A circular multilayer plastic approach for value retention of end-of life multilayers films

D1.2: Circularity by design guidelines for the next generation of recyclable films capable of substituting today's difficult to recycle multilayers

WP1: Systemic transition to the circularity of multi-layer filmsT1

Project Information

Grant Agreement n°	101003864
Dates	1st June 2021 – 31st May 2024

Document status

Document information

Deliverable name	**D1.2: Circularity by design guidelines for the next generation of recyclable films capable of substituting today's difficult to recycle multilayers**
Responsible beneficiary	6-LEY
Contributing beneficiaries	1-IPC 2-TNO 3-VTT 8-EVERSIA 10-BARBIER 11-PAPREC
Contractual delivery date	31/05/2023
Actual delivery date	29/05/2023
Dissemination level	PUBLIC

Document approval

Name	Position in project	Organisation	Date	Visa
C.Chevallier	Coordinator	IPC	30/05/2023	OK

Document history

Version	Date	Modifications	Authors
V1	01/05/2023	Document created	S.Zakarya
V2	02/05/2023	Revision	J.M.Brouillat
V3	16/05/2023	Revision	M. Lions, R. Josselin, C. Chevallier, C. Rambaud, A. Runstraat, M. De Lint
V4	29/05/2023	Revision	R. Salcedo Santana, A. Bourely, I. Rytöluoto
VF	30/05/2023	Final review	C. Chevallier

Table of content

Abbreviations

PE: Polyethylene

PET: Polyethylene Terephthalate

PA: Polyamide

PP: Polypropylene

EVOH: Polyethylene vinyl alcohol

HDPE: High Density Polyethylene

LDPE: Low Density Polyethylene

PVC: Poly vinyl chloride

PVDC: Polyvinylidene chloride

AlOx: Aluminum Oxide

SiOx: Silicon Oxide

PO: Polyolefins

SEBSgMAH: Poly (styrene ethylene butylene styrene) grafted maleic anhydride

BOPPmet: Metallized bioriented polypropylene

PETmet: Metallized polyethylene terephthalate

MNL: multi nano layer

TD: Transverse direction

MD: Machine direction

NIR: Near Infra Red

rPE: Recycled Polyethylene

DfR: Design for Recycling

List of illustrations

List of tables

Executive summary

The CIMPA project aims at developing a circular value chain for post-consumer multilayers, multimaterial packaging films and agricultural films. With this purpose, partners study and develop sorting, recycling, decontamination and upgrading technologies but they also have to consider the first step of the value chain, that is the design of the structure of the packaging.

Indeed, for several years now European countries have developed their own ways of assessing recyclability, based on their specific consumption mode, collection and recycling model, to give guidelines for their internal market and valorization path, leading European industries to be forced to consider each country, one by one when it comes to recyclability rules. These guidelines are necessary for the countries as specific consumption will lead to a specific mix of waste and composition into the bale and that is why most of DfR recommendations are the same in all countries. But sometimes there are specific rules depending of the collection & sorting streams, thus recycling processes & applications (color sorting, mix flexible PO, opaque PET…). This non-harmonized European map of guidelines led the industrial players to find new certification paths like RecyClass, that also have their own point of view on the different components of a packaging, but that may answer the question of recyclability more objectively and considering only one country.

Today as the Plastic and Plastic Waste Regulation is being revised, we see a will of harmonization in the European Commission to be able to treat a common flow of waste that comes from packaging and every kind of plastics in the market. To face this challenge, the standards that will be created need some strong assessment of recyclability and sorting ability of the different kinds of structures and materials present today, considering the actual technologies, but they also need to have an opinion on the possibilities that come with innovation. Moreover, this revision and the general European legislation on recycling rate and use of recycled material gives high challenges to the industry, that needs to have some guidance on how to design a product to be recycled and reused in a sustainable, closed loop.

In this context, the partners of the CIMPA project collected the information from the different work packages consisting of sorting and recycling technologies to build some guidelines on the use and the limits of the materials studied. To do so, a first map of European guidelines was drawn to depict the context and the actual differences that represent the points of interest to be studied. Then these guidelines were compared to the results obtained using the CIMPA technologies step by step (lab scale, pilot and industrial scale) to determine the added value of these innovations and finally new guidelines with some typical structures were explored.

Deliverable report

Introduction

According to the article 6 of the proposed revision of Packaging and Packaging Waste Regulation, a packaging is considered recyclable if it complies with the following:

"(a) it is designed for recycling;

(b) it is effectively and efficiently separately collected in accordance with Article 43(1) and (2);

(c) it is sorted into defined waste streams without affecting the recyclability of other waste streams;

(d) it can be recycled so that the resulting secondary raw materials are of sufficient quality to substitute the primary raw materials;

(e) it can be recycled at scale. "[1]

Following this definition, the present guidelines for recyclability will focus on **three main steps of the value chain**: sorting, recycling, reprocessing. The process of designing the packaging will be addressed by the guidelines and the collection is a topic that should be addressed separately as it is a matter of communication, education of the consumer and logistics of collection.

As stated earlier, this report will first draw a picture of actual national or industrial guidelines for recycling that may exist in the EU. Once this basis is set an overlook on the work done in the CIMPA project according to the three topics of sorting, recycling and reprocessing at lab, pilot and industrial scale will be made. All this information will allow to finally define guidelines for the design of recyclable multilayer multimaterial films for food packaging.

Considering the scope of the CIMPA project that is about the multimaterial structures used in agricultural and food packaging application, this report will focus on guidelines for recycling when it comes to choosing the base material of the design and the barrier material added to this structure. No work has been done over closure devices, labels, lamination adhesives, printing or other parts.

The information contained in this report includes the synthesis of the technical work realized by the CIMPA partners from lab scale with virgin grade polymers to industrial scale with recycled material from post-consumer waste. To demonstrate the results while keeping confidential results hidden, every numerical value will be normalized to the ones of virgin grade PE film.

[1] According to «Proposal for a
REGULATION OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL
on packaging and packaging waste, amending Regulation (EU) 2019/1020 and Directive (EU) 2019/904, and repealing Directive 94/62/EC » Article 6 paragraph (2) p56

This report is the first phase report and will be updated at the end of the project with the last values of the project, mainly considering the industrial scale trials.

Description

Map of the European Guidelines for Recycling

The different countries of the EU do not all have specific guidelines for recycling of the flexible packaging. In this context and with the need of the industry to have indication on the recyclability of different designs, many entities tried to answer the issue. The Table 1 is summarizing the different guidelines existing at European or national level with official guidelines and more theoretical ones (compilation of scientific knowledge and empirical trials).

		France (COTREP)	Austria (FH Campus de Vienne)	Central Agency Packaging Register	Recyclass	CEFLEX	Suez Circpack	network for circular plastic packaging
Base material	PE, all kind	recyclable	recyclable	recyclable	recyclable	recyclable	recyclable	recyclable > 90%
	PE Composites (mixture of different PE)	recyclable	recyclable	recyclable	recyclable	recyclable		
	Oriented PE	recyclable				recyclable		
	PE mixed with PP	limited recyclability	limited recyclability	limited recyclability	limited recyclability (<5%)	limited recyclability	limited recyclability	recyclable
	Other material	not recyclable				not recyclable	not recyclable	not recyclable
	PS, PVC, PLA, PET, PETG	not recyclable	not recyclable	not recyclable	not recyclable	not recyclable	not recyclable	not recyclable
Barrier	EVOH	Recyclable < 5% in	limited recyclability <	Recyclable	limited	recyclable <	limited	recyclable <2%,
	Metallisation Al	limited recyclability (must not be on the external layer)	limited recyclability (must not be on the external layer)	Recyclable	not recyclable	recyclable < 5%	limited recyclability	recyclable
	AlOx, SiOx	limited recyclability	Recyclable (without topcoats or precoat)	Recyclable	Recyclable	recyclable < 5%	recyclable	recyclable
	carbon coating using		Recyclable				recyclable	
	PA	Not recyclable	Not recyclable	Considered recyclable with	limited recyclability (2)	recyclable < 5%	Not recyclable	Not recyclable
	Aluminium lamination	Not recyclable	Not recyclable		Not recyclable	Not	Not recyclable	Not recyclable
	Other material	Not recyclable		Not recyclable	Not recyclable	Not	Not recyclable	Not recyclable
	PVOH	limited recyclability		Not recyclable	Not recyclable	recyclable <	Not recyclable	recyclable
	PVC	Not recyclable	Not recyclable	Not recyclable	Not recyclable	Not	Not recyclable	Not recyclable
	PVDC	Not recyclable	Not recyclable	Not recyclable	Not recyclable	Not	Not recyclable	Not recyclable

(1): nylon 6 or co-polyamide 6-66 in coextruded PE/PA films without EVOH, combined with MAH-grafted PE as an adhesive promoter at a ratio of at least 0.5 g of adhesive per 1 g of PA

(2): ≤15% PA 6/66 copolymer with melting temperature < 192 ºC and incorporating ≥ 10% PE-g-MAH tie layers

Table 1. Summary of European guidelines on flexible packaging concerning base and barrier material[2,3,4,5,6,7,8].

[2] According to COTREP recommendations (https://www.cotrep.fr/etapes/films-souples-complexes/fs-pebd/)

[3] According to FH CAMPUS WIEN, **CIRCULAR PACKAGING DESIGN GUIDELINE,** DESIGN RECOMMENDATIONS FOR RECYCLABLE PACKAGING; Version 04, October 2021; APPLIED LIFE SCIENCES

[4] According to Zntrale Stelle VERPACKUNGSREGISTER, Minimum standard for determining the recyclability of packaging subject to system participation pursuant to section 21 (3) VerpackG

[5] According to Recyclass recomendation, (https://recyclass.eu/recyclability/design-for-recycling-guidelines/)

[6] According to CEFLEX recomendation, (https://guidelines.ceflex.eu/guidelines/)

[7] According to SUEZ CIRCPACK, Design for recycling guidelines 23.04.2019

[8] According to the network for circular plastic packaging, Reuse and recycling of plastic packaging for private consumers

This summary helps CIMPA partners to understand which points in the design of a film are already confirmed as being recyclable or not recyclable and on which material there is no consensus in the market.

First of all, it has been identified that these guidelines are based on the polyethylene stream, which makes sense as it is the main component of the flexible packaging film. Although some of these guidelines also consider polypropylene, the guidelines are similar to those of PE. This is mostly because the collection, sorting and recycling produce a mixture of PO (PE & PP) in the country.

When it comes to the points of agreement of all these guidelines, it appears that a base material consisting of any kind of PE and in different mixes (HDPE + LDPE) is accepted if there is at least 90% of polyethylene (or polypropylene if the stream exists). All the other materials consisting generally of PET or paper are recognized as not recyclable.

For the barrier layers, there are also common results. Indeed, limited compatibility for the metallization for example as long as the packaging is not natural, if the metallization does not represent a too large amount of the structure or if it is not placed on the external part of the packaging (skin layer) for sorting issues. The main issue for metallization is the color of the final product and, if on the external layer, can also add sorting issues. Other kinds of surface treatment as aluminum oxide, silicone oxide or carbon oxide deposition are accepted if they are not used with coatings used for protection or adhesion promotion (topcoat or precoat). Other agreements are found over PVC, PVDC, aluminum foil or other kinds of material that are considered as not recyclable. About EVOH, it appears that all the guidelines indicate this material as recyclable as long as it represents less than 5% of the total structure. However it is to be noted that some also consider the EVOH content not in the film itself but in the total stream to allow some structures with > 5% EVOH and avoid a switch with less EVOH and additional other resins.

When it comes to the points of disagreement in these guidelines, they are limited to three interesting points:

- PE/PP mixability: Respecting the base material, it looks that a consensus was not found on the mixability of the polypropylene with polyethylene. This disagreement surely comes from the variety of grades and molecular architectures existing in the market of these two materials and final applications and requirements in different countries;
- PVOH barrier material: Some recommendations do not consider it as recyclable and other do, but this surely comes from the new applications in which this material is used. Indeed, it is more and more used as a coating in different mixtures to give barrier properties to new designs. This material is also often soluble in water and may interfere with the recycling or sorting processes that use water (cleaning, density sorting, cooling). Deeper studies should be led to determine the recyclability of these new coatings;
- PA barrier layer: Polyamide barrier layers is surely the most discussed point when it comes to recycling of the flexible films. Depending on the kind of polyamide and polyethylene used, the recycling results (in terms of quality

and reusability) can change a lot, and this makes the choice difficult. This family of barriers needs to be more studied and specified. Moreover, the polyamide is hardly recognized in sorting processes (preventing it from being sorted out of the mixed stream) and this material is constantly improved by the suppliers making this stream very heterogeneous in Europe and in the different applications where it is used.

To conclude on existing guidelines, they all recommend **to change from multimaterial to mono material structures**. This change in design would definitively help to recycle most of the common structures. Nevertheless, many of the food packaging or agricultural films need oxygen barriers to protect its content against oxidation or avoid fumigation gases to escape. This need of oxygen barrier cannot be achieved with the same polymer as the base one (polyolefins) and some specific applications also need high thermal properties (pasteurization, high production rates) that cannot be achieved using mono material structures. In this context, flexible packaging and agricultural film designers need to have recyclable resins to achieve these high demanding properties. As regard of oxygen barrier, EVOH seems to be the best option as it is recognized as recyclable under 5% rate of the structure. Mixtures of PP and PE could, if proved to be recyclable, achieve the high temperature resistance properties. The study of the real impact of PA could also be important as it has interesting properties as oxygen barrier and mechanical performance, as well as PET-PE structures recycling could allow to keep this very efficient structure for high demanding applications.

CIMPA results on recycling

For this purpose, the CIMPA partners have studied 4 streams of flexible multimaterial polymers consisting of:

- PE/EVOH/PE stream
- PE/PA streams
- PE/PET stream
- Metallized or metal laminated polyolefinic based structures.

According to the aforementioned definition of recycling stated by the EU commission, a recyclable polymeric film should be sortable, recyclable and be able to be used in the same application as the first product, all of the above must be demonstrated at scale.

Sorting of the film:

In CIMPA, partners worked on the sorting ability of the streams mentioned before.

At lab scale, for instance, each one of these materials could be detected using thermal analysis or spectrometry. When it comes to industrial scale sorting, the most common technology is NIR spectrometry as it handles high industrial throughputs. Even though this technology is very efficient, it cannot differentiate all multilayered structures. Within the scope of CIMPA, the company PELLENC ST has been able to develop its NIR technology, using both virgin and post-consumer material. Therefore, thanks to CIMPA's study, they succeeded in separating PE/PA (all kinds of PA are mixed) and PE/PET films from the actual post-consumer waste stream.

CIMPA sorting trials also demonstrated that the combined deployment of PELLENC ST NIR technology and FILIGRADE's CurvCode digital watermark enables effective sorting of complex multi-layer materials. Digital watermarking also makes it possible to sort on criteria not-related to material-structure like 'food contact versus non-food contact'.

These trials were performed at industrial scale and allowed CIMPA partners to define PE/PA and PE/PET as sortable. Moreover, another learning is that all the metallized or metal laminated layers can also be sorted using induction separation even though they will be sorted with other metallic contaminants such as aluminum cans (additional sorting step or technologies needed).

The last structure consisting of PE and EVOH cannot be separated yet. This is not an issue today as EVOH is recognized as recyclable with PE under 5% use rate (usual rate in present structures). Even if it is not an issue, EVOH could be sortable using new technologies. Indeed, the CurvCode technology from FILIGRADE studied in CIMPA has proven with the sorting demonstrator that PE/EVOH used in food packaging applications could be sorted out from the stream and used in a specific recycling path like the one designed in the project.

Recycling technology:

This first version of the design for recycling report is only based on the recycling technologies applied to virgin material. The real packaging wastes are actually being studied, however, the results are not available yet.

PE/EVOH, PE/PA and PE/PET streams are studied in mechanical recycling path consisting of the steps below:

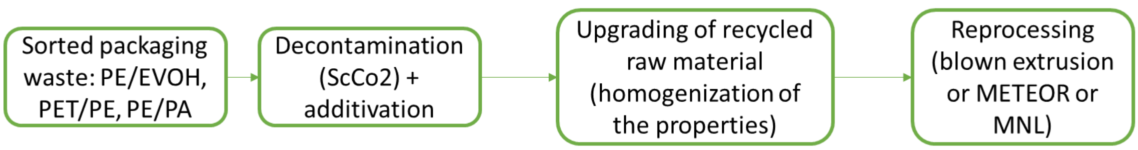
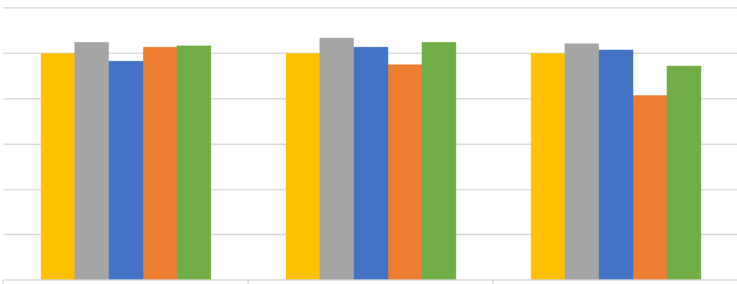


Figure 1. Flow chart of the packaging waste streams in mechanical recycling.

When it comes to metallized PO, the chosen technology is dissolution, considered as physical recycling because metallized coloration cannot be removed with mechanical recycling. Furthermore, dissolution is able to separate the PO from the other polymers present in the structures. Some trials were also run to establish the effect of polyamide, EVOH and PET.

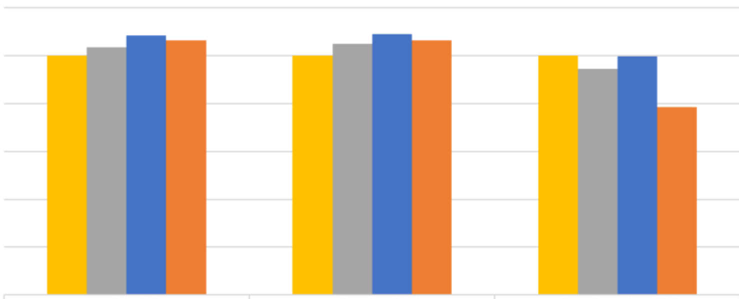
Mechanical recycling

Mechanical recycling was first studied over virgin material in the upgrading step at VTT with their VAREX compounding process followed by lab scale cast film extrusion. The used material did not go through washing, densification nor flotation which are also important steps that will be studied further in the project (washing and densification only). The process used here was based on blending different granulates of the studied material: EVOH 32%mol of ethylene, PA (6/66) and PET with different rates of LDPE and possibly compatibilizers. The used EVOH is a 32% ethylene commercial grade while the PA is a copolyamide 6-6;6 also a commercial grade. The utilized commercial PET copolymer grade was formulated for sheet/film extrusion. The films obtained by cast extrusion were then biaxially stretched 3.5x3.5 times. The mechanical properties of the obtained films were finally analyzed.



	Modulus (%)	Strain at break (%)	Elongation at break (%)
Pure PE	100	100	100
3.1% EVOH	105	107	104
5% EVOH	97	103	102
6.9% EVOH	103	95	81
7.5% EVOH	103	105	94

Figure 2. Evolution of mechanical properties in function of EVOH content, normalized to PE film



	Modulus (%)	Strain at break (%)	Elongation at break (%)
Pure PE	100	100	100
7.5 % PA	103	105	94
15 % PA	108	109	99
22.5 % PA	106	106	78

Figure 3. Evolution of mechanical properties in function of PA content normalized to PE film

These trials demonstrated that adding secondary materials to virgin PE does not influence the mechanical properties a lot even if we can see a little decrease in blends containing 5% and more EVOH and a slight decrease in elongation at break for all the samples containing PA. These first results are in line with actual European guidelines knowing that PA is impacting the film's elongation at break, even at low concentration (even if upgrading the other properties) while EVOH does not impact the properties under 5% rate.

All these studies were performed using actual compatibilizer agents (LLDPE grafted maleic anhydride). The PE/PET stream has also been studied using compatibilizer being SEBS-g-MAH.

The PET/PE recycling with compatibilizer also showed good mechanical properties even if it was about 95% of the one of PE/PA films. However this blend also showed a very white appearance, surely because of the PET phase and the number of defects was also higher than the ones present in PE/PA blends (on some samples twice more).

To conclude on this recycling part, the results from CIMPA looks aligned with actual guidelines knowing **that EVOH is indeed miscible in PE under 5% rate of the formulation**, PA can impact some properties but needs deepen study. The only point that is astonishing, when compared to state-of-the-art studies, is that PET does not seem to impact PE recycling quality. These results come from the fact that the trials were led with virgin material. We think that a mixture of different PET and PE, added to the degradation due to the life of the packaging should really change these results. It is also to be noted that the use of compatibilizers in the blends was beneficial to reach these properties.

The conclusions on these results must be considered carefully because:

- Only one extrusion step has been done here without considering the other recycling steps (washing, floatation, drying, filtration Etc.). Moreover, only basic mechanical analysis on films has been studied (modulus, strain at break and elongation at break) whereas many more parameters should still be considered: floatation, humidity uptake, filtration behavior (clogging, more losses), sealing ability.
- Only virgin material with known and constant mixture of material was used with only one kind of PA6/66 or EVOH 32%.
- Studied process was about blending virgin pellets and not recycling actual films.

Physical recycling:

The first studies on physical recycling were performed on post industrial waste, meaning that the source of the wastes containing metallized PO (BOPPmet/PP and PETmet/PE) come from the partners (Leygatech, Barbier Group and Eversia). Both the pure streams as mixes of these have been studied at lab scale on the installation TNO Möbius 1.0 at TNO. This stream may of course also contain PET or other materials as the goal here is to dissolve and recover only the PO fraction (mix of PE and PP).

The results obtained were very satisfactory as the yield of recovery of 84% and 85 % with respect to PP and PE fraction respectively were achieved. Moreover, no Aluminum nor PET could be found in the recovered PO material which indicates a good purity even though recompounding induced a bit of discoloration of the material.

Other materials have also been studied in physical recycling to see what could happen if the stream with metallized foils contains EVOH or PA. **It appears that even at low level of EVOH (5-10%), PE and EVOH cannot be separated through dissolution while for the PA the threshold is more around 15% and also mainly affecting the yield while the PO product is relatively pure PO.**

Reprocessing:

The mechanically recycled streams have only been reprocessed at lab scale for now, using virgin material over innovative technologies that are METEOR (extensional flow mixing coupled with cast extrusion) and multi nano layering also coupled with cast extrusion. In the second period (that has begun), partners are studying the real packaging wastes of these streams, after upgrading, over these innovative technologies, at pilot scale but they will also use these streams with a real industrial process that are blow film extrusion at industrial partner's plant.

PE/PA

As mentioned before PE/PA (6/66) was studied using METEOR and MNL technologies and with virgin material consisting of the same grades than the one used in upgrading process.

With METEOR, the results that we faced were similar to the ones after upgrading. Indeed, PE/PA blends have their elongation at break in machine direction decreasing while increasing the PA rate, with or without METEOR even if it allows to have better results (see figure below). When talking about the elongation at break in transverse direction this property decreases until 15% of PA where it improved by a factor of 20 before a slight decrease at 22.5% of PA.

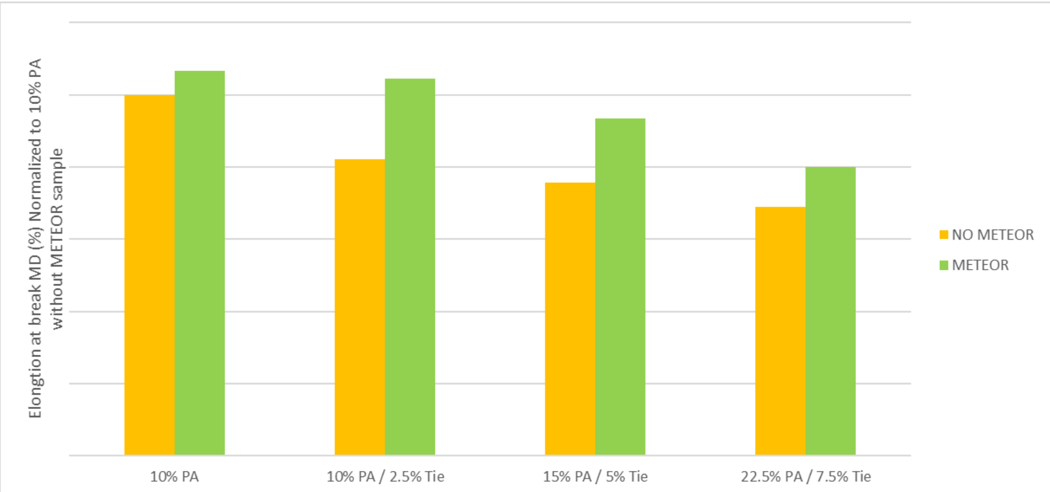


Figure 4. Elongation at break of PE/PA blends in machine direction, with and without METEOR (normalized to PE/PA blend at 10% without METEOR).

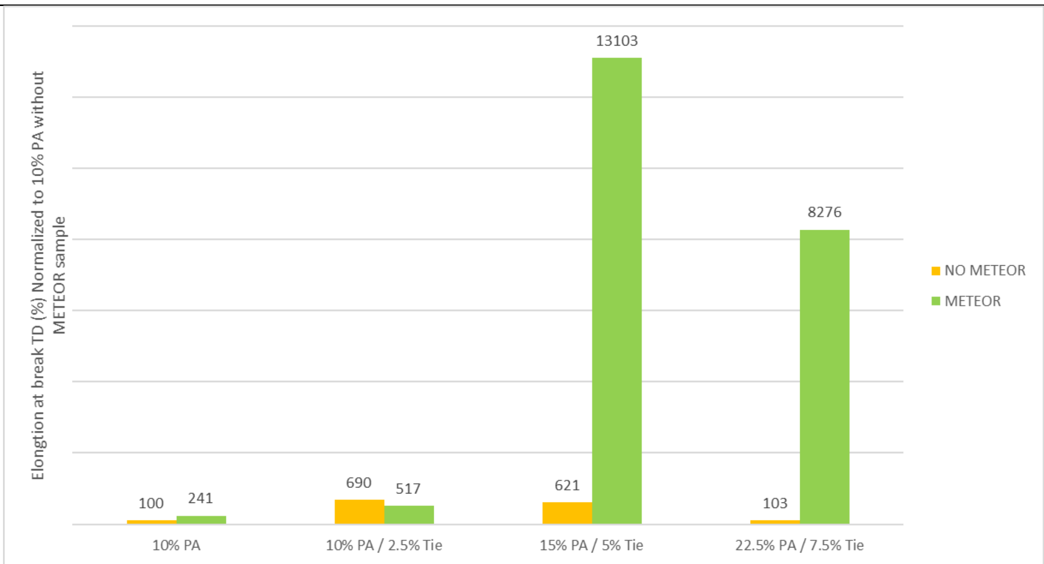


Figure 5. Elongation at break of PE/PA blends in transverse direction, with and without METEOR (normalized to PE/PA blend at 10% without METEOR).

PE/PA has also been reprocessed using MNL technology where it performed in a totally different way. Indeed, here all of the mechanical properties decreased with increasing PA content. Nevertheless, thanks to this technology we obtained good barrier properties, in the range of what is used in the market today.

This difference in properties according to the material is well understandable as in MNL PA layers are very thin (decreased mechanical properties) but continuous (improved oxygen properties). It is almost sure that trials with real packaging waste from PE/PA will give different results as the PA will not be in a separate matrix but it is interesting to see here that, in a blend, PA 6/66 still allows PE recycling thanks to METEOR process and the next study with MNL and real wastes should also be interesting.

PE/EVOH

Cast and METEOR experimentations with PE/EVOH (32%mol of ethylene) blends showed similar results as the aforementioned ones knowing that the mechanical properties of the films are stable until we reach a rate of EVOH of 5% where it begins to decrease. Still, it is interesting to notice that we can clearly see an influence of tie resin that clearly helps PE/EVOH blend to get compatible.

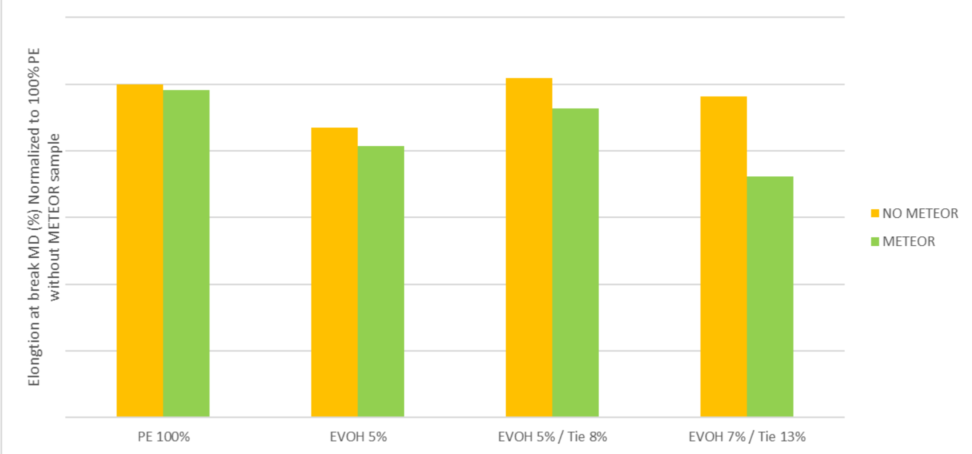


Figure 6. Elongation at break of PE/EVOH blends in machine direction, with and without METEOR (normalized to 100% PE without METEOR).

When it comes to MNL technology the results are similar to the ones achieved by PE/PA. Indeed, the reduction in mechanical properties becomes more pronounced when the number of layers increase.

PE/PET:

Finally, PE/PET blends went through the same process as PE/EVOH and PE/PA. In METEOR, first, the results showed an important decrease in elongation at break when not using compatibilizer, of about 70% for 10% PET in the blend. When using some coupling agent, the results were more stable (while still decreasing) with a loss of about 15% of the elongation.

A multinanolayering process has also been used for PET and PE but for now it has not been successful and other trials should follow.

Guidelines

No reprocessing was conducted yet over the material coming from physical recycling. The results of this study will be taken into account in the update of this report at the end of the project.

The studies previously described allowed us to demonstrate, first of all, that PET/PE blends, even if it showed astonishing results after upgrading, are for now the most difficult ones to recycle and will surely be even more a challenge at industrial scale and with real waste. The preferred path for this stream will surely be the physical recycling. This recycled material will not be used for the two demonstrators as considered with poor processing properties.

Moreover, partners have also seen that PO can be recovered from metallized PO structures, that are already very difficult to replace with the same barrier properties, via physical recycling.

For the PE/PA, it seems that the stream is difficult to handle with mechanical recycling but some PE/PA could give good properties (puncture -> TD resistance) and could, according to the results of industrial scale trials with real wastes, follow a path of upcycling using innovative technologies studied in CIMPA. The next challenge would be to be able to sort recyclable PE/PA (copolymers 6/66) from non-recyclable PE/PA.

Finally, the PE/EVOH stream is actually not sortable using NIR technologies but might be so if using new tracing technologies such as CurvCodes. When looking at its recyclability with PE it is assessed as long as EVOH rate is under 5%, which is the common use rate of actual designs.

Based on these studies, the guidelines for design that address this report are the following ones:

1. As long as it is possible, a flexible packaging or agricultural film should always be designed as a mono-material.
2. If barrier and/or particular thermal properties are necessary it should be addressed by one or more EVOH layer(s) that consist in less than 5% weight rate of the final structure as long as there is at least 1.5 times the quantity of EVOH in the structure that is a compatibilizer or tie layer.
3. If the mechanical properties cannot be achieved using the aforementioned solution (2) it is recommended to use one or more layer of PA 6/66 that consist in less than 15% (to be confirmed with updated report at the end of the project) of the final structure as long as there is at least 1.5 times the quantity of PA in the structure that is a compatibilizer or tie layer.
4. If barrier properties (water vapor barrier particularly) cannot be achieved then the structure should be realized using a complex of metallized polyolefins that do not contain the metal layer on the outer side of the packaging.

Moreover, as regard of sustainability, the project should also consider guidelines about the use of recycled material:

1. For common applications, when no particular properties are needed, PE or PE containing less than 5% EVOH recycled material should be used.
2. If high mechanical properties are needed, PE containing less than 15% PA (to be confirmed with updated report at the end of the project) recycled material should be used.
3. For applications where high purity recycled material is needed, a PO recycled grade from physical recycling should be used.
4. No recycled material containing other materials than PE should be used with a third material (for example R-PE/EVOH should not be used in a structure with PA).

New designs

Considering these guidelines, new designs can be established that will be used as use cases in the demonstration phase. The first one will be the demonstrator for food packaging application. This application will need oxygen barrier and should be, as much as possible, a mono material. The necessary properties that need to be achieved are:

- a sufficient differential in thermal resistance between inner and outer layer to enable a good sealing;

- good barrier properties (to oxygen and water vapor);
- good optical properties;
- no major defect that would risk to break the barrier to gas.

And to answer the need of the guidelines the choice is to design a mono PE complex film, like an oriented PE film laminated to a barrier (EVOH) sealant film. If possible, this film should also contain some recycled material. From what it has been established earlier, it could use physically recycled PE in the oriented PE film as it has good purity. It is also possible to use some PE/EVOH recycled material for the laminated layer of the sealant as this one does not need any thermal properties.

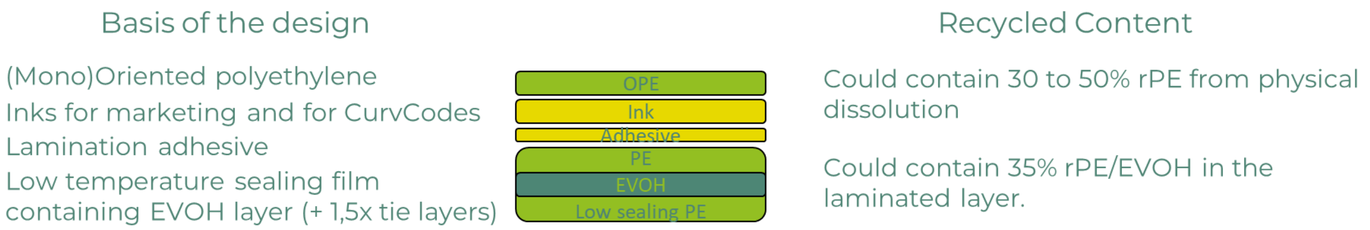


Figure 7. Design of a food packaging structure in agreement with CIMPA's guidelines.

The second design is the one of an agricultural film that should be used for silage application. This film will need to have good mechanical properties regarding the puncture and the tear resistance. It should also be stable for long term application and UV exposure.

The choice here is to have a three-layer PE film (to keep the benefits from mono material) and have PE/PA recycled content in the core layer at least at around 50% that will make a 30% recycled content film (knowing that the core layer represents 60% of the structure).

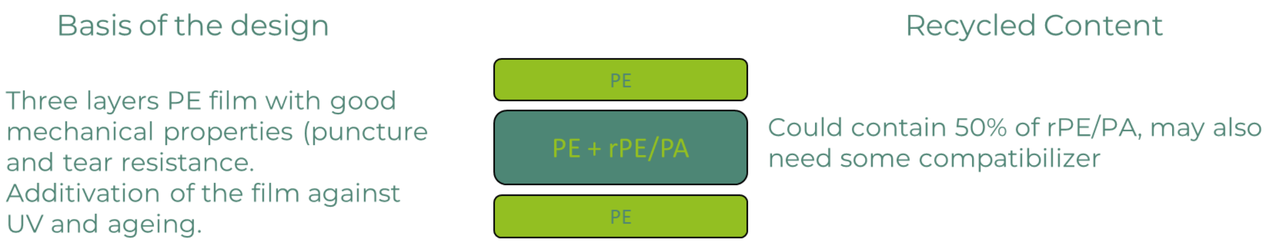


Figure 8. Design of a silage film structure in agreement with CIMPA's guidelines.

These two designs will be demonstrated at pilot and pre-industrial scale within CIMPA project. During the pilot scale demonstration, attention should be given to the upgraded properties. Indeed, for the first example one of the goals of the new technologies used would be to obtain some barrier properties using METEOR with rPE/EVOH material or at least having an adhesion effect to reduce tie layers in the structure (keeping enough of tie layer for recyclability). This could lead to the reduction of EVOH layer thickness or to the replacement of layers 2 and 4 in the PE/EVOH/PE structure (cost savings). For the second design, MNL technology using rPE/PA and virgin PE could lead to a substantial reduction in thickness and maybe also give some barrier effect (useful for some applications).

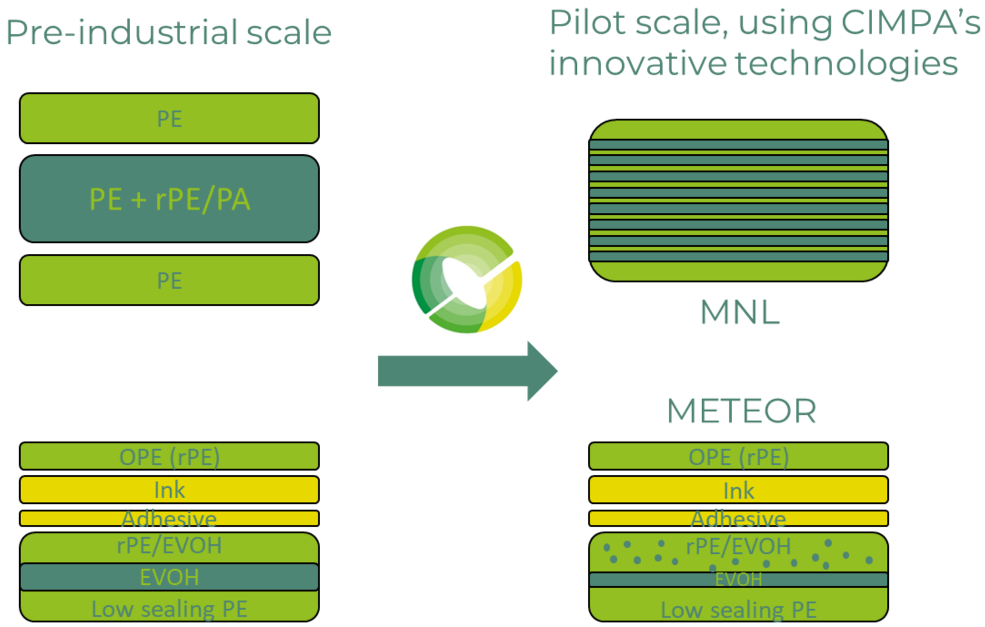


Figure 9. Possible achievement in innovative design with CIMPA technologies.

Conclusion and next steps

Today the best option when it comes to the design of a flexible plastic packaging is, definitively, the mono material structure. Nevertheless, when highly demanding properties are needed the other resins as EVOH or specific PA may be used. To keep the value of this materials, particular recycling paths should be followed and new structures will be created with sustainable secondary raw material.

The first part of the work done in CIMPA allowed the consortium to set two demonstrators that will be studied towards the end of the project. These two demonstrators should prove that recycling well-designed multimaterials with the smartest recycling path can enable a real circularity in agricultural films and food packaging applications.

As a reminder this first version of the study does not consider real packaging waste with real recycling path meaning that the studied material are virgin pellets that are blended directly in the extruder and not a mix of actual packaging waste that would be washed, densified and recycled. Moreover, only one grade of each kind of material has been studied (copolyamide 6/66 for PA, EVOH 32% for EVOH and PET Copolymer, sheet extrusion grade) which is not representative of actual packaging waste streams. The study at pilot and industrial scales will follow in the coming months leading to an updated report with the new results at the end of the project.