H2020-NMBP-HUBS-2019

FlexFunction2Sustain

Open Innovation Ecosystem for Sustainable Nano-functionalized Flexible Plastic and Paper Surfaces and Membranes

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Duration: 48 months

= Deliverable D3.1 =

Upgrades of "Circularity by design" facilities demonstrating processing of 95% Polyolefin based film (PE or PP based film)

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Executive Summary

FlexFunction2Sustain project aims at developing a network of upgraded lab-to-fab facilities, able to tackle challenges such as the replacement of non-recyclable multilayers by recyclable mono-material structures. This is part of the "Circularity by design" services to be offered through the FlexFunction2Sustain Open Innovation Test Bed (OITB). Figure 1 shows the main steps used for the design of novel flexible packaging materials that are recyclable.

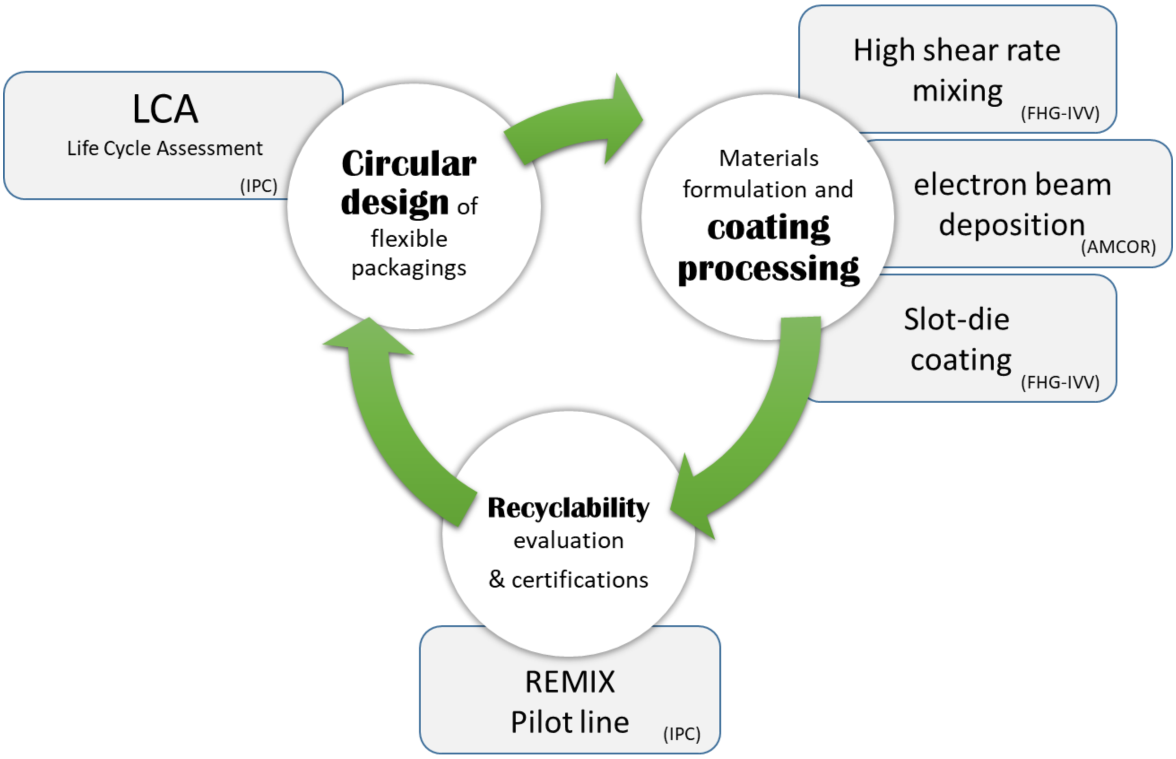

Figure 1. Schematic sketch of the main steps that belong to "Circularity by design". The lab-to-fab facilities refer to the thin film coating formulation and processing, i.e. high shear mixing, e-beam deposition and slot-die coating

This deliverable focuses on the "Circularity by design" of mono-material based laminates for flexible packaging applications, their production by means of the upgraded lines, and the evaluation of the process limits. FHG-IVV has upgraded two facilities required for Circularity by design of materials (in particular for the design of materials with a mono-material multilayer concept). The upgrades of the FHG-IVV machines have been performed within Work package 2, Task 2.2 "Atmospheric pressure processes for film extrusion, coatings and lamination". The following upgraded lines have been implied in the course of this deliverable:

- Homogeneous dispersion formulations through high shear rate mixing
- Slot-die coating unit for the coating of the nanolacquer

This deliverable presents the novel mono-material multilayer laminate structures designed for their recyclability, their production processing, and the experimental characterisation results. Three different types of novel laminate structures have been designed and produced by AMCOR and FHG-IVV within this project. The main facilities used for the production of these mono-material based laminates is demonstrated in Figure 2. These novel laminates consist of 95% polypropylene (PP), which belongs to the polyolefin (PO) group of polymers. The produced mono-material based laminates were then successfully recycled at IPC, and the PP recyclates have been extruded. The circularity of the novel 95% PO based multilayer laminates has been successfully demonstrated. The possibility of extruding PP based flat films with more than 95 mass.% PP has been verified both at IVV and IPC facilities.

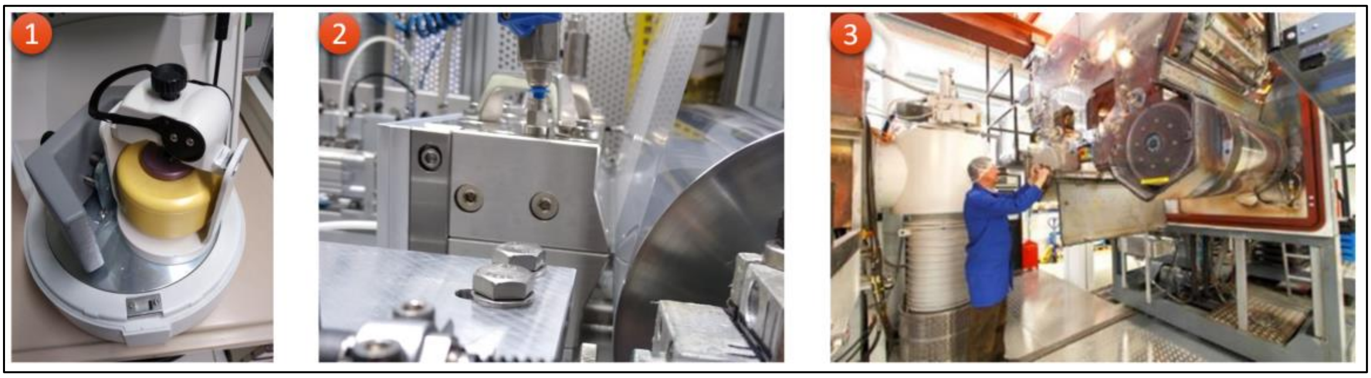

Figure 2. Upgrades of lab-to-fab facilities. (1) High energy ball mill to disperse the nanoparticles properly. (2) Slot-die coating unit for the coating of nanoparticle consisting barrier lacquers. (3) Electron-beam deposition unit for the deposition of thin SiO_x layers

IPC will assess the novel structures by means of environmental Life Cycle Assessment (LCA). The LCA results are used in an iterative approach to 1) guide the development towards more sustainable solutions, 2) quantify the potential benefits of products compared to traditional products (baseline), and 3) help future suppliers and clients to make more informed decisions. IPC uses the SimaPro software in order to gather and analyse technical and environmental inputs from all members.

In a nutshell, D3.1 includes the commissioning protocols and a proof of successful processing of 95% polyolefin based films. This deliverable addresses the set-up of facility cluster 5 for “Circularity by Design” services.

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Abbreviations

Al	Aluminum
cPP	Cast-polypropylene
BOPP	Bi-oriented polypropylene
EVOH	Ethylene vinyl alcohol
LCA	Life Cycle Assessment
LCC	Life Cycle Cost
MD	Machine Direction
MMT	Montmorillonite
PE	Polyethylene
PET	Polyethylene terephthalate
PO	Polyolefin
PP	Polypropylene
R2R	Roll to roll
SiOx	Silicon oxide
SOA	State of the art
TD	Transverse Direction

1. Introduction

Deliverable D3.1 includes the commissioning protocols for recycling of flexible packaging materials and a proof of successful processing of 95% polyolefin based films. This deliverable addresses the set-up of facility cluster 5 for "Circularity by Design" services. "Circularity by Design" refers to the structure of a flexible packaging material. A flexible packaging material includes several layers in order to fulfil the requirements of the packed product. However, it is not possible to recycle such a multi-material packaging with the established state-of-the-art industrial recycling technologies. In WP3, one of the objectives is to develop a "Circularity by Design" service, and propose different solutions towards recyclable packaging materials. One proposed solution for recyclability is to design a mono-material based laminate for flexible packaging.

This deliverable describes various novel laminate structures, which are based on a mono-material based multilayer concept. In this deliverable, the mono-material is selected to be the polypropylene (PP), which belongs to the group of polyolefin (PO). Its properties are similar to polyethylene, but it is slightly harder and more heat resistant. It is a mechanically stable material and has chemical resistance. These properties make it a very suitable candidate for mono-material based flexible packaging. We focused on the production of the novel PP based mono-material laminate structures by means of the upgraded lines, and the evaluation of the process limits. The used processes were mainly the coating of the PP-based films by the thin nanoparticle consisting barrier formulation at FHG-IVV and by the thin SiO_x layer at AMCOR via e-beam vacuum deposition process. Both of the coated PP films were then laminated to another sealing film, which was again PP based.

The following upgraded lines have been implied in the course of this deliverable:

- Homogeneous dispersion formulations by means of high shear rate mixing (FHG-IVV)
- Slot-die coating unit for the coating of the so-called "nanolacquer" barrier formulation (FHG-IVV)

The upgrades of the relevant FHG-IVV pilot lines have been performed in WP2, Task 2.2 "Atmospheric pressure processes for film extrusion, coatings and lamination".

There have been three different types of novel mono-material based laminate structures designed and produced by AMCOR and FHG-IVV. These novel laminates consist of 95% polyolefin. The mechanical recycling of these laminates has been performed successfully by IPC and the produced polypropylene recyclate granules have been extruded at FHG-IVV. Chapter 2.1 describes the novel mono-material based multilayered laminate structures designed for recyclability. The upgraded pilot lines used in the framework of this Deliverable are described in Chapter 2.2. The laminate production and the results of the experimental characterisations are discussed in Chapter 2.3 and 2.4, respectively. This deliverable demonstrated the circularity of the novel 95% PO based multilayer laminates as discussed in Chapter 2.5. The possibility of extruding PP based flat films with > 95 mass.% PP has been verified both at IVV and IPC facilities.

Furthermore, IPC assesses the novel mono-material based laminate structures by means of environmental Life Cycle Assessment (LCA). The LCA results are used in an iterative approach to 1) guide the development towards more sustainable solutions, 2) quantify the potential benefits of products compared to traditional products (baseline), and 3) help future suppliers and clients to make more informed decisions. IPC uses the SimaPro software in order to gather and analyse technical and environmental inputs from all members.

In summary, FlexFunction2Sustain has the competence and initiated the facility cluster 5 for "Circularity by Design" services. The project partners; Fhg-IVV, IPC, AMCOR, are in a position to offer their services for providing guidelines towards design for circular economy for flexible products and also for providing their relevant production and characterisation facilities thereof.

2. Results and discussion

2.1. Circularity by design

2.1.1. Motivation

According to the strategy of the European Commission, all packaging materials need to be re-useable or recyclable by 2030 (Commission, 2016). Currently the ratio of non-recyclable flexible multi-material-films is 20%, of all non-recyclable packaging (Nonclercq, 2016). To consider a plastic multi-layered flexible packaging material as recyclable, according to CEFLEX guidelines (guidelines, 2019), only 10% of its weight is allowed to be another type of material; the rest is required to be only one material class, preferably a polyolefin based material. These allowed different materials in a mono-material multilayer film are, for example, the lamination adhesive or the barrier layer, which can either be laminated on one of the films or co-extruded or deposited via physical vapour deposition. Thus, the goal in FlexFunction2Sustain project is to develop a mono-material solution that provides similar or better properties, than a comparable currently used state-of-the-art multi-material solution, and fulfils these CEFLEX guidelines.

2.1.2. State-of-the-Art and the novel mono-material based laminates

One of the typically used state-of-the-art structures is a multi-material laminate that contains an Aluminum (Al) foil to provide a sufficient oxygen and water vapor barrier (see Figure 3). This structure, as an example of several flexible packaging structures used in food packaging industry, containing several material types, is not easy to recycle, since the different materials cannot be separated easily during the recycling process. Nevertheless, these types of multi-layered structures are used, because the combination of the different materials allows tailor-made property profiles to be created with low material consumption (Dixon, 2011). For example, one of the layers provides a barrier against oxygen, the other against water vapor, the next one provides printability, and another one the mechanical stability.

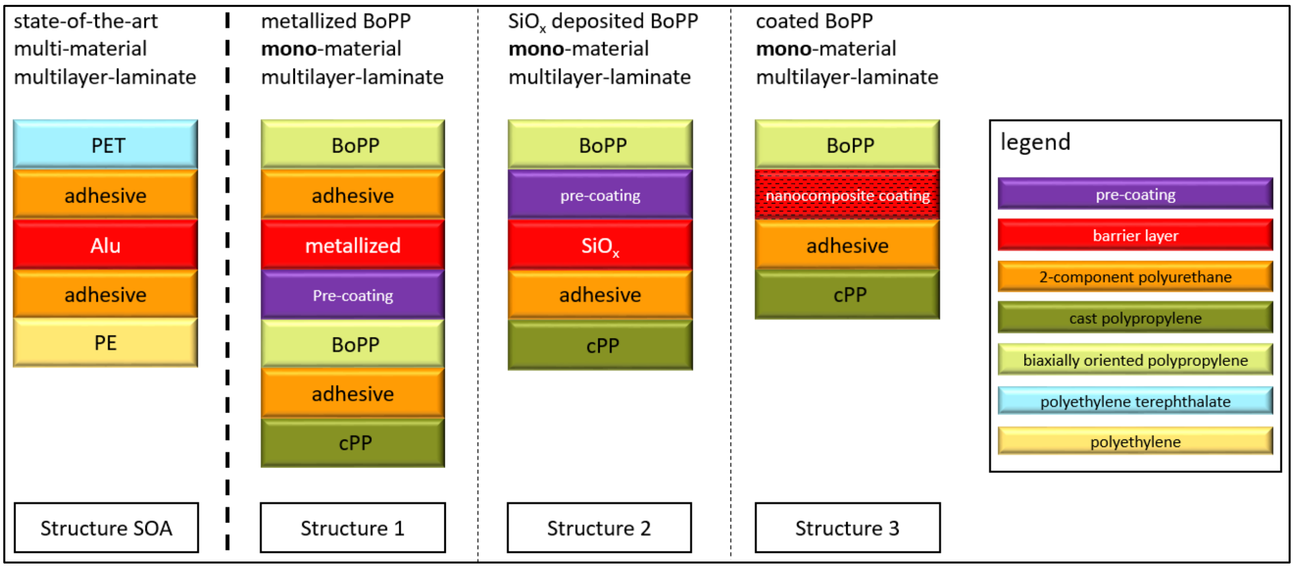

Figure 3. Structure of a State-of-the-Art multi-material laminate (the very left picture) in comparison to the novel mono-material based laminates, i.e. Structure 1, Structure 2, and Structure 3 with different thin barrier technologies of metallization, SiO_x deposition, and nanocomposite coating. This illustration is not to scale.

It is necessary to design flexible packaging films suitable for collection, sorting and recycling after use to create a circular economy for flexible packaging. Polyolefin based consumer flexible packaging makes up the largest proportion of flexible packaging waste-stream (70-80%) and the ability to sort and mechanically recycle polyolefin-based flexible packaging is already proven in industrial scale in Europe[1]. Thereby, one strategy for

[1] CEFLEX, Technical report, Designing for a circular economy, Recyclability of polyolefin-based flexible packaging

circular design of flexible packaging materials is to increase the polyolefin content in the laminate structure. This means, for instance, to replace the polyethylene terephthalate (PET) by PP.

Furthermore, the multi-material laminates consisting of Al foil (thicker than 5 µm) can theoretically be identified by eddy current separation technology, regardless of the other materials in the laminate structure. However, Al foil is not compatible with a plastic mechanical recycling process, although the sorted Al foil consisting structures can be identified and removed in the sorting process. Such sorted structures can be recycled via a pyrolysis process and the plastic proportion cannot be recycled due to the incineration.

Based on these, mono-material packaging is currently desirable since these structures are easier to recycle and should contribute to a better quality, and therefore value, of the recyclate produced. Some possible solutions, of how the structure of a high barrier film for packaging of the fruit juice can look like, are shown in Figure 3.

Structure 1 (Al Metallization): It is, for example, possible to metallize PP films (Structure 1), if the surface is sufficiently smooth. This is typically achieved by a pre-coating, also called as planarization. Afterwards the PP is metallized via physical vapor deposition (PVD) process. Flexible packaging structures with metallisation are remarkably different to structures containing aluminum foil. The permeation barrier layer provides a good barrier against oxygen and water vapor and is opaque (see Section 2.4). The thickness of the metallization layer is typically 50-80 nm; the thickness of the pre-coating is 1 µm in maximum. The typical layer thickness of an adhesive in that kind of multilayer-laminates is 3 µm.

In summary, this would mean 7.05 µm of foreign material (including the adhesive, the pre-coating, and the metallised layer), which is 5.8 wt% of the investigated mono-material film laminate. Laminated and printed mono-material based laminates do not cause any sortability problems. Metallisation is compatible to the plastics mechanical recycling process since it is very thin to contribute to the allowed foreign content share. One important point, which is worth to mention here is that the metallised layer in our design is inside of the laminate, and we do not have any surface metallisation, which might have caused some issues. However, the FlexFunctin2Sustain mono-material laminates with the metallised Al layer do not have this issue.

Structure 2 (SiO_x deposition): Silicon oxide (SiO_x) is another possibility for the achievement of barrier performance against gas and water vapour permeation into the product. SiO_x is applied as a very thin layer coating to give additional barrier properties. It is often at a layer thickness of 80-90 nm within a structure so fall under the maximum of 5% by weight of the total structure.

In comparison to the metallized film, the SiO_x solution (Structure 2) in Figure 3 is transparent, but provides a good barrier against oxygen and water vapor (see Section 2.4). The total thickness of the foreign material, i.e. the SiO_x, the planarization and the adhesive layers, is typically not more than 4,05 µm. For the example structure used during this recycling test the amount of foreign material was 3.3 wt%.

Structure 3 (nanocomposite lacquer coating): This one is a mono-material laminate, which is produced at ambient conditions, whereas structure 1 and 2 need to undergo a vacuum deposition process step. Structure 3 provides a good oxygen barrier, but only a little barrier against water vapor (see Section 2.4). The film can be considered as transparent. The amount of foreign material within the used example structure was 3.3 wt%.

All structures shown for the different barrier technologies (metallization, SiO_x deposition and nanocomposite) are examples that can vary in the thickness of the respective layers and the order and number of layers. The examples chosen are all sealable. The different amount of layers results from the fact that not every surface of every structure can be printed. For example printing and metallization on the same film is not possible. This is why Structure 1 consists of 3 layers, which is called a “3-ply-structure” while Structure 2 and 3 are 2-ply-structures. Depending on the used polyolefin type, preferably PP or PE, the layer thicknesses of the used films, the barrier technology, the order of the layers, the total number of the layers, and more, these novel designed laminate structures can be applied as stand-up pouches, or as a sealing film on a tray, or as tube pouches or as simple walletpack.

2.1.3. Life Cycle Assessment

The environmental performance of the FF2S materials, surfaces and products will be assessed by means of environmental Life Cycle Assessment (LCA). This chapter describes the tools that will be used for the LCA of the novel designed recyclable mono-material based laminates for circular economy.

LCA is a quantitative tool used to identify, characterise and analyse the environmental impacts a product or a system generates across its entire life cycle from raw material extraction ('cradle') to final disposal ('grave'). By taking such a perspective, LCA helps businesses understand the flows of matter and energy involved in the material, processing, distribution and packaging elements of their products (Figure 4).

Figure 4. Life cycle phases of a product

LCA methodology

The LCA framework is defined by the International Organization for Standardization (ISO) (Figure 5). This framework defines four key stages: goal and scope definition, inventory analysis, impact assessment and interpretation. The process is iterative; stages might be revisited to accommodate eventual changes in study goals and data availability.

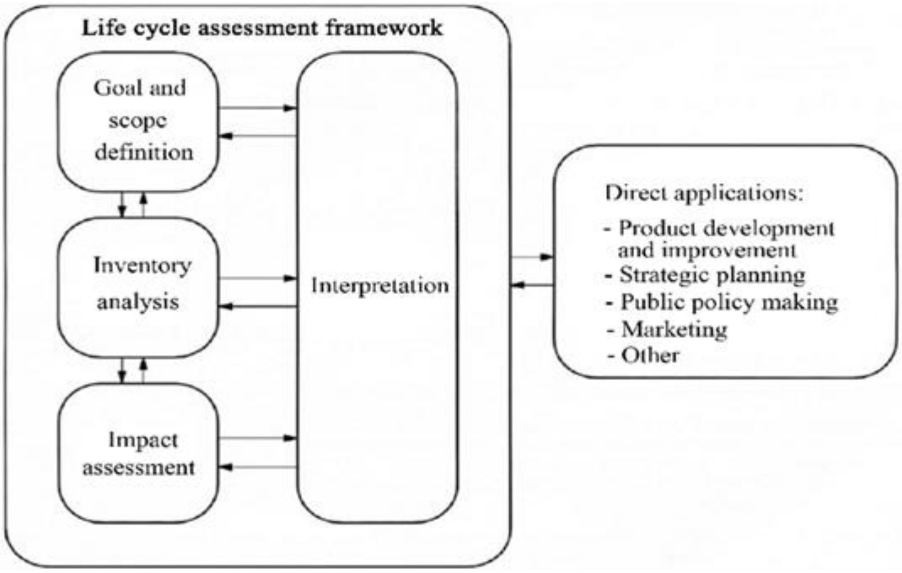

Figure 5. LCA Framework – ISO 14040:2006

Phase 1 - Goal and scope definition

In this phase, the application and type of LCA is described, the product systems are defined, as well as the geographical and temporal scope. This step also includes the definition of the functional unit, which acts as the reference for the subsequent steps.

Functional unit

The functional unit is a quantitative description of the function for which the assessment is performed, and the basis of determining the reference flow of product that scales the data collection in the next LCA phase (the inventory analysis).

> **The functional unit**
> *« Quantified performance of a product system for use as a reference unit » (ISO 14040 :2006)*
>
> *Example: Orange Juice Pouch*
> *FU = "Contain, protect, store 200 ml of orange juice during its lifetime"*

System boundaries

The system boundary defines which processes should be included in (or excluded from) the system. The definition of system boundaries allows identifying the data required to fulfil the objectives of the study. An example of system boundaries for a flexible packaging life cycle is shown in Figure 6.

Types of LCA

Cradle to grave LCA is an assessment of the full Life Cycle Assessment from resource extraction ('cradle') to use phase and disposal phase ('grave').

Cradle to cradle LCA is a specific kind of cradle-to-grave assessment (the full Life Cycle Assessment from resource extraction ('cradle') to use phase and disposal phase ('grave')), where the end-of-life disposal step for the product is a recycling process.

Cradle to gate LCA is an assessment of a partial product life cycle from resource extraction (cradle) to the factory gate (i.e., before it is transported to the consumer). The use phase and disposal phase of the product are omitted in this case.

Gate-to-gate LCA is a partial LCA looking at only one value-added process in the entire production chain.

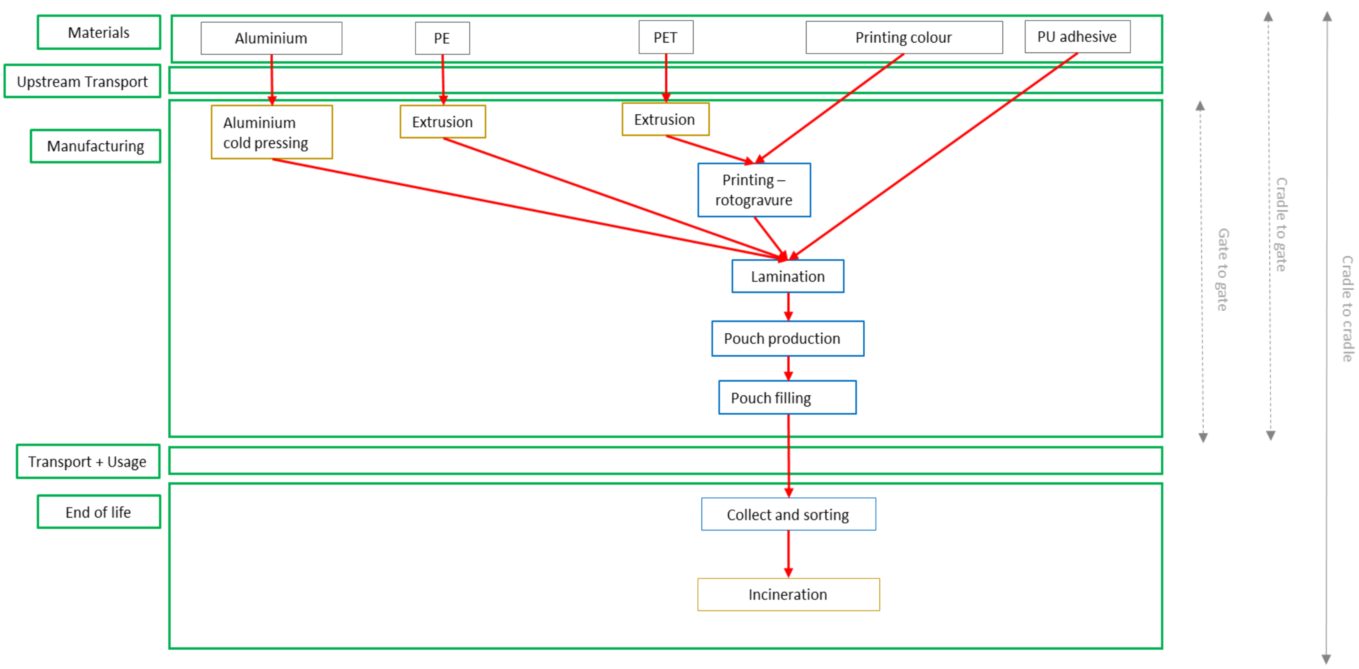

Figure 6. Example of the life cycle and system boundaries for a flexible packaging for liquid products

Phase 2 - Inventory analysis

This second phase is the most time-consuming part of an LCA. The analysis is guided by the goal and scope definition, and it consists of collecting and compiling input and output data from all processes in the studied product. Three types of data can be collected in this phase:

- **Primary or specific data:** data measured or calculated by the company.

Examples: Product composition, process energy consumption, etc.

- **Semi-specific data:** average data of the sector used in the absence of primary data.

Example: Volatile Organic Compounds emissions, truck loading rate...

- **Secondary data:** Data from generic databases

Example: Impact of materials, Impact of the electric kWh in France, Impact of the ton-kilometre in 3.5t trucks...

Phase 3 - Life cycle impact assessment

This step consists in evaluating the environmental impact of the inputs and outputs identified in the previous phase, translating them into environmental impacts (Midpoint) and potential damages (Endpoint) as shown in Figure 7. This step of translation is called "Characterisation" and is generally performed using an LCA software.

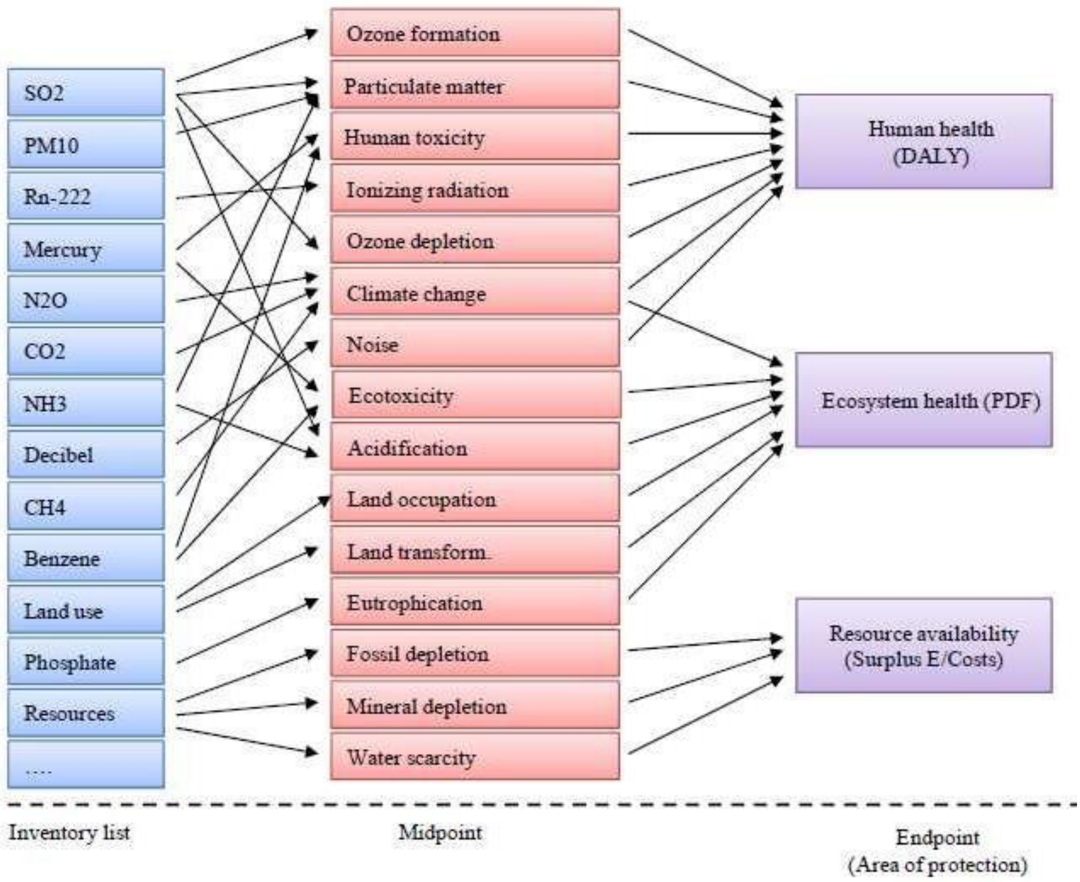

Figure 7. List of impact categories for characterization at midpoint and endpoint level

To carry out this classification (or characterisation) step, several calculation methods can be used. The most known methods are: ReCipe Midpoint, ReCipe Endpoint, ILCD 2011, CML 2002 and Environmental Footprint.

Phase 4 - Interpretation

This step aims at analyzing the results, at each step of the LCA, in order to establish conclusions in a transparent way and always in relation to the set scope. The limits of the study must be explicit and clear. The principle of this phase is to identify the most significant processes of the assessment in order to establish priorities for action. This can be done in several steps:

- Analysis using the 3 whys: stage, process, substance
- Analysis by PARETO diagram
- Sensitivity analysis: vary different sensitive parameters (in %, set maximum and minimum limits, by choice of hypotheses) to test the robustness of the study. Sensitivity analysis can be applied on different elements such the functional unit, study assumptions (transportation, uncertain data, end-of-life scenario), study boundaries, allocation, data and method choice.

In the SimaPro software, the interpretation step is performed using several visualization tools. The environmental impacts as a single score applied to each unit process enable to quickly identify which process is the major contributor to the overall impacts, and pinpoint on which impact to focus.

Phase 5 (Optional) – Critical review

The use of LCA results to compare similar products or to communicate on the environmental profile of a product requires a critical review.

A critical review is a process to verify whether a Life Cycle Assessment has met the requirements for methodology, data, interpretation and communication, and whether it is consistent with the principles of the methodology as indicated by the applicable standards.

A critical review may be performed by an internal/external expert or by a stakeholder committee. The review statement, the expert's comments and any responses to the expert's recommendations should be included in the LCA report.

2.2. Upgrades for circularity by design of materials

There have been two upgrades involved for the production of the novel designed mono-material laminates for flexible packaging, which have been described in Chapter 2.1.2. These upgrades are implemented for the production of the Structure 3 laminate consisting of a nanocomposite lacquer coating. Section 2.2.1 describes the high shear rate mixing process for the production of the homogeneous dispersion formulation used as a coating for oxygen barrier functionality. Section 2.2.2 describes the upgrade of the slot die coating unit implementation for its homogenous coating.

2.2.1. Homogeneous dispersion formulations through of high shear rate mixing

The first - and the most challenging - step during the production of a nanoparticle-containing lacquer is the homogeneous and stable dispersion of the nanoparticles. The dispersion needs to be stable in the solvent, the polymer solution is based on. In the scope of this project, the nanocomposite lacquer is based on ethylene vinyl alcohol (EVOH) copolymer, which is soluble in water.

Figure 8. (1) Zirconium dioxide cup with zirconium dioxide balls; (2) filled with montmorillonite pre-dispersion; (3) planetary ball mil; (4) final montmorillonite dispersion after milling

Therefore, the nanoparticles need to be dispersed homogeneously, without agglomerations, in water. The nanoparticles used are platelet-shaped montmorillonite (MMT) particles. They are pre-dispersed slightly with a magnetic stirrer and this brownish pre-dispersion (see Figure 8), with a solid content of 5 wt%, is ball milled with a planetary ball mill and zirconium oxide balls for 1 h. After the ball milling the dispersion changed its colour to white. The solid content of 5 wt% is the maximum solid content that is possible to process with the chosen materials. A further increase of the solid content would lead to a strong increase in viscosity of the nanoparticle dispersion and the pasty appearance cannot be processed further. Afterwards the dispersion is mixed with EVOH granulate, this is described in Section 2.3.1.

2.2.2. Slot-die coating unit

Coating techniques are separated in self-metered and pre-metered techniques. Self-metered coating techniques are, for example, the k-bar coating, the roller coating, or the dip coating. It is difficult to produce thin films with self-metered coatings, since the thickness varies with process parameters. The coating fluid is constantly in contact with air, which leads to changes in viscosity, density or surface tension over time.

The advantage of pre-metered coating techniques, like slot-dies operating in bead or curtain coating mode, is that the wet film thickness can be controlled easily via the volume flow. In some set-ups, with closed reservoirs, the coating liquid has no contact to air during the whole coating process; i.e. there is no dependency of the coating layer thickness on other process parameters.

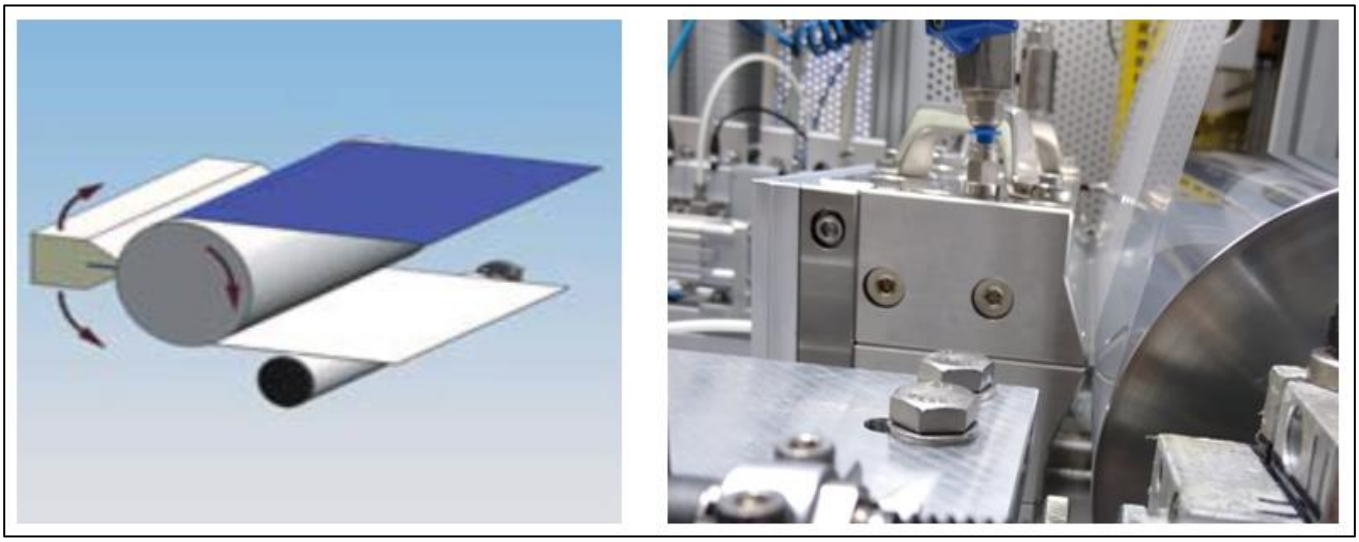

Figure 9. Schematic drawing of a horizontal set-up for slot-die coating operating in extrusion- or bead-mode (left) and a picture of 260 mm slot-die at the R2R machine at the FHG-IVV operation in bead-mode (right)

The installed slot-die coating system at the R2R machine at FHG-IVV enables several coating possibilities. Figure 10 shows the different steps from coating liquid reservoir to coated film. The coating liquid can be pre-heated before coating in a pressure tank. This tank also enables the possibility to de-gas the coating liquid previously to the coating and to stir it during the whole coating process. However, it is also possible to pump the liquid directly out of the cup where it was mixed. This is especially useful if only small amounts of coating liquid are available or if only a few meters need to be coated.

The alternating syringe pump pumps the coating liquid through the hoses. Before filling the slot die, there is an option to filter the coating liquid to get rid of agglomerations and/or gel-particles. The last step is the filling of the slot-die and the coating.

By knowing the solid content of the lacquer and the dimensions of the slot-die, it is possible to calculate the required volume flow for a respective wet and – after drying – dry layer thickness. An example for a set of coating parameters is given in Section 2.3.2.

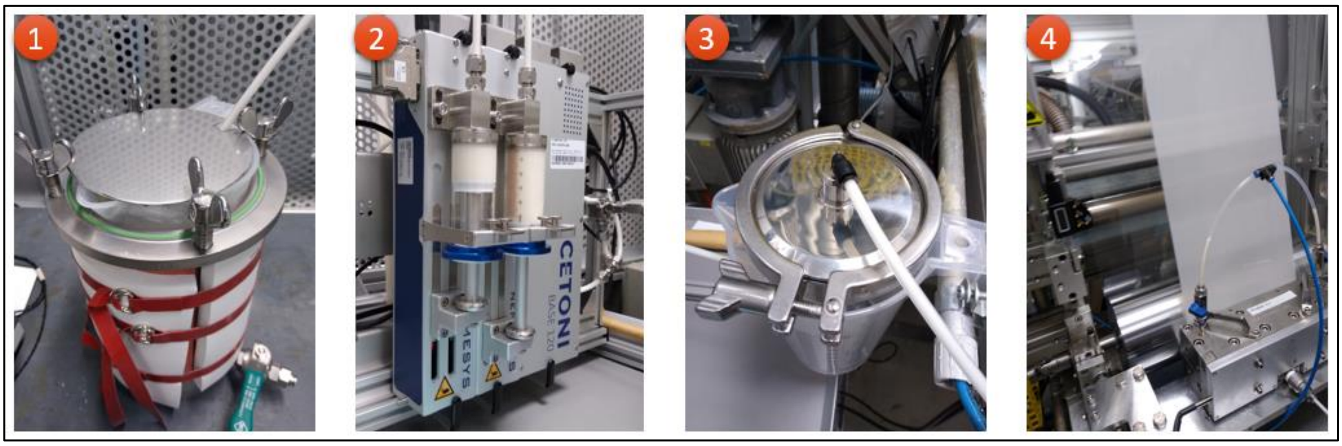

Figure 10. (1) Lacquer reservoir (2) alternating syringe pump (3) filter option (4) coating of lacquer

2.3. Production of the novel laminates

2.3.1. Nanolacquer formulation for Oxygen barrier

The EVOH is added in the form of granulate to the nanoparticle dispersion produced as described in Section 2.2.1. The dispersion, together with the EVOH granulate, is stirred and heated up to 90°C for 1.5 h to dissolve the EVOH. After cooling down, the nanocomposite lacquer is ready to use. By changing the amount of EVOH granulate, nanoparticle dispersion and addition of water (for dilution), it is possible to adjust several mixing ratios of nanoparticles to EVOH and to adjust the total solid content of the lacquer. In the scope of this project, a nanoparticle-containing lacquer with a total solid content of 6 wt% and a mixing ratio of EVOH to nanoparticles of 1:1 by weight was used. The total solid content of 6 wt% provides a lacquer with a suitable viscosity and a good coatability with either reverse gravure process or slot-die coating process.

2.3.2. Slot die coating process on BOPP substrate

Before laminating against a cPP sealing film, the BoPP substrate film needs to be coated with the nanocomposite lacquer to build up the structure as it is shown in Figure 3 (Structure 3). The formulation of the nanolacquer is described in Section 2.2.1 and 2.3.1, where the Upgrades were described.

Table 1. Parameter set for slot-die coating process of nanocomposite lacquer at R2R machine of FHG-IVV

Gap width	µm	300	**Web speed**	m/min	5
Slot length	mm	45	**Volume flow**	ml/min	4.,3
Slot width	mm	260	**Shear rate**	1/s	62
Solid content	%	6	**viscosity**	Pas	0.05
Dry layer thickness	µm	2	**Pressure loss**	bar	0,028
Wet layer thickness	µm	33.3	**Distance slot-die to web**	µm	150
Temperature pressure tank	°C	50	**Temperature slot-die**	°C	40
Curing temperature	°C	80	**Curing time**	s	50
Unwinding	N	20	**Controlled winding**	N	20

The gap width, the slot length and the slot width W_{slot} describe the geometry of the slot-die. The higher the viscosity of the coating fluid, the bigger should the gap-width be, to avoid too much pressure loss within the slot-die. The dry layer thickness, d_{dry} is typically the parameter to set. By knowing the solid content of the lacquer, c and the web-speed, v_{web} of the film, the necessary volume flow Q can be calculated:

$$Q = \frac{d_{\text{dry}}}{c} \cdot v_{\text{web}} \cdot W_{\text{slot}}$$

The volume flow can be adjusted very accurately with the syringe pump. To avoid possible changes in room temperature, the lacquer is coated at a temperature slightly above room temperature, at 40°C. The increased temperature also reduces the viscosity of the lacquer. The coating parameters applied during the coating process for the production of Structure 3 laminate are listed in Table 1.

2.3.3. Electron beam deposition of SiO_x and Al metallisation

The e-beam deposition of the silicon oxide, SiO_x (Ceramis process) has been performed at AMCOR using the e-beam deposition unit. The upgrades of this system are being performed in Work package 2, Task 2.1. Figure 11 shows the high barrier thin film deposition pilot line of AMCOR. Further details of this process will be described in Deliverable D2.1. The process uses a vacuum chamber, unwinding and rewinding units. The process starts with a plasma pre-treatment stage where the surface of the substrate film is cleaned and prepared for the coating process. The process uses two high voltage electron beam guns to evaporate the silicon oxide materials and build up a cloud above. The film substrate runs over a chilled reel, at ~ -15°C and through the temperature difference the SiO_x condensate on the surface of the film. The applied coating thickness of the SiO_x is in nanometer scale.

A similar type of Biaxially Oriented PP (BOPP) film used for the production of Structure 3 at a thickness of 18 µm was used for the SiO_x deposition at AMCOR. At the coating speed applied, the thickness of the inorganic barrier layer on top of BOPP substrate was in nanometer scale (ca 60 nm).

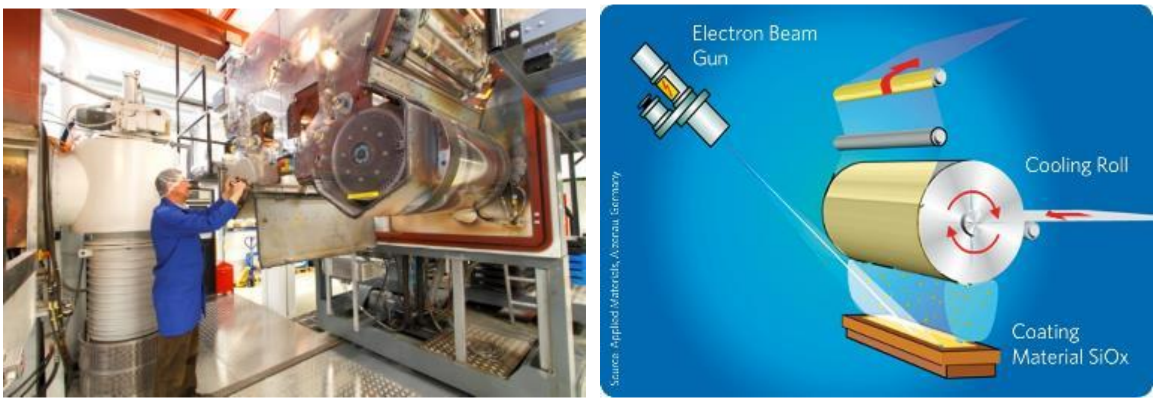

Figure 11. High Barrier Thin Film Deposition Pilot Line at the thin film deposition centre of Excellence of AMCOR

The Al metallised BOPP film has been provided to FHG-IVV from the project partner CapriSun and used for the production of Structure 1 as discussed further in Section 2.3.4.

2.3.4. Lamination process

The lamination process is similar for all the structures described in Section 2.1.2. The adhesive, which is used, is a typical lamination adhesive with food contact approval. It is a two component polyurethane system.

Figure 12 shows the different process steps during a typical lamination at the roll-to-roll (R2R) machine of FHG-IVV. The adhesive is transferred from a pan via an engraved roll to the film. The film, where the adhesive is applied, is usually the more resistant one. For example, if a metallized BoPP film is laminated against a cPP sealing film, then the adhesive is applied on the sealing film to avoid possible stresses on the metallized layer due to rolls or heat at the drying station. After the application of the adhesive to the film, a doctor blade removes the excess adhesive. The coated film then moves through the dryer. The heat activates the adhesive and evaporates the solvent. After the dryer, the adhesive needs to be tacky, but should still be able to wet the lamination film. The coated film and the lamination film are pressed together with two rolls and the lamination is done. Table 2 shows the parameters used during the lamination processes at the R2R machine of FHG-IVV.

Table 2. Parameter used during the lamination process of FHG-IVV

Convection oven		Web-tension			Coating unit			
Frequency	**Temperature**	**Unwinding**	**Controlled winding**	**Lamination unit**	**UV-power**	**Solid content**	**Speed coating head**	**Film floating speed**
Hz	°C	N	N	N	%	%	m/min	m/min
40	50	30	50	30	44	30	7	5

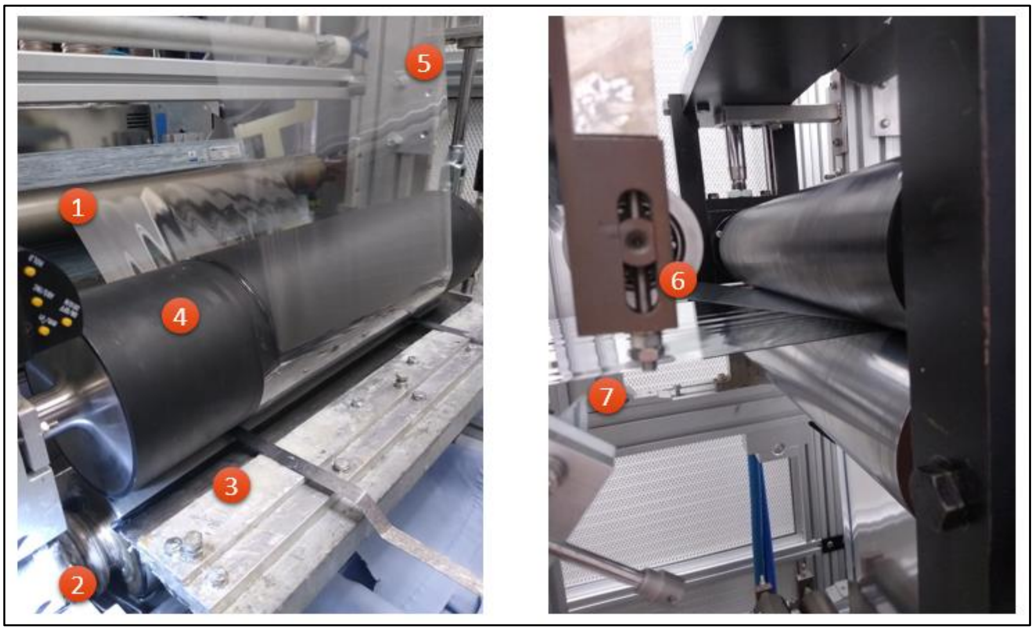

Figure 12. Lamination process at FHG-IVV: (1) uncoated film (2) pan with lamination adhesive and engraved roll (3) doctor blade (4) rubber backing roll (5) coated film (6) coated film after drying in a convection oven (7) lamination web

The three structures; Structure 1, Structure 2 and Structure 3 have been produced at about 1000 m at a width of 300 mm. Figure 13 shows the rolls produced.

Figure 13. The rolls of the three mono-material based laminates: Left: Structure 1 with Al metallisation, Middle: Structure 2 with SiO_x deposition, Right: Structure 3 with nanocomposite lacquer, all 1000 m at 300 mm

2.4. Characterisation of the laminates

Table 3 shows typical properties that are measured for a flexible film used in the food packaging industry. The three novel laminates have been characterised in terms of these properties and the results are presented in Table 3. The exact structure and layer thicknesses are confidential information, but the structures can be used as stand-up pouches for fruit juices, as an application example.

Table 3. Typical properties measured for a flexible film in food packaging. Compared are examples for the film structures shown in Figure 3 and produced as shown in Figure 13. The abbreviations MD and TD stand for machine and transverse direction, respectively. The seal strength is measured at cold conditions, meaning at room conditions.

	Structure code	Oxygen transmission rate	Water vapour transmission rate	bond strength	Seal strength	Tensile strength	Elongation at tear	thickness	weight
Condition		23 °C / 50 %RH	38 °C / 90 %RH	TD	cold	MD	MD	/	/
unit		$cm^3/(m^2 \cdot d \cdot bar)$	$g/(m^2 \cdot d)$	N/15mm	N/15mm	MPa	%	µm	g/m^2
State-of-the-art	SOA	<0.05	<0.00005	8,5	66	59	105	102	112
Metallized BoPP	1	0,05	0,7	1,9	22	102	36	127	117
SiO_x deposited BoPP	2	0,07	0,6	1,7	17	55	29	121	110
Coated BoPP	3	0,01	2,3	0,2	19	54	38	127	117

The oxygen transmission rate is similar for all samples within the range of 0,01 to 0,05 $cm^3/(m^2 \cdot d \cdot bar)$. Since the basis polymer systems, PET, PE and PP do not provide a high oxygen barrier; the oxygen transmission is provided by the respective barrier layer. For the case of oxygen barrier, the metallization, the SiO_x deposition and the coating with nanocomposite coating are suitable substitutes for the Al foil of the state-of-the-art structure (SOA).

Structure SOA provides a water vapour barrier below the measurement range of the device, <0.00005 $g/(m^2 \cdot d)$, since Al foil provides a very high barrier. The WVTR values for Structure 1 and 2 are 0.7 and 0.6 $g/(m^2 \cdot d)$, respectively. Only Structure 3 does not provide a sufficient water vapour barrier with a value of 2.3 $g/(m^2 \cdot d)$. The reason, therefore, is that the other structures contain a closed inorganic barrier layer, which provides high barrier against water vapour, whereas the water vapour barrier of the nanocomposite structure only comes from the polyolefin. EVOH is sensitive to water, because contact to water weakens the intermolecular hydrogen bonds of EVOH. Therefore, EVOH does not provide a water barrier.

Structure SOA hast the highest bond strength (8.5 N/15mm) and seal-strength (66 N/15mm). Indeed, a bond strength of more than 1.5 N/15mm is sufficient for most of the food packaging applications. This is fulfilled by Structure 1 and Structure 2 with a value of 1.9 and 1.7, respectively. Structure 3 provides a bond strength of 0.2 N/15mm. The required sealing strength strongly depends on the product and packaging. The values for the mono-material laminates are here in the same range of around 20 N/15mm and about 60 % lower than for structure SOA with a value of 66 N/15mm.

During the tensile test structure SOA provided a high elongation (105 %), since the structures itself (PE, PET, and Al) are quite ductile. The elongation of the mono-materials is lower, and, within them, similar (29 – 38 %). The tensile strength is highest for Structure 1 (102 MPa) and similar for the others (54 – 59 MPa). Regarding the thickness and weight of the films, all films are within the same order of magnitude.

As so often, the choice of the right film depends on the requirements of the packaging and the food product. If there is no peeling force on the film and only a slight water vapour barrier is necessary, then Structure 3 might be sufficient. If a light blocking is requested only structure SOA and 1 are opaque and will be suitable for the considered application.

2.5. Extrusion of films using PP recyclates

The examples of the structures given in Figure 3 that have been characterized in Section 2.4 have been sent to IPC for mechanical recycling. The whole mechanical recycling process is described in detail in Deliverable D3.3 of this project. The resulting recyclates, rPP, have been sent to FHG-IVV for film extrusion.

Table 4. Composition of extruded films according to RecyClass protocol and parameters used during their extrusion process at FHG-IVV

Extruded films	Barrier film rPP	Control film rPP	Virgin PP pellets	Total amount of PP	Melt temperature	Chill roll temperature	Film thickness
	wt%	wt%	wt%	Wt%	°C	°C	µm
Ref-B0	0	50.0	50.0	100	260	50	50
Structure 1-B25	12.5	37.5	50.0	99.6	260	40	49
Structure 1-B50	25.0	25.0	50.0	99.3	260	40	50
Structure 2-B25	12.5	37.5	50.0	>95	260	50	51
Structure 2-B50	25.0	25.0	50.0	>95	260	40	48
Structure 3-B25	12.5	37.5	50.0	99.5	260	50	52
Structure 3-B50	25.0	25.0	50.0	99.0	260	50	53

The recyclates themselves consist of different composition of barrier film, control film and virgin PP pellets. The control film was the pure cast PP that has been used as the sealing film in the Structures 1, 2, and 3, and the recyclates of this control film were first mixed in different ratios with recyclates of each barrier structure. Then, the pellets resulting from this initial mix have been further mixed with virgin PP pellets (50% in all the cases). The final (theoretical) PP content of the recyclates, as was used for film extrusion at IVV, is summarised in Table 4.

The extrusion of the granulate mixtures was performed at FHG-IVV according to the process conditions as shown in Table 4. The film thickness was set to 50 µm, the extrusion process was performed successfully at the extrusion line (see Figure 14) at this layer thickness of 50 µm. An attempt to target a thinner film thickness of 30 µm would lead to cracks, holes and film break during its processing.

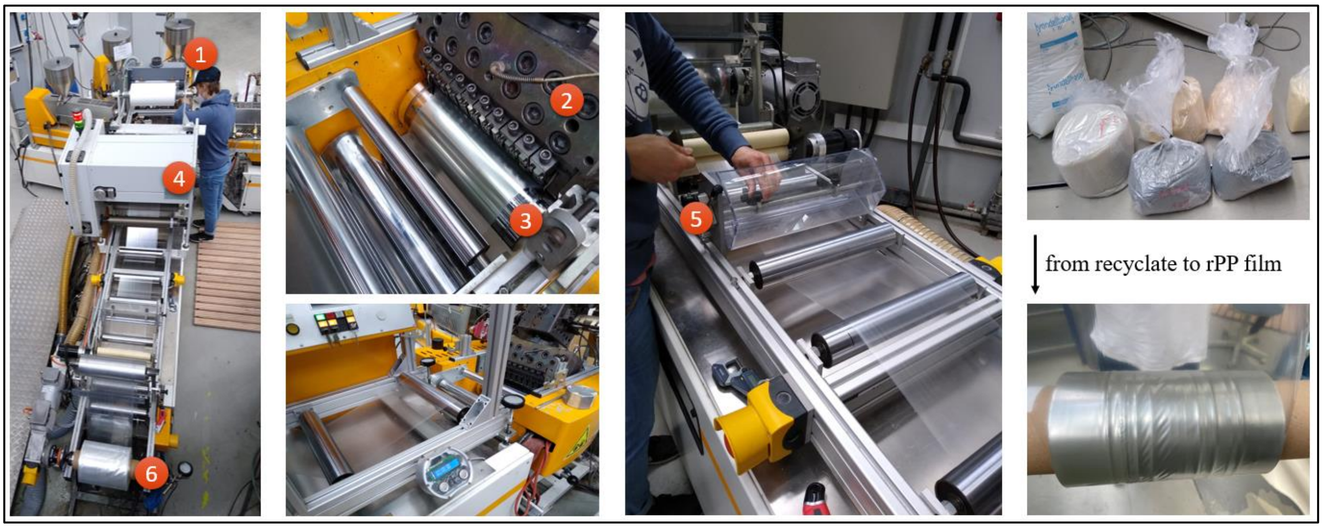

Figure 14. (1) Hopper for filling granulate, in that case a mixture of 50 % virgin PP and 50% recyclates (2) die head for extrusion (3) chill roll (4) thickness control (5) cutting station (6) up-winding. The roll on the right hand side is an example for the rPP film of Structure 1-B25

The extrusion process (see Figure 14) starts with filling the granulate mixture in the hopper. The granules are moved through the heated extruder by a excentric screw and are melted and heated up to 260 °C. The melt is pumped through the die and formed to a film. The film is cooled down by hitting the chill roll and is then moved over rolls to the thickness control unit, cutted, and up-winded at the end.

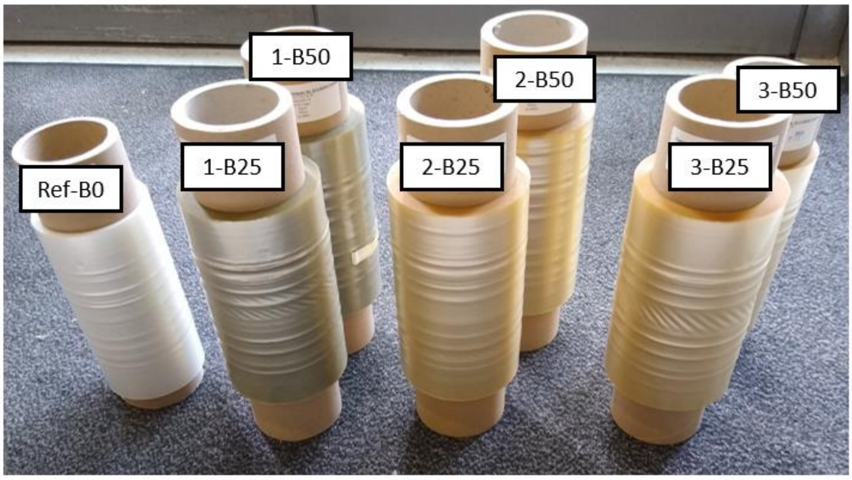

Figure 15. Recycled films (rPP); composition listed in Table 4

The mechanical and visual properties of the extruded PP films are characterized.

As shown in Figure 16, the elongation at yield for all samples have a value of around 12 %. The Young's Modulus is for all samples at around 550 MPa and the tensile strength at around 23 MPa for all samples. The thickness of all films is around 50 µm as shown in Table 4. The mechanical properties of the produced films are close to the control film's (Figure 16).

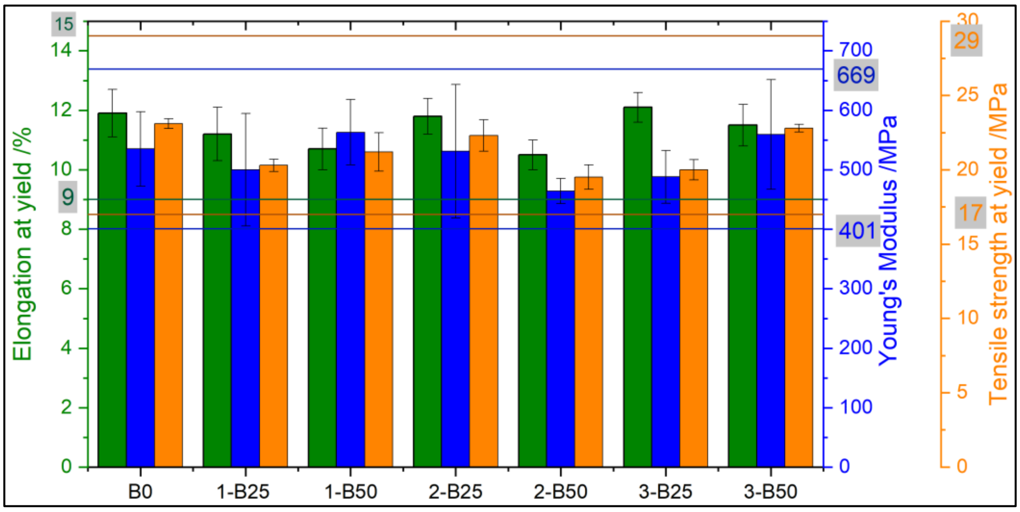

Figure 16. Young's Modulus, elongation at yield and tensile strength at yield for the recycled films in machine direction. The numbers in grey represent the 25 % delta value from the control film.

During the investigations of the surface, several 10 µm high surface spikes have been found (see Figure 17) in particular for Structures 1 and 2. The number of these spikes and inhomogeneity increased with higher percentage of recycled material.

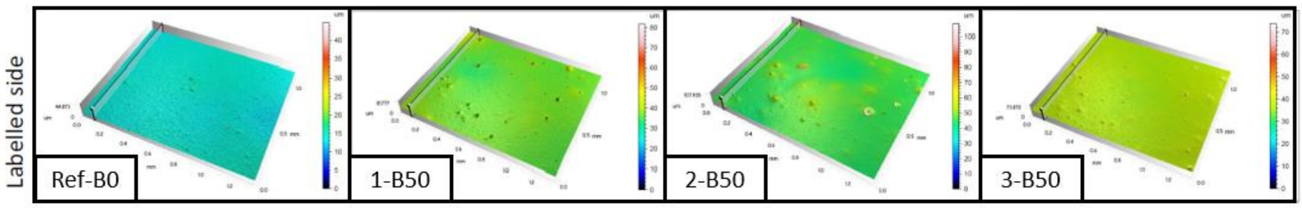

Figure 17. Surface images of recycled PP films acquired with WhiteLight Interferometry

The optical appearance of all samples is similar. As shown in Table 5 all samples provide a transmittance for the whole visible range of more than 90 %.

Table 5. Transmittance of the recycled films in the visible range

Sample	Ref-B0	1-B25	1-B50	2-B25	2-B50	3-B35	3-B50
Transmittance (%)	93	91	90	92	93	92	92

Summarizing the characterizations of the recycled films lead to the finding that the recycling of the novel mono-material laminates introduced in Section 2.1.2 is possible, indicating the possible contribution of the flexible packaging films to the circular economy through the appropriate design of the flexible packaging materials.

3. Conclusions

First of all, novel mono-material laminate structures to substitute non-recyclable multi-material structures have been found. Multilayer disassembly was therefore not relevant in that case, as the structures were designed to be almost exclusively mono-material.

The challenge is to keep the functional properties that the state-of-the-art structures provide. One of the most challenging properties to achieve is a sufficient barrier against oxygen and water vapour. The solutions that we suggested in this project provide different barrier technologies. On the one hand, there are structures based on vacuum deposition of Al or SiO_x, on the other hand, there is the new option with an EVOH based nanocomposite. The achieved barrier against oxygen and water vapour could only be achieved because of the updates of the several production lines. The upgrade of the E-beam machine allowed to deposit a homogeneous SiO_x- or Al-layer on a polyolefin film without any cracks. The change from a dispersing device to a ball mill enabled a homogeneous dispersion of nanoparticles in water without agglomeration. The upgrade to the slot-die coating device allowed to coat the nanolacquer in thin layers on polyolefin films. Depending on the requirements, the right structure can be chosen, because every structure has its advantages and disadvantages.

The metallized option, for example, provides very good barrier against oxygen and water vapour, but is not transparent. The SiO_x deposited option also provides very good barrier, but is not very resistant against bending. The nanocomposite structure can be produced at ambient conditions and no vacuum process step is necessary, but the barrier against water vapour is not too high. By knowing the requirements and the usage of the film, the right structure can be designed accordingly for circular economy.

All suggested structures are recyclable according to CEFLEX guidelines (guidelines, 2019). The recyclability has been tested. The films were sent to IPC after production. At IPC the films were mechanically recycled (Deliverable 3.3). The achieved recyclates have been used at FHG-IVV to extrude a rPP film. The films were characterized and there was no significant difference between a control film and the rPP films measureable, thus, the films can be considered as recyclable.

The recyclable structures will be compared with a LCA, and the results will be part of the dedicated Deliverable in Work Package 5. A Life Cycle Cost assessment could also be performed to provide additional help for deciding on the best structure.

The rPP films can then be re-used in several applications. In the scope of this project, they will be tested as substrate material for flexible electronics. Therefore, the r-PP films (the film extruded using the recyclates of Structure 3) have already been sent to FHG-FEP by FHG-IVV with and without a planarization layer for electrode and barrier deposition. OET will be the final recipient of those films, in order to test them for photovoltaic applications, demonstrating the circularity for flexible applications.

4. Degree of progress

Deliverable 3.1 is completed by 100%

5. Dissemination level

This Deliverable is public.

6. References

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