cross-section micrographs) of the components (lid or tray) and recycling simulation (cold-wash, grinding, and extrusion), followed by material testing (MFR, E_t, ε_b, and a_{tN}).

In the first step, the compatibility of the components was investigated. The structure analysis revealed that both PET- and PP-based MAP comprised functional layers that provided stability (PET or PP homopolymer) and barrier layers (PE/PP-EVOH-PE/PPE) to block oxygen or moisture. The PET-based MAP, which contained highly incompatible PET and PE, resulted, however, in highly brittle material (*MFR*: 27.9–82.3 g/10 min; E_t: 890–2286 Mpa; ε_b: 2–3%, a_{tN}: 5–10 kJ/m^2), which is, subsequently, not suitable for products of similar economic value to packaging films. In contrast, the secondary material from the PP-based lids and trays exhibited a property profile closely resembling virgin PP (*MFR*: 2.7–4.5 g/10 min; E_t: 1100–1300 Mpa; ε_b: 832–915%; a_{tN}: 44–64 kJ/m^2).

In a further step, the ability to recycle was investigated. Due to the poor mechanical properties and difficult processability (high *MFR*) of PET-based MAP, the repeated reprocessing of this waste stream was not possible. However, the PP-based packaging (lid + tray) was recycled 10 times to demonstrate recycling capability and remained widely intact as the *MFR* increased from 3.7 g/10 min to 11.3 g/10 min, the E_t decreased from 1245 MPa to 944 Mpa, and ε_b and a_{tn} were maintained within the standard deviation at average values of 888% and 69 kJ/m^2. Good melt strength (pronounced strain hardening at strain rates of 0.1 and 1 s^{-1}) for the PP-based MAP after being reprocessed 10 times was estimated by extensional rheology measurements and was confirmed by film-blowing experiments on a micro lab unit.

Even though the manufactured films did not have a flawless surface, for which a high degree of purification that would demand the consumption of resources (energy, chemicals, and water) would have been necessary prior to processing, the appearance had little to no effect on the mechanical properties and, thus, functionality, which allows for its reuse as a polymer film for applications such as secondary packaging.

The PP-based MAP has clearly demonstrated its superiority in terms of "design for" and "design from" recycling characteristics when compared to the PET-based MAP, which is, unfortunately, the predominant packaging source in the Austrian market. These findings make the structure design of PP-based MAP an attractive prospect to improve mechanical recycling rates within this packaging segment (mandatory contribution to the EU recycling targets) to prevent incineration, which is the common end-of-life treatment for multilayer films that combine PET and PE.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/polym15132966/s1, Detailed wt.% calculation; Figure S1: FT-IR spectra of inner and outer PET tray layers.; Figure S2: DSC melt region of PET tray.; Figure S3: FT-IR spectra of inner and outer PP tray layers.; Figure S4: DSC melt region of PP tray.

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Design from recycling: Overcoming barriers in regranulate use in a circular economy

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ABSTRACT

Despite government and consumer attitudes, industry's use of recycled material remains low and narrowly scoped. Critical advances in recycling practices and technology depend on increased scope of application for regranulates, which can be achieved through design from recycling principles. We investigated regranulates and laboratory reprocessed material in the context of processing, ability to meet functional requirements, viability of closed loop recycling, design strategies and co-dependent industry reforms. Reprocessing results in polymer degradation, changes in melt flow rate (MFR) and tensile impact strength (a_{tN}) and restricts polymer processing options. Contamination and multilayer packaging cause changes in MFR, a_{tN}, elastic modulus (E) and elongation at break (ε_b) affecting ability to meet application-specific functional requirements. The problem is complex, whereby the preferences and requirements of recyclers, designers, industry, and consumers are contradictory. New high value regranulate applications using clever design practices are necessary to finance new sorting and processing technology in addition to industry and consumer tolerance and conservative product expectations to circumvent these competing interests.

1. Introduction

The importance of recycling and its relevance to achieving a sustainable future for humans on Earth has been indisputable for over 50 years (Jody et al., 2023). Recycling has an exceptionally positive image; It is often a focal point in new sustainability-related policies and is valued by consumers to the point that it has become highly marketable (Grębosz-Krawczyk and Siuda, 2019). However, despite all the hype surrounding recycling, which would seem to be ubiquitous, the reality is a lot bleaker than one might expect. Only 8.5% of new products are made using recycled polymers (Plastics Europe, 2022). Additionally, 86% of all use of recycled polymers are in just three applications: building and construction (45%), packaging (30%) and agriculture, farming and gardening (11%) (Plastics Europe, 2022).

Manufacturer design preference for virgin over recycled polymers is easy to understand; Recycled polymers often exhibit poorer mechanical properties, can be more difficult to process, less aesthetically and olfactorily appealing and cannot be used for food contact applications (Karaagac et al., 2021a, 2021b). Industry reluctance to work with regranulates, however, retards widespread adoption and implementation of recycled materials; It is not possible to justify investment in improving recycling technologies and infrastructure while regranulates remain useful for only selected low value products, such as bin liners. The growth of recycling in practice, as opposed to as a philosophy, is directly linked with the scope of application for which regranulates can be used (Raghuram et al., 2023).

'Design for recycling', which considers the impact of the design process on the recyclability of materials at their end of life, is a well-known research area (Roos et al., 2019; Sudheshwar et al., 2023; Thompson et al., 2020). Advances include cradle-to-cradle design, where products are designed with disassembly and recyclability in mind (Bjørn and Hauschild, 2018; Hansen and Schmitt, 2021), eco-informed material selection during design (Jones et al., 2022; Law and Narayan, 2022), extended producer responsibility, which holds the manufacturer responsible for the recyclability of their product (Leal Filho et al., 2019) and improved public awareness and education on recycling (Smol et al., 2018; Wang et al., 2020). Other eco-informed design strategies focus on resource pressure and circulation (Desing et al., 2021; Toxopeus et al., 2018) and life cycle assessment (LCA) (Broeren et al., 2016; Rigamonti et al., 2018). However, 'design from recycling', which focuses on the concept of circular economy and examines the extent to which a new product can be produced from existing recycled polymer

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streams and the design specifications required to do so (Ragaert et al., 2020) is highly underrepresented in the literature, yet arguably critical, timely and widely applicable.

Fundamentally, 'design from recycling' is limited by the 'quality' of regranulates, an important yet vague and undefined term pertaining to technical considerations linked but not directly correlated with issues, such as contamination and degradation of polymers (Tonini et al., 2022). Post-consumer polymer waste may be contaminated with inorganic material, such as glass or metal, and organic material, such as food residue or even other polymers, due to imperfect sorting practices (Gazzotti et al., 2022). Polymers also degrade into smaller molecules or fragments under oxidative, hydrolytic, thermal, photo or microbial stress (Hinsken et al., 1991). These processes result in changes in the molecular mass, morphology, and mechanical properties of regranulates, meaning that they no longer meet the design requirements of a given application. Contamination and polymer degradation effects in recycled material are well documented in the literature (Archodoulaki et al., 2022; Chamas et al., 2020; López et al., 2014) which typically focuses on characterizing property changes and their relevance within the context of polymer science and polymer processing, i.e., changes in melt mass-flow rate (MFR), increased stiffness and more brittle fracture behavior. How these changes affect the combination of abstract (e.g., aesthetics, user experience, etc.) and technical (e.g., material selection, manufacturing processes, etc.) aspects associated with design and ramifications for stakeholders along the value chain are, however, rarely considered.

Even moderate changes in polymer processing and mechanical properties result in downcycling (or cascading) i.e., conversion into products of lower quality or value, rather than recycling of the material i.e., conversion into products of equal or higher quality or value for the same (closed-loop recycling) or a different (open-loop recycling) application to the original application of the polymer (Jehanno et al., 2022). The relationship between material 'quality', recycling process and destined application can be modelled in the context of economics (Nelen et al., 2014), LCA (JRC, 2018) and technical metrics (Hummen and Sudheshwar, 2023). However, while these studies do attempt to join their corresponding parameters with sector and process analysis, their scopes are often very narrow, and they offer little big picture perspective on recycling reform or the roles of and interactions between the key stakeholders.

We investigated the processing and mechanical properties of common commercial regranulates in Austria, laboratory reprocessed material and corresponding virgin polymers as a reference. Results were interpreted within the context of design from recycling and used to assess design from recycling considerations, such as processing restrictions, issues with polymer degradation and contamination, polymer ability to meet functional requirements, the viability of closed loop recycling, design strategies and co-dependent industry reforms to promote increased use of recycled polymers. With a technical focus on the recycler and designer, the polymer processing issues they encounter, their individual and shared property preferences and requirements, while considering ramifications down the supply chain for the manufacturing industry and consumers, we achieve a diverse technical perspective that is holistic and broad scoped yet well linked with the roles of key stakeholders and critical polymer science and processing fundamentals.

2. Experimental section

2.1. Materials

LD 310 E low density polyethylene (PE-LD) was purchased from Dow Chemical Company, (Michigan, U.S.A), while MB6561 high density polyethylene (PE-HD) and BC918CF polypropylene (PP) copolymer were kindly provided by Borealis (Vienna, Austria). H7058–25R PP was kindly provided by Braskem (Rotterdam, Netherlands). PE-LD and Dipolen S PE/PP regranulates were purchased from Rissland (Katzhütte, Germany) and MTM Plastic (Niedergebra, Germany) while Systalen 7002 PE-HD regranulate and RPPC03GR PP regranulate were kindly provided by Duales System Holding (Cologne, Germany) and Total (Paris, France), respectively. All regranulates had an ash content < 2 wt.% and no other known impurities. These materials were used as received. PE-LD snack packaging and shrink film contaminated with PE-LLD, PE-HD cosmetic bottles and PP film (modified atmosphere packaging), yogurt cups and buckets were also purchased or collected from post-consumer waste bins in eastern Austria. All laboratory recycled materials had an ash content < 5 wt.% and contained no polymeric contamination, except for PP films which contained < 5 wt.% PE. Collected materials were washed and milled prior to use.

2.2. Reprocessing of virgin PE-LD, -HD and PP

Virgin PE-LD, -HD and PP were reprocessed to simulate the recycling cycle. Virgin granulates were extruded using a single screw extruder (EX-18–26–1.5, Extron Engineering Oy, Finland) with a screw diameter of 18 mm and length to diameter ratio of 25:1 at 240 °C and 70 rpm screw speed. The extruded material was then ground into flakes using a mill (Fritsch Pulverisette 19, FRITSCH GmbH, Germany) to obtain the reprocessed samples. Recent standards, such as DIN SPEC 91,446 focus on the classification and trading of recycled plastics, indirectly relating to the recycling process by standardizing the classification of recycled plastics based on Data Quality Levels (DQLs) and promoting better understanding of the quality and characteristics of the materials being traded and used (Deutsches Institut für Normung, 2021). However, no standards currently exist directly governing the reprocessing of material by recyclers. The described process is nonetheless analogous to typical industry practices (Shamsuyeva and Endres, 2021). The reprocessing and grinding process was repeated ten times to simulate ten recycling cycles with samples collected and tested after 1, 3, 5, 7 and 10 cycles. 10 × reprocessing of a material is an extreme case and represents the upper limit or an unlikely scenario based on currently available infrastructure.

2.3. Compression moulding of PE-LD, -HD and PP virgin material and regranulate and preparation of the mechanical test specimens

PE-LD, -HD and PP virgin material and regranulates were compression moulded (Collin P 200 P, Germany) at 180 °C and 50 bar with a cooling rate of 20 K/min to prepare mechanical test specimens for all materials. At least ten dog-bone tensile (thickness 1.8–1.9 mm) and tensile impact test specimens (thickness 1.1–1.2 mm) were cut from each compression moulded sheet in accordance with ISO 527–2-A5 (The International Organization for Standardization, 2012) and ISO 8256/1A (The International Organization for Standardization, 2004), respectively. Tensile impact test specimens were notched with a Notch-Vis tool (Ceast, Germany).

The MFR was measured for at least ten replicates of each sample according to ISO 1133–1 (The International Organization for Standardization, 2011) at 230 °C under 2.16 kg load on the MeltFloW basic (Karg Industrietechnik, Germany). These conditions were selected to enable comparison of PE and PP at the same temperature.

2.4. Tensile (impact) testing of PE-LD, -HD and PP virgin material and recycled samples

A universal testing system comprising a Zwick 050 frame, 1 kN load cell and extensometer (Zwick Roell, Germany) was used to perform tensile tests on the prepared specimens at a constant velocity of 10 mm/min. The elastic modulus E, tensile strength σ_{UTS} and elongation at break ε_b were calculated using the ZwickRoell testXpert II software (v. 3.6) across five replicate tests. An Instron 9050 impact pendulum (Ceast, Germany) was used to establish the tensile impact strength a_{tN} of the notched samples across at least ten replicates.

3. Results and discussion

3.1. Fundamental technical problems with recycling and recycled materials

3.1.1. Restricted processing options based on polymer degradation and contamination

The high temperatures and shear forces associated with re-extruding polymers during recycling results in increased crosslinking and branching (primary mechanism for PE) and chain scission (primary mechanism for PP) associated with changes in molecular mass and weight distribution (Hinsken et al., 1991). These changes in chain length affect polymer crystallinity and shrinkage in turn, necessitating tool changes by the manufacturer. They also affect the MFR and melt strength making undesired changes to machine parameters e.g., injection speed and pressure in injection moulding necessary for processing or worse, restrict polymer processing options and subsequently the products and applications for which the polymer mixture can be used (Demets et al., 2021). Pipe extrusion and extrusion blow moulding, which is used to produce hollow vessels such as bottles, require viscous blends with low MFRs (Table 1). Conversely, film extrusion often used to produce packaging requires a considerably higher MFR (1.5–5 g/10 min) and injection moulding, which is used to produce parts with more intricate geometries such as screw on caps, requires an even higher MFR (6–16 g/10 min). These polymer processing options are also restricted to use with polymers exhibiting a property profile in line with the requirements of the end product e.g., PE-LD or -LLD for film extrusion and PE-HD for pipe extrusion, extrusion blow moulding and injection moulding.

Extrusion-associated crosslinking and branching typically results in a reduced MFR in PE while chain scission results in the opposite effect in PP (Yin et al., 2015). The processing time associated with extrusion is the most influential factor associated with this change in MFR, with longer processing times associated with greater reductions in the MFR of PE and increases in the MFR of PP (Martey et al., 2021; Schall and Schöppner, 2022). Temperature and shear rate (screw speed) also promote changes in MFR but to a lesser extent. Contamination of PE with PP and vice versa, a common problem in recycled polyolefins due to their similar densities and hence more challenging sortability, also affects MFR in all applications detailed in Table 1 but especially in injection moulding (Karaagac et al., 2021b). Multilayered structures, which are used in ~30% of polymer packaging, by definition also comprise multiple polymers, such as PE (PE-LD, PE-LLD) and PP (homopolymer, random and block copolymer), and are not easily separated using common sorting technologies (Schmidt et al., 2022). A single processing step can see the MFR of PP contaminated PE increase by as much as 10% (Karaagac et al., 2021a). With up to 10 wt.% PP contamination common in PE recycled mixtures this can cause an otherwise reducing MFR to remain constant or increase slightly (Juan et al., 2021). While the MFR of virgin polymers is very easily modified using highly reactive additives e.g., peroxides or cross-linking agents accurately dosed based on the virgin polymer type it is much more challenging in recycled polymers due to organic and inorganic contaminants which might either act as reaction accelerators or inhibitors making the effects of additives unpredictable (Maris et al., 2018). Polymer degradation during re-extrusion affects the recycling yield since crosslinked or branched gel is typically removed by the recycler and discarded at an early stage in the recycling process (Schyns and Shaver, 2021).

Table 1
MFR measured at 230 °C for 2.16 kg of granulate.

Process	Polymer	MFR (g/10 min)	Products Virgin	Recycled
Pipe extrusion	PE-HD	0.2–0.6	Pipes	Pipes
Extrusion blow moulding	PE-HD	0.6–2.7	Hollow vessels	Hollow vessels (coloured)
Film extrusion	PE-LD/LLD	2.7–9	Films, packaging	Bin liners (coloured)
Injection moulding	PE-HD	10–20	Complex geometries	PE/PP injection moulded parts

3.1.2. Closed-loop recycling: ambitions and paradoxes in a post-consumer world

Closed loop recycling can be seen as a best-case scenario in recycling terms, i.e., a separately collected waste stream of known polymers with little contamination sourced from e.g., battery housings, car bumpers (Kozderka et al., 2017) or beverage bottles. It represents a tiny fraction of the recycling industry and practical realizations remain limited to PET bottle-to-bottle recycling and some examples utilising PE-HD milk containers (Gaduan et al., 2023).

The main reason for the stunted adoption of closed-loop recycling is a lack of suitable input material. ~90% of recycled material is post-consumer waste, which is typically dirty, mechanically deformed and derived from widely used packaging materials with short service lives (Archodoulaki and Jones, 2021; Soares et al., 2022; Tukker, 2012). Colour and odour problems in recycled polyolefins coupled with legal requirements associated with food packaging and reductions in their mechanical properties often make closed-loop packaging-to-packaging recycling impossible. Volatile organic compounds migrating into the polymer matrix (Cabanes and Fullana, 2021) downgrade the use of this material to products that do not use white or natural colours (Golkaram et al., 2022). (Hot and cold) washing procedures help to target unpleasant odours but do not improve mechanical properties, which makes their inclusion less economically and ecologically viable (Bashirgonbadi et al., 2022).

Closed-loop recycling also has limitations in terms of long-term viability, due to the limitations on the number of times the material can be recycled before it degrades e.g., 2–3 times for PET (La Mantia and Vinci, 1994). Most PE regranulates also have a lower MFR than virgin polymers due to the predominance of crosslinking mechanisms and finding a regranulate that is suitable for injection moulding is subsequently very challenging (Mendes et al., 2011). This restricts the design options for recycled PE-HD to pipe extrusion and extrusion blow moulding, effectively downgrading injection moulding grade PE-HD for use in these more restricted applications as its MFR reduces (Oblak et al., 2015; Yin et al., 2015).

Mixing ‘cleaner’ waste streams with virgin polymers can help to combat the effects of polymer degradation and dilute contamination but often relies on large quantities of virgin material to do so, consequently failing to promote dominant use of recycled material in products. A mixture comprising 30% regranulate and 70% virgin material statistically contains only 0.8% material that has be processed ≥ 5 times, which may yield satisfactory properties for many applications (Niessner, 2022). However, mixtures comprising 70% regranulate and 30% virgin material contain ~5% material that has been processed ≥ 5 times, which will limit the applications for which the material can be used.

In addition to most post-consumer waste and consequently most recycled material not being suitable for closed-loop recycling, it is very challenging to repurpose it for another application. This is because the dominance of packaging in this stream results in the presence of a narrow range of polymers used for packaging e.g., PE-LD, PE-HD, PP, PET designed with a property profile to meet this single application and not others (Horodytska et al., 2018). For example, the mechanical properties of PE-LD- and -LLD are most suitable for production of films and there are limited options other than to use them for their original application – packaging (Franz and Welle, 2022).

Despite these limitations of mechanical recycling, the aim should be to retain and maintain quality within the primary recycling loop and integrate secondary materials as a design standard prior to chemical recycling or energy recovery.

3.1.3. Impaired ability to meet functional requirements due to polymer degradation and contamination

Processing limitations aside, the other fundamental technical challenge when designing with recycled polymers is 'material functionality', i.e., the specific properties and characteristics of a material that determine its suitability for a particular application or purpose. Important mechanical properties of recycled polymers, such as E, σ_{UTS}, a_{tN} and ε_b, are inferior to virgin polymers (Golkaram et al., 2022; Thoden van Velzen et al., 2021). They also can't be modified using additives such as plasticizers (as is possible in virgin polymers) due to the risk that substances within the degraded polymer matrix will leak (Shi et al., 2022). This makes design from recycling more challenging since the restricted property profiles of recycled polymers may not match the functional requirements of an application or product.

Contamination in recycled mixtures has arguably the greatest effect on their mechanical properties. Post-consumer waste streams may contain a wide range of different materials, both inorganic and organic. Inorganic matter, such as glass, metal and even organic contaminants, such as food and beverage residues or paperboard can often be easily separated from polymers using a range of different sorting and processing technologies, including hot wash, air classifier systems, magnetic separation and sink-float sorting (Lange, 2021). But the inability to accurately sort polymers from each other results in polymer-based contamination of one polymer within a mixture with another (Qu et al., 2022). Since these polymers all have different properties, including MFR, E, ε_b and a_{tN} even a small quantity of polymer contamination in a mixture predominantly comprising another polymer can result in considerably altered mechanical properties. PP contamination in PE is an especially good example of this with the presence of <10 wt.% PP in a PE mixture resulting in a considerably higher E and much lower ε_b and a_{tN} in the mixture than would be present in virgin PE (Karaagac et al., 2021a). This can affect the suitability of a mixture in both cases where it is intended for use as a film and in cases where it is intended for use as a rigid vessel (Fig. 1**a, b**).

PE-LD and -HD regranulates exhibit lower MFRs and PP regranulates higher MFRs than virgin material (Fig. 1**c**). PE-LD regranulate exhibits considerably lower MFR and a_{tN} than virgin material but little difference in E and ε_b. PE-HD and PP regranulates exhibit considerably lower E, a_{tN} and ε_b than virgin material. Laboratory processed samples typically exhibit greater differences in properties compared to virgin material than commercial regranulates (Fig. 1**d**). This is likely due to the absence of technologies used in industry, such as degassing, melt filtration and stabilizing processing aids e.g., additives. Re-extrusion based polymer degradation (crosslinking and chain scission) with respect to processing time—the most influential factor affecting crosslinking—also greatly affects the MFR and mechanical properties of regranulates (Fig. 1**e**). The MFR of PE-LD and -HD regranulates decreases with increasing reprocessing cycles, while that of PP homo- and copolymers increase. a_{tN} decreases before stabilizing with increasing reprocessing cycles in PE-LD and PP copolymer and a_{tN} is largely unaffected by reprocessing cycles in PE-HD. However, PP copolymers are far more susceptible to a_{tN} losses with increasing reprocessing cycles. It should be noted that these effects are only the result of material damage due to mechanical stress and heat (processing) and neglect the influences of contamination and degradation that occur during the service life of a product, such as irradiation and oxidation (weathering).

The concept of material functionality is critical to design from recycling, since the properties of regranulates will always diverge from what would be considered optimal or desirable for design purposes. Achieving the property profile of virgin material isn't possible, and even if it was, it would be prohibitively expensive to do so. Consequently, the economic and successful use of regranulates can only be considered within the context of suitability for a given application. Expression of the material functionality of regranulates using a single aggregate metric alongside LCA or environmental impact assessment (EIA) data would enable designers to maximise environmental benefit in products. The establishment of simple yet insightful and accurate metrics to aid design decisions must be a priority.

3.2. A necessary compromise: designing with recycled materials

One of the biggest problems currently hindering the expanded use of recycled materials is consumer perception (Polyportis et al., 2022). Or at least the lack of willingness from companies to risk consumers rejecting their products if they were to be made from recycled material due to issues associated with reduced material properties, less desirable colours or odours (Park and Lin, 2020). This is perhaps a baseless fear given the exceptionally positive image that environmentally friendly and recycled products currently enjoy in society (Ketelsen et al., 2020). These factors are in fact typically considered highly marketable selling points these days (Gelderman et al., 2021).

While marketing is obviously important in shaping consumer perceptions, designers play the most active and fundamental role in product image and function (Michelini and Razzoli, 2004). Whether a product is successful or not largely depends on how it looks and how well it works (Kumar and Noble, 2016). Whether a product is perceived as 'cool' also largely depends on these factors (Tiwari et al., 2021). Designers will play a pivotal role in reinventing the norm – setting new unwritten standards on acceptable colour deviations, odours and material properties (Mugge, 2018). Less than perfect needs to be okay with the redeeming factor that the product is more environmentally friendly (Ehrenfeld, 2008). Expanding the range of accepted applications for recycled materials will be the key factor in increasing recycling rates and improving the utilisation of recycled streams (Perry et al., 2012). Recycled materials must be associated with cool products, not just bin liners and similar.

Of course, that is easier said than done. Issues with extrusion- and contamination-based mechanical property degradation in recycled polymers do make their property profiles very different from virgin polymers – more limited and much less easily modified **(**Fig. 1**)**. This causes issues in both processing, where the MFR plays a considerable role in how the recycled material can be processed, in addition to the ability of the recycled material's mechanical properties to service product function (Hopewell et al., 2009). Managers and designers must manage their expectations and tolerance for the time and human resources required to achieve optimal processing conditions, profit margins and product quality when working with recycled polymers (Kumar et al., 2021). Optimisation of extrusion processes to minimise processing time (and less importantly shear rate and temperature) and reduce crosslinking and improving sorting capabilities to reduce contamination without affecting production viability will constitute a considerable investment of time and money (De Weerdt et al., 2022). These compromises are perhaps more palatable when considered in the knowledge that they will play an important role in creating a more sustainable society and that since climate change affects us all, they will ultimately be marketable.

Designing with recycled polymers is more challenging, but possible (Preka et al., 2022). MFR is a strong and useful indicator of polymer degradation for designers and can be used to gauge when special attention must be afforded to the design process. To ensure products meet safety standards and expected minimum service life using degraded material that is less durable than virgin polymers and exhibits varied and unpredictable material properties, designers must over-engineer e.g., thicker walls and avoid notches, sharp curves and edges and filigree details e.g., flap hinge closures (Martínez Leal et al., 2020). There is massive potential for improved design practices across many products and industries (Dokter et al., 2021; Rauch et al., 2022). Minimising the time required to achieve a design (often prioritised) comes at the expense of any real investigation or understanding as to how well a design really fulfills its requirements (Stechert and Franke, 2009). Many designs using virgin polymers considerably outperform their baseline requirements (Hauschild et al., 2020). Finite element analysis and other

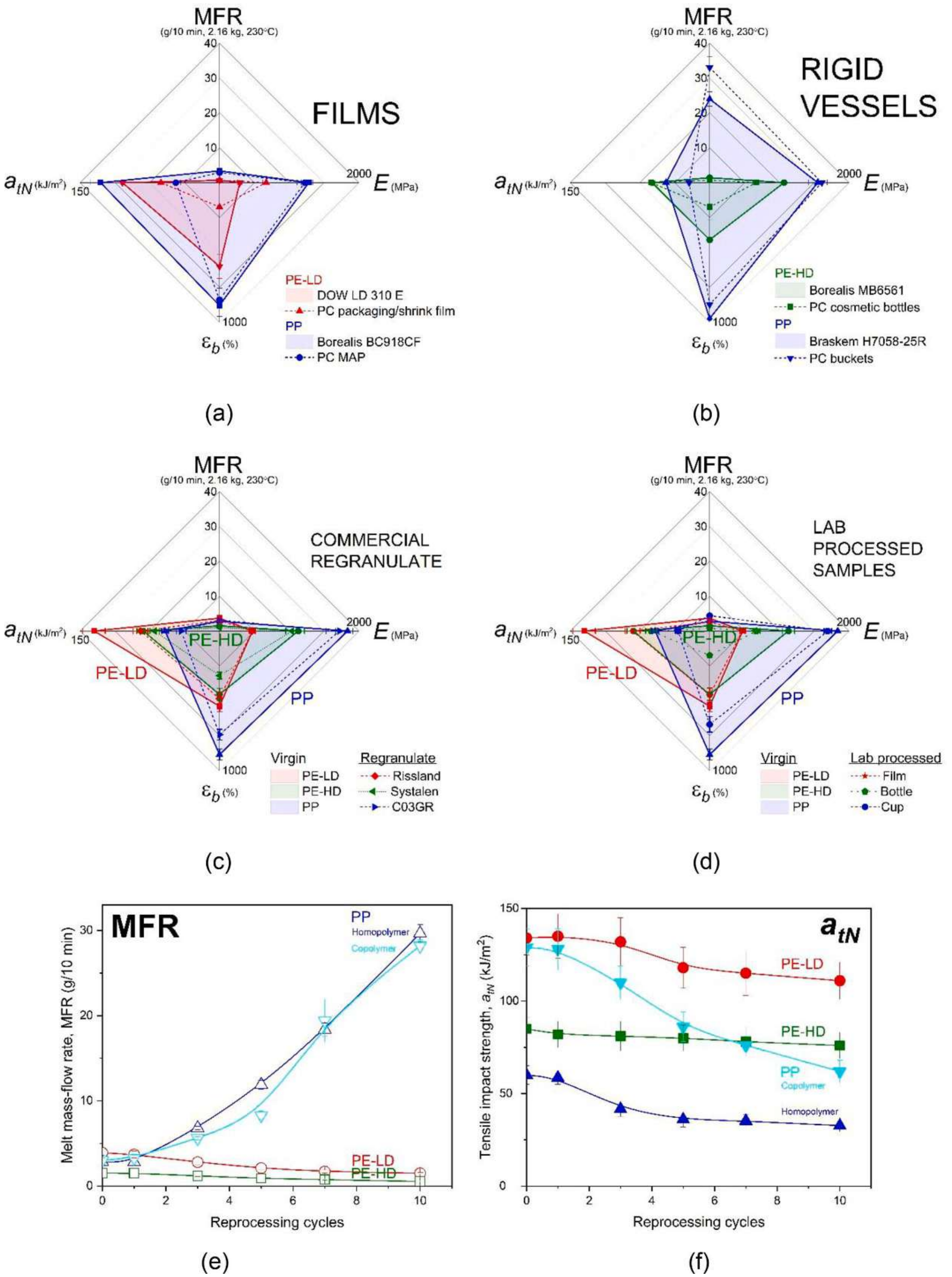

Fig. 1. Melt mass-flow rate MFR, elastic modulus E, elongation at break ε_b and tensile impact strength a_{tN} property profiles of virgin and post-consumer (PC) low (PE-LD) and high (PE-HD) density polyethylene and polypropylene (PP) based (a) films and (b) rigid vessels; differences between (c) commercial regranulates and (d) lab processed samples compared to virgin material; and the effect of reprocessing cycles on polymer (e) MFR and (f) a_{tN}.

simulation and modeling is readily available to assist in design and could be used to generate designs suitable for recycled polymers (Mulakkal et al., 2021). Reduced mechanical properties and other recycling-derived polymer degradation effects, such as altered degree of crystallinity (Vilaplana and Karlsson, 2008) can also be offset by over-engineering, which is as described an already common practice – increasing product thickness to fulfill function (Maris et al., 2014). Ironically, the concept of overengineering using recycled material does, however, necessitate the use of far more plastic than would be needed to fulfill the same function if virgin polymers were used. Notably, not all properties are affected equally by degradation and subsequently the suitability of recycled material for a product does depend on how it will be loaded e.g., flexural modulus is most important in plastic sheets (Golkaram et al., 2022). Smarter design practices could also include the separation of products into parts (supporting design for disassembly practices), each with unique functional requirements and the use of material combinations and synergies to produce multi-material structures (Abuzied et al., 2020). Suitable parts could be produced using recycled polymers while parts with more demanding functional requirements could be produced using other more suitable materials (Martínez Leal et al., 2020). Similarly, crosslinking-derived gels in films that affect their esthetic can be used as the middle layer in three layered structures to offset use of virgin material (Radusin et al., 2020).

The final important consideration for designers is the cyclic nature of the material stream and design when it comes to recycling (Cândido et al., 2011). Design from recycling is rather unsurprisingly closely linked with design for recycling. If products are designed to be more easily sorted and recycled (design for recycling) then designers will also see these benefits carry through to the design from recycling stage in the form of higher quality recycled mixtures with less contamination and superior mechanical properties, making the design from recycling process easier (Svanes et al., 2010). The same is true of diversification of the material stream. If designers want to work with certain polymers at the design from recycling stage then they obviously need to introduce those polymers into circulation during the design stage (Venkatachalam et al., 2022).

3.3. Co-dependent strategies for recycling industry reform

Contamination is the main problem in recycling streams (Fig. 1). Design from recycling, while critical, relies on ‘fully compatible’ regranulates, with < 5–10% contamination. Even clever technology assisted design practices have their limits with ‘limited compatibility’ regranulates ($\geq$ 30% contamination) currently effectively useless for design from recycling. Achieving better quality (or even useable) recycled polymer mixtures for designers is going to depend heavily on improved sorting and recycling practices that achieve lower levels of mixture contamination (Zelenika et al., 2018). Current sorting technology typically relies on ballistic and density separation methods and near infrared (NIR) technologies, which are limited in precision and can be disrupted by product designs that utilise multiple layers or coatings that confuse infrared sensors. More sophisticated technologies utilising precision object identification, intelligent adaptive systems and machine learning algorithms are under development but again the interlinked nature of design and recycling must be emphasised: the viability of more complicated and expensive sorting and recycling technologies relies on greater market demand for recycled polymers and more applications with higher product values to compensate for the additional costs associated with generating superior recycled polymer mixtures (Fig. 2). Designers drive these changes but are reluctant to do so due to the inferior quality of recycled compared to virgin polymers (Demets et al., 2021).

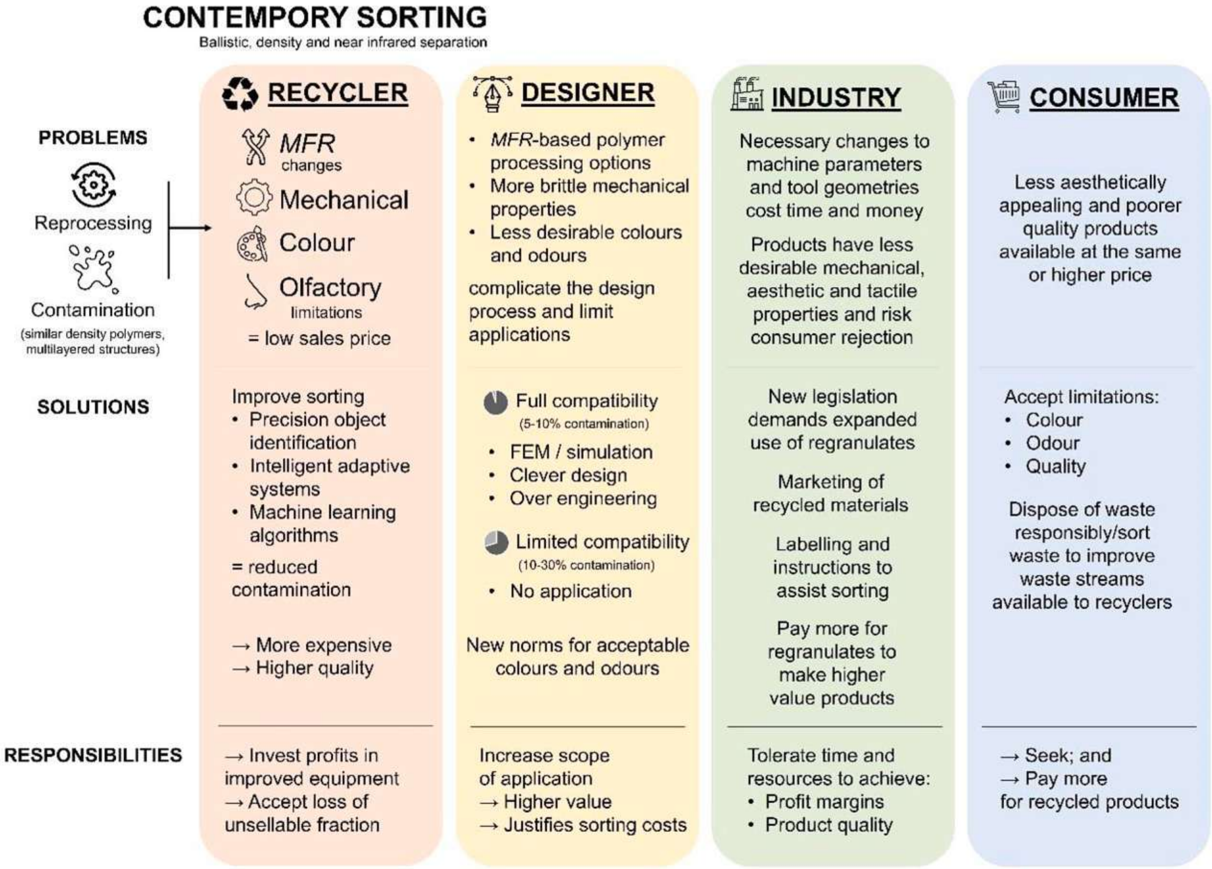

Fig. 2. Interlinked and co-dependent problems, solutions and responsibilities of recyclers, designers, manufacturers (industry) and consumers currently hindering design from recycling practices with opportunities for improvement through collaboration.

Future policy and legislation must focus on establishing and adopting new protocols and standards, improving operational feasibility, minimising associated administrative burdens and enforcing compliance with new legal frameworks. Recycling industry reforms targeting contamination levels $< 5\%$ can be achieved through more accurate sorting based on the tagging of products with detailed recycling information that can then be scanned on collection. This is demonstrated in the European Union-funded initiative ‘Holy Grail’, which is aimed at improving the efficiency of plastic waste sorting and recycling using digital watermarks and artificial intelligence, to create a "digital twin" of plastic packaging that can be read by sorting machines to identify the composition and recyclability of the packaging (Taneepanichskul et al., 2022). Other more generic options include the sorting of flakes using near infrared technologies to identify PP contamination or better, deliberate avoidance of the use of similar density polymers, such as PE and PP for the same application (design for recycling) (Martin De et al., 2010). That said, superior recycling would almost certainly change the type of recycled products available and their distribution (Tonini et al., 2022). Rather than a wide range of fairly uniformly mediocre recycled mixtures there would be a stark divide between high-quality, purer and obviously more expensive recycled mixtures and very low quality, highly contaminated mixtures of polyolefins that would potentially be without use (Jacobs et al., 2022).

On a final note, it is important to remember that design from recycling can help when dealing with contaminated and degraded materials but that we should be attempting to eliminate the root cause of the problem rather than the consequence. Our priority should be to design systems which avoid losses in quality in the first place. This is more representative of system change than product design as evidenced by PET bottle-to-bottle recycling, which largely owes its success to a clean, separate collection system in combination with appropriate product design. That said, the interconnected nature of the recycling sector and all its stakeholders, as discussed in this article, does mean that system change is reliant on a common goal and strategy acted upon by all stakeholders based on careful consideration of mutual and mutually exclusive priorities, requirements, and limitations.

4. Conclusion

While virgin polymers provide consistent, desirable, and customisable processing, mechanical properties and esthetic that make them popular with designers, regranulates are undisputedly more difficult to work with. High temperatures and shear forces used during recycling result in polymer degradation and subsequent change in MFR and a_{tN}. Polymer-based contamination in recycling streams and deliberate use of multilayer packaging affect MFR, E, a_{tN} and ε_b, meaning regranulates may fail to meet the functional requirements of many applications. PE-LD regranulates exhibit considerable differences in MFR and a_{tN} to virgin material. PE-HD and PP regranulates also notably differ from virgin material in MFR E, a_{tN} and ε_b. Current sorting technologies cannot effectively combat these issues and in many cases the use of additives may not be desired due to risk of leakage. More advanced technologies are being developed; however, the future of recycling lies in the hands of designers, industry and consumers: the key players in a vicious circle. Regranulate value must be high enough to warrant investment in new sorting and processing technology, which is only the case if designers actually use regranulates. Better quality regranulates depend on reduced contamination, which can be aided by designing products to be more easily disassembled and sorted in the first place. Polymers that designers want to work with need to be introduced into circulation during design if they are to reappear later in the material life cycle. Industry must be willing to manage expectations and tolerance for time and human resources required to achieve optimal processing conditions, profit margins and product quality when working with recycled materials. Consumers must be willing to trade product esthetic for sustainability. The scope of application, value and interest in recycled material must increase for investment and advances in recycling. This can be achieved by clever design practices, finite element analysis, simulation, and modeling to use regranulates for interesting new products, not bin liners.

CRediT authorship contribution statement

M.P.J. and V-M.A. conceptualised the manuscript. J.R and M.S. completed the experimental work. M.P.J visualised the data. All authors contributed to formal analysis, writing of the original draft and review and editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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SKILLS

LANGUAGES

German	● ● ● ● ●
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French	● ● ○ ○ ○

IT-SKILLS

MS Office	● ● ● ● ●
OriginPro	● ● ● ● ○
SAP	● ● ● ○ ○
Programming	● ● ○ ○ ○
CAD	● ● ○ ○ ○
Lab software (RheoPlus, testXpert, TA Universal Analysis)	● ● ● ● ○

ABOUT ME

Impassioned to make the future more sustainable

I am a goal oriented, determined and ambitious person with a passion for problem solving of complex tasks, sustainable and efficient production circles and construction materials (especially polymers) in all aspects. An active lifestyle and curiosity for the unknown shape my work-life balance.

WORK EXPERIENCE

Project assistant **since 02.2020**

TU Wien - Research Group for Structural Polymers, Vienna

- Research mechanical packaging recycling (project partnership with Borealis)
- Publication of research findings (scientific articles, conferences)
- Supervision of scientific theses (bachelor and master)

Student employee (part time) **11.2015 - 11.2019**

TU Wien - University's Development and Quality Management, Vienna

- Organizational support tasks (proof reading, statistical analyses)
- Monitoring and actualization of key processes (SAP, MS Visio)
- Assistance with the preparation of reports and quality metrics

Internship Railway (1 month) **2015, 2016, 2017**

TSA - Traktionssysteme Austria, Wr. Neudorf

- Employment in the sales and engineering department
- Risk analyses, updating documents and projects (SAP), department head assistance, assistance product catalog revision and redesign

EDUCATION

TU WIEN **10.2011 - today**

- Doctoral studies: Materials science (polymer packaging recycling)
- Master: Mechanical Engineering - Management
- Bachelor: Mechanical Engineering - Management

Secondary School **09.2007 - 06.2011**

Don Bosco Gymnasium Unterwaltersdorf

- Language focus
- Matura: 1.0