



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

D3.5: Publishable report on production of multilayered films from mechanically recycled ML films

WP3: Mechanical recycling

Project Information

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Abbreviations

MNL	MultiNanoLayering
Ca, Ca_c	Capillary number, critical Capillary number
(LD)PE	(Low Density) Polyethylene
PA	Polyamide
PE-g-MAH	Polyethylene grafted Maleic Anhydride
PET	Polyethylene Terephthalate
SEBS-g-MAH	Styrene-Ethylene-Butylene-Styrene grafted Maleic Anhydride
TSE or TS	Twin Screw Extruder
SSE or SS	Single Screw Extruder
L/D	Length/diameter (used to qualify an extrusion screw)
WVTR	Water Vapor Transmission Rate
OTR	Oxygen Transmission Rate
MD or TD	Machine Direction or Transverse Direction
DSC	Differential Scanning Calorimetry
MFI	Melt Flow Index
OIT	Oxidation Induction Time
DFS	Dynamic Frequency Sweep
DTS	Dynamic Time Sweep
SR	Strain Relaxation
PIR	Post-Industrial Recyclate
PCR	Post-Consumer Recyclate

Executive summary

In this deliverable, IPC aims to demonstrate the efficiency of two innovative processes for polymer recycling: the MultiNanolayering (MNL) process its elongational flow mixer called METEOR®.

In the first step, several virgin material blends were studied, according to the sorting families developed in the project: PE/EVOH, PE/PA and PE/PET. From the METEOR® study, all virgin materials blends revealed particularly promising mechanical results (see Figure 13 of this deliverable).

Indeed, the properties reach the level of a twin-screw extruder, and thus using single-screw extruder coupled with METEOR® process. The rheological analysis indicated improved interfacial energies for PE+PA and PE+EVOH blends: either the dispersion is thinner, or some interactions are created at the interface. Thus, METEOR® might be a good way to reduce the compatibilizer content for such blends.

With the MNL process, the structure PE/PA, an extensive study was conducted, comparing different numbers and positions of LMEs within the MNL modules (see Figure 21 and Figure 26).

From the first graph, it can be concluded that the MNL structure was efficient up to 129 layers. However, with a higher number of layers (ranging from 513 to 1025 layers), both mechanical properties, such as tensile modulus, and the Oxygen Transmission Rate (OTR) showed degradation. The second figure examined the influence of LME positioning. It was assumed that the relation with the viscosity ratio was significant. This structure likely requires layers to be multiplied at the beginning of the process (as in configuration 1) because the viscosity ratio between PE and PA is close to 1 at this stage.

When the recycled materials are processed (Post-Industrial Recyclates (PIR) and Post-Consumer Recyclates (PCR)), with MNL or METEOR® processes, a decline in mechanical properties was observed, as well as a degradation.

For all the blends, virgin and recycled ones, the Water Vapor Transmission Rate were satisfying, but the Oxygen Transmission Rate were higher than the industrial target. Unfortunately, the attempt of stretching of METEOR® films at VTT did not improve these properties. Although a deeper study of the stretching parameter would be highly beneficial, it was not within the scope of the CIMPA project.

Deliverable report

1. Introduction

This deliverable aims to present the work done on the METEOR® and MNL (Multi NanoLayering) processes at IPC as mechanical recycling methods for multilayer films. METEOR® is a unique process developed and patented by IPC (FR3054159A1), that utilizes elongational mixing, instead of the conventional shear mixing found in standard extruders. MNL is a process available at Nordson, which aims to multiplying the layer of a structure (up to more than 4000 layers), while keeping the thickness of the film unchanged. The layers are thus of nanometric size, giving interesting properties, especially with respect to oxygen or water barrier performance.

An extensive review of the state of the art is included in this deliverable to explain why elongational mixing and MNL are studied in the CIMPA project. In the first stage, the trials made conducted using virgin materials are presented. Three types of blends were tested, PE+EVOH, PE+PA and PE+PET, following the classification by families established within CIMPA (see Table 1). This table examines “simple multilayers” through the lens of mechanical recycling.

	Models	Multilayers	Usage (sector)	Partner
Simple multilayers	PA/PE	PA/PE	Vaccum packaging (food)	LEY
		PE/PA/PE	Barrier fumigation film (agri)	BAR
	PE-EVOH-PE	PE/EVOH/PE	Barrier fumigation film/ cover silage film (agri)	BAR
		PE/EVOH/PE	Flowpack line (coextrusion film) (food)	LEY
	PET/PE	PET/LDPE	Snacks (food)	EVE
PET / Alu / PE		Coffee pouch packaging (food)	EVE	
Complex multilayers	BOPPmet-BOPP	BOPPmet-BOPP	Chips (food)	EVE

Table 1: Proposed classification by CIMPA. Extracted from the Grant Agreement, part B, Figure 4

A focus was made on PE+EVOH blends processed through METEOR® and PE/PA structures processed through MNL. To characterize such blends, mechanical, rheological and barrier properties were investigated.

In the last stage, real wastes (post-industrial and post-consumer) were processed. These wastes came from the partners (Paprec for post-consumer waste, and Leygatech, Barbier and Eversia for post-industrial wastes). After a recompounding step at VTT, using the VAREX technology (see deliverable 5.1), the blends arrived additivated and in a pellet form at IPC. A final dilution was made at IPC, directly during the METEOR® or MNL processes, adding PE and barrier material, to meet the target of recycled content in packaging.

2. The METEOR® Concept

To gain a comprehensive understanding of the METEOR® mixing technology from both operative and conceptual perspectives, it is highly recommended to review some fundamental concepts of polymer mixing. Key concepts such as capillary number (Ca), and especially mixing efficiency within the context of shear and/or elongational flow fields should be revisited.

a. Capillary number

The mixing process, in big lines, rests on the competence of two opposing forces: viscous vs interfacial or cohesive forces. On the one hand, the viscous forces tend to deform the dispersed domains and, ultimately, lead those to break-up conditions and, on the other hand, the interfacial or cohesive forces tend to keep the domain, i.e., droplet, at its original size and shape in order to minimize the interfacial energies. Based on these opposing phenomena, a dimensionless indicator called *capillary number* (Ca) is able to describe quantitatively the state of a determined mixing process in terms of dispersive and distributive character and, overall, mixing quality or efficiency. Accordingly, Ca is mathematically represented as:

$$C_a = \tau R / \sigma = \eta_m \dot{\gamma} R / \sigma \quad (1)$$

Where,

τ = Shear stress (Pa); η_m = Viscosity of the major phase (Pa.s); $\dot{\gamma}$ = Shear rate (s^{-1})

R = droplet radius (m)

σ = Surface tension (N/m)

At the earlier stages of a mixing operation, since the system is rather constituted of large components' domains, deformation readily occurs because of the little opposition of interfacial forces, which are largely surpassed by the viscous ones (shear or elongational stress) at this point. One can thus say that this initial mixing regime is more characterized by a distributive mixing mechanism, where the interfacial area increases under the deformation process. Progressively, Ca continues to increase by virtue of the increase of the stress trying to overcome a more opposing interfacial forces as long as the dispersed domain reaches higher deformation (Figure 1a). At a critical balance in the competition between the stress and the interfacial energy, dispersion mechanisms start to dominate based on the appearing of interfacial instabilities. From these, mainly Rayleigh-type disturbances¹ finally lead to the drop break-up phenomenon (Figure 1b). At this point, Ca is rather known as critical Ca (Ca_c).

¹ Ica Manas-Zloczower, E. (2009). *Mixing and compounding of polymers*. Vienne: Hanser.

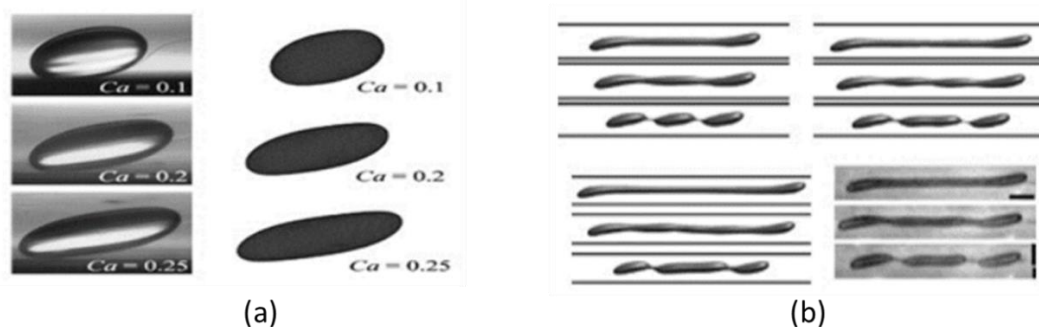


Figure 1 a) Evolution of Ca as deformation increases and, b) deformation and break-up phenomenon in the vicinities of Ca_c^3

Ca_c depends on the viscosity ratio (λ) of the system, which is defined as:

$$\lambda = \eta_d / \eta_m \quad (2)$$

Where, η_d = Viscosity of the dispersed phase; η_m = Viscosity of the major phase

Grace², depicted the referred dependency, in his well-known work on the matter, in the framework of two major types of flow fields, simple shear and extensional one. The next Figure presents an adaptation of this central contribution:

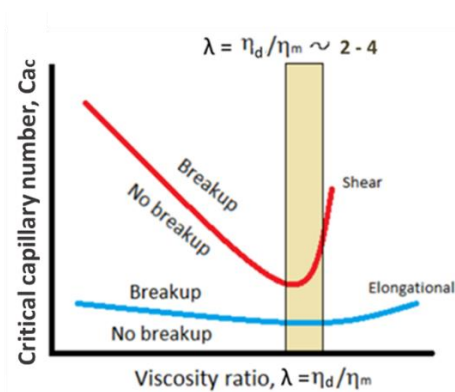


Figure 2: Critical capillary number, Ca_c as function of the viscosity ratio for two flow field regimes during the mixing of two immiscible fluids.

From Figure 2, several fundamental facts can be highlighted:

1. In the practical range of λ , Ca_c values for simple shear are always higher than those for elongational flow. The difference is noteworthy at low λ .
2. For both flow regimes, Ca_c decreases in a quasi-linear fashion as function of λ until around $\lambda=2$, where simple shear fails to generate dispersion any

² Grace, H. P. (1982). Dispersion phenomena in high viscosity immiscible fluid systems and application of static mixers as dispersion devices in such systems. Chem. Eng. Commun., 14, 225-277.

longer (asymptotic trend). On the other hand, elongational flow is able to promote dispersive phenomena after that point.

3. Around $\lambda=1$, both simple shear and elongation present the minimum Cac .

It is important to point out that Grace's work and, thus, the precedent statements are circumscribed to *dispersive mixing* since Cac characterizes only dispersion phenomena. This is in fact the central scenario when it comes to blends of immiscible polymers because it is related, from the practical point of view to the concept of mixing efficiency, primarily characterized by the dispersion quality. Particularly, the facts about Grace results point to the higher efficiency of elongational flow fields to sustain dispersive mixing in comparison to shear flow, especially when $\lambda > 2$. Indeed, a basic explanation for this within a spatial context is that, whereas shear involves a significant rotational component, in the z direction, which does not contribute to strain, elongation acts in only one axial direction, contributing entirely to strain and dispersive effect.

b. Batch mixers and/or attachments units with predominantly elongational flow fields

The difficulty to generate striking elongational flow effects in traditional mixer systems lies in:

- 1) The prohibitive axial dimension needed to generate enough dispersed droplet elongation leading to break-up.
- 2) The limited residence time or number of passages of the material through points of relatively intense elongational stress.

The descriptions of several developments in batch elongational mixers can be found in literature in the last decades (Figure 3)³.

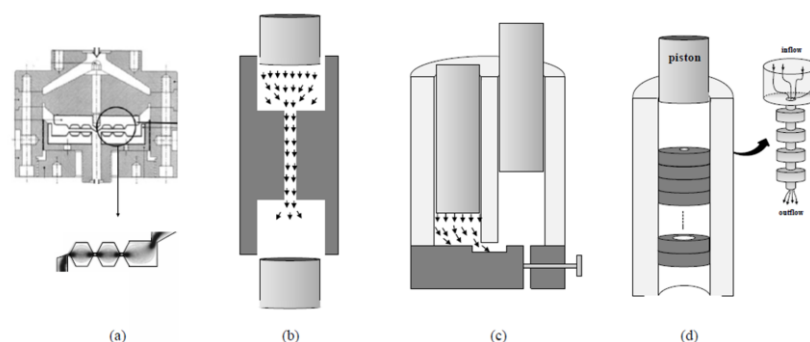


Figure 3: Schematics of extensional mixers: (a) Extensional Flow Mixer⁴; (b) RMX@mixer⁵; (c) Son's internal mixer⁶; (d) Ferrás et al⁷

³ L.L. Ferrás, C. F. (2021). Dispersion of graphite nanoplatelets in polypropylene by melt mixing: the effect of hydrodynamic stresses and residence time. *Polymers*, 13, 102-116.

⁴ Bourry, D., Godbille, F., Khayat, R., Luciani, A., Picot, J., & Utracki. (1999). Extensional Flow of Polymeric Dispersions. *Polym. Eng. Sci*, 9, 1072-1086.

⁵ J. Rondin, M. B. (2014). Dispersive mixing efficiency of an elongational flow mixer. *Polym. Eng. Sci.*, 54, 1444-1457.

⁶ Son, Y. (2009). Development of a novel microcompounder for polymer blends and nanocomposite. *J. Appl. Polym. Sci.*, 112, 609-619.

In the devices of Figure 3, there is a common approach to generate elongational flow and compensate for the lack of dimensions: the application of contraction/expansion or converging/diverting flow to maximize accelerated flows in reduced spaces. On the other hand, in order to increase the contact of the material with the points of elongational stresses, a and d , in Figure 3, uses multiple contraction/expansion elements which number is limited by the overall size of the device. In the case of b and c , RMX® and Sons', respectively, the multiple passage is obtained by a back-and-forth movement of pistons, forcing the mass to go through the contraction/expansion zone repeatedly. The obvious advantage of the latter systems in comparison to fixed elongational elements is the control of the number of cycles or passages, which can be as high as desired. Nevertheless, when envisaging the implementation of these principles to continuous mixing operations the back-and-forth approach presents more significant technical difficulties and the multiple fixed elements option becomes, by far, more compatible, as it will see in further sections.

c. Continuous elongational flow mixer METEOR®

IPC METEOR® design was inspired by the high dispersive mixing efficiency originally found in the elongational batch-type mixer RMX® (Figure 3b) developed by Muller et al⁹. As briefly described in the referred Figure, the RMX® is based on a simple geometric approach involving abrupt flow contractions and forced passage of the mixture through the convergent/divergent zones by means of pistons exerting a back-and-forth displacement. In this way, the mixing sequence efficiency is mainly characterized by the linear speed of the piston (directly related to shear and elongational rates) and the number of back-and-forth passages (cycles) across the converging/diverging zone. Accordingly, the RMX® is able to generate quite fine morphologies in polymer blends even at mild mixing conditions i.e., low number of cycles and low pistons' speed. However, as also aforementioned, the batch character of some highly efficient elongational mixers like the RMX® limits their employment at large scale, particularly preventing a potential use in industrial environment where continuous operation is quite the privileged scenario. Based on this premise, METEOR® was designed and developed aiming to provide the industry with a continuous elongational mixer equipment making use of the strong mixing efficiency principles found in the RMX® (Figure 4). For this to happen, a screw with a series of axial convergent/divergent elements were designed, exerting accelerated flow and keeping the shear component as low as possible. In this way, besides privileging the elongational mixing action, it is possible to prevent the excessive viscous heating inherent to shear flow mixings. In shear flow, perpendicular friction effects are implied in the driving force for shear deformation to happen, whereas in elongational flow the entire axial deformation does not involve other stress components.

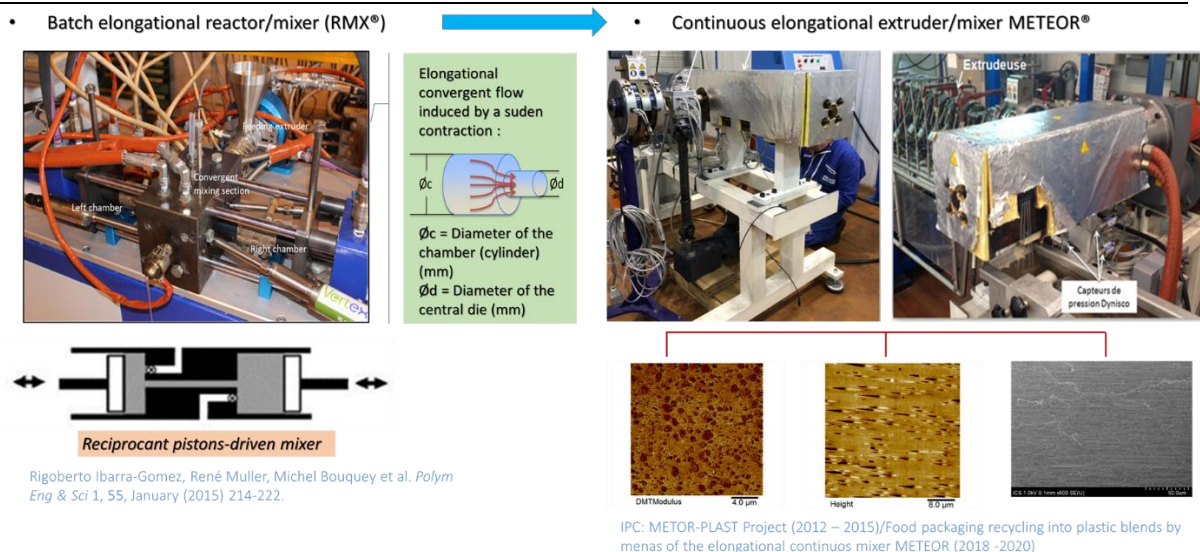


Figure 4: METEOR® design and development following transposing of elongational mixing principles of batch-type mixers.

Regarding the processing set up Meteor®, the mixer was thought to be attached downstream a single-screw extruder or a twin-screw extruder at mild mixing capacity. Figure 5 and Figure 6 presents the processing configuration of the METEOR® employment, particularly for cast film production, objective of the CIMPA project.

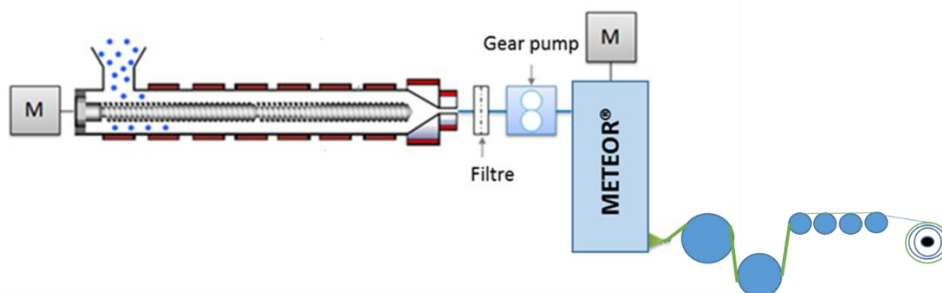


Figure 5: Diagram of the METEOR® processing set up in the CIMPA project.

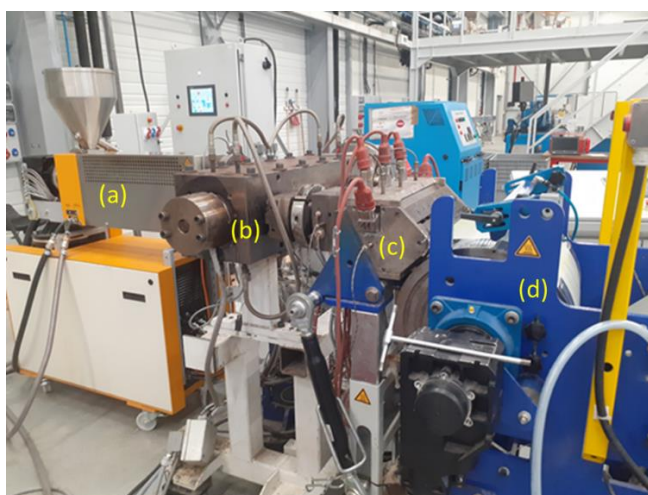


Figure 6: Actual METEOR® processing set up for the CIMPA project: a) single-screw extruder, b) METEOR®, c) cast film die head, d) film cooling and wind-up unit.

d. Role of METEOR® in the development of functional morphologies in polymer blends and recycling

METEOR® is designed to enhance the dispersive mixing of polymer blends, particularly those derived from the recycling sector. This sector is characterized by a wide range of viscosity ratios among plastic wastes due to the inherent heterogeneity of their thermomechanical histories and sources. METEOR®, with its predominance of elongational flow mixing, surpasses the deformation and dispersion limits of shear flows. This capability enables the creation of added value by achieving fine nodular dispersions and fine elongated morphologies. Fine nodular morphology generally contributes to homogeneous physical bulk properties while fine elongated morphologies enhance gas barrier properties, which are highly valued in packaging applications. This is well illustrated by Banerjee et al⁷ in Figure 7. The primary goal of achieving fine dispersions is demonstrated in the case A. Furthermore, the process can create very small dispersed domains and fine elongated structures as shown in Figure 7b.

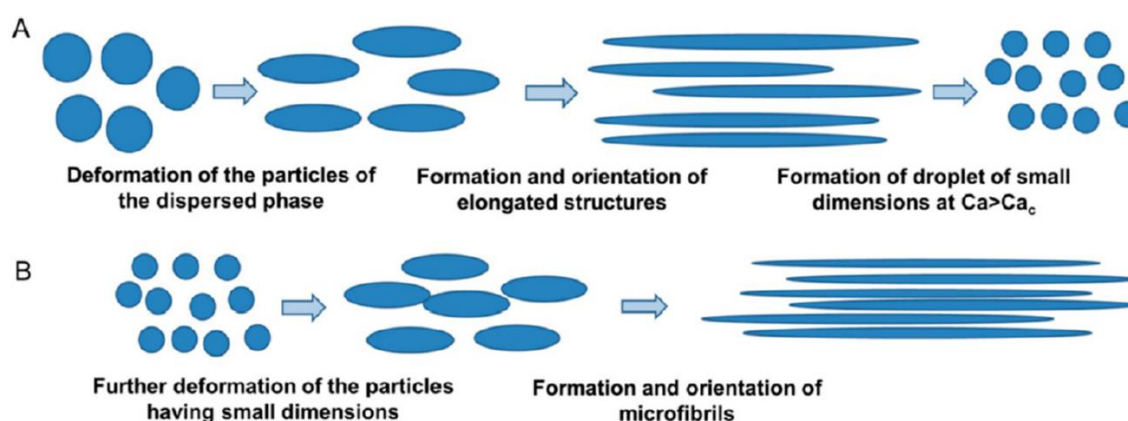


Figure 7: Schematic of the possible mechanism of the droplet-to-fibril transition in immiscible blends subjected to elongational flow: (a) deformation of the original particles of the dispersed phase and (b) formation of microfibrillar structures²⁹

This concludes the state-of-the-art of elongational flow mixing and its interest in polymer recycling. The following paragraphs present the trials made within the CIMPA project.

⁷ R. Banerjee, S. S. (2023). Role of Rheology in Morphology Development and Advanced Processing of Thermoplastic Polymer Materials: A Review. ACS Omega, 8, 27969-28001.

3. PE/EVOH virgin materials blends through METEOR®

a. Materials and Formulations

The materials were chosen in collaboration with WP5 trials, and sent by Leygatch, due to availability, delays and prices.

For the LDPE, Leygatch uses several equivalent grades, with the same density, the same Melt Flow Index (2.0g/10min), and all polymerized using radical polymerization.

The four grades of LDPE used during these trials are: DOW 352E, SABIC NC1013, REPSOL ALCUDIA 2320FG and BASSELL LUPOLEN 2426H.

The EVOH was also supplied from Leygatch: KURARAY EVAL J171B, as well as the compatibilizer, a PE-g-MAH from SK, OREVAC 18300, typically used as a tie-layer in multilayer films.

All the technical data sheets can be found in Annex1.

For the formulations tested (see Table 2), the standard composition of a PE/EVOH packaging film was taken as a basis: 5 wt.% EVOH, 8 wt.% tie layer (which is the same as the compatibilizer, i.e., PE-g-MAH), and 87 wt.% LDPE., corresponding to the formulation n°3.

		Formulation			
		1	2	3	4
PE	wt.%	100	95	87	80
EVOH	wt.%	0	5	5	7
OREVAC	wt.%	0	0	8	13

Table 2: Formulations of PE/EVOH blends

For these formulations, trials were made using a single-screw extruder (SS) Collin E45E (L/D=25), the same single-screw extruder followed by the METEOR® process (SS+METEOR), a twin-screw extruder (TS) Coperion ZSK 32 MC (L/D=40) and the same twin-screw extruder followed by the METEOR® process (TS+METEOR). The flat die used was the Scamex 350, giving films of 350mm width. The extruders and METEOR® temperatures were between 210 and 230°C (viscosity ratio EVOH/PE = 1.4-1.7, ratio of complex viscosities between 6 and 10 rad/s from Dynamic Frequency Sweep tests, see Annex 2), and the throughput was 5kg/h.

b. Mechanical properties

The results of tensile measurements can be found in Table 3. The entire test method can be found in Annex 2.

The measured tensile moduli are all in the same range, whatever the direction (transverse and machine ones), and whatever the set-up (single-screw versus twin-screw extruders, with and without METEOR®). The same goes with the stress

at break property. There was no yield in machine direction for almost all the blends. In the transverse direction, elongation and stress at yield are also in the same range.

		Machine direction					Transversal direction				
		Tensile Modulus (MPa)	Stress at yield (MPa)	Elongation at yield (%)	Stress at break (MPa)	Elongation at break (%)	Tensile Modulus (MPa)	Stress at yield (MPa)	Elongation at yield (%)	Stress at break (MPa)	Elongation at break (%)
PE 100%	SS	311 ± 8	/	/	17.5 ± 1.8	600 ± 30	341 ± 8	9.14 ± 0.12	14 ± 0	14.9 ± 1.3	600 ± 40
	SS+ METEOR	299 ± 22	8.92 ± 0.52	15 ± 4	16.1 ± 4.4	470 ± 190	311 ± 11	8.75 ± 0.25	15 ± 1	15 ± 1.4	600 ± 50
	TS	280 ± 8	/	/	15.9 ± 1.5	430 ± 30	331 ± 8	8.27 ± 0.26	11 ± 1	12.8 ± 0.6	540 ± 20
	TS+ METEOR	329 ± 13	/	/	16.1 ± 1.5	360 ± 60	362 ± 5	8.78 ± 0.15	9.8 ± 0.2	12.7 ± 0.3	530 ± 20
PE/EVOH 95/5	SS	409 ± 16	10.1 ± 0.1	16 ± 1	17.8 ± 0.8	560 ± 20	443 ± 16	10.2 ± 0.3	8.8 ± 0.3	8.53 ± 1.06	11 ± 1
	SS+ METEOR	400 ± 25	9.87 ± 0.5	15 ± 1	18.6 ± 0.6	540 ± 20	333 ± 5	8.54 ± 0.05	12 ± 0	9.97 ± 0.32	430 ± 10
	TS	335 ± 27	/	/	13.4 ± 2.3	390 ± 60	350 ± 11	8.58 ± 0.12	11 ± 0	10.5 ± 1.3	470 ± 50
	TS+ METEOR	381 ± 90	/	/	16.1 ± 2.2	340 ± 90	390 ± 40	9.19 ± 0.21	9.9 ± 0.2	9.4 ± 1.13	390 ± 40
PE/EVOH/Comp 87/5/8	SS	369 ± 10	/	/	15.4 ± 0.6	470 ± 20	411 ± 30	9.62 ± 0.57	11 ± 0	9.19 ± 0.58	170 ± 60
	SS+ METEOR	317 ± 2	/	/	18.2 ± 0.9	480 ± 20	316 ± 9	8.64 ± 0.17	15 ± 1	12 ± 1.1	460 ± 50
	TS	319 ± 9	/	/	15.6 ± 2	440 ± 50	603 ± 32	13.4 ± 0.5	11 ± 0	19.8 ± 1.8	520 ± 40
	TS+ METEOR	292 ± 13	/	/	15.1 ± 1.9	400 ± 30	352 ± 7	8.78 ± 0.1	12 ± 0	13.2 ± 1	500 ± 40
PE/EVOH/Comp 80/7/13	SS	374 ± 19	9.72 ± 0.76	20 ± 2	15.5 ± 1.1	410 ± 20	450 ± 27	9.86 ± 0.49	9.8 ± 0.2	9.79 ± 0.44	230 ± 40
	SS+ METEOR	395 ± 3	/	/	18.1 ± 0.6	450 ± 20	433 ± 5	9.8 ± 0.07	11 ± 0	12 ± 0.2	440 ± 10
	TS	318 ± 22	/	/	16.3 ± 1.3	400 ± 50	417 ± 11	9.01 ± 0.18	10 ± 0	15.1 ± 1	550 ± 30
	TS+ METEOR	315 ± 16	/	/	13.3 ± 1.2	350 ± 30	365 ± 12	8.69 ± 0.28	11 ± 0	12.3 ± 1.4	440 ± 60

Table 3: Mechanical properties of PE/EVOH films

The most interesting property is the elongation at break. In the machine direction, this property seems to be a bit lower when the blends were made using the twin-screw extruder (from around 400 to 600% to 300-400%), but they remain quite satisfying, being in the same range of the pure LDPE (see first 4th row). In the transverse direction, when the single-screw extruder is used without METEOR® (see Figure 8, the solid pink bars), the elongation at break is quite low as soon as EVOH is introduced. This indicates that the dispersion is probably poor. This property increases a bit when compatibilizer (OREVAC) is added, showing its efficiency. When METEOR® is used after the single-screw extruder (solid blue bars), the elongation at break increases a lot, reaching the properties observed with the twin-screw process (TS: dotted pink bars). However, the use of METEOR® process after a twin-screw extruder (TS+METEOR: dotted blue bars) shows no improvement. At this stage, the hypothesis is that the twin-screw extruder dispersed the EVOH into the PE efficiently, and thus, the METEOR® process makes no real added-value, while following a single-screw extruder, the added-value is obvious, given the elongation at break observed.

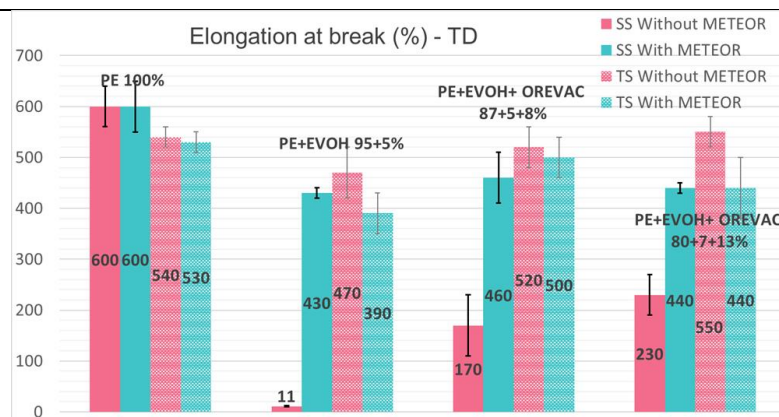


Figure 8: Elongation at break in transverse direction (%) for PE/EVOH films

With this scenario, the property of high interest becomes the study of the degradation: indeed, the elongational mixing advantage is that there is a limited shear, thus less degradation.

c. Degradation study

To measure the degradation, several tests have been done: Melt Flow Index measurements, Oxidation Induction Times, and finally Dynamic Time Sweep (DTS) tests. The methods used can be found in Annex 2. Both MFI and OIT values measured are so low that no conclusions can be made. For DTS, the modulus (G' and G'') crossover times are compared: they are very close to each other, and very comparable to the PE one.

Unfortunately, the main advantage of the use of an elongational mixer like METEOR®, i.e., avoid the degradation occurring with shear during mixing, is not proven. This might be due to the residence time of the METEOR® process, which is very long compared to an extruder (30-40 min with METEOR® versus 15-20 min without). However, the use of METEOR®, even with these high residence times, does not seem to degrade the material.

An update of this process, aiming to reduce the residence time, could give a very nice improvement in the matter of material degradation.

d. Barrier properties

The methods used to measure the Water Vapor Transmission Rate and the Oxygen Transmission Rate can be found in Annex 2, and the results are summarized in Figure 9.

For the water permeability (Figure 9a), all results are satisfying, being below 10g/m².day, as set by the industrial partners. However, the METEOR® process does not seem to improve this property, whether it is used in combination with a single-screw or a twin-screw extruder.

For the oxygen permeability (Figure 9b), the first thing to be explained it that the results with twin-screw extruder (TS and TS+METEOR) exceeded the sensor IPC had at this time (the limit was 500mL/m².day). Given resources needed, and

especially that the results will not be below the target set by the industrial partners (i.e., 3mL/m².day), the measurements were not done with the new sensor able to reach up to 4000mL/m².day. Thus, only the results for the trials made using the single-screw extruder (SS and SS+METEOR) are available.

The second point to be highlighted is that the EVOH does not seem to act like an efficient barrier in these samples: adding EVOH does not improve the OTR. Usually, EVOH is very efficient in a multilayer structure to obtain good oxygen barrier properties. But here, the structure is not a multilayer but a blend, and the EVOH is not present in a layer form but dispersed in a matrix. The aim of METEOR® was to disperse this phase in a fibrillar shape, or even better, form plaques of EVOH that could increase the diffusion path, and thus, decrease OTR. But, given the poor OTR results:

- Either the dispersion is not in the preferred shape, despite the good elongation at break obtained,
- Or the plaques are formed, but they are not big enough to increase the diffusion path of oxygen in the film.

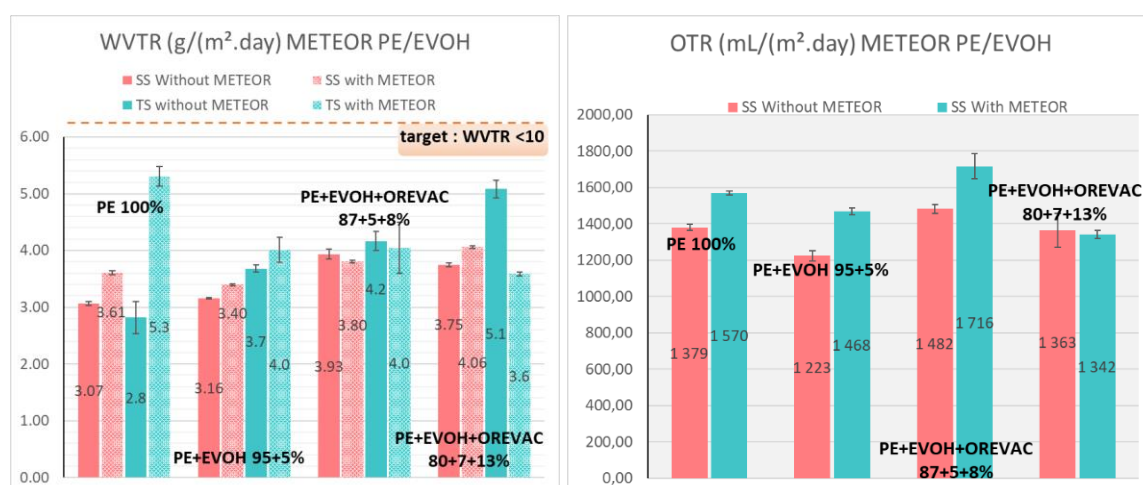


Figure 9: a) Water-vapor transmission rate (g/(m².day)) and b) Oxygen transmission rate (mL/(m².day)) for PE/EVOH films

In order to improve these results, attempts of biaxial stretching of the films were made at VTT, as proposed in the Grant Agreement. The hypothesis is that if the fibril or plaque are stretched even more, then the diffusion path of oxygen could be increased, and thus OTR decreased. The bi-axial stretching method used is available in Annex 3.

For the first test, a full study was conducted: all films made using twin-screw with and without METEOR® (TS and TS+METOR) were stretched, and then observed through SEM at VTT, with an analysis of the average size of EVOH domains and tested with DSC at VTT.

Every film was stretchable at ratio 1.5x1.5 at 145°C reaching films of thickness between 40 and 50µm. More details about the stretching process can be found in Annex 3.

The SEM pictures after bi-axial stretching can be found in Figure 10.

When pictures taken from films processed by twin-screw extruder are compared to ones processed by twin-screw extruder + METEOR®, the dispersion of EVOH does not seem to be in fibrillar or plaques form. But, from the mechanical property results, it was concluded that METEOR® does not show a great effect after a twin-screw extruder, probably because the dispersion after the twin-screw extruder is so good that METEOR® does not improve it more. The effect of OREVAC as compatibilizer is clearly seen, because the films containing it are more homogeneous, and the EVOH domains are smaller.

When the films processed by single-screw extruder are observed by SEM, the average size of particles is better when using METEOR with a high amount of compatibilizer, but worse when no compatibilizer is added. The conclusion are then unclear regarding the improvement of dispersion induced by METEOR after a single-screw extruder.

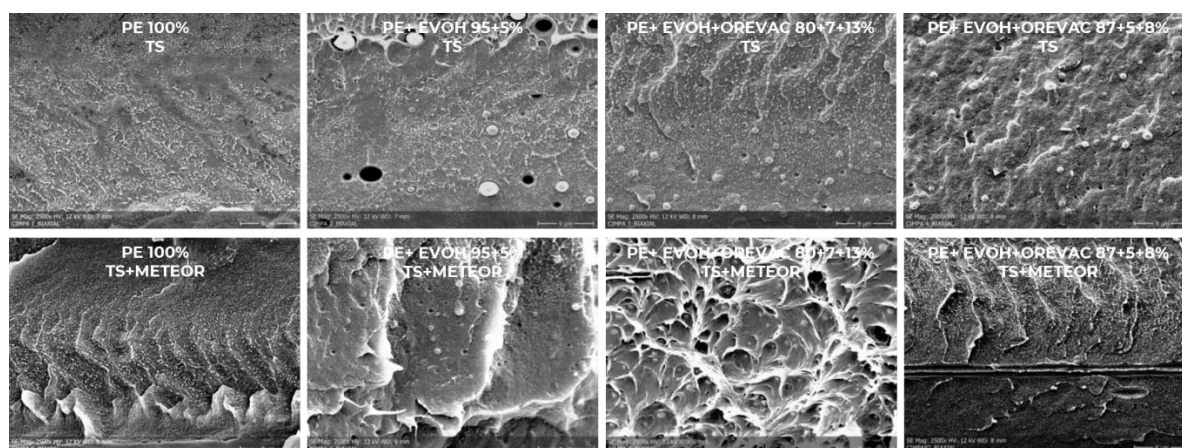


Figure 10: SEM observations of the samples after bi-axial stretching from the PE/EVOH films (x2500)

The barrier properties were measured after bi-axial stretching to see if this treatment decreases the transmission rates. The OTR (Oxygen Transmission Rate) data could not be compared due to insufficient data from IPC. However, the results are unsatisfactory compared to the industrial partners' target of 50mL/m².day target. The WVTR are, on the contrary, all good and below the target (10g/m².day).

The bi-axial stretching can be a powerful tool to use to increase barrier properties, but it needs a full study to establish the right temperatures and the right stretching parameters, which was not the purpose of the project. LDPE and EVOH have differing melting points, and a compromise in the biaxial stretching temperature must be made to be able to stretch a film comprising of PE matrix and a dispersed EVOH phase. Hence, the stretching temperature of 145 °C (dictated by the LDPE matrix phase) might be too limited to achieve proper elongation of the EVOH droplets during biaxial stretching. Thus, due to resources constraint, the full study cannot be made within CIMPA.

e. Rheological properties

For the rheological properties, IPC made Dynamic Frequency Sweep (DFS) and Stress Relaxation (SR) tests. The methods are available in Annex 2.

From the Dynamic Frequency Sweep tests, the frequency at which the storage modulus G' and the viscous modulus G'' cross over is studied (see Figure 11). At a first sight, all the cross over frequencies are very close to each other. The crossover seems to happen at a bit lower frequency when the twin-screw extruder is used with and without METEOR® (dotted bars), especially for the blend PE+EVOH+OREVAC 87+5+8%. The opposite of the frequency corresponds to the relaxation time of a blend. So, it seems that when the twin-screw extruder is used, the relaxation time are higher. Either more interactions are created at the interface (chemical reactions) or there is more interface (dispersion improved). In both conclusions, the blending seems to be more efficient with a twin-screw extruder than a single-screw one.

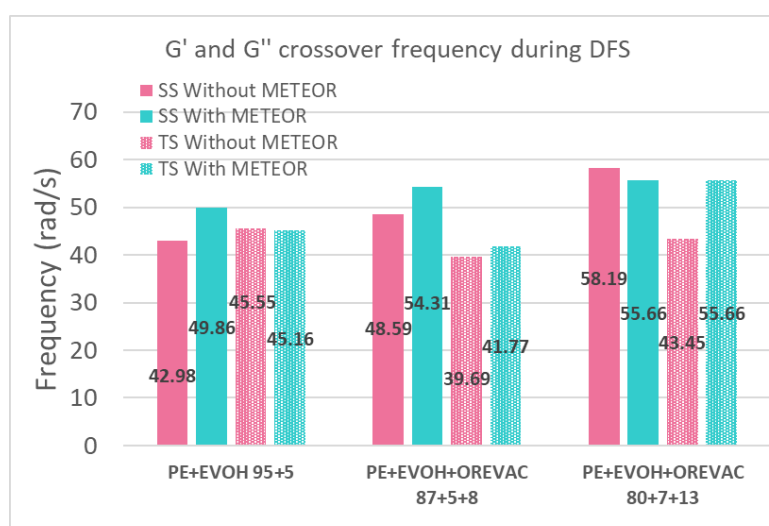


Figure 11: G' and G'' crossover frequency during Dynamic Frequency Sweep (DFS) test for PE/EVOH films at 220°C

In order to study more deeply the relaxation of the different blends, Strain Relaxation (SR) tests were made (see the method in Annex 2). All the curves are displayed in Figure 12.

For the blend without compatibilizer (first graph), the trial made with twin-screw extruder and METEOR® (dark green) show a bump at the end of the curve, especially compared to the one without METEOR® (light green). This bump usually means that there is more interfacial energy than in the blends. Again, there might be two reasons explaining this: either the blend is better dispersed, creating more surface of interface, or some chemical bonding happened. In both cases, the blend is stronger, showing an effect of METEOR®. For the single-screw extrusions, with and without METEOR®, this effect is not as clear.

When a compatibilizer is added (second graph), then the METEOR® effect after the twin-screw extrusion is not as clear. The compatibilizer probably worked efficiently, and no more interface energy was created through the METEOR®

process. With the single-screw extrusion, the effect of METEOR® is shown: again, a bump appeared, showing more interfacial energies.

Finally, when more compatibilizer and EVOH are introduced (third graph), the effect is hidden by the strong interactions created by both process (twin-screw and single-screw extruder) and with or without METEOR®.

In that sense, and for this type of blend, the use of METEOR® might reduce the need of compatibilizer.

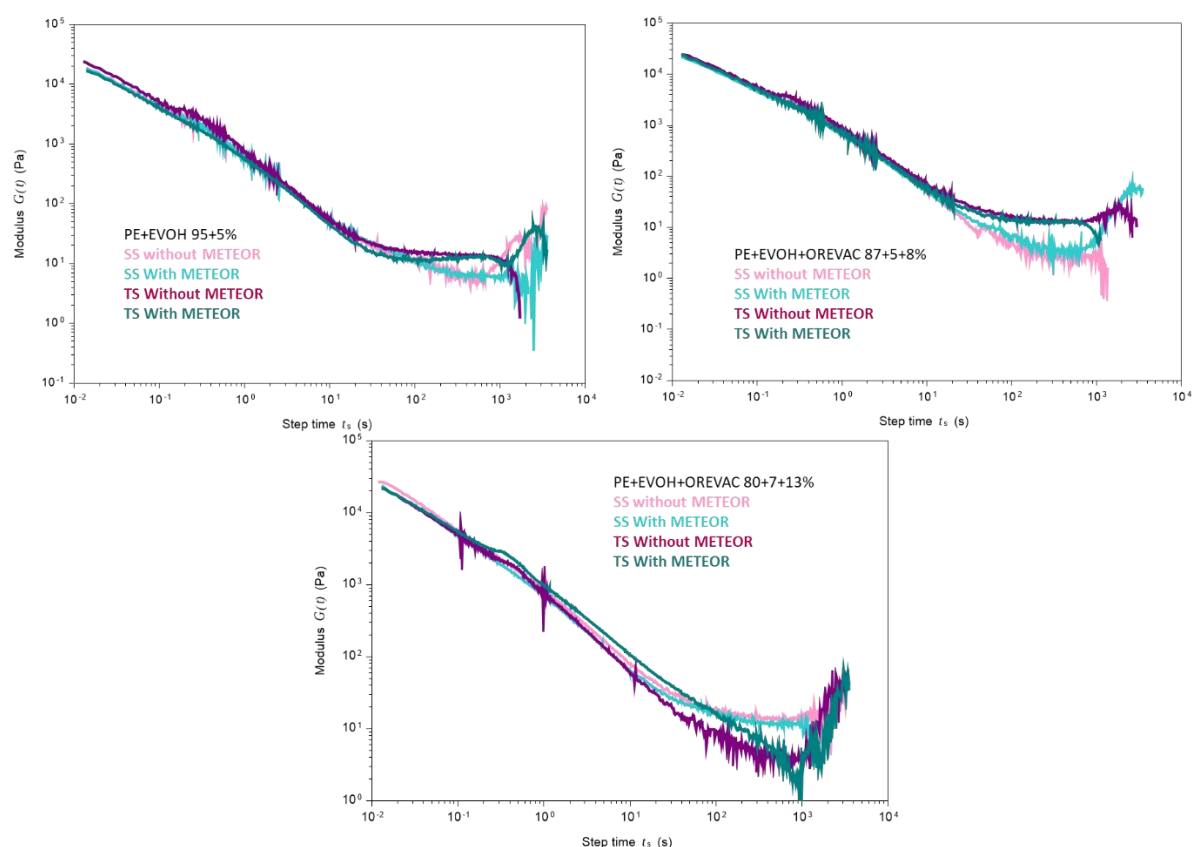


Figure 12: Strain relaxation (SR) curve for PE/EVOH films at 220°C

f. Conclusion for PE/EVOH blends with METEOR®

In this study, several PE/EVOH blends were made:

- With and without compatibilizer
- Using a twin-screw extruder, without and with METEOR®
- Using a single-screw extruder without and with METEOR®

From mechanical properties, the elongation at break in the transversal direction show the effect of METEOR® after a single-screw extruder for all the formulations, but no improvement after a twin-screw extruder.

The barrier properties were satisfying for water-vapor transmission rates, but exceeded the target set up by the industry for oxygen transmission rates. The bi-

axial stretching attempt show no real improvement of the barrier properties. The observation of the structures does not show elongation of the dispersed phase.

The degradation study fails to prove that METEOR® degrades less the materials than a twin-screw extruder. But however, even though the residence time is much increased, the materials are not degraded more than the same trial without METEOR®.

Finally, the rheological properties show that up until a compatibilizer content, the interfacial energy is increased (either by a better dispersion, or the creation of reaction at the interface). When the compatibilizer percentage is higher, then the effect is not seen anymore. In that sense, METEOR® could be used in such blends to reduce the proportion of compatibilizer.

4. Summary of results obtained with PE/PA and PE/PET with METEOR®

All the results of the following trials are available in deliverable 3.3. Only a summary is presented here.

a. PE/PA blends

Several PE/PA blends were prepared with different ratios of PA and compatibilizer. All blends were processed using a single-screw extruder, both with and without the addition of METEOR®

From the mechanical properties analysis, it was observed that the elongation at break in the transversal direction demonstrated the impact of METEOR® following a single-screw extrusion for all formulations containing compatibilizer. This impact was not confirmed with the optical microscopies couples with Infra-Red, which did not show a clear trend.

While the barrier properties met satisfactory levels for water-vapor transmission rates, they exceeded the industry-established target for oxygen transmission rates. Bi-axial stretching experiments revealed no significant improvement in barrier properties.

Observation of the bi-axially stretched structures indicated elongation of the dispersed phase post-stretching, though this might be attributed to the stretching process rather than the METEOR® treatment itself.

The degradation study did not conclusively demonstrate that METEOR® results in lesser degradation compared to a twin-screw extruder. However, employing METEOR®, despite its longer residence times, did not appear to introduce additional degradation to the material.

Furthermore, rheological analysis revealed that at lower compatibilizer proportions, there was an increase in interfacial energy, possibly due to improved dispersion or formation of reactions at the interface. However, this effect was not

observed at higher compatibilizer percentages. Thus, METEOR® could potentially be utilized in such blends to reduce the requirement for compatibilizer.

The conclusion for this blend was very close to the PE+EVOH's one. This might be due to the very similar viscosity ratios ($PA/PE = 1.7$).

b. PET/PET blends

For PE/PET blends, the study utilized only single-screw extruders (SS and SS+METEOR). Various blends were created, with different ratio of PET and a compatibilizer was introduced. Two PET drying methods were tested, including a standard method and a specialized one.

Regarding mechanical properties, the elongation at break in the transverse direction demonstrated the impact of METEOR® following single-screw extrusion across all formulations, particularly with standard drying conditions.

While the barrier properties proved satisfactory for water-vapor transmission rates, they surpassed the industry's target for oxygen transmission rates. Attempts at bi-axial stretching did not significantly enhance barrier properties. Observation of the bi-axially stretched structures revealed no elongation of the dispersed phase post-stretching, due to insufficient temperature to melt the PET phase.

The degradation study did not definitively establish that METEOR® results in less degradation compared to twin-screw extrusion. However, the use of METEOR®, despite its extended residence times, did not appear to introduce additional degradation to the material.

Analysis of rheological properties did not provide conclusive evidence regarding the efficiency of METEOR®. This ambiguity may stem from differences in viscosity behaviours between PE and PET. The viscosity ratio transitioned from 0.3 in the extruder to 100 in the METEOR® process, resulting in a challenging-to-predict behaviour.

c. Conclusion on virgin material METEOR® blends

For all the virgin blends tested, METEOR® proves its efficiency, with the favorable elongation at break shown in the Figure 13.

Indeed, the properties reached the level of a twin-screw extruder (standard extrusion in the graph), and thus using single-screw extruder coupled with METEOR® process.

The extensive rheological study showed that the interfacial energy increased when METEOR® was used for virgin PE/PA and virgin PE/EVOH blends. This indicated that the dispersion was thinner, or that some interactions were created at the interface. In that sense, METEOR® process could be used to reduce the compatibilizer content in such blends.

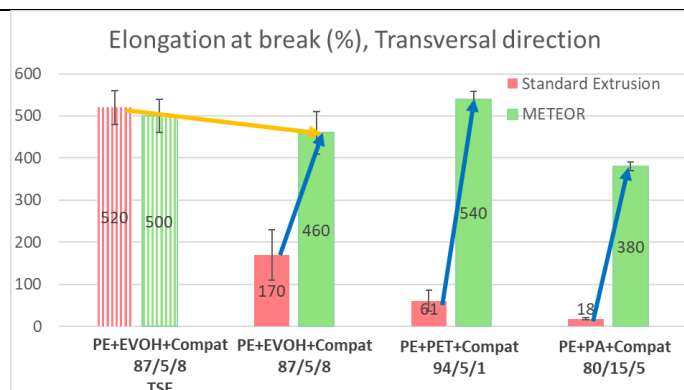


Figure 13: Elongation at break in TD for all the virgin material METEOR® blends

For virgin PE/PET blends, the conclusion was unclear. This was probably due to the high differences in viscosities between these two materials, leading to difficulties to blend efficiently.

However, the study failed to prove that METEOR® gave less degradation than twin-screw extruder. On the other hand, despite its high residence time (30-40 min), the materials were not degraded more than when a twin-screw extruder was used.

For all the blends, the Water Vapor Transmission Rate were satisfying, but the Oxygen Transmission Rate were higher than the industry's target. Attempts at bi-axial stretching did not significantly enhance barrier properties. A deeper study of the stretching parameter would be highly beneficial, but was not the objective of the CIMPA project.

5. MultiNanolayering (MNL) Concept

Regarding both material eco-design and processing, adaptation of existing plastic conversion methods to meet the recycling purposes is currently the main applied approach; particularly aiming to take advantage of the enormous and evolving polymer processing techniques. In this regard, MNL is a relatively recent and powerful extrusion technique for the production of structured polymer films envisaging for the most part the packaging sector. As it is known, plastic packaging applications demands a series of physical properties, particularly food packaging, which should reunite principally strength and barrier properties. Although, traditional cast or blown co-extrusion is extensively present in the packaging sector as main multilayer film production techniques, these require well-defined material characteristics meeting specific functions in the film. Subsequently, a successful film performance is entirely based in the actual component's physicochemical properties. Regarding MNL, although it is also true that the physical properties of the individual layers play a significant role, the final film characteristics are also dictated at a great extent by the dynamics of the layer multiplying process. As illustrated in Figure 14, the MNL, in its basic configuration, involves initially the same procedure as cast film coextrusion, merging two different materials, coming each from one extruder, into a feed block. Afterwards,

the resultant bilayer system is led to a multiplier unit, consisting of a series of multiplier elements (LME) each one being capable of splitting the incoming flux and remerging it, and thus, increasing the number of layers in an exponential way along the whole unit, according to:

$$\text{Number of layers} = 2^{n+1} \quad (1)$$

Where, n is the number of multiplier elements.

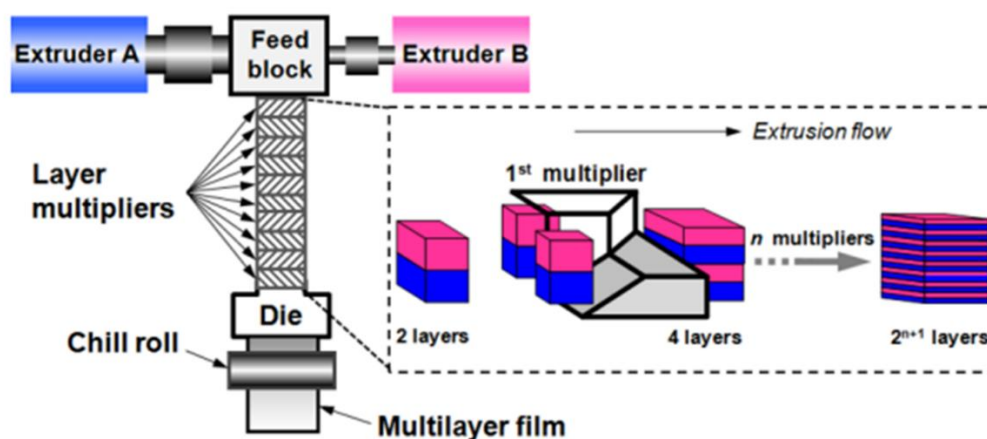


Figure 14: Schematic illustration of the multilayer coextrusion process⁸

Once the desired theoretical number of layers obtained, depending on the number of multiplier elements, the final film is shaped by a die head at the end of the MNL unit and recovered as usual. From a structural point of view, MNL allows targeting layers' thickness depending on both the polymer mass ratio and the number of layers or multiplication steps. By controlling and making interact these parameters in the right balance, the system is capable to attain layers' dimensions in the orders of nanometres. In the proximities of those dimensions, when using semi-crystalline polymers, the quite thin and adjacent layers do not allow the crystallites to grow in the perpendicular direction any longer because of the generated structural hindrances. Therefore, the only available direction for the spherulitic and/or lamellar evolution is along the layer, i.e., the planar direction. This phenomenon is called *confinement* and represents one of the highest potentials of MNL since it promotes a high density of anisotropic crystalline regions, which favours the gas barrier resistance in the application plane of the film. On the other hand, the same phenomenon may also enhance the mechanical behaviour of the film.

It can be observed that, higher layer thickness (in the order of nanometric scale though) at low Tx allows enough room for isotropic lamellar growing of crystalline regions. When thickness is reduced while Tx is increased, more spherulitic grow

⁸ Bo Lu, Pierre Alcouffe, Guillaume Sudre, Sébastien Pruvost, Anatoli Serghei, Chuntai Liu, Abderrahim Maazouz, Khalid Lamnawar (2020). Unveiling the Effects of In Situ Layer–Layer Interfacial Reaction in Multilayer Polymer Films via Multilayered Assembly: From Microlayers to Nanolayers. *Macromol. Mater. Eng.*, 2000076

is favoured and, at very thin layer thickness the in plane lamellar crystalline growing is practically the solely route since other dimensions are no longer available.

Baer et al⁹ emphasize the morphology changes in the crystallinity scale and patterns as the layer thickness allows for confinement phenomenon. This is illustrated in Figure 15, where, as approaching critical layer thickness of PCL (nanometric order in dimensions), by increasing the number of layers, crystalline arrangements change from spherulites to lamellae and finally to single crystals at a thickness well within nanometric scales.

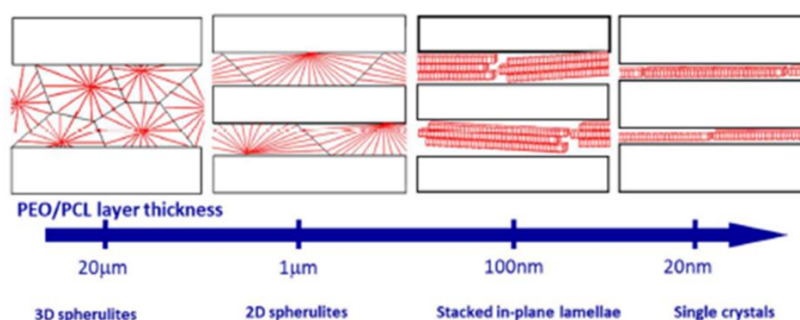


Figure 15: Evolution of the crystalline layer morphology toward highly confined structures as its thickness decreases as function of layers multiplication.

From the same authors as in Figure 15, the impact of the confinement phenomenon in the barrier properties of the multilayer films seems to be obvious in Figure 16. As the layer's thickness of the semi-crystalline component in any of the three polymer pairs of the Figure 16 reaches nanometric dimensions, the permeability significantly decreases because of the homogeneous crystalline barrier built as a result.

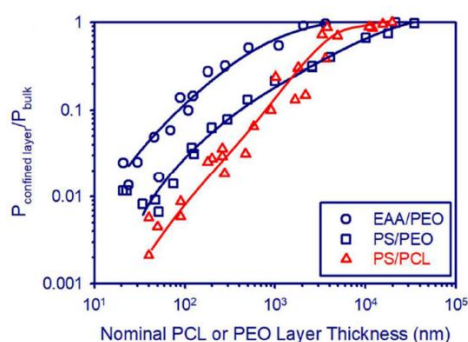


Figure 16: Normalized permeability as function of the semi-crystalline layer in a multilayer film by MNL.

⁹ Zhenpeng Li, Andrew Olah, Eric Baer (2020). Micro- and nano-layered processing of new polymeric systems. Progress in Polymer Science, 102, 101210.

At higher dimensions, less compact and homogenous structures, including amorphous zones, allows for a higher perpendicular diffusion of gas molecules. In addition, it is interesting to observe that, in the opposite direction, as layer's thickness increased beyond 1 μm , the permeability tends to reach constant values and dimensions are no longer a factor affecting this property.

Other attracting approaches in MNL targeting at the same level both barrier properties and mechanical properties are characterized by the inclusion of fillers or reinforcing charge in one of the components of the film. Rivollier et al¹⁰ exfoliated and compatibilized montmorillonite in LLDPE, which subsequently was shaped into film by MNL along with non-filled LLDPE layer as a second component. In addition, bi-axial stretching was carried out on the resultant films. Multilayer films from 17 to as much as 1025 layer were produced just by MNL, reaching for the latter a layer thickness of about 100 nm. Although, montmorillonite platelets were not fully aligned "in plane" into the filled layer during the MNL processing, the auxiliary bi-axial stretching complemented this target, getting in the end highly oriented nanoplatelets at even lower layer's thickness.

Although the potential of the MNL processing technique in the tailor-made elaboration of multilayer film is well documented, essential requirements regarding raw material properties should not be overlooked in order to ensure an efficient process and targeting the desired films characteristics. In this matter, a central aspect concerns the rheological properties of the individual components to be shaped into a multilayer film by MNL. For a successful processing, it is recommended that the viscosity ratio between the film components be kept around 1. The farther the viscosity ratio from this value, either upwards or downwards, the higher the risk of failing the multilayer structure. Particularly, high difference of viscosities leads to what is known as "encapsulation", where the most fluid phase tends to encapsulate the more viscous one, preventing layer-to-layer mechanisms. The described undesired phenomenon is clearly shown in the work by Huang et al¹¹, Figure 17, where viscosity ratio around 1 (matched) promotes multilayer configuration whereas high viscosity ratio (mismatched) generates kind of mixing patterns, including encapsulation phenomenon, which lead to a failed film.

¹⁰ N. Rivollier, R. Schwidessen, G. Cabrera, C. Combeaud, S. Schorr, G. Dennler (2024). Montmorillonite Exfoliation in LLDPE and Factors Affecting Its Orientation: From Monolayer to Multi-Nano-Layer Polymer Films. *Polymers*, 16, 200.

¹¹ R. Huang, J. Silva, B. A. Huntington, J. Patz, R. Andrade1, P. J. Harris, K. Yin, M. Cox, R.T. Bonnezeze, J.M. Maia (2015). Co-Extrusion Layer Multiplication of Rheologically Mismatched Polymers: A Novel Processing Route. *Intern. Polymer Processing*, 3, 317 - 330.

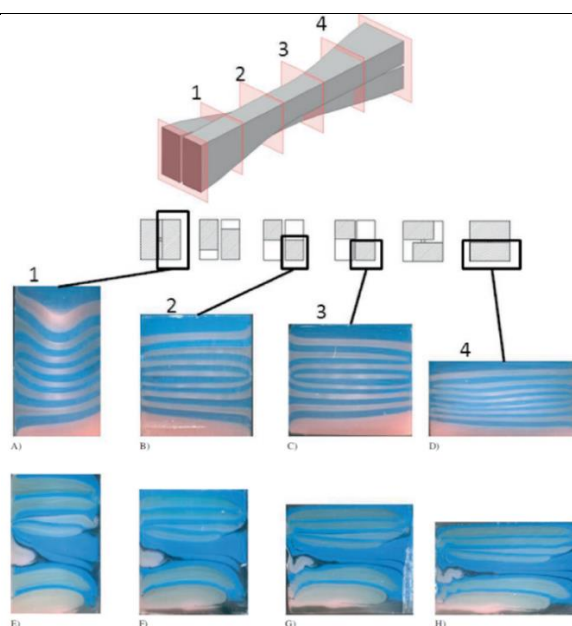


Figure 17: Progression of viscosity matched (A to D) and mismatched (E to H) flow along multiplier die for 32 layer extrudates.

Other important physicochemical factors to take into account for successful films point to the interface stability between adjacent layers. Firstly, poor compatibility between constituents, even at the right viscosity balance, may lead to kind of phenomena known as *interfacial slippage*¹². This undesirable condition can be translated into either processing troubles or later when the poor adhesion favours the readily delamination of the film. As suggested for polymer blends in general, the use of compatibilizers suppresses interfacial issues and ensures the structured building of the multilayer. In this regard, works by Lu et al⁶ and Lozay et al¹³ analysed the effect of compatibilizers in the elaboration of multilayer films by MNL. On the other hand, it is certain that the MNL technique focuses on a great extent in the development of interfaces in non-compatibilized systems by various mechanisms of intermolecular diffusion, thus, favouring stables interfaces¹³.

This concluded the state-of-the-art MNL study, and its interest in polymer recycling. The following paragraphs present the trials made within the CIMPA project.

¹² Khalid Lamnawar, Abderrahim Maazouz, Huagui Zhang (2013). Encyclopedia of Polymer Science and Technology. John Wiley & Sons.

¹³ Quentin Lozay, Quentin Beuguel, Nadège Follain, Laurent Lebrun, Alain Guinault, Guillaume Miquelard-Garnier, Sylvie Tencé-Girault, Cyrille Sollogoub, Eric Dargent, Stéphane Marais (2021). Structural and Barrier Properties of Compatibilized PE/PA6. Membranes, 11(75).

6. PE/PA virgin materials MNL structures

a. First campaign summary

The materials were chosen in collaboration with WP5 trials, and sent by Leygatch, due to availability, delays and prices.

For the LDPE, Leygatch uses several equivalent grades, with the same density, the same Melt Index (2.0g/10min), and all polymerized using radical polymerization.

The four virgin LDPE grades used during these trials are: DOW 352E, SABIC NC1013, REPSOL ALCUDIA 2320FG and BASSELL LUPOLEN 2426H.

The PA was a PA6/6.6 (film grade) also supplied from Leygatch: UBE NYLON 5033B.

All the technical datasheets can be found in Annex 1.

For the first PE/PA campaign, two formulations were tested across four layer configurations: 0 LME (3 layers), 4 LME (33 layers), 8 LME (513 layers), and 9 LME (1025 layers). Based on IPC's experience, the addition of a compatibilizer tends to disrupt the MNL structures. Therefore, it was decided not to introduce a compatibilizer during the virgin polymer trials.

Regarding mechanical properties, the elongation at break in the transversal direction decreased when LMEs were introduced. Conversely, the barrier properties, particularly the Oxygen Transmission Rate (OTR), showed an improvement. Notably, a satisfactory OTR value was achieved with 33 layers, i.e., 4 LME.

This specific configuration was selected to reach the 100 kg production milestone. The same process parameters were maintained, with the only change being the width of the flat die. Unfortunately, the OTR obtained from this production exceeded the industrial target.

Attempts to biaxially stretch the MNL structure revealed that MNL films are not suitable for this process. The layers delaminated due to the absence of a tie layer and a mismatch between the melting temperatures of LDPE and PA, resulting in the formation of voids during biaxial stretching.

After this initial campaign and the production of 100 kg of film using the MNL process, IPC decided to further study two parameters of the PE/PA 85/15 structures:

- The number of LMEs and consequently the number of layers in the final films
- The position of the LMEs in the MNL process, as well as the rolling speed

b. PE/PA 85/15 MNL structure: LME number influence

Materials and Formulations

The formulation chosen was PE/PA 85/15, due to the good OTR result obtained during the first campaign. The point of this trial is to add the data for 5 and 6 LME. The trial of 4 LME was done again, but with a different localization of the LME. The extensive study of the influence of the localization of the LME is examined in the next paragraph (c.).

These trials were made using 2 single-screw extruders:

- Collin E45E (L/D=25) for the layer A, i.e., PE at 220°C
- Collin E30E (L/D=25) for the layer B, i.e., PA at 270°C

The throughputs of both extruders were set to respect the proportion and the throughput of 5kg/h. The flat die used was the Scamex 350, giving films of 350mm width. The MNL temperature was set at 250°C. The chill roll speed was set between 1.5 to 2.0m/min, to obtain films with thickness between 200 and 250µm.

The LME were located as indicated in Table 4.

	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10
4 LME										
5 LME										
6 LME										

Table 4: LME position for PE/PA 85/15 structures, 4, 5 and 6 LME. Green square indicates that the LME is positioned, grey square that it is not

The viscosity ratio of the materials was 0.4 at the beginning of the MNL process (extruders temperatures) and 1.17 at the MNL temperature.

Mechanical properties

The results of tensile measurements can be found in Table 5 (for the methods used see Annex 2).

		Machine direction					Transversal direction				
		Tensile Modulus (MPa)	Stress at yield (MPa)	Elongation at yield (%)	Stress at break (MPa)	Elongation at break (%)	Tensile Modulus (MPa)	Stress at yield (MPa)	Elongation at yield (%)	Stress at break (MPa)	Elongation at break (%)
PE/PA 85/15	0 LME 3 layers	531 ± 13	13.1 ± 0.1	13 ± 0	16.6 ± 1	550 ± 40	483 ± 51	16.4 ± 0.9	14 ± 1	33.5 ± 3.2	390 ± 20
	4 LME 33 layers	329 ± 15	9.97 ± 0.52	15 ± 0	28.7 ± 1.7	530 ± 10	301 ± 20	8.86 ± 0.56	13 ± 0	19.5 ± 1.1	420 ± 20
	5 LME 65 layers	338 ± 22	10.4 ± 0.6	16 ± 0	29.6 ± 1.9	520 ± 10	328 ± 29	9.77 ± 0.36	11 ± 1	17.7 ± 1.1	360 ± 20
	6 LME 129 layers	261 ± 14	8.55 ± 0.6	16 ± 1	21.7 ± 2.3	460 ± 20	291 ± 6	8.23 ± 0.2	11 ± 2	7.8 ± 3.64	90 ± 47
	8 LME 513 layers	354 ± 47	10.6 ± 1.5	14 ± 2	19.6 ± 4.7	460 ± 50	196 ± 22	1.64 ± 0.4	2.1 ± 0.6	1.38 ± 0.44	2.8 ± 1.1
	9 LME 1025 layers	378 ± 30	11.1 ± 1.4	13 ± 2	19.4 ± 5.8	430 ± 60	330 ± 37	/	/	3.29 ± 1.14	1.9 ± 0.4

Table 5: Mechanical properties of PE/PA 85/15 structures (0, 8 and 9 LME from first campaign, and 4, 5 and 6 LME from second campaign)

All properties seem to be impacted by the introduction of LME in the process. They all decrease, a bit for the machine direction, and more in the transversal direction. The most impacted is the Elongation at break (see Figure 18). An abrupt loss of elongation is observed from 5-6 LME.

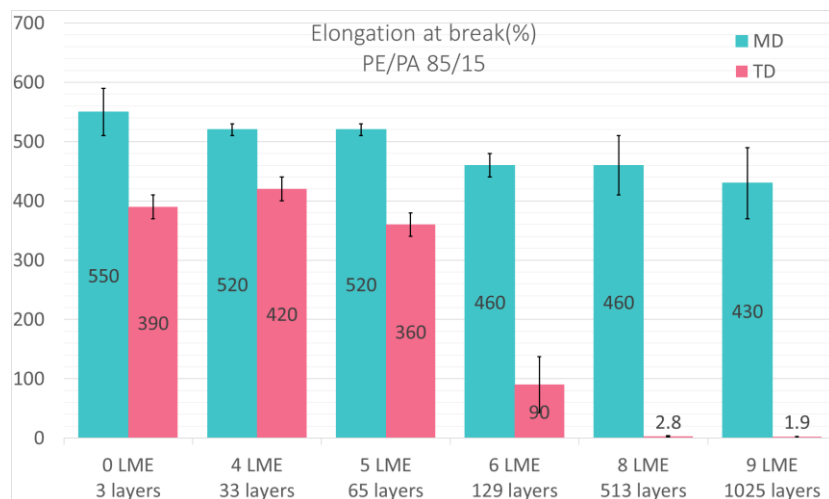


Figure 18: Elongation at break (%) in transversal direction for PE/PA 85/15 structures (0, 8 and 9 LME from first campaign, and 4, 5 and 6 LME from second campaign)

Barrier properties

The methods used to measure the Water Vapor Transmission Rate (WVTR) and the Oxygen Transmission Rate (OTR) can be found in Annex 2, and the results are summarized in Figure 19.

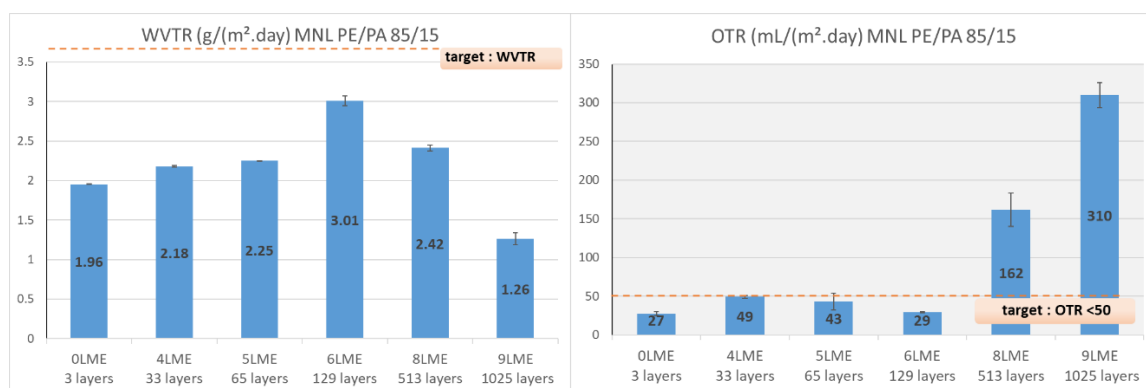


Figure 19: Water-vapor transmission rate (g/(m².day)) and Oxygen Transmission Rate (mL/(m².day)) for PE/PA 85/15 structures (0, 8 and 9 LME from first campaign, and 4, 5 and 6 LME from second campaign)

All the WVTR values are below the target set by the industrial partners in D1.1. For the OTR, the values are satisfying, being under the target set by the industrial in D1.1 (50mL/m².day) until 8 LME (513 layers). When the elongation at break and the OTR are superposed, it seems that between 6 and 8 LME (129 and 513 layers), the structure broke, and the confinement effect is lost, as well as the barrier properties. The loss in mechanical properties is likely due to the absence of compatibilizer between PE and PA phases.

Optical microscope coupled with InfraRed detection

The methods used to see the MNL structures can be found in Annex 2, and the pictures are gathered in Figure 20.

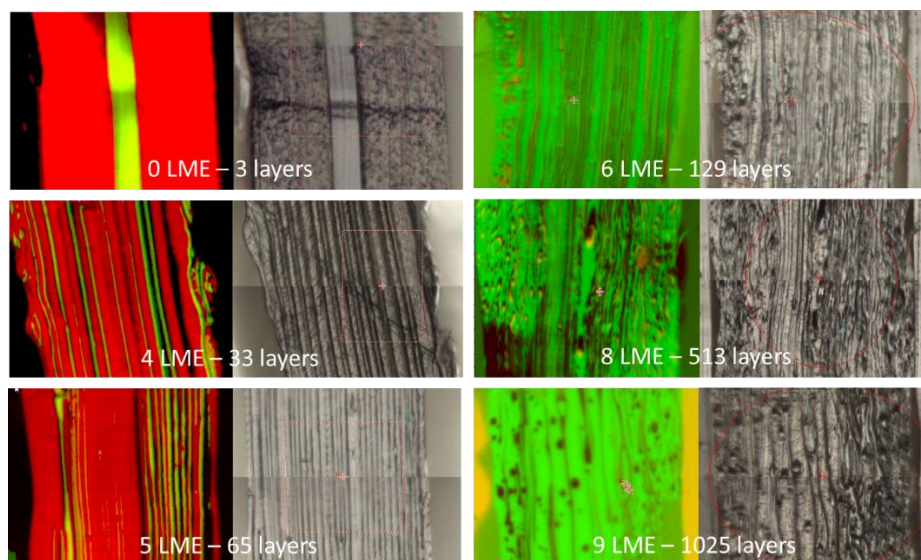


Figure 20: Optical microscopies coupled with Infra-Red detection of PE/PA 85/15 MNL structures. the PE phase is in red in the first column, and in green in the second column

As suspected from mechanical and barrier properties discussions, the structure seems to be regular with a nice stacking of layers until 6 LME (129 layers). With 6 LME, one can see that on the left, the layers broke and start to recombine. In the middle of the film, layers still appears. At 8 LME (513 layers), one can see few layers are seen in the middle of the film's thickness, but on the right and left side, the structure is completely broken. Finally, with 9 LME (1025 layers), no layers are seen anymore.

This is perfectly in line with the mechanical and barrier properties.

Conclusions for PE/PA MNL structures: influence of LME number

For the LME number influence trial, only one formulation was tested (PE/PA 85/15), with three different amounts of LMEs: 4 LME (33 layers), 5 LME (65 layers) and 6 LME (129 layers). These results were combined with those obtained in the first campaign, featuring 0 LME (3 layers), 8 LME (513 layers) and 9 LME (1025 layers).

In Figure 21, elongation at break in the TD, OTR and optical microscopies are overlaid. From this graph, the hypothesis, supported by the optical microscopies, suggests that the layer multiplication was effective up to 6 LME (129 layers). Indeed, the elongation at break is favorable for 0, 4 and 5 LME (3, 33 and 65 layers), but diminishes at 6 LME (129 layers). Conversely, the OTR property follows the opposite trend.

From 8 LME (513 layers) onwards, it appears that the multi-nanolayer structure faltered, leading to layer recombination and a subsequent loss of OTR and mechanical properties.

It is noteworthy that WVTR properties are satisfactory for every structure, even for 8 and 9 LME (513 and 1025 layers).

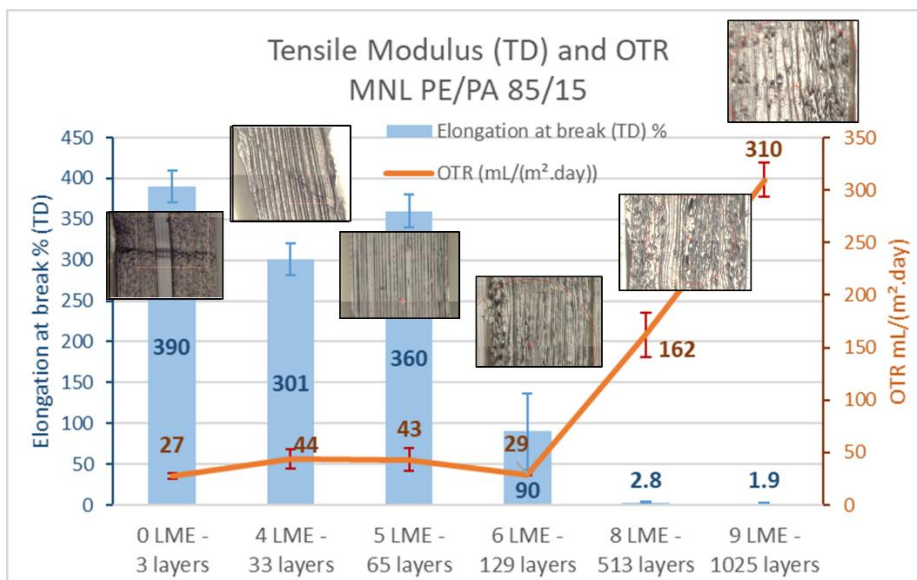


Figure 21: Superposition of Elongation at break (% TD) and OTR for PE/PA 85/15 structures in function of the LME number

c. PE/PA virgin materials structures: LME's position influence

Materials and Formulations

The formulation chosen was PE/PA 85/15, to pursue the analysis of this structure. The point of this trial is to study the influence of the LME position in the MNL modules. The number of layers chosen is 33 layers (4 LME), due to the good results obtained in both elongation at break (TD) and OTR.

These trials were made using 2 single-screw extruders:

- Collin E45E (L/D=25) for the layer A, i.e., PE at 210°C
- Collin E30E (L/D=25) for the layer B, i.e., PA at 270°C

The throughputs of both extruders were set to respect the proportion and the throughput of 5kg/h. The flat die used was the Scamex 350, giving films of 350mm width. The MNL temperature was set at 250°C. The chill roll speed was set between 1.5 to 2.0m/min, to obtain films with thickness between 200 and 250µm.

The LME were located as indicated in Table 6.

	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10
Config 1										
Config 2										
Config 3										

Table 6: LME position for PE/PA 85/15, 4 LME (33 layers) with different configurations. Green square indicates that the LME is positioned, grey square that it is not.

The viscosity ratio of the materials remains 0.3 at the beginning of the MNL process and 1.17 at the MNL temperature.

The 4 LME trial made in the first campaign corresponds to the Configuration 2 and is added to the results table for comparison purpose.

The influence of rolling speed was also studied (speed of the chill roll after the MNL process). This speed influences the final thickness of the films, but also the orientation of the polymer chains within the layers.

Mechanical properties

The results of tensile measurements can be found in Table 7 (for the methods used see Annex 2). To complete the study, the 4 LME trial made during the first campaign is included in the table, and corresponds to the Config 2, 1.6m/min (4th row).

		Machine direction					Transversal direction				
		Tensile Modulus (MPa)	Stress at yield (MPa)	Elongation at yield (%)	Stress at break (MPa)	Elongation at break (%)	Tensile Modulus (MPa)	Stress at yield (MPa)	Elongation at yield (%)	Stress at break (MPa)	Elongation at break (%)
PE/PA 85/15 4 LME : 33 layers	Config 1 1.5m/min	329 ±15	9.97 ±0.52	15 ±0	28.7 ±1.7	530 ±10	301 ±20	8.86 ±0.56	13 ±0	19.5 ±1.1	420 ±20
	Config 1 1.7m/min	327 ±12	10 ±0.3	15 ±0	30.6 ±0.9	520 ±10	324 ±9	9.64 ±0.27	13 ±1	22.1 ±0.7	420 ±20
	Config 1 2.0m/min	257 ±23	8.42 ±0.64	15 ±1	24.3 ±1.5	470 ±20	247 ±29	6.71 ±0.75	7.9 ±1.7	12.3 ±1.2	320 ±20
	Config 2 1.6m/min	400 ±35	12.5 ±0.7	15 ±0	28.2 ±1.8	520 ±20	408 ±27	9.96 ±0.65	6.2 ±0.1	6.87 ±0.66	13 ±3
	Config 2 2.0m/min	232 ±46	7.31 ±1.44	12 ±3	18.6 ±4.9	420 ±70	223 ±76	3.62 ±1.54	3 ±1.1	3.43 ±2.75	33 ±22
	Config 3 2.0m/min	285 ±35	9.16 ±1.09	16 ±0	24.2 ±2	470 ±20	322 ±17	9.66 ±0.59	13 ±0	17.6 ±0.7	360 ±20

Table 7: Mechanical properties of PE/PA 85/15 structures, 4 LME (33 layers) with different configurations and rolling speeds

The properties in machine direction does not seem to be affected by the different configuration, or the rolling speed: they remain stable. In the transversal direction, they follow a U shape: they are good for the first rows, decrease and then increase again. For the visual, Elongation at break is shown in Figure 22.

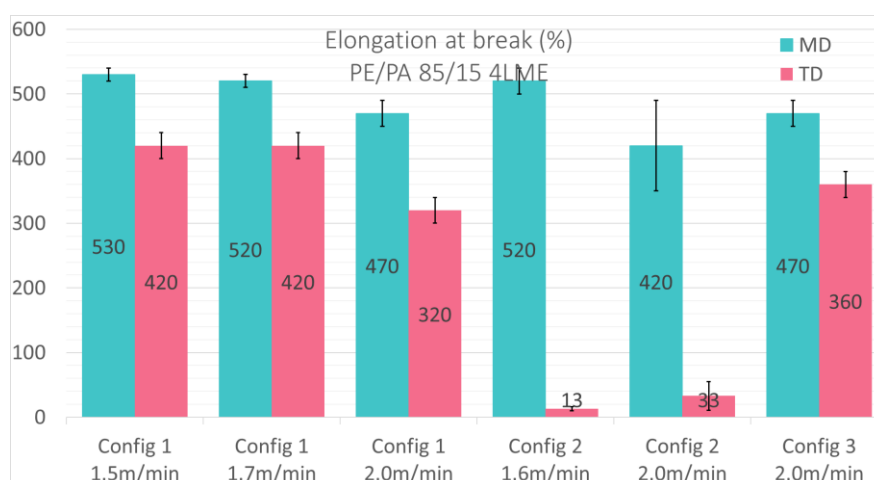


Figure 22: Elongation at break of PE/PA 85/15 structures, 4 LME (33 layers) with different configurations and rolling speeds

At a first sight, it seems that the Config 2 is not favourable to the mechanical properties: the elongation at break in TD are very low. The Config 1 with the highest rolling speed (2.0m/min) is a bit affected too, but this might be due to the thickness of the film, which decreases when the rolling speed increases.

To see if the MNL structure is affected, barrier properties are studied.

Barrier properties

The methods used to measure the Water Vapor Transmission Rate and the Oxygen Transmission Rate can be found in Annex 2, and the results are summarized in Figure 23).

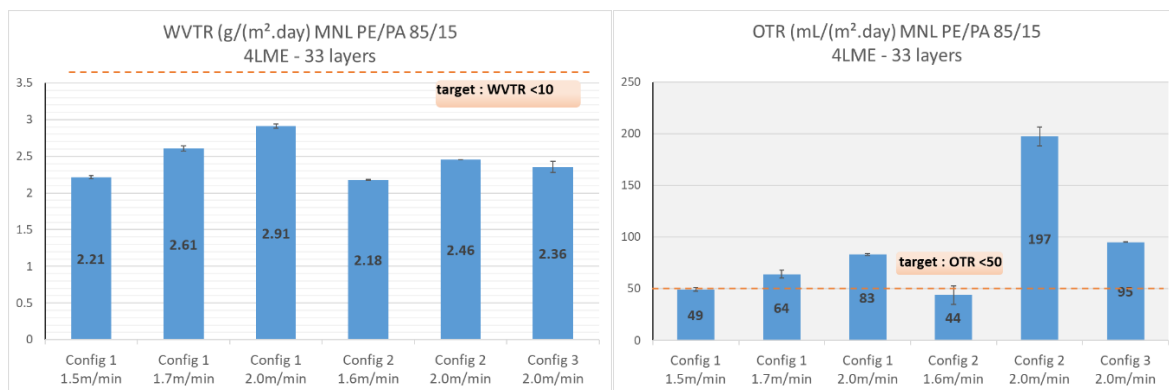


Figure 23: Water-vapor transmission rate (g/(m².day)) and Oxygen Transmission Rate (mL/(m².day)) for PE/PA 85/15 structures, 4 LME (33 layers) with different configurations and rolling speeds

The WVTR are, as usual, all satisfying. For the OTR values, with Config 1, the OTR are increasing with the rolling speed. Since the thickness of the film decreases, the Figure 24 shows the OTR multiplied by thickness result, in order to take into account this factor.

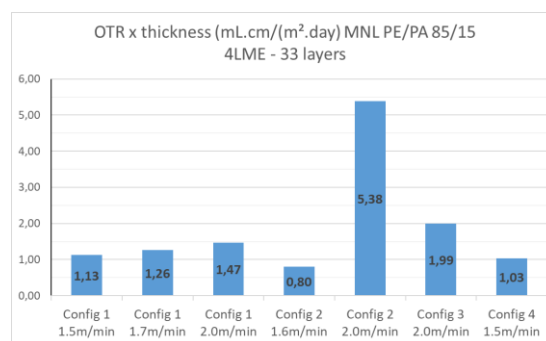


Figure 24: Oxygen Transmission Rate x thickness (mL.cm/(m².day)) for PE/PA 85/15 structures, 4 LME (33 layers) with different configurations and rolling speeds

In this figure, the config 2 is not very favourable to the barrier properties, especially at 2.0m/min. The config 3 presents also high values.

To respect the target set by the industrial partners, the config 1 and 4 are the most suitable. Taking into account mechanical properties results, the config 2 and 3 are probably showing broken layers, which compromise the confinement effect and the good barrier properties.

Optical microscope coupled with InfraRed detection

The methods used to see the MNL structures can be found in Annex 2 and the pictures are gathered in Figure 25.

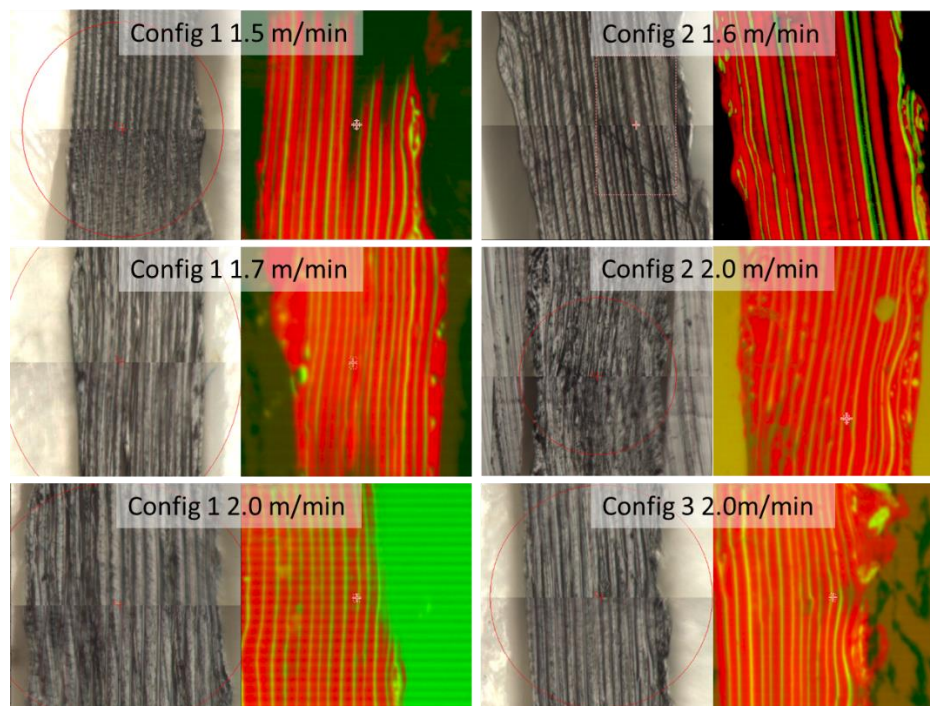


Figure 25: Optical microscopies coupled with Infra-Red detection of PE/PA 85/15 MNL structures, influence of LMEs' positions. The PE phase is in red.

Conversely to what was seen in mechanical and barrier properties, no real differences are seen between these structures. In all images, the external layers seems to be more fragile for configuration 2 and 3, starts to broke and recombine. But this is seen for the 2 external layers only, in the core of the film, the layers seems regular and homogeneous.

Conclusions for LME position influence

For the LME's position influence trial, only one formulation was tested (PE/PA 85/15), with the number of layers set at 33 (4 LME).

The LME were inserted either at the beginning of the process (Config 1), at the end (Config 2) or dispersed all along the modules (Config 3).

In the Figure 26, elongation at break in the TD, OTR and optical microscopies are overlaid. From this graph, the hypothesis, sustained by the results, suggests that the multiplication of the layers did not work for Config 2 and 3. Indeed, the elongation at break is very low for these configurations, as well as the OTR. From the first 3 bars, one can see the influence of the rolling speed. If the elongation at break drop can be explained by the reduction of film thickness, the OTR were multiplied by the film thickness. The increase is then limited for these trials.

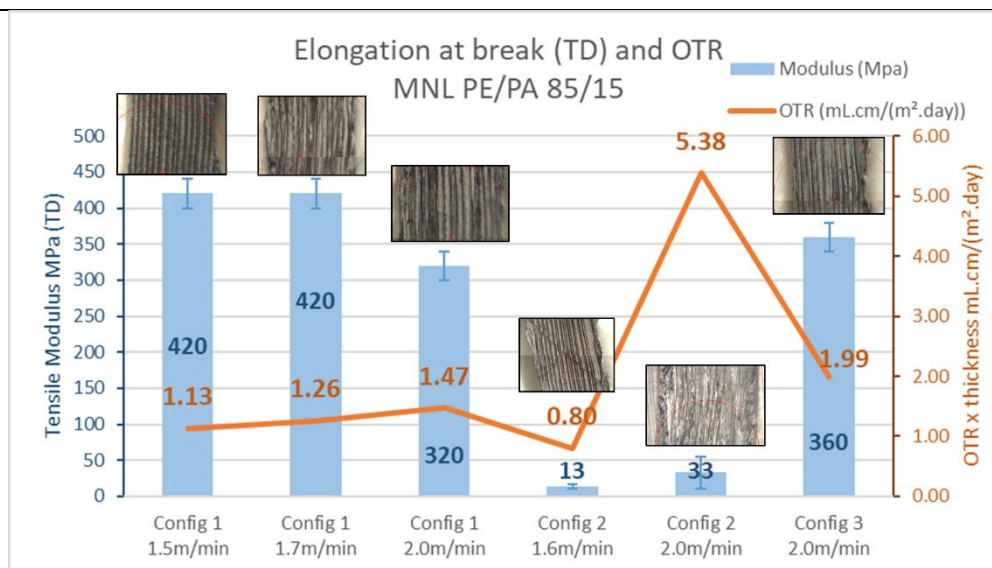


Figure 26: Superposition of Elongation at break (% TD), OTR x thickness and optical observations for PE/PA 85/15 structures with different configurations and rolling speeds

To conclude, it seems favourable for the MNL structures to have the LME placed at the beginning of the MNL process (Config 1). This might be due to the viscosity ratio or the material temperatures of this trials: when the PE arrived in the MNL process, it is at the extruder temperature (210°C), and the PA at 270°C. The viscosity ratio is then of 0.3. But as the MNL process temperature is set at 250°C, the viscosity ratio increased all along the modules to reach the 1.17 value. The favourable viscosity ratio being 1 (see the state-of-the-art), it seems that this ratio is achieved at the beginning of the MNL process. This might explain the influence of the LME position for the PE/PA 85/15 structures.

7. Summary of results obtained for PE/EVOH and PE/PET virgin materials MNL structures

All the results of the following trials are available in deliverable 3.4. Only a summary is presented here.

a. PE/EVOH MNL structures

For PE/EVOH structures, two formulations were tested across three different layer configurations: 0 LME (3 layers), 8 LME (513 layers) and 9 LME (1025 layers).

In terms of mechanical properties, the elongation at break in the transversal direction decreased with the introduction of LME.

Conversely, the barrier properties, particularly the Oxygen Transmission Rate, exhibited an opposite trend. However, the melt flow index indicated no evidence of polymer degradation.

It appears that the number of LMEs tested (or the targeted number of layers) was too high for this structure, considering the outcomes.

Ideally, the study should have been conducted with a lower number of layers.

Unfortunately, due to a equipment breakage, and the extensive study already conducted with the elongational flow mixer on PE+EVOH blends (as detailed in deliverable 3.3), this adjustment will not be feasible within the CIMPA project.

b. PE/PET MNL structures

For PE/PET structures, two formulations were tested across three different layer configurations: 0 LME (3 layers), 4 LME (33 layers) and 8 LME (513 layers). Notably, for the virgin polymer trials, the decision was made not to introduce a compatibilizer.

Regrettably, these trials failed. Inhomogeneous films were obtained, with light and opaque areas visible with the naked eye.

Laboratory investigations conclusively revealed that these opaque and light regions did not conform to the intended layering specifications. Furthermore, infrared spectroscopy confirmed that the opaque areas exhibited a higher concentration of PET chemical functionality, while the lighter areas were enriched with PE functions. This stark differentiation strongly indicates the occurrence of PET layer breakage and subsequent recombination, leading to segregation on one side of the film at a macroscopic level.

Analysis of the rheological properties of PE and PET revealed markedly disparate behaviors. Coupled with the evident lack of compatibility between the two polymers, it became evident that the MNL process and design should be optimized for such complex structures.

8. Post-industrial and post-consumer wastes blends

a. Materials and Formulations

For this trial, IPC received from VTT the materials resumed in Table 8. VTT analysed the composition after their recompounding (see deliverable 5.1).

	Code	Description	Composition (wt.%) from VTT analysis			
PE+PA	PIR		LDPE+tie	PA6-66	Compatibilizer	Stabilizer
	CIMPA-095	Upgraded r-PE/PA	75.70	15.90	8.00	0.40
	PCR		LDPE+tie	PA6-66	Compatibilizer	Stabilizer
	CIMPA-063	Upgraded r-PE/PA	93.32	2.68	3.6	0.4
PE+EVOH	PIR		LDPE+tie	EVOH	Compatibilizer	Stabilizer
	CIMPA-102	Upgraded r-PE/EVOH	88.10	5.80	5.80	0.30
PE+PET	PIR0		LDPE	PET	Compatibilizer	Stabilizer
	CIMPA-105	Upgraded r-PE/PET	85	15.00	0.00	0.00
	PIR1		LDPE	PET	Compatibilizer	Stabilizer
	CIMPA-107	Upgraded r-PE/PET	89.4	7.50	2.50	0.60
	PIR2		LDPE	PET	Compatibilizer	Stabilizer
	CIMPA-108	Upgraded r-PE/PET	87.20	9.00	3.00	0.80
	PIR3		LDPE	PET	Compatibilizer	Stabilizer
	CIMPA-109	Upgraded r-PE/PET	81.95	12.75	4.20	1.10
	PCR		LDPE+tie	PET	Compatibilizer	Stabilizer
CIMPA-068	Upgraded r-PE/PET	94.24	2.96	2	0.8	

Table 8: Blends received from VTT (PIR: post-industrial recyclates, PCR: post-consumer recyclates)

IPC then diluted these recyclates with 2 targets in mind: the final recycled content (12 wt.% for METEOR® blends and 10 wt.% for MNL structures), and the barrier material content (5 wt.% for EVOH and 15wt.% for PA6/6.6) following the RecyClass guidelines for recyclability. For PET, the target was set at 10 wt.%. The virgin materials used are the same as the ones used in the previous trials, sent by Leygatech (see the Technical Data Sheet in Annex 1).

For the METEOR® blends formulations (see Table 9), the barrier contents were multiplied by 2, in order to enhance their effect.

For the METEOR® formulations, trials were made using a single-screw extruder Collin E45E (L/D=25) followed by the METEOR® process (corresponding to the process referred as SS+METEOR in the virgin materials trials). The flat die used was the Scamex 350, giving films of 350mm width. The extruder and METEOR® temperatures were set at 240°C, and the throughput was comprised between 3.5 and 4kg/h.

	Target % Waste	Blends made with METEOR®			Final Composition (wt%)				% Barrier
PE+PA	0.12	CIMPA-095	PE	PA	LDPE+tie	PA6-66	Compatibilizer	Stabilizer	PA6-66
	0.12	0.752	0.128	0.84284	0.14708	0.0096	0.00048	0.15	
PE+PA	0.12	CIMPA-063	PE	PA	LDPE+tie	PA6-66	Compatibilizer	Stabilizer	PA6-66
	0.12	0.584	0.296	0.695984	0.299216	0.00432	0.00048	0.3	
PE+EVOH	0.12	CIMPA-102	PE	EVOH	LDPE+tie	EVOH	Compatibilizer	Stabilizer	EVOH
	0.12	0.84	0.04	0.94572	0.04696	0.00696	0.00036	0.05	
PE+PET	0.12	CIMPA-107	PE	PET	LDPE	PET	Compatibilizer	Stabilizer	PET
	0.12	0.792	0.088	0.89928	0.097	0.003	0.00072	0.1	
	0.12	CIMPA-108	PE	PET	LDPE	PET	Compatibilizer	Stabilizer	PET
	0.12	0.792	0.088	0.89664	0.0988	0.0036	0.00096	0.1	
	0.12	CIMPA-109	PE	PET	LDPE	PET	Compatibilizer	Stabilizer	PET
	0.12	0.792	0.088	0.89034	0.1033	0.00504	0.00132	0.1	
	0.12	CIMPA-068	PE	PET	LDPE+tie	PET	Compatibilizer	Stabilizer	PET
	0.12	0.68	0.2	0.793088	0.203552	0.0024	0.00096	0.2	

Table 9: Compositions blended in METEOR® process

For the MNL structures, virgin LDPE and barrier materials (EVOH, PA and PET) were added by IPC for the process, following the formulation in Table 10. PE was added in layer A, alone, and the layer B consisted in a blend of the barrier materials and the compounds from VTT. The final composition of each blend (taking into account the composition of the blends from VTT) is also indicated in this table.

	Code	Target % Waste	Layer composition			Final Composition (wt.%)				% Barrier
PE/PA	PE/PA PIR	0.1	Layer A	Layer B		LDPE	PA6-66	Compatibilizer	Stabilizer	PA6-66
			PE	CIMPA-095	PA					
	PE/PA PCR	0.1	Layer A	Layer B		LDPE	PA6-66	Compatibilizer	Stabilizer	PA6-66
			PE	CIMPA-063	PA					
PE/EVOH	PE/EVOH PIR	0.1	Layer A	Layer B		LDPE	EVOH	Compatibilizer	Stabilizer	EVOH
			PE	CIMPA-102	EVOH					
PE/PET	PE/PET PIRO	0.1	Layer A	Layer B		LDPE	PET	Compatibilizer	Stabilizer	PET
			PE	CIMPA-105	PET					
	PE/PET PIR1	0.1	Layer A	Layer B		LDPE	PET	Compatibilizer	Stabilizer	PET
			PE	CIMPA-107	PET					
	PE/PET PIR2	0.1	Layer A	Layer B		LDPE	PET	Compatibilizer	Stabilizer	PET
			PE	CIMPA-108	PET					
	PE/PET PIR3	0.1	Layer A	Layer B		LDPE	PET	Compatibilizer	Stabilizer	PET
			PE	CIMPA-109	PET					
	PE/PET PCR	0.1	Layer A	Layer B		LDPE	PET	Compatibilizer	Stabilizer	PET
			PE	CIMPA-068	PET					

Table 10: Compositions processed through MNL

These trials were made using 2 single-screw extruders:

- Collin E45E (L/D=25) for the layer A, i.e., PE at 215°C
- Collin E30E (L/D=25) for the layer B, i.e., PA at 270°C, EVOH at 215°C and PET at 280°C, as virgin material trials

The throughputs of both extruders were set to respect the proportion and the throughput of 5kg/h. The flat die used was the Scamex 350, giving films of 350mm width. The MNL temperature was set at 250°C for PA and PET trials, and 220°C for EVOH one. The chill roll speed was set between 3.1 to 3.3m/min, to obtain films with thickness around 100µm.

The LME were located as indicated in Table 4.

	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10
4 LME										

Table 11: LME position for MNL recyclates. Green square indicates that the LME is positioned, grey square that it is not

b. Mechanical properties

METEOR® blends

The results of tensile measurements of METEOR® blends can be found in Table 12. The test method can be found in Annex 2.

For comparison purposes, the properties of virgin material blends are reported in the Table 12 in blue. They are coming from the previous section of this deliverable (PE+EVOH+OREVAC 87+5+8%, PE+PA+OREVAC 80+15+5% and PE+PET 90+10% with the standard drying protocol).

		Machine direction					Transversal direction				
		Tensile Modulus (MPa)	Stress at yield (MPa)	Elongation at yield (%)	Stress at break (MPa)	Elongation at break (%)	Tensile Modulus (MPa)	Stress at yield (MPa)	Elongation at yield (%)	Stress at break (MPa)	Elongation at break (%)
PE+PA	SS	308 ± 5	9.48 ± 0.12	22 ± 1	13.3 ± 0.6	340 ± 10	349 ± 11	8.58 ± 0.15	10 ± 0	7.79 ± 0.56	18 ± 2
	SS+ METEOR	370 ± 8	11.3 ± 0.1	17 ± 1	17.6 ± 0.3	420 ± 0	403 ± 7	11.2 ± 0	14 ± 0	13.9 ± 0.3	380 ± 10
	PIR	380 ± 11	/	/	13.5 ± 0.6	420 ± 30	414 ± 16	10.6 ± 0.4	13 ± 0	8.92 ± 0.52	87 ± 39
	PCR	414 ± 5	12.7 ± 0.2	19 ± 1	25.8 ± 2.2	410 ± 20	445 ± 8	/	/	10.7 ± 0.2	6.6 ± 0.3
PE+EVOH	SS	409 ± 16	10.1	16 ± 1	17.8 ± 0.8	560 ± 20	443 ± 16	10.2 ± 0.3	8.8 ± 0.3	8.53 ± 1.06	11 ± 1
	SS+ METEOR	400 ± 25	9.87	15 ± 1	18.6 ± 0.6	540 ± 20	333 ± 5	8.54 ± 0.05	12 ± 0	9.97 ± 0.32	430 ± 10
	PIR	389 ± 22	/	/	18 ± 1.2	480 ± 60	418 ± 8	10 ± 0.2	11 ± 0	13.3 ± 1.2	530 ± 10
PE+PET	SS	734 ± 28	16.3 ± 0.3	8.4 ± 0.4	11.2 ± 1	160 ± 40	598 ± 40	/	/	12.2 ± 0.3	5.6 ± 0.5
	SS+ METEOR	469 ± 9	13 ± 0.3	16.5 ± 0.6	16.8 ± 1.5	440 ± 40	543 ± 8	12.4 ± 0.1	11 ± 0	10.8 ± 0.2	240 ± 120
	PIR1	447 ± 8	10.7 ± 0.2	13 ± 1	16.2 ± 1.1	500 ± 40	357 ± 22	7.5 ± 0.5	6.5 ± 0.3	6.3 ± 0.37	30 ± 23
	PIR2	452 ± 17	10.6 ± 0.3	12 ± 1	16.8 ± 1	530 ± 20	349 ± 24	7.05 ± 0.4	5.4 ± 0.3	6.14 ± 0.51	38 ± 17
	PIR3	437 ± 29	10.2 ± 0.6	12 ± 0	15.8 ± 1.5	520 ± 30	368 ± 29	7.32 ± 0.52	5.2 ± 0.2	6.71 ± 0.56	29 ± 13
	PCR	425 ± 34	8.73 ± 0.6	6.4 ± 0.6	9.28 ± 0.9	360 ± 30	301 ± 19	/	/	4.01 ± 0.37	2.1 ± 0.3

Table 12: Mechanical properties of recyclates materials METEOR® films. The values in blue were taken from the previous trials with virgin materials

When analysing the results, several tendencies can be found:

- In machine direction, the properties are maintained. This is due to the cast film process just after SS+METEOR: the materials are oriented during this process, orienting the materials in the machine direction.
- In transversal direction, elongation at break and at yield decrease when PIR are processed, and even more when PCR are (see Figure 27 for an example). Meanwhile, tensile modulus and stresses (at break and at yield) increase, especially for PE/PA and PE/EVOH materials. The final film is then stiffer, with a probable degradation of the material during its lifetime. For PE/PET blends, all the properties decrease when recyclates are introduced: the final film is more fragile. This might be due to the poor compatibility between PET and PE.

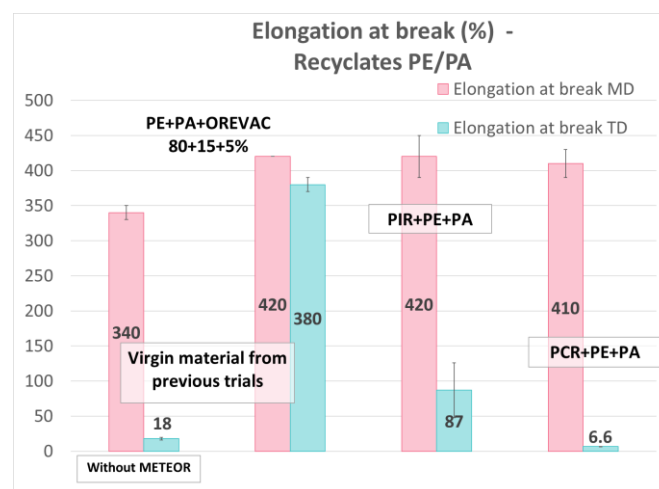


Figure 27: Elongation at break for PE/PA recyclates films

MNL structures

The results of tensile measurements can be found in Table 13. The test method can be found in Annex 2.

For comparison purposes, the properties of virgin material blends are reported in the Table 13 too (in blue). They are coming from the previous trials.

		Machine direction					Transversal direction				
		Tensile Modulus (MPa)	Stress at yield (MPa)	Elongation at yield (%)	Stress at break (MPa)	Elongation at break (%)	Tensile Modulus (MPa)	Stress at yield (MPa)	Elongation at yield (%)	Stress at break (MPa)	Elongation at break (%)
PE/PA	4 LME Config 1 2.0m/min	257 ± 23	8.42 ± 0.64	15 ± 1	24.3 ± 1.5	470 ± 20	247 ± 29	6.71 ± 0.75	7.9 ± 1.7	12.3 ± 1.2	320 ± 20
	PIR	201 ± 32	/	/	10.9 ± 4.6	320 ± 100	189 ± 19	/	/	3.44 ± 0.46	7.3 ± 2.4
	PCR	315 ± 23	9.48 ± 0.07	18 ± 1	20.9 ± 2.1	390 ± 20	332 ± 27	8.17 ± 0.6	9.8 ± 0.9	7.22 ± 0.83	12 ± 1
PE/EVOH	PE/EVOH 90/10 0 LME	698 ± 48	16.1 ± 0.5	14 ± 0	24.4 ± 2.6	280 ± 10	829 ± 30	16.5 ± 0.3	6.3 ± 0.2	13.6 ± 0.9	55 ± 17
	PE/EVOH 90/10 8 LME	412 ± 37	/	/	10.5 ± 1.4	170 ± 60	424 ± 27	/	/	4.19 ± 0.55	1.4 ± 0.3
	PE/EVOH PIR	299 ± 16	/	/	17.2 ± 1.1	440 ± 20	291 ± 13	7.4 ± 0.31	12 ± 1	11.3 ± 1.0	500 ± 50
PE/PET	PIR0	371 ± 250	8.64 ± 1.07	11 ± 1	15.9 ± 1.7	520 ± 20	419 ± 53	8.37 ± 1.2	4.7 ± 1.1	7.84 ± 0.93	9.3 ± 5.1
	PIR1	349 ± 26	8.54 ± 0.74	13 ± 1	15.9 ± 1.8	520 ± 30	315 ± 31	7.28 ± 0.63	8.4 ± 2.2	7.98 ± 0.76	220 ± 20
	PIR2	369 ± 32	8.33 ± 0.73	11 ± 1	13.2 ± 2.7	430 ± 40	302 ± 36	5.41 ± 0.78	3.5 ± 0.4	5.55 ± 0.88	13 ± 7
	PIR3	368 ± 21	8.59 ± 0.42	12 ± 1	14.8 ± 1.6	490 ± 40	326 ± 16	6.18 ± 0.41	4.8 ± 1.1	6.19 ± 0.72	36 ± 25
	PCR	398 ± 15	9.10 ± 0.52	12 ± 1	15.8 ± 0.7	510 ± 20	350 ± 17	8.15 ± 0.43	7.8 ± 0.8	7.58 ± 0.21	210 ± 50

Table 13: Mechanical properties of recyclates MNL structures. The values in blue were taken from the previous trials with virgin materials

For the PE/PA recyclates, the stresses at break and at yield, as well as the elongation at break (see Figure 28) decrease. The tensile moduli remained stable. This shows the degradation of the recycled materials, which might have happened during its lifetime or the recycling processing (decontamination, VAREX, MNL...).

For the PE/EVOH recyclates, the conclusion is quite the same: all mechanical properties decreased. The main difference is for elongation at break, which increased (see Figure 28). This confirms that 8 LME (i.e., 513 layers) were too much for this type of blends, as indicated in the conclusion of the paragraph 7a.

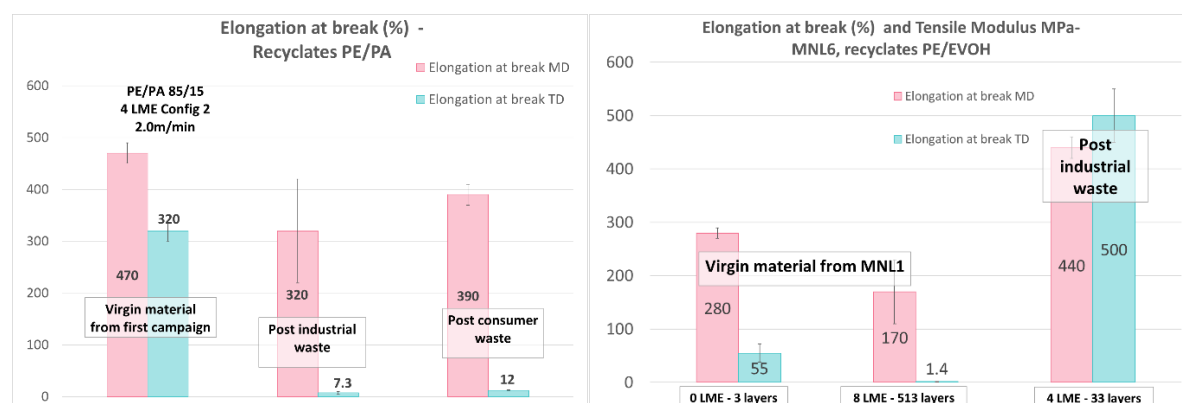


Figure 28: Elongation at break of PE/EVOH and PE/PA recyclates MNL structures

For the PE/PET recyclates, since the trial with virgin materials failed, the only conclusion that can be made is that the mechanical properties are quite equivalent between post-industrial (PIR) and post-consumer (PCR) recyclates.

c. Barrier properties

METEOR® blends

The methods used to measure the Water Vapor Transmission Rate and the Oxygen Transmission Rate can be found in Annex 2, and the results are displayed in Figure 29.

The results are presented in comparison with virgin materials from previous campaign (PE+EVOH+OREVAC 87+5+8%, PE+PA+OREVAC 80+15+5% and PE+PET 90+10% with the standard drying protocol). For the PE/PA blends, all the WVTR value are below the target. For OTR, they are all way higher than the target. The same conclusions can be made for PE/EVOH blends and PE/PET blends.

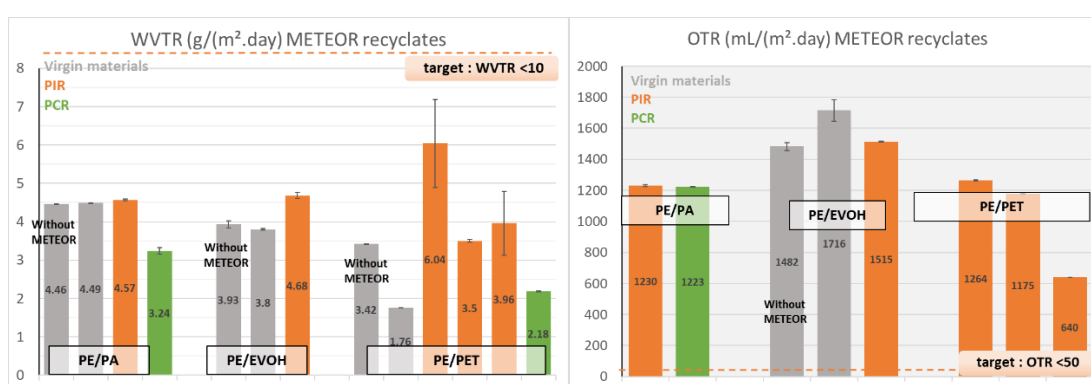


Figure 29: a) Water-vapor transmission rate (g/(m².day)) and b) Oxygen transmission rate (mL/(m².day)) for recycles films

As for PE/EVOH and PE/PA films, attempts of biaxial stretching of the films were made at VTT, as proposed in the Grant Agreement following the same hypothesis of increasing the diffusion path (the methods used is available in Annex 3).

Every film was stretchable at ratio 1.5x1.5, and pictures of the films can be found in Annex 3. On this bi-axially stretched films, observations were made, using optical microscope (see Figure 30a) and SEM (see Figure 30b).

From the optical microscopies, several observations can be made. For PE/PA blends, the one containing PIR seems to be more homogeneous than the one with PCR. This seems understandable, because PIR commonly present less pollution and inks than PCR. For the PE/PET blends, a lot of dark spots are visible. They are printing residues from the PIR received from Eversia. Again, the PE+PET+PCR blend seems to be rougher than the PIR ones.

This is confirmed by the SEM observations, all at the same scale (x5000). Bigger particles can be seen when PCR are used, demonstrating a worse dispersion probably due to pollution from the consumer waste which is not present in the PIR.

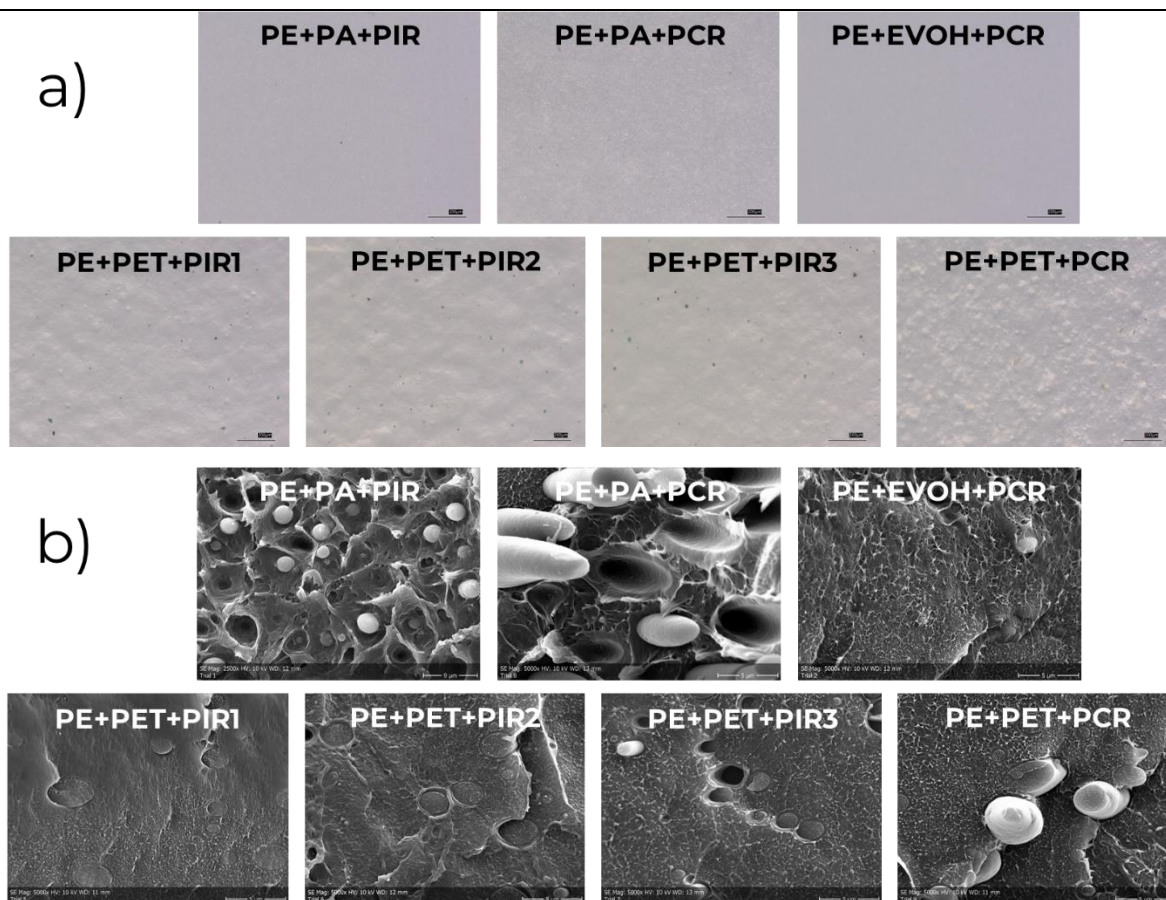


Figure 30: Optical microscopies (a) and SEM observations x5000 (b) of the recyclates METEOR® films after bi-axial stretching

For comparison purpose, the WVTR and OTR measured after bi-axial stretching were multiplied by the thicknesses of the films (see Table 14). Indeed, after the stretching, the films were thinner, and so the rates were bigger. Taking into account this factor, the OTR and WVTR are improved by bi-axial stretching.

		WVTR x thickness (g.cm/(m ² .day))		OTR x thickness (mL.cm/(m ² .day))	
		Before stretching	After stretching	Before stretching	After stretching
PE+PA	SS+METEOR	0.052	0.045	NA	11.44
	PIR	0.049	0.038	13.13	9.16
	PCR	0.043	0.05	8.38	7.98
PE+EVOH	SS+METEOR	0.04	0.035	15.36	NA
	PIR	0.061	0.024	15.94	10.42
PE+PET	SS+METEOR	0.019	0.039	NA	11.81
	PIR1	0.038	0.034	14.41	10.84
	PIR2	0.042	0.038	14.08	14.21
	PIR3	0.043	0.039	15.35	14.02
	PCR	0.028	0.023	NA	NA

Table 14: WVTR and OTR values multiplied by film thickness for recyclates blends. The values in blue were taken from the previous trials with virgin materials

But the values of OTR are still very high. To reach the target of 50mL/m².day, the thickness of the film would be 2mm for an value equal to 10 mL.cm/m².day.

MNL Structures

To see if the MNL process was well adapted to the recyclates used, barrier properties are studied (see Figure 31).

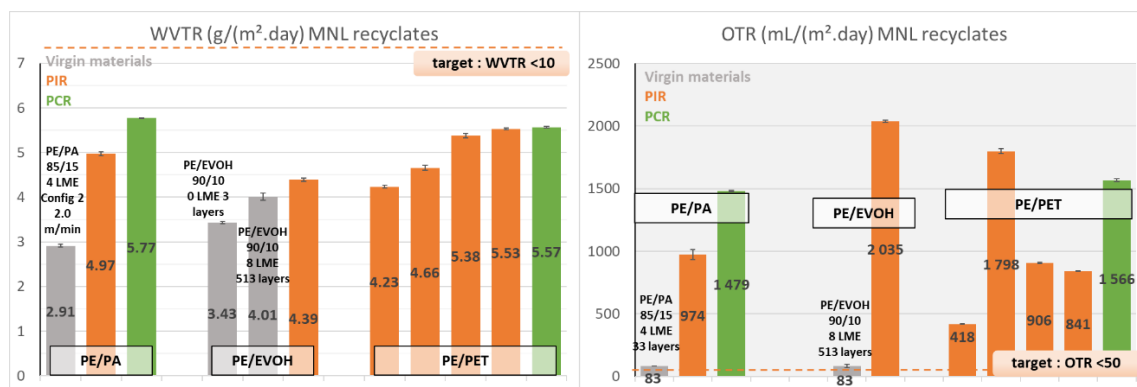


Figure 31: Water-vapor transmission rate (g/(m².day)) and Oxygen Transmission Rate (mL/(m².day)) for recyclates MNL structures. The grey bars were taken from the previous trials with virgin materials

The WVTR are, as usual, all satisfying, even though an increase is observed between virgin material and PIR, and then PIR and PCR, indicating the degradation of the barrier material during its lifetime and its recycling. This might also be due to a higher level of impurities in PCR, such as other polymers (for example PP).

The OTR of the recyclates are not satisfying. But one can see that they are better when EVOH or PET are involved. Indeed, this material present a higher oxygen barrier intrinsic property than PA. The PIR present lower OTR values than the PCR: the materials are less degraded, given they did not age during their lifetime.

Some adjustments are needed to achieve satisfying barrier properties, but due to the limited availability of recycled materials, and a breakage of the equipment, the investigation could not go further.

d. Conclusion for recyclates films production

To process the recyclates, IPC diluted the batches received from VTT with two specific targets in mind:

- Recycled content of 10wt.% for MNL structures, and 12wt.% for METEOR® blends, to follow the target set by PPWR at that time.
- Barrier content of 5wt.% for EVOH, 15wt.% for PA and 10wt.% for PET with Post-Industrial Recyclates (PIR). This target doubled when utilizing Post-Consumer Recyclates (PCR) blended with METEOR®.

Regarding mechanical properties, for PE/PA and PE/EVOH blends, the elongation at break and at yield in the transverse direction decrease when PIR is processed, further decreasing with PCR. Meanwhile, tensile modulus and stresses (at break

and at yield) increase. This results in a stiffer final film, indicating probable material degradation over its lifetime. For PE/PET blends, all properties decrease when recyclates are introduced, rendering the final film more fragile, possibly due to poor compatibility between PET and PE.

For the MNL structures, in terms of mechanical properties, a decrease was observed in PE/PA and PE/EVOH structures when PIR or PCR were processed. However, for PE/PET structures, since IPC lacked a virgin reference, it can only be stated that properties were comparable between PIR and PCR.

While the barrier properties were satisfactory for water-vapor transmission rates, they exceeded the industry-set target for oxygen transmission rates. Although bi-axial stretching attempted on METEOR® blends to improve the barrier properties, it fell short of the target set by industrial partners.

Conclusion and next steps

For all the virgin blends tested, METEOR® proves its efficiency, with the excellent elongation at break shown in the Figure 13.

Indeed, the properties reach the level of a twin-screw extruder (standard extrusion in the graph), and thus using single-screw extruder coupled with METEOR® process.

Extensive rheological studies indicate that the interfacial energy increases when METEOR® is applied to virgin PE+PA and virgin PE+EVOH blends. This suggests that the dispersion is finer, or that some interactions are created at the interface. Therefore, the METEOR® process could potentially reduce the amount of compatibilizer needed in these blends. If IPC did not manage to prove that METEOR® coupled with a single-screw extruder degrades less the material than a twin-screw extruder, despite the METEOR® process having a high residence time (30-40 minutes), materials are not more degraded compared to those processed with a twin-screw extruder due to the lower shear during the process.

An extensive study was conducted on virgin PE/PA structures, focusing on the influences of LME number and position. A notable formulation (PE/PA 85/15) was established, depicted in Figure 21 and Figure 26.

It appears that introducing 6 to 8 LMEs (129 to 513 layers) led to the breaking of the multi-nanolayered structure: elongation at break decreased while OTR increased. Optimal results were observed when LMEs were positioned at the beginning of the process (Configuration 1), possibly due to viscosity ratio proximity to unity at that process location.

When recycled materials are processed (Post-Industrial Recyclates (PIR) and Post-Consumer Recyclates (PCR)), with MNL or METEOR® processes, a loss in mechanical properties and some degradation are observed.

For all blends, virgin and recycled, the Water Vapor Transmission Rate is satisfactory, but the Oxygen Transmission Rate is higher than the industrial target. Unfortunately, attempts to improve these properties by stretching the METEOR® films at VTT were unsuccessful. A deeper study of the stretching parameters would be beneficial but was not within the scope of the CIMPA project.

In conclusion, the elongation flow mixer METEOR® presents a promising opportunity for blending recycled materials. The controlled degradation in the METEOR® module by reducing the residence time will allow greater flexibility in mixing parameters. Since recyclates are already degraded from their first life, this process might enable effective mixing at low shear rates, thereby protecting these highly sensitive materials.

On the other hand, the potential of layer multiplication to achieve nanolayer structures holds promise for multilayer film recycling. However, due to equipment malfunction and limited availability of recycled materials, not all parameters, such as layer number or LME position, could be fully explored when processing recyclates.

Optical	Nominal Value (English)	Nominal Value (SI)	Test Method
Gloss (20°, 1.97 mil (50.0 µm))	60	60	ASTM D2457
Haze (1.97 mil (50.0 µm))	8.00 %	8.00 %	ASTM D1003

Extrusion	Nominal Value (English)	Nominal Value (SI)
Melt Temperature	320 to 347 °F	160 to 175 °C

Extrusion Notes
Film Blow-Up ratio 1:2.5

Notes

These are typical properties only and are not to be construed as specifications. Users should confirm results by their own tests.

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LDPE SABIC NC1013

SABIC[®] LDPE NC1013

LOW DENSITY POLYETHYLENE

DESCRIPTION

SABIC[®] LDPE NC1013 is a general purpose grade, containing anti block and slip agent. This grade offers good optical properties and a very good draw down ability.

Application

SABIC[®] LDPE NC1013 is typically used for packaging films for food and industrial goods and for lamination films. This grade is typically used when high draw down is required.

SABIC[®] LDPE NC1013 can typically be used for food applications due to very low migration levels.

This product is not intended for and must not be used in any pharmaceutical/medical applications.

TYPICAL PROPERTY VALUES

Revision 20200923

PROPERTIES	TYPICAL VALUES	UNITS	TEST METHODS
POLYMER PROPERTIES			
Melt Flow Rate			
at 190 °C and 2.16 kg	2.5	dg/min	ISO 1133
Density	921	kg/m ³	ASTM D1505
FORMULATION			
Slip agent	☑	-	-
Anti block agent	☑	-	-

HEALTH, SAFETY AND FOOD CONTACT REGULATIONS

Detailed information is provided in the relevant Material Safety Datasheet and or Standard Food Declaration, available on the Internet (www.SABIC.com). Additional specific information can be requested via your local Sales Office.

QUALITY

SABIC is fully certified in accordance with the internationally accepted quality standard ISO 9001.

STORAGE AND HANDLING

Polyethylenes resins (in pelletised or powder form) should be stored in such a way that it prevents exposure to direct sunlight and/or heat, as this may lead to quality deterioration. The storage location should also be dry, dust free and the ambient temperature should not exceed 50 °C. Not complying with these precautionary measures can lead to a degradation of the product which can result in colour changes, bad smell and inadequate product performance. It is also advisable to process polyethylene resins (in pelletised or powder form) within 6 months after delivery, this because also excessive aging of polyethylene can lead to a deterioration in quality.

ENVIRONMENT AND RECYCLING

The environmental aspects of any packaging material do not only imply waste issues but have to be considered in relation with the use of natural resources, the preservations of foodstuffs, etc. SABIC considers polyethylene to be an environmentally efficient packaging material. Its low specific energy consumption and insignificant emissions to air and water designate polyethylene as the ecological alternative in comparison with the traditional packaging materials. Recycling of packaging materials is supported by SABIC whenever ecological and social benefits are achieved and where a social infrastructure for selective collecting and sorting of packaging is fostered. Whenever 'thermal' recycling of packaging (i.e. incineration with energy recovery) is carried out, polyethylene -with its fairly simple molecular structure and low amount of additives- is considered to be a trouble-free fuel.

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CHEMISTRY THAT MATTERS™

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LDPE REPSOL ALCUDIA 2320FG



LDPE REPSOL ALCUDIA 2320FG EXP1

The grade REPSOL ALCUDIA 2320FG EXP1 is a low density polyethylene grade produced by high pressure tubular technology, suitable for thin blown film or cast film applications. This material offers easy processability and good balance of mechanical and optical properties. It contains slip (550 ppm) (erucamide), antiblock (1125 ppm) and thermal stabiliser additives.

Applications

- Shrink film.
- Automatic packaging
- Lamination.
- Film coextrusion with good optical properties

Recommended melt temperature range from 160 to 200°C. Processing conditions should be optimised for each production line.

PROPERTIES	VALUE	UNIT	TEST METHOD
General			
Melt Flow Index (190°C, 2.16kg)	2.0	g/10 min	ISO 1133
Density at 23°C	923	kg/m3	ISO 1183
Mechanical			
Dart drop (F50)	100	g	ISO 7765-1
Tear resistance (Elmendorf) (MD/TD)	215 / 230	cN	ISO 6383-2
Tensile stress at break (MD/TD)	19 / 15	MPa	ISO 527-3
Elongation at break (MD/TD)	400 / 250	%	ISO 527-3
Coefficient of friction (Dynamic)	<0.1	-	ISO 8295
Gloss (45°)	72	-	ASTM D-2457
Gloss (60°)	105	-	ASTM D-2457
Haze	6.0	%	ASTM D-1003
Others			
Vicat softening temperature (load 10 N)	95	°C	ISO 306
* Provisional values to be confirmed upon statistical data (1) 30 µm thickness film, blow up ratio 2.5:1			FIL-18032

The grade REPSOL ALCUDIA 2320FG EXP1 complies with the European Directives regarding materials intended for contact with foodstuffs. The product mentioned herein is not intended to be used for medical, pharmaceutical or healthcare applications and we do not support their use for such applications. For further information, please contact our Technical Service and Development Laboratory or our Customer Care Service.

Storage

The grade REPSOL ALCUDIA 2320FG EXP1 should be stored in a dry atmosphere, on a paved, drained and not flooded area, at temperatures under 50°C and protected from UV radiation. Storage under inappropriate conditions could initiate degradation processes or undesired migration of additives included in its formulation which may have a negative influence on the processability and properties of the transformed product.

April 2018

This information is offered in good faith and meant only as a guide. The transformer or user will be, in each case, responsible for the processing conditions and the final use of the product. Freedom under patents, copyright and registered designs cannot be assumed.

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LDPE BASELL LUPOLEN 2426H

Technical Data Sheet

Lupolen 2426H

Low Density Polyethylene



Product Description

Lupolen 2426 H is an additivated, low density polyethylene. It contains slip and anti-blocking agent. It is characterized by a good balance between processability and mechanical properties. Films made from Lupolen 2426 H exhibit good optical properties. It is delivered in pellet form.

This product is not intended for use in medical and pharmaceutical applications.

Regulatory Status

For regulatory compliance information, see [Lupolen 2426H Product Stewardship Bulletin \(PSB\) and Safety Data Sheet \(SDS\)](#).

Status	Commercial: Active
Availability	Africa-Middle East; Asia-Pacific; Europe
Application	Bags & Pouches; Food Packaging Film; Hygiene Film; Liner Film; Shrink Film
Market	Flexible Packaging
Processing Method	Blown Film; Cast Film
Attribute	Good Heat Seal; Good Optical Properties; Good Processability; Low Friction; Unspecified Antiblocking; Unspecified Slip

Typical Properties	Nominal Value	Units	Test Method
Physical			
Melt Flow Rate, (190 °C/2.16 kg)	1.9	g/10 min	ISO 1133-1
Density	0.924	g/cm ³	ISO 1183-1
Mechanical			
Tensile Modulus	260	MPa	ISO 527-1, -2
Tensile Stress at Yield	11	MPa	ISO 527-1, -2
Film			
Dart Drop Impact Strength, F50	110	g	ASTM D1709
Tensile Strength			
MD	25	MPa	ISO 527-1, -3
TD	21	MPa	ISO 527-1, -3
Tensile Strain at Break			
MD	250	%	ISO 527-1, -3
TD	600	%	ISO 527-1, -3
Coefficient of Friction	<0.2		ISO 8295
Impact			
Failure Energy	4	J/mm	DIN 53373
Thermal			
Vicat Softening Temperature, (A/50 N)	94	°C	ISO 306
Peak Melting Point	111	°C	ISO 11357-3
Optical			
Haze, (50 µm)	<8	%	ASTM D1003

Gloss		
(20")	>50	ASTM D2457
(60")	>100	ASTM D2457
Additive		
Slip, Erucamide	500 ppm	LYB Method
Antiblock, Natural Silica	1000 ppm	ISO 3451-1
Additional Information		
Test Specimen	Film	
Film properties tested using 50 µm thickness blown film extruded at a melt temperature of 180°C and a blow-up ratio of 2.5:1.		
Processing Parameters		
Extrusion Temperature	160-200 °C	

Notes

These are typical property values not to be construed as specification limits.

Processing Techniques

Users should determine the conditions necessary to obtain optimum product properties and suitability of the product for the intended application.

In cases where higher temperatures are required, please contact your appropriate technical contact for support.

Further Information

Health and Safety:

The resin is manufactured to the highest standards, but special requirements apply to certain applications such as food end-use contact and direct medical use. For specific information on regulatory compliance contact your local representative.

Workers should be protected from the possibility of skin or eye contact with molten polymer. Safety glasses are suggested as a minimal precaution to prevent mechanical or thermal injury to the eyes.

Molten polymer may be degraded if it is exposed to air during any of the processing and off-line operations. The products of degradation may have an unpleasant odor. In higher concentrations they may cause irritation of the mucus membranes. Fabrication areas should be ventilated to carry away fumes or vapours. Legislation on the control of emissions and pollution prevention should be observed.

The resin will burn when supplied with excess heat and oxygen. It should be handled and stored away from contact with direct flames and/or ignition sources. While burning, the resin contributes high heat and may generate a dense black smoke.

Recycled resins may have previously been used as packaging for, or may have otherwise been in contact with, hazardous goods. Converters are responsible for taking all necessary precautions to ensure that recycled resins are safe for continued use.

For further information about safety in handling and processing please refer to the Safety Data Sheet.

Conveying:

Conveying equipment should be designed to prevent production and accumulation of fines and dust particles that are contained in polymer resins. These particles can under certain conditions pose an explosion hazard. Conveying systems should be grounded, equipped with adequate filters and regularly inspected for leaks.

Storage:

The resin is packed in 25 kg bags, octabins or bulk containers protecting it from contamination. If it is stored under certain conditions, i. e. if there are large fluctuations in ambient temperature and the atmospheric humidity is high, moisture may condense inside the packaging. Under these circumstances, it is recommended to dry the resin before use. Unfavorable storage conditions may also intensify the resin's slight characteristic odor.

Resin should be protected from direct sunlight, temperatures above 40°C and high atmospheric humidity during storage. Higher storage temperatures may reduce the storage time.

The information submitted is based on our current knowledge and experience. In view of the many factors that may affect processing and application, these data do not relieve processors of the responsibility of carrying out their own tests and experiments; neither do they imply any legally binding assurance of certain properties or of suitability for a specific purpose. This information does not remove the obligation of the customer to inspect the material on arrival and notify us of any faults immediately. It is the responsibility of those to whom we supply our products to ensure that any proprietary rights and existing laws and legislation are observed.

Company Information

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EVOH KURARAY EVAL J171B




Technical Data Sheet

EVAL™ J171B

32 mol% Ethylene-Vinyl Alcohol Copolymer

Typical Properties	Unit	Test Method	Metric/(English)
MFR	g/10min	ISO1133 (190°C)	1.7
Density	10 ³ kg/m ³	ISO1183-3	1.18

Thermal Properties	Unit	Test Method	Metric/(English)
Melting Temperature	°C (°F)	ISO 11357	183 (361)
Crystallization Temperature	°C (°F)	ISO 11357	158 (316)
Glass Transition Point	°C (°F)	ISO 11357	60 (140)
Vicat Softening Point	°C (°F)	ISO 306	162 (324)

Mechanical Properties	Unit	Test Method	Metric/(English)
Tensile stress at break	MPa (psi)	ISO 527	30 (4,400)
Elongation at break	%	ISO 527	15
Young's Modulus	GPa (psi)	ISO 527	4.3 (620,000)
Flexural Modulus	GPa (psi)	ISO 178	4.2 (610,000)
Charpy Impact Strength	kJ/m ² (ft.lbf/in ²)	ISO 179-1	8 (3.8)
Rockwell Hardness	HRM	ISO 2039-2	88

Barrier Properties (cast film)	Unit	Test Method	Metric/(English)
Oxygen Transmission Rate	cm ³ .20µm/m ² .day.atm (cm ³ .mil/100in ² .day.atm)	ISO 14663-2 Annex C	0.4 (0.02)

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PE-g-MAH compatibilizer SK OREVAC 18300



Technical Data Sheet
Date Prepared: March 2020

OREVAC[®] 18300

OREVAC[®] 18300 is a maleic anhydride grafted linear low-density polyethylene.

- OREVAC[®] 18300 has been designed to develop a reliable bonding strength in coextrusion processes between polyethylene or ethylene copolymers and different materials among which polyamides and EVOH.
- OREVAC[®] 18300 is recommended for cast or blown film coextrusion.

Typical Properties

	Test Method	Unit	Typical Value
Melt index (190°C/2.16kg)	ISO 1133 / ASTM D1238	g/10min.	2.3
Melting Point	ISO 11357-3	°C	120
Vicat Softening Temperature (10N) ¹	ISO 306 / ASTM D1525	°C	85
Density	ISO 1183 / ASTM D1505	g/cm ³	0.91

¹: On compression molded samples.

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Processing

OREVAC® 18300 is to be processed like a standard polyethylene resin. Typical extrusion temperature settings could be:

Zone 1	Zone 2	Zone 3	Zone 4	Exit	Fittings-Channels	Die
190-120°C	200°C	200-210°C	210-220°C	220-230°C	220-240°C	220-240°C

Final profile and settings will depend on the line and the multi-layer structure being run.

Storage, Handling & Safety

OREVAC® 18300 should be stored in dry conditions protected from UV-light. Improper storage conditions may cause degradation and have consequences on physical properties of the product.

Safety data sheet as well as information on handling and storage of the OREVAC® 18300 is available upon request to your SK Functional Polymer representative.

Shelf Life

Three years from the date of delivery, in unopened packaging. For any use above this limit, please refer to our technical services.

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PA6/6.6 UBE NYLON 5033B



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UBE NYLON 5033B

Technical Product Information: CAST - PA monolayer film properties

UBE NYLON 5033B is a basic, high viscosity Polyamide 6/6.6 copolymer. The grade is suitable for a wide range of applications. This material has the following features:

- Excellent processability
- Excellent mechanical properties
- Excellent puncture and pin hole resistance
- Deep drawability

Basic Properties ⁽¹⁾	Method	Unit	Value
Polymer	-	-	PA6/6.6
Melting Point	ISO 11357	°C	196
Relative Viscosity (96% H ₂ SO ₄)	JIS K6810	-	4,05
Density	DIN 53479	g/cm ³	1,14

Mechanical Properties ⁽²⁾	Method	Unit	Value
Tensile strength at yield	ISO 527-3	MPa	22 - 26
Tensile strength at break		MPa	105 - 125
Tensile elongation at break		%	430 - 450
Tensile modulus		MPa	450 - 550
Tear resistance	ASTM D 1922	N	2,1 - 2,3
Puncture energy	JAS P-1019	mJ	37 - 39
Puncture deformation		mm	10,0 - 11,0
Spencer impact resistance	ASTM D 3420	mJ	850 - 950
Flex crack (23°C, 1000 cycles / 5°C, 100 cycles)	Mil B-131C	Holes/0,04m ²	< 20 / < 5

Optical Properties ⁽³⁾	Method	Unit	Value
Haze	ASTM D 1003	%	< 0,5
Gloss	ASTM D 523	%	145 - 165

Gas Barrier ⁽⁴⁾	Method	Unit	Value
Oxygen (T=23°C, RH=0%)	ASTM D 3985	ml/m ² .day	33 - 35
Water Vapour (T=40°C, RH=90%)	JIS Z-0208	g/m ² .day	180 - 200

UBE

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Slip Properties ⁽¹⁾	Method	Unit	Value
Coefficient of friction (static)	ASTM D 1894	-	>1
Coefficient of friction (dynamic)		-	>1

Regulation	Method	Unit	Value
FDA / EC	-	-	I&D / I&D

I: Approved for indirect food contact D: Approved for direct food contact

(1) Measured on base resin
 (2) All tests carried out with a monolayer cast line, Die width = 350 mm (Coat hanger type)
 Structure: PA monolayer
 Layer thickness distribution: PA = 50 µm
 Cooling conditions: Chiller roll temperature = 20 °C
 Sample conditioning and testing conditions: T = 23°C, RH =50%

Processing conditions

	Extruder					Adaptor	Die
	Hopper	Zone 1	Zone 2	Zone 3	Zone 4		
Temperature (°C)	40 - 120	180 - 200	200 - 220	210 - 230	220 - 240	230 - 250	230 - 250

Drying conditions

UBE NYLON is supplied dry (moisture content < 0,1%) and packed in high barrier films. However, as polyamide is a hygroscopic material, the user should take a special care of the possible moisture absorption once the bag or liner box has been opened. In case of moisture absorption, the material should be dried with dehumidified air at 80°C for more than 4 hours.

Storage

Well-sealed packages could be stored in cool and dry conditions over long periods of time. Protect the packages from heat and direct sunlight to prevent possible damages.

Health & environmental data

Please refer to the corresponding **UBE NYLON** grade SDS.

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TDS VERSION (01/01/2022)

PET RAMAPET N1S



Technical Datasheet RAMAPET N1(S)

RAMAPET N1(S) is a general purpose, non-reheat PET resin for bottles, film, thermoformed containers and other transparent applications. It has good optical properties; the mechanical properties are representative for a PET resin with IV of 0.80. RAMAPET thermoplastic polyesters are condensation polymers produced by a continuous melt-phase polymerization process followed by a solid-state polymerization process.

RAMAPET N1(S) has a spherical (round) pellet shape.

Applications and use:

- Carbonated soft drinks
- Water bottles
- Juices, beer, wine and spirits
- Household products
- Food packaging
- APET sheet and thermoformed containers
- Pharmaceutical and medical applications (European and US Pharmacopoeia approved)

Sales specification:

Property	Unit	Test method	RAMAPET N1(S) value
Intrinsic Viscosity	dl/g	IVP-01-PL-04P	0.80 ± 0.02
Acetaldehyde	ppm	IVP-01-PL-06P	Max 1
Colour L*	-	IVP-01-PL-01P	Min 82
Colour a*	-	IVP-01-PL-01P	-2.5 to 0
Colour b*	-	IVP-01-PL-01P	-3 to 0
Pellet weight	Pieces/gram	IVP-01-PL-05P	65 ± 5
Fines	Wt%	IVP-01-PL-08P	Max 0.05

The specification describes a grade of clear co-polyester in the form of pellets which meets all of the requirements when tested as directed by the referenced methods.

Product shipments are not tested for acetaldehyde. Samples that are representative of product are tested in a monitor program to ensure that the process capability for acetaldehyde in the polymer is less than the specification limit.

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Typical properties:

Property ^a	Test ^b Method	Typical Value, Units ^a
Pellet Properties		
Crystalline Density Solid Stated pellets	D 1505	1.39 - 1.4 g/cm ³
Bulk Density Poured	D 1895	850 ± 30 kg/m ³
Bulk Density Vibrated	D 1895	920 ± 30 kg/m ³
Melt Density @ 285°C	D 1238 (Note A- Table 2)	1.2 g/cm ³
Crystalline Peak Melting Point (T _m) ^d	D 3418	245 ± 5°C
Glass transition point (T _g) (dry)	D 3418	78 ± 2°C
Heat of Fusion ^e	E 793	56 kJ/kg (13 cal/g)
% crystallinity solid Stated pellets		50 ±5 %
Pellet Shape		Spherical
Moisture content pellets		<0.3%
^a Unless noted otherwise, all tests are run at 23°C (73°F) and 50% relative humidity. ^b Unless noted otherwise, the test method is ASTM. ^c Units are in SI or US customary units. ^d Determined by DSC on the second heating cycle. ^e Determined by DSC on the first heating cycle.		
Properties reported here are typical of average lots. Indorama makes no representation that the material in any particular shipment will conform exactly to the values given.		

Food Contact Position:

RAMAPET N1(S) is a copolymer made from terephthalic acid (PTA), isophthalic acid (IPA) and monoethylene glycol (MEG) complies with the requirements of the European Legislation (EU) No 10/2011 for plastics used in contact with food.

This product is produced under good manufacturing practices in compliance with EU Regulation 2023/2006 and is intended for use to manufacture articles in compliance with the general requirements (in Article 3) of Regulation (EC) 1935/2004.

Under the regulations administered by the U.S. Food and Drug Administration (FDA),

RAMAPET N1(S) may lawfully be used as articles or components of articles intended for the use in contact with food subject to provisions of CFR 177.1630 and 21 CFR 174.5.

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Processing:

PET is hygroscopic, and since moisture adversely affects the IV during melt processing of the polymer, it must be dried prior to molding. Un-dried PET pellets can contain up to 0.2 - 0.3% (2000 - 3000 ppm) of moisture. After drying the moisture content typically is below 50 ppm.

In the drying process, there are four variables that should be considered:

- Drying temperature 150 - 170°C
- Residence time in the dryer silo 6 hours
- Dew Point of Drying Air < - 20°C
- Airflow Rate 2.2 m³/h/kg PET

PET has a relatively high melting point of around 245°C. Processing PET pellets on an extruder or injection molding machine requires temperature setting of 30 – 40°C above the melting point.

A typical temperature profile (from hopper to nozzle) is: 295 – 285 – 280°C and hot-runners can be set at 275 to 285°C. Temperatures above 300°C should be avoided.

The melt residence time should be limited to < 2 minutes to avoid discoloration. During machine stops the temperature setting should be reduced to avoid the material to degrade or decompose.

To find the latest version of this document visit our website: www.indoramapolymers.eu

Revision date January 2019

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SEBS-g-MAH compatibilizer KRATON FG1901 G

KRATON

K0127 North America 10/9/2019		KRATON™ FG1901 G Polymer		Data Document	
Identifier : K127DDm19U					
Description					
Kraton FG1901 G is a clear, linear triblock copolymer based on styrene and ethylene/butylene with a polystyrene content of 30%. It is supplied from North America in the physical form identified below.					
<ul style="list-style-type: none"> • FG1901 GT - supplied as a dusted pellet 					
Kraton FG1901 G is used as a modifier of bitumen and polymers. It is also suitable as an ingredient in formulating compounds for footwear applications and may be used in formulating adhesives, sealants, and coatings.					
Sales Specifications					
Property	Test Method	Units	Sales Specification Range	Notes	
Melt Flow, 230C/5000g	ASTM D1238	g/10 min	14 TO 28		
Maleic anhydride, Bound	BAM 1026	%w	1.4 TO 2.0		
Water	BAM 1024	ppmw	≤ 500	a	
a At time of packaging					
Typical Properties (These are typical values and may not routinely be measured on finished product)					
Property	Test Method	Units	Typical Value	Notes	
Melt Index 230C, 5 kg	ASTM D 1238	gms/10 min.	22		
Tensile Strength	ASTM D 412	psi	5000	a	
Specific Gravity	ASTM D 792	g/cc	0.91		
Styrene / Rubber ratio	n/a		30/70		
Hardness, Shore A	ASTM D 2240	Shore A (10 sec)	71	b	
Elongation at Break	ASTM D 412	%	500	a	
Solution Viscosity	BAM 922	cps	1000	c	
a Typical properties determined on film cast from toluene solution.					
b Typical values on polymer compression molded at 300Å°F.					
c 25%w toluene solution at 25Å°C, measured at time of packaging.					
Packaging					
Kraton FG1901 G is packaged in sealed, foil-lined bags and typically does not require drying before melt process. However, if the bag is left open for an extended period of time, the product may absorb moisture and should be dried under vacuum at 80 degree C for a minimum of six hours before processing.					
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End Use Requirements

If the finished article is intended for use in food contact and packaging applications, toys, or human contact areas, manufacturers of the final product should observe all relevant regulations. Some of these regulations require tests to be carried out on the final product, e.g. migration. These are the responsibility of the final product manufacturer. Information on the food packaging clearances of individual products is available from Kraton Polymers.

Safety and Handling Precautions

Read the Safety Data Sheet carefully and thoroughly before beginning any work. Additional information relating to the health, safety, storage, handling and processing of Kraton Polymers products can be found in "Health and Safety Aspects of Kraton D and Kraton G Polymers" (Document K0155), available from your local Sales Representative or the company website. Kraton Polymers also recommends that customers or users consult other sources of safety information, for example, the current edition of the "Code of Practice on the Toxicity and Safe Handling of Rubber Chemicals," British Rubber Manufacturers Association Limited. Kraton Polymers products and compounds can accumulate electrostatic charges when rubbed, chafed or abraded. Processing and storage equipment for use with Kraton Polymers products should provide a means of dissipating any charges that may develop.

When processing Kraton Polymers products, maintain a fire watch if the material reaches 225Å°C (437Å°F) for Kraton IR and Kraton D (polymers and compounds), and 280Å°C (536Å°F) for Kraton G (polymers and compounds). The temperatures listed above are indicated only for safety reasons (risk of fire and product degradation) and are not necessarily recommended for processing. Degradation of the polymer (polymer breakdown) will start at lower temperatures depending on the specific processing conditions. Therefore, operating below these temperatures does not guarantee the absence of product degradation.

Kraton Polymers products (the neat resin or the base product) are high molecular weight polymers which are non-toxic and biologically inactive.

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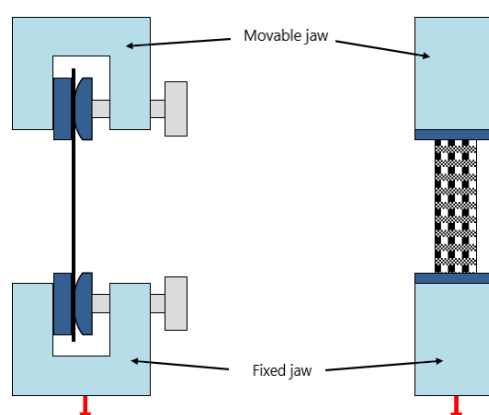
For Further Information

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Annex 2: Tests methods

Tensile properties

The tensile tests were performed with an electromechanical testing machine (ZwickRoell, Germany), following the ISO 527-3/2 at IPC. The specimens used had a rectangular shape with dimensions of 15 mm in width, and 150 mm in length. As shown in the scheme below, each sample was clamped into the machine with one end held at a fixed position and the other end displaced at a constant rate of 100 mm/min, using a load cell of 1 kilo Newton (kN). Data was collected with a chart that monitored the force as a function of the displacement.



This experiment gives the tensile modulus, the elongation and stress at yield and finally the elongation and stress at break for every sample.

Barrier properties: WVTR

Water Vapour Transmission Rate (WVTR) were performed at IPC according to the standard ASTM F1249. Samples were studied using the PERMATRAN® model 3/38 H (Mocon, USA).



Distilled water is poured into a glass, leaving a small air gap between the sample and the water. The container is then sealed, except for the test sample, to prevent vapour loss. The instrument is initially weighed, then at regular intervals before the effects become linear. The experiment must be carefully monitored to ensure that any weight loss is due to water vapour passing through the sample.

Therefore, the sample cell is constantly flushed by a stream of 6.0 purity nitrogen. The water vapour transmitted through the film is analysed by the specific IR (Infra-Red) detector instrument. The results are expressed in (mL)/(m².day).

The following testing conditions were applied:

- The surface area analysed: 50 cm²
- Relative humidity: 90%
- Temperature: 38°C

Barrier properties: OTR

Oxygen Transmission Tests were performed at IPC according to the standard ASTM D3985. Samples were studied using the OX-TRAN® model 2/28H (Mocon, USA).



When gaseous oxygen enters the nitrogen carrier gas through the film, it is transported to the coulometric detector, where it generates an electric current whose magnitude is proportional to the quantity of oxygen entering the detector per unit of time. Gaseous oxygen works as a “test gas”, and gaseous nitrogen is used as a “carrier gas”. Since there is a concentration difference between the two sides of the sample, the concentration of gaseous oxygen in the upper chamber is higher than that in the lower chamber. During the conduction process, gaseous oxygen is transmitted from the upper chamber to the lower chamber through the sample.

Temperature and humidity are critical parameters that affect the final test data. Therefore, the cell is conditioned at 23°C. The results are expressed in (ml)/(m².day).

The following testing conditions were applied:

- The surface area analysed: 50 cm²
- Test gas used: 100% O₂
- Carrier gas used: a mixture of H₂/N₂ (5/95)

Rheological measurements

The rheological studies were performed using a stress-controlled AR2000ex rheometer (TA instruments) with a parallel plate geometry of 25 mm in diameter. In case of the neat polymers, sample's disks were prepared by compression moulding process at 210°C for 3 minutes. For the films, samples were cut out and put directly on the rheometer without additional preparation. Several films were tacked to obtain a suitable thickness (16 layers).

In a first step, dynamic strain sweep tests were made to choose a deformation at which the samples remain in the linear viscoelastic region, and where the thermal stability was validated. The strain chosen must be in the plateau of the curve, indicating that the measures are independent of the strain applied.

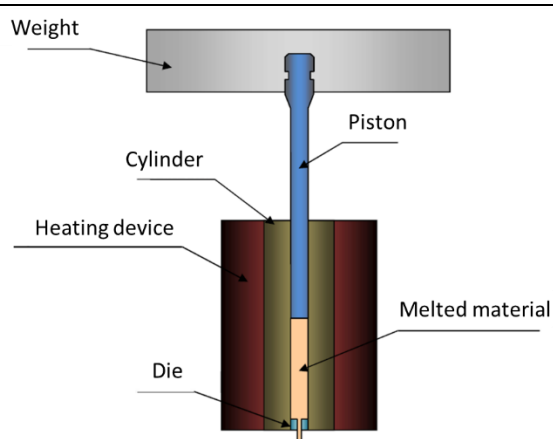
Then, several tests were made:

- Dynamic time sweep (DTS) tests: the sample is put under fixed strain (depending on the sample 3% for PE/EVOH and PE/PA blends, and 1% for PE/PET blends) and frequency (1Hz) and its response is recorded over time (3600s). The cross over between the G' (storage modulus) and G'' (viscous modulus) is noted, as it is an indication of the thermal stability of the film. When G' modulus becomes higher than the G'' modulus, the material starts to degrade.
- Dynamic frequency sweep test (DFS): the sample is put under fixed strain (the same as for the DTS) and temperature, and the frequency varies from 0.05rad/s to 628 rad/s. The modulus G' and G'' as well as the intrinsic viscosity are monitored: their evolution allows to draw conclusions on the viscoelastic behaviour of the blends, and thus, the quality of the dispersion and interfacial interactions. The DFS was used to determine the viscosity ratio of the virgin materials (average between 5 and 10rad/s at extrusion temperature).
- Stress Relaxation (SR) tests: the sample is put under a strain (the same as for DTS) at a constant frequency of 1Hz, and the modulus $G(t)$ is recorded during 3600s. The difference with DTS is that the strain is not maintained over time, therefore the way the material relaxes over time is observed.

Melt Flow Index

The Melt Flow Index (MFI) is measured using a GOTTERT mi40 equipment, according to the NF EN ISO 1133-1, method B standard.

This test measures the quantity of material flowing through a 2mm diameter die under a weight and a temperature depending on the material tested.

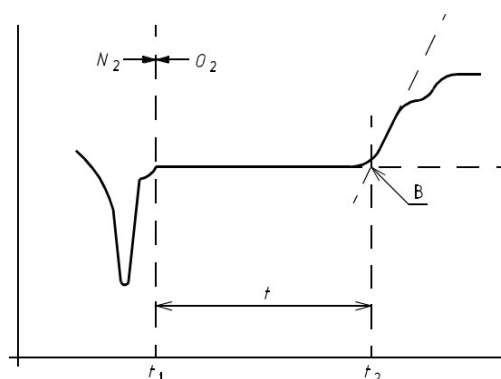


Oxidation Induction Time

The Oxidation induction time was measured using a DSC Q100 TA Instrument equipment. The test measures the time within the sample is stable to oxidation when it is maintained under a specific temperature in a flow containing oxygen.

The oxidation is monitored by measuring the heat flow difference between the sample cell and a reference cell and by recording it over time.

The material is put in the equipment under N_2 and then heated up to $200^\circ C$. Then the atmosphere is changed from N_2 to Air (N_2/O_2 80/20) at t_1 and the Oxidation induction time is the time t_2 when the material changes its mass (increase or loss). It is an indication of the stability overheat and oxidation of the material.



FTIR analysis

Infra-Red analysis was performed using a FTIR iS10 Thermo Nicolet, with the SMART ITX ATR accessory and the diamond crystal.

The spectra was obtained with absorbance method, 32 scans and a resolution of 4 cm^{-1} .

The wave numbers went from 525 cm^{-1} to 4000 cm^{-1} .

The film samples analysis was compared to neat polymer.

Optical microscope analysis

The equipment used is a Keyence VHX7100.

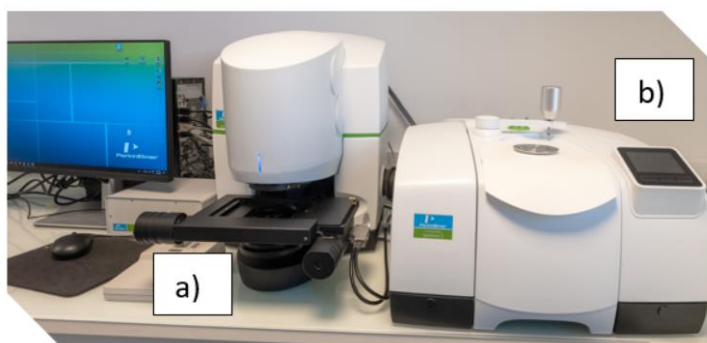
The samples were cut using a scalpel and then observed through the film thickness.

The magnification used were from x300 to x700.

Optical microscopy coupled with FTIR

The study of the multilayer film's morphology was performed using optical microscopy.

These observations were studied at IPC using the Spotlight 400 FT-IR Imaging System, as shown in the figure below. This system is a binocular magnifier linked to image processing software, M80 ergonomic (LEICA, Germany). Additionally, the microscope is linked to an Infrared micro spectrometry Spectrum 3_Spotlight 400 (PERKIN ELMER, USA), which allow us to measure the thickness and identify the materials at the same time.



Spotlight 400 FT-IR Imaging System at IPC

Annex 3: Bi-axial stretching at VTT

The biaxial stretching took place at VTT using a Brückner KARO IV laboratory sheet stretching equipment.

METEOR® blends

Due to resources available, a priority needed to be set because all samples could not be tested. The list of the samples tested is in the following table:

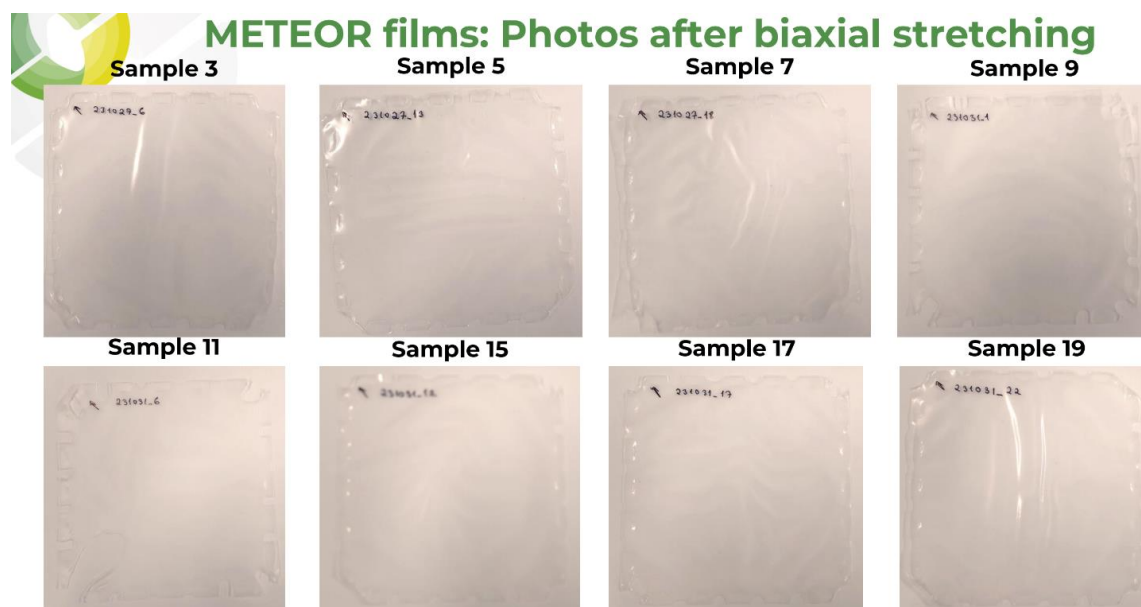
	Formulation	Extruder	Sample Code	Cast thickness [um]
Round 1	PE 100%	TS	PE TS	119 ± 10
	PE+EVOH 95+5%	TS	PE/EVOH 95/5 TS	111 ± 18
	PE+EVOH+OREVAC 87+5+8%	TS	PE/EVOH/OREVAC 87/5/8 TS	113 ± 3
	PE+EVOH+OREVAC 80+7+13%	TS	PE/EVOH/OREVAC 80/7/13 TS	108 ± 6
	PE 100%	TS+METEOR	PE METEOR	73 ± 3
	PE+EVOH 95+5%	TS+METEOR	PE/EVOH 95/5 METEOR	95 ± 9
	PE+EVOH+OREVAC 87+5+8%	TS+METEOR	PE/EVOH/OREVAC 87/5/8 METEOR	106 ± 10
	PE+EVOH+OREVAC 80+7+13%	TS+METEOR	PE/EVOH/OREVAC 80/7/13 METEOR	115 ± 9
Round 2	PE+EVOH 95+5%	SS+METEOR	Sample 3	111 ± 8
	PE+EVOH+OREVAC 87+5+8%	TS+METEOR	Sample 5	109 ± 14
	PE+EVOH+OREVAC 87+5+8%	SS+METEOR	Sample 7	95 ± 7
	PE+PA+OREVAC 80+15+5%	SS+METEOR	Sample 9	123 ± 8
	PE+PA+OREVAC 70+22,5+7,5%	SS+METEOR	Sample 11	122 ± 16
	PE+PET 90+10%	SS+METEOR	Sample 15	110 ± 9
	PE+PET+KRATON 94+5+1%	SS+METEOR	Sample 17	112 ± 12
	PE+PET 95+5%	SS+METEOR	Sample 19	116 ± 13

Round 3	PE+PA PIR	SS+METEOR	Trial 1	103 ± 10
	PE+PA PCR	SS+METEOR	Trial 8	122 ± 9
	PE+EVOH PIR	SS+METEOR	Trial 2	87 ± 12
	PE+PET PIR1	SS+METEOR	Trial 5	117 ± 18
	PE+PET PIR2	SS+METEOR	Trial 6	123 ± 17
	PE+PET PIR3	SS+METEOR	Trial 7	126 ± 12
	PE+PET PCR	SS+METEOR	Trial 9	152 ± 34

Square-shaped specimens (dimensions 90 x 90 mm²) were cut from the cast films and biaxially stretched (simultaneous stretching) at 145 °C. This temperature range was selected after optimization, being an intermediate temperature between the melting points of PE vs. melting points of PA, EVOH and PET. Pre-heating time of 20 s (before stretching), stretching ratio of 1.5x1.5 and stretching speed of 417 %/s were used, unless otherwise stated. For the 1.5x1.5 stretched films the final dimensions were approximately 129mmx129mm and the film thickness was in the range of 40-60 µm.

All the samples were able to be stretched in a smooth process. Machine directional and transverse directional forces recorded during the stretching were low, in the range of ~0...0.7 N; the low force levels indicate that the PE phase was completely molten during stretching.

Round 2:



Round 3:



The observation through optical microscope and SEM are presented in the main part of the deliverable. OTR and WVTR measurements were made on the biaxially stretched samples as described in Annex 2.

MNL structures

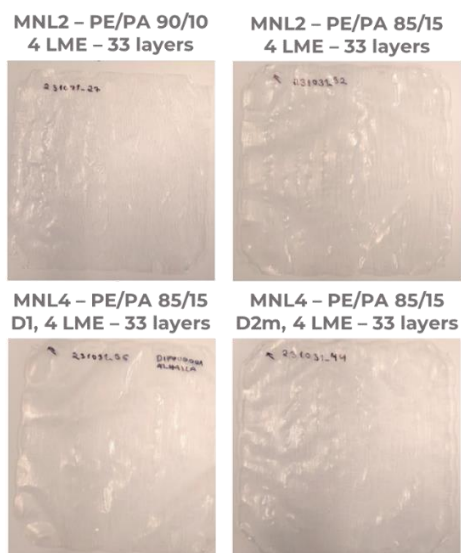
Due to resources available, a priority needed to be set because all samples could not be tested. The list of the samples tested is in the following table:

	Formulation	Cast thickness [um]
First campaign	PE/PA 90/10 4 LME – 33 layers	227 ± 46
	PE/PA 85/15 4 LME – 33 layers	238 ± 45
100kg production	Beginning Day 1	203 ± 31
	Middle Day 2	207 ± 27

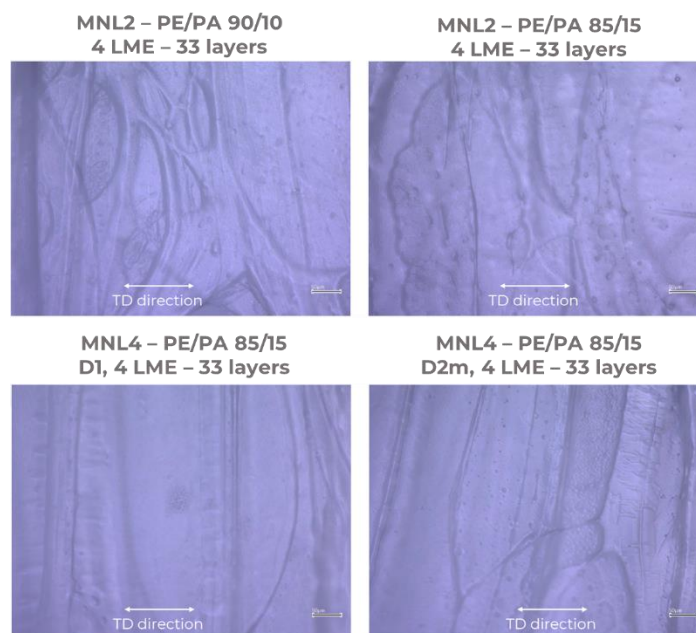
Square-shaped specimens (dimensions 90 x 90 mm²) were cut from the cast films and biaxially stretched (simultaneous stretching) at 145 °C. This temperature range was selected after optimization, being an intermediate temperature between the melting points of PE and of PA. Pre-heating time of 20 s (before stretching), stretching ratio of 1.5x1.5 and stretching speed of 417 %/s were used, unless otherwise stated. For the 1.5x1.5 stretched films the final dimensions were

approximately 129mmx129mm and the film thickness was in the range of 80-100 μm .

Pictures of the films show inhomogeneous surfaces. The films present holes and rough surfaces.



The observations through high resolution optical microscope are displayed below. The layers are not clearly seen.



The observations through SEM are presented in the main part of the deliverable. OTR and WVTR measurements were made on the biaxially stretched samples as described in Annex 2.