CIRCULAR FoodPack

# D5.2 DELIVERY OF TEST MODEL SAMPLES TO TASK 4.5 FOR MIGRATION TESTS, AND CHARACTERISATION RESULTS

## WORK PACKAGE 5

Associated Task(s): T5.2
Functional barrier layer combinations.
Assessments with model contaminants

**Lead Partner**: Fraunhofer IVV
**Partners involved**: AMCOR-K, AMCOR-G, Siegwerk
**Dissemination Level**: Public
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## HISTORY LOG

| Version number | Date | Organization name | Comments |
|---|---|---|---|
| 0.1 | 29-03-2022 | Fraunhofer IVV | Initial version |
| 0.2 | 06-04-2022 | BayFOR | 1st review |
| 0.3 | 06-04-2022 | Fraunhofer IVV | Final version |
| 1.0 | 05-04-2023 | Fraunhofer IVV | Revised for resubmission |

## QUALITY REVIEW

| Quality check | Date | Status | Comments |
|---|---|---|---|
| Diana Kemmer, Petra Schmid | 29-03-2022 | Reviewed | Corrected and complemented |
| Pieter Cools | 01-04-2022 | Reviewed | For confidentiality |
| Verena Bürger-Michalek | 06-04-2022 | reviewed | Corrected, comments |
| Esra Kucukpinar | 06-04-2023 | reviewed | Corrections |
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## DISCLAIMER



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## PUBLISHABLE EXECUTIVE SUMMARY

The main objective of Work Package (WP) 5 is to design, develop and upscale sortable mono-material packaging films, suitable for recycling and containing post-consumer recyclates (PCR). The developed film shall be suitable for the packaging of food and non-food products. The incorporated PCR will be the outcome of the improved sorting and treatment technologies of WP2 and WP3, and solely be sourced from food packaging. This is required to fulfil the premise of the Commission Regulation (EC) No 282/2008 of 27 March 2008 on recycled plastic materials and articles intended to come into contact with foods and amending Regulation (EC) No 2023/2006.

The films designed within this WP shall comply with the existing Design 4 Recycling (D4R) guidelines and contain more than 90% polyethylene (PE). Additionally, the novel film structure will contain at least 70% of PCR in one single layer. The development of high performance functional barriers is required, to effectively prevent migration of any residual contaminants from the PCR, and to keep them below the migration limits set out by legislations and European Food Safety Authority (EFSA).

The developments of WP2 and WP3 will be implemented in the mono-material packaging design, so that the novel material can be collected and sorted. Especially, the inclusion of tracers for improved sorting and an adequate choice of inks to be well deinkable will be respected. Moreover, the novel material will fulfil all gas barrier and mechanical requirements defined by the end users. As well all requirements regarding sealability and machinability at the end users' packaging lines will be satisfied. Any possible effect on production efficiency or energy consumption should be reduced or avoided.

While Task 5.1 is dealing with the requirement collection from the end users, namely from the partners Nestlé, Amcor-Gent and Amcor-Kreuzlingen, Task 5.2 is targeting a screening of potential functional barriers to be used in the novel film structures. High-performance functional barrier combinations of inorganic or polymeric coatings will considered. Moreover, barrier lacquer formulations and top coating possibilities will be investigated. Commercially available barrier films will be benchmarked.

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## ABBREVIATIONS

| Abbreviation | |
|---|---|
| $AlO_x$ | Aluminium oxide |
| Amcor-G | Amcor Flexible in Gent |
| Amcor-K | Amcor Flexible in Kreuzlingen |
| D4ACE | Design for a circular economy |
| D4R | Design for recycling |
| EFSA | European Food Safety Authority |
| EU | European Union |
| EVOH | Ethylene vinyl alcohol |
| GC-FID | Gas chromatograph - Flame Ionization Detector |
| LDPE | Low density polyethylene |
| PCR | Post-consumer recyclates |
| PE | Polyethylene |
| PET | Polyethylene terephthalate |
| $SiO_x$ | Silicon oxide |
| WP | Work package |

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# 1. INTRODUCTION

The main objective of WP5 is to design, develop and upscale sortable mono-material packaging films, suitable for recycling and containing post-consumer recyclates (PCR). The developed film shall be suitable for food and non-food products.

The films designed within this WP shall comply with the existing Design 4 Recycling (D4R) guidelines and target at least 70% of PCR in one single layer. The PCR integration into food packaging items will require functional barriers. For the development of high performance functional barriers it is required to screen potentially suitable barrier film structures, which will fulfil the migration limit set by EFSA.

While Task 5.1 is dealing with the general requirement collection from the end users, namely Nestlé and Amcor-G (representative for a personal care product producer), Task 5.2 is specifically targeting potential functional barriers to be used in the novel film structures. Therefore, Amcor-K and Amcor-G supply different barrier films. High-performance functional barrier combinations of inorganic or polymeric coatings/layers are considered. Moreover, barrier lacquer formulations and top coating possibilities from Siegwerk are investigated and other commercially available barrier films are to be benchmarked.

These supplied barrier films or barrier lacquers are laminated to an artificially contaminated virgin cast film. The contamination solution and the procedure follow the general EFSA approach of challenge tests, which are usually performed to approve novel processes or materials. Details on the lamination process and virgin material contamination are described in this deliverable.

The permeation barrier testing will be performed in WP4. Based on the results obtained from these barrier tests, the structures for the targeted food and non-food applications will be designed with the selected functional barriers, including the developments of WP2 for an improved sorting and respecting the suitability for the recycling process developed in WP3.

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# 2. BARRIER FILM SELECTION

The objective of Task 5.2 is to screen different barrier films regarding their suitability as functional barrier against possible contaminants from PCR. Hereinafter, the general composition of the investigated barrier concepts will be described. The CEFLEX D4ACE Guidelines[1] will serve as recyclability reference. There, it is stated that the addition of lacquers, adhesives or coatings at a layer thickness range of 20 nm to 10 µm do not change the definition of mono-material. Furthermore, for the recycling of PE or PP based packaging materials, it is specified that the lacquers, adhesives, barrier coatings, or tie-layers should not exceed the threshold value of 5 weight-% of the total packaging structure for the packaging to be considered recyclable.

## EVOH barrier film

EVOH is a common barrier polymer used in food packaging providing an excellent gas barrier. It is applied in thin layers, often as co-extrusion for example with polyethylene (PE). With a layer thickness thin enough to be below 5 weight-% of the total film structure, such a film structure can hence be considered as barrier material within this screening. A typical EVOH barrier film structure was supplied by Amcor-G, considered *EVOH1* in this deliverable.

## Barrier film with thin inorganic coating

A thin deposition of inorganic coating materials like $AlO_x$ or $SiO_x$ without additional coatings are considered as fully compatible with the PE recycling streams and are potentially suitable for a closed loop recycling. Therefore, four different thin barrier films are supplied by Amcor-K to Fraunhofer IVV; they will be considered as follows:

- CFP00098: thin inorganic coating on polyethylene/polyolefin film
- CFP00100: thin inorganic coating with primer A
- CFP00116: thin inorganic coating with primer B

Later in the project, additional film structures with inorganic coating might be considered and tested, especially in the work related to Task 5.4.

## Barrier lacquers

Barrier lacquers at sufficiently thin layers are also being investigated within this task. The preparation of these samples is ongoing with Siegwerk. The lacquers will be considered *BL1* and *BL2*.

## Benchmark films

To put the novel film structures in context, commercially available barrier films will also be benchmarked, like an oriented PET film or an $AlO_x$ based barrier film. Therefore, samples will be prepared with benchmarked films and/or literature and the involved experts will be consulted.

[1] CEFLEX Technical report, designing for a circular economy, June 2020

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# 3. CONTAMINATION OF VIRGIN PE WITH MODEL SUBSTANCES

## 3.1 ARTIFICIAL CONTAMINANTS-COCKTAIL

The evaluation of the barrier concepts, which will be developed within WP5, is done by intentional contamination of virgin PE material in pellet form. A set of surrogates is chosen in Task 4.5 of WP4. This set is already well established for the authorization of recycling processes for the manufacturing of recycled PET. Moreover, the combination is based on extensive experimental and analytical experience of the Fraunhofer team working in WP4. The substances cover a wide spectrum of volatility and polarity and a wide range of migration-relevant molecular weights and serve as representatives for contaminants present in real post-consumer waste.

- Toluene, CAS 108-88-3
- Chlorobenzene, CAS 108-90-7
- Phenyl cyclohexane, CAS 827-52-1
- Benzophenone, CAS 119-61-9
- Methyl stearate, CAS 112-61-8
- Methyl salicylate, CAS 119-36-8

The surrogates are chemically stable and can be quantified analytically by GPC-FID.

## 3.2 CONTAMINATION PROCESS DESCRIPTION OF VIRGIN PE

For the measuring of the permeation barrier of films, virgin material is contaminated with the surrogate-cocktail in liquid form in this case. Therefore, the mix is pumped into the virgin material melt and homogeneously distribution through shearing in the extruder. By laminating the contaminated material with the barrier film of interest, they will be in direct material contact, and hence represent the scenario of PCR material in contact with the barrier film.

The contamination process takes place in a lab sized Dr. Collin twin screw extruder at Fraunhofer IVV. Figure 1 shows the set-up of the contamination process. The surrogate-cocktail is filled into a syringe, which is installed in a syringe pump. A dosing speed can be adjusted to the targeted concentration in the material. Since the spiked material undergoes two extrusion processes subsequently after the dosing, initial contamination-compounding and film extrusion, the initial concentration is set higher than the finally required. With a dosing of 1000ppm, the final concentration after the spiking becomes around 700ppm, which allows for a sufficient concentration of surrogates in the material also after film extrusion. Anyway, before any permeation barrier testing, the initial concentration is always determined.

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*Figure 1: Contamination set-up at Fraunhofer IVV, from left to right: pelletizer, full extruder set-up with dry ice cooling bath, syringe system to pump the contamination solution into the extruder. The process flow starts with the dosing of the contamination solution, extrusion and then pelletizing.*

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# 4. PREPARATION OF MODEL SAMPLES

## 4.1 EXTRUSION OF CONTAMINATED PE FILM

To avoid any volatilisation of the added contaminants, the spiked material was processed into cast film the day after the contamination. The equipment used for this extrusion is a Dr. Collin seven-layer co-extrusion line, which is shown in Figure 2. The extrusion parameters (temperature profile in the extruder, process pressure or screw speed) were the same for the spiked PE as for the virgin PE used to set up the machine. No gels or discoloration were observed. 300µm mono-layer cast film of contaminated PE is extruded at a total length of 180m and film width of 235mm. This film was stored in a sealed aluminium-PE laminate bag, to avoid any volatilisation of the surrogates until the laminate production with the barrier films supplied by Amcor-G.


*Figure 2: virgin (left) and contaminated (middle) PE film extruded at IVV on a Dr. Collin extruder (right)*

## 4.2 LAMINATION CONCEPT OF CONTAMINATED FILM TO BARRIER FILMS

The contaminated PE film was subsequently laminated to the chosen barrier films described in Chapter 2. A scheme of the lamination/coating line at Fraunhofer IVV is given in Figure 3 on the left side. The contaminated film is put into the unwinding station 1 (position 1 in the scheme) and the barrier film in the second unwinding station 2 (position 8 in the scheme). This allows for an easier and quicker change of barrier films, since the substrate PE film does not change. Additionally, a third unwinding unit is installed in the line (position 13 in the scheme). It is required to unwind a 20µm aluminium film, which is rewound with the produced laminate, as protection against set-off migration from the contaminated side to the virgin side.

Figure 4 shows a scheme of the winding setup. (A) shows the single components, which are combined. The lamination adhesive is applied on the spiked PE film in the coating unit. After the drying in the drying tunnel, it is laminated to the barrier film as shown in (B). This laminated web is then winded with the **aluminium foil**, fed from the roll positioned in (13) (ref. Figure 3), as shown in (C). This prevents contact between the virgin barrier film and the spiked PE film on the roll.

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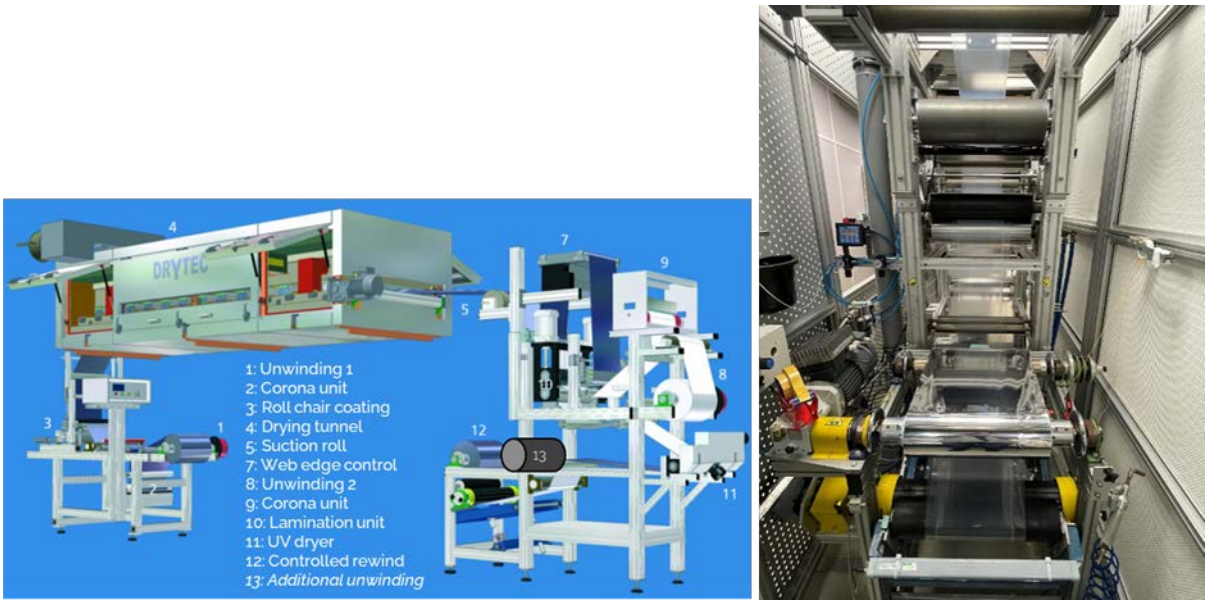


*Figure 3: Pilot lamination line scheme at Fraunhofer IVV (left), actual view towards (12) (right)*

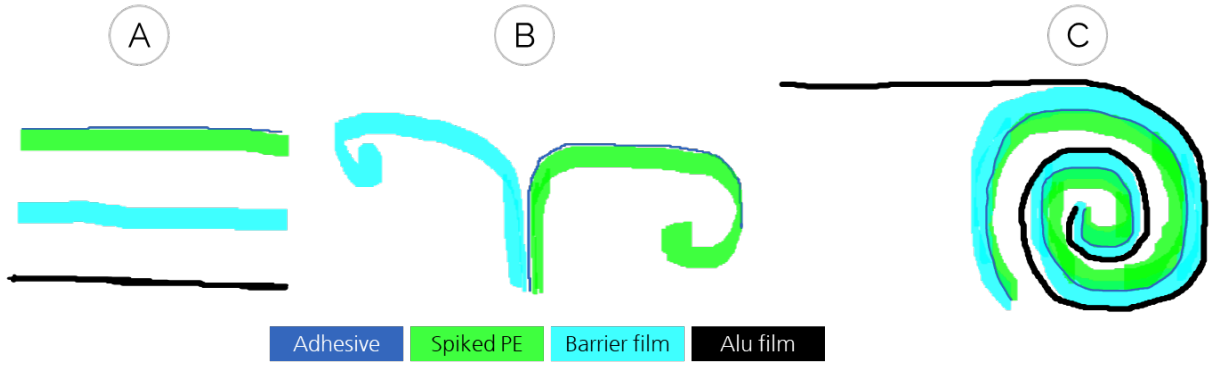


*Figure 4: Rewinding principle of laminate with aluminium film*

## 4.3 FINAL MODEL LAMINATE SAMPLES

The described process has been performed for the final laminate structures of CFP00101, CFP00102, CFP00103, and CFP00104, as shown in the Table 1 below. These film rolls of the first batch produced are shown in Figure 5. After the production and the curing process according to the adhesive requirements, the film rolls were packed into aluminium bags and sealed. They are only opened when the migration barrier measurements start.

*Figure 5: First batch of laminated model samples for WP4*

The second batch is currently under preparation. A new batch of spiked PE (Batch 2) has to be produced and the respective laminate production will be repeated. The second batch comprises the structures listed in

Table 2. The procedure of production will be the same as described in this deliverable. The results from this initial screening will be the base for the design of a mono-material film with incorporated PCR.

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*Table 1: First set of laminates with ongoing permeation measurements*

| Laminate No | Barrier film | Permeation measurements - Status |
|---|---|---|
| CFP00101 | with EVOH1 | Ongoing |
| CFP00102 | with CFP00098 | Ongoing |
| CFP00103 | only with primer A | Ongoing |
| CFP00104 | with CFP00100 | Ongoing |

*Table 2: Next laminate combinations to be prepared*

| Barrier film | Laminated to | Status |
|---|---|---|
| CFP00116 | Batch 2 | Planned |
| BL1 | Batch 2 | Planned |
| BL2 | Batch 2 | Planned |
| PET benchmark | Batch 2 | Planned |
| $AlO_x$ benchmark | Batch 2 | Planned |

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## 5. CONCLUSION

To conclude, the contamination of the virgin material can be performed by using a set of surrogates covering a wide spectrum of volatility and polarity. These substances serve as representatives for contaminants present in real post-consumer waste. A process methodology has been developed for the preparation of the laminates using the barrier films and the contaminated PE. The adhesive lamination process with the contaminated PE and the chosen barrier films was completed for the first set of samples and is planned for the next set of samples. This first set of laminates (4 different laminates) has already been delivered to Work Package (WP) 4.

All produced samples could be stored until the permeation measurement started in WP4. This shows that the handling worked well and that it can be repeated for the future samples.

Based on the results obtained by the WP4 team, the design of the novel film structures in D5.1 as part of WP5 will be performed.

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