



Coextruded PE/PA multilayer films are recyclable!

Dr. Rolf-Egbert Grützner*, Dr. Roland Bothor**

* BASF SE, D-67056 Ludwigshafen/Rhein; ** Institut cyclos-HTP GmbH, D-52076 Aachen

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1. Introduction

Over the course of decades, aliphatic polyamides such as polyamide 6 (PA6) as well as copolyamides from PA6 and PA6.6 monomers (PA6/6.6) have become essential components of flexible packaging solutions, e.g., for fatty, oxygen-sensitive food or various medical items.

Although significantly smaller quantities of polyamides (PA) are used in this segment compared to conventional packaging polymers such as polyethylene (PE), polypropylene (PP) or polyethylene terephthalate (PET), making them more like specialties,^(#1) integrating PA can bring about ecologically advantageous packaging structures as a result of the material's mechanical strength and its barrier effect in polyolefin-based packaging. These are thinner (downgauging) and therefore require fewer resources and smaller amounts of polymers; it is thus logical that these structures also generate a smaller carbon footprint while cutting down on packaging waste, all without having to compromise the long-term protection of packaged foods.

Although polyamides (PA) have positive effects on the sustainability of multilayer composite films, they are still deemed disturbing factors in the mechanical recycling of PE film fractions from household plastic waste collections. Simply put, it is assumed that polyamides are not compatible with the PE waste stream, and therefore represent contamination and need to be separated out; they should generally disappear from packaging structures in the medium and long term, as they are unacceptable for today's mechanical recycling processes.^(#2-4)

As the classification of packaging materials with regard to their recyclability is very closely intertwined with political and economic interests, a holistic approach for evaluating the advantages and disadvantages of individual polymers and the associated packaging solutions while considering pragmatic constraints is not only impossible, but in individual cases also not wanted.

If associations immediately tout "traffic light solutions" that require the presence of 95% polyolefin (RecyClass^(#5)) as a basic criterion for achieving the "green" category for full recyclability, without linking this demand to any technical basis, objective framework conditions for equal competition must be in doubt.

These classifications are not only questionable from a technical point of view (as they do not realistically reflect the current state of the art) – they also support the replacement of optimized PE/PA-based multilayer film structures with often less sustainable polyolefin solutions or plastic-coated paper structures. Moreover, the classification of PA as "incompatible" already exerts a massive influence on statutory recommendations (e.g., the minimum standard pursuant to VerpackG issued by the Central Agency Packaging Register (*Zentrale Stelle Verpackungsregister*, ZSVR)^(#2)) or design guidelines issued by associations (e.g., RecyClass, CEFLEX, Ellen MacArthur Foundation^(#5-7)). In turn, many of these recommendations are used as guidance by the retail sector (discount supermarket chains, chain stores, etc.^(#8-9)), with the design guidelines of companies in this sector exerting enormous pressure on the supply chain to create packaging solutions in accordance with their wishes.

The basis for rating recyclability as per section 21 (3) VerpackG (Packaging Act) is the minimum standard of the ZSVR.^(#2) Appendix 3 of this minimum standard forms the basis for determining incompatibilities in accordance with no. 4.3. "PA layers" are currently listed as an "incompatibility" in the "Film and LDPE" and "Rigid PP" packaging groups. There is no differentiation, e.g., by PA type, tie layers, etc.

This classification is based on various properties of polyamides when used in PE-based packaging. In particular, the melting points of PA and PE are significantly different and, in contrast to PE, PA provides a strong polarity, which theoretically leads to repulsion effects between these two materials and gives PA hygroscopic properties.

For any deviating determination (that PA does not negatively impact recyclability), individual evidence currently has to be produced in accordance with no. 4.3 of the minimum standard.



In order to provide such individual evidence, comprehensive investigations have been conducted to classify and certify the recyclability of packaging and materials at both BASF and Institut cyclos-HTP GmbH as an independent company; this is reported below. The concept of these investigations is based on the following principles:

- PE and PA are thermodynamically incompatible, but can be dispersed homogeneously and processed further using suitable treatment and extrusion technology for PA concentrations of < 10%.^(#10)
- PA concentrations of $\geq 10\%$ can be dispersed into homogeneous blends by establishing covalent bonds with the polyethylene using compatibilizers.^(#11–13)
- Commercially available processing methods were used as standard for regranulation (combination of shredder/densifier with single-screw extruder and underwater pelletizer system).
- PA accounts for < 4% of mixed plastic packaging waste from households in the EU^(#14) and < 3% in Germany^(#15), and including PA6 and the lower-melting PA6/6.6 copolyamides.
- As well as evaluating typical PE/PA multilayer film structures with a PA share of 20–30%, these original recipes were adapted to realistic concentrations comparable with those in mixed plastic waste using both systematic serial dilutions and independent test standards that reflect current recycling practice.^(#13)
- Alongside the PA concentration, the investigations focused on temperature profile(s), as well as moisture absorption and necessary drying steps during regranulation and the final processing to create blown films and/or injection-molded items.

2. Basic investigations of the BASF joint project with Dow, RW Plast and W&H

The work, which was originally intended to support our customers in closing their internal material loops, quickly became the basis for further, independent investigations. As a result, the basic statements of this phase 1 are summarized in advance.

Figure 1 illustrates the testing concept of this work.

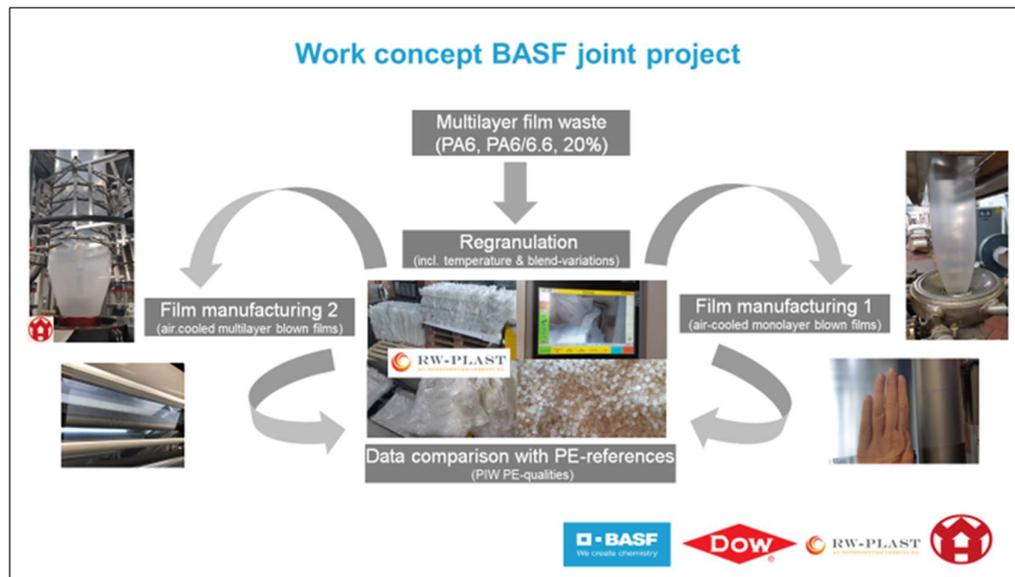


Fig. 1: Overview of work concept for internal BASF investigations

The base materials were coextruded PE/PA multilayer films with a PA share of 20%, with this being coated by both polyamide 6 (PA6, Ultramid B40LN) and a copolyamide 6/6.6 (PA6/6.6, Ultramid C40L). The investigations focused on PA6, as this represents the “worst-case” scenario: melting point = 220°C, highly viscous and highly crystalline, because a nucleated recipe (Ultramid® B40LN with N = nucleation additive) was used. In addition to the coextruded basic formulation (structure B, Fig. 2), two different compatibilizers were already integrated in the primary film structure here in order to assess their effectiveness with respect to a homogeneous morphology with undiluted PA concentrations of >10%. Figure 2 illustrates this variety of formulations based on the example of the applied PE/PA6 film structure.

PE/PA6 – Multilayer film structures

- **Reference:** PE/PE/PE/PE/PE/PE/PE/PE/PE
- **Structure B:** PE/PE/PE/tie#/B40LN/tie#/PE/PE/PE (20 % Ultramid® B40LN*)
- **Structure R:** PE/PE(inkl. CompR)/PE/tie/B40LN/Htie/PE/PE(inkl. CompR)/PE (20 % B40LN, 2 x 2,5 % CompR = Retain 3000**)
- **Structure F:** PE/PE(inkl. CompF)/PE/tie/B40LN/tie/PE/PE(inkl. CompF)/PE (20 % B40LN, 2 x 2,5 % CompF = Fusabond E226***)

*tie = tie layer; *Ultramid® B40LN = high viscosity (RV = 4,0), nucleated PA6, Source: BASF SE
 ** & *** different compatibilizer grades, Source: Dow/DuPont

Fig. 2: Selected examples of multilayer film structures investigated

All regranulations took place at RW Plast GmbH & Co. KG, using an S GRAN 95 (NGR) under processing conditions with practical relevance. This machine has a shredder / densifier / single-screw extruder combination, which was originally designed for processing residual polyolefins. The melt is exposed to a vacuum of max. – 1 bar at throughput rates in the region of 350 kg/h. The film regranulate extracted through underwater granulation was not explicitly dried, nor stored in damp-proof packaging.

Using suitable microscopic investigation methods, severe differences can already be determined regarding the morphology of the respective regranulate blends. To this end, microtome cut sections of the regranulates were dyed, examined under a microscope with polarized light and the color differences made visible using a special contrast method. All polyolefin components are shown in green, while all polyamide domains appear as reddish-orange areas in this and the following figures.

Figures 3a to 3c are compared directly to illustrate these effects.

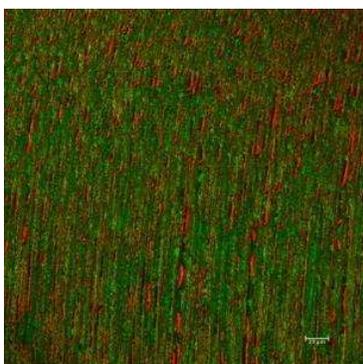


Fig. 3a: Morphology of regranulate B

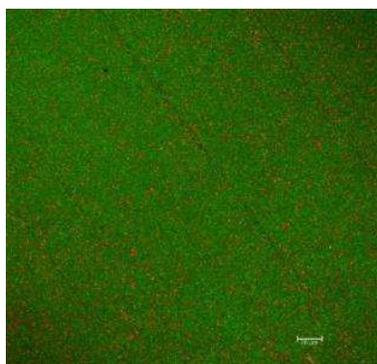


Fig. 3b: Morphology of regranulate R

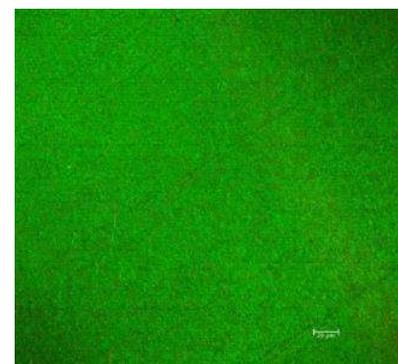


Fig. 3c: Morphology of regranulate F

While the incompatibility of the 80 % PE / 20 % PA6 mixture is clearly visible in Figure 3a through the inhomogeneous distribution of the PA blend component (orange) in the PE matrix (green), Figures 3b and 3c already show two examples of highly effective compatibilization during the process of standard regranulation on the basis of excellent component distribution and bonding in a homogeneous morphology of the existing PE/PA blend.

The systematic serial dilutions performed following this with a focus on the PE/PA blend regranulates with 20 % PA6 (Ultramid B40LN) and 20 % PA6/6.6 (Ultramid C40L) in each case built on these initial findings. To this end, simple physical (“salt and pepper”) mixtures were processed directly into mono blown films. Table 1 summarizes the recipes used in these tests.

While the concentration figures in black (e.g., RegrB or RegrC) show the absolute share, the figures in red represent the absolute share of the respective polyamide (PA) with respect to the overall structure (also in %).

Table 1: Test overview for PE/PA serial dilutions – film blends

| Nr. | RegrB* (PA6 ^{***}) % (%) | RegrR* (PA6 ^{***}) % (%) | RegrF* (PA6 ^{***}) % (%) | RegrC** (PA6/6.6 ^{***}) % (%) | PE Ref (%) | Comp [#] (%) |
|------|---------------------------------------|---------------------------------------|---------------------------------------|--|---------------|--------------------------|
| 1.0 | - | | - | - | 100 | - |
| 1.1 | 100/20 | | - | - | - | - |
| 1.2 | - | 100/20 | - | - | - | - |
| 1.3 | - | - | 100/20 | - | - | - |
| 2.5 | - | - | - | 100/20 | - | - |
| 3.1 | 50/10 | - | - | - | 50 | - |
| 3.2 | 37,5/7,5 | - | - | - | 62,5 | - |
| 3.3 | 25/5 | - | - | - | 75 | - |
| 1.4 | 95/19 | - | - | - | - | 5 |
| 4.2 | 50/10 | - | - | - | 45 | 5 |
| 4.3 | 37,5/7,5 | - | - | - | 57,5 | 5 |
| 4.4 | 25/5 | - | - | - | 70 | 5 |
| 3.8 | - | - | - | 50/10 | 50 | - |
| 3.9 | - | - | - | 37,5/7,5 | 62,5 | - |
| 3.10 | - | - | - | 25/5 | 75 | - |
| 4.10 | - | - | - | 97/19 | - | 3 |
| 4.12 | - | - | - | 50/10 | 27 | 3 |
| 4.13 | - | - | - | 37,5/7,5 | 59,5 | 3 |
| 4.14 | - | - | - | 25/5 | 72 | 3 |

* see Fig. 1; ** 80 % PE /20 % Ultramid[®] C40L; *** % PA in overall structure; # compatibilizer: Fusabond E226

All monofilms were processed using a Weber 30 blown film machine, die Ø 80 mm, BUR of 1:2.5 and throughputs of approx. 5–7 kg/h. The temperature profiles used were modified and optimized in connection with changed mixing ratios. While melt temperatures of 240°C (PA6) and 220°C (PA6/6.6) were necessary for the original recipes with 20 % PA, melt temperatures up to 20°C lower could be achieved on the extruder for mixing ratios with PA shares <= 10 %.

The resulting mono blown films were primarily analyzed in terms of mechanical characteristic values, while optical performance was assessed visually. The rheology (melt pressure, etc.) was adjusted on the basis of the

run logs. A comparison or monitoring of the melt index data of different PE/PA blend recipes, for example, was not performed at this early phase.

The tensile test was performed in accordance with ISO 527-3:2017 and provided information on the tensile strength, elongation at break and the tensile modulus of elasticity.

The measurements for puncture resistance were conducted as per DIN EN 14477 and for tear resistance (Elmendorf test) in line with DIN EN ISO 6383-2:2004.

Figures 4–6 below illustrate the measured mechanical characteristic values using representative comparative scenarios (a: machine direction (longitudinal); b: transversal direction). This is done through a percentage comparison to a 100% reference value, measured against the polyamide-free PE reference.

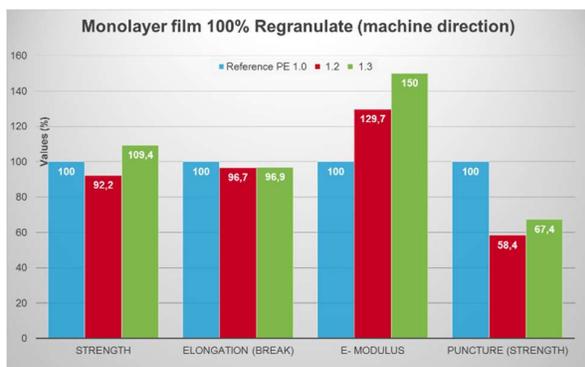


Fig. 4a: Mechanical film properties for 3 basic recipes (longitudinal)

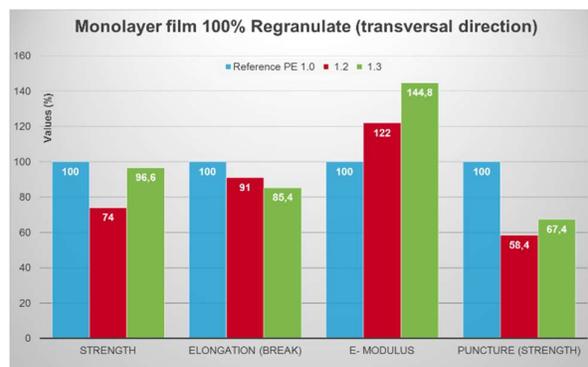


Fig. 4b: Mechanical film properties for 3 basic recipes (transversal)

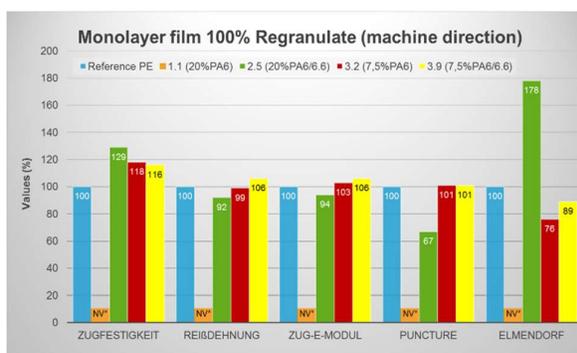


Fig. 5a: Mechanical film properties, dilution without compatibilizer (longitudinal)

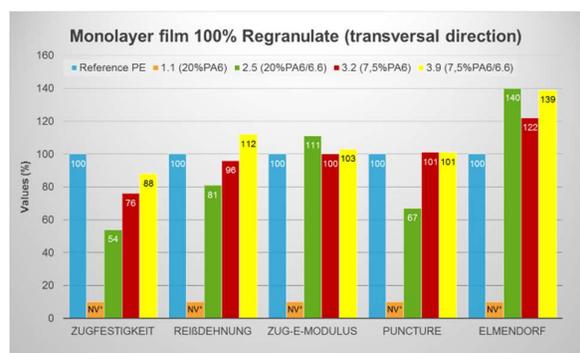


Fig. 5b: Mechanical film properties, dilution without compatibilizer (transversal)

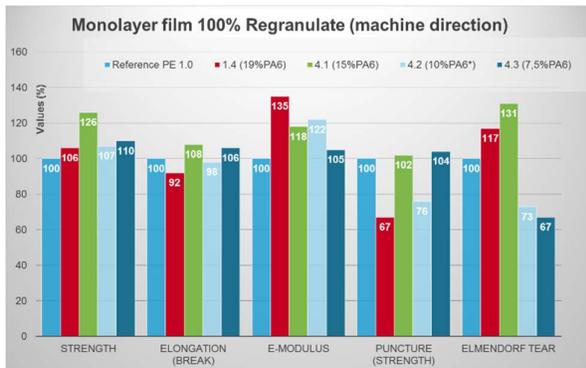


Fig. 6a: Mechanical film properties PE/PA6, dilution with added compatibilizer (longitudinal)

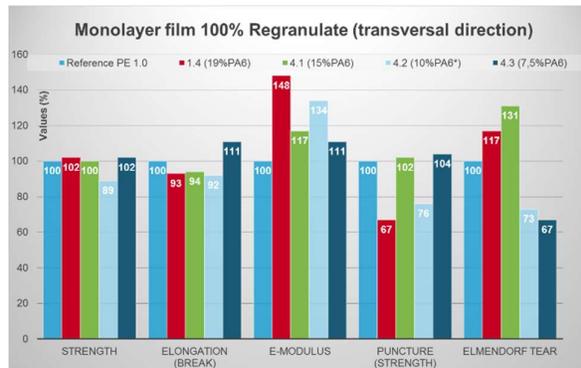


Fig. 6b: Mechanical film properties PE/PA6, dilution with added compatibilizer (transversal)

Figures 4a and 4b illustrate the effect of precompatibilization in the primary film and confirm its effectiveness. In spite of the high PA share of 20 %, films made from 100 % regranulate recorded almost identical strength and strain values, with the modulus of elasticity even showing significantly increased values. Although the puncture resistance declines significantly, this is the “worst-case scenario” without any optimization or dilution and is far away from reality or practical relevance in terms of processing post-consumer waste.

Figures 5a and 5b represent key mechanical characteristic values of diluted PE/PA blends without the use of compatibilizers. It is impressively demonstrated that, in the region of < 10 % PA (in this case 7.5 %), the data generated were very close to that of the reference sample made from 100 % PE virgin material for practically all measured characteristic values, including the more sensitive parameters of puncture resistance and tear resistance (Elmendorf test). This is the case for both PA6 and PA6/6.6 copolyamides, although PA6/6.6 is clearly easier to homogenize: In contrast to the tested PE / PA6 blend, it can be processed with a PA share of as little as 20 % without additional compatibilizers. This corresponds to and confirms expectations due to the lower melting point and crystallinity content of PA6/6.6 copolyamides.

If an effective compatibilizer is used, PE/PA mixtures can also be homogenized very well with PA concentrations of > 10 %. Figures 6a and 6b illustrate this effect on the basis of the selected PE / PA6 blends.

Although the measured mechanical characteristic values shown in the previous figures permit indirect conclusions to be drawn as to the quality of the blend morphologies created, microscopic examinations using the proven dyeing/contrast technology were also added to the evaluation program. Figures 7–12 show various morphology structures using representative mono blown film samples.

In the figures below, a different display variant has been chosen from that used for the regranulate morphologies presented above. While the reflection image in the left-hand section of each figure only visualizes the green PE matrix, the separate fluorescence image in the right-hand section shows the polyamide component as red domains.

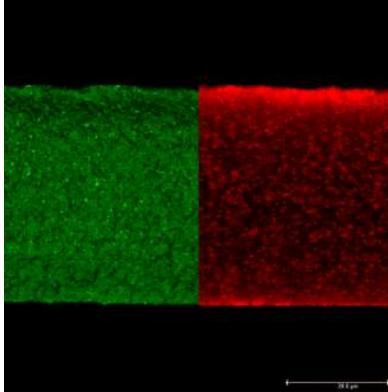


Fig.7: Morphology of film 1.2
(contains 20 % PA6 and 5 % E226,
precompatibilized)

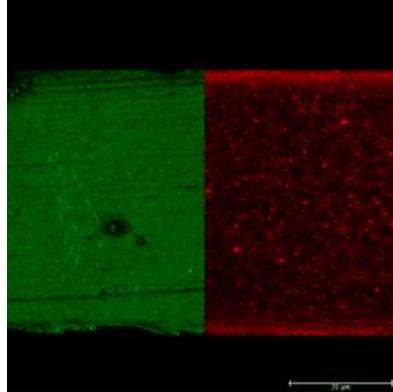


Fig. 8: Morphology of film 4.2
(contains 10 % PA6 and 5 % E226,
compatibilized via premixing)

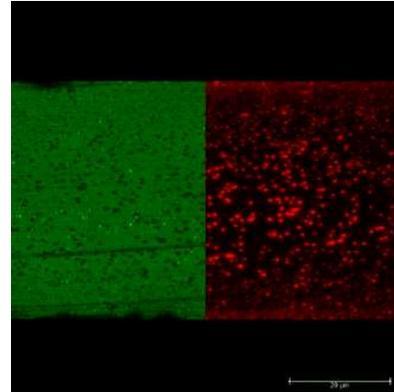


Fig. 9: Morphology of film 3.3
(contains 5 % PA6/6.6)

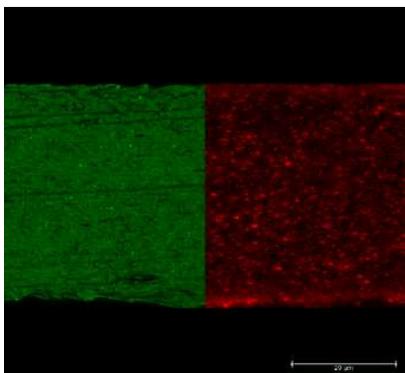


Fig. 10: Morphology of film 4.10
(contains 15 % PA6/6.6 and 3 % E226)

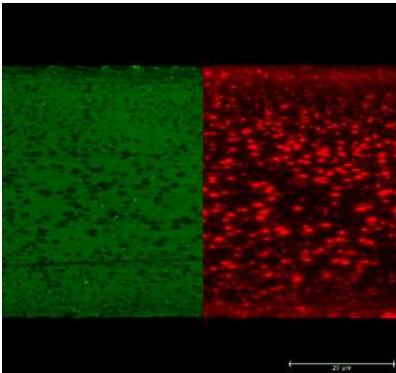


Fig. 11: Morphology of film 3.8
(contains 10 % PA6/6.6)

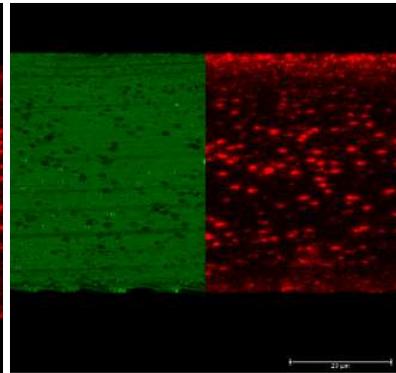


Fig. 12: Morphology of film 3.10
(contains 5 % PA6/6.6)

While the excellent homogeneity of the monofilm precompatibilized in the primary film (see Fig. 7) is not surprising, in these cases, too, various recipes prepared immediately prior to blown film manufacturing through simple physical mixing (“salt and pepper blends”) also show a highly effective compatibilization of polyamide components ≥ 10 % through Fusabond E226 (see Fig. 8 and 10).

Despite the lack of chemical coupling of the PA to the polyethylene, dilution to bring about PA concentrations in the region of 5–10 % results in an even dispersion and distribution of the minority component(s) in the matrix (see Fig. 9 (5 % PA6) and Fig. 11 and 12 (10 % and 5 % PA6/6.6).

Multilayer films containing regranelate were manufactured with a three-layered structure with a total thickness of 70 μm and the structure



on both a Varex II blown film line from Windmöller & Hölscher (die Ø 400 mm, haul-off film width 1700 mm dfl, 400 kg/h throughput, melt temperature approx. 245°C, film samples multi 0–5), and a multi-layer blown film line from Dr. Collin (haul-off film width 800 mm dfl, throughput 30 kg/h, melt temperature approx. 225°C, multi 6). The PA component was varied exclusively in the middle layer by means of different regranulate types and concentrations, with the non-precompatibilized settings multi 4–6 also being realized through simple premixing of the regranulate and compatibilizer components in the respective concentration ratio (“salt and pepper blends”).

Furthermore, a reference film based on 100 % PE ran on both lines, which was used as a 100 % basis in order to assess the mechanical characteristic values.

Table 2 presents an overview of representative recipes when using PE/PA regranulates. While the concentration figures in black (e.g., Re gr **B** or Re gr **C**) quantify the absolute share of the selected regranulate in the middle layer (in %), the figures in red represent the absolute share of the respective polyamide (PA) with respect to the overall structure (also in %).

Tab. 2: Overview of selected multilayer film structures with PE/PA regranulates in the middle layer

| Nr. | Re gr B * (PA6 ^{***}) (%) (%) | Re gr R * (PA6 ^{***}) (%) (%) | Re gr F * (PA6 ^{***}) (%) (%) | Re gr C ** (PA6/6.6 ^{***}) (%) (%) | PE Ref (%) | Comp [#] (%) |
|---------|---|---|---|--|---------------|--------------------------|
| Multi 0 | - | - | - | - | 100 | - |
| Multi 1 | 32/4 | - | - | - | 68 | - |
| Multi 2 | - | 32/4 | - | - | 68 | - |
| Multi 3 | - | - | 32/4 | - | 68 | - |
| Multi 4 | 78/10 | - | - | - | 20 | 2 |
| Multi 5 | 98/12,5 | - | - | - | - | 2 |
| Multi 6 | - | - | - | 78/10 | 20 | 2 |

* see Fig. 2; % only in middle layer; ** 80% PE / 20% C40L; *** % PA in overall structure; # compatibilizer: Fusabond E226

Figures 13a and 13b show selected mechanical characteristic values for representative multilayer films (in the longitudinal and transversal direction) with regranulates of varying concentrations integrated in the middle layer.

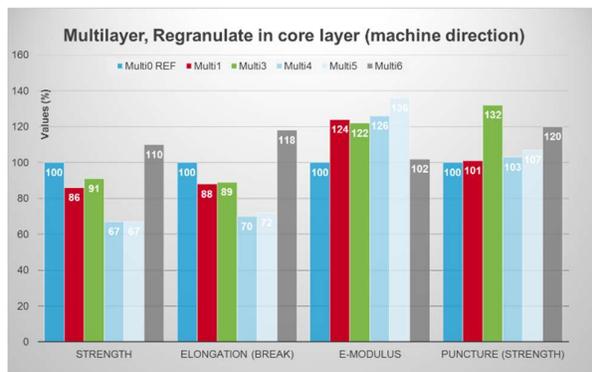


Fig. 13a: Mechanical values for multilayer films containing regranulate (longitudinal)

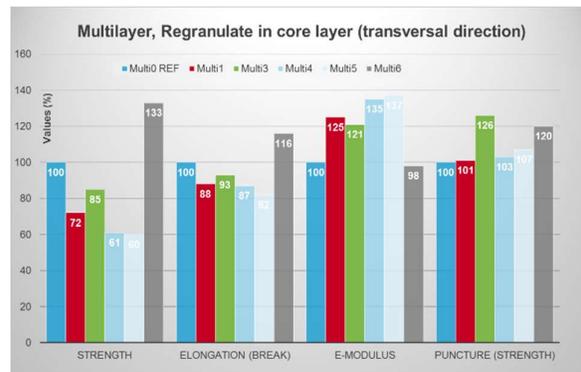


Fig. 13b: Mechanical values for multilayer films containing regranulate (transversal)

The charts for comparing the measured mechanical characteristic values confirm the results already generated on the basis of monofilms made of 100 % regranulate: Precompatibilized PE/PA6 blends (multi 3) can be integrated into a new multilayer film very well; the same also applies for PE/PA6/6.6 mixtures (multi 6), although the latter experiment contains a significantly higher share of PA6/6.6 and the compatibilizer was only added during final film manufacturing. This is further confirmation that the lower-melting and lower-crystalline copolyamides of the PA6/6.6 series are easier to homogenize. If no compatibilizer is available, a simple dilution of the PA component to < 10 % (here: multi 1 with 4 % PA6) is also sufficient for achieving acceptable mechanical characteristic values.

In the case of multilayer film structures, too, it is possible to gain a representative visual impression of the morphologies of the PE/PA blends in the middle layers by examining dyed microtome sections through contrast light microscopy. The microscopic images in Figures 14–16 show this. Accordingly, Fig. 15 very clearly shows the effectiveness of precompatibilization in direct comparison to Fig. 14. With the same PA6 concentration in the middle layer, the domains are virtually invisible when using the compatibilizer (multi 3, Fig. 15). The PA6 share in the middle layer can be increased to almost 20 % very effectively when using a compatibilizer, even when it is only added during the final film manufacturing stage (multi 5, Fig. 16).

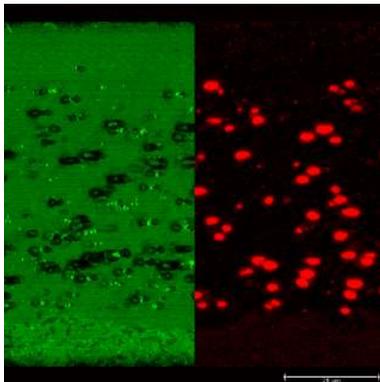


Fig. 14: Morphology of multi 1
(6.4 % PA6 in corelayer)

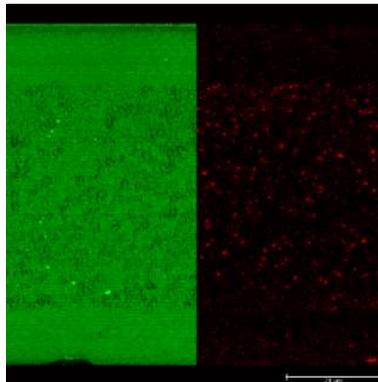


Fig. 15: Morphology of multi 3
(6.4 % PA6 in core, precompatibilized)

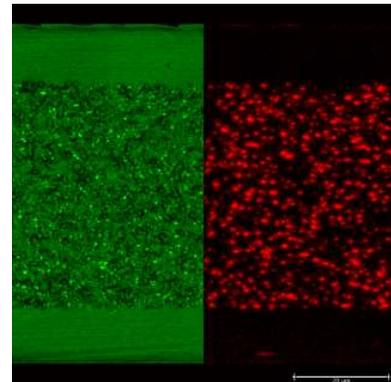


Fig. 16: Morphology of multi 5
(19.6 % PA6 in core, 2 % Retain 3000)

Because the optical quality of films in general and of recycled structures or those containing regranulate in particular are of special interest for the applications, the gel level or the ascertainable contaminations (black spots) in various multilayer films containing regranulate were quantified during the experiments on the Varex II from W&H. Furthermore, the gel level was compared when integrating the compatibilization step at various stages of the process (precompatibilization of the primary film, addition of the modifying agent during regranulation, “salt and pepper” premixtures of the individual components immediately prior to manufacturing of the final blown films).

Figures 17 and 18 below show the optical film quality in the photograph, while Figure 19 correlates the gel level measured with different compatibilizer addition concepts. While the pure dilution in multi 1 (Fig. 17) still produces gels and streaks, the positive effect of precompatibilization is also evident in the visual appearance of the resulting film (multi 2, Fig. 18a and 18b).

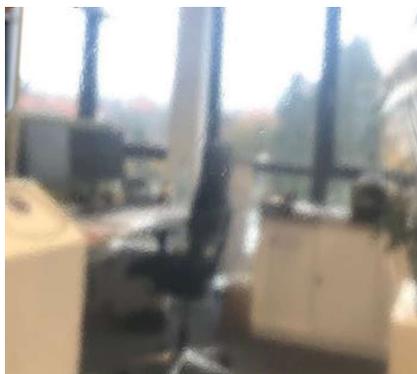


Fig. 17: multi 1 visual

Fig. 18a: multi 2 visual

Fig. 18b: multi 2, bubble

(Source of Figures 17 and 18: With the kind permission of Windmöller & Hölscher)

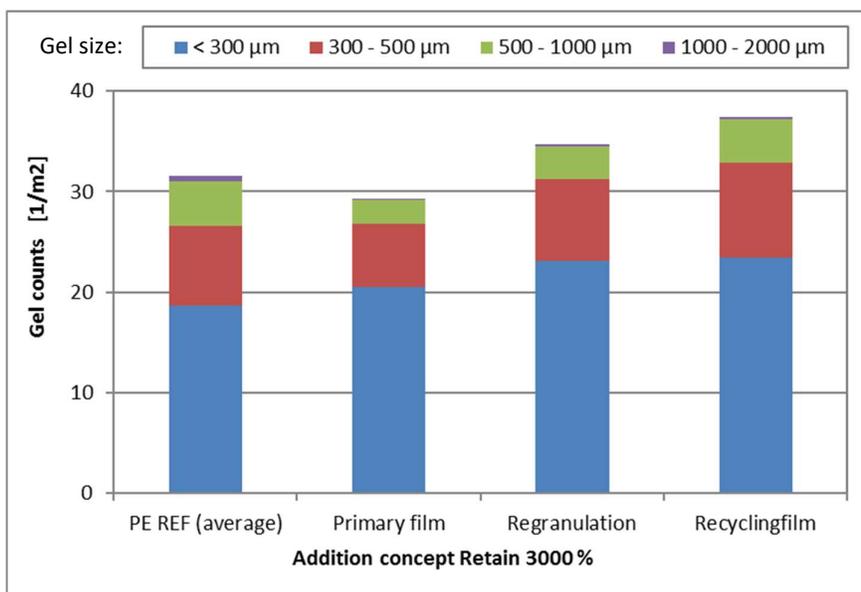


Fig. 19: Number of gels in multilayer films containing regranalate, as a function of the place of addition of Retain 3000 compatibilizer

(Source of Figure 19: With the kind permission of Windmöller & Hölscher)

Figure 19 shows the development of the gel level in the manufactured films as a function of the place at which the compatibilizer was added, in both absolute figures and with respect to various gel size classes. It is revealed that precompatibilization even reduces the gel level compared to the reference, while a “delayed” addition of compatibilizer results in slightly raised gel values. However, all of the measured gel values are in a



fundamentally comparable range, which means that these three compatibilization options can be considered for the practical implementation.

For the successful mechanical recycling of coextruded PE/PA multilayer film waste, the following reproduced constraints can thus be summarized:

1. When diluting PE/PA waste mixtures in PA concentrations (PA6 or PA6/6.6) < 10 %, PE/PA film waste can easily be dispersed without additional compatibilizers even when using standard regranulation technology with single-screw extruders.
2. If the polyamide share in the mixtures to be recycled increases to ≥ 10 %, a compatibilizer needs to be added. Maleic anhydride-grafted, PE-based modifiers such as Fusabond E226 or Retain 3000 have proven especially effective here. Adding a compatibilizer share of 2–5 %, depending on the type and concentration of PA, is recommended.
3. The necessary amount of compatibilizers can be added using various methods. Integration in the primary film – a process known as precompatibilization – is an elegant solution, as an effective homogenization of intrinsically incompatible components takes place during regranulation (even when using the simplest equipment). Furthermore, the additives can also be mixed in during regranulation (provided that even dosing of the target concentration throughout the process is ensured) or as simple physical blends (“salt and pepper” mixtures) prior to final processing, e.g., into blown films.
4. The temperature profiles to be selected depend on the existing polyamide concentration and the types to be used in each case. While PA6 shares of 20 % and more should ideally be extruded or processed with melt temperatures of at least 240°C, the essentially lower-melting PA6/6.6 copolyamides can be processed from temperatures of 210–220°C. If one considers the presence of a mixture of PA6 and PA6/6.6 in a ratio of approx. 1:1 and at concentrations of < 4 % in mixed packaging waste from households, the use of typical temperature profiles for PE recycling processes must be considered highly realistic.
5. As PA is known to absorb moisture, it may also be necessary to consider predrying. As is the case with the temperature profiles mentioned previously, this is also dependent on the PA types and concentrations present. Within the scope of the experiments summarized here, mixtures with PA concentrations of 20 % and more were predried, while PE/PA regranulates or blends of these with PA shares ≤ 10 % were generally not dried (i.e., obtained in the same manner as from regranulation or from a necessary interim storage in simple octabins/boxes with normal PE liners) before being processed into blown films. Figures 20 and 21 illustrate basic statements from the first phase of the investigation in concise form.

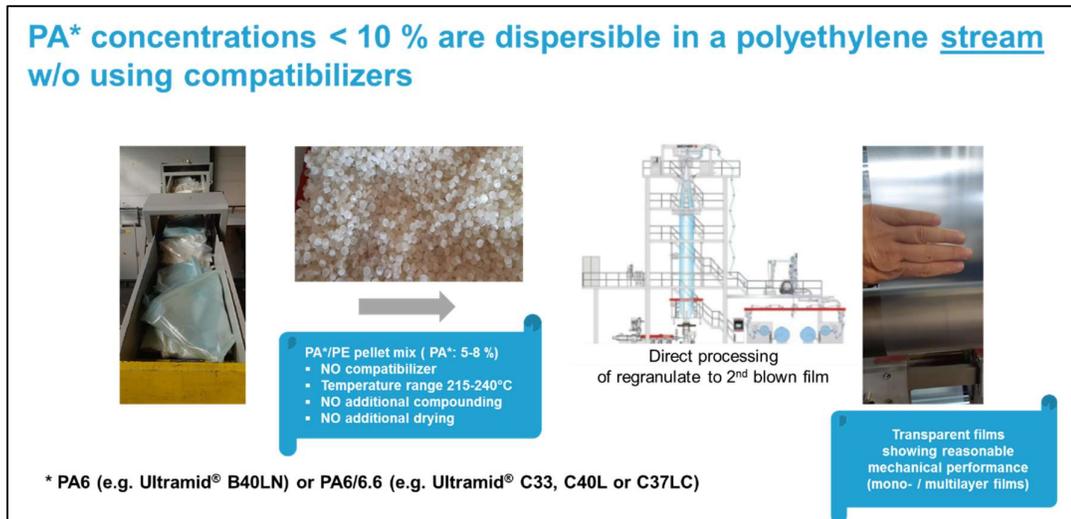


Fig. 20: Summary of process recommendation for PE/PA mixtures with < 10 % PA

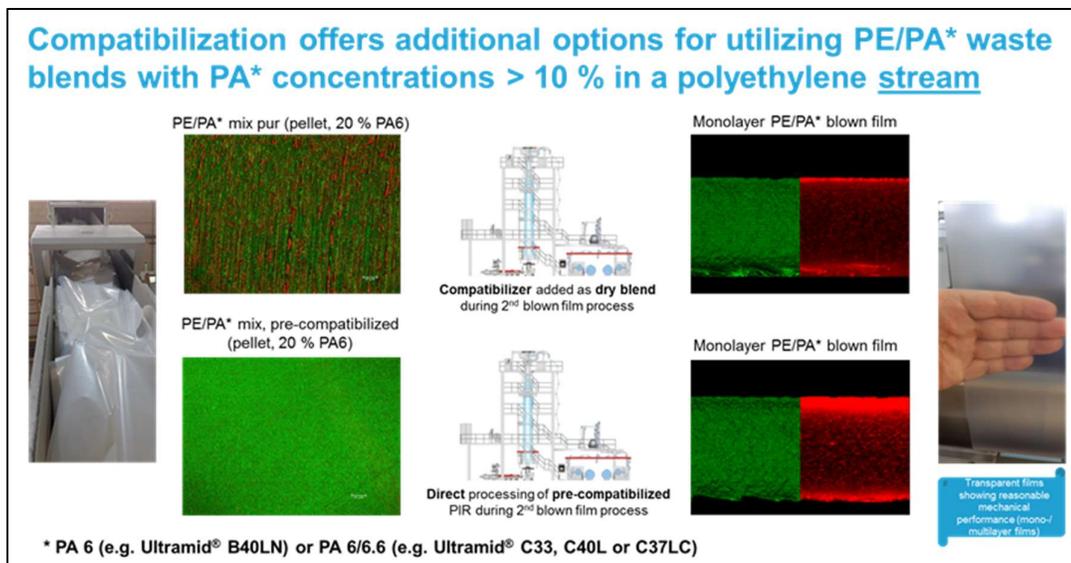


Fig. 21: Summary of process recommendation for PE/PA mixtures with >= 10 % PA

These results were the basis for initiating further collaborations with the goal of confirming and ideally certifying recycling compatibility and recyclability through independent and certified institutions in accordance with officially recognized testing standards.



We create chemistry



Institute cyclos-HTP GmbH in Aachen was ultimately commissioned to independently evaluate the possibilities for recycling flexible multilayer film waste containing polyamide. The results of these investigations are described below.

3. Certification project at Institute cyclos-HTP GmbH

Polyamides are a small – but essential – component of flexible multilayer films for applications including food, medicine or technical packaging, both in what are known as coextruded (joined through the melting process), multi-component composite and laminated (i.e., bonded) structures and in combination with other polymers such as PE, PP, EVOH, PET, etc. This is the only way to achieve optimum properties in the target application and also take advantage of the “downgauging” potential, i.e., the ability to create ever thinner and yet higher-performance film structures.

The investigations on recyclability when using mechanical recycling procedures summarized below focused on coextruded PE/PA multilayer films. Alongside the main components PE (with different variants, such as LDPE, LLDPE or mLLDPE) and PA, such coextrudates also contain what are known as tie layers. These are chemically modified polyethylenes that contain maleic anhydride groups grafted onto the main PE chain. Through hydrolysis and the release of carboxylic acid groups, these functional groups enable a chemical coupling of the tie layer to the neighboring PA layer, thereby guaranteeing stable and firm adhesion between neighboring PE and PA layers with intrinsically poor adhesion properties.

Figure 22 illustrates the basic principle of tie layer adhesion between nonpolar (e.g., polyethylene – PE) and polar (e.g., polyamide – PA6 or PA6/6.6) polymers.

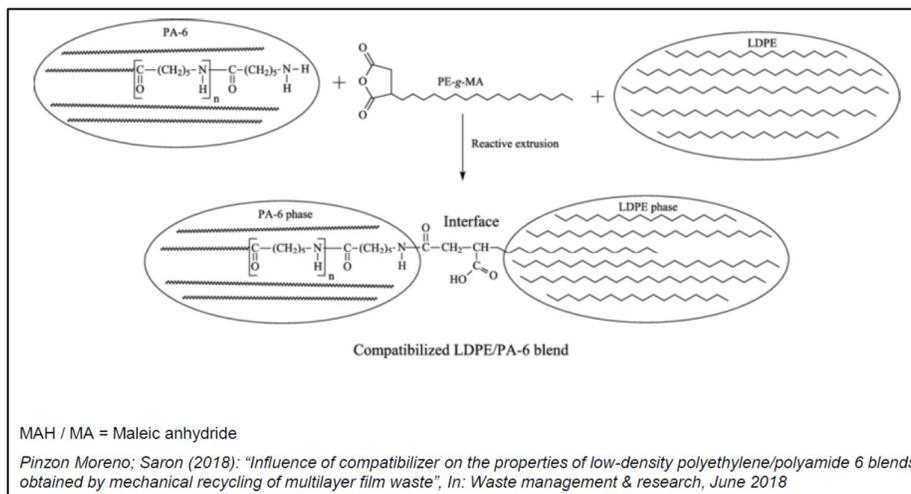


Fig. 22: Basic principle of tie layer adhesion between polyethylene and polyamide

The principal mechanical recyclability of flexible packaging containing PA (coextruded PE/PA multilayer films with PA6 and PA6/6.6) was assessed at Institute cyclos-HTP GmbH in accordance with the CHI test method CHI-C8-PEF-1 “Recycling Application Compatibility Test for PE-based Flexible Packaging.” This is a transparent testing and evaluation method for packaging and materials which takes into account basic scientific principles, the state of the art and practical experiences during the industrial recycling process.^(#16) The evaluation method from cyclos-HTP includes all European countries in which the infrastructure and the recycling of valuable materials is established on an industrial scale. The relevant countries will be named on the certificates on recycling compatibility or recyclability from cyclos-HTP.

This is important for initiating further activities (where applicable), for example changing the classification of PA from coextruded PE/PA multilayer composite films in the standards and guidelines of the relevant institutions in these countries.

Figure 23 shows the basic test sequence, while Figure 24 presents the used PE/PA multilayer film structures with up to 30 % PA6 (Ultramid® B40LN) and PA6/6.6 (Ultramid® C40L or C33L) using the PA6 test structures as an example.

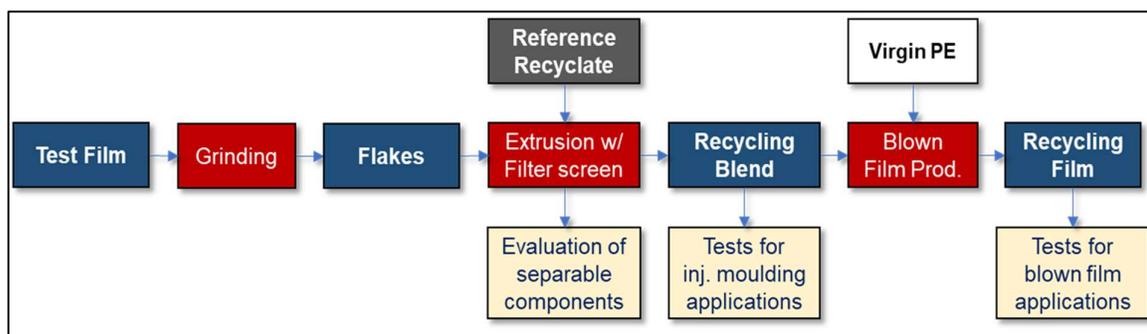


Fig. 23: Schematic diagram of the CHI standard test method CHI-C8-PEF-1

In accordance with the testing procedure shown in Fig. 23, recyclability is investigated on the basis of recyclate mixtures compared to the reference material (REF):

- **CHI 5** – “Realistic scenario” representing a slightly raised concentration of a packaging material in the recycling stream
= 5 % sample + 95 % reference | comparable with REF ✓ → compatibility of PA in PE films?
- **CHI 30** – “Worst-case scenario” representing a very high concentration of a packaging material in the recycling stream
= 30 % sample + 70 % reference | comparable with REF ✓ → 100 % recyclability of the entire packaging?
- **REF** – “Reference,” commercial PCR recyclate for the corresponding applications (injection molding, blown film) from the recycling stream to be investigated
- Optional: **PE 30** – “PE reference” for a typical PE-based packaging material without PA in the recycling stream
= 30 % reference sample without PA + 70 % reference

Target structure: symmetric medium barrier 7-layer-blown film (100 µm)

FB0 – PE / PE / PE / PE / PE / PE / PE

FB1 – PE* / tie** / PA6*** / tie / PA6 / tie / PE

FB2 – PE + Comp**** / tie / PA6 / tie / PA6 / tie / PE + Comp

“Recycling-critical” materials to be tested:

LDPE-based film with **30 % PA6** und **15 % tie layer**

LDPE-based film with **15 % PA6** und **7.5 % tie layer**

LDPE-based film with **30 % PA6** und **15 % tie layer** and **5 % compatibilizer**

* Typical mixture of 70 % LLDPE / 30 % LDPE (e.g. Dowlex GM8090/Dow 410E)

** maleicanhydrid-grafted PE (standard type for PE/PA6 coextrusion, e.g. ByneI 41E687)

*** PA6, Ultramid® B40LN, BASF SE

**** maleicanhydrid-grafted PE (specific type, e.g. Fusabond E226, Dow etc.)

Fig. 24: Overview of test structures for PE/PA6 program section (Ultramid® B40LN)

The three film structures defined in Figure 24 were produced on a Vorex II (air-cooled blown film line, die Ø 400 mm, BUR 1:2,5, haul-off film width 1400 mm dfl, throughput 450 kg/h) from Windmüller & Holscher using typical temperature profiles for the individual polymers (PE types: approx. 210, PA6: approx. 250°C) and subsequently reworked into PE/PA6 regranulates at RW Plast using again the S GRAN 95 recycling equipment of NGR, that is designed for processing film waste which is only feedable to a limited degree (throughput approx. 350 kg/h, temperature profile for PE/PA6 blends 235–240°C, vacuum max. –1 bar, underwater granulation). The created regranulates had (surface) moisture values in the region of 0.2–0.3 %, were not additionally dried, were temporarily stored in octabins with normal PE inliners and were processed further without additional predrying after various storage periods. Figure 25 summarizes detailed information from the regranulation step.



Fig. 25: S GRAN 95 impressions (left: material feed; middle: shredding unit; right: screw)

Here, too, it is easy to verify and compare the homogeneity of the individual morphologies realized during regranulation by means of contrast light microscopy on the basis of dyed microtome cut sections of the regranulates. Figure 26 shows a direct comparison of the pure PE reference, the non-compatible sample with a PA6 share of 30 % and the precompatibilized comparison variant with an identical share of PA6 in the regranulate.

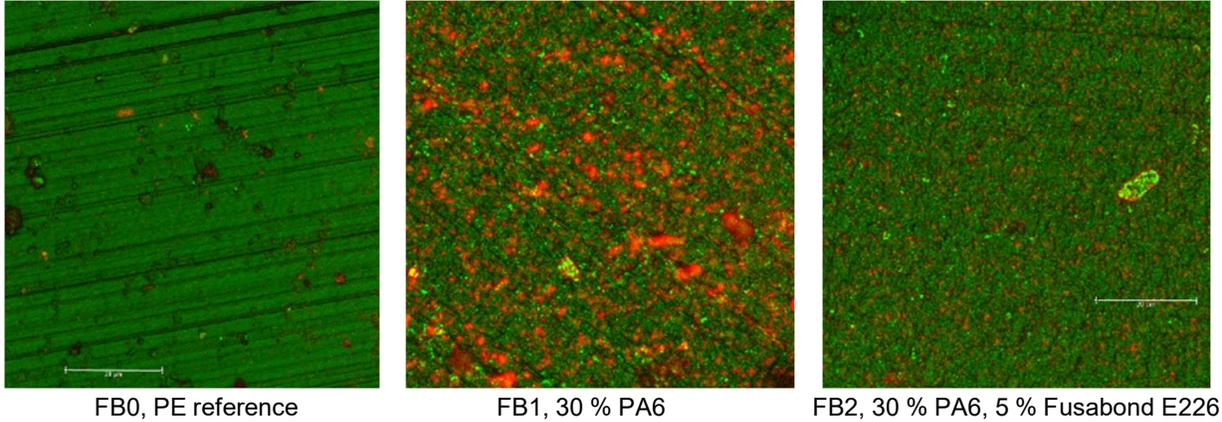


Fig. 26: Comparison of regranulate morphologies (for nomenclature, see Fig. 24) The remarkable effect of precompatibilization (here, e.g. 5 % Fusabond E226 in the primary film) when high concentrations of PA are present in the mixture (here, e.g. 30 % PA6) has also been proven impressively and reproducibly with these experiment settings.

A key component of the CHI test protocol was compounding to create the final material mixtures with a focus on defined “benchmark” recipes CHI 5 and CHI 30 (see Fig. 23). These experiments took place at Polymer Service GmbH, on a twin screw extruder (from Leistritz) using the three basic regranulates, an industrial polyolefin recyclate (PCR, Ecoplast NAV 104) from a real film fraction based on sorted residual films from mixed household plastic waste and a virgin product mix consisting of 70 % LLDPE / 30 % LDPE with the same type and composition as for the starting films based on a PE/PA6 combination in order to realize dilution levels that are typical of a standard procedure. All mixtures were processed without additional predrying at a predetermined temperature profile that is specified in the CHI standard as representative for the extrusion of polyethylene film waste fractions. (#16)

Figure 27 shows the equipment used for the tests and summarizes these process conditions.



Processing parameter:

- Extruder temperature (nozzle) = 210°C
- Melting temperature measured = 220-224°C
- Melt pressure = 50-60 bar
- Vacuum degassing after 6 of 8 zones
- Use of a melting pump

Fig. 27: Overview of extruder and processing conditions for compounding

Table 3 below provides a detailed overview of the recipes used to realize the defined key samples (see also definitions in Fig. 23 and Fig. 24).

Tab. 3: Overview of recipes for key samples CHI 5 and CHI 30

| Nr. | Blend components | | | | Composition | | |
|-----------------------|------------------|---------|---------|------------|-------------|-----------|--------------|
| | FB0* (%) | FB1 (%) | FB2 (%) | RECY** (%) | PA6 (%) | Comp# (%) | PCR RECY (%) |
| REF (PCR Recyclate) | - | - | - | 100 | 0 | 0 | 100 |
| CHI5: 30% PA6 | - | 5 | - | 95 | 1,5 | 0 | 95 |
| CHI5: 30% PA6 + Comp | - | - | 5 | 95 | 1,5 | 0,3 | 95 |
| PE30: PE film | 30 | - | - | 70 | 0 | 0 | 70 |
| CHI30: 30% PA6 | - | 30 | - | 70 | 10 | 0 | 70 |
| CHI30: 30% PA6 + Comp | - | - | 30 | 70 | 10 | 1,6 | 70 |
| CHI30: 15% PA6 | 15 | 15 | - | 70 | 5 | 0 | 70 |
| CHI30: 15% PA6 + Comp | 15 | - | 15 | 70 | 5 | 0,8 | 70 |

* For nomenclature, see Fig. 24, ** PCR reference: Ecoplast NAV 104; # compatibilizer: Fusabond

E226

No problems occurred during compounding – neither with moisture nor with separation or other inhomogeneities as a result of the temperature profile, which was representative but significantly reduced for the processing of PA6. Other measured characteristic values with process engineering relevance are summarized in Table 4.

Tab. 4: Overview of physical characteristic values of representative compound samples

| Rezyklatblend | Extrusion | Tests of recyclates | | C o m p o s i t i o n | | |
|---------------------------|-----------------------|---------------------|------------------------------|-----------------------|-----------|--------------|
| | Pressure-increase (%) | MFR (190°C/5kg) | Density (g/cm ³) | PA (%) | Comp# (%) | PCR RECY (%) |
| REF (PCR Recyclate) | 51 | 2,63 | 0,934 | 0 | 0 | 100 |
| PE30: PE film | 42 | 2,36 | 0,93 | 0 | 0 | 70 |
| CHI30: 30% PA6 | 30 | 1,86 | 0,947 | 10 | 0 | 70 |
| CHI30: 30% PA6 + Comp | 13 | 1,9 | 0,947 | 10 | 1,6 | 70 |
| CHI5: 30% PA6/6.6 (C40L) | - | 2,66 | 0,932 | 1,5 | - | 95 |
| CHI30: 30% PA6/6.6 (C40L) | - | 2,46 | 0,942 | 10 | - | 70 |

compatibilizer: Fusabond E226

The assessment standard CHI-C8-PEF-1-2.0 used for the investigations discussed here is not only oriented toward real-life practice in terms of the selection of reference material, but also takes into account relevant applications in connection with the technologies and procedures that are deployed as standard.

In specific terms, this means that the material samples summarized in Table 3 were evaluated with regard to their suitability for both injection molding applications (e.g., buckets) and typical blown film applications (e.g., garbage bags).^(#17) While the available CHI 5 and CHI 30 recyclates were used directly for the injection molding process, an additional dilution step with virgin PE at a ratio of 1:1 was conducted for the final blown film manufacturing. This corresponds to the state of the art and the procedure used in the recycling industry and is currently absolutely necessary in order to generate a recyclate quality from mixed household plastic waste that is suitable for blown films.

Figure 28 shows a selection of mechanical properties determined on the basis of standardized injection molding test specimens, while also illustrating the basis for assessing the available samples in terms of their usability for injection molding applications

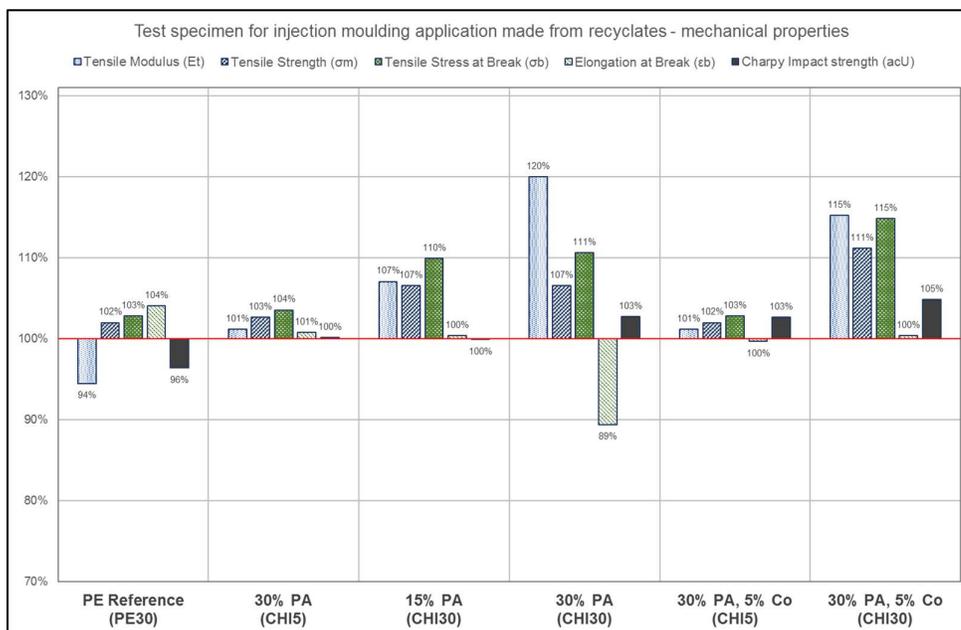


Fig. 28: Mechanical characteristic values of various recyclate samples, measured on the basis of injection molding specimens

While the CHI 5 samples passed the test in both cases (with and without Fusabond E226), this was only the case for the compatibilized variant of the CHI 30 samples with an initial concentration of 30 % PA6. Of the tested CHI 30 samples without pre-integrated compatibilizers, only the variant with a starting level of 15 % PA6 passes; the version with 30 % exhibits significantly worse values for elongation at break and thus did not pass the test. These results confirm the findings of the internal investigation phase (see chapter 1 of this publication), which revealed that, when the polyamide fraction in the PE material stream is 10 % or higher, the use of a compatibilizer is unavoidable in order to ensure that the resulting polymer mixture has a usable homogeneous morphology.

Due to the comprehensive program of tests for generating mono blown films from 100 % recycled material of varying compositions, this overview will not be provided within the scope of this publication. If required, the details can be found in the project report.^(#18) Instead, visual impressions of the blown film processing are provided in Figures 29a to 29c.

While PA6 concentrations <10% can be processed at typical PE extrusion temperatures of approx. 215°C (Fig. 29a), a temperature adjustment is necessary above this level (from 215°C in Fig. 29b to at least 235°C, Fig. 29c). This once again demonstrates the easier processability of PA6/6.6 copolyamides on the basis of a CHI 5 blown film sample (Fig. 29d). However, these necessary temperature adjustments do not represent a fundamental problem, as the realistic PA concentrations processed in post-consumer waste mixes are significantly below 10 % and are thus not affected.



Fig. 29a–d: Visual appearance of the monofilm bubble (melt temperature):
 a) 5 % PA6 (215°C) b) 15 % PA6 (215°C) c) 15 % PA6 (235°C) d) 0.75 % PA6/6.6 (210°C)

In addition, Figures 30 to 32 show the morphology of various mono blown films on the basis of the proven contrast light microscopy method using dyed cross-sections of the respective recycle film. Here, too, the effective influence of dilution and/or compatibilizing on the morphologies of the resulting PE/PA blends is demonstrated convincingly in the manner proven and discussed previously (see, for example, Fig. 10–12 or 14–16). Interestingly, significant red domains are also visible in the fluorescence image on the right-hand side of the microscopic image for the officially PA-free reference sample (PCR material, Fig. 30), which can be attributed to polar polymer fractions (e.g., PA) and definitely do not represent PE or PP.

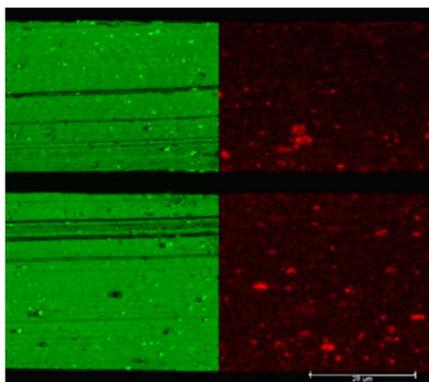


Fig. 30: Ref. PCR recyclete (0 % PA6)

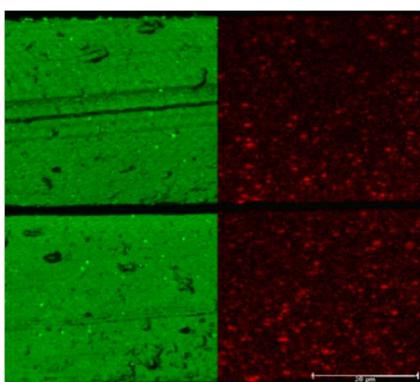


Fig. 31: CHI 5 – 30% PA6 (0.75 % PA6)

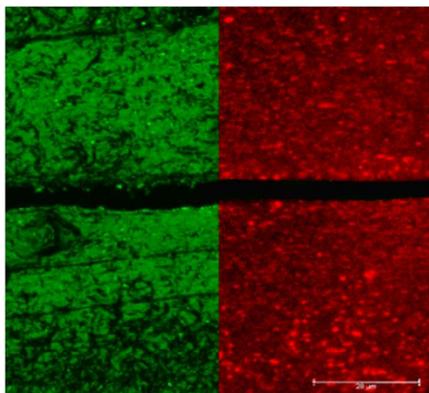


Fig. 32: CHI 30 – 30 % PA (5 % PA6)

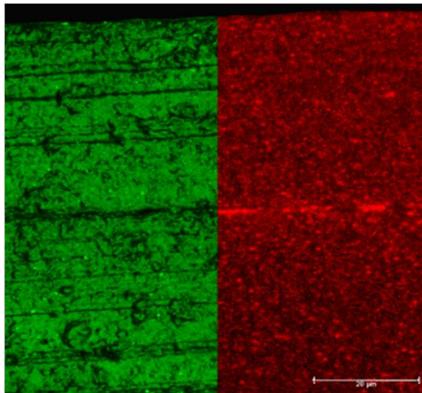


Fig. 33: CHI 30 – 30 % PA + comp. (5 % PA6)

As part of blown film manufacturing, the compounded recyclate samples (see Table 3 for recipes) were once again mixed with fresh PE virgin material with a ratio of 1:1. This corresponds to both the standardized evaluation method^(#17) and the practiced state of the art in the recycling industry. This resulted in the polyamide concentrations specified in Figures 29–33, which correspond to those in the manufactured monofilm after the two aforementioned dilution steps.

A comparative overview of representative mechanical and characteristic values, incorporating the seal strength of bags manufactured from the blown film samples, can be found in Figure 34.

The comparison of various mixtures containing PA in Fig. 34 once again shows that reproducibly excellent mechanical parameters were measured on the resulting blown films when PA concentrations of less than 10 % were ensured, even without the use of an additional compatibilizer.

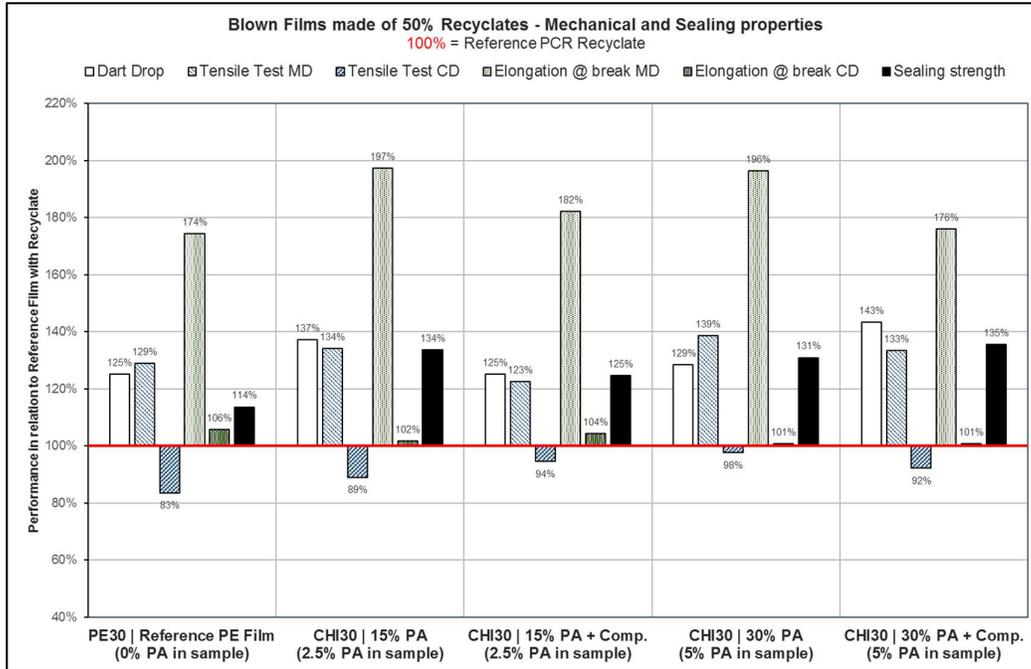


Fig. 34: Mechanical characteristic values of various recyclate samples, measured on the basis of blown films

One very important test that should be examined in somewhat greater detail is the tightness test for the seal strength of heat-sealed recyclate films, which is also part of the CHI standard for assessing blown film quality. (#19) To this end, the film samples to be tested were heat-sealed to form bags with a sealed seam width of 2.4 mm and were then filled with approx. 1.8 l of water corresponding to their diameter of 11 cm and a filling height of 20 cm. The filled bags were hung up and monitored over at least 10 minutes in order to determine whether water escaped due to leaks or the sealed seam tore open.

If less than two drops of water escaped per minute, the test is deemed to have been passed. All tested blown films passed this tightness test, regardless of their polyamide or compatibilizer component. Table 5 below presents an overview of the test results for the measured samples.

Tab. 5: Tightness test for mono blown films containing recyclate – overview:



| Recyclate blend in mixture* | Composition | | | Leak test (drops/min) |
|-----------------------------|-------------|-----------|--------------|-----------------------|
| | PA (%) | Comp# (%) | PCR RECY (%) | |
| REF (PCR Rezyklat) | 0 | 0 | 50 | 0 |
| CHI5: 30% PA6 | 0,8 | 0 | 47,5 | 0 |
| CHI5: 30% PA6 + Comp | 0,8 | 0,15 | 47,5 | 0 |
| PE30: PE film | 0 | 0 | 35 | 0 |
| CHI30: 30% PA6 | 5 | 0 | 35 | 1 |
| CHI30: 30% PA6 + Comp | 5 | 0,8 | 35 | 0 |
| CHI30: 15% PA6 | 2,5 | 0 | 35 | 0 |
| CHI30: 15% PA6 + Comp | 2,5 | 0,4 | 35 | 0 |
| CHI100**: 30% PA6 + Comp | 15 | 2,5 | 0 | 0 |
| CHI5: 30% PA6/6.6 (C40L) | 0,8 | 0 | 47,5 | 0 |
| CHI30: 30% PA6/6.6 (C40L) | 5 | 0 | 35 | 1 |

* 50% mixed with 50 % PE virgin material in each case, ** base sample at an extrusion temperature +20°C (235°C), for comparison purposes, # compatibilizer: Fusabond E226

Copolyamides consisting of PA6 and PA6.6, known as random PA6/6.6 copolyamides, have a far lower crystallite melting point than the most important polyamide, PA6. In addition to slower crystallization and a lower crystallite content in the manufactured film (at a comparable cooling rate during processing), this also results in significantly better mechanical flexibility properties such as puncture resistance, ductility and elongation. Figure 35 shows the correlation between the comonomer content and the resulting melting points of the copolyamide variants, with all technically available PA6/6.6 copolyamides having melting points between 182 and 196°C.

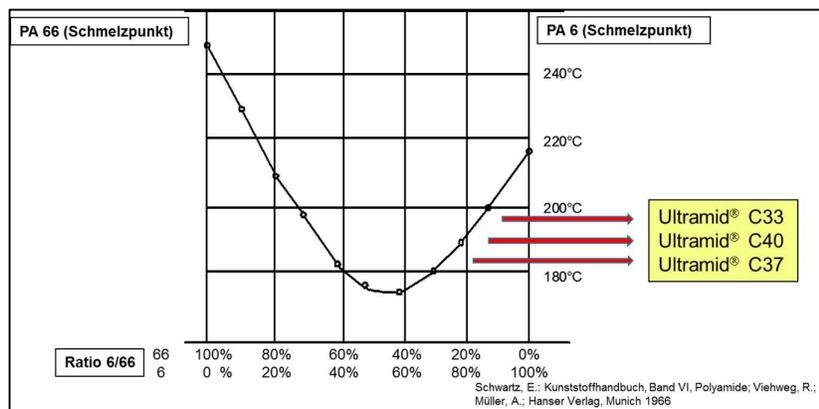


Fig. 35: Correlation of melting points of PA6/6.6 copolyamides

As such, numerous factors indicate easier processing and dispersion in a PE matrix. They are very well suited to the normal temperature profile for extrusion (180–210°C), they melt faster and are easier to disperse due to the lower crystallite share and, ultimately, they contribute significantly better mechanical flexibility properties to the PE waste film stream.

For this reason, two representative PA6/6.6 copolyamides were evaluated with a slightly simplified test procedure. Ultramid C33 (melting point 196°C, medium viscosity) and Ultramid C40L (melting point 189°C, high



viscosity) were also dispersed in a PE matrix with a concentration of 30 % in combination with 15 % of a comparable tie layer. Instead of the regranulated multilayer films, however, a 30 % “root compound” was produced, which, using the CHI 5 and CHI 30 test mixtures, was subsequently processed into standard-compliant injection molding test specimens using the same sequence in accordance with CHI-C8-BFPE-2.0,^(#17) and into final mono blown films in combination with an additional dilution step.

As expected, and like the investigated mixtures containing PA6, the manufactured mono blown films passed the evaluation of their mechanical characteristic values.

Representative examples of various measurement results and processing ratios have been integrated in the respective figures and tables, e.g., the image of a mono blown film in Fig. 29d, the density and MFI values in Table 4 and the tightness values of manufactured bags in Table 5.

An examination of the possible moisture absorption of waste or recycle containing PA will round off the investigation presented here.

In connection with the assessment of the recyclability of PE/PA multilayer films, whether coextruded using tie layers or laminated using adhesive, the argument of polyamides’ water absorption is repeatedly mentioned as a knock-out criterion. Specifically, this refers to two different approaches:

1. Film flakes containing PA absorb too much moisture during the washing and density separation steps of the recycling process; as a result, effective drying is no longer guaranteed in the normal recycling process, leading to problems during manufacturing and a poorer recycle quality.
2. Under normal storage conditions, recycles containing PA absorb so much moisture that final processing to create injection molding items and, in particular, blown films is no longer possible.

With regard to the first approach, it is apparent that the residence time during the washing and density separation step is not long enough to permit a complete migration of water into the PA layer(s), which are also usually enclosed in PE layers. As such, the moisture is predominantly surface moisture, which is largely removed in the also integrated drying process BEFORE extrusion. In addition, the latest recycling extruders possess (vacuum) degassing functions, which are also used to remove volatile components.

Table 6 shows the resulting moisture absorption of PE/PA6 regranulates with very high shares of PA6 (20 or 30 %) when immersed directly in water for one hour(!) as a practically unrealistic worst-case scenario.

Tab. 6: Water absorption of different granulates when immersed in water for one hour

| Nr. | Typ | Rezept | Moisture (%) | |
|-------|-------------|------------------|--------------|--------------------|
| | | | start point | 1h in water (23°C) |
| FB0 | regranulate | PE | 0,093 | 0,1 |
| FB1 | regranulate | 30%PA6/PE | 0,11 | 0,3 |
| FB2 | regranulate | 30%PA6/PE + Comp | 0,11 | 0,31 |
| B40LN | virgin PA6 | PA6 (B40LN) | 0,1 | 0,65 |

Regarding the second approach, it is apparent that recycling plants with a focus on PE-based film recycles generally have underwater pelletizers. This technology produces spherical pellets without visible cut edges. This study has demonstrated that the PA shares form what are known as domains in the PE matrix of the recycle, which are always completely surrounded by hydrophobic PE. The good dispersing properties of the low PA shares (<5 %) in practically relevant PE recycles, combined with the geometry of a spherical pellet,



impedes moisture absorption during storage under normal climactic conditions and does not present a risk with regard to the downstream processing steps for manufacturing the end products.

During the course of the investigations, some of the materials were stored in standard packaging units (cardboard boxes with simple PE inliners) under various climactic conditions over longer periods of time, but they were never dried.

Nevertheless, this topic and the theoretical approach presented here will be investigated in greater detail through further test series on realistic PE/PA recyclates, in some cases considering longer storage cycles.

4. Conclusions and outlook

To sum up, the recycling compatibility of PA as a material in general and the recyclability of PE film structures containing PA in particular have been proven in a reproducible manner, regardless of the detailed procedure and the reference sample quality used, on the basis of both polyamide 6 (PA6) and copolyamides consisting of PA6 and PA6.6 (PA6/6.6).

Various constraints must be adhered to depending on the polyamide concentration in the respective mixture, with the dispersing limit of 10 % in the recycle playing a particularly important role. It is highly improbable that a typical recycling stream made up of household waste will even come close to this limit. When this aspect is disregarded, a relatively simple homogenization into a PE matrix is possible below this limit. Above 10 %, the use of a compatibilizer is essential in order to realize a homogeneous morphology of the blend. The most elegant way to add a compatibilizer is by integrating it into a multilayer primary film, which is also known as precompatibilization. However, the compatibilizer can also be added to this blend by means of a simple physical preblend (“salt and pepper”) and through direct dosage – both when integrated during regranulation and immediately prior to final processing for a new, recycle-based application.

The main result of the systematic investigations by the Institute cyclos-HTP in accordance with the CHI standard is the official confirmation of recycling compatibility and recyclability through a current total of five certificates, as well as the practical use of these new evaluation guidelines when evaluating the recyclability of coextruded multilayer film structures containing PA. PA layers in coextruded structures are newly classified while taking into account certain prerequisites described here and independently of the current classification status in the recommendations and design guides of a wide range of institutes.

Figures 36 and 37 show the certificates confirming recycling compatibility of both PA6 (Fig. 36) and PA6/6.6 copolyamides with melting points < 200°C (Fig. 37).



Fig. 36: Certificate for recycling compatibility of PA6 Fig. 37: Certificate for recycling compatibility of PA6/6.6



Figures 38–40 show the certificates confirming the recyclability of the two named polyamide groups; in this case, different grades apply depending on the used recipe or film structure. Regarding the confirmation of 100 % recyclability in the case in which compatibilizer is present in the primary film, the feasibility when using PA6 has been proven during the course of the investigations and confirmed with an associated certificate. The transferability of the results to PA6/6.6 copolyamides is clearly and automatically derived when considering the lower melting points, the lower level of crystallization and the comparable homogenization success when processing non-compatibilized PE/PA blends proven during the course of the investigations; as a result, this was only examined on a spot-check basis.

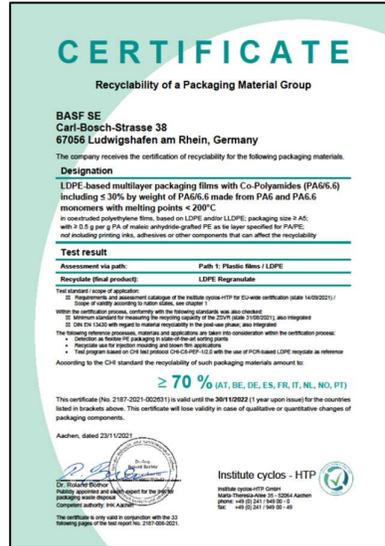
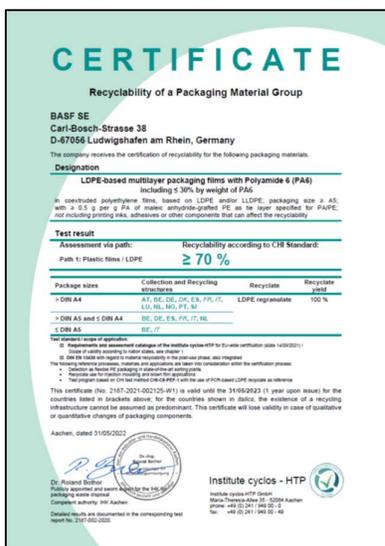


Fig. 38 Certificate for recyclability of PE/PA ≤ 30 % PA6 Fig. 39: Certificate for recyclability of PE/PA ≤ 30 % PA6, compatibilized Fig. 40: Certificate for recyclability of PE/PA ≤ 30 % PA6/6.6

The investigations clearly demonstrated that both PA6 and PA6/6.6 copolyamides in realistic proportions can be tolerated in PE film waste streams for mechanical recycling of the LDPE film fraction without any problems or additional effort. Conventional temperature profiles for remelting material mixtures that predominantly contain PE (180–210°C) were used and it was not necessary to additionally dry even regregranates with a high PA6 content before or during further processing.

The realistic scenario with a low content of PE/PA films was examined on the basis of recyclate mixtures with a PE/PA film fraction of 5 % and a PA fraction of less than 2 %. Here, the results show that the homogeneous dispersion of PA particles in the PE matrix is generally possible without the addition of compatibilizers.

The tie layer concentrations in coextruded multilayer films already have a positive function here as “diluted” compatibilizers and, in addition to guaranteeing dispersability in practical applications, also ensure that the resulting polymer blend has a stable morphology.

In the worst-case scenario with a significantly increased PE/PA film fraction of 30 % linked to a PA concentration of appr. 10%, a compatibilizer needs to be added in order to turn an inherently incompatible mixture of PE and PA components into a stable and homogeneous blend. On the basis of the result under these conditions, polyamide layers in coextruded structures with tie layer and compatibilizer in specified shares can even be classified as a recyclable material in mechanical recycling, as they clearly make a positive contribution to recyclate quality here.



These basic statements have been reproduced several times and with different film recipes or polyamide types and concentrations, and are also consistent with other investigations that have been produced in the meantime, such as those conducted by individual plastics manufacturers. One of these is the evaluation of a symmetrical PE/PA five-layer film with an integrated PA6/6.6 copolyamide by RecyClass.^(#19) Further examples include an investigation into the influence of PA or PP in polyethylene conducted on behalf of RecyClass at the Spanish plastics institute AIMPLAS^(#20) and studies by raw materials manufacturers both without^(#21) and including various compatibilizers.^(#22)

As such, there is a varied and robust body of technical evidence that refutes the initially mentioned and proven classification of all polyamides as non-recyclable contamination; this evidence provides a sound basis for reclassification.

The findings of the investigations, in combination with the properties of the barrier polymers, allow the conclusion that the compatibility of polyamide must be far higher than that of EVOH.

The proof of significant PA and EVOH fractions in “officially PA-free” commercial LDPE recyclates is worthy of note. The NIR sorting practiced is currently geared more toward identifying polyolefins and accumulations thereof in the respective target fractions, and less toward rejecting polyamide components. Accordingly, the results of this investigation support the retroactive official acceptance of multilayer film structures containing PA in a mechanical recycling process that is already being practiced!

In line with the requirements and evaluation standard of the Institute cyclos-HTP, the following evaluation of polyamide in PE-based packaging films will be conducted.

Coextruded film layers made of polyamide 6 (PA6) or copolyamide 6/6.6 (PA6/6.6) are “compatible for PE film recycling” into recyclates for injection molding and blown film applications if the following prerequisites are met:

- PA is used in combination with a maleic anhydride-grafted PE as a tie layer in a ratio of ≥ 0.5 g tie per g PA in a coextruded film.
- The tie layer must be specified by the manufacturer for use in the coextrusion of PA and PE.

Packaging films with a polyamide 6 (PA6) layer are “fully recyclable” when *all* of the following conditions are met:

- PA6 is used in combination with a maleic anhydride-grafted PE as a tie layer in a ratio of ≥ 0.5 g tie per g PA in a coextruded film.
- The film structure must *also* contain a compatibilizer such as Dow Fusabond E226 (or a comparable alternative) with a content of ≥ 0.15 g per g of PA.

This applies for film structures without print inks or other components that compromise recyclability. The corresponding materials were tested with ≤ 30 wt% PA6 in coextruded PE films, based on LDPE and LLDPE.

A corresponding recommendation for inclusion in the minimum standard has been formulated and submitted to the responsible committees.

In addition, further work regarding long-term studies of moisture absorption under various conditions, coextruded multilayer films based on PE/PA/EVOH and PA/PP and laminated composite structures containing PA are planned or already in progress. The expected results will be reported where appropriate in due course.



5. Literature/references

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