Thermal and Chemical recycling of textile waste

CircTex Interreg project

Interreg North-West Europe CIRCTEX

European Regional Development Fund

EUROPEAN UNION

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Contents

1 CircTex project

CircTex focuses on the development of recycling and production techniques in a closed-loop process textile chain for polyester (PET) workwear. The textile industry in the NWE zone consumes 17 million tonnes of non-renewable raw materials annually. That is why the transition to a circular economy within the textile industry is urgent.

Circularity within the textile industry can be best initiated through industrial clothing, especially Personal Protective Equipment (PPE). Given the prohibition on reusing PPE garments, this sector is entirely dependent on the procurement of new materials. Additionally, the quality standards and usage impact in this particular domain exceed those observed in other textile sectors. Consequently, the outcomes achieved through the CircTex project will equally benefit and apply to the broader textile industry.

The following technologies and processes for circular PET workwear will be developed:

- Mechanical and chemical recycling
- Spinning and weaving
- No-waste production
- Wear2 yarn and industrial microwave for assembly and disassembly
- Recyclable laminates, logos and accessories
- Collection and sorting systems

CircTex provided a proof-of-concept for fully recyclable workwear within the NWE region by developing technologies for large-scale processing of PET workwear, which was validated and demonstrated in a closed chain pilot.

2 different fabrics were used to produce PET workwear:

- A fabric of 100 % polyester form PET bottles dyed in high-vis orange.
- A fabric from 20% production waste and 80% PET bottles

The material and yarns were produced by means of a thermomechanical recycling process.

After use the garments were disassembled, the polyester material, that should be as pure as possible were chemically recycled via a depolymerization process.

Research was done on how to improve the thermomechanical process and on the feasibility of a chemical process to create the same garment in a High-visibility version with the ability to protect against rain.

This report resumes the lessons learned on the recycling process of polyester and on closing the loop.

2 Recycling options for polyester

Recycling end-of-life material

The first step to recycle a material is to choose a suitable technology for the waste stream to be recycled. To recycle a textile there are several options. In the study on the technical, regulatory, economic, and environmental effectiveness of textile fibres recycling from the European Commission (Duhoux, et al., 2021) the recycling categories are defined as in Figure 1.

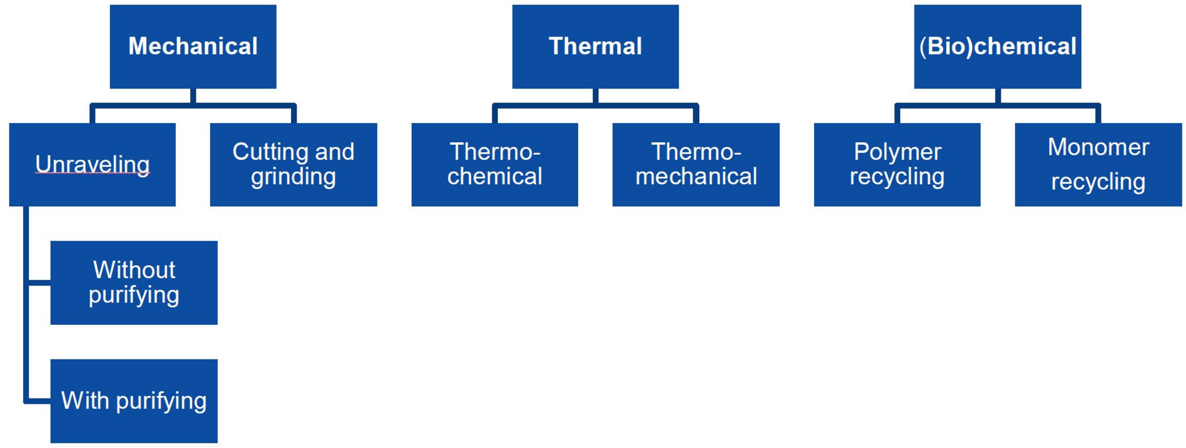


Figure 1 Categorization of textile recycling technologies (Duhoux, et al., 2021)

Depending on the conditions, various techniques can be employed to handle the waste stream, including mechanical recycling, thermo-mechanical recycling, thermo-chemical recycling, enzymatic monomer recycling, and chemical monomer recycling. As the Interreg CircTex project applies circular-design principles a near 100% polyester waste stream is created. The fabric, buttons, sewing yarn and labels are made from 100% polyester. The membrane used to laminate the fabric to make the garment compliant with EN 343 – "Protective clothing - Protection against rain" is made from a polyester layer and co-polyester glue. The reflection striping and zippers are sewn on with the Wear2Go yarns so that there was still a very low % of PU material presence because it was not possible to create a 100% polyester-based transfer print and seam tape within the foreseen timeframe.

The CircTex partners intended to obtain a garment made from used garments preserving their original function. There was also the desire to close the loop of the fibre content in the garment. This means all polyester present is re-processed in polyester fibres for yarn and fabric production. This calls for a technology that must meet the following criteria:

- All material in the process is converted into a polyester fibre.
- Contaminations are tackled by removing dyestuff, small amounts of PU and possible contaminations from the user phase.
- The output needs to be a polyester material or fibre with a virgin-like output so it can be certified according to EN 343 – "Protective clothing - Protection against rain" and EN ISO 20471 – "High visibility clothing - Test methods and requirements".

- The output needs to be colourless in order to dye the fabric in a high-vis colour and certify the garment according to EN ISO 20471 – "High visibility clothing - Test methods and requirements".
- All chemicals from both production and contamination from the user phase need to be removed to be compliant with the OEKO-TEX® standard 100 requirements.

Most polyester is currently recycled mechanically (textile to textile from different sources) and thermos-mechanically (PET bottles to fibres or polyester production waste leading to a partial recycled fibre). Many startups are developing capacity in chemical monomer recycling, enzymatic monomer recycling and thermomechanical recycling. It is considered that the technology will improve, and the capacity will increase soon. (Textile exchange, 2020)

As stated in the study on textile recycling technologies (Duhoux, et al., 2021) mechanical recycling is unable to yield 100% spinnable fibres from polyester waste material. In addition, some products contained laminated fabrics and taped seams (CE-certified protective garment according EN 343:2019 – "Protective clothing - Protection against rain"). Even if the seams can be removed the remaining fabric with the laminated membrane would not lead to spinnable fibres when mechanically processed. In addition, the study on textile recycling technologies (Duhoux, et al., 2021) pointed out that both mechanical and thermomechanical technologies result in lower quality of materials so that an amount of virgin material must be added. The presence of certain materials like PU and Co-polyester glue would also hinder the thermomechanical recycling process. In addition, all non-removable contamination from the use phase, dyestuff and other chemicals would stay inside the polyester material as currently there is no dyestuff removal technology available, nor can all contaminations be removed prior to recycling. This would mean that the material would not lead to a fabric that could be CE-certified as intended.

This reduced the choice to enzymatic monomer recycling, chemical monomer recycling or thermochemical recycling. Since thermochemical recycling leads to feedstock for polyester production and enzymatic monomer recycling is not fully developed yet, chemical monomer recycling was considered the only suitable technology.

CircTex development of a no waste production process

In the Interreg CircTex project test garments were produced leading to production waste. We obtained the following production waste:

- **Spinning**: the initial output of the extruder is extracted while the yarn is guided around the cooling rolls in the designated area. During this process, the yarn is also elongated, until it is wound onto the cones that collect the cooled down extruded yarn. Additionally, the texturizing processes contribute to minimizing waste.
- **Warping and weaving**: waste is generated in the form of yarn ends and selvedges.
- **Finishing**: possible small amounts of fabric waste
- **Garment production** involves the creation of waste during the cutting and sewing stages. Waste is generated when the parts of the garment are cut out, and there is also a small amount of waste produced during certain sewing processes.

All this waste is 100% polyester and there is a zero chance of forbidden legacy chemicals because it is newly produced. Moreover, the content of this waste stream is known so it is possible to estimate whether the material can be processed via a thermophysical recycling process. The consortium selected this recycling technology for the production waste.

3 Thermomechanical recycling

Technology

The thermomechanical recycling process is very similar to a normal thermoplastic yarn extrusion process. Figure 2 show the general thermo-mechanical recycling process scheme. Because polyester is a thermoplastic material this is an option for polyester.

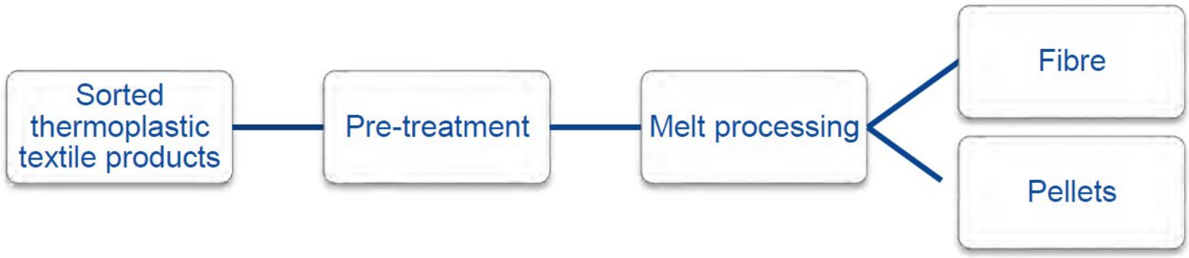


Figure 2 General process scheme for a thermo-mechanical recycling process ((Duhoux, et al., 2021)

A disadvantage of the thermo-mechanical recycling process is the degradation of the polymer due to hydrolysis resulting in viscosity issues. Depending on the polymer this is more or less problematic. In the case of PET (owing to polymer degradation, which is inherent to the thermal process) suppliers of thermo-mechanical recycling machinery offer solutions to improve recyclate quality such as:

- High-level (vacuum) degassing for removal of volatile contaminants and moisture as well as viscosity stabilization for PET.
- High-performance filtration for removal of non-melting particles.
- Solid state or liquid state polymerization units (SSP or LSP) to increase PET intrinsic viscosity (IV).

Intrinsic viscosity of PET can also be enhanced by adding chain extenders. The chemical chain-extending method is widely used to modify the IV of PET because it has a low energy consumption and high efficiency compared to solid-state polycondensation and melt polycondensation. (Zhoa, et al., 2020) Several chain extenders are commercially available from companies such as BASF, Sukano, Nexam Chemicals, etc.

Project partner Antex applies a polycondensation process in its recycling line. Therefore, the CircTex project examined the influence of viscosity boosters.

In addition to viscosity there is an issue with chemical content when a thermo-mechanical recycling process is used to recycle thermoplastic materials. All chemicals, except probably the partial release (removal) of volatile chemicals during the process due to the heating up of the materials, remain inside the materials (Duhoux, et al., 2021). Thus, colorants remain inside the material. In many cases, a continuous operation must be maintained to obtain a critical mass. This leads to the combination of polyester in different colours with various dyestuffs and pigments.

CircTex also investigated the possibility of colour sorting of dye processes to obtain a uniform colour.

Improving the viscosity

Fluorescent jacket inspection

The composition of the different layers of the CircTex fluorescent jacket is determined to ensure that we would test 100% polyester-based materials. This is required by the thermo-mechanical recycling process of the material.

<u>Jacket construction</u>

- The outside of the jacket is orange and contains reflective strips.
- The inside is lined with a black fabric.
- Once the lining is removed, a knitted net structure is visible.
- Between the mesh structure and the orange layer of the jacket is a polyester film.
- Where the seams are reinforced, a reinforcement band made of polyester is observed, in addition to the mesh structure and foil.
- The reinforcement band itself is glued onto the mesh structure with a polyurethane-based adhesive.
- A cross-section of the jacket was heated using melting microscopy. From 255°C onwards, all layers melt away completely.

<u>Photos of the received jacket</u>

Figure 3 Received sample.

Figure 4 Received sample detail.

Microscopy results

Microscopic examination reveals the presence of a transparent film underneath the orange top layer followed by a textile layer in net structure visible on Figure 5. Over the seams in the orange fabric there is a black seam tape glued on top of the mesh structure.

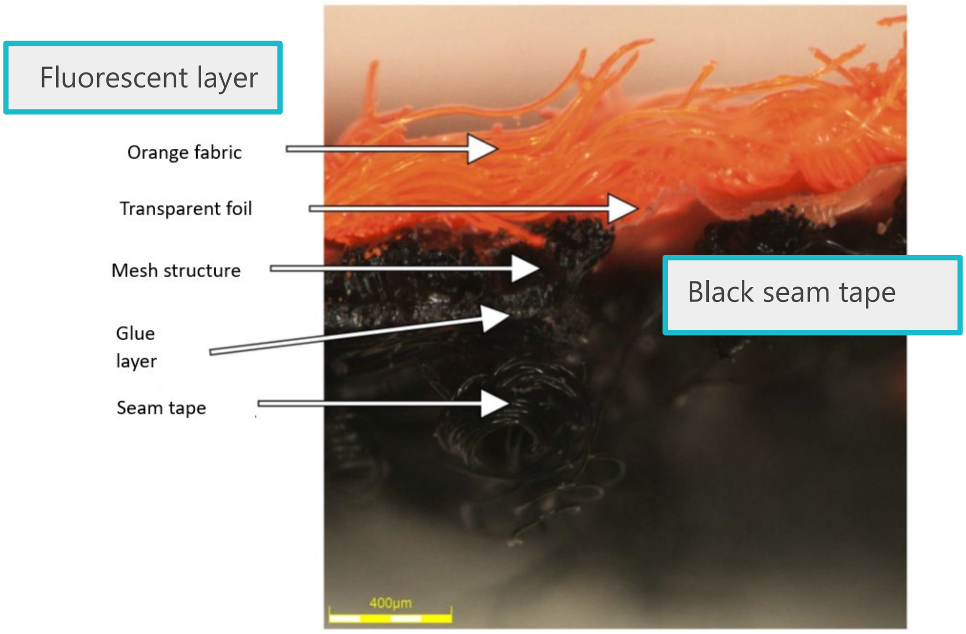


Figure 5 Cross section of the receive sample.

Figure 6 is a diagonal cut of the seam structure to show the built up. From outside to inside, the orange top layer, the transparent foil, the mesh structure the glue (adhesive) layer and the black seam tape can be seen. The area on which this cut was made did not contain a reflective strip on the orange textile. The black liner was removed to clearly visualise the different layers.

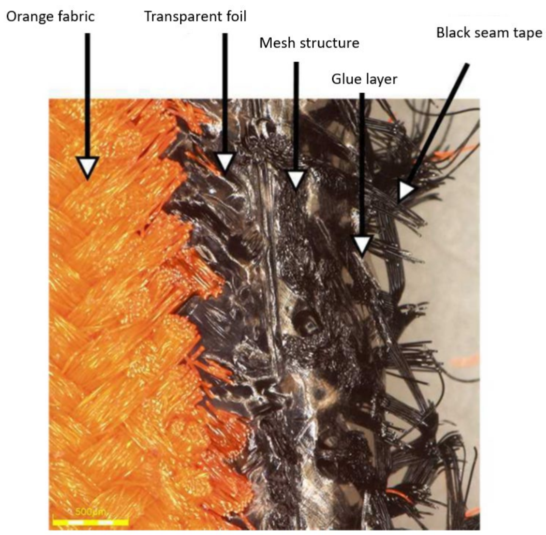


Figure 6 Diagonal cut of the sample.

FTIR analysis

The orange fabric, transparent film, mesh structure and reinforcement tape show similarity to reference spectrum of polyester. The adhesive layer between the mesh structure and the reinforcement tape shows similarity to a polyurethane.

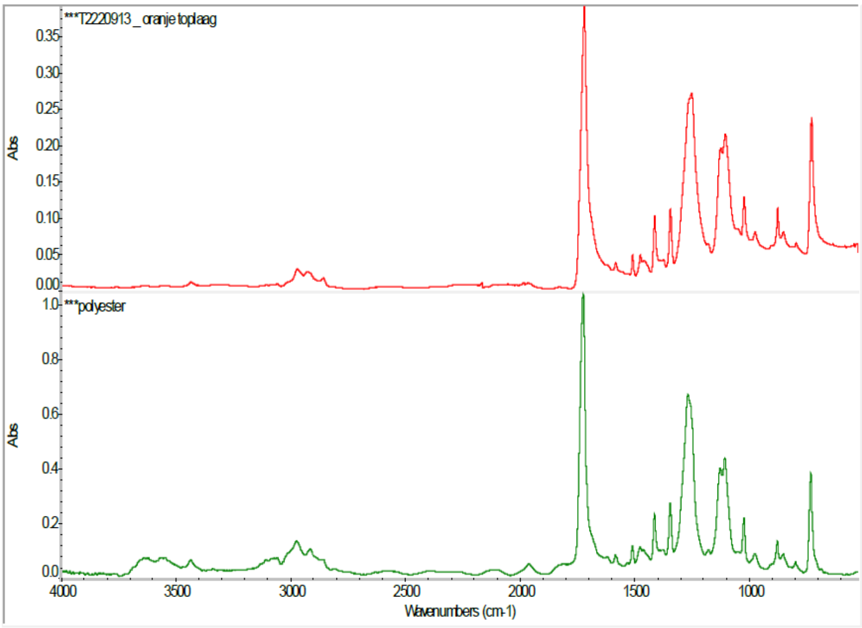


Figure 7 FTIR spectrum of the orange fabric

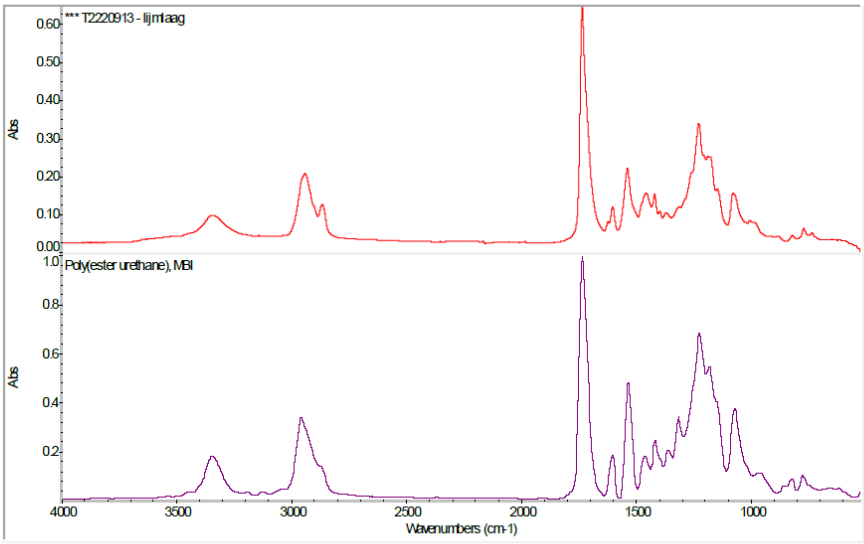


Figure 8 FTIR spectrum of the adhesive (glue) layer

Conclusion

All the material studied is polyester-based, except for the polyurethane-based adhesive layer of the seam tape used to make the seams watertight. For this reason, the jacket could not be used for the trials. Instead, a virgin grade polyester was used for the viscosity trials.

It is possible to chemically recycle the vest after removal of the reflection stripes and zippers (See Chemical recycling).

Influence of PET-reprocessing on the viscosity and the application of booster additives

Virgin PET reprocessing

Virgin PET RT20 (melt spinning grade) was repeatedly processed via compounding to study the viscosity decrease with each (re)processing step. Compounding was done on a Leistritz twin-screw extruder. This pilot-scale compounder, with a throughput up to 25 kg/h, has all the specific parameters of the large-scale production machines. The diameter and the total length of the screw are 27 mm and 120 mm, respectively. The polymer melt was visually more liquid from the 3rd cycle onwards and very liquid after cycle 5 making it more difficult to pelletize. Intrinsic viscosity measurements were performed and revealed indeed a considerable decrease in viscosity with each step and especially after the third reprocessing cycle (see Figure 3 Intrinsic viscosity of PET RT20 upon each reprocessing step)

Cycle	IV (ml/g)
0	63
1	60
2	57
3	51
4	51
5	45

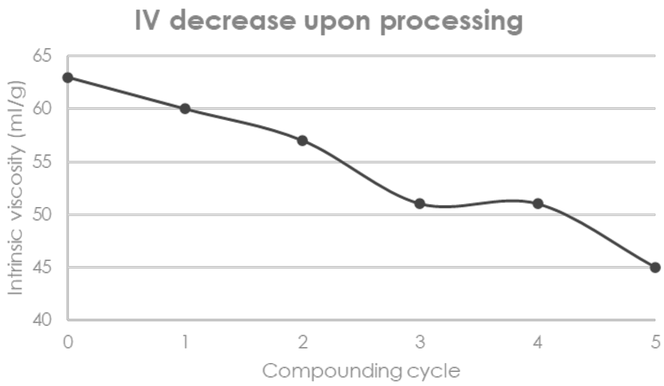


Figure 9 Intrinsic viscosity of PET RT20 upon each reprocessing step

Comparison of viscosity boosting additives

Three different viscosities enhancing or stabilizing additives were selected to study their effect on PET reprocessing. An overview of these additives is presented in Figure 1. Compounding PET RT20 was performed to simulate a first processing step. In a second compounding step, the different types of viscosity boosters were added with a concentration of 5%.

Booster	Producer/supplier	Info
VIBATAN PET 03933	Viba/WENR plastics	Contains a "chain extender" additive carried on a special copolymer polyester Recommended dosage: 10%
ADK STAB PEP-36	Adeka	Phosphite that should prevent and compensate the degradation of polyester
Nexamite M021200	Nexam Chemical	PET-based masterbatch containing a branched chain-extender for polyesters (pyromellitic dianhydride) Recommended dosage: 1-3%

Table 1 Selected viscosity boosters for PET recycling.

Compounding was successful with the Vibatan and ADK STAB additives but failed for the Nexamite M021200 because of a too high viscosity, even after lowering the concentration to 3%. The latter booster seemed too strong for the limited decrease in viscosity after 1 processing cycle.

Next, a tape extrusion trial was performed to compare the effect of the different boosters on processability, and filament properties Extrusion was performed on the tape/monofilament extrusion line at CTB. As illustrated in Figure 4, this line is equipped with a water bath for quenching, a set of cold drawing rolls, one intermediate oven, a set of heated drawing rolls and relaxation rolls.

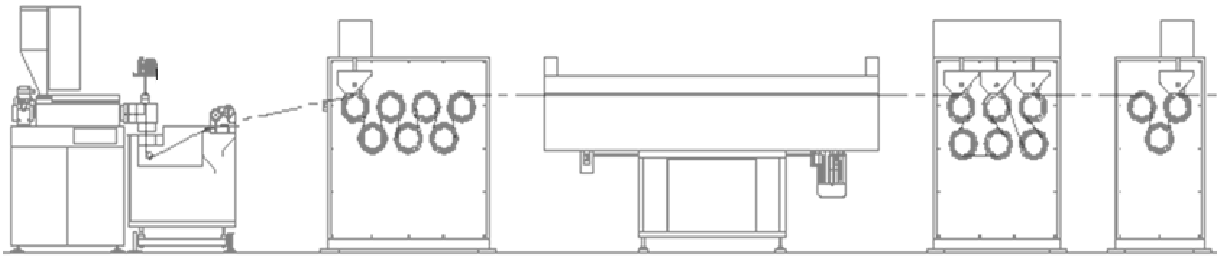


Figure 10 Schematic representation of Centexbel's monofilament/tape line

As illustrated in Table 2 , neither the Vibatan or the ADK STAB additive had an enhancing effect on the filament extrusion process. It can be argued that the viscosity decrease after 1 processing step was not sufficient to have a significant effect on the reprocessing into filaments.

Material	Extrusion	Filaments	Max. DR
PET RT20	Stable	OK	5.65
PET RT20 + VIBATAN compound	Stable	OK	5.25
PET RT20 + ADK STAB compound	NOK	jagged	/
PET RT20 compounded twice	Stable	OK	5.25
PET RT20 compounded twice + VIBATAN dry blend	Stable	OK	5.25

Table 2 Results of the filament extrusion process: indication of extrusion process, filament quality and maximum draw ratio.

A search for additional enhancing additives was performed and grades selected from Sukano, BASF and Nexam Chemicals, will be tested on post-industrial rPET. This will be tested outside the CircTex project.

Dyeing process of thermo-mechanically recycled polyester

Polyester can be dyed by:

- Using disperse dyestuff in a separate dyeing process. This type of dyeing is applicable in small and large batches.
- Dope-dyeing using a pigment that is distributed via a masterbatch during the spinning process. This type of dyeing is applicable in large batches due to the high material loss in the case of switching colours. This is both costly and environmentally unfriendly because it produces too much waste.

The attempt to dye recycled polyester using disperse dyes in order to achieve a uniform colour with various shades was not successful. The dyeing process did not adequately cover the different colours, resulting in varying shades.

Colour sorting was tried to obtain uniform colours by processing a same-colour batch. Production waste of different shades of the same colour was processed together. It was noticed that since some colorants are thermochromic, rising temperatures can change or even remove the colour (colour change or disappearance). Colour sorting does not lead to a uniform colour after extrusion.

The only solution is to dope-dye the polyester with carbon black pigment into black to obtain a uniform black. This dyeing process was used to produce the black recycled yarns that were 80% recycled content of pet bottles and 20 % recycled content of production waste.

4 Chemical recycling

Technology

The polymer is dissolved and then depolymerised via a reaction into the initial building blocks of the material. These monomers are purified of impurities and then used in a polymerisation process to synthetise again a polymer. The different steps of this process are illustrated in Figure 5 General process scheme for a synthetic fiber chemical recycling process. (Duhoux, et al., 2021)(Duhoux, et al., 2021)

Figure 11 General process scheme for a synthetic fiber chemical recycling process (Duhoux, et al., 2021)

In general, three different major reaction pathways can be identified: glycolysis, methanolysis and hydrolysis. The glycolysis pathway is the most developed as several technologies are reaching, or have reached, commercial demonstration, while hydrolysis is currently the least developed one: (Hann & Connock, 2020)

- Glycolysis: several demonstration plants constructed and even running
- Methanolysis: first plant in operation, more underway
- Hydrolysis: pilot stage, moving onto industrial/demo stage

There are some additional pathways, example via enzymes, but they are currently not well developed. The consortium has chosen the glycolysis pathways because it is the most developed technology.

Chemical recycling of polyester end-of-life garments

The chemical recycling, by catalytic glycolysis, of a polyester waste sample and the necessary purification steps to obtain a pure and colourless BHET (bis(2-hydroxyethyl) terephthalate) product useful for the manufacture of PET pellets was examined in a trial recycling process.

The PET waste sample consists of a conditioned polyester waste from work clothes (Figure 1).

Figure 12 Conditioned polyester waste from work clothes.

Catalytic glycolysis reaction

The chemical recycling process of the material is carried out by solvolysis reaction, using a glycol as an agent (called glycolysis) which consists of the chemical depolymerization of the polymer (PET) to obtain the minimum unit that composes it, called monomer (BHET), which after purification allows it synthesis.

The reaction process, carried out in a nitrogen atmosphere, consists of the degradation of the PET by the action of a solvent (ethylene glycol, EG) in presence of a catalyst and a near boiling point temperature of the EG. The reaction product is the monomer BHET (bis(2-hydroxyethyl) terephthalate) with a low proportion of oligomers such as dimmers.

Reaction 1. PET depolymerization reaction:

$$\mathrm{PET} + \mathrm{EG} \xrightarrow[\text{catalyst and heat}]{} \mathrm{BHET} + \mathrm{Oligomers} + \mathrm{EG}$$

Figure 13 Reaction 1 Pet depolymerization reaction.

The reaction has been conducted on a laboratory scale in specific conditions. The laboratory reaction equipment consists of a 1 L reactor, a heating system, a PTFE thermocouple, a cooling tower and shoveltype stirrer. The following equations (1-3) are used to obtain the yields of the glycolysis reaction, the results are presented in Table 3.

$$\% \text{ Oligomers} = \frac{\text{mass oligomers obtained}}{\text{mass PET waste fed}} \cdot 100$$

Equation 2. Production to BHET:

$$\% \text{ Production to BHET} = \frac{\text{mass BHET obtained}}{\text{mass PET waste fed}} \cdot 100$$

Equation 3. BHET yield:

$$\% \text{ BHET yield } \frac{\frac{\text{mass BHET}}{\text{MW BHET}}}{\frac{\text{massPET}}{\text{MW PET}}} \cdot 100$$

Figure 14 Equation - Oligomers obtained.

Oligomers (%)	Production to BHET (%)	Yield to BHET (%)
0.6	100.9	76.3

Table 3 Results of the glycolysis reaction obtained at lab scale.

The resulting products of the post-treatment steps are shown next, in Figure 8 to Figure 11. Figure 8 shows the retained unreacted material. Figure 3 shows the oligomers obtained. Figure 10 shows the liquid obtained from this filtration step, and Figure 11 shows the coloured BHET after the crystallization.

Figure 15 Retained unreacted material after glycolysis

Figure 16 Oligomers retained

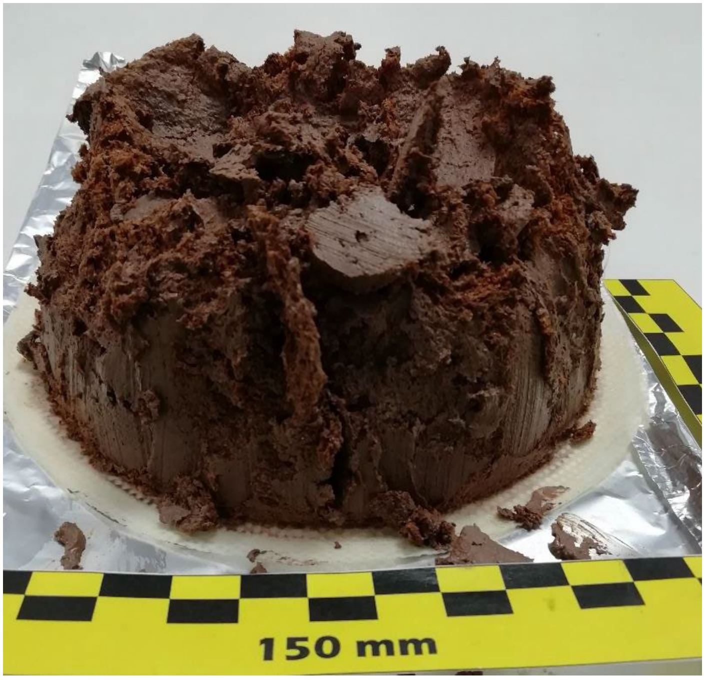


Figure 17 Liquid obtained after filtration of oligomers.

Figure 18 Coloured BHET obtained.

FTIR and DSC analyses have been carried out for the coloured BHET and the oligomers obtained.

The results of BHET analysis are shown in Figure 12 and Figure 13. The results of oligomers analysis are shown in Figure 14 and Figure 15.

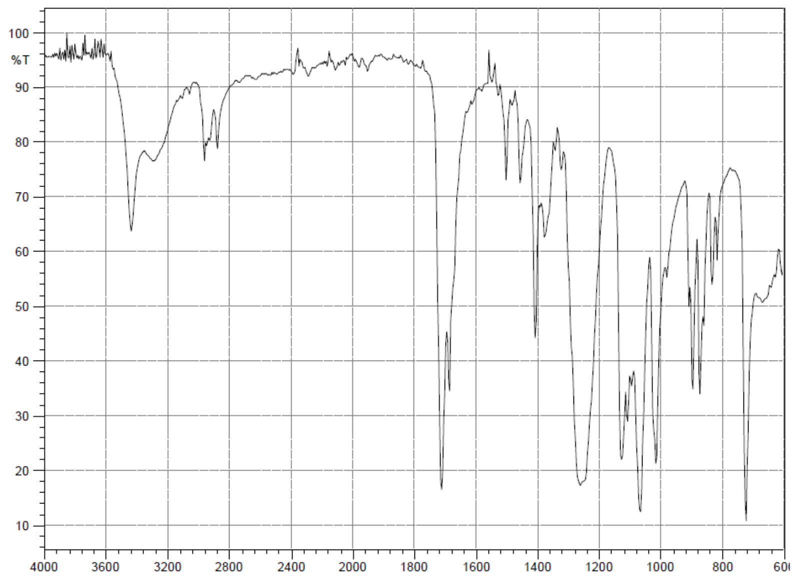


Figure 19 FTIR spectrum of the obtained coloured BHET.

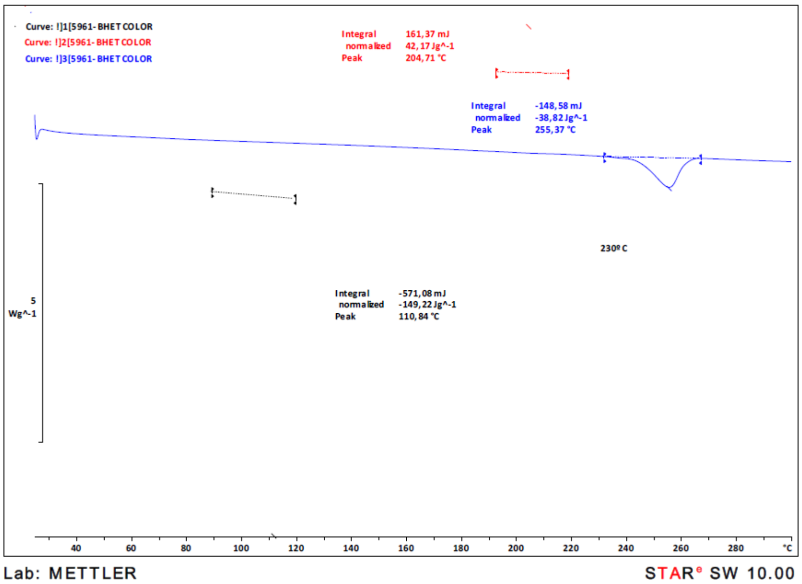


Figure 20 DSC thermogram of the obtained coloured BHET.

Regarding the DSC spectra, the peak obtained at 110 °C belongs to BHET monomer.

Therefore, the final solid recovered is BHET. Regarding the FTIR spectra, the results indicate that the spectral bands found are consistent with those of the BHET.

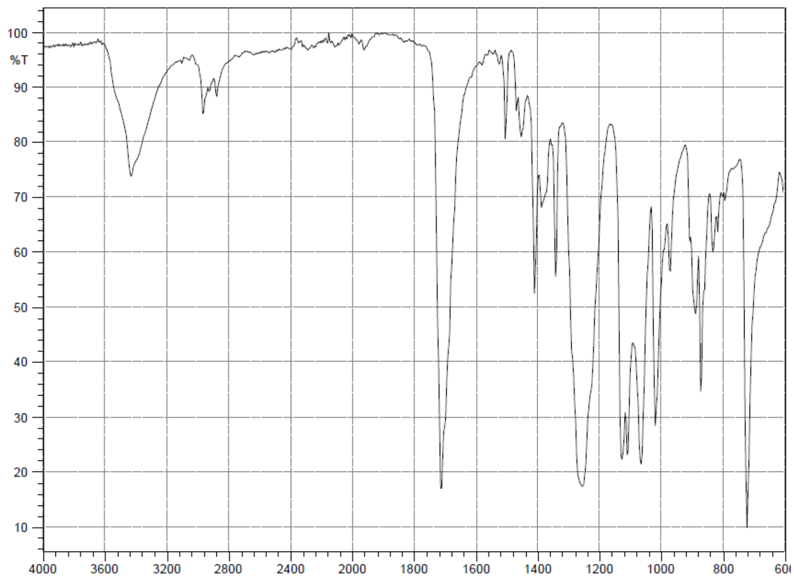


Figure 21 FTIR spectrum of the oligomers.

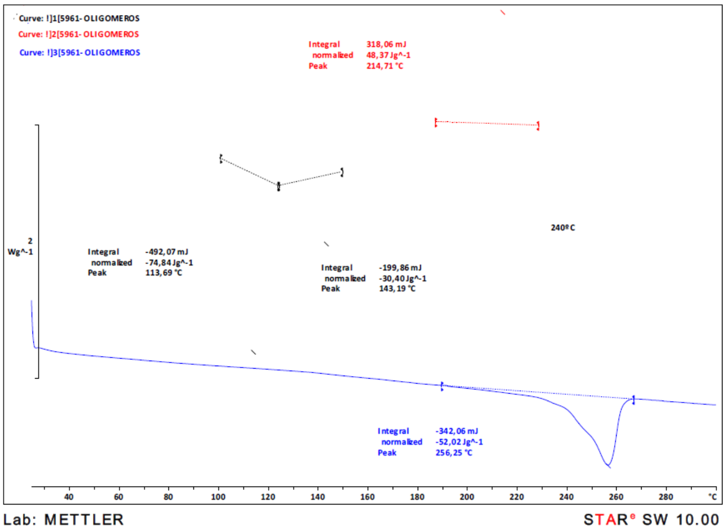


Figure 22 DSC thermogram of the oligomers.

Regarding the DSC spectra, the peak obtained around 110 °C belongs to BHET monomer. Peaks between 110 °C (BHET peak) and 250 °C (PET peak) are due to oligomers. Therefore, oligomer filtration retains mainly dimmers (peak at 143 °C). Regarding the FTIR spectra, the results indicate that the spectral bands found are consistent with those of the BHET and dimmers.

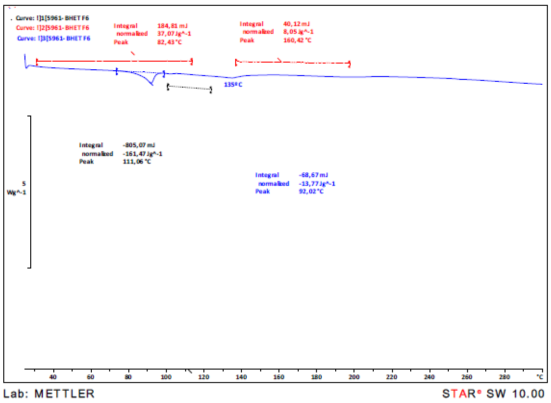
BHET Colour removal

After the reaction process and the post-treatment stages, the obtained product (BHET) was purified with the aim to get a colourless BHET. Colour removal operation has been performed using two steps because of the complexity of the material. Figure 16 shows the first step obtained BHET. Figure 17 shows the second step obtained BHET.

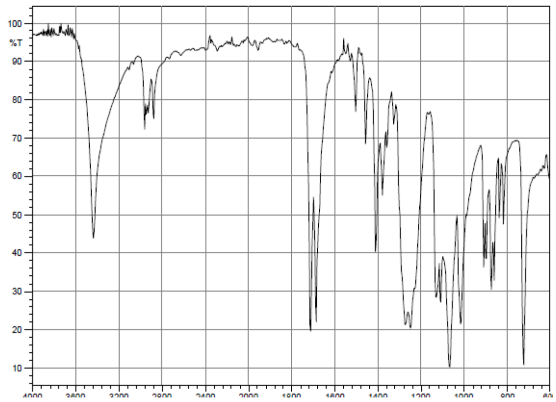
Figure 23 BHET obtained by colour removal (first step).

Figure 24 BHET obtained by colour removal (second step).

Colourless BHET has been obtained, this BHET has been characterized by FTIR and DSC analysis. The results are shown in Figure 18.



DSC thermogram of the colourless BHET



FTIR spectrum of the colourless BHET

Figure 25 DSC and FTIR results of the colourless BHET obtained.

Regarding the DSC spectra, the peaks obtained around 110 °C belongs to BHET monomer. Therefore, the final solid recovered is BHET. Regarding the FTIR spectra, the results indicate that the spectral bands found are consistent with those of the BHET.

Polymerisation

The decoloured BHET has been polymerised. FTIR spectrum of synthesized PET pellets was carried out Figure 19. Ash content of the PET pellets was determined by TGA. Two characteristic bands are observed: one at 1100-1000 cm-1 wave number and other at 800-700 cm-1 wave number, belong to silicon oxide (SiO_2), and antimony oxides (Sb_2O_3), respectively.

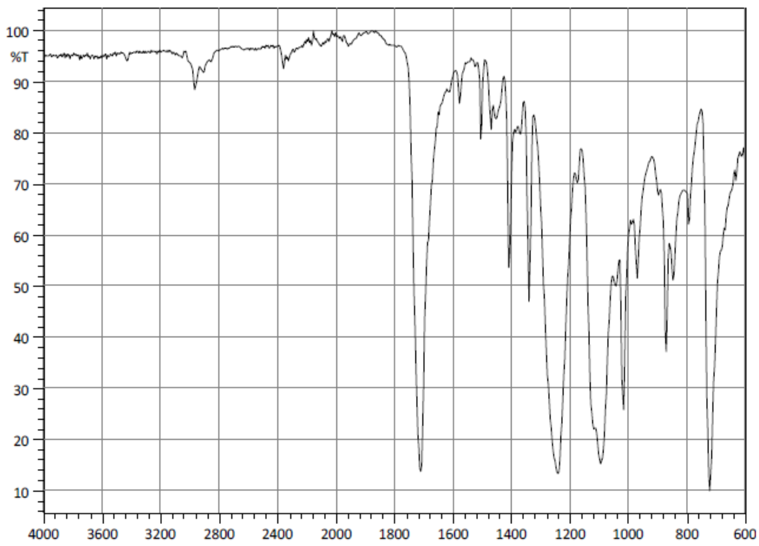


Figure 26 FTIR spectrum of the chemically recycled PET pellets produced.

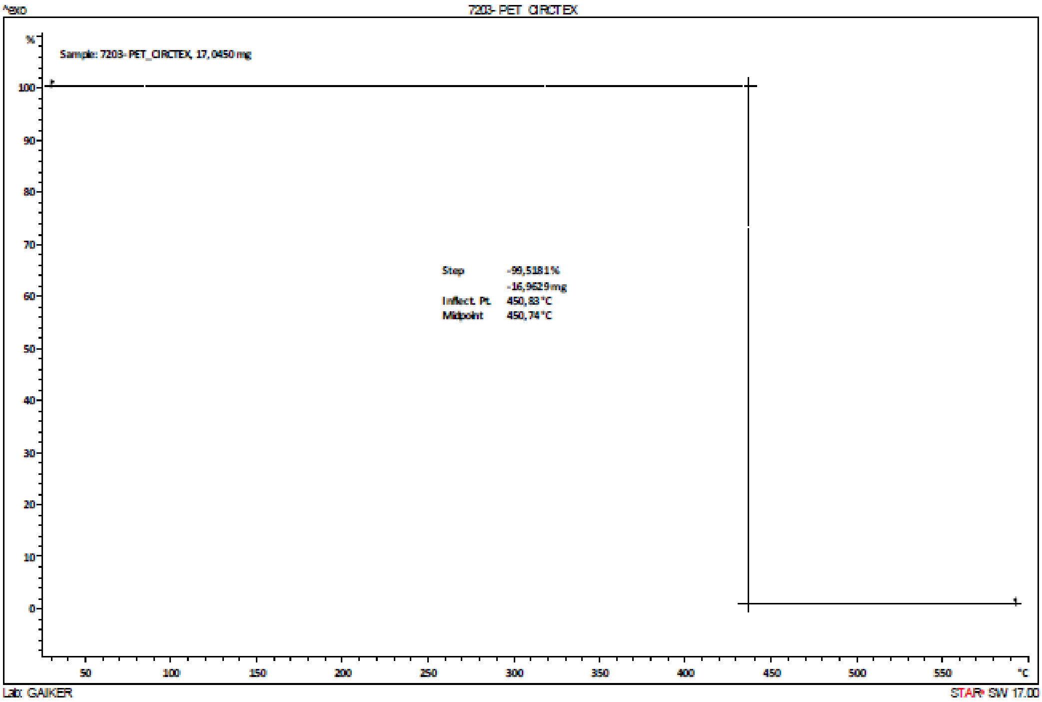


Figure 27 TGA spectrum of the chemically recycled PET pellets produced.

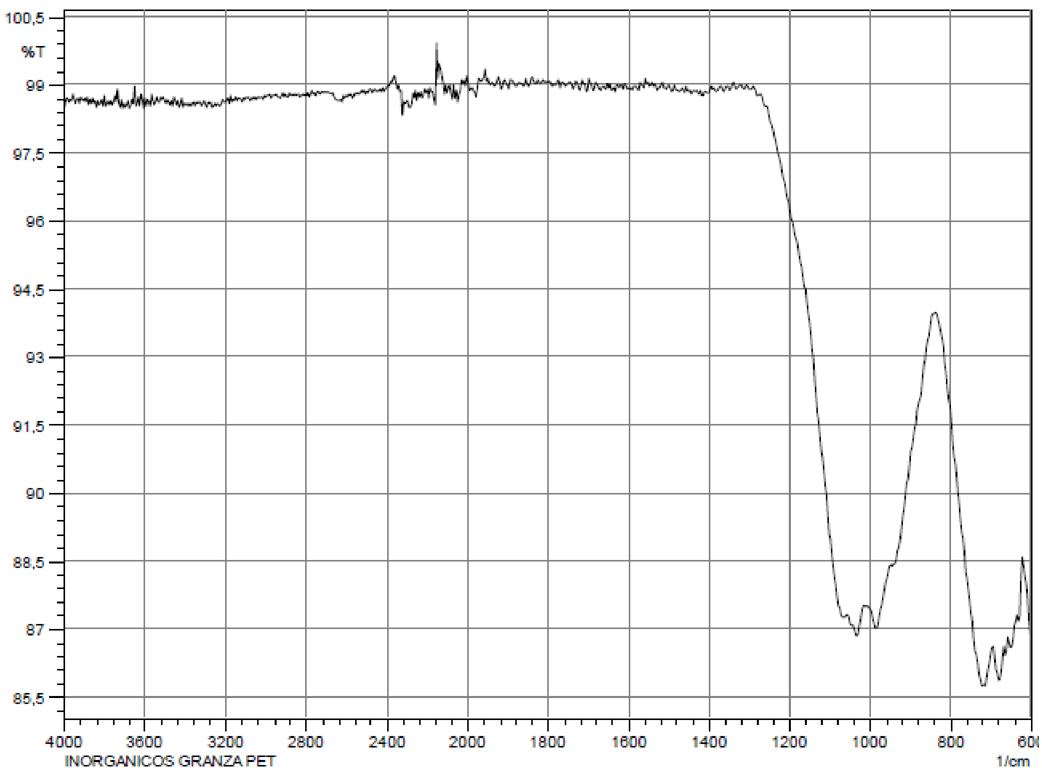


Figure 28 FTIR spectrum of the rPET ashes.

Inorganic content of the chemically recycled PET pellets. This is further analysed (see Chemical testing of the chemical recycled material)

Sample	Inorganic content (%)
rPET pellets	0.482

Table 4 Inorganic content of the chemically recycled PET pellets.

The intrinsic viscosity (IV) and colour coordinates of PET pellets (CIELAB space coordinates) were also determined. The results can be found in Table 5 and Table 6. This is in line with the results of a virgin polyester.

Sample	IV (dL/g)
rPET pellets	0.62

Table 5 Viscosity of the chemically recycled PET pellets.

Sample	CIELAB space coordinates (ISO 11664-4:2020)		
	L*	a*	b*
rPET pellets	59.78	0.52	0.95

Table 6 Colour of the chemically recycled PET pellets.

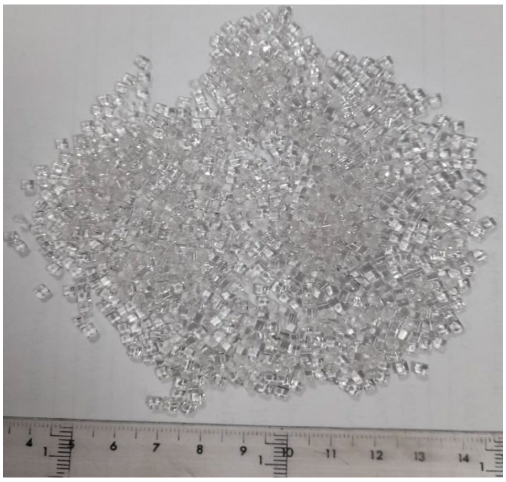


Figure 29 Chemically recycled PET pellets.

Yarn production

The pellets, with the addition of TiO_2 obtained a dull effect, have been extruded in the Antex Spinning plant.

The polymer has been successfully spun into a yarn with a dpf of 3.47, without any process incidents, and presenting mechanical parameters similar to those of yarn produced directly from virgin PET.

This POY yarn has been textured (Figure 20) and the resulting yarn has been delivered to IBQ to develop the final demonstrator of the project.

Figure 30 Textured yarn obtained.

Using the yarn supplied by ANTEX, obtained from the chemical recycling of garments, IBQ has manufactured four fabrics:

- Two 2-layer fabric (black and orange high-visibility)
- Two 3-layer fabric (black and orange high-visibility)

5 Weaving chemically recycled yarn

The weaving efficiency was not perfect. The usual efficiency on a fabric of polyester in continuous filament like this is around 98%, and in 600 m the loom has maybe 10 or 20 stops.

Since we noticed a slightly lower tensile strength of the yarn the production speed was lowered to decrease the tension of the warp on the loom to avoid breaks and to minimize tension peaks in weft. In addition, an extra sizing process was applied on the warp to compact the yarn and minimize the friction on the weaving. In this way the efficiencies were improved.

The weaving of the recycled fabric was monitored on the length of 600 m that was produced with the recycled yarn from used garments, and the following figures were obtained:

- Efficiency of the loom: 95,9%
- Total stops in warp: 47
- Total stops in weft: 12

6 Quality assessment of recycled polyester fabric

General quality assessment

We compared the fabrics made from thermo-mechanically recycled polyester yarn with the fabrics made from chemically recycled yarns. In this chapter we only assess the original state of the fabric, the influence of the maintenance processes is described in the report "Washing of recycled polyester fabrics"

- T2309444 3L JACKET BLACK ALTERNATIVE MEMBRANE + RY PES TUL (chemical recycled)
- T2309445 3L JACKET ORANGE HV CO-PES MEMBRANE + RY PES TUL (chemical recycled)
- T2309778 3L JACKET ORANGE HV ALTERNATIVE MEMBRANE + RY PES TUL (thermo-mechanical recycled)

The results of the quality tests of the different fabrics were compared with the requirements in the following standards:

- ETSA requirements for workwear garments (referred to further as ETSA requirements)
- EN 343: 2019 Protective clothing - Protection against rain

<u>The results are assessed per test method.</u>

ISO 3801-5 (1977) Determination of the mass per unit area

This was determined to check if we would test a similar fabric since different membranes were used.

T 230944 with an average 264 g/m² and T2309778 with an average of 261 g/m² are similar while T2309445 is slightly off with 256 g/m².

EN ISO 1421-1 (2016) Determination of the breaking strength and elongation

For all 3 fabrics the results are high. The ETSA requirements has for class 3 ••• low limitations in use expected a minimum of 400 n. With the results shown in Table 7.

EN ISO 1421-1 (2016)	**Determination of the breaking strength and elongation**	
	Length direction	**Width direction**
T2309444	1213 N	973 N
T2309445	1221 N	970 N
T2309778	1404 N	1302 N

Table 7 Determination of the breaking strength and elongation results.

We can see that although the results are very good the results of T2309778 are considerably better, especially in the width (warp) direction.

EN ISO 4674-1A (2016) Determination of tearing resistance - method with three legs

The ETSA requirements did not take up the tearing resistance requirement in contrast to the PPE standard EN 343 protection against rain. The minimum requirement is 20 N. All samples scored a lot higher than this minimum requirement. We can consider all samples to score very good on this parameter. The results can be found in Table 8.

EN ISO 4674-1A (2016)	**Determination of tearing resistance - method with three legs**	
	Lengt direction	**Width direction**
T2309444	Warp 130 /	Weft N 100 N
T2309445	94 N	100 N
T2309778	92 N	100 N

Table 8 Determination of tearing resistance - method with three legs results

ISO 12947-2 (2016) Determination of the abrasion resistance – Martindale

There are no minimum requirements set in the ETSA requirements nor in PPE standards. The assessment is done on the experience of CENTEXBEL. The result of T2309444 and T2309445 differ from T230778 which with 90000 cycles is much better. The results of 50000 and 60000 cycles are considered average and acceptable. However, the result of 90000 cycles probably results in less customers complaints and a longer lifetime.

ISO 12947-2 (2016)	**Determination of the abrasion resistance - Martindale**
T2309444	50 000 cycles
T2309445	60 000 cycles
T2309778	90 000 cycles

Table 9 Determination of the abrasion resistance - Martindale

ISO 105-X12 (2016) Determination of the colour fastness to rubbing

All results 4-5 for both wet and dry rubbing are excellent and meet all minimum requirements that are set by standards (5 is the best result possible). This means that the colour bleeds out very little due to rubbing but will probably never cause an issue.

ISO 105-X11 (1994) Determination of the colour fastness to ironing

The results of all fabrics are good (4) to very good (4-5 and 5). The only sample that has a lower result is T2309444 for staining in wet condition, with 3-4. This means the colour migrates if it is ironed when the sample is still wet. This can be avoided. The results can be found in Table 9, Table 10 and Table 11.

Temperature	Staining	Change in colour	
110 °C		Immediately after testing	After conditioning
Dry	5	4-5	4-5
Humidified	4	4-5	4-5
Wet	3-4	4	4-5

Table 10 Determination of the colour fastness to ironing results of T2309444

Temperature	Staining	Change in colour	
110 °C		Immediately after testing	After conditioning
Dry	5	4-5	5
Humidified	5	4-5	5
Wet	4-5	4-5	5

Table 11 Determination of the colour fastness to ironing results of T2309445

Temperature	Staining	Change in colour	
110 °C		Immediately after testing	After conditioning
Dry	5	4-5	5
Humidified	5	4-5	5
Wet	4-5	4-5	5

Table 12 Determination of the colour fastness to ironing results of T2309778.

ISO 12945-2 (2020) Determination of the resistance to pilling, matting and fuzzing – Martindale

All samples scored the best result (5) on pilling, matting and fuzzing. This means none of the effects occurs on the fabrics.

EN ISO 11092 (2014) Determination of the water vapour resistance

The result informs us on the ability to transmit water vapour coming from the human body and to transfer it through the fabric to keep the wearer dry. The result depends mainly on the membrane used but also on the construction of the fabric, build-up of the different layers, the method of binding the membrane to the fabric and the used fibres. Switching the membrane will probably have the biggest effect in this case since the other parameters are similar. In addition the amount of glue that is used plays a factor. Too much glue can act as a barrier.

- For T2309444 result is 43.72 m^2. Pa/W. This does not even comply with the lowest class of EN 343 protection against the rain. It is considered a bad result.
- For T2309445 result is 25.08 m^2.Pa/W. This complies with class 2 in EN 343 protection against the rains and this is considered an average result.

- For T2309778 result is 11.46 m^2.Pa/W. This corresponds to class 1 of EN 343 protection against the rain and this is considered a very good result.

Changing the membrane and amount of glue could improve the result. The latter can be concluded coming from the result of the adhesion strength tests.

ISO 2411 (2017) Determination of the adhesion strength

The results of T2309778 and T2309445 are lower than the ones of T2309444. This is the opposite for the RET. This might indicate that the conclusion on the RET result referring to the glue is correct.

- For T2309444 the result is length direction 27.4 N/ 50mm and width direction 23.3 N/ 50mm
- For T2309445 the result is length direction 10.2 N/ 50mm and width direction 8.09 N/ 50mm
- For T2309778 the result is length direction 10.1 N/ 50mm and width direction 9.50 N/ 50mm

OEKO-TEX® (2023) Determination of the colour fastness to water

All colourfastness to water were very good with a result of 4-5 for all samples, this means that the colour bleeds under the influence of water to other items in only a limited way. This will probably not lead to customers complaints. The ETSA requirements ask a minimum of 4-5 for class 3.

ISO 105-E04 (2013) Determination of the colour fastness to perspiration

The colour of all samples doesn’t change under the influence of sweat, both alkaline and acid sweat as a human can produce both alkaline or acid sweat. It bleeds out in a limited way depending on the material that is in contact with the sample. For the results of 3-4 this is a low result. All results can be found in Table 12, Table 13 and Table 14.

[3], Alkaline solution

Numerical rating	
Change in colour	5
Staining on diacetate	3-4
Staining on cotton	4-5
Staining on polyamide	4
Staining on polyester	4-5
Staining on acrylic	4-5
Staining on wool	4

[3], Acid solution

Numerical rating	
Change in colour	5
Staining on diacetate	4
Staining on cotton	5
Staining on polyamide	4
Staining on polyester	4-5
Staining on acrylic	5
Staining on wool	4-5

Table 13 Determination of the colour fastness to perspiration result of T2309444

[3], Alkaline solution

Numerical rating	
Change in colour	5
Staining on diacetate	4
Staining on cotton	4-5
Staining on polyamide	3-4
Staining on polyester	4-5
Staining on acrylic	4-5
Staining on wool	4

[3], Acid solution

Numerical rating	
Change in colour	5
Staining on diacetate	4
Staining on cotton	4-5
Staining on polyamide	3-4
Staining on polyester	4
Staining on acrylic	4-5
Staining on wool	4

Table 14 Determination of the colour fastness to perspiration result of T2309445.

Multifibre Type DW , Alkaline solution

Numerical rating	
Change in colour	5
Staining on diacetate	4-5
Staining on cotton	5
Staining on polyamide	4-5
Staining on polyester	5
Staining on acrylic	5
Staining on wool	4-5

Multifibre Type DW , Acid solution

Numerical rating	
Change in colour	5
Staining on diacetate	4-5
Staining on cotton	5
Staining on polyamide	5
Staining on polyester	5
Staining on acrylic	5
Staining on wool	4-5

Table 15 Determination of the colour fastness to perspiration result of T2309778.

ISO 105-N01 (1993) Determination of the colour fastness to bleaching – Hypochlorite.

All colourfastness to water were very good with a result of 4-5 for all samples. implying that the colour will bleed when hypochlorite is used under the influence of water onto other items in a limited way. This will probably not lead to customers complaints. The ETSA requirements ask a minimum of 4 for class 3. All samples scored better.

OEKO-TEX® (2023) Determination of the colour fastness to sweat and saliva.

No colour is released when the samples were brought info contact with an artificial sweat and saliva solution. This is a requirement for the OEKO-TEX® class I – Babies.

In general, all samples can be considered fit for use, except for sample T2309444 that fails to EN ISO 11092 (2014) Determination of water vapour resistance.

Personal Protection Equipment

The objective was to produce a garment having the same features as the original garment. The garment should protect against rain and have a high visibility aspect. The standards to assess the compliance to the PPE regulation and corresponding CE-marking are:

- EN ISO 20471: 2013 (Cor. 2013-06)/A1:2016 - High visibility clothing - Test methods and requirements
- EN 343: 2019 Protective clothing - Protection against rain

High visibility clothing (EN IS 20471)

The garment is compliant with this standard, see the below results and the pass and fail, except for the RCT. The RCT was not assessed and should be measured additionally in order to show full compliance.

- Dimensional stability max 3 %: pass
- Breaking strength & elongation min 100 N: pass
- Tear 20N: pass
- Chromaticity and luminance before and after Xenon: pass
- Chromaticity and luminance after washing: pass
- Colourfastness rubbing min 4: pass
- Colourfastness perspiration: change in colour min 4: pass; staining min 4: pass.
- Colourfastness laundering: change in colour min 4-5: pass; staining min 4: pass

Protection against the rain (EN 343)

The garment is not compliant with the standard because of several fails which can be seen below. An additional layer to protect the membrane might already improve the results. However then the seam tapes will not be compliant with the standard. Further development is needed on the membrane or seam tapes in order to become compliant.

- Dimensional stability max 3 %: pass
- Breaking strength & elongation min 450 N: pass
- Tear min 20 N: pass
- ISO 811 original: class 4
- ISO 811 after influence with oil: fail
- ISO 811 after influence with gasoil: class 4
- ISO 811 after crumple flex: fail
- ISO 811 after sand paper: fail
- ISO 811 after washing: class 4
- ISO 811 on seams original: fail
- ISO 811 on cross seams after washing: fail
- Seam strength: min 200 N: pass
- RET 16.83 m2.Pa/W: class 3

Conclusion on the quality

It is possible to produce a 100% closed-loop workwear garment (same textile to textile product) from recycled content that has more or less the same life expectation. It is even possible to obtain a CE-Marking for High-visibility but not yet for protection against rain. With further adaptations to the design, it might be possible to obtain this certification as well in the near future.

7 Chemical testing of the chemical recycled material

Contamination due to production.

To assess what chemicals and materials might be left behind after the chemical recycling process including the purifying step, additional tests were performed followed by an assessment. First the new material was tested even though it was OEKO-TEX® compliant. This was done to assess if there might be an issue with future changes in the REACH legislation. The fabric that was tested was the thermo-mechanical recycled fabric.

Results

No carcinogenic azo dyes present as expected.

DHS has shown an overall low emission. However, non-negligible peak of benzyl benzoate, which can be irritating. Benzyl benzoate:

Consumer Use

This substance is used in the following products: washing & cleaning products, air care products, polishes and waxes, perfumes and fragrances, cosmetics and personal care products and biocides (e.g. disinfectants, pest control products).

Other release to the environment of this substance is likely to occur from: indoor use as processing aid and outdoor use as processing aid.

These substances are not restricted in REACH annex XVII or in the SVHC-list. So it can be used in REACH (legal) without any problem. Concerning benzyl benzoate: it has been put on the agenda to be evaluated by Oekotex100, but as of today, it is not on this list. This substance can be used in the dyeing process. Benzyl benzoate makes the fibre swell and undraw so that the fibre will be dyed easily. So this can still be found in textiles.

In one of the samples additional some paraffins were present, which may precipitate possible transfer to skin with benzylbenzoate that could cause skin irritation. However, this is very individual depended.

Further DHS has shown amounts of dipropyleneglycol as a test result

Consumer Uses

This substance is used in the following products: anti-freeze products, lubricants and greases, air care products and biocides (e.g. disinfectants, pest control products).

Other release to the environment of this substance is likely to occur from: outdoor use, indoor use (e.g. machine wash liquids/detergents, automotive care products, paints and coating or adhesives, fragrances and air fresheners), indoor use in close systems with minimal release (e.g. cooling liquids in refrigerators, oil-based electric heaters) and outdoor use in close systems with minimal release (e.g. hydraulic liquids in automotive suspension, lubricants in motor oil and break fluids).

Some paraffins were also found together with a small amount of benzylbenzoate.

Conclusion

No traces of substances that would not be ok with REACH were found in DHS. This was expected since the material is Oekotex compliant. Of course, REACH is more than a DHS test. Only REACH VOCs can be excluded there. Hard to predict if there will be a health problem, it depends on exposure and person. Benzyl benzoate in combination with paraffin (in one sample) could possibly.

Analysis of the possible legacy chemicals due to the use phase:

Used clothing from different activities was subjected to a full REACH analysis to see if there were things present that were not ok with legislation. The use fabric in these clothing was the thermos-mechanical fabric also analysed in 7.2.

The following tests were carried out:

- Centexbel LCMS screening (Reach SVHC)
- Centexbel Determination of the elemental composition (screening)
- Centexbel Determination of the limited FR products (REACH Annex XVII)
- DIN 54231 (2005) Determination of limited allergenic dyes Annex XVII CMR
- Centexbel Determination of the composition using XRF-screening
- Centexbel Determination of the emission profile by thermal extraction.
- Centexbel Determination of the emission profile by using dynamic headspace
- EN 16711-2 (2015) Determination of heavy metals Annex XVII CMR
- EN 14362-1 (2017) Aromatic amines derived from azo colorants in textiles
- OEKO-TEX® (2022) Determination of per and poly fluorinated compounds (PFCs)
- ISO 14184-1 (2011) Determination of formaldehyde
- Centexbel Determination of low phthalates concentration – Content for Reach SVHC
- Forbidden Azo-Dyes

Results

in addition to what was found from the production phase PFAS substances were found (not tested on the production samples).

- 0.059 mg/kg PFHxA found: C6 chem. (0.025 mg/kg)
- 0.19 6:2 FTOH: C6 chem. (1 mg/kg)
- 0.13 10:2 FTOH: C6 chemistry (1 mg/kg)

These substances are currently still permitted but will soon be restricted. Protective clothing will get another exception. Limit value is 0.025 mg/kg and 1 mg/kg. So will be above it for first, but only applies to products that will be produced after a certain date.

The PFAS chemicals originate from the water repellent finish that is applied. The finish was changed for the production of the chemically recycled fabric as an alternative PFAS free finish was used. However, it industrial laundering company might continue to apply PFAS finishes as long as they are not prohibited.

Additional testing of phthalates as thermal extraction indicated no presence of DBP, DIBP, BBP and DEHP.

In Regulation (EU) 2018/1513, which introduces a restriction on certain carcinogenic, mutagenic or toxic for reproduction (CMR) substances in apparel, footwear and other textile consumer products there are 4 extractable metals listed. Cd, Cr, As and Pb. In one stain Pb was found above the detection limit: 0.49 mg/kg Pb. Limit for legislation is 1 mg/kg so in principle there is no issue. However, this must come from the use faze.

Another solvent from the CMR list was found. NMP was found in small quantities, but this will never lead to more than 3000 mg/kg, which is the limit in CMR.

Conclusion

There are no problems concerning the legislation. C6 chemistry are present in certain samples, but this is allowed for now. There is a restriction coming up, with limits as mentioned in summary. But this will be only applicable after a certain date.

DHS: benzyl benzoate (known irritant) is problematic in combination with substances that accelerate skin transfer: palmitate or silicones. Obviously, there must be prolonged skin contact and the reaction may differ from one person to another. Benzyl benzoate is known to colour PES, which is why this was encountered on many samples. A couple of years back, OEKO-TEX® contemplated adding it to its list of restricted items, but until now, it has refrained from doing so.

Analysis of samples of BHET and repolymerised polyester

A small quantity of used, unwashed clothes was recycled to determine what would remain unaffected by the purification process.

During this analysis, Benzylbenzoate, which is associated with dyeing residue, was discovered. In addition, small traces of fats (such as esters or paraffins) and plasticizers (contaminants) were also detected.

Furthermore, remnants of laundry procedures, including fragrances, emulsifiers like softeners, and additives like octocrylene, were present.

However, all these additives were effectively eliminated through depolymerization and repolymerization, as neither significant amounts of benzyl benzoate nor smaller residues were found. The only remaining substance was squalene, a type of skin fat. This issue could easily be resolved by washing all waste prior to recycling it.

OEKO-TEX® standard 100 annex 6

The chemical recycled material was tested according to OEKO-TEX® standard 100 annex 6. Annex 6 excludes chemicals listed in up in the detox list of Greenpeace. Being compliant with this standard means that no chemicals are present that harm the human health or that Greenpeace wants to avoid.

The granulates and fabrics were tested an no chemicals were found that would prohibit the material being conform with OEKO-TEX® Standard 100. Therefore, it is conform to the requirements of OEKO-TEX® standard 100 annex 6.

8 Economic feasibility assessment

The circular business model will only succeed if 2 conditions are met:

- Sufficient volume of suitable waste.
- Willingness to pay a premium price.

Sufficient volume of suitable waste.

There is a sufficient volume of collected suitable waste that can feed the business model, in this case a recycling process. The same goes for the other R-strategies including refurbish, remanufacture, and even reuse. If there are not enough products of a good enough quality the second-hand shop might lose the interest of the audience. In interview with a second-hand organisation in Belgium, 'the kringloopwinkel', it was mentioned that they need clothes of premium brands to attract customers; If they won't find these brands any more in the second hand store, they will sell also clothes from lesser known brands.

Regarding to recycling:

Currently, there are few recycling technologies available that can effectively handle large quantities of waste in a cost-effective manner. This is partly due to technical challenges that require further research and development. Additionally, for a company to establish a large-scale chemical recycling process, there needs to be a consistent supply of materials and a market demand for these materials. To achieve the necessary volumes, certain measures must be implemented. Initiatives such as the separate collection of textile waste and the introduction of a digital product passport through legislative actions will contribute to this goal. However, it is equally important to properly classify the waste to avoid mixing valuable waste streams if efficient sorting is not possible. Furthermore, there should be incentives for consumers to participate in these efforts. While legislation can help, it may not be sufficient on its own. This can be observed in other industries, such as the deposit schemes for PET bottles in countries like Germany. Despite the separate collection of PET bottles, consumers still make mistakes, whether consciously or unconsciously. These deposit schemes serve to motivate consumers, although some may perceive them as burdensome.

Willingness to pay a premium price.

The consumer shows little to no interest in paying extra for recycled materials. This is likely true for other approaches as well, given the abundance of inexpensive new products. Even when it comes to reusing items, the consumer is willing to pay less compared to buying new. Therefore, it is crucial to keep all expenses associated with collecting, sanitizing, and presenting second-hand products as low as possible, in order to avoid making them more expensive than new products. This is challenging as long as a new t-shirt costs only €5 at certain retailers.

Regarding recycling, the premium includes different elements to support the business model of a circular polyester garment:

- collecting sorting and disassembly.
- digital product passport
- various logistics
- recycled content (if needed).

Surcharge for collecting, sorting and disassembly.

The surcharge covers the temporary storage of waste until there is enough to make transportation economically feasible. Additionally, sorting may be necessary to separate different waste streams that require different treatment methods. Apart from the costs associated with collection and sorting, there will be an additional cost for disassembly, which varies depending on the material content and disassembly features. For example, a 100% cotton towel can be easily identified and recycled as a whole if it is assembled with cotton labels and no other materials. However, a PPE garment that contains various materials will require disassembly. Products that are **not** made entirely of a single material or a blend for which recycling technology exists need to be disassembled into recyclable mono materials or separate blends. It is rare to find mono-material products in the market, as small items like sewing yarn, buttons, zippers, and prints are often overlooked. Implementing a disassembly line incurs additional costs, which are ultimately borne by the consumer. This is because a business cannot sustain itself if it is not profitable. Creating a whole new logistical system is necessary to make this process feasible. The premium charged for this service varies greatly depending on the product and the specific use case.

Surcharge for a digital product passport

Chips, software, and hardware all come with their own expenses. Developing software requires time and resources, and there may be additional costs for licenses. Storing data on hardware also incurs expenses, as the necessary hardware is needed to access the data at various stages. These costs should be taken into consideration.

Surcharge for the various logistics

In this scenario as well, the premium is determined by whether the product requires disassembly or can be utilized in bulk or by an individual consumer. Is there a convenient and effortless method for disposal or does it require some effort. Possible logistics flows are:

- From consumer to sorting
- From consumer or sorting to disassembly
- From consumer, sorting or disassembly to recycling.

This could potentially be the premium where a substantial incentive could be incorporated. The premium payment could be divided into two components:

- The actual logistics cost, which varies depending on the product's route.
- An incentive to encourage consumers to return the goods. A significant deposit fee will motivate consumers to dispose of the products correctly.

Incentivizing the consumer has a significant advantage. It can establish a consistent local supply of materials that can compete with long-distance transportation of new materials. The premium for transporting new materials depends on global economics. Prices fluctuate, whereas prices for local transportation are generally more stable, especially when a consistent supply is established. Global disruptions in the supply chain easily lead to price increases. Additionally, potential disruptions caused by global events can potentially be avoided.

Surcharge for the end-product due to its recycled content (if needed).

It is estimated that the chemically recycled yarn might be ± 2,5 to 3 €/kg more expensive than the virgin material. It is estimated to be ± 2 to 2,5 €/kg more expensive than thermomechanical recycled yarn. However, as the production process scales up, it is anticipated that this price premium will decrease. In fact, over time, it is possible that this premium may even disappear, although the scarcity of recycled content could still influence supply and demand dynamics.

Extended producer responsibility scheme (EPR)

To ensure the success of this initiative, it is crucial to pay the highest premium for any product that cannot be recycled with a valuable outcome. However, in certain cases, a lower premium may be considered. It may also be necessary to explore other strategies to achieve the desired results.

Implementing this approach within an ERP scheme is essential, as it may otherwise be challenging to recover specific materials. The fashion industry is likely to be subject to the highest premium for logistics, as they do not benefit from the scale advantage that other sectors, such as workwear, hotel, and hospital linen, may enjoy.

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Index of figures

Index of Tables