ACT GROUP 2528

CREATING A CIRCULAR CHAIN FOR EUROPEAN MOSO BAMBOO

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CREATING A CIRCULAR CHAIN FOR EUROPEAN MOSO BAMBOO



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Executive summary

ArtEZ Centre of Expertise Future Makers (ArtEZ FM) has reached out to Wageningen University & Research for a group of master students to improve the circularity and sustainability of the European based bamboo value chain. Over the years, bamboo has become increasingly interesting as a sustainable source due to its fast growth rate and adaptability to harsh environments. Even more sustainability can be gained by reusing the residues of the bamboo processing system by extracting one of the natural most abundant substances: lignin. A compound that can embody many purposes, e.g. fuels, plastics and resins (binders). However, its complex structure poses a difficult challenge to define sufficient extraction methods. Still, to decrease the dependency on exhaustible sources, it is necessary to both shift towards sustainable options as to strive towards a more circular system by giving value to our waste.

The aim of this project is to evaluate different lignin extraction methods on their technical feasibility and their environmental, social and economic sustainability. In addition, the feasibility for lignin to be applicated as a binder will be researched. This is done by executing literature review and conducting interviews. This study is limited by time, making the performance of experimental research not possible. The main research question that will be addressed is: *'Can lignin from European-based Moso Bamboo residues be used as a binder for lamination to improve the sustainability of the European-based bamboo value chain?'*

The following extraction methods are analysed: organosolv, soda process and ionic liquids (ILs). While the soda process embodies a very simple process, organosolv and ILs involve many different options to choose from. The overview of the analysis on these extraction methods is given in table 1. It is concluded that for a short-term perspective, the soda process is the most suitable due to the available knowledge and large-scale applicability as well as environmental friendliness and economic profitability. For a long-term perspective, IL can be preferred as they show great potential for delivering high purity lignin and being environmentally friendly and financially profitable, though utilisation is still at an early stage.

Process	Simpleness	Tailor made potential	Purity	Environmental/Economical potential	Large scale	Applicated to bamboo
Organosolv	2	2	2	3	2	2
Soda	1	3	3	2	1	1
ILs	3	1	1	1	3	3

Table 1: Overview of results analysis extraction methods (1 being high score, 3 low score)

This short term/long term perspective is also mirrored in the conclusions of this report on using lignin from bamboo residues as a binder. On the short term, it might be preferred to choose a binder that partly still uses petrol derived chemicals for economic and technical feasibility, as the lignin formaldehyde resin. On the long term, complete biobased resins are advised for their sustainable

potential. Even though knowledge is limited nowadays, it will develop with adequate research conducted. The simplicity of the process and large scale will become more attainable through the development and availability to new techniques and accessibility to technology. To what circularity concerns, a lignin binder from bamboo residues must be favoured over a petrochemical one. When it comes to sustainability, it might be tempting to choose a petrochemical one over a biobased one. As their production process has been optimised over time, they often consume less energy, water, prime materials, etc. per unit produced. However, the difficult recyclability of these products and their (very) long degradation cycle makes of them a non-sustainable alternative on the long term, without forgetting that petrol-based products originate from a finite resource.

Table of contents

Contact details	ii
Executive summary	iii
Chapter 1: Introduction	1
Lignin: from a broad and close view	2
Knowledge and research gaps	3
Research questions	5
Methodology	6
Defining sustainability	7
Chapter 2: Technical feasibility extraction methods	9
Introduction	9
The organosolv process	10
Extraction method specification	11
Bamboo specific extraction	12
The soda process	14
Extraction method specification	14
Bamboo specific extraction	15
Ionic liquids	17
ILs families and properties	17
Extraction method specification	19
Conclusion	21
Chapter 3: Lignin as a binder	23
Lignin-formaldehyde resin	23
Completely biobased resins	25
Lignin-furfural resin	26
Lignin (alone) as a binder	27
Conclusion	28
Chapter 4: Sustainability and economic feasibility extraction methods	29
Description of the value chain	29
Asian-based bamboo value chain	29
European-based bamboo value chain	
Description of stakeholders included in value chain	33
Bamboo producer	
Local Portuguese population	
Non-Portuguese bamboo farmers	

Governmental institutions	34
Research institutes	35
Suppliers and transporters	35
Media	35
Customers and consumers	36
Environmental and economic performance of extraction methods	37
Organosolv	37
Soda Method	40
Protic Ionic Liquids	44
Sustainability of a binder	48
Comparison	49
Environmental impact	49
Economic feasibility	51
Social sustainability	53
Conclusion	53
Conclusion	55
SWOT Analysis of the recommendation	58
Strengths	58
Weaknesses	58
Opportunities	59
Threats	60
References	62
Appendixes	74
Appendix 1: Stakeholder longlist	74
Appendix 2: Comparison of organosolv and soda extraction for all impact categories	75
Appendix 3: comparison of organosolv and soda extraction per impact category	76

Abbreviations

A = Acidification AE = Aquatic Ecotoxicity ArtEZ FM = ArtEZ Centre of Expertise Future Makers BambooLogic = BambooLogic Europe BV DES = Deep Eutectic Solvents EOL = Ethanol Organosolv Lignin FEC = Fossil Energy Consumption FEP = Freshwater Eutrophication Potential FECP = Freshwater Ecotoxicity Potential FPMFP = Fine Particulate Matter Formation Potential FRSP = Fossil Resource Scarcity Potential GW = Global Warming **GWP = Global Warming Potential** HT = Human Toxicity HCTP = Human Carcinogenic Toxicity Potential HNCTP = Human Non-Carcinogenic Toxicity Potential IL = Ionic Liquid **IRP** = Ionising Radiation Potential LCA = Life Cycle Assessment LF = Lignin Formaldehyde LPS = Lignin Process System LUP = Land Use Potential MECP = Marine Ecotoxicity Potential MEP = Marine Eutrophication Potential MRSP = Mineral Resource Scarcity Potential NPV= Net present Value NVP = Net Value Product **OFP = Ozone Formation Potential** PF = Phenol Formaldehyde PIL = Protic Ionic Liquid SODP = Stratospheric Ozone Depletion Potential SPS = Switchable Polarity Solvents SQ = Sub-research Question SWB = Strand Woven Bamboo SWOT = Strengths Weaknesses Opportunities Threats RTIL = Room Temperature Ionic Liquid **TAP = Terrestrial Acidification Potential**

- TEP = Terrestrial Ecotoxicity Potential
- Tg = Glass Temperature
- WCP = Water Consumption Potential

List of Figures

Figure 1 Main monomers of lignin (Harmsen, 2016)	3
Figure 2: Knowledge and research gaps of this research	5
Figure 3: The 'Triple Bottom Line' (University of Wisconsin, n.d.)	7
Figure 4: Schematic outline of the ethanol organosolv process for lignin extraction	10
Figure 5: Process scheme of soda extraction (based on Osman and Ahmad (2018)	15
Figure 6: Various classes of Ionic Liquids (Yinghuai, 2013)	17
Figure 7: The imidazolium salts are only defined as TSILs when functional group is covalently bond	ed
to the cation/anion of the salt, which behaves as a reaction medium and reagent/catalyst (Yinghua	i,
2013)	18
Figure 8: Examples of CILs (Yinghuai, 2013)	18
Figure 9: Shear strength of lignin resins compared to Phenol formaldehyde (Kalami, Somayyeh et	
al.,2018)	25
Figure 10: Effect of pH on lignin-furfural resins tensile properties. The adhesives were applied at	
180°C and under 1.9Mpa pressure. Blue bars represent the tensile strength whereas the orange on	ie
the elastic modulus. PF= Phenol-Formaldehyde, the commonly used petrol derived resin as a	
standard comparison	27
Figure 11: Asian-based bamboo value chain (Vögtlander, van der Lugt & Brezet, 2010)	29
Figure 12: Eco-costs of plybamboo (Van der Lugt, 2008)	30
Figure 13: Eco-costs of bamboo stems (Van der Lugt, 2008)	30
Figure 14: Eco-costs SWB (Van der Lugt, 2008)	31
Figure 15: European-based bamboo value chain	32
Figure 16: Power-interest diagram or stakeholder matrix	36
Figure 17: Environmental impact sulfuric acid (Marwa et al., 2017)	42
Figure 18: Production costs liquid ions (Passos, Freire & Coutinho, 2014).	46
Figure 19: Total costs per kg of biomass protic ionic liquids compared to acetone and glycerol	
(Baaqel et al., 2020)	47
Figure 20: Global warming potential organosolv and soda	50
Figure 21: Several other indicators environmental impact organosolv and soda	51
Figure 22: SWOT-analysis	61

List of tables

Table 1: Overview of results analysis extraction methods (1 being high score, 3 low score) iii
Table 2: List of interviewees consulted in this research
Table 3: List of academic advisors (WUR) consulted in this research7
Table 4: Comparison of lignin extraction methods (adapted from Tribot et al. (2019)9
Table 5: Le (%) amount of lignin extracted in the organosolv extraction method with ethanol and
different catalyst (percentage of EOL with respect to the lignin content in the original raw material).
Modified from de la Torre et al. (2013)11
Table 6: Characteristics of lignin resins (Kalami, Somayyeh et al., 2018)
Table 7: Potential environmental impact ethanol and sulfuric acid for organosolv method. 38
Table 8: Net Present Value organosolv (Moncada et al., 2018) 39
Table 9: Potential environmental impact, per impact category, of sodium hydroxide and sulfuric acid.
The total was calculated by summing the values of sodium hydroxide and sulfuric acid
Table 10: Life cycle impact assessment results and the contribution of most significant process to the
mid-point scores. Values are presented per functional unit. (Hong et al., 2013)42
Table 11: Comparing environmental impact organosolv and soda50
Table 12: NVP Value comparison organosolv and soda 52
Table 13:: Overview of results analysis extraction methods (1 being high score, 3 low score)

Chapter 1: Introduction

The word 'bamboo' creates, for many Europeans, images of lush Chinese bamboo forests inhabited by many giant pandas. Unbeknownst to many of them, however, is the frequent usage of bamboo products in the everyday European life, like floors, cups and bowls and even essential parts of modern buildings can be made from bamboo wood. Even more strange to most of the European population is the recent development of bamboo plantations opening on European soil. This development has two main causes: the growing concern on importing increasingly scarce hardwood and the environmental burden associated with transporting bamboo from the other side of the world (Van der Lugt *et al.*, 2015).

Related to the former concern, it is well known that the demand for natural resources has been rising rapidly. Not only when it comes to energy production but also, for instance, regarding building materials. Due to promising characteristics such as a fast growth-rate and adaptability to harsh environments, bamboo has been increasingly investigated as a possible substitute to hardwoods (Van der Lugt, Vogtländer & Brezet, 2009). This being said, it is worth mentioning that lignin in monocotyledon, from which bamboo is part of, is structurally different from softwood and hardwood lignin. Knowing the lignin composition is key to determine which refinery pathway is most adapted and key to understanding the properties of lignin-based end-products. But the availability to information on lignin composition finds itself limited to a few species and plant components (Lourenço & Pereira, 2017). Due to this reason, the demand for bamboo, in the West, has been rising steadily in the past decades.

Related to the latter concern, it is seen that the main producers of bamboo are still based in Asian countries, such as China, which hold a big share of the market (Peng *et al.*, 2013). Importing bamboo like this has a big environmental footprint, due to the shipping processes, which diminishes the sustainability prospects of the bamboo value chain for European consumers. Furthermore, this chain is yet to be a circular process, as there is a production of residues. This entails that there is still room to improve the sustainability of the bamboo value chain. This can be done by investing in the production chain's circularity. This means that, by putting already existing waste to use, waste is reduced, and more value is added to the chain, which makes the system more sustainable. This is also emphasised by Mateusz Wielopolski, an expert in the field of sustainability and bamboo with whom an interview was conducted. He stated that 'not many people are dealing with residues' (personal communication with Mateusz Wielopolski, 2020).

ArtEZ FM is an organisation that initiates and realises 'design-driven innovation and research projects that contribute to a diverse, inclusive and sustainable society'. The organisation applies its knowledge on sustainable fashion, product design and interior architecture (ArtEZ FM, n.d.). For this project, ArtEZ FM is collaborating with BambooLogic Europe BV (BambooLogic), and together, they aim to create a sustainable European-based bamboo value chain. BambooLogic is the owner of a start-up European-based bamboo plantation, in Portugal, Alcoates, where Moso Bamboo is grown for a variety of applications. ArtEZ FM is interested in using the bamboo for interior and exterior applications whilst simultaneously making the European-based bamboo production processes to create a binder for laminated bamboo. By using this lignin, the 'waste' becomes useful, which improves the circularity of the

European-based bamboo value chain. Ultimately, this can translate into a more sustainable value chain for European bamboo products.

ArtEZ FM has reached out to Wageningen University & Research to organise a group of master students, who together will work on the problem at hand. This serves two purposes: firstly, the group has a high level of interdisciplinarity, with both natural and social sciences well represented. This is vital for this project, since expertise and knowledge are needed in order to understand the social problems that might arise as well as the implication for the natural ecosystems. Furthermore, it is crucial to have a technical insight on the structure and function of the lignin molecule. Secondly, the students serve as a connection between academia and society and can address societal issues by using information available to and from academia.

This study will examine three different extraction methods to remove lignin from bamboo residues: organosolv extraction, soda extraction and IL extraction. The choice to include the organosolv method is based on it being proposed by ArtEZ FM and the fact that it is a well-established method that produces high-value lignin with possibly economic and environmental advantages (Mcdonough, 1992). Soda extraction is a well-known method for delignification which seems promising in terms of low production costs and limited environmental impact (Carvajal, Gómez, & Cardona, 2016). IL is a novel and promising method which seems very effective for lignin extraction, but there is uncertainty about its economic performance (Muhammad *et al.*, 2011). Before delving into the specifics of this project, a closer look at the characteristics of lignin and lignin in bamboo is required.

Lignin: from a broad and close view

Lignin forms a large stock for phenolic compounds on our planet, and it embodies the second most abundant macromolecule after cellulose (Tribot *et al.*, 2019). On an annual scale, plants produce lignin in a quantity of 150 billion tons, in which 0.082% of the solar energy received by the earth's surface is stored (Hu, Zhang, & Lee, 2018). According to Hu *et al.* (2018), this corresponds to 5.4 times the energy consumption rate currently maintained across the world. Out of 70 million tons of lignin that is annually extracted by humans, only 5% is being used to produce bio-adhesives and other added-value products. The remaining 95% is still being burned as low-quality fuel as energy production (Laurichesse and Averous, 2013).

Furthermore, out of 50 million tons of lignin produced as a by-product, by the pulp and paper industry, merely 2% is used as a recycled material, while the other part is, again, burned for heat and energy production (Upton & Kaslo, 2016; personal communication with Mateusz Wielopolski, 2020). Burning lignin for heat is useful and sustainable as stored carbon for plant growth is just returned to the atmosphere. However, the potential for lignin to be used in high quality products is huge. This entails that more research is still necessary to understand how such an abundant compound can be more efficiently used.

On the other hand, lignin is increasingly being researched as a promising part in the production of value-added and high-performance materials (Khan, Colmenares & Gläser, 2020). In 2014, the worldwide lignin market was estimated to be worth €660 million, with a steady increase of 2.5% per year, between 2014 and 2020. At the time, it was anticipated an increase up to €770 million by 2020 (Khan, Colmenares & Gläser, 2020).

Lignin can, for instance, be applied in the field of fuels, plastics, chemicals and composites (Ragauskas *et al.*, 2014; Ghaffar & Fan, 2013). However, finding a sufficient extraction method for lignin is highly challenging, because of its complex structure and the fact that it forms lignin-carbohydrate complexes, including bindings with cellulose and hemicelluloses (Tribot *et al.*, 2019). Lignification evolves through polymerization during which lignin macromolecules are synthesized via free radical coupling mechanisms (Lu & Ralph, 2010). These monomers are primarily coupling 'endwise' (Ralph, Lapierre, & Boerjan, 2019), which means that monomers are added one-by-one to the polymer (Wang, Chantreau, Sibout, & Hawkins, 2013). The main three monomers that form the 'building bricks' of lignin, can be seen in Figure 1. Thorough understanding of these structures is not necessary, but it provides some background information, and it might be valuable when this report is presented to an expert.



Figure 1 Main monomers of lignin (Harmsen, 2016)

It is worth mentioning that different types of plants have different lignin concentrations and structure. The lignin polymer is built with three monomers (p-coumaryl alcohol (G), coniferyl alcohol (H) and sinapyl alcohol (S)). Gymnosperms have a HG-type of lignin and angiosperms a GS-type of lignin. Bamboo, being a monocotyledon, has a HGS type of lignin and varies from 25-30% lignin content, which is more comparable to ranges reported in softwoods (24-37%) and hardwoods (17%-30%) instead of other non-woody species (11-27%) (Wen et al., 2010). However, research lacks the content and characteristics of lignin in bamboo and the extraction methods, specifically, from bamboo residues. Further research on lignin will contribute to the understanding of lignin nature and mechanisms within the plant as well as support the development and perfection of lignin extraction methods providing valorisation routes for chemicals, materials and energy.

Knowledge and research gaps

This report addresses many forms of knowledge gaps. These are vital pieces of information that need to be retrieved to fulfil the commissioner's wishes, which are, at the moment, under-researched. Four main knowledge gaps are identified below, and a research gap is presented.

Technical feasibility of extracting lignin from bamboo residues

The first knowledge gap that needs to be addressed is on how to extract lignin from bamboo residues effectively. While much literature exists on lignin extraction methods of conventional materials like hardwood, this is not the case for bamboo. The challenge here is to either find literature on bamboobased lignin extraction methods, or to use information based on hardwood lignin extraction, to explain the technical feasibility of extracting lignin from bamboo residues. This knowledge gap is one of the cornerstones in this research project, since in order to build a biobased binder out of lignin, it is first necessary to extract it successfully. This relation is shown in Figure 2.

Sustainability of extracting lignin from bamboo residues

The second knowledge gap is related to the sustainability aspect of this research. The commissioners have expressed their interest regarding the sustainability of these extraction processes. This means that any recommended method cannot have a higher significant environmental impact or do social harm than conventional extraction methods. This knowledge gap exists due to lack of information on bamboo-based lignin extraction methods and consequently on their environmental footprint. This knowledge gap is vital to fill, and thus, one of the cornerstones of this research: if the sustainability of the chosen extraction method cannot be determined, it is unlikely that the commissioner will use the information on how to extract lignin (Figure 2).

Economic feasibility of extracting lignin from bamboo residues

The third knowledge gap that will be focused on is the economic feasibility of extracting lignin from bamboo residues. Scientists often focus on the 'how-to' and its environmental impacts, but they fail to address the economic feasibility of their methods. This lack of connection between academia and society is a widely known issue, which this interdisciplinary group tries to overcome. This knowledge gap will therefore not so easily be filled through information from academia, but rather through societal consultation. Certain businesses or institutions might have the economic knowledge gap is the last cornerstone of this research since it can be established that businesses will not use a method where costs outweigh profits. This ultimately means that it is crucial to find information on the economic feasibility of the extraction methods

A lignin-based binder for laminating bamboo products

The fourth knowledge gap is one that builds upon the three previously described ones. Once the feasibility of the different extraction methods is established, information needs to be found on how to make a binder from the extracted lignin. While information is already available on this subject in academic literature, one question remains regarding the similarity of lignin from bamboo residues compared to that from other hardwoods. Furthermore, there is a lack of information on how this lignin-based binder differs from other petrochemical binders that are already being produced. This knowledge gap is best addressed by consulting industry stakeholders that are already working on finding bio-replacements. This knowledge gap is relevant, since this lack of information might be the factor keeping companies from wanting to include a lignin-based binder as part of the circular European-based bamboo value chain (Figure 2).

Involving stakeholders

While a knowledge gap concerns the lack of information, a research gap is about how to put this new information into practice. The research gap that is addressed in this report is about how to move stakeholders from the European-based bamboo value chain to uptake and embrace the use of a ligninbased binder. One research gap that is important to address here as well, is the consequences that this development might have on certain stakeholders in or related to the European-based bamboo value chain and Asian-based bamboo value chain. In doing so, it is important to sketch the Europeanbased bamboo value chain, which is very new, and describing its stakeholders.

In Figure 2, both the knowledge and research gaps are illustrated. It can be established that the knowledge gaps on the technical, economic and environmental side of the extraction methods serve as three 'cornerstones' in the research, since without one of these the research questions about creating a lignin-based binder cannot be answered. Furthermore, if the research gap on stakeholder involvement or action is not filled, there will be a limited uptake. If all the knowledge and research gaps are addressed, this might lead to a relatively more circular European-based bamboo value chain.



Figure 2: Knowledge and research gaps of this research

Research questions

This project contributes towards the goal of ArtEZ FM to develop a sustainable and circular European based bamboo value chain, specifically to improve the sustainability of laminated bamboo.

The aim of this project is to evaluate different lignin extraction methods in terms of technical feasibility, environmental & social sustainability, and economic performance and to study the application of lignin as a biobased binder. Therefore, the main research question is '*Can lignin from European-based Moso Bamboo residues be used as a binder for lamination to improve the sustainability of the European-based bamboo value chain?*'

In order to answer the main research questions, the following three sub-research questions (SQ) are formulated.

SQ 1: How can lignin be extracted from Moso bamboo residues?

- a. Is organosolv extraction a technically feasible extraction method for bamboo residues?
- b. Is soda extraction a technically feasible extraction method for bamboo residues?
- c. Is extraction through IL a technically feasible extraction method for bamboo residues?

SQ 2: How can lignin from Moso bamboo residues be used as a binder for laminating?

- a. How do petrochemical binders work?
- b. Does lignin share the same chemical and mechanical properties with some of the petrochemical binders?

c. Does the lignin need modifications in order to function as a binder?

SQ 3: To which extent does this process and product of bamboo binder really result into sustainable bamboo value chain for bamboo products?

- a. What is the environmental impact of organosolv, soda and IL extraction?
- b. What is the economic performance of organosolv, soda and IL extraction?
- c. What impact has using lignin as a binder on the stakeholders?

Methodology

To provide answers for the SQs posed above, different research methods are used. Information is mostly acquired through literature research and document analysis, through the library of Wageningen University & Research or open peer-reviewed articles available on Google Scholar. Unstructured interviews serve a two-fold purpose in this research: firstly, to fill the knowledge gaps unanswered by academic literature with a more practice-based view. Secondly, to discuss and verify the findings of this report and how this relates to their own experience in their business or institute. This combination of literature review and interviews are suitable to answer the posed research questions since it contributes towards a triangulation of results. This means that results found with one research method, like literature review, can be confirmed through interviews with an expert, which increases the validity of this report. The following table presents a list of people that were contacted for an interview to either fill knowledge gaps or to verify found information.

Interviewee	Organisation	Date
Aart van Vuure	KU Leuven	21-11-2019
Joost Borneman	Bamboologic	21-09-2020
Marina van der Zee	Stichting Hout Research	02-10-2020
Mateusz Wielopolski	Aevolution Tech	07-10-2020
Karsten Brast	Nature2Need	12-10-2020

Table 2: List of interviewees consulted in this research

Furthermore, the authors of this report have the opportunity to discuss the findings with two academic advisors. This serves multiple purposes: firstly, to preserve the academic integrity of the report. Even though the report is meant to be applicable for societal purposes, it is vital that this does not suppress the academic quality of the result. Secondly, the academic advisors are experts in their own field related to this report, which provides the authors with new insights on how to fill the knowledge gaps. The following table presents the two academic advisors that were consulted for this report

Table 3: List of academic advisors (WUR) consulted in this research

Academic Advisor	Policy group
Paulien Harmsen	Biorefinery & Sustainable Value Chains Group
Ellen Slegers	Operations Research and logistics Group

While this report strives for complete accuracy and completeness, it should be acknowledged that the results presented in this report were limited due to several constraints. Firstly, due to the COVID-19 pandemic, the authors of this report are not able to meet in person to discuss findings related to the report. The authors are dependent on online programs to communicate between one another, which sometimes limits creative brainstorming or discussion. However, the authors utilize these online programs in the best way possible, to imitate an offline discussion session. Secondly, due to time and money constraints, the authors are not able to experiment with bamboo and the discussed extraction method in this report. The report is therefore not able to go beyond the theoretical, and further practical analysis should be done by stakeholders that can do so.

Defining sustainability

Sustainability plays a central role in this report. The commissioners shared, from the beginning, their wish to create a more circular and sustainable value chain for European-based Moso bamboo, and ultimately, value-added products from that chain.

Before ensuing this report's contents, however, it is vital to first discuss what this report defines as 'sustainable'. Sustainability is a word that caries many meanings, which means it is important to define it for the project.



Figure 3: The 'Triple Bottom Line' (University of Wisconsin, n.d.)

The term first got its real policy meaning in 1987 in the Brundtland report, which was concerned with the drive for mankind to have a 'better' life and how to balance this with the limitations of the natural world (Kuhlman & Farrington, 2010). While this environmental perspective of sustainability persisted most prominently until today, an economic and social perspective of sustainability has also risen. This

is especially apparent when talking about sustainability in a capitalistic paradigm, where without profit there cannot be sustainability. This interdependency has been coined as the 'Triple Bottom line' or the 'People, Planet, Profit' of sustainability (Norman & MacDonald, 2004). Therefore, without environmental sustainability, there cannot be social sustainability or profit, and vice versa, as depicted in Figure 3.

This Triple Bottom Line has also been found to be applicable to review the sustainable performance of value chains (Balkau & Sonneman, 2010; Mol, 2015). To analyse the sustainable added value of a lignin-based binder, therefore, does not just mean to look at its environmental impact. This also involves looking at the social impact of localizing the European-based bamboo value chain, not just for people directly involved, but also for people who are indirectly affected. This will be discussed at large in the report. Lastly, the profits in the European-based bamboo value chain should at least be maintained, to prevent a lack of usage from stakeholders in the chain. This interdependency is depicted in Figure 3.

Based on this interdependency of the environmental, economic, and social aspects of sustainability, this report will analyse to what extent the lignin-based binder will contribute to the sustainability of the European-based bamboo value chain.

Chapter 2: Technical feasibility extraction methods

Introduction

Over the years, a certain amount of methods emerged to extract lignin from biomass while dealing with its complex amorphous structure (Rahim *et al.*, 2018). The most common method is called the Kraft process that entails sulphate pulping, but this process requires high temperatures and high pressures (Rahim *et al.*, 2018). In addition to this, delignification is incomplete (Al-Kaabi *et al.*, 2018) and the recovery of kraft lignin chemicals is not sufficiently developed (Tribot *et al.*, 2019). Other conventional methods are the soda process, lignosulfonates and organosolv. The comparison of these processes is illustrated in Table 4. It can be seen in this table that the organosolv method has relatively low and constant ash content, meaning low levels of impurity. Soda's ash content has the potential to be lower than Kraft's and is definitely lower than lignosulfonates. Lastly, sulphur is not used in the extraction stage for soda and organosolv, keeping the lignin molecule closer (almost) intact.

Lignins	Ash content (%)	Sulfur content (%)
Kraft	0.5–3.0	1.0-3.0
Soda	0.7-2.3	0
Lignosulfonates	4.0-8.0	3.5-8.0
Organosolv	1.7	0

Table 4: Comparison of lignin extraction methods (adapted from Tribot et al.(2019)

After extraction from the raw material, an effluent called 'black liquor' is obtained. The chemical composition of black liquor depends on the chosen raw material (Cardoso, de Oliveira, & Passos, 2009). However, for all types of raw material it can be concluded that the derived black liquor forms a complex aqueous solution, containing organic materials, such as lignin, polysaccharides (cellulose, hemicellulose) and inorganic compounds, which are mainly soluble ionic salts (Cardoso *et al.*, 2009). Lignin must be isolated from this black liquor to reach a certain lignin purity extractant. Such a method will differ between lignin recovery methods. Furthermore, these processes do not result in high purity lignin as cellulose, hemicelluloses and other products are obtained as impurities. This means that to transform lignin into added-value products, further purification is needed (Toledano *et al.*, 2010a; Toledano *et al.*, 2010b). Therefore, other methods are considered to ensure a technically feasible lignin extraction while maintaining environment and economic integrity.

Upcoming extraction methods are also being explored, among which IL showed to be a very promising method. These substances can be task specific and extract lignin directly from lignocellulosic biomass and no black liquor is produced. ILs have a wide range of applications in diverse application domains due to their outstanding combination of properties. One of these applications is its specificity for lignin extraction from lignocellulosic biomass, with advantages for recovery and recyclability (Yungiao & Nan Jand, 2007). Besides, it is the most widely investigated extraction methods nowadays, particularly in the biomass area (Gordon CM, 2001).

Having considered all the above-mentioned processes, the continuation of this chapter will hold a more detailed regard on the most promising current extraction processes: organosolv and soda. The novel method IL will also be discussed, which provides great potential for being the next generation of lignin extractants.

The organosolv process

The 'organosolv process' is characterised by the usage of organic solvents. The development of this technique allowed the full use of the raw material, contrary to former procedures. Their utilisation could be used for obtaining hydrolysable cellulose, phenolic polymers of lignin and sugars (Muurien, 2000; Pala, 1999; Varshney & Patel 1988). There are many solvents that may be used for organosolv pulping: glycols, phenols, ester, organic acids, acetone, ammonia, and amines. Nevertheless, alcohols are the most popular solvent with ethanol being one of the most effective (Rodriguez *et al.*, 2018).

In the organosolv process the lignin is dissolved in organic solvents. Originally, the organic solvents were used to break down wood components to study its composition as well as lignin more specifically. In 1992, two full-scale operations were already using organosolv, namely organocell and the ASAM process (alkaline sulphite-anthraquinone-methanol process), both using methanol in the extraction process. Two more processes were being tested at a pilot-scale which used acetic acid and peroxyformic acid (Muurien, 2000). Despite the promising outlook for the organosolv process, it had only limited commercial success. This is largely due to the economic competitiveness towards more established processes like the kraft process. The paper and pulp industry had no interest in changing their systems. More recently, the organosolv process has become more attractive due to the potential use for bio-refineries. The process has the advantage that both lignin and the solvent can be recovered in addition to a relatively pure cellulose fraction, making it more sustainable and circular. In order to make this process more competitive, optimisations have to be done, and its use should be limited to reasonable and abundant feedstocks that contain preferably low-density lignocellulosic compounds (Schmetz, Q. *et al.* 2016). A schematic overview of this process can be seen in Figure 4.



Figure 4: Schematic outline of the ethanol organosolv process for lignin extraction.

The organosolv process is done by the hydrolysis of the linkages between the lignin and the cellulose, hemicellulose and pectines. This means they are broken down by the addition of water. Because there are bonds within the lignin itself that can also be hydrolysed, it is impossible to extract lignin in its original, pure form. The lignin that is recovered by means of this process is purer than former extraction methods. Because of this, the lignin that is recovered by an organosolv process can have multiple uses. While it will yield lignin with a wide range of lengths and molecular weights of the lignin,

there is limited sulfonating happening in this extraction process. This makes it possible to refine the lignin into three main products; aromatic compounds, additives for polymers or to turn the lignin into a binder itself. (Schmetz, Q. *et al.*, 2016 & Tian, D. *et al.*, 2017)

Extraction method specification

Even though the organosolv process is described above as a single process, it should be considered as a set of processes. Indeed, 'organosolv' is an umbrella term for a multitude of different extraction methodologies using different solvents, concentrations, catalysts, temperatures, reaction times and pressures applied to specific raw materials (Zhao *et al.*, 2009; Muurinen *et al.*, 2000). Insights on the various available options will be provided. Furthermore, examples focused only on bamboo lignin extraction will also be presented.

Acetic acid, acetone, methanol, and ethanol are the main solvents used for the organosolv lignin extraction (Borand *et al.*, 2018). Moreover, these are also the substances for which a large-scale process is operating or has at least been tested and proven to be technically feasible (Harmsen *et al.*,

Table 5: Le (%) amount of lignin extracted in the organosolv extraction method with ethanol and different catalyst	
(percentage of EOL with respect to the lignin content in the original raw material). Modified from de la Torre et al. (2013))

Catalyst	L _e (%)
Hydrochloride acid	61.17
Sulfuric acid	60.64
Nitric acid	58.71
Phosphoric acid	58.78
Formic acid	49.55
Acetic acid	50.84
Oxalic acid	48.03
Calcium chloride	51.20
Aluminum chloride	51.60
Ferric chloride	49.42

2007, unpublished). Out of all these methods, and many others, the one using ethanol seems to be the most promising. Even if it may not be the most efficient solvent with the highest amounts or purity of lignin extracted, its negligible toxicity and easy recovery leaves no room for doubts (Ethanol 2020, PubChem; Zhao et al., 2009). Methanol and acetone are easy to recover and can be used at low temperatures, consequently saving on energy costs. However, they are more poisonous to humans and harmful for the environment. Therefore, more sustainable and safer options are preferred. Acetic acid on the contrary is safe for both human beings and natural ecosystems. However, its recovery can be technically challenging and energy demanding, leaving a lower margin for profit. Ethanol is claimed to be safe for the environment and to have limited toxicity for humans (PubChem, 2020). In addition, thanks to its high vapour pressure and, therefore, low boiling point, it is technically feasible and not too costly in terms of energy need. Next to the solvent's choice, its concentration should also be evaluated. For ethanol organosolv lignin (EOL) extraction usually ethanol-water mixtures are used (Zhao et al., 2009). Looking at values, literature reports a concentration of ethanol between 35% and 70% (v/v) (volume/volume) depending on the extraction efficiency and lignin purity needed for further processing (Borand et al., 2018; Zhao et al., 2009). Finally, the solvent's quantity, i.e. the solid-liquid ratio, can also play a key role in the extraction process. Literature reports ratios varying from 1/4 (w/v) (weight in kg / volume in L) to 1/10 (w/v), again according to the producer's needs and investments opportunity /disposable income (Borand et al., 2018; Bahera et al., 2014; Kumar et al., 2009). Despite the choice of the solvent, its concentration and quantity, the choice of the catalyst is also crucial. There are many possible reaction catalysts enhancing lignin extraction efficiency and purity (see table 5). The most used catalysts are HCl, hydrochloride acid, or H₂SO₄, sulfuric acid, being the most efficient, as depicted in the table above (de la Torre *et al.*, 2013; Sun and Cheng, 2002), However, it has to be kept in mind that the more substances (how many and how much of each) are added to the reaction chamber, the more components should be removed afterwards. Hence, the benefit (i.e. much higher extraction efficiency) of adding these substances should over-compensate the costs for its addition and for its recovery, otherwise it will not be worth the investment. Finally, before choosing, a careful evaluation on the needed quality of lignin should be performed. This assessment will be done according to lignin's future use, as it will be extensively discussed in the chapter about the binder characteristics.

Reaction conditions are important for a successful EOL extraction as well. Literature reports reaction temperatures varying from 120°C to 200°C, but only over 150-160°C high efficiency extraction is obtained (Fan *et al.*, 2015; Bahera *et al.*, 2014; Bai *et al.*, 2013; Kumar *et al.*, 2009). Extraction pressures will increase accordingly to temperatures until a maximum after which residual or excessive pressure may be released through service or emergency valves. Furthermore, higher temperatures and pressures enhance the extraction strength, diminishing the time needed for the process (Borand *et al.*, 2018; Fan *et al.*, 2015). In other words, harsher treatment conditions lead to less required time needed for the same level of extraction efficiency. Even though this is true in most cases, this correlation does not always stand. For example, too low temperatures will not enable lignin extraction (usually lower that 120°C), whereas too high temperatures will lead to thermal degradation of lignin and therefore to lower quality values (Zhao *et al.*, 2009). Here again, the characteristics of the starting material, the target lignin quality and the investments opportunity will determine the extraction conditions.

Although the organosolv method can be applied on large scale and is technically feasible, it still has important limitations. First, ethanol, and even more ethanol/catalyst mixture, can become very dangerous when brought to high temperatures and kept under pressure. At these conditions the compound, or compounds mixture, becomes very aggressive and corrosive, therefore highly expensive instrumentation should be implemented (Personal communication with Paulien Harmsen, 2020; Muurinen *et al.*, 2000). Secondly, the recovery of the solvent is key. Recovery rates lower than 95% will cause high ethanol wastes, determining the costs to be higher than any possible income (Muurinen *et al.*, 2000). The following examples show if organosolv is suitable to be applied for EOL extraction from bamboo.

Bamboo specific extraction

There is uncertainty about the existence of any large scale organosolv processes using bamboo as main starting material and with lignin as aimed end product. Nevertheless, literature reports many experimental studies in small scale using organosolv as method to extract lignin from bamboo. Wu *et al.* (2020) report EOL obtained by processing bamboo with 75% ethanol solution and 1.0% (w/w) sulfuric acid in a solid to liquid ratio of 1:7 (w/v) in a high-pressure autoclave at 170 °C for 60 min. After the extraction, the reactor was cooled down in a water bath. The slurry (or black liquor) was separated into a solid fraction (rich in cellulose) and a liquid fraction by filtration. Ethanol was then recovered, from the remaining liquid phase, by applying low pressure and heat. The resulting liquor

was then washed with an excess of cold water in order to separate lignin from the rest of the matrix. The precipitated EOL is then washed by warm water, whereas the remaining supernatant rich in hemicellulose was discarded or reused to produce biogas and recover energy. Fan *et al.*, 2015 examined different temperatures, time frames and solvents. Out of the tested conditions, the experiment using 180°C for 2h, with 50% ethanol and a solid to liquid ration of 1:8 (w/v) was the most reliable one. Moreover, Bai *et al.* (2013) report working conditions similar to the Fan's article: 180°C for 2h but with 70% ethanol and a solid to liquid ration of 1:10 (w/v). Many other publications, providing slightly different protocols, can be found for EOL extraction from bamboo. Some of them provide slightly higher lignin quality, whereas others were identified to optimise the costs maintaining a decent lignin purity. Therefore, there is no perfect protocol. They are all good starting points from which the most suitable conditions for each purpose can be defined

Concludingly, organosolv was proven to be a potential method to extract EOL from bamboo. However, the different variables playing an important role in this extraction process have to be properly optimised in order to avoid huge failures, as happened in the past (Harmsen *et al.*, 2010).

The soda process

The soda pulping process was already introduced in 1851 before the Kraft process to process nonwoody materials or produce a high yield of hardwood pulps required for paper packaging and boards (Al-Kaabi *et al.*, 2018). The advantage of this process is, in contrast to the Kraft and sulphite process, the absence of sulphur utilisation and residue in the lignin structure (Al-Kaabi *et al.*, 2018). The basic process entails the heating of biomass in a reactor under pressure, maintaining a temperature between 140-170°C (Al-Kaabi *et al.*, 2018). With this, sodium hydroxide (NaOH) is added to a concentration of 13-16 wt% (weight percent) (Al-Kaabi *et al.*, 2018). The effluent of this process is the aforementioned black liquor, of which the lignin should be isolated from. This is done by performing acidification during which lignin is precipitated, enabling separation of lignin from the liquid (Lora, 2002). A challenge of this procedure is the separation of silica from lignin, a substance that can lower the quality of the end product as it can co-precipitate with lignin (Lora, 2002). One example of an acidification process is the Lignin Process System (LPS) in which the lignin slurry is produced after the pH of the black liquor is decreased with a mineral acid (Belgacem & Gandini, 2011). A lignin cake is formed after this slurry is filtered, which is washed and dried in order to obtain lignin powder of high purity and less than 5% moisture (Belgacem & Gandini, 2011).

A recovery of 84% of chemicals of the soda process can be reached, according to Garcia *et al.* (2011). However, the paper of Garcia *et al.* (2011) describes a recovery system in which lignin is burned and used as an energy source, which is standard in paper industries. The burning of the liquid fraction (including lignin) results in solid residue containing sodium carbonate (Na₂CO₃), which is dissolved with water and extra sodium carbonate (Garcia *et al.*, 2011). Treating it with calcium hydroxide will convert the Na₂CO₃ back into NaOH and calcium carbonate (CaCO₃) (Garcia *et al.*, 2011). However, a method which does preserve lignin, can be found in the report of Chen *et al.* (2020), which describes a method that separates sodium hydroxide from alkaline lignin based on molecular weight. This was done by applying an ultra-highly crosslinked resin (Mn270) on which sodium hydroxide absorbs, allowing separation (Chen *et al.*, 2020). Besides recovery of sodium hydroxide, this causes the acid consumption and associated produced waste salts (NaSO₄) to decrease with 37.86% (Chen *et al.*, 2020).

Extraction method specification

The soda process is often accompanied with the term 'anthraquinone' in literature. This is a catalyst that increases the rate of delignification (Hart & Rudie, 2014). However, according to the review article of Hart and Rudie (2014), economic justification of adding this catalyst is difficult as its yield improvement is modest and therefore often gets lost in the profits of a producer. In combination with its potential to be carcinogenic (Hart & Rudie, 2014), anthraquinone is not included in this recommendation of the soda extraction method.

As partly illustrated by the ranges mentioned above, the soda concentrations, reaction times and temperatures differ across methodologies. This depends on aspects such as the substrate and the acid used for the soda process (Carvajal, Gómez, & Cardona, 2016; Hubbe, Alén, Paleologou, Kannangara, & Kihlman, 2019). Acidification can be performed with acids such as carbondioxide (CO₂), sulfuric acid, phosphoric acid, hydrochloric acids, and organic acids, according to Gilarranz *et al* (1998). The use of CO₂ can be beneficial for the recovery cycle when recovered from the system's own emissions.

However, CO₂ causes the pH to drop to only 8-9, resulting in a rather low precipitation yield of 60-80% (Gilarranz *et al.*, 1998). Application of sulfuric acid is not expensive and delivers a higher lignin yield (Hubbe *et al.*, 2019). For example, a study noted a 90% lignin yield with sulfuric acid in comparison to a 77% yield with CO₂, according to the review article of Hubbe *et al.* (2019). Besides this, from the retrieved research papers that studied lignin extraction from soda processes with non-woody species, most of them used sulphuric acid for precipitation (Cardoso, de Oliveira, & Passos, 2009; Fernández-Rodríguez *et al.*, 2018; García *et al.*, 2009; Mussatto, Fernandes, & Roberto, 2007; Zhang, Chen, & Peng, 2017). Some researchers propose a two-staged procedure of respectively adding CO₂ and sulfuric acid (Gilarranz *et al.*, 1998; Hubbe *et al.*, 2019). It can be concluded from this paragraph that these choices can depend on many factors and more extensive study can potentially be done in a later stage if needed.

Bamboo specific extraction

An example of lignin extraction of bamboo using the soda process can be found in the study of Osman and Ahmad (2018), in which lignin was extracted from two Malaysian bamboo species that aged for three years. First, the bamboo was treated with hot water maintaining a temperature of 100°C for one hour in an autoclave, a metal container used for reaction occurring at high temperatures and pressures (Osman & Ahmad, 2018). After this, the lignin extraction was performed in a closed vessel with 13% w/v NaOH solution for one hour using a solid to liquid ratio of 1:4 (Osman & Ahmad, 2018). Next, the lignin was extracted with a sulfuric acid until the pH of 3 was reached (Osman & Ahmad, 2018). The soda process executed by Mousavioun and Doherty (2010) on sugarcane also entailed sulfuric acid to lower the pH to 3. The scheme represented in Figure 5, is based on the methodology of Osman and Ahmad (2018). According to Osman and Ahmad (2018), the obtained bamboo lignin



Figure 5: Process scheme of soda extraction (based on Osman and Ahmad (2018)

from the soda process, formed a good alternative to phenol when a low glass temperature (Tg), a critical value between the solid and liquid state at which material shows rubberlike elasticity (Irvine, 1985), is combined with high stability at high temperature during the synthesis of adhesives.

Concludingly, research on the soda process to extract lignin combined with bamboo remains limited. Some additional papers were found, but these papers combined the soda process with ethanol treatment (Dagnino *et al.*, 2018; Li *et al.*, 2010; Sun *et al.*, 2012). In other words, the soda process is combined with the organosolv extraction method. The adoption of these systems' combinations should be taken with caution, as the more complexity is attributed to lignin extraction, the more potential is created for the process to be expensive and unsustainable (Personal communication with Paulien Harmsen, 10-02-2020).

Ionic liquids

ILs are salts that are in a liquid state. Sodium chloride, for example, commonly known as *salt*, or *table salt* (NaCl), can become an IL when it melts at 801°C with sodium cations (Na+) and chloride anions (Cl⁻). However, it is not easy to work with such high temperatures (Yinghuai Z. *et al.*, 2013). In literature, it is spoken of ILs for salts that can be in a liquid form at room temperature (<100°C).

At a molecular level, ILs are characterised by powerful electrostatic interactions. Because of this, Rogers & Seddon (2005) state that bulk ILs can be considered as pure electrolytes¹. The electrostatic interactions have an impact on many properties at a macroscopic level, viscosity, negligible vapour pressure, poor conductivity. Such aspects are conditioned by the complexity of the molecular components and to their asymmetry, which defy the natural tendency of ionic compounds to form crystals (Bodo & Migliarati, 2011). These properties make ILs unique both as neat liquids and as solvents (Mocci *et al.*, 2014), giving ILs proper technologically exciting properties and characteristics that distinguish them from other molecular solvents. Indeed, ILs can be easily, quickly, and efficiently recycled due to their negligible vapour pressure and good thermal stability (Yinghuai Z. *et al.*, 2013). This being said, Ludwig and Kragl (2007) stated that vapour pressures, enthalpies² of vaporisation and boiling points are physical chemical properties that depend on the cation–anion combination of every IL. As a result, the extraction methods and recovery procedures depend on the IL being used and adjustments must be made for perfecting the extraction method.

ILs families and properties

It is not easy to separate and differentiate every IL there is. Due to the existing freedom in designing the cation in combination with an anion, a colossal number of ILs can be generated. Around six hundred conventional solvents are being used for industrial and synthetic processes. ILs, on the other hand, exist in more than a million pure forms, making it possible to make up to, and possibly more, than a trillion mixtures (Bodo & Migliarati, 2011). Many classifications can be found; as seen in Figure 6.



Figure 6: Various classes of Ionic Liquids (Yinghuai, 2013).

¹ Disclaimer: this would imply that ILs are entirely composed of ions. However, it may occur that some ionised molecules become de-ionised and become ionised again. This phenomenon occurs randomly and makes it technically impossible for the solution to be 100% ionised all the time, but rather very close to it.

² Enthalpy: a thermodynamic quantity equivalent to the total heat content of a system. It is equal to the internal energy of the system plus the product of pressure and volume.

1. Task-Specific ILs/ (Multi)-Functional ILs

The incorporation of additional functional groups characterises this family into functional ILs. This is made, in order to introduce or enhance specific capacities, such as catalyst reusability. The main aim being the obtaining of task specific ILs through the addition of specific functionalities, which can be achieved with the addition of a functional group to the branch. One of the most common ones being imidazolium cation. Some examples are shown in Figure 7. The imidazolium salts are only defined as TSILs when a functional group is covalently bonded to the cation/anion of the salt, which behaves as a reaction medium and reagent/catalyst (Winkel, Reddy & Wilhelm, 2008).



Figure 7: The imidazolium salts are only defined as TSILs when functional group is covalently bonded to the cation/anion of the salt, which behaves as a reaction medium and reagent/catalyst (Yinghuai, 2013).

2. Chiral ILs

Chiral ILs are characterised by having a chiral³ centre on either its cation, anion, or both. (Baudequinm *et al.*, 2003). They have been gaining in popularity thanks to the ease of synthesis (Yanghuai *et al.*, 2013). Some examples can be observed in Figure 8.



Figure 8: Examples of CILs (Yinghuai, 2013).

3. Protic Ionic Liquids

Protic Ionic Liquids (PILs), also known as liquid salts, are a unique group of ILs. They are produced via a simple acid-base neutralisation (Achividu *et al.*, 2014). Indeed, the major difference between PILs and other ILs is the presence of exchangeable protons (Yanghuai *et al.*, 2013).

³ A chiral centre is defined as an <u>atom</u> in a <u>molecule</u> that is bonded to four different chemical species, allowing for optical isomerism. It is a stereocenter that holds a set of atoms (ligands) in space such that the structure may not be superimposed on its mirror image.

The use of PILs for lignin extraction is a relatively new area of research. However, it has proved to have an excellent extraction efficiency (Achividu *et al.*, 2014; Gschwend *et al.*, 2016); besides, they are economically accessible and have an excellent potential for recyclability (Achividu *et al.*, 2014).

4. Switchable Polarity Solvent

Switchable polarity solvents tend to have a higher polarity. Nevertheless, this can be changed into a low polarity when a trigger is applied. This is useful when two different polarities are needed for two different steps (Yanghuai *et al.*, 2013). Recently, secondary amines have been used as SPS, with carbon dioxide as the trigger, forming carbamate⁴ salts (Lin & Vasam, 2005).

5. Metal salts

Metal ILs figure within the first room temperature ionic liquids (RTILs). Metal-based salts can be subdivided into three groups: transition metal, p-block, and f-block metal salts. These can be synthesized by the reaction of phosphonium/imidazolium halides with metal halides or metathesis reaction with alkali salts of metal-based anions. The recently developed chlorometalate salts are not water sensitive, unlike the chloroaluminates, but are generally more viscous. The introduction of the metal ions inside the ILs can immobilize catalysts while it is being an integral part of the ILs' structure (Yanghuai *et al.*, 2013).

Deep eutectic solvents

Deep eutectic solvents (DES) are a subgroup of room temperature ionic liquids (RTILs) (Marcus, 2019). DES refers to a two-component or three-component mixture systems, in comparison to RTILs which are single substances (Lyu *et al.*, 2019). DES are very ionic in nature, they should therefore have very appreciable electrical conductivity (Marcus, 2019). Otherwise, their physicochemical properties resemble much to those of ILs, so it can be found being classified as a new kind of IL or IL analogues (Paiva *et al.* 2014). Indeed, it can be stated that DES are a parallel classification that goes across all the other classes of IL. In other words, DES can be found in different IL families. Just as PILs, DES are more economical, environmentally friendly, and easy to be prepared in comparison with other ILs (Pena and Namiesnik 2014). As DES are considered a green solvent, they are now wildly used for lignin isolation and other utilisations (biomass pre-treatment or fractionation) (Loow *et al.*, 2017; Tang *et al.*, 2017; van Osch *et al.*, 2017).

Extraction method specification

As mentioned above, many kinds of ILs can be used as solvents, catalysis, reagents, and many other applications. The liberation and properties of lignin highly depend on the biomass source and on the lignin extraction method (pre-treatment) (Wang *et al.*, 2017; Kumar *et al.*, 2009). For what concerns lignin extraction, the ideal IL should have the following properties: 1) high dissolution capacity for lignin, 2) low melting point, 3) good thermal stability, 4) non-volatile, 5) non-toxic, 6) chemically stable,

⁴ Carbamate: a category of organic compounds that is formally derived from carbamic acid. The term includes organic compounds, formally obtained by replacing one or more of the hydrogen atoms by other organic functional groups; as well as salts with the carbamate anion H $_2$ NCOO⁻

7) no lignin decomposition, 8) easy lignin regeneration and 9) low cost and simple process (Olivier-Bourbigou et al., 2010).

Conventional ILs can certainly extract lignin from biomass. However, these processes require high temperatures (>100°C), it extracts only a fraction of the present lignin (<50%) and creates soluble residuals such as sugars, extractants, soluble lignin derivatives, among others. The latter proves one of the most problematic economic aspects: since ILs are expensive to produce, any losses during the extraction process and thermal degradation of the ions due to the high handling temperature for extended periods of time represent a need to renew the ILs stock (Achividu, 2018). After thorough research in literature on lignin extraction with ILs, there is mainly one family of ILs (Figure 6) that fulfils most of the aforementioned aspects: PILs, which check at least points 1, 2, 3, 7, and 9 previously listed, and are promising on checking the remaining points. After thorough research in literature on lignin extraction you family of ILs that fulfils most of the aforementioned aspects: PILs, which check at least points 1, 2, 3, 7, and 9 previously listed, and are promising on checking the remaining points. After thorough research in literature on lignin extraction with ILs, there is mainly one family of ILs that fulfils most of the aforementioned aspects: PILs, which check at least points 1, 2, 3, 7, and 9 previously listed, and are promising on checking the remaining points. After thorough research in literature on lignin extraction with ILs, there is mainly one family of ILs that fulfils most of the aforementioned aspects: PILs, which check at least points 1, 2, 3, 7, and 9 as previously listed, and are promising on checking the remaining points. Because of this, a more in-depth look into this specific IL family will be held.

PILS utilisation for lignin extraction processes is relatively new, but they have proven to have a relatively high lignin extraction efficiency (Achividu *et al.*, 2014; Gschwend *et al.*, 2016; Rocha *et al.*, 2017; Rashid *et al.*, 2016). PILs are a unique kind of ILs that can be produced via a simple acid-base neutralisation, like acetic acid and amine reagents. As PILs melt at relatively low temperatures (<100°C), they can be used as an alternative to conventional chemical intensive lignin removal methods. Besides, they possess the particularity of being able to extract selectively large amounts of lignin from biomass and preserving its quality, along with the possibility of being recycled (Achividu *et al.*, 2018). Furthermore, two main aspects allow a nearly complete recovery of PILs: their reversible exothermic reactions, and their large difference in volatility compared to that of lignin, which means that PILs can easily and efficiently be recovered after the extraction process via methods such as simple distillation. For full PILs recovery, the anions and cations used for their synthesis must be carefully selected (Achividu *et al.*, 2014).

One of the most promising PILs for lignin extraction is pyrrolidinium acetate [Pyrr][Ac]-PIL. Its high ionicity gives it an extraction efficiency of up to 76%. There are no conclusive studies concerning bamboo extraction with the use of PILs. But several lignin extraction analyses using PILs were made in other similar growth rate plants, such as corn stover. The lignin functionality and composition appear to be largely retained. Besides, after the PIL distillation process, it is possible to separate the sugars and polysaccharides from the lignin-rich solids via a simple wash step as the lignin in insoluble. Rendering an even purer lignin extract; over 90% of the sugar/polysaccharides were recovered, and about 75% of the lignin in the extractive-free corn stove (Achividu *et al.*, 2014). All of this leads to the conclusion that it is possible to use relatively inexpensive PILs to selectively extract lignin from biomass by using simple processes at near room temperatures (<100°C) and near atmospheric pressure.

Even though the mechanisms for lignin dissolution and regeneration in the PILs are not yet fully understood, several hypotheses have been postulated (Achividu *et al.*, 2018). Consequently, an extraction procedure has not been established, yet. Nevertheless, attention can be given on studies showing which properties favour lignin extraction. In order to test PILs efficiency on lignin extraction, Achividu *et al.*, (2014) used several commercially available biopolymers with a range of PILs and amine reagents; lignin, cellulose and hemicellulose with low, medium and high ionicity. Results showed that

ionicity had a direct influence on lignin extractability; the more ionic, the more lignin was extracted. This suggests that the xylan and lignin solubility in the PILs may originate from the interactions with the salt ions. Furthermore, the results verified that PILs can dissolve large amounts of lignin, but little to no cellulose. Therefore, it proves an exciting approach for the selective extraction of lignin (Achividu *et al.*, 2014). After the extraction process, PILs present another significant advantage; easy recovery/recyclability. This can be done through simple vacuum distillation. Through this process, the PILs were recovered, and the leftover lignin was mostly unmodified. Besides, a separation between lignin and cellulose can easily be done due to the difference in solubility in water of the different remaining compounds (Achividu *et al.*, 2014). However, it is worth mentioning that there is a compromise to be done regarding the choice of PIL; PILs with strong ionicity have very good lignin extractability, but contained some amide impurities after recovery/recyclability, while compounds with lower ionicity where recovered as near pure PILs, but are less efficient on lignin extractability (Achividu *et al.*, 2014).

Conclusively, it can be stated that even though ILs are a novelty, they are already attracting a lot of attention from researchers and industries for their wide range of applications, outstanding performances and for being considered as a green chemical with little or no consequences on the environment. So far, research shows that PILs are the most promising when it comes to lignin extraction. Their relatively easy and cheap obtention and recyclability, without forgetting a high-steady performance throughout every extraction cycle, make them a very suitable economic alternative to common solvents. In addition, they are safe from an environmental and user-manipulation perspective which is not only good for the wellbeing of workers and the environment today, but also most probably be the answer to future EU regulations. Nevertheless, further research is needed in order to better understand the nature and mechanisms of bamboo lignin within the plant. Even further research to determine which IL, in their vast availability and countless combinations, are most adapted for bamboo lignin extraction. And last but not least, trials that will allow an adjusted extraction procedure at an industrial scale, resulting in the most sustainable procedure possible.

Conclusion

It can be concluded that it is indeed technically feasible to extract lignin through a wide range of methods. Purity of the recovered lignin increases respectively with soda, organosolv and ILs. Among extraction methods, there are a lot of choices to be made concerning scale of the process, desired purity, preferred availability of knowledge on application of both process and in combination with bamboo, *et cetera*. Also, within extraction methods, there are a lot of options to choose from; soda appears to be the most basic process; has the benefit on being practiced at large scale and its applicated on bamboo has been proved; knowledge on organosolv is also available on both process and application to bamboo, and the process delivers lignin with high purity but it comes with environmental drawbacks and may be health-threatening for its users; ILs extract lignin with high purity and seem promising on environmental, health and economical aspects. But their utilisation is still at an early stage and a lot of knowledge is still missing at different levels; the nature and mechanisms of lignin within the bamboo plant structure, the relationship PILs and bamboo lignin have during and after the extraction process as well as the technological and environmental implications if this extraction procedure was to be implemented on an industrial scale. Indeed, the most adapted ILs must be determined out of the wide range of available ILs and even wider possible combinations,

without forgetting the adjustments in time of extraction, quantities, temperatures, etc. in order to make it effective and cost-efficient.

Chapter 3: Lignin as a binder

Lignin is naturally present in all plant material where its structure helps to keep cells together and provides support. Besides, support prevents degradation and rotting of cellulose. When lignin is recovered it could be used in the same way. Since the lignin is altered and cut loose from the cell walls during the extraction process, it needs to be altered in order to work again. The original lignin structure can be seen as a big network of molecules and in order for it to be removed from the cell walls, this network needs to be cut down into smaller chunks. This network, in turn, needs repairing in order for the lignin to become solid again so it can hold its structure and be used as a binder. One of the ways this is done is to let the lignin react with binding molecules, which can bind 2 separated lignin molecules together, reforming the network, usually called the matrix. These binding molecules are called 'cross linkers'. This reaction solidifies the resin, but there is no way to reverse it easily. Resins that solidify this way are known as thermoset polymers. Because the formed network is large, it will become rigid and stay this way even at higher temperatures (Personal communication with Aart van Vuure, Professor KU Leuven 2019).

The length of the lignin (or polymer) chains is important. If the lignin chains are too short, the resin will be too fluid and it will be harder to completely solidify, leaving the matrix malleable in the end. On the other hand, if the chains are too long, the resin will be too viscous, and it will be harder to form a good bond with the wood. Longer chains will also cause the matrix to be more rigid, which makes it more prone to breaking. In an optimal combination of matrix with fibre, the matrix is less rigid then the fibres. If this is the case the fibres will take the stress from bending or stretching, since this is the strong point of the fibres. If the matrix is not rigid enough it will allow the fibres to move more freely from each other. When the fibres can move like this, they will transfer the stress from themselves on to the matrix. This will negatively impact the end composite material, since the fibres normally can endure much more stress than the matrix making it a weaker composite overall. (Personal communication with Aart van Vuure, Professor KU Leuven 2019)

Currently on the market the most similar resin to lignin is phenol-formaldehyde (PF). This resin consists of a large quantity of phenols, also present in lignin, and could, therefore, work in a similar way. PF is currently employed in two-thirds of the wood industry, which makes it the most used commercial adhesive. PF is, just like lignin, resistant to moisture which gives it a good weather resistance. It also is very stable under elevated temperatures. First, lignin-formaldehyde resins will be discussed after which completely biobased resins will be analysed, namely the lignin-furfural resin and the possibility of a pure lignin resin.

Lignin-formaldehyde resin

The extraction method of the lignin alters the structure of the lignin as discussed in chapter two. These alterations change the way lignin will behave when used as a binder. Besides the extraction methods, the origin of the lignin also influences the binding capacity. In the study by Kalami, Somayyeh *et al.* (2018) different extraction methods from different lignin sources were compared. In this paper they compared the lignin against a phenol formaldehyde resin, which is the resin that lignin could potentially compete against due to its similarities. In the study the lignin was combined with formaldehyde as cross linker in the resin. A cross linker is used in combination with a resin – in this

case lignin – and binds two different lignin molecules together. This way the lignin is made into what is effectively a net in which the fibres are bound.

In the study by Kalami, Somayyeh *et al.* (2018), the lignin from hardwood, softwood, and corn stover were extracted to compare. The extraction methods that were tested in this paper exceed the scope of what is discussed in this report. Besides organosolv and soda extractions, this paper also looks at the Kraft process as well as enzymatic hydrolysis and sulphite extractions. This report did not look at lignin recovered from bamboo, but these results can give some insights in what the results could be when using bamboo.

Lignin was prepared by mixing it with 1 M sodium hydroxide and 37% formaldehyde solution. The amount of solution that was added to the lignin was calculated beforehand. The amount of phenolic hydroxyl groups in the lignin was determined for every lignin variant. The molar ratio of phenolic lignin to formaldehyde was set at 2:1. This means that for every two phenolic hydroxide groups, there is one formaldehyde group to connect them both. The more formaldehyde crosslinks are formed in the resin, the more rigid and strong the composite will be in the end. Not every formaldehyde will be able to bind to two lignin molecules together, so there is a minimum threshold of formaldehyde required to ensure that there are enough cross linkages in the final product. On the other hand, if there is too much formaldehyde present in the resin it will reduce the amount of crosslinking in the overall resin. This is because the place at which the formaldehyde can bind to the lignin could already be occupied by another formaldehyde, thus obstructing the crosslinking.

With the lignin resins prepared in the way described above, several tests were performed. In these tests, the gelation time as well as curing temperature were tested. While the lignin binders were all very comparable to the phenol formaldehyde resin, they were generally less viscous to begin with, but had a shorter gelation time. This means that the time required for the resin to turn from a viscous liquid into a semi-solid gel is faster. This increases the process time, but also has the downside that the time to apply the binder to the bamboo is significantly shorter. After the resin is gelated, it needs to cure at elevated temperatures to properly glue the bamboo together. In these tests, the lignin showed very comparable results to the phenol formaldehyde resin, namely a curing temperature around 160°C as shown in table 6. The only real noteworthy exceptions are the hardwood lignin

Sample ID	Solid Content (%)	pН	Viscosity (mPa. s)	Main Curing Temperatures ("C)	Enthalpy (j/g)
L1- Kr-HW	30 (0.1)	13.0	340	121	230
L2- EH-CS	34 (0.1)	13.3	530	168	116
L3- Kr-SW	29 (0.1)	13.2	380	166	175
L4- Os-SW	33 (0.2)	13.1	1100	167	60
L5- Os- HW	26 (0.2)	13.0	4610	127	110
L6- Su-SW	35 (0.5)	13.2	140	137	165
1.7-0s-CS	27 (0.3)	13.4	570	161	47
L8- Kr-SW	29 (0.1)	13.1	610	162	80
L9- So-WS	29 (0.1)	13.1	320	145	127
Lab PF	42 (0.6)	13.1	2179	160	137

Table 6: Characteristics	of lignin resins	(Kalami, Soma	yyeh et al.,2018)
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Note: SW = Softwood, HW = Hardwood, CS = Corn Stover, WS = Wheat Straw, Kr = kraft, EH = Enzymatic Hydrolysis, OS = organosolv, Su = Sulfite, and So = Soda. samples, which cured at a significantly lower temperature. It is improbable that lignin from bamboo will show the same properties as hardwood seeing as bamboo is a grass. This hypothesis, however, must be confirmed by testing.



Figure 9: Shear strength of lignin resins compared to Phenol formaldehyde (Kalami, Somayyeh et al., 2018)

What is also noteworthy is that the solidifying reaction of a phenol formaldehyde resin undergoes an endothermic reaction before it goes exothermic and solidifies. This means that heat is required to start the crosslinking reaction. The lignin samples did not have this endothermic reaction and their solidifying process is purely exothermic. Due to this it is quite likely that the crosslinking reaction will take place at any temperature and an elevated temperature will just increase the reaction speed. This means that the lignin-resins need to be stored in cold conditions to slow down their curing speed before it is applied, and the resin will only be able to be stored for shorter amounts of time.

Another interesting test that was done was the testing of the shear strength of lignin resins in comparison to the phenol formaldehyde resin. While the lignin resins in general came out weaker than the petrochemical resin, the results were very comparable, as can be seen in Figure 9, which uses the same abbreviations as described in table 6. It should be noted that performance of the two best results in this test were limited by failure of the wood rather than the resin itself. This means that the break happened in the wood and not in the part where the two wood panels were glued together. These two adhesives could probably have performed better.

While these tests cannot be compared one to one with lignin from bamboo, they do show that it is possible to make a resin out of lignin. As of yet, it the mechanical properties are not as good as the petrochemical standard, but for non-loadbearing constructions it should be more than good enough.

Completely biobased resins

Besides wood adhesives partially based on lignin and partially on oil/petrol derived chemicals, much has been done to find completely biobased resins in the last years. The first trials happened while trying to reproduce the well-known PF resins. Therefore, lignin was used instead of the phenolic compounds and many biobased crosslinkers were investigated. The most promising seem to be furfural, polymethylene polyphenyl isocyanate, polyethyleneimine, tannins and glyoxal, being proven to be non-toxic and environmentally friendly (Ang *et al.*, 2019; Dongre *et al.*, 2015). A second research area that was explored, was instead detached from the traditional idea of petrol derived binders. Indeed, lignin was found to be used as adhesive without the need of a crosslinker (Ang *et al.*, 2019; Dongre *et al.*, 2015).

In the next section, lignin-furfural resins will be discussed more into depth to give an example of complete biobased two-components-resin. Furthermore, the approach of using lignin alone as binder will be discussed extensively pointing out technical limitations and feasibility.

Lignin-furfural resin

As mentioned above, a potential adhesive derived from bamboo lignin can be the lignin-furfural resin. Furfural was chosen out of the listed crosslinkers because it is the most studied. Even though this is true, the availability of studies focusing on this topic is still very limited. However, the studies that are available describe it as a promising adhesive (Dongre *et al.*, 2015; Sakostschikoff *et al.*, 1934). As compared to formaldehyde, used for lignin-formaldehyde (LF) resins, furfural or furfuryl alcohol can be completely biobased (Fusaro *et al.*, 2015; Agirrezabal-Telleria *et al.*, 2013; Hu *et al.*, 2012; Binder *et al.*, 2010; Dias *et al.*, 2005). Apart from the typical synthesis, based on pentose and/or hexose sugars, this phenolic compound can also be purified from depolymerized lignin, obtaining an even more circular and sustainable chain (Xie *et al.*, 2012).

From a technical point of view, Donger *et al.* (2015) provided an extensive study comparing different furfural concentrations and different conditions. Reporting the findings of these experiments and other articles, lignin-furfural resins should be applied in acidic condition in order to be effective (Donger *et al.*, 2015). Being more specific, a pH of 0.3, 0.6 and 1 were found to be suitable. However, the optimal pH must be defined also according to the furfural concentration. Indeed, an interaction between the two factors was found. At low pH (0.3 mainly but also 0.6) acidolysis reactions occur producing short chain molecules that can work as crosslinker, which diminishes the need of furfural (Li *et al.*, 2008, Roberts *et al.*, 1988). In practical terms, the mechanical properties of the resin were better in low pH for equal furfural concentration, specifically 5% (see Figure 12). However, by increasing furfural concentration (up to 8-16%) at pH1, the binding capacity of that mixture was comparable to the one at lower pH and lower crosslinker content (see Figure 12).



Figure 10: Effect of pH on lignin-furfural resins tensile properties. The adhesives were applied at 180°C and under 1.9Mpa pressure. Blue bars represent the tensile strength whereas the orange one the elastic modulus. PF= Phenol-Formaldehyde, the commonly used petrol derived resin as a standard comparison.

(Codes interpretation: $Fx_y \rightarrow F=$ lignin-furfural resins; x=furfural concentration (0%, 5%, 8%, 16%); y=pH). From Donger *et al.* (2015)

Looking at the other factors playing a role in the adhesive mechanical properties, Donger *et al.* (2015) tested two different temperatures, i.e. 150°C and 180°C, and two different pressures, i.e. 1.9Mpa and 3.8Mpa, and the combination of these two factors. In contrast with what described for the pH and furfural content, there seems to be no interaction between temperature and pressure. Lower pressures and higher temperatures favoured better mechanical properties both alone and combined. Here again, no optimal conditions were found but these data provide an idea of which conditions may suit better for a lignin-furfural resin production, namely 180°C at 1.9Mpa.

Lignin (alone) as a binder

Despite two-components-resins, lignin as a biomolecule can also be used alone as adhesive in the lamination process. Unfortunately, literature is not consistent regarding this topic. As a brief contextualisation, this biobased polymer can be used as such or it can undergo an activation process. There are many different activation processes; methylolation (hydroxymethylation), glycosylation, oxidation, phenolation, and demethylation are some examples (Azarpira *et al.*, 2014; Malutan *et al.*, 2008; Chakar *et al.*, 2004; Zhao *et al.*, 1994). Despite the different procedures, the common goal is to make lignin more reactive, by removing inert functional groups or adding reactive ones. The effective increase in reactivity and therefore better performance as a binder is undoubted. However, these benefits come at a cost, both economic and environmental, usually making it a less valuable option (Mansouri *et al.*, 2011).

As it can be noticed from Figure 10 Donger *et al.*, (2015) claims that unmodified lignin, without any addition of furfural (F0-0.65 \rightarrow lignin with 0% furfural at pH 0.65), can have mechanical characteristics similar to the commercial PF resins. Similarly, Hu *et al.*, (2011) proved that unmodified lignin can be used as a binder, with good tensile properties, but it requires longer pressing times and higher temperatures as compared to activated lignin. Many other articles instead claim that EOL, kraft lignin, soda lignin as well as other types of lignin, have a too low reactivity and therefore need an activation process before being used as binders (Ang *et al.*, 2019).
This literature's inconsistency can be derived by different lignin origins. However, many articles report in the material and methods section, for example, just 'kraft lignin' or 'EOL' instead of the plant species used for the extraction. Furthermore, when this information is specified, it usually varies a lot, which limits the capability to execute adequate comparisons. Unfortunately, these results imply that further research should be performed to determine the boundaries of the use of unmodified lignin as an adhesive.

Conclusion

There are three main ways in which lignin can be used as a binder. Either with petrol derived, biobased crosslinkers or alone. In this range of options there are some, like the LF resin are technically and economically feasible but are still using petrol derived chemicals, raising environmental issues. More sustainable options, like complete biobased resins, seem promising but it is not consistent across literature. Therefore, doubts about their practical feasibility arise. Furthermore, economic analyses of these methods are lacking. In conclusion, there are some ready-to-use options on the market to exploit lignin as wood adhesive. However, they are not the most sustainable options. On the contrary, the most promising options proposed above lack the full scientific community support.

Chapter 4: Sustainability and economic feasibility extraction methods

In the last two chapters, organosolv, soda and PILs were introduced as a lignin extraction method. Furthermore, the possibilities of forming bamboo-based lignin into a binder were presented. In this third and last SQ, the environmental and economic impact of these extraction methods will be discussed to determine if a lignin-based binder does indeed result in a more sustainable bamboo value chain. The sustainability – conceptualised below – of the traditional Asian-based bamboo value chain and novel European-based bamboo value chain will first be analysed. Secondly, a stakeholder analysis will be performed as a critical element of both value chains with two different origins. This stakeholder analysis will later be used to analyse the social sustainability of using bamboo-based lignin as a binder. After the description of the bamboo value chain and its stakeholders, and economic and environmental assessment will be conducted on the organosolv, soda and PILs extraction methods. This chapter will end with a conclusion on the different extraction methods in terms of their economic feasibility and environmental impact.

Description of the value chain

Asian-based bamboo value chain

Before diving into the most promising lignin extraction methods and in order to understand how such methods could or could not improve the sustainability of the European-based bamboo value chain, the current Asian-based bamboo value chain must be understood. The scheme in Figure 11 is an adaptation of the production system suggested by Vogtländer, Van der Lugt & Brezet (2010) for Asian-based bamboo imported to The Netherlands, to which the waste streams and their possible applications have been added. This scheme starts at the plantation in China until the warehouse in Rotterdam, the Netherlands. What comes forward in this scheme is the many kilometres bamboo



Figure 11: Asian-based bamboo value chain (Vögtlander, van der Lugt & Brezet, 2010).

must travel from point A to point B in this scenario, and that residues from the plybamboo factory seem to be excluded as a by-product in the value chain.

Bamboo can be transported in many forms and application: as raw material (stems), as plybamboo after being processed for floors and Strand Woven Bamboo (SWB) as a building material. All these distinct applications have a different impact on the environment and on the bamboo value chain. For example, plybamboo and panels are more efficiently stacked than bamboo furniture, which enables bigger volumes being transported per transportation unit. Notwithstanding, it is still possible to find commonalities. The following graphs by Van der Lugt (2008), show the eco-cost of plybamboo (Figure 12), bamboo stems (Figure 13) and SWB (Figure 14), respectively, throughout its entire value chain. Eco-costs are costs that should be made to reduce the environmental pollution and are thus the environmental burden of a certain phase in a product's value chain (Van der Lugt, 2008).



Figure 12: Eco-costs of plybamboo (Van der Lugt, 2008).



Figure 13: Eco-costs of bamboo stems (Van der Lugt, 2008)





Figure 14: Eco-costs SWB (Van der Lugt, 2008).

While the Figure s presented above cannot be compared directly with each other, they are helpful to identify the activities in the bamboo's life cycle that have the highest eco-costs. It is clear that steps of the value chains of different bamboo applications like stems, plybamboo and SWB have the greatest eco-costs. Firstly, transportation from harbour to harbour presents one of the greatest eco-costs in the bamboo value chain as shown in Figure s 12 and 13. The reason for this is the emission of greenhouse gasses linked to transportation of the bamboo by trucks and ships. Indeed, most of the bamboo used in Europe nowadays comes from Asia. Secondly, for some bamboo applications like SWB and plybamboo, the glue application also presents considerable eco-costs. SWB, for example, is a relatively new industrial material; it is very dense due to a combination of bamboo stripes and a high resin content (Van der Lugt, 2008). Being these resins petrochemical compounds, they have evident burdens onto the environment as seen with the eco-costs in Figure 14.

By having bamboo plantations within the European Union, the European bamboo market could effectively reduce the environmental costs derived from CO₂ emissions, as well as the economic costs relative to the harbour to harbour transportation. This development has already been set in motion by companies like BambooLogic. However, further research needs to be conducted to properly assess the impact of bamboo cultivation in Europe and to see whether its environmental costs are indeed lower than bamboo cultivated in other continents.

The second eco-costs driver in the traditional Asian-based bamboo value chain is the glue application that are often made of petrochemical binders. This is, of course, the main focus in this report, since it is being researched whether lignin from bamboo residues can create a sustainable binder even though possibilities remain open for alternative utilisations of pure and transformed lignin. Firstly, if using lignin as a biobased binder proves feasible for such bamboo material, the impact associated with its

production would be significantly lower, making the bamboo value chain more sustainable. Secondly, by utilising bamboo residues, waste is prevented (Figure 15), which improves the bamboo value chain circularity.

European-based bamboo value chain

This report has incorporated all the formerly mentioned aspects into a relatively more sustainable and circular European-based bamboo value chain as depicted in Figure 15. It should be noted that the assembly of the bamboo product at this moment is assumed to be happening in the warehouse of ArtEZ, but that a shift could also occur when ArtEZ will import bamboo furniture directly as opposed to making it themselves. This is further discussed in the stakeholder analysis.





Several aspects of this European-based bamboo value chain differ from its Asian counterpart depicted in Figure 15. Firstly, while exact distances were unable to be retrieved by these authors, it can be established that the distance between the bamboo plantation (BambooLogic) and the warehouse of ArtEZ is considerably lower (2300 km on land) than that between Shanghai and Rotterdam (19000 km by sea) as seen in Figure 15. This, in turn, leads to lower eco-costs due to reduced emitted greenhouse gasses. Secondly, this European-based bamboo value chain incorporates retrieving, transporting, and re-using bamboo residues that result in a more circular value chain. The bamboo residues can be used as a lignin binder, that in turn can be transported to the warehouse of ArtEZ to be used in their interior and exterior design products. Furthermore, raw materials are already being processed close to the plantation, which increases the efficiency of transportation.

Description of stakeholders included in value chain

To assess the impact of introducing this lignin-based binder into the European-based bamboo value chain, it is also essential to look at the consequences for the various stakeholders in it, with a stakeholder being: 'any person, group or organisation that is impacted in some way by the action or inaction of another' (OpenLearn, n.d.). In this case, it is anyone concerned about the introduction of a lignin-based binder and the specific extraction method chosen for this. Based on the previously described European-based bamboo value chain, all relevant stakeholders will be discussed, and a stakeholder matrix will be presented.

Bamboo producer

For this project, the European-based bamboo value chain begins in Southern Portugal – Alcoutim - at the bamboo plantation owned by BambooLogic. This Dutch-owned organisation aims to 'make a positive impact with bamboo' that serves several Sustainable Development Goals from the UN 30 Agenda for Sustainable Development (BambooLogic, 2020). The organisation is expanding from its start-up phase of 150 hectares bamboo shoots towards 2000 hectares. It already has a factory on its grounds where the harvested bamboo is processed. Then, transported to, for example, ArtEZ, in Arnhem. BambooLogic has the ambition to add more value to their bamboo products, by wanting to introduce a workshop on their property that could make furniture for consumers (Personal communication with Joost Borneman, 2020). The binder discussed in our report can be of great importance for BambooLogic's plans, since it would allow them to re-use their waste streams and, ultimately, increase their profits. While right now waste streams are used by external companies for 'Bamboepoeder', this innovation would allow bamboo producers to process bamboo residues by themselves. Every hectare of bamboo on their plantation generates around 4 tons of waste every year (Personal communication with Joost Borneman, 21-09-2020). This entails that as BambooLogic proceeds to further stages of the project, the amount of waste produced increases accordingly. For instance, the next phase would bring a yearly production of 8000 tons of bamboo residues.

While the profitability of the binder should be secured, to ensure economic feasibility for companies, this should not happen at the expense of the sustainability of the extraction method. It can, thus, be established that BambooLogic has much interest in this project, and it also has a considerable amount of power through its business relations with ArtEZ Future Makers, as illustrated in the stakeholder matrix (Figure 16).

Local Portuguese population

While the plantation is owned by a Dutch company, it is worked on by the local Portuguese community of Alcoutim county. The county is remote and hard to access, which has led to economic stagnation in the past decades (Carrasqueira, 2006). BambooLogic's prospects of expanding their plantation to 2000 hectares and localising the bamboo value chain in Alcoutim county offers job opportunities for local residents (Personal communication with Joost Borneman, 2020). Furthermore, by generating new work posts and making the local economy circular, BambooLogic receives state subsidies from the Portuguese government, stimulating them even more to have a more local bamboo value chain (BambooLogic, 2020). The local workforce is represented by the ADPM, a non-governmental

organisation specialised in local development. Creating a fully local chain of bamboo production and utilisation – by bringing ArtEZ FM close to the harvest and processing areas - would not only increase the ecological sustainability but also potentially lead to the creation of local employment.

Caution is advised for the 2000 hectares expansion phase that BambooLogic has planned since poor landowners might feel pressure to give up their land. If they would not do so, they risk the plantation moving elsewhere and thus taking away employment opportunities in the region (Rao, 2019). In this sense, it could be established that the local population of Alcoutim county has a big interest in this project, but only a moderate amount of power, as illustrated in the stakeholder matrix.

Non-Portuguese bamboo farmers

Another stakeholder group that should be held into account in this project are bamboo farmers from low-income countries. While they are not included in the European-based bamboo value chain depicted in Figure 16, they should be held into account as the introduction of a lignin-based binder might impact them. The production of bamboo is seen as an excellent opportunity to promote propoor livelihood development in these countries. Countries like Vietnam, Ethiopia and Tanzania have, in recent years, seen a tremendous increase in bamboo exports to the European Union (INBAR, 2018). For example, 35% of the total bamboo exports from Vietnam went to the EU, with a total value of over US\$100 million (VOV, 2017).

Moreover, even though the majority of the bamboo trade is domestic, high-income countries increasingly initiate development programs aimed at building the bamboo industry in low-income countries. This is visible in the EU-IFAD agriculture research for development program and the Dutch-Sino East Africa Bamboo Development program (European Commission, 2020; Pegasys, 2020). These bamboo development programs are meant to mitigate climate change and to upscale the bamboo value chain in the country. Thus, addressing SDG Goal 1 (no poverty), 7 (affordable and clean energy), 11 (sustainable cities), 12 (responsible consumption and production, 13 (climate action), 15 (life on land), 17 (partnerships for the goals) (European Commission, 2020). In other words, localising the European-based bamboo value chain is possibly environmentally more sustainable can mean a decrease in bamboo social sustainability in other parts of the globe. If the BambooLogic pilot proves a success, growth in European-based bamboo can be expected, which will lead to a decline of the EU bamboo imports from low-income countries. These non-Portuguese bamboo farmers have very little power. However, they could, in the long-term, have a significant interest in the development of European-based bamboo, as illustrated in the stakeholder matrix.

Governmental institutions

Governmental institutions can make or break the profitability of a certain value chain, with regards to restrictions or subsidies of certain products (Cabri, 2019). There are multiple levels of governance involved in the European-based bamboo value chain: local (municipal/provincial), national (Portuguese) and transnational (European Union). The local government can, oftentimes, be a restricting factor, due to their overly bureaucratic and difficult procedures (Cabri, 2019). This could be a hindrance with respect to BambooLogic expanding plans and for the space needed to build a workshop where lignin can be extracted and used to build a binder. However, BambooLogic cooperates with a local Alcoutim organisation, that helps in bridging with the local government

(Personal communication with Joost Borneman, 2020). The national Portuguese government would have great interest in localising the European-based bamboo value chain in Portugal, since it would increase employment of its rural citizens. BambooLogic is already working together with the Portuguese public employment institute (IEFP), which provides the former with financial incentives when hiring locals. They are, furthermore, working together with a company that links them with subsidies from the Portuguese government (BambooLogic, 2020). The transnational government institution, the EU in this case, also plays an important role in the European-based bamboo value chain. The free transportation of goods is a cornerstone of the EU and will continue to reveal itself important for the bamboo value chain. If the bamboo value chain is localised in Portugal, for example, it would be unprofitable if import duties needed to be paid for these products. In this sense, it could be established that governmental institutions have a lot of power for the future of the European-based bamboo value chain, but their interests vary between different levels of government.

Research institutes

There are many knowledge gaps that this report is addressing, which means that research institutes could also benefit from the information presented in this report. While many academic articles have been written about lignin extraction in all sorts of biomass, lignin extraction from bamboo is still underexposed. Furthermore, little academic attention has gone to creating a lignin-based binder from bamboo residues. After all, the European-based bamboo value chain is very young, and main bamboo producing countries happen to not have the means to carry out extensive research. In this sense, research institutes could be crucial to European bamboo producers and consumers since they can further help the development of a circular European-based bamboo value chain. These research institutes, which are described in the appendix, therefore have a medium amount of power and interest in developing a lignin-based binder from bamboo.

Suppliers and transporters

While suppliers and transporters will not be directly affected by the outcomes of this research, it could mean the beginning of a value chain shift through the localising efforts of BambooLogic. While suppliers right now are occupied with the transporting processed bamboo with train or truck to, for example, ArtEZ FM, this could change in the future. BambooLogic has expressed their ideas of localising the European-based bamboo value chain in Portugal, which would mean that they would make furniture in Portugal for ArtEZ FM (Personal communication with Joost Borneman, 2020). This could entail a change of transportation for suppliers since to fill trucks or train wagons with furniture is less efficient than filling it with just processed bamboo. This could lead to an increase in trucks or train wagons that would need to be used. In the end, the transporters have a medium level of interest in this new development, but not much power since they are simply a 'middleman' between the producer and consumer.

Media

In the past decade, the European Union has encountered a tendency from consumers for sustainable products produced within the EU (European Parliament, n.d.). While bamboo is until now considered a 'foreign' product to many Europeans, this could change when the general public is aware of bamboo grown in Portugal. The lignin-based binder, in this regard, can be considered as a catalysator for

localising the European-based bamboo value chain. Offline media like newspapers could inform the Portuguese population effectively, due to their proximity to the bamboo plantation. Online media, however, could reach throughout the European Union through online newspapers, YouTube, and other internet mediums (Nguyen, 2010). However, this relies entirely on the advances of other stakeholders in the European-based bamboo value chain make. In essence, 'the media' might not be interested in this new development; however, through smart marketing from European bamboo producers, this could change. If this works, it will have a significant impact on the European-based bamboo value chain since the media has much power in reaching the minds of people, as seen in Figure 16.

Customers and consumers

All the stakeholders in the value chain would not be interested in this new development if there was not anyone to buy the bamboo-based product. ArtEZ FM can be considered as a customer of BambooLogic when acquiring bamboo products that have a lignin-based binder. Consumers are individuals who in their turn, would buy bamboo interior and exterior design products of ArtEZ that use a lignin-based binder. Consumer preference for these new products require additional consumerbased research in the future, but it is possible to predict some future aspects. As mentioned before, the consumption of EU-based sustainable products is on the rise and is predicted to rise more in the future. However, consumers are also concerned with living conditions worldwide, and the potential effect a shifting bamboo value chain might have on non-Portuguese bamboo farmers is not to be underestimated. Therefore, it can be predicted that two types of sustainable bamboo consumers will develop: firstly, more environmentally sustainable bamboo consumers that will be more inclined to buy European-based bamboo because of its low environmental footprint. Secondly, more socially sustainable bamboo consumers that will keep importing bamboo from low-income countries in order to foster the set Sustainable Development Goals in these countries. In essence, customers like ArtEZ can anticipate this behaviour of its consumers by framing their products as such to attract certain environmentally and/or socially sustainable consumers. Due to the wide range of power and interest



Figure 16: Power-interest diagram or stakeholder matrix

levels of customers and consumers, it is not included in the power-interest diagram in Figure 16. However, it can be established that without this stakeholder group, there would be no feasibility of a bamboo-based binder.

Environmental and economic performance of extraction methods

After having assessed the bamboo value chain overall, and key stakeholders that might be of importance when analysing the impact and process of a lignin-based binder from European-based bamboo, the environmental impact and economic feasibility of using organosolv, soda and PILs as extraction method will be analysed.

As mentioned in Chapter 1, the extraction method is very much dependent on the feedstock that is being used and on the desired outcome, amongst others. As such, the available literature showed significant variation, regarding the amounts necessary to perform these methods most efficiently. An initial disclaimer would, then, be that it is not this project's aim to stablish the input and output quantities for each extraction method. The many values provided bellow will serve to compare the extraction methods under analysis, and caution is advised when drawing conclusions from isolated values.

Organosolv

This section will describe the organosolv lignin extraction method in terms of environmental sustainability and economic performance. For the discussion of the environmental assessment, the inputs ethanol, sulfuric acid, and energy (heat and electricity) are considered. Water is also an important input, but it is assumed that the water can be reused in the process or for other purposes, so its impact is negligible (Weiner et al., 2016). For the assessment of ethanol and sulfuric acid, the software SimaPro is used to generate the impacts per kg of ethanol and sulfuric acid. The database used was 'Ecoinvent 3 – allocation at point of substitution – system.' The analysis is based on the method 'ReCiPe 2016 Midpoint (H) V1.03 / World (2010) H.' The potential environmental impact will be discussed according to the following impact categories: global warming potential (GWP), stratospheric ozone depletion potential (SODP), ionising radiation potential (IRP), ozone formation potential (OFP), fine particulate matter formation potential (FPMFP), terrestrial acidification potential (TAP), freshwater eutrophication potential (FEP), marine eutrophication potential (MEP), terrestrial ecotoxicity potential (TEP), freshwater ecotoxicity potential (FECP), marine ecotoxicity potential (MECP), human carcinogenic toxicity potential (HCTP), human non-carcinogenic toxicity potential (HNCTP), land use potential (LUP), mineral resource scarcity potential (MRSP), fossil resource scarcity potential (FRSP) and water consumption potential (WCP). To analyse the environmental performance in terms of energy use, the electricity losses will be evaluated according to literature.

In order to evaluate the organosolv method in terms of economic performance, a literature analysis will be done.

Environmental assessment

This section will discuss the findings of the basic calculations of the inputs and the literature search on the environmental impacts.

Several studies discuss the quantity of ethanol used for the organosolv extraction of lignin from bamboo residues. Based on the articles of (Bai, Ziao, Shi and Sun, 2013; Wu, Shi, Yang, Liao and Yang, 2017; Choi *et al.*, 2019; Fan, Ruan, Liu, Wang and Tu, 2015; Kim, Jang, Hong, Choi and Choi, 2016; Li *et al.*, 2012 and Mabrouk, Erdocia, Alriols and Labidi, 2018), together with the help of the technical experts working on the project, the quantities for ethanol use were determined. The quantity of ethanol used in the organosolv process varies between 5 and 8 litres per kilogram of biomass. This number is the total ethanol used, both for the mixing step as for the washing process. In the calculations, 6.5 litres of ethanol are used. Using a range of the ethanol quantity is considered, however since these calculations function more as an illustration this is left out for clarity reasons. If ArtEZ FM would decide to use the organosolv method, they can easily calculate the potential impact again with their optimised quantities for the inputs.

The same studies from the last paragraph were used to find the quantity of sulfuric acid used per kg of biomass. The quantities found varied between 2.74 ml and 10.95 ml. However, the most studies used 5.44 ml of sulfuric acid, so for the impact calculations, the mode of 5.44 ml will be used.

With the aforementioned input quantities, the potential environmental impact was calculated with SimaPro for both ethanol and sulfuric acid. The results can be seen in table 7.

 Table 7: Potential environmental impact ethanol and sulfuric acid for organosolv method.

 CO_2 = carbon dioxide; CFC11 = trichlorofluoromethane; Co-60 = cobalt 60; NO_x = nitrogen oxide; PM2.5 = particles with a diameter less than 2.5 micrometres; SO₂ = sulfur dioxide; P = phosphorus; N = nitrogen; 1,4-DCB = dichlorobenzene; Cu = copper

Impact category	Unit	Ethanol	Sulfuric acid	Total
Global warming	kg CO2 eq	4,7080	0,0012	4,7092
Stratospheric ozone depletion	kg CFC11 eq	0,0000	0,0000	0,0000
Ionizing radiation	kBq Co-60 eq	0,1178	0,0001	0,1180
Ozone formation, Human health	kg NOx eq	0,0118	0,0000	0,0119
Fine particulate matter formation	kg PM2.5 eq	0,0099	0,0000	0,0099
Ozone formation, Terrestrial ecosystems	kg NOx eq	0,0121	0,0000	0,0122
Terrestrial acidification	kg SO2 eq	0,0322	0,0001	0,0323
Freshwater eutrophication	kg P eq	0,0011	0,0000	0,0011
Marine eutrophication	kg N eq	0,0042	0,0000	0,0042
Terrestrial ecotoxicity	kg 1,4-DCB	11,6880	0,0289	11,7169
Freshwater ecotoxicity	kg 1,4-DCB	0,0794	0,0001	0,0795
Marine ecotoxicity	kg 1,4-DCB	0,1021	0,0002	0,1023
Human carcinogenic toxicity	kg 1,4-DCB	0,1124	0,0001	0,1125
Human non-carcinogenic toxicity	kg 1,4-DCB	3,0848	0,0051	3,0899
Land use	m2a crop eq	5,7684	0,0001	5,7685
Mineral resource scarcity	kg Cu eq	0,0118	0,0000	0,0118
Fossil resource scarcity	kg oil eq	0,8451	0,0016	0,8466
Water consumption	m3	0,6560	0,0003	0,6563

Due to the variety of units, it is not possible to make conclusions about the values in the table yet. However, in the conclusion of this chapter, the table will be used again to compare the extraction methods. What can be seen in the table, however, is that ethanol has more environmental impact than sulfuric acid. The greatest difference is seen for global warming, terrestrial ecotoxicity, human non-carcinogenic toxicity and land use.

Yadav *et al.* (2020) indicated the source of electricity and the use of chemicals as the main hotspots of environmental impact. Thus, to further analyse the environmental impact of the specific case it might be interesting to look more into the sources of energy and ethanol. Regarding the impact categories for ethanol, the main potential environmental impact lies in the toxicity (human toxicity and terrestrial

toxicity potential) according to Carvajal, Gómez and Cardona (2016). Furthermore, they found high values for photochemical oxidation potential, which they think is caused by the use of ethanol as it consists of hydroxyl radicals. Hydroxyl radicals (OH) contribute greatly to the production of photochemical smog (Carvajal, Gómez & Cardona, 2016).

The energy requirements for the organosolv process are quite high, mainly caused by the utilities. The utilities consist primarily of cooling and heating during the process (Carvajal, Gómez & Cardona (2016). In terms of electricity the organosolv process requires an input of 13 TJ/y for a biomass input of 1111 kt/y according to the study of Moncada *et al.* (2018). The other energy requirements are as follow: 998 TJ/y for cooling water, 1025 TJ/y for the LP steam and 350 TJ/y for the MP steam. This indicates that most energy is needed for the steam or utilities (58%). Per kilogram of biomass, this means that the treatment consumes 2.4 MJ. The biomass considered in the study is woodchips (Moncada *et al.* (2018). Nitzsche, Budzinski and Gröngröft (2016) found a value of 2.5 MJ/kg biomass for beech wood.

Organosolv has good results on circularity. As mentioned before, the water that is used can be recycled and reused in the process or can be used for other purposes like hydrothermal carbonisation, generating biogas or as fertiliser (Weiner *et al.*, 2016). Furthermore, the solvent can be recovered and reused in the process (Ferreira & Taherzadeh, 2020; Park, Kim & Kim, 2018).

Economic assessment

The organosolv treatment is a relatively new technology, which causes it to be somewhat unclear regarding the (investment) costs for large scale production. However, Moncada *et al.* (2018) calculated the total initial capital investments to be €210 million for a production plant that uses organosolv to extract C6-sugars from spruce and corn. Table 8 shows how this NPV is calculated. The amount of biomass the plant processes is 1111 kt/year, thus 126,742.25 kg/h. According to Mabrouk, Erdocia, Alriols and LAbidi (2018), the total cost of the equipment needed for organosolv extraction is

Table 8: Net Present Value organosolv (Moncada et al., 2018)					
Feature	Organosolv (System I)				
	M€/y	Share (%)			
Operating costs					
Raw materials	121.7	62%			
Utilities	36.0	18%			
Maintenance	14.5	7%			
Labor	2.0	1%			
Fixed & general	13.4	7%			
Overhead	8.6	4%			
Total	196.1	100%			
Revenues					
C6 sugars	107.6	49%			
Lignin	100.5	46%			
Furfural	10.6	5%			
Digestate	-	_			
Electricity	-	_			
Gluten feed	-	-			
Germ	-	_			
Gluten meal	-	_			
Total	235.8	100%			
Fixed capital investment					
M€ 210					
Net present valu	e after t	axes "			
M€	-119				

€250,862.54 for a biomass stream of 106.5 kg/h. Important to note is the difference in the amount of biomass that the two studies use. It shows that having a larger plant is relatively cheaper and thus more efficient.

Furthermore, Moncada *et al.* (2018) performed Net Present Value (NPV) calculations and found a negative NPV. The NPV after taxes was found to be \notin -119 million, which means that that the organosolv method turns out to be economically unfeasible. These calculations were based on a lignin price of \notin 630/ton of lignin. In the study it is stated that the price of lignin should be raised to \notin 751/ton of lignin to reach the break-even point.

Carjaval, Gómez and Cardona (2016) found even worse NPV numbers. They calculated the NPV for two different types of biomass, sugarcane bagasse and rice husk. The NPVs turned out to be \notin -366,09 million and \notin -286,14 million for sugarcane bagasse and rice husk, respectively. Furthermore, they found that the cost of utilities contributes most to the total costs (45%). More details of the data used in this study can be found in the appendix.

Thus, according to the NPV, which is a strong tool to measure economic feasibility, it is not advisable to use the organosolv extraction method based on its economic performance. However, the studies mentioned above are not necessarily using Moso bamboo as a biomass. Therefore, it is recommended to further look into the specifics of the costs when the design of the process is clear. This means for example looking into the specifics of Moso Bamboo, the location of extraction and minimal wages. Furthermore, there is still some uncertainty about the economic performance when organosolv is applied on large scale.

Soda Method

The soda extraction method is a commonly used method due to its simplicity and low financial costs. Its only required inputs are sodium hydroxide (NaOH) – a versatile chemical used by a variety of industries including manufacture of petroleum products, pulp and paper, alumina, textiles, and soaps and detergents (MarketWatch, 2020) – and an acid, which choice can vary from a range of acids such as CO₂, sulfuric acid, phosphoric acid, hydrochloric acids and organic acids (Gilarranz, Rodriguez, Oliet, and Revenga, 1998). Sulfuric acid is the most used chemical for being cheap and more environmentally friendly than other chemicals. Likewise, most available literature is on sulfuric acid, as a precipitation agent.

This process can be carried out using different temperatures, soda concentrations, precipitating agents (acids) and reaction times (Sipponen *et al.*, 2013; Jonglertjunya *et al.*, 2014; Carvajal *et al.*, 2016), which will translate into different environmental and economic impacts. One important remark is that when used to extract lignin, this method produces sulphur-free lignin which adds value to the material and, ultimately, to the process (Van den Bosch *et al.*, 2020).

In order to assess the economic and environmental feasibility of the alkaline extraction method, it is important to address the inputs and outputs of the procedure. Whilst the former will be measured by the solvent, water, acid, and energy consumptions (heat and electricity), for the latter the analysis will focus on lignin and pulp yield and waste stream production. Furthermore, although a comprehensive analysis will be done on sodium hydroxide and acid, for they are the main inputs of this method, the

remaining factors will be considered through a comparative analysis with those used on the organosolv extraction method.

Environmental Assessment

As done for organosolv, the same analysis using SimaPro software was conducted for the soda process. From this program, the values of each impact category per kg of sodium hydroxide and sulfuric acid produced were extracted. In addition, information found in scientific articles, regarding the amount of sodium hydroxide and acid necessary for the soda extraction method, was used to calculate the environmental impact of each component when used for that purpose. In this report, the amounts of sodium hydroxide and sulfuric acid used are 0.4kg per kg of biomass and 2.28kg per kg of biomass, respectively (Carvajal *et al.* 2016; Barros *et al.* 2018). Table 9 shows, for every impact category addressed for the organosolv method, the impact of both sodium hydroxide and sulfuric acid regarding the soda method.

Later in this chapter, these numbers will also be compared with those of organosolv in order to be able to make conclusive remarks on the relative sustainability of soda.

The results shown it table 9 will allow a comparison between the soda and organosolv method. As of now, it is not possible to compare between different impact categories, since they have different units. It is, however, possible to say that sulfuric acid presents a bigger environmental impact than sodium hydroxide for 12 out of 17 categories with a striking difference on TEP.

Table 9: Potential environmental impact, per impact category, of sodium hydroxide and sulfuric acid. The total was calculated by summing the values of sodium hydroxide and sulfuric acid.

 CO_2 = carbon dioxide; CFC11 = trichlorofluoromethane; Co-60 = cobalt 60; NO_x = nitrogen oxide; PM2.5 = particles with a diameter less than 2.5 micrometres; SO₂ = sulfur dioxide; P = phosphorus; N = nitrogen; 1,4-DCB = dichlorobenzene; Cu = copper

Impact category	Unit	NaOH	Sulfuric acid	Total
Global warming	kg CO2 eq	0,5128	0,2810	0,7938
Stratospheric ozone depletion	kg CFC11 eq	0,0000052	0,0000019	0,0000071
Ionizing radiation	kBq Co-60 eq	0,0565	0,0310	0,0874
Ozone formation, Human health	kg NOx eq	0,0013	0,0020	0,0033
Fine particulate matter formation	kg PM2.5 eq	0,0012	0,0046	0,0058
Ozone formation, Terrestrial ecosystems	kg NOx eq	0,0013	0,0021	0,0034
Terrestrial acidification	kg SO2 eq	0,0022	0,0152	0,0174
Freshwater eutrophication	kg P eq	0,0003	0,0002	0,0005
Marine eutrophication	kg N eq	0,0000	0,0000	0,0000
Terrestrial ecotoxicity	kg 1,4-DCB	1,6613	6,6271	8,2884
Freshwater ecotoxicity	kg 1,4-DCB	0,0189	0,0288	0,0477
Marine ecotoxicity	kg 1,4-DCB	0,0266	0,0440	0,0707
Human carcinogenic toxicity	kg 1,4-DCB	0,0249	0,0220	0,0470
Human non-carcinogenic toxicity	kg 1,4-DCB	0,5544	1,1731	1,7275
Land use	m2a crop eq	0,0127	0,0148	0,0275
Mineral resource scarcity	kg Cu eq	0,0016	0,0036	0,0052
Fossil resource scarcity	kg oil eq	0,1279	0,3580	0,4859
Water consumption	m3	0,0134	0,0591	0,0725

Categories Unit ReCiPe (midpoin		ReCiPe (mid	point E)	Impact 2002+	TRACI	USEtox
		Value	Process			
Climate change	t CO ₂ eq	1.59	NaCl (19%) + Electricity (72%)	1.58	1.80	
Ozone depletion	kg CFC-11 eq	3.03×10^{-5}	NaCl (74%) + transport (10%)	3.03×10^{-5}		
Human toxicity	kg 1,4-DB eq	4.43×10^{3}	NaCl (28%) + Electricity (69%)			1.14×10^{-4} CTUh/kg
Photochemical	kg NMVOC	13.13	NaCl (27%) + Electricity (65%)			
oxidant formation						
Particulate matter formation	kg PM ₁₀ eq	3.42	NaCl (33%) + Electricity (57%)	2.16 PM _{2.5} eq	1.31 PM _{2.5} eq	
Ionising radiation	kg U235 eq	27.36	NaCl (33%) + transport (40%) + other chemicals (17%)			
Terrestrial acidification	kg SO ₂ eq	10.24	NaCl (27%) + Electricity (60%)	10.19		
Freshwater eutrophication	kg P eq	0.08	NaCl (29%) + Electricity (59%) + other chemicals (11%)			
Marine eutrophication	kg N eq	2.01	NaCl (39%) + Electricity (56%)			
Terrestrial ecotoxicity	kg 1,4-DB eq	1.84	NaCl (91%)			
Freshwater ecotoxicity	kg 1,4-DB eq	2.17	NaCl (33%) + Electricity (61%)			1.69×10^3 CTUe/kg
Marine ecotoxicity	kg 1,4-DB eq	4.10×10^3	NaCl (25%) + Electricity (69%)			
Agricultural land	m ² a	2.90	NaCl (31%) +other chemicals (16%) +steam (35%) + transport (11%)	2.82		
Urban land occupation	m ² a	5 51	NaCl (41%) \pm Transport (33%) \pm Electricity (16%)			
Natural land	m ²	8.81	NaCl (99.7%)			
transformation		0.01	Hact (33.7%)			
Water depletion	m ³	7.26	NaCl (53%) + other chemicals (31%)			
Metal depletion	kg Fe eq	19.24	NaCl (62%) + other chemicals (11%) + transport (20%)			
Fossil depletion	kg oil eq	445.63	NaCl (23%) + Electricity (74%)	19.06 GJ primary		

Table 10: Life cycle impact assessment results and the contribution of most significant process to the mid-point scores. Values are presented per functional unit. (Hong et al., 2013)

This analysis can the supplemented with others found in academic literature. Thannimalay, Yusoff and Zawawi (2013) studied the environmental impact of sodium hydroxide production. They measured this impact based on the following factors: Fossil Energy Consumption (FEC), Global Warming (GW), Aquatic Ecotoxicity (AE), Acidification (A) and Human Toxicity (HT). Indeed, in their analysis, the authors found a common element translating into environmental burden in almost all categories: energy consumption. An LCA done by Hong *et al* (2013) had similar results – energy consumption summed the vast majority of the overall environmental burden (Table 10). As presented in the table, almost all categories attribute a considerable percentage of the environmental impact to electricity consumption. This impact of energy consumption is due to the fact that most energy production worldwide still runs on fossil fuels and, hence, going from a petrochemical binder to a biobased one, as intended by ArtEZ FM, this does not necessarily mean a reduction of that impact. This entails that energy consumption must be considered when analysing and choosing from the extraction methods presented in this report.

Furthermore, a literature review on sulfuric acid production was also conducted. Sulphuric acid production is divided into two subunits: sulphur treatment and sulphuric acid process (Marwa *et al.*, 2017). According to the authors, the former contributes the greatest to 13 out of 16 impact categories



Figure 17: Environmental impact sulfuric acid (Marwa et al., 2017)

assessed in that research - Climate Change (GWP), Ozone Depletion (ODP), Water Resource Depletion (WRDP), Mineral and Fossil and Renewable Resource Depletion (MFRRDP), Acidification (AP), Terrestrial Eutrophication (TEP), Freshwater Eutrophication (FEP), Marine Eutrophication (MEP), Photochemical Ozone Formation (POFP), Human Toxicity (cancer effects) (HTTP,CE), Human Toxicity (non-cancer effects) (HTTP, NCE), Particulate Matter (PM), Ionising Radiation (human health) (IR, HH), Ionising Radiation (ecosystem) (IR, E), Freshwater Ecotoxicity (FETP) Land Use, Cumulative Energy Demand (CED) and Cumulative Exergy Demand (CExD). The sulphuric acid process, on the other hand, is most responsible for the acidification impact category. This is evident in Figure 17 where the most relevant impact categories are place on the X axis and different factors influencing them are compared – transport, equipment, utilities, process, and chemicals. The process is, thus, the most environmentally harmful in all categories except for the Water Resource Depletion. This category was not considered since the water used, mainly as a cooling system, concerns seawater and was considered an infinite recourse (Marwa *et al.*, 2017).

Economic Assessment

For the economic assessment, an example taken from the literature will be provided together with an interpretation fitting this report's aim. This strategy was chosen since, as mentioned already, there is a lot of variation that can influence an economic assessment, including the desired characteristics for this new biobased binder, information which this team was not provided with. There are, however, studies that aimed at comparing the economic feasibility of various extraction methods. By drawing from these, it is possible to achieve an illustrative outline that can afterwards be used in order to get even more tailored information.

Before delving into the case study, it is important to mentioned that, despite the variation found within literature, it is commonly agreed that soda extraction method presents an optimal solution when it comes to financial investment and monetary return. This is due to the low material costs required. This will be further looked into in the comparison section that follows.

In a comparative study where different (lignin) extraction methods were analysed, the following categories were considered: raw material cost, utilities cost operational costs and depreciation (Carvajal et a., 2016). According to Carvajal *et al.* (2016) the greatest financial burden associated to soda extraction is the raw materials' costs. They estimated that around 70% of the overall cost of this process was mainly due to the transportation of these materials, that include the feedstocks (rice husk and sugarcane bagasse, in that case) and reagents, among others. Furthermore, heating, and cooling services express around 25% of the total costs. The expenses associated with transportation are significantly diminished if the extraction took place on a workshop near the plantation, as is indented by the plantation owners (Personal communication with Joost Borneman, 21/09/2020).

Carvajal et al. (2016) provided insightful data regarding production costs of Soda depicted both in USD per kg of lignin and USD per year (Table 12, page 52 of this report). In order to make it more comparable and significant all the USD values were converted into euros. Firstly, when using rice husk and sugarcane bagasse, the production cost per kg of lignin showed to be \leq 4.34 and \leq 2.98 respectively. Secondly, regarding the annualised costs, the value for rice husk was \leq 30.06 million and that for sugarcane bagasse was \leq 29.84 million.

An interesting addition was the payment period, which translates into the time when the profit overcomes the initial investment. Carvajal *et al.* (2016) state that the payment period for soda when using sugarcane is 3 years, while in rice husk scenario, the payment period is only 1 year (Table 12, page 52 of this report).

Although some of the values presented above do no differ much, an analysis specific on Moso bamboo residues should be carried out, in order to have a clearer view on the necessary investment and potential profit. Further analysis will be presented below, where soda and organosolv will be put side-to-side, regarding their environmental and economic feasibility.

To sum up, the soda extraction method is a commonly used method which is simple to apply and can bring profit. This is mainly due to the fact that it requires only two components - sodium hydroxide and an acid – which production is optimised and hence, very cheap. These very components, however, still rely of fossil fuels for its production translating into climate change potential and have shown to have a negative influence on terrestrial and human non-carcinogenic toxicity.

Protic Ionic Liquids

In this section, the environmental impact and economic feasibility of using ILs for extracting lignin from Moso bamboo will be analysed. More specifically, the PILs in general and Pyrrolidinium Acetate will be analysed, as described before. The research on PILs are thought to grow rapidly the coming years, due to its unique properties, having a 'negligible vapour pressure, low nucleophilicity, miscibility with organic solvents, and good extractability for the organic compounds' (Xiao, Chen & Li, 2018). These properties could work together towards extracting lignin from bamboo biomass residues (Greaves & Drummond, 2008). The question that will be mainly addressed is whether PILs processes, and particularly into pyrrolidinium acetate, are more environmentally friendly and economically competitive with extraction methods that use volatile organic compounds (VOCs). In this way, this report does not disregard the potential environmental externalities that PIL's have.

Environmental assessment

Current extraction methods that are used presently in the industry have several features that, in the end, lead to high energy demands. Furthermore, the volatile organic solvents used in other extraction methods often have a considerable environmental footprint. Using VOC's for extracting biomass is often associated with the loss of solvent to the atmosphere, which is environmentally unfriendly as it contributes to climate change (Venture *et al.*, 2017). For this reason, researchers have been looking into extraction methods that are more environmentally benign and sustainable, with ionic liquids receiving increasing attention (Venture *et al.*, 2017). Using ILs as an extraction method implies no loss of solvents into the atmosphere, which therefore reduces its impact on global climate change (Yang *et al.*, 2011).

However, environmental concerns are increasing on ILs because of questions raised arounds its toxicity and biodegradability (Neumann *et al.*, 2010). Furthermore, it is theorised that the usage of IL's at the extraction phase as opposed to VOCs will push the usage of VOCs 'upstream to the production phase' since the VOCs can be used elsewhere (Alviz & Alvarez, 2017). Overall, ILs solvents are considered to have a lower environmental footprint in extraction processes compared to more

traditional volatile solvents. For this reason, ILs have been used increasingly in extracting natural products from plants, like herbal tea (Xiao, Chen & Li, 2018).

When looking more specifically at the sub-family of protic PILs, it can be said that the sustainability of a certain PIL depends on the source of its bioactive compounds. In the case of bamboo residue biomass, it is found that its sustainable character varies. However, PILs are, compared to their aprotic counterparts, more environmentally friendly and, as mentioned before, easier to use (Venture *et al.*, 2017).

Using PILs for extracting biomass is still quite novel, which leads to little environmental assessments being conducted on this. However, one study by Baaqel *et al.*, (2020) conducted an environmental assessment on the usage of 2 types of PILs: [TEA][HSO4] and [HMIM][HSO4] and compared it with the traditional solvents of acetone and glycerol. The results are mixed and show that there is a great variety of environmental impacts that using PILs could have. It is found that the PILs have a lower impact on the ecosystem quality than glycerol, and that they have a lower impact on resources than acetone. Regarding human health, [TEA][HSO4] has the least impact, while [HMIM][HSO4] has a relatively high negative impact on the human health. The authors have monetised the externalities on the environment and human health of the discussed solvents and found that the [TEA][HSO4] PIL has the least direct (see economic assessment) and indirect costs of the four discussed solvents, but that one should pay attention to the components of the PILs as their specific characteristics can result in great efficiency or barely any technical and economic interest.

When looking specifically at the environmental impact of using pyrrolidinium-based ILs, like pyrrolidinium acetate as discussed before, academic literature falls short. It has been found that using pyrrolidinium as an extraction method has a relatively low toxicity level compared to other ILs like phosphonium-based ILs (Seiler *et al.*, 2020; Bubalo *et al.*, 2014). Pyrrolidinium-based ILs are often considered to be 'green solvents' due to their non-volatility which means they have a low vapour pressure and a high boiling point (Moity *et al.*, 2012). Lastly, it was found by Syguda *et al.* (2016) that pyrrolidinium ILs were more environmentally friendly than other ILs due to its high biodegradability, which would lead to less pollution. A further environmental assessment on the usage of pyrrolidinium acetate should be done professionally by a company with the means to assess this.

One advantage of using ILs as extractants over VOCs is that that new regulations in the European Union increasingly restrict and target VOCs because of its environmental impact. This means that, while VOCs might be economically more viable at the short-term, long-term governmental regulations could limit the profitability of these investments (European Council regulation 1907/2006/EC).

Economic assessment

It has been found that some ILs that can be used for extraction are very financially costly, which would prevent a large-scale industrial usage. Researchers have therefore coined the development and research into ILs importance (Xiao, Chen & Li, 2018). These developments are coined as promising, since studies have shown that using ILs as extraction method increases the efficiency of the extraction, which lowers the cost if used repeatedly (Passos, Freire & Coutinho, 2014).

Economic analysis assessments on the usage of ILs are not abundant, but a recent study showed that their application is only economically feasible when the extracted compounds either have a high value or when the concentration of extracted compounds is at least 5 wt%, which is quite high (Passos, Freire & Coutinho, 2014). Furthermore, the high production costs of standard ILs are mainly dependent on its raw materials, which can be seen in the following Figure :



Figure 18: Production costs liquid ions (Passos, Freire & Coutinho, 2014).

PILs, however, are considered to have a lower cost when extracting biomass, which would also count in the case of bamboo residues. Another way that the costs of using PILs as extraction method could be reduced by employing integrated strategies that would allow re-using. One solution that is coined for this, is to re-use PILs in the purification stage that could follow upon extracting the lignin from bamboo residues (Venture *et al.*, 2017). It has been found that re-using ILs does not lead to losses in extraction yields, which increases their cost-effectiveness (Ma *et al.*, 2011).

Passos, Freire and Coutinho (2014) present a way of analysing the economic viability of using PILs for lignin extraction from bamboo residues:

$$R = [C_{\text{prod}} \times \$_{\text{prod}} - \$_{\text{biom}}] - [V_{\text{IL}} \times \$_{\text{IL}} \times r_{\text{IL lost}} \times \alpha + \beta]$$

This formula shows the return (*R*) for extracting lignin from bamboo residues with the extracted concentration of lignin (*Cprod*) times its price per kg (*\$prod*) minus the costs or worth of the bamboo residues (*\$biom*). This gets deducted from the costs of the extraction method, with the volume of PILs needed to treat one kg of bamboo (*VIL*) times the price of PIL per kg (*\$IL*) times the ratio of PIL lost during the process of recycling (*rIL lost*). A represents the proportional costs and β represents other constant costs. Due to limited information on the exact variables for this formula, it is not possible to convert it to the Net Present Value as discussed before. However, it is encouraged to do so when more variables become known.

This formula on the economic feasibility of using PILs as extraction method shows several interesting aspects to keep in mind when analysing the economic feasibility of extracting lignin from bamboo using PILs. Firstly, this extraction method only seems viable when the concentration of extracted lignin (*Cprod*) is sufficiently high; at least 5 wt %, as discussed before. Secondly, it is vital that the lignin has a high added value (*\$prod*) to ensure good returns (Passos, Freire & Coutinho, 2014). While the latter aspect is more easily reached, since the lignin will be processed as a high-value binder for bamboo products. The former requirement depends on the contents of the bamboo residues, of which the

authors of this report do not have enough information. However, the lignin concentration of the bamboo residues can be assumed to be higher than the beforementioned threshold (Bai *et al.*, 2013).

Baaqel *et al.*, (2020) have conducted an economic assessment on the usage of 2 types of PILs. It was found that PILs that need a lot of pre-treatment steps before they can be used are more costly than those that are readily usable. Their prices per kg range from 0.66 for [TEA][HSO₄] to 1.24 for [HMIM][HSO₄]. It was found that 90% of these costs are day-to-day operating expenses needed to keep the operation running, like obtaining the raw materials needed for the PILs and energy needed for the machines. In this sense, it can be established that PILs are not necessarily cheaper or more expensive than traditional solvents like glycerol, but it depends on the specific contents of the PILs and the amount of pre-treatment steps. As described before, the authors also monetised externalities in their economic assessment, as seen in Figure 19. It can be seen here that when direct economic costs and indirect environmental costs are combined, PILs require a lower total cost compared to other solvents like acetone and glycerol used in traditional non-IL extraction methods.



When looking more specifically at the economic feasibility of using pyrrolidinium acetate (PyrrAc) as an extraction method, several authors have found that pyrrolidinium-based PILs have a relatively low costs compared to other ILs (Anouti, 2008). As described before, re-using PILs can have a great impact in lowering the costs for large-scale industry usage. This has also been confirmed for the case of pyrrolidinium ILs (Sobrinho *et al.*, 2020). Pyrrolidinium acetate has been found to be quite efficient in extracting lignin, with studies reporting an extraction rate between 70% and 76% (Achinivu *et al.*, 2013; Achinivu, 2018). Pyrrolidinium acetate has been found to be economically more attractive than other PILs (Jiang *et al.*, 2017), and is now being studied by several biorefineries on different biomass types (Shi *et al.*, 2013; Cetinkol *et al.*, 2010). It is clear, however, that the economic feasibility of using PyrrAc as an extraction method for bamboo residues specifically, should be further analysed.

Concludingly, while much is unknown about the environmental impact and economic feasibility of using PILs, and more specific on pyrrolidinium acetate, several lessons can be drawn to foster further research on PILs. First and foremost, it was found that the recovery and reusability of PILs are vital issues to support the economic viability and to minimize the environmental footprint of the proposed processes. PILs can be very efficient in extracting lignin from biomass, and it does not lose its capacities when re-used, besides having very little losses during the extraction process, which makes PyrrAc and other PILs very suitable. Secondly, to ensure economic feasibility of using PILs for extracting lignin from biomaboo residues, a high level of extracted lignin and a high-value lignin product should be

ensured. Lastly, compared to other PILs, pyrrolidinium acetate can be considered relatively environmentally friendly due to its low toxicity levels and high biodegradability.

Sustainability of a binder

The focus in this chapter has primarily been on the three chosen methods that can extract lignin from bamboo residues. When considering the binder, it can be established that the extraction method has the greatest environmental and economic consequences for the producer. Once the lignin is extracted, the steps towards a binder are described in the previous chapter. To understand if extracting and using lignin from bamboo residues to create a biobased binder results into a more sustainable bamboo value chain, it needs to be compared with the current production chain which uses petrochemical-based binders. This, however, can be grasped from two perspectives: from a circular point of view and a sustainable one. Although complementary, these views bring forward slightly different aspects.

From a circularity point of view, the lignin-based binder might be perceived as better compared to the petrochemical binder. While the latter requires additional, non-renewable resources, the former re-uses natural components that otherwise would be designated as 'waste'. This essentially implies that the lignin-based binder 'closes' the bamboo value chain, while the petrochemical binder 'opens' the bamboo value chain further creating lose ends, which ultimately translate into waste. Since ArtEZ FM is focused on '... the development of completely new, closed value chains based on renewable raw materials...', (ArtEZ FM, n.d.), the lignin-based binder should be preferred over a petrochemical binder.

From an environmental point of view, other arguments can be brought forward. Academic literature is divided between the positive sustainable impact of using a lignin-based binder over a petrochemical based binder. The production of petrochemical binders, which are based on conventional fossil resins has been being 'optimised' by the industry over the past decades, making them more efficient and, in the end, potentially more environmentally friendly then its biobased counterparts (Arias *et al.*, 2020). However, this links to the fact that more research needs to be conducted on lignin-based binders in order to make them environmentally competitive with petrochemical-based binders. It is important to further highlight that the lignin provided from bamboo residues is a waste stream, meaning that it is not coming from plants being specifically grown for 'lignin production'. This makes it more environmentally friendly (Yuan & Guo, 2017). Furthermore, questions can be asked to whether 'optimisation' in an unsustainable system is something to strive for with regards to petrochemical binders. Instead, it is better for the environment in the long-term to invest in novel, sustainable ways.

In essence, this 'dilemma' between circularity and sustainability can be compared of the plastic and linen bags debate. The Denmark's ministry of environment concluded in 2018 that plastic shopping bags have less environmental impact when compared to linen shopping bags (Ministry of Environment and Food, 2018). What seems to be important to consider, is that, in order to reduce waste, single use materials should be discouraged as much as possible. Therefore, in this scenario, it would be better to re-use fabrics to make linen bags than to produce single-use plastic bags. This relates to the sustainability of biobased binders: while a petrochemical binder might be more energy

efficient and thus environmentally more sustainable, re-using 'waste streams' and reducing waste is vital to increase the sustainability of a value chain in the long term.

Comparison

In this chapter, it was analysed to which extent the process and product of bamboo binder really result into a more sustainable bamboo value chain for bamboo products. First, the Asian-based and European-based bamboo value chain were presented. Secondly, a stakeholder analysis was conducted, and a power-interest diagram was drawn to show the impact of introducing a lignin-based binder into the European-based bamboo value chain. Thirdly, the environmental impact and economic feasibility of using organosolv, soda and PILs as extraction methods were analysed. Fourthly, the overall sustainability of a lignin-based binder was discussed. Now, the environmental impact and economic feasibility between organosolv, soda and IIs will compared.

Environmental impact

Regarding the environmental impact for PILs and specifically for Pyrrolidinium acetate, it is not possible to directly compare this with the soda and organosolv extraction methods. Once again, PILs are a novel approach with a limit amount of research conducted on them, making it difficult to employ them in a large scale. However, PILs can be more sustainable than other traditional solvents when emphasis is laid upon re-using them for extracting lignin. Furthermore, for Pyrrolidinium acetate specifically it can be said that it is a 'green solvent' with a relatively low toxicity level and high biodegradability. The usage of ILs compared to other solvents like organosolv and soda can become more favourable in the future, which can be accentuated by new regulations from the EU.

As for the comparison between organosolv and soda extraction, it can be concluded that soda extraction is more environmentally friendly as shown in table 11; calculated as *difference environmental impact = organosolv value - soda value*. The values are calculated for a functional unit of a kg of biomass. The table shows that the soda methods performs better for every category, as all the values are positive.

Impact category	Unit	Comparison
Global warming	kg CO2 eq	3.9154
Stratospheric ozone depletion	kg CFC11 eq	0.0000
Ionizing radiation	kBq Co-60 eq	0.0305
Ozone formation, Human health	kg NOx eq	0.0085
Fine particulate matter formation	kg PM2.5 eq	0.0042
Ozone formation, Terrestrial ecosystems	kg NOx eq	0.0088
Terrestrial acidification	kg SO2 eq	0.0149
Freshwater eutrophication	kg P eq	0.0006
Marine eutrophication	kg N eq	0.0042
Terrestrial ecotoxicity	kg 1,4-DCB	3.4285
Freshwater ecotoxicity	kg 1,4-DCB	0.0318
Marine ecotoxicity	kg 1,4-DCB	0.0316
Human carcinogenic toxicity	kg 1,4-DCB	0.0655
Human non-carcinogenic toxicity	kg 1,4-DCB	1.3624
Land use	m2a crop eq	5.7410
Mineral resource scarcity	kg Cu eq	0.0066
Fossil resource scarcity	kg oil eq	0.3608
Water consumption	m3	0.5838

Table 11: Comparing environmental impact organosolv and soda

Some of the most interesting conclusive ones in table 11 are discussed further in this section. The other graphics can be found in the appendix for a more complete overview.

Figure 20 shows the global warming potential of the two extraction methods. It clearly shows that organosolv has a much higher global warming potential than soda.



Figure 20: Global warming potential organosolv and soda

Other relatively large differences are found for stratospheric ozone depletion, ozone formation (human health), ozone formation (terrestrial ecosystems), marine eutrophication, land use and water consumption as can be seen in Figure 20.



Figure 21: Several other indicators environmental impact organosolv and soda

Concludingly, the soda extraction method is preferred over the organosolv in terms of potential environmental impacts. However, ILs can be favourable over organosolv and soda extraction if an efficient industrial implementation can be established.

Economic feasibility

In previous sections the economic and environmental characterisation of each method was put forward. Due to gaps in the available literature, it was not possible to refer only to articles that compared these methods directly. Instead various articles were used in order to present the most comprehensive analysis possible. This being so, the comparison between different methods analysed by different authors is sometimes impossible due to the unmatched utilisation of parameters. Therefore, for the following comparison, literature is chosen that offered a direct comparison. Most literature compares soda and organosolv and these with other methods such as kraft and sulphite processes. Harder it is, however, to find suitable comparison with ILs, since this is a very recent chemical process with a lot of possible variations, as discussed earlier in this chapter.

Overall, the available literature agrees that the best extraction methods, when considering both economic and environmental sustainability, are those in which lignin is removed without traces of sulphur (Carvajal *et al.*, 2016; Fernández-Rodríguez *et al.*, 2017; Van den Bosch *et al.*, 2020). Both soda and organosolv produce high-value sulphur-free lignin (Carvajal *et al.*, 2016) which lowers the total costs when compared with other methods (such as kraft), where the lignin is exposed to sulphur during extraction. It is, however, still possible to distinguish these two methods by its economic features.

Table 12 shows the production costs per kg of lignin produced. In the table by Carvajal *et al.* (2016) two different feedstocks were used: sugarcane bagasse and rice husk. The production cost using soda is always significantly lower than when organosolv is applied, with values of 3.34 (kg lignin and 2.98) kg lignin, for sugarcane and rice husk, respectively, when using soda, and value of 13.6 (kg lignin and 8.67) kg lignin, for sugarcane and rice husk, respectively, when using organosolv. The high values registered for organosolv are caused mainly by lower yields of organosolv when compared to soda. Additionally, the total cost in one year of production is ≤ 30.06 million for sugarcane and ≤ 29.84 million USD for rice husk, when using soda, while the values when using organosolv, decrease to ≤ 24.01 million for sugarcane and ≤ 22.47 million USD when using rice husk.

Other interesting feature would be the profitability index and NPV, presented by the same authors. These indicators 'aim to establish and express the extent of any economic entity to generate income measuring the increase of the investment per each dollar spent' (Carvajal *et al.*, 2016). Soda shows higher profitability than organosolv for both feedstocks tested. However, it is important to realise that the feedstock choice has high impact on theses parameters, being of great importance to further extend this research to Moso bamboo. As shown in table 12 the NVP value strongly favours the Soda method. In fact, organosolv has negative NVP values for both feedstocks implying strong inefficiency of this method.

	Soda		Organosolv		
	Sugarcane bagasse	Rice husk	Sugarcane bagasse	Rice husk	
Production cost (€/kg lignin)	3.34	2.98	13.6	8.67	
Annualized cost (million €/year)	30.06	29.84	24.01	22.47	
NVP (millions €)	19.04	61.88	-420.41	-374,77	
Profitability index	1.13	1.4	0.16	0.2	
Payment period	3	1	-	-	

Table 12: NVP Value comparison organosolv and soda

Another comparative article, by García *et al.* (2011) states as well that 'the soda process energy requirements were found to be considerably lower than the organosolv-ethanol ones, as the heating utilities (low and medium pressure steam) were generated within the process and the only consumption was water at 20°C. Therefore, the total utilities cost calculated by the authors were 566 000€ per year and 4160€ per year when using organosolv and soda, respectively. Important to mention, however, is that the organosolv method allows for the recovery of 93% and 89% of ethanol and water, respectively, whilst during the soda process only 84% and 81% of NaOH and water is

respectively recovered (García *et al.*, 2011). This entails that a way of improving the soda method even further would be to work on the solvent recovery and ultimately reduce the total production cost.

The comparison of the economic feasibility of PILs with organosolv and soda forms also a difficult task due to the lack of economic information on this novel approach. However, it can be established that the economic feasibility of using PILs as extraction method relies on a high concentration of extracted lignin and a high value of this lignin (as a binder). It can be hypothesized that this concentration and value is high enough, which makes PILs interesting to compare in further research with soda and organosolv. Furthermore, the main production costs of PILs are their raw materials, which means that re-using PILs will be vital for an economically feasible extraction method. This novel method should be further researched in-depth in the future, to be able to compare its true economic feasibility.

To sum up, in light of current knowledge, the soda extraction method it the one that provides better indicators from both economic and environmental feasibility. In a direct comparison between soda and organosolv, the former should always be chosen over the latter. When adding the PILs, however, more research should be done, since it is a promising methodology which is currently not fully understood, but that could potentially translate into an even better environmental and economic approach.

Social sustainability

As discussed in the introduction of this chapter, the 'people, planet, profit' aspects of the lignin-based binder are important to assess its sustainability. While it has become clear that the soda extraction method on the short term is most feasible regarding the 'planet' and 'profit' dimension of sustainability, it is less clear what the general impact of the lignin-based binder would be on the social sustainability. Here the three extraction methods themselves will not be compared as such but be generally looked at as a whole. As mentioned in the stakeholder analysis, different stakeholders will be impacted differently. While Portuguese locals might benefit from this development due to improved employment in the region, this development and the localisation of the European-based bamboo value chain could mean less exports for non-Portuguese bamboo farmers exporting to the EU. The authors of the report therefor urge the importance of knowledge and technology transfers to low-income countries, since these farmers could then also potentially benefit from this development in the future. Close relations need to be upheld with the local Portuguese community to ensure their participation in localising the European-based bamboo value chain. If Bamboologic would be expanding their plantation and building workshops, the approval of the local population needs to be ensured.

Conclusion

To see whether the lignin-based binder from bamboo residues results into a more sustainable bamboo value chain, the following points can be put forward:

Firstly, it was shown here that the European-based bamboo value chain has a clear advantage over its Asian counterparts because of its lower environmental costs related to reduced transportation distances. Moreover, it was concluded that the incorporation of bamboo residues into the value chain could make it more sustainable and circular. This shows then that for the European-based bamboo

value chain, a lignin-based binder is positive for its sustainability. More research, however, needs to be conducted for other aspects of the European-based bamboo value chain, like the impact of bamboo cultivation in Europe.

Secondly, the stakeholder analysis and its power-interest diagram showed the many different impacts a lignin-based binder could have on the European-based bamboo value chain. While on the one hand there are many powerful actors that could steer towards the adoption of a lignin-based binder in the European bamboo value chain, like bamboo producers, governmental institutions and the media, there are also many actors that do not have a lot of power in this regard. It is therefore important to involve all stakeholders of the bamboo value chain to move towards a participatory, sustainable value chain. This also involves looking at stakeholders that might be affected indirectly, like non-Portuguese bamboo farmers in low-income countries. Furthermore, it was established customers might favour circularity over sustainability or vice versa, which is a development ArtEZ should hold into account.

Thirdly, it was found that the soda extraction method is the most suitable candidate for immediate large-scale lignin extraction from bamboo residues. It is relatively environmentally friendly compared to other extraction methods like organosolv, and it is economically feasible to set this up with the soda extraction method. When using soda as an extraction method, it is recommended to use sulfuric acid. More research should be conducted on the environmental impact and economic feasibility of extracting lignin with protic ionic liquids. This method is promising from an environmental point of view, especially with Pyrrolidinium acetate, but to make it economically feasible at large scale more test trials need to be done. Furthermore, it was found that overall a lignin-based binder is superior in circularity over a petrochemical binder. However, environmentally seen there is still much to win for lignin-based binder since they are not yet as optimised compared to petrochemical binders.

Concludingly, it can be said that a lignin-based binder from bamboo residues, extracted with the soda method can lead to a more sustainable bamboo value chain for its products. However, only when great efforts are done by all stakeholders among the European-based bamboo value chain to guarantee the people, planet and profit side of its sustainability this is possible. Furthermore, it is vital that more research and experiments need to be conducted specifically on lignin extracted from bamboo.

Conclusion

This report aimed to evaluate different bamboo lignin extraction methods in terms of technical feasibility, environmental and social sustainability, economic performance and to understand the application of lignin as a biobased binder. The first step was to determine the problems regarding the bamboo production cycle, where can it be improved and what role ArtEZ FM has in this matter. Soon after that the report introduced the state-of-the-art regarding lignin and its current applications, as well as important knowledge gaps that the development of this report would aim to fill. The definition of sustainability as the 'Triple Bottom Line' were provided, highlighting the fact that the report would be considering a multidimensional perspective that includes people, planet and profit.

After having properly defined the background of this project, different extraction methods were analysed. The first goal was to explain the technicalities of organosolv, soda and ILs processes. Thus, this information allowed this report to shed some light on the feasibility of using these methods to extract lignin from bamboo residues. What can be inferred reading the dedicated section, is that all these three methods will enable lignin extraction from bamboo residues, even though each of them have their own drawbacks. By comparing the different extraction processes, this report highlights the fact that the choice is very much dependent on the wishes and demands of the interested parties. If the plan would be to apply this methodology in a short-term, the best and ready-to-use solution would be the soda process. It is the oldest method exposed in this review and, as such, most is already known. Therefore, the soda process is the easiest to apply from the technical point of view and the variables that need optimisation are few. Furthermore, in the conducted research, the soda process ranked higher in terms of environmental friendliness and economic feasibility, when compared with the other methods in a short-term perspective. For this reason, the soda extraction method is recommended to extract lignin from bamboo residues on the short-term. Organosolv will not be recommended, instead, for being technically challenging, when it comes to its application and optimisation, even though it is known to be applied in large scale. On the contrary, ILs seem very promising: they are efficient and can be biobased. However, this methodology is still very recent, and more research is necessary in order to make it economically feasible in large scale. For this reason, this process is recommended as a long-term investment, for it brings the opportunity of improving the sustainability of the bamboo value chain even further.

Following these considerations, the feasibility of using extracted lignin from bamboo residues as a binder were investigated. The presence of contaminants, namely its purity, the average length of the lignin chains and the distribution of these lengths are key factors for a good quality binder production. Soda lignin quality is not as high as that of other methods, but, as explained in the third chapter, this will probably have a limited effect on the mechanical properties of the biobased binder. Notwithstanding, further support for this statement should be provided by additional research. EOL and ILs lignin were found to show higher levels of purity compared to soda lignin. However, it was found that any of the extraction methods are suitable to create a decent lignin-based binder. Anyhow, it should be kept in mind that, being a biobased product, its purity level will never be as high as in petrol-based chemicals. Moreover, a certain degree of variation in lignin composition should be expected, given variables such as different plant age and different growing and environmental conditions. Nevertheless, the impact of these factors will be the same regardless of the type of resin used. Therefore, choosing from the mentioned types of binders will be driven by other considerations.

Once again, a ready-to-use solution, like the lignin LF resin, has proven to be effective but not as sustainable as other options. Lignin (alone) binders are completely biobased, but the lack of consistency across literature limits the authors to suggest it as a 'plug and play' option. LF resins and other lignin-biobased crosslinker resins can be, instead, seen as a middle ground. Ligning (alone) binders are more sustainable than the LF resins but less than lignin used alone. They are better known and have better mechanical properties as compared to lignin from bamboo residues alone but still less when compared to LF resins. Therefore, depending on the wishes of the commissioner, **it is recommended using lignin-formaldehyde resins for the production of a binder proven to be effective. However, for a long-term goal it is suggested to focus on lignin alone as an adhesive.** Table 1 can be consulted for an overview of the analysis on the three extraction methods in this report.

Process	Simpleness	Tailor made potential	Purity	Environmental/Economical potential	Large scale	Applicated to bamboo
Organosolv	2	2	2	3	2	2
Soda	1	3	3	2	1	1
ILs	3	1	1	1	3	3

Table 13: Overview of results analysis extraction methods (1 being high score, 3 low score)

It became clear that, by using lignin from bamboo residues, the production chain became more circular and waste was reduced. The report made evident that distinguishing the terms circularity and sustainability was also important. While investing on the creation of European bamboo plantations seems to improve the sustainability of the chain, using the residues and, hence, reducing the waste, benefits its circularity. This, in turn, might have a positive impact on the sustainability of this system. Using the lignin present in these residues to create a binder can, however, backfire when considering the overall sustainability of the value chain. This is so, since such processes are very novel when compared to the petrochemical industry and thus, are not yet optimised. The same applies to cost efficiency, which, although disregarded in most academic literature, is of great importance. Indeed, the market will not welcome a solution which is four to five times more expensive than the currently applied alternatives (Personal communication Dr. Karsten Brast, 2020). However, this does not mean that investment on these methods should be discouraged. It is through further investigation that they can be optimised and perhaps surpass the effectiveness of their petrochemical counterparts. Thereby, one recommendation should be to invest on the circularity of the production chain, for it contributes greatly to waste reduction and, ultimately, to oppose one of the five main threats to biodiversity loss, i.e. pollution (Tilman et al., 2017). Although on the short-term this might result into an overall less sustainable production, it is a necessary step to move away from the fossil fuel industry.

Finally, it is important to keep in mind the long list of stakeholders that are affected by changes in the bamboo value chain. To properly hold into account the social sustainability of the soda extraction method, the stakeholder analysis is a good starting point. Furthermore, in the appendix a long-list of stakeholders can be found

Unfortunately, literature and experts' knowledge do not, yet allow the authors to design a perfect solution containing a direct answer to this project's main research question. Indeed, lignin from European-based Moso bamboo residues can be used to form a biobased binder for laminated products. This, in long term and after further research is conducted, might result in an improvement of the value chain' sustainability. Having this said, the authors recommend that the available lignin types, i.e. EOL lignin, soda lignin and ILs lignin, should be tested for the following features:

- 1. preparation of a lignin (alone) based binder;
- 2. laminate bamboo using this resin;
- 3. mechanical properties and its resistance towards external conditions (e.g. the weather).

If a lignin (alone) resin is not suitable, investigation should be conducted for the other available polymers. They should be explored in the following order: lignin-furtural resins, other lignin biobased compounds, and lastly, LF resins. This is not very time-consuming, and it will provide the interested parties with necessary insight - which resins' type is best for them and which extraction method provides the better lignin for the adhesive production. This will allow to fill in the remaining knowledge gaps and help to achieve the wished goal of getting more circular and sustainable value chains.

SWOT Analysis of the recommendation

This SWOT-analysis will be focusing on the short-term recommendation to use soda as an extraction method to extract lignin from bamboo residues, as mentioned in the conclusion. In this analysis, the strengths, weaknesses, opportunities and threats of using soda as an extraction method for lignin from bamboo will be laid out, in order for ArtEZ FM and other parties to make balanced decisions on their course of action. At the end of the section, Figure 22 is presented which shows a graphical representation of the SWOT analysis.

Strengths

Well-established and straightforward method that is adaptable for Moso Bamboo

The soda method is a well-established method for lignin extraction, which is the first strength of the method. As mentioned earlier in the report, the process was already used in 1851. This provided the opportunity to gain a lot of knowledge about the process and thus being able to optimise it. Furthermore, the process is relatively simple which can be a great advantage as no very complex knowledge is needed to perform the process. The soda process has the strength of being easily adaptable. The process consists only of a few steps: heating and pressurising, filtration, separation and washing and drying. This implies that alterations, for example specifically for Moso Bamboo, are relatively easy to test and implement in order to get the results that are requested.

Good economic performance

The soda method is a relatively cheap method for delignification. The process does not require a lot of inputs. The most important input is the acid that is used. Most studies use sulfuric acid, because it produces high lignin yields and since it is one of the cheapest acids. Chapter 4 discussed the economic performance of the extraction methods and showed good prospects for the soda process. The report showed that the profitability index for the soda method is promising and the costs are relatively low. Furthermore, the NPV shows promising numbers, which can lead to profitability when used in large-scale applications.

Sustainable

As discussed in previous chapters of the report, the soda method is quite a sustainable approach, which is a great strength. Especially if the soda method is compared with other methods, it shows its sustainability potential.

Weaknesses

Technological development limitations

Even though there are several methodologies within the soda extraction method, the soda process finds itself limited in terms of the number of applications. Indeed, being a well-known and established process, without forgetting its simplicity, the soda process is a victim of its former success. Little alternatives exist for altering the process or make it any more specific towards lignin extraction from bamboo biomass. In other words, the soda extraction method has been very well developed over the years, but it does not entirely fit lignin extraction from bamboo biomass at this moment in time.

Besides the lignin extraction method, it is also worth mentioning the implications concerning the nature of the lignin itself. Generally speaking, natural products do not excel in specific tasks, since they are rather versatile. Lignin can therefore be used for a wide range of applications. Lignin binds and holds the plant cells together to make it more rigid. It also keeps water out and prevents degradation and rotting. Comparable substances like phenol formaldehyde are better at holding things together and making the product rigid, but it does not repel water, nor does it prevent rotting effectively.

Difficult recovery of substances

Some of the substances used during the extraction process, such as sodium hydroxide, are very watersoluble. In other words, it is difficult to recover these substances, which happen to be acids for the most part. This translates in a high acid consumption, that often leads to economic and environmental consequences. Other substances can be added to the process in order to recover the lignin extracting substances, but this increases costs and extends the demanded time to carry out the extraction procedure.

Compromise on chemical choices

Often with the soda process, a compromise must be reached when it comes to the choice of the solvents used during the lignin extraction. Indeed, even though most of them are economically feasible and have high lignin extractability, they can be aggressive towards the environment and potentially harmful to humans. Sulphuric acid, for instance, can allow interesting results of up to 90% lignin yield but is volatile. Other more friendly substances are available and result in a much lesser acidification, but present lower economic profitability. CO₂ for instance has a yield of 77%. Furthermore, catalysts, such as anthraquinone, can be employed to increase lignin extraction yields. However, these have proven to do so only by a small percentage, resulting in little or no economic benefits. Besides, it presents other drawbacks, like its potential to be carcinogenic.

Opportunities

Governmental drive towards sustainability

One opportunity for soda as an extraction method is the drive of national governments to move away from the world's overdependency on non-renewable resources, like fossil fuel. As mentioned in this report, the European Union, for example, is creating more tightening regulations for using fossil fuels. This would impact petrol-based binders negatively, which means that the focus could be put more on the possibilities that a lignin-based binder extracted with the soda method could do. Furthermore, this would implore more public research funds to go to the further development and modernization of the soda method. This governmental drive towards sustainability is also beneficial for European-based bamboo, due to the big demand of bamboo in the EU and the great environmental costs associated with importing bamboo from other continents.

Market potential

The market potential for bamboo products from a circular bamboo value chain is rising, as mentioned in the report. There is a growing demand for products from a sustainable and circular bamboo value chain. Using soda to extract lignin from bamboo residues and making it into a binder for bamboo products is in line with these customer's wishes. As discussed, a growing segment of circular-aware and sustainable-aware customers is on the rise in the EU. Therefore, there is a great market potential for swaying these customers for bamboo products that come from a circular bamboo value chain. Furthermore, if the market for European-based bamboo increases, the knowledge on using soda as an extraction method for lignin from bamboo residues will also increase due to investments.

(Social) media

To make the 'new' generation enthusiastic about bamboo products is vital to create a long-lasting impact on the European market. Opportunities arise with this in new social media like 'TikTok', (mentioned in the stakeholder analysis) where already tons of accounts exist promoting bamboo as a plant and product. These videos oftentimes get millions, if not tens of millions, of views. To jump into this trend could mean an increase into the demand of European-based bamboo products and would teach millions of young Europeans the importance of a circular and sustainable European-based bamboo value chain, and what place the soda extraction method has within this.

Threats

Petroleum competition

The petroleum industry has been established for many years and during this time the processes have been optimised. Because of this, the cost of producing petroleum-based binders is very low. New products, or new developments, are rarely optimised. This makes the process less efficient and more expensive, which makes it hard for new products to compete economically. Besides the process cost, it is also not uncommon for newer biobased products to be more environmentally harmful. This might sound contradictory, but this is due to lower efficiency. Everything outside of the product is oftentimes still mostly powered by fossil fuels, so the less efficient process will usually do worse since it needs to process more raw materials in order to produce the same amount of product. This will use more electricity and heat as well are more transportation.

Knowledge gap

There is a certain lack of information in literature concerning the binding properties of bamboo lignin specifically. Because of this, relatable lignin biomass was used for this research. Therefore, lignin from bamboo might have particularities that would result in different results than expected. If the lignin from bamboo is significantly different, the extraction methods might not work as effectively. These extraction methods work by breaking the lignin free from the rest of the biomass, but if the lignin in bamboo is bound differently, other methods might be required. The same problem applies to the binder part. The binder works by binding lignin together again at specific places in the molecule. If the regions where the crosslinker binds with the lignin are less abundant in the structure, the final binder might be significantly weaker than when wood lignin was used.

Optimisation

The aforementioned threats can be overcome by researching and optimising the process. While it might improve the production and it also might make it more economically and environmentally feasible than the established binders, it also might not. Doing research into new products as well as optimising the production of these products cost money. While it can be estimated whether or not a process can be profitable, this is never certain when the project is starting. It might cost a lot of money, with the only result being in the end that the process cannot be as profitable as the established alternatives.



Figure 22: SWOT-analysis

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Appendixes

Appendix 1: Stakeholder longlist

1. Governmental institutions

- a) Local: Municipality of Alcoutim
- b) National: Portuguese Government, ICNF
- c) Transnational: European Union

2. Media

- a) Offline: Portuguese newspapers like 'Correio da Manha' and 'Expresso'
- b) Online: Online global newspapers such as BBC and New York Times. Youtube channels such as 'Vice' and other documentary making Youtube Channels. Online media such as Twitter, Facebook, Linkedin and Tiktok.

3. Research institutes

- a) Public research institutes: Circular Fashion Lab WUR and other University Research departments
- b) Private research institutes: Stichting Hout Research, Kiemt, GIST

4. European bamboo producers

- a) Bamboologic's plantations
- b) Moso Europe
- c) Private farmers transitioning to bamboo plantations

5. Local Portuguese inhabitants

- a) ADPM: local non-governmental organization
- 6. Suppliers
 - a) Trucking companies
 - b) Shipping companies

7. Non-portuguese bamboo farmers

- a) Farmers in low-income countries previously farming in bamboo and exporting to EU
- b) Local NGOs in low-income countries in regions where bamboo is cultivated

8. Customers

- a) Fashion and design labs: ArtEZ FM
- b) Private companies: MOSO, Wilhelmina winkel, Bamboe informatie centrum
- 9. Consumers
 - a) Circular-oriented consumers that want to purchase bamboo products that have a closed value chain
 - b) Sustainable-oriented consumers that want to purchase bamboo products that are the most socially and environmentally sustainable



Appendix 2: Comparison of organosolv and soda extraction for all impact categories.



Appendix 3: comparison of organosolv and soda extraction per impact category



























