Article

Mechanical recycling of LDPE films containing PA6/66 by three different, independent testing protocols

0. Introduction

Polyethylene is one of the most widely used polymers in plenty of packaging applications, such as films, bottles, etc… However, in food packaging applications, which consists in 23% of the whole PE flexible film market, it is necessary to combine polyethylene with other materials to achieve the requirements of the product.

What makes polyamide so attractive in the film packaging industry is its long list of beneficial properties, noteworthy its toughness, high mechanical resistance, thermoformability, excellent puncture resistance, good oxygen barrier and chemical resistance to oils, greases, and aliphatic and aromatic hydrocarbons [A].

Polyamide 6 and the copolymer Polyamide 6/6.6 are the most common polyamide types used in flexible packaging, mainly in coextrusion and lamination. This study is focused on coextruded films, where there will be always present a tie layer, a PE-g-MAH providing cohesion between the LDPE layers and PA layers.

Recycling evaluation protocols have been presented by different initiatives and institutions. The protocols are not harmonized and there are important differences between them.

In this article, results according RecyClass, APR and COTREP recyclability evaluation protocols will be exposed. Structure tested in all protocols is shown in next table:

Sample	Layer 1 (PE)	Layer 2 (Adh.)	Layer 3 (PA)	Layer 4 (Adh.)	Layer 5 (PE)	Total thickness
SC15	37,5µm	5µm	15µm	5µm	37,5µm	100µm

Table 1. *Layer thickness distribution of selected film for recycling study.*

1. Methods

In this article, results according RecyClass, APR and COTREP recycling evaluation protocols will be exposed. As a first view, below is shown a comparison between the different protocols.

Table 2. Protocol characteristics.

Method	APR	RecyClass	COTREP
Protocol	FPE-CG-01	V1.0 Last update v5.0	PE-1 & PE-2
Reference Material	Virgin Resin	Virgin resin	PCR
Washing Flakes			YES
Flake Blend Composition	A: 100% Control B: 50/50 Control/Innovation C: 100 % Innovation	A0: 100% Control A25: 75/25 Control/Innovation A50: 50/50 Control/Innovation A100: 100% Innovation (optional)	Control: 100% PCR Innovation: Dilutions applied to simulate market penetration
Densification	yes	No (v1.0) Optional (v5.0)	Yes
Pelletization equipment	Single screw	Twin Screw (v1.0)	Typical extruder (define main parameters)
Drying before pelletization	Up to 90degC	90degC@1h	130degC (moisture <5%)
Filter Pelletization	40/150/40 mesh 400/104/400 µm	110 µm	150µm (one filter switch allowed)
Pelletization Temperatures	Tm: 190-245degC	Tm: 200-230degC (v1.0) Tm: 230±5degC (v5.0)	Minimum one zone at 250ºC
Pellet Blend Composition	No additional Blend	B0: 50/50 Control / virgin PE B25: 50/50 A25 / virgin PE B50: 50/50 A50 / virgin PE B100: 100% Innovation (optional)	To be defined depending on market penetration data
Effective Innovation Composition	B: 50% C: 100%	B25: 12.5% B50: 25% B100: 50%	To be defined depending on market penetration data
Drying Before Extrusion	Up to 90degC @ 1h	60degC @ 10min	Yes (moisture content <1%)
Film Extrusion Temperatures	190-245degC	200-230degC	Record (not specified)
BUR	2.5	>2.5	2.5-3.0
Film Thickness	1.5 – 2.5 mills 38-63,5 µm	<25 µm (v1.0) 25 ± 5 µm & 50 ± 5 µm (v5.0)	50 µm

2. Materials and Evaluation Methods

2.1. Film structure and materials

The selected structure is a film with polyamide in the core layer: PE/Tie/PA/Tie/PE.

The adhesive used in the layers between polyamide and polyethylene is a tie resin that bonds both materials thanks to maleic anhydride (MAH) grafted in a polyethylene matrix. The unreacted MAH will act as a compatibilizer during the recycling process of the film.

Figure 1. Selected film for recycling study.

PE (LDPE), grade name 410E by Dow Chemicals. Adhesive material (LLDPE-g-MAH), grade name NF498E by Mitsui Chemicals. PA (PA6/6.6), grade name UBE NYLON 5033B by UBE for APR or UBE NYLON 5034B by UBE for the other protocols.

2.2. EVALUATION WITH RecyClass PROTOCOL

The original films to be studied were produced in a Dr. Collin airblown extrusion line at UBE Corporation Europe S.A.U. R&D department.

Sample	Layer 1	Layer 2	Layer 3	Layer 4	Layer 5	Total thickness
Reference	LDPE 37,5µm	LDPE 5µm	LDPE 15µm	LDPE 5µm	LDPE 37,5µm	100µm
SC15	LDPE 37,5µm	Tie 5µm	PA6/66 15µm	Tie 5µm	LDPE 37,5µm	100µm

Table 2. Initial films for RecyClass protocol.

Both films were then sent to Aimplas technology center (accredited laboratory by RecyClass) to evaluate the recyclability according to RecyClass protocol[1] (v1.0).

[1] Current version of the protocol is v5.0: RecyClass Recyclability Evaluation Protocol for PE Films.

2.2.1. Pre-treatment

Each film was then shredded on a Lidem blade mill obtaining 5mm diameter flakes. Shredding was successfully performed. The resulting flakes of each film were diluted with the reference material according to the protocol.

Sample	Composition
A.0	100% reference film
A.25	75% reference + 25% SC15
A.50	50% reference + 50% SC15

Table 3. Dilution and composition of each flakes blending.

2.2.2. Pellet extrusion

Each flake blend was later repelletized in a Leistritz ZSE 27 MAXX co-rotating twin screw extruder. The extruded strand was cooled down in a water bath and cut in a pelletizer to obtain the recycled pellets. The temperature profiles used are described in Table 4.

Temperature profile (ºC)	Q (kg/h)
60/100/210/210/210/205/205/205/200/200/200/200	15

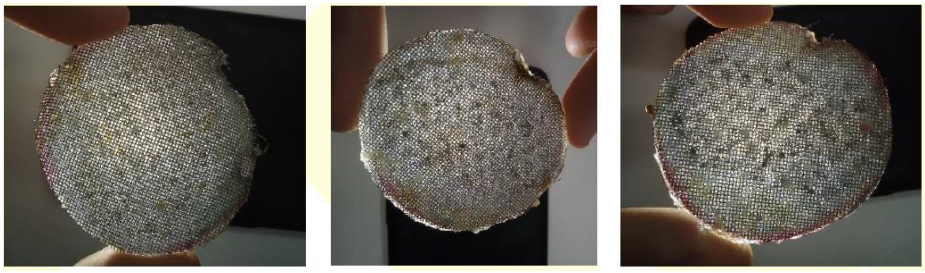
Table 4. Compounding parameters for blended compounds.

Response parameters were recorded as it is described in Table 5. Melting temperature should have a value between 200-230degC according to RecyClass Protocol v1.0.

Sample	ΔP0	ΔP15	ΔP30	Tm (degC)
A.0 (100% control)	ref	ref	ref	203
A.25 (75% control + 25% innovation)	2.1	8.6	4.1	203
A.50 (50% control + 50% innovation)	6.3	16.1	11.3	203

Table 5. Pressure variations and Melting temperature.

In between each trial, filter was changed in order to detect particles and build-up of each sample. In all samples including Control film, some dark specks were observed trapped in the filters, possibly corresponding to contamination from the milling process, but no appreciable polymeric build-up in filters were detected.



A.0 A.25 A.50

Figure 2. Aspect of filters after each trial.

Pellets obtained were characterized according to the RecyClass protocol specification. Results summarized in Table 6.

Test	Properties	A.0	A.25	A.50	Requirement	Judgement
Bulk density	Bulk density (Kg/m^3)	519	518	524	≥500	OK
Melt flow rate	Melt flow rate (g/10min)	1.52	1.23	1.06	<Δ0.5g/10min to control	OK
Gas content and ash content by TGA	Gas Content (%)	0.05	0.02	0.04	-	No threshold
	Ash Content (%)	0.08	0.17	0.09	-	No threshold
DSC	T_{m1} (degC)	112.7	113.1	113.2	<150degC	OK
	ΔH_{m1} (J/g)	112.6	108.4	102	-	No threshold
	Tm_2 (degC)	-	190.6	189.7	<150degC	OK [A] Negligible peaks
	ΔH_{m2} (J/g)	-	1.5	2.9	-	No threshold
Impurities Content	Visual Inspection	No impurities	No impurities	No impurities	-	No threshold
Surface Appearance	Visual Inspection	Uniform	Uniform	Uniform	-	No threshold
Volatiles	Weight Loss (%)	0.02	0.09	0.10	<1.0	OK
Colour	L*	65.41	65.46	66.63	-	No threshold
	a*	-0.59	-0.84	-0.97	-	No threshold
	b*	-3.17	0.69	-0.93	Δb*<0.5 for natural stream	OK
Delta Pressure	ΔP (%)	-	4.1	11.3	<Δ25% to control	OK

Table 6. Pellet properties

[A] The enthalpy of the melting peak observed at 190.6degC for A.25 and at 189.7degC for A.50, corresponding to PA6/66, have a value of 1.5J/g and 2.9J/g respectively, which are very low and even lower than the uncertainty value. According to this, the observed peaks could be considered as negligible.

Below are shown the DSC curves of A25 and A50:

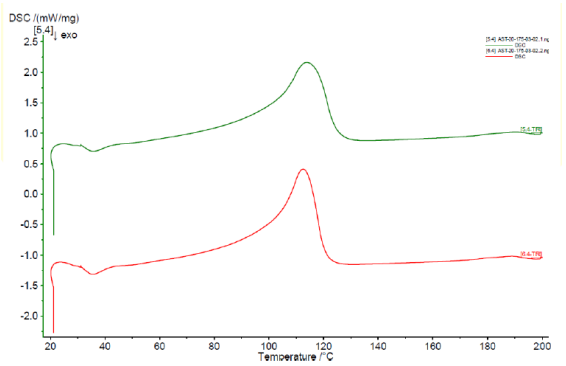


Figure 3. DSC sample A25.

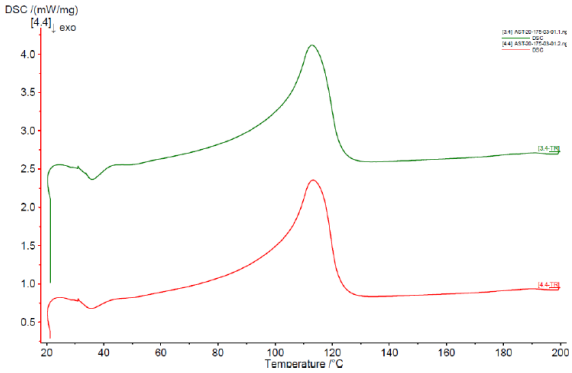


Figure 4. DSC sample A50.

2.2.3. Converting

From previous materials different blends were prepared by blown extrusion process. The blends were prepared by manual mixing at indicated ratios as it is shown in Table 7.

Sample	Composition
B.0	50% A.0 + 50% virgin pellet
B.25	50% A.25 + 50% virgin pellet
B.50	50% A.50 + 50% virgin pellet

Table 7. Samples obtained by blown film extrusion.

The materials were processed in a blown film extrusion pilot line to evaluate the processability and obtain films to be characterized.

Table 8 shows the conditions of the film extrusion defined in RecyClass protocol (v1.0):

- Blow up ratio > 2.5.
- Melt temperature of 200-230degC.
- Thickness : <25 µm (v1.0) / 25±5 µm (v5.0)
- 30-minute run time

T (degC)					Die (degC)	Screw speed (rpm)
185	195	205	210	205	195	55

Table 8. Temperature for blown film extrusion

All samples were extruded without any problem, reaching the stationary state during 30minutes. **All samples were stable** and none of them presented holes or visual imperfections.
Output parameters from film extrusion are shown in Table 9.

Sample	B.0	B.25	B.50
Melt Temperature (degC)	199	199	199
Melt Pressure (bar)	69	73	76
Stretching (mm/min)	6.8	6.8	6.8
Extrusion Output (Kg/h)	3-4	3-4	3-4

Table 9. Outputs in blown film extrusion.

2.2.4. Film characterization

Results of the film characterization are summarized in Table 10.

Test	Properties		B.0	B.25	B.50	Requirement
Thickness	Thickness (µm)		20	20	21	< 25 µm
Elmendorf tear resistance	**Tear resistance (N)**	**Longitudinal**	0.491	0.595	0.492	No more than a 25 % delta decrease to B.0
		Transverse	1.291	1.057	1.044	
Tensile Properties	**Tensile Strength (MPa)**	**Longitudinal**	8.71	9.67	11.8	
		Transverse	9.08	9.40	9.51	
	Elongation at Yield (%)	**Longitudinal**	13	12	15	
		Transverse	8.9	8.9	9.4	

Dart Impact	**Dart Impact (g)**	<47	60	62	No more than a 25% delta decrease to B.0
Haze	**Haze (%)**	7.4	17.4	25.5	Record
Gels and Specks	**Number of gels and specks**	11	13	19	<100 Gels & Specks
Surface Appearance	**Visual inspection**	Uniform shape and colour	Uniform shape and colour	Uniform shape and colour	Record

Table 10. Film Properties.

2.2.5. Conclusion Evaluation following RecyClass protocol.

All materials were shredded without problems.

During pelletizing step, higher torque values of the extureder for the Innovation Film was observed compared with the Control Film but maintained stable during whole processing time. Equivalent behaviour was observed for pressure values in the extureder.

No significant polymeric build-up in the filters was observed, and pressure rise was low during the run-time, reachin a maximum pressure difference respect control of 11.3% for A.50.

Nonconformities regarding the melting peaks observed for samples A.25 and A.50 could be considered negligible, since enthalpy of respective peaks are very low and even lower than the uncertainty value.

Film extrusion was highly stable, reaching the stationary state for all samples. Melt pressue were higher for Innovation film blends (B.25 and B.50), compared to control. Low gel-specks content was detected in all samples, and neither presented holes or other significant visual imperfections, meeting the requirements of dimensions and weight defined.

Regarding to properties obtained for pellets and films, all requirements were meeting or exceeding protocol values. Suchs as higher dart impact resistance and higher tensile resistance.

2020-PO-011-UBE-technology-approval-letter.pdf (recyclass.eu)

2.3. EVALUATION WITH APR PROTOCOL

The original films to be studied were produced in a Dr. Collin airblown extrusion line at UBE Corporation Europe S.A.U. R&D department.

Sample	Layer 1	Layer 2	Layer 3	Layer 4	Layer 5	Total thickness
Reference	PE 37,5µm	PE 5µm	PE 15µm	PE 5µm	PE 37,5µm	100µm
SC15	PE 37,5µm	Tie 5µm	PA 15µm	Tie 5µm	PE 37,5µm	100µm

Table 11. Initial films for APR protocol.

Both films were then sent to Aimplas technology center (accredited laboratory by APR) to evaluate the recyclability according to APR protocol FPE-GC-01[2], for consideration of Guidance Recognition.

2.3.1. Pre-treatment

Each film was then shredded on a Lidem blade mill obtaining 5mm diameter flakes. Shredding was successfully performed.

Next step was a Flotation Test according APR Sink-Float Screening Test (APR-O-S-01-PO), to determine if the flakes can be separated by density in the float/sink tank used in recycling operation. As result, 100% of the flakes floated in cold and hot conditions:

Sample	Method	Required Value (100% Flotability)	Result
Innovation Film	O-S-01 (cold conditions)	100	100
Control Sample	O-S-01 (cold conditions)	100	100
Innovation Film	O-S-01 (hot conditions)	100	100
Control Sample	O-S-01 (hot conditions)	100	100

Table 12. Sink-Float summary results.

A thermal densification was carried out according to FPE-P-04 at 15min 190degC.

2.3.2. Pellet extrusion

Blends of densified control material and densified innovation films were melt filtered per FP-P-00 Film Processing Practices to create the samples according to table 13.

Sample	Composition	Control	Innovation

[2] APR Design Recognition Programs Critical Guidance Operation Procedures.

		(%)	%)
A pellet	100% densified Control	100	0
B pellet	50/50 blend of densified control + innovation	50	50
C pellet	100% of densified innovation	0	100

Table 13. Film Samples.

The palletization process was carried out using a single screw extruder (Collin 30mm (30L/D)), following processing parameters described in Table 14.

Temperature profile (ºC)	V (rpm)
165/190/195/200/200/195	60

Table 14. Compounding parameters for blended samples.

Response parameters were recorded as it is described in Table 15.

Sample	T_0 (%)	T_{45} (%)	P_0 (bar)	P_{45} (bar)	Tm_0 (ºC)	Tm_{45} (ºC)
A pellet	1,4	2,2	45	49	194	196
B pellet	1,4	1,4	40	38	193	195
C pellet	1,6	1,5	35	48	194	196

Table 15. Blending response parameters.

Table 16 shows variation of pressure running for each blend:

Sample	Av P first 5min. (bar)	Av P last 5 min. (bar)	ΔP_{45}(%)
A pellet	45,5	50	10
B pellet	38,5	42,50	10
C pellet	33,5	46	37

Table 16. Pressure variations during extrusion.

Visual inspection of filters showed some specks and build up for all samples A, B, C:

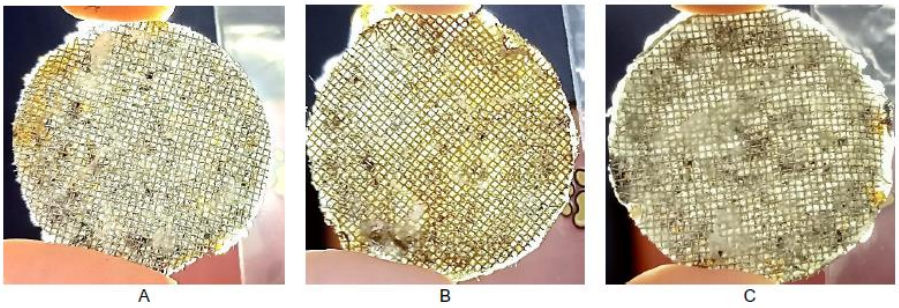


Figure 5. Filter aspect after each trial.

Extruded pellets of samples A, B, C were evaluated, results are summarized in Following table:

Test	Method	Sample A	Sample B	Sample C*	Preferred Values
Screen Pack Pressure Build	Practice FPE-P-06	10	10	37	ΔP over run <25%
Melt Flow Rate (g/10min)	ASTM-D1238	1,49 ±0,05	0,85 ±0,04	0,38 ±0,02	0,2 to 3 g/min@ 190ºC 2, 16Kg
Density (g/cc)	ASTM-D792	0,922 ±0,01	0,941 ±0,01	0,951 ±0,01	<0,966
Ash (% w/w)	ASTM-D5630	0,008 ±0,002	0,014 ±0,00	0,013 ±0,00	Record
Volatiles/ moisture (%)	ASTM-D6980	0,04 ±0,00	0,10 ±0,01	0,12 ±0,01	<0,5
Bulk density (Kg/m3)	ASTM-1895	520 ±3	532 ±8	520 ±3	>480
FTIR for PP homopolymer detection	ASTM-D3799	PE	PE+PA	PE+PA	Characteristic bands
DSC (ºC)	ASTM-D3418	112,4 ±0,1	112,5 ±1	112,1 ±0,1	Primary peak <150ºC
		-	195,2 ±0,4	195,5 ±0,1	Secondary peak
Extrusion Process	Fumes, odour, build-up	Ok	Ok	Ok	Record
Pellet irregularity	Visual inspection	Ok	Ok	Ok	Uniform in sape, colour and size

Table 17. Extruded pellets evaluation.

* APR Guidance is for A (0%) and B (50%) preferred values only – C (100%) to be recorded and reported.

Property	Method	Coordinate values	Sample A	Sample B	Sample C*
Pellet Colour	ASTM D6290-13	L	68,5 ±0,5	68,0 ±0,5	69,7 ±0,5
		a	-0,21 ±0,5	-1,1 ±0,5	-1,1 ±0,5
		b	-0,9 ±0,5	5,9 ±0,5	10,2 ±0,5
		X	37,9 ±0,5	36,9 ±0,5	39,3 ±0,5
		Y	38,7 ±0,5	37,9 ±0,5	40,4 ±0,5
		Z	46,6 ±0,5	39,7 ±0,5	38,1 ±0,5
		x	0,308 ±0,002	0,332 ±0,002	0,332 ±0,002
		YI	-2,6 ±0,5	12,5 ±0,5	23,1 ±0,5

Table 18. Pellet colour.

2.3.3. Converting

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All samples A, B, C, were extruded into blown film per FP-P-06 Film processing practices for required blown film Testing.

The blown-film extrusion line consists of a Collin single-screw extruder E25M, 25mm of diameter and a length to diameter ratio of 30. A melt filter of 250µm was used at the end of the extruder, assemble between 400µm grids as filter holder, on the breaker plate, and installed in a screen changer.

Following tables show input and outputs parameters of blow film extrusion:

Temperature profile (ºC)	Die (ºC)	Screw speed (rpm)
190/210/215/220/200	200	43

Table 19. Inputs in blown film extrusion.

Sample	Melt Temperature (ºC)	Melt Pressure (bar)	Screw Speed (rpm)
A Film	203	76	1,7
B Film	202	78	1,8
C Film	203	79	1,8

Table 20. Outputs in blown film extrusion.

All samples were extruded without any problem, reaching the stationary state in all cases and a blow-up ratio of 2,5 and run at this state for 45 minutes.

2.3.4. Characterization

Films obtained were characterized according to APR protocol specifications. Results are summarized in following table:

Property		Method	Sample A	Sample B	Δ to control	Sample C*	Preferred Values
Thickness (mils)		ASTM-D6988-21	2,2	1,96	-12%	2,12	1,5 to 2,5
Tensile Properties MD	Tensile Strength (MPa)	ASTM-D882-18	13,6 ±0,8	17 ±1	20%	16 ±2	>25% Δ drop to the control
	Elongation at yield (%)		12 ±1	17 ±7	29%	14 ±4	
	Percent elongation at break (%)		390 ±30	360 ±40	-8%	320 ±60	
Tensile Properties TD	Tensile Strength (MPa)	ASTM-D882-18	15 ±2	13 ±2	-15%	12 ±1	>25% Δ drop to the control
	Elongation at yield (%)		11 ±1	10 ±1	-10%	4,3 ±0,3	
	Percent elongation at break (%)		480 ±40	460 ±60	-4%	170 ±0	
Elmendorf tear resistance	(MD)	ASTM D1922-15	658 ±122	777 ±144	15%	639 ±123	>25% Δ drop to the control
	(TD)		1523 ±51	1552 ±31	2%	4129 ±12	
Dart Impact		ASTM D1709	112 ±8	90 ±16	-24%	57 ±5	
Surface appearance detection		According to FPE-S-01, FAR rating	0	1(A)	Preferred	1(A)	

Table 21. Film properties.

Defects description	Rating	
	Sample B	Sample C
Texture	1	1
Gels > 0.8mm	0	0
Carbon black particles/ Specks/Unmelts	0	0
Fisheyes	0	0
Holes	0	0
Total	1	1
Grade	A	A

Table 22. FAR rating.

2.3.5. Conclusion evaluation following APR protocol.

All materials (Control film and Innovation films) have been shredded without problems. Flotation test was performed to control and Innovation film, resulting in clear water and 100% flotablity. Thermal densification was done to the flakes with no remarks on the process.

Druing pelletization step, some pressure and torque peaks were esperienced for all samples over running time. Regarding melt pressure values in the extruder, blend samples B and C experiences slighter lower values than sample A. Regarding screen pack pressure build, samples A and B did not overreach the superior benchmark of 25% over starting pressure value.

In respect of extruded pellet properties, no benchmarking protocol deviations were recorded for both samples A and B.

Blown film extrusion also showed some pressure peaks (3-4 bar), nonetheless the stationary state was reached for all samples and no production issues (e.g. line rupture) ocurred. Melt pressure and torque showed a different trend than in pelletizing process and experienced slightly higher values than control sample A. Melt filters inspection revealed no signs of build-up, neither black specs were observed. Regarding surface appearance, none of the samples presented holes or any other significant visual imperfections and met the requirements of dimensions and weight defined.

Regarding mechanical properties of film sample B, no disconformities were found, and all the values met the requirements established by protocol, being in all cases under the benchmark of 25% Δ drop to the control. Regarding FAR rating, value of 1A (preferred) was obtained as low gels and no holes were observed.

Based on the excellent recyclability performance of film tested, Performance SC 15 obtained “Critical Guidance recognition” from APR:

APR-CGR-PEFilm-barrier-UBE-2023.pdf (plasticsrecycling.org)

2.4. EVALUATION WITH COTREP PROTOCOL

The original films to be studied were produced in a Dr. Collin airblown extrusion line at UBE Corporation Europe S.A.U. R&D department.

Sample	Layer 1	Layer 2	Layer 3	Layer 4	Layer 5	Total thickness
SC15	PE 37,5µm	Tie 5µm	PA 15µm	Tie 5µm	PE 37,5µm	100µm

Table 23. Initial film for COTREP protocol.

Innovation film was sent to IPC center (accredited laboratory by COTREP) to evaluate the recyclability according to COTREP Test Protocol PE-1 and PE-2.

2.4.1. Raw material balance

KIND	Thickness (µm)	Supplier	Name	Amount
Film (coil)	50	COTREP	Reference-rPE	56,4Kg
Film (coil)	100	UBE	LDPE/PA	10,1Kg

Table 24. Film raw materials for trial.

Figure 6. Reference Film

Figure 7. Innovation Film.

2.4.2. Grinding

The size reduction step is carried out in the single-shaft shredder with a 20 mm calibrated screen. Mechanical dust removal is carried out with the vibrating screen to remove dust less than 3 mm. The results of this processing appear in the following table:

SAMPLE	FLAKES >3mm	FLAKES<3mm	THROUGHPOUT EVALUATION
Reference	51.8Kg/94,4%	3,1Kg/5,6%	≈ 21Kg/h
UBE	9,4Kg/97%	0,3Kg/3,0%	≈ 19Kg/h

Table 25. Grinding result.

Conclusion of Grinding:

- Visually the crushed films are homogeneous, there are no agglomerates.
- The UBE sample has a very slightly lower dust content than the reference used.

2.4.3. Washing

Procedure to perform washing was done according following the protocol steps:

- Introduction of 1 kg sample into the tank.
- Fill the tank with clear water at a temperature of 23°C (volume of 20L).
- Wash with stir (1000 rpm) for 5 minutes.
- Unload and filter through a 1 mm mesh sieve.
- Collect a bottle of washing water for visual examination (note any changes in color
- and/or transparency of the water).
- Check the moisture content of the flakes leaving the dryer (Tx < 5%).

Conclusion of Washing:

- The washing water is not colored.
- It was observed a few fines particles on the surface of the water sample.

SAMPLE	Moisture content after washing
UBE	2,1%

Table 26. Moisture content after washing.

2.4.4. Blends preparation

The blends are made in a simple rotary mixer; it has three double mixing blades for optimal homogenization. Blends were prepared as follows, 1% to simulate PA content in the bale and 4% to simulate bale peaks, where PA content can be higher.

BLENDS	REFERENCE	UBE	PA RATE
BATCH Nº 1	100%	-	-
BATCH Nº 2	93,3%	6,7%	1%
BATCH Nº 3	73,3%	26,7%	4%

Table 27. Blends preparation.

2.4.5. Flotation

Flotation was performed according to COTREP protocol:

The blends resulting from the previous step are processed according to the procedure described below:

- Introduce the prepared flakes into the tank containing clear water at room temperature without additives. A vibrating conveyor and two spray bars facilitate the introduction. The flakes are sinking 5 times successively by rollers on the surface of the tank.
- Collect floating flakes and weigh it.
- Collect sinking flakes and weigh it.
- Note if there is a change in the color and transparency of the floatation water.
- Examine the 2 fractions of sequins and note the possible presence of glue, paper, ink, etc. on the sequins.

Conclusion of Flotation:

- No observation of any change in the appearance of the flotation water.
- No observation of any sinking fraction or suspended fraction.
- There are material losses inherent in the treatment process, this explains that the floating fraction percentage is not 100%.

BLENDS	AMOUNT BEFORE FLOTATION	AMOUNT AFTER FLOTATION	FLOTATION FRACTION PERCENTAGE
BATCH Nº 1		NOT CONCERNED	
BATCH Nº 2	15,0Kg	14,3Kg	95,3%
BATCH Nº 3	15,0Kg	14,6Kg	97,3%

Table 28. Batch weight before and after flotation/drying.

2.4.6. Drying

The drying stage of the different batches is achieved in a ventilated oven for a minimum of 4 hours, the temperature of the air circulating in the equipment is 40°C. The control of the moisture content is carried out on 3 samples of one gram at a temperature of 105°C.

SAMPLE	MOISTURE AFTER DRYING
BATCH Nº 2	0,71%
BATCH Nº 3	0,70%

Table 29. Moisture content.

Conclusion of drying process:

Moisture content obtained is lower than 1%.

2.4.7. Film densification

The film densification is achieved by means of the thermogranulator. This allows the regular feeding of shredded films by means of an auxiliary screw in addition to the plasticizing screw. The densification temperature is 135°C, slightly above the melting temperature of the material.

Conclusion of densification process:

- No difficulties in feeding the batches into the thermogranulator hopper.
- The process is stable in automatic mode with a screw speed control imposed by the head pressure.

BLENDS	AMOUNT AFTER FILM DENSIFICATION
BATCH Nº 1	14,5Kg
BATCH Nº 2	13,7Kg
BATCH Nº 3	14,3Kg

Table 30. Amounts after film densification.

2.4.8. Extrusion with melt filtration

Densified pellets were extruded in a corotating twin-screw extruder characterized by a screw diameter of 32 mm and an L/D ratio of 44. Filtration of the melted material was carried out by a manual two-position system located at the head of the extruder.

Below tables summarize respectively the parameters of the extrusion process and the output parameters:

INPUT PARAMETERS	VALUES
Feed rate	10Kg/h
Extrusion time	From 1h to 1h30min
Screw Speed	200rpm

Barrel temperature	10°C/180°C/190°C/210°C/220°C/250°C/250°C/220°C/220°C/220°C/220°C/230°C
Degassing to ambient	Section nº10
Filter screen size	Superposition of 500 µm and 150 µm filters
Filtration area	19,6cm^2

Table 31. Input parameters extrusion.

OUTPUT PARAMETERS	BATCH Nº1	BATCH Nº2	BATCH Nº3
Engine Torque (%)	41	42	43
Specific mechanical energy (Wh/Kg)	372	387	394
Melt pressure at starting (bars)	7	10	17
Melt pressure at end (bars)	18	23	28
Delta pressure (bars)	11	13	11
Number of filters used (pce)	1	1	1
Amount produced (Kg)	11,5	11,5	12,7
Filter clogging ratio (bars/Kg)	0,96	1.13	0,87

Table 32. Output parameters extrusion.

Conclusion of Extrusion process:

- Drying of the batches before extrusion in a ventilated oven at 40°C for several hours (2 hours minimum).
Stable extrusion conditions for each batch, no need to change the filters.
- The LDPE base of the UBE sample appears to be slightly more viscous than the reference used.

2.4.9. Blown film extrusion

The pellets from the regeneration were processed by blown extrusion with 50% in weight of virgin material, DOW LDPE 535E.

Blown extrusion was performed with a CMG single-screw extruder, model 4530/HTM, designed to produce plastic films from 10 to 250 µm.

Below tables summarize the parameters of the blown extrusion process and the output parameters:

INPUT PARAMETERS	VALUES
Extrusion machine	Diameter 45 mm, ratio L/D = 30
Tools	Die = 100 mm and punch = 98 mm
Barrel temperature (Z01/Z07)	180°C/185°C/190°C/195°C/200°C/210°C/205°C
Filter size	150µm
Gap	2mm
Target Thickness	45µm
Cooling Type	Air
Bubble width	370mm

Table 33. Input parameters blown extrusion.

OUTPUT PARAMETERS	BATCH Nº1	BATCH Nº2	BATCH Nº3
Screw speed	44rpm	42rpm	44rpm
Engine Torque	38 A	32 A	36 A
Head pressure	212 bars	194 bars	199 bars
Cooling Air	35%	35%	35%
Drawing speed	10m/min	10m/min	10m/min
Blown pressure	42mbars	42mbars	42mbars
Swelling rate	2,5	2,5	2,5
Produced Film	300mm	300mm	300mm

Table 34. Output parameters blown extrusion.

Below pictures show aspect of filters after blown extrusion process:

Figure 8. Filter Batch Nº1

Figure 9. Filter Batch Nº2

Figure 10. Filter Batch Nº3

Conclusion of Film Extrusion process:

Laboratory report concluded on stable processing from BATCH Nº1 to 3. No smoke observed during processing. Perception of characteristic smell of the rPELD. Presence of black and metallic particles on the filter for all batches. Regarding bubble stability, it was concluded Stable bubble during the production of 30min. Some weeping movements were observed on the bubble at the die exit for the batch 1 without knowing the reason.

About Film aspect, BATCH Nº1 showed presence of the defect named “tears” probably due to the VOC’s or humidity in the rPELD, and presence of black spots. BATCH Nº2 and 3, showed good quality appearance with presence of back spots on the film.

Die showed slight material deposit along the lips of the extrusion die for BATCH Nº1. BATCH Nº2 and 3, also showed some slight deposit of degraded material along the lips of the extrusion die.

There was no additional dry of the batches before blowing extrusion.
The appearance of the reference film was visually worse than the evaluated package. Indeed, **the rate of PA does not seem to influence the processability**, moreover the UBE packaging being on a virgin LDPE base, so the part of recycled material in the batches Nº2 and Nº3 is less important than in the batch Nº1 (reference).

Below figures show Film appearance for the different batches:

Figure 11. Film Batch Nº1

Figure 12. Film Batch Nº2

Figure 13. Film Batch Nº3

2.4.10. Blown film properties

After film extrusion, film properties (dimensional, mechanical and optical properties) were analyzed. Below tables show all results obtained during film evaluation:

PROPERTIES	NORM	BATCH Nº1	BATCH Nº2		BATCH Nº3		VARIATION
Width (mm)	ISO 4592	379,6 ±0,3	396,0 ±0,5	OK	409,1 ±0,3	OK	<10%
Dimensional variation after heating _ MD (%)	NFT 54-115	79,8 ±0,5	79,6 ±0,5	OK	77,9 ±0,6	OK	
Dimensional variation after heating _ TD (%)		18,8 ±2,9	20,8 ±1,1	NO OK	26,6 ±0,8	NO OK	
Thickness by mechanical scanning (µm)	ISO 4593	41,4 ±1,7	40,7 ±3,0	OK	40,2 ±2,9	OK	(40-50) µm
Average tickness by gravimetric techniques (µm)	ISO 4591	36,2 ±0,4	34,6 ±1,5	OK	35,5 ±3,0	OK	<10%

Table 33. Dimensional properties.

PROPERTIES	NORM	BATCH Nº1	BATCH Nº2		BATCH Nº3		VARIATION
Elongation at break _ MD (%)	ISO 527-3	150 ±10	190 ±30	OK*	190 ±40	OK*	<10%
Elongation at break _ TD (%)		370 ±60	520 ±30	OK*	510 ±10	OK*	
Stress at break _ MD (MPa)		16,8 ±2,0	17,5 ±1,2	OK	16,4 ±1,7	OK	
Stress at break _ TD (MPa)		13,2 ±2,9	18,3 ±1,9	OK*	19,4 ±1,6	OK*	
Static coefficient of friction	ISO 8295	0,389 ±0,002	0,386 ±0,017	OK	0,370 ±0,008	OK	
Dynamic coefficient of friction		0,337 ±0,015	0,325 ±0,008	OK	0,322 ±0,008	OK	
Peeling force in a heated state (Temp (°C) / Peak Force (N))	KOPP	Not satisfying	Not satisfying		Not satisfying		

Table 34. Mechanical properties.

(*) Variation is >10%, in all cases showing a higher value (improved) compared to reference material BATCH Nº1.

PROPERTIES	BATCH Nº1	BATCH Nº2		BATCH Nº3		VARIATION
Spectrobolometry	L*=55,55 ±0,14	L*=56,41 ±0,04		L*=57,69 ±0,24		NA
	a*=-3,22 ±0,02	a*=-3,22 ±0,02		a*=-3,22 ±0,02		
	b*=2,85 ±0,03	b*=2,90 ±0,01		b*=2,99 ±0,03		
		ΔE=0,86	OK	ΔE=2,15	OK**	AE ≤ 2
Visuel observations (appearance, surface irregularity, unmelted)	Conforme	Conforme	OK	Conforme	OK	Variation/Reference
Odours	Recycled material Odour. No fumes	Same	OK	Same	OK	Variation/Reference

Table 35. Optical properties.

(**) ΔE>2, due to higher L value (lighter film obtained in BATCH Nº3 compared to reference BATCH Nº1.

2.4.11. Conclusion evaluation following COTREP protocol

All described processes evaluated according COTREP protocol, were successfully performed: Grinding, washing, flotation, drying, densification, extrusion and blown film were performed without any issues and good performance according to laboratory judgement.

Film extrusion process concluded that PA did not affect the processability of the recyclate.

After all steps pellet and film properties were also evaluated. All evaluations show good results, in some of the properties even exceeding the performance of control material.

As a conclusion, PA6/66 evaluated according to COTREP recycling protocol did not have any negative effect on the recyclate.

2.5. CONCLUSION MECHANICAL RECYCLING FILM CONTANING PA6/66

Results according RecyClass, APR and COTREP recycling evaluation protocols obtained for film containing PA6/66 are meeting or exceeding protocol thresholds. All evaluations have been performed by independent laboratory accredited by the different institutions.

Each protocol presents unique advantages and challenges, by assessing the recyclability of PA6/66 with different and non-harmonized protocols, it brings further pathways for repurposing PA6/66-based films which prove their good recyclability in PE-flexibles stream.

The recyclability of films containing PA6/66 has been proven by the positive assessment through three different protocols from three different institutions.