GREEN-LOOP

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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

D1.2 – Scientific and technical plan and execution report

Version 1.0

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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 second D1.2 (M12) 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 pressing to panels are investigated in **WP3**. Research is mainly focused on two aspects of rubber composite manufacturing. On one hand, the recycling of rubber from tyre waste is being demonstrated by UBRIS (T3.2). On the other hand, lignin extraction from biomass waste is being investigated by NIC. Up to M12, the following results were achieved: Ultrasound-assisted lignin extraction was proved in the 1 kg scale by NIC. IRIS has also shown the advantage of its ultrasound system to devulcanise recovered tyre waste to enable its reuse (T3.5.3). Both activities are in progress. However, the lignin compatibility with the rubber is not been proven yet. A characterisation test matrix was developed by ISQ (T3.5). Moreover, UBRIS demonstrated a vibration transmissibility rig using test rubber pieces which will be used to assess the performance of the rubber panels once produced. Compounding to achieve lab-scale formulations will be accomplished by UBRIS and NCC with the help of a selected external partner by mixing pre-devulcanized rubber. The extrusion of bio-rubber compounds will not further be followed up, since up-scaling capabilities to large quantities are not available in the project.

Three risks that have been identified within the work package are now considered to be solved: suitable compounding facilities were identified. Ultrasound improves the lignin extraction and enhances the devulcanisation of rubber. Other risks that remain open are all considered to be either of low or moderate risk levels. These include the devulcanisation of the rubber feedstock, although this is looking to be mitigated with the source of pre-devulcanized rubber. Samples from three different suppliers have now been received in suitable quantities for lab-scale formulation. The transfer of the process from the lab scale (g's) to the upscale (kg's) remains a general risk to WP3. It is unknown, how the material will behave when it is processed in larger quantities.

On the other hand, novel bioplastic material for bottle closures is the focus of the **WP4** value chain. Product specification definition was successfully done in D4.1. MYX, as the supplier, produced the first blends based on polymers and fillers (T4.3), which are foreseen for injection moulding at LBRT facility (T4.4). Samples were sent to ISQ to test their properties. Microwave tests at IDE showed that the developed compound can be heated by MW coupling. Within T4.3, MYX and IDE are currently collaborating to create an AI model of the material based on scientific studies of the carriers that compose the bioplastic. A bioplastic composite is needed with a long shelf-life and low permeability.

One of the next steps within WP4 will be the use of the MW enhancement as an improved microwave heating system. The design of this MW equipment is nearly completed and ready to be reviewed by LBRT before being manufactured. The final design permits the conversion of a conventional hopper into a microwave- and hot-air-based preheating system.

Finally, the goal of **WP5** is to develop novel Wood Composite (WC) material for friction application, particularly for sliding bearings. Within the first project year, progress was made in the field of manufacturing and tribological testing of specimens from two material loops. In the first material loop, commercially available bio-compounds were used as reference material. In the 2nd loop, novel mixtures based on pre-selected biopolymers and fillers were investigated. Materials from both loops were sent to the partners UBRIS (T5.2.1) and ISQ (T5.4) for their further characterisation of mechanical, physical and chemical properties. However, it is not clear yet, if the defined KPIs (see D1.6) could be reached.

The extrusion process is working in a lab-scale environment, and samples could be achieved. Samples from the 2nd loop showed promising sliding performance (T5.5). Further improvements in the recipe and compounding are needed to increase the homogeneity of the composite. Simple geometries were chosen for the pin-to-disc tests. However, the final design of the sliding bearing, which will be integrated into Labrenta´s injection moulding machine, is still not fixed yet. The demonstration of the WC bearings at Labrenta requires a careful re-design of a machine component since a rolling bearing shall be replaced by a slide bearing. Deeper discussions between FHF, Labrenta and NCC about the most suited design are ongoing in T5.3.

The modification of the pilot plant extruder at FHF will mostly be finished in September within T5.2.3. In parallel several iteration steps were done to find out the most suitable design for the microwave equipment (T5.2.2), which will be integrated into the extruder system. Another issue was the search for a material with the required dielectric properties to replace the steel barrel. Some transparent materials could be identified and will be manufactured soon by FHF.

Up to now, the identified risks in WP5 are low or moderate. Some delay can be expected since the integration and qualification of the MW system is challenging and will take some more time than expected.

Upscale production and demonstration of all use cases will be performed in WP6. This work will be started earliest in M20 when the newly developed materials fulfil the defined KIPs. Results from WP3-WP5 will be the technical basis for scaling up the processes and to establish a viable process chain.

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Abbreviations

CMC - Carbomethyl cellulose

DX.X – Deliverable X.x

KPI – Key Performance Indicator

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 D1.2 and will be subjected to a set of two more revised versions.

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 every 6 months

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 (IDE). 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 will be done to set up feasible process chains for producing GREEN-LOOP 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"

Materials concept for bio-rubber and evolution of the process

Work has focused on two aspects of the rubber composite – 1) Recycled rubber and 2) Lignin extraction from biomass waste. The recycled rubber currently being explored is collected from tyre waste, this is a mix of natural and synthetic rubber. To reuse tyre waste, it must go through a devulcanization process. Two streams of devulcanization are being accessed in the project, initially work is ongoing using devulcanized rubber sourced from the commercial supply chain. Alongside this work is being carried out by IRIS and UBRIS to assess other devulcanization methods using ultrasound and green chemistry principles which may be more sustainable than the devulcanized rubber currently on the market. Lignin extraction work is ongoing with trials being carried out to utilise ultrasound technology to enhance this process.

Technology Readiness Level

Upscaled production runs are not scheduled until month 15 and therefore the technology is currently at TRL level 4. This is true of both the rubber composite production and the ultrasound-assisted lignin extraction.

Technical progress and highlights up to month 12

Lignin extraction to a total of 1 Kg has been completed by NIC using conventional methodologies. Alongside this ultrasound-assisted lignin extraction has been trialled by NIC and IRIS. This has shown a modest benefit to using the ultrasound process and further work is ongoing to improve the efficacy of this. IRIS has also shown the use of ultrasound to devulcanized recovered tyre waste to enable its reuse. UBRIS has developed and demonstrated a vibration transmissibility rig using test rubber pieces which will be used to assess the performance of the rubber panels once produced.

Technical gaps and challenges

During the initial phase of the research, the investigation into the potential utilisation of ultrasound in the rubber extrusion process for the purpose of augmenting devulcanization was undertaken. After conducting an inquiry, it was determined that this option was not feasible due to difficulty in obtaining a suitable extruder and technological limitations in utilising ultrasonic probes at elevated temperatures. Currently, there are ongoing investigations on two alternative approaches to devulcanization. 1) IRIS is currently conducting experiments with the application of ultrasound technology to facilitate the devulcanization process of rubber particles while immersed in an oil medium. 2) The UBRIS is currently investigating a devulcanization technique that employs principles of green chemistry. In addition, they have obtained commercially accessible devulcanised rubber, which is currently being utilised for the ongoing compounding experiments. After UBRIS/IRIS successfully produces adequate amounts of devulcanised rubber, it becomes possible to conduct a comparative analysis of its technical and environmental attributes in relation to the existing commercial product.

Critical process steps

There will be continued focus on ultrasound-assisted lignin extraction. The lignin will then be used in the formulation and upscaled production runs in Task 3.4 and 3.5.

Initial formulation trials at external compounding partner will commence on w/c 24th July. The results of this are necessary to inform the scale up in Task 3.5.

Pending results of the formulation trials an initial upscaled run will begin in month 15. This will provide sufficient material for the testing campaign to begin.

Delays and their reasons

Several samples have been generated, and UBRIS/NCC are currently anticipating a comprehensive report from the compounding supplier. The initial formulations proved to be ineffective; nevertheless, subsequent modifications were made to the formulation by using a certain quantity of virgin natural rubber. This adjustment enabled the creation of a compounded substance. The aforementioned samples are currently undergoing testing at UBRIS. Additional issues pertaining to the curing of materials have to be resolved, and these will be addressed in the subsequent phase of compounding.

Technology focus in next semester for each task

The general status of the process steps can be derived from table 1. The following work is planned in the next semester:

Task 3.3 – Trials will continue between NIC and IRIS to develop the ultrasound-assisted lignin extraction.

Task 3.4 – Formulation trials are due to commence imminently. Testing will be carried out on these samples to inform further optimisation trials.

Task 3.5 – Outcome of the trials run in Task 3.4 will directly feed into the upscaled material production outlined in Task 3.5. Once trials are completed material production will begin on the kilogram scale to allow the testing outlined in task 3.6 to begin.

Task 3.6 – As samples begin to be produced, testing will begin according to the test matrix already developed by ISQ.

***Table 1:** Status of process steps and responsibilities in WP3 [M12].*

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** ➢ **Sensors (smart feature 1)** ➢ **Ultrasound enhancement (smart feature 2)** ➢ **Compounding** ➢ **Press moulding** ➢ **Testing**	**NIC** **IRIS** **IRIS** **NCC** **NCC** **ISQ**	**O** **O** **O** **P** **P** **P**

2.2. SOA of WP4 "Bio-plastic material production"

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

MIXCYCLING has defined some blends to test different materials. The biomaterial has been compounded in MYX and samples have been produced. Specimens of the most promising blend (PLA blended with PHB and bio filler) have been sent to ISQ to test their properties such as strength modulus, flexural modulus, and tensile strength. Another production of the material is planned to select the best supplier for the polymer part.

For the microwave heating system, there are no advances in the SOA external of Green Loop compared with the previous report. Internally, in the project, the design is complete and ready to be reviewed by LABRENTA before being sent to manufacture.

Technology Readiness Level

Samples in MYX are developed at TRL4. Production of the material has run smoothly without any difficulties to highlight.

Regarding the microwave process MIXCYCLING has made a set of pellets that have been analysed by IDE, demonstrating the feasibility of the microwave usage. Milestone 6 will include these findings. The material is able to be heated through microwaves. But the hopper will also be ready to heat up with hot air in case the pellets need to be activated before being heated in the microwave. The final design will reach the TRL6 as expected.

Technical progress and highlights up to month 12

Material characteristics have been defined in D4.1. The characterization of the first samples sent to ISQ will give actual data to compare with the KPIs set in the proposal.

Task 4.2 "Microwave enhancement": The simulations have concluded, yielding favourable results for electromagnetic distribution and microwave shielding. The final design permits the conversion of a conventional hopper into a microwave- and hot-air-based preheating system. This innovation is currently being documented in a utility model to be submitted in month 12. In the same month, production of the system will also commence. It is anticipated to be completed by the end of month 13.

Technical gaps and challenges

For MYX an eventual technical gap is to be found once all the characterization analyses have been run.

For LBRT the challenge may lie in the production of the fruit juice cap since currently there is not a mould that can be adapted to the required shape. Probably a new mould will have to be ordered, produced and tested LBRT facilities.

Critical process steps

The microwave manufacturing will commence in September. Then, when everything is ready, the microwave control system will be adapted to account for the dielectric properties of pellets.

Production of samples at LBRT should start at M16 once the moulding for the project has arrived. It must be tested if the prototypes produced satisfy the requirements.

Delays and their reasons

Biodegradability analysis of the final product can take up to 9 months. Hence, D4.2 “Detailed description of final bioplastic and eco-design to be manufactured” may suffer a slight delay until MYX have all the key information regarding the bioplastic from the supplier.

Since the Deliverable of the microwave system for WP4 is in month 20 we do not expect any delay.

Similarly, D4.4 “Report for bioplastic validation trials and key findings” by LBRT should be delivered on time on M20.

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: *Subtask 4.2.1 Sensors Installation and data management:* planning between LBRT and IRIS hasn’t yet started. Subt*ask 4.2.2 Microwave enhancement:* The manufacture of microwave will be finalized and preliminary test of the system operating at LBRT facilities will be performed. *Subtask 4.2.3 Tooling designs and adaptations:* ongoing.

Task 4.3: MYX and IDE are currently collaborating to create an AI model of the material based on scientific studies of the carriers that compose the bioplastic.

Table 2: *Process steps and responsibilities in WP4 [M12].*

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** **LABRENTA** **IRIS** **IDE**	**O** **P** **P** **O**

2.3. SOA of WP5 "Wood composite material production"

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.

FHF performs many retrofit activities with the extruder to integrate the MW system from IDE.

For the microwave heating system, there are not advances in the SOA external of Green Loop compared with previous report. Internally, in the project, the system is already on manufacture, and it is expected to be ready in September 2023. However, since the barrel manufactured by FHF will not be ready until end of October, it is possible that the delivery will be delayed 2 months. The reason is that the system must be tested with all its components to ensure health and safety issues, before delivering ton FHF.

Two series of WC samples were produced and provided for various material tests including their chemical and physical characterisation. Tribological properties were determined in lab testing environment at FHF and these results were promising. For further optimisation of these materials, their thermal and wear stability was improved by using additional fillers. A second material series has already been prepared and experimental studies are on-going. Further materials characterisation at partners ISQ and UBRIS is in preparation as well. The design of WC bearings for Labrenta is intensively discussed. However, the final design is not fixed yet.

Technology Readiness Level

Until now the WC materials and processing parameters have been optimised. First tribological studies were conducted. The current TRL is estimated to be 2 or 3. Higher TRL levels will be reached when bearing performance data will be generated towards the end of the project.

Due to several restrictions on the practical applications of electromagnetic field on metal rods, the adaptation of the extrusion machine to a microwave cavity has been difficult. However, a suitable design has been developed to surmount these constraints. Currently, the system is being manufactured, and TRL6 will be reached as presented in the GA.

Technical progress and highlights up to month 12

Requirements for WC bearings have been defined.

Subtask 5.2.2 The microwave system is already in the manufacturer. The final fine tuning is being performed by IDE and FHF. The microwave system will be most probably ready in month 14. In this case, it is expected that there will be a delay in the final testing at the FHF facilities as explained before.

A first series of different WC material types was developed by using certain filler materials and by varying their concentration. Tribological results have been quite promising. Different WC bearing designs are already being discussed and proposed in D5.1.

Technical gaps and challenges

The first series of WC showed good tribological properties. Compared to state-of-the-art materials, which were represented by PA-based composite materials, the wear coefficients are higher and should be improved. The approach is to use base materials with higher thermal stability as fillers.

Due to a delay in the delivery of the samples to perform investigations on the dielectrical behaviour, barrel manufacture is delayed.

Critical process steps

Regarding the optimisation of WC material type delay can be expected if the second material series cannot fulfil the expected tribological properties. In that case another bio-based polymer needs to be purchased by FHF which allows the optimisation of the WC materials.

Regarding microwaves, the most crucial phase to date has been the finalisation of the design and manufacture. The dielectric laboratory test is also crucial. However, delays are expected.

Delays and their reasons

Regarding microwave activities the laboratory testing of samples manufactured by FHF has been delayed. The transporter misplaced the samples, resulting in their nearly two-month-late arrival at IDE facilities.

Technological focus in next semester for each task

The general status of the process steps can be derived from table 3. The following work is planned in the next semester:

T5.1: Concluded

T5.2: Optimisation of WC material types and processing by parameter studies. Sub task 5.2.2 IDE will have ready the microwave in September. The delivery will depend on the barrel manufacture at FHF facilities. It is estimated to be delivered by October. Then the set-up the microwave (MW) assisted extruder system will be installed. FHF will finish the refurbishment of the extruder machine to incorporate the microwave part. The retrofit activities with the MW-extruder at FHF need more resources than planned, however this effort in WP5 will contribute to achieve high quality parts in the planned series WC production in WP6.

T5.3 Conclusion of design of WC bearings for demonstration at LBRT.

T5.4: Characterisation of a selection of WC composites and their base materials at ISQ/UBRIS according to the proposed test matrix by ISQ.

T5.5: Tribological characterisation of second loop of WC materials and identification of promising candidates for bearing component testing.

Table 3: Key-process steps and responsibilities in WP5 [M12]. 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**	**UBRIS**	**O**
			➢ **Extrusion WC**	**FHF**	**O**
			➢ **Adapted microwave system (smart feature 1)**	**IDE**	**O**
			➢ **Inline monitoring w. sensors (smart feature 2)**	**IRIS**	**P**
			➢ **Press moulding WC**	**NCC**	**P**
			➢ **Surface modification / Machining**	**FHF**	**O**
			➢ **Quality control WC**	**FHF**	**O**
			➢ **Tribo-testing WC**	**FHF**	**O**
			➢ **Characterisation**	**ISQ/ UBRIS**	**O**

SOA of WP6 "Upscale production and demonstration"

There is no change in the planning or progress up to month 12. As foreseen in the proposal the activities in W6 relating to all use-cases will start in month 20.

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 M20 and to be initiated, it needs the outcome and results from WP3-WP5 at an appropriate TRL. Thus, no information will be reported or updated before month 20.

***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 risks are still effective during the whole running time of the project and cover all WP's.

3.1.1. Risk analysis WP3

Three risks that have been identified within the work package are now considered to be solved, with suitable compounding facilities identified and the rescope of work to adopt ultrasound within the lignin extraction and enhancing the devulcanization of rubber.

Other risks that remain open are all considered to be either of low or moderate risk. These include the devulcanization of the rubber feedstock, although this is looking to be mitigated with the source of pre-devulcanized rubber. Samples from three different suppliers have now been received in suitable quantities for lab scale formulation.

The transfer of the process from lab scale (g’s) to the upscale (kg’s) remains a risk to the work package, it is possible modification will need to be made to the process as it is currently unknown how the material will behave when processed in larger quantities. Countermeasures are being put in place to do this transfer in systematic steps with a gated test plan. A suitable commercial compounding supplier has been identified of which they will be sharing their experiences in compounding with similar materials.

The lignin compatibility with the rubber is yet to be proven, although it is hoped that the lab scale tests being performed in the short term will be give some indication to this. The mitigation to this, should the materials not be compatible, is to look at fractionation/modification of the lignin.

3.1.2. Risk analysis WP4

Identified risks in WP4 are low or moderate. The moderate risks are relating to compounding, product test time and achievement of the KPI´s. Possible delay might occur, since some of the standardized tests are time consuming. The performance and shape of bioplastic prototype parts will be tested at LBRT facilities. If the products do not meet the requirements countermeasures will be undertaken to achieve the final design and material´s performance.

3.1.3. Risk analysis WP5

Although first results from pin-to-disc tests were quite promising, it is still uncertain if the tribological properties of the materials may be sufficient. Maybe further optimisation cycles will be necessary.

Possible delay on the final design and set-up of the microwave heating system due to delay in laboratory test. Variability on dielectric properties of wood composites by variation of their composition. The system will be able to operate also with low microwave absorption due to the susceptor adaptation.

The demonstration of the WC bearings at Labrenta requires a careful re-design of a machine component, since a rolling bearing shall be replaced by a slide bearing. As a possible alternative demonstration, a general bearing design with a DIN standard geometry will be realised and tested as well.

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 and completion of the KPI list was done in D1.6 (see Annex A, table 2-4) by AIMEN within WP1 Task 1.3

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

Smart manufacturing – Work is ongoing to assess, how the rubber panel pressing can be monitored, no trials have yet been carried out. The ultrasound enhancement has been applied to the lignin extraction process. Further trials are planned to maximise its efficacy.

Environmental impacts – No recycling trials have been carried out yet, these are scheduled for WP6, however the product is produced with recycled rubber, and it is anticipated that a similar recycling process can be applied to the product. Reduction of waste and reduction of emissions will be assessed in the next semester when larger scale production is carried out.

Material properties – No testing for compressive strength, smoke production, thermal conductivity or shear bond strength has been carried out yet. Testing is scheduled to commence this semester.

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

Smart manufacturing – The KPIs regarding microwaves cannot be measured or estimated at this stage of the project. Sensors to monitor the production of samples with bioplastic have not been installed yet, nor the production itself has started.

Environmental impacts – All the environmental assessments shall be done once actual production starts.

Material properties – Tests to determine material properties are being currently carried out on different specimens. Additional analysis could be done in M12.

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

Smart manufacturing – Up to month 12 the defined KPIs cannot be measured, since the equipment is not completely developed and working yet. The MW (IDE) and sensors (AIMEN) are not integrated in the FHF extruder system. Therefor KPIs regarding microwave enhancement cannot be measured or estimated at this stage of the project. Moreover, the recycling rate cannot be determined, since the composite is still under development.

Environmental impacts – The process for WC production is running in lab-scale environment to manufacture test specimens. Products with defined design can be earliest expected in M20 when the MW assisted extruder is working properly in WP6. Then it will be possible to calculate the waste and CO_2 emissions.

Material properties - Wear and friction coefficients need to be further improved to achieve the required properties of the state-of-art material. Improvements will be done in iterative steps by changes in the manufacturing followed by tribological testing. So far two material loops were generated.

5. Literature and patents

5.1. Literature review update M12 (WP3)

State-of-art for bio-rubber material production

5.1.1. Lignin Extraction & treatments (NIC)

The impact of lignin extraction on the overall energy and chemical balances of the industrial Kraft mills has been determined. Black liquors and lignin cakes precipitated by CO2 were characterized to evaluate the organic and mineral balances. Lignin was precipitated at a pilot scale to quantify the consumption of chemical commodities. The chemicals and energetic costs were calculated for different production scale scenarios (tons per day of lignin). [BER23]

The recovery of kraft lignin from black liquor provides a valuable material that can be used as chemical or energy feedstock. The environmental consequences from a life cycle perspective of lignin extraction were under discourse. The results revealed that lignin used as a chemical feedstock is environmentally advantageous while a newly developed chemical recovery strategy was assessed. [MAR23]

References

[BER23] F. Bertaud, P. Ottenio, A. Aubigny, A. Dufour, "Recovering lignin in a real-case industrial kraft pulp mill: Pilot-scale experiment and impact on the mill commodities", ACS Sustainable Chem. Eng., vol. 11, 6311-6318, 2023.

[MAR23] A. Marson, J. S.M. Samec, A. Manzardo, "Consequential life cycle assessment of kraft lignin recovery with chemical recycling", Science of the Total Environment, vol. 882, 163660, 2023.

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

Lignin has been utilised as a filler in rubber composites in conjunction with silica nanoparticles. By modifying the lignin through a Mannich reaction with diethylamine the hydrophobisity of the lignin was increased to improve its intersection with the rubber. It was shown that up to 20% carbon black could be replaced with lignin without a reduction in mechanical performance [QUI23]. Similarly lignin has been incorporated into SSBR rubber as a replacement for silica, a limit of 20wt% lignin was found before reagglomeration was seen in the composites [CHO23].

Amine functionalised lignin has been explored as a replacement for a typical antioxidant 6PPD which is environmentally hazardous. While not a focus of the Green-Loop project this could be a tangential benefit to the rubber composites produced. The addition of the modified lignin also promoted the curing rate and cross-link density of the rubber [CHU23].

Lignocellulose has been used to replace carbon black in natural rubber composites to reduce the flammability and smoke emissions from natural rubber composites, this was achieved without impacting the mechanical properties. Unlike many other studies this paper used lignocellulose rather than extracting the lignin first [RYB23].

Lignin was incorporated into a SBR and BR rubber blend and a comparison between untreated and silane modified lignin was carried out. It was found that the silane modified lignin composites showed improved properties compared to the unmodified composites owing to the improved interaction between the lignin and the rubber [SEK23]

References

[QUI23] Jiabao Qui et. al., “Study on lignin amination for lignin/SiO2 nano-hybrids towards sustainable natural rubber composites”, International Journal of Biological Macromolecules, Volume 233, 2023

[CHO23] Soumya Ghosh Chowdhury et. al., “Evaluation of lignin as potential green filler in an optimally designed solution grade styrene–butadiene rubber (SSBR) based tyre tread compound”, Plastics, Rubber and Composites, 52:2, 63-74, 2023

[CHU23] June-Young Chung et. al., “Amine-Functionalized Lignin as an Eco-Friendly Antioxidant for Rubber Compounds”, ACS Sustainable Chem. Eng., 11, 2303−2313, 2023

[RYB23] Przemysław Rybinski et. al., “Effect of Hybrid Filler, Carbon Black–Lignocellulose, on Fire Hazard Reduction, including PAHs and PCDDs/Fs of Natural Rubber Composites”, Polymers, 15, 1975, 2023

[SEK23] Priyanka Sekar et.al., “Hydrothermally Treated Lignin as a Sustainable Biobased Filler for Rubber Compounds, ACS Appl. Polym. Mater., 5, 2501−2512, 2023

5.1.3. Ultrasound Enhancement of Rubber Manufacturing (IRIS)

The implementation of the ultrasound technology in the rubber manufacturing chain was carried out through the acoustically assisted devulcanization of wasted rubber. Chemical devulcanization includes the use of chemical agents (deep eutectic solvents, paraffin oil) to break in C–S and S–S bonds (e.g. cross-links) in the vulcanized network [SHA22]. Probe sonication is a simple method that involves higher energies to the rubber particles needed to promote cross-link breakage [VAH1]. The effects of sonication media, swelling media, sonication, power-time and chemical agent content on devulcanization percent, cross-link density, sol/gel fractions, it is being evaluated by IRIS.

References

[SHA22] Shabdani Marzie et al., “Recycling NR/SBR waste using probe sonication as a new devulcanizing method; study on influencing parameters”, RSC.adv.,12, 26264-26276, 2022

[VAH21] Vahdabtin M et al., “Using chemical agent in microwave assisted devulcanization of NR/SBR blends: An effective recycling method”, Resour. Conserv. Recyl. Adv., 179, 106045-106056, 2021

5.1.4. Patent review update M12 (WP3)

Filed patents

Patent search tool Lens was used to search for relevant patents using the key words: rubber, devulcanization, lignin, vibration, fire, damping

In [HON21] a rubber damping material is proposed using hydroxyethyl methacrylate modified lignin and natural rubber latex. Their approach is to graft the lignin and rubber together during the processing phase to produce a copolymer. In [CHE21] a reinforced rubber composite is produced using lignin as the filler. This lignin is modified with a molecule containing a carbon-carbon double bond. This modification allows for an improved interaction between the rubber matrix and the lignin filler. Their claim also includes an improved curing mechanism due to the lignin modification. This reduces the use of additional vulcanizing agents and allows for a greater replacement to lignin from carbon black.

Lignin has been applied as a sustainable filler into road tyre constructions in [MI21]. This includes incorporation of unmodified lignin and lignin modified with an amine containing compound. This utilises the lignin as an antioxidant in the composition. Lignin loadings up to 20 pbw have been utilised. [TAK15] describes the extraction of lignin for using in rubber moulding compositions. It describes the lignin extraction by a chemical extraction process. The lignin is then compounded into a rubber composite using at least one of natural rubber, butadiene rubber, and styrene-butadiene rubber. These are the rubbers typically using in tyre construction so will be present in the recycled tyre rubber used in WP3.

There are several patents which cover the devulcanization of rubber waste. [MOR22], [ARS21] and [DAM21] describe chemical methods for devulcanization. These focus on using a chemical agent to selectively break the cross-linking bonds in the rubber product without breaking the main chain bonds. [MAR22] uses a swelling agent and the application of heat (170°C) to achieve the same effect. This is applied on relatively large particle sizes between 5- and 10-mm. Rubber devulcanization can also be achieved using biotechnology. [DAV15] describes a method for rubber devulcanization using an aerobic bacterial process.

References

[HON21] Liu Hongchao et. al., “High-performance rubber damping material and method for preparing the same”, US 11661467 B2, filed 2021

[CHE21] Zhu Chenjie, “Modified Lignin Reinforced Rubber and Preparation Method Therefor”, US 2023/0135725 A1, filed 2021

[MI21] Jeong Da Mi et. al., “Rubber composition for sustainable tire having lignin compounds”, US 2021/0395497 A1, filed 2021

[TAK15] Murai Taketoshi et. al., “Lignin Derivative, Lignin Resin Composition, Rubber Composition, and Molding Material”, EP 3192825 A1, filed 2015

[MOR22] Morimoto Shouhei, “Devulcanized Rubber, Rubber Composition, and Pneumatic Tire”, US 2023/0192990 A1, filed 2022

[ARS21] Bin Mohd Ali Jinnah S Arshad, “Process for Preparing Devulcanized Rubber”, WO 2023/059179 A1, filed 2021

[DAM21] Justin Michael Damster et. al., “A process for devulcanizing tyre rubber particles”, NL 2029863 B1, filed 2021

[MAR22] Wirtz Marcel et. al., “Rubber Product Devulcanization Method and Product”, EP 4116336 A1, filed 2022

[DAV15] Staedler Davide et. al., “Process for Bacterially Devulcanizing Sulphur-Vulcanized Rubber Particles”, EP 3438244 B1, filed 2015

5.2. Literature review update M12 (WP4)

State-of-art for bio-plastic material production

The state of the art for microwave heating: A study [KIM22] analysed the energy utilisation of a pilot-scale microwave hopper dryer used to dry plastic pellets. The microwave-generated electromagnetic field was used to heat the plastic granules, and the results demonstrated that the microwave hopper dryer was more energy-efficient than conventional hot air dryers. At higher TRL levels, we have found also relevant information focused on Lumber, wood, chemicals, fossil fuels, foam, fibre, textiles, and other sheets or solids. Several procedures, including microwave-assisted drying, microwave-assisted vacuum drying, and microwave pasteurization/sterilization, utilise this technology [MAR23]. In reference [JEO22] authors introduce a microwave cavity on a large scale for uniform and effective plastic heating. The lower feeder measured 272 mm in height, 430 mm in upper diameter, and 58 mm in lower diameter. To assure uniform heating, the height of the cavity was adjusted based on the dielectric properties of the plastic.

<u>**References**</u>

[KIM22] Jae Kyung Kim et al. (2022), Analysis of energy consumption for microwave hopper dryer on a pilot scale. Drying Technology 41(7):1-12

[MAR23] Francesco Marra (2023), Combining computer aided food engineering and electro-heating applications as contribution to food processing sustainability. Front. Sustain., 17th April 2023

[JEO22] Sangjun Jeon (2022), Design of Large-Scale Microwave Cavity for Uniform and Efficient Plastic Heating. Polymers 2022, 14(3), 541.

5.2.1. Patent review update M12 (WP4)

No new patents regarding microwave processing of bioplastic have been found.

5.3. Literature review update M12 (WP5)

State-of-art wood composite material production

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

No significant progress was published elsewhere.

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

No significant progress was published elsewhere.

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

The combination of microwave heating and extrusion has yielded promising results in the last 12 months. Microwave Processing of Polymer Matrix Composites has been studied [NAI22]. Authors describes how microwave energy provides rapid and uniform heating, enhanced processing efficiencies, and the capability to create distinct material properties. In other research, Extrusion-based additive manufacturing (AM) technologies have been included. Microwave heating method is favoured over powder-based additive manufacturing techniques. The precise and controlled extrusion of materials is made possible by the high-power heat source, such as electromagnetic energy [ALT22]. In terms of wood materials, reference [MAS23] summarise the current state of knowledge regarding microwave treatment for wood modification and investigate the application of microwave energy to drying. This paper provides an overview of microwave technology's applicability in wood treatment.

References

[NAI22] Tejas Naik et al. (2022) Processing of polymer matrix composites using microwave energy: A review. Composites Part A: Applied Science and Manufacturing, 156, 106870.

[ALT22] Sadettin Cem Altıparmak et al. (2022) Extrusion-based additive manufacturing technologies: State of the art and future perspectives. Journal of Manufacturing Processes, 83, 607-636.

[MAS23] Fernando Mascarenhas et al. (2023). Microwave technology and its applications to wood treatment and modification. World conference on Timber Engineering, Oslo.

Patents review update M12 (WP5)

Patent search tool PatBase was used to search for latest patents using the key words such as extrusion, wood, lignin, fiber, fillers, bioplastic, composite, wear. No new patents regarding microwave processing of WC have been found.

6. Conclusions

At the very beginning of the project, Task 1.1 was responsible for kicking off the process of scientific and technical coordination. The D1.2 report conducts an analysis of the ongoing research projects that are being carried out in WP3–WP5 as well as the technological progress made in each of the use cases. Additionally, the successes and developments of key performance indicators (KPIs) within each technical work package (WP) are assessed and evaluated. In addition, a thorough analysis of the research literature and patents that have been filed is carried out as part of the Green-Loop project in order to determine the potential influence that these items may have on the research that is being carried out there. Through the TEAMS meetings, management was kept up to date on the latest technical developments on a regular basis. The leaders of the WP (Work Package) provide updates on the state of their efforts, including major technical outcomes, ongoing work, impending tasks, difficulties, and potential risks. The management team has identified several minor setbacks during the M12 phase and determined that the related risks are of a low-to-medium degree. Even if these difficulties exist, it is not believed that they will be a significant obstacle in the way of the project's completion.