PE / PA – Multilayer films are mechanically recyclable!

Dr. Rolf-Egbert Gruetzner, BASF SE
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......

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**Disclaimers**

**UBE**

**RECYCLASS TECHNOLOGY APPROVAL**

Brussels, 23 March 2021

**Disclaimer**

RecyClass recognition applies only to UBE "PERFORMANCE PA SC15" technology reported in Annex I. It, therefore, does not concern to a recyclability assessment of specific packaging using this film. Any specific packaging using this film would need to be tested individually to demonstrate that the system of resin, adjuvants, label, closure, and printing conforms to the RecyClass Recycling Compatibility Evaluation Protocol for PE films, and that it is sorted in the PE flexible stream at the state-of-art sorting plants in Europe.

Publication of results of testing of this technology MUST clearly include all the conditions listed in the approval letter. Partial reporting of the conditions is forbidden.

Additionally, any change in the formulation of the technology must be communicated to the Technical Committee which will reassess the approval of the technology.

Taken from [https://recyclaseu.eu](https://recyclaseu.eu); published March 23rd, 2021


Red
- Not to be used as customers cannot easily recycle (UK)
- Materials: Compostable/Bio degradable
- Formats & Designs: Black Plastic
- Composite Drums
- Expanded/ Foamed/Density Modified Plastics
- Paper/board coated or laminated on both sides
- Glitter

Amber
- When functional requirements mean green materials are not an option
- Materials: Non PE Flexible film incl. OPP
- Foiled paper
- New material innovations
- Formats & Designs: Beverage Cartons
- Multilayer Films/Complex Laminates
- Shrinks sleeves (perfector, include messaging to remove, max coverage 40%)

Green
- Preferred for UK recycling via kerbside or store
- 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

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

<table>
<thead>
<tr>
<th>Group/sort</th>
<th>Incompatibilities</th>
</tr>
</thead>
<tbody>
<tr>
<td>Film and LDPE</td>
<td>PA layers</td>
</tr>
</tbody>
</table>
- Glass cellulose-based labels that cannot be removed in cold washing; PET layers with a density of <1 g/cm³; |
| Rigid PE | 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/metalisations) |
- Silicone components; components of formalized non-thermo-plastic elastomers; glued cellulose-based labels that cannot be removed in cold washing, PET layers with a density of <1 g/cm³; |
- Silicone components; components of formalized non-thermo-plastic elastomers; glued cellulose-based labels that cannot be removed in cold washing, PET layers with a density of <1 g/cm³; |
- Non PE plastics with a density of <1 g/cm³; |

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

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

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

VI. Anhang

Übersicht zu den gängigsten Verpackungsmaterialien

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

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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 lifes 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!
Agenda

- Status Quo of Polyamide acceptance – Current status
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What has been investigated?

- Coextruded multilayer reference & waste films (PA6, PA 6/6.6 content up to 34%)
- Regranulation (incl. temperature & blend variation)
- Data comparison to PE references (PIW PE qualities)

Film manufacturing 1 (mono blown film)
Film manufacturing 2 (multilayer blown film)

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

* 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)

Monolayer film of regranulate blends (MD)

*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)

Monolayer film of regranulate blends (TD)

Comparison of mechanical properties in transversal direction (TD)
(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!!
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)

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

Compatibilizer added as dry blend during 2nd blown film process

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

- Blend morphology of PA6/PE mix visualized as polyolefine = green / polyamide = red_orange

Monolayer PA*/PE blown film

- Blend morphology of PA6/PE mix visualized as polyolefine = green / polyamide = red_orange

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

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

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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
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

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Test Phase 1 – Production of Sample Blown Films – Raw materials

Multilayer Blown Films produced at Windmöller & 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
15% PA, 2.5% Comp.*

* 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

100% Level = Film with 50% Reference PCR PE Recyclate and 50% virgin PE

PE Reference

<table>
<thead>
<tr>
<th>PA: 0%</th>
<th>Comp.: 0%</th>
<th>Recy: 70%</th>
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<th>Comp.: 0.8%</th>
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<th>Comp.: 0%</th>
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Blown Film Application Tests – Seam Tightness Test (CHI-C8-BFPE)

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
<table>
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<tr>
<th>Status</th>
<th>Categorization of PA6 &amp; PA6/6.6 (acc.CH1 standard)</th>
<th>CHI Assessment of a Packaging structure</th>
<th>Material (PA6) according to ZSVR Minimum Standard</th>
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</thead>
<tbody>
<tr>
<td><strong>Current</strong></td>
<td>CAT 3 (PA in any structure)</td>
<td>Not recyclable (0%)</td>
<td>Contaminant / Incompatible (“PA layers”)</td>
</tr>
<tr>
<td></td>
<td><strong>Future</strong> (Based on results)</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>CAT 2</strong></td>
<td>CAT 2 (PA6 layers with tie layer)</td>
<td>Recyclable (≤ 90%)**</td>
<td>Recycling compatible (“PA 6 layers with tie layer”)*</td>
</tr>
<tr>
<td></td>
<td>Valuable material (PA 6 layers with tie layer and Compatibilizer)</td>
<td>Fully Recyclable (≤ 100%)** (Structure with Compatibilizer)</td>
<td></td>
</tr>
</tbody>
</table>

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……
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)

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