THESIS FOR THE DEGREE OF LICENTIATE OF ENGINEERING

ASSESSING THE FUTURE ENVIRONMENTAL IMPACT OF LIGNIN-BASED AND RECYCLED CARBON FIBRES IN COMPOSITES USING LIFE CYCLE ASSESSMENT

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ABSTRACT

Carbon fibre reinforced polymers (CFRPs) are composite materials that are gaining attention for their lightweighting and strengthening properties in a wide range of applications. However, using them instead of conventional materials (such as steel or other composites) does not automatically lead to a decrease in life cycle climate impact or energy use. This is the result of the energy-intensive production of the carbon fibres. Two routes that could mitigate this problem are: 1) the use of lignin for carbon fibre production and 2) the use of recycled carbon fibres. This thesis assesses how these two routes could decrease the environmental impact of carbon fibres in composites, and how challenges connected to assessing these emerging technologies can be handled using life cycle assessment (LCA). The two routes were assessed by conducting a meta-analysis of earlier LCAs of CFRPs and lignin production and three different LCA case studies. Results show that both using lignin as a raw material and using recycled carbon fibres have good potential to decrease the environmental impact of CFRPs, making them more environmentally competitive than other materials. It was found that the transition from polyacrylonitrile (PAN) to lignin as a raw material has good potential to decrease the environmental impact of future carbon fibres. However, the extent of this potential depends on both internal factors, such as process development, and external factors, such as the development of the lignin market and the future energy supply system.

Keywords: LCA, carbon fibres, lignin, recycling, bio based, prospective, environmental assessment

LIST OF PUBLICATIONS

This thesis is primarily based on the work described in the following two papers:

PAPER I
Hermansson, F., Janssen, M., & Svanström, M. (2019). Prospective study of lignin-based and recycled carbon fibers in composites through meta-analysis of life cycle assessments. Journal of Cleaner Production, 223, 946-956. doi:10.1016/j.jclepro.2019.03.022

PAPER II
Hermansson, F., Janssen, M., & Svanström, M. (2020). Allocation in life cycle assessment of lignin. The International Journal of Life Cycle Assessment. doi:10.1007/s11367-020-01770-4

Other related publications:

PUBLICATION A
Hermansson, F., Janssen, M., & Gellerstedt, F. (2016). Environmental evaluation of Durapulp bio-composite using LCA: Comparison of two applications. J-FOR-Journal of science & technology for forest products and processes, 5(2), 68-76.

PUBLICATION B
Hermansson, F., Janssen, M., & Svanström, M. (2018, 5-7 March). Recommendations for Routes to Sustainable Exploitation of CFRP Materials. Presentation presented at the ECO-BIO 18, Dublin, Ireland. https://research.chalmers.se/publication/501178.

PUBLICATION C
Hermansson, F., Janssen, M., & Svanström, M. (2018, 23-26 September). Life cycle assessment of lignin-based carbon fibers. Poster presented at the SETAC Europe 24:th LCA Symposium, Vienna, Austria. https://research.chalmers.se/publication/505183.

PUBLICATION D
Hermansson, F., Janssen, M., & Svanström, M. (2019, 7-11 July). Environmental challenges and opportunities of lignin. Presentation presented at the 10th International Conference on Industrial Ecology, Beijing, China. https://research.chalmers.se/publication/511316.

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1. INTRODUCTION

Carbon fibre reinforced polymers (CFRPs) are composite materials that are gaining attention in a wide range of applications, primarily due to their strengthening and lightweighting properties. The lightweighting properties can lower the energy consumption in a vehicle's use phase and, as a consequence, the related environmental impacts connected to fuel use (Duflou et al., 2012). When used for reinforcements, such as reinforcing beams in a building, the motivation for using CFRPs is often to prolong the lifetime of a structure and to avoid replacing structural components. While the intentions for using CFRPs instead of conventional materials, such as metals and other types of composites, are often to either decrease energy use and related emissions during use, or to prolong the applications' lifetime to avoid producing new components, a change of materials does not always lead to an overall reduction in environmental impact. This is primarily a consequence of the very energy intensive carbon fibre production process (Das, 2011). In light of this, some measures must be taken to increase the CFRPs´ environmental competitiveness. To achieve this, the literature suggests two main routes: 1) the use of lignin as a raw material for carbon fibre production (see e.g. Das (2011)), and 2) using recycled carbon fibres (see e.g. Meng et al. (2017)). Both of these technology routes are, however, still in the early stages of their process and market development, and production data are not yet available. This thesis aims to assess the potential future environmental impacts of carbon fibres in composites with a special focus on lignin-based and recycled carbon fibres. The purpose is to contribute both to the technical development of the two technology routes and to the methodological development of assessing the future environmental impact of such emerging technologies using life cycle assessment (LCA). The assessment was done by a combination of a meta-analysis of earlier LCA findings and explorative scenarios to assess how the future impact of carbon fibres is influenced by internal factors, such as the consequences of production process development, and external factors, such as changes of the energy system and in the demand for lignin, applied in a prospective LCA.

2. BACKGROUND

2.1 The LIBRE project

The research was conducted within the project LIBRE: Lignin-based carbon fibres for composites (2016-2021). The project received funding from the Bio-Based Industries Joint Undertaking under the European Union's Horizon 2020 research and innovation programme under grant agreement No 720707. The work described in this thesis was performed within a work package that dealt with the environmental and economic performance of lignin-based carbon fibres. The intention was to guide material developers in the project consortium to identify routes for decreasing the environmental impact of lignin-based carbon fibres. As there is currently no industrial-scale production of lignin-based carbon fibres, a future-oriented assessment approach using different explorative scenarios in combination with close collaboration with the material and technology developers within the project was needed. The focus of the collaboration was on collecting data and information for the LCA and delivering results that were helpful for guiding the material developers in efforts to minimize the environmental impact of LIBRE carbon fibres.

2.2 Carbon fibres and CFRP production

Carbon fibres are most often made from polyacrylonitrile (PAN), which is a fossil-based polymer. This polymer is spun into a precursor fibre before being processed into a carbon fibre. This is done in a series of steps including thermosetting in an oxidizing environment, carbonization in an inert nitrogen atmosphere, and lastly, surface treatment. When producing a composite, the carbon fibres are arranged in a way specific for each application, and a polymer matrix material and any additives necessary to obtain the desired material properties are added. The composite is formed into the shape needed for the intended application, e.g. by sheet moulding (Das, 2011). At the end of its use phase, the composite is most often sent to landfill or incineration (possibly with energy recovery), but it can also be recycled. Recycling can be achieved by either reusing the whole composite by reshaping it (see e.g. Suzuki and Takahashi (2005)) or by separating the fibres from the matrix material to reuse them (see e.g. the work by La Rosa et al. (2016), La Rosa et al. (2018) and Dong et al. (2018)). Figure 1 outlines the main parts of the carbon fibre composite life cycle.

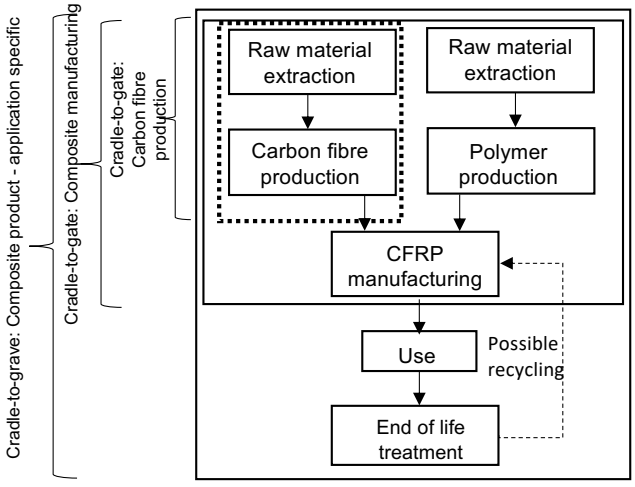

Figure 1: Outline of the carbon fibre composite life cycle

2.3 Production of lignin-based carbon fibres

Lignin has been suggested as a possible alternative raw material to PAN for carbon fibre production (see e.g. Sudo and Shimizu (1992), Kadla et al. (2002) and the LIBRE (2016) and GreenLight (2016) projects). Lignin is found in most terrestrial plants and is responsible for 15-40 % of their dry weight (Ragauskas et al., 2014). Lignin can be made available from the process streams of pulp mills, e.g. from the black liquor of a Kraft pulp mill (Culbertson et al., 2016), and from biorefineries, e.g. ethanol plants (Modahl et al., 2015), once the different wood components, i.e. cellulose, lignin, hemicelluloses, and extractives (Henriksson et al., 2004), have been separated from each other. Lignin must be purified, dried, and sometimes blended with a polymer when it is to be used for carbon fibres (Das, 2011). Blending with a polymer can reduce brittleness of the lignin-based carbon fibre as well as improve the thermoplastic behaviour of lignin (Collins et al., 2019). In contrast to the PAN precursor fibre, which is spun using wet-spinning, the lignin precursor fibre is spun using melt-spinning, which unlike wet-spinning does not require any solvents (Das, 2011). The subsequent steps, i.e. turning the precursor fibre into carbon fibre and the composite manufacturing, would typically be the same for lignin-based carbon fibres and PAN-based carbon fibres. For an in-depth description of the processing of lignin-based carbon fibres, see Collins et al. (2019).

2.4 Market development of carbon fibres and CFRPs

The modern era of carbon fibre production started in the mid 1950s. The development was supported by the U.S. Air Force Materials Laboratory in an effort to develop high-strength composites for, e.g., air craft structures (American Chemical Society, 2003). Generally, the life cycle cost of CFRPs is higher compared to other materials, such as steel and glass fibre reinforced polymers (GFRPs), which is due to the cost of raw materials, but also because of the composite manufacturing processes (Witik, 2011). The production cost can be reduced by choosing a cheaper raw material than PAN for the fibre production, such as lignin (Fang et al., 2017), or to use recycled carbon fibres instead of primary fibres, given that the recovered fibres are of high enough quality and that the fibre recycling capacity and recovery rate is high enough (Dong et al., 2018). The higher price of high-performance carbon fibres compared to other materials has kept production and use to a minimum for a long time; however, efforts are being made to reduce production cost (Gorss, 2003), for example by implementing the suggestions listed above. The successive lower cost of CFRPs has resulted in that the use of carbon fibres in recent years has expanded to a wider range of applications in which they replace conventional materials, such as metals or GFRPs in, e.g. vehicles, structures, and sports equipment (see e.g. Witik (2011), Maxineasa et al. (2015) and Subic and Paterson (2006) respectively). The reason for this substitution is primarily due to the lighter weight of carbon fibres compared to most metals and, to some extent, GFRPs (Witik, 2011). In addition to a slightly lower density, CFRPs generally have a higher specific strength and impact resistance than GFRPs (Liu et al., 2013), as well as withstanding erosion better (Tewari et al., 2003). As the application area recently began to expand and disperse into a growing range of applications, CFRPs can still be seen as an emerging material even if they have been available for some for 60+ years. As a consequence, the increase in application areas and demand for carbon fibres and CFRPs can be expected to continue to grow. For example, Sauer and Kuhnel (2017) write that the theoretical mid-to-long-term growth of carbon fibre production capacity is 29% higher than the production capacity in 2019. In addition to this, they state that the CFRP market can be expected to grow by 39% from 2019 to 2023.

Predicting future market development is a difficult task connected with large uncertainties. However, there are strong indications that the production of carbon fibres and carbon fibre composites is expanding rapidly. Further, PAN-based carbon fibres dominate the world market today, (Gorss, 2003) but with growing concerns about the environment and the depletion of oil resources, efforts are being made to find new means of producing high-performance carbon fibres with lower environmental impact. As mentioned in the introduction, two different routes to address these problems (as well as lowering the cost of CFRPs) are lignin-based carbon fibres and using recycled carbon fibres in composites. As the demand for carbon fibres is expected to grow, it can be assumed that the market for these new types of carbon fibres will grow as well, possibly taking over a part, or in a very long-term future, the majority share of the primary PAN-based carbon fibre market.

While using lignin as a raw material for carbon fibre production and the use of recycled carbon fibres have been suggested as possible routes for decreasing the environmental impact of CFRPs it is not fully known to what extent such transition would in fact reduce their environmental impact. One method that can be used to assess a material's environmental impact is life cycle assessment (LCA) (described in Section 2.5). However, literature on LCAs of lignin-based and recycled based carbon fibres is scarce. Consequently, there are still areas that must be examined further, especially considering that these are materials that by no means is mature yet. In order to assess how and if using lignin as a raw material in carbon fibre production and the use of recycled carbon fibres will reduce the environmental impact of CFRPs, more efforts must be put into assessing which phases in the carbon fibre product life cycle are environmental hotspots (i.e. intensive in terms of environmental impact) and how a transition to lignin-based and recycled carbon fibres could influence this, as well as what challenges will be faced when assessing the impact of carbon fibre products using LCA.

2.5 Life cycle assessment

Life cycle assessment (LCA) was developed to assess potential environmental impacts related to the life cycle of products and services. LCA can be used to identify opportunities for improving the environmental performance of products and as an analysis tool for decision support. There are four main stages in an LCA study: goal and scope definition, inventory analysis, life cycle impact assessment, and interpretation (International Organization of Standardization, 2006). The LCA procedure is outlined below, as described by Baumann and Tillman (2004), and visualized in Figure 2.

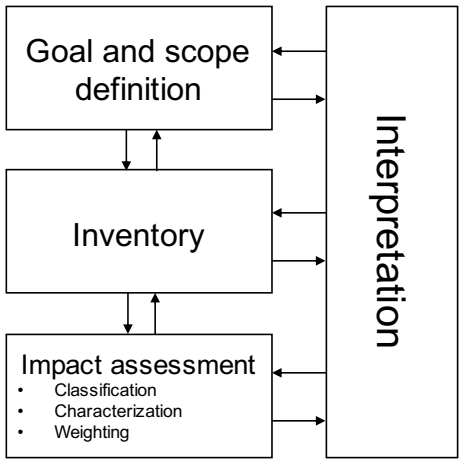

Figure 2: The LCA procedure as described by Baumann and Tillman (2004)

The first step of an LCA is **the goal and scope definition.** This phase describes the purpose of the study, the intended application, and its modelling aspects. The goal of an LCA clarifies the reason for carrying out the study and the intended use of the results. The scope includes modelling aspects, such as the choice of functional unit, which reflects the function of the product or service being assessed, and the scope includes describing the technical system with flowcharts, system boundaries (e.g. geographical, technical, or temporal), allocation procedures, and which environmental impacts to consider (e.g. acidification or climate impact).

An **inventory analysis** follows the goal and scope of an LCA. An inventory analysis inventories the inputs to the system, such as raw material or energy used, and the outputs from the system, such as emissions, wastes, and products.

The third step is the **impact assessment**. This step includes classification, i.e. what substance in the inventory causes what environmental impact, and characterization, i.e. each substance's relative contribution to the environmental impact of the product. The impact assessment employs different characterization factors as defined in the impact assessment method for each of the inputs and outputs. In some contexts, the resulting environmental impacts are aggregated into a single number that includes several impact categories by using a weighting procedure.

Throughout the assessment, the findings are **interpreted,** and the goal and scope, inventory, and impact assessment can be changed. This means that an LCA can be quite iterative.

As mentioned in Section 1, the literature suggests that the use of lignin as a raw material for carbon fibre production and using recycled carbon fibres are two possible routes to decrease the environmental impact of CFRPs. As these two routes are still rather unexplored and by no means mature yet there is a problem with data availability. In fact, one of the major challenges in assessing emerging materials using LCA is the lack of data. The trade-off between the lack of data when there are still possibilites for alterations and enough data when alterations typically are no longer possible is referred to as the Collingridge dilemma (Collingridge, 1980). In short, this means that while the lack of data is problematic for an assessment, the early phase of material development also poses an opportunity to influence decision makers and process developers to improve the environmental performance of a product. To assess emerging materials, such as lignin-based and recycled carbon fibres, using LCA, a future-oriented approach is therefor needed. Such an approach is further described in Section 2.5.1.

2.5.1 Assessing emerging materials using LCA

As mentioned in Section 2.5, assessing emerging materials in LCA requires a future-oriented approach. While future-oriented LCAs cannot predict the future, they can help explore it by assessing different scenarios for technology and systems development (Cucurachi et al., 2018). Villares et al. (2017) state that the outcomes of a future-oriented assessment should not be seen as final results but rather as a contribution to technology development, raising strategic questions, introducing systems perspectives, and environmental concepts in the beginning of technology development. Similarly, Buyle et al. (2019) write that future-oriented LCAs have the potential to influence technology from the start and to guide efforts to minimize the environmental impact of a material.

One type of future-oriented LCAs is prospective LCAs. Arvidsson et al. (2018) defines on p. 1287 prospective LCAs as "studies of emerging technologies in early development stages, when there are still opportunities to use environmental guidance for major alterations". In line with this definition, the assessments done in this thesis have been classified as prospective as they assess both a material with a market that recently began to grow significantly (i.e. the PAN-based carbon fibre), possible future technical systems (i.e. the production of lignin-based carbon fibres and the use of recycled carbon fibres) and because there are still opportunities for major alterations in the production processes for lignin-based carbon fibres and in the recycling of carbon fibres. Life cycle assessments can either be attributional or consequential (i.e. change-oriented), where the attributional assessments focus on describing the environmental impact from direct physical flows and consequential assessments focuses on how decisions change environmentally relevant flows (see e.g. Curran et al. (2005) and Finnveden et al. (2009)). The assessments done in this thesis have been conducted using an attributional methodology, focusing on the direct environmental impact resulting from physical flows to and from the carbon fibre production system, using average and not marginal data. It should be acknowledged that the prospective assessments in this thesis include some consequential features in that those assessments examine changes made both to the production system and to the surrounding system, which will change environmentally relevant flows. However, as the assessments in this thesis focuses on exploring and describing the direct environmental impact from the different elements of the technology based on possible future developments, rather than describing how flows will

change in response to any possible decisions being made, in combination with the large uncertainties in the future surrounding systems, the assessments done follow the attributional methodology.

A way to assess the future life cycle environmental impact of a material is by examining different scenarios (Pesonen et al., 2000). Börjeson et al. (2006) identifies three main types of scenario categories that are based on the type of question about the future that the user wants to answer: 1) What will happen? 2) What can happen? and 3) How can a specific target be reached? They write that Question 1 is answered with predictive scenarios that often have short timeframes and focus on external factors. Question 2 is often answered with explorative scenarios that generally have a longer timeframe and focus on external or internal factors, where internal factors are under the influence of external ones. Finally, Question 3 is answered with normative scenarios that often have a long timeframe and focus on both external or internal factors. This thesis aims to answer the question "what can happen?" in terms of changes to both internal factors in the technology system, such as process development, and external factors in the technology system such as market development with a long time frame. Therefor the scenarios used in the assessments in this thesis have been classified as explorative.

Material developers within the LIBRE project sought guidance to improve the environmental impact of future lignin-based carbon fibres. As described above, this was done in the early stages of material development when process-specific and/or large-scale data are still missing, but there is still an opportunity for environmental guidance. In the assessments, we focused on introducing the systems perspective, identifying environmental hotspots, and assessing possible future scenarios in terms of both internal factors related to technology development (i.e. the carbon fibre production process) and external factors related to world development (e.g. the energy system). In addition to this, the project was also interested in assessing if and how recycled carbon fibres could be an option for decreasing the environmental impact of CFRPs. While not included in the material development activities in the project, we included it in the to provide a systems perspective to this choice as an alternative or complement to the lignin-based carbon fibres

2.5.2 Distributing the burdens

Lignin is always the product of a multi-output process. According to the guidelines stipulated by ISO 14044:2006 (provided by the International Organization of Standardization (2006)) and the International Reference Life Cycle Data System (ILCD) (provided by the European Commission Joint Research Center (2010)), any distribution of environmental burden among co-products and a multi-output process should be avoided if possible. This can be done by means of subdivision (i.e. increasing the level of detail in the modelling) or system expansion in which the system is expanded to include the production of all functions of the multi-output system. The ILCD guidelines suggest an alternative to this, where the system is expanded to include the functions replaced by the co-products, thus giving the co-products a negative impact. If these approaches are not applicable or feasible, the environmental burden of the system may need to be distributed among the different products. This is referred to as allocation in LCA. Both ISO 14044:2006 and ILCD guidelines state that if allocation cannot be avoided, the inputs and outputs of the system should be allocated among the products based on physical relationships between them (e.g. mass or energy). If this is not possible, the inputs and outputs should be allocated in a way that reflects other relationships, e.g. the economic value of the products.

When evaluating the environmental impacts of recycling using LCA, allocation is also something that needs to be considered if it cannot be avoided, in order to distribute the impacts of both product production and recycling processes between the primary and recycled products. The procedure for allocating in recycling follows the same basic guidelines as for allocation in multi-output processes described in this section, but is however not addressed further in this thesis.

3. AIM OF THESIS AND RESEARCH QUESTIONS

The overall aim of this thesis is to assess the possible future environmental impacts of carbon fibres in composites with a special focus on the transition from primary PAN-based to lignin-based and recycled carbon fibres. To fulfil this aim, two main research questions and a set of sub-questions were addressed:

1. What is the life cycle environmental impact of carbon fibres in composites?
    a. How would a transition to lignin-based carbon fibres or recycled carbon fibres change the environmental impact of carbon fibres in composites?
    b. Which are the environmental hotspots in the life cycle of lignin-based carbon fibres, now and in the future, and is lignin an important contributor to these hotspots?
2. How can challenges associated with assessing the life cycle environmental impact of lignin-based and recycled carbon fibres be addressed?
    a. How can the lack of data for these emerging technologies be dealt with in LCA?
    b. How can allocation be dealt with for lignin production, and does the choice of allocation approach have a major influence on the environmental impact of lignin?

4. METHODOLOGY

The core methodology applied in this thesis was: 1) screening of LCA literature for useful methods and relevant data (such as LCA results) that could be directly used or adapted, and 2) the subsequent application of the data mined from literature in new contexts in combination with using primary data gathered within the LIBRE project. Finally, in order to test the usefulness of the data and methods and to generate results to answer the research questions, 3) three different case studies were performed: i) one that examined how environmental impact changes when lignin is used as a raw material for carbon fibres instead of PAN or when fibres are recycled, ii) one that assessed the climate impact of 1 kg of lignin-based carbon fibres produced today and in a long-term future to identify hotspots and explore the effect of possible improvement opportunities and changes in the surrounding system, and iii) one that examined how the allocation approach applied to lignin production influences the environmental impact of lignin, now and in a long-term future.

This chapter is divided into two sections: Section 4.1 describes how data and methods from the literature were selected, and if necessary adapted, further developed, and sometimes even complemented with new data or approaches for the application in new contexts. Section 4.2 contains a description of the three case studies.

4.1 Mining from LCA literature and adapting for new contexts

Information was extracted from the LCA literature for two main purposes: 1) to find LCA assessments of the transition to lignin-based or recycled carbon fibres, and 2) to find allocation approaches that could be used in the context of assessing the environmental impact of lignin from a multi-output process. The information, i.e. LCA results and allocation methods for these two purposes, was collected using Google Scholar in combination with Scopus and Summon (Chalmers Library, 2010). While the data collection method can be grouped into literature reviews, it was neither exhaustive nor systematic as this was deemed unnecessary for the purpose of the studies. The intention was not to generate a full account of earlier work but to extract a sufficient amount of information. The process was highly iterative; when new areas of interest were found or when a gap in knowledge was identified, the scope of the initial study was expanded to include those areas. Once the information was collected, it was either

recalculated to be assembled into new contexts or adapted to better fit the system under study.

The process of collecting information to be further recalculated and assembled into the context of a transition to lignin-based and recycled carbon fibres is described in Section 4.1.1. The collection, adaptation, and development of allocation approaches for lignin production are described in Section 4.1.2. Additional data for the case studies described in Section 4.2 were also collected from literature using standard approaches. Sources are provided for each study in Section 4.2, in Papers I-II, and in Publications A-D.

4.1.1 Recalculating, adapting, and assembling LCA data in new contexts

The data for assessing possible changes in energy use and climate impact when transitioning from primary PAN-based carbon fibres to lignin-based carbon fibres or recycled carbon fibres were collected between June 2017 and April 2018. The methodology for this (developed in an iterative process) is seen as a methodological contribution to this field and is described in detail in Paper I and briefly in this section. The methodology is also briefly summarized in Section 5 Results and Discussion in relation to each research question that it addresses and is further discussed in Section 5.2.1.

Initially, only cradle-to-gate LCAs of the production of lignin-based carbon fibres were sought, but as only one such study was found (a study by Das (2011)), the scope of the search was expanded to include fossil-based carbon fibre production as well as the application of carbon fibres in CFRPs. As the literature suggested that the recycling of carbon fibres from composites was a possible route for decreasing the environmental impact of CFRPs, the recycling of carbon fibres and of carbon fibres in composites was also included in the search. The scope was, therefore, expanded from cradle-to-gate to cradle-to-grave. Studies presenting overly aggregated results (e.g. when the impact of the CFRP material could not be separated from other parts of the life cycle) were directly excluded. As only one study on lignin-based carbon fibres was found, more data were needed to allow for identifying trends, hotspots, possibilities, and challenges related to the use of lignin as a raw material. Therefore, the search was expanded to include LCAs of systems where lignin is an output product. Studies where lignin was not

reported as a quantified and separate output were directly excluded from further assessment.

To identify which LCA results from the studies collected should be used in the further analysis, and how the results should be dealt with, the studies were evaluated based on: 1) functional units, system boundaries, and life cycle impact assessment categories; and 2) how the results were presented in relation to different life cycle phases. One of the purposes of this step was to identify which impact categories and life cycle phases had sufficient information overlaps across studies to allow for the identification of trends or hotspots. While LCA studies are seldom directly comparable because of differences in system boundaries, functional units, and impact assessment methods, an attempt was made to normalize results on a common basis in order to identify overall patterns and the orders of magnitudes of impacts. This means that when needed, the results reported in different studies were recalculated for one common functional unit. Only when comparing the environmental impact of a CFRP to the impact of a conventional material in a specific application, were the original functional units kept. The greatest impact category overlap between the studies was for climate impact and energy use. As a consequence, only studies considering these impact categories were selected for further assessment. The results were also categorized and sorted into modules based on system boundaries. In this way, the findings were divided into five different categories: cradle-to-grave CFRP (i.e. carbon fibre production, composite production, use phase, and end of life), cradle-to-gate CFRP (i.e. carbon fibre and composite production), only carbon fibre production, only lignin production, and only end-of-life treatment (including recycling).

The resulting modules were then assembled in a case study (i.e. Case study 1, further described in Section 4.2.1) to illustrate the possible changes in energy use and climate impact that a transition from primary PAN-based to lignin-based carbon fibres, or to recycled carbon fibres, could lead to.

4.1.2 Collecting, adapting, and developing allocation approaches

During the search for LCA results for lignin production, as described in Section 4.1.1, the allocation approaches used in the studies were also collected and listed, as these were suspected to potentially have a major effect on the life cycle environmental impact of lignin. As the allocation methods were eventually applied in a case study (see Section 4.2.3), their applicability and usefulness are commented on in the discussion (see Section 5.2.2). To complement the allocation approaches and methods found during the search for lignin generating processes, an additional search for allocation approaches used in biorefinery systems was conducted. This search was done in a manner similar to what was described in Section 4.1 and Section 4.1.1, i.e., neither exhaustively nor systematically but iteratively. The search was ended when a sufficient number and range of allocation approaches had been found.

Ten allocation approaches and methods deemed appropriate for a lignin-generating system were found in the literature. When necessary, they were adapted to the specific model system and context that was selected for the case study in terms of for example reference products. Two new allocation methods were also developed in addition to the ten found in the literature, and these were designated *changes made to the mill* and *mass- and energy-based allocation*. As these two allocation approaches were the results of methodological development work done in the context of this thesis, they are also described as results in Section 5.2.2. Table 1 includes a short description of all twelve allocation methods, collected or developed, and the order of presentation roughly follows the hierarchy postulated by relevant guideline documents (as described in Section 2.5). For an in-depth description of each approach, see Paper II.

Table 1: Allocation procedures used for assessing the climate impact of lignin production (Hermansson et al., 2020). The table continues on the next page.

Method	Approach	Reference
Changes made to the mill	Subdivision is simulated in a pragmatic way. Considers only the impact of the lignin extraction process added and any internal energy loss related to lignin removed.	Proposed by the authors of Paper II
Marginal approach	Considers the difference in impacts of the whole system before and after lignin extraction.	Bernier et al. (2013)
Main product bears all burden	A main product of the system is selected to carry the entire environmental burden.	Sandin et al. (2015)
System expansion by substitution	The system boundaries are expanded to include the replacement of other products on the market.	European Commission Joint Research Center (2010)
Mass-based allocation	The impacts of the system are partitioned based on the mass of each co-product flow.	International Organization of Standardization (2006) and European Commission Joint Research Center (2010)
Energy-based allocation	The impacts of the system are partitioned based on the energy content of each co-product flow.	International Organization of Standardization (2006) and European Commission Joint Research Center (2010)
Exergy-based allocation	The impacts of the system are partitioned based on the exergy content of each co-product flow.	Cherubini et al. (2011)
Energy- and mass-based allocation	The impacts of the system are first partitioned between the energy streams and the mass streams based on energy efficiency, followed by either energy allocation (for energy streams) or mass allocation (for material streams).	Njakou Djomo et al. (2017)

Mass- and energy-based allocation	The impacts of the system are first partitioned between the energy streams and the mass streams based on the mass conversion rate, followed by either energy allocation (for energy streams) or mass allocation (for material streams).	Proposed by the authors of Paper II
Economic allocation	The impacts of the system are partitioned based on the economic value of each co-product flow.	International Organization of Standardization (2006) and European Commission Joint Research Center (2010)
Allocation based on substituted impacts	The impacts of the system are partitioned based on the impacts of replaced products.	Cherubini et al. (2011)
Allocation based on inversed substituted impacts	The impacts of the system are partitioned based on the inversed impacts of replaced products.	Sandin et al. (2015)

The twelve allocation methods were tested for applicability and were used to illustrate the span of results when employing different approaches and timeframes in a case study on lignin production, see Section 4.2.3.

4.2 Case studies

Three case studies were performed within the scope of this thesis: Section 4.2.1 describes Case study 1, which assessed the changes in environmental impact when lignin was used as a raw material for carbon fibres instead of PAN, or when fibres were recycled. Section 4.2.2 describes Case study 2, which assessed the environmental impact of the production of 1 kg of lignin-based carbon fibres and potential improvement opportunities, and Section 4.2.3 describes Case study 3, which assessed the production of lignin using different allocation methods. The order of presentation reflects the chronology of the studies being done. As they were part of research conducted both to provide assessment results and to improve assessment methodology, the case studies built on work that had been conducted in previous studies (see Papers I-II and Publications A-D).

4.2.1 Case study 1: Assessing the transition to lignin-based or recycled carbon fibres

The goal of this case study was to assess how the transition from primary PAN-based carbon fibres to lignin-based or recycled carbon fibres could influence the environmental impact of CFRPs. This was done to obtain input to the material development within the LIBRE project in early stages of the project. Data availability only allowed for the consideration of energy use and climate impact. The functional units, system boundaries, and allocation methods used in the different parts of the case study are shown in Table 2.

Table 2: The functional units, system boundaries and allocation methods used in Case study 1

	Functional unit	System boundaries	Allocation methods
Carbon fibre reinforced polymer (CFRP)	1 kg CFRP	cradle-to-gate or cradle-to-grave	n/a
Carbon fibre production	1 kg carbon fibre	cradle-to-gate	n/a
Recycling of carbon fibres	1 kg recycled carbon fibre	grave-to-gate[1]	n/a
Lignin production	2 kg of lignin[2]	cradle-to-gate	Economic allocation assuming price of 0.3€/kg, mass allocation and marginal approach

The data used for this study were collected as described in Section 4.1.1, and the results are presented in Section 5.1.1; both data and results are described in more detail in Paper I.

4.2.2 Case study 2: Assessing the climate impact of lignin-based carbon fibres

The goal of this case study was to identify environmental hotspots in the production of lignin-based carbon fibres as well as to assess how future developments could influence the fibres´ future impact by means of a prospective LCA. Production was assumed to take place in Europe both in present time and in a future with a different energy system

[1] i.e. only the recycling process
[2] i.e. the theoretical amount needed to produce 1 kg of pure lignin-based carbon fibres based on the assumption that there is a 50% material loss in the carbonization and stabilization phase and that other material losses are insignificant

to account for changes in external factors. The assessment also explored different technology development opportunities to account for changes in internal factors. This case study only assessed climate impact. This was primarily due to data availability, but as climate impact often correlates well with other impact categories (Janssen et al., 2016) this was deemed sufficient for fulfilling the aim of this explorative case study.

The functional unit of the study was 1 kg of lignin-based carbon fibres. The carbon fibres were assumed to be made from 50% Kraft pulp lignin and 50% bio-thermopolyurethane (bio-PU). The allocation method used in the lignin production was economic allocation, assuming a price of 0.3 €/kg lignin (González-García et al., 2016). The dataset for bio-PU was constructed by taking the dataset for polyurethane rigid foam and replacing the dataset for the fossil-based polyols with a dataset for rapeseed-based polyols for polyurethane production by Fridrihsone-Girone (2015). The production of lignin was assumed to take place in Sweden, and the production of polymer and carbon fibres was assumed to take place in Germany. Transportation was excluded due to uncertainties and its expected negligible contribution to the overall climate impact, unless it was already part of background datasets.

Five different explorative scenarios were generated to assess the future climate impact of the fibres. These scenarios included three scenarios related to technology development dependent on internal factors and two scenarios related to technology development dependent on external factors. The scenarios were selected after screening the literature in the meta-analysis and after discussions within the LIBRE project. Based on the assumption that the original carbon fibres are produced using PAN in Germany today, the explorative scenarios were defined as follows: 1) Carbon fibres are produced from bio-polymers (50% lignin and 50% bio-PU, referred to as lignin based carbon fibres from now on); 2) The carbonization and stabilization of the lignin-based fibres require less energy than PAN; 3) The spinning of the lignin-based fibres is further developed and made more efficient than it is today; 4) The demand for and hence price of lignin increases; and 5) The German electricity system transitions to a more climate neutral one. Note that Scenarios 2-5 for the lignin-based carbon fibres all include Scenario 1, i.e. the fibres in these scenarios are all based on the blend of 50% lignin and 50% bio-PU.

The climate-neutral electricity system was approximated by the current Swedish electricity system. While simplistic, the assumption in Scenario 5 is based on an electricity mix for the German system where the trend is to decrease the amount of fossils and increase the amount of renewables as the fossil electricity plants are replaced due to age (see e.g. Schumacher and Sands (2006)). The climate neutral electricity system was applied to all direct electricity use in the carbon fibre production system as well as to the bio-PU production system. It was not possible to alter the electricity mix for the PAN-precursor fibre production dataset due to its construction. For the same reason, the electricity mix was not altered for the production of electrolytes used for precursor fibre spinning, or for the polymers used for cleaning the precursor fibre spinning machine. As the production of lignin already was set in a low-carbon energy system, this was not altered either. The scenarios are further described in Table 3.

The time setting for the prospective assessment and all scenarios was assumed to be 20 years from now, i.e. in 2040. While it can be assumed that the transition from PAN to lignin and the increase in process efficiency (as described in Scenarios 1, 2 and 3) will happen sooner than that, the long-term frame was necessary to allow for significant changes in the German electricity mix (e.g.; one third of Germanys´ fossil-based electricity generating capacity may be retired by 2026 (Schumacher & Sands, 2006)) and the market demand for lignin as changes in these systems are slower.

Table 3: The different explorative scenarios applied for assessing the impact of future production of lignin-based carbon fibres in Case study 2

Scenario #	Current system	Future scenario	Type of factor	Assumption
1	Carbon fibres are produced using PAN.	Carbon fibres are produced using bio-based polymers.	Internal	Carbon fibres are produced using 50% lignin and 50% bio-PU.
2	The lignin-based fibres require as much energy as PAN for carbonization and stabilization.	Due to inherent material properties, lignin requires less energy than PAN for carbonization and stabilization.	Internal	The lignin-based fibres require 25% less energy for carbonization and stabilization than PAN (Das, 2011).[3]
3	The energy use for precursor fibre spinning was measured to be 1.7 kWh/kg fibre.	The spinning of the lignin-based fibres is further developed.	Internal	Lignin fibre spinning is further developed and made more efficient using 0.10 kWh/kg fibre (Das, 2011).
4	The global demand for lignin is low.	The global demand for lignin is high.	External	The price of lignin increases to 3 €/kg.
5	Production processes uses a German electricity mix.	German electricity mix transitions to be more climate neutral.	External	A future low-carbon energy system is approximated with a Swedish electricity mix.

The data used in Case study 2 was gathered as much as possible from participants in the LIBRE project using email questionnaires and discussions during project meetings within the consortium, and complemented with direct personal communication. When specific data for lignin-based carbon fibres were not available, data for PAN-based carbon fibres were used. The LCA software OpenLCA was used in combination with the LCA databases in Ecoinvent 3.3 (Wernet et al., 2016) and the ELCD (European Platform on Life Cycle Assessment, 2018) to generate results. The results are presented

[3] It is assumed that the blending of bio-PU does not change this value and that it is applicable to the lignin- and bio-PU based carbon fibres

in Section 5.1.2 and are an updated version of results available in Publication C and in internal project reports within the LIBRE project.

4.2.3 Case study 3: Assessing impacts of lignin using different allocation approaches

The goal of this case study was to assess how different allocation approaches can influence the environmental impact of lignin. Only climate impact was considered because of low data availability and because climate impact typically correlates well with other impacts (Janssen et al., 2016)

The functional unit of the study was the production of 1 kg of lignin at a Kraft pulp mill located in Sweden. Co-products of the system were pulp, soap, and heat, and the inventory for the Kraft pulp mill was primarily based on a dataset provided by Culbertson et al. (2016). All twelve allocation methods listed in Table 1 were applied. Some of the methods were applied in different variants reflecting a future setting to assess the cradle-to-gate climate impact of 1 kg of lignin-based carbon fibres. The future temporal setting of Case study 3 was set to 2040 (i.e. 20 years from now) for the same reason as in Case study 2; market changes related to lignin demand are expected to be rather slow, so to account for any significant changes, the timeframe must be long-term. The basic assumption is that the global demand for lignin will grow due to an increase in both the production of lignin-based carbon fibres and other products, e.g. lignin-based fuels. As a consequence, pulp mills could in extreme cases transition from producing pulp as their main product to producing lignin, and a higher price for lignin is expected as the demand increases. However, it was also assumed in variants of some allocation approaches that the price of lignin would remain constant at today's assumed value in spite of a growing supply of lignin.

As in the Case study 2, see Section 4.2.2, the results were generated using OpenLCA in combination with the Ecoinvent 3.3 (Wernet et al., 2016) and ELCD (European Platform on Life Cycle Assessment, 2018) databases.

5. RESULTS AND DISCUSSION

5.1 What is the life cycle environmental impact of carbon fibres in composites?

Case study 1 assessed the climate impact and energy use of carbon fibres in composites based on a meta-analysis of published LCAs. In Paper I, the impact of cradle-to-gate CFRP production in relation to the CFRP cradle-to-grave life cycle was examined for climate impact with 14 studies and for energy use with 11 studies. Results show that the CFRP production was responsible for a significant share of the cradle-to-grave climate impact and energy use. The cradle-to-gate climate impact of CFRP for the different studies found in the literature was found to be in the range of 13-56 kg of CO_2 eq./kg CFRP and the energy use range of 160-1080 MJ/kg CFRP (see Fig 4 in Paper I). The carbon fibre production for the cradle-to-gate of CFRP was found to be responsible for at least, but often more than, half of the total energy consumption in all cases (six studies), and it was responsible for more than half of the climate impact in all but one of five studies. This was likely due to a lower share of fibre in the composite in the divergent study (13-27% carbon fibre compared to 29-60% carbon fibre in the composite in other studies). Case study 1 also identified the energy needed (and its associated emissions) for the conversion of the precursor fibre into a carbon fibre as the main driver of the energy use and climate impact of CFRPs. To address this issue and to lower the environmental impact of CFRPs, two main routes were identified: the shift to lignin as a raw material for carbon fibre production and the use of recycled carbon fibres in composites. These two routes are further discussed in Section 5.1.1.

5.1.1 How would a transition to lignin-based or recycled carbon fibres change the environmental impact of carbon fibres in composites?

Figure 3 illustrates how a transition from primary PAN-based to lignin-based and recycled carbon fibres could change the environmental impact as explored in Case study 1. The climate impact and energy use for carbon fibres (both primary PAN- and lignin-based) are compared to the energy use of 1 kg of recycled carbon fibres and the production of 2 kg of lignin (assumed to be needed for the production of 1 kg of 100% lignin-based carbon fibres), using two different allocation methods and a marginal approach (in Figure 3 referred to as a consequential approach).

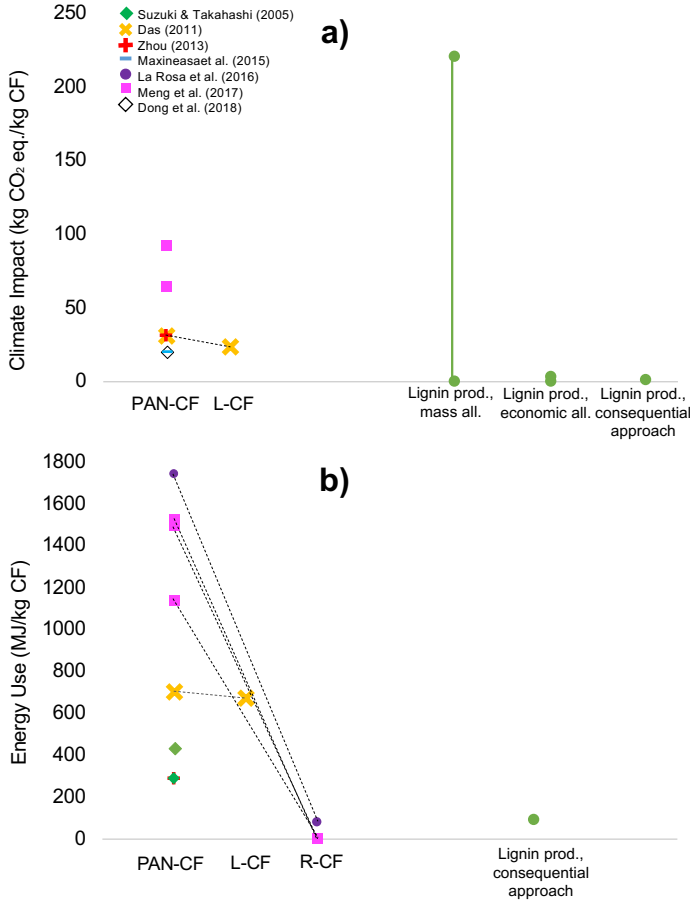

Figure 3: The climate impact (a) and energy use (b) for PAN-based carbon fibres (PAN-CF), lignin-based carbon fibres (L-CF), recycled carbon fibres (R-CF), and the production of lignin using different allocation methods. The different coloured markers for the carbon fibre production (PAN-CF, L-CF and R-CF) represent data from different sources, and the green lines represent the range of the climate impact of lignin production systems found in the literature using different allocation methods, with the green dots showing maximum and minimum values . Lines that connect PAN-CF and either L-CF or R-CF highlight potential savings as indicated in studies in the literature. Adapted from Hermansson et al. (2019).

Paper I showed that the climate impact of PAN-based carbon fibres was in the range of 20-92 CO_2 eq./kg of carbon fibres. Note that the comparison in Figure 3 does not consider any potential quality differences between the lignin-based, the recycled, and the primary PAN-based carbon fibres. Note also that the production technology of the primary PAN-based carbon fibres can be assumed to be much more mature than the production technology for the lignin-based carbon fibres or carbon fibre recycling process as there is no large-scale industry for these. This means that the impact could be lowered further as the production processes for lignin-based carbon fibres and carbon fibre recycling processes develop.

While the climate impact and energy use in Figure 3 for lignin production not includes any fibre production processes (actually only one data point, the yellow cross, in Figure 3 shows the climate impact of lignin-based carbon fibres), it is evident that the use of lignin as a raw material is a possible route for decreasing the environmental impact of future carbon fibres and, as a consequence, CFRPs. The two main reasons are connected to inherent properties of lignin: lower energy use in carbonization and a possible higher material yield in carbonization. Das (2011) writes that the use of lignin as a raw material could reduce the environmental impacts of carbon fibres as it could decrease the energy needed for carbon fibre production by 25% compared to the carbonization of PAN-based carbon fibres. This is due to the relatively high content of aromatics and oxygen in lignin, leading to less time required for both carbonization and stabilization (Das, 2011). The higher carbon content in lignin could also (in theory) increase the material yield of the carbonization process, but it is not known to what extent. Apart from decreasing the energy use, using lignin as a raw material could also come with other benefits, such as aiding the transition from a fossil-based to a bio-based economy as well as lowering the costs of carbon fibres and possibly lowering toxic emissions from the carbon fibre production; the carbonization of PAN leads to hydrogen cyanide emissions, which are toxic. However, emissions from the carbonization of lignin will still require monitoring due to the release of volatile organic carbon (Das, 2011). While there seem to be many benefits to using lignin as a raw material for carbon fibre production, there are processing challenges that must be overcome where the main difficulties are connected to the processability of lignin (Collins et al., 2019).

As shown in Figure 3, using recycled carbon fibres is a possible route for decreasing the environmental impact of CFRP. This route may actually be even more promising for decreasing climate impact and energy use for carbon fibres than producing carbon fibres using lignin as a raw material. The recycling of carbon fibres from composites, however, requires that the carbon fibres can be recovered from discarded composites and that the recycled fibres can be used in a second application. When assessing the environmental impact of recycled carbon fibres, a credit is often given for the impact of the avoided product (i.e. the product the recycled carbon fibre replaces; this is referred to as system expansion by substitution in LCA practice). Li et al. (2016) applied mechanical recycling to carbon fibre composites, which generated a recycled carbon fibre that was deemed suitable for replacing glass fibre. This gave the recycled carbon fibre a credit corresponding to the environmental impact for producing the glass fibres. In a study by La Rosa et al. (2018), the recycled carbon fibres were deemed to be of sufficiently high quality to replace virgin carbon fibres and, therefore, are rewarded a higher credit (i.e. 'removing' more emissions by replacing a product emitting more in the production phase) using the same approach. This highlights the importance of generating recycled carbon fibres with sufficiently high quality for this route to be viable, and the importance of choosing reasonable substitution products as this will impact final results of the LCA.

As both lignin-based and recycled carbon fibres are promising routes in terms of decreasing climate impact and energy use of carbon fibres and CFRPs, I cannot help but wonder what the combined gains would be. It is too early to draw any conclusions on what such a combination could lead to as the technology for both carbon fibre recycling, lignin extraction, and lignin-based carbon fibre production are still immature, and the generated volumes of both lignin-based and recycled carbon fibres are small. The possible future impact of lignin-based carbon fibres based on different possible scenarios will be further discussed in section 5.1.2, while the possible impact of recycled carbon fibres in composites is an area of future research.

5.1.2 Which are the environmental hotspots in the life cycle of lignin-based carbon fibres, now and in the future, and is lignin an important contributor?

As there is no large-scale production of lignin-based carbon fibres today, explorative scenarios were used to assess their possible impact today and in the future in Case study 2. The results were compared to the impact of PAN-based carbon fibres today and in the same external future. Preliminary results and data have previously been published in internal project reports and project meetings within LIBRE as well as in Publication C. The results presented in this thesis are an update of those preliminary results based on new data reported within the project and on assumptions made. The climate impact of producing 1 kg of PAN-based carbon fibres was based on a dataset provided by Romaniw (2013) and related to production in Germany today. A scenario where PAN-based carbon fibres are produced using a low carbon electricity mix was also generated based on this dataset. Both cases for PAN-based carbon fibres are shown with black bars in Figure 4.

The five future scenarios for lignin-based carbon fibre production in Table 3 were each applied in a prospective LCA to assess their possible future climate impact if nothing else is changed in the technical system, and a case that explored the combination of all scenarios was also generated. The results are presented in Figure 4 with grey bars.

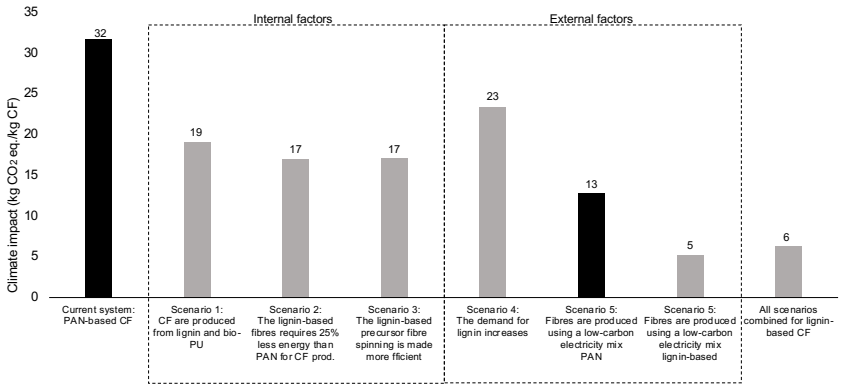

Figure 4: The climate impact of lignin-based (grey bars) and PAN-based carbon fibres (black bars) for different technology development routes, where the Scenarios 2-5 all are based on lignin- and bio-PU-based carbon fibres (i.e. Scenario 1).

The results indicate that the climate impact of the future lignin-based carbon fibres could be between 5 and 23 kg CO_2 eq./kg carbon fibre for the different scenarios. The scenario showing the lowest climate impact is Scenario 5 where the German electricity mix transitions towards a Swedish mix over time. The scenario with the highest impact is Scenario 4 with an increase in demand for lignin and, as a consequence, an assumed price increase. This is because a larger share of the pulp mill total burden would be allocated to the lignin as an economic allocation method was used. The difference between implementing all scenarios simultaneously and only Scenario 5 was minor. This is because the climate impact of carbon fibre (both for PAN- and lignin-based) is largely dependent on the electricity mix used in the production process. This means that when using a low-carbon electricity mix, changes in climate impact due to a change in energy use are not as significant as when using a high-carbon electricity mix. Note that the electricity mix was not varied for any chemicals or processing materials in the prospective case except for the electricity used in nitrogen production (see Section 4.2.2 for a full account of this). As mentioned in Section 4.2.2, it was not possible to alter the electricity used in the PAN-based precursor fibre production. As a consequence, this value would most probably be lower in the future if a more climate neutral electricity mix is used.

Figure 5 shows the climate impact of the lignin-based carbon fibres from Case study 2 divided into the following life cycle phases: bio-PU production (for blending with the lignin), lignin production, precursor fibre production (includes compounding, pelletizing, and spinning), and carbon fibre production (includes stabilization, carbonization, and surface treatment).

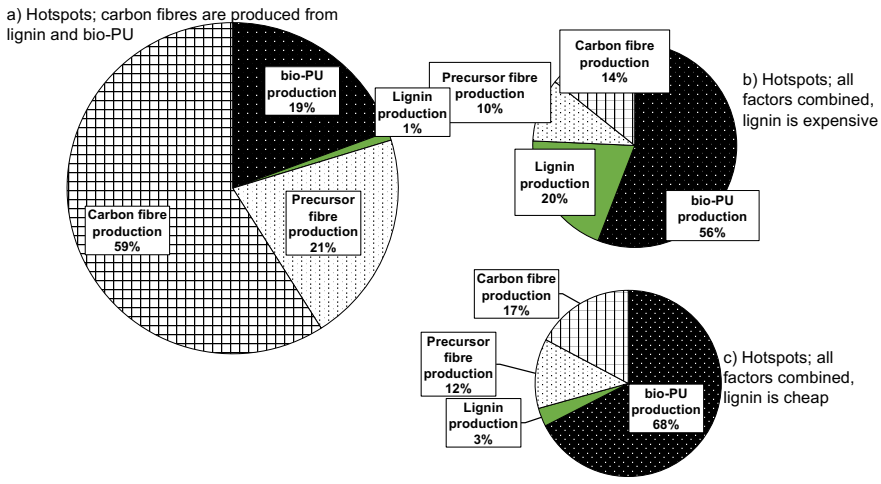

Figure 5: The different life cycle phases´ contribution to the climate impact of 1 kg of lignin-based carbon fibre for three scenarios in Figure 4: a) Carbon fibres are produced from lignin (Scenario 1) and b) all factors combined for lignin-based carbon fibres for lignin is expensive (as in all factors combined in Fig 4), and c) all factors combined and lignin is cheap (i.e. Scenarios 1,2,3, and 5). The size of the circles represents the relative size of the total climate impact of 1 kg of lignin-based carbon fibres.

Figure 5 shows that the main contribution to the climate impact of the current system is the carbon fibre production phase. In this phase, the energy use for the stabilization and carbonization of the precursor fibre is the main source of the environmental impact (the same is true for the PAN-based carbon fibres, as described in Paper I), and the impact of lignin is minor. The bio-PU in the prospective cases contributes the most to the impact, and lignin production is responsible for 3% in b) and 20% in c). In both of these cases, economic allocation was used to distribute environmental loads between products in the lignin production system. The price was assumed to be 3 €/kg for b) and 0.3 €/kg lignin for c) due to an assumed increase in lignin demand and supply in the future that might or might not affect the price. The possible influence a change in allocation method could have on these results is discussed further in Section 5.2.2.

As mentioned above, the largest contributor to the climate impact of the prospective cases is the production of the bio-PU used to blend with lignin, which is half of the weight

of the precursor fibre. This main contributor to the climate impact is the methylene diphenyl diisocyanate (MDI) production, which is responsible for 14% of the total climate impact of the carbon fibres shown in Figure 5 b) and 18% in Figure 5 c). While the prospective case uses a low-carbon electricity mix in the production of both the polymer and the polyol, the electricity mix was not altered for the production of MDI as it was not possible due to the construction of the dataset. It can be assumed that this value will drop in a future where the electricity mix has a lower carbon content. Another opportunity for decreasing the impact of the bio-PU in carbon fibre is by increasing the share of lignin as much as possible while still achieving the required fibre quality.

The results presented in Figures 4 and 5 have large uncertainties connected to the assumed scenarios. For example, in Scenario 5, in which the electricity mix is changed, the changes could not be made to the entire system. Any changes in emissions caused by varying the energy use in the PAN precursor fibre production or the MDI production are not accounted for. This means that the fibres' climate impact when the electricity mix is changed in the future may be even lower in reality. Other uncertainties related to the scenarios are market development, both for the carbon fibres and CFRPs. A larger market share for lignin-based carbon fibres could both increase the price of lignin due to a higher demand or lower the price (or as in the present study, be assumed to be a constant 0.3 €/kg) due to a larger supply as a consequence of a higher demand. This would influence the share of the pulp mills' burden to the lignin as economic allocation has been used. Both of these cases regarding changes in lignin demand are assessed in Figures 4 and 5, however, there remain uncertainties related to the actual future price of lignin. There are also uncertainties connected to the actual future electricity mix, and what any possible transitions of a German mix will look like in terms of carbon intensity.

5.2 How can challenges associated with assessing the life cycle environmental impact of lignin-based and recycled carbon fibres be addressed?

Two main challenges to assessing the life cycle environmental impact of lignin-based and recycled carbon fibres in composites have been identified: data availability in the early stages of material development and how to handle allocation. While data availability is always a problem in LCA, it is especially accentuated for emerging materials. A way to mitigate this is to do a meta-analysis of relevant results found in the literature, as described in Section 4.2.1 and further discussed in Section 5.2.1. Allocation was identified as a main challenge for both possible routes for decreasing the impacts of carbon fibre production, i.e. using lignin as a raw material and the recycling of carbon fibres. Allocation issues for lignin production are explored in Paper II, while allocation in the recycling of carbon fibres in composites is an area that requires further research. How allocation can be dealt with in lignin production is described in Section 5.2.2.

5.2.1 How can the lack of data for these emerging technologies be dealt with in LCA?

Sections 4.1 and 4.1.1 describe a method for mining information from LCA literature and building result packages in new contexts that could be used for guiding materials and process developments in the early stages of the design phase. The method was applied in Case study 1 (see Section 4.2.1). The method resulted from an iterative exploration conducted in the context of assessing how different technology routes can decrease the environmental impact of CFRPs (see Paper I). The approach of a quantitative meta-analysis is similar to the one used by Nordelöf (2017) but differs in that no qualitative assessment was made. Nordelöf (2017) conducted both qualitative and quantitative meta-analyses of electrified road vehicles, and found that the goal and scope formulation is crucial for the interpretation of the LCA results, and that in a sense, divergent results can be a consequence of the studies essentially answering different question. This issue was not addressed in detail in Paper I as the goal of Paper I and the method developed was to identify trends and hotspots of the production system; carefully assessing the goals and scopes of the studies was therefore unnecessary. However, it should be acknowledged that the studies compiled in Paper I likely answer different questions as a result of different goals and scopes and that this should be

considered when evaluating the results of a meta-analysis like the one presented in this thesis if this is important for the specific context.

The method developed in paper I proved useful for identifying hotspots in the life cycle of the material, as well as identifying the challenges and opportunities of different technology routes. As such, a meta-analysis of LCA results as described in Sections 4.1 and 4.1.1 can be a way of dealing with challenges related to assessments with the purpose of guiding materials and process developments in early stages of technology development. When using this method in other studies to obtain information for emerging materials, the amount and resolution of the available information in the literature will determine if and how data from different literature sources can be extracted and compiled. For understanding the environmental challenges and opportunities of a shift from primary PAN-based carbon fibres to lignin-based or recycled carbon fibres in composites, the influence of the allocation method needs to be studied carefully. In the specific cases of lignin-based and recycled carbon fibres, questions related to how to handle allocation in both the use of one material flow in several consecutive life cycles (i.e., recycled carbon fibres) and the use of one product from a multi-output process (i.e., lignin) are of significance. Note that other methodological issues than allocation may appear in other contexts.

A limitation of a meta-analysis method of the kind described here is that it would rarely allow for any sensitivity or statistical analysis, as the extracted data would typically not be sufficiently detailed for this to be done. Published data are in many cases aggregated, and LCA results tend to be quite opaque despite the fact that standards and recommendation documents emphasize the importance of transparency. Furthermore, the difference in scope and system boundaries among studies in combination with a relatively low number of studies also make a sensitivity or statistical analysis difficult and less meaningful in a specific case. As a consequence, the method is useful for providing a sense of the order of magnitudes by showing possible variations rather than generating mean values with standard deviations.

5.2.2 How can allocation be dealt with for lignin production, and does the choice of allocation method have a major influence on the environmental impact of lignin?

As mentioned in Section 4.1.2, ten allocation methods were found in the literature, either specific to lignin production or to biorefinery systems, and the latter were adapted for the context of this study to better fit a lignin-generating system (see Table 1). In addition to the methods found in the literature, two new allocation methods, *changes made to the mill* and *mass- and energy-based allocation*, were developed within the study as explained in Paper II. The rationale behind the allocation method *changes made to the mill* was that when lignin is introduced to an existing process, the impact of lignin should not be higher than the additional impact of the added extraction process plus the impact of replacing lost energy (as lignin is traditionally burnt for internal energy use). The idea was to keep the main product at approximately the same level of impact after the change, in this case pulp, and only punish the new product for all changes made. The method resembles subdivision, and as a consequence can be seen to follow, in principle, the ISO 14044:2006 and ILCD guidelines. The second allocation method developed by the authors of Paper II was the *mass- and energy-based allocation method*. This method is a version of the *energy- and mass-based allocation method* developed by Njakou Djomo et al. (2017), which is an allocation method combining different physical relationships. The rationale behind the original allocation method was that neither mass- nor energy allocation captures all flows of a mill, and exergy allocation can be difficult for the intended audience to grasp. As a consequence, a method combining mass- and energy flows was developed where the impact of the system is initially divided between the energy and material flows based on energy efficiency, followed by a classical mass allocation of the impacts distributed to the material stream, and energy allocation of the impacts distributed to the energy streams. However, the initial focus on energy efficiency leads to that using this method in an extreme case where there is no, or very little, energy content in the co-products, such as ash, carbon dioxide, and salts, all impacts are allocated to the energy streams, meaning that these co-products are left without any environmental impact. In response to this, the authors of Paper II developed an alternative method where the total impacts of the system are initially allocated between the material and energy streams based on the mass conversion rate (see Paper II for a more in-depth description of this method). This allows for a new perspective where the

mill (or biorefinery) is seen primarily as a mass conversion facility rather than an energy conversion facility.

The different allocation methods were applied to a case of lignin extraction from a Kraft pulp mill (Case study 3) by means of the LignoBoost process with a varying timeframe (now and in a long-term future, defined as 2040) by varying the lignin demand (and hence price) to examine how this could influence the future impact of lignin.

The LignoBoost process separates lignin from the black liquor by acidification with CO_2 and precipitation (Tomani, 2010). The extraction of lignin not only generates a product that can bring more revenue to the mill. It also increases the production capacity for co-products by debottlenecking the recovery boiler (Axelsson et al., 2006; Culbertson et al., 2016). Figure 6 shows the climate impact of 1 kg of lignin using the 12 different allocation approaches with different timeframes (now and in a long-term future, i.e. 2040) described in Table 1.

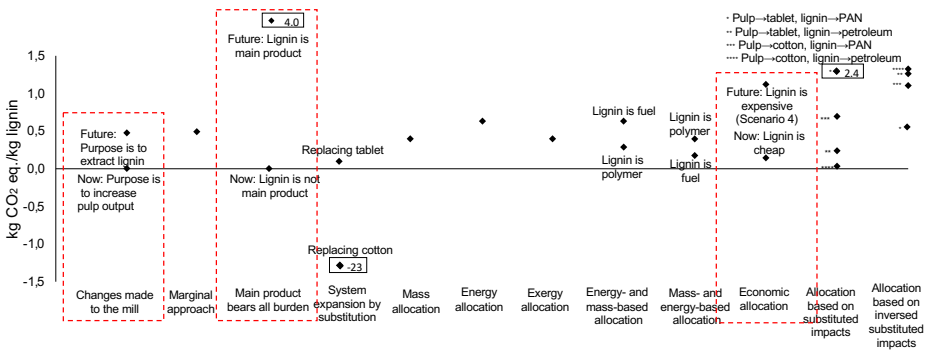

Figure 6: The climate impact of producing 1 kg of lignin using 12 different allocation approaches. Outliers not fitting the y-scale are marked with black boxes. The red boxes mark the allocation methods identified as most sensitive to changes in lignin market development and temporal settings. Adapted from Hermansson et al. (2020).

The results, as illustrated in Figure 6, show that the climate impact of lignin varies significantly with the choice of allocation method and timeframe. It especially varies with what products are considered to be replaced with the co-products of the system, the price of lignin, and how the price varies due to demand, and the drivers behind the

operation of the system (i.e. what is the system's main product now and in the future?), or behind why changes are made to the system (i.e. to extract more pulp or to extract lignin). The results highlight the importance of identifying the drivers both for the current system and a possible future system as well as considering how this could influence the allocation method. The results also show that it is important to consider what possible future changes there might be in lignin demand and, as a consequence, in the price of lignin and its co-products. Identifying the system's drivers and selecting appropriate allocation methods are particularly important for lignin and lignin-based products as the drivers of the system are expected to change with time as pulp mills and biorefinery technologies develop and also respond to developments in lignin technologies and lignin markets; in extreme cases, the reason for running the mill (or biorefinery) could change. For a more in-depth description and analysis of these drivers, see Paper II.

The highest climate impact of lignin occurs from using the method *main product bears all burden*, which represents a situation where lignin is considered the main product of the mill (4.0 kg CO_2 eq./kg lignin). This might be the case in a future with a large global lignin demand. The lowest impact occurs when using *system expansion by substitution* when pulp replaces cotton (-23 kg CO_2 eq./kg lignin), and thereby the lignin receives a credit for a highly impacting alternative. While the choice of cotton as a substituted product for pulp is rather extreme, it is included to illustrate that the choice of substituted product is very important. The choice of cotton as a replacement product is based on the assumption that pulp products could today be substituted by cotton in for example fluff in sanitation products and tote bags. A more reasonable alternative for the future might be plastics in composite products (see Publication A for such an example), but as this is a rather unexplored application, it was not included in the assessment. The choice of substituted products is described and discussed in more detail in Paper II.

Applying these two extreme methods to the base case of lignin-based carbon fibres in Case study 2 changes the results of the lignin-based carbon fibre's climate impact significantly. This variation was put in relation to the impact of lignin-based carbon fibres, as shown in Figure 5, to explore the significance of this variation in the impact of lignin resulting from the choice of allocation method. Figure 7 shows how the lignin

contribution to the total climate impact of the carbon fibre could change dramatically depending on allocation method, assuming that 1.1 kg of lignin is needed for the production of 1 kg of carbon fibre consisting of 50% lignin and 50% bio-PU due to material losses in the production process.

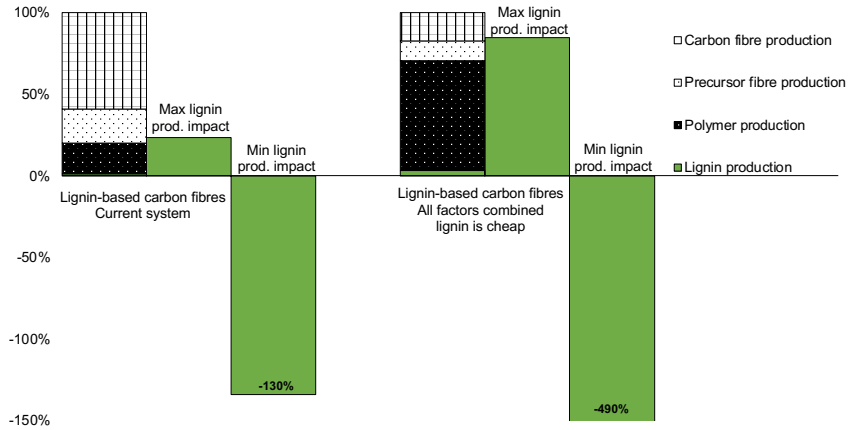

Figure 7: The influence of the choice of allocation method for lignin production on the climate impact of lignin-based carbon fibres of the current system (same as shown in Figure 5a) and a prospective case including a combination of Scenarios 1, 2, 3, and 5 in Table 3. Note that the scale is relative. The green bars represent the range that the lignin impact (green part) could have using different extreme allocation methods and timeframes.

Figure 7 also shows that depending on the allocation approach used, the production of lignin could very well be a major hotspot for the climate impact of lignin-based carbon fibres, both now and in the future. Lignin production could also decrease the total impact of lignin-based carbon fibres considerably when using the system expansion by substitution approach. However, as the resulting climate impact of this approach is highly dependent on the choice of products being replaced by the co-products of the system, the results are highly sensitive to assumptions made concerning these. The future system of the substituted products (e.g. related to production efficiency or to changes in the background system) are hard to foresee, especially in a prospective setting.

6. CONCLUSIONS

This thesis explored two possible routes to decreasing the climate impact of carbon fibres in composites: the use of lignin as a raw material for carbon fibre production and the recycling of carbon fibres. Two main challenges to assessing the climate impact of these routes were identified and addressed: 1) the lack of data availability in the early stages of material development and 2) allocation among different co-products in lignin production. I suggest that data scarcity in the early stages of material development can be dealt with by repurposing LCA results from the literature and recalculating them for new contexts in a meta-analysis. This repurposing would make it possible to identify trends and hotspots in the early phase of technology or material development. The results presented in this thesis show that transitioning from producing PAN-based carbon fibres to lignin-based, or the use of recycled carbon fibres, could decrease the climate impact and energy use of CFRPs and that the allocation method used in the lignin production system is important, especially in a future setting. For the ten allocation methods found in the literature and the two that were developed in the context of Paper II, results show that the timeframe of the assessed system and any changes this leads to are important to identify and consider, especially when assessing emerging materials. These two main findings contribute to a better understanding of how to assess emerging materials using LCA.

7. FUTURE RESEARCH

Among other things, this thesis attempts to suggest the future environmental impact of lignin-based carbon fibres by means of a prospective assessment of a case of cradle-to-gate lignin-based carbon fibre production. However, very little research has been conducted on assessing such a material in various real applications from a cradle-to-grave perspective; little is known of how that material compares to conventional materials, where the use of such a material would lead to an environmental gain, when it would not, and under what conditions. Therefore, more effort must be put into this.

This thesis only explores the climate impact and energy use of carbon fibres. This was mainly due to the fact that emissions data for lignin-based carbon fibres were insufficient to be able to do a fair comparison with the PAN case or other materials, such as metals. More effort must be put into collecting and publishing emissions data, so that more impact categories can be included in future research. There is also a need to develop relevant impact categories and impact characterization factors for, e.g. the use of biotic resources. For example, the transition to bio-based materials is often motivated by the fact that bio-based materials are seen as renewable and carbon neutral. However, depending on various factors, such as the time frame, geographical scale, and ecosystem management practices, these assumptions may not be true. An increase in global demand for bio-based raw materials along with excessive and irresponsible use can lead to impacts related to the loss of ecosystem services, such as climate control, and many other detrimental effects on human health, ecosystems and resource availability can also be imagined. Therefore, decision makers need support in choosing routes with low impact, e.g. in terms of biotic resource use. There is currently no widely accepted indicator in LCA for biotic resource use, which makes assessing the resource use for forestry products difficult.

This thesis identifies the use of recycled carbon fibres in composites as a route to decreasing the environmental impact of CFRPs. However, the recycling of carbon fibres was not explored further. Technical challenges to the recycling of carbon fibres in terms of process development along with methodological challenges to assessing the environmental impact of recycled carbon fibres need to be addressed. Methodological challenges to assessments are especially connected to the allocation of impacts between

the first-time use of carbon fibres and the use of recycled carbon fibres and this is identified as an area that needs further research.

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