CIRCULAR FoodPack

D3.5 REGRANULATES DELIVERY WITH TECHNICAL DATA SHEETS (TDS) TO PARTNERS FOR FURTHER PROCESSING

WORK PACKAGE 3

Associated Task(s): T3.5
Mechanical recycling tests
and application development

Lead Partner: UM
Partners involved: SUEZ SA, Amcor
Dissemination Level: Public
Date: 19.12.2023

HISTORY LOG

Version number	Date	Organization name	Comments
1	2023-11-15	UM- Kim Ragaert	draft
2	2023-11-28	UM- Kim Ragaert	Final draft

QUALITY REVIEW

Quality check	Date	Status	Comments
1	2023-11-30	OK	Checked by SUEZ SA
2	2023-11-30	OK	Checked by AMCOR
3	2023-12-05	OK	Fraunhofer IVV
4	2023-12-19	Formatting	BayFOR

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

TABLE OF CONTENTS

LIST OF FIGURES

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

PUBLISHABLE EXECUTIVE SUMMARY

The EU-funded project CIRCULAR FoodPack comprises different loops, demonstrating the circular use of plastic packaging material. One of those loops aims for the circularity of PE-based mono-material food-grade packaging items, running two cycles of packaging production, sorting, and recycling.

Films marked with tracers and subsequently sorted tracer-based in a sorting line, have been washed, de-inked and were processed to regranulates for use by a convertor. This process included additional drying, densification and extrusion to regranulates, consecutively. During extrusion, 5wt% anti-oxidant stabilizer was added, as is common for poly-ethylene. The resulting granulates were of good quality and suitable for film blowing into 25 μm film at the downstream convertor. This deliverable contains the characterisation of this recyclate and the produced film.

Points of attention are material losses for each step of the process (also preceding this work) and deposition of colourant on the equipment, which must be further investigated.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

ABBREVIATIONS

Abbreviation	
TDS	Technical data sheet
TBS	Tracer based sorting
PE	Polyethylene
AO	Anti-oxidant
OIT	oxidative-induction time
DSC	Differential scanning calorimetry
MFI	Melt flow index
TSY	Tensile yield strength
EAY	Strain at yield
UTS	Ultimate tensile strength
EAB	Strain at break
TSB	Tensile break strength
E	E-modulus

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

1. INTRODUCTION

The main objective of Work Package (WP) 3 is the purification and the recycling of flexible packaging, including the characterisation of the recycled outcome. Task 3.5 more specifically targets mechanical recycling tests. Shredded and cleaned flakes are the input for re-melting, melt filtration and re-compounding. The subsequent characterisation to indicate further processing steps are also considered as part of the task. In this deliverable, the material produced as part of WP6 in a first recycling cycle will be investigated. The results are shared with the respective WP5 and WP6.

Figure 1 displays a schematic view of the entire process chain (so-called Loop 3):

First, PE-based mono-material laminates are produced by AMCOR. During the lamination process, Tracers provided by Polysecure are integrated into the food packaging items as part of the printing ink. Food packaging pouches are produced at NESTLE using these laminates. The produced flexible food packaging waste material is then mixed with the real household waste from Poitiers, France, provided by SUEZ SA. Tracer-Based-Sorting (TBS), a process developed by Polysecure, serves as the sorting protocol and takes place at the Steinert Technical Center in Pulheim, Cologne, Germany.

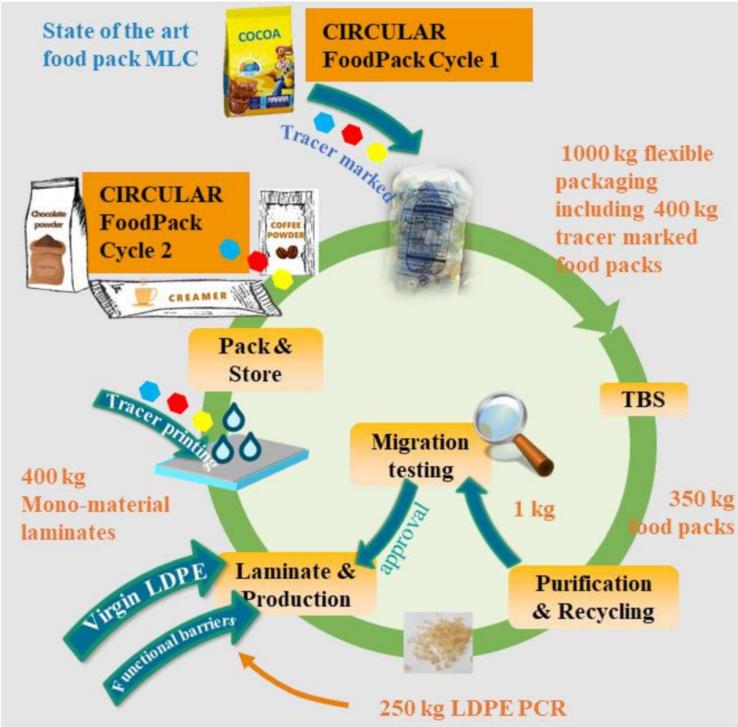


Figure 1: Overview of Loop3 as performed in CIRCULAR FoodPack. This deliverable reports on the Purification&Recycling step.

From there, the TBS sorted material is shredded and washed at Herbold (DE) and subsequently deinked and dried at NTCP (NL). The washing at Herbold and the deinking process at NTCP have been subcontracted by UGENT. The deinked flakes are then dried, densified and regranulated by SUEZ SA. It is these granulates which are the subject of this deliverable D3.5, which comprises a report of their production and a TDS. The granulates are then sent onwards to KREYEN for deodourization and finally to AMCOR for production of new laminates, which is discussed in (confidential) deliverable D5.4.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

2. MATERIALS & METHODS

2.1 Material flows

The material flows of this deliverable are shown in Figure 2. The material flowing into this process is 315 kg of Loop3 flakes coming from Deliverable D2.6, having been shredded and washed at Herbold.

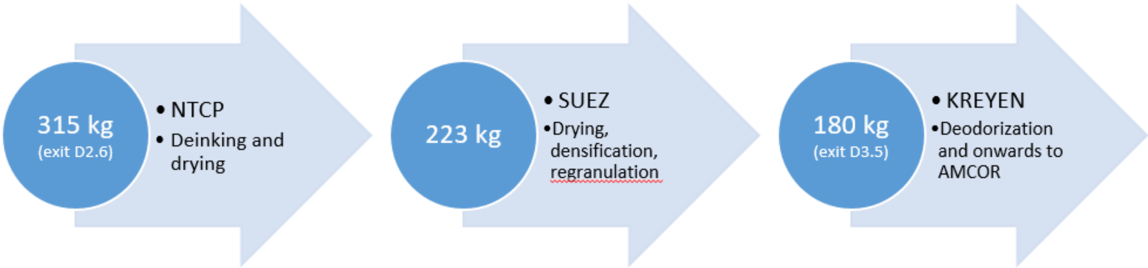


Figure 2: Material flows surrounding D3.5

223 kg of deinked material was received from NTCP, meaning a significant material loss in this step. The material was not as dry as expected, so extra drying was included at SUEZ SA, where the material was then densified (flakes to densified pellets) to enable feeding into the extruder for regranulation by extrusion. After this regranulation step, the final product is 180 kg of regranulate (pellets), which are sent on to KREYEN for deodorization and finally to AMCOR for production of new laminates.

Figure 3 gives an overview of the different shapes of the material throughout this process and the terminology used.

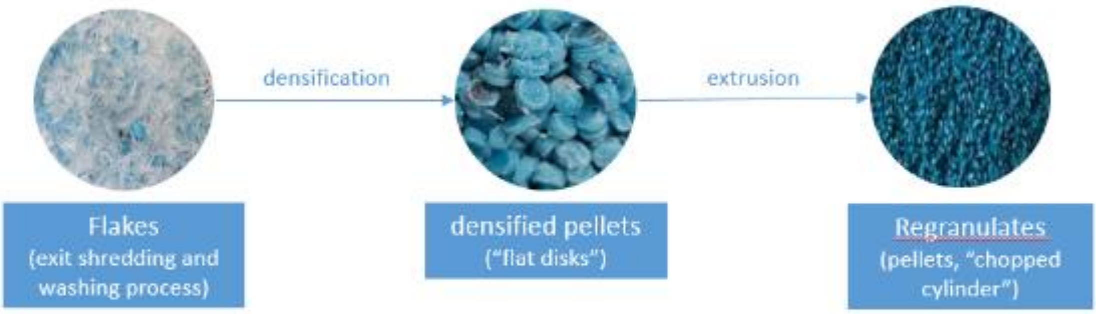


Figure 3: Nomenclature of material shapes in D3.5

2.2 Drying and densification

The materials received from NTCP were wet at around 11% of moisture, which obliged to dry all the quantities received in several batches and days to reach less than 1% of moisture (0.8% measured by thermobalance before starting the densification). Each drying treatment was 1 night at 60°. This drying step is shown in Figure 4.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

Figure 4a (left): overnight drying of flakes to reduce moisture content and 4b (right) Densifier Intarema 504K at SUEZ SA.

The dried materials were then densified at SUEZ SA over 5 different dates in 2023 (07/04, 17/04, 19/04, 21/04 and 02/05), using a Densifier Intarema 504K from EREMA as shown in Figure 4b (densification at 90°C, barrel and die at 170°C and 160°C respectively) .

2.3 Granulation

The densified pellets were extruded to regranulates at SUEZ SA over 5 different dates in 2023 (26/06, 27/06, 28/06, 04/07 and 05/07), using a co-rotating twin-screw Extruder LTE26-44 from Labtech (Diameter 26, 44D) with degassing. The extruder barrel temperature was 200°C and melt filtration was done with a mesh size of 112 µm. 5 wt% of antioxidant (AO) stabilizer Polybatch AO25 (Schulman) was added gravimetrically. This selected AO is a combination of the stabilizers Irganox 1010 and Irgafos 168. The TDS of the AO is added in Appendix 2. The operating settings of the extruder and the extrudate leaving the extruder are shown in Figure 5.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

Figure 5: Operating settings and die exit of the Labtech extruder at SUEZ SA.

2.4 Film blowing

The regranulates were successfully blown into film at AMCOR using standard processing parameters on a 1-Layer blown line (Dr. Collin 25 E, Die 50mm) for characterisation on film properties. A film sample is shown in Figure 6 and the processing parameters are shown in Figure 7.

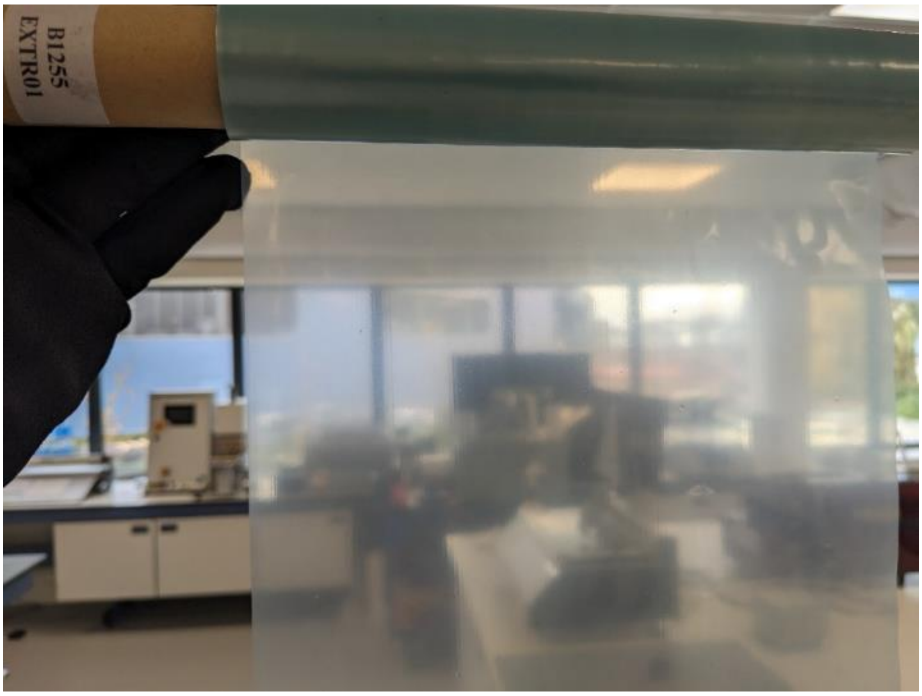


Figure 6: Film extruded by AMCOR, with the recycled granulates.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

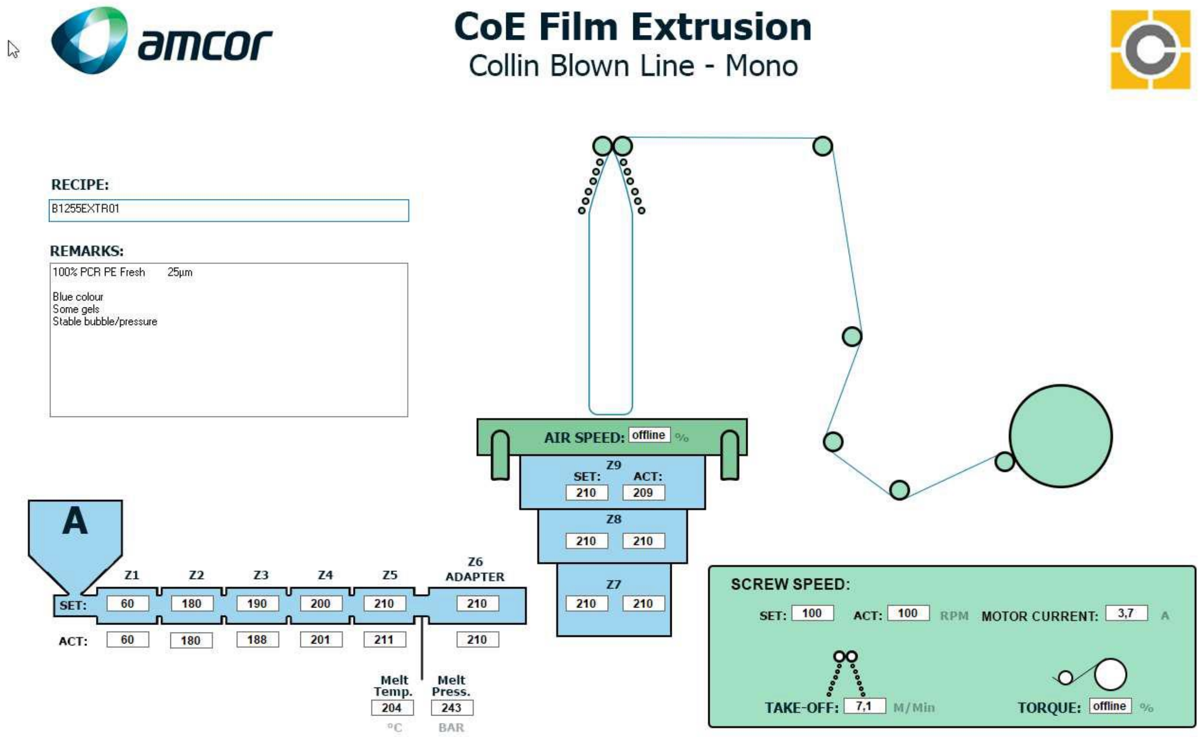


Figure 7: Processing parameters on the CFP Loop3-Cycle1 generated PCR

2.5 Characterization and TDS

OIT (oxidative-induction time) was measured at Fraunhofer using DSC 200F3 Maia from Netzsch, at 200°C and under an oxygen atmosphere. It is a measure of the material's stability in downstream processing. Melt temperature was determined on the same device, at a heating rate of 10°C/min under a nitrogen atmosphere.

Melt flow index (MFI) was measured at SUEZ SA using a CEAST MF20 from INSTRON at 190°C and using a weight of 2.16kg, in accordance with EN ISO 1133-1.

Coefficient of Friction was measured using the 32-06 friction tester from Testing Machines Inc. following the ASTM D-1894. Films were tested inside to inside and outside to outside to display the effect of the calendering when the bubble is nipped on the film blowing line.

Tensile properties were measured using a Houndsfield 1 ST tensile tester using a load cell of 250 N at 500mm/min (method for samples displaying elongation >100%). Sample length of the films was 50mm with a width of 15mm. Samples were conditioned for 24h at 50% RH and 23°C before tesing. Film thickness was measured at least 3 points in the testing zone of which the minimum thickness was taken to proceed with the calculation of stres and strain.

Gel count was performed following the Recyclass protocol described in the Recyclability Evaluation protocol for PE films. Here, gels with a diameter larger than 200µm were counted. Image J was used as software tool to threshold for the gels in the scanned area of 100x100mm squares.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

Tear resistance was measured using a Lorentzen & Wettre SE009ED tearing tester using the Elmendorf method with precuts with a radius of 43 mm and precut made. The ASTM D1922 norm was followed for the analysis. The pendulum used for the TR measurement was pendulum B.

The TDS of the regranulate is built using the property data gathered in this D3.5 and product (film) data, as tested downstream by AMCOR. Details of the film production process are confidential and part of D5.4.

3. DISCUSSION OF THE RESULTS

3.1 Processing-related properties of the regranulates

The evolution of OIT and MFI is shown in Table 1.

Table 1: OIT and MFI of the flakes, densified pellets and regranulates.

	flakes	densified pellets	regranulates
OIT [min]	3.35	1.25	14.12
MFI [g/10 min]	NA	0.89	0.94

OIT is an indicator of how stable the material will be in further processing. As regranulates are sent on to a convertor, where they will be melt processed again into the final product, this is an important quality marker. Without sufficient OIT values, the polymer will degrade during the conversion process, leading to inferior properties. For this reason, AO stabilizers are added during the granulation, which is a common industrial practice also for virgin PE. By adding 5 wt% of the AO masterbatch, the OIT is increased to over 14 minutes, which is considered acceptable (a lower acceptance limit of 12 minutes was used). Previously, the OIT is reduced by the densification process (compared to flakes), due to the thermo-mechanical stresses occurring in this process.

The MFI (190 °C, 2.16 kg) of the regranulates is just below 1 g/10 min, which is a typical value for film grades. The material is therefore suitable for the film blowing process, which is to follow at Amcor.

Furthermore, the DSC measurement (graph attached in Appendix 3) showed two melting peaks, at 108°C and 125°C, which are representative for LDPE and LLDPE/HDPE respectively, thereby confirming the expected composition of the material.

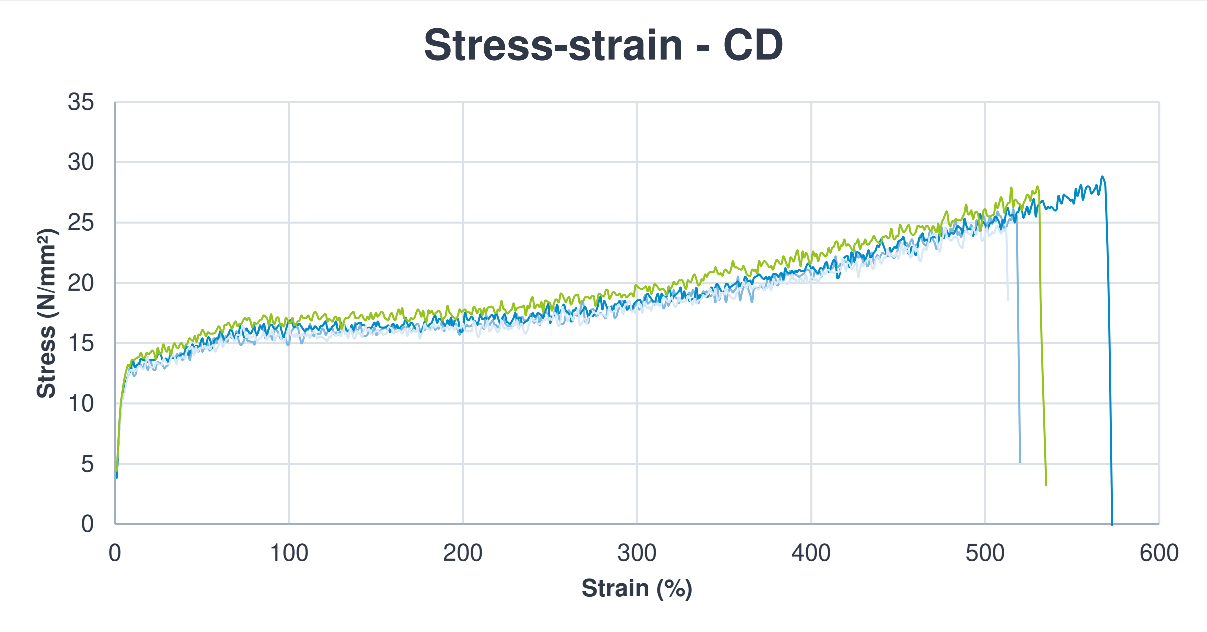
3.2 Properties of the films produced from the regranulates

The properties measured on these films are tabulated in Table 2. Figure 8 displays a typical stress-strain curve measured on the film samples.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

Table 2: Film properties determined from the tensile curves above.

		Thickness (µm)	TSY (N/mm²)	EAY (%)	UTS (N/mm²)	EAB (%)	TSB (N/mm²)	E (MPa)
MD	**Average**	**25**	**8,9**	**2,8**	**27,0**	**478**	**26,3**	**253**
	STD D	**1**	**0,5**	**0,1**	**1,4**	**19**	**1,4**	**5**
TD	**Average**		**10,5**	**3,0**	**24,3**	**553**	**24,0**	
	STD D		**0,2**	**0,3**	**2,4**	**34**	**2,6**	



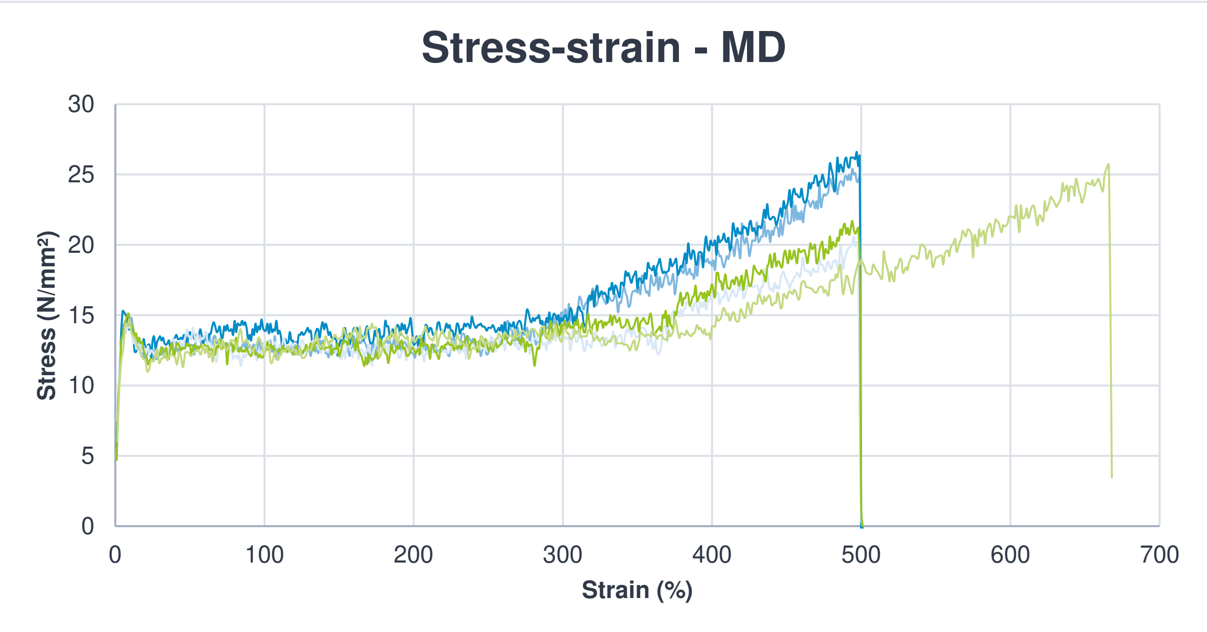


Figure 8: Stress-strain curves of the blown films of Loop3-Cycle1 PCR

Overall, one can conclude that the properties of the PCR film are performing on par with some of the virgin PE resins commercially available. The film displays **good elongation**, both in the MD and TD, which already indicates that the presence of the gels are not decreasing the mechanical performance notably. The deviation on the film thickness measured remains acceptable indicating again low influence of the gels present. In case of thick prominent gels the film thickness would vary much more prominently. Finally, the **gel counts** measured and displayed

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

in Figure 9 and further tabulated in Table 3 show a minor presence of gels which meet the acceptance criteria of the Recyclass evaluation protocol (<100 gels in 100x100mm square), although the material could be processed at 100 % without diluting it with virgin grades.

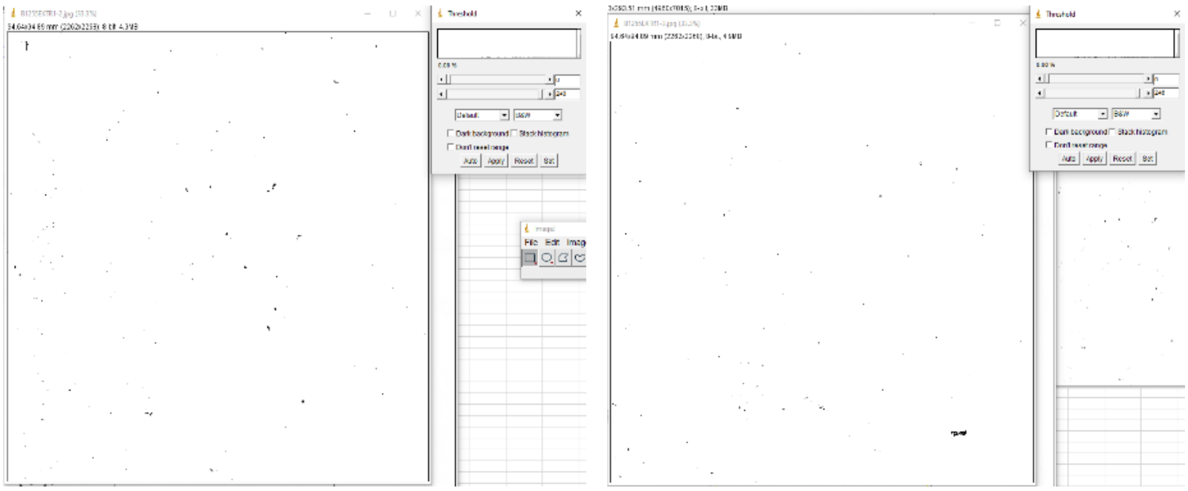


Figure 9: Gel counts on the extruded PCR films

Table 3: Gel count on the analysed films

gels/cm²	2,31
gels/cm² (APR proto	0,41
average gel area (m	0,025
max gel area (mm²)	1,908
min gel area (mm²)	0,002

There are two more properties that indicate well the performance of a PCR film for later processing: the **coefficient of friction** and **the tear resistance**. The former is a measure for the smoothness of a film where a lower coefficient indicates that the surfaces are slicker - there is less resistance to the sliding motion. This gives a good indication whether or not the film will be easy to manipulate over the packaging line without further blocking of clinging. Ideally the values for a virgin film would be 0,35 or lower in the dynamic mode and 0,4 or lower in the static mode. The latter indicates the resistance to the propagation of a tear. Although the orientation of a film contributes to a large amount to this factor, it is facilitated by the presence of stress concentrators, which gels could represent. COF values are displayed in Table 4 while the tear resistance values are displayed in Table 5.

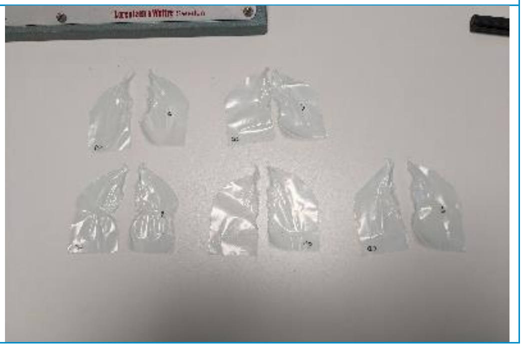
Table 4: COF values of the PCR film

COF inside / inside					
Static			Dynamic		
Average	SD	95% CI	Average	SD	95% CI
0,39	0,01	0,01	0,32	0,01	0,01
COF outside / outside					
Static			Dynamic		
Average	SD	95% CI	Average	SD	95% CI
0,42	0,01	0,01	0,34	0,01	0,01

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

Table 5: Elmendorf Tear resistance values

Tear resistance (mN)					
MD			TD		
Average	SD	95% CI	Average	SD	95% CI
1346	226	237	6098	272	285



Tear resistance on the measured films do not indicate any facilitation of tear propagation by the gels present as the results line up with those typically measured for virgin PE's. The extra orientation in the MD, which is also apparent in the strain hardening visible in the tensile curves results in a lower tear resistance.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

3.3 Points of attention

3.3.1 Colour contamination of equipment

After both the densification and extrusion steps, it was noted during the cleaning of the equipment that there was a deposition of blue colour. This was the most significant for the knives inside the reactor of the densification machine, as can be seen in Figure 10a.

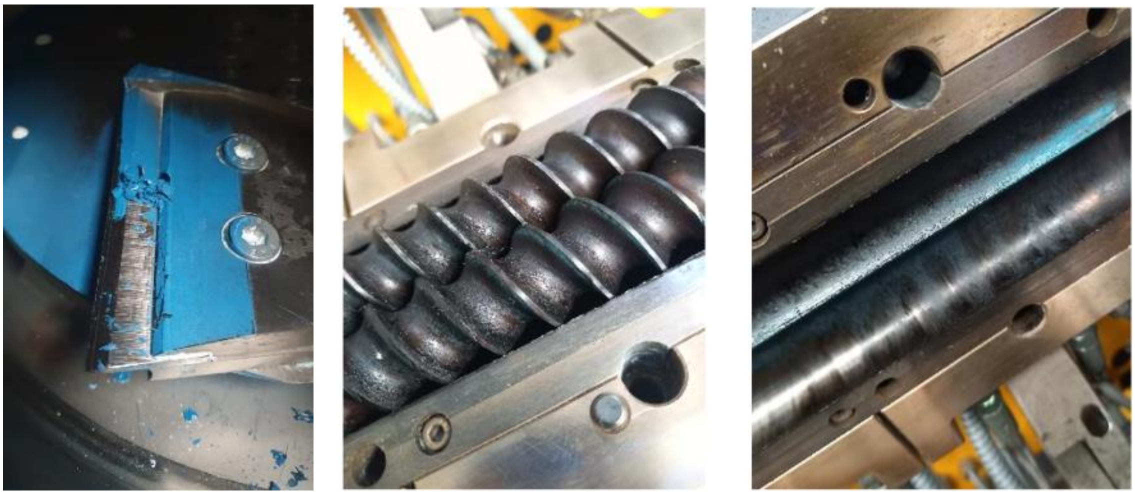


Figure 10a (left): significant blue colour deposition on the blades in the densifier and 10b (middle and right): minor blue residue on screws and shaft of extruder.

The extruder screws and shaft were also affected with the blue colour (Figure 10b), but to a much lesser degree.

This is a major point of attention for further trials and potential upscaling to industrial scale. The cause remains to be investigated. One avenue of thought is that the de-inking process did weaken the bond between inks and polymer, thereby making it easy for them to agglomerate (and deposit) under the friction induced in the densifier. The de-inking improvements in the project are ongoing, and will follow up with the materials of Loop3-Cycle2 and Loop2 in WP6 of the project.

3.3.2 Losses

For every material processing step, losses are noted. 5 kg was lost in the densification and a further 38 kg in extrusion, as shown in Figure 11. The losses in extrusion are relatively high, due to the experimental nature of this trial. It was the first time processing of this material at SUEZ SA and therefore, ideal settings needed to be determined. Furthermore, extrusion is a process where (for small batches like this) 10% is a typical mass loss due to start-up and end-of-process cleaning. Within the bounds of CIRCULAR FoodPack, it is not likely that these losses could be reduced, but looking forward to continuous industrial operation; it should not be an issue.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

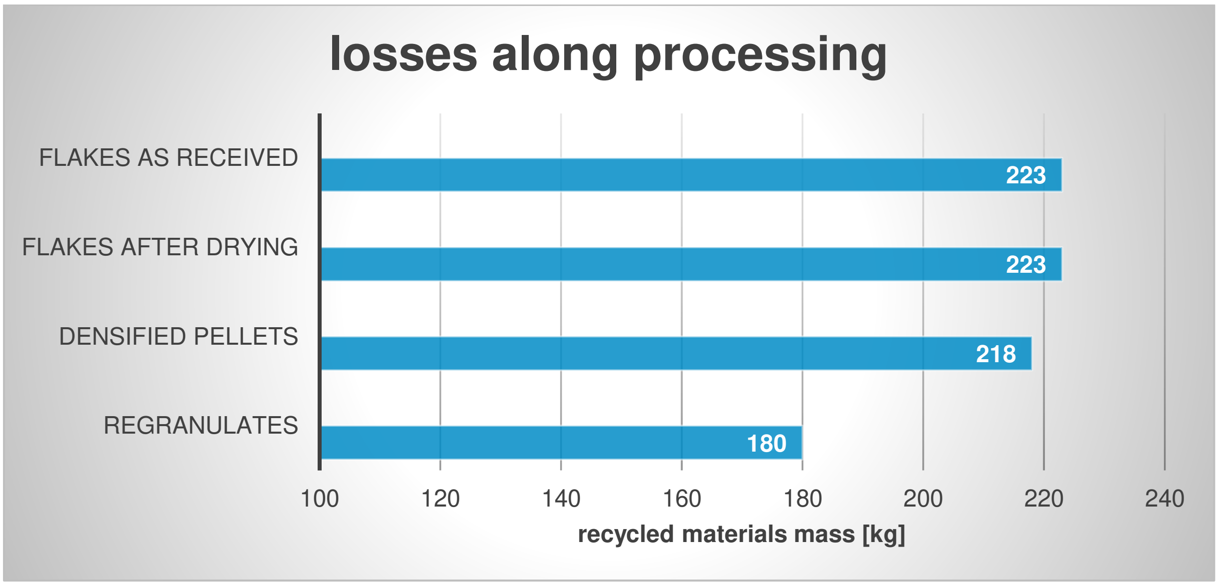


Figure 11: Material mass after each processing step, giving insight into the material losses throughout the process.

4. CONCLUSION

The de-inked recycled plastics from TBS in Loop 3 were successfully extruded to quality regranulates for downstream conversion to film. A TDS was drafted and is added to this deliverable. It confirms the regranulates is suitable for film blowing.

A remaining point of attention is the observed deposition of colorant on the densification and extrusion equipment, which requires further investigation. Therefore, further deinking process optimisations are still ongoing in the project.

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

APPENDIX 1 – REGRANULATE TDS

PROPERTY		UNIT	TYPICAL VALUE	COMMENTS	TEST METHOD
GENERAL					
Name		-	CFP_Loop3_Cycle1		
Type			PE regranulate		
APPEARANCE					
Color		-	Blue-green		
Shape		-	pellets		
PHYSICAL					
Melt Flow Index (MFI)		g/10 min	0.94 2.62	2.16 kg, 190 °C 5 kg, 190 °C	ISO 1133
OIT		Min	>12	200°C, oxygen	DSC
Melt peak temperature		°C	125	10°C/min, nitrogen	DSC
MECHANICAL – blown film, specimen type 5 (Thickness= 25 µm)					
Modulus	MD	MPa	258 ± 6		ISO 527
Tensile strength	MD	MPa	27 ± 2		ISO 527
Strain at break	MD	%	530 ± 48		ISO 527
Tear strength	MD	N	1346 ± 226	200 mm/min	ISO 6383

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

APPENDIX 2 – TDS OF POLYBATCH AO25

A. Schulman

POLYBATCH® AO 25
Masterbatch

Product Description

POLYBATCH® AO 25 and AO 25 S are antioxidant masterbatches for the thermal stabilisation of LDPE, HDPE, LLDPE and EVA. They should be added during the extrusion process where high temperatures or long residence times cause degradation, cross-linking, gel formation, carbonised particles and/or loss of mechanical properties.

POLYBATCH® AO 25 : is the preferred stabiliser system for blown film, profile extrusion, etc. where the processing temperature does not exceed 280°C.

General

Material Status	• Commercial: Active		
Availability	• Asia Pacific	• Europe	• North America
Uses	• Masterbatch		
Agency Ratings	• EU 2002/72/EC [1] • FDA 21 CFR 177.1520	• FDA 21 CFR 177.1520(c) 2.1 • FDA 21 CFR 177.1520(c) 2.2	
Appearance	• Translucent		
Forms	• Pellets		
Processing Method	• Blown Film • Cast Film • Double-Bubble Film	• Extrusion Blow Molding • Pipe Extrusion • Profile Extrusion	• Sheet Extrusion

Physical	Nominal Value (English)	Nominal Value (SI)
Specific Gravity	0.932	0.930 g/cm³
Bulk Density	34.3 lb/ft³	550 kg/m³
Moisture Content	< 1500 ppm	< 1500 ppm

Usage

The addition rate of the POLYBATCH® AO 25 and/or AO 25 S depends on the processing temperature and the residence time of the material in the machine.

Stabilisation improvement: 1 - 2%
Extrusion of rework: 2 - 3%
During recycling: 2 - 3%
Stabilisation over a week-end shut-down: 5 - 10%

Regulatory

FDA: max. 5%
BGA: max. 5%
EEC: Section 2 max. 60 mg/kg food, SML = 6 mg/kg

Packaging & Storage

POLYBATCH® AO 25 and/or AO 25 S are packed in 25 kg bags on shrink-wrapped pallets. POLYBATCH® AO 25 and/or AO 25 S can be stored up to maximum 6 months at 25°C for optimum performance. Higher temperatures might reduce storage time considerably.

Notes

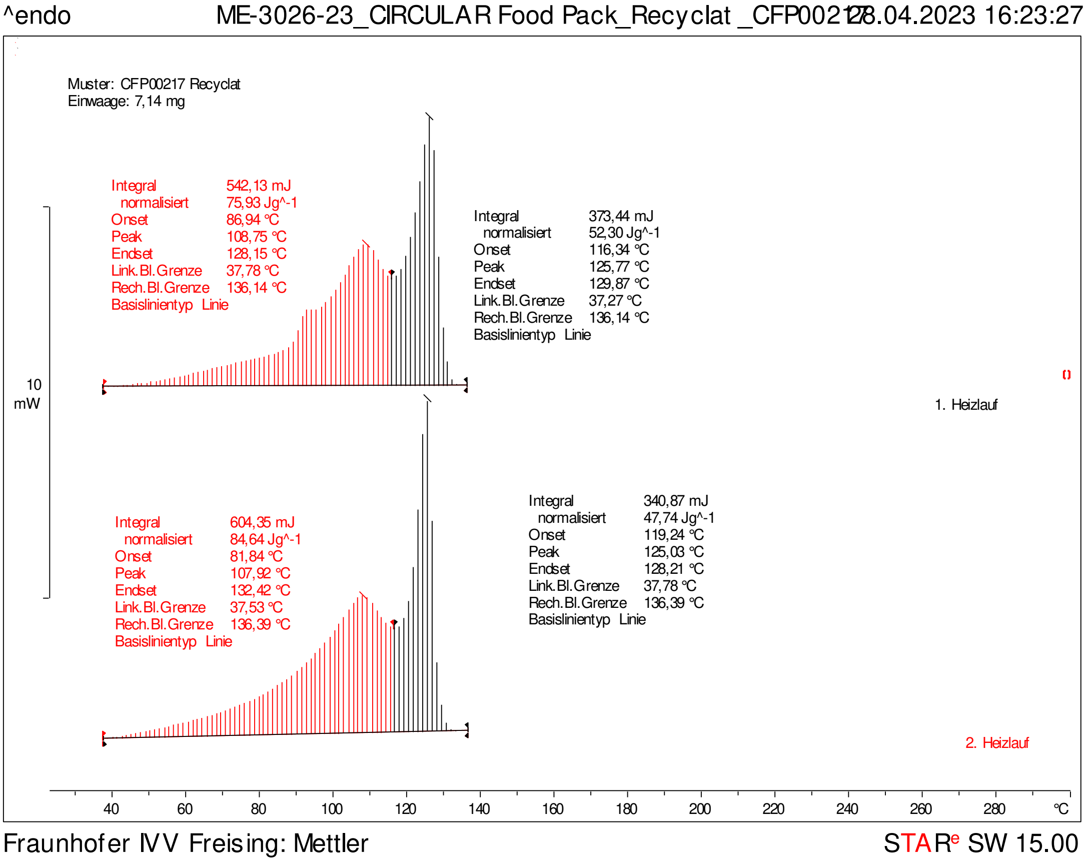
[1] Commission Directive 2002/72/EC and its successive amendments up to and including 2009/975/EC.

Page: 1 of 1

www.aschulman.com
Publish Date: 2016-09-08

This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.

APPENDIX 3 – DSC OF THE REGRANULATE



This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No. 101003806.