MultiRec: Effects of multiple extrusion in cast and blown film applications

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Agenda

- Problem Description
- Application 1: stretch wrapping films
- Application 2: blown extrusion of sealing films

Problem description

- Stretch wrapping of pallets: prevents sliding + tipping over
- Seal films (non-food): protects during storage + handling
- Legislation & eco-modulation
  - Packaging and packaging waste directive sets targets for recycling[1]
    - Minimum percentages of post-consumer recycled content in 'other plastic packaging' 2030 ≥ 35%; 2040 ≥ 65%
  - Plastic tax[2]
    - Applies to plastic packaging with less than 30% recycled content produced or imported in the UK
  - Bonus myRecycledContent[3]
    - €50 bonus/ton recycled material, ≥ 30% PCR content
- ≠multiple cycles x ≠recycled content: Impact?
  - Cast/blown film extrusion, composition, film performance

Use of multi-processed polyethylene is inevitable

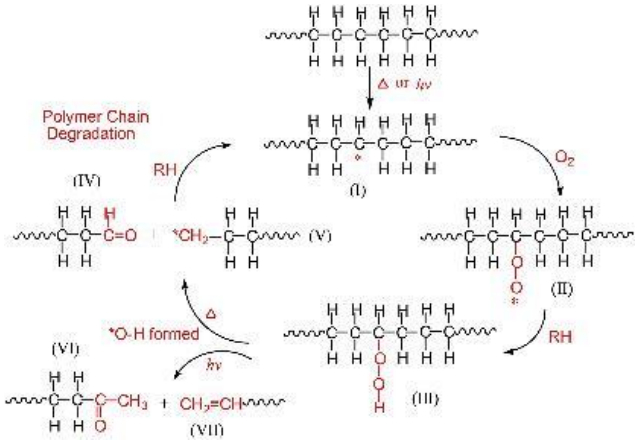
[1]: https://www.europarl.europa.eu/RegData/etudes/BRIE/2023/745707/EPRS_BRI(2023)745707_EN.pdf + https://valipac.academy/en/modules/gaining-a-better-understanding-of-the-legislation/packaging-and-packaging-waste-directive/
[2]: https://www.gov.uk/guidance/check-if-you-need-to-register-for-plastic-packaging-tax#:~:text=The%20tax%20came%20into%20force,tonne%20from%201%20April%202024
[3]: https://valipac.academy/nl/de-modules/je-aangifte-invullen/bonus-myrecycledcontent/

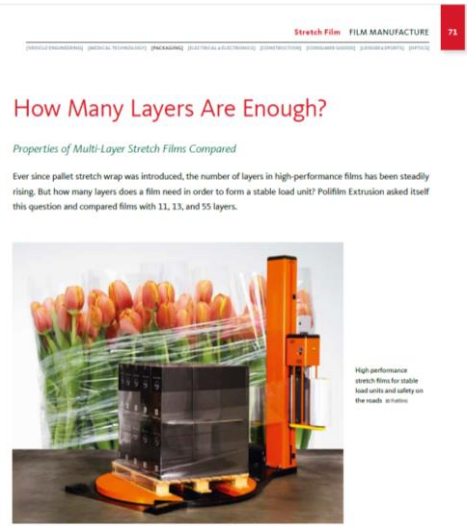
Application 1
Stretch wrapping films

State of the art

- Mitigation strategies (currently applied in the industry) are *trial-and-error* based, relying on individual experiences
  - Addition of antioxidants to prevent thermal degradation
  - Blending with virgin materials or increasing film thickness
  - Using virgin material in outer layers to improve machinability

- Stretch film = multilayer structure of max. 23 µm
  Cast coextrusion of LLDPE




Stretch Film FILM MANUFACTURE 71

How Many Layers Are Enough?

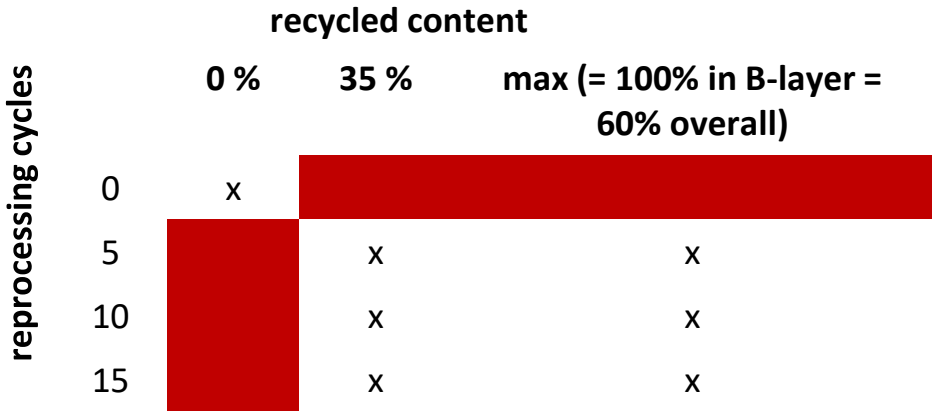
Properties of Multi-Layer Stretch Films Compared

Ever since pallet stretch wrap was introduced, the number of layers in high-performance films has been steadily rising. But how many layers does a film need in order to form a stable load unit? Polifilm Extrusion asked itself this question and compared films with 11, 13, and 55 layers.

https://res.cloudinary.com/sternwald-systems/raw/upload/v1/hugoprd/ARTIKEL_ATTACH/002425B0_F135DF8CFF24/a994e7a653f58a618fcd2c6f3fcd049f52ecf027/KUint_2017_10_How-Many-Layers-Are-Enough.pdf; 10.1002/app.40239, 10.1177/875608702035146; https://doi.org/10.1016/j.polymer.2018.05.019, Morris B. Thermoforming, Appendix B: Examples of Flexible Packaging Film Structures. In: The Science and Technology of Flexible Packaging. 1st ed. Elsevier; 2016:697-709; MulitRec-supervisory group

Aim of the study & defined multilayer structure

- *Systematical* study of common mitigation strategies, open science
  - Depletion/degradation of additives → Influence on physicochemical properties + film/transport performance
  - Polymer mixtures and impurities such as sometimes found in reality were not tested

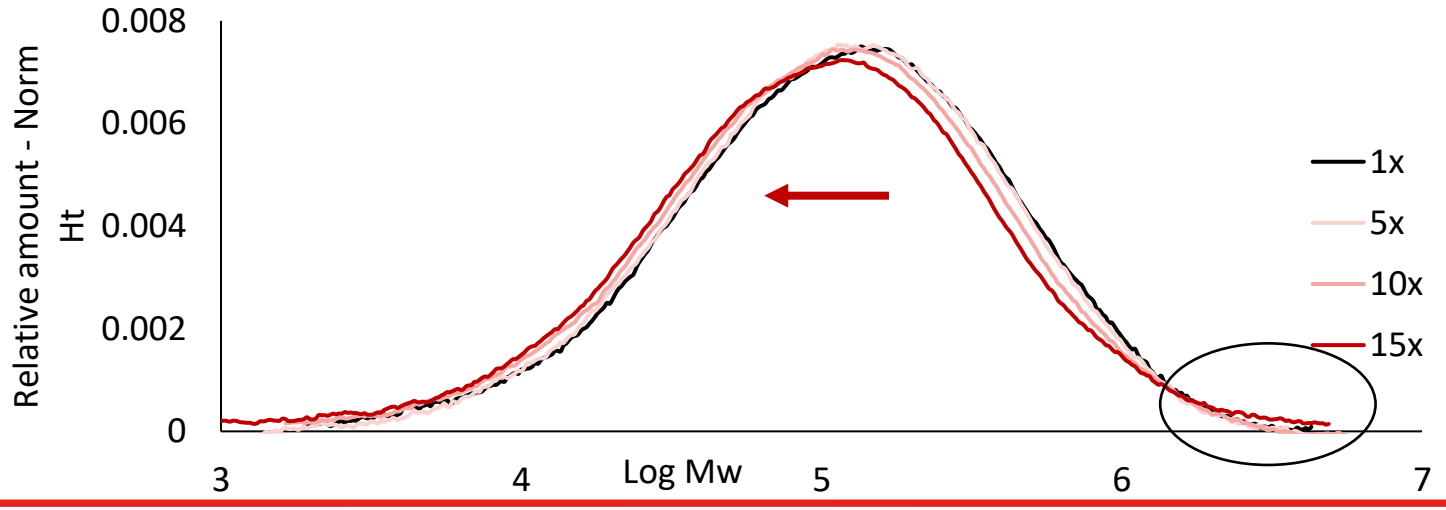


- ABC: simple structure
  - A: Cling layer (5 µm)
  - B: Core layer, with reprocessed content (15 µm)
  - C: Outer layer (5 µm)

Results and discussion – granulate

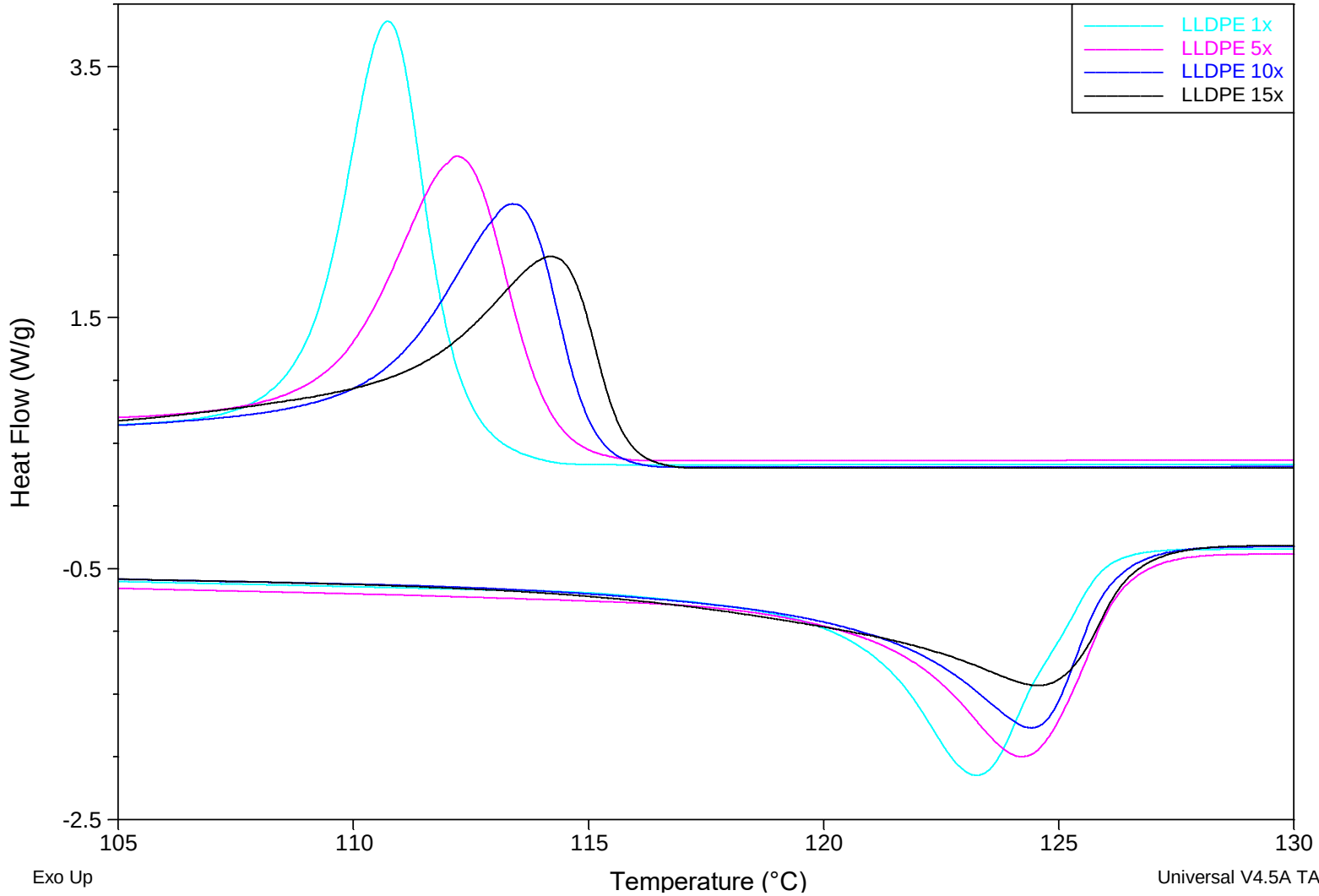
- LLDPE: d: 0.918 g/cc; MFI: 2.0 g/10 min; M_w: 240357 g/mol; PDI: 4.49; T_m: 123.4 °C; Contains primary and secondary antioxidants; Hindered amine light stabiliser (HALS) added to increase UV-stability
- GC/MS: Primary + secondary antioxidants: depletion + degradation;
  E.g. primary antioxidant: 1022 ppm (1x) → 570 ppm (5x) → 387 ppm (10x) → 79 ppm (15x)
- GPC
  - Reprocessing reduces M_N and M_W, reduction more clear in M_N
  - Dispersity (Đ) increases

Sample Name	M_N	M_W	Đ
1x	47308	238757	5.047
5x	48614	230334	4.738
10x	41321	217072	5.253
15x	31394	228621	7.282



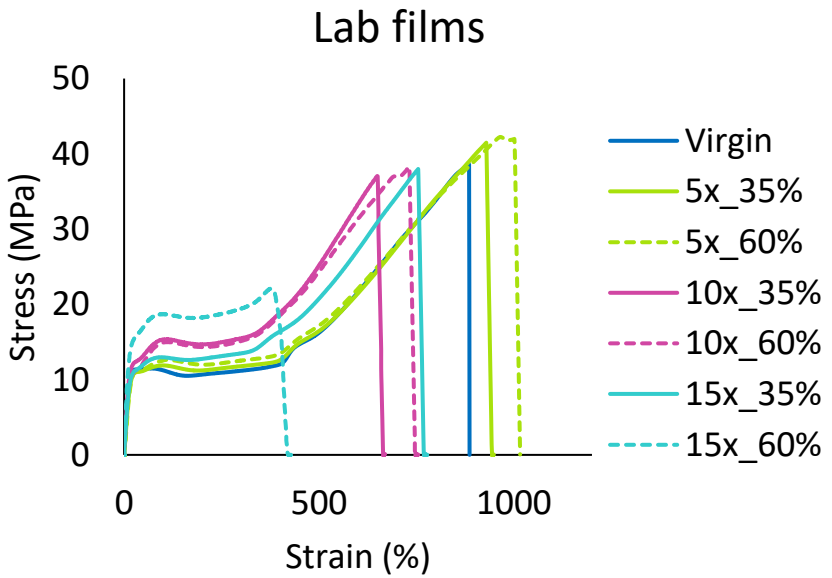
Results and discussion - granulate

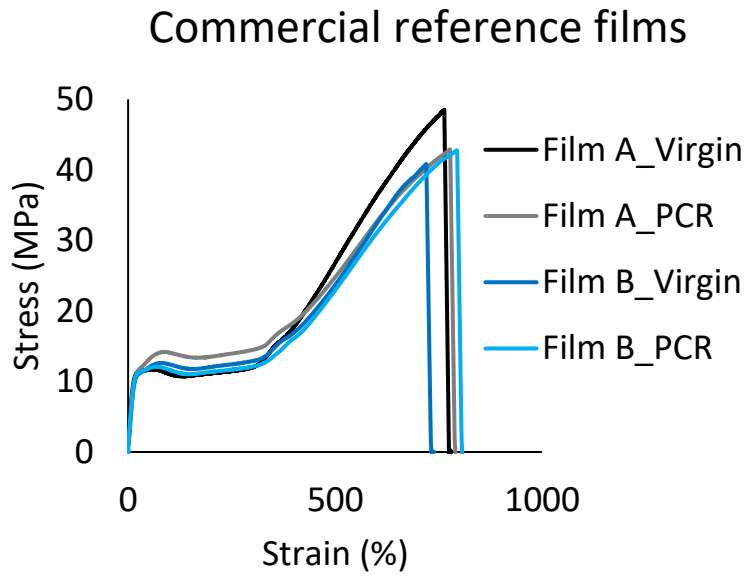
- DSC (cooling + second heating)
  - Reprocessing
    - Increases melting and recrystallisation T_{peak}
    - Broadens heat flow peak
    - Decreases peak height



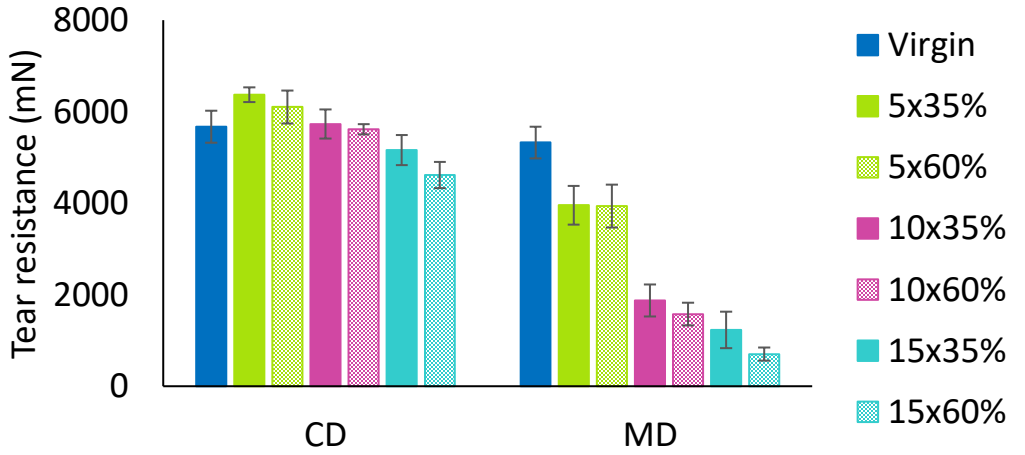
Results and discussion - film

- Tensile properties (ISO 527-3)
  - 15x_60% <<<< other films
  - ≈ commercial reference films



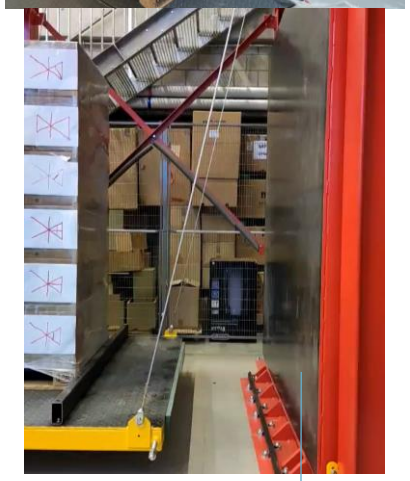


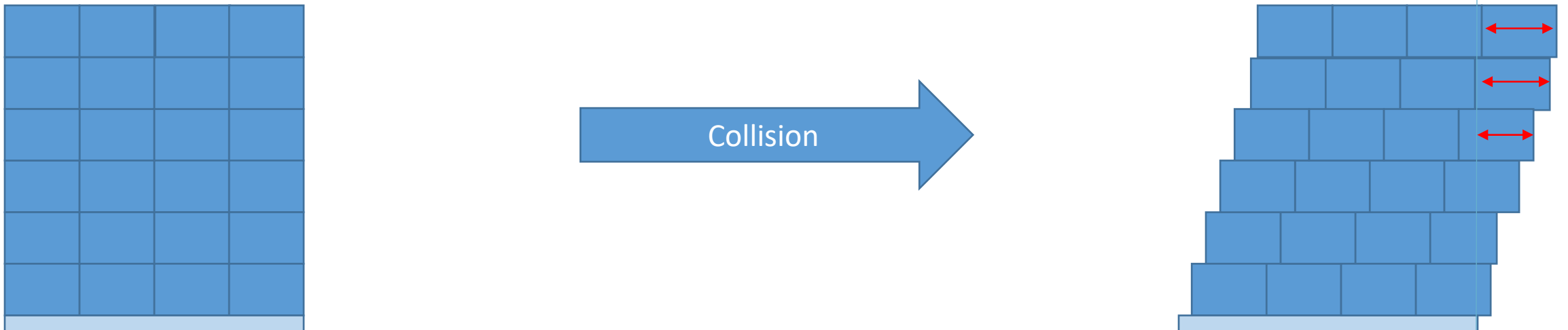
- Tear resistance (ISO 6383-2)
  - Clear differences in MD
    - 15x < 10x < 5x < virgin
    - 60% < 35%
  - Commercial reference films (1000-6000 mN)
    - MD << CD
    - PCR << virgin



Transport simulation tests

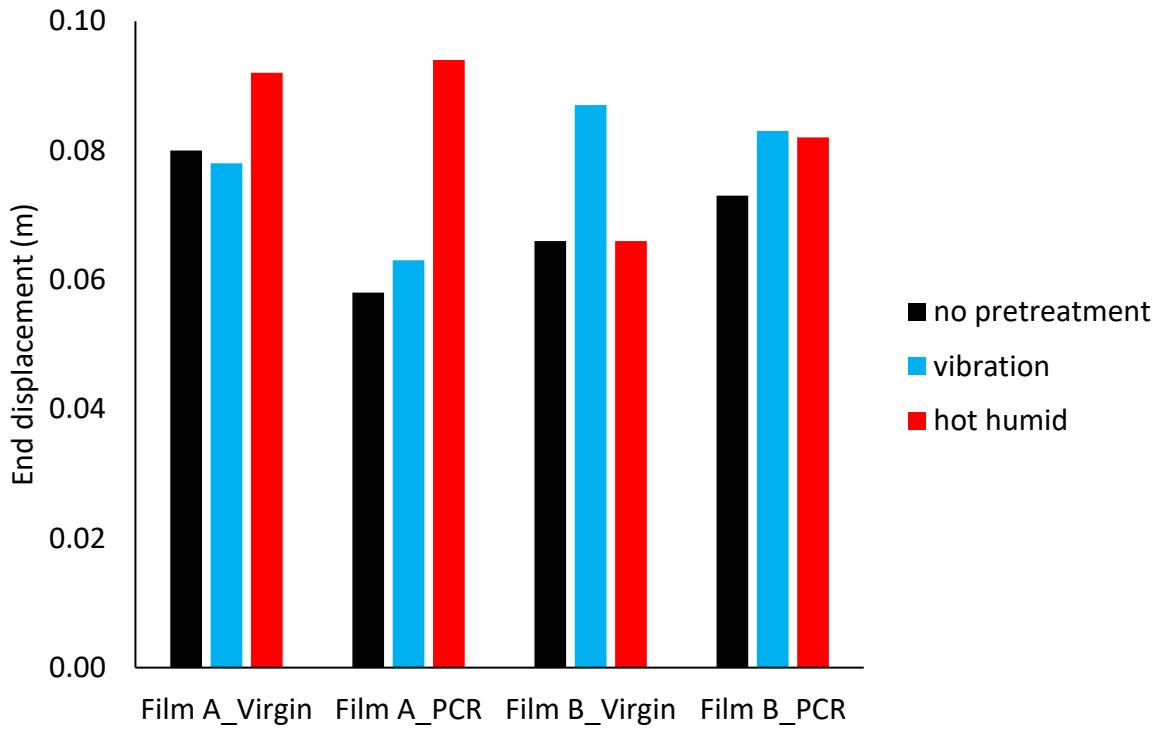
- Palletised load = constant
  - EUR-pallet, edge protectors, top sheet, 6 layers column-stacked boxes
  - Wrapped with 7 base windings + 3 top windings
- Simulations assess sudden horizontal shocks, vibration effects, conditioning, and extreme tilting
  - Swing test evaluates impact after a horizontal shock, either tested 10 minutes after wrapping or after pre-treatment
    - Vibration table (ISTA 3E), 3 hours random vibration
    - Climate chamber: 72 h at 38°C/50% RH, then 6 h at 60°C/30% RH
  - Tilt test: Pallet lifted at 26° for 1 min.





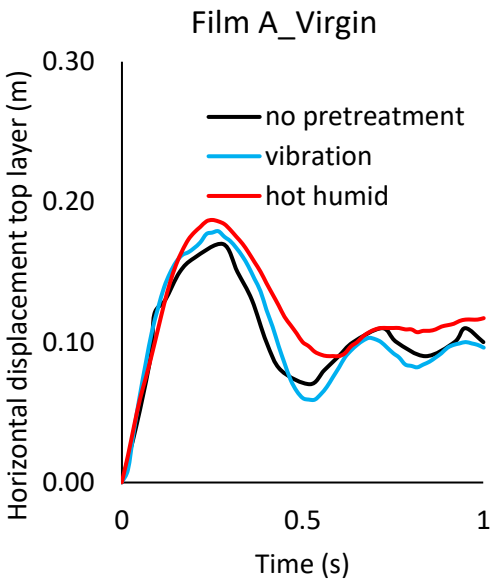
Transport simulation tests
First results of commercial reference films

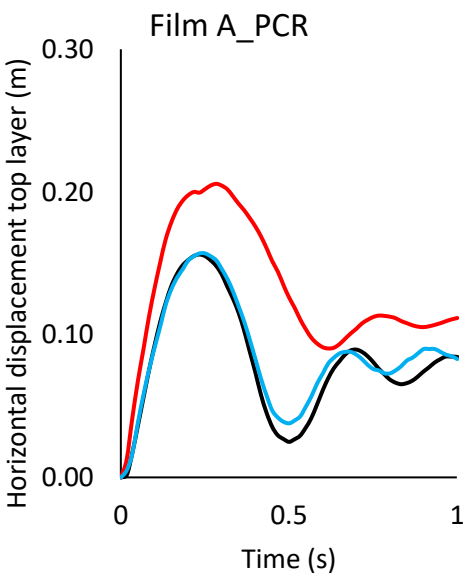
- 26° tilting: no displacements observed ✓
- Horizontal displacement
  - Calculated for top layer (distance to wall: before – after)
- No clear trend, slight tendency of less top-layer shifting in pallets without pre-treatment
- Shifting/tilting during collision?

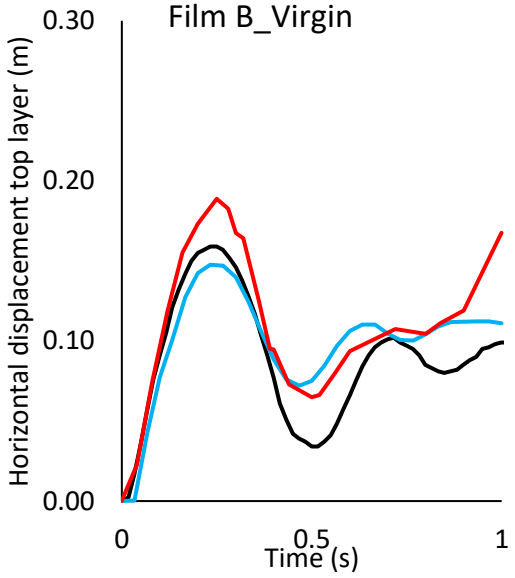


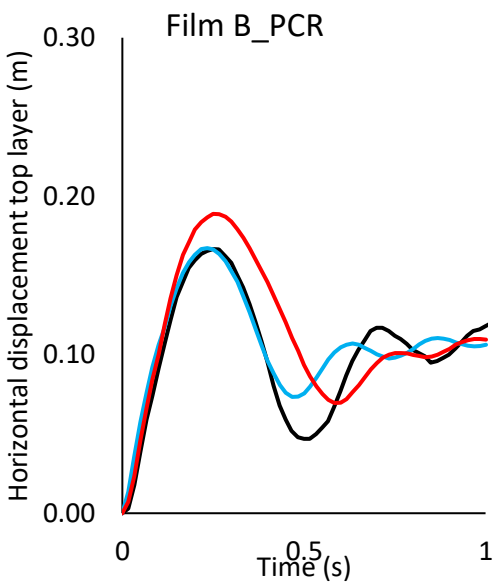
Transport simulation tests
First results of commercial reference films

- Horizontal displacement top layer during collision: initial displacement twice the value of final position (0.2 m > 0.1 m)
- Hot humid conditioning increased displacement; no effect of vibration









Conclusion

- Reprocessing of LLDPE granulate
  - Depletion of antioxidants
  - Decreased molecular weight
  - Increased polydispersity
  - Broadening DSC heat flow peaks

} Decreased mechanical film performance

- To do
  - Further chemical analysis
    - LC/MS: to screen HALS depletion
    - NMR: to study branching
    - …
  - Check lab film performance (transport + UV-stability)

Application 2
Blown extrusion of sealing films

Department of Polymer Engineering and Technology, Faculty of Chemistry

Results – granules for sealing application

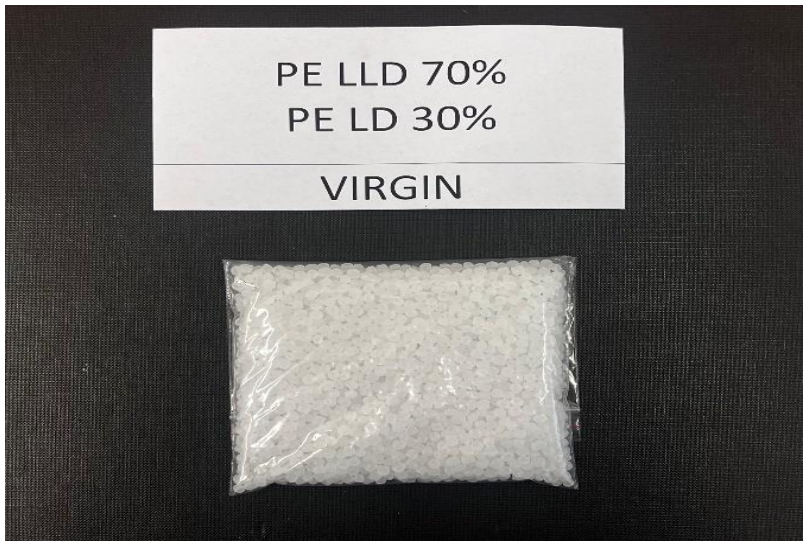
Samples designation

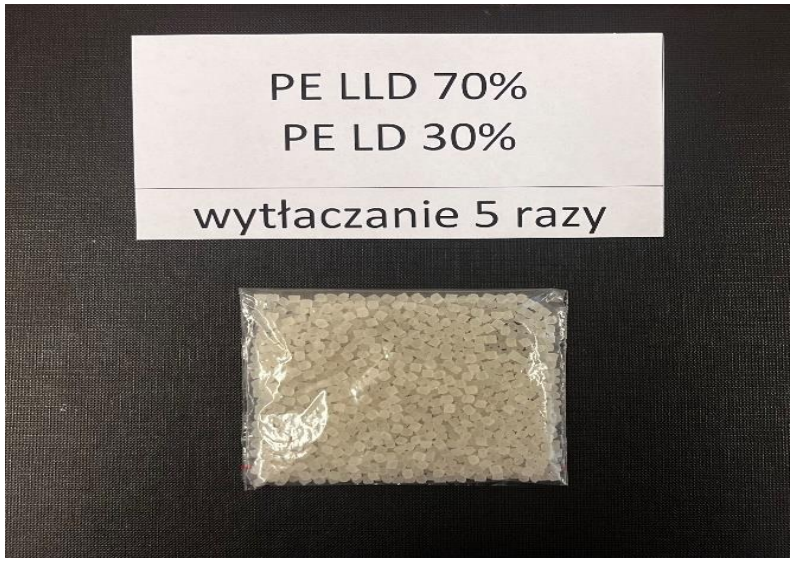
LDPE/LLDPE x1 ref.

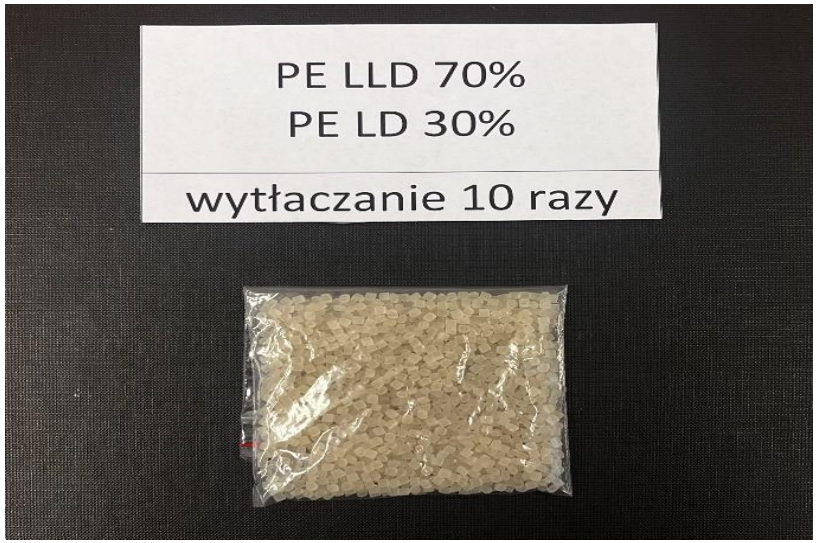
LDPE/LLDPE x5

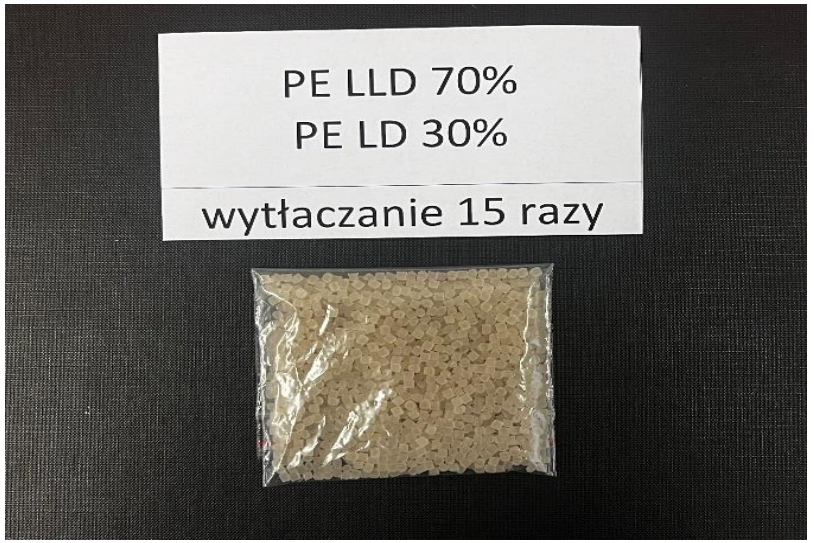
LDPE/LLDPE x10

LDPE/LLDPE x15



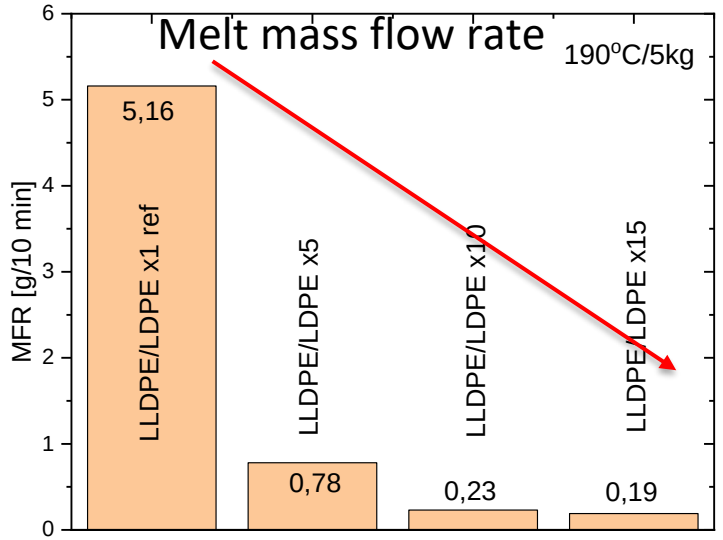




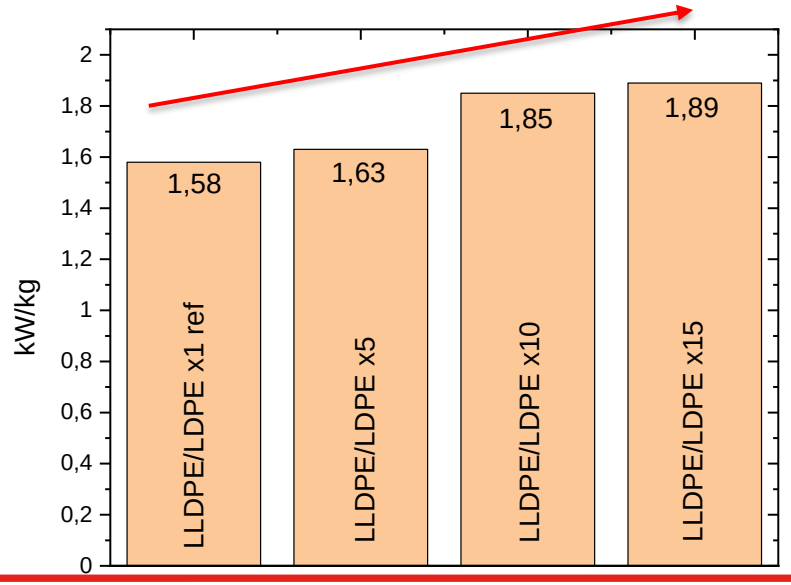


- Results – processing films for sealing application

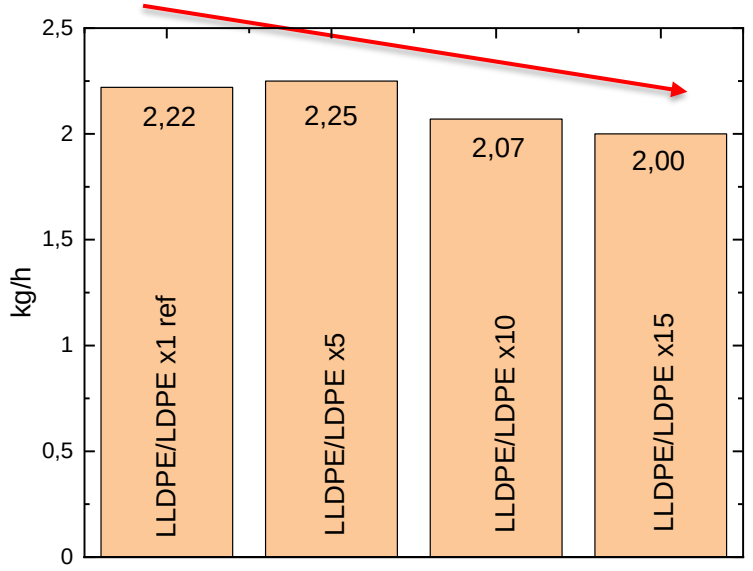
Results – MFR, efficiency and energy consumption



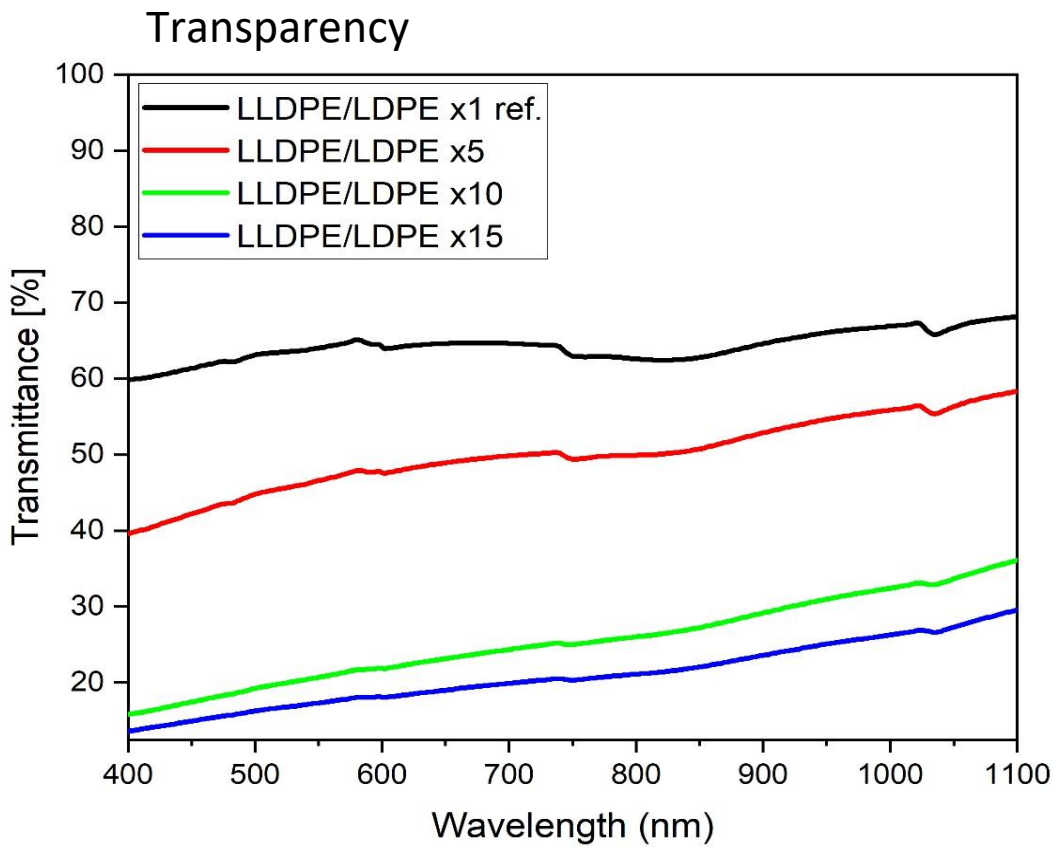
Energy consumption for film production



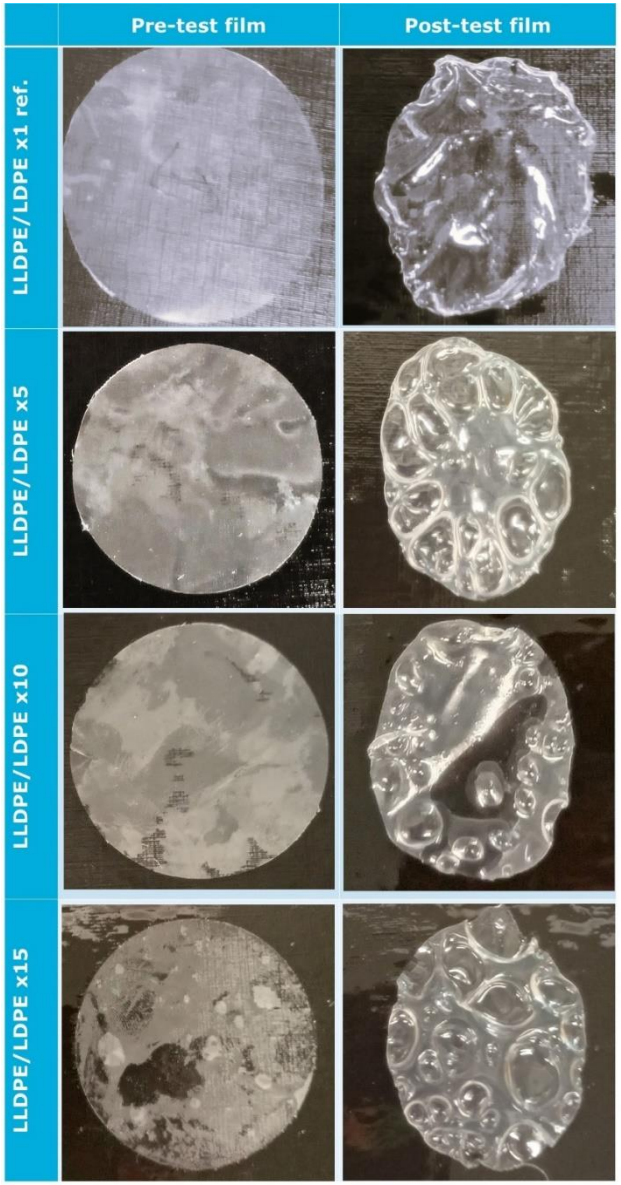
Efficiency of extrusion



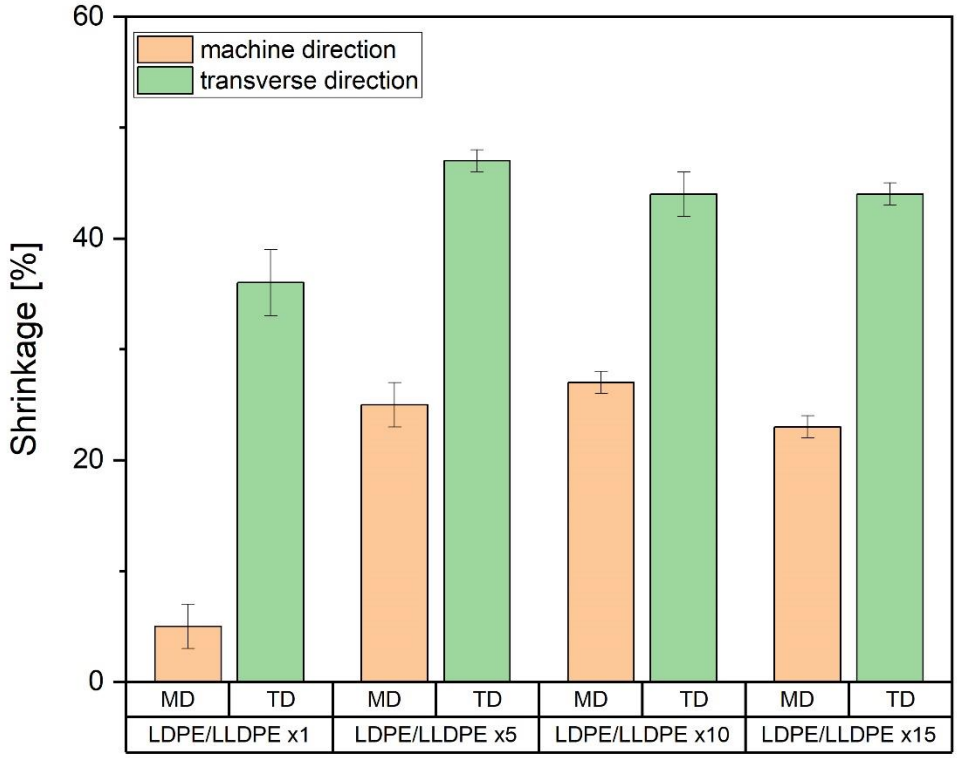
Results – Properties – UV-Vis



Results - shrinkage



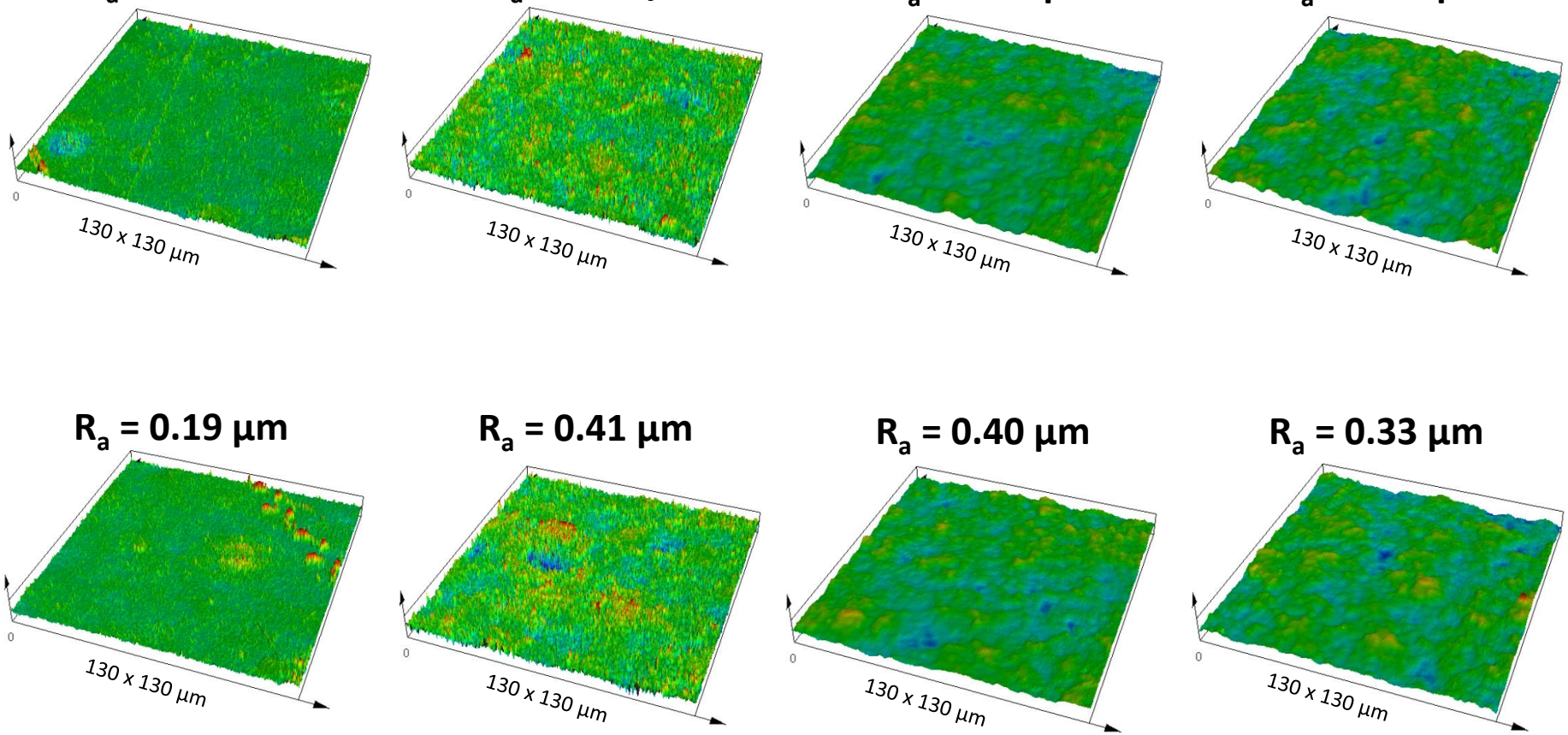
Photographs of the tested films before and after shrinkage measurements



Sample	Shrinkage [%]	
	machine direction	transverse direction
LDPE/LLDPE x1 ref.	5	36
LDPE/LLDPE x5	25	47
LDPE/LLDPE x10	27	44
LDPE/LLDPE x15	23	44

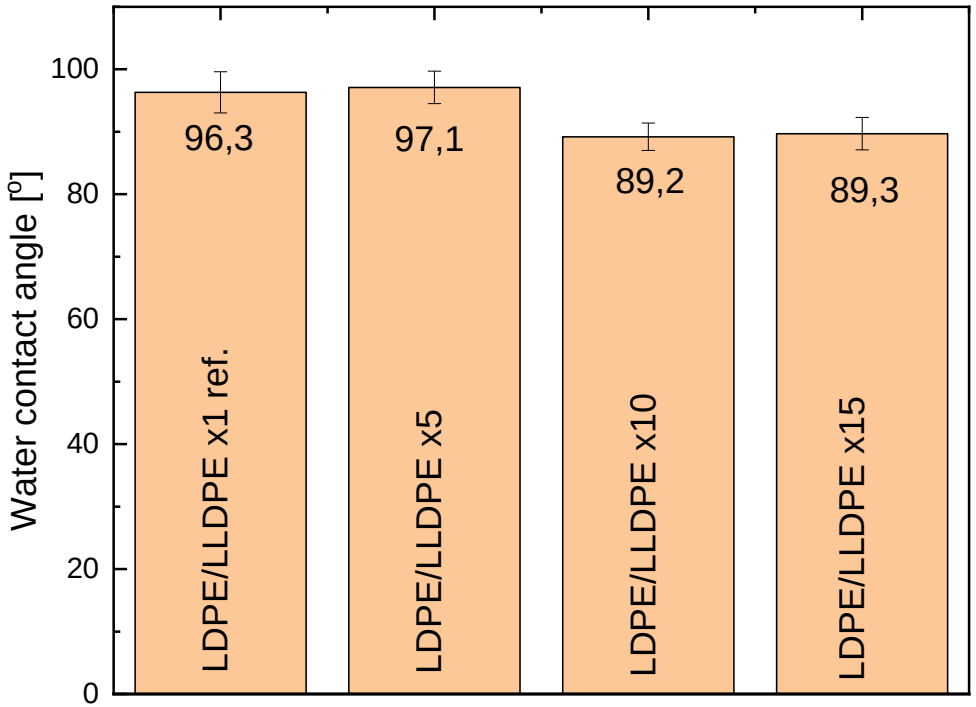
Results – Surface topography (optical profilometry)

	LDPE/LLDPE x1 ref.	LDPE/LLDPE x5	LDPE/LLDPE x10	LDPE/LLDPE x15
Inner surface	R_a = 0.19 µm	R_a = 0.34 µm	R_a = 0.27 µm	R_a = 0.31 µm
Outer surface	R_a = 0.19 µm	R_a = 0.41 µm	R_a = 0.40 µm	R_a = 0.33 µm



Results – Water Contact Angle

LDPE/LLDPE x1 ref. LDPE/LLDPE x5 LDPE/LLDPE x10 LDPE/LLDPE x15



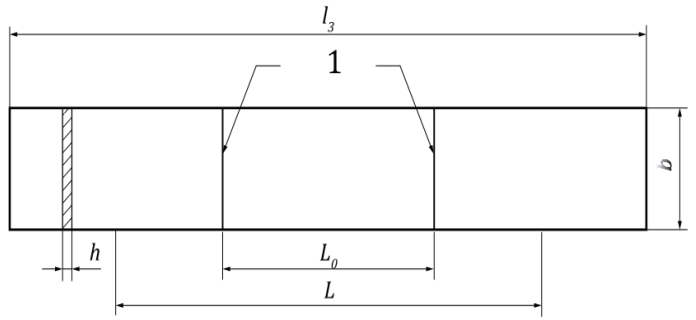
Results – Mechanical Testing (tensile testing (ISO 527-3:2018)

Machine Direction

Sample	E [MPa]	σ_M [MPa]	ε_B [%]
PE	169 ±11	28.1 ±2.8	654 ±42
PE@5	185 ±5	24.0 ±3.2	475 ±55
PE@10	188 ±3	14.3 ±0.7	232 ±33
PE@15	186 ±7	17.6 ±2.7	376 ±66

Transverse Direction

Sample	E [MPa]	σ_M [MPa]	ε_B [%]
PE	179 ±11	29.5 ±3.8	670 ±54
PE@5	200 ±10	25.2 ±4.4	527 ±64
PE@10	212 ±12	15.6 ±1.2	277 ±41
PE@15	213 ±10	17.1 ±2.2	346± 61



Key

1	gauge marks
b	width: 10 mm to 25 mm
h	thickness: ≤1 mm
L_0	gauge length: 50 mm ± 0,5 mm
L	initial distance between grips: 100 mm ± 5 mm
l_3	overall length: ≥150 mm

Results – tear resistant – trouser tear method (ISO 6383-1:2015)

Machine Direction

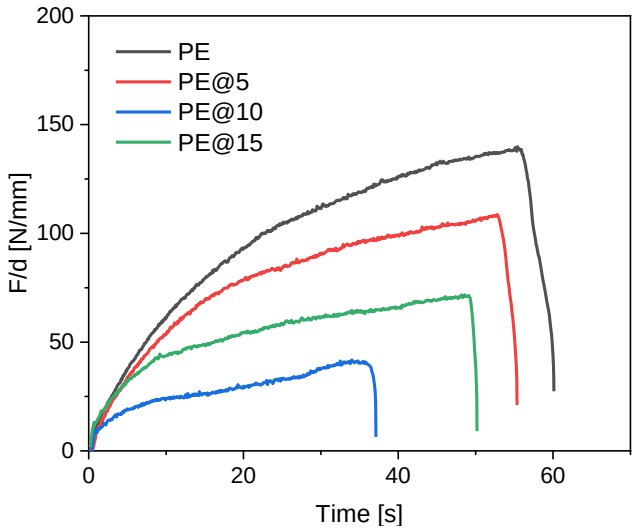
Sample	F_{max}/d [N/mm]	Type of break
PE	143 ±8	*
PE@5	110 ±9	*
PE@10	42 ±6	*/#
PE@15	67 ±5	*/#

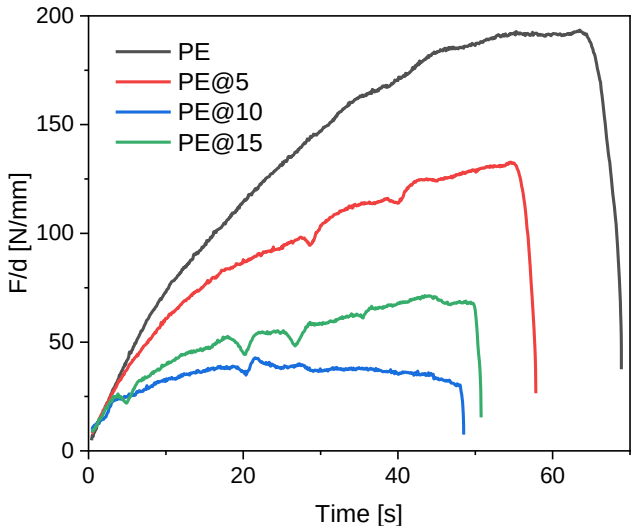
Transverse Direction

Sample	F_{max}/d [N/mm]	Type of break
PE	196 ±7	*
PE@5	133 ±5	*
PE@10	41 ±1	*
PE@15	70 ±7	*

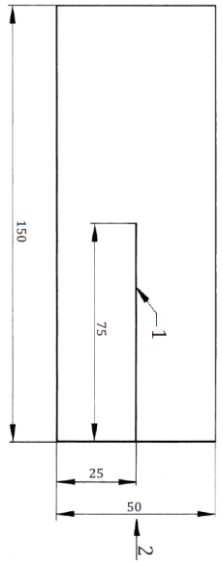
* tear parallel to the cut in the sample

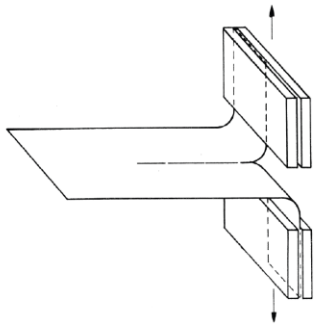
\# tear changes direction without reaching the end of the sample





Methodology





Summary:

1. In our research, we have shown that it is possible to produce a blown film from LLDPE/LDPE blend extruded 15 Times;
2. Repeated processing of the LLDPE/LDPE blend increases its surface roughness and affect the wettability of the film, as well as affects the transparency of the films
3. No significant changes of shrinkcage were measured between samples extruded 5,10 and 15 Times.

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https://www.uhasselt.be/en/instituten-en/imo-imomec/material-analysis/materials-packaging

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Thank you for your attention

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