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FlexFunction2Sustain

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

Starting date of the project: 01/04/2020
Duration: 48 months

= Deliverable D3.2 =

Upgrade of compounding and formulation facilities: 50-300 µm PLA based composite film extruded with OTR < 100 cm³/(m²dbar)

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Executive Summary

Deliverable D3.2 presents the results of the upgrades of compounding and formulation facilities at IPC and FHG-IVV. It demonstrates the feasibility of the processing of bio-based plastics, namely PLA, into a film with an oxygen barrier suitable for packaging applications. The upgrades were performed on the METEOR® line at IPC and the high-shear mixing equipment of FHG-IVV for design of novel formulations and their applications as bio-based, bio-degradable materials.

IPC upgraded its METEOR® line with a film cast line, consisting of a flat die in association with a take-off station, to be able to produce films of 50 to 100 µm in thickness and 350 mm in width. This upgrade was validated by the successful production of PLA-based films, combining compounding and converting in a single step.

IVV upgraded the formulation equipment in order to produce PLA films with an oxygen transmission rate of below 100cm^3/(m^2*d*bar). It is shown in this deliverable that the used nanocomposite lacquer produced with the upgrades of high-shear mixing within Task 3.2 provides an OTR of below 0.01cm^3/(m^2*d*bar) when coated on the extruded PLA film.

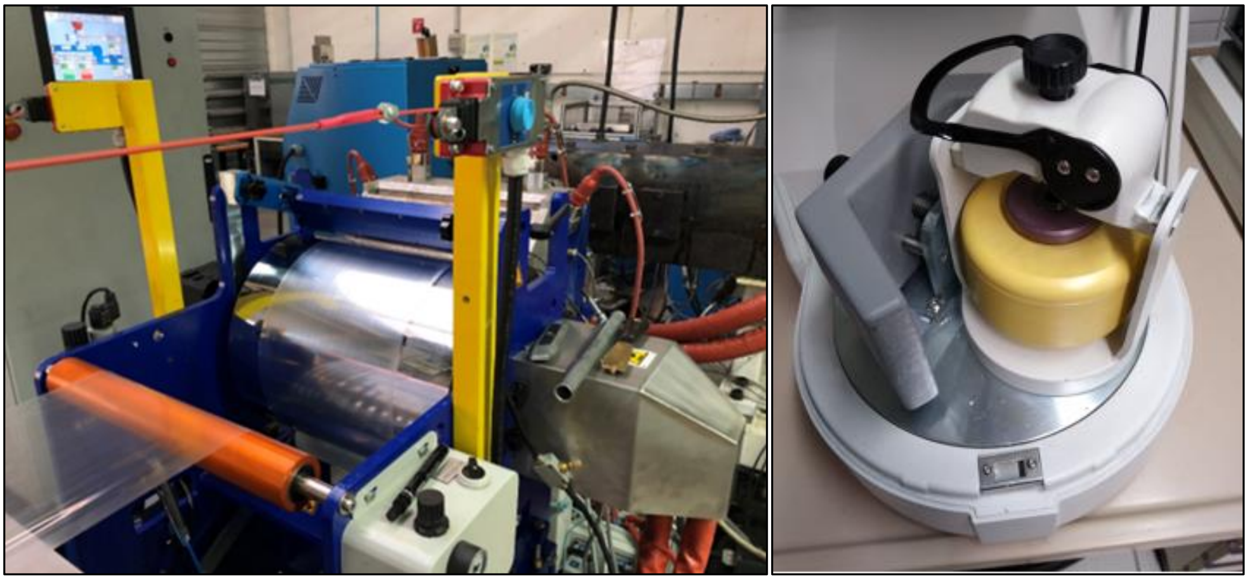

Figure 1: Upgraded lines: left: film cast-line associated to the METEOR line at IPC; right: high shear rate mixing at FHG-IVV

The results on the mechanical and thermal properties of the PLA films will be used in work package 5, use case 1, to give a bio-based alternative to Hueck Folien for their currently fossil-based film applications.

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1. Introduction

In order to support the targeted Open Innovation Test Bed (OITB) clients, Work package (WP) 3 aims at implementing holistic Circular Economy approaches serving the different client needs. This deliverable, therefore, describes the upgrade of the compounding and formulation facilities at IPC and IVV. The targeted upgrades of the compounding line at IPC and the high-shear mixing unit at the lacquering and film converting machines of IVV were installed and commissioned. The upgrades are demonstrated in this report by the production of a 50 – 300 µm thick PLA based film, which is coated to reach an oxygen transmission rate of less than 100 $cm^3/(m^2.d.bar)$.

This deliverable is linked to Task 3.2 of WP 3, Compounding and Formulation Facilities for Recyclable, bio-based and biodegradable materials. This task aims at optimising the METEOR® equipment of IPC in order to better control the process parameters and to produce bio-based polymer films of 50 to 100 µm thickness at 350 mm width. Bio-based polymers are often sensitive and tend to degrade at high temperatures during the extrusion process. So, the sensitivity of the polymers to higher temperatures requires an improved compounding process, a mixing with less sensitive polymers, or a mixing with cross-linkers to provide higher thermal stability. PLA is one of the mostly used bio-based polymers and it exhibits high oxygen and water vapour permeability. It, however, has low thermal and mechanical stability during its coating processing, which is why it is targeted in this deliverable.

Task 3.2 includes the optimization of homogeneous dispersion formulations. Fraunhofer-IVV achieved this goal by using high shear rate mixing for the preparation of nano-lacquer formulations, useable for the coating of PLA for improved oxygen barriers.

Within task 3.2 of FlexFunction2Sustain, which seeks to set up an OITB, the involved partners JOA, FHG-FEP, FHG-IVV and IPC have set up an experimental study to assist Hueck Folien in the development of a sustainable biodegradable optical film for security.

In this deliverable, the upgrades are described in Chapter 2, Section 2.1. Section 2.2 describes the PLA film production process both at IPC on the METEOR line and at FHG-IVV, for comparison purposes. The METEOR line allowed to perform the compounding and film-extrusion of the film in one single step. This one-step method is compared to the more classical sequence of compounding the PLA first at IPC and then performing the film-extrusion step at FHG-IVV. To obtain a film with a low oxygen permeability as defined above, the extruded PLA film is coated with a nanocomposite lacquer. For a homogeneous barrier layer, a specific mixing unit is required as also described in this section. Finally, Section 2.3 compares the results of mechanical and thermal characterisation of the films produced in the two above-mentioned ways and comprises the reduced oxygen permeability. The conclusions are presented in Chapter 3.

These upgrades will support potential OITB customers in the development of sustainable biodegradable plastic products such as for optical films for security or food packaging films.

2. Results and discussion

The extensional flow mixer METEOR® at IPC was upgraded both, to improve the dispersion of cross-linking additives in PLA and prepare films in a single step, along with compounding. IVV produced cast films from the different compounds delivered by IPC in a pellet form and sent these films to FHG-FEP for e-beam treatment. The overall goal is to develop a biodegradable film substrate with sufficiently high glass transition temperature and better imprint process stability. This could allow for instance Hueck Folien (HF), an industrial use case partner in the FlexFunction2Sustain project, to switch from fossil-based materials to more sustainable options. These trials, hereafter, aim at producing cast film at a thickness of 50 µm from the received PLA compounds and characterize the material thermally and mechanically.

Furthermore, the newly developed permeation barrier coating formulation for oxygen barrier enhancement has been applied on the extruded PLA and the Oxygen transmission rate was measured. Here the aim was to reach a sustainable barrier concept for packaging applications. Therefore, the high shear-mixing tool was upgraded for improved dispersion to allow for optimal coating of PLA films with the upgrade slot-die at IVV.

2.1. Upgrades for Compounding and Formulation Facilities

2.1.1. Upgrade 1- Compounding and extrusion by extensional flow mixer METEOR® (IPC)

The METEOR® Pilot line (patented by IPC) is based on an innovative extensional flow mixing technology used to develop specific material formulations with an efficient dispersion of additives while reducing the thermo-mechanical degradation of the material.

This pilot line is dedicated:

- to reach good dispersive and distributive mixing, the two key conditions for a successful plastics compounding;
- to reduce the shearing forces, compared to the traditional extrusion machines like the twin-screw extruder;
- to reduce the shear heating (also called the overheating of the polymer melt), which will avoid the thermal degradation of the polymer melt (for sensitive polymers like bio-based polymers);
- to reduce indeed the power consumption during the compounding process.

The used Continuous Extensional Flow Mixer METEOR® is based on:

- an original design of screw and barrel to provide a strong extensional strain and a continuous division and then recombination of the polymer melt flow: to reach a high quality of dispersive and distributive mixing;
- a modular screw and barrel design that offer wider flexibility for compounding and recycling;
- real-time supervision and exploring of the process data;

With this machine, working at high temperatures, up to 350°C, and at throughput rates from just a few kgs/h up to 100 kg/h are feasible.

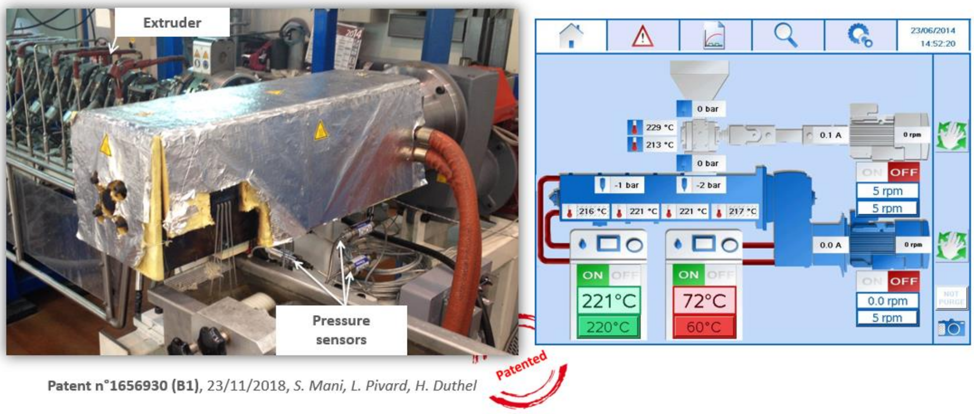


Figure 2: The Continuous Extensional Flow Mixer METEOR®

Moreover, IPC has upgraded its METEOR® Pilot line in order to:

(1) Develop specific material formulations with controlled morphology and efficient dispersion of polymer phases and/or additives for bio-degradable and polyolefin based polymers;

(2) Reducing, or avoiding, the overheating and indeed the thermomechanical degradation during the compounding, extrusion, and recycling process. For this, an inline rheometer has been selected, in order to control the melt flow evolution during processing trials;

(3) Implement co-extrusion, flat die, and film take-off station to produce films of 25 to 250 µm in thickness and 300-330mm in width. This equipment was installed and used to prepare the samples presented in this deliverable.

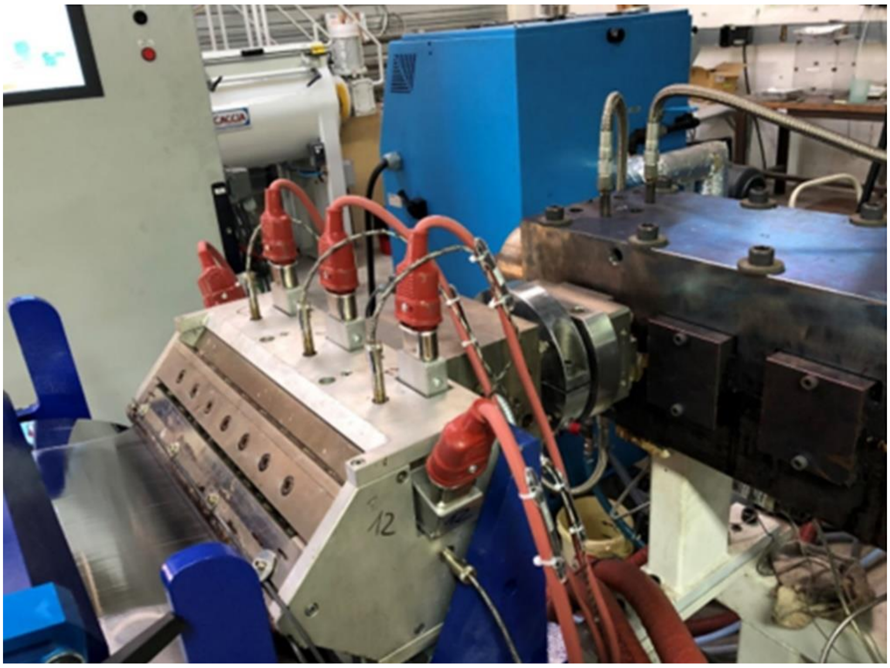


Figure 3: METEOR equipped with the flat die

Figure 4: Film take-off station in line with METEOR

2.1.2. Upgrade 2- Homogeneous dispersion by high shear mixing (FHG-IVV)

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 5. (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 (see Figure 5) pre-dispersion, 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 Deliverable 3.1, D3.1, and has been coated on the PLA films for the gas barrier enhancement, as described in Section 2.2.3.

2.2. PLA Film Production Processes

The experimental study addresses compounding and cast film extrusion for selected suitable PLA and additives to develop a biodegradable film substrate with higher glass transition (Tg) in order to improve process stability for further imprinting processes.

The extensional flow mixer METEOR® is used to improve the cross-linking additives dispersion and to prepare the compounded PLA for cast film extrusion, both at IPC and FHG-IVV. These films are further to be irradiated by e-Beam at FHG-FEP, and will be imprinted with hologram structures at JOA in the course of the project. The most promising formulations will be up-scaled for roll-to-roll coating.

The trials described hereinafter consist in preparing:

(4) Compounded PLA pellets, with and without METEOR®, a part of which being sent to FHG-IVV (5kg/sample)

(5) Compounded PLA converted into thin films, at IPC and FHG-IVV, which will be sent to FHG-FEP

2.2.1. Compounding of PLA with crosslinkers (IPC)

Upstream

- Feeder 1: Mono-screw feeder for matrix pellets, feeding zone 1.
- Feeder 2: Peristaltic pump for liquid additives, feeding zone 6, by the top.

Extruder

- Screw profile is designed to be highly shearing.
- Every zone is closed, except: zone 1 (feeding zone), zone 6 (additive introduction), and 10 (degassing zone).

METEOR© technology (for second phase)
The METEOR® pilot line was upgraded and equipped with a flat die, film take-off station, and tested for producing films of 50 to 100 µm in thickness and 300mm in width

Downstream
- Water cooling bath for filament extrusion die
- Standard pelletizer
- Flat die with film take-off station

Used materials
The matrix material used for these trials is PLA. Two kinds of additives: TAIC and TMPTMA were selected and compounded with the PLA. Further information is presented in the Table 1 below:

Table 1: Raw materials used for the preparation of samples

Role	Provider	Reference	Form	Drying step
Matrix	TOTAL CORBION	Luminy L175 (PLA L175)	Pellets	24h at 60°C or 4-6h at 80°C
Additives	Acros Organic	TAIC	Liquid	NO
	Startomer/Arkema	TMPTMA	Liquid	NO

Produced samples
As agreed previously, three combination of formulations (PLA L175, PLA L175+3%TMPTMA, PLA L175+3%TAIC) and two processes (Extruder, Extruder + METEOR) were used to prepare six different types of pellets. These samples formulation and processes are summed up in the Table 2 below:

Table 2: Samples formulation and parameters set

Trial	Sample Name	Weight fraction of PLA over additive w/w	Process. temp. T (°C)	Through-put (kg/h)	Extruder speed (rpm)	METEOR® speed (rpm)	Pellets quantities
1	PLA L175	100	170---190	10	200		5,5 kg
2	METEOR PLA L175	100	170---190	10	200	60	
3	PLA L175 + 3%TMPTMA	97/3	170---190	10	200		
4	METEOR PLA L175 + 3%TMPTMA	97/3	170---190	10	200	60	
5	PLA L175 + 3%TAIC	97/3	170---190	10	200		
6	METEOR PLA L175 + 3%TAIC	97/3	170---190	10	200	60	

All samples were successfully produced without the need to significantly modify the process parameters. The residence time was estimated at about 45-60 seconds for the pellets production with the extruder alone. The residence time within METEOR equipped with the flat die was estimated about 10 min in these conditions.

Temperatures recorded at the exit of METEOR® seem to indicate that no self-heating has been induced, as summed up in Table 3. During processing, the addition of TMPTMA and TAIC did not lead to significat impact on the melt's processability or apparent viscosity.

Extruder's motor intensity, which gives indications about the torque value, was recorded generally at around 29% and decreased slightly to around 26% with TAIC introduction, which could indicate a minor decrease of viscosity. For all samples, melt pressure was within 27 to 37 bars, regardless of their formulation, but melt pressure seemed to be lower at the exit of METEOR® (Table 3).

Melt temperatures showed an overheating of PLA-based formulations during the extrusion process without METEOR, up to 212°C for PLA alone, when it was measured with an external probe. The measured melt temperature at the die output was slightly lower for PLA with additives, when measured with an external probe, which could indicate they act as plasticizers. However, additives introduction generated smoke, in the degassing zone.

Table 3: Processing observations during the compounding step at IPC

Sample name	T_{melt}[°C] (external probe)	T_{melt} [°C] (internalsensor)	$P_{at\ die}$ [bar]	Motor intensity [%]
PLA L175	212	195	37	28
METEOR PLA L175	-	174	27	29
PLA L175 + 3% TMPTMA	205	196	37	28
METEOR PLA L175 + 3% TMPTMA	-	175	31	29
PLA L175 + 3% TAIC	209	198	37	27
METEOR PLA L175 + 3% TAIC	-	177	31	26

2.2.2. PLA Film Extrusion (FHG-IVV, IPC)

The pellets prepared as described in the previous part were converted in films both at IPC in one single compounding-extrusion step and at FHG-IVV.

2.2.2.1. Collin cast line (FHG-IVV)

For the film production with the different types of PLA at FHG-IVV, a 7-layer cast extrusion line from Dr. Collin is used. The extruder line consists of four single screw extruders, which can be used either individually or connected together for coextruded multilayer films. The screw of the main extruder has a diameter of 30mm and a length of 900mm (L/D = 30). This extruder serves for the production of the PLA monolayer cast film in the following trials. A chill-roll/calendar-unit and a winder unit follow the extruder as depicted in Figure 6. The installed flat die width measures 300mm, which results in a maximum film width of 250mm, and has an adjustable range of film thickness from 20µm to 1,5mm, depending on the material. A capacitive sensor tracks this film thickness. Before the extrusion, a pre-drying of the PLA at 60°C through the night was performed as recommended by IPC.

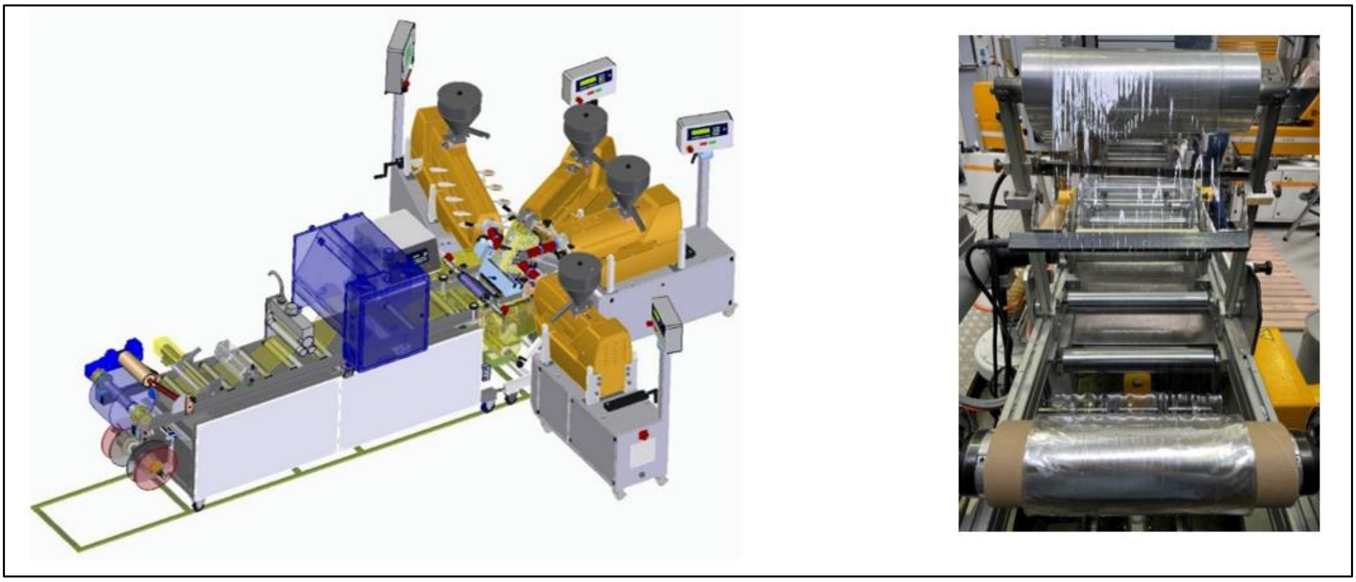


Figure 6. Scheme of the lab Collin-line (left) and picture from the winding unit (right)

2.2.2.2. IPC Cast line

The films are prepared on a cast line (Scamex) composed of a heated flat die and a film take-off station. The flat die's width is 350 mm and its lips can be adjusted to prepare films with a thickness from 25 to 250 µm. Several heating zones ensure that the matter is properly melted, when passing through the die. First, a co-extrusion block covers zone 1 to 4, leading the matter from the extruder (or METEOR) to the flat die.

Then, the flat die itself includes several heating elements positioned as following: 3 heating zones on the top, 3 heating zones on the bottom, 1 on each side, 1 for the upper lip and 1 for the lower one.

The film exiting the flat die is then brought to the film take-off station, first in contact with the chill roll, of which the temperature can be adapted from 20 to 160°C, up until the end of the line where it is winded. The speed of this equipment ranges from 0.1 to 20 m/min.

Figure 7: Film take-off station (IPC)

This whole cast line can be equipped directly at the exit of the extruder or of the METEOR technology, to allow both the compounding and converting to occur in a single step.

2.2.2.3. Extrusion and Observations

The equipment described was above was used for the production of 50µm thick PLA films at a width of 250mm (IVV) and 350mm (IPC).

Cast film extrusion at IVV
The first temperature profile is shown in Table 4 for the first extrusion (PLA L175). However, the pellets fed into the extruder showed different sizes and agglomerations (see Figure 8), which made it more difficult to feed them continuously. It was necessary to push in the pellets manually from time to time. However, a smooth film production was feasible at the conditions of row 2 given in Table 4.

Table 4: Temperature profiles in the extruder for PLA extrusion

Zone	Infeed	Zone 1	Zone 2	Zone 3	Zone 4	Zone 5	Adapt.
PLA L175	30	180	200	200	195	190	190
METEOR PLA L175 & others	30	160	175	195	205	205	205

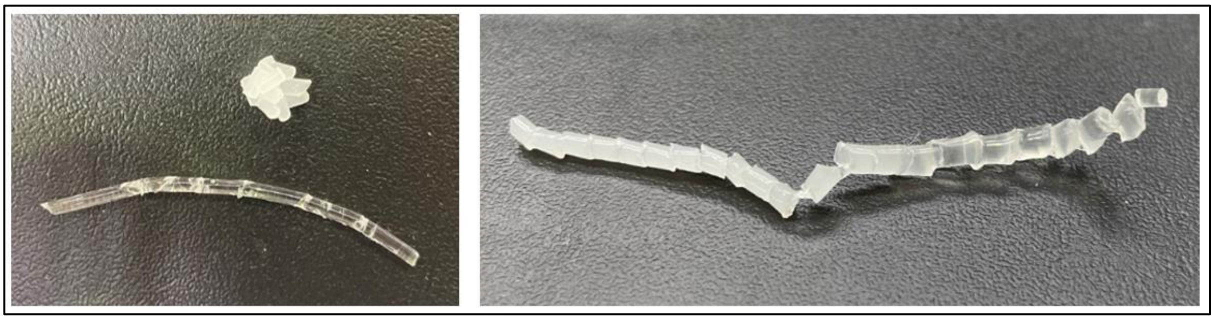


Figure 8. Agglomerations of pellets found in the material bags (METOR® PLA L175)

Overall, the production of the film from the METEOR® PLA L175 was less stable with and without cross-linking additives. On the one hand, the higher MFI of the materials from the METEOR® process required a higher screw speed (set from 25rpm to 48rpm (see Table 5)) and pull-off speed, which was adjusted from 3,3m/min to 6,3m/min. Moreover, the chill roll temperature was decreased from 60°C to 50°C to avoid sticking of the film to the chill roll. On the other hand, at current temperatures, many gels occurred in the film. As a measure, the temperature was increased, but at 210°C, the melt started pulsing at the die what led to an uneven thickness distribution. Therefore, a middle temperature of 205°C was set for the further production of the films (ref. Table 4). Figure 9 shows a comparison of the PLA L175 and the METEOR® PLA L175. The difference in the amount of gels is clearly visible.

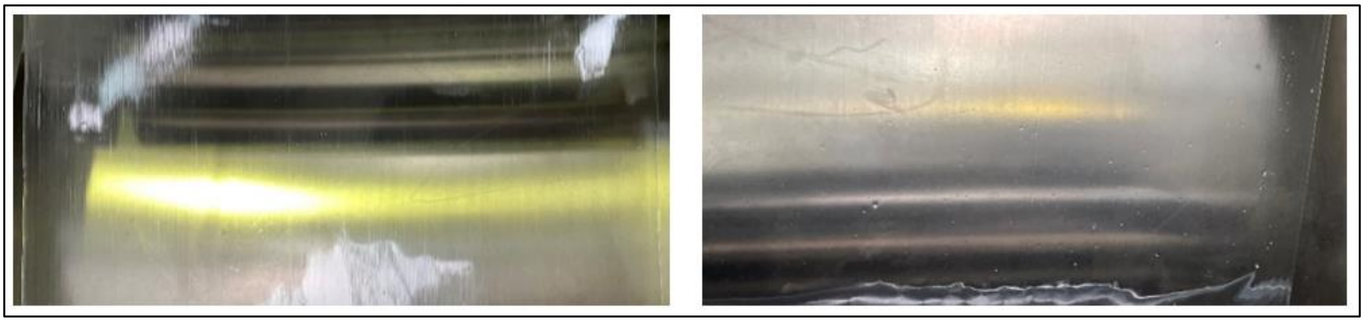


Figure 9. PLA L175 film (left) and METEOR® PLA L175 film (right) during the extrusion process

The following Table 5 sums up all the process parameters observed during the extrusion of the different films at FHG-IVV.

Table 5: Adjusted parameter sets per material

Sample name	MFI [g/10min]	T_{melt} [°C]	P at die [bar]	Screw [rpm]	$T_{Chillroll}$ [°C]	T_{mass} [°C]	I_{motor} [A]
PLA L 175	6,85	176	56	**25**	60	201	4,4
METEOR PLA L 175	10,745	176	55	**48**	50	216	7,5
PLA L175 + 3% TMPTMA	5,3	174	68	48	50	214	7,5
METEOR PLA L175 + 3% TMPTMA	18,7	176	32	48	50	209	3,3
PLA L175 + 3% TAIC	6,81	173	68	48	50	215	7,4
METEOR PLA L17 + 3% TAIC	11,44	174	37	48	50	215	6,3

After the last extrusion of METEOR® PLA L175 + 3% TMPTMA, a liquid was found on the chill roll next to the film (see Figure 10).

Figure 10. METEOR® PLA L175 on chill roll, liquid marked in red circle

The following four points sum up the key observations:

- Increase of the screw speed for a more stable process was necessary, especially for the METEOR® films.
- 60°C on the chill roll let to a sticking on the roll, which is why the temperature was decreased to 50°C.
- Overall, the METEOR® material ran less stable, with more pulsing at the die.
- An unidentified liquid was found on the chill roll during the extrusion of METEOR® PLA L175 + 3% TMPTMA.

However, six different cast films could be produced (ref. Figure 11 below).

Figure 11: PLA-based film rolls produced at IVV

In Figure 12, METEOR® PLA L175 + 3% TMPTMA and the PLA L175 + 3% TMPTMA films produced at IPC and IVV respectively are shown next to each other against daylight, to make the difference in film quality better visible. More pictures can be found in

Appendix I – Pictures of the produced films). The films made from the METEOR® pellets display more gels, and the films from IPC display less gels overall.

Figure 12. Cast films produced at FHG-IVV (PLA + TMPTMA crosslinker)

Cast film extrusion at IPC
All samples were processed at a temperature of 170 to 190°C, with a throughput of 10kg/h, and the extruder screw speed set at 200rpm. METEOR technology was combined to extruder for the corresponding samples, with a processing temperature of 183 to 185°C, leading to a temperature of 175°C at the exit for all samples. Regarding the cast line, the slot die temperature was set to 170 to 180°C, and the lips gap to 700µm. The chill roll temperature was 60°C, and the chill and winder rolls' speeds were adjusted depending on the samples, aiming a thickness of 50µm, with a width of 250-260mm.

The following table sums up all process parameters observed during the extrusion of the different films at IPC.

Table 6: Adjusted parameter sets per material (IPC)

Sample name	T_{melt} (internal sensor) [°C]	$P_{at\ die}$ [bar]	Screw [rpm]	$T_{Chillroll}$ [°C]	Motor intensity (%)
PLA L 175	190	24	200	60	30
METEOR PLA L 175	175	30	200	60	28
PLA L175 + 3% TMPTMA	190	24	200	60	30
METEOR PLA L175 + 3% TMPTMA	175	31	200	60	29
PLA L175 + 3% TAIC	Not measured	36	200	60	25
METEOR PLA L17 + 3% TAIC	175	30	200	60	26

All samples were successfully produced, without the need to significantly modify the process parameters. For the flat die extrusion, the residence time with the extruder alone was estimated at about 90-170 seconds, based on the first sighting of the melt and the time it took to fill entirely the flat die. The residence time within METEOR equipped with the flat die was estimated about 10 min in these conditions.

Temperatures recorded at the exit of METEOR seem to indicate that no self-heating has been induced. During processing, the addition of TMPTMA and TAIC did not lead to significat impact on the melt's processability or apparent viscosity.

Extruder's motor intensity, which gives indications about the torque value, was recorded generally at around 29% and decreased slightly to around 26% with TAIC introduction, which could indicate a minor decrease of viscosity. For all samples, melt pressure was within 24 to 36 bars, regardless of their formulation, the pressure was generally higher for samples processed with METEOR.

As for the pellets processing, the melt temperatures showed an overheating of PLA-based formulations during the extrusion process without METEOR. However, it was not possible to measure the melt temperature with an external probe at the exit of the flat die.

Additives introduction generated smoke, in the degassing zone and directly emanating from the films at the flat die output.

It was observed that the film borders could not be cut by a knife, due to its deformation in contact with the blade. In addition, when cut trials were performed, the film breaks along its entire width like a brittle material. Nevertheless, the quality in the middle of the films is good.

Apart from this sample, no gels were observed in the samples, neither at the flat die output nor on the obtained rolls. Samples of PLA with additives did not exhibit any exudation, or oil condensation at the films' surface, or oil inclusions within the films.

2.2.3. Slot-die coating of PLA film for oxygen barrier

To create an oxygen barrier on the extruded PLA-based film, the film is coated with the nanocomposite lacquer, which is described in Deliverable 3.1 "Upgrades of Circularity by design facilities". This nanocomposite lacquer contains ethylene vinyl alcohol (EVOH) copolymer and montmorillonite (MMT) nanoparticles in a mixing ratio of 1:1 by weight. The total solid content of the lacquer is 6 wt%.

The lacquer is coated via a slot-die coating method, which is being developed as part of the upgrades performed in WP2, Task 2.2. **Error! Reference source not found.** shows the different steps from coating l iquid reservoir (1) to coated film (4). The alternating syringe pump (2) pumps the coating liquid through the hoses. Before filling the slot die, there is an option to filter (3) 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 (4).

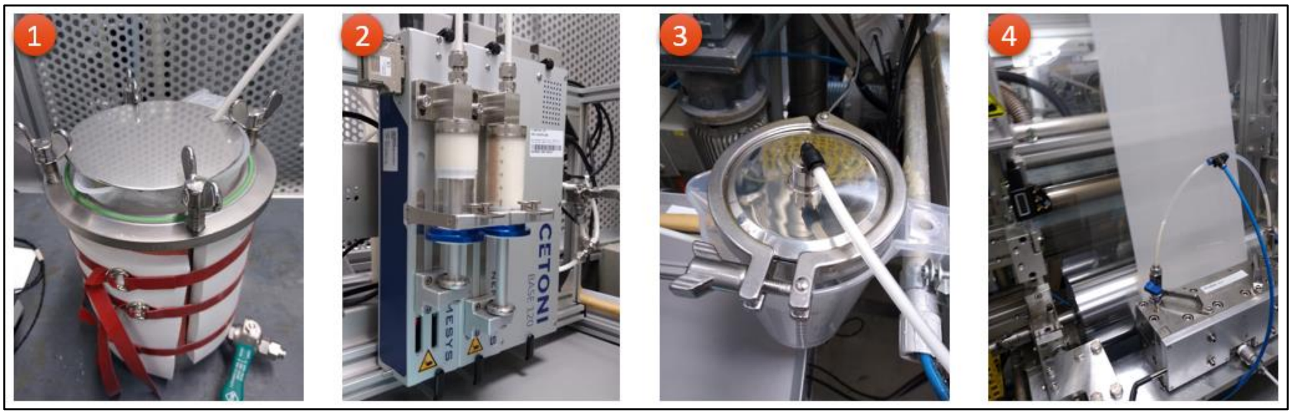


Figure 13. (1) Lacquer reservoir (2) alternating syringe pump (3) filter option (4) coating of lacquer

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. The coating parameters are shown in Table 7.

Table 7: Parameter set for the slot-die coating process of nanocomposite lacquer on PLA at the R2R machine at Fraunhofer IVV

Gap width	µm	300	**Web speed**	m/min	2.5
Slot length	mm	45	**Volume flow**	ml/min	16.67
Slot width	mm	100	**Shear rate**	1/s	62
Solid content	%	6	**viscosity**	Pas	0.05

Dry layer thickness	µm	4	Pressure loss	bar	0.028
Wet layer thickness	µm	66.67	Distance slot-die to web	µm	150
Temperature pressure tank	°C	50	Temperature slot-die	°C	40
Curing temperature	°C	55	Curing time	s	100
Unwinding	N	30	Controlled winding	N	30

The gap width, the slot length and the slot width Wslot 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, ddry is typically the parameter to set. By knowing the solid content of the lacquer, c and the web-speed, vweb 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.

2.3. Characterisation results

All produced films from IPC and IVV were characterised. The tensile properties were measured according to the ISO 527-3 standards on 50µm thick film test specimen. Oxygen permeation was determined according to DIN 53380 – pt.3 (ISO 15105-2).

2.3.1. Mechanical properties

Figure 14 shows the mechanical properties, such as the tensile strength and elongation at break of the films produced at FHG-IVV in comparison to the films produced at IPC. No significant differences in tensile strength are visible for most of the films, except for the METEOR® PLA L175 + 3% TMPTMA. The elongation at break does not differ significantly neither, although for the METEOR® PLA L175 and PLA L175 + 3% TMPTMA the difference is found to be higher.

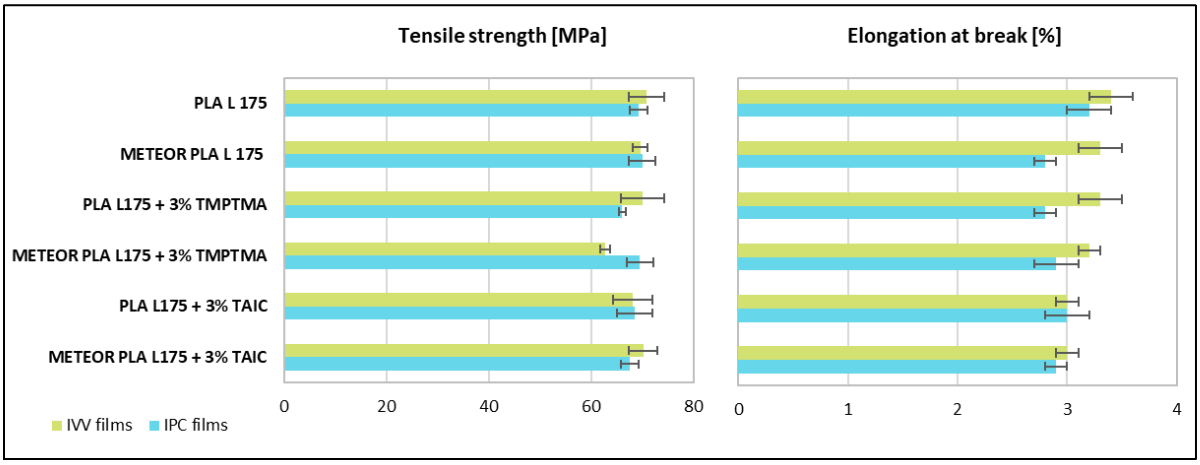


Figure 14: Tensile strength (left) and elongation at break (right) of films from FHG-IVV in comparison to the films from IPC, both measured in MD and normalised taking in account the films thicknesses

These results do not indicate any significant property difference between the films from the two different extrusions. Figure 15 depicts the modulus of the films. The measured values are lower and the standard deviations become higher for the IVV produced films when the PLA L175 and PLA L175 + TMPTMA pellets are from the METEOR® line.

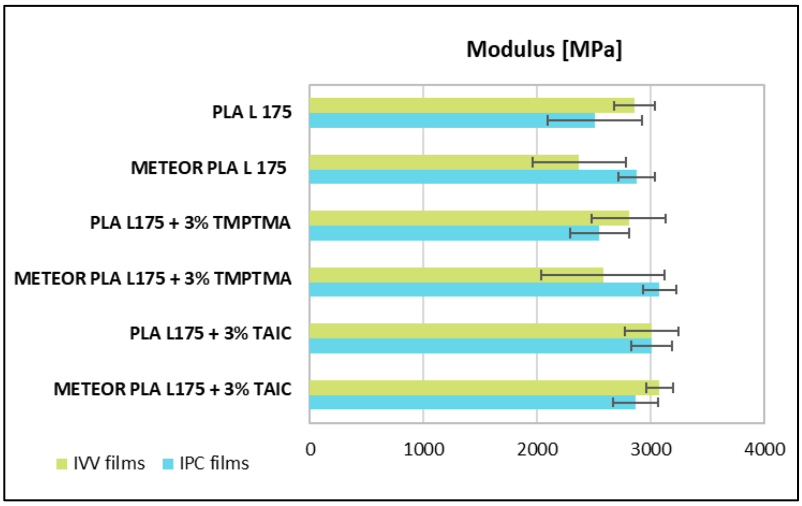


Figure 15: Modulus of films produced at FHG-IVV and IPC, measured in MD and normalised

2.3.2. Thermal properties

The compounded PLA arrived from IPC at FHG-IVV in form of pellets. As preparation for the extrusion trials, the melt flow index (MFI) and the melt temperature (Tm) have been determined, by a melt flow indexer and Differential Scanning Calorimetry (DSC), respectively. Table 8 shows the mean values of two MFI (and MVR) measurements for all investigated PLA types. It can be observed that the material produced with the METEOR® process has a significantly higher MFI and, hence, a lower viscosity than the material which has not gone through the METEOR® process.

Table 8: Melt flow index and melt volume rate measured at 190°C with 2.16 kg

Description	**MFI** [g/10min]	**MVR** [cm³/10min]
PLA L 175	6.85	6.07
METEOR PLA L 175	10.745	9.65
PLA L175 + 3% TMPTMA	5.3	4.8
METEOR PLA L175 + 3% TMPTMA	18.7	16.75
PLA L175 + 3% TAIC	6.81	6.25
METEOR PLA L17 + 3% TAIC	11.44	10.2

By the DSC measurement, melt temperature Tm and glass transition temperature Tg are determined. The results are shown in Table 9. No significant difference stands out when comparing the temperatures of the materials.

Table 9: Melt temperature, Tm and glass transition temperature, Tg resulting from DSC

Description	T_m [°C]	T_g [°C]
PLA L 175	176	61
METEOR PLA L 175	176	61
PLA L175 + 3% TMPTMA	174	57
METEOR PLA L175 + 3% TMPTMA	176	57
PLA L175 + 3% TAIC	173	58
METEOR PLA L17 + 3% TAIC	174	60

2.3.3. Oxygen permeability

The aim of the upgrades of the shear mixer and the slot-die coating[1] at FHG-IVV are an improvement of the oxygen permeability of the bio-based plastic films. The targeted OTR to reach is below 100 cm3/(m2*d*bar) for PLA layer thicknesses of 50-300 µm. The following table sums up the oxygen transmission rates of the 50µm PLA L175 film with and without 4µm barrier-lacquer produced on the upgraded ball-mill and coated via the slot die as described in Section 2.2.3. The barrier improvement by the nanocomposite lacquer is clearly visible in the values given below in Table 10.

- From these results of 50µm films it can be derived that the oxygen transmission rate of 200µm thick films would range between 70 and 80 $cm^3/(m^2*d*bar)$, which would already be within the targeted range.
- The coating of the nanocomposite at a dry layer thickness of only 4 µm significantly improves the oxygen barrier properties of the PLA films by a factor of 4000.

Table 10: Oxygen transmission rate of 50µm films from IVV and IPC and one coated IVV film

Description	Thickness (µm)	IVV film	IPC film
		OTR [$cm^3/(m^2*d*bar)$]	OTR [$cm^3/(m^2*d*bar)$]
PLA L 175	50	336	287
PLA L 175 coated with 4µm lacquer	54	0,084	

[1] Upgrades still ongoing in work package 2

3. Conclusions

In this deliverable it could be shown that with the upgrade of the METEOR® line at IPC, it was possible to produce six different PLA-based films at IVV and IPC each. The installation of a film cast line directly associated with METEOR now enables to perform compounding and converting of thermally sensitive material in a same step. It is now possible to produce films of 25 to 250 µm in thickness and 300-330mm in width, directly after the optimized distribution of additives by METEOR. With the upgraded high-shear mixing of the nanocomposite lacquer at FHG-IVV, it was moreover possible to show a significant improvement of the oxygen permeability of the PLA film by coating, significantly below the targeted value of 100 $cm^3/(m^2*d*bar)$.

The key findings can be summarised as following:

- Extrusion was possible, METEOR® lead to a less stable cast film extrusion process at FHG-IVV extrusion, however the film production directly on the METEOR® line at IPC did work without problems
- Mechanical properties do not vary significantly between the films produced at IPC in one-step and the ones produced in IVV in two-steps.
- However, the gel amount of the films from IPC seems lower as seen on Figure 16.

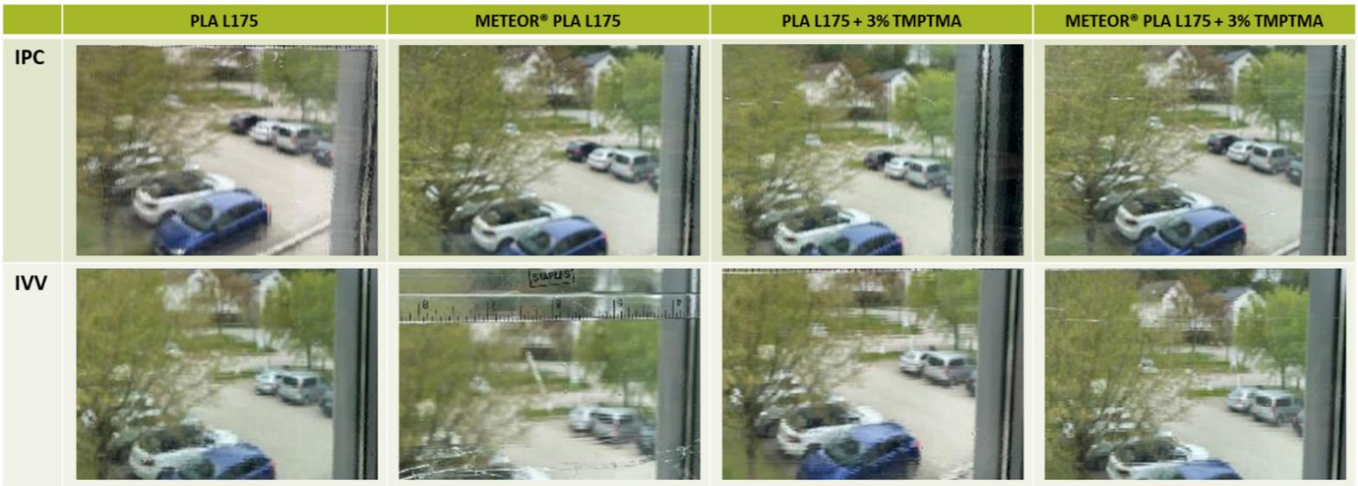


Figure 16: PLA-films obtained both, at IPC and FHG-IVV, comparing the impact of METEOR

The films produced at IPC as well as the films produced at FHG-IVV were sent to FHG-FEP for e-beam treatment. As a next step, IVV will address the characterization of these treated films and compare the results to the initial non-treated film properties. The findings and demonstrations of this deliverable will be implemented in the work package 5 use case of Hueck Folien.

4. Degree of progress

The task activities were successfully fulfilled for IPC's METEOR upgrade and IVV's high-shear mixing and slot-die coating upgrades. The extrusion of PLA was successfully demonstrated as well as the deposition of a EVOH based nanolayer that reduced the oxygen transmission rate to < 0,1 $cm^3/(m^2d\ bar)$. Deliverable 3.2 therefore is completed by 100%.

5. Dissemination level

This Deliverable is public.

6. Appendix I – Pictures of the produced films

Table 11: Cast films produced at IPC and IVV (PLA)

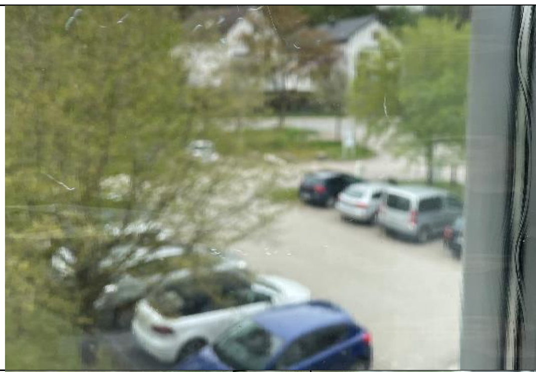
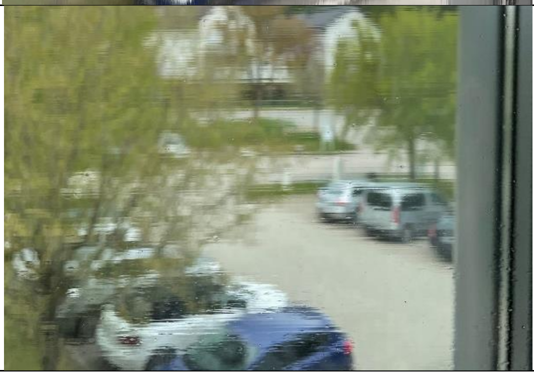
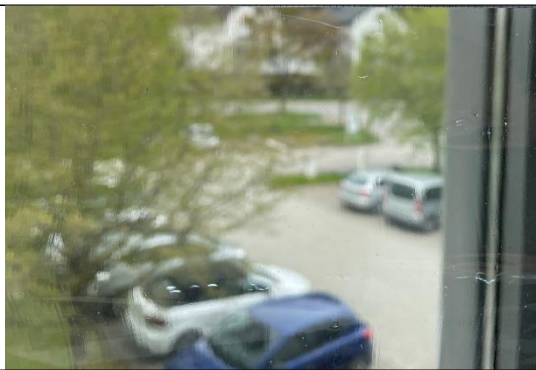
	PLA L175	METEOR® PLA L175
IPC		
IVV		

Table 12: Cast films produced at IPC and IVV (PLA + TAIC)

	PLA L175 + 3% TAIC	METEOR® PLA L175 + 3% TAIC
IPC		
IVV