Master's thesis

NTNU
Norwegian University of Science and Technology
Faculty of Engineering
Department of Energy and Process Engineering

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Life cycle assessment of hard carbon production in Norway using biomass (sawdust) as precursor

Master's thesis in Sustainable Energy
Supervisor: Professor Steven Tyler Boles
Co-supervisor: Dr. Long Lin
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Project Thesis

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Acknowledgment

This article is dedicated to a thesis study at Norwegian university of science and technology (NTNU). The study program is ”Sustainable energy in buildings” which is a two year international study program in Energy and Process Engineering (EPT) department of NTNU. The subject of this thesis study is defined by WAI and Byonder companies and the thesis position was given to the author of this study.

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Mojtaba Firoozkoohi

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Abstract

Hard carbon production is getting more attention in the recent years due to its value in alkali-ion batteries. Its beneficial features in sodium-ion batteries enables the battery manufacturing to be more sustainable and cheaper compared to current Lithium-ion battery technology which has some limitations regarding the scarcity of lithium and the geographical concerns of Lithium sources in the earth crust.

However, the hard carbon production is in its starting point compared to graphite as a dominantly used anode material in the current state-of-the-art technology. There are many aspects to be improved, and investigated, regarding the utilization of hard carbon as an alternative. The environmental impacts of producing hard carbon is one the several aspects that is the focus of this study.

A literature study is done in this thesis work that contributes to creating a foundation for further analysis. The literature review provides basic conclusions about the structure of hard carbon and the underlying influence it has on the electrochemical performance. It is shown in the literature review that there is evidence that the structure of the hard carbon precursors have a large impact on the final structure of the hard carbon. Furthermore, life cycle assessment is introduced as the main tool for environmental impacts analysis. ReCiPe 2016 is used as the method for analyzing the impacts. Four main midpoint impact categories are deeply investigated, namely, global warming potential, acidification potential, ozone depletion potential, and eutrophication potential. The manufacturing model is implemented in SimaPro software which is a professional life cycle assessment software.

The manufacturing model for hard carbon production from sawdust consists of four main processes. These processes are carbonization, acid washing with drying, heat treatment (pyrolysis), and milling. These processes have sub-processes and more focused information is provided in the method section.

Several different scenarios for the hard carbon production are investigated. It has been shown that using sawdust as a precursor for hard carbon production has the lowest global warming potential impacts.

Regarding the manufacturing processes, it is shown that the highest global warming potential impacts are attributed to acid washing process. Also, it is evident that the excessive heat production in carbonization process reduces the total environmental impacts and its impacts are negative. Heat treatment process has the highest impacts after acid washing process due to its energy use and high temperature requirement.

Sammendrag

Hard karbonproduksjon har fått mer oppmerksomhet de siste årene på grunn av verdien i alkali-ion-batterier. Dens fordelaktige egenskaper i natriumion-batterier gjør det mulig for bat- produksjonen for å være mer bærekraftig og billigere sammenlignet med dagens Lithium-ion batteriteknologi som har noen begrensninger når det gjelder knapphet på litium og geografiske bekymringer for litiumkilder i jordskorpen.

Den harde karbonproduksjonen er imidlertid i sitt utgangspunkt sammenlignet med grafitt as et dominerende brukt anodemateriale i dagens toppmoderne teknologi. Det er mange aspekter som skal forbedres og undersøkes når det gjelder utnyttelse av hardt karbon som et alternativ. Miljøpåvirkningene ved å produsere hardt karbon er én av flere aspekter som er fokus i denne studien.

Det gjøres en litteraturstudie i dette oppgavearbeidet som bidrar til å skape et grunnlag for videre analyse. Litteraturgjennomgangen gir grunnleggende konklusjoner om strukturen til hardt karbon og den underliggende påvirkningen det har på den elektrokjemiske ytelsen. Det er vist i litteraturgjennomgangen at det er bevis for at strukturen til det harde karbonet forløpere har stor innvirkning på den endelige strukturen til det harde karbonet. Dessuten, livssyklusvurdering er introdusert som hovedverktøy for miljøkonsekvensanalyse. ReCiPe 2016 brukes som metode for å analysere påvirkningene. Fire hovedmidtpunkt påvirkningskategorier er dypt undersøkt, nemlig global oppvarmingspotensial, forsuring potensial, ozonnedbrytningspotensial og eutrofieringspotensial. Produksjonen modellen er implementert i SimaPro programvare som er en profesjonell livssyklusvurdering programvare.

Produksjonsmodellen for hardkarbonproduksjon fra sagflis består av fire hovedtyper prosesser. Disse prosessene er karbonisering, syrevasking med tørking, varmebehandling (pyrolyse), og fresing. Disse prosessene har delprosesser og mer fokusert informasjon er gitt i metodedelen.

Flere ulike scenarier for den harde karbonproduksjonen undersøkes. Det har vært vist at bruk av sagflis som en forløper for produksjon av hardt karbon har den laveste globale potensielle oppvarmingseffekter.

Når det gjelder produksjonsprosessene, er det vist at den høyeste globale oppvarmingspo-foreløpige påvirkninger tilskrives syrevaskeprosessen. Dessuten er det tydelig at overdreven varmeproduksjon i karboniseringsprosessen reduserer den totale miljøpåvirkningen og dens virkningene er negative. Varmebehandlingsprosessen har størst effekt etter syrevask prosess på grunn av sin energibruk og høye temperaturkrav.

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Abbreviations

CH_4 Methane.

CO_2 Carbon dioxide.

NH_x Reduced nitrogen.

NO_x Nitrogen oxides.

N_2O Nitrous oxide.

SO_2 Sulfur dioxide.

${PO_4}^{-3}$ Phosphate.

AP Acidification Potential.

B Boron.

C Carbon.

CFC chlorofluorocarbons.

EES Electrochemical Energy Storage.

GHG Green House Gas.

GWP Global Warming Potential.

ICE Initial Coulombic Efficiency.

IOA Input output analysis.

K Potassium.

LCA Life Cycle Assessment.

LCI Life Cycle Inventory.

LCIA Life Cycle Inventory Assessment.

Li Lithium.

LIB Lithium Ion Battery.

N Nitrogen.

Na Sodium.

NDC Nationally Determined Contributions.

O Oxygen.

ODS Ozone-depleting Substances.

P Phosphorus.

PBA Process based analysis.

pH Potential of hydrogen.

PIB Potassium Ion Battery.

S Sulfur.

SEI Solid Electrolyte Interface.

SIB Sodium Ion Battery.

SSA Specific Surface Area.

UV Ultraviolet.

1. Introduction

1.1. Background

The Paris agreement was adopted by 196 parties in December 2015 to mitigate the climate change crisis. This was the first time that many countries were unified to legally obligate themselves to reduces their greenhouse gas emissions. The Paris agreement goal was to reduce the Green House Gas (GHG) emissions to a point where the global temperature increase of less than $2°C$ is achieved [1]. However, it has been stated that even more ambitious goals should be pursued in order to prevent dramatic environmental hazards by the end of this decade. Since 2020, countries have submitted their plans and strategies as their Nationally Determined Contributions (NDCs) to the paris agreement goals. In order to effectively persue the $1.5°C$ increase limit, it is requested from countries to update their NDCs and target by 2023 [1, 2].

Consequently, many efforts have been made and probably continue in the future in order to protect the environment. For instance, the electrification of industrial processes and incorporation of sustainable energy sources such as wind and solar energy are some of the viable solutions for the environment protection. However, wind and solar energy are not a consistent source hence, not sufficiently reliable on their own. Therefore, electric energy storages (EESs) such as lithium-ion batteries (LIBs), sodium-ion batteries (SIBs), and potassium-ion batteries (PIBs) are necessary to store the renewable energy. LIBs have been the most successful until now, although the SIBs and PIBs are being investigated and have shown promising features and development potentials [3].

Furthermore, carbonaceous materials have been most widely used as precursors for production of anodes for the LIBs, SIBs and PIBs. Graphite has been shown to have favorable charcateristics for LIBs while hard carbon is most preferable for SIBs and PIBs. The production of hard carbon from biomass can be cheaper and more environmentally friendly than that of graphite. Additionally, production of SIBs and PIBs can be less expensive than LIBs due to the abundance of sodium and potassium in earth crust.

The hard carbon production from biomass is not commercialized yet, although it is important to simulate the manufacturing process prior to actual production to assess the environmental impacts associated with the production processes and find the possible improvement strategies.

1.2. Objectives

The main objective of this study is to perform a Life Cycle Assessment (LCA) on hard carbon production in Norway. The analysis goal is to find the related environmental impacts (mainly global warming) of hard carbon production from sawdust and find the highest contributors. The analysis results should be compared to previous LCA studies on hard carbon production from similar biomass precursors. The production of hard carbon from fossil-based precursors is also compared to the results of this study as a benchmark. Also, the probability of using alternative processes in the production to reduce the environmental impacts is also included in the objectives of this thesis.

1.3. Limitations

Several limitations are associated with this study which can be similar to most of the life cycle assessments that focus on a production scenario where the actual manufacturing process does not exist. Firstly, the production of hard carbon for sodium-ion batteries is relatively new compared to Li-ion battery technology. Therefore, available literature on the subject is also limited. Additionally, lack of existing technology for large-scale manufacturing is a limiting factor on the validation of the results.

1.4. Structure of study

This study consists of seven chapters starting with the following chapter where the background and objectives of the project are provided. The next chapter creates the fundamental theory around the topic of the study and is taken from the available literature and academic articles. Chapter 3 provides the details about conducting the life cycle inventory and impact assessment and the references and data sources are presented. The 4^{th} chapter is dedicated to the life cycle analysis generated outcome from the created Life Cycle inventory (LCI) and different impact categories values are provided. Chapter 5 focuses on the interpretation of the results and the comparison between the generated results between this study and other related LCA studies on hard carbon production. Chapter 6 summarizes the results and the interpretations of the thesis as the conclusion of this study. Lastly, further possible improvements and objectives are explained in chapter 7 followed by the references section at the end of this thesis.

2. Literature review

2.1. Introduction to Life Cycle Assessment (LCA)

According to ISO 14040 standard, environmental management is necessary for manufactured goods in order to reduce the potential environmental hazards associated with the products. Therefore, Life cycle assessment (LCA) tool has been developed during the recent years to assist achieving this goal. This method's goal is to quantify the total environmental impacts generated by a certain product or service throughout its lifetime. The lifetime of a product usually consists of extraction of raw materials, manufacturing, products use phase, and their end of life (cradle-to-grave LCA) as shown in Figure 2.1. In some cases, the use phase and end of life can be too extensive to study and therefore, not regarded in certain studies (cradle-to-gate LCA).

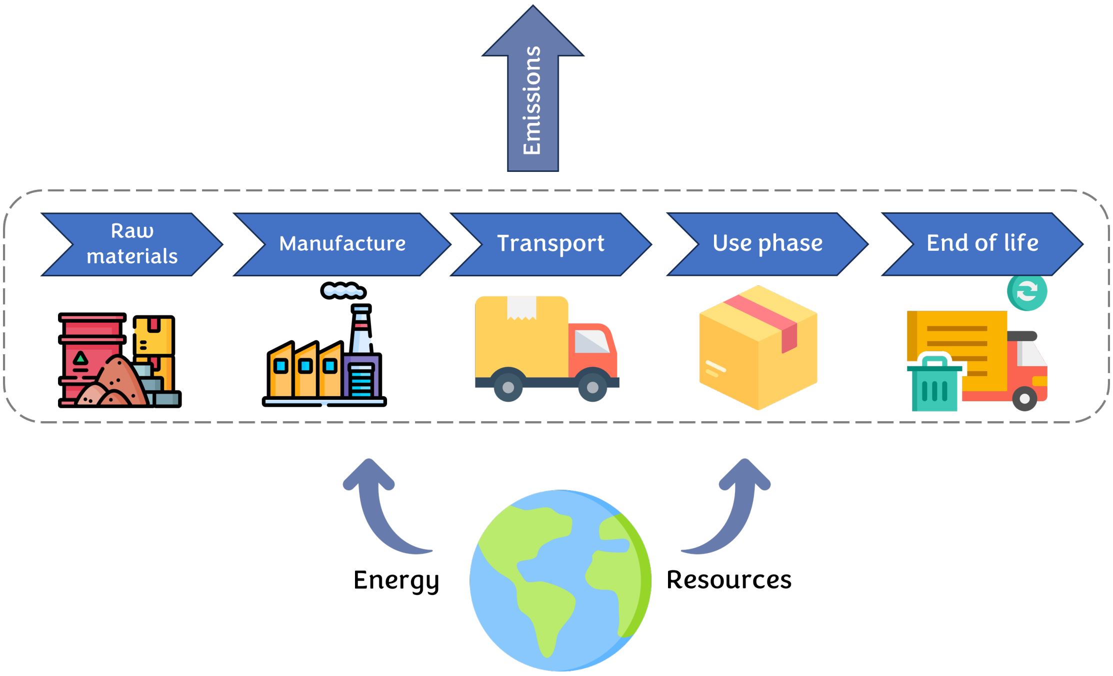

Figure 2.1: Demonstration of the life cycle of a product

Every LCA should consist of four stages, those being: Goal and scope, inventory modelling, impact assessment, and interpretation. These stages and the interconnections of information between them can be seen in Figure 2.2. The goal and scope determines the system boundaries and the functional unit of the model. The system boundary defines the main focus area of the LCA where the main manufacturing processes are, and the inputs and outputs to each individual processes are determined in details. The outer part of this area can be regarded as the background processes where their interconnections are not the main concern of the LCA study but they are represented as inputs from available libraries which contain extensive background data. The functional unit defines the quantity of the demand that an LCA expert wants to put on the system. For instance, considering milk production, the functional unit can be $1l$ of milk produced or $1gr$ of pro-

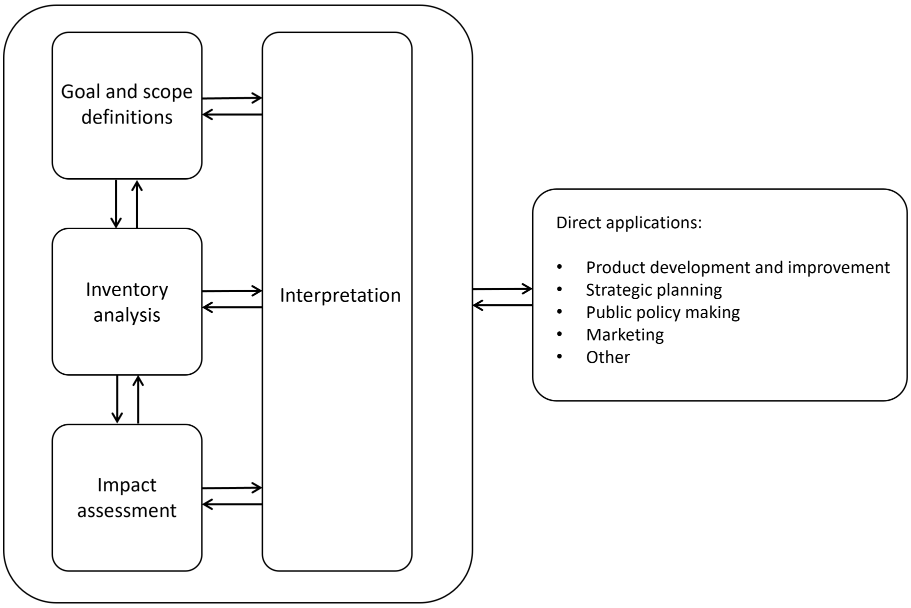

Figure 2.2: Schematic demonstration of LCA stages and information flow between them

tein contained by the milk. The functional unit shows how much environmental impacts are generated according to the demand of 1 functional unit from a certain product. Furthermore, inventory modelling is the most time-consuming stage of every LCA where the details of inputs and outputs to each manufacturing process and their respective emissions are determined. The 3rd stage, impact assessment considers the impact categories that are relevant to the study and defines the mid-point (Global warming potential (GWP)) and/or end-point (Human health) indicators that explain how the emissions impact the environment. Finally, the interpretation phase explains the information resulted from the previous stages in a proper way that accommodates the targeted audience whether its the public or manufacturers etc.. Overall, LCA aims to assist in reducing the environmental burdens, decision making in the industry and marketing for a product.

2.2. Life cycle impact assessment (LCIA)

As mentioned previously, after modelling the LCA inventory, the next step is to characterize the emissions resulted from the system. There can be an extensive list of emissions that are generated from the life cycle of a product. In order to clearly demonstrate the impacts of the emissions to the environment, there are characterization methods to reduce the list of emissions to a limited number of impact indicator scores [4]. These indicators can be midpoint or endpoint. Midpoint indicators such as GWP and Acidification Potential (AP) focus on single environmental impacts. However, the endpoint indicators focus on three upper aggregated levels of environmental burdens such as human health, biodiversity, and resource scarcity. The midpoint indicators are connected closely to the environmental stressors. Therefore, midpoint indicators suffer from less uncertainty compared to endpoint indicators. However, it is easier to interpret the LCIA results using the endpoint indicators [4, 5]. The reason can be that the endpoint indicators are related to protection areas that are more understandable for average public. This can be more difficult if an LCA practitioner tries to publish the results in midpoint indicators since it may require more academic background for the audience to understand some impact categories such as eutrophication, ozone depletion, etc., and their underlying influence on the environment.

2.2.1. ReCiPe method

The ReCiPe method provides harmonized characterization factors at midpoint and endpoint levels [5]. These factors consist of 18 midpoint and 3 endpoint indicators which can be seen in Figure 2.3. The figure shows an overview of the midpoint and endpoint indicators included in ReCiPe2016 methodology [6]. The characterization factors show the environmental impacts per unit stressors such as per kg of resource used or emissions generated. As can be seen in Figure 2.3, there are several different areas of protections and each midpoint impact category is attributed to several damage pathways. For better understanding, ozone depletion potential, eutrophication potential, global warming potential, and acidification potential is explain in the following sections section and these impact categories will be considered in the results section of this study.

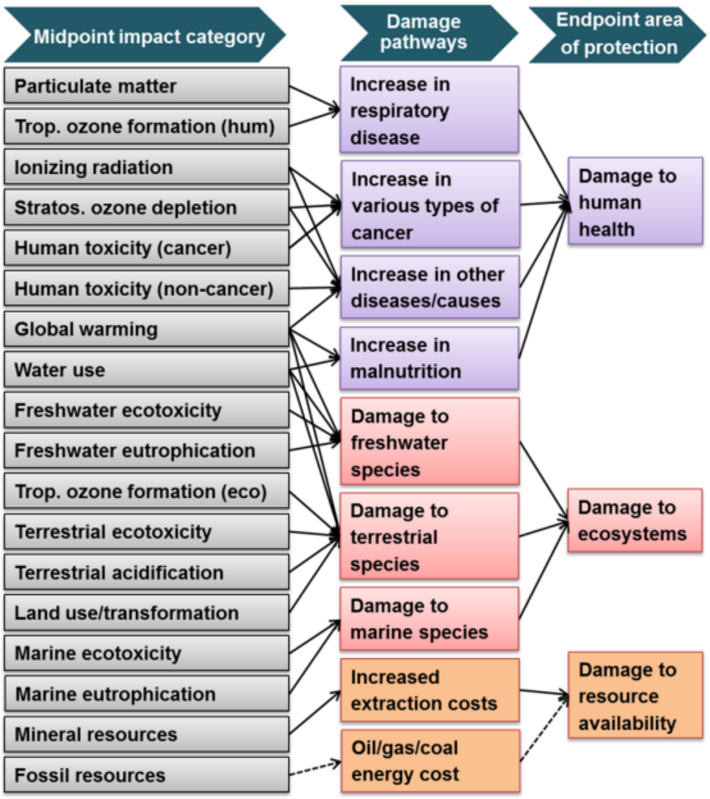

Figure 2.3: Midpoint and endpoint environmental impacts included in ReCiPe2016 method for Life cycle impact assessment

2.2.2. Global warming potential

As the name suggests, GWP considers the underlying effects of overall temperature increase of the earth. The reason has been mainly attributed to the overuse of fossil fuels during the last few decades. This global warming happens due to generation of greenhouse gases such as CO_2, CH_4, N_2O, water vapor, and CFCs. These chemical substances concentration can increase and subsequently trap the sun's radiation into the earth atmosphere. This can happen over and over and the overall temperature of the planet increases which has several adverse effects on the environment, flora and fauna, and the human health. This can lead disease and increased death from heat waves and drought in developing countries. Also, several areas can suffer from increased floods and many species will be on the edge of extinction [7]. The global warming potential impact is measured in CO_2 equivalents.

2.2.3. Acidification potential

The acidification potential is related to the possibility of increasing the concentration of acid contaminants that have hazardous effects on the environment. The main contributing materials that increase the acid concentration are SO_2, NO_x, and NH_x [8]. When the acid concentration increases, the pH of the environment decreases which can lead to deaths in fish ecosystems and decline in the coniferous forests. The acidification potential is measured as SO_2 equivalents.

2.2.4. Ozone depletion potential

The ozone layer is sensitive to chlorine and bromine substance in such way that these substances can deplete the ozone layer which protects the earth surface from ultraviolet beams of sun. Some chemicals such as chlorofluorocarbons (CFCs) consist of these substances and when they are in the stratosphere, they release chlorine and bromine due to being exposed to the ultra violet (UV) light. The ozone depletion effects on the amount of UV lights that penetrates the earth stratosphere and reaches the humans. This exposure can have detrimental effects on the human health with adverse effects on the human eye and skin [9]. The depletion effect of the ozone-depleting substances (ODSs) on the stratospheric ozone have been shown to have faster reactions than the recreation of ozone layer. Therefore, production of some of these materials such as CFCs have been banned in several countries including United states. The ozone depletion potentials are shown in CFC11 equivalents.

2.2.5. Eutrophication potential

Eutrophication is the process of overgrowth of algea in the coastal areas where nutrients are dischard into the water. These nutrients usually consist of nitrogen and phosphorus that is discharged to the water. Due to the lack of proper waste management, the amount of nutrients increase dramatically that leads to excessive growth of algea in the shallow waters and phytoplankton in deeper waters [10]. The problem that arise from this phenomenon is that light can no longer reach to deeper areas of the water which leads to eradication of sea grass and oxygen content. This can eventually have detrimental effects on the fish life. This can be especially important in coastal areas such as Norway and the effects should be mitigated by effective waste management. Other causes such as the global warming can also support the nutrient enrichment and lead to higher eutrophication potentials. Furthermore, eutrophication can also cause the increase in production of CO_2 due to large amount of organic matter decomposition [11]. Eutrophication potential impact is measured in ${PO_4}^{-3}$ equivalents.

2.3. Types of LCA models

There are two types of LCA methods namely: 1-Process based analysis (PBA) and 2-Input output analysis (IOA). PBA considers the flow of material and energy between the processes. Therefore, the relations between the processes are described using physical units such as mass, energy, volume, and etc.. On the other hand, IOA considers the monetary values flowing between sectors of economy. In this type of analysis, the data usually is gathered from national statistics which contains the transactions between different economy sectors. In comparison to PBA, IOA method is usually less detailed and it does not consider the use and end-of-life phase of the product. However, the information collection is easier in IOA compared to PBA method.

Another categorization for LCA methods based on the goal and scope of the project can be named as: Consequential modelling, and attributional modelling. In consequential modelling, the LCA tool is used to analyze a change into system processes and compare it with a baseline situation. In this scenario, based on the similarities between two processes an LCA practitioner can substitute a process by another and subtract the environmental impacts of the latter from the baseline situation to see if the total impacts change or stay the same. Moreover, attributional modelling happens when a process has two or more by-products where only some of them is the interest of the LCA study. Therefore, the impacts associated with each by-product must be attributed reasonably. For instance, the production of wooden planks in a sawmill process produces sawdust as a by-product. The amount of impacts attributed to sawdust can be based on its mass percentage out of the total mass of products. Alternatively, the attribution can be based on the monetary value of the sawdust and wooden planks. However, the ISO 14040 suggests that the monetary value attribution should be selected as the last option.

2.4. SimaPro

SimaPro is a professional software for life cycle analysis and it can be said that SimaPro is the most widely used tool by many LCA studies. This LCA tool was created in 1990 and currently is used by different academic institutions, consultancies, and companies in more than 80 countries. In comparison to other LCA tools, SimaPro can be more favorable since it is a subscriber to Ecoinvent database which is one the most comprehensive LCA databases available. Also, the software is based on the ISO 14040 recommendations. Additionally, SimaPro has several LCIA methods such as ReCiPe method which is introduced in previous sections. The software is easy to use and it provides many possibilities for further analyzing the results that are excellent for better interpretations.

2.5. Introduction to Hard carbon: Advantages and disadvantages

Carbonaceous materials such as graphite, hard carbon, soft carbon etc. have been widely used as promising anode materials for alkali metal ion batteries such as LIBs, SIBs and PIBs. Some of the advantages of these materials are their relatively low cost and abundance in the earth crust as well as their various structures and properties [12, 13]. Although graphite has been most commercially used anode in the current state-of-the-art LIB technology, it has some impractical issues such as low rate capability, low capacity at low temperatures and limited fast charging performance. Also, the accelerating rise of demand for LIBs and the increasing interest in electric vehicles has led to increase in Li demand and due to uneven distribution of Li other alternatives such as SIBs and PIBs are being investigated [13, 14]. On the other hand, hard carbon as anodes have shown superior cycling performance and higher rate capability compared to graphite [12].

Hard carbon is one of the promising alternatives to graphite, although it has not yet been sufficiently improved to commercially compete with graphite mainly due to high capacity loss in the first cycle [12]. Some interesting features of hard carbon compared to graphite can be better cycling ability, higher capacity at lower temperature and fast charging conditions. Additionally, hard carbon has been shown to be viable anode material in Sodium-ion and Potassium-ion batteries [13, 15]. This makes Na -ion and K -ion batteries promising alternatives to Li -ion batteries due to lower cost of Na and K alkali metals and their abundance in earth crust. In case of SIBs, it has been shown that the cost can be reduced by 18% according to [14]. Meanwhile, in order to understand these opportunities fundamentally, it is reasonable to dive into some key factors influencing the electrochemical performance of hard carbon such as structure, Li /Na storage mechanisms, hard carbon precursors, heteroatom doping and prelithiation/presodiation.

2.5.1. Hard carbon structure

Hard carbon is a carbonaceous material that cannot be graphitized even at $3000°C$. This means that, hard carbon precursors cannot change to an ordered crystalline structure such as graphite. The reason might be due to cross-linking C-O-C bonds formed during the pyrolysis of the precursors. Pyrolysis is the process in which the materials go through a thermal decomposition usually is done in an inert gas. Therefore, hard carbon microstructure can be described as disordered graphitic sheets consisting of micropores, defective sites and oxygen-containing functional groups [12, 13, 15]. The microstructure of hard carbon, soft carbon, and graphite are compared in Figure 2.4. As can be seen in the figure, the graphite structure is properly structured and has almost no defective sites. Soft carbon has defects to some extent although it has several graphitic zones. However, the hard carbon structure is much more disordered compared to the former two. This makes hard carbon unable to graphitize even in high temperatures. The disordered structure of hard carbon allows the Li $^+$ ions to store in different sites such as pores, defective sites and via intercalation into the graphitic domains. This can reduce the Li $^+$ transport distance which leads to better rate capability of hard carbon anodes. Also, hard carbons have shown higher capacity compared to graphite due to larger surface area [15]. However, the larger the surface area, the higher portion of Li $^+$ ions react with the

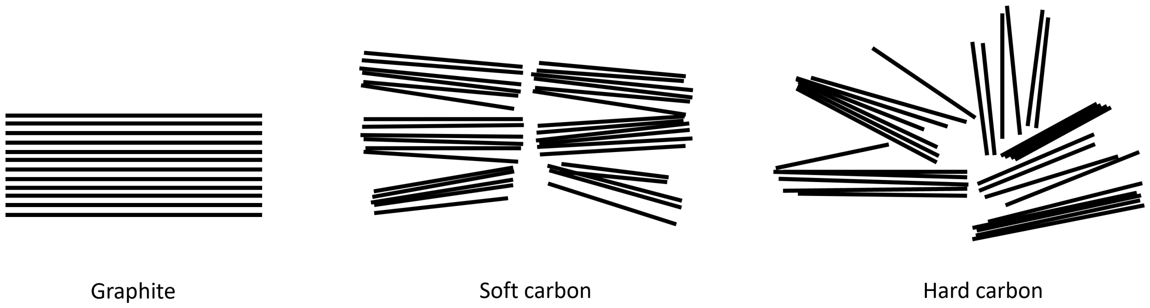

Figure 2.4: The schematic representation of the microstructure in graphite, soft carbon, and hard carbon

electrolyte and form the solid electrolyte interface (SEI) layer which leads to irreversible loss of capacity in the first cycle. Therefore, current hard carbons suffer from low initial coulombic efficiency (ICE) [16].

The structure of hard carbons are heavily dependent on the precursors and the pyrolysis conditions. There are a wide range of precursors for hard carbon which can be mainly categorized into three groups i.e. i) Resin-based, ii) Pitch-based, and iii) Biomass-based [12]. The variety of the precursors creates many possibilities for improvements in hard carbon electrochemical performance but at the same time, makes it difficult to find a generalized method for manufacturing hard carbon anodes with optimal performance. Each precursor has characteristics that demand different manufacturing conditions to reach the peak performance. Resin-based precursors found to be the most expensive alternatives, although they have shown the best electrochemical performance. The reason is the controllable structure of these materials in terms of pore characteristics, and active sites [12]. The pitch-based precursors are cheap and posses high quality among the hard carbon precursors. However, during the pyrolysis process these materials tend to create structures similar to graphite [12]. Finally, biomass-based precursors such as cellulose, lignin, and starch can be promising alternatives due to their abundance, availability and sustainability. However, the diversity of biomass-based precursors demands large amount of research since the performance of the hard carbons is directly affected by the precursor structure [12, 15].

2.5.2. Storage mechanisms in LIB and SIB

The storage mechanism of Li and Na ions into the hard carbon structure is crucial to be understood in order to find underlying methods that can improve the hard carbon electrochemical performance. Overall, there are three main storage mechanisms namely, intercalation, adsorption and pore-filling mechanisms [13, 15]. In the following, the differences between LIB and SIB storage are provided based on the literature.

It has been shown that the voltage variation with respect to the capacity consists of two regions [12]. Regarding LIBs, it has been proved that the sloping region is related to the adsorption of Li ions into the defective sites where the Li ions are placed on the abundant surface of hard carbon microstructure [13]. Afterwards, the intercalation of Li ions into the graphitic sheets happens which corresponds to the low voltage plateau region of the voltage vs capacity diagram.

As for the Na ions, it should be noted that the atomic radius is larger compared to Li ions. This means that the intercalation is limited due to the small layer distance. Also, the intercalation of the Na ions is shown to be thermodynamically unstable [13]. In comparison to Li ion storage mechanism, it has been indicated that the Na ions adsorption happens during the sloping region and the plateau can be associated with the intercalation of Na ions into the nanographitic domains.

2.6. Hard carbon manufacturing

Hard carbon fabrication mainly consists of preparation of precursors, and a two-step pyrolysis [17, 18]. The preparation of the precursors depends on whether the raw material is an origin biomass or a by-product from other industries such as agriculture or forestry. The focus of this study is on the by-product biomass (sawdust) since the use of origin raw materials make the hard carbon manufacturing more complex and expensive. Therefore, it is assumed that the sawdust is bought as a by-product from another industry. The by-products go through drying, acid washing and low temperature pyrolysis (300-900°C) which creates a carbonized material called biochar [16]. It is also possible to produce biochar by hydrothermal carbonization where the carbon content of the precursors increase by chemical reaction at elevated temperature and pressure [15, 19]. Acid washing process is performed on the raw materials to reduce the portion of inorganic substances in the precursors. Afterwards, during another pyrolysis with high temperature (up to 1400°C[16]) hard carbon with high carbon content is created. The structure of the created hard carbon is directly linked to the structure of the precursors. Therefore it is beneficial to understand the main aspects about the structure of biomass precursors which is provided in the next section.

2.6.1. Impact of precursors-biomass

The biomass structure largely impacts the final structure of the hard carbon hence, its electrochemical performance [13, 16, 18]. Additionally, there is a wide range of biomass precursors which differ in terms of their structure, even between the same species in different areas [16]. This leads to large variation in electrochemical performance even with the same carbonization procedure. Also, the inorganic content of the precursors are influential on the final hard carbon performance. The inorganic portion leads to higher distance between the garphene layers, and higher surface area, and consequently, lower ICE [16]. However, the main contents of biomass can be lignin, cellulose and hemicellulose [12].

2.6.2. Impact of pyrolysis

The pyrolysis temperature has been shown to have large impact on the hard carbon structure. It is indicated that the increase in the pyrolysis temperature leads to decrease in the number of defective sites, larger graphitic domain, and decrease in the pore volume. Overall, the higher carbonization temperature creates more ordered structure [16]. Moreover, it has been shown that the graphene layer spacing becomes smaller with higher pyrolysis temperature. Also, the surface area of most biomass materials tends to converge to an acceptable range by increasing the temperature although at low temperatures the surface area can widely change between different precursors. Additionally, higher pyrolysis temperature improves the graphitic domains by making them larger. It has been shown by some studies that the pyrolysis temperature of $1200 - 1300°C$ might be better regarding the electrochemical performance [19, 20]. However, the higher temperature leads to lower yields [19].

2.6.3. key factors influencing the electrochemical performance

According to the information in previous sections, there are two important factors contributing to the performance of hard carbon, 1-The precursors structure, and 2- Carbonization temperature. These factors influence the final structure of hard carbon and consequently the performance is dependent on the hard carbon structure. The important structure parameters are the following:

d_{002}: The distance between the graphene layers where the Li or Na ions can intercalate

SSA: The specific surface area of the hard carbon

V: The nanopores volume

Some studies have shown that the d_{002} impacts the capacity and ICE of the hard carbon. The optimal range for d_{002} in regard to intercalation alkali metal ions can be 3.75Å-4Å [20]. The higher temperature of pyrolysis can lead to excessive reduction in d_{002} and the Na ions can no longer intercalate into the hard carbon structure [20]. The SSA values might be very dependent on the structure of the precursor although the increase of pyrolysis temperature leads to decrease in SSA [15]. The porosity of the hard carbon is also important. There are two type of pores that can be formed in the hard carbon structure, namely, open pores and closed pored. The accessibility to closed pores is limited for gas adsorption. The increase of temperature can increase the open pore size and decrease the closed pore size at high pyrolysis temperatures [15].

2.6.4. Measures to improve electrochemical performance of hard carbon

Several methods can be implemented in structural design of hard carbons to improve their performance. For instance, heteroatom doping is where heteroatoms such as B, N, O, S, and P are added to the hard carbon structure. This method can increase the specific capacity of hard carbon [15]. The study suggests the reason can be due to improvements of electronic conductivity, defect concentration and porosity of the final product. Also, some studies suggest that microwave activation where the hard carbon is subject to microwave radiation can improve the structure by creating open pores [20, 21]. Moreover, presodiation of the hard carbon can improve the ICE since it can recover the relatively high amount of sodium ions that are lost during the first cycle [13]. Another method for improving the hard carbon structure is the exfoliation of 2-D materials that can provide short Na diffusion paths that improves high rate capability of the product [20].

3. Method

3.1. Introduction

In this chapter, the methodology of the study consisting of LCA mathematical procedure, LCA inventory building, and goal and scope of the project is presented. It is important to point out that the method utilized in this study is a cradle-to-gate LCA for the production of hard carbon from sawdust. The aim of this study is to investigate the environmental impacts of a large-scale hard carbon manufacturing in Norway.

3.2. LCA mathematics

In each production model that is the focus of an LCA study, there are interconnections of material and energy flow between each individual process. These interconnections should be defined mathematically as an start point for calculating the total impacts. These interconnections are modelled as a matrix which is named A matrix. The structure of A matrix is shown in the following equation:

$$A = \begin{bmatrix} Aff & Afb \\ Abf & Abb \end{bmatrix} \tag{3.1}$$

. The A matrix is the requirement matrix and shows the requirements of each process from other processes. This can be seen in Figure 3.1 as it shows the different sub parts in the A matrix. The processes numbered 1 to 6 are the foreground processes and their interconnections are shown. Moreover, we can see the background processes in Abb part that have both forward and backward flows to each other, despite the foreground processes. Usually, this is because different industry processes have requirements from each other. The middle group of flows shows the connection between background and foreground processes and usually there is no flow from foreground to background. The corresponding A matrix for the system shown in Figure 3.1 can be formulated as shown

Figure 3.1: The schematic representation of an LCA inventory and the flows between different processes. Different categories for each group of processes is defined on top and they are separated by dashed lines

in the following equation.

$$A = \begin{bmatrix} 0 & a12 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & a26 & 0 & 0 & 0 & 0 \\ 0 & a32 & 0 & 0 & 0 & a36 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & a46 & 0 & 0 & 0 & 0 \\ 0 & a52 & 0 & a54 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ a71 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & a79 & a710 \\ 0 & 0 & a83 & 0 & 0 & 0 & 0 & 0 & a89 & a810 \\ 0 & 0 & a93 & 0 & 0 & 0 & a97 & a98 & 0 & 0 \\ 0 & 0 & 0 & 0 & a105 & 0 & a107 & a108 & 0 & 0 \end{bmatrix} \tag{3.2}$$

To better explain the A matrix, a_{12} shows the required amount of process 1 to produce one unit of process 2. The sub parts in the A matrix, are related to the foreground and background processes of the model and their relations. For instance, Aff is the requirements of foreground processes from other foreground processes and Abf is the requirements of background processes into the foreground processes. Abb is related to the interconnections between background processes and usually is provided by the LCA libraries. Afb, is usually zero since in many cases there are no flows from the foreground to the background. This can be seen in the A matrix in Equation 3.2 in the top right corner with a 6x4 box of zeros. It basically means that background processes only provide for the foreground processes and do not require any flow from the foreground. Furthermore, another important characteristic which needs to be modelled mathematically is the emissions matrix which is named S matrix and is defined as follows:

$$S = \begin{bmatrix} Sf & Sb \end{bmatrix} \tag{3.3}$$

As shown in Equation 3.8 similar to A matrix, Sf is the direct emissions of the foreground processes and Sb emissions from background processes. Moreover, in order to calculate the impacts of the emissions of the system, there should be a characterization matrix that defines the severity of each emission with respect to every impact category. Another important information is the demand which is put on the system model and is usually is defined based on the functional unit. Therefore, a demand vector (y vector) is defined by how much is demanded from each process. Finally, finding the total impacts of the production starts with calculating the total production of each process which results in finding the x vector. This vector can be calculated as the following equation:

$$x = (I - A)^{-1}.y \tag{3.4}$$

where I is the Identity matrix, A is the requirement matrix, and y is the demand vector. Moreover, after finding the total outputs, the total emissions of the system can be calculated using the next equation:

$$e = S.x \tag{3.5}$$

The e vector shows the total emissions of whole system. Finally, the d vector shows the total impacts of the system which is calculated as follows:

$$d = C.e \tag{3.6}$$

It is also possible to find the contribution of each process in the emissions and the impacts in order to have deeper understanding of the system. This can be done by the following equations:

$$E = S.\hat{x} \tag{3.7}$$

$$D_{process} = C.E \tag{3.8}$$

The E matrix calculates the emissions of each process in different emission types. Subsequently, we can calculate the impacts of each process after multiplying E matrix with characterization matrix (C) and it results in $D_{process}$ matrix.

3.3. Goal and scope

The goal is to assume a large-scale hard carbon production system using the available information in the literature and supplementary information from Beyonder and WAI companies. The reason is the lack of a large-scale manufacturing of hard carbon from biomass at the moment. Moreover, this study tries to perform a comprehensive investigation of the sustainability of the modelled production system. The scope of the study is to model the production phase of sawdust-derived hard carbon and it excludes the preliminary processes of sawdust production from other industries. Therefore, the sawdust is considered as a market commodity that can be purchased. The relevant inventory data is taken from ecoinvent libarary database which is explained in more details in the following sections. The functional unit is set to be $1kg$ of hard carbon produced from sawdust. Additionally, the production system is compared to a commercial hard carbon production from fossil-based materials in order to provide a comparative analysis with respect to a reference system. Additionally, it should be noted that the large-scale production of hard carbon from sawdust is not commercially viable yet. Therefore, the production scenario in this study is an assumption incorporating related technologies that can be a probable candidate for a large-scale hard carbon manufacturing system. This is done based on the available information in the literature and experience from experts working in the industry.

3.4. Life cycle inventory (LCI)

The LCI modelled based on the literature is provided in this section. Firstly, the hard carbon from sawdust manufacturing processes are presented in detail. Next, the inventory modelled for production of hard carbon from fossil sources is provided for further comparative analysis.

3.4.1. Biomass-based hard carbon production model

The modelled large-scale manufacturing of sawdust-based hard carbon is shown in Figure 3.2. The boundary of the foreground system can be seen in Figure 3.2 and inputs and outputs of the system are shown as well. The production starts off with the transportation of sawdust to the factory. Subsequently, the dry sawdust goes through a carbonization process where the carbon content of the biomass increases in elevated temperatures (300 $°C$-600 $°C$). The process utilises internal heat as its source and the sawdust is carbonized under low-oxygen-content atmosphere. The excess heat in this process can be used in the subsequent processes and the remaining energy can be sold to industrial district heating. The biocarbon generated from this process is cooled and the flue gas is treated by addition sodium bicarbonate and the gases are ventilated to air. The cooled biocarbon is washed and rinsed with acid and deionized water and subsequently dried in a drying step. Acid washing of the biocarbon is done in order to reduce the inorganic content of the material since it can effectively improve the electrochemical performance of the final products. The solution of acid and water need to be neutralized. Therefore, the washed-off solution is neutralized with sodium hydroxide and can be discharged to the regular sewage system. Later on, the cleaned biocarbon is subject to a heat treatment (at 1000 $°C$-1400 $°C$) process where the hard carbon is created. Similar to carbonization process, some emissions are discharged to air due to generation of flue gas in the heat treatment process. Next, the hard carbon is milled after being cooled. Finally, the milled hard carbon is transported to the customer site.

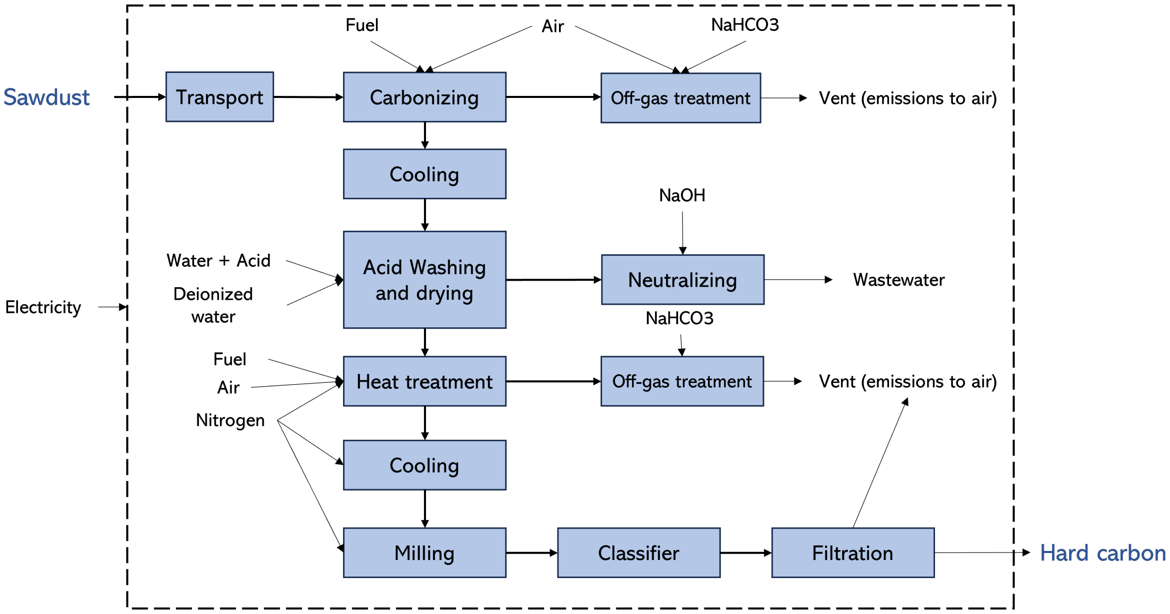

Figure 3.2: Hard carbon manufacturing processes

3.4.2. fossil-based hard carbon production model

It is also possible to hard carbon from fossil based fuels such as petroleum coke [22]. The process is modelled based on the work of peters et al [22] where petroleum coke is subject to a pyrolysis process in a rotary kiln. However, the production yield of the pyrolysis process is higher than that of biomass-based hard carbon due to higher carbon content of petroleum in comparison to biomass [22]. The life cycle inventory modelled in SimaPro software is shown in Table 3.1 which is the same as that of Peters et al [22] study supplementary information.

Table 3.1: Life cycle inventory of hard carbon production from petroleum coke

Dataset	Value	Unit
Inputs		
Petroleum coke GLO— market for petroleum coke	1.14E+00	kg
Water, deionised EU market for water, deionised	1.78E-01	kg
Nitrogen, liquid RER— market for nitrogen, liquid	9.00E-01	kg
Transport, freight, lorry 16-32 metric ton, EURO5 RER	1.59E-01	tkm
Transport, freight train EU	7.77E-01	tkm
Heat, central or small-scale, natural gas EU	1.89E+00	MJ
Electricity, medium voltage EU	1.83E-02	kWh
Outputs		
Hard carbon	1.00E+00	kg
Emissions		
Heat, waste	1.95E+00	MJ
Carbon dioxide, fossil	8.39E-02	kg
Nitrogen monoxide	4.66E-02	kg
Nitrogen dioxide	3.76E-03	kg
Sulfur dioxide	5.03E-02	kg

3.5. Data collection

The LCI modelling requires intensive information about the inputs and outputs of assumed production processes. These inputs and outputs are categorized as materials and energy flows. The life cycle inventory of different process are provided in this section. These datasets are mostly taken from the ecoinvent database and some inventory values are exclusively created for this study.

3.5.1. Carbonization

The details of inputs and outputs for production of carbonized biomass can be seen in Table . The data values are mostly taken from reported values from WAI company. In order to produce biocarbon in the plant materials such as woody biomass, diesel, and sodium bicarbonate for flue gas treatment are needed. The heat input for this process is negligible since the process uses the internal heat of the biomass throughout the process. The outputs of the process by percentage are 23% biocarbon, 77% syngas which is partially used in the carbonization process and the remaining is either used in the subsequent processes or sold as heat. In order for the syngas to fully burn for heating purposes, 9.8 kg air per kg of hot syngas is needed to provide enough oxygen for burning reaction. The flue gas emissions are also provided in the life cycle inventory as shown in Table 3.2. The carbonization process takes place at a temperature of 600°C. The residence time of the feed materials is suggested to be 30 to 60 minutes.

Table 3.2: Life cycle inventory of carbonization process in hard carbon manufacturing model

Dataset	Value	Unit
Inputs		
Woody biomass (sawdust)	4.35E+00	kg
Diesel EU— market for diesel	2.18E-03	kg
Sodium bicarbonate GLO	8.70E-03	kg
Electricity, medium voltage NO	8.70E-01	kWh
Outputs		
Biocarbon	1.00E+00	kg
Heat, district or industrial, other than natural gas EU	3.55E+01	MJ
Emissions		
Wood (dust)	2.18E-04	kg
TOC, Total Organic Carbon	2.18E-04	kg
Hydrochloric acid	2.18E-04	kg
Hydrogen fluoride	4.35E-05	kg
Sulfur dioxide	1.13E-03	kg
Nitrogen oxides	4.57E-03	kg
Carbon dioxide, biogenic	4.71E+00	kg

3.5.2. Acid washing and drying

Acid washing process inventory is provided in Table 3.3. The process yield is 90%. The process requires deionized water and acid for the washing and rinsing process. The reaction is assumed to take place in a continuous reactor where 30% concentration hydrochloric acid and biocarbon mix and the temperature is controlled to stay at $60 - 80°C$. Heat requirement for this process consists of 114.78 Wh per kg of Acid and biocarbon plus 714.5 Wh per kg of H_2O evaporated [23]. The heat required for this process can be provided by the heat generated from carbonization process. The heating requirements can be seen in the inventory processes in Table 3.3. Also, in order to account for the electricity supply to the industrial equipment 0.2 kWh per kg of biocarbon is considered for this process. Additionally, sodium hydroxide is added to the washed acid as shown in the inventory as the neutralizing agent. The drying heating requirements are also subtracted from the heat provided by carbonization process.

Table 3.3: Life cycle inventory of acid washing and drying processes in the hard carbon manufacturing model

Dataset	Value	Unit
Inputs		
Biocarbon	1.12E+00	kg
Hydrochloric acid, without water, in 30% solution state RER	2.31E-01	kg
Water, deionised EU	2.79E+01	kg
Neutralising agent, sodium hydroxide-equivalent GLO	2.54E-01	kg
Electricity, medium voltage NO	2.23E-01	kWh
Heating for acid reaction	2.03E-01	kWh
Heating for drying	7.97E-01	kWh
Outputs		
Washed biocarbon (dried)	1.00E+00	kg
Emissions		
Waste water, to water	2.24E+01	kg

3.5.3. Heat treatment

In this section, the life cycle inventory of heat treatment process is provided as shown in Table 3.4. This process yield is assumed to be 85%. The heating need of this process is assumed to be the same as that of Liu et al [19] where 0.8396 kWh of heat is needed for producing 1 kg of heat treated hard carbon. The pyrolysis process takes place at a temperature of $1300°C$. Furthermore, same as previous section, the process heating need is considered to be covered by heat generation of carbonization process. Meanwhile, electricity need for running the systems is considered in the inventory. Also, due to production of syngas (5%wt of washed biocarbon), flue gas treatment is needed and the same emissions as the carbonization process should be considered in the inventory.

Table 3.4: Life cycle inventory of heat treatment process in the hard carbon manufacturing model

Dataset	Value	Unit
Inputs		
Washed biocarbon	1.18E+00	kg
Nitrogen, liquid RER— market for nitrogen, liquid	2.11E-01	kg
Sodium bicarbonate GLO— market for sodium bicarbonate	1.53E-04	kg
Electricity, medium voltage NO	2.36E-01	kWh
Heat requirement for pyrolysis	8.40E-01	kWh
Outputs		
Hard carbon (not milled)	1.00E+00	kg
Emissions		
Wood (dust)	3.83E-06	kg
TOC, Total Organic Carbon	3.83E-06	kg
Hydrochloric acid	3.83E-06	kg
Hydrogen fluoride	7.66E-07	kg
Sulfur dioxide	1.99E-05	kg
Nitrogen oxides	8.04E-05	kg
Carbon dioxide, biogenic	8.30E-02	kg

3.5.4. Milling

The milling process inventory is shown in Table 3.5. The milling equipment should be specialized for the hard carbon material to prevent increase of impurities in the hard carbon. It is assumed that the milling process has to take place in inert atmosphere and nitrogen gas is the selected gas for this process. The electricity and nitrogen requirements for the milling process are based on Hjaila et al [23]. It is assumed that the milling process for the activated carbon is similar to that of hard carbon.

Table 3.5: Life cycle inventory of milling process as the last process in the hard carbon manufacturing model

Dataset	Value	Unit
Inputs		
Hard carbon (not milled)	1.02E+00	kg
Nitrogen, liquid RER— market for nitrogen, liquid	3.89E-03	kg
Electricity, medium voltage NO	2.47E-01	kWh
Outputs		
Hard carbon	1.00E+00	kg
Emissions		
Wood (dust)	2.04E-02	kg

4. Results

The current chapter provides the environmental impacts associated with the production models of hard carbon comprising two different scenarios for the precursors. The viability of the hard carbon manufactured with biomass precursor and its potentials can be evaluated by comparison between the two scenarios environmental impacts.

4.1. Hard carbon from biomass (sawdust)

Life cycle impacts associated with biomass-derived hard carbon can be provided in several impact categories although in this study, main focus is on categories such as climate change, acidification potential, eutrophication,and ozone depletion potential. Extensive attention is on climate change impact category due to its importance considering the environmental concerns of Paris agreement [1].

4.1.1. Climate change (GWP)

The global warming potential (GWP) of the four processes involved in the hard carbon manufacturing can be seen in Figure 4.1. As shown in the bar chart, the major contributing processes are carbonization and acid washing with 6.19 and 0.537 kg $CO_2 - eq$ respectively. The heat treatment process is the third contributing process with 0.135 kg $CO_2 - eq$ GWP impact per functional unit which is 1 kg of hard carbon. The milling process contribution is very low and is barely visible in the diagram. Therefore, it is acceptable to consider milling process impacts negligible. Also, there is a large negative value of -10.3 kg $CO_2 - eq$ (green bar) which is associated to the precursor provided by Bergene Holm AS report [24]. Therefore, the total GWP impacts of the system is -3.43 kg $CO_2 - eq$ as can be seen in Figure 4.1 in the top grey bar.

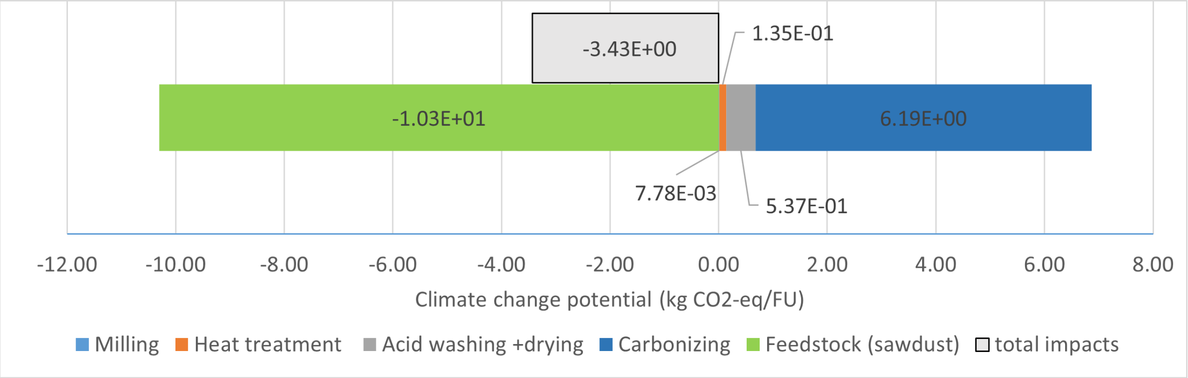

Figure 4.1: Climate change impacts associated with the foreground processes of hard carbon manufacturing from biomass

Furthermore, it is better to observe more deeply into the primary contributors. Figure

4.2 shows the climate change impacts of sub-processes of carbonization process and its direct emissions. As can be seen the figure, the highest impact stems from the direct CO_2 emission in carbonization process where the carbon-content biogenic material is burned and the flue gas contains CO_2. However, the excessive heat which is sold to district heating and the dry sawdust indicate -0.283 and -10.3 kg $CO_2 - eq$. The impacts from electricity, sodium bicarbonate, and diesel are 0.0328, 0.0147, and 0.00269 kg $CO_2 - eq$ respectively. Also, the impacts of transportation of materials to the carbonization site is 0.125 and 0.00791 kg $CO_2 - eq$ which are attributed to road and boat transport.

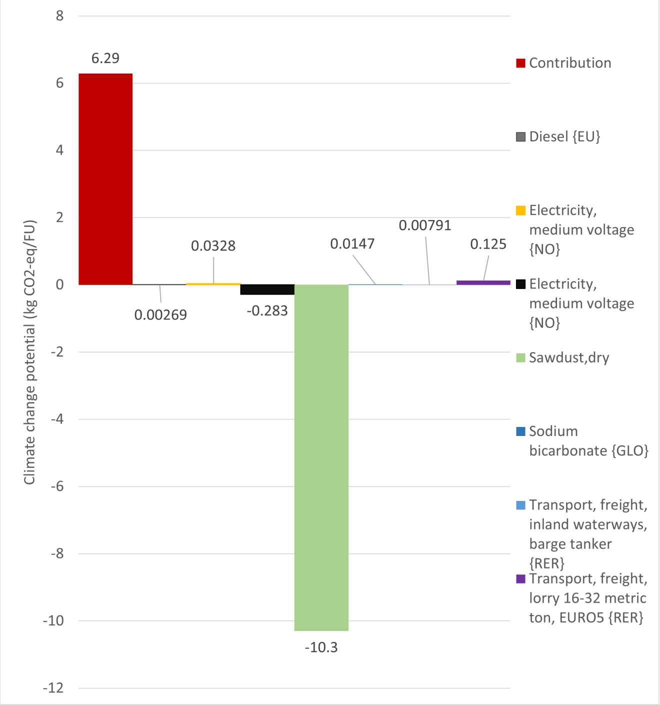

Figure 4.2: Climate change impacts associated with the carbonization process and the contriburtion of its sub-processes

Moreover, the impacts from acid washing process is shown in the bar chart in Figure 4.3. The highest impact is related to sodium hydroxide as the neutralizing agent with 0.37 kg $CO_2 - eq$ followed by that of hydrochloric acid with 0.144 kg $CO_2 - eq$. The GWP impacts associated with deionized water and electricity are relatively lower than that of NaOH and HCl with values of 0.015 and 0.00755 kg $CO_2 - eq$ respectively.

4.1.2. Other impact categories

The results of life cycle impact assessment (LCIA) in several impact categories linked to hard carbon manufacturing from biomass precursor is provided in this section. The impacts are presented separately due to difference in the order of values in each impact category. Firstly, the acidification potential impacts are presented in Figure 4.4. As can be seen in the figure, the total acidification potential impact of the system is $1.21E - 02$ kg SO_2 eq. The main contributors to acidification potential are carbonization and acid washing and drying processes indicating $9.23E-03$ and $2.62E-03$ kg SO_2 eq respectively. Despite the climate change impact of carbonization process, its contribution is positive

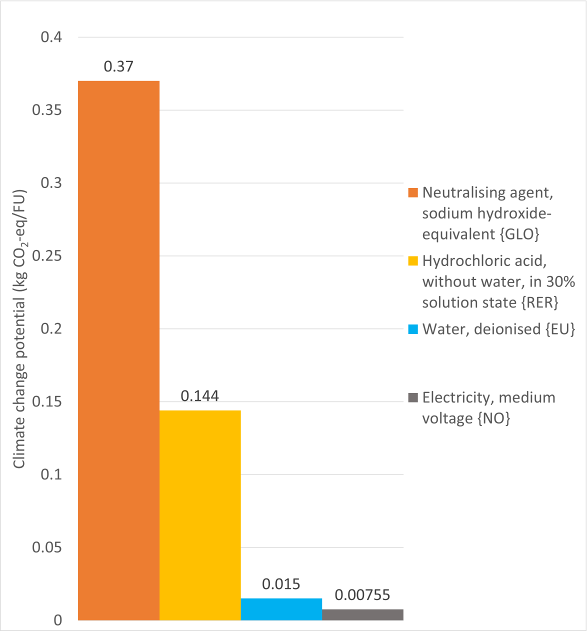

Figure 4.3: Climate change impacts associated with the acid washing process and the contribution of its sub-processes

in acidification potential followed by the heat treatment process with $2.18E-04$ kg SO_2 eq.

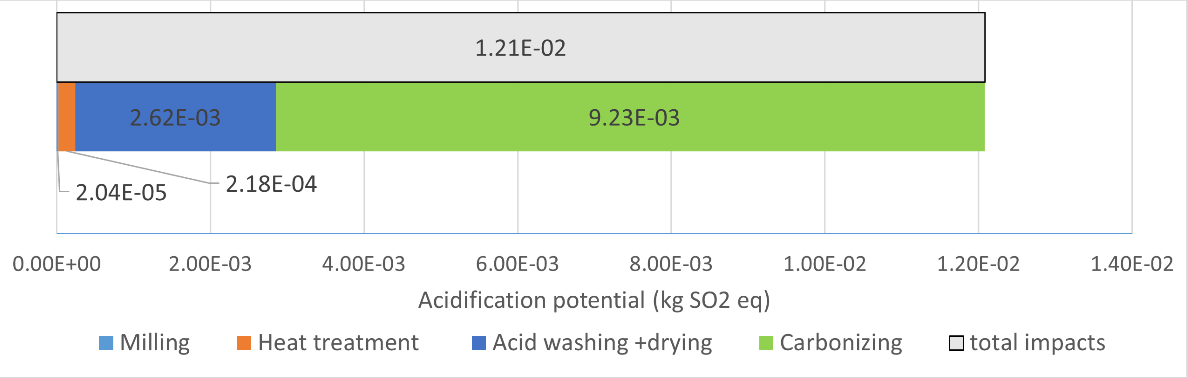

Figure 4.4: Acidification potential impacts linked to the foreground processes in hard carbon manufacturing from biomass precursor

Next, the ozone depletion impacts are presented in Figure 4.5. The highest values are linked to carbonization and acid washing processes with $-4.85E-07$ kg CFC11 eq and $5.81E-07$ kg CFC11 eq respectively. The negative impacts of carbonization process is due to the excess heat production. The other two processes are much less influential as can be seen in the bar chart. The total ozone depletion impact of the system is $1.52E-07$ kg CFC11 eq.

Finally, eutrophication potential impacts are presented in Figure 4.6. Similar to previous results, it can be seen that the acid washing process is again the most contributing process in eutrophication potential impacts followed by the heat treatment process with $2.77E-04$ and $4.16E-05$ kg ${PO_4}^{-3}eq$ respectively. The carbonization process excess electricity production gives credits to the total amount of impacts by $-8.65E-05$ kg ${PO_4}^{-3}eq$. The

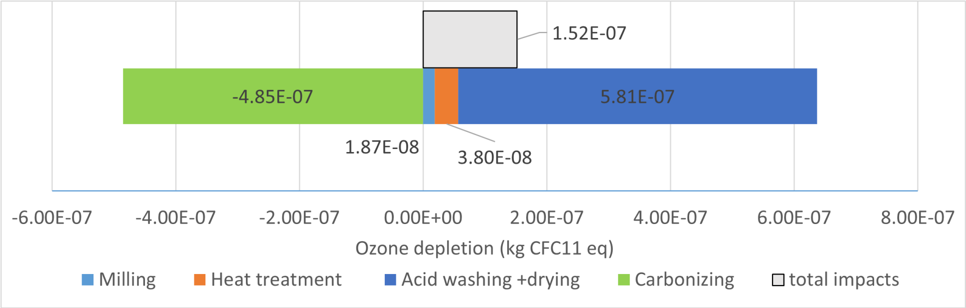

Figure 4.5: Ozone depletion impacts linked to the foreground processes in hard carbon manufacturing from biomass precursor

total eutrophication potential of the production model is $2.35E-04$ kg ${PO_4}^{-3}$ eq.

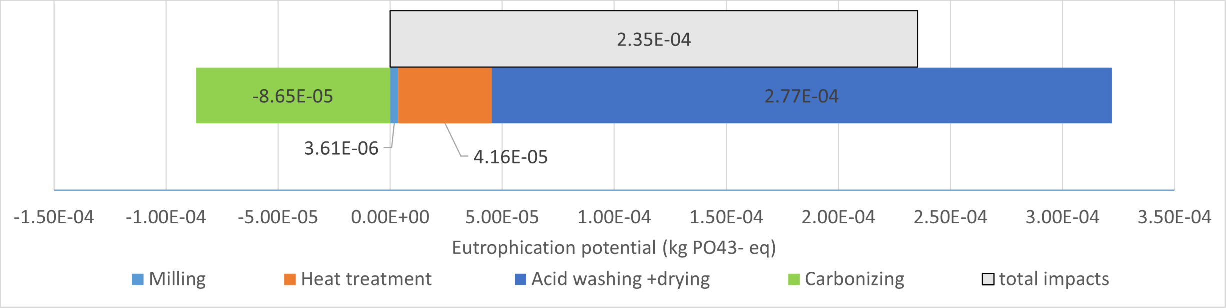

Figure 4.6: Eutrophication potential impacts linked to the foreground processes in hard carbon manufacturing from biomass precursor

4.2. Alternative scenarios

In order to make proper comparison, alternative scenarios have been modelled in the SimaPro software and the corresponding results are provided in the following sections. Firstly, the production model from Ecoinvent database sawdust is provided for having perspective to the previous generic model. Afterwards, other hard carbon sources and other studies results are provided for better comparison and further discussions in the next chapter.

4.2.1. Hard carbon modelled with Ecoinvent database precursor

In this section, GWP impacts of hard carbon production from predefined precursor by Ecoinvent is provided since majority of LCA studies use Ecoinvent as their database and this improves the comparison with other studies results. The GWP impacts of different processes are presented in Figure 4.7. As can be seen from the figure, highest impact stems from acid washing and drying process followed by the precursor (sawdust) with values of 0.537 and 0.359 kg CO_2-eq. Heat treatment process impact is relatively lower than mentioned processes and milling process contribution is lowest among other processes. Also, the credits from carbonization process due to the excessive heat production is shown as the green bar which has negative value of -0.131 kg $CO_2 - eq$. The total GWP impacts of this scenario is shown in the grey top bar with value of 0.908 kg $CO_2 - eq$.

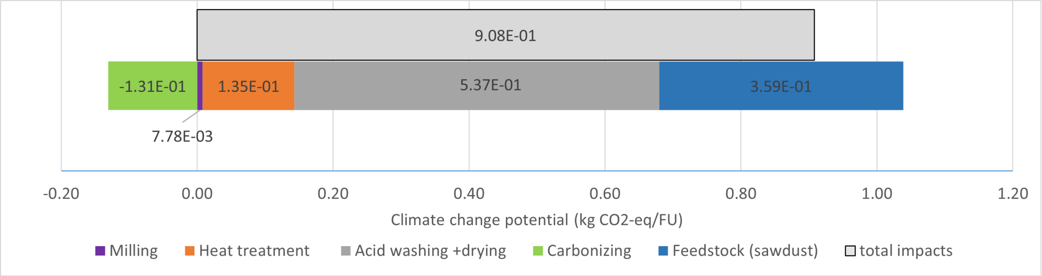

Figure 4.7: Climate change impacts of hard carbon production from Wood chips, dry, measured as dry mass RER as a predefined precursor in Ecoinvent database

4.2.2. Hard carbon from fossil source

The climate change potential of hard carbon manufacturing from fossil source is provided in this section for the sake of comparison to the base scenario. The results of this section are reproduced from the study of Peters et al [22] and the inventory model is exactly taken from their study. However, the provided results from this section is not provided in their study [22]. As can be seen in Figure 4.8. The figure shows that the highest impact is associated with the precursor with value of 0.77 kg $CO_2 - eq$ followed by nitrogen and heat from natural gas with GWP impacts of 0.185 and 0.149 kg $CO_2 - eq$.

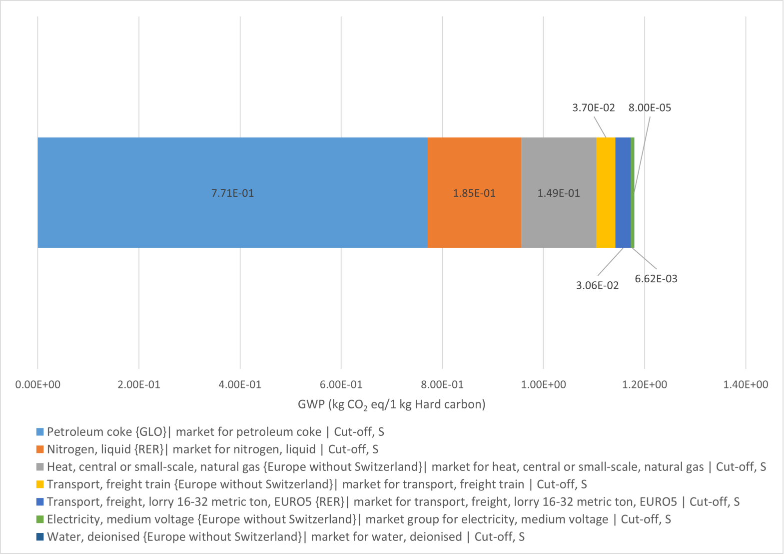

Figure 4.8: Climate change impacts of hard carbon production from petroleum coke, based on Peters et al LCA inventory data [22]

4.2.3. Hard carbon from sugar based source

In this section, the GWP impacts of sugar based hard carbon production is presented based on the study of Peters et al [22], same as previous section. As shown in Figure 4.9, the climate change potential of sugar based source has the highest impact which is 11.7 kg $CO_2 - eq$. Also, nitrogen and heat have large contributions of 1.43 and 0.748 kg $CO_2 - eq$ in comparison to other processes. The electricity impact is 0.0387 kg $CO_2 - eq$ which is important for comparison to the base scenario.

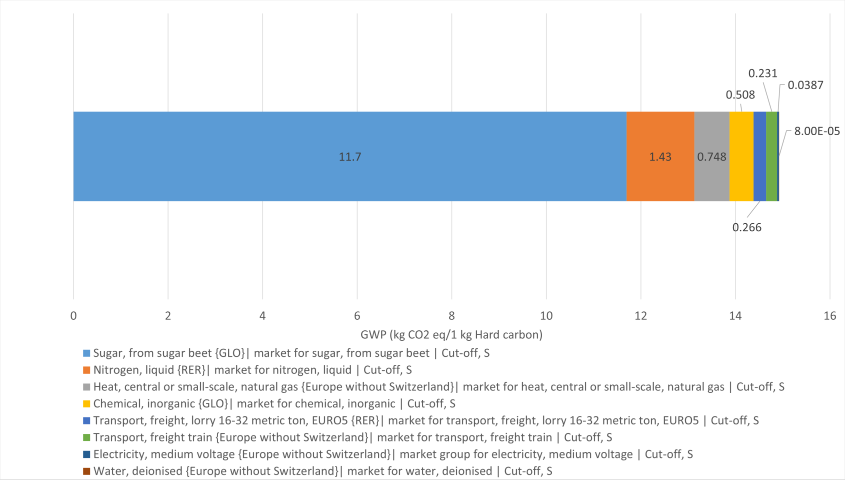

Figure 4.9: Climate change impacts of hard carbon production from sugar source, based on Peters et al [22]

4.2.4. Comparison of different scenarios

This section provides a summary of the total global warming potentials of previously mentioned hard carbon manufacturing scenarios. This can be seen in figure 4.10. The first blue bar on the left hand side shows the total GWP impacts of hard carbon manufacturing using sawdust dataset in Ecoinvent library which is associated with 0.908 kg CO_2 eq. Next, same manufacturing processes use sawdust dataset using values reported by Bergene Holm AS [24] which show a negative value of -3.43 kg CO_2 eq. Sugar based and petroleum coke hard carbons have shown 14.9 and 1.18 kg CO_2 eq. The last two bars show the results from Liu et al [19] with 4.07 and 4.61 kg CO_2 eq for two step hard carbon production and direct pyrolysis respectively.

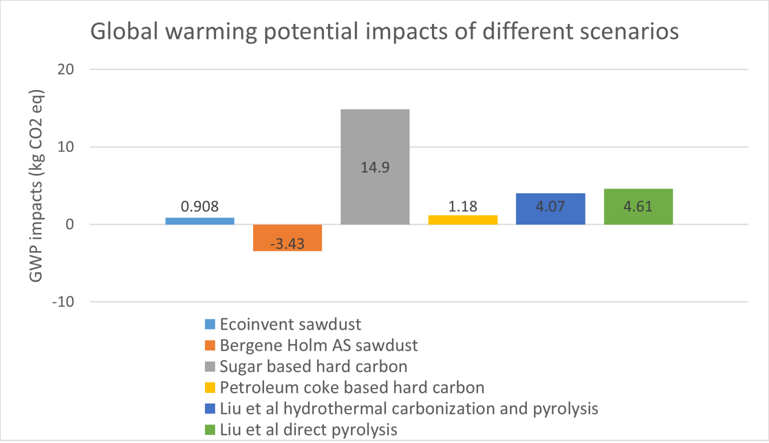

Figure 4.10: Comparison of different hard carbon manufacturing scenarios;

4.3. Sensitivity analysis

4.3.1. Neutralizing agent

Based on the GWP impact contribution of consumables in the entire process of hard carbon manufacturing as shown in Figure 4.11, it can be seen that hydrochloric acid stands out as the highest contribution with 0.37 kg $CO_2 - eq$ followed by hydrochloric acid with 0.144 kg $CO_2 - eq$. However, sodium bicarbonate contribution is much less than previously mentioned consumables by one order. Also, sodium bicarbonate can be utilized as a neutralizing agent as well. Therefore, it is also interesting to find out how much decrease can be achieved in the climate change impacts of consumables in the production. Consequently, the analysis was done on replacing the neutralizing agent with sodium bicarbonate and the results can be seen in Figure 4.12. The figure shows that unlike what was expected the GWP impacts for neutralizing the acid doubled. Consequently, the total GWP impacts of the consumables increases from 0.645 kg $CO_2 - eq$ in base scenario to 1.085 kg $CO_2 - eq$ using sodium bicarbonate.

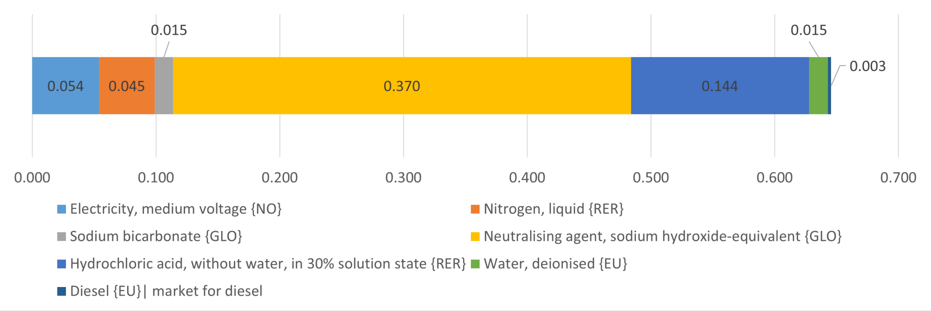

Figure 4.11: Climate change impacts of consumables in hard carbon production from sawdust

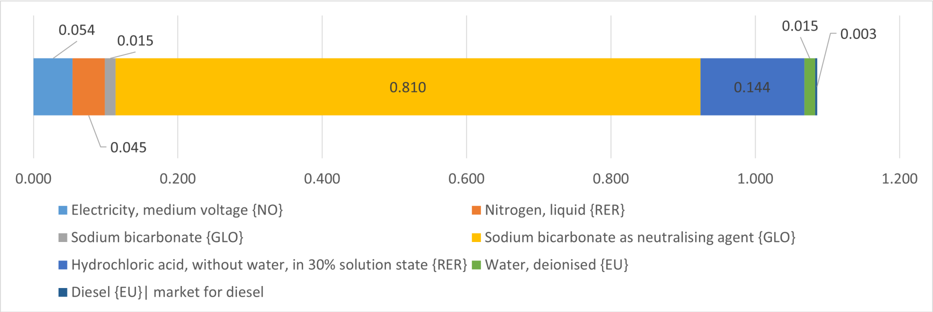

Figure 4.12: Climate change impacts of consumables in hard carbon production from sawdust, using sodium bicarbonate as the neutralizing agent

4.3.2. Change of yield

In this section, the focus is put on the GWP impacts of the system. It may be interesting to look at the impact of improvements done in each process to assess which one can have the best influence on reducing the GWP impacts. Therefore, a sensitivity analysis is done on the yield percentage of namely, carbonization, acid washing and drying, and heat treatment processes. The milling processes is excluded in this section due to its relatively negligible contribution on the GWP impacts. This can be seen in Figure 4.13. The sensitivity analysis was done by changing the yield percentage in each process by ±10%. Subsequently, the life cycle inventory was adjusted accordingly and the total GWP impacts were calculated. As shown in Figure 4.13, the green bars show the desired decrease in the total GWP impacts in percentage of the actual total value. On the other hand, the red bars show the undesirable outcome of decreasing the yield in different processes which subsequently leads to increase of total GWP impacts. Overall, it can be seen that the increase in yield can achieve lower GWP impacts and better results. Moreover, the system climate change impacts change by -1.32% and 3.3% when there is a change of ±10% in carbonization yield. However, heat treatment has the highest changes of -10.01 % and 12.3 %. Also, acid washing process has comparable values to heat treatment process with -7.71 % and 1.12 %.

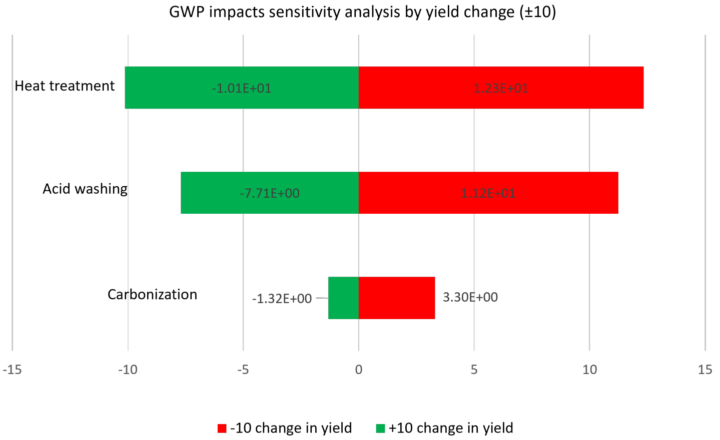

Figure 4.13: GWP impact sensitivity to different processes yields change, comparison between carbonization, acid washing, and heat treatment impacts by changing their yield by ±10%

5. Discussion

5.1. Production model

As provided in chapter 3, the production of hard carbon from sawdust is devised based on several studies and the suggestions from experts in the industry who have related knowledge to similar processes. Some of the mentioned studies are more deeply discussed here and their similarities and difference to the subject of this study is investigated.

In Peters et al [22] study, they investigated the environmental impacts associated with the production of sodium-ion batteries using hard carbon as the anode. In this study, carbohydrate is the source of hard carbon. They have shown that the GWP impacts per 1 kwh energy of sodium-ion battery produced from sugar source is nearly 140 kg CO_2-eq where almost 26% of that was associated with anode production. Also, they compared other sources for the production of hard carbon such as starch, cellulose, and fossil source. The interesting evidence in their study was that the GWP impacts of production of Na-ion battery from fossil had approximately 20% decrease compared to sugar precursor [22]. However, cellulose showed a small increase in terms of GWP impacts compared to sugar based anode. The reason for the large decrease was reported to be the high yield of hard carbon from fossil source compared to that of sugar precursor [22]. However, there was no result presented regarding the GWP impacts only associated with 1 kg of hard carbon production. Therefore, in order to achieve that, the provided inventory for hard carbon manufacturing was modelled in SimaPro in this study. Their production model consists of a carbonization process under ambient atmosphere at $300°C$ plus a pyrolysis process for 3 hours at $1100°C$ [22]. Similarly, in this study the production model incorporates a carbonization process (at $600°C$ based on WAI) and a pyrolysis process at $1300°C$. However, it is evident that sawdust-based hard carbon production model in this study has lower GWP impacts compared to Peters et al study (0.81 kg $CO_2 - eq$ compared to almost 15 kg $CO_2 - eq$). The reason for this large difference can be attributed to the low yield of hard carbon production from sugar to a large extent where 20 kg of sugar is needed for producing 1 kg of hard carbon. In comparison to this, in this study the product yield 1 kg of hard carbon from 5.8 kg of sawdust. Therefore, the GWP impacts associated with the sawdust material is nearly 0.36 kg $CO_2 - eq$, approximately 11 kg $CO_2 - eq$ lower than sugar source hard carbon. Another relatively large contribution comes from nitrogen gas in Peters et al study which can be due to the large amount of feed material to the pyrolysis process and the nitrogen need is a function of that feed material input. This can be due to lower carbon content of sugar compared to sawdust. However, other factors such as efficiency of the manufacturing processes and different assumptions can be the reason as well. Also, it is worth mentioning that the electricity source for Peters et al [22] study is a European mix which leads to higher GWP impacts than Norway electricity. This can be due to clean electricity of Norway that has surplus renewable electricity production compared to its consumption[25].

Furthermore, the study of Liu et al [19] was regarding the comparison of hard carbon manufacturing by hydrothermal carbonization plus a second pyrolysis, and a direct pyrolysis method. Their study was a lab-scale investigation and some of their energy requirements could be lower if the production was large scale due to better efficiency of the processes. The reported GWP impacts by Liu et al study is just over 4 kg CO_2eq for the two-step production. The alternative scenario with direct pyrolysis process is not the interest of this study due to no similarity. It can be verified that a very large contribution (almost 3 kg CO_2eq) to the GWP impacts in Liu et al study [19] was attributed to the electricity input. This is interesting when compared to this study results since clean electricity of Norway grid has much lower impacts (0.054 kg CO_2eq) to the climate change. The source of electricity was not reported in Liu et al study and it is not possible to properly compare the GWP impacts.

5.2. Main findings

Overall, several hard carbon production scenarios have been presented that mainly were different in the precursor used in the hard carbon. Firstly, the sawdust material modelled by the information given by Bergene Holm AS [24] where the production of carbonized sawdust has a negative value of -1.77 kg CO_2-eq per kg of product. Secondly, preexisting sawdust material from Ecoinvent database was added to have better comparison to the literature available results since most of investigated results used the predefined datasets available in libraries. Next, the study provides the GWP impacts associated with sugar-based and petroleum-coke-based hard carbons. The base scenario shows that a very large credit is achieved by the use of sawdust from Bergene Holm AS [24] since it has negative CO_2-eq emissions and this can be counter intuitive to some extent since this means that more usage of this type of feedstock leads to lower GWP impacts. However, this means that the efficiency of the manufacturing system is decreased which is not satisfactory.

Moreover, looking at Figure 4.7 we can see that largest impact comes from acid washing and drying process, this can be due to use of chemicals such acids and neutralizing agents. This can be represented in Figure 4.3 where neutralizing agent and hydrochloric acid have the largest impacts respectively. The credits that are shown in green in Figure 4.7 are due to export of extra heat produced in the carbonization process. However, it is important to note that this heat should be credited based on an energy type which is used in the geography of Norway and not other countries such as Europe. This is especially important because if this heat is assumed to get credits of replacing heat in the Europe area, it is given excessive credits. Therefore, it is assumed that the heat is used as electricity in Norway. The climate change impact of the sawdust bought from the market has the second highest value followed by heat treatment. The reason for high impact of heat treatment can be associated with the high amount of heat needed for consistent high temperature of 1300$°C$. This is based on the provided information by Liu et al in their published work [19]. It is interesting to point out that carbonization process has almost zero GWP impacts as can be seen in Figure 4.7. This is basically because the sawdust is assumed to be biogenic material and the carbon emission of the carbonization is a biogenic type emission. This means that the carbon emission barely have GWP impacts on the environment.

The total impacts of the production in the base scenario and the alternative with predefined sawdust are approximately -3.56 and 0.81 kg CO_2-eq respectively. The latter result is relatively comparable to similar studies in literature such as Liu et al study [19]. However, the difference is large to some extent. Firstly, it is important to note that the electricity source for the manufacturing plant plays an essential role in the total impacts as it is used in different processes. The mentioned study did not provide the geography for their electricity dataset. Therefore, one can argue that the difference in high impact of electricity usage is due to the carbon-intensive electricity grid in Liu et al study [19].

Looking at four impact categories, we can understand that acid washing process has very high contribution to eutrophication and ozone depletion potentials although it is not as influential in global warming and acidification potentials. The reason can be due to usage of acid and the waste water discharge that has been considered in the LCI. However, one can argue that the acidification impacts of this process should also be relatively higher than other processes. An argument against the former could be that there was no direct emission of SO_2 in acid washing process although there are direct SO_2 emissions in carbonization process which makes it relatively more influential than acid washing acidification impact.

Meanwhile, by taking a look at Figure 4.11, we can understand neutralizing agent used in acid washing process has the highest GWP impacts followed by hydrochloric acid. The GWP impacts of neutralizing agent were not mitigated by gchanging the material to sodium bicarbonate due to higher mass of the sodium bicarbonate compared to sodium hydroxide. This is due to the fact that 1 mole of each neutralizing agent is needed for 1 mole of hydrochloric acid although the molar mass of sodium hydroxide is much higher than that of sodium bicarbonate.

The sensitivity of climate change impacts to different processes were analyzed in the last section of chapter 4. The main finding in that analysis is that main focus for improvements can be firstly attributed to heat treatment process and acid washing. The reason is that increasing efficiency in these processes will lead to higher reductions in the GWP impacts. Compared to the former two processes, carbonizing process is less influential on the GWP impact reduction. It is also interesting to look at Figure 4.13, where it shows that lowering the efficiency in each process has more adverse effect than achieving benefits of increasing the efficiency. This can imply that heat treatment process is more critical and its efficiency should be at least consistent if there is no possibility of improvements.

6. Conclusion

A literature review was done on the hard carbon manufacturing and structure analysis to place fundamental knowledge for further life cycle analysis. The manufacturing insights of technical experts in the industry was also utilized for more specific design of a manufacturing model in order to incorporate in the life cycle inventory. Using a combination of both sources, this study provides a detailed model of the hard carbon production based on sawdust as a precursor. The details about the temperatures, storage times, and equipment energy and material requirements were established. The processes modelled in the system were mainly categorized in four different stages namely, carbonization, acid washing and drying, heat treatment, and lastly the milling process. These processes were modelled for a large-scale manufacturing capacity of 1000 metric tons as the yearly production. However, the functional unit of the model was set to 1 kg of hard carbon. This was based on the other available LCA studies.

Furthermore, a life cycle assessment study was established using the mentioned information. The data was modelled in SimaPro as the LCA professional software. Several scenarios were investigated in order to give perspective and foundation for reasonable comparison. Meanwhile, it is important to note that some of the inventories are completely based on another study supporting information. This was done due to the fact that the functional unit of this study is different.

The results of the life cycle assessment were provided in four different impact categories namely, global warming potential, acidification potential, ozone depletion potential, and eutrophication potential. The results showed that considering the GWP impacts, the highest impacts were attributed to carbonization and acid washing processes. However, it is also important to emphasize the sensitivity of GWP impacts to other processes such as heat treatment. This process might have significant influence on the results if it yield could be higher or lower. Regarding that the yields of several processes despite carbonization are assumptions based on the literature and industry experts. This can imply that future changes to yields can have large effects on the climate change potentials impacts.

Overall, the total GWP impacts of the LCA model in this study were relatively smaller in comparison to other LCA results from other studies available in the literature. The underlying reasons could be the usage of clean electricity in Norway and different yields and efficiencies compared to other studies. The difference in the assumptions for energy requirements for each process could also be the reason as well. This implies that hard carbon manufacturing in Norway could be a clean production and a viable source for alkali-metal ion batteries.

7. Furhter Work

More details regarding the manufacturing processes can be investigated after the building of factory with the real data taken from the actual process energy requirements. This can improve the reliability of the results for further planning.

The production of hard carbon can be further analyzed by establishing a lab-scale fabrication of sodium-ion batteries to understand how the structure of sawdust can affect the electrochemical performance and investigate the best conditions in every production step to reach maximum performance for the battery. This can influence the production rate to a large extent since it can be a function of the efficiency of the battery performance.

It can be suggested that an economic analysis can be done on the hard carbon manufacturing from sawdust to understand which processes have the highest economical values. This can improve the planning of the manufacturing plant for improvements in the production steps. In this regard, a sensitivity analysis can be done on the processes to see which processes can have the highest impacts on the overall costs and try to see trade offs of lowering the environmental impacts to the cost of improving the processes.

Further analysis can be done on the other impact categories in order to understand if there are any burdens that are shifted in other impact categories due to having very small impacts in the GWP category. Furthermore, the study can be improved by analyzing the endpoint indicators and find out how different impact categories have influence on different areas of protection such as human health, land use, and marine ecosystems. This can give proper perspective to the reader for better comparison between different impact categories.

Moreover, more in depth research can be done the possible equipment that can be used in each process to gather more reliable assumptions and designs for the manufacturing process. This was not possible during the current study since the equipment specifications were not a focus and the process could be too much time consuming. This could affect the main focus of the study which was the life cycle analysis.

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