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GREEN-LOOP

Sustainable manufacture systems towards novel bio-based materials

WP1 – Set-up of GREEN LOOP biobased products, green and smart solutions

D1.4 – Scientific and technical plan and execution report

Document information

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GREEN LOOP Consortium Partners

	Partner	Acronym	Country
1	IDENER RESEARCH & DEVELOPMENT	IDE	ES
2	NATIONAL INSTITUTE OF CHEMISTRY	NIC	SI
3	SLOVENIAN NATIONAL BUILDING AND CIVIL E. I.	ZAG	SI
4	FRAUNHOFER GESELLSCHAFT ZUR FOERDERUNG DER ANGEWANDTEN FORSCHUNG E.V	FHF	DE
5	LABRENTA SRL	LBRT	IT
6	MIXCYCLING SRL	MYX	IT
7	NEROSUBIANCO	NSB	IT
8	TERRE DI ZOE’	TDZ	IT
9	IRIS TECHNOLOGY SOLUTIONS, SOCIEDAD LIMITADA	IRIS	ES
10	GLOWNY INSTYTUT GORNICTWA	GIG	PL
11	AACHEN UNIVERSITY: PROCESS CONTROL ENGINEERING / AACHEN UNIVERSITY: INSTITUTE OF SOCIOLOGY	AAU	DE
12	AUSTRIAN STANDARDS INTERNATIONAL	ASI	AT
13	INSTITUTO DE SOLDADURA E QUALIDADE	ISQ	PT
14	AXIA INNOVATION UG	AXIA	DE
15	ASOCIACIÓN DE INVESTIGACIÓN METALÚRGICA DEL NOROESTE	AIMEN	ES
16	NATIONAL COMPOSITE CENTER	NCC	UK
17	UNIVERSITY OF BRISTOL	UBRIS	UK

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Executive Summary

In WP1, the monitoring and controlling of the technical development will be performed in Task1.1. This third D1.3 (M24) provides an update on the technological development in the GREEN-LOOP project. It is primarily focused on the progress of Work Package (WP) 3-6. These four WPs cover the complete technology of the three value chains. Fraunhofer (FHF), supported by IDENER (IDE), are both in charge of the technical coordination.

Bio-rubber materials and their processing by the steps formulation, compounding and compression moulding to multifunctional rubber panels are investigated in **WP3**. The bio-rubber value chain is aiming to demonstrate the manufacture of panels with a reduction in vibro-acoustics transmission and an improvement in fire-retardant properties. The lignin extraction trials have been successfully completed. The rubber formulation has been increased to TRL 5 at M24 as production has moved from lab scale to 10-20 Kg batches. Ultrasound application only focus on rubber devulcanisation and not lignin extraction. Energy monitoring sensors provided by IRIS were installed to observe the energy consumption of the devulcanisation process. A novel layer-by-layer (LbL) method was performed by coating the rubber plates with a biobased coating solution at UBRIS and fused together using compression moulding. Both, the devulcanisation process as well as the multilayer coating process is at lab scale, TRL 4 and will be upscaled in WP6. Finally, the panel size 350 x 350 x 3 mm could be produced by compression moulding to be tested at ZAG and ISQ.

On the other hand, novel bioplastic material for bottle closures is the focus of the **WP4** value chain. First prototypes of limoncello caps and olive oil dispensers were achieved by extrusion and injection moulding process. Viable material blends based on PLA/PHBH biopolymers were developed at MYX by extrusion. The pellets were used in the injection moulding (IM) unit at LBRT. The integration of the microwave system to the hopper will be performed end of M24. It is further planned to collect all data from the preheating of the pellets and to transfer it to the ICT platform. The sensors installation and data management are still ongoing at LBRT facilities. TRL5 was finally reached, and it is planned to go on with IM process by using selected blends in WP6 to reach TRL6. Moreover, final product tests are planned to assess the biodegradability, compostability, and permeability of the products.

Finally, the goal of **WP5** is to develop novel Wood Composite (WC) material for friction application, particularly for sliding bearings. The smart manufacturing has been approved by integration of the MW system provided by IDE to the extruder at FHF. Thus, TRL5 could be achieved. The new process work and the smart feature power meter is already connected to the device to collect the energy consumption during extrusion. The data from the first MW assisted extrusion process will be transferred to the ICT platform at IRIS and the energy consumption will be calculated until end of M25. The achieved WC material showed a high potential to be used as sliding bearings. Low Wear and friction coefficients could be achieved. Environmental related issues will be mainly investigated in WP6 when the process runs in larger scale. WP6 will focus on small series production and increase the output of the extrudate. Other issues such as circularity of the wood composite product, reduction of waste and emissions as well as recyclability will be addressed.

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Abbreviations

CMC - Carbomethyl cellulose

DX.X – Deliverable X.x

ICT – Information and Communications Technology

IM – Injection Moulding

KPI – Key Performance Indicator

LbL – Layer by Layer

M - Month

MW - Microwave

SLRP - Sequential Liquid-Lignin Recovery and Purification

SOA – State of the Art

TX.x – Task X.x

TRL - Technology readiness level

US – UltraSound

WC – Wood Composite

WP - Work Package

1. Introduction

1.1 Objectives of Task 1.1

In this task, the technical developments will be monitored and controlled. This comprises the coordination of technological development and the technical progress, supported by IDENER. Moreover, technical reports and deliverables will be approved regularly. The task covers all the activities to be carried out in the GREEN-LOOP three value chains during the whole running time of the project starting in month 1. The following specific objectives are pursued:

- Following the state-of-the-art and state of practice
- Progress of scientific and technical development
- Identification of technical problems and remedial actions

Deliverables: Periodic reports on the state of the technical issues will be performed in months 4, 12, 24 and 36, like the ones presented here as deliverable D1.3 [M24] and will be subjected to one more revised version in [M36].

1.2 Methodology

The methodology of Task 1.1 is based on three major actions:

- Monitoring the progress of production, sample manufacturing and risks
- Monitoring progress of KPIs (values), related to Task 1.3 'Validation KPIs definition'.
- Continuous patent and literature update

All GREEN-LOOP partners should provide their input on the technical progress to the task leader (FHF), which is the technical coordinator. Especially, input is required from partners doing the technical work in the value chains (WP3-WP5). These partners should provide the following items:

- Update of technical advance of key-process steps every 3 months.
- Update of risk table every 3 months
- Update list of KPI's every 3 months
- Literature study and patent survey regularly

The collection of these inputs from the partners will be done in form of questionnaires. The partners who provide the information will be especially the WP leaders from the value chains WP3 (NCC), WP4 (LBRT) and WP5 (FHF). The expected output of the provided technical information is the following:

- Punctual identification of risks in the value chains
- Identification of technical progress or technological gaps
- Recommendations for the further planning
- Prioritisation of technical work

The output will be performed in quarterly feedback rounds after assessing the technical information.

2 State-of-the-art (SOA) technology of value chains

Research in GREEN-LOOP project is focused to set up feasible process chains for producing biomaterials (WP3-WP5) to obtain Technology Readiness Levels TRL 6. A fine-tuning for the retrofit of tooling and facilities to prepare the equipment for relevant environment production and upscaling activities is finally planned in WP6 ('Upscale production and demonstration'). The TRL's defined by the EC are used to assess the TRL of each value chain WP3-WP6 at the beginning of the project and will be updated in the following reports.

2.1 SOA of WP3 "Bio rubber material production"

2.1.1 Materials concept for bio-rubber and evolution of the process

The bio-rubber value chain is aiming to demonstrate the manufacture of multifunctional rubber panels with a reduction in vibro-acoustics transmission and an improvement in fire-retardant properties. The target use case for the panels is within the construction industry as internal panelling. The work will utilise recycled and bio-derived material feedstocks to reduce the environmental impact of the final product. There are two main components to the formulation – rubber which is reclaimed from waste tyres, and lignin powder which is extracted from waste biomass. The formulations are compounded within an internal mixer before being compression moulded into the desired final geometry.

For waste rubber to be suitable for reprocessing it must first go through a process called devulcanisation to break the cross-linking bonds formed during its first manufacture. Commercially available devulcanised rubber has been used in the bio-rubber formulation trials. Alongside this work is continuing within Greenloop to develop a new method of devulcanisation using ultrasound and deep eutectic solvents (DES) to minimize the environmental impact of the devulcanisation step. The lignin extraction has been carried out using the Kraft extraction method. This lignin powder then undergoes a purification step prior to compounding into the rubber.

2.1.2 Technology Readiness Level

The rubber formulation has been increased from TRL 4 at M12 to TRL 5 at M24 as production has moved from lab scale (60g per batch) to 1.2 Kg batches. 10-20 Kg batches are planned during M24.

The ultrasound/DES devulcanisation process is at lab scale, TRL 4, pending upscale in WP6.

The multilayer coating process is at lab scale, TRL 4, pending upscale in WP6.

2.1.3 Technical progress and highlights up to month 24

NIC have produced 5 Kg of Kraft lignin for use in formulation trials. NMR and FTIR testing have been carried out to confirm successful lignin isolation and to characterise the lignin structure. Fractionation of the lignin to remove lower molecular weight fractions improved the lignin powder’s fire resistance by up to 20 %.

A new method of rubber devulcanisation has been explored by IRIS and UBRIS combining ultrasound technology and deep eutectic solvents. Currently, this is at TRL 4; production is at 50 g per batch, but the scale will be increased within WP6. Testing has shown a devulcanisation percentage of 72% has been achieved which compares favourably with the baseline commercial product result of 75%. Compounding trials are ongoing to assess how well this material can be reprocessed.

Compounding trials (60 g per batch) have been completed to establish the optimum formulation for the rubber-lignin composite. An experiment design was carried out to determine the best ratio of devulcanised rubber, natural rubber, lignin, and curatives. The optimum formulations achieved so far contain 10-20 wt % lignin and the following mechanical properties have been achieved: break stress of 7.7-9.7 MPa, break strain of 311-330 %, Shore A hardness of 56-62 and a modulus at 100 % strain of 2.12-2.35 MPa. Compounding batch size has been successfully increased from 60g to 1.2 Kg with no impact on material. The material has been compression moulded into test panels at NCC for further testing at ZAG and ISQ. These can be seen in Figure 1. A further increase in production to 10-20 Kg batches will be completed in M24.

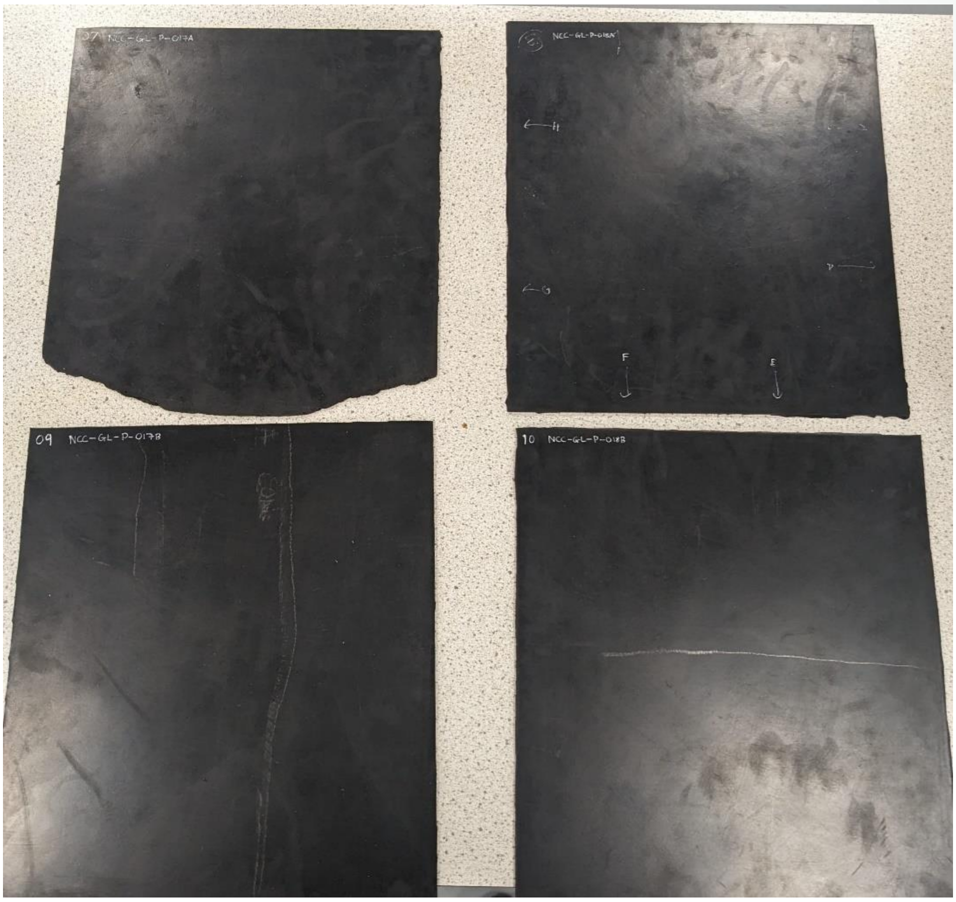

Figure 1. Compression moulded panels (350 x 350 x 3 mm) using the Greenloop developed formulation.

An adjacent piece of work is being carried out by UBRIS to assess the efficacy of incorporating lignin into a coating layer and apply the coating to the rubber panel, rather than mixing the lignin into the bulk of the rubber. Here, commercially sourced rubber sheets have been coated using a layer-by-layer (LbL) method. In this process, multiple rubber panels are coated with a biobased coating solution (polyacrylic acid (PAA), lignin, and sepiolite), they are then fused together using compression moulding. Trials so far have been on commercially available rubber panels and their fire resistance will be compared against the Greenloop produced rubber panels.

2.1.4 Technical gaps and challenges

Tests to apply ultrasound technology to lignin extraction were carried out in order to enhance this process. However, as previously communicated, after testing different ultrasound applications: before high-pressure extraction, during atmospheric-pressure extraction; and after assessing the effect of several variables involved in the process (solvent to solid ratio, extraction time, acoustic amplitude and energy) these trials showed that using ultrasound in kraft lignin extraction offers no significant benefit over conventional methods. Thus, further trials with ultrasound have been halted. Further upscale of this enhancement to the manufacturing line (use of the 2KW Ultrasound) will not take place in T3.3.3. An amendment was delivered to address this change. As a result, research to enhance the chemical devulcanisation of recycled rubber utilizing ultrasound will be carried out. IRIS will apply ultrasound using the 800 W prototype during the lab scale chemical devulcanization of recycled rubber using paraffin oil or urea/choline chloride as solvents. Some resources were transferred from WP3 to WP1, specifically into Task 1.4 Inline monitoring, with the inclusion of the near infrared (NIR) technology (VISUM) from IRIS for the determination of the lignin content in biomass prior to extraction (Task 1.4). IRIS will also extend its efforts towards T3.3.3 Adaption of ultrasound prototype to manufacturing line, increasing participation in the Green Chemistry Rubber Devulcanization.

This will not impact the final bio-rubber production, as lignin extraction can proceed without ultrasound via conventional extraction methods.

Initial compounding work on commercially sourced devulcanised rubber which was in a crumb format highlighted a challenge when using a small crumb size. The particle size was too small so did not experience sufficient shear force when compounded. This led to the crumb not mixing sufficiently and a final product with very poor mechanical strength. This was overcome by transitioning to another supply of commercially available devulcanised rubber which was supplied as a solid material rather than as a crumb. This could be compounded and resulted in a greatly improved performance. The results described above are for the formulations using the solid commercially available devulcanised rubber.

The learnings from compounding with the commercial material have been taken into the Greenloop devulcanisation trials which combine ultrasound technology and deep eutectic solvents. These trials had originally been carried out on a small crumb size to maximise the rubber surface area and therefore increase the chance of a high rubber devulcanisation percentage. However, now it was understood that the small crumb size would not be suitable for compounding, the focus shifted to trialling the devulcanisation on larger crumb sizes. The devulcanisation experiments have been repeated with larger crumb sizes and the results of the corresponding compounding tests are still outstanding.

2.1.5 Critical process steps

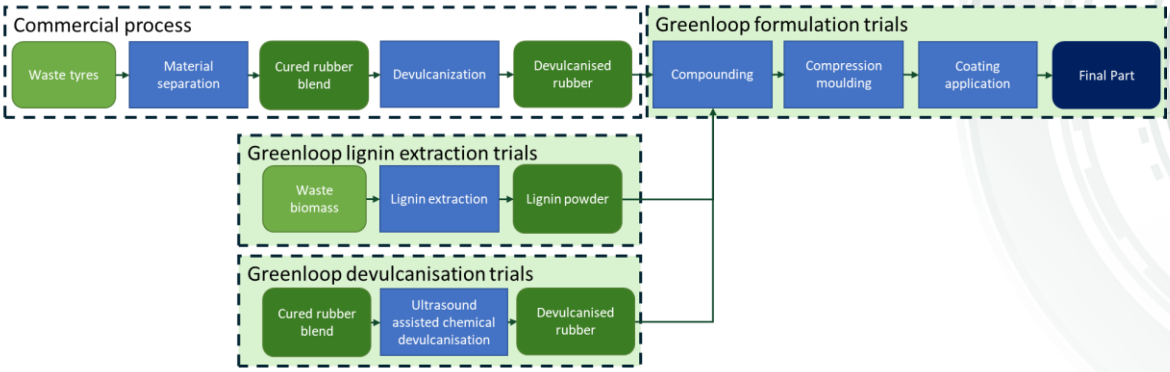

Figure 2: Bio-rubber critical process steps.

1) Lignin extraction trials – The extraction trials have been successfully completed and 5Kg of lignin has been produced. This is sufficient to produce 35 Kg of the rubber composite.
2) Devulcanisation trials – Devulcanisation trials have been carried out using ultrasound and deep eutectic solvents. Initial measurements have shown devulcanisation percentages of up to 72%. Compounding trials are underway to assess how well the material can be processed.
3) Formulation trials – Compounding – Trials completed to identify optimised formulation and batch size has been increased from 60 g to 1.2 Kg. Further scale up planned to 10-20 Kg per batch in M24.
4) Formulation trials Compression moulding – Initial compression moulding completed to produce 350 x 350 x 3 mm panels for testing at ZAG and ISQ. Thicker panel production (up to 13mm) will be carried out in M24 once further material has been compounded. This will allow for further testing at ZAG and ISQ.
5) Formulation trials – Coating application – A preliminary multi-layer rubber panel has been manufactured (100 x 100 x 10 mm). Further optimisation will be carried out before undergoing fire testing at ZAG.

2.1.6 Delays and their reasons

Deliverable 3.3 and Deliverable 3.4 were delayed from M20 to M23 to allow for additional work on the material formulation and ultrasound enhanced processes following the agreed change in ultrasound application to only focus on rubber devulcanisation and not lignin extraction. Both D3.3 and D3.4 were delivered in M23.

2.1.7 Technology focus in next semester for each task

The general status of the process steps can be derived from Figure 2. The following work is planned in the next semester:

Task 3.2 – Completed.

- All design work and preparation for manufacturing has been carried out. Tooling design and manufacture has been completed.

Task 3.3 – Ongoing

- Task 3.3.1 – Energy monitoring sensors to be installed in M24. Collaboration ongoing with IRIS to integrate into Greenloop ICT platform
- Task 3.3.2 – Devulcanisation trials completed on 50g scale using 800W ultrasound prototype. Awaiting results from compounding trials. Upscale activity to be carried out in WP6
- Task 3.3.3 – Preparation for transfer from 800W to 2kW prototype completed. Awaiting results of compounding trials.

Task 3.4 – Completed.

- All material procured (100Kg of commercially available devulcanised rubber purchased) and lignin extraction completed for WP3. Further lignin extraction will take place in WP6.

Task 3.5 – Ongoing

- Task 3.5.1 – Complete. Lab scale formulation trials completed
- Task 3.5.2 – Upscaled compounding and compression moulding to be completed in month 24.

Task 3.6 – Ongoing

- Characterisation of physical properties (mechanical, thermal), fire performance (ZAG), acoustic testing (UBRIS). Report with characterization results and key findings will be reported in D3.5 in M24.

Table 1. Status of process steps and responsibilities in WP3 [M24]. Legend: P=planned; O=ongoing; V=validated; I=Implemented, F=Finished

Work package	Resp.	Material	Core Technologies / process steps	Partner	Status
WP3	**NCC**	**Bio-rubber**	➢ **Lignin modification and production**	**NIC**	**C**
			➢ **Sensors (smart feature 1)**	**IRIS**	**O**
			➢ **Ultrasound enhancement (smart feature 2)**	**IRIS**	**O**
			➢ **Compounding**	**NCC**	**O**
			➢ **Press moulding**	**NCC**	**O**
			➢ **Multi-layer coatings**	**UBRIS**	**O**

			➢ **Testing**	**ISQ/ZAG**	**O**

2.2 SOA of WP4 "Bio-plastic material production"

2.2.1 Materials concept for bio-plastic composites and evolution of the process

In the initial stages, MYX experimented with various biopolymers, including PLA, PBAT, and Starch, and fillers such as calcium carbonate, titanium dioxide, cork, and eggshell. These fillers were meticulously micronized by MYX to achieve the desired particle size, facilitating their integration into the biopolymer matrix. Through rigorous testing, MYX determined that the combination of PHBH and PLA offered superior compatibility and dispersion of the biofiller in the polymer matrix. This combination not only ensured a consistent and high-quality final product but also benefited from excellent market availability and favourable processing characteristics.

After extensive testing, cork was identified as the most suitable filler for the melt extrusion process. Cork demonstrated superior compatibility with the PLA/PHBH biopolymer blend due to its excellent wettability, which ensures uniform interaction between the filler and the polymer matrix. During compounding, cork’s use resulted in minimal increases in torque and melt pressure, indicating a smooth integration with the biopolymer melt. This seamless integration is vital for producing high-quality, homogeneous granules, a key factor in the material's performance and processability.

The rigorous development process led to the creation of the T23089 blend, comprising PHBH, PLA, and cork. This blend stands out for its excellent processability, environmental sustainability, and suitability for applications requiring bioplastics that closely replicate the properties of traditional materials like aluminium and conventional plastics. The use of cork as a filler, combined with the innovative NTP treatment and the careful selection of biopolymers, resulted in a biocomposite that not only meets the project’s environmental goals, but also offers practical advantages in manufacturing and end-use performance.

MYX has also developed blend T23088 (PHBH/PLA) to use in the project.

2.2.2 Technology Readiness Level

MYX has developed many blends that were tested at TRL4 by MYX and ISQ. TRL5 has been reached with injection moulding trials at LBRT's facilities with materials T23088 (PHBH, PLA) and T23089 (PHBH, PLA, Cork). Further up, demonstration at TRL6 is ongoing for the olive oil cap, whereas it is momentary in stand-by for the limoncello bottle closure.

The microwave technology has increased from TRL4 in month 12 to TRL5 in month 24. Pending upscale in WP6 to TRL6.

2.2.3 Technical progress and highlights up to month 24

By month 24 MYX has successfully developed a bio- based plastic in accordance with GA requirements. The final blend T23089, composed of PLA, PHBH, and cork, stands out for its remarkable characteristics that cater to a variety of needs in the packaging industry.

Thanks to the blend of PHBH and PLA, the material excels in processability. This combination ensures smooth extrusion and injection moulding, making it easier to handle during production. The inclusion of cork as a filler integrates seamlessly with the biopolymer matrix, resulting in minimal increases in torque and melt pressure, which contributes to efficient processing.

The blend also demonstrates excellent chemical compatibility among its components—PLA, PHBH, and cork. This ensures that the cork is evenly dispersed throughout the biopolymer matrix. The natural wettability of cork enhances its interaction with the biopolymers, resulting in a consistent and homogeneous final product.

Moreover, the blend maintains good thermal stability, allowing it to endure the temperatures required during processing without compromising its integrity. Balancing mechanical strength with flexibility, the blend is well-suited for various applications, especially in packaging, where these attributes are essential.

This blend, along with T23088, has been tested also at LBRT facilities to produce TDZ’s olive oil dispenser and bottle closure. The trials revealed some difficulties in the extraction of the samples from the mould, creating cracks and deformities.

Figure 3. Olive Oil Bottle Closure Samples produced with T23089.

Up to month 24, the microwave equipment for the preheating of pellets has been conceptualised, designed, manufactured, built, and tested. The system consists of a specialised hopper designed by IDE that allows the inclusion of a waveguide as a microwave input, two pneumatic valves to stir the pellets, four temperature sensors to effectively control the heating procedure, an input on top for a hopper loader, and several microwave leakage preventive measures, such as a specially designed cap, metallic chokes, absorbing gaskets and leakage detectors. The system also contains the microwave generator (magnetron) and its power source, a control box that contains elements such as a PLC, relays and thermomagnetic protections, and a water chiller to prevent the magnetron from overheating. The box also presents an HMI screen that allows an easy and intuitive manipulation of the system, as well as an emergency stop button.

The control system manages the different actuators to automatically preheat the pellets homogeneously, in a precise way, and fast, while also ensuring the safe operation of the microwave equipment by stopping every component if either microwave leakage, a short circuit, or overheating of the magnetron is detected. Lastly, the system also contains a microwave tuner to ensure maximum delivery of microwave power to the pellets, while minimising the reflected power towards the magnetron.

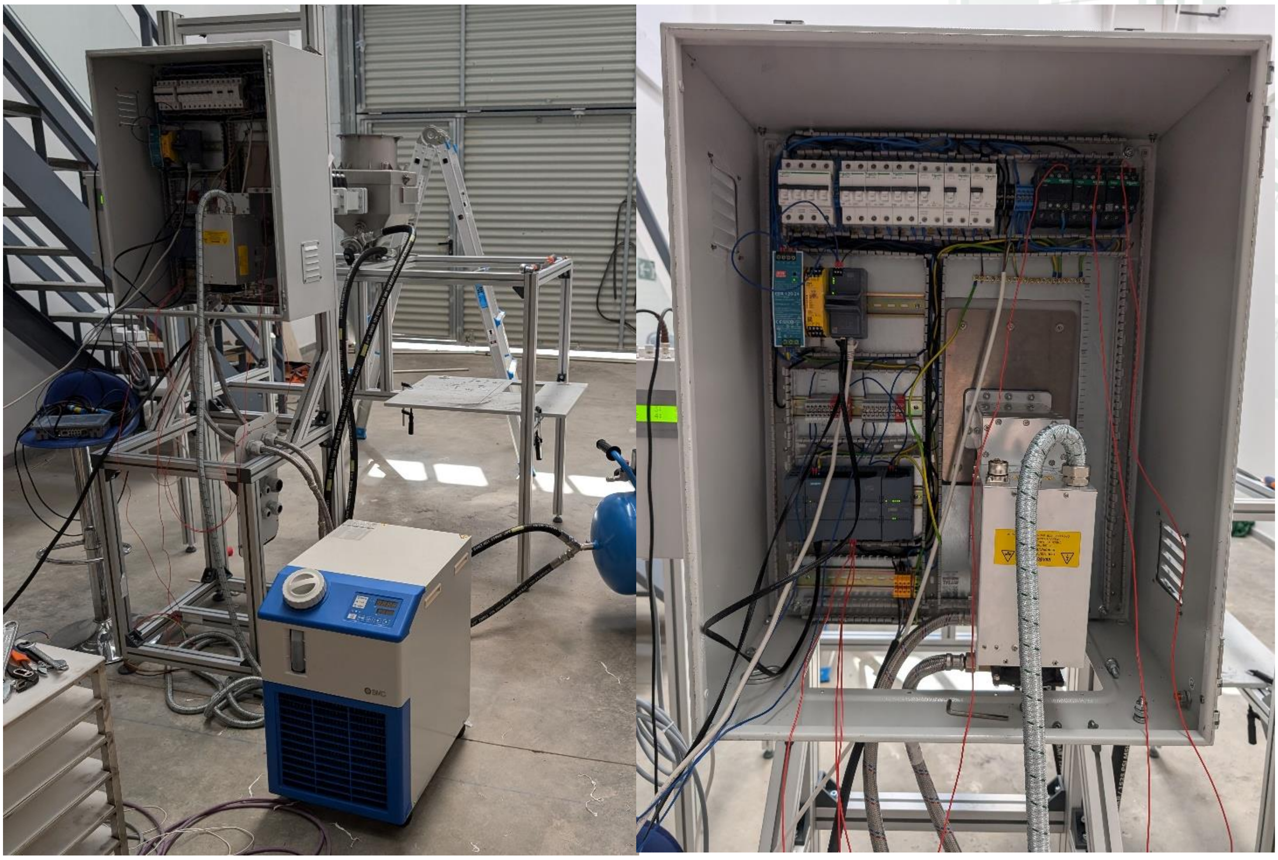

Figure 4. Whole WP4 MW setup including the control box, hopper, magnetron and water chiller (left), and closeup of the box (right).

2.2.4 Technical gaps and challenges

LBRT conducted injection moulding tests using material T23089 provided by MYX, focusing on the pourer component of the olive oil bottle, typically made from LDPE. The tests revealed significant challenges due to the material's excessive rigidity and low fluidity, which affected the moulding process. These issues caused difficulties in removing the pourer components from the mould, leading to deformation and breakage due to high pressure requirements. The test results indicate that T23089's current formulation is to be revised for the pourer, prompting MYX to adjust the PLA content to achieve a softer, more mouldable material.

Similarly, LBRT has done sample production of the screw cap of the bottle of oil, facing unique challenges due to different processing requirements compared to standard materials. Specifically, material T23089 proved too rigid for the screw cap, typically made from more flexible polymers like PP or aluminium. Additionally, MYX materials required processing at a lower temperature (140°C) and a longer cycle time than LBRT's standard setup. The existing core pin system in the mould, designed for higher temperatures and shorter cycles, cooled too quickly, causing blockages and incomplete filling. These issues resulted in suboptimal component quality, though T23088 showed slightly better aesthetic results.

Due to the criticises seen during the production of the oil bottle closure, prototypes of the Limoncello cap are yet to be produced.

2.2.5 Critical process steps

1) Definition of the Blend: MYX has successfully formulated T23089, a blend with a polymeric base made by PHBH + PLA, and a natural filler of cork, in line with the proposal requirements. (TRL 4)
2) Characterization: ISQ conducted mechanical and physical characterization test on the blend. (TRL4)
3) Injection trials at LBRT: LBRT conducted an injection moulding test to verify the printability of T23089, obtaining a positive output. (TRL5)
4) Olive oil dispenser and cap sample production: even with difficulties, samples of the pourer and the bottle closure for TDZ’s olive oil have been produced. (TRL6)

2.2.6 Delays and their reasons

Due to challenges in injection moulding EVO oil bottle closures, and after MYX adjusted the T23089 formulation, final product testing is set to begin in M25. Concurrently, the mould for the limoncello cap will be developed. The recent acquisition of Labrenta Srl by Guala Closures Group has caused some delays in the planned activities. In the next phase, LBRT will conduct critical food contact tests on the samples according to UNI EN 13342 and MOCA standards to ensure safety and compliance. Additionally, LBRT will assess the biodegradability, compostability, and permeability of the closures in collaboration with project partners to verify their environmental impact and performance. These tests aim to confirm that the closures meet food safety regulations and sustainability goals.

With respect to the microwave system, it will be finished and delivered to LBRT facilities by the end of M24.

2.2.7 Technological focus in next semester for each task

The general status of the process steps can be derived from table 2. The following work is planned in the next semester:

Task 4.1: Concluded

Task 4.2: On going

Subtask 4.2.1 Sensors Installation and data management: On going. Sensors will be installed at LBRT facilities by M25.

Subt*ask 4.2.2 Microwave enhancement:* system manufacturing and setup finished. Finishing touches to the control algorithm and sending everything to LBRT facilities.

Subtask 4.2.3 Tooling designs and adaptations: Concluded.

Task 4.3: Concluded, even if some adaptations are still made to carry out a better output. See Deliverable 4.2 for specifications.

Task 4.4: On going. Trials for the bottle of olive oil have been made, Limoncello caps are yet to be tested. See Deliverable 4.4 for specifications.

Task 4.5: On going. Tests and validations have been performed on the material, but they are pending on the final product.

Table 2. Process steps and responsibilities in WP4 [M24]. Legend: P=planned; O=ongoing; V=validated; I=implemented, F=Finished

Work package	Resp.	Material	Core Technologies / process steps	Partner	Status
WP4	**LBRT**	**Bio-plastic**	➢ **Biocomposites optimization / production** ➢ **Injection moulding** ➢ **Sensors (smart feature 1)** ➢ **Microwave enhancement (smart feature 2)**	**MYX** **LBRT** **IRIS** **IDE**	**V** **O** **P** **V** **I**

2.3 SOA of WP5 "Wood composite material production"

2.3.1 Materials concept for wood plastic composites and evolution of the extrusion process

The requirements for wood-based composite materials for bearings have been defined in D5.1. For the demonstration of the project results in WP5 a bearing was chosen, which will be used in an injection moulding machine at Labrenta. Moreover, a general cylindrical bearing shape was chosen that will also enable benchmark testing and comparison with state-of-the-art materials and bearings.

2.3.2 Technology Readiness Level

The microwave technology has increased from TRL4 in month 12 to TRL5 in month 24. Pending upscale in WP6 to TRL6. Moreover, the tribological behaviour of several developed wood composites (loop 3) derived from the combined extrusion-press moulding process showed good sliding performance and will be further used in WP6.

2.3.3 Technical progress and highlights up to month 24

Up to month 24, the microwave equipment for the extrusion of the wood composite has been conceptualised, designed, manufactured, delivered and successfully tested. The system consists of a microwave cavity specially designed to include an extrusion barrel and a susceptor system. The susceptor system contains a graphite/SiC half-cylinder that moves up and down: when up, the susceptor helps kick-off the preheating of the barrel; when down, the barrel and material heat up by themselves. The system contains several protection measures, such as sensors to detect the successful closure of the door, a stop mechanism activated when the water chiller isn’t working properly, etc.

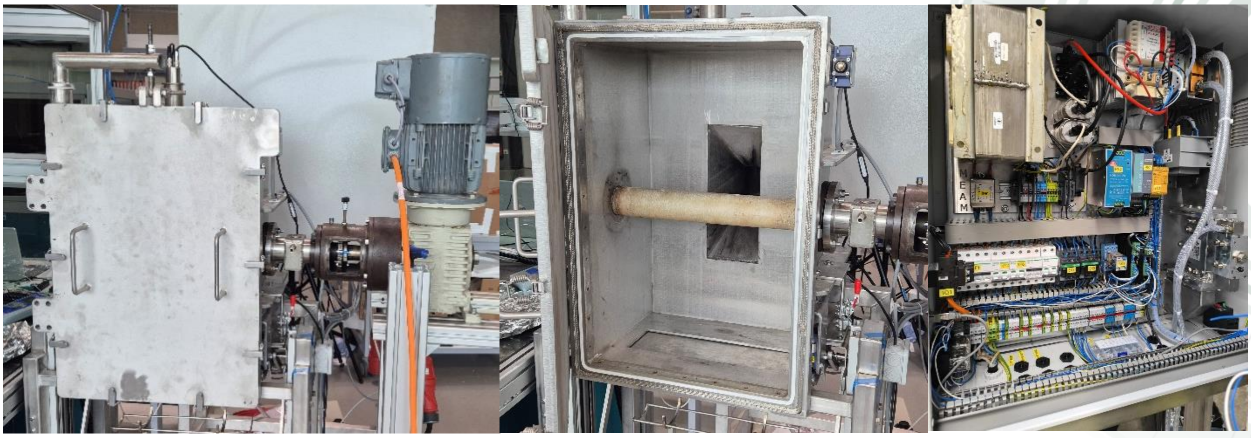

Figure 5. Whole WP5 MW setup, including the MW cavity with the screw motor (left), the open MW cavity with the specialised MW-transparent barrel (middle), and the inside of the control box (right).

During the testing phase, it was found that the SiC susceptor heats the barrel in a more uniform way with less power, and the material inside the barrel reaches a temperature so that the extrusion process can be performed successfully. Moreover, a first run with the wood-composite material was successfully performed.

2.3.4 Technical gaps and challenges

Up to M24 technical gaps cannot be identified during production in lab scale environments. The challenge pre-products and finally moulded components will be further investigated in WP6.

2.3.5 Critical process steps

Up to M24 critical process steps cannot be identified. A complete process chain could be setup in lab scale. However, the reproducibility has not been investigated so far. This will be done in WP6.

2.3.6 Delays and their reasons

With respect to the microwave system, it was delayed due to complications in the manufacturing process, as well as in the shipment to FHF’s facilities. Finally, the prototype was delivered in July.

2.3.7 Technological focus in next semester for each task

The general status of the process steps can be derived from table 3. Up to M24 all planned activities were finished, since all single tasks were completed. The process to produce wood composites by extrusion and press moulding is viable and TRL5 is achieved. The work planned in the next semester will be done within WP6 and will concentrate on the upscale of the process and recyclability of the wood composite material. Moreover, tests of wood composite parts in industrial related environments are planned at LBRT facility.

Task 5.1: Wood composites materials specification definition

Subtask concluded and results are described in [D5.1].

Task 5.2: Upgrades and modifications of equipment in manufacturing lines

Subtask 5.2.1 Development of new compounds using mostly renewable resources:

Subtask completed. More than 30 different compounds were investigated within 3 different material loops. The best suited mixtures showing good performance will be selected for further trials and upscaled in WP6.

Subt*ask 5.2.2 Microwave enhancement:*

Subtask completed. A description on the MW design can be found in report [D5.2]. The MW system was successfully delivered and connected to the extruder at FHF. The sensors provided by IRIS collect the data of energy consumption, both motor and the microwave. These data will be transferred to the ICT platform.

Subtask 5.2.3 Tooling designs and adaptations:

Subtask completed. Tools for the press moulding of plates and net-shape cylinders were designed and manufactured by NCC. The moulds were approved by using a selected wood compound mixture in the process. Morte details on the press moulding can be derived from [D5.3]. The moulding process and final machining will be used to make the sliding bearing parts for LBRT.

Task 5.3: Conclusion of design of WC bearings for demonstration at LBRT:

Subtask completed: Design for WC bearing to be tested at LBRT is defined.

Task 5.4: Characterisation of a selection of WC composites and their base materials at ISQ/UBRIS according to the proposed test matrix by ISQ:

Subtask completed: Testing of selected wood composites was performed according to the test matrix by using standards. A comprehensive summary on the properties can be derived from [D5.4].

Task 5.5: Tribological characterisation:

Subtask completed: Three material loops of WC materials investigated in pin-to-disc tests. Identification of promising wood composites material candidates for slide bearing components. Results can be derived from D5.5.

Table 3. Key-process steps and responsibilities in WP5 [M24]. Legend: P=planned; O=ongoing; V=validated; I=Implemented, F=Finished.

Work package	Resp.	Material	Core Technologies / process steps	Partner	Status
WP5	**FHF**	**Wood composite** **(WC)**	➢ **Modification raw materials / Compounding** ➢ **Extrusion WC** ➢ **Adapted microwave system (smart feature 1)** ➢ **Inline monitoring w. sensors (smart feature 2)** ➢ **Press moulding WC/Sample machining** ➢ **Quality control WC** ➢ **Tribo-testing WC**	**UBRIS** **FHF** **IDE** **IRIS** **NCC** **FHF/NCCFHF FHF** **ISQ/**	**F** **F** **F** **F** **F** **F** **F**

			Characterisation	**UBRIS**	**F**

2.4 SOA of WP6 "Upscale production and demonstration"

Due to some delays in the setup of all value chains and integration of smart features, Task 6.1 has been extended until M27 (as described and justified in the last GA amendment).

The objective of WP6 is to achieve reproducible manufacturing and to provide prototypes for the testing and characterisation by the end-users under relevant environment (ZAG, TDZ, LBRT). Another topic will be the analysis of the product's end of life as well as their next use to ensure the circular economy aspect of the project. Investigations will be performed on recycling and refurbishment of the new developed bio-based materials.

This work package will set up a viable process chain for all use cases to obtain TRL6. It starts in M25 and to be initiated, it needs the outcome and results from WP3-WP5 at TRL 5. Thus, no information will be reported or updated before month 27.

Table 4. Key-process steps and responsibilities in WP6.

Work package	Resp.	Material	Core Technologies / process steps	Partner	Sector/ Application
WP6	**IDE**	**Improved materials from WP3-WP5**	**Process Scale-up** **Demonstration** **Viable process chains WP3-WP5 to achieve TRL 6** **Recycling, refurbishment process**	**Partners from WP3-WP5**	**All referred in WP3-WP5 tables**

3 Risk analysis and Countermeasures planned

3.1 Description of risks

The risk management is one tool to identify critical technical issues early in the project which might cause delays or deviation from the original project scope. A table of general risks was already reported in the GREEN-LOOP proposal and is a general list with preliminary risks and mitigation measures. The risk categories cover technical risks, the demonstration of materials, organisational risks and management risks. After a review, it is stated that these general risks are still effective during the whole running time of the project and cover all WP's.

3.1.1 Risk analysis WP3

The active risks associated with WP3 are shown in Table 5. Risks that have been resolved are not included for brevity.

Table 5. WP3 risk table, correct at M24

No	Potential Risk	Possible Damage	Occurrence Probability		Consequence		Risk Level	Countermeasure	Resp.
1	Not expanding on SOA	Using existing work to potentially steer the work to be conducted	Unlikely	3	Negligible	2	Low	Ensure that an in depth lit search is conducted in all areas	All
2	Engagement of partners	Risk of losing partner or suitable engagement within the WP	Very Unlikely	1	Significant	5	Low	Monthly drumbeats	All
3	Saleable/Viable product for material	Material not able to be used within any applications	Unlikely	4	Marginal	3	Low	Engagement with C&I and other external stakeholders	All
10	Clash between LCA & performance considerations	Best formulation yields low sustainable characteristics	Very Unlikely	2	Negligible	2	Low	KPI's to drive decisions and sustainability focus and performance requirements	All
14	Ultrasound adaptations	Low performance in the process	Very Likely	7	Significant	5	Moderate	Identified other applications for the ultrasound	IRIS, NCC
15	Test methods not best suited for some parameters	Test results are not sufficiently meaningful	Very Unlikely	1	Significant	5	Low	Modify test method	ZAG
16	Not able to deliver to Grant Agreement	Material formulation path diverges to GA, KPI's not met	Very Unlikely	2	Significant	6	Low	Recoping activity and critical path identified to identify materials to suit	NCC
19	External compounding partners timescale not aligned to projects	Delay in trials	Likely	5	Critical	7	Moderate	Other partners identified, but at a higher cost and timescale. Mitigate by keeping in regular contact with current partner.	NCC, UBRIS
21	Rubber crumb from devulcanisation	Limits the adoption of US to the	Very Likely	8	Critical	8	High	Potential application in production of	IRIS

	trials not suitable for compounding	manufacturing process					🟥	coatings to rubber panel	

Most risks are judged to be low a low risk level. Those at moderate or high risk are discussed below:

Risk 14 – Adoption of ultrasound into the manufacturing process has presented challenges when applied to lignin extraction. The mitigation was to focus solely on applying ultrasound to rubber devulcanisation and not lignin extraction. The increased effort from IRIS on rubber devulcanisation has reduced UBRIS’s involvement in this task and allowed UBRIS to investigate a second method for utilising lignin within multilayer coatings.

Risk 19 – To move to larger scale compounding a new external partner was required with an internal mixer of sufficient scale to produce 10-20 Kg per batch size. A risk was identified that the progress of the compounding work would then be dependent on a stakeholder outside of the Green-Loop consortium. Effort was undertaken to develop a solid relationship with the compounding partner through regular email and phone contact. In this way the work has been keep on schedule and no delays have occurred to date.

Risk 21 – The compounding work highlighted that using a small rubber crumb as an input material was not suitable for compounding within an internal mixer. This had a knock-on effect to the ultrasound/DES devulcanisation trials which had been a feedstock material with a small crumb size. Experiments have now been carried out with larger crumb sizes to try and mitigate this risk. The compounding trials to test these materials are still ongoing. If the compounding trials are successful then this risk can be closed. If they are unsuccessful then this could limit how ultrasound can be incorporated into the WP3 bio-rubber value chain.

3.1.2 Risk analysis WP4

The active risks associated with WP4 are shown in Table 3. Risks n° 5 and n°6 have been resolved, thus are not analysed.

Table 6. WP4 risk table, correct at M24

N°	Risk Category	Potential Risk	Possible Damage	Occurrence Probability		Consequence		Risk Level	Countermeasure	Resp.
1	Technical	Microwave integration is not fully non invasive process	Possible modification on injection machine is needed	Likely	5	Marginal	3	Moderate	Detailed study and advanced simulations to find different and innovative solutions	IDE, LBRT
2	Technical	MW coupling to compound is not possible	No improvement in heating system	Unlikely	3	Crisis	9	Moderate	Search for alternative additive, vary concentration	IDE
3	Technical / Field Experience	Bio material with lower performance characteristics than expected (ex. flexural modulus)	KPIs cannot be reached	Unlikely	3	Crisis	9	Moderate	Investigate, how to improve	MYX
4	Technical / Field Experience	Long time required for some tests/ redo some tests (ex. flexural modulus)	Mid-deadlines are not respected, can lead to late in the final deadline	Likely	5	Critical	7	Moderate	Search for equivalent alternative tests, change laboratory to run test	MYX
5	Pilot and Delivery/Material	Polymers or fillers are not available in the time expected	Some tests are not possible to run	Unlikely	3	Marginal	3	Low	Provide alternative materials	MYX

		due to shortage of supply								
6	Technical / Field Experience	Compounding process is not possible	Set-up or extruder modification	Unlikely	3	Critical	7	Moderate	Identify a possible innovative solution	MYX
7	Equipment/Material	Samples produced present some defects	Aesthetic / functional	Very Likely	7	Critical	7	High	Redo production of samples	LBRT
8	Project Management	Budget Allocation	Resource to reallocate	Likely	5	Significant	5	Moderate	Review options	MYX / LBRT
9	Technical / Field Experience	Bio material is not biodegradable or is not for food contact	KPIs cannot be reached	Likely	5	Crisis	9	High	Investigate, how to improve	MYX / LBRT

Made exception by two High Level Risk, all the remaining risks are considered Moderate:

Risk 1 – The hopper enhanced with MW technology has been designed on the LBRT one. Final evaluation will be done once the MW system will be installed at LBRT facilities.

Risk 2 – The MW demonstration has shown that the material formulated by MYX can absorb microwaves. Confirmation will arrive once LBRT will produce a sample with the use of MW system.

Risk 3 – MYX has formulated two performing materials currently used for testing in LBRT, T23089 and T23088. Some difficulties arose during production in LBRT due to the rigidity of the material. For this reason, MYX is actively recalibrating the % of the components in the blends. However, the material are complaint to the KPIs.

Risk 4 – MYX has already performed several mechanical ad physical characterizations along with ISQ, obtaining relatively rapid results.

Risk 7 – LBRT has encountered numerous difficulties in extracting the olive oil dispenser and bottle closures from their mould, due to the low flexibility of the material. If the problem persists, the shelf life of the product cannot be guaranteed.

Risk 8 – LBRT is considering moving some budget within category B but is unlikely.

Risk 9 – Due to delays, biodegradability, composability and permeability tests haven’t been made yet. The poor aesthetic of the samples should not compromise the testing; thus, the analysis can be done regardless.

3.1.3 Risk analysis WP5

The technical risks can be derived from the following risk table. Only one moderate risk can be stated in WP5 in M24.

Risk 1 – MW coupling to compound during extrusion was recently solved due to the specific adaption of the extruder barrel by using ceramic material instead of steel.

Risk 2 – This risk is still active since the enhancement of MW extrusion has not been approved yet. Risk 2 will be re-valuated end of M25 when the energy consumption is calculated by IRIS with the data derived from the process.

Table 7. Table 6: WP5 risk table, correct at M24.

No	**Risk Category**	**Potential Risk**	**Possible Damage**	**Probability**		**Consequence**		**Risk Level**	**Counter measure**	**Resp.**	**[illegible]**	**Status**
1	Technical	[illegible]	[illegible]	Unlikely	3	[illegible]	9	Moderate	[illegible]	IDE	**31.08.2024**	solved
2	Technical	[illegible]	[illegible]	Unlikely	3	[illegible]	9	Moderate	[illegible]	FHF/ IDE	**Moved to 30.09.2024**	active

3.1.4 Risk analysis WP6

No risks are identified, since the WP didn´t start yet.

4 KPI's achievements and progress

The technical management will also review the progress on the KPI's, especially with a focus on material properties and manufacturing. The specific results from the value chains and material's characterisation will prove if the KPI's can be achieved. The validation of the KPI list was accomplished in D1.6 by AIMEN within WP1 Task 1.3.

4.1 Review KPIs for use case 1: bio-rubber (WP3)

The relevant KPIs for the bio-rubber value chain can be found in Table 8 along with any comments about the progress achieved towards those KPIs.

Table 8. KPIs achievements in WP3.

Smart Manufacturing/ Environmental Impact/ Properties	Target Value acc. D1.6	Results/Evaluation/Remarks
Monitorization of production	>80%	Lab scale compounding and compression moulding energy monitoring completed. Results of this have been included in the WP2 LCA. Work ongoing to monitor energy consumption on larger scale processing equipment.
Ultrasound enhancement: (lignin production & rubber manufacture).	R: 15%	Current trials matching devulcanisation of commercially available product.
Circularity and sustainable ratio measurement	>90%	This will be address in WP6.
Reduction of waste	>85%	This will be address in WP6.
Reduction of CO2 emissions	25%	WP2 LCA has identified some hotspots in the manufacturing process. It is anticipated these will be reduced once consideration of industrial scale is carried out. This will be address in WP6.
Valorisation yield	>95%	Less than 1% waste in compounding and compression moulding at lab scale.
Recyclability – 80%	80%	Product uses ~50% already recycled material so high degree of confidence this can be recycled again. This will be established in work package 6.
Final product refurbishment rate	>50%	This will be address in WP6.
Compressive strength	> 80 MPa	Not yet tested. Requires larger scale compounding to be completed first to allow sufficient material to be produced.

Smoke production	<1.3 [m2/s]	To be assessed in large scale fire testing in WP6.
Thermal conductivity	< 0.3 W/mK	Not yet tested.
Shear bond strength	> 4.5 MPa	Not yet tested.

4.2 Review KPIs for use case 2: bioplastic (WP4)

Smart manufacturing – Will be demonstrated in WP6 when the data transfer works, and the ICT platform is established.

Environmental impacts – Will be demonstrated within WP6.

Material properties – Will be further evaluated when the pellets based on new formulations are processed via Injection Moulding.

Table 9. KPIs achievements in WP4.

Smart Manufacturing/ Environmental Impact/ Properties	Target Value acc. D1.6	Results/Evaluation/Remarks
Monitorization of production	**>80%**	**Tbd in WP6**
Manufacture enhancement with microwaves	**>20%**	**TbD in WP6**
Circularity and sustainable ratio measurement	**>90%**	**Tbd in WP6**
Reduction of waste	**>60%**	**Tbd in WP6**
Reuse of biomaterial	**50%**	**Tbd in WP6**
Recyclability	**100%**	**Tbd in WP6**
Compressive Elastic Modulus	**>2.5 GPa**	**Diverge slightly from original values**

Compressive Strength	**>80 MPa**	**Diverge slightly from original values**
Flexural Modulus	**>2.3 GPa**	**Diverge slightly from original values**
Flexural Strength	**>71 MPa**	**Diverge slightly from original values**
Tensile Elastic Modulus	**>2.3 GPA**	**Diverge slightly from original values**
Tensile Strength	**>49 MPA**	**Diverge slightly from original values**

4.3 Review KPIs for use case 3: wood composites (WP5)

The KPI achievements within WP5 can be derived from table 7.

Smart manufacturing – The smart manufacturing has been approved by integration of the MW system provided by IDE to the extruder (FHF). Thus, TRL5 could be achieved. The new process work and the smart feature power meter is already connected to the device to collect the energy consumption during extrusion. However, the data from the first MW-assisted extrusion process has so far not been transferred to the ICT platform (IRIS), and the energy consumption has not been calculated yet.

Environmental impacts – Environmental related topics will be mainly investigated in WP6 when the process runs in larger scale. WP6 will focus on small series production and increase the output of the extrudate. Other issues such as circularity of the wood composite product, reduction of waste and emissions as well as recyclability will be addressed.

Material properties – The mechanical and tribological properties were determined. The wear and friction coefficients without lubrication are very good as well as the compression strength. However, lubricated samples could not be tested so far. The best suited material from the type V270 showed the highest application potential and will be further investigated.

Table 10. KPIs achievements in WP5.

Smart Manufacturing/ Environmental Impact/ Material Properties	Target Value acc. D1.6	Results/Evaluation/Remarks
Monitorization production extrusion w/wo microwave	**>80%**	Data from extrusion, both with el. heating and with MW heating are available. However, the data is not transferred to the ICT platform.
Manufacture enhancement with microwave	**>20%**	Energy consumption has not been calculated yet. Data is available.
Circularity and sustainable ratio measurement	**>90%**	Tbd in WP6
Reduction of wastes	**>40%**	Tbd in WP6
Reduction of CO2 emissions	**28%**	Tbd in WP6
Recyclability	**70%**	Tbd in WP6
Refurbished level	**90%**	Tbd in WP6
Homogeneous mechanical properties (compression strength)	**-**	Sample V270: 62 MPa, s < 1 %
Retention of bio-lubricant	**5-10%**	Improvement cannot be determined compared to dry lubrication.
Wear coeff. (dry lubrication)	**< 10-6 mm^3/Nm**	Loop 3: < 0.005-0.002 mm^3/Nkm
Friction coeff. (dry lubrication)	**< 0.05**	Loop 3: < 0.2-0.15

5 Literature and patents

5.1 Literature review update M24 (WP3)

State-of-art for bio-rubber material production

5.1.1 Lignin Extraction & treatments (NIC)

The possibility of using guanidine-based DESs to enhance the microwave-assisted extraction of lignin from oil palm empty fruit bunches has been explored. Following the promising results, a screening has been performed to investigate the influence of pretreatment parameters such as time and temperature on the structure and properties of the extracted lignin. [YAA24]

The Organosolv method was used to extract lignin from *Reseda luteola L.*, achieving excellent thermal stability, radical scavenging activity and anti-UV protection upon optimising the extraction conditions. [ZOU24]

In order to convert extracted lignin into nanoparticles, a new method has been developed consisting of water steam explosion pretreatment to extract pure lignin, followed by antisolvent precipitation to form the nanoparticles. The results with Kraft and Organosolv lignin have been compared, and showed significant differences. [GIR24]

The potential of lignin extracted from *Pinus montezumae lamb*. after Kraft pretreatment toward the production of biofuels was explored. [GAR24]

Lignin extracted from coconut shell biomass via Kraft and Soda methods were compared as corrosion inhibitors in polyurethane. The Soda method provided both a higher extraction yield and better performance. [RAM24]

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5.1.2 Recycled rubber and devulcanisation with bio-derived additives (UBRIS)

Since the previous report in month 12, work has now focused on rubber devulcanisation by combining deep eutectic solvents and ultrasound technology. Below is a summary of available literature on these topics.

Ultrasonic methods

This technique involves using wave energy vibrations to induce cavities in the polymer matrix, leading to the scission of crosslinks (C-S and S-S bonds) in vulcanised rubbers. A study conducted by Isayev et al. in 2014 found that the efficiency of devulcanisation is influenced by the particle size or surface area of the rubber. They discovered that rubber particles with a size of 30 mesh exhibited a greater extent of devulcanisation and lower gel content compared to 10 mesh particles. This relationship was observed under the same ultrasonic wave amplitude and a temperature of 250 °C [ISA14]. Another interesting research by Sun and Isayev in 2008 showed a positive correlation between ultrasonic amplitude and the degree of devulcanisation. They also investigated the effect of processing oil on the devulcanisation of isoprene rubber (IR) and natural rubber (NR). In addition, they compared the devulcanisation of CB-filled IR and NR and found that the impact of CB loadings on the devulcanisation process differed between the two rubbers, suggesting that the rubber's stereoregular structure could affect the extent of devulcanisation [SUN08].

Chemical methods

The chemical method for rubber reclamation involves the use of various chemicals to break the crosslinks between rubber chains or prevent the recombination of sulphur linkages. Several chemicals have been identified as effective devulcanising agents, including sulfides, peroxides, amines, deep eutectic solvents, and ionic liquids. For example, the use of diphenyl disulfide as a devulcanising agent in combination with microwave treatment was reported by Vega et al. in 2007. The inclusion of DPDS facilitated the devulcanisation process, resulting in the production of squalene and triterpene compounds [VEG08]. In another study, thiosalicylic acid was used by Thaichaoroen et al. to reclaim vulcanized natural rubber (NR) through the mechano-chemical method. They found that 1 phr (parts per hundred rubber) of thiosalicylic acid achieved optimal devulcanisation, and its efficiency was comparable to that of DPDS [THA10]. Hydrogen peroxide (H_2O_2) was employed by Zhang et al. in 2021 for the devulcanisation of styrene-butadiene rubber (SBR). The combination of SBR powder, soybean oil, and H_2O_2 resulted in complete devulcanisation, as indicated by the 100% sol fraction obtained [ZHA21]. Benzoyl peroxide was investigated by Sabzekar et al. in 2015 as a devulcanising agent for sulphur-cured NR. They found that selective cleavage of crosslink bonds occurred at lower concentrations of benzoyl peroxide and a shorter reaction time of 2 hours was recommended to prevent main chain scission. The authors also discovered that a reaction temperature of 110 °C with a low benzoyl peroxide content of up to 4 phr significantly reduced the crosslink density of the rubber [SAB15]. Walvekar et al. in 2018 studied the devulcanisation of waste tire rubber using amines in combination with ultrasonic treatment. Tertiary amine (3-aminopropyltrimethoxysilane) demonstrated better results compared to primary amine [(n-diethyl-3-aminopropyl) trimethoxysiloxane] at a sonication temperature of 50 °C, with the treated rubber showing gel content ranging from 63% to 77% and 75% to 87%, respectively [WAL18]. Walvekar et al. also investigated the use of deep eutectic solvents (DES) as devulcanising agents. The DES comprised of zinc chloride:urea at mole ratios of 2:7 and 1:4, and the devulcanisation was carried out via ultrasonic treatment at temperatures of 30, 130, 150, and 180 °C. The DES with a $ZnCl_2$:urea ratio of 2:7 required a temperature of 130 °C for optimal devulcanisation, while a higher temperature resulted in bond reformation and decreased devulcanisation efficiency. On the other hand, a $ZnCl_2$: urea ratio of 1:4 required a temperature higher than 130 °C for better devulcanisation. The authors concluded that DES containing $ZnCl_2$ and urea was highly effective for the desulphurisation of rubber, with sol fractions exceeding 85% [WAL18]. Pyrrolidinium hydrogen sulphate ionic liquid (IL) was used in combination with microwave treatment by Seghar et al. in 2015 to devulcanise SBR. They observed a positive correlation between the sol fraction and microwave energy, and the addition of the IL further increased the sol fraction at microwave energy levels above 220 Wh/kg, confirming its role as a devulcanising agent [SEG15].

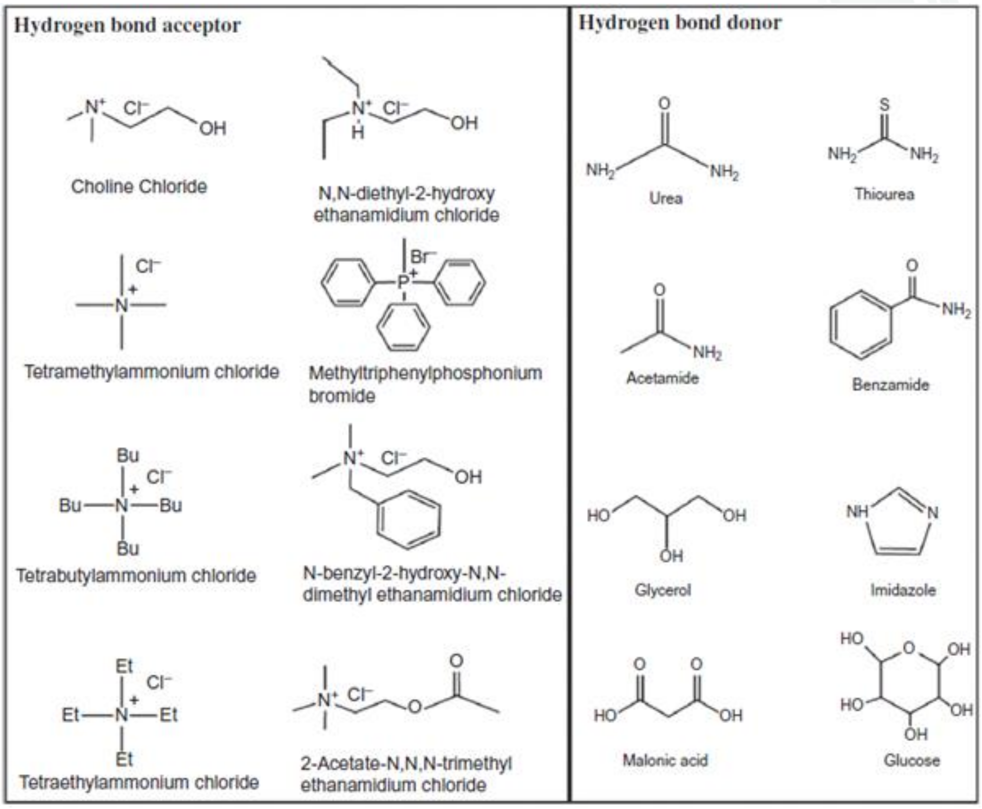

Figure 6. Most widely used HBD and HBA for producing DESs (reprinted [ARR19]).

Deep eutectic solvents

Abbot et al. in 2003 showed that when hydroxyethyltrimethylammonium (choline) chloride and urea were combined they remained in a liquid state even at normal room temperature. This feature was present despite the melting points of choline chloride and urea which are 302°C and 133°C, respectively; the mixture forms a eutectic system that melts at a significantly lower temperature than the melting point of its constituents. This substantial reduction in terms of melting point arises from the distribution of charges facilitated by hydrogen bonding between the molecules of urea and the chloride ion. The resulting liquid was discovered to possess intriguing solvent properties akin to those observed in ionic liquids (ILs). To distinguish these liquids from ILs, Abbot et al. came up with the term "Deep Eutectic Solvents" (DESs) which is still being used today [ABB03, ABB04].

DESs are comprised of a blend of organic substances, involving both a hydrogen-bond acceptor (HBA) and a hydrogen-bond donor (HBD), demonstrating a melting point substantially below that of either individual component. The formulation of numerous DESs is a straightforward process, achieved through the combination of an uncharged HBD (which could be an amino acid, sugar, carboxylic acid, amine, amide, or alcohol) and an HBA, for example, a quaternary ammonium salt. The common HBA and HBD options utilized for creating DESs are depicted in Figure 6. Among the available choices, choline chloride has been a frequent choice for the HBA in DESs due to its minimal toxicity [ARR19].

The resemblance between DESs and ILs encompasses traits such as being non-flammable, maintaining high chemical and thermal stability, being recyclable, having low volatility, and displaying significant solubilisation potential for various compounds. Additionally, much like ILs, DESs offer considerable adjustability through the manipulation of constituent compositions and their respective molar ratios. Noteworthy advantages of DESs over ILs include more cost-effective production owing to economical raw materials, streamlined preparation procedures devoid of intricate purification steps, and an environmentally conscious profile encompassing reduced toxicity and heightened biodegradability. As of now, DESs are emerging as a promising category of sustainable solvents that cater to a diverse range of applications, showcasing their rapid growth and potential [ARR19].

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5.1.3 Patent review update M24 (WP3)

Lignin Extraction & treatments (NIC)

In [FEL20], a method for extracting high-purity nano-sized lignin has been proposed, using as solvent a DES formed by an organic salt and an organic acid. The method consists in disrupting the biomass mechanically so it can create a mixture with the solvent, filtrating to separate the solids and liquids in it and letting lignin nanoparticles precipitate from the liquid portion.

In [PEN24], a method to extract and purify lignin from *dendrocalamus* l*atiflorus* under mild conditions is proposed. The method uses the synergy between organic acids and zinc chloride to carry out the extraction.

In [GUO24], a method of extracting lignin from corn straw is proposed. The authors have used a DES formed by 1,4-butanediol and the ionic liquid [Emim][OAc] and worked under reduced pressure and extracted the lignin from the primary filtrate via ethyl alcohol absolute. The resulting lignin has high purity and good free radical scavenging rate, which makes it a possible antioxidant additive for polymers.

References

[FEL20] Alban Felicity et. al., »Methods for Lignin Extraction«, US 2024/0158637 A1, filed 2020.

[PEN24] Zhang Peng et. Al., »Method for Separating and Purifying Dendrocalamus Latiflorus Lignin at Normal Temperature and Normal Pressure«, CN 117843989 A, filed 2024.

[GUO24] Zheng Guoxiang et. Al., »Extraction and Separation Method of Corn Straw Lignin«, CN 117866227 A, filed 2024.

Rubber-lignin composites and rubber recycling

Patent search tool Lens was used to search for relevant patents using the key words: rubber, lignin, vibration, fire, damping. The search time frame was limited to Aug 2023-Aug2024 to give an update on the patent search presented in Deliverable 1.2.

[WIL24] proposed as system for repurposing rubber crumb from ground rubber tyres. However, this process uses a very small crumb size (5 micron) which Greenloop has determined is not suitable for our production method via an internal mixer.

[ALE24] outlines a continuous rubber devulcanisation method to reprocess sulfur-cured rubbers. This process uses additives such as calcium carbide, silica, resin, oil, nucleophilic catalysts and co-catalysts. It does not use ultrasonic technology or deep eutectic solvents.

[CON24] describes a material system which is a rubber blend with one filler being a devulcanised rubber material. In this case the devulcanised rubber is used as a filler material rather than as the matrix.

References

[ALE24] Alerta Plateia, “Rubber Devulcanisation Method”, EP 3892674 B1, filed 2024

[CON24] Continental Reifen Deutschland Gmbh, “Vulcanisable Rubber Blend and Rubber Product That Can Be Produced from Same”, WO 2023/213363 A1, filed 2024

[WIL24] Coe William, “Inter-Penetrating Elastomer Network Derived from Ground Tire Rubber Particles”, US 2024/0101798 A1, filed 2024

5.2 Literature review update M24 (WP4)

State-of-art for bio-plastic material production

The global plastic sector is under increasing pressure to address performance, sustainability, and circularity in materials, especially in response to growing environmental concerns and the push towards a circular economy. This is particularly evident in Europe, where strategies like the European Strategy for Plastics in a Circular Economy have set ambitious goals for reducing plastic waste and promoting the use of sustainable materials. Among the various sectors affected, beverage packaging has emerged as a critical area for innovation due to its high consumption and environmental impact. In this context, biocomposites have gained significant attention as a promising solution for achieving these goals.

In 2024, biocomposites, which combine natural fibres or bio-based polymers with traditional or novel matrices, represent a cutting-edge approach to addressing the plastic sector's challenges. Recent advancements have focused on enhancing the performance of biocomposites to ensure they meet or exceed the functional requirements of traditional plastics. Key developments include:

1. **Material Performance**: Researchers have made significant strides in improving the mechanical properties of biocomposites, such as tensile strength, flexibility, and thermal stability. Innovations in the use of nano-fillers and advanced processing techniques have played a crucial role in these enhancements.
2. **Sustainability**: The incorporation of natural fibres like flax, hemp, and jute, as well as bio-based polymers like polylactic acid (PLA) and polyhydroxyalkanoates (PHA), has contributed to the sustainability of biocomposites. These materials are not only renewable but also biodegradable, reducing the environmental impact of end-of-life disposal.
3. **Circularity**: Biocomposites are increasingly being designed with end-of-life considerations in mind. This includes the development of materials that can be easily recycled or composted, thus supporting circular economy principles. The use of bio-based and recyclable matrices has been particularly influential in this regard.
4. **Non-Harmful Utility**: The shift towards non-toxic, bio-based additives and coatings has further enhanced the appeal of biocomposites. These additives ensure that the materials are safe for both human health and the environment, addressing concerns related to leaching of harmful substances.

The beverage packaging sector has seen significant integration of biocomposites as part of broader efforts to reduce plastic use and enhance sustainability. Biocomposite materials are being used to create bottles, caps, and other packaging components that are not only durable and functional but also environmentally friendly. Key trends include:

- **PLA and PHA-Based Bottles**: These bio-based polymers are increasingly used for producing bottles that are compostable under industrial conditions, reducing plastic waste.
- **Natural Fiber Reinforcement**: The inclusion of natural fibres in packaging materials has improved the mechanical strength and durability of packaging, making it suitable for widespread commercial use.
- **Recyclability and Compostability**: Companies are focusing on developing packaging solutions that can be easily recycled or composted, aligning with circular economy goals.

References

[ACQ21] Maria Assunta Acquavia et al. (2021), Natural Polymeric Materials: A Solution to Plastic Pollution from the Agro-Food Sector. Polymers (Basel). 13(1): 158.

[BAL22] Bala A. et al. (2022), Life Cycle Assessment of Biocomposites in the Automotive Industry. Journal of Cleaner Production. 367: 132917.

[CHO23] Cho S. et al. (2023), Advances in PLA-Based Biocomposites for Packaging Applications. Packaging Technology and Science. 36(4): 183-202.

[DEF22] Defossez A. et al. (2022), Biodegradable Biocomposites Reinforced with Natural Fibers for Food Packaging. Food Packaging and Shelf Life. 34: 100850.

[ELO23] Elomaa L. et al. (2023), Circularity in Biocomposite Materials: Challenges and Opportunities. Resources, Conservation & Recycling. 192 : 106968.

[FAZ23] Fazey F.M.C. et al. (2023), Assessing the Environmental Impact of Biocomposites: A Comprehensive Review. Materials Today Sustainability. 19: 100385.

[GRU21] Grubnic M. et al. (2021), A Review of Natural Fiber Composites for Structural Applications. Composites Part A: Applied Science and Manufacturing. 140: 106168.

[HAS24] Hassan M.A. et al. (2024), Biocomposites: Emerging Trends and Future Prospects. Composite Structures. 324: 116301.

[JOH23] Johnson S. et al. (2023), Recyclability of Biocomposite Packaging: Current Status and Future Directions. Sustainable Materials and Technologies. 30: e00399.

[KAR22] Karimah A. et al. (2022), A Review on Biocomposite Materials for Industrial Applications. Journal of Polymer Research. 29: 382.

[LIN21] Lin T. et al. (2021), PLA-Based Biocomposites: Recent Developments and Future Perspectives. Polymer Composites. 42(6): 2871-2890.

[MAR23] Martínez-Abad A. et al. (2023), Development of Sustainable Biocomposite Packaging Materials: A Critical Review. Industrial Crops and Products. 189: 115690.

[NOR23] Noreen A. et al. (2023), Advances in Biocomposites for Environmental Applications. Science of the Total Environment. 875: 162680.

[PET24] Petinakis E. et al. (2024), Durability and Performance of Biocomposites in Beverage Packaging. Materials and Design. 239: 134878.

[TAN22] Tan B.H. et al. (2022), Enhancing the Mechanical Properties of Biocomposites Using Nano-Additives. Advanced Materials Interfaces. 9(12): 2200051.

5.2.1 Patent review update M24 (WP4)

Search regarding PLA, PHBH, Cork patents has been made on Google Patents and on WIPO Patentscope.

In 2024, the use of polylactic acid (PLA), polyhydroxybutyrate-co-hydroxyhexanoate (PHBH), and cork in beverage packaging is advancing rapidly due to the industry's focus on sustainability and reducing reliance on traditional plastics. Patents filed this year highlight innovative applications of these materials that address both performance and environmental concerns.

PLA has been widely adopted in beverage packaging for its biodegradability and bio-based origins. Recent patents focus on enhancing its mechanical properties and extending its applicability in bottle production and caps, emphasizing fully compostable solutions that meet industry standards for sustainability.

PHBH is being explored for its flexibility and durability, particularly in flexible pouches and coatings for paper-based containers. Patents indicate a growing interest in PHBH due to its superior barrier properties, which are crucial for maintaining beverage quality over extended periods.

Cork is being revisited not just for traditional wine stoppers but as an innovative material in composite packaging. Recent patents highlight cork's potential in improving the sustainability of closures and as an insulating material in beverage containers, making it an attractive alternative to synthetic materials when combined with PLA and PHBH.

These biocomposites are pivotal in the shift towards more sustainable beverage packaging, offering solutions that are environmentally friendly without compromising on performance.

[WIPO24] WIPO Patentscope (2024), Patent WO2024112996 - PLA-based biodegradable packaging material. WIPO. Accessed from WIPO Patentscope.

[WIPO23] WIPO Patentscope (2023), Patent WO2023104928 - PHBH-coating for paper-based beverage containers. WIPO.

[WIPO22] WIPO Patentscope (2022), Patent WO2022100033 - Cork composite closures and caps for beverages. WIPO.

[GGL23] Google Patents (2023), US20231123478B2 - Biodegradable beverage bottles using PHBH. Google Patents.

[GGL24] Google Patents (2024), US20240123456A1 - Sustainable packaging using PLA and cork composites. Google Patents.

[GGL24] GlobalData (2024), "Patent activity in the packaging industry decreased in Q2 2024," GlobalData Patent Analytics. Accessed from GlobalData.

For Microwaves, a patent survey was performed through WIPO Patentscope.

In [ABI23], the use of microwave plasma treatment is discussed to enhance the properties of cellulose films, specifically to impart hydrophobicity. Oleic acid is utilised as a hydrophobic agent, and this treatment's effectiveness is evaluated by measuring the dynamic contact angle of water on the cellulose film surfaces. This innovative approach aims to improve the functionality of bioplastics, making them suitable for various applications, including packaging and agricultural uses.

5.3 Literature review update M24 (WP5)

A literature review was performed in the database Google Scholar by using search terms such as tribology, sliding bearings, biopolymer and extrusion.

5.3.1 State-of-art polymers for tribological applications (FHF)

In [BROI20] the nanomechanics and tribology of biopolymer called Arboblend V2 Nature (100 % biopolymer) was investigated. The material was manufactured by injection moulding. Tribological characterization against a bearing-steel counterface showed that for different processing temperatures, the increase of the applied load or the increase of sliding speed will produce an increase of the friction coefficient (µ) and wear. For a load of 10N the friction coefficient is 0.21-0.23 after 3000 cycles (fig. 6). These results showed that the chosen biopolymer is a candidate to substitute some popular fossil-based thermoplastics in numerous tribological industrial applications.

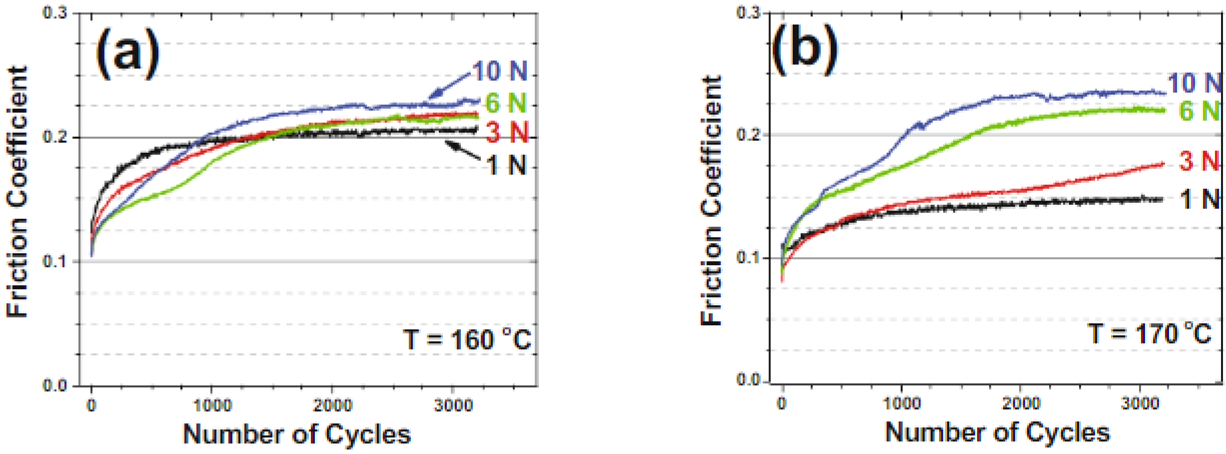

Figure 7. Friction coefficient vs the number of cycles for samples made at 160°C (a) and 170°C (b) at different applied loads [BROI20]

[BROI20] Esteban Broitman, Dumitru Nedelcu, and Simona-Nicoleta Mazurchevici (2020) Tribological and nanomechanical properties of a lignin-based biopolymer, e-Polymers; 20: 528–541.

5.3.2 State-of-art manufacturing of wood plastic composites via extrusion (FHF)

The more and more extruded filaments are further processed to samples by 3D printing besides the conventional injection moulding process. In [PAR24] the study focuses on the bio composite material which is developed from polylactic acid (PLA) and wood dust (NF). Wood dust was employed as fillers. PLA served as the matrix material, supplemented with the plasticizer polyethylene glycol (PEG) to enhance PLA processing. The PLA/NF filament was produced through the extrusion process, and specimens were prepared using 3D printing techniques. The test results demonstrated improvements in enhanced mechanical robustness and tribological performance of the PLA composite. The tensile strength of the PLA/wood composite was enhanced by 11%. The hardness value of the PLA/wood composite increased by 27% with the addition of wood as filler material. However, the introduction of wood as filler material led to degradation in physical properties. The water absorption test revealed a thickness change of approximately 14% for the PLA wood composite, whereas for pure PLA, it was 9%. Thermal stability tests showed that the temperature resistance of PLA improved to 250 °C with the introduction of wood fillers. Biodegradability tests indicated that PLA wood composite has the potential to address environmental concerns associated with conventional materials.

[PAR24] H.H. Parikh, S. Choksi, C. Prakash (2024), Development and characterization of eco-friendly extruded green composites using PLA/wood dust fillers. Sage Journals, Proc. Of the Institution of Mechanical Engineers, Part J: Journal of Engineering Tribology, Vol. 238, Issue 6, p. 676-686, https://doi.org/10.1177/13506501241233628.

5.3.3 State-of-art microwave curing process (IDE)

Microwave curing of wood composites represents a cutting-edge approach in material processing, offering significant advancements over traditional thermal curing methods. In the study by [KHO24], microwave irradiation was used to cure modified benzoxazine resin/ethylene–propylene–diene monomer rubber/Kevlar composites, demonstrating that microwave curing significantly reduces curing time while enhancing the thermal stability, glass transition temperature, and uniformity of the cured composites. In a different context, the use of vacuum low-temperature microwave-assisted pyrolysis was explored for substituting phenol with lignin-derived products in phenol-formaldehyde resins for wood modification [KAR23]. This research showed that microwave-assisted methods could improve the environmental footprint of wood modification by enabling the use of renewable resources without compromising the quality of the wood treatment. Another study [KAR24] extended this investigation to multiple wood species, confirming that up to 30% of phenol could be substituted by lignin-based products in phenol-formaldehyde resins using microwave-assisted techniques, achieving comparable results across various wood species.

References

[ABI23] N. Abidi, “METHOD OF MAKING CELLULOSE BIOPLASTICS”, US20230235136, 2023.

[KHO24] Hamidreza Khodaeian et al. (2024) Microwave curing of modified benzoxazine resin/ethylene-propylene-diene monomer rubber/Kevlar composite. Polymer Engineering and Science, vol. 64, no. 4, pp. 1887-1899, doi: 10.1002/pen.26679.

[KAR23] Johannes Karthäuser et al. (2023) Substituting phenol in phenol–formaldehyde resins for wood modification by phenolic cleavage products from vacuum low-temperature microwave-assisted pyrolysis of softwood kraft lignin. Cellulose, vol. 30, issue 11, pp. 7277-7293, doi: 10.1007/s10570-023-05295-5.

[KAR24] Johannes Karthäuser et al. (2024) Utilizing pyrolysis cleavage products from softwood kraft lignin as a substitute for phenol in phenol-formaldehyde resins for modifying different wood species. European Journal of Wood and Wood Products, vol. 82, issue 3, pp. 761-771, doi: 10.1007/s00107-024-02056-4.

Patents review update M24 (WP5)

A patent search was accomplished with PatBase program by searching for different key words and their combinations to identify relevant patents.

The search was covering the publication date between 1st August 2023 and 14th August 2024 (PD=20230801:20240814)

The search was done in the in the in the section title, abstract, claims (TAC):

TAC = bioplastic+extrusion+wood and PD=20230801:20240814 (3 hits)

Assessment: no relevance

TAC = bioplastic+extrusion+wear and PD=20230801:20240814 (no hits)

TAC = bioplastic+extrusion+ composite* and PD=20230801:20240814 (4 hits)

Assessment:

In IN202121013015A ("PROCESS OF MANUFACTURING BIOCOMPOSITE PELLETS AND MANUFACTURING METHODS OF BIODEGRADABLE PRODUCTS AND USES THEREOF") a biocomposite material is described which is a combination of thermoplastic biopolymer and agricultural residues such as wheat straws, paddy straws, bagasse etc. The said process follows two methods for making biocomposite pellets that are further used as a raw material for manufacturing different biodegradable products. The invention also comprises of different methods of manufacturing biodegradable products such as Extrusion, Manufacturing filaments, 3D Printing, Sheet/Film extrusion, Thermoforming, Blow Moulding, Injection Moulding. CN116874888A ("PREPARATION METHOD OF POLYLACTIC ACID-STARCH-BASED BIOPLASTIC COMPOSITE MATERIAL") relates to a production process and a production system of polylactic acid (PLA)-added starch-based bioplastic. The production process comprises the following steps: mixing and stirring the corn starch and the glycerol according to a certain proportion to prepare the starch-based bioplastic. And extruding the starch-based bioplastic molded particles at a certain rotating speed at a certain temperature by using a single-screw extruder. And extruding the prepared starch-based bioplastic and polylactic acid particles with a certain concentration in a molding press to prepare the composite board. A blending-polymerization-extrusion process is adopted, the production process is simple, the time is short, the operation is easy, and the product performance is excellent. CN114605795B ("PREPARATION METHOD OF BIOCHAR POLYLACTIC ACID COMPOSITE") describes bioplastics to be used for 3D printing.

Assessment: Especially biopolymers with agricultural residues are used, however, no anorganic fillers are used, material used for further 3D printing, composites cannot be used for friction applications

TAC = bioplastic+extrusion+wood and PD=20230801:20240814 (3 hits)

Assessment: no relevance

TAC = bioplastic+extrusion+fib* and PD=20230801:20240814 (5 hits):

In US2022204774 AA ("MELT EXTRUSION OF HEMP-BASED THERMOPLASTICS") the method of preparing a lignocellulosic biomass-based thermoplastic composition is described.

Assessment: no use for friction applications

TAC= extrusion and friction and composites and PD=20230801:20240814 (13 hits)

Assessment: no relevance

TAC= extrusion and sliding and bearing* and biomaterial and PD=20230801:20240814 (no hits)

TAC= extrusion and sliding and bearing* and biocomposite* and PD=20230801:20240814 (no hits)

6 Conclusions

The D1.3 report conducts an analysis of the technical status of Green Loop project especially focusing on the technical progress performed within work packages 3-5.

It can be stated that Technology Readiness level 5 was achieved within all value chains to achieve smart manufacturing. The integration of smart features such as the ultrasound system (IRIS) to WP3 and microwave systems (IDE) to WP4 and WP5, respectively, were successfully performed. Composites based on new developed formulations could be achieved. Moreover, the process lines were equipped with sensors to collect process data and to transfer it to the ICT platform. However, further work will be needed to set-up the platform and to use the data for energy calculations. In WP6 all processes will be upscaled to TRL6 with the pre-defined composites. Another focus will be on circularity as well as the environmental impact of the enhanced manufacturing.

The viability of all process lines could be approved by the manufacturing of composite prototype parts. Multifunctional rubber panels could be derived from the moulding at NCC. Bottle closures such as caps and dispensers were achieved by pellet extrusion followed by injection moulding at LBRT. Wood composite specimens for sliding bearing application could be produced by extrusion at FHF and final press moulding process at NCC. However, trials to improve the basic formulations of each process chain must be further developed in WP6 to optimize the material properties.

To provide the materials data, specific tests will be performed in each use-case to reach end users' specifications, which are part of the defined KPIs. WP3 will focus on vibro-acoustics transmission and fire-retardant properties. In WP4 tests are planned to assess the biodegradability, compostability, and permeability of the products. The friction and wear properties using cylinder shape as a standard will be in the focus in WP5.