COTREP

Comité Technique pour le Recyclage des Emballages Plastiques

Resin

Flexible PE

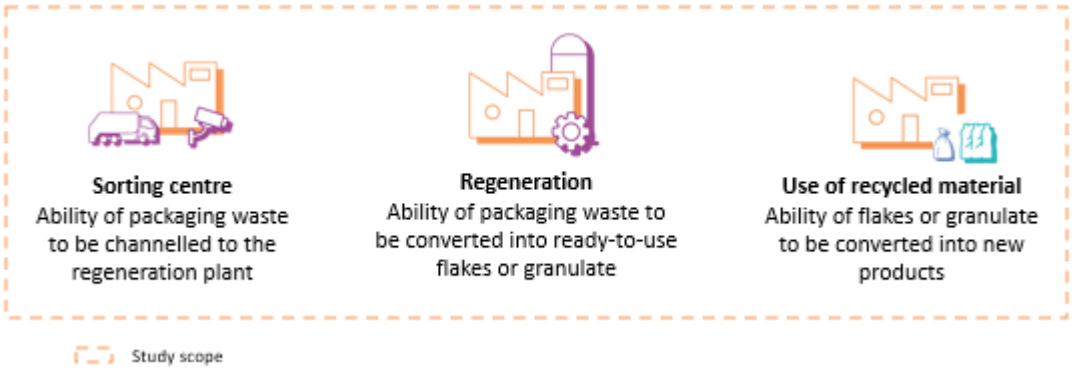
GENERAL NOTICE 70

Impact of PE/PA structures on regeneration by mechanical recycling of flexible PE household packaging

SUMMARY

The aim of this general notice is to assess the impact on regeneration of flexible PE household packaging containing a polyamide (PA) barrier.

In 2021, COTREP issued an initial notice regarding the impact of PE/PA structures on the generation by mechanical recycling of flexible PE household packaging based on structures available on the market at that time. Since this notice, new solutions have been developed in Europe. COTREP therefore decided to carry out a further study to take account of developments in packaging structures since the 2021 study and assess some of these structures.



PA is a polymer mainly used as a barrier in flexible packaging structures to meet product protection requirements.

During sorting, it is not possible to separate all PE/PA structures from the PE stream, which means that a certain quantity of PA enters the PE stream. COTREP therefore tested the potential impact of this packaging on the regeneration of flexible PE packaging.

The results of these tests show that the impact of PE/PA structures varies depending on the types and quantities of PA, tie layer or compatibilizer used. Depending on the structure used, PA can cause instability during the blow extrusion process and/or lead to changes in the properties of the rPE granulate produced. The additional use of PE compatibilizer grafted with MAH has no significant impact on co-extruded structures using PA 6 or PA 6/6.6.

In conclusion, given the current state of equipment and techniques used, COTREP has classified certain PE/PA structures as offering limited compatibility, as shown in the table below. COTREP may review this notice in light of developments in PE/PA structures available on the market, sorting and recycling technologies, or quality requirements for recycled material.

PE/PA STRUCTURES	COTREP'S OPINION
PE/PA structure produced by lamination with ≤ 25% PA 6 and ≥ 0.15 g of PE compatibilizer grafted with ≥ 1% MAH per gram of PA	**Limited compatibility**

PE/PA structure produced by co-extrusion with ≤ 15% PA 6 and ≥ 0.9 g of PE tie layer grafted with between 0.2% and 0.5% MAH per gram of PA	**Limited compatibility**
PE/PA structure produced by co-extrusion with ≤ 15% PA 6/6.6 with a melting point ≤ 192°C and ≥ 0.7 g of PE tie layer grafted with ≥ 0.1% MAH per gram of PA	**Limited compatibility**
Other PE/PA structures	**Non-compatible**

1. CONTEXT

Polyamide (PA) is a polymer mainly used in packaging due to its oxygen barrier properties that improve product protection and preservation and other properties that facilitate packaging forming while also offering superior perforation resistance. PA-type barriers are currently used for product preservation in the market for chilled products such as smoked salmon, grated cheese, cooked pressed cheeses and cured meats. In France in 2024, 20,000 tonnes of flexible packaging contained a PA barrier.

PA 6 is commonly used in PE/PA structures produced by co-extrusion or lamination. The '6' refers to the number of carbon atoms in the polyamide's constituent monomer. Various PA copolymers, such as PA 6/6.6, are available for use in co-extrusion. PA 6/6.6 is a copolymer of PA 6 and PA 6.6 and is the most commonly used copolymer for co-extruded packaging structures. Its comparatively low melting point closer to the conversion temperature of PE is advantageous when manufacturing films.

Polyethylenes grafted with maleic anhydride (MAH) may be used as a tie layer between the PE and PA or as a compatibilizer blended with the PE layer. Maleic anhydride enables polyolefins to bind with polar polymers such as PA. The aim of adding this compound as a compatibilizer is to facilitate the homogenisation of different polymer types, thus limiting the impact of PA content on flexible PE packaging regeneration by mechanical recycling. The effectiveness of maleic anhydride as a binder is dependent on the manufacturing process used to graft it onto the PE and the quantity of MAH actually grafted. Methods used to measure MAH quantities are not yet standardised or disclosed in detail by manufacturers for reasons of confidentiality.

The melting point of PA varies between 190°C and 220°C depending on copolymer type. This is much higher than the melting point for PE, which is between 110°C and 130°C depending on the grade. Due to its chemical properties, PA tends to capture moisture. Regenerators seeking to discard flexible PE/PA packaging do so for valid reasons, since the difference in the melting points of PA and PE and the potential for added moisture due to PA content can adversely affect their regeneration processes and/or the quality of recycled material produced.

In 2021, COTREP issued an initial notice regarding the impact of PE/PA structures on the generation by mechanical recycling of flexible PE household packaging based on laminated and co-extruded structures available on the market at that time.

Given the challenges of omitting a PA barrier for certain market applications mentioned in this notice, new solutions have been developed in Europe since 2021. Certain proportions of tie layer or compatibilizer in the packaging structure and/or different types of PA may be considered to improve the compatibility of PE/PA structures with flexible PE regeneration.

COTREP therefore decided to carry out a further study to take account of developments in packaging structures available since the 2021 study and assess these structures.

This notice seeks to assess the impact of PE/PA structures in flexible PE packaging on regeneration.

2. IMPACT ON REGENERATION

2.1. Principle and analytical criteria

In its study, COTREP assessed the impact of multilayer flexible PE packaging with a PA-type barrier on the process of regeneration by mechanical recycling and on the quality of recycled PE (rPE[1]) produced from flexible household packaging.

These tests were performed on a pilot scale based on protocols defined by COTREP for recycling flexible PE packaging. The protocols are representative of industrial practices applied by regenerators processing streams in France.[2]

Various physical-chemical criteria were measured during the test phases and compared to those of a standard sample composed of 100% rPE.

2.2. Test samples

Distinctions can be drawn between different packaging structures containing PA based on the type of PA used and the manufacturing process applied to bind the PE with the PA.

PE/PA structures can be manufactured using two processes:

Co-extrusion: Different polymers are extruded to form one film. A tie layer is needed to provide an interface between the PE and PA layers ensuring that the two layers bind properly. The tie layer is a PE polymer grafted with maleic anhydride (MAH). The proportion of maleic anhydride grafted varies depending on the tie layer type.

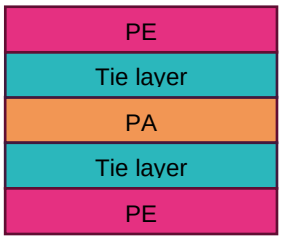
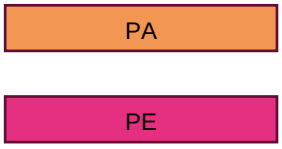


Diagram showing a PE/PA structure produced by co-extrusion

Lamination: Several films are first extruded then joined by lamination using an adhesive to join the PE and PA layers.

Step 1: PA film and PE film produced



Step 2: PE/PA structure created by lamination

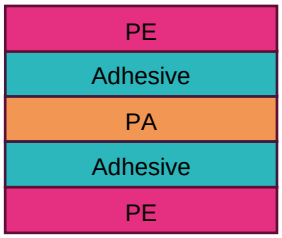


Diagram showing a PE/PA structure produced by lamination

[1] *rPE means recycled PE, i.e. recycled material from the regeneration of flexible PE household packaging*

[2] *For further information, see protocols Flexible PE-1 and Flexible PE-2 on the COTREP website: www.cotrep.fr*

Based on a market assessment, various structures were selected including PA 6 or PA 6/6.6 combined with different tie layers and compatibilizers. These were procured from several suppliers to create a representative sample of various possible PE/PA structures with a view to improving compatibility with the mechanical recycling of flexible PE packaging. The films were tested without any food residue.

The tested samples are listed in the table below.

Polyamide type	Manufacturing process	Tie layer	PE with MAH (maleic anhydride) compatibilizer
PA 6	Lamination	/	/
PA 6	Lamination	/	0.15 g of PE grafted with > 1% MAH per gram of PA
PA 6	Co-extrusion	0.2 g of PE grafted with < 0.2% MAH per gram of PA	/
PA 6	Co-extrusion	0.5 g of PE grafted with between 0.1% and 0.5% MAH per gram of PA	/
PA 6	Co-extrusion	0.5 g of PE grafted with between 0.1% and 0.5% MAH per gram of PA	0.15 g of PE grafted with > 0.7% MAH per gram of PA
PA 6	Co-extrusion	0.9 g of PE grafted with between 0.2% and 0.5% MAH per gram of PA	/
PA 6/6.6 with a melting point of 196°C	Co-extrusion	0.5 g of PE grafted with between 0.07% and 0.5% MAH per gram of PA	/
PA 6/6.6 with a melting point of 196°C	Co-extrusion	0.5 g of PE grafted with between 0.07% and 0.5% MAH per gram of PA	0.15 g of PE grafted with > 0.3% MAH per gram of PA
PA 6/6.6 with a melting point of 192°C	Co-extrusion	0.7 g of PE grafted with ≥ 0.1% MAH per gram of PA	/

PE/PA structures tested for the study

A 100% rPE film was produced exclusively as the standard film for the study from granulate manufactured from waste generated by the French selective collection (flexible PE standard).

Flexible PE packaging items containing PA are not separated as rejects at sorting centres. The optical signature of the PA between two PE layers can be difficult to detect. It can therefore be challenging to detect PA content in PE/PA structures, hindering efforts to separate these structures from other flexible PE packaging. As a result, some PE/PA films may enter the flexible PE packaging stream sent for regeneration.

Tests were therefore performed with 1%, 4% and 7% PA by mass to respectively reflect effective separation of a majority of structures by optical sorting, packaging placed on the market in 2024, and peak concentrations in bales.

2.3. Results

IMPACT OF PE/PA 6 STRUCTURES MANUFACTURED BY LAMINATION WITH OR WITHOUT COMPATIBILIZER ON REGENERATION AND CONVERSION PROCESSES FOR FLEXIBLE PE PACKAGING

RECYCLING PROCESSES	IMPACT	*DESCRIPTION*
SHREDDING	✓	*No impact on shredding*
WASHING AND SPINNING	✓	*No impact on washing or spinning.* *Note that, due to their PA content, all the tested samples exhibited higher moisture content than the standard sample tested on exiting the washing process.*
FLOTATION AND DRYING	✓	*No impact on flotation.* *Note that the presence of PA makes drying prior to extrusion more difficult as PA tends to reabsorb moisture easily*
EXTRUSION/ GRANULATION	⚠	*No impact on the extrusion process.* *At 7% PA using laminated PE/PA with ≥ 0.15 g of PE compatibilizer grafted with an MAH content ≥ 1% per gram of PA, granulate properties, in particular MFI, changed slightly.*
BLOW EXTRUSION	⚠	*From 1% PA using certain compatibilizer-free laminated PE/PA structures, blow extrusion was unstable, with the emergence of porosity invisible to the naked eye requiring the bubble to be supplied with air to maintain a constant blow ratio.* *For laminated PE/PA structures with ≥ 0.15 g of PE compatibilizer grafted with an MAH content ≥ 1% per gram of PA, blow extrusion was stable up to 7% PA.* *No impact on the properties of the films produced.*

⚠ Caution ✓ No impact

PA type	Manufacturing process	Tie layer	PE with MAH compatibilizer	Impact of the structure
PA 6	**Lamination**	**/**	**/**	From 1% PA, blow extrusion was unstable, with the emergence of porosity invisible to the naked eye requiring the bubble to be supplied with air to maintain a constant blow ratio. No impact on the properties of the films produced.
PA 6	**Lamination**	**/**	**0.15 g of PE grafted with > 1% MAH per gram of PA**	At 7% PA, granulate properties, particularly MFI, varied slightly. Blow extrusion was stable up to 7% PA. No impact on the properties of the films produced.

Impact of PE/PA 6 structures tested for the study manufactured by lamination with or without compatibilizer

Adding compatibilizer to the tested PE/PA 6 structure manufactured by lamination enabled blow extrusion process stability without affecting film properties but slightly modifying granulate properties.

IMPACT OF PE/PA 6 STRUCTURES MANUFACTURED BY CO-EXTRUSION WITH OR WITHOUT COMPATIBILIZER ON REGENERATION AND CONVERSION PROCESSES FOR FLEXIBLE PE PACKAGING

RECYCLING PROCESSES	IMPACT	*DESCRIPTION*
SHREDDING	✓	*No impact on shredding*
WASHING AND SPINNING	✓	*No impact on washing or spinning.* *Note that, due to their PA content, most of the tested samples exhibited higher moisture content than the standard sample tested on exiting the washing process.*
FLOTATION AND DRYING	✓	*No impact on flotation.* *Note that, for most of the tested samples, the presence of PA makes drying prior to extrusion more difficult as PA tends to reabsorb moisture easily.*
EXTRUSION/ GRANULATION	⚠	*Using certain co-extruded PE/PA structures with ≥ 0.5 g of PE tie layer grafted with between 0.1% and 0.5% MAH at 7 % of PA, there was no impact on the extrusion process but granulate properties were impacted.* ⇨ *Impact on MFI* *At 4% PA using a PE/PA structure with 0.9 g of PE tie layer grafted with between 0.2% and 0.5% MAH, there was no impact on the extrusion process or granulate properties.*
BLOW EXTRUSION	⚠	*From 1% PA using certain co-extruded PE/PA structures with ≤ 0.2 g of PE tie layer grafted with < 0.2% MAH per gram of PA, blow extrusion was unstable, with the emergence of porosity invisible to the naked eye requiring the bubble to be supplied with air to maintain a constant blow ratio.* *For co-extruded PE/PA structures with ≥ 0.5 g of PE tie layer grafted with between 0.2% and 0.5% MAH, blow extrusion was stable up to 7% PA.* *For the co-extruded PE/PA structure with 0.9 g of PE tie layer grafted with between 0.2% and 0.5% MAH, blow extrusion was stable up to 4% PA.* *For the co-extruded PE/PA structure with 0.9 g of PE tie layer grafted with between 0.2% and 0.5% MAH, some properties of the films produced were slightly impacted. No impact on the properties of the films produced with the other tested structures.*

⚠ Caution ✓ No impact

PA type	Manufacturing process	Tie layer	PE with MAH compatibilizer	Impact of the structure
PA 6	**Co-extrusion**	**0.2 g of PE grafted with < 0.2% MAH per gram of PA**	**/**	From 1% PA, blow extrusion was unstable, with the emergence of porosity invisible to the naked eye requiring the bubble to be supplied with air to maintain a constant blow ratio.
PA 6	**Co-extrusion**	**0.5 g of PE grafted with between 0.1% and 0.5% MAH per gram of PA**	**/**	At 7% PA, there was no impact on the extrusion process but granulate properties were impacted. Blow extrusion was stable up to 7% PA.
PA 6	**Co-extrusion**	**0.5 g of PE grafted with between 0.1% and 0.5% MAH per gram of PA**	**0.15 g of PE grafted with > 0.7% MAH per gram of PA**	
PA 6	**Co-extrusion**	**0.9 g of PE grafted with between 0.2% and 0.5% MAH per gram of PA**	**/**	At 4% PA, there was no impact on the extrusion process or granulate properties. Blow extrusion was stable up to 4% PA. Some properties of the films produced were slightly impacted.

Impact of PE/PA 6 structures tested for the study manufactured by co-extrusion with or without compatibilizer

Adding compatibilizer to the tested PE/PA 6 structures manufactured by co-extrusion did not improve the results obtained.

IMPACT OF PE/PA 6/6.6 STRUCTURES MANUFACTURED BY CO-EXTRUSION WITH OR WITHOUT COMPATIBILIZER ON REGENERATION AND CONVERSION PROCESSES FOR FLEXIBLE PE PACKAGING

RECYCLING PROCESSES	IMPACT	*DESCRIPTION*
SHREDDING	✓	*No impact on shredding*
WASHING AND SPINNING	✓	*No impact on washing or spinning.* *Note that, due to their PA content, all the tested samples exhibited higher moisture content than the standard sample tested on exiting the washing process.*
FLOTATION AND DRYING	✓	*No impact on flotation.* *Note that the presence of PA makes drying prior to extrusion more difficult as PA tends to reabsorb moisture easily*
EXTRUSION/ GRANULATION	⚠	*Between 1% and 7% PA using certain co-extruded PE/PA 6/6.6 structures with ≤ 0.5 g of PE tie layer grafted with between 0.07% and 0.5% MAH, granulate properties were impacted.* ⇨ *Impact on MFI* *No impact on the extrusion process or granulate properties up to 4% PA for certain PE/PA 6/6.6 structures with ≥ 0.7 g of PE tie layer grafted with ≥ 0.1% MAH per gram of PA.*
BLOW EXTRUSION	⚠	*Blow extrusion stable for all PE/PA 6/6.6 structures.* *At 7% PA using PE/PA 6/6.6 structures with ≤ 0.5 g of PE tie layer grafted with between 0.07% and 0.5% MAH per gram of PA, the properties of the films produced were impacted, particularly in terms of tensile elongation.* *Up to 4% PA using a PE/PA 6/6.6 structure with ≥ 0.7 g of PE tie layer grafted with ≥ 0.1% MAH per gram of PA, the properties of the films produced were compliant.*

⚠ Caution ✓ No impact

PA type	Manufacturing process	Tie layer	PE with MAH compatibilizer	Impact
PA 6/6.6 with a melting point of 196°C	**Co-extrusion**	**0.5 g of PE grafted with between 0.07% and 0.5% MAH per gram of PA**	**/**	From 1% PA, granulate properties were impacted. Blow extrusion stable. At 7% PA, the properties of the films produced were impacted, particularly in terms of tensile elongation.
PA 6/6.6 with a melting point of 196°C	**Co-extrusion**	**0.5 g of PE grafted with between 0.07% and 0.5% MAH per gram of PA**	**0.15 g of PE grafted with > 0.3% MAH per gram of PA**	
PA 6/6.6 with a melting point of 192°C	**Co-extrusion**	**0.7 g of PE grafted with ≥ 0.1% MAH per g of PA**	**/**	No impact on the extrusion process or granulate properties up to 4% PA. Blow extrusion stable. Up to 4% PA, the properties of the films produced were compliant.

Impact of PE/PA 6/6.6 structures tested for the study manufactured by co-extrusion with or without compatibilizer

Adding compatibilizer to the PE/PA 6/6.6 structures manufactured by co-extrusion tested for this study did not improve the results obtained.

TECHNICAL CONCLUSIONS

Through tests performed by COTREP, it was possible to assess the impact of PE/PA structures in PE films on the regeneration by mechanical recycling of flexible PE household packaging.

Results obtained show that:

- The impact of PE/PA structures varies depending on the types and quantities of PA, tie layer or compatibilizer used.

- For PE/PA 6 structures manufactured using a lamination process:
  - From 1% PA 6, the blow extrusion process was unstable: the tested PA/PA 6 structure produced by lamination without compatibilizer led to porosity invisible to the naked eye.
  - As regards the tested structure with ≥ 0.15 g of PE compatibilizer grafted with ≥ 1% MAH per gram of PA, the blow extrusion process was stable under test conditions, though granulate properties were slightly modified.
  - Adding compatibilizer to the tested PE/PA 6 structure manufactured by lamination enabled blow extrusion process stability without affecting film properties but slightly modifying granulate properties.

- For PE/PA 6 structures manufactured using a co-extrusion process:
  - From 1% PA 6 with ≤ 0.2 g of PE tie layer grafted with < 0.2% MAH per gram of PA, the blow extrusion process was unstable: the PE/PA 6 structure produced by co-extrusion with ≤ 0.2 g of PE tie layer grafted with < 0.2% MAH per gram of PA led to porosity invisible to the naked eye.
  - As regards the tested co-extruded PA 6 structure with ≥ 0.5 g of PE tie layer grafted with between 0.2% and 0.5% MAH per gram of PA, the blow extrusion process was stable up to 7% PA, though granulate properties were impacted at 7% PA.
  - As regards the tested co-extruded PA 6 structure with 0.9 g of PE tie layer grafted with between 0.2% and 0.5% MAH per gram of PA, the blow extrusion process was stable and granulate properties were not impacted up to 4% PA. Some properties of the films produced were slightly impacted at 4% PA.

- For PE/PA 6/6.6 structures manufactured using a co-extrusion process:
  - From 1% PA **with a melting point of 196°C** using the tested structure manufactured by co-extrusion with ≤ 0.5 g of PE tie layer grafted with between 0.07% and 0.5% MAH per gram of PA, granulate properties were impacted.
  - Up to 4% PA **with a melting point of 192°C** using the tested co-extruded PA 6/6.6 structure with ≥ 0.7 g of PE tie layer grafted with ≥ 0.1% MAH per gram of PA, the PA had no impact on the mechanical regeneration of flexible PE packaging.

- The additional use of PE compatibilizer grafted with MAH had no significant impact for tested co-extruded structures using PA 6 or PA 6/6.6.

Multiple tests were performed using different types of PE/PA structures composed of various grades of PA, tie layer and compatibilizer.

A wide variety of results was obtained, proving that a diverse a range of solutions with PE/PA structures can be developed and highlighting the complexity of drawing up a general notice on the impact of these structures on the flexible PE household packaging stream.

In conclusion, given the current state of equipment and techniques used:

- **Flexible PE/PA packaging produced by lamination with ≤ 25% PA 6 and ≥ 0.15 g of PE compatibilizer grafted with ≥ 1% MAH per gram of PA offers limited compatibility with the flexible PE household packaging stream.**
- **Flexible PE/PA packaging produced by co-extrusion with ≤ 15% PA 6 and 0.9 g of PE tie layer grafted with between 0.2% and 0.5% MAH per gram of PA offers limited compatibility with the flexible PE household packaging stream.**
- **Flexible PE/PA packaging produced by co-extrusion with ≤ 15% PA 6/6.6 whose melting point is ≤ 192°C and ≥ 0.7 g of PE tie layer grafted with ≥ 0.1% MAH per gram of PA offers limited compatibility with the flexible PE household packaging stream.**
- **Other PE/PA structures are incompatible and disrupt the regeneration by mechanical recycling of flexible PE household packaging placed on the market in France.**

It should be noted that the appearance of the films produced was not assessed in this test. A further study may be performed to supplement this notice.

COTREP may review this notice if needed in light of developments in PE/PA structures available on the market, sorting and recycling technologies, or quality requirements for recycled material.