



We create chemistry

PE / PA – Multilayer films are mechanically recyclable !

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AMI Multilayer Flexible Packaging Europe 23.11.-25.11.2021

Melia Sitges, Barcelona, Spain

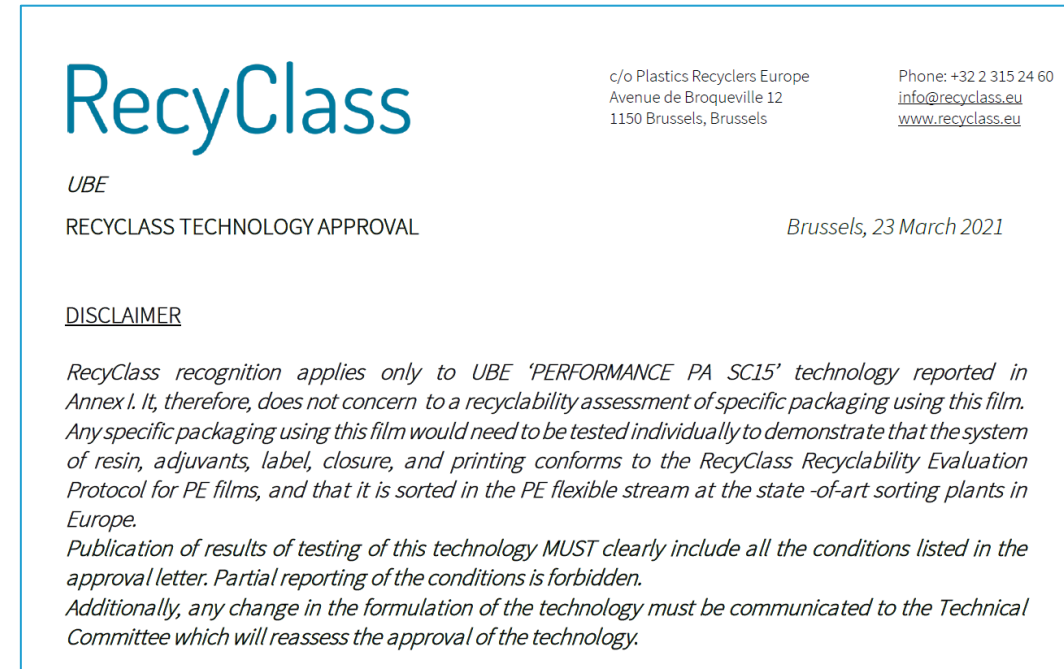
Agenda

- **Status Quo of Polyamide acceptance – Current status**
- **BASF evaluations overview**
- **Independent PA 6/PE multilayer film study carried out by cyclos-HTP**
- **Conclusions**
- **Status Quo of Polyamide acceptance – Outlook**

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Despite of available info confirming recyclability of PE/PA multilayer film structures.....



Taken from <https://recyclclass.eu>; published March 23rd, 2021
<https://recyclclass.eu/wp-content/uploads/2021/03/2020-PO-011-UBE-technology-approval-letter.pdf>

.....insufficient public / legislation opinions & rules do exist !

Packaging Preferred Materials & Formats Guidelines 2020. DATE: 13/02/20

Red <small>Not to be used as customers cannot easily recycle (UK)</small>		Amber <small>When functional requirements mean green materials are not an option</small>		Green <small>Preferred for UK recycling via kerbside or store</small>	
Materials Compostable/Bio degradable Oxy/Oxo Degradable PLA Polystyrene PVC PVdC (incl. layer within Complex Laminates) Plywood and MDF Water Soluble plastics	Formats & Designs Black Plastic* Composite Drums Expanded/Foamed/Density Modified Plastics Paper/board coated or laminated on both sides Glitter *Brands may choose to use black plastic for drink bottle caps	CONTROLLED USE: CONTACT THE PACKAGING TEAM FOR APPROVAL packaging.team@tesco.com Materials Non PE Flexible film incl. OPP Foiled paper New material innovations	Formats & Designs Beverage Cartons Multilayer Films/Complex Laminates Shrink sleeves (perforated, include messaging to remove, max coverage 40%)	Rigid Materials Glass (clear or of light colour) PET HDPE/LDPE pp** Steel & Aluminium Cardboard Flexible Materials Paper Glassine PE (recyclable at front of store)	Formats & Designs Cardboard with plastic: Max 15% plastic, one side of board only and be easily separated Mono lidding film matched to tray material when permanently attached Glass colour: Clear/Light colour preferred

Underlined denotes red material/format added in 2020 (Production to cease by end of 2020)
 **PP is widely recyclable, however may have a packaging tax implication for food grade if recycled content cannot be included



source: www.circularonline.co.uk/uploads

Appendix 3: overview of packaging groups/sorts and material-specific recycling incompatibilities

Group/sort	Incompatibilities
Film and LDPE	Glued cellulose-based labels that cannot be removed in cold washing; PA layers, PE-X components, PVDC layers, other non-PE polymeric layers (excluding adhesion promoters, adhesives, PP, EVA and EVOH), non-polymeric layers (excluding SiOx/AlOx/metallisations)
Rigid PE	Silicone components; components of foamed non-thermoplastic elastomers; glued cellulose-based labels that cannot be removed in cold washing; PET sleeves with a density of < 1g/cm ³ ; PA layers, PE-X components, PVDC layers; non-PE plastics with a density of < 1 g/cm ³ .
Rigid PP	Silicone components; components of foamed non-thermoplastic elastomers; glued cellulose-based labels that cannot be removed in cold washing; PET sleeves with a density of < 1g/cm ³ ; PA layers; PVDC layers; non-PE plastics with a density of < 1 g/cm ³ .

Excerpt from „Minimum standard for determining the recyclability of packaging subject to system Participation pursuant to section 21 (3) VerpackG“ of Stiftung Zentrale Stelle Verpackungsregister, Germany 2021

VI. Anhang Übersicht zu den gängigsten Verpackungsmaterialien

Barriere- und Siegelmaterialien

Bezeichnung *	Erscheinungsbild des Ausgangsmaterials (d.h. ungefärbt bzw. unlackiert)	mechanische Eigenschaften	Barriere Eigenschaften			Verwendung
			H ₂ O	O ₂	CO ₂	
PA Polyamid 6 und Polyamid 12	* • Transparent bei geringer Wandstärke	• Steif • Flexibel und dehnbar	-	-	-	Getränkflaschen, thermogeformte Schalen, auch als Deckelfolie
EVOH Ethylenvinylalkohol Copolymer	✓ • Transparent bei geringer Wandstärke	-	✗ bis ○	✓	✓	Barriere-Schicht in PP- oder PE-Verpackungen (Flaschen, Dosen, Schalen, etc.)

Excerpt from „Leitfaden für nachhaltigere Verpackungen, Version 2.0“
 Aldi / Reclay Group, Germany 2020



Why it's important to demonstrate recyclability of PE/PA blends ?

Polyamide (PA) containing flexible films

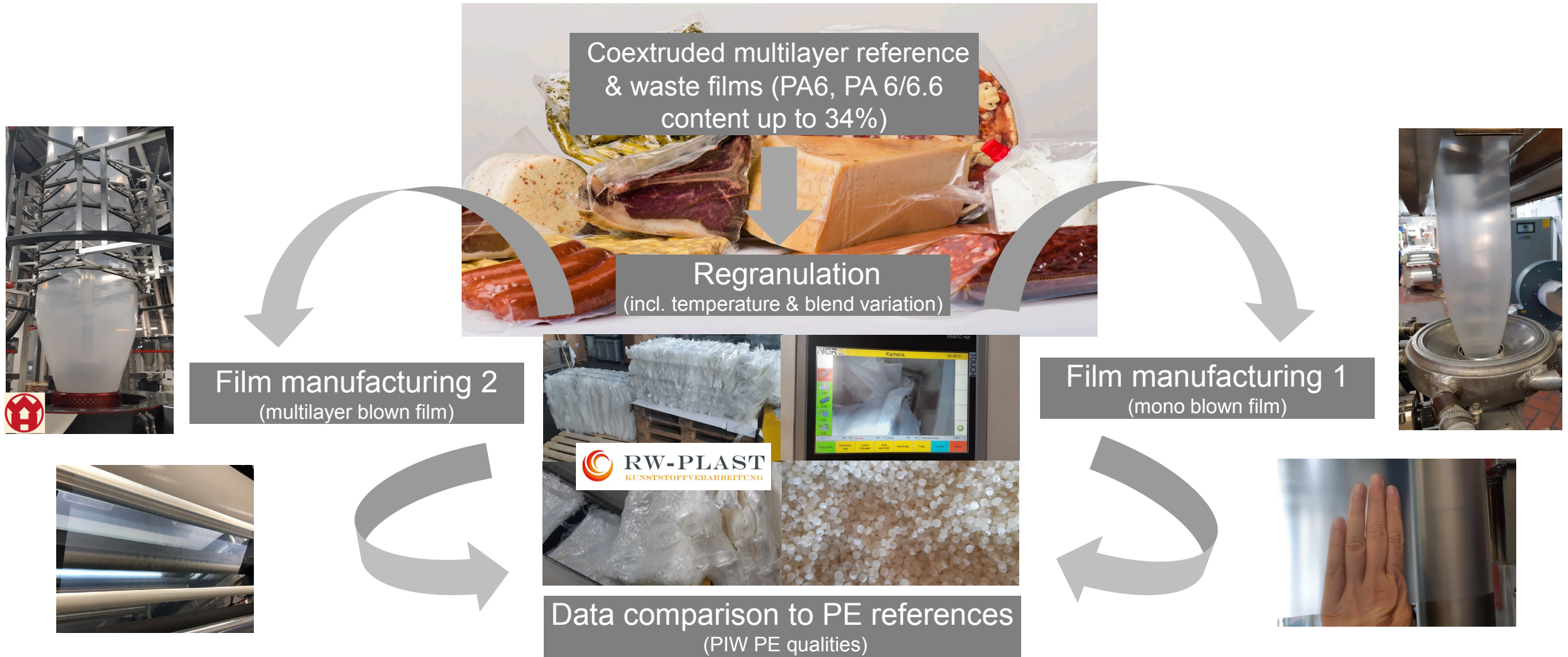
- provide unique performances for many applications
- offer significant down gauging potentials linked to
 - less use of resources
 - less use of plastics packaging
 - less waste
- Allow optimized shelf lives for saving and protecting food
- represent real sustainable packaging solutions !

It does not make any sense to sacrifice these advantages for a „(mechanical) recycling for the sake of (mechanical) recycling only“ approach !

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What has been investigated ?



partially a joint project



Film structures I (example PA 6)

- **Trial 1.0 (Reference): PE/PE/PE/PE/PE/PE/PE/PE/PE**
- **Trial 1.1 (StructureA): PE/PE/PE/tie/B40LN/tie/PE/PE/PE (20% B40LN*)**
- **Trial 1.2 (StructureR): PE/PE(incl. CompR)/PE/tie/B40LN/tie/PE/PE(incl. CompR)/PE (20% B40LN, 2 x 2,5% Retain 3000**)**
- **Trial 1.3 (StructureF): PE/PE(incl. CompF)/PE/tie/B40LN/tie/PE/PE(incl. CompF)/PE (20% B40LN, 2 x 2,5% Fusabond E226***)**

*** Ultramid® B40LN = high viscosity (RV = 4,0), nuleated PA6, source: BASF SE**

**** & *** 2 different types of compatibilizers, source: Dow/DuPont**

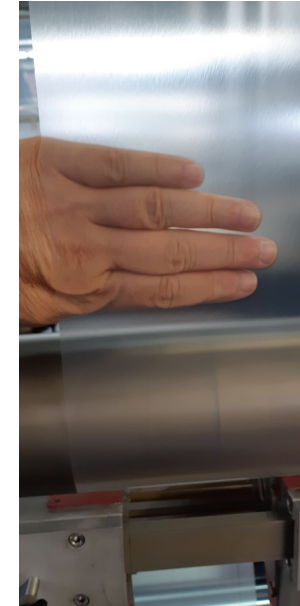
PA* concentrations < 10% are dispersible in a polyethylene stream w/o using compatibilizers



- PA*/PE pellet mix (PA*: 5-8%)
- NO compatibilizer
- Temperature range 215-240°C
- NO additional compounding
- NO additional drying



Direct processing
of regranulate to 2nd blown film

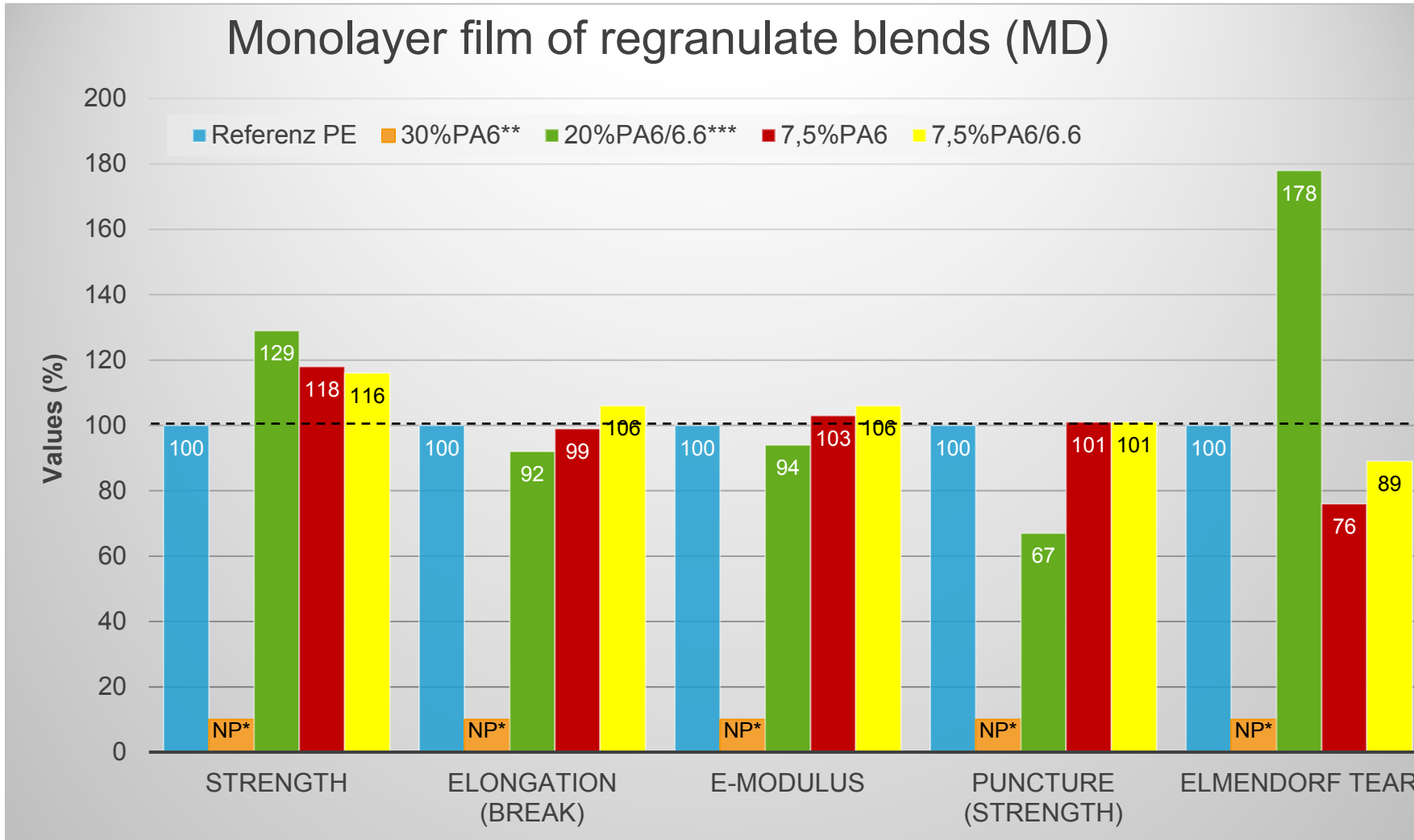


Transparent films
showing reasonable
mechanical performance
(mono- / multilayer films)

* including PA 6 (e.g. Ultramid® B40LN) or PA 6/6.6 (e.g. Ultramid® C33, C40L or C37LC)

Comparison of mechanical properties in machine direction (MD)

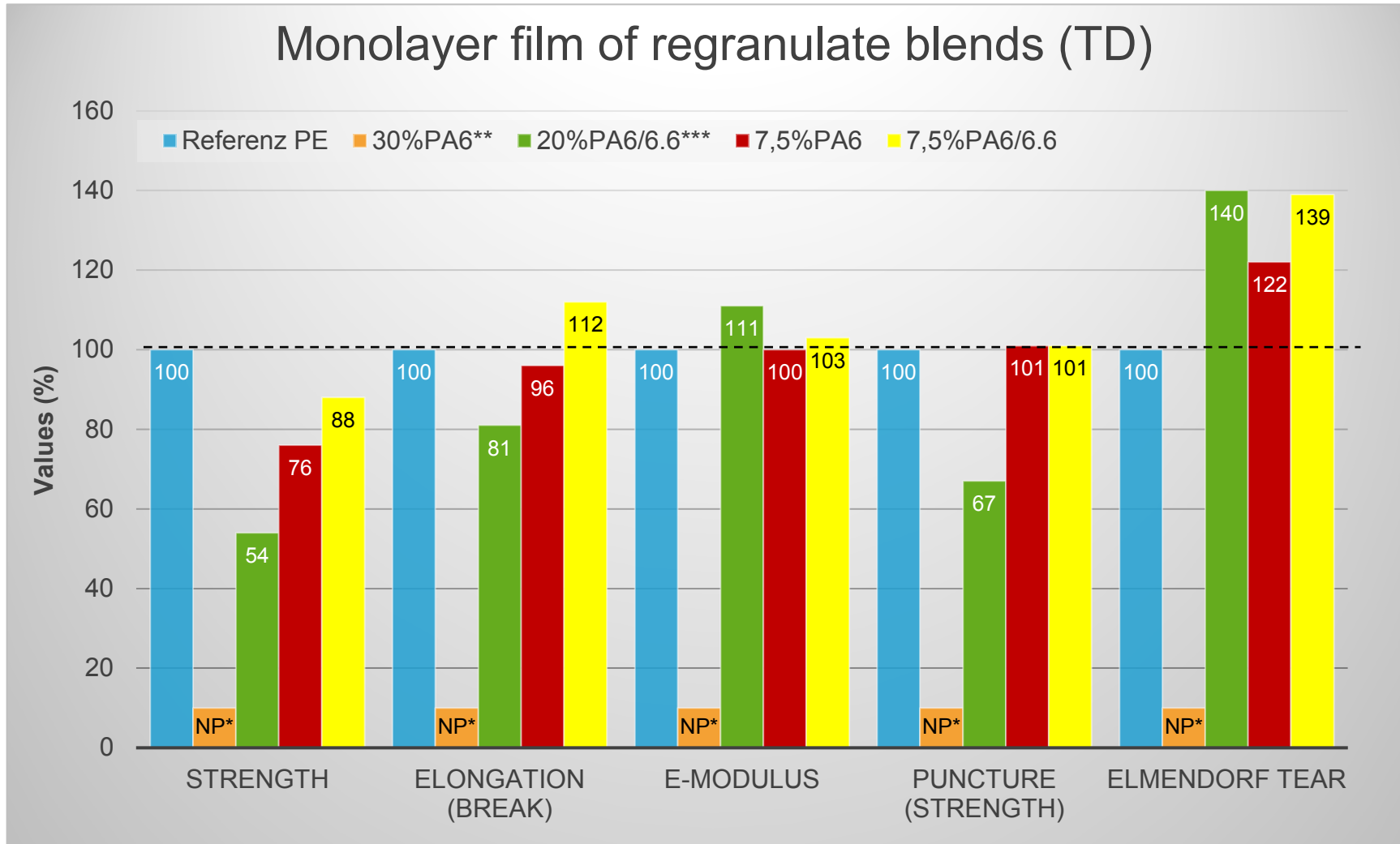
(Dilution w/o compatibilizer, just an example)



*NP = not processable
 **PA 6 = Ultramid® B40LN
 ***PA 6/6.6 = Ultramid® C40L
 All components mixed as salt & pepper blends prior to blown film process directly!!

Comparison of mechanical properties in transversal direction (TD)

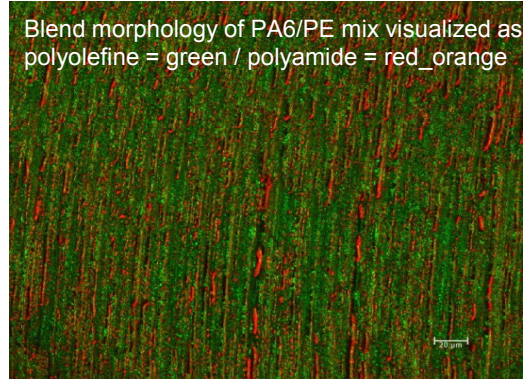
(Dilution w/o compatibilizer, just an example)



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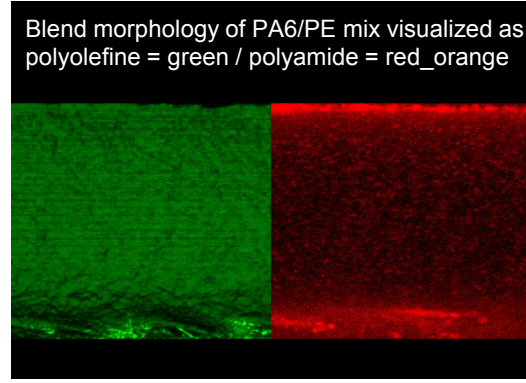
Compatibilization offers additional options for utilizing PA*/PE waste blends with PA* concentrations > 10% in a polyethylene stream

PA*/PE mix pur (pellet, 20% PA6)

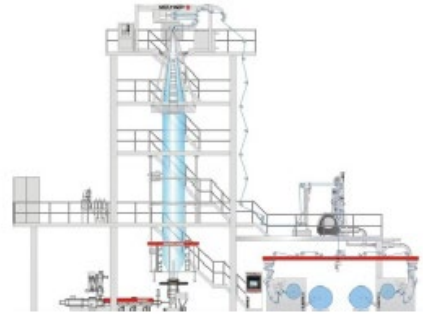
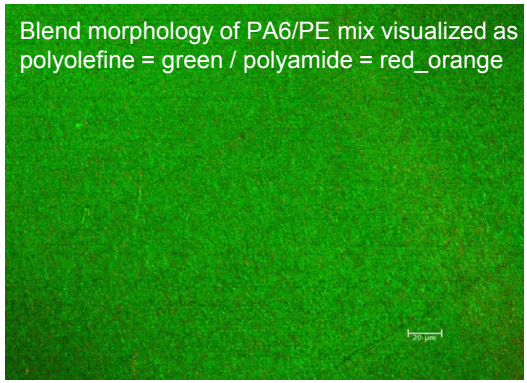


Compatibilizer added as **dry blend** during 2nd blown film process

Monolayer PA*/PE blown film

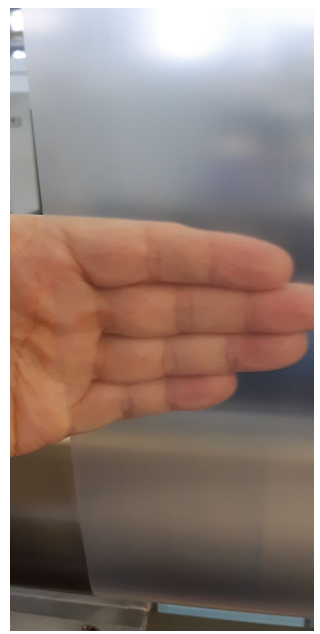
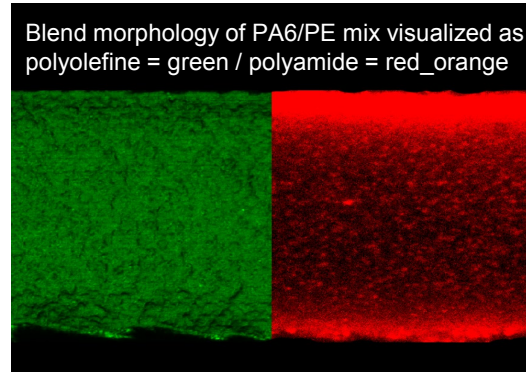


PA*/PE mix, pre-compatibilized (pellet, 20% PA6)



Direct processing of **pre-compatibilized** PIR during 2nd blown film process

Monolayer PA*/PE blown film



Transparent films showing reasonable mechanical performance (mono-/multilayer films)

* including PA 6 (e.g. Ultramid® B40LN) or PA 6/6.6 (e.g. Ultramid® C33, C40L or C37LC)



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✓ **Targets of this project:**

- Investigation of the effects of PA6 (most critical PA grade) on the relevant properties of recyclates in LDPE film recycling
 - PA6 is incorporated **as coextruded layer with tie layers** in a LLDPE/LDPE film
 - Additional tests of films with PA6 and a commercial **compatibilizer** blended in PE
- Tests of the reference applications for LDPE recyclates:
 - Blown films
 - Injection moulding

→ **Assessment of PA6 as “Recycling Compatible” or “Recyclable” in LDPE films (by demand with use of a compatibilizer)**

✓ **Reference Processes**

Relevant Processes in the corresponding Recycling Path incl. **Recyclate Applications**

✓ **Reference Material**

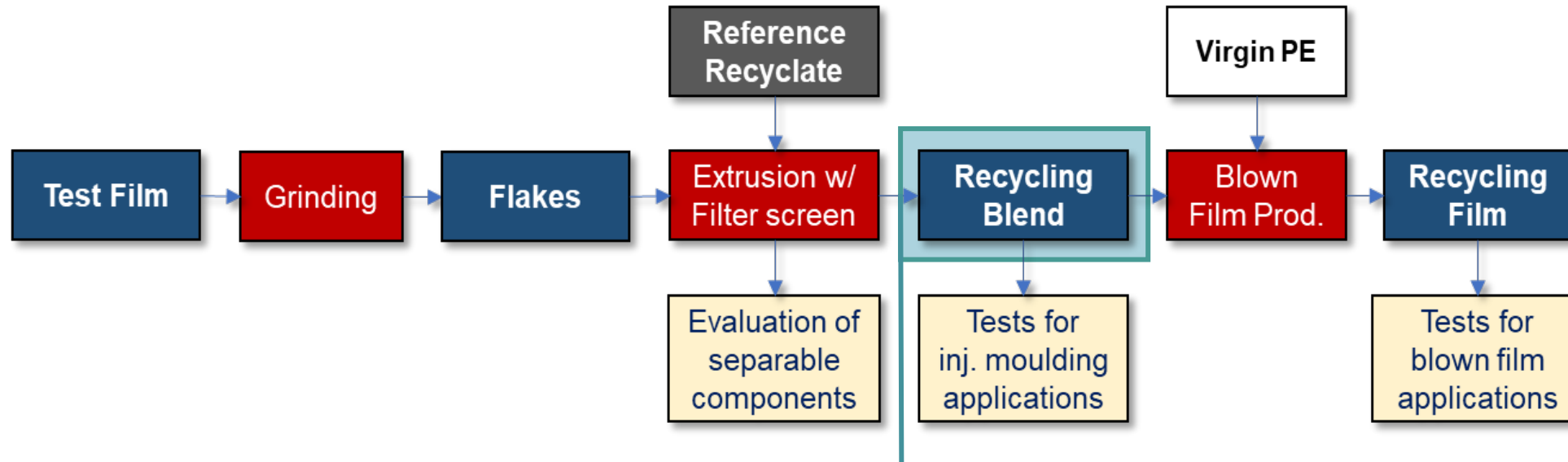
Commercial PCR recyclate commonly used for the same reference applications in the Recycling Path

✓ **Sample**

Packaging/Material containing a component with an **unknown recycling compatibility**



Recycling Application Compatibility Test for PE-based Flexible Packaging (CHI-C8-PEF-1)



Assessment of Recycling Blends *against Reference material*:

- CHI5** “Realistic Scenario” for the concentration of a packaging material in the recycling stream = **5% Sample** + **95% Reference** → **Compatibility of Packaging or Material?**
- CHI30** “Worst Case Scenario” for the concentration of a packaging material in the recycling stream = **30% Sample** + **70% Reference** → **100% Recyclability of Packaging ?**
- REF** Commercial **PCR Recyclate** from the recycling stream

Multilayer Blown Films produced at Windmüller & Hölscher

7 layer film structures:

FB0 – PE / PE / PE / PE / PE / PE / PE (100 µm)

FB1 – PE / Tie layer / **PA6** / Tie layer / **PA6** / Tie layer / PE

FB2 – PE + **Comp** / Tie layer / **PA6** / Tie layer / **PA6** / Tie layer / PE + **Comp**

“Recycling-critical” materials to be tested:

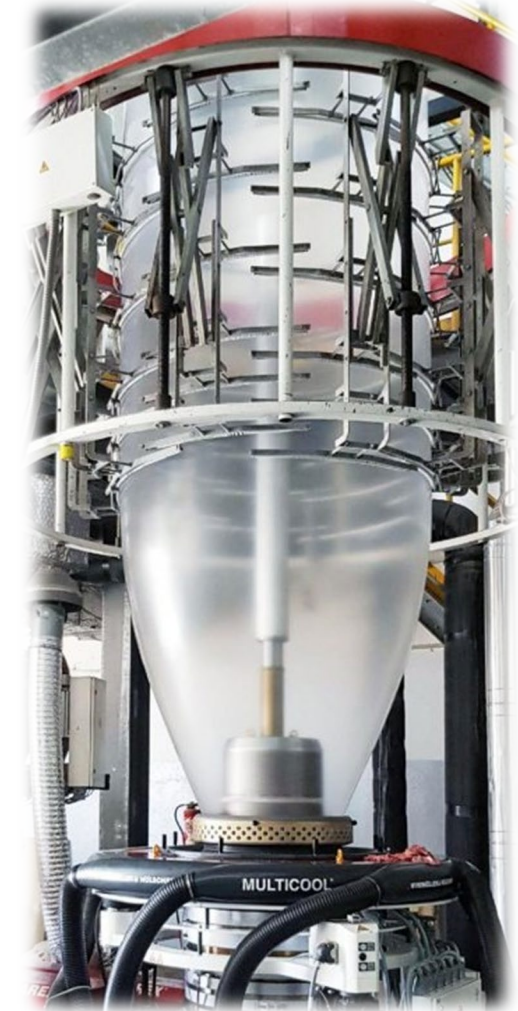
LDPE-based film with **30% PA6** and **15% tie layer resin ***

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

LDPE-based film with **30% PA6** and **15% tie layer resin *** and **5% Compatibilizer ****

** Maleic anhydride grafted PE (Standard grade for PA/PE coextrusion)*

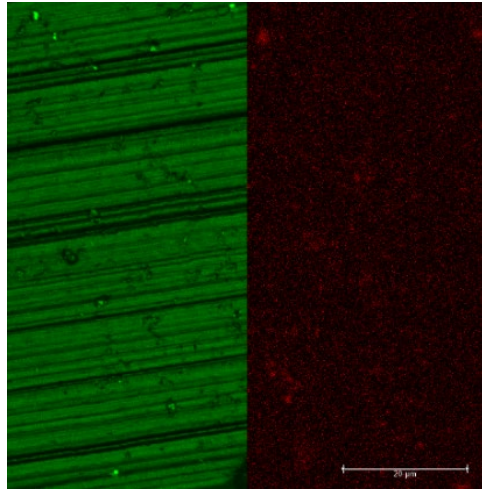
*** Maleic anhydride grafted PE (high MAH content)*



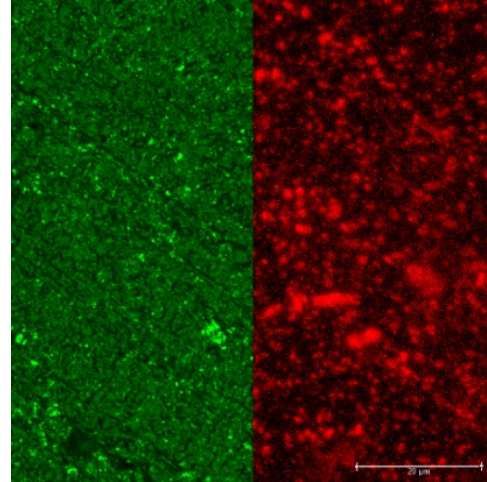
Morphology of Regranulate pellets after Regranulation on a NGR line



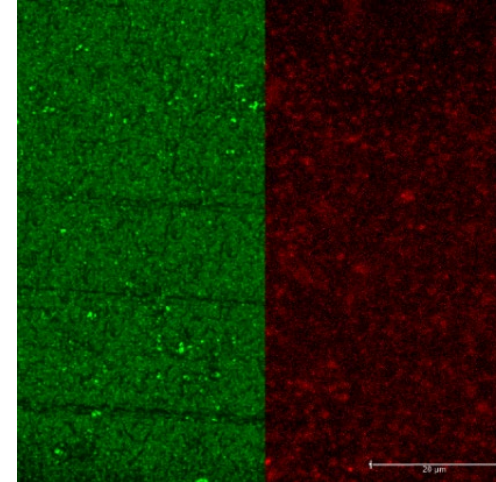
Regranulate,
100% PE reference



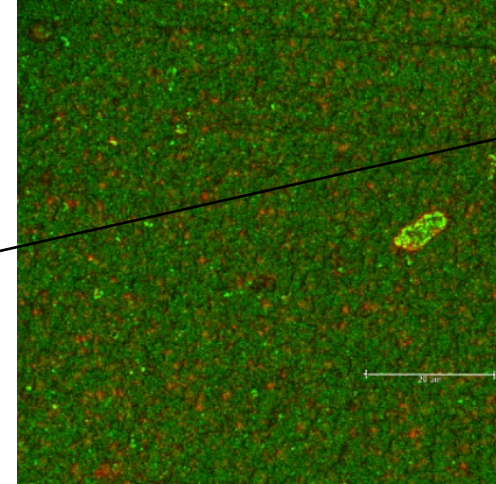
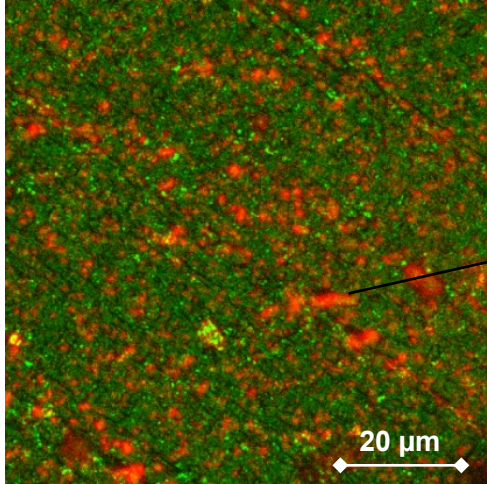
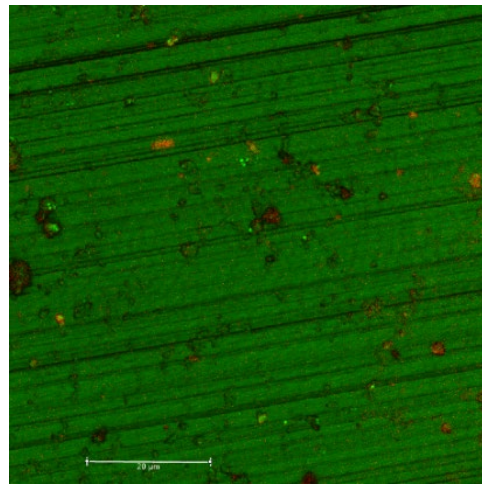
Regranulate w/
30% PA6, 15% Tie



Regranulate w/
30% PA6, 15% Tie,
5% Compatibilizer



Pre-compatibilization generates a **more homogeneous morphology** in comparison with 30% PA6 without compatibilizer



PA phases up to 10 µm

Application Test for Blown Films



- Recyclate materials not dried !
- 50% virgin LD/LLDPE added for each film (Standard in industry)



5% PA, no Comp.*, 215°C

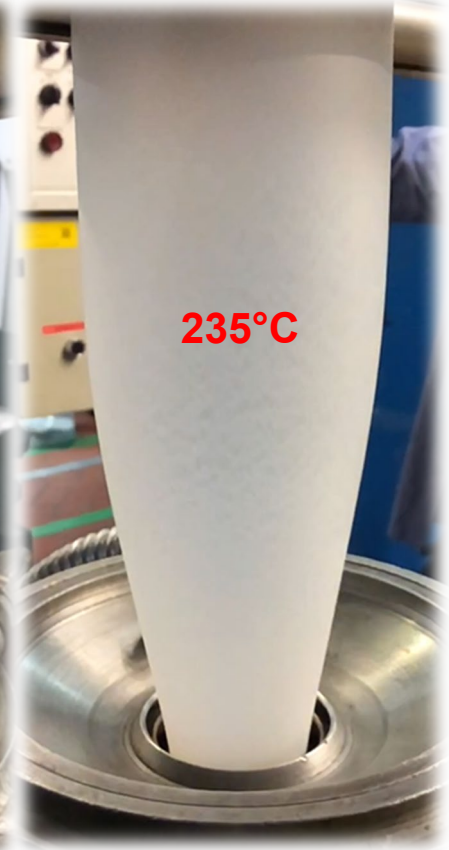


15% PA, no Comp.*, 215°C



215°C

15% PA, 2.5% Comp.*

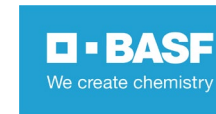


235°C

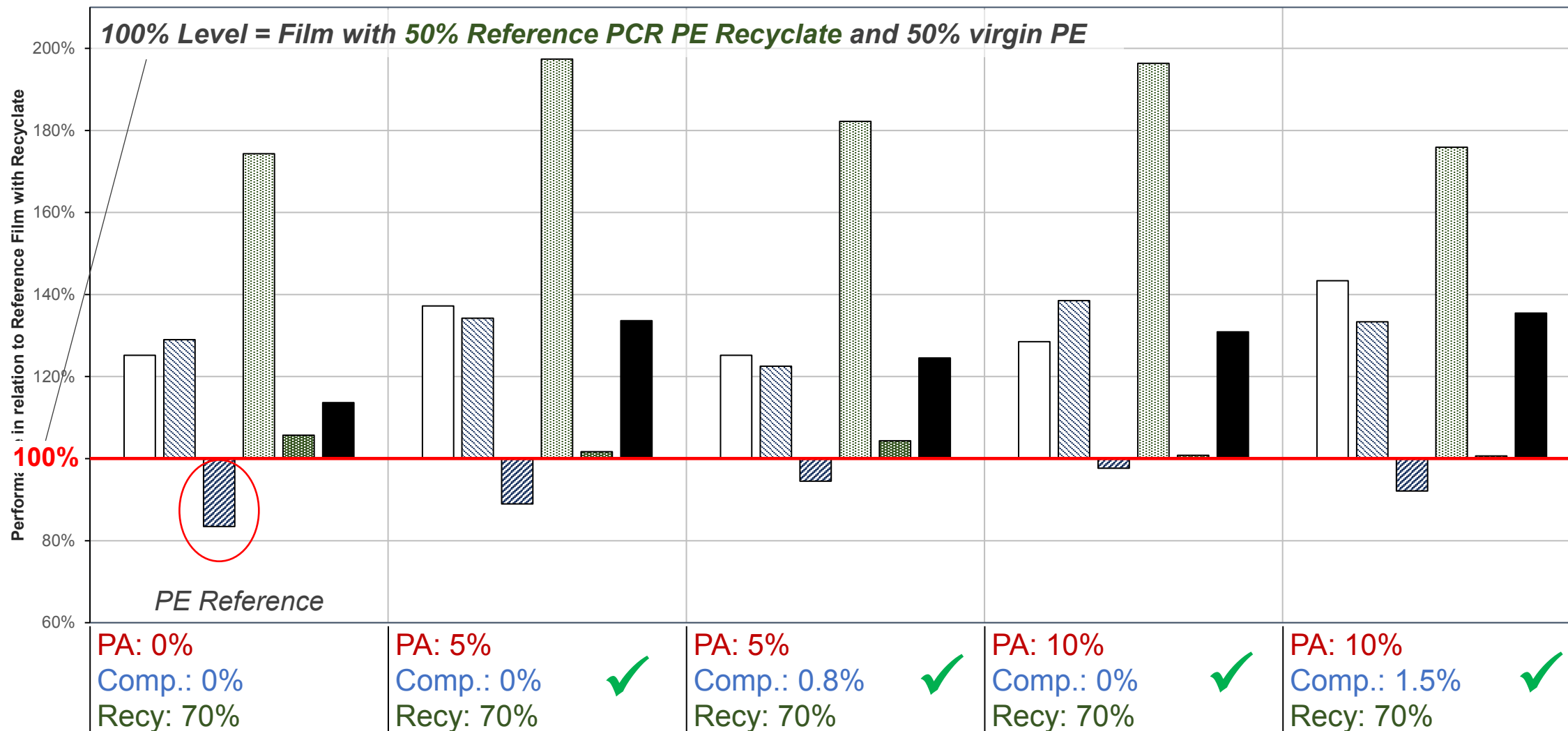
* Comp. = Compatibilizer

Blown Film Application Tests – Mechanical and Sealing Properties

Blown Films made of 50% Recyclates and 50% virgin PE - Mechanical and Sealing properties



□ Dart Drop
 ▨ Tensile Test MD
 ▩ Tensile Test CD
 ▧ Elongation @ break MD
 ▦ Elongation @ break CD
 ■ Sealing strength



Blown Film Application Tests – Seam Tightness Test (CHI-C8-BFPE)

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1. Films sealed with seam width of 2 - 4 mm
2. Filled with 1.3 L of water (with 16 cm seam length and 20 cm fill height)
3. Test duration: min. 10 minutes
4. Count the number of drops per minute due to leakages in the seam
5. Test is passed with < 2 drops per minute

**All Sample Films
passed the test ✓**



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Major conclusions

- PE/PA* waste mixtures are recyclable with PA* contents of **< 10% without using compatibilizers**
- **Compatibilizers** are an efficient way to consistent PE/PA* mixtures with PA* contents of **>> 10%**
- Compatibilizers might be incorporated
 - already in original multilayer film structures (**pre – compatibilization**)
 - **during regranulation** by using standard single screw machinery
 - by adding as individual component **on final processing**
- While PE/PA6 films need a regranulation temperature of 220-240°C, **realistic PA blends** in packaging streams (PA6 + several lower melting CoPolyamides) **are processable at about 210°C**
- **NO drying steps** were necessary during our trials
- PE/PA* mixtures are suitable for manufacturing mono-layer and multilayer structures, e.g. using highly concentrated PE/PA* mix core layers, **showing sufficient mechanical and optical properties while using standard single screw extrusion equipment !**

* including PA 6 (Tmelt 215-220°C) and different CoPolyamides (Tmelt 182-200°C) in coextruded multilayer structures

Classification of Materials and Packaging structures for Recycling



Status	Categorization of PA6 & PA6/6.6 (acc. CHI standard) 	CHI Assessment of a Packaging structure Example: PE film with 10% PA6 + ≥ 5% Tie layer	Material (PA6) according to ZSVR Minimum Standard
Current	CAT 3 (PA in any structure)	Not recyclable (0%)	Contaminant / Incompatible (“PA layers”)
Future (Based on results)	CAT 2 (PA6 layers with tie layer)	Recyclable (≤ 90%)** (no Compatibilizer)	Recycling compatible (“PA 6 layers with tie layer”)*
	Valuable material (PA 6 layers with tie layer and Compatibilizer)	Fully Recyclable (≤ 100%)** (Structure with Compatibilizer)	



Still in progress.....!!

* Application still in progress for the next Minimum Standard 2022

** Structure without printing inks or other components that affect recyclability, practiced by Institut cyclos-HTP for individual evidence test according to chapter 4.3 of Minimum standard 2021

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First official certificates received.....

CERTIFICATE

Recycling Compatibility of Packaging Material

BASF SE
 Carl-Bosch-Strasse 38
 D-67056 Ludwigshafen am Rhein, Germany

The company receives the certification of recycling compatibility for the following plastic material.

Designation

Polyamide 6 (PA6)
 as layer in coextruded PE films, based on LDPE and/or LLDPE;
 in combination with ≥ 0.5 g per g PA of maleic anhydride-grafted PE as tie layer specified for PA/PE
 tested with $\leq 5\%$ by weight of PA6 in a PCR LDPE recyclate

Test result

Assessment via path/specification: Path 1: Plastic films / LDPE

Recyclate (final product): LDPE Regranulate

Test standard / scope of application:
 Requirements and assessment catalogue of the Institute cyclos-HTP for EU-wide certification (state: 07.10.2019) /
 Scope of validity according to nation states, see chapter 1
 DIN EN 13430 with regard to material recyclability in the post-use phase; also integrated
 The following reference processes, materials and applications are taken into consideration within the certification process:
 • Detection as feasible PE packaging in state-of-the-art sorting plants
 • Recyclate use for injection moulding and blown film applications
 • Test program based on CHI test protocol CHI-C8-PEF-12.0 with the use of PCR-based LDPE recyclate as reference

According to the CHI standard the plastic material is no contaminant in the tested application and in the above-mentioned material combination and can be considered as:

Recycling Compatible for PE Film Recycling
 (AT, DE, ES, FR, IT, NL, NO)

This certificate (No. 2187-2021-002124) is valid until the **31.05.2022** (1 year upon issue) relating to the countries identified in the assessment report. This certificate will lose validity in case of qualitative or quantitative changes of packaging components.

Aachen, dated 17.05.2021

Dr. Joachim Christiani
 Publicly appointed and sworn expert for the IHK for packaging waste recycling
 Competent authority: IHK Aachen

Dr. Roland Bothor
 Technical supervisor of the Investigations

Institute cyclos - HTP
 Institute cyclos-HTP GmbH
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CERTIFICATE

Recyclability of a Packaging Material Group

BASF SE
 Carl-Bosch-Strasse 38
 D-67056 Ludwigshafen am Rhein, Germany

The company receives the certification of recyclability for the following packaging materials.

Designation

LDPE-based multilayer packaging films with Polyamide 6 (PA6)
 including $\leq 30\%$ by weight of PA6;
 in coextruded polyethylene films, based on LDPE and/or LLDPE; packaging size $\geq A5$;
 with ≥ 0.5 g per g PA of maleic anhydride-grafted PE as tie layer specified for PA/PE;
 not including printing inks, adhesives or other components that can affect the recyclability

Test result

Assessment via path/specification: Path 1: Plastic films / LDPE

Recyclate (final product): LDPE Regranulate

Test standard / scope of application:
 Requirements and assessment catalogue of the Institute cyclos-HTP for EU-wide certification (state: 07.10.2019) /
 Scope of validity according to nation states, see chapter 1
 DIN EN 13430 with regard to material recyclability in the post-use phase; also integrated
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 • Detection as feasible PE packaging in state-of-the-art sorting plants
 • Recyclate use for injection moulding and blown film applications
 • Test program based on CHI test protocol CHI-C8-PEF-12.0 with the use of PCR-based LDPE recyclate as reference

According to the CHI standard the recyclability of the packaging component amounts to:

$\geq 70\%$ (AT, DE, ES, FR, IT, NL, NO)

This certificate (No. 2187-2021-002126) is valid until the **31.05.2022** (1 year upon issue) relating to the countries identified in the assessment report. This certificate will lose validity in case of qualitative or quantitative changes of packaging components.

Aachen, dated 17.05.2021

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 with ≥ 0.5 g per g PA of maleic anhydride-grafted PE as tie layer specified for PA/PE;
 with ≥ 0.15 g per g PA of Dow Fusabond E226 or chemically comparable compatibilizer in a neighbouring PE layer of the structure; not including printing inks, adhesives or other components that can affect the recyclability

Test result

Assessment via path/specification: Path 1: Plastic films / LDPE

Recyclate (final product): LDPE Regranulate

Test standard / scope of application:
 Requirements and assessment catalogue of the Institute cyclos-HTP for EU-wide certification (state: 07.10.2019) /
 Scope of validity according to nation states, see chapter 1
 DIN EN 13430 with regard to material recyclability in the post-use phase; also integrated
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 • Detection as feasible PE packaging in state-of-the-art sorting plants
 • Recyclate use for injection moulding and blown film applications
 • Test program based on CHI test protocol CHI-C8-PEF-12.0 with the use of PCR-based LDPE recyclate as reference

According to the CHI standard the recyclability of the packaging component amounts to:

100% (AT, DE, ES, FR, IT, NL, NO)

This certificate (No. 2187-2021-002126) is valid until the **31.05.2022** (1 year upon issue) relating to the countries identified in the assessment report. This certificate will lose validity in case of qualitative or quantitative changes of packaging components.

Aachen, dated 17.05.2021

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.....more will follow

Appendix 3: overview of packaging groups/sorts and material-specific recycling incompatibilities

Group/sort	Incompatibilities
Film and LDPE	Glued cellulose-based labels that cannot be removed in cold washing; PA layers, PE-X components, PVDC layers, other non-PE polymeric layers (excluding adhesion promoters, adhesives, PP, EVA and EVOH), non-polymeric layers (excluding SiOx/AlOx/metallisations)
Rigid PE	Silicone components; components of foamed non-thermoplastic elastomers; glued cellulose-based labels that cannot be removed in cold washing; PET sleeves with a density of < 1g/cm ³ ; PA layers, PE-X components, PVDC layers; non-PO plastics with a density of < 1 g/cm ³ .
Rigid PP	Silicone components; components of foamed non-thermoplastic elastomers; glued cellulose-based labels that cannot be removed in cold washing; PET sleeves with a density of < 1g/cm ³ ; PA layers; PVDC layers; non-PO plastics with a density of < 1 g/cm ³ .

Excerpt from „Minimum standard for determining the recyclability of packaging subject to system Participation pursuant to section 21 (3) VerpackG“ of Stiftung Zentrale Stelle Verpackungsregister, Germany 2021

Changes for minimum standard 2022 still in progress !!!!

Certification of Recyclability of PE/PA multilayer film structures available by cyclos-HTP !!!

(individual evidence test acc. to chapter 4.3 of minimum standard)



4.3 Recycling incompatibilities

The declaration of the recyclability of a packaging requires that no combinations of materials or substances are used that can impede a successful recycling. **Appendix 3 ('Overview of packaging groups/sorts and material-specific recycling incompatibilities')** provides the basis for determining incompatibilities. For any deviating determination in the sense that incompatible substances do not negatively affect recyclability, individual evidence produced through analytical testing must be provided.

Excerpt from „Minimum standard for determining the recyclability of packaging subject to system Participation pursuant to section 21 (3) VerpackG“ of Stiftung Zentrale Stelle Verpackungsregister, Germany 2021



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