WOOD2WOOD

A Wood-to-Wood Cascade Upcycling Valorisation Approach

» Deliverable 7.2

New mix between wood, glass and composites for wallboards

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GLOSSARY OF ACRONYMS

Acronym	Extended Definition
ATR-FTIR	Attenuated Total Reflect Fourier Transform Infrared Spectroscopy
CI	Carbonyl Index
CNF	Cellulosic nanofibers
DSC	Differential Scanning Calorimetry
DSV	Dilute Solution Viscometry
GPC	Gel Permeation Chromatography
LCNF	Lignocellulosic nanofibers
MFR	Melt flow rate
MW	Molecular weight
OOT	Oxidative Onset Temperature
PLA	Poly(lactic acid)
SEM	Scanning Electron Microscopy
TGA	Thermogravimetric analysis
$T_{d5\%}$	Degradation temperature of 5% mass loss
T_d	Maximum degradation temperature
UV	Ultra-violet
W2W	Wood2Wood

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EXECUTIVE SUMMARY

The Deliverable 7.2. includes the developments regarding Task 7.2: New mix between wood, glass and composites for wallboards (M08-M18). In the early steps of this task, the NTUA team investigated the PLA market by assessing various commercially available PLA grades, based on a comparative analysis of their datasheets. Ingeo 4043D from NatureWorks was selected, since it was pointed out as suitable for 3D-printing, cost-effective and readily available. Following, neat PLA twin-screw extrusion conditions in terms of temperature profile, feeding rate and screw rotation speed were identified, so as to ensure low thermomechanical degradation extent of the biobased matrix. Once concluding on the PLA extrusion parameters, a first batch of lignocellulosic materials extracted from eucalyptus tree was received from UHE, and the biocomposite compounds production of 1 wt.% filler concentration was initiated. All biocomposite compounds were characterized with a series of techniques, in order to assess filler dispersion, thermal stability and extent of thermomechanical degradation of the matrix, as well as, mechanical performance. The following methods were conducted: ATR-FTIR, DSC/TGA-OOT, DSV, MFR, SEM, tensile testing and density measurements. Once the second batch of lignocellulosic materials from W2W wood waste was received from UHE, the aforementioned steps were repeated with a higher concentration of 5 wt.% fillers in the PLA matrix. The biocomposite compounds were successfully prepared with the objective of identifying one or two optimal formulations to serve as the base material for the next development phase. In this stage, a suitable additivation strategy was also considered, to produce hydrolytically stabilized PLA-based materials, suitable for long-term applications, such as in the construction sector. To this end, a literature-based review has been performed on commercially available anti-hydrolysis additives. In parallel, a preliminary study using UV-accelerated ageing was also conducted on neat PLA and the developed biocomposite compounds of 5 wt.% fillers: degradation was primarily evaluated through the monitoring of mechanical properties, with a focus on 3-point flexural bending tests. Further investigation will be carried out on more PLA formulations for delivering the optimal biocomposite compound. Finally, a processability study was also conducted by the NTUA team, using a heated plates hydraulic press, to determine the most appropriate set of compression molding parameters (temperature, heating time, cooling time and pressure) for PLA plaques production. These parameters will guide larger-scale manufacturing by BLOOM, ahead of the final transformation and application testing to be carried out by LEVERY in Task 7.3.

1. INTRODUCTION

In 2023, over 413 Mt of plastic materials were generated, 90% of which were fossil-based [1]. The extensive use of fossil-based plastics has raised concerns regarding their lack of biodegradability and biocompatibility, along with the depletion of oil reserves. Concurrently, there is an increasing demand for renewable and biodegradable materials for single-use plastics and longer life cycle applications. Furthermore, efforts have focused on the search for sustainable fillers to replace traditional minerals and petrochemicals. Within this context, biocomposite materials consisting of a biodegradable polymeric matrix and reinforced with natural fillers, have emerged as a strong prospect for reducing plastic waste and utilizing untapped resources such as wood waste [2-3].

Several biobased and compostable polyesters have been studied as matrices for biocomposites, including poly(lactic acid)(PLA), poly(butylene succinate)(PBS) and polyhydroxybutyrate (PHB) [2-9]. PLA specifically, has gained significant attention due to its biobased origin, biocompatibility and biodegradability. Furthermore, PLA exhibits rigidity, excellent optical transparency, and good barrier properties (low oxygen permeability) comparable to LDPE [2,4]. However, its brittleness, slow crystallization rate and low thermal resistance can limit its potential applications, especially in demanding fields such as 3D-printing [2,3]. In this context, the incorporation of fillers can be beneficial in increasing ductility, providing nucleation sites to aid crystallization, and improving thermal properties.

Glass fibre (GF) is widely accepted as reinforcing agent in polymer matrices to improve mechanical strength (tensile strength, impact resistance). However, due to its brittle nature, the incorporation of GF can lead to reduced ductility and toughness, especially when the interfacial bonding between the fibres and the matrix is not good [10]. Even though the existing literature is limited, there have also been a few efforts to incorporate glass waste or recycled glass (in the form of fibres or powder) in thermoplastic matrices such as PET and PLA. Generally, an increase in thermal stability has been reported, and a decrease in tensile strain as the material becomes more brittle and rigid [11-12]. It is important to note here, that compatibilization issues between glass fibre and the matrix are frequently encountered [10].

Lignocellulosic materials (primarily composed of cellulose, hemicellulose, and lignin) are often used to reinforce these biodegradable matrices, as biobased and biodegradable fillers. These materials are renewable, lightweight, inexpensive, and frequently originate from plant biomass such as agricultural residues (wood flour, straw residues), wood waste from construction and demolition, and paper waste (e.g. old newspapers) [2]. Incorporating lignocellulosic materials into a polymeric matrix can reduce the manufacturing cost of biocomposites while also achieving environmental and sustainability targets. Furthermore, lignin and lignocellulosic nanofibers have been found to enhance the mechanical properties of PLA and PHB matrices (improved durability, toughness, tensile strength, Young's Modulus) due to the formation of strong intermolecular bonds between aromatic rings of lignin with PLA and PHB [2-3,6,8-9]. In addition, PLA/lignin, PHB/lignin, and PBS/lignin composites often demonstrate excellent UV-shielding and antioxidant properties, which can hinder their oxidative degradation over time [2,6-9,13]. This combination of properties renders biocomposites suitable for a broad range of applications, such as food packaging, additive manufacturing, construction, and outdoor applications.

Despite their potential and desirable properties, biocomposites face significant challenges. Due to the poor interfacial compatibility between the hydrophilic lignocellulosic fillers and the hydrophobic matrices, it is challenging to obtain composites with high lignocellulosic content (>10%) without compromising mechanical properties [2,4,14]. This obstacle can be addressed by reducing lignocellulosic materials to the nanoscale (nanospheres or nanofibers) to improve dispersion, adhesion, and compatibility. Another route could be the addition of plasticizers (like polyethylene glycol (PEG) or glycerol) and coupling agents/ compatibilizers (based on maleic anhydride) [2].

Additionally, the hydrophilic nature of lignocellulose renders biocomposites more sensitive to hydrolytic degradation, potentially limiting their use in long-term applications [2]. As PLA is inherently prone to hydrolysis, several anti-hydrolysis agents (epoxy copolymers, isocyanates, carbodiimides (CDI), and aliphatic and aromatic polycarbodiimides (PCDI)) have already been studied for their stabilizing properties (Table 1. Commercially available anti-hydrolysis agents.**Table 1**) [15-17]. Anti-hydrolysis agents function through various mechanisms. Chain extenders re-connect the degraded polymer chains and minimize molecular weight decrease. On the other hand, carbodiimides and polycarbodiimides specifically react with the free carboxylic end groups and/or water molecules and forming urea derivatives, therefore both shielding the material from hydrolysis and binding and deactivating water molecules (**Figure 1**) [17]. In previous literature, the addition of aromatic carbodiimide in PLA/wood flour composites has proven to stabilize effectively the biocomposite against hydrolytic degradation [18].

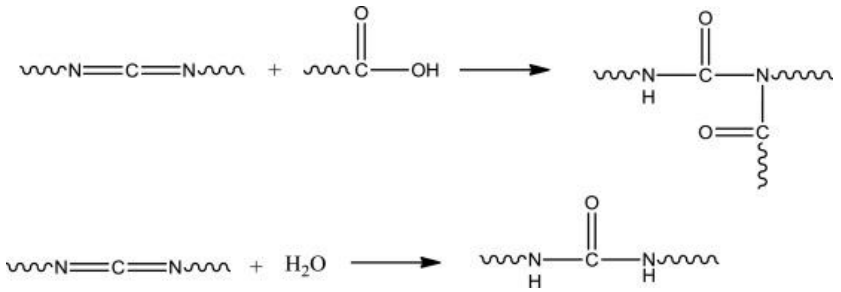
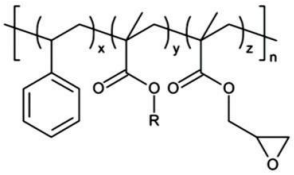
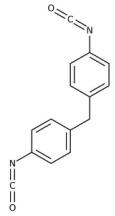
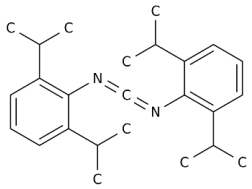


Figure 1. Anti-hydrolysis function of carbodiimides [17].

Table 1. Commercially available anti-hydrolysis agents.

Commercial name	Chemical structure	Chemical Name	Description - action
Joncryl® ADR 4468		Epoxy-containing oligomeric copolymer based on glycidyl ethacrylate, styrene and other acrylates	Epoxy-based chain extender, able to modify and control the intrinsic viscosity of the polyester plastics to restore and improve mechanical properties
		4,4'-Methylenebis(phenyl isocyanate) (MDI)	Aromatic diisocyanate chain extender mainly used in polyurethane synthesis.
BioAdimide® 100		Bis(2,6-diisopropylphenyl) carbodiimide	Aromatic carbodiimide which prevents polymer degradation due to exposure to humidity and acids
Stabaxol® P110		Oligomer of 2,6-diisopropylphenyl carbodiimide	Polymeric carbodiimide which acts as an anti-hydrolysis agent for polyester polymers (PET, PBT, TPE), polylactide (PLA) and polyamides (PA).
Carbodilite HMV-15CA		poly(4,4-dicyclohexylmethane carbodiimide)	Polymeric carbodiimide which improves anti-hydrolysis performance for various types of polyester resins including biobased polyesters

Another popular approach to address both compatibilization and hydrolytic degradation, is modifying the lignin segments to obtain products with different surface properties than lignin macromolecules, reduced hydrophilicity and improved thermal stability. The chemical modification of lignin includes reactions of the hydroxyl groups present in lignin and the addition of new chemically active sites to its structure [19]. One prominent modification technique is the silanization process during which silicon-containing moieties replace with the hydroxyl groups of lignin. Consequently, silanized lignin is less polar and more hydrophobic, and therefore more compatible with hydrophobic matrices, such as PLA. Finally, silanization can enhance thermal stability of lignin (higher $T_{d,5\%}$) [20].

The present work investigates the formulation of high-performance, biodegradable PLA-based biocomposites reinforced with chemically modified lignocellulosic fillers. The primary objective is to simultaneously overcome the inherent interfacial incompatibility and hydrolytic instability associated with high lignocellulosic filler content in PLA matrices. The final goal is to offer a novel, sustainable materials strategy for producing durable biocomposites suitable for moisture-sensitive and long-service-life applications, particularly in the construction and infrastructure sectors.

2. SELECTION OF PLA GRADE AND COMMERCIAL ANTI-HYDROLYSIS AGENTS

Starting T7.2, the NTUA team researched the PLA market, in order to identify different commercially available PLA grades which can serve within the scope of W2W, as a biobased thermoplastic matrix for biocomposites manufacture. Based on the datasheets, the main properties were registered for each of the materials in **Table 2**, e.g. optical purity, crystallinity, glass transition temperature (T_g), melting temperature (T_m) and the melt flow rate (MFR), while all PLA grades characteristics can be found in **Appendix 1**.

Table 2. PLA grades available in the market and main characteristic properties.

Trademark	Company	Grade	Optical purity	Crystallinity	*Tg* (°C)	*Tm* (°C)	MFR (g/10 min)
Ingeo 2003D	NatureWorks	Extrusion	4.5 % D	semi-crystalline	58	140 - 155	6, 210 °C, 2.16 kg
Ingeo 2500HP			0.4 % D	amorphous/semi-crystalline	55-60	165-180	8, 210 °C, 2.16 kg
Ingeo 4032D			1.6 % D	semi-crystalline	55-60	155-170	7, 210 °C, 2.16 kg
Ingeo 4043D			**4.3 % D**	**semi-crystalline**	**55-60**	**145-160**	**6, 210 °C, 2.16 kg**
Ingeo 4060D			12 % D	amorphous	55-60	150-180	n.d.
Ingeo 6302D			10 % D	amorphous	55-60	125-135	15-20, 210 °C, 2.16 kg
Ingeo 6752D			Mid D	semi-crystalline	55-60	145-160	15, 210 °C, 2.16 kg
Ingeo 3001D		Injection	1.5 % D	semi-crystalline	55-60	150-180	22, 210 °C, 2.16 kg
Ingeo 3100HP			<2 % D	amorphous/semi-crystalline	55-60	165-180	24, 210 °C, 2.16 kg
Luminy L175	Total-Corbion	Extrusion	≥99 % L	semi-crystalline	60	175	8, 210 °C, 2.16 kg
Luminy LX175			96 % L	amorphous	60	155	6, 210 °C, 2.16 kg
Luminy LX575			98 % L	semi-crystalline	60	165	7, 210 °C, 2.16 kg
Luminy LX975			88 % L	amorphous	60	130	10, 210 °C, 2.16 kg
Luminy D120			≥99% D	semi-crystalline	60	175	23, 210 °C, 2.16 kg
Luminy L105		Injection	≥99 % L	semi-crystalline	60	175	70, 210 °C, 2.16 kg
Luminy L130			≥99 % L	semi-crystalline	60	175	23, 210 °C, 2.16 kg
Luminy D070		Nucleating agent	≥99 % D	semi-crystalline	60	175	20, 190 °C, 2.16 kg
PLE005	NaturePlast	Extrusion	99 % L		n.d.	n.d.	23, 210 °C, 2.16 kg
PLE005-A			95 % D	amorphous	n.d.	n.d.	4, 190 °C, 2.16 kg
PLE005-1			n.d.		n.d.	n.d.	8, 210 °C, 2.16 kg
PLI005		Injection	PLLA	amorphous	48-50	170-180	25-35, 190 °C, 2.16 kg

The chosen grade was **Ingeo 4043D**, due to the fact that it consists of an extrusion grade biobased PLA that is generally recommended for 3D printing. It is also thoroughly studied in the literature for corresponding applications [21]. Thus, PLA Ingeo 4043D was purchased from NatureWorks (25 kg). Datasheet available here.

3. DETERMINATION OF EXTRUSION CONDITIONS FOR PLA COMPOUNDING

Before the biocomposites' production, it was important to conduct a preliminary study on the extrusion process, in order to determine the conditions for the chosen PLA grade. The extrusion was studied using a twin-screw extruder (HAAKE Thermo Fischer PTW16, **Figure 2**) at the Laboratory of Polymer Technology (LPT).

Figure 2. HAAKE Thermo Fischer PTW16 twin-screw extruder at NTUA LPT.

As a first step, the NTUA team dried some small amount of the Ingeo 4043D at 80 °C for 4 h. Then, the focus turned to matching the throughput of the volumetric feeder with the extruder's throughput. This step was necessary in order to prevent overfeed, which would by extend, lead to increased torque (stress), increased pressure and inconsistent flow, as well as, to prevent underfeed, which would result in inconsistent melting and mixing and air pockets in the product. In other words, the goal was to achieve **stable conditions and product uniformity**. The following parameters were tested and the results are presented below:

- Feeder screw rotation speed (1-40 rpm)
- Extruder screw rotation speed (30, 50, 80, 100 rpm)
- Extruder temperature/all heating zones (185, 195, 205 °C)

In **Table 3**, the three aforementioned parameters tested in different combination sets are reported. The closest matching of the feeder's throughput with the extruder's throughput was for 80/3 (Extruder/Feeder rpm) with ≤ 1% throughput difference at all tested temperatures. The difference was calculated based on the following **Equation 1**:

$$D\ (\%) = \frac{|\text{Feeder throughput} - \text{Extruder throughput}|}{\text{Extruder throughput}} \times 100\% \quad \text{(Eq. 1)}$$

Based on prior results at 185 °C and 195 °C, 80 rpm was identified as the optimal extruder screw rotation speed. Therefore, subsequent testing at 205°C focused solely on verifying that at 80 rpm, the D% remained ≤1%. Although other rpm values at 205 °C might yield similar D% results, they were not applicable across all temperatures, rendering further testing at that stage unnecessary.

Table 3. Extruder-feeder throughput study using PLA Ingeo 4043D.

Extrusion temperature (°C)	Extruder rpm	Feeder rpm	Extruder throughput (g/min)	Deviation (g/min)	RSD (%)	Feeder throughput (g/min)	D (%)
185	30	1.5	3.26	0.26	0.08	3.67	12.7
		3	6.42	0.26	0.04	7.15	11.4
	50	3	7.26	0.26	0.04	7.15	1.62
		4.5	10.25	1.06	0.1	10.52	2.6
	80	**3**	**7.11**	**0.18**	**0.03**	**7.15**	**0.5**
		4.5	10.44	0.55	0.05	10.52	0.8
	100	3	6.97	0.15	0.02	7.15	2.6
		4.5	10.88	0.58	0.06	10.52	3.3
195	30	1.5	3.53	0.25	0.07	3.67	4.1
		3	6.65	0.34	0.05	7.15	7.4
	50	1.5	3.51	0.13	0.04	3.67	4.6
		3	6.88	0.06	0.01	7.15	3.9
	80	1.5	3.89	0.44	0.11	3.67	5.6
		3	**7.06**	**0.17**	**0.02**	**7.15**	**1.0**
		4.5	10.83	0.16	0.01	10.52	2.9
	100	1.5	3.50	0.12	0.03	3.67	4.9
		3	7.0	0.37	0.05	7.15	2.1
205	**80**	1.5	3.16	0.1	0.03	3.67	16.1
		3	**7.08**	**0.08**	**0.01**	**7.15**	**1.0**
		4.5	10	0.21	0.02	10.52	5.2

The effect of the extrusion temperature was then tested on the extrudates' properties, i.e. thermal properties and molecular weight, when operating at 80/3 rpm. The extrusion outputs, such as average torque and pressure are given in **Table 4**, and it is evident based on their RSD% values, that the temperature of 195°C provided the most consistent and moderate extrusion parameters, in terms of material strain and thermomechanical stress, in a total extrusion cycle of approximately 15 min.

Table 4. Output parameters of 80/3 rpm extrusion at 185/195/205°C.

Extrusion temperature (°C)	Torque (Nm)	RSD (%)	Pressure (bar)	RSD (%)
185	60.4 ± 12.3	20.4	9.3 ± 2.3	25.1
195	55.7 ± 7.3	13.1	6.9 ± 1.2	16.8
205	49.1 ± 9.5	19.3	4.9 ± 1.1	22.5

In order to have a complete overview of the temperature effect, the thermal properties were also studied, *via* DSC/TGA, using a Mettler Toledo TGA/DSC 1 STARe System. To begin with, the TGA was performed in a temperature range between 30 - 600°C with a heating rate of 10°C /min and N_2 flow of 20 ml/min. As shown in the following graphs (**Figure 3**), no significant differences were detected between the materials prior or post to the extrusion cycle. Furthermore, the characteristic thermal stability properties (**Table 5**) remained around the following values: $T_{d,5\%}$ = 330°C, T_d = 370°C and Residue < 2 %.

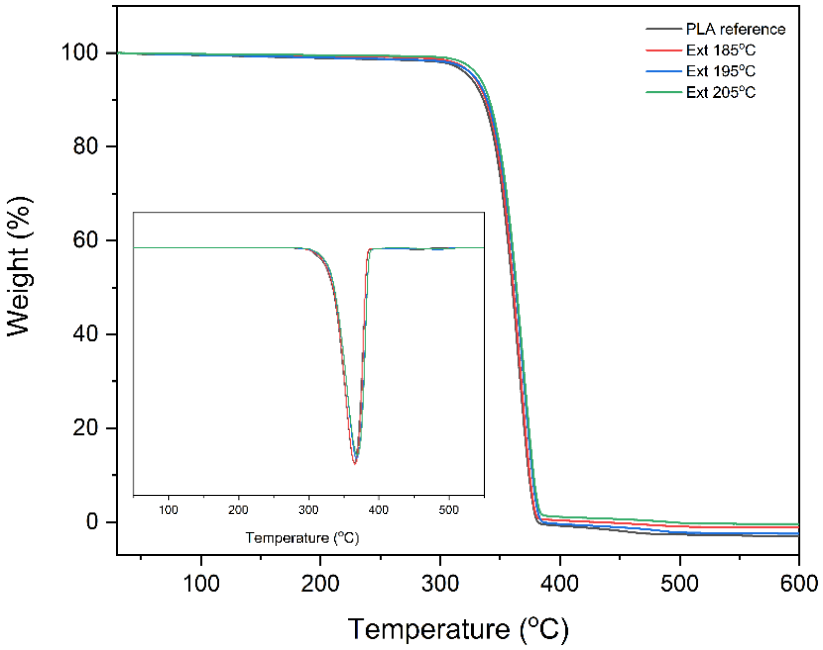


Figure 3. TGA curves for PLA Ingeo 4043D prior and post extrusion at 185/195/205°C.

Table 5. TGA characteristic temperatures for PLA Ingeo 4043D thermal stability prior and post extrusion at 185/195/205°C.

Materials	$T_{d5\%}$ (°C)	RSD (%)	T_d (°C)	RSD (%)	Residue (%)
PLA Ingeo 4043D	328.3 ± 5.5	1.7	368.3 ± 4.2	1.2	< 2 %
PLA ext@185°C	327.8 ± 0.0	0.0	366.2 ± 0.0	0.0	
PLA ext@195°C	330.0 ± 2.4	0.7	369.5 ± 1.4	0.4	
PLA ext@205°C	331.5 ± 1.0	0.3	370.7 ± 1.6	0.4	

Regarding the DSC, the method included heating-cooling-heating cycles, in the range of 30 - 210°C, with a heating rate of 10°C/min and N_2 flow of 20 ml/min. Once again, no significant differences were detected for the melting properties between the materials prior or post to the extrusion cycle and all of them presented a T_g around 60°C (amorphous phase), as shown in **Figure 4(a)**. However, it is worth highlighting that for the material as received (PLA Ingeo 4043D), a melting peak was detected only during the 1st heating (semi-crystalline regions), while afterwards no crystallization and no melting during the 2nd heating were reported. All the aforementioned observations, proved the semicrystalline nature of the material. In contrast, the extruded materials at the three different temperatures, they all presented *cold crystallization* during the 1st and 2nd heating cycles, right before the melting, while during the cooling, no *melt crystallization* took place.

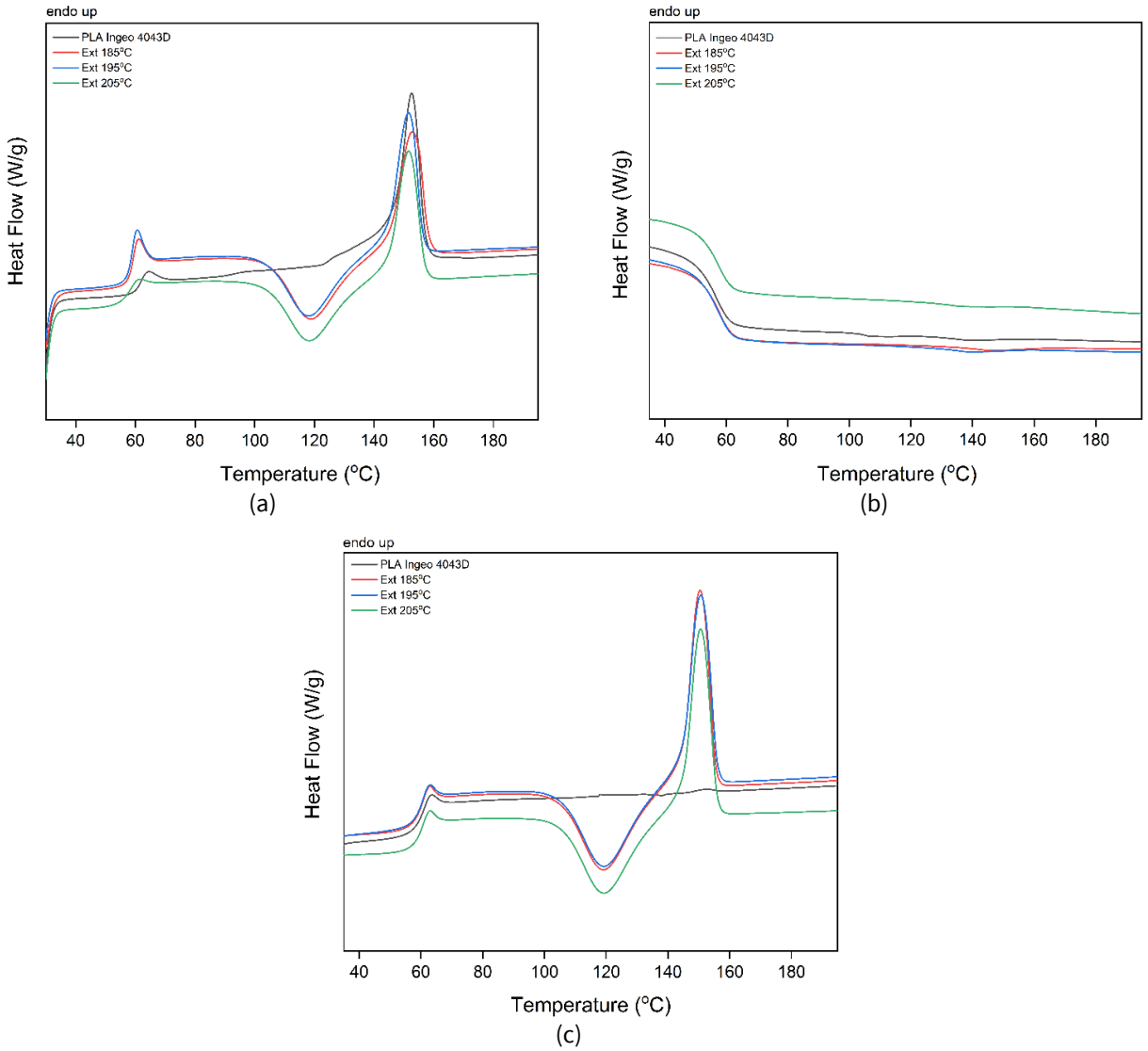


Figure 4. DSC curves of (a) 1st heating, (b) cooling, (c) 2nd heating of PLA Ingeo 4043D prior and post extrusion at 185/195/205°C.

Finally, dilute solution viscometry (DSV) was conducted based on ISO1628 (for thermoplastic polyesters). The measurements were performed in chloroform at a concentration of 0.5 g/dL in an Ubbelohde 0C capillary viscometer at 30 ± 1°C. All samples were dissolved at room temperature and filtered prior to the measurement. The viscosity number (VN, dL/g) and intrinsic viscosity ([η], dL/g) values were calculated based on the following equations:

$$VN\ (dL/g) = \eta_{sp} - 1 \qquad \text{Eq.2}$$

$$[\eta]\ (dL/g) = \frac{\sqrt{1+1.5\eta_{sp}}-1}{0.75\ C} \qquad \text{Eq.3}$$

where C is the solution concentration (g/dL), η_{sp} the specific viscosity of the solution. The intrinsic viscosity was converted to viscosity-average molecular weight (M_v, g/mol) through the equation: $[\eta] = K\overline{Mv}^a$, where the values of the parameters K and a were 1.31 x 10^{-4} (dL/g)(g/mol)$^{-0.777}$ and 0.777 respectively [22].

The results are presented in **Table 6** and **Figure 5**, and in combination with the standard, in which it is stated that a maximum 3% deviation is allowed between the VN values, it is found that the 205 °C is not within the acceptable margin due to large VN deviations and thus it is rejected.

Table 6. DSV results for PLA Ingeo 4043D prior and post extrusion at 185/195/205°C.

Materials	VN (dL/g)	[η] (dL/g)	M_v (g/mol)
PLA Ingeo 4043D	2.295 ± 0.025	1.732 ± 0.006	200600 ± 1000
PLA ext@185°C	2.237 ± 0.001	1.697 ± 0.000	196200 ± 100
PLA ext@195°C	2.248 ± 0.003	1.704 ± 0.002	197200 ± 300
PLA ext@205°C	2.162 ± 0.003	1.651 ± 0.002	189300 ± 300

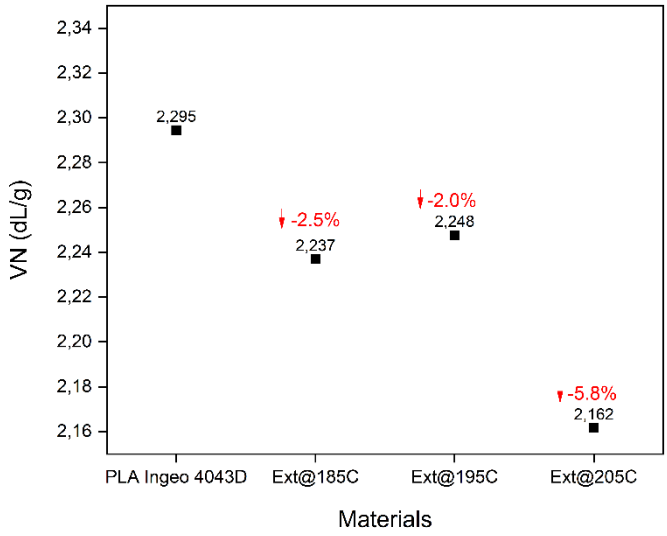


Figure 5. VN (dL/g) results for PLA extrudates at 185/195/205°C and % deviations from PLA Ingeo 4043D (reference).

Based on all of the aforementioned, in order to achieve a steady and uniform flow rate during twin-screw extrusion and, a consistent material flow with low extent of thermomechanical degradation, as evidenced by molecular weight decrease, the optimal extrusion parameters set is: **3 rpm feeder screw rotation speed, 80 rpm extruder screw rotation speed, temperature of all heating zones at 195°C, under vacuum**, reaching a PLA throughput equal to 0.2-0.3 kg/h.

4. BIOCOMPOSITES OF PLA WITH LIGNOCELLULOSIC MATERIALS FROM EUCALYPTUS TREE

Once the study of the extrusion parameters for the compounding was complete, the biocomposites' production was initiated. The NTUA team focused on determining a protocol for the production and further, the characterization methods to be performed, based upon waste origin and application criteria. The main characterization methods included thermal, melt flow rate and mechanical properties tests of biocomposite compounds prepared with lignocellulosic materials extracted from the eucalyptus tree (received from UHE). In detail, the following methods were conducted: ATR-FTIR, TGA-OOT, DSV, MFR, SEM, tensile and density measurements. The **lignocellulosic materials extracted from the eucalyptus tree** at this phase of the project are given below:

1. Lignocellulose nanofiber (LCNF): 5.8 g (bulky fluffy mass, **Figure 6.a**)
2. Cellulose Nanofiber (CNF): 6 g (bulky fluffy mass, **Figure 6.b**)
3. Lignin: 26 g (powder, **Figure 6.c**)

Figure 6. Right to left: (a) LCNF, (b) CNF and (c) Lignin materials for UHE.

For the incorporation of the LCNF and CNF into the PLA matrix, it was important to prepare a premix. Thus, PLA + 5 wt.% LCNF (or CNF) was prepared *via* cryogenic centrifugal milling (Fritsch Variable Speed Rotor Mill, FT-14) under liquid N_2 at 15000 rpm, using a 500 µm sieve. Finally, the materials received from the mill in a powder form were diluted with additional PLA, also in a powder form, to the final concentration of 1 wt.% LCNF (or CNF). Due to the lignin's initial form (received as powder) this intermediate step was not needed and the 1 wt.% concentration was achieved directly, using PLA, also in powder form (**Figure 7**).

Figure 7. (a) PLA (Ingeo 4043D) in powder form, (b) PLA + 5 wt.% LCNF.

The production of the **biocomposite compounds in the form of pellets** was conducted *via* twin-screw extrusion. Three biocomposite compounds were prepared at an average amount of **250 g per formulation**:

(1) PLA + 1 wt.% LCNF
(2) PLA + 1 wt.% CNF
(3) PLA + 1 wt.% Lignin

The experimental output parameters of the extrusions are presented below. The main focus was on the average torque and pressure, as recorded in **Table 7**. Notably, once incorporating even this small ratio of lignocellulosic materials, **more stable torque** and **much lower pressure** were observed throughout the extrusion processes. This effect could possibly be attributed to the lubricant effect of the materials, as found in the literature [23].

Table 7. Output parameters of 80/3 rpm extrusion at 195°C for the PLA neat and its 1 wt.% biocomposites, including density.

Materials	Torque (Nm)	Pressure (bar)	Density (g/cm³)
PLA Ingeo 4043D	-	-	1.238 ± 0.000
PLA neat @195°C	46.0 ± 6.8	4.6 ± 1.8	1.243 ± 0.003
PLA + 1% LCNF	38.9 ± 1.2	1.0 ± 0.3	1.244 ± 0.002
PLA + 1% CNF	40.4 ± 1.0	2.0 ± 0.4	1.243 ± 0.000
PLA + 1% Lignin	n.m.	n.m.	1.242 ± 0.003

***not measured**

Regarding the density measurements, they were conducted using a Density Kit on a scale (Mettler Toledo) with solid samples of ca. 5 g. The results as presented in **Table 7**, indicated a density around 1.24 g/cm^3 with the PLA Ingeo 4043D being the one with slightly lower value, most likely due to the fact that it was not subjected to an extrusion cycle, which typically involves melting and can lead to the formation of a more compact and dense material by enhancing particle packing, improving dispersion, and reducing porosity through the elimination of air pockets.

Following the biocomposite compounds' production, a complete characterization was conducted, using several analytical and non-analytical methods. The main conclusions of this phase of the study on the PLA biocomposite compounds of 1 wt.% lignocellulosic materials derived from the eucalyptus tree, indicated that they overall had similar properties and behaviour. However, **the formulation of 1 wt.% Lignin** proved to have a more positive impact on some of the properties studied, when compared to the other formulations, with respect to the properties of the initial neat PLA material. Explicitly, it indicated **the lowest carbonyl index (CI),** attributed to a slightly reduced oxidative degradation during processing. In terms of thermal stability, the PLA + 1% Lignin demonstrated **the highest onset degradation temperature ($T_{d5\%}$)** among the tested formulations, when in oxidative environment, indicating a noticeable antioxidant effect, even at this small concentration, likely due to lignin's inherent radical-scavenging properties. Dilute solution viscometry (DSV) showed **no molecular weight loss** for PLA + 1% Lignin, unlike the LCNF and CNF compounds, which experienced evident degradation. The MFR measurements were conducted in duplicates, at the standard conditions (ISO 1133): 210°C, 2.16 kg and 60 sec melt time, using a Dynisco model 4000 capillary rheometer. The **MFR** of PLA + 1% Lignin **remained nearly unchanged** compared to the reference PLA Ingeo 4043D, verifying the positive outcomes of the DSV and TGA experiments. The tensile properties were determined using an Instron 4400 machine (10 κN). 10 specimens of type 1BA were prepared by injection (Xplore injection micromolder) with the following dimensions: 55 mm length, 5 mm width, 2 mm thickness. The tensile testing proved that lignin offered the most balanced effect on the mechanical properties of the material, moderately enhancing tensile stress at yield. Finally, in terms of morphology monitored by SEM, the **biocomposites exhibited a notably rough surface, in contrast to the more uniform morphology of the neat PLA**.

5. PLA WITH LIGNOCELLULOSIC MATERIALS FROM WOOD WASTE

Wood waste from furniture feedstock, as supplied by Ecomaison (France) within W2W, was treated by UHE in the scope of T.7.1, and a new batch of lignocellulosic materials was produced and shipped to NTUA. Then, in light of the study in chapter 4, the activities by NTUA (**Figure 8**) described in chapter 5 involved:

1. The production of the new biocomposite compounds at **higher filler concentration, i.e. 5 wt.% content**, in order to improve performance and at **higher extrusion scale** (500 g per formulation), so as to cover a more systematic series of characterization methods.

2. Focus was given on the durability potential of the W2W biocomposite compounds through subjecting the materials to **accelerated ageing in a UV chamber** and using flexural bending as the key evaluation parameter since this was defined by the project end user (façade construction).

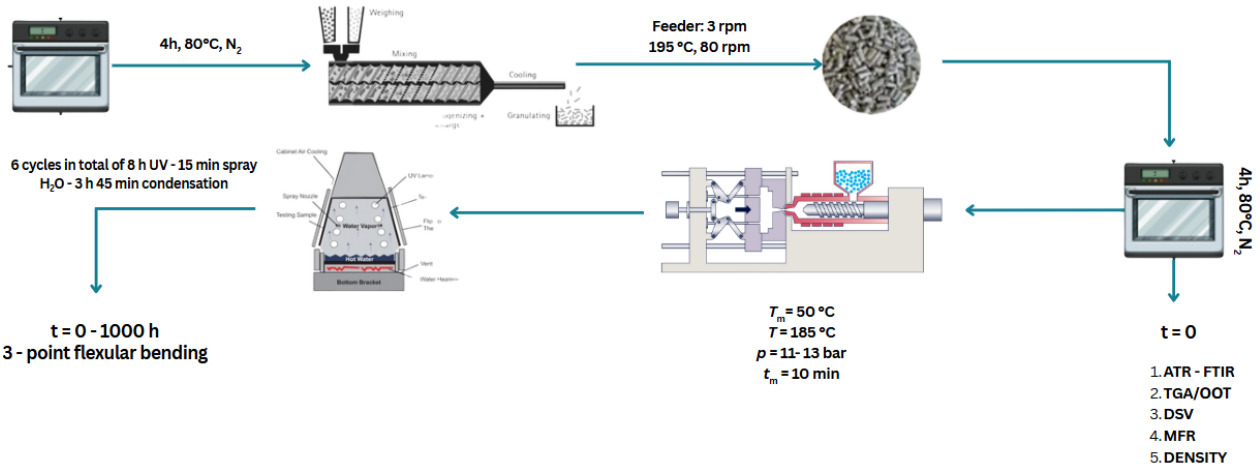


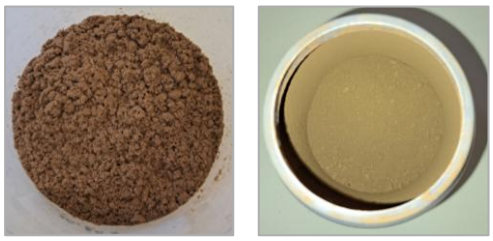
Figure 8. Flow chart of the processes of the main study with the lignocellulosic materials derived from wood waste.

3. A preliminary **processability study** of the compounds was initiated by NTUA, in order to investigate the optimal parameters for PLA panels production *via* a heated-plates hydraulic press. Different sets of parameters were tested (heating temperature, heating time, cooling time etc.) for both the neat PLA Ingeo 4043D and the PLA + 2.5 wt.% LCNF + 2.5 wt.% Lignin (50:50) formulation, and based on the results, the optimal set was chosen in terms of optical purity, texture and brittleness. The conclusions were shared with the BLOOM partner.

Additionally, 4 kg of PLA Ingeo 4043D and 2 kg of PLA + 2.5 wt.% LCNF + 2.5 wt.% Lignin (50:50) were produced by NTUA and delivered to BLOOM for panels' production *via* compression molding in a heated-plates press (scale-up).

5.1. PRODUCTION AND CHARACTERIZATION OF PLA BIOCOMPOSITES COMPOUNDS

For the production of biocomposite compounds with the new lignocellulosic materials from wood waste, the PLA (Ingeo 4043D) was dried for 4 h at 80 °C in a N_2 atmosphere, while LCNF and Lignin (**Figure 9.a** and **Figure 9.b**, respectively) were dried for 2 h at 80 °C.



a) *b)*

Figure 9. (a) LCNF and (b) Lignin extracted from W2W wood waste (T.7.1).

All the materials were processed under the selected conditions of the extrusion study in chapter 4. Following, the compounds were also dried for 4 h at 80°C in N_2 atmosphere. Based on the extrusion output parameters (**Table 8**), it is worth commenting on lower pressure while compounding the PLA with 5 wt.% Lignin, which can be attributed to a reduced friction between material and screw. Regarding density, no significant variation was noted.

Table 8. Output parameters of 80/3 rpm extrusion at 195°C for the PLA neat and its 5 wt.% biocomposites, including density.

Materials	Torque (Nm)	Pressure (bar)	Density (g / cm³)
PLA Ingeo 4043D	-	-	1.238 ± 0.000
PLA neat@195°C	46.0 ± 6.8	4.6 ± 1.8	1.243 ± 0.003
PLA + 5% LCNF	36.0 ± 8.8	7.4 ± 3.3	1.239 ± 0.016
PLA + 5% Lignin	31.7 ± 5.6	0.6 ± 0.5	1.244 ± 0.003
PLA + 2.5% Lignin + 2.5% LCNF	38.5 ± 4.7	2.0 ± 0.7	1.239 ± 0.013

The new biocomposite formulations were also thoroughly characterized. The ATR-FTIR revealed consistent spectral patterns across the different formulations, with semi-quantitative analysis of the carbonyl region suggesting minor oxidative degradation. Notably, **formulations containing lignin exhibited slightly lower CI**, indicating improved oxidative stability. TGA showed no significant shifts in degradation temperatures among the formulations, suggesting that thermal stability was retained. DSV experiments indicated that **PLA + 5% Lignin better preserved molecular weight**, implying reduced polymer chain scission. MFR results further highlighted the influence of additives: **PLA + 5% LCNF demonstrated reduced flow**, due to poor dispersion of the filler, whereas lignin-enhanced formulations showed improved flow behaviour, likely due to lignin's lubricating characteristics. Overall, while LCNF alone posed some challenges, the incorporation of lignin contributed to enhanced performance across multiple metrics, particularly in oxidative resistance, molecular weight retention and processability. Therefore, the **PLA + 5% Lignin and PLA + 2.5% LCNF + 2.5% Lignin** were considered as the two most promising formulations for the next steps of the project.

5.2. DURABILITY EVALUATION OF PLA BIOCOMPOSITES COMPOUNDS

In order to study the durability of the PLA biocomposites with lignocellulosic materials extracted from W2W wood-waste, UV accelerated ageing was performed and then, the mechanical properties were evaluated *via* three-point flexural bending. In detail, the work executed in the frame of Task 7.2 involved: (1) production of specimens to be subjected to accelerated ageing, (2) accelerated ageing trials with systematic monitoring of a critical mechanical property. All of the steps are described in detail below.

5.2.1. Samples preparation

The materials examined in this preliminary durability study were the following:

- PLA Ingeo 4043D *(only for t = 0)*
- PLA neat extruded at 195°C
- PLA + 5 wt.% LCNF
- PLA + 5 wt.% Lignin
- PLA + 2.5 wt.% LCNF + 2.5 wt.% Lignin (50:50 LCNF/Lignin)

First, proper drying of the materials was performed at 80 °C for 4 h under N_2 atmosphere. Then, the injection molding was conducted, using an Xplore injection micromolder. The specimens were prepared according to ISO 178, with a rectangular shape and the dimensions: L = 80 mm, W = 10 mm, h = 4 mm, as presented in **Figure 10**. Finally, the standardized specimens were left for 40 h at 23°C and 50% humidity atmosphere prior to testing, according to the preconditioning standard.

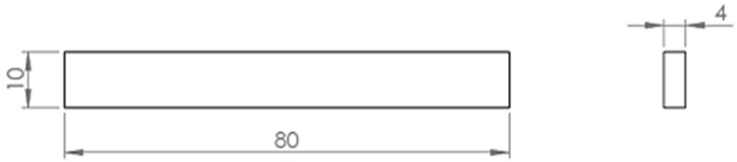


Figure 10. ISO 178 three - point bending test standards (in mm).

5.2.2. UV accelerated ageing

Once prepared, the specimens were placed in the UV chamber and the monitoring of the accelerated weathering under the chosen conditions was initiated. The experiments were performed in a QUV Accelerated Weathering Chamber (Q-Lab), based on the standard ISO 04892-3-2016. The method chosen was *Method A: Artificial accelerated weathering with UVA-340 lamps (type 1A)* and it included 6 exposure cycles of 8 h UV (irradiance 0.76 W/m^2 x nm^{-1} at 340 nm, 50°C ± 3°C), 15 min spray H_2O (not controlled temperature) and 3 h 45 min condensation (50°C ± 3°C). The sampling was performed until 1000 h.

5.2.3. Three - point flexural bending testing

The three - point flexural bending test was performed according to ISO 178, under room conditions and with a strain rate of 2 mm/min, using an Instron 4466 tester (10 kN). At t = 0, a distinctive increase of the flexural modulus was monitored, which indicated a beneficial effect of the lignocellulosic fillers in the PLA. Preliminary results for times up to 1000 h showed no significant degradation in terms of flexural modulus properties. Therefore, the durability studies will continue and the UV ageing will be in progress until 2000 h.

5.3. PROCESSABILITY STUDY OF PLA BIOCOMPOSITES COMPOUNDS

In order to prepare for T.7.3, a processability study was conducted using a heated-plates hydraulic press, for determining the optimal parameters for scaling-up panel fabrication from selected biocomposites *via* compression molding. First, preliminary tests were performed with the neat PLA, in different conditions, based on the literature [24]. More specifically, the parameters that were tested are shown below:

- Heating temperature (190, 180, 170°C)
- Heating time (20, 25, 30 min)
- Cooling time (15, 20 min)

The early steps of the processability study were conducted with the neat PLA Ingeo 4043D, after drying at 80°C for 4 h. A square mold (15 cm x 15 cm x 1 cm) was filled with ca. 30 g of the dried PLA and then inserted into the hydraulic press at 190°C and 170 bar for 20 min. After the 20 min passed, it cooled for 15 min and then the plaque was procured. At 190°C the PLA plaque of 1 cm thickness, was very flexible as it was removed from the mold, resulting in an uneven shape after it completely harden. The temperature of the hydraulic press was decreased at 180°C, resulting in a better end shape. To decrease the flexibility of the plaque as it was removed from the mold, the cooling time was increased at 20 min, resulting in smoother surface of the plaque. The heating temperature was then decreased at 180°C, because the fillers that will be added in later time are temperature sensitive. It was observed that by further lowering the temperature to 170°C, the PLA pellets would not melt. Finally, in order to improve the clarity of the plaque, the heating time was increased at 180°C. For these reasons, the experimental set N°7 was considered the most appropriate: 180°C for 35 min, steadily increased pressure until 170 bar and 20 min cooling time. An overview of the experimental sets of parameters, with corresponding results are presented in **Table 9**.

Table 9. Working conditions of hydraulic press and end-dimensions of films.

Experiments	EXPERIMENTAL CONDITIONS					RESULTS	
	m (g)	Heating temperature (°C)	Heating time (min)	Pressure (bar)	Cooling time (min)	Dimensions (cm x cm)	Ave. Thickness (mm)
1	28	190	20	170	15	15 x 15	0.8
2		190	20		20		1.0
3		180	20		20		0.8
4		170	20		20		1.0
5		180	25		20		1.0
6		180	30		20		0.9
7		180	35		20		1.0

Once the compression molding parameters for the neat PLA Ingeo 4043D were established, a subsequent experiment was performed using the biocomposite compound PLA + 2.5 % LCNF + 2.5 % Lignin. The results confirmed that the selected parameters: 180°C for 35 min, 170 bar and 20 min cooling time, were effective for the PLA biocomposites compound. Additional repetitions will be conducted in the future with newly developed formulations. The plaques of 1 mm thickness produced are presented in **Figure 11**.

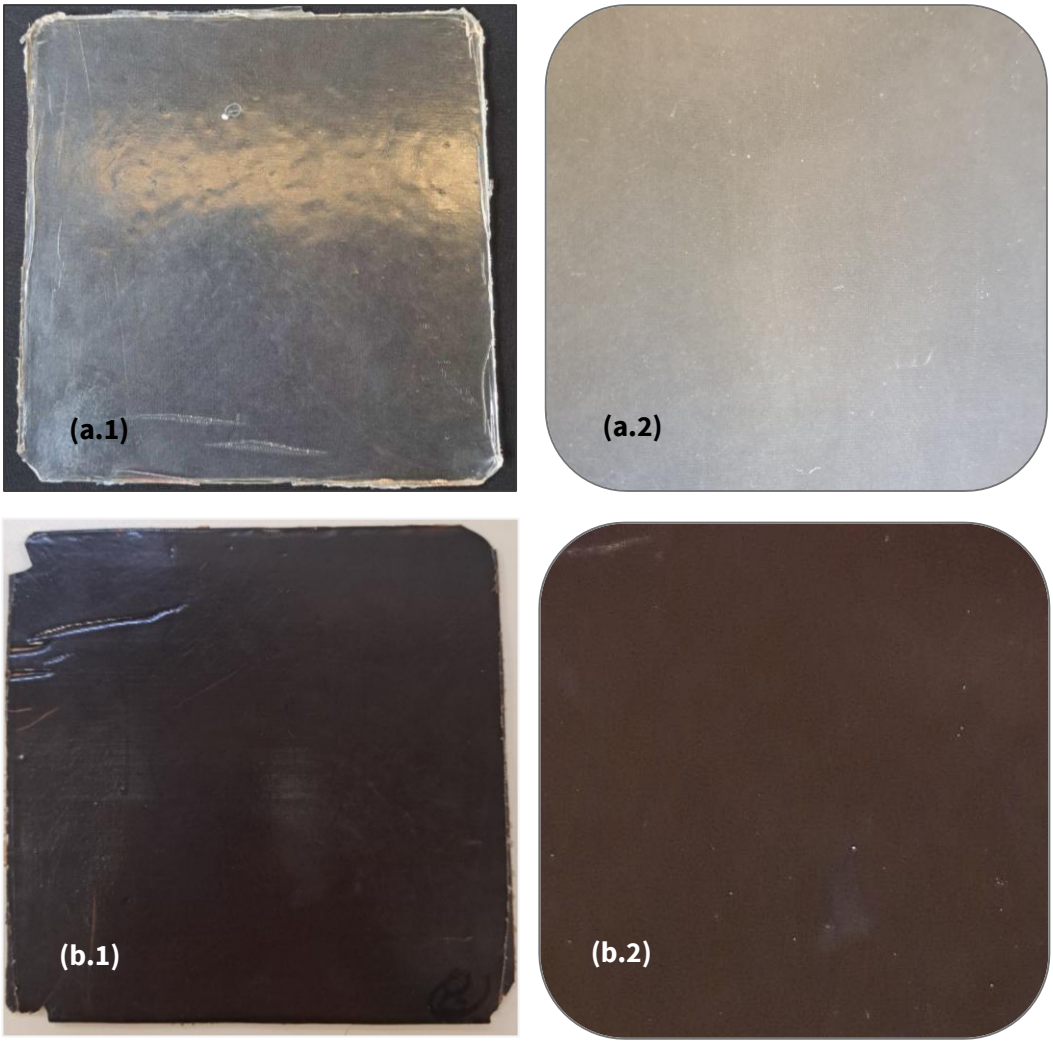


Figure 11. Heated plates press plaques produced from (a.1) neat PLA Ingeo 4043D and (a.2) closer view (b.1) PLA + 2.5%LCNF + 2.5% Lignin and (b.2) closer view.

6. PANEL PRODUCTION

After the detailed processability assessment and preparation of biocomposites, neat PLA Ingeo 4043D and a W2W biocomposites compound containing 2.5 wt.% LCNF + 2.5 wt.% Lignin, were shipped to BLOOM (4 kg and 2 kg respectively). The pellets were then analysed for key thermal properties (glass transition temperature and decomposition temperature) which are key for their processing steps. The pure PLA Ingeo 4043D analysis results were identical to what is reported above, while the PLA biocomposite (2.5 wt.% LCNF + 2.5 wt.% Lignin) showed an unchanged T_g at 58°C, a cold crystallisation at 88 to 124°C (typical for PLA) and two melting peaks, one at 147°C and one at 155°C indicative of the two crystalline phases of PLA.

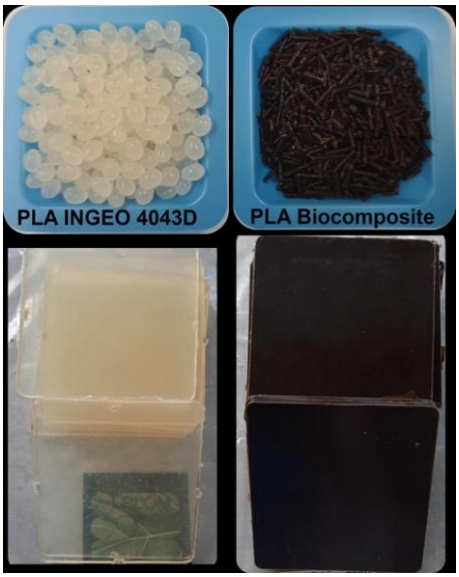


Figure 12. Image: (top left) commercial PLA pellets, (top right) W2W PLA 2.5 wt.% LCNF + 2.5 wt.% lignin biocomposite panels, and the panels made from each (below respective pellets).

The panel preparation of the pellets was performed on a hydraulic 20 tons press (Qnubu Press Pro Lion, plates 12x12 cm^2) using an aluminium mold of size 7 x 8 cm^2 with a thickness of 5 mm. Prior to pressing, the pellets were dried overnight at 45°C at 10 mbar. The pellets were placed in a sandwich mold, placed in the pre-heated press and left at this temperature for 10 min to ensure full melting of the pellets. A temperature of 190°C and a pressure of <2 MPa were used for the final pressing for 60 seconds. The mold was then removed and cooled outside of the press.

Using this approach, 12 small rectangular panels were made, weighing approximately 50 g each. Clear colour and homogeneity differences between the neat PLA and the W2W biocomposite' s panels were noted (**Figure 13**). For the latter, small black dots in the PLA matrix proved non-homogeneous dispersion.

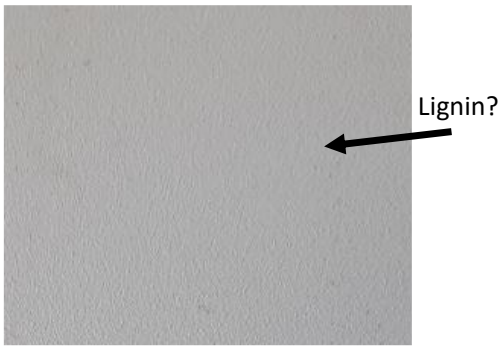


Figure 13. Photograph of a close-up of a W2W biocomposite hot pressed film which shows black dots dispersed within the PLA matrix.

The samples were shipped to LEVERY for acoustic tests according to their specifications of panel requirements of 39- and 99-mm diameter. The larger panels are still under preparation as different molds are needed (ordered but awaiting delivery).

For further acoustic tests and façade creation, larger plates (28 x 28 x 0.5 cm^3) are needed. Partners for such larger panels have been identified at BLOOM and the use of this compression molding press is planned for the end of June. Panels larger than 28 x 28 cm^2 require a different technology,

typically film extrusion followed by calendaring, to achieve homogeneous panels. This requires a significant amount of polymer >1 kg.

3D-printing is another processing method to make panels for façades, which allows more elaborate and lighter structures to be made. Contrary to the compression-molded panels detailed above, this manufacturing allows the creation of honeycomb panels which use less material and are more light weight. Given the weight of the 7 x 8 x 0.5 cm^2 sample above (50 g), a solid panel of 50 x 50 x 0.5 cm^3 size would weigh 2.2 kg. This is comparable to 12.5 mm thick gypsum panels or 15 mm OSB3 boards, highlighting the need for light weight solutions to be developed.

For this, the PLA and W2W composite can either be drawn into a filament before 3D-printing or the pellets can be directly printed using an adapted 3D-printer. The filament creation is an additional processing step which can induce degradation of polyester materials (i.e. PLA), while also requiring additional equipment and more material. A partner able to perform pellet 3D-printing has been identified and discussions are ongoing to detail the execution. As a back-up, a filament drawing machine is also available to BLOOM, as well as a conventional 3D-printing machine.

LEVERY provided the 3D models of such panels to BLOOM and two sizes are being targeted: 39 mm diameter and 99 mm diameter (**Figure 14**).

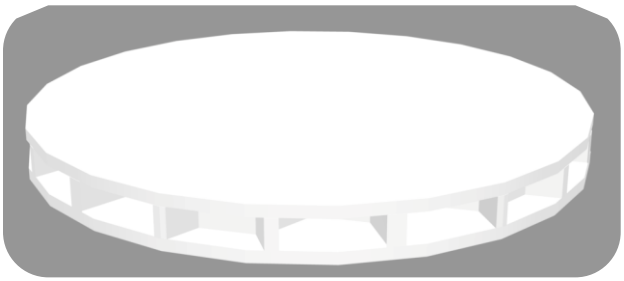


Figure 14. 3D-rendered image of the honeycomb-type panel.

To summarize, this phase of the study focused on the processability and preliminary application development of neat PLA and a W2W biocomposite compound containing 2.5 wt.% LCNF + 2.5 wt.% lignin. Thermal characterization confirmed that the PLA-based biocomposite compound retained a stable glass transition temperature (around 58 °C) and typical cold crystallization and melting behavior, indicating that the addition of LCNF and lignin did not negatively affect thermal processability. Compression molding revealed key visual differences between neat PLA and the biocomposite. The latter exhibited a non-homogeneous dispersion of fillers, evidenced by black dots and some migration onto the PET sheets during pressing. Despite these challenges, 12 panels were successfully fabricated and sent for acoustic performance evaluation. For even larger formats, or more material-efficient structures, 3D-printing is being explored as a viable alternative. Initial planning includes either direct pellet printing or filament extrusion, though filament preparation poses additional technical risks such as PLA degradation. A partner for pellet-based 3D-printing has been identified, and panel designs have been shared to facilitate production of both solid and honeycomb-style geometries in 39 mm and 99 mm diameter formats.

7. CONCLUSIONS

The current work examined the production and evaluation of PLA-based biocomposite compounds reinforced with lignocellulosic fillers, derived from eucalyptus tree and W2W wood waste. Biocomposite compounds were produced by NTUA, in various formulations and assessed through a series of physicochemical, thermal and mechanical tests. The aim was to determine the impact of the fillers on the PLA properties and to identify promising formulations for upscaled production.

At low filler loadings (1 wt.%) derived from eucalyptus tree, the lignin formulation consistently exhibited the most positive influence across several performance metrics. It showed the lowest CI, indicating reduced oxidative degradation, and exhibited the highest $T_{d5\%}$ under oxidative conditions, an effect attributed to lignin's inherent radical-scavenging properties. Additionally, DSV analysis revealed no molecular weight loss for PLA + 1% Lignin, in contrast to the degradation observed in CNF- and LCNF-containing composites. MFR remained nearly unchanged, confirming good processability, and tensile testing showed that lignin conferred the most balanced mechanical performance, modestly improving tensile yield strength. In the subsequent phase, biocomposites with higher lignocellulosic content (5 wt.%) derived from W2W wood waste were investigated. ATR-FTIR analysis across all formulations confirmed comparable chemical structures and minor oxidative degradation, with lignin-containing samples again showing slightly better oxidative stability. Thermal analyses showed that lignin helped to preserve molecular weight and thermal stability, with PLA + 5% Lignin showing the best retention of polymer chain length, and improved MFR due to lignin's lubricating behavior. Conversely, PLA + 5% LCNF posed challenges in dispersion and processability, evident by reduced flow and visual non-homogeneity, highlighting the importance of filler distribution within the matrix.

The combined formulation of PLA + 2.5% LCNF + 2.5% Lignin emerged as one of the most promising options, offering a balanced performance and compatibility in processing. It was successfully used in panel production *via* compression molding, where optimal processing parameters (180 °C for 35 min at 170 bar, with a 20 min cooling phase) were established. Though some dispersion issues and filler migration were observed, over 12 panels were successfully fabricated by BLOOM and sent for acoustic testing to LEVERY. Larger-scale panel production was initiated by BLOOM, using industrial compression molding equipment. Future plans include the fabrication of larger ($28 \times 28\ cm^2$) panels and potential transition to film extrusion and calendaring techniques for even larger formats. In parallel, 3D-printing has been identified as a promising alternative manufacturing route, especially for lightweight, structured panels such as honeycomb geometries. Two approaches are being explored: direct pellet printing and filament-based printing, with the former being prioritized to avoid additional thermal degradation risks. Partners have been identified, and CAD models have been provided to support prototype development.

Overall, the study conclusively demonstrated the technical feasibility and added value of incorporating lignin – particularly at both 1 wt.% and 5 wt.% concentration – into PLA for the development of more durable, processable and sustainable biocomposite materials. While LCNF presented some limitations in terms of dispersion and flow behavior, its synergistic use with lignin showed promise. These results provide a strong foundation for the continued upscaling of PLA-based biocomposites materials for real-world applications, such as façade systems production for exploitation within the construction market.

8. ANNEXES

8.1. ANNEX A: NATURE WORKS INGEO SERIES PLA GRADES

Supplier	Nature Works Ingeo Series										
Trademark	4060D	6302D	2500HP	2003D	4032D		4043D		6752D	3100HP	3001D
Grade	extrusion	extrusion	extrusion	extrusion	extrusion	extrusion	extrusion	extrusion	extrusion	injection	injection
Crystallinity	amorphous	amorphous	amorphous / semi-crystalline	semi-crystalline	semi-crystalline	semi-crystalline	semi-crystalline	semi-crystalline	semi-crystalline	amorphous / semi-crystalline	semi-crystalline
Optical purity	12 % D	10 % D	0.4 % D [2]	4.5 % D	1.6% D	1.6 % D	4.3 % D [3]	4.3 % D [3]	Mid D [5]	<2 % D [1]	1.5 % D [2]
Applications	seal layer between coextruded films	nonwoven	neat resin-need agents	sheet extrusion, blending resin	Profiles	films-packaging	3D-printing monofilament	films-packaging	Nonwoven, fibers	neat resin-need agents	clear bulk products
Datasheet [y/n]	y	y	y	y	y	y	y	y	y	y	y
Density (g/ml)	1.24	1.24	1.24	1.24	1.24	1.24	1.24	1.24	1.24	1.24	1.24
Tg (°C)	55-60	55-60	55-60	58	55-60	55-60	55-60	55-60	55-60	55-60	55-60
Tm (°C)	150-180	125-135	165-180	140 - 155	155-170	155-170	145-160	145-160	145-160	165-180	150-180
MFR (g/10min)	n.d.	15-20	8	6	7	7	6	6	15	24	22
MFR conditions	n.d.	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg
Tensile strength @ Break (Mpa)	n.d.	n.d.	n.d.	53	53	n.d.	53	53	n.d.	65	62
Tensile Yield Strength (Mpa)	n.d.	n.d.	64(am) / 65.5(cr)	60	60	15 kpsi MD / 21 kpsi TD	60	16 kpsi MD / 21 kpsi TD	n.d.	65 (Am) / 64 (Cr)	62
Tensile Modulus (Gpa)	n.d.	n.d.	n.d	3.5	3.5	500 kpsi MD / 550 kpsi TD	3.6	480 kpsi MD / 560 kpsi TD	n.d.	n.d.	n.d.
Tensile Elongation (%)	n.d.	50	3,6(am) / 4,3(cr)	6	6	180 MD / 100 TD	6	160 MD / 100 TD	Окт-70	3.4 (Am) /2.2 (Cr)	3.5
Notched Impact [Izod (j/m), Charpy (kJ/m²)]	n.d.	n.d.	18,7(am) / 40(cr) [Izod]	16 [Izod]	16 [Izod]	n.d.	16 [Izod]	16 [Izod]	n.d.	18,2 (Am) / 32 (Cr) [Izod]	16 [Izod]
Molded linear shrinkage (%)	n.d.	n.d.	0,4(am) / 1,8(cr)	similar to PET	n.d	n.d.	n.d.	n.d.	<8%	0,2-0,4 (Am) / 1,7-1,8 (Cr)	n.d.
Heat Distortion Temperature (°C)	n.d.	n.d.	54(am) / 144(cr)	55	n.d	n.d.	55	55	n.d.	3.4	55
Drying	4h, 45 °C	8-12h, 40 °C	10h, 50 °C (for amorphous)	2h, 90 °C	2h, 90 °C	4h, 80 °C	4h, 80 °C	4h, 80 °C	4-6h, 70-80°C	10h, <50°C (am) / 3h, 100 °C (cr)	10h, <50°C (am) 3h, 100 °C (cr)
Technique	extrusion, 24:1-30:1 (L/D)	extrusion, 24:1-30:1 (L/D)	extrusion, 24:1-32:1 (L/D)	extrusion, 24:1-32:1 (L/D)	extrusion, 24:1-32:1 (L/D)	extrusion, 24:1-30:1 (L/D)	extrusion, 24:1-30:1 (L/D)	extrusion, 24:1-30:1 (L/D)	extrusion, 24:1-30:1 (L/D)	injection	injection
Conditions	180-210 °C	220-240 °C	190-210 °C	180-210 °C	180-210°C	180-210 °C	180-210 °C	180-210 °C	220-240 °C	185-200°C, mold 120°C	165-205°C, mold 25°C
Enzyme								DmPETase/LCC/MoPE			
ΔMn, ΔMw (%)								0/0/10,4			
Mass loss (%)								<10/<10/0			

8.2. ANNEX B: TOTAL CORBION LUMINY SERIES PLA GRADES

Supplier	Total - Corbion Luminy Series							
Trademark	LX175	LX975	L175	LX575	L105	L130	D070	D120
Grade	extrusion	extrusion	extrusion	extrusion	injection	injection	nucl. agent	nucl. Agent
Crystallinity	amorphous	amorphous	semi-crystalline	semi-crystalline	semi-crystalline	semi-crystalline	semi-crystalline	semi-crystalline
Optical purity	96 % L	88 % L	≥99 % L	98 % L	≥99 % L	≥99 % L	≥99 % D	≥99% D
Applications	film extrusion, thermoforming, fiber	heat-seal layer-film	film extrusion, thermoforming, fiber	film extrusion	thin wall injection, fiber spinning	injection, fiber spinning	nucleating agent for heat resistance	nucleating agent for full stereocomplex PLA
Datasheet [y/n]	y	y	y	y	y	y	y	y
Density (g/ml)	1.24	1.24	1.24	1.24	1.24	1.24	1.24	1.24
Tg (°C)	60	60	60	60	60	60	60	60
Tm (°C)	155	130	175	165	175	175	175	175
MFR (g/10min)	6	10	8	7	70	23	20	23
MFR conditions	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	210°C, 2.16kg	190°C, 2.16kg	210°C, 2.16kg
Tensile strength @ Break (Mpa)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Tensile Yield Strength (Mpa)	45	40	50	50	50	50	50	50
Tensile Modulus (Gpa)	3.5	3.5	3.5	3.5	3.5	3.5	3.5	3.5
Tensile Elongation (%)	≤5	≤5	≤5	≤5	≤5	≤5	≤5	≤5
Notched Impact [Izod (j/m), Charpy (kJ/m²)]	≤5 [Charpy]	≤5 [Charpy]	≤5 [Charpy]	≤5 [Charpy]	≤5 [Charpy]	≤5 [Charpy]	n.d.	≤5 [Charpy]
Molded linear shrinkage (%)	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
Heat Distortion Temperature (°C)	60	60	60	60	105	105	n.d.	n.d.
Drying	4-6h, 85 °C	24h, 40 °C	4-6h, 100 °C	4-6h, 85 °C	4-6h, 100 °C	4-6h, 100 °C	4-6h, 100 °C	4-6h, 100 °C
Technique	extrusion, 24:1-32:1 (L/D)	extrusion, 24:1-32:1 (L/D)	extrusion, 24:1-32:1 (L/D)	extrusion, 24:1-32:1 (L/D)	injection, mold 90-100°C	injection, mold 90-100°C	n.d.	n.d.
Conditions	170-210 °C	180-210 °C	180-210 °C	180-210 °C	155-220°C, slow speed	155-220°C, slow speed	n.d.	n.d.

8.3. ANNEX C: NATURE PLAST PLA GRADES

Supplier	Nature Plast					
Trademark	**PLE005**	**PLE005-A**	**PLE005-1**	**PLI005**	**PLI211**	**NP HT 201**
Grade	extrusion	extrusion	extrusion	injection	injection	injection
Crystallinity		amorphous		amorphous		
Optical purity	99 % L [4]	95 % D [4]	0	PLLA		Opaque
Applications	n.d.	n.d.	n.d.	n.d.		high temperature resistance, high impact
Datasheet [y/n]	n	n	n	y	n	y
Density (g/ml)	1.25	1.24	1.25	1.25	1.25	1.28
Tg (°C)				48-50		n.d.
Tm (°C)				170-180		170-180
MFR (g/10min)	23	4	8	25-35	30	26
MFR conditions	210°C, 2.16kg	190°C, 2.16kg	210°C, 2.16kg	190°C, 2.16kg	190°C, 2.16kg	190°C, 2.16kg
Tensile strength @ Break (Mpa)				60		31.2
Tensile Yield Strength (Mpa)				27.8		15.2
Tensile Modulus (Gpa)	3.5			3.5	3.5	3.6
Tensile Elongation (%)				4.7		17.2
Notched Impact [Izod (j/m), Charpy (kJ/m^2)]	20 [Charpy unnotched]	21 [Charpy unnotched]		25 [Charpy unnotched]	20 [Charpy unnotched]	85 [Charpy unnotched]
Molded linear shrinkage (%)						1.3
Heat Distortion Temperature (°C)	61	60	55-60	54	60	127
Drying				3h, 60 °C		4h, 60 °C
Technique				Injection, mold 20 °C		Injection, mold 105 °C
Conditions				25-195°C		25-190°C

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