

THESIS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

Advancing prospective life cycle assessment
- Experiences from guiding carbon fibre composite development

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Cover: An example of a life cycle assessment of a car. Picture by me, built using my kids Lego, with some help from my father Jonas Hermansson.

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ABSTRACT

Life cycle assessment is a powerful tool for quantifying the environmental impacts of goods and services. While most easily done for well-defined and known product systems, there is also a need for a prospective (that is, a future-orientated) approach to assessing developing technologies. This is because choices made in the early stages of technology development greatly influence the environmental impact of final product systems.

An increasingly popular range of materials on the market is carbon fibre composites, with a common application being in vehicles. The relatively light and strong carbon fibre composites can significantly reduce vehicles' weight and, therefore, the impact of the use phase. However, carbon fibres are based on a fossil raw material and their production is highly energy-intensive. This means that if used in a vehicle, there may be no overall environmental benefit from a life cycle perspective. In fact, the use of carbon fibre composites might even increase the environmental impact of vehicles' life cycles. Thus, the carbon fibre composite manufacturing process needs to be changed.

In this thesis, I present an environmental screening method to be used in the early stages of material development research projects when primary data is scarce. I also provide recommendations on how to allocate benefits and burdens between life cycles and co-products in prospective studies. Moreover, I reconceptualise the composite recycling process into a multi-output separation process, which leads to an allocation between both composite constituents and life cycles. I also provide three consistent future scenarios developed for carbon fibre composites. Finally, I include advice on how to compare immature to mature technologies in prospective life cycle assessments.

Keywords: life cycle assessment, prospective, carbon fibre composites, climate impact, energy use

SAMMANFATTNING

Livscykelanalys är ett kraftfullt verktyg för att kvantifiera produkters och tjänsters miljöpåverkan. Enklarest görs det för ett väldefinierat och känt produktsystem, men det finns också ett behov av en prospektiv, det vill säga framåtblickande, ansats för att utvärdera ny teknik i tidiga utvecklingsskeden. Detta på grund av att val som görs i de tidiga skedena av teknikutveckling har ett stort inflytande på det slutgiltiga teknisksystemets miljöpåverkan.

Ett material som blir mer och mer populärt på den större marknaden är kolfiberkompositer, där en vanligt förekommande tillämpning är i fordon. Det relativt lätta och starka materialet kan minska fordonets vikt, och därmed miljöpåverkan under användarfasen. Kolfibrer är emellertid traditionellt sett baserade på fossila råmaterial, och produktionsprocessen är mycket energiintensiv. Detta innebär att bytet till kolfiberkompositer i fordon inte per automatik leder till en miljövinst ur ett livscykelperspektiv. Det kan till och med leda till att fordonets totala miljöpåverkan ökar. Detta innebär att tillverkningen av kolfiberkompositer behöver utvecklas.

I denna avhandling föreslår jag en screeningmetod som kan användas i tidiga skeden av materialutvecklingsprojekt, när det är ont om primärdata. Jag ger också rekommendationer för hur miljöpåverkan ska fördelas mellan livscykler, vid återvinning, och mellan produkter vid processer som genererar flera produkter, i prospektiva livscykelanalyser. Dessutom konceptualiserar jag kompositåtervinningen till en separationsprocess med flera flöden, vilket leder till allokering mellan livscykler såväl som mellan de utgående produkterna. Jag tillhandahåller också tre framtidsscenarier som har utvecklats för kolfiberkompositer. Slutligen ger jag råd om hur man kan jämföra mogen med omogen teknik i prospektiva studier.

Nyckelord: *livscykelanalys, framåtblickande, kolfiberkomposit, klimatpåverkan, energianvändning*

LIST OF PUBLICATIONS

This thesis is based on the work described in the following five 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.

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PAPER II

Hermansson, F., Janssen, M., & Svanström, M. (2020). Allocation in life cycle assessment of lignin. *International Journal of Life Cycle Assessment*, 25(8), 1620-1632.

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PAPER III

Hermansson, F., Ekvall, T., Janssen, M., & Svanström, M. (2022). Allocation in recycling of composites - the case of life cycle assessment of products from carbon fiber composites. *International Journal of Life Cycle Assessment*, 27(3), 419-432.

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PAPER IV

Hermansson, F., Heimersson, S., Janssen, M., & Svanström, M. (2022). Can carbon fiber composites have a lower environmental impact than fiberglass? *Resources, Conservation and Recycling*, 181, 106234.

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PAPER V

Hermansson, F., Edgren, F., Xu, J., Asp, L., Janssen, M., & Svanström, M. (2023). Climate impact and energy use of structural battery composites in electrical vehicles - a comparative prospective life cycle assessment. *International Journal of Life Cycle Assessment*, 28(10), 1366-1381. <https://doi.org/10.1007/s11367-023-02202-9>

CONTRIBUTION REPORT

Paper I: All authors developed the idea and designed the methodology and framework. FH performed data collection and analysis and wrote the initial draft, which MJ and MS supervised and reviewed.

Paper II: All authors developed the idea, methodology and framework. FH performed data collection and analysis and wrote the initial draft, which MJ and MS supervised and reviewed.

Paper III: All authors developed the idea, methodology and framework. FH performed data collection and analysis and wrote the initial draft, which MJ, TE and MS supervised and reviewed.

Paper IV: FH developed the concept and all authors contributed to the methodology. FH and SH collected the data. FH wrote the initial draft and all authors reviewed it.

Paper V: FH developed the initial idea. FH, FE, JX and LA collected the data. FH, MJ and MS developed the methodology. FH wrote the initial draft and all authors contributed to rewriting and editing.

OTHER PUBLICATIONS

Work related to this thesis has also been presented in the following publications:

Journal papers:

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Hermansson, F., Janssen, M., & Svanström, M. (2023). Life Cycle Assessment as a Decision Tool in Material Development - Experiences from a Multi-year Carbon Fibre Composite Development Project. *Materials Circular Economy*, 5(1).

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Conference papers:

Hermansson, F., Berg, I., Sandberg, K., Asp, L., Janssen, M., & Svanström, M. (2021). The environmental benefits and challenges of a composite car with structural battery materials. *REV 2021 Proceedings. Resource Efficient Vehicles Conference*, Online.

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Conference presentations:

Hermansson, F., Janssen, M., & Gellerstedt, F. (2013). Environmental evaluation of bio-composites using LCA - Comparison of two different applications. *Proceedings of the 63rd Canadian Chemical Engineering Conference*, 20th-23rd October, Fredericton, Canada, <https://research.chalmers.se/en/publication/188110>

Hermansson, F., Janssen, M., & Svanström, M. (2018). Recommendations for Routes to Sustainable Exploitation of CFRP Materials. *ECO-BIO 18*, 5th-7th March, Dublin, Ireland. <https://research.chalmers.se/en/publication/501178>

Hermansson, F., Janssen, M., & Svanström, M. (2019). Environmental Challenges and Opportunities of Lignin. *10th International Conference on Industrial Ecology*, 7th-11th July, Beijing, China, <https://research.chalmers.se/en/publication/511316>

Janssen, M., Hermansson, F., & Svanström, M. (2021). Prospective life cycle assessment for biorefinery concept development. *Canadian Chemical Engineering Conference 2021*, 24th-27th October, Montreal, Canada.

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<https://research.chalmers.se/en/publication/533020>

Hermansson, F., Janssen, M., & Svanström, M. (2023). A Procedure for Prospective LCA in Materials Development - the Case of Carbon Fibre Composites. SETAC Europe 33rd annual meeting, 30th April - 4th May, Dublin, Ireland.
<https://research.chalmers.se/en/publication/535703>

Hermansson, F., Janssen, M., & Svanström, M. (2023). Lessons learned when assessing emerging composite materials using life cycle assessment. 23rd International Conference on Composites Materials (ICCM 23), 30th July - August 4th, Belfast, Northern Ireland. <https://research.chalmers.se/en/publication/536848>

Conference posters:

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Brunklaus, B., Hermansson, F., Armbrecht, J., & Lundberg, E. (2015). Turistens klimatpåverkan - modell och beräkning för Västsverige.
<https://research.chalmers.se/en/publication/226288>

Wickerts S, Hermansson F, Arvidsson R, Nordelöf A, Svanström M. Screening resource assessment of next-generation battery chemistries. 2021.
<https://research.chalmers.se/en/publication/528218>

Licentiate thesis:

Hermansson, F. (2020). Assessing the future environmental impact of lignin-based and recycled carbon fibres in composites using life cycle assessment. Chalmers Tekniska Högskola (Sweden).
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1. INTRODUCTION

A system may be defined as a set of interconnected elements organised to achieve something (Meadows, 2008). Single elements, such as raw materials or products, are not systems. However, production processes with their different elements of raw material input, refining and manufacturing are; they thus constitute a product system. One way of analysing and understanding product systems is by using life cycle assessment (LCA). In LCA, the environmental impacts of the system generating a product or service are quantified. It can help in such things as identifying environmental hotspots in the life cycle of products and services is therefore an essential part of product development. This is especially true when LCA results can be used by, say, technology developers to influence a product system and thus lead to environmental improvements. While an LCA is more easily done for a well-defined and known system, choices in the early stages of product development will often have a more significant influence on the environmental impacts of that product than decisions taken later (Moni et al., 2020). This prompts efforts to develop methodology and practice for LCA in the early stages of product design and/or process development.

This thesis has been written in the context of using LCA to assess the environmental impact of carbon fibre composites. Carbon fibre composites consist of carbon fibres in a polymer matrix and are often used for lightweighting of vehicles, resulting in reduced fuel consumption in the use phase (Duflou et al., 2012). However, carbon fibre production is energy-intensive, meaning that it does not automatically lead to a reduced overall environmental impact. Paradoxically, conventional lightweight materials that are heavier than carbon fibre do reduce environmental impacts (cf. Das (2011) and Witik (2011)). Therefore, if the environmental impact of vehicles with carbon fibre composites is to benefit from reduced fuel consumption in the use phase, measures must be taken to reduce the environmental impacts of producing the material. Two possible routes that have been suggested in the literature are: *i*) using bio-based raw material in carbon fibre production instead of fossil raw materials (Das, 2011; Janssen et al., 2019) and *ii*) recycling and recovering carbon fibres for use in new carbon fibre composites (Meng et al., 2017a; Zhang et al., 2020).

Another application of carbon fibre composites is in structural battery composites (SBCs). SBCs are carbon fibre composites that store and deliver energy while simultaneously carrying a mechanical load. In SBCs, the carbon fibres function as reinforcement (just as in conventional carbon fibre composite applications) but also host lithium ions and conduct electrons. The polymer matrix distributes load and provides ionic conductivity (Asp et al., 2021). Therefore, using SBCs in battery electric vehicles (BEVs) can reduce the lithium-ion battery (LIB) size and/or prolong vehicle drive range, while also decreasing the weight of the car's structural parts (Carlstedt & Asp, 2020). Material developers wanting to highlight the benefits of the new technology often refer to SBCs as multifunctional materials, as they have dual functionality; energy storage and provision of structural integrity. This addition of a function without adding weight has also given rise to the expression “massless energy storage”.

Producing lignin-based carbon fibres, recycling carbon fibre composites and manufacturing SBCs have yet to achieve industrial-scale production. Consequently, a prospective approach is needed when assessing the environmental impact of these technologies (Arvidsson et al., 2018). In this thesis, I identify and address methodological challenges related to the assessment of future composites using LCA. This was done using an iterative approach, in which lessons learned from LCAs performed on carbon fibre composites feed into the development of LCA methodology which, in turn, provides new case studies. The methodological development provides new case study layouts eventually better input to carbon fibre composite developers.

2. THESIS CONTEXTS

The research presented in this thesis was financed by two projects: The *LIBRE: Lignin-based carbon fibres for composites* (2016-2021) project and the *Carbon fibre composites and structural batteries in vehicles - when is it a good idea?* (2022) project. Papers I-V are built upon results from the work I undertook in these projects.

The *LIBRE* project was funded by the Bio-Based Industries Joint Undertaking under the European Union's Horizon 2020 research and innovation programme, grant agreement no. 720707. This large, multi-year, transdisciplinary project included representatives of academia, research institutes and industry. Their roles varied from carbon fibre and composite developers to vehicle developers. The work, reported in Papers I-IV, was undertaken as part of a work package dealing with the environmental and economic performance of lignin-based carbon fibres. Our main role as LCA practitioners was to guide the material developers in the *LIBRE* project in identifying routes for decreasing the environmental impact of carbon fibres. Thus, Papers I-IV are primarily results of the information sought by the consortium.

The project *Carbon fibre composites and structural batteries in vehicles - when is it a good idea?* was funded by Chalmers Transport Area of Advance. It was a collaboration between Chalmers researchers, who work using either LCA methodology or composite development. Compared to the *LIBRE* project, this consortium was significantly smaller. The aims were twofold: *i)* to provide input to material and vehicle developers from a life cycle perspective early in the technology development phase; and *ii)* to identify the main challenges and opportunities for decreasing the environmental impact of SBCs in vehicles. The work presented in Paper V is based on data collected through laboratory observations and input from material and vehicle developers.

3. ENVIRONMENTAL SYSTEMS ANALYSIS USING LIFE CYCLE ASSESSMENT

LCA is a tool that can be used by, say, material developers to include environmental considerations in their design and development decisions. It is a quantitative analysis method based on natural process models. It also serves as a decision support tool, evaluating the importance of different life-cycle stages and emissions in relation to concerns about the environment (Hertwich & Hammitt, 2001). LCA is specified in such documents as the ISO:14040 and ISO:14044 standards (International Organization for Standardization, 2006a, 2006b). Its operating procedure comprises four main phases: goal and scope definition, inventory analysis, impact assessment and interpretation. The procedure, as visualised by Baumann and Tillman (2004), is outlined in Figure 1.

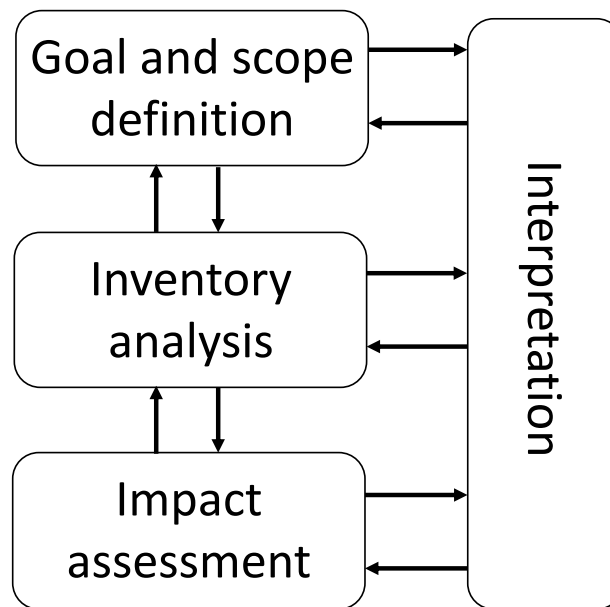


Figure 1. The work procedure for life cycle assessment, as visualised by Baumann and Tillman (2004).

The product or service to be studied and the purpose of the study are determined in the **goal and scope definition**. The ISO:14044 standard (International Organization for Standardization, 2006b) states that the following items must be included in the goals of an LCA: the intended application; the reason for carrying out the study; the intended audience; and whether the results are intended for use in a comparative declaration to be publicly disclosed. The scope, on the other hand, must include such things as: a description of the system to be studied (including its function); the functional unit (a reference whereby the input and output data is normalised); system boundaries; any

partitioning between products in multioutput processes or between life cycles in recycling; and the impact assessment methodology and impact categories. After the goal and scope definition comes the **life cycle inventory (LCI) analysis**, in which all the flows (such as raw materials, energy, products and emissions) entering and exiting the systems are inventoried.

In a **life cycle impact assessment**, the results from the LCI are translated into more environmentally relevant information. This includes sorting the inventory parameters according to what environmental impacts they contribute to (referred to as “classification”). After this comes the characterisation, which involves calculating the relative contributions of the emissions and the resource consumption for each type of environmental impact stated in the goal and scope definition (Baumann & Tillman, 2004).

The **interpretation** is ongoing throughout the study. Its goal is to identify significant issues and evaluate the completeness, sensitivity and consistency of the results. The main output of the interpretation is conclusions, limitations and recommendations for use in, say, product development, policy-making, or marketing (International Organization for Standardization, 2006b).

3.1 Prospective life cycle assessment

Future-orientated LCAs are often referred to as ex-ante LCAs or prospective LCAs. They explore the future by scaling up as-yet immature technologies of interest. This usually means exploring various future scenarios in which the technology could operate. Often, the scaled-up emerging technologies are compared to an (evolved) incumbent technology on the same scale (Cucurachi et al., 2018). Arvidsson et al. (p. 1287, 2018) define prospective LCAs as “studies of emerging technologies in early development stages, where there are still opportunities to use environmental guidance for major alternations”. In line with these definitions, the work underlying this thesis is called “prospective LCA” because the materials under consideration have not yet reached the large-scale production stage and are only just emerging onto the global market.

3.2 Dealing with multifunctionality

A big challenge of LCA is the distribution of burdens and benefits between product flows in multioutput processes and between product life cycles in recycling. In LCA, this is referred to as “allocation”. The ISO: 14044 standard (International Organization for Standardization, 2006b) specifies how to handle allocation for multi-output processes. The guidelines state that whenever possible, allocation should be avoided by: *i*) dividing the unit process into two or more sub-processes; and *ii*) expanding the product system to include additional functions of the co-products. In addition to the ISO: 14044 standard, the ILCD framework (European Commission’s Joint Research Center, 2010) suggests that the latter may be solved using system expansion by substitution. This means that the additional functions of the co-products are subtracted from the process. For cases in which subdivision and system expansion (by substitution) are not possible, the documents suggest that the allocation of system inputs and outputs should be based on the underlying physical relationships (such as mass or energy content). When physical relationships cannot be established, other relationships may be used, such as economic ones.

According to the ISO: 14044 standard (International Organization for Standardization, 2006b), when the system includes recycling, the allocation of inputs and outputs follows the same procedure as for multioutput processes. However, more elaboration is needed in the following situations: reuse and recycling leading to flows being shared by more than one product system; and reuse and recycling changing the material’s inherent properties. The ISO: 14044 standard distinguishes between systems in which the material’s inherent properties are not changed during recycling (most often closed-loop recycling but can sometimes be open-loop) and those in which the material’s properties are changed (always open-loop). In closed-loop recycling, allocation is avoided, since the use of secondary materials may be considered as a replacement for primary materials. Guidelines provided by such organisations as the British Standards Institute (2011) and WBCSD & WRI (World Business Council for Sustainable Development and World Resource Institute) (2011) provide advice on what allocation approach to use in different situations. In general, the cut-off approach, in which a credit is given to the product for using recycled material, should be used when the recycled material is not of the same high quality as the primary material. It should also be used in open-loop

recycling systems with inputs and outputs of recycled material. When recycled material maintains its inherent properties and can replace primary material, the end-of-life recycling approach should be used. In this approach, a credit is given for providing recycled material to another life cycle. In some cases, both allocation approaches are applicable. The Greenhouse Gas Protocol (WBCSD & WRI, 2011) stipulates that the cut-off approach is to be used: when the product contains recycled input but is not being recycled; when the use phase is long or uncertain; and when the content of recycled material is influenced by the company's activities alone. On the other hand, the end-of-life recycling approach is to be used when: the product's recycled content is unknown; when the product's use phase is short or well-known; or when the market for recycled materials is not saturated. Another recycling allocation approach that is likely to be essential for LCA practitioners in the future is the Circular Footprint Formula (CFF). This is a part of the Product Environmental Framework (PEF) provided by the European Commission (2018). The CFF is an approach to modelling recycling, energy recovery and waste disposal. It was developed through a comprehensive consensus process involving researchers, industry and authorities, with the aim of increasing the reproducibility and comparability of LCA results of goods and services. When using CFF, both impacts and credits from using and providing recycled materials are included. Impacts are distributed between the product life cycles using an allocation factor A, based on the recyclable materials' market supply and demand balance. The values for factor A are defined in Annex C of the Product Environmental Footprint (PEF) Category Rules Guidance (European Commission, 2021). A low factor A reflects an insufficient supply but a high demand for recyclable material (and vice versa if the factor A is high). When no A factor can be found in the list of default values, 0.5 should be used.

4. CARBON FIBRE COMPOSITES

Carbon fibre composites consist of carbon fibres, which provide structural integrity, plus a polymer matrix that distributes the load between the fibres. Due to its mechanical properties and low density, the composite is often used for lightweighting purposes in vehicles (Duflou et al., 2012). Traditionally, the carbon fibres are produced from the fossil-based raw material polyacrylonitrile (PAN). The PAN-precursor fibre is prepared using a solvent-based polymerisation process, generally followed by wet-spinning or air-gap spinning. The precursor fibre is then converted into carbon fibre through a series of processing steps, including carbonisation and stabilisation (Das, 2011). The composite is manufactured by arranging the fibres in a specific way for each application and the polymer and any additives are added before the composite is shaped by, say, injection or compression moulding. Figure 2 shows an image of a steering wheel made from carbon fibre composites.



Figure 2. A steering wheel made from carbon fibre composites. The checked pattern illustrates the woven fibre, which is impregnated with a polymer. Reprinted under licence CC BY 2.0¹.

¹ The image “CFRP steering wheel” by VK-Sportsman is licensed under CC BY 2.0. To view a copy of this licence, visit <https://creativecommons.org/licenses/by/2.0/?ref=openverse>.

4.1 Lignin-based carbon fibres

One possible route for decreasing the environmental impact of carbon fibres is using the bio-based precursor material lignin instead of the fossil-based PAN (cf. Das (2011) and Janssen et al. (2019)). Lignin is a macromolecule found in wood that provides structural integrity (Ragauskas et al., 2014). Currently, its main use is as an internal energy source in pulp mills and biorefineries (cf. Culbertson et al. (2016)). The production process for lignin-based carbon fibres resembles that of conventional carbon fibres. Nevertheless, it differs in that the lignin sometimes needs to be blended with another polymer to reduce brittleness (Collins et al., 2019). This is unnecessary for PAN. Another difference is that the precursor fibre is produced using melt-spinning rather than wet-spinning (which requires the use of solvents). Theoretically, the naturally aromatic structure and oxygenated chemical make-up of lignin could also lead to lower energy consumption in the carbon fibre production phase, as less time is needed for carbonisation (Das, 2011). There is currently no large-scale production of lignin-based carbon fibres, so the assessment requires a prospective approach.

4.2 Carbon fibre composite recycling

Fibres and polymers can be recovered through recycling at the end of the composite's life. There are three main types of carbon fibre composite recycling: mechanical techniques, whereby the composite is treated without chemicals or heat (by shredding, for example); thermal techniques, whereby the matrix is decomposed using heat or microwave radiation; and solvolysis techniques, whereby the matrix is decomposed using chemical reactions (Dong et al., 2018). In the two latter cases, fibres are separated from the matrix, with the polymer matrix being recovered in varying degrees. It has been shown that recycling the composite and recovering the fibres is a successful route for decreasing the environmental impacts of carbon fibre composites (Meng et al., 2017a; Meng et al., 2017b; Zhang et al., 2020). There is currently no industrial-scale recycling of carbon fibre composites but many efforts are targeting this, as described by Rybicka et al. (2016). Thus, carbon fibre composite recycling may be considered an emerging technology. A prospective approach to the LCA is needed.

A major challenge regarding LCA modelling in composite recycling is that composites consist of at least two materials that have been merged. Depending on the quality degradation when they are recycled, these materials may meet very different fates. Moreover, the recycled composite constituents may have different recycling rates (theoretically, from 0 to 1), further complicating the assessment. While many products consist of multiple materials, the impacts relating to dismantling and separating them are typically minor and have little influence on the LCA results. However, with composites, these processes are often energy-intensive and must be considered.

4.3 Structural battery composites

The main difference between SBCs and conventional carbon fibre composites is that in SBCs, the carbon fibres act as a host for lithium ions and also conduct electrons. The polymer matrix also provides ion conductivity (Asp et al., 2021). However, the ability of commercially available PAN-based carbon fibres to host lithium comes at the cost of ultimate tensile strength. Lithium is permanently trapped, resulting in the development of strain (albeit reversible) in the fibres (Jacques et al., 2014). A challenge for the polymer is that the two properties (distributing load and providing ionic conductivity) often counteract each other. This makes it possible to achieve high ionic conductivity without mechanical rigidity, or vice versa. Nevertheless, it is difficult to accomplish both functions (Asp et al., 2019).

The SBC's electrodes are insulated from each other by a separator. A thinner separator with appropriate pore properties increases the elastic modulus and energy density of SBCs compared to a thicker separator. This is because the thinner one enables a higher fibre volume fraction while also reducing the resistance (Asp et al. (2021) and Xu et al. (2022)). However, too thin a separator may lead to short-circuiting and there may also be problems withstanding processing (Asp & Greenhalgh, 2015). Figure 3 shows a schematic image of an SBC battery cell with a cellulose-based separator and aluminium and copper-based current collectors.

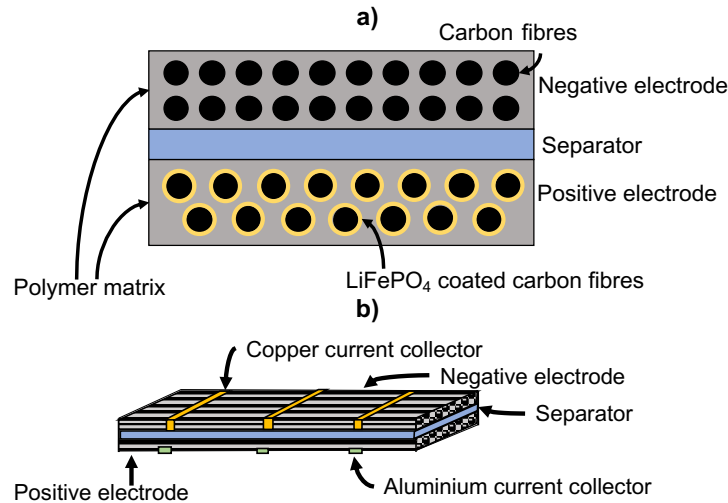


Figure 3. Schematic image of a structural battery composite in a) cross-section and b) from above. Reprinted from Hermansson et al. (2023a).

The multi-functionality of SBCs provides even more opportunities for lightweighting vehicles, as the mass of the LIB can be reduced while maintaining the same drive range for the BEV. An alternative approach would be for the vehicle's mass to remain constant but have an extended drive range. For example, a total vehicle weight reduction of 20-30% is possible with only a minor decrease in vehicle driving range. Alternatively, a 70% longer drive range can be achieved if the vehicle weight is maintained (Carlstedt & Asp, 2020). However, it is a challenge to capture such changes in material and/or vehicle functionality in a comparative LCA.

5. AIM OF THESIS AND RESEARCH QUESTIONS

Prospective LCA introduces opportunities for decreasing the environmental impacts of products by targeting hotspots and exploring possible paths early in the technology development process. However, there are many challenges in assessing a product system that does not yet exist. In response to these challenges, this thesis aims to contribute to the development of prospective LCA as a tool for environmental systems analysis. By assessing systems that are relevant to carbon fibre composite and vehicle developers, this thesis also provides these stakeholders with input on how to decrease the environmental impacts of carbon fibre composites.

Five research questions (RQs) have been formulated to fulfil the aim of contributing to the advancement of prospective LCA.

- RQ 1. How can usable information from available LCA studies be extracted and made relevant to new contexts?
- RQ 2. How can allocation in multi-output processes capture future changes in systems, particularly in lignin-generating processes?
- RQ 3. How can allocation be handled when assessing the recycling of materials with different fates in prospective contexts?
- RQ 4. How can future product systems and, in particular, carbon fibre composites be assessed using consistent scenarios with regard to foreground and background systems, including a differentiated selection of methodological choices?
- RQ 5. What are the key considerations when comparing immature to mature technologies in prospective LCAs?

The research questions in this thesis are answered in the appended papers, as shown in Figure 4.

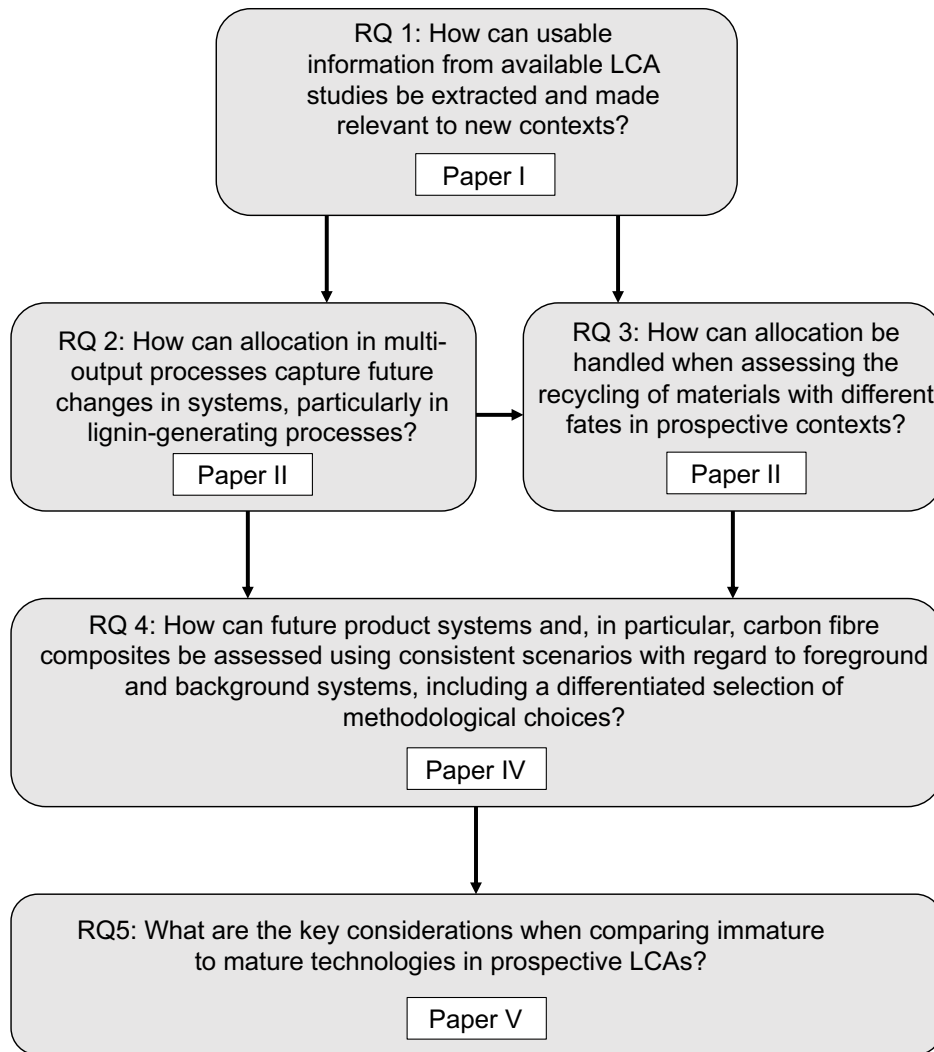


Figure 4. Outline of how the five research questions are related and which publications are primarily used to answer these.

6. METHODOLOGY

In this thesis, I apply a systems perspective to comprehend and capture the complexity of product life cycles. The methodology of this thesis builds on the notion that case studies are useful when there is a need for an in-depth understanding of a complex issue in an applied setting (Crowe et al., 2011). To quantify the environmental impacts of product life cycles, the case studies in this thesis use the LCA framework. There are different kinds of case studies; Stake (1995) distinguishes three: intrinsic, instrumental and collective. Intrinsic case studies are used to learn about the case and may be given to us by a stakeholder in a project. In instrumental case studies, the case study is used to gain a general understanding of an issue. Collective case studies, in turn, include studying multiple cases (instrumental and/or intrinsic) to generate an even broader understanding. Given these definitions, the overall methodology of this thesis could be described as a collective case study, aimed at shedding light on the complexity of assessing the environmental impacts of emerging products using prospective LCA. In turn, the collective case study consists of some primarily intrinsic case studies (Case studies 1, 4 and 5, done on behalf of project partners) and some that are primarily instrumental (Case studies 2 and 3, done to understand a phenomenon in LCA methodology).

In practice, this thesis was created using an iterative approach, in which lessons learned from LCA case studies of different types of carbon fibre composites feed into LCA methodology development. The methodological development, in turn, provides new case studies and new composite material knowledge. The iterative procedure of this thesis is illustrated in Figure 5.

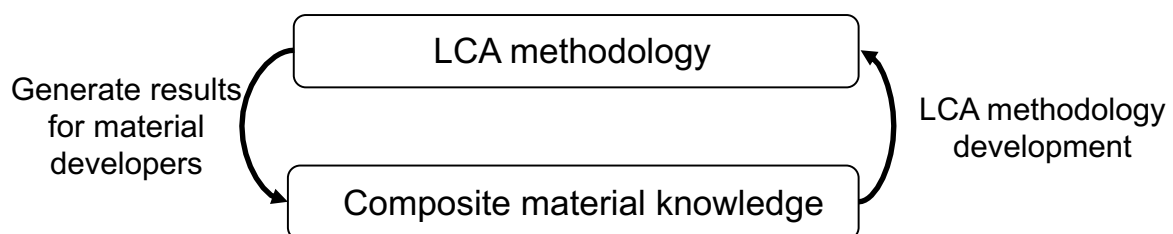


Figure 5. Outline of the research design used in this thesis.

The Methodology section is divided into two parts: *i*) Data collection (Section 6.1) and *ii*) Case studies (Section 6.2). The data for the LCA case studies was collected using various methods, including various forms of literature studies and discussions with experts from different disciplines. These discussions focused on capturing the stakeholders' need for knowledge about the life cycle environmental impacts of carbon fibre composites and how they could meet my data needs as an LCA practitioner. In the context of this thesis, including experts in the case study helped meet their need for decision support regarding what design or development choice to make, to improve the environmental impacts of carbon fibre composites. Moreover, the relevance of the results was enhanced by primary data from experts in the carbon fibre composite life cycle. The collected data was used in case studies, as shown in Figure 6. It should be noted that the case studies presented in the summary of this thesis are not always exactly the same as those described in each paper. Sometimes, the case studies were further developed to generate even more targeted results or illustrations (c.f. 6.2.1 and 6.2.2)

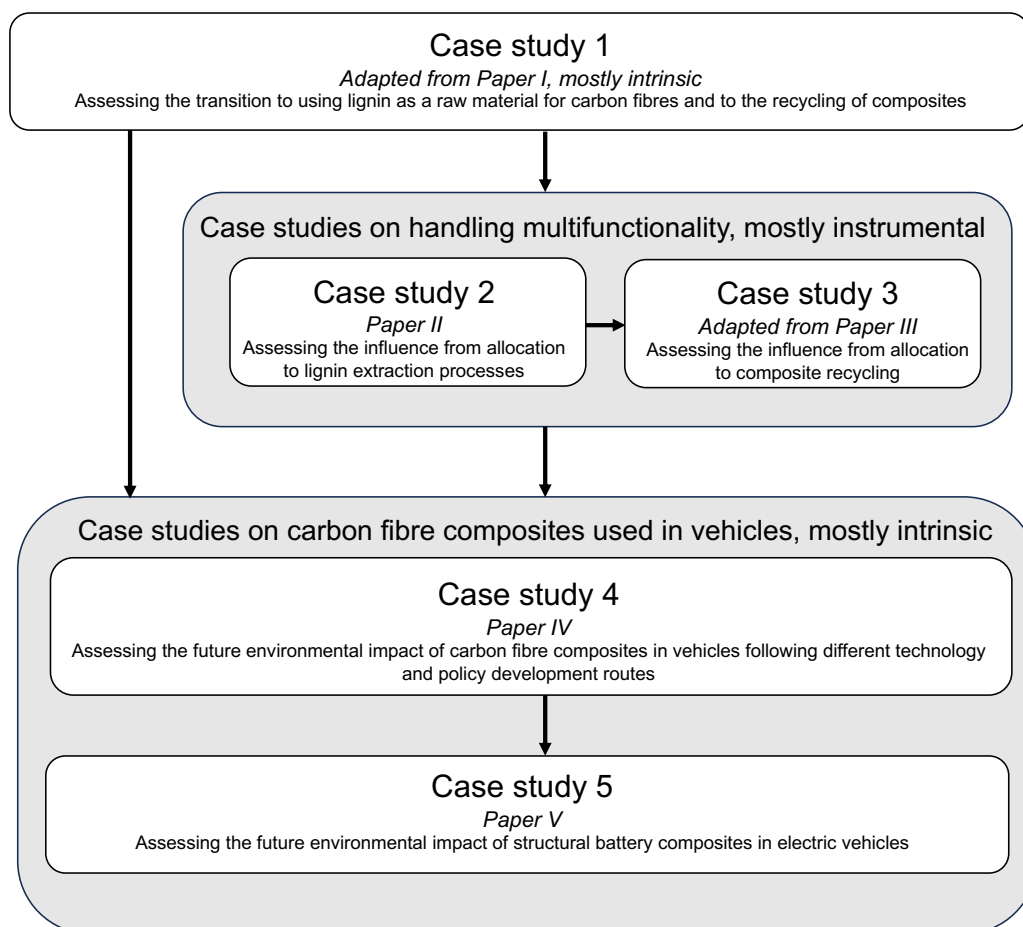


Figure 6. Outline of how the intrinsic and instrumental case studies are connected in the collective case study.

6.1 Data collection

Data for the case studies was collected from three main sources: literature, discussions with experts and study visits. Additional databases were used (primarily for the background system modelling), such as Ecoinvent 3 (Wernet et al., 2016) and the ELCD database (European Platform on Life Cycle Assessment, 2018).

The literature was used to collect relevant LCAs of carbon fibre composites and lignin extraction processes. It was also used to identify allocation approaches suitable for lignin-generating processes and for composite recycling. The literature was primarily found using Google Scholar in combination with Summon (Chalmers Library, 2010) and Scopus. Most of the collected literature consisted of peer-reviewed journal papers. However, reports, dissertations and conference proceedings were also collected. While the data collection that used the literature could generally be grouped under classical literature reviews, it was usually neither exhaustive nor systematic (that was deemed unnecessary for the purposes of the case studies). Rather, the criterion was to extract enough information to allow the RQs to be answered. The process was highly iterative. When new areas of interest were found or a knowledge gap identified, the scope of the initial search was expanded to include those areas.

Data was also gathered through discussions with various experts. This was done during physical or online project meetings, by telephone, email and sometimes via data collection questionnaires. The experts spanned researchers, carbon fibre manufacturers, composite manufacturers and vehicle manufacturers. The discussions were initially based on what type of input the experts needed from the LCA case studies. This was followed by discussions on what data the experts needed to provide for the case studies.

In addition to discussions, expert data was also gathered during study visits to laboratories and production plants. In these cases, the experts' information was complemented by observations. Visits were made to: *i*) precursor fibre production plants (2018); *ii*) carbon fibre production plant (2018); and *iii*) a structural battery composite manufacturing laboratory (2022).

6.2 Case studies

This section describes basic information on the case studies that were conducted using the data that was gathered (as described in Section 6.1). The case studies are briefly outlined in Table 1. In cases when additional work for this thesis was done (beyond what is included in Papers I-V for Case Studies 1 and 3), the case study methodology was described in more detail, see Sections 6.2.1 and 6.2.2.

Table 1: A brief outline of case studies 1-5.

Case study #	Paper	Goal	Functional unit	System boundaries	Allocation	Data sources
1	I	Assess influence from using lignin as a fibre raw material and of recycling of composites	<ul style="list-style-type: none"> - Production of 1 kg carbon fibre composite - Production of 2 kg lignin - Recycling of 1 kg carbon fibre composite 	<ul style="list-style-type: none"> - Cradle-to-grave - Cradle-to-gate - Grave-to-gate 	<ul style="list-style-type: none"> - Mass allocation - Economic allocation 	<ul style="list-style-type: none"> - Literature
2	II	Assess influence of allocation approach for lignin extraction	Extraction of 1 kg lignin	Cradle-to-gate	- 12 different, see Table 3	<ul style="list-style-type: none"> - Literature - Ecoinvent 3.3
3	III	Assess influence of allocation approach in composite recycling	Service provided by one piece of passive carbon fibre composite product	Cradle-to-grave	<ul style="list-style-type: none"> - Cut-off approach - End-of-life recycling approach - Circular footprint formula - Mass allocation 	<ul style="list-style-type: none"> - Literature - Ecoinvent 3.3
4	IV	Explore if and when carbon fibre composite can outcompete fibreglass in vehicles	Service provided by one pair of car mirror brackets for 100,000 km	Cradle-to-grave	<ul style="list-style-type: none"> - Cut-off approach - End-of-life recycling approach - Economic allocation 	<ul style="list-style-type: none"> - Literature - Discussions with experts - Study visits - Ecoinvent 3.3
5	V	Assess future impact of using structural battery composites in battery electric vehicles	The vehicle's roof, doors and bonnet with maintained flexural stiffness, used for 200,000 km and the lithium-ion battery	Cradle-to-grave	<ul style="list-style-type: none"> - Cut-off approach - End-of-life recycling approach - Mass allocation 	<ul style="list-style-type: none"> - Literature - Discussions with experts - Study visits - Ecoinvent 3.8

6.2.1 Case study 1

While Paper I presented the idea of using LCA results as building blocks for assessing future product systems, the actual building blocks were not generated. In this thesis, I therefore complement the original case study by creating the building blocks for the production/recycling of 1 kg carbon fibre composite as well as 2 kg of lignin (assumed amount needed to generate 1 kg of carbon fibres). The composite is assumed to consist of 20 wt. % fibres and 80 wt. % polymers. Data for the cradle-to-gate carbon fibre composite production assessment is based on findings in Das (2011); La Rosa et al. (2016); Maxineasa et al. (2015); Suzuki and Takahashi (2005); Zhou (2013) and Meng et al. (2017a). For the recycling process and substitution credits, data is based on Witik et al. (2013), Li et al. (2016), Dong et al. (2018) and Khalil (2018) (no substitution values are included for the last one). For the lignin-generating process, the data came from González-García et al. (2011), Culbertson et al. (2016), González - García et al. (2016), Nascimento et al. (2016), González - García et al. (2017) and Gullón et al. (2018). For the lignin-generating processes, the average value of the data in Paper I is used for both a mass-based and an economy-based allocation (assuming a price of EUR 0.3/kg lignin). All values used to generate the building blocks are found in the [Supplementary Material for Paper I](#).

6.2.2 Case study 3

In Paper III, we applied mass allocation to distribute the impact of the separation process between the composite's constituents. To assess the influence of the choice of allocation approach in the separation process, I here also use the *main product bears whole burden* approach in two different ways. In one case, the fibre is seen as the main product, whilst in the other it is the polymers. The functional unit is the service provided by one piece of fictitious product φ used for σ years and weighing 1 kg. The product consists of 30 wt.% fibre and 70 wt.% polyamide. A passive product type is considered to allow for a simplification in terms of the use phase being disregarded. At the end-of-life, three different composite recycling techniques are considered: mechanical recycling using grinding; thermal recycling using pyrolysis; and chemical recycling using supercritical water. Details on the recycling, allocation factor α_i , the A factor (used in CFF only) and quality degradations for the respective materials are provided in Table 2 and details on how these were selected are found in Paper II.

Table 2. Values for the variables used in the calculations. * Assumed to substitute polymer, **Assumed to substitute petroleum.

Variable	Constituent	Grinding	Pyrolysis	Supercritical water
Recycling rate, incoming material	Fibre	0	0	0
	Polymer	0	0	0
Recycling rate, outgoing material	Fibre	0.8	0.8	0.8
	Polymer	0.8	0.8	0.8
Allocation factor, separation process	Fibre	0.3, 0 and 1	0.3, 0 and 1	0.3, 0 and 1
	Polymer	0.7, 0 and 1	0.7, 0 and 1	0.7, 0 and 1
Factor A (only CFF)	Fibre	0.5	0.2 and 0.8	0.2 and 0.8
	Polymer	0.5	0.5	0.5
Quality correction factor, outgoing materials	Fibre	n/a*	0.82	0.98
	Polymer	0.9	n/a**	0.9

The assessed impact category is the CED in CML2001, provided by Ecoinvent 3.3 (Wernet et al., 2016). All data came from the Ecoinvent APOS 3.3 database (Wernet et al., 2016), except for the PAN-precursor fibres, which are an adapted version of the data from Fazio and Pennington (2005) (see Paper III for details of the adaptation). All LCA modelling was done using the OpenLCA software.

7. RESULTS AND DISCUSSION

7.1 How can usable information from available LCA studies be extracted and made relevant to new contexts?

The developed method (as described in this section) resembles traditional meta-analyses in LCA, in which results from different studies are normalised to the same functional unit (cf. Archer et al. (2018) and Lorenz et al. (2019)). However, it also differs in that this method allows results from different studies to be combined to generate an understanding of systems that do not yet exist on a large scale. While the method, as presented here, appears straightforward, the process was highly iterative and each new step depended on the findings in the preceding one. The major steps of the method include:

1. Compiling relevant published LCA studies that jointly contain sufficient knowledge of the studied field.
2. Mining, recalculating and assembling information from the studies.

The first step includes a search for LCA results that can be extracted from the literature to provide an understanding of a new field that has yet to be assessed or has only been nominally assessed before. Thus, to obtain sufficient coverage of the new life cycle, studies covering parts of the new conceptual product life cycle need to be found and combined. It is preferable (but not essential) that several studies for each life cycle phase should be included, as this allows cross-validation. Hotspot identification and order-of-magnitude comparisons can still be done but their results need to be interpreted carefully.

The second step is based on: *i*) an initial screening of the compiled literature, based on functional units, system boundaries and common environmental impact categories (including differences between the impact assessment methods that were used); and *ii*) presenting results concerning life cycle phases. How and what data is mined is based on the need for information about emerging technologies and what questions the study aims to answer.

The approach that was developed enabled the identification of hotspots in carbon fibre composites' cradle-to-grave and cradle-to-gate life cycles. The main steps of the procedure are outlined in Figure 7. Note that this outline is specific to the case of assessing carbon fibre composites and the transition to using lignin as a precursor material in carbon fibre production.

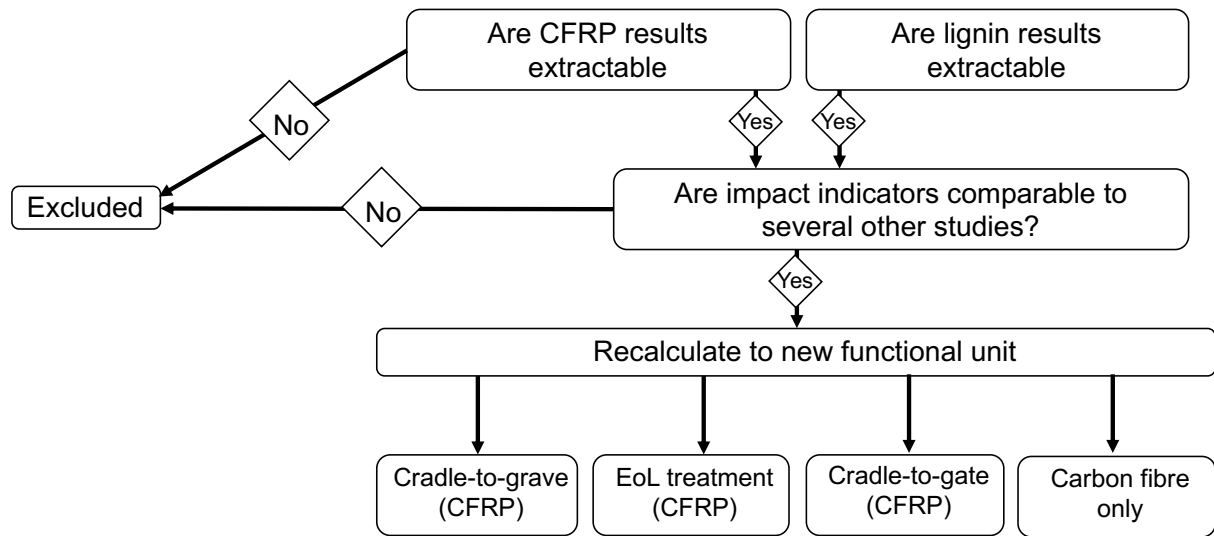


Figure 7. Mining and recalculation procedure outlined in Paper I. CFRP is an abbreviation for carbon fibre reinforced polymers (i.e., carbon fibre composites). Adapted from Hermansson et al. (2023c).

As well as enabling hotspot identification, the method also allows for the creation of “building blocks” for different parts of product systems. These building blocks may be combined into new product systems to indicate what the new systems will look like and what their future environmental impact may be. Methodological challenges related to the assessment of different technological routes using LCA can also be identified. The procedure concept and building blocks for future carbon fibre composites (20 wt. % fibres and 80 wt. % polymer) are shown in Figure 8.

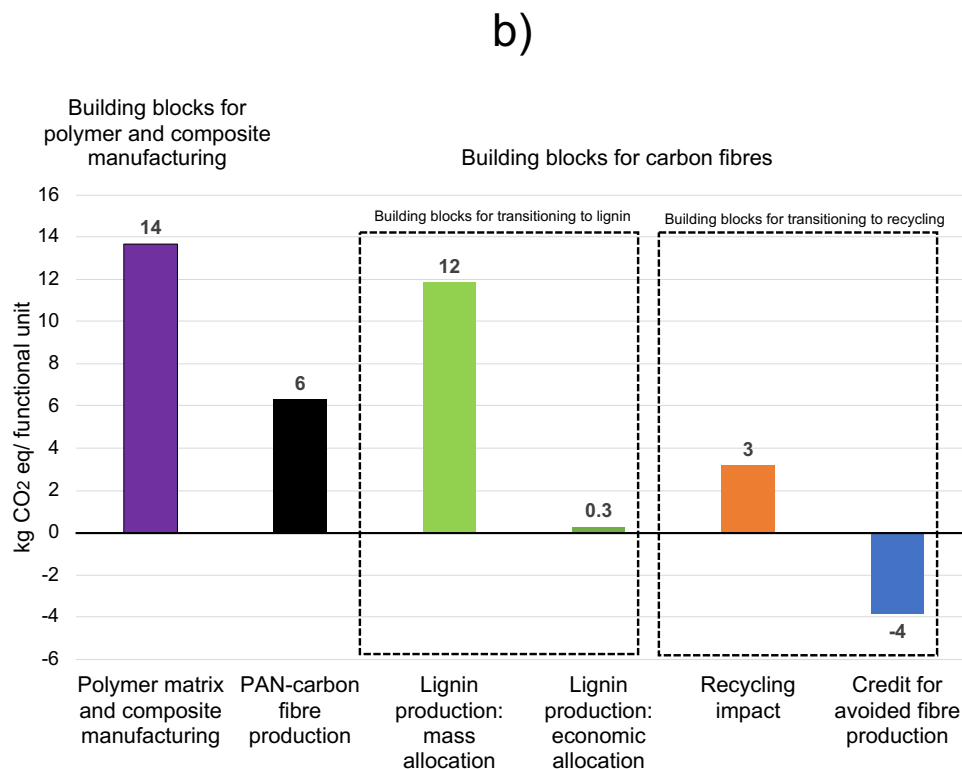
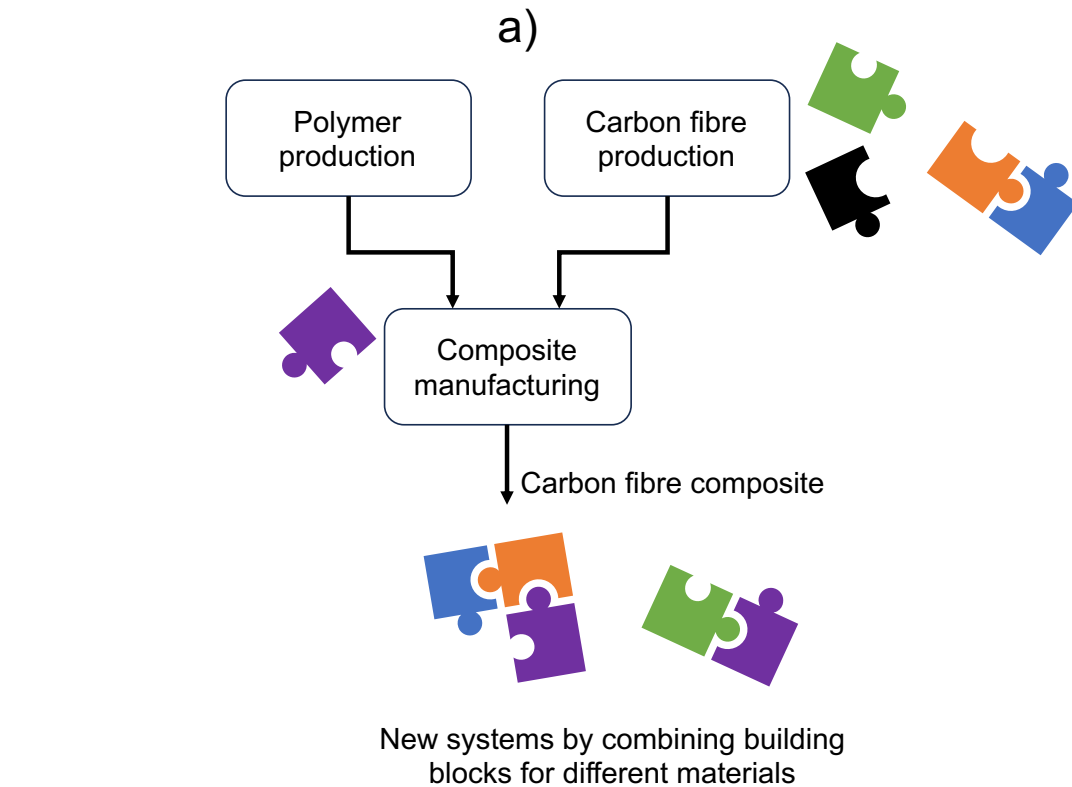


Figure 8. a) A conceptual illustration of using building blocks (extracted from literature) for different parts of the carbon fibre composite's life cycle to generate new systems and b) size of the different building blocks for a conceptual future carbon fibre composite product system. The purple block represents polymer matrix and composite manufacturing; the black block is conventional carbon fibres; the green block is lignin; and the blue and orange blocks are the recycling and recovery of fibres.

Hermansson et al. (2023b) compared the building blocks for PAN-based carbon fibres in composites with the results based on primary data collected during the LIBRE project. This includes composites made from PAN-based carbon fibres using contemporary data as well as a prospective assessment of carbon fibre composites which includes several technology development routes (such as using lignin as a raw material as well as microwave heating). We found that the impact for the PAN-based carbon fibre production using the meta-analysis results and the contemporary primary data was of the same order of magnitude but that there were large differences between the polymer matrix impacts. This was partly due to different polymers being used in the matrices but also because different LCA studies have different system boundaries and present their results differently. Thus, although considerable effort was made to discover and compensate for (or discuss) differences, there was likely some double counting of impacts, primarily related to composite manufacturing processes (such as injection moulding) and transportation. Original credits for avoided carbon fibre production were recalculated as corresponding to 0.2 kg of carbon fibres. As the recalculation did not allow for any heat recovery, the credit is likely higher in some cases due to a higher proportion of polymer in the matrix (>80 wt.%)

Using the meta-analysis approach allowed information to be extracted from the literature and assembled into new systems. This approach identified carbon fibre composite manufacturing as the cradle-to-grave hotspot and carbon fibre production as the cradle-to-gate hotspot. It also facilitated an understanding of the challenges of assessing a transition from primary, PAN-based carbon fibres to recycled or lignin-based ones. In this specific case, this approach identifies allocation in prospective markets as a key challenge (as seen in Figure 8). This is because the choice of allocation approach strongly influences the results of lignin production and recycling. This means that differences in results will not be due to the inherent properties of the technologies but rather to the methodological choices of the LCA practitioner.

7.2 How can allocation in multi-output processes capture future changes in systems, particularly in lignin-generating processes?

Section 7.1 showed that the allocation approach used in the lignin-generating system could significantly influence the resulting impact. This means that the influence of the allocation approaches needs further assessment. This is especially important in a prospective study such as lignin production and utilization, as lignin's primary use is still mainly for internal energy in pulp mills and biorefineries. If the market for lignin changes, demand might increase. For some allocation approaches, this would change the resulting environmental impact. Therefore, not considering this transition could generate skewed results and flawed decision support for technology and material developers.

A literature search revealed ten allocation approaches suitable for lignin-generating processes. Two allocation approaches were also developed in Paper II to address gaps in the literature, connected to the need to capture future changes in lignin-generating systems. These approaches are: *changes made to the mill* and *mass and energy-based allocation*.

The rationale behind the allocation approach *changes made to the mill* is that when lignin extraction is introduced to an existing process, the lignin's environmental impact should not exceed the impact of the added extraction process and any impact from replacing lost energy within the system. Essentially, this means that the impact of the main product of the existing mill is unchanged and any additional impacts from extracting the lignin are only allocated to the lignin product.

The *mass and energy-based allocation* was developed as a response to the energy and mass-based allocation approach developed by Njakou Djomo et al. (2017). The original allocation approach builds on the rationale that neither mass nor energy-based allocation captures all flows of a mill and that exergy allocation can be challenging for the intended audience to understand. To address this issue, Njakou Djomo et al. (2017) suggest that the mill's flows should first be allocated based on energy efficiency. This should be followed by a classical mass allocation for the flows allocated to the material streams and energy allocation for the flows allocated to energy streams. A drawback of

using this approach is that there might be cases in which there is very little or no energy content in some of the co-products (such as carbon dioxide, ash, or salts). In these situations, all impacts from the mill are allocated to the energy streams and other co-products are left with no environmental impact. To address this, Paper II developed an alternative approach, allocating the flows between material and energy streams based on the mass conversion rate rather than the energy conversion rate. This allows for a different perspective, whereby the pulp mill or biorefinery is seen as a mass conversion facility rather than an energy conversion one, something which could become the primary purpose of biorefineries in a bioeconomy future.

Table 3 shows the collected and developed allocation approaches. It includes a short description of these and an assessment of their sensitivity to changes in temporal settings. For an in-depth description of each approach, see Paper II.

Table 3. Allocation procedures used for assessing the climate impact of lignin production. Adapted from Hermansson et al. (2020).

Method	Approach	Reference	Sensitivity to temporal changes
Changes made to mill	Subdivision simulated pragmatically. 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	High
Marginal approach	Considers the difference in impacts of the whole system before and after lignin extraction.	Bernier et al. (2013)	Low
Main product bears whole burden	A main product of the system is selected to carry the entire environmental burden.	Sandin et al. (2015)	High
System expansion by substitution	System boundaries expanded to include replacement of other products on the market.	European Commission's Joint Research Center (2010)	Medium
Mass-based allocation	Impacts of the system partitioned based on mass of each co-product flow.	ISO 14044 and ILCD*	Low
Energy-based allocation	Impacts of the system partitioned based on energy content of each co-product flow.	ISO 14044 and ILCD*	Low
Exergy-based allocation	Impacts of the system partitioned based on exergy content of each co-product flow.	Cherubini et al. (2011)	Low
Energy and mass-based allocation	Impacts of the system first partitioned between the energy streams and the mass streams based on energy efficiency. This is followed by either energy allocation (for energy streams) or mass allocation (for material streams).	Njakou Djomo et al. (2017)	Low
Mass and energy-based allocation	Impacts of the system first partitioned between the energy streams and the mass streams based on the mass conversion rate. This is followed by either energy allocation (for energy streams) or mass allocation (for material streams).	Proposed by the authors of Paper II	Low
Economic allocation	Impacts of the system partitioned based on the economic value of each co-product flow.	ISO 14044 and ILCD*	High
Allocation based on substituted impacts	Impacts of the system partitioned based on the impacts of replaced products.	Cherubini et al. (2011)	Medium
Allocation based on inverse substituted impacts	Impacts of the system partitioned based on the inverse impacts of replaced products.	Sandin et al. (2015)	Medium

*International Organization for Standardization (2006a) and European Commission's Joint Research Center (2010)

Table 3 shows that allocation approaches based on physical relationships have low sensitivity to changes in temporal settings. This is supported by such other scholars as Coelho et al. (2022), who argue that mass allocation is more appropriate in prospective assessments as the outcome will remain over time. The allocation hierarchy in the guidelines also supports this claim. Table 3 also shows that allocation approaches based on substitution have medium sensitivity to temporal changes. This is primarily related to the fact that the replaced product type can change with time (or even between studies) and that the substitution product significantly influences the assessment results.

In Table 3, it is evident that the approaches: economy-based allocation (*main product bears whole burden* and *changes made to mill*) are related to a high degree of sensitivity to changes in the temporal settings of the study and are thus likely to introduce a higher degree of uncertainty in prospective contexts. Economy-based allocation is at the bottom of the guidelines' allocation hierarchy. However, there are arguments stating that economy-based allocation better reflects the reason for a system's existence and thus makes more sense to users (Huppes & Schneider, 1994). The economy-based allocation also reflects the quality aspects of the process and the products generated. Thus, this may be more appropriate to use than the mass allocation, in situations where one or more co-products have significantly higher market prices than others (Ardente & Cellura, 2012). However, future market prices are difficult to foresee. This means there is a trade-off in prospective LCAs of multi-output processes, between generating robust results and reflecting a change in the system. Likewise, when there is increased demand for lignin to perhaps be considered the main product of the process, the outcome of using *main product bears whole burden* would change. The *changes made to mill* approach is sensitive to the future reason for extracting lignin, as lignin extraction can also increase the efficiency of the recovery boiler, thus generating more cellulose (Axelsson et al., 2006). When the reason for lignin extraction is to debottleneck the recovery boiler and generate more cellulose, the impacts of the extraction process should be allocated to the cellulose. When the reason is to extract and sell lignin, the impacts should be allocated to that. This means that when using these allocation approaches in prospective studies, the driver of the product's existence needs to be carefully considered, plus how this might change over time.

Arguably, when assessing future lignin extraction, the allocation approach *main product bears whole burden* (with lignin as the main product), or *changes made to mill* (with the intention of selling the lignin) are the most appropriate as a proxy for a future in which there is relatively high demand for (and thus price of) lignin. This avoids economy-based allocation and detailed future supply and demand analyses. This reasoning stems from lignin still being underused as a raw material in the context of material production. Sometimes, lignin is even considered a waste product and left free of burden in LCA calculations. However, “there is no such thing as a free lunch” and if a material such as lignin is seen as a hidden gem for the future², it makes little sense to consider it as a waste product or by-product, in the future.

7.3 How can allocation be handled when assessing the recycling of materials with different fates in prospective contexts?

The reconceptualisation of the composite flow by modelling the recycling of composites into multiple parallel flows proved helpful in capturing the different fates of the composite’s constituents. While this conceptual split of the composite into different material flows seems obvious in hindsight, it is not intuitive for a composite to be thought of as several material constituents that will have different fates at their end-of-life and in subsequent life cycles. If the composite is modelled as a single material being recycled, the constituents’ various properties, quality degradations and differences in supply and demand are lost. This could mean that one constituent carries too significant a burden from the recycling process, or vice versa, thus creating skewed results.

² In fact, I took a summer course in 2019 at Lund University called “Lignin - a hidden gem for biorefineries?”

It is impossible to provide general advice on what allocation approach to use, as this depends on the context and purpose of each LCA study. However, as society changes and technologies develop, variables in the allocation approaches are expected to change. One way of coping with this in future-orientated studies is to investigate the extremes. This would mean that both the cut-off and end-of-life recycling approaches should be included in prospective assessments. Thus, the maximum range of possible changes in market saturation and technology changes will be captured. An example of a type of technology development that could be important to consider is that the quality degradation of the composite constituents in recycling would decrease over time.

The adapted allocation approaches were applied in a case study assessing three cases of carbon fibre composite recycling. The results are shown in Figure 9. The impacts generated using the cut-off approach are identical for all recycling options. This means there is no incentive to send the product to recycling, nor to improve the recycling process. What cannot be seen is that it generally does provide an incentive to use recycled materials. However, this was not considered in the study as adding an input of recycled material as a parameter would have complicated the modelling of the different scenarios and made them hard to interpret. The results generated using the end-of-life recycling approach in Figure 9 show that, even though Case C displays a relatively high environmental impact from the recycling process, there are clear benefits to recovering high-quality constituents. This demonstrates that this allocation approach provides clear incentives for recovering high-quality materials (especially those with the highest environmental impact associated with primary material production) and for improving the recycling process. However, it does not provide any incentives for using recycled materials. The CFF approach, on the other hand, strongly depends on the supply and demand balance of the market (factor A in CFF). The results in Figure 9 suggest that when there is a high demand for recycled carbon fibres, these should be recycled separately, especially if the quality of the recycled carbon fibres can remain high. Nevertheless, a low-impact recycling method is preferable when demand for recycled carbon fibres is small.

A drawback of the CFF is that it is complex and challenging to apply in prospective studies. This is because it considers not only the recycling rate but also the market supply and demand balance, the specification of the substituted product and the quantified quality changes of each constituent. In cases where the future market is unknown, PEF stipulates that an A factor of 0.5 is to be used (European Commission, 2018). This automatically makes the approach a compromise between the cut-off approach and the end-of-life recycling approach in those cases. It also makes CFF superfluous if both other allocation approaches are used to explore the range of possible outcomes.

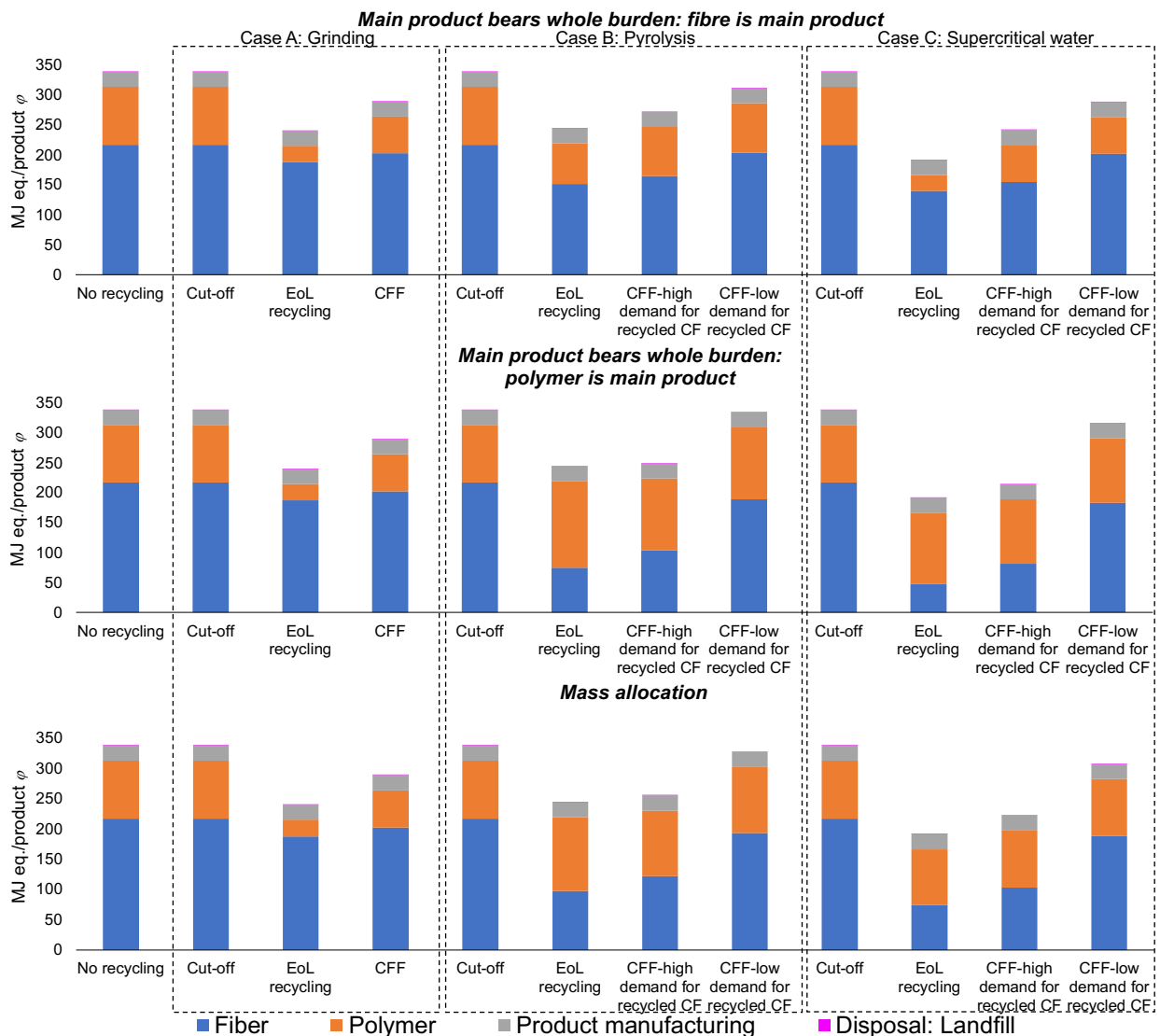


Figure 9. Influence of the recycling allocation approach and allocation factor used to distribute the separation process for three different recycling systems. Adapted from Hermansson et al. (2022a).

Figure 9 also shows how the choice of multi-output allocation approach to the recycling process affects the results (using either the *main product bears whole burden* approach or mass allocation). Not surprisingly, we find that when the fibre is the main product of the system, its impact increases and when the polymer is the main product of the system, the impact of the polymer increases. This is because the full separation process impacts are attributed to the main product.

A somewhat counterintuitive finding is that the total impact of the CFF approach varies as the allocation factor for the separation process, α , changes. This is due to the A factor, which is based on the supply and demand of recycled materials and which scales the related benefits and burdens. Consequently, the impact of the separation process is scaled to the A factor. When there is low demand for the constituent that has been allocated a large share of the impacts from the separation process and when there is no input of recycled materials, the impact of the separation process decreases. In essence, this means there is a risk of greenwashing. Greenwashing can be avoided by identifying the driver behind the recycling process and either allocating the full impact of the separation process to the driving flow, or using economic allocation, the latter however introduces large uncertainties.

7.4 How can future product systems and, in particular, carbon fibre composites be assessed using consistent scenarios with regard to foreground and background systems, including a differentiated selection of methodological choices?

Discussions with experts, study visits and literature searches identified several possible future development routes for carbon fibre composites. These include changes to the foreground system: *i*) using lignin as a raw material in carbon fibre production; *ii*) using microwave heating in fibre production; and *iii*) recycling composites and recovering fibres. Some changes to the background system were also identified: *iv*) changes in lignin demand and thus price; *v*) decarbonisation of the energy system; and *vi*) moving away from internal combustion engine (ICE) vehicles to BEVs. Since many technology development routes are likely to be implemented in parallel due to such things as vehicle funding synergies, policies and legislation, it can be meaningful to create consistent scenarios. This means that assumptions in future development are combined using cross-consistency assessments. This is followed by a distinctness-based selection to keep the number of assumptions and scenarios to a reasonable level (Langkau & Erdmann, 2021). The benefit of creating consistent scenarios is that it enables the LCA practitioner to assess which future is likely to achieve the lowest environmental impact.

By combining the technology development routes identified for carbon fibre composites, plus background system changes (such as demand for bio-based raw materials and energy mixes) following the approach by Langkau et al. (2023), three consistent scenarios for carbon fibre composites were developed: a bioeconomy future, a circular economy future and a circular bioeconomy future. In essence, the bioeconomy future includes routes and background system changes related to minimising the use of fossils; the circular economy future includes routes focusing on recycling; and the circular bioeconomy future combines the previous two to illustrate an ideal future technical system.

When developing scenarios for comparing future technology development routes, I suggest including the allocation approach as a parameter. This may be done by connecting the allocation approaches to aspects of future world development through, say, the concept of weak or strong sustainability (see Paper IV). This connection is based on Frischknecht (2010), which proposes that the cut-off approach should be connected

to the strong sustainability concept (natural capital remains constant). The end-of-life recycling approach is instead related to the weak sustainability concept (total capital remains constant). In line with this, the cut-off approach was used in the bioeconomy to reflect a society focusing on minimising the extraction of fossils from the ecosphere. In the circular economy, the end-of-life approach was chosen because it provides an incentive to recycle (this was deemed to be aligned with the ideas of a circular economy). In the circular bioeconomy, the cut-off approach was used as this was deemed to be more strongly connected to the strong sustainability concept. Moreover, in the bioeconomy, the allocation factors of multioutput processes for bio-based products can be varied. An example would be capturing the transition of something that is currently considered a by-product into a high-demand future product. These considerations may also be connected to strong or weak sustainability, whereby the *main product bears whole burden* approach for bio-economies is in line with the strong sustainability concept (decreased fossil extraction from the ecosphere). The scenarios for the technology development routes being considered are shown in Table 4.

Table 4. Description of the changes considered in each future scenario. Adapted from Hermansson et al. (2022b).

	Changes to the foreground system	Changes to the background system
Scenario 1: Bioeconomy future	<ul style="list-style-type: none"> - Fibres produced from bio-based raw materials - Fibres made using microwave heating - Composites sent to landfill 	<ul style="list-style-type: none"> - Lignin price increases - Energy mix transitions towards being fossil-carbon lean - Composites used in battery electric vehicles - Legislation to reduce extraction of fossils from the ecosphere
Scenario 2: Circular economy future	<ul style="list-style-type: none"> - Fibres produced using fossil-based raw materials - Fibres produced using conventional technologies - Composites recycled and materials recovered - Recycled fibres used to manufacture composites 	<ul style="list-style-type: none"> - Lignin price remains the same - Energy mix stays constant - Composites used in internal combustion engine vehicles - Legislation to promote recycling and recovery of materials; end-of-life recycling approach
Scenario 3: Circular bioeconomy future	<ul style="list-style-type: none"> - Fibres produced using bio-based raw materials* - Fibres produced using microwave heating - Composites recycled and materials recovered - Recycled fibres used to manufacture composites 	<ul style="list-style-type: none"> - Lignin price increases - Energy mix transitions towards being fossil-carbon lean - Composites used in battery electric vehicles - Legislation to reduce extraction of fossils from the ecosphere; cut-off allocation approach

The connection of the allocation approaches to the different sustainability concepts enables a selection based on various future environmental concerns. These concerns would, in turn, influence legislation and policy instruments that may impact technological development by such means as R&D funding. This means that when creating consistent scenarios based on the likelihood of different sub-scenarios happening simultaneously, allocation approaches may also be chosen based on the overall focus of the system. This may involve either decreasing the extraction of fossils from the ecosphere or recycling of materials.

7.5 What are the key considerations when comparing immature to mature technologies in prospective LCAs?

Assessing the environmental impact when transitioning from a conventional material to an emerging one in a future setting brings many challenges. One major such challenge is that a comparative prospective assessment requires that the product systems should be equipotent, so that the LCA practitioner is not “comparing apples and pears”. The challenge becomes even more obvious when more functions are added to the emerging technology or the view of the functionality shifts. The result is that the selection of functional units for product systems and the drawing of system boundaries will need careful consideration.

When comparing dissimilar product systems, defining the functional unit poses a challenge. This is because there is a fundamental mismatch between the materials’ functions, which needs to be captured by the functional unit. One way to solve this is to enlarge the system boundaries to include all functions provided by the system. According to the ISO:14044 standards set by the International Organization for Standardization (2006b) and the guidelines of the European Commission’s Joint Research Center (2010), this is the recommended action for handling multifunctionality and avoiding allocation in classical LCA. This aligns with the suggestions made by Hetherington et al. (2014) who write that new materials are not necessarily direct replacements for conventional ones, as their functions may not be equivalent. They suggest mitigating this by expanding the system boundaries to achieve functional equivalence between product systems. Likewise, Heimersson et al. (2019) suggest considering a “basket of functions” to make different systems comparable. An example

of product systems with dissimilar functions is the comparison of SBCs (which provide both mechanical integrity and energy storage) with metals (which only provide mechanical integrity) or batteries (which provide only energy storage). In this case, the functional unit needs to cover both the structural parts of the vehicles as well as the energy storage capacity equivalent to what the SBCs can manage. For a BEV, this would be a fraction of the LIB. See Paper V for details.

Another challenge when comparing mature and immature technologies is that conventional materials will probably have established recycling systems. This means that for all routes considered, there will be a cascade of allocation issues to handle, even when recycling the immature technology is not considered. In the case of the assessment of SBCs compared to metals and LIB (Paper V), this becomes very apparent. In this situation, there is already an established recycling system for metals and LIBs, whereas the recyclability of SBCs is yet to be implemented. This means that allocation to recycling needs to be considered in all situations for SBC, even when the SBC itself is not recycled. This is because the recycling allocation approach significantly influences the outcome of the study, and the selection of allocation approach is difficult in prospective studies. This further highlights the importance of: *i*) carefully selecting the allocation approach and being transparent with this choice; and *ii*) including both the cut-off approach and end-of-life recycling approach in prospective studies, as two future extremes.

Another challenging aspect when comparing immature to mature technologies is that there is probably a difference in repairability. This may originate from such things as an extended functionality of an emerging technology (as with the SBC), which may be difficult to restore after an accident. It may also result from products becoming more complex and integrated, making it harder to replace damaged parts. One example is laminating smartphone glass covers together, which makes changing broken screens difficult (Knight, 2017). This means that, for some technologies, there is a need to include certain incidents or accidents in the assessment. While the inclusion of accidents in LCA is usually dismissed in most guidelines (cf. the European Commission's Joint Research Center (2010)), excluding them can lead to essential flows being overlooked in product systems where they are more likely to occur (Fries & Hellweg, 2014). In the specific case

of the assessment of SBCs compared to metals and LIBs, the potential need to include the impacts of accidents is significant. This reasoning is initially based on the study by Ishfaq et al. (2023). They write that SBCs may need complete replacement if damaged, as they could be too difficult to repair. This may mean that, even if the part is damaged such that most of its mechanical integrity is maintained, the function of energy storage may have been compromised. However, for a conventional material, the accident may not lead to any changes other than aesthetic ones. This means that, if the accident is not included, the comparative assessment no longer compares equivalent functions. This was shown in Paper V to potentially influence the results and conclusions of the study. This may be seen as one of the crucial flows mentioned by Fries and Hellweg (2014) that risk being overlooked if accidents are not included in the assessment.

8. CONCLUSIONS

To address the lack of data in the early stages of material development projects, LCA results may be extracted from the literature, recalculated to a new functional unit and merged to form systems that do not yet exist. When assessing these routes, this is a helpful approach for identifying hotspots, identifying different potential technology routes for decreasing environmental impacts. It also allows for identifying methodological challenges.

Economy-based allocation is more sensitive than physical allocation to the influence of future system changes. This may be seen as a benefit of economic allocation but it also introduces significant uncertainties related to future market prices and demand. To avoid these large uncertainties related to predicting future markets, the *main product bears whole burden* approach (with the product currently underutilised as a main product) can be used as a proxy. This allows for capturing future changes in, say, a pulp mill that transitions to selling lignin as a product alongside cellulose. Another possible solution is to consider the impacts of the process added to the system due to the change. However, this requires the LCA practitioner to reflect on why the change is implemented in the first place, as the reason might change the outcome of this approach.

I suggest considering the recycling process of composites as a multi-output separation process. This means that the benefits and burdens from recycling should be allocated between life cycles and between the constituents generated by the recycling process. In doing so, differences in quality degradation and market demand between the constituents may be captured. I also recommend including both the cut-off and end-of-life recycling approaches in prospective studies to capture extremes concerning future market saturation and demand.

To fully capture the differences between potential futures, I recommend including the choice of allocation approach as a parameter when developing scenarios when assessing multifunctional systems. In doing so, the underlying worldview that drives the direction of future policies and legislation that might influence R&D and the background system can be included in the scenario and, thus, the LCA.

When comparing mature technologies to immature technologies, a useful approach is to expand the system boundaries to capture all functions. However, this does come with challenges such as cascading allocation due to mandated recycling of mature technologies. Another challenge is the potential loss of function for more complex immature technologies if there is an accident or unplanned event that would not affect simpler mature technologies in the same way. This highlights: *i)* the need for including both the cut-off and end-of-life allocation approaches as two extremes to avoid skewed results; and *ii)* the need to include the influence of accidents or unplanned events.

9. FUTURE RESEARCH

This thesis has identified methodological challenges related to prospective LCA. While some are addressed in this thesis and in the appended papers, some remain to be solved through more research. These challenges are both related to LCA methodology in general, but also more specifically for future LCAs of carbon fibre composites.

As described in this thesis, it proved difficult to assess carbon fibre composites with dual functions from a life cycle perspective. This problem will likely become more prominent in general as complexity of products increase. The challenge mainly stems from expanding the system boundaries to capture all the functions provided. While Paper V shows how this is possible in a cradle-to-grave assessment, it introduced uncertainties as to what products to include in the expansion as well as cascading allocation issues. The approach in paper V also hinders comparative cradle-to-gate studies of complex products, which I also see a need for. Comparative studies on cradle-to-gate basis would aid developers in the early stages of screening technology when details on the intended application of the product is uncertain. However, this brings the challenge of identifying the function of the cradle-to-gate system. In other words, is it, say, providing mechanical integrity, energy storage or both? More research is needed on how to define a meaningful functional unit in such cases, alternatively if and how the flows should be allocated between the multiple functions. An additional challenge for comparative prospective LCAs of products with different complexity levels is when an accident happens. An example that is discussed in the thesis is when a car crashes, that would not change the functions of one product but could significantly change the functions of the other. Replacement materials might be needed in the latter case, leading to a higher environmental impact. While Paper V introduces a simple calculation exercise for how these types of events can be included in LCAs, more research is needed to establish guidelines for when and how accidents should be included in LCA. In line with this, there is also a need for assessing the possible toxicity of damaged carbon fibre composites and SBCs, and what type of accident that would generate what type of emission (for example a crash or fire). Partly because damaged carbon fibres are known to behave like asbestos (Hertzberg, 2005), but also due to the possible toxicity of the matrix, and especially the SBC electrolyte.

A significant problem when assessing the environmental impact of carbon fibre composites in the development stages as early as done in this thesis, is the need for more data, especially for carbon fibre production³. To generate more accurate LCA results, more efforts should also be put into generating open-access datasets for production of carbon fibres with different intended use, as the temperature required in the heat treatment depends on the desired properties of the carbon fibres (Johansen, 2023). Furthermore, it is essential to create a specific life cycle inventory for carbon fibres to be used in SBCs for future research on the environmental impacts of SBCs. Future research on SBCs also includes expanding the LCA done in Paper V to include toxicity, as the production of the SBC includes the use of many (some nasty) chemicals. This does however require development of appropriate characterization factors. More open-source data is also needed that describes the recycling of the composites and recovery of fibres. This has proved the most promising route for reducing the environmental impacts of carbon fibre composites in the near future. However, large scale, primary data for the recycling processes is lacking, which means that there is a risk of generating skewed results. This also includes generating accurate data on the quality of the recovered materials, as Paper III showed that this was essential for the outcome for the recycling allocation approaches. In line with this, efforts should also be made to understanding to what extent fibres may be recovered from SBCs for use in other applications, especially regarding the lithiation.

³ The lack of data for carbon fibre production, plus variances in the environmental impacts of the few different datasets available were discussed intensively in the “Life Cycle Assessment for Composite Materials” session at the *International Conference on Composite Materials* in Belfast, Northern Ireland, 2023.

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