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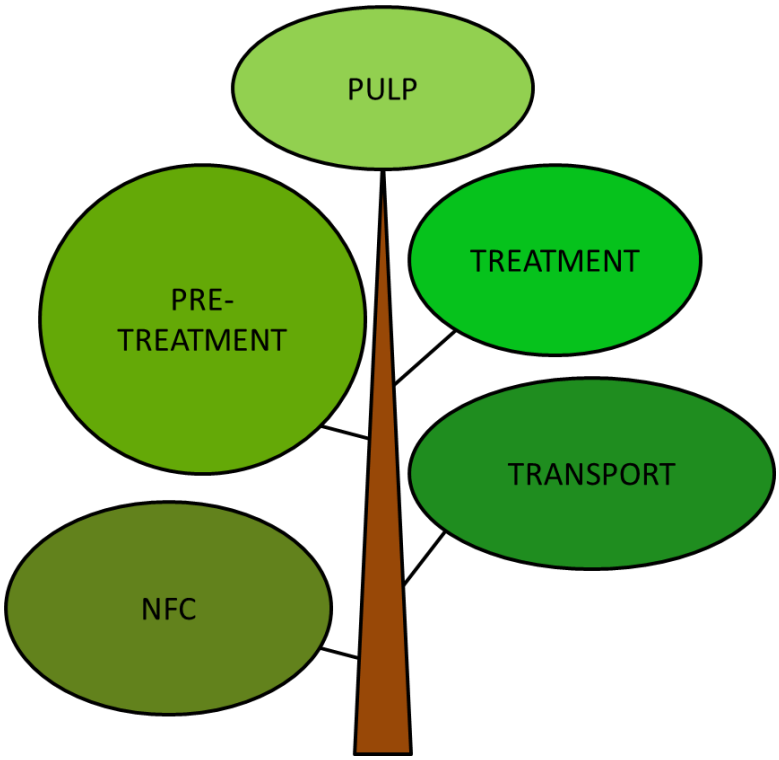

Life cycle energy assessment of wood-based Nano Fibrillated Cellulose

Master of Science Thesis in Industrial Ecology

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Department of Chemical and Biological Engineering
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In collaboration with Innventia
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Cover: Tree of factors influencing energy use in nano fibrillated cellulose production.

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Abstract

Nanocellulose is a new ‘super material’, stronger than steel, stiffer than Kevlar, transparent and light, absorbent and conducting electricity, biodegradable and anti-bacterial. As a result, it attracts a lot of attention from companies and research institutes around the world. It is likely that commercial nanocellulose and its application products enter the market in the near future.

However, this new material has an energy use which relates directly to both production cost and environmental impacts. Therefore, information about the energy use of nanocellulose is needed as soon as possible to help decision makers in choosing appropriate production processes from the very early stage of production.

This master thesis aims to analyze the energy use of wood-based nano fibrillated cellulose (NFC), probably the most common nanocellulose type, from a life cycle perspective. In this study, two production routes - the enzymatic route and the carboxymethylation route - are outlined, assessed and compared for a number of different scenarios. The system boundaries include the extraction of cellulosic materials, the processing of NFC and input materials, and the transport of materials. The functional unit is 1 kg of NFC. Data is obtained from three main sources: the Ecoinvent database, presentations and studies from NFC’s projects and the general scientific literature.

The findings indicate that the total energy use of NFC production for the carboxymethylation route is much higher than that for the enzymatic route. This higher demand relates mostly to the chemicals production, especially the organic solvent production. Solvents may not be perceived as having an energy use by NFC producers, but this is clear when seen from a life cycle perspective.

This study points out five main factors that strongly affect NFC’s energy demand, namely pulp properties, pretreatment, treatment, transport and NFC product’s properties. Efforts to reduce NFC’s energy use should be based on a comprehensive view of both separate factors and the successful combinations of factors.

Suggestions of future work in the field include a comprehensive LCA (life cycle assessment) of NFC, and a cradle-to-grave life cycle (energy) assessment of a specific NFC application product.

Keywords

Life cycle assessment, Life cycle energy assessment, Energy use, Nanocellulose, Microfibrillated cellulose, Nano fibrillated cellulose.

Acknowledgements

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Finally, I would like to thank my family, my friends and my teachers for all the love and support.

Table of contents

1. INTRODUCTION

This chapter provides an overview of the new material nanocellulose and recent publications related to the topic. The study's aims and research questions are also described.

An invention of a new material can open a new era in manufacturing industries. By using a new material with new properties, new functions can be fulfilled. However, a new material also has an energy requirement which impacts both production cost and the environment. The question is whether this energy use of the new material exceeds that of existing materials. Early awareness of the total energy use can help decision makers, including producers, consumers and policy makers, to avoid energy intensity in new materials and production processes from very early stages of technological development.

This study investigates a new bio-nano material, namely nanocellulose. It is stronger than steel, stiffer than Kevlar, transparent and light, absorbent and conducting electricity, bio-degradable and anti-bacterial. Therefore, this new material promises many potential applications in the fields of nanotechnology, energy, medicine, food and many others. Because of the increasing number of companies and research institutes starting projects related to manufacturing of nanocellulose, the material will probably be produced at industrial scale in the near future. That is the reason why further understanding of the material and its production, especially the energy use, is of high interest.

To fulfill this research need, the study analyzes the life cycle energy use of one common type of nanocellulose: Nano-Fibrillated Cellulose (NFC) made from wood. In this study, the total energy use of NFC's production is assessed during three life cycle phases: pulp production, pretreatment and treatment. Because it is a cradle-to-gate life cycle assessment, it covers the steps from input raw material extraction to output product at the factory gate.

1.1. Context

These recent years have witnessed a booming development in nanocellulose production and applications, shown by recent publications about new cellulose sources, new methods to produce nano-sized fibrils and new functions discovered. The term "nanocellulose" was first introduced in 1983 (Wågberg et al., 2008). In 2006, nearly 600 scientific documents about nanocellulose were published. It doubled and reached 1,200 papers after 5 years (Lavoine et al., 2012).

However, there still remain knowledge gaps related to the life cycle performance of the material and its products at industrial scale. From producers' perspective, energy-intensive manufacturing acts as one of the biggest barriers for up-scaling nanocellulose production. The mechanical treatment phase, which requires up to 27,000 kWh/tonne, is considered an energy-intensive step in the process (Ankerfors, 2012). By applying pretreatment processes, recent process developments have enabled energy use to be reduced by 93% (2,000 kWh/tonne is reported in projects of Innventia, Ankerfors, 2012 and projects of SUNPAP, Meyer et al., 2012) and even a 98% reduction is projected (Ankerfors, 2012). However, this energy use is only the tip of the iceberg from a life cycle energy perspective. In 2014, Li and colleagues calculated the lowest total energy use of NFC at 34.7 MJ/10 g NFC at lab scale, equal to approximately 963,889 kWh/tonne, even when using pretreatments. The differences originate from how to define the concept of "energy", which in life cycle energy assessment

(LCEA) implies not only the manufacturing energy but also energy for producing input materials, energy in raw materials, and energy in transport.

There exist a number of studies on energy demand of nanocellulose production, such as a comparative study of energy use for different NFC processing routes (Spence et al., 2011), energy use in several different NFC production routes produced by Innventia (Ankerfors, 2012), energy use in NFC production using TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl) oxidation pretreatment in SUNPAP, an European project about Scaling Up Nanoparticles in Modern Paper Making (Meyer et al., 2012), and approximate energy use for NFC production in general (Klemm et al., 2011). However, these works provided only energy use for the production phase, which is the life cycle phase of highest concerned for producers. Until recently, there are very few academic studies on the topic LCA of nanocellulose, including a study by Li and colleagues on LCA of NFC (2013), and a study on LCA of nanowhiskers (Figueiredo et al., 2012).

In this thesis, the energy use of the whole NFC production process is calculated, using a life cycle perspective. Therefore, it provides more comprehensive results for the whole life cycle and also specific results for each phase in each production route, which is useful for producers.

1.2. Aim and research questions

This study aims to assess the total energy use of NFC production. Two different production routes are described; their energy demands are compared and contrasted in order to provide useful information for decision makers, including producers, consumers and policy makers.

The research further attempts to answer the following two research questions:

- Which production route in the two selected routes requires less energy for producing 1 kg of NFC?
- What are the main factors that affect energy use of NFC along the whole life cycle?

1.3. Thesis outline

This thesis contains the following chapters. Chapter 1 provides a brief introduction to the study, including context, motivation and research questions. Chapter 2 brings in background information of the material nanocellulose, describing its chemical structure, classification, production, application, safety and market. Chapter 3 presents research methods and materials. Chapter 4 highlights the main results obtained from the study, in this case the total energy use and scenario analysis. After that, the results are interpreted and discussed in Chapter 5. Finally, a conclusion and suggested future work are presented in Chapter 6.

2. BACKGROUND

This chapter is about background information on nanocellulose. Its content covers chemical structure, classification, production, application, safety and market of nanocellulose.

2.1. Chemical structure

The strong, cheap, and light material of nanocellulose, which has been called a "super-material", was introduced the first time by Turbak and his colleagues in the 80s (Turbak et al., 1983). However, it has been revealed that Rayonier and colleagues were the first producers of cellulose fibrils in nanosize 6 years earlier, according to a report of Turbak on Over The Wire, in an online magazine by TAPPI (Technical Association of the Pulp and Paper Industry) (Turbak, undated).

The name correctly indicates the structure of cellulose fibrils at nano-size scale. Cellulose, a main fibrous component of plants' cell walls, is a polysaccharide consisting of several hundred to over ten thousand β-1,4-D-glucan molecules linked in a linear chain (Crawford, 1981). When the size is reduced from micro-size (normal size of cellulose fibers) to nano-size in one or more dimensions, nanocellulose is created (see figure 1).

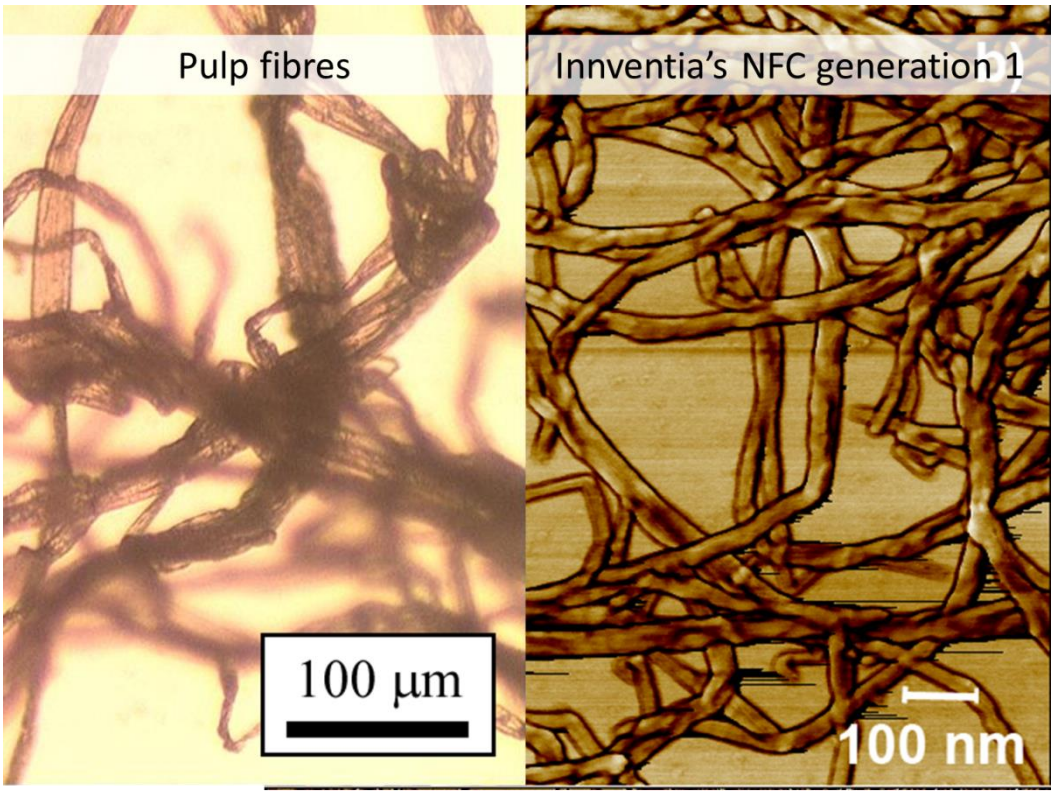

Figure 1: NFC's fiber structure.
Image provided by Ålander, 2014, and are reprinted here with permission.

2.2. Classification

There are several types of nanocellulose, separated by their size, shape and cellulose source. Table 1 shows different types and terms of nanocellulose, as proposed by Klemm and colleagues in 2011. Because of the lack of official international standards for

nanocellulose up until now, there are many other ways to classify this new material, such as in the work of Siro and Plackett (2010) and Khalil et al. (2014). In 2012, Lavoine and colleagues mentioned 20 different terms describing different types of nanocellulose in publications from 1993 to 2011. To facilitate the commercial transaction of this promising material, TAPPI developed a roadmap for the international development of nanocellulose, which also proposed a naming hierarchy (TAPPI, 2011).

Table 1. Types of nanocellulose (Klemm et al., 2011)

Types of nanocellulose	Synonym	Size
Nano fibrillated cellulose (NFC)	Microfibrillated cellulose (MFC), nanofibrils, microfibrils	Diameter: 5-60 nm Length: several micrometers
Nano crystalline cellulose (NCC)	Cellulose nanocrystals, crystallites, whiskers, rodlike cellulose microcrystals	Diameter: 5-70 nm Length: 100-250 nm (made from plant) or 100 to several micrometer (made from bacteria, algae, tunicates)
Bacteria nanocellulose (BNC)	Bacterial cellulose, microbial cellulose, biocellulose	Diameter: 20-100 nm

Despite the differences in terminology, in general, on the basis of production methods and raw material origin, nanocellulose is divided into three main subcategories: (1) Nano fibrillated cellulose (NFC), a spaghetti-shaped elongated fibril form prepared from wood or other natural cellulose sources via chemical and/or mechanical treatments, (2) Nano crystalline cellulose (NCC), a rice-shaped crystalline form prepared from plant, animal or bacteria-originated cellulose produced via acid hydrolysis, and (3) Bacteria nano-cellulose (BNC), a network form of nanocellulose produced by bacteria or genetically transformed algae (Klemm et. al, 2011, Li et. al, 2013). Figure 2 illustrates NFC products of Innventia, a Swedish NFC producer.

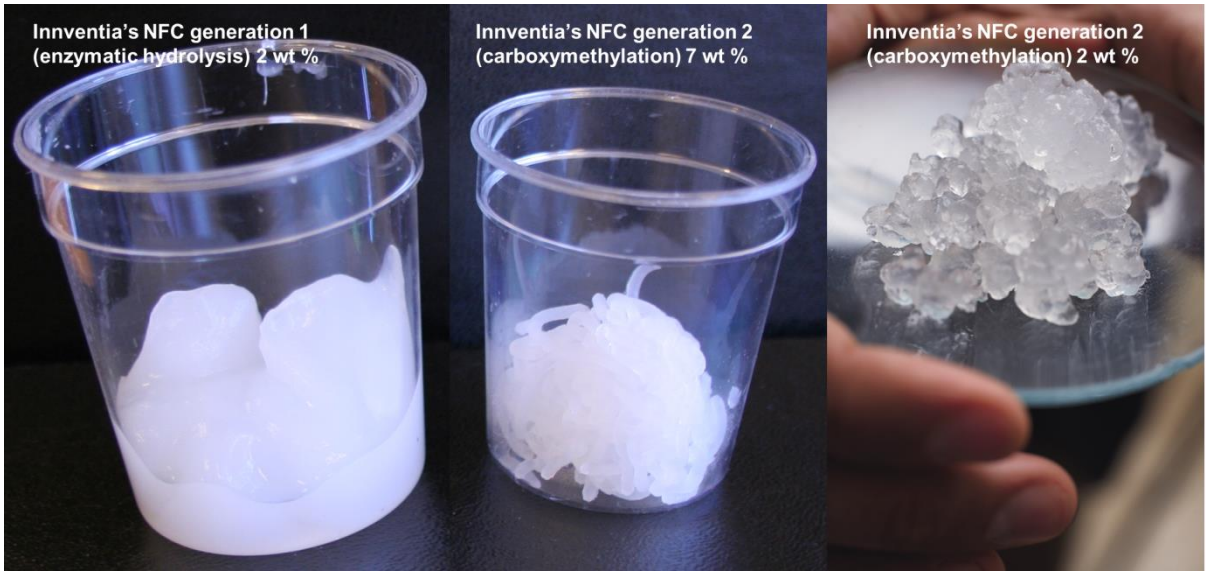

Figure 2. NFC products produced by Innventia: (from left to right) (1) NFC generation 1, produced by enzymatic process, 2 wt %; (2) NFC generation 2, produced by carboxymethylation process, 7 wt %; (3) NFC generation 2, produced by carboxymethylation process, 2 wt %. Images are provided by Ålander, 2014, and are reprinted here with permission.

2.3. Production

Theoretically, nanocellulose can be produced from any cellulose source from wood, other crops and bacteria. While NFC and NCC can be created by the disruption of hydrogen bonds in the cellulose until it reaches nano-scale (top down process), BNC can be directly produced from certain bacteria (bottom up process).

Recently, a brand new method to produce BNC was reported. Dr. Brown from University of Texas introduced a process of engineering nanocellulose genes of *A. xylinum* (vinegar bacterium) into blue-green algae (Algae Industry Magazine, 2013). By doing that, these algae (Cyanobacteria) had the potential to release nanocellulose directly into their surroundings, making it easier to harvest. However, natural generation rates of cyanobacterial nanocellulose are low, which limits the potential of producing BNC in commercial scale. When this problem is solved, the process for making BNC will be "cheap, fast and involved nothing else than water, sunshine and time", according to the Huffington Post UK (2013). Scientists expect the successful mass production of both biofuels and nanocellulose made by these algae.

When it comes to NFC and NCC, among many types of sources, wood pulp is one of the most commonly used. Pulp is normally manufactured by breaking down the bulk structure of cellulose fibers; therefore it is already half way to create nanofibrils. The complete process requires further steps to split up pulp fibers into smaller, nano-sized fibrils. Figure 3 shows an example of a life cycle of NFC from cradle (wood extraction) to grave (end of life, which could involve recycling, incineration, landfilling or other waste management methods).

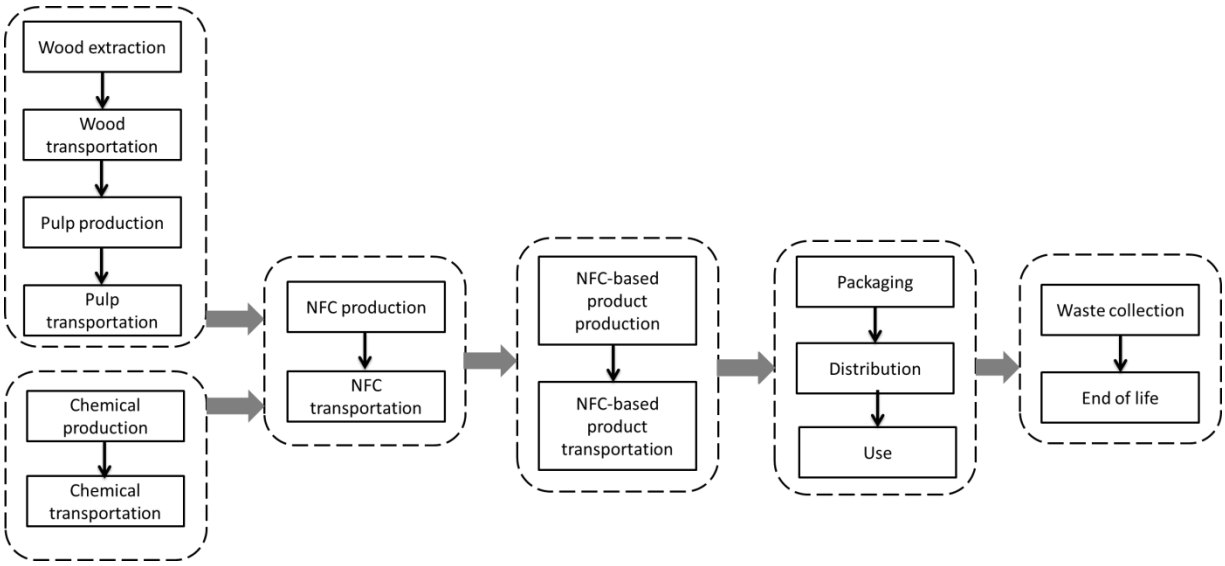

Figure 3. Generic life cycle of wood-based NFC.

In general, one could divide the production process of nanocellulose (NFC and NCC) into three main stages. First is the transformation phase from wood to pulp, which has been done for years in pulp mills. This process aims to break down the bulk structure of the wood fiber source into the constituent fibers in pulp. It can be done either by chemical processes in which lignin and hemicellulose (other parts of plants' cell walls) are degraded and washed away, or mechanical processes when cellulose fibers are simply torn apart. There are also hybrid methods with supplement chemical and thermal treatments in pulp making.

Next are the two main phases of nanocellulose production: pre-treatment and treatment, when pulp fibers are separated in serial steps to make nanocellulose. In early times, the process included only the treatment phase in which the manufacturer produced nanocellulose by a high pressure homogenizer (Turbak et al., 1983). It is a machine widely used in food and paint industries to disrupt bulk input materials into small uniform particles (Spence et al., 2011). In the homogenization process, cellulose fibers are subjected to a large pressure drop with shearing and impact forces (Nakagaito and Yano, 2004). The process' disadvantages are energy intensity and clogging problems, particularly in the moving parts (Spence et al., 2011). Homogenization is the conventional mechanical treatment in the nanocellulose production at industrial scale. After that, new technological solutions, such as fluidization and sonication, were brought in. Micro-fluidization is commonly used in the cosmetic, biotechnology, and pharmaceutical industries (Microfluidics Corporation, 2010). In this process, cellulosic pulp is passed through an intensifier pump, followed by an interaction chamber which defibrillates the fibers by shear forces and impacts against the channel walls and colliding streams. Because the micro-fluidizer has no in-line moving parts, it reduces the likelihood of clogs, compared to the homogenizer (Spence et al., 2011). Sonication is a mechanical process which uses oscillating power to isolate cellulose fibrils by hydrodynamic forces of ultrasound (Cheng et al., 2009). Khalil and colleagues suspected that mechanical methods with high energy use can cause a dramatic decrease in both the yield and fibril length (Khalil et al., 2014). The treatment phase, which requires up to 27,000 kWh/tonne product when using homogenization (Turbak et al., 1983), is often considered the most energy intensive step in the whole process.

To reduce the total energy use and thereby obtain cost savings, scientists have proposed several mechanical and chemical pre-treatment processes. Applying these processes decreases the energy use for treatment to about 2,000 kWh/tonne product, equal to 93% reduction (2,000 kWh/tonne was reported in projects of Innventia - Ankerfors, 2012 and Meyer et al., 2012). Some common and feasible processes in the pre-treatment and treatment phases are shown in Table 2.

Table 2. Methods for nanocellulose production (NFC and NCC)

Phase	Process	Source
Pretreatment	Physical refining	Ankerfors, 2012 Spence et al., 2011 Pääkkö et al., 2007
	TEMPO oxidation	Li et al., 2013 Klemm et al., 2011
	Carboxymethylation	Li et al., 2013 Ankerfors, 2012 Klemm et al., 2011
	Enzymatic hydrolysis	Ankerfors, 2012 Klemm et al., 2011 Pääkkö et al., 2007
Treatment	Homogenization	Li et al., 2013 Isogai et al., 2011 Klemm et al., 2011 Spence et al., 2011 Pääkkö et al., 2007
	Micro-fluidization	Ankerfors, 2012 Spence et al., 2011
	Micro-grinding/shearing	Spence et al., 2011 Pääkkö et al., 2007
	Sonication	Li et al., 2013

Different combinations of pretreatment and treatment processes can create different production routes. After the treatment phase, the NFC product has a gel-like shape containing a major fraction of water (Figure 2), for example standard 2% gel-like MFC product (Pääkkö et al., 2007). Further processes such as purification, dilution, etc. can be applied to make nanocellulose with different concentrations. The material can then be functionalized through specific processes to produce nanocellulose-based products, including films, nanocomposites, coatings, etc.

Researchers continuously propose additional processes and new combinations of feasible methods in order to reduce energy demand on one hand and improve material properties on the other hand. The fact that many solutions give successful results at lab scale but fail at industrial scale implies the need of further investigation in this field.

2.4. Application

Manufacturers expect a big success in nanocellulose-based products thanks to its special properties. Nanocellulose is suitable for food and emulsion applications, medical, cosmetic

and absorbent products. It can also be used in various nanocomposites, packaging, coating additives and gas barriers (Klemm et al., 2011).

According to VTT Technical research center of Finland, in 2011, composite material contributed a 38% share of the total nanocellulose applications' market, becoming the largest market segment. It was followed by non wovens adsorbent web (18%), paper and board (16%) and food products (13%). The smaller market segments (less than 10% of total) were paper and board coating (8%), filter material (4%) and cosmetics and toiletry (3%) (Koskinen et al., 2011).

Abundant applications of this new material motivate manufacturers to enter the market as fast as they can. However, pioneers have to face high risks, including the lacking policy, limited feasible technology and the doubts about how good this material actually is.

2.5. Safety

Novel substances and materials may come with new hazards. A recent study indicated that another novel nanomaterial, graphene, has considerable toxicity which may cause adverse environmental and health effects (Arvidsson et al., 2013). In fact, a number of environmental and safety concerns have been raised for nanomaterials in general. Nel and colleagues, in 2006, mentioned the ability of harmful interactions between nanomaterials and biological systems and the environment. In 2011, Arvidsson et al. outlined challenges in exposure modeling of nanoparticles in aquatic environments.

Therefore, when nanocellulose is expected to be mass produced, it requires further research into health, safety and environmental burdens. However, no immediate environmental and safety concern related to exposure of wood-based nanocellulose has been reported yet. In 2011, Vartiainen and colleagues noticed no toxicity in the friction grinding and spray drying processes of NFC. In their study, no evidence of inflammatory effects or cytotoxicity on mouse and human macrophages were observed, and ecotoxicity tests showed high NOEC (no effect concentration) values. Although no significant effects were detected, additional hazard studies were recommended to dig deeper in the field (Rouhiainen, 2011).

Furthermore, it is necessary to understand the safety of each type of nanocellulose and each modification. Potential risks should also be considered in each step of the production of nanocellulose, and also in the production of input materials needed to manufacture nanocellulose. For instance, bleaching is an important part of the pulp production. Evidences showed that bleaching of pulps may cause significant environmental impact through the release of organic components into waterways (Sonnenfeld, 1999). This negative impact may also contribute to environmental impacts of NFC made from bleached pulp. Such environmental impacts along the life cycle of nanocellulose imply consideration of the whole life cycle by the method of LCA, described further below.

2.6. Market

People have called nanocellulose a "super material" due to its plentiful outstanding properties in comparison with other available materials on the market. That is the reason why more and more companies and institutes make efforts in developing commercial scale production of this material. According to ArboraNano, The Canadian Forest Nanoproducts Network, until 2012, the biggest production capacity at current rate, reaches around 10 kg NCC/day (Bio Vision Technologies Inc., Canada) and 500 kg NFC/day (The US Forest

Service). Innventia, a Swedish company, produces approximately 100 kg NFC/day. A Canadian company, Celluforce Inc. projected to have 1 tonne product/day. Table 3 lists some nanocellulose suppliers who have announced their production capacity. In total, these numbers indicate an annual production rate of 200 – 300 tonnes/year of nanocellulose (excluding the projected future capacity), with most being in the form of NFC.

Table 3. Nanocellulose suppliers (ArboraNano, 2012, Rebouillat and Pla, 2013).

Producers	Production capacity	Country
Alberta Innovates – Technology Futures	NCC, 100 kg/week, projected rate	Canada
Bio Vision Technologies Inc.	NCC, 4 tonne/year	Canada
Celluforce Inc.	NCC, 1 tonne/day, projected rate	Canada
FP Innovations	NCC, 10 kg/week	Canada
Innventia	NFC, 100 kg/day	Sweden
The US Forest Service	NCC, 35-50 kg/day, projected rate	USA
The US Forest Service in collaboration with University of Maine	NFC, 500 kg/day	USA

In addition, there are many other big companies around the world that started a project in nanocellulose, for example J. Rettenmaier & Söhne GmbH, BASF and Zelfo Technology in Germany, Cellucomp Ltd. in UK, CTP/FCBA in France, Melodia in Israel, Borregaard Chemcell in Norway, Stora Enso and UPM-Kymmene in Finland, Nippon paper, Asahi Kasei and Daicel corporation in Japan, Verso paper and FMC biopolymer in USA. Moreover, nanocellulose projects have increased in numbers: EU projects such as SUNPAP and SustainComp, ArboraNano in Canada, EMPA in Switzerland, Agenda 2020 in USA and many projects at universities. Considering this high number of companies, the 200-300 tonnes/year is probably an underestimation, since it does not include universities and companies that do not report their production rates. Besides, it further seems that European actors focus more on NFC, while North American ones focus more on NCC.

3. METHOD

In this chapter, the research method is presented in steps. Life cycle energy assessment, goal and scope, system boundary and database are defined. After that comes the system description including the explanation of phases and routes used in the research.

3.1. Method description

3.1.1. Life Cycle Assessment (LCA)

In the umbrella document ISO 14040:2006, life cycle assessment (LCA) is defined as a technique for assessing the environmental aspects and potential impacts associated with a product or service throughout its life cycle, from the cradle, where the raw materials were extracted, to the grave, the disposal. An LCA is implemented by compiling an inventory of relevant inputs and outputs of a product system, evaluating the potential associated environmental impacts, and interpreting the results in relation to the research objectives (Baumann and Tillman, 2004). LCA typically follows the main steps of goal and scope definition, inventory analysis, and impact assessment.

3.1.2. Life Cycle Energy Assessment (LCEA)

Life cycle energy assessment is an approach that accounts for all energy inputs to the product or service, not only energy use during manufacturing and other processes but also energy needed to produce input materials and services during the whole life cycle. In other words, the total energy use during the production is analyzed from a life cycle perspective. An LCA study often includes several different impact categories, such as global warming potential, acidification potential, and eutrophication potential, which are assessed in the impact assessment step. However, in a life cycle energy assessment, energy use is the only included impact category. This is similar to the method of carbon footprint assessment, in which global warming potential is the only included impact category.

3.1.3. Goal and scope

Functional unit

The functional unit of this study is defined as 1 kilogram equivalent dry mass of NFC at the factory gate. This study focuses on the common NFC final product with 2% w/w concentration. Energy use is calculated in the unit of megajoules per kilogram NFC (MJ/kg NFC).

System boundary

This study applies a cradle-to-gate perspective (Baumann and Tillman, 2004), considering the aim to assess the energy required to produce the NFC material. The time horizon considered is approximately ten years from now, making this a prospective LCA study, similar to the prospective LCA of the nanomaterial graphene by Arvidsson et al. (2014).

The total energy use of NFC production in this study is categorized in three different ways, shown in figure 4. The first divides the energy use into the three main phases of pulp making, pretreatment and treatment. The second divides the energy use in another way,

namely into pulp production, chemical production, manufacturing (including both manufacturing in pre-treatment and treatment), and transports. The third divides the energy use into different types of energy: biomass, resource, heat, electricity and fuel. Biomass is the energy content of bio-based feedstock such as wood and corn. Resource includes the fossil resources (crude oil, natural gas and hard coal) and other energy resource (energy in hydropower reservoir).

Heat is the energy for heating from different sources. Fuel includes both the energy listed as 'fuel' in LCI of each input material (in the Ecoinvent database) and the fuel for transports within the processes and between the processes. In order to enable comparability between electricity and heat, electricity use is converted to its corresponding heat requirements by the heat-to-electricity conversion factor. For electricity produced by combustion of fossil fuels, the factor is 0.3, thereby accounting for energy losses in such electricity production (Arvidsson et al., 2014). For electricity produced by renewable sources, the factor is 1. Put differently, energy carriers are recalculated into their corresponding primary energy. Note that the energy indicator in this study is the total extracted energy as defined by Arvidsson et al. (2012). It is the sum of renewable energy and nonrenewable energy and includes feedstock energy.

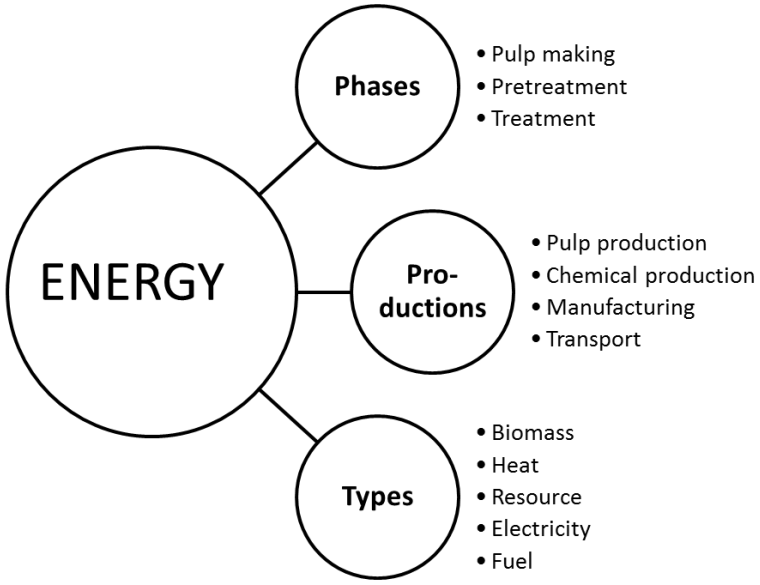

Figure 4. Energy analysis approaches and categories.

Cradle-to-gate assessment covers the steps from extracting resources to the factory gate. In particular, the system boundary (figure 5) contains the extraction of cellulosic materials, the processing of cellulose into NFC, input materials required, and the transport of materials, but not the manufacturing of NFC application products, the use and disposal phases. The making of NCF-based products (NFC application products) differs for specific applications. As a result, cradle-to-grave assessment connects directly to a specific NFC application product while cradle-to-gate is generic for the NFC material. The system model of NFC material could, however, be expanded to include the use and end-of-life phases of certain NFC application products in future studies. Note that the two different main production routes in this study are the enzymatic route and the carboxylation route.

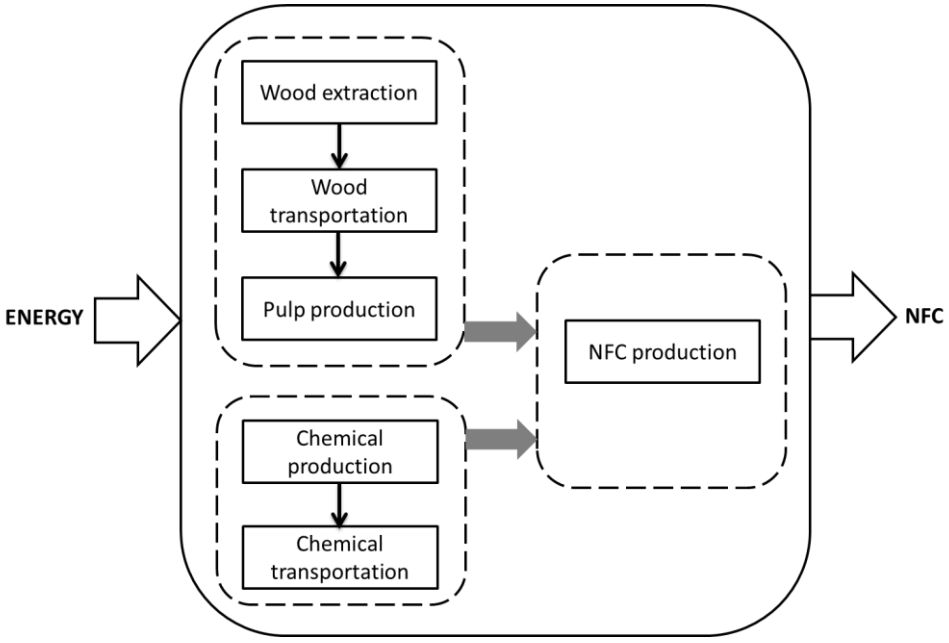

Figure 5. System boundary of this study.

Transportation of input materials includes the transport of wood from the forest or plantation, and the transport of enzyme and chemicals to the NFC factory (figure 6). The transport of wood includes a local production scenario and an import scenario. The transport of other materials includes import scenarios with different exporter regions. In this study, it is assumed that the NFC factory is inside the existing pulp mill due to the fact that wood-based NFC producers are likely pulp producers.

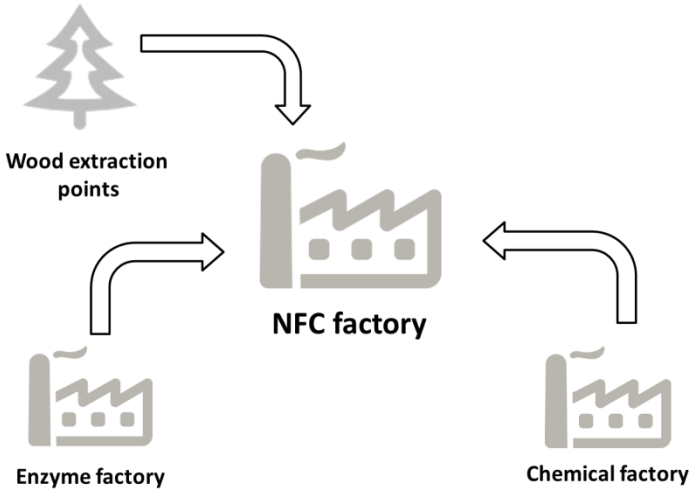

Figure 6. Transport routes
(enzyme factory is only included in the enzymatic production route).

3.1.4. Sources and software

Data is obtained from three main sources: the online Life Cycle Inventory (LCI) database Ecoinvent version 2.2 (2010), presentations and research about NFC production in Innventia's project (Ålander, 2014, Ankerfors, 2012) and SUNPAP project (Meyer et al., 2012), and articles from the scientific literature. Several software and online tools were used,

such as Excel for calculating energy use, and Searates.com for calculating the transport distances.

3.1.5. Research process

This study uses a three-step research process (figure 7) that includes three sources of data: the Ecoinvent database, project literature and scientific literature.

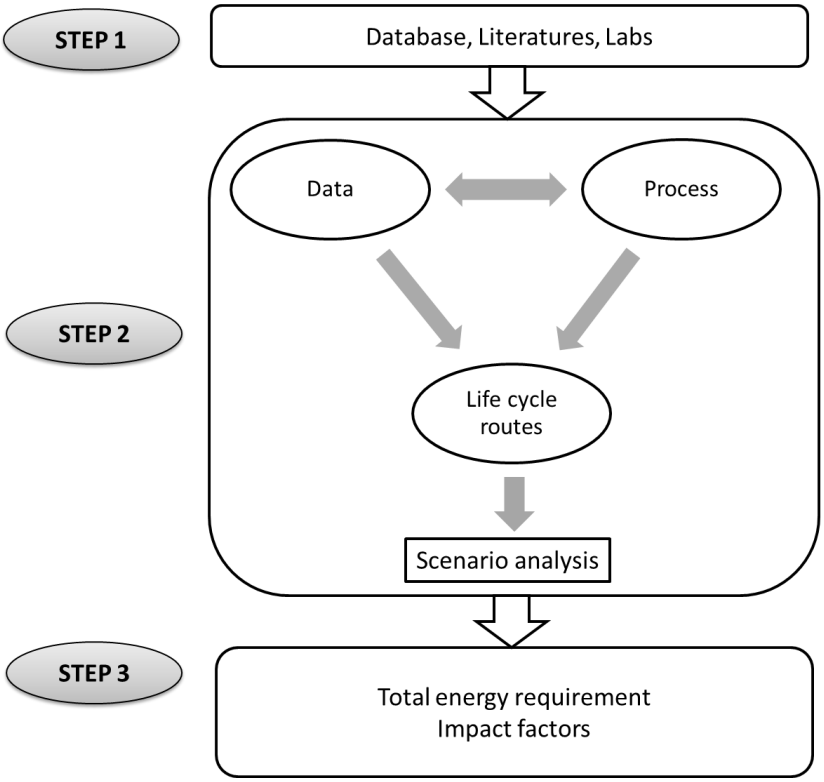

Figure 7. Three-step research process applied in this study.

1. Step one is a data collecting step.

- Data in the pulp making phase is collected from the Ecoinvent database. In the study, four types of pulp mentioned in studies by Ankerfor et al. (2012), Li et al. (2013), Pääkkö et al. (2007), Spence et al. (2011), and Wågberg et al. (2008) are selected.
- Data for the pretreatment and treatment phases is collected from multiple sources. Research literatures show feasible NFC production processes. Project literatures (presentations of NFC producers) indicate which processes are being applied. In this study, carboxymethylation and enzymatic processes are selected because they have detailed step descriptions and manufacturing energy calculation. Steps in these processes identify which input materials need to be analyzed. Data of these input materials is searched in the Ecoinvent database and relevant literatures.

2. Step two is an analyzing step.

- Data of the input materials in the two processes mentioned by Pääkkö et al. (2007) and Wågberg et al. (2008) is re-calculated for 1 kg NFC. From the process, the data needed is identified. After calculation, the energy data is filled in the process. Three phases (pulp making, pretreatment and treatment) are combined to make the life

cycle routes. Note that NFC production processes in most literatures illustrate only two phases (pretreatment and the treatment phases).

- A baseline scenario is made for the enzymatic life cycle route and the carboxymethylation route. In this scenario, total energy is analyzed in three ways: energy in different phases, energy in different productions and energy in different types (see figure 4).
- Other scenarios are made concerning the differences in the types of pulp, the sources of ethanol and the transport options (routes and vehicles).

3. Step three is a conclusion step.

- Total energy use is calculated. The worst case and the best case show the range of energy use in NFC production.
- Factors influencing the energy use of the NFC production are identified.

Some assumptions are made during the process. They are described in the section: Key assumptions.

3.2. Production routes

This section describes the production routes in each phase and in each route.

3.2.1. Phase description

This study only includes NFC made from wood pulp. The cradle-to-gate route can be described in three main phases, where each phase contains several detailed options (transport is included in each phase):

- Pulp making (four different types): Bleached sulfite, bleached sulfate ECF (Elemental Chlorine Free), bleached sulfate TCF (Totally Chlorine Free) and unbleached sulfate.
- Pretreatment (two different processes): Carboxymethylation and enzymatic hydrolysis.
- Treatment (one process only): Micro-fluidization.

Pulp making

This study calculated the total energy use of NFC with four choices of input pulp used in never-dried form. Pulping is the process of separating wood fibers. The two main types of chemical pulps are the more common sulfate pulp, also known as Kraft pulp, and the sulfite pulp. The major difference between the two types of pulps is the chemicals used to dissolve the lignin (Brady et al., 1998).

Bleaching is the chemical process of whitening pulp, mostly used in paper making. The process may cause notable environmental effects, primarily through the release of organic materials into waterways. Conventional bleaching using elemental chlorine produces a large amount of chlorinated organic compounds, especially chlorinated dioxins which are highly toxic and persistent in the environment. Concerning this issue, many producers have applied more environment friendly bleaching processes such as ECF and TCF. TCF removes chlorine completely from the process while ECF can substantially reduce but cannot fully eliminate chlorinated organic compounds from effluent.

This study includes alternatives of bleached and unbleached pulps, sulfite and sulfate pulps, produced from a mixed source of softwood and hardwood, in Scandinavian and European

forests or plantations. There are four types of pulps in this study; energy use for each type of pulp is calculated.. Data is available in the Ecoinvent database (version 2.2) for the following pulps:

- Bleached sulfite pulp: 11% European industrial hardwood, 8% Scandinavian industrial hardwood, 48% European industrial softwood, 33% Scandinavian industrial softwood (% in volume).
- Sulfate pulp (bleached sulfate pulp ECF, bleached sulfate pulp TCF, unbleached sulfate pulp): 14% European industrial hardwood, 19% Scandinavian industrial hardwood, 20% European industrial softwood, 27% Scandinavian industrial softwood, 9% industrial residue softwood, 11% Scandinavian softwood chips (% in volume).

Table 4. Energy and Fuel Data (Staffell, 2011)

	Net calorific value (MJ/kg)
Crude oil	43.1
Gasoline	44.2
Diesel	42.9
Fuel oil	40.9
Coal	25.8
Natural gas	45.9

Energy in the pulp making phase includes the energy use of input chemical production and energy for pulp transportation. Data of input chemicals in the pulp making phase is also taken from the Ecoinvent database. The energy use of such input chemical is calculated using the energy content i from table 4. The relation 1kWh = 3.6 MJ was used to convert energy units. This data is applied in all three phases: pulp making, pretreatment and treatment.

Pretreatment

The enzymatic production route requires a commercial cellulase named monocomponent endoglucanase (Novozym 476, Novozym A/S) to weaken the fibre walls (Pääkkö et al., 2007). This enzyme degrades only the cellulose but not the hemicelluloses, which facilitates refining. Refining is the step before the main enzymatic process because it swells the cell walls, enabling the enzyme to penetrate them. After that, the endoglucanase facilitates the disintegration of cellulosic fiber pulp by increasing its swelling in water. After the enzymatic hydrolysis step comes the second refining to delaminate the fibers. This refining step avoids the clogging during the treatment phase (Ankerfors, 2012).

In the carboxymethylation process, high charge density in the cell wall increases the electrostatic repulsion which leads to fiber swelling. The swelling, in turn, creates lower delamination resistance in the fibers. As a result, cellulose fibrils can be easily separated (Ankerfors, 2012).

In this study, energy use in the pretreatment phase contains energy for input chemical production, direct manufacturing energy and energy in the transport of input materials used in the pretreatment. The energy for chemical production is calculated based on data for each chemical in the Ecoinvent database, the energy content in table 4 (Staffell, 2011) and the conversion factors which will be explained later. The energy use of enzyme production is

based on a study of Liptow et al. (2013). The mass of the material is re-calculated for 1 kg of NFC based on the ratio in two experiments by Pääkkö et al. (2007) and Wågberg et al. (2008).

Detailed calculations of some specific input chemicals are described below. In the enzymatic route, trisodium phosphate is substituted for both monopotassium phosphate and disodium phosphate in phosphate buffer used by Ankerfors, 2012. Monopotassium phosphate has the concentration of 1.44 g/L and disodium phosphate has the concentration of 0.24 g/L (Wikipedia, 2014). Thus the concentration of phosphate buffer is equal to 1.68 g/L, corresponding to 0.042 kg in 25 L (the phosphate buffer's volume is calculated from a study by Pääkkö et al., 2007). That study also mentioned the volume of enzyme as 170 µL, without any information about enzyme concentration. In this study, the mass of enzyme is assumed to be 170 mg. The amount of enzyme, in both volume and mass, is very small compared with other input chemicals, thus the assumption likely does not affect the final results.

In the carboxymethylation route, the concentration of organic solvents (ethanol, isopropanol and methanol) is assumed to be 100% pure. Mass of these solvents is calculated by the equation $m = D*V$, in which m is mass, D is density, and V is volume. Density of ethanol is 0.789 kg/L, that of isopropanol is 0.786 kg/L and of methanol is 0.792 kg/L (Wikipedia, 2014). Volume of all input chemicals is calculated for 1 kg of NFC based on the ratio in the experiment of Wågberg et al. (2008). The mass of acetic acid is also calculated by the equation $m = D*V$, in which D (kg/L) is converted from the concentration 0.1 M (Wågberg et al., 2008). Sodium carbonate is substituted for sodium bicarbonate; its mass is calculated by the equation $m = C*V/(1-C)$; C is the concentration of 4% w/w (Wågberg et al., 2008).

Concerning the manufacturing energy, it covers both electricity and heat. Energy in refining and re-refining steps (in the enzymatic route) is considered to be electricity. Electricity is reported as the corresponding heat needed to produce that amount of electricity, converted with the heat-to-electricity conversion factor of 0.3 for nonrenewable energy source and the factor of 1 for renewable energy source (Arvidsson et al., 2014). A general percentage of renewable and nonrenewable electricity production for Europe of 20.9% and 79.1%, respectively, is applied in the study. This ratio is taken from a report of the electricity production and use in Europe, according to European Environment Agency (2013). Heat use includes incubation and enzyme denaturation (enzymatic route) and energy for heating isopropanol (carboxymethylation route). These conversion factors are used in all three phases: pulp making, pretreatment and treatment.

To calculate the heat needed to change the temperature of the solution, the heat equation is used: $Q = c*m*\Delta T$; Q represents the amount of heat, c is the heat capacity per unit mass of the solution, m is the mass of the solution and ΔT is the change in temperature before and after heating (see table 5). In this study, the temperature before heating is assumed to be equal to standard room temperature at 21°C (Wikipedia, 2014).

Table 5. Heat calculation in the study

Step	c (J/gK)	m (kg)	ΔT	Q (MJ/ kg NFC)
Incubation (enzymatic route)	4.1818 (c for water, Wikipedia)	25 kg buffer + 1 kg pulp = 26 kg (mass of enzyme is 170 mg, being negligible)	50°C – 21°C = 29	3.15
Enzyme denaturation (enzymatic route)	4.1818 (c for water, Wikipedia)	25 kg buffer + 1 kg pulp = 26 kg (mass of enzyme is 170 mg, being negligible)	80°C – 21°C = 59	6.42
Heating isopropanol (carboxy-methylation route)	2.68 (c for iso-propanol, Wikipedia)	m = D*V = 0.768 kg/L*18.18 L = 13.96 kg (D is density of isopropanol, V is volume of isopropanol per kg NFC, calculated from a study of Wågberg et al., 2007)	82.5°C – 21°C = 61.5°C (82.5°C is the boiling temperature of isopropanol)	2.31

Treatment

Micro-fluidizer is a machine to liberate the nanofibrils from the fiber wall fragments. In the machine, pulp is passed through an intensifier pump, followed by an interaction chamber which defibrillates the fibers by shear forces and impacts against the channel walls and colliding streams. Processing with a micro-fluidizer reduces the likelihood of clogs to compare with a homogenizer because it has no in-line moving parts (Spence et al., 2011).

In the study, the energy data in treatment is taken from Ankerfors' experiments in 2012 (2,344 kWh/tonne and 2,221 kWh/tonne in the enzymatic and carboxymethylation routes, respectively). The energy is converted to MJ/kg NFC, and then re-calculated using the heat-to-electricity conversion factor and the ratio in European electricity production.

Transport

Energy for transport covers transport within processes and transport between processes. Data of the transport within processes is calculated for each input material, using the Ecoinvent database version 2.2. Data of the transport between processes depends on each scenario of transport. Transport of pulp and input materials for producing pulp is included in the pulp making phase. Transport of input chemicals for producing NFC is included in the pretreatment phase. The treatment phase does not contain transport.

Table 6 shows the types of vehicles assumed in the study. Trucks are convenient vehicles for both short and long distance. This study focused on the use of two types of truck: medium sized, regional distribution truck and truck with semi-trailer, long distance transport. Railway transport was considered in some scenarios, including two types: diesel and electric trains. Ship was used to transport products oversea, thus large ship was chosen among types of ships. All types of mentioned vehicles and their energy use per metric tonne km (MJ/tkm) were taken from the book *The Hitch Hiker's Guide to LCA* (Baumann and Tillman, 2004).

Table 6. Types of vehicles in this study (Baumann and Tillman, 2004)

Type of vehicle	Characteristic	Energy use (MJ/tkm)
Medium sized truck	14-24 tonnes weight, 8.4-14 tonnes pay load capacity, 10 m length, regional distribution.	1.87
Truck with semi-trailer	40 tonnes weight, 26 tonnes pay load capacity, 18 m length, long distance transport.	0.72
Electric rail car goods train		0.151
Diesel train		0.245
Large ship	> 8000 dwtonnes*, 14 knots** speed	0.216

*Deadweight tonnage (dwtonne) is a measure of how much weight a ship is carrying or can safely carry (Wikipedia, 2014).
**Knot: The knot is a unit of speed equal to one nautical mile (1.852 km) per hour, approximately 0.51 m/s (Bartlett, 2003).

When using truck, the distance was doubled because the truck was assumed to travel two ways, one way full of products and one way empty. However, only one way was considered for train and ship since their load capacity was much higher than truck. Beside the concerned input materials, many other products are transported simultaneously in the containers in both ways, resulting in no empty transport. Data in the transport within processes is collected from Ecoinvent which does not show the exact types of vehicles and their energy use but only some descriptions of the vehicles. Assumptions of the vehicles' types are made (see the section 3.3. Key assumptions). All the vehicles' types and their energy use are taken from the book *The Hitch Hiker's Guide to LCA* (Baumann and Tillman, 2004), see table 6.

In transports between processes, specific distances were presented in each scenario (see table 8). The 8 combinations under table 7 desmonstrate the 8 scenarios. Specific vehicles were assumed for each case (see table 7). Table 7 shows the route way for different scenarios of transport between processes. Transports within processes are the same as in the baseline scenario. Transports between processes, however, are different depending on the assumptions of location and types of vehicle used.

In the transport between processes, Stockholm was chosen as a pulp and NFC manufacturing location in this study. Assumptions of locations are made to calculate the distances (see table 8). Distances are calculated by Searates.com and Google map.

Table 7: Transport between processes

Product	Route way	Vehicle	Code
Wood	Swedish (local) wood	Train, electric	A
		Truck, semi-trailer	B
	Import from Russia	Truck, semi-trailer Train, average Ship, large	C
		Truck, semi-trailer	D
Chemical	Import from Germany	Truck, semi-trailer Ship, large	E
	Import from China	Truck, semi-trailer Ship, large	F
Enzyme (in enzymatic route)	Import from Denmark	Truck, semi-trailer Ship, large	G

Different combinations of distances and vehicles give a number of scenarios:

- Scenario 1: transport within processes + A + E (+G)
- Scenario 2: transport within processes + A + F (+G)
- Scenario 3: transport within processes + B + E (+G) (baseline scenario)
- Scenario 4: transport within processes + B + F (+G)
- Scenario 5: transport within processes + C + E (+G)
- Scenario 6: transport within processes + C + F (+G)
- Scenario 7: transport within processes + D + E (+G)
- Scenario 8: transport within processes + D + F (+G)

The distance of transporting wood was calculated from the wood extraction point in the forest or plantation to the NFC gate (with an assumption that the pulp factory and the NFC factory are the same). In the import case, the distance was the sum of separate roads from Russian forest or plantation to wood factory, from wood factory to the port, from Russian port to Swedish port, and finally from that port to the NFC factory gate. Similar procedures were used for the scenarios of importing chemicals and enzyme from Germany, China and Denmark. Details of the selected locations and the specific distances in each scenario are shown in table 8.

Table 8. Assumptions of transport routes in the transport between processes

Transport route	Description	Assumption	Distance (km)	Note
From local forest to NFC factory	From Swedish forest to NFC factory	Length of Sweden from North to South	1,574 (train) 1,574*2 (truck)	Maximum value
From foreign forest to NFC factory (import)	From Russian forest to Russian factory	From Ukhta to Saint Peterburg	1,715 (train) 1,715*2 (truck)	
	From Russian factory to Russian port	Equivalent to the distance from Stockholm to Nynäshamn port	60*2 (truck)	Nynäshamn is a port near Stockholm, Sweden.
	From Russian port to Swedish port	From Saint Peterburg port to Nynashamn port	730 (ship)	Saint Peterburg is one of the major ports in Russia.
	From Swedish port to NFC factory	From Nynäshamn port to Stockholm	60*2 (truck)	
From enzyme factory to NFC factory (import)	From Danish factory to Danish port	Equivalent to the distance from Stockholm to Nynäshamn port	60*2 (truck)	
	From Danish port to Swedish port	From Copenhagen port to Nynäshamn port	630 (ship)	Copenhagen is one of the major ports in Denmark.
	From Swedish port to NFC factory	From Nynäshamn port to Stockholm	60*2 (truck)	
From chemical factory to NFC factory (import)	From German factory to German port	Equivalent to the distance from Stockholm to Nynäshamn port	60*2 (truck)	
	From German port to Swedish port	From Hamburg port to Nynäshamn port	948 (ship)	Hamburg is one of the major ports in Germany.
	From Swedish port to NFC factory	From Nynäshamn port to Stockholm	60*2 (truck)	
	From Chinese factory to Chinese port	Equivalent to the distance from Stockholm to Nynäshamn port	60*2 (truck)	
	From Chinese port to Swedish port	From Shanghai port to Nynäshamn port	20,651 (ship)	Shanghai is one of the major ports in China.
	From Swedish port to NFC factory	From Nynäshamn port to Stockholm	60*2 (truck)	

3.2.2. Route description

Enzymatic route

To produce 1 kg of dry NFC requires 1 kg pulp, 0.042 kg trisodium phosphate, 1.70E-04 kg of enzyme cellulase and 133 kg deionized water (calculated for 1 kg NFC, based on the experiment by Pääkkö and colleagues, 2007 and assumptions – details are in the section:

Key assumptions). Cellulosic pulp was treated in a nine-step process (Pääkkö et al., 2007) (see figure 8):

1. Cellulose pulp was refined the first time by an Escher-Wyss refiner (Angle Refiner R1L, Escher-Wyss) with 33 kWh/tonne.
2. Enzyme was added to a solution of refined pulp dispersed in phosphate buffer. Enzyme cellulase was used without further purification.
3. The solution was incubated at 50°C for 2 h. To do that, the temperature needs to be increased from room temperature (21°C) to 50°C.
4. After that, the sample was mixed manually every 30 min.
5. Deionized water was used to wash the sample.
6. Because enzyme needs to be denaturated at 80°C for 30 min, the temperature was increased from 21°C to 80°C.
7. The sample was washed again with deionized water.
8. The treated pulp was refined a second time by an Escher-Wyss refiner with 90 kWh/tonne.
9. The solution was added microbicide 5-chloro-2-methyl-4-isothiazolin-3-one in order to prevent bacterial growth.

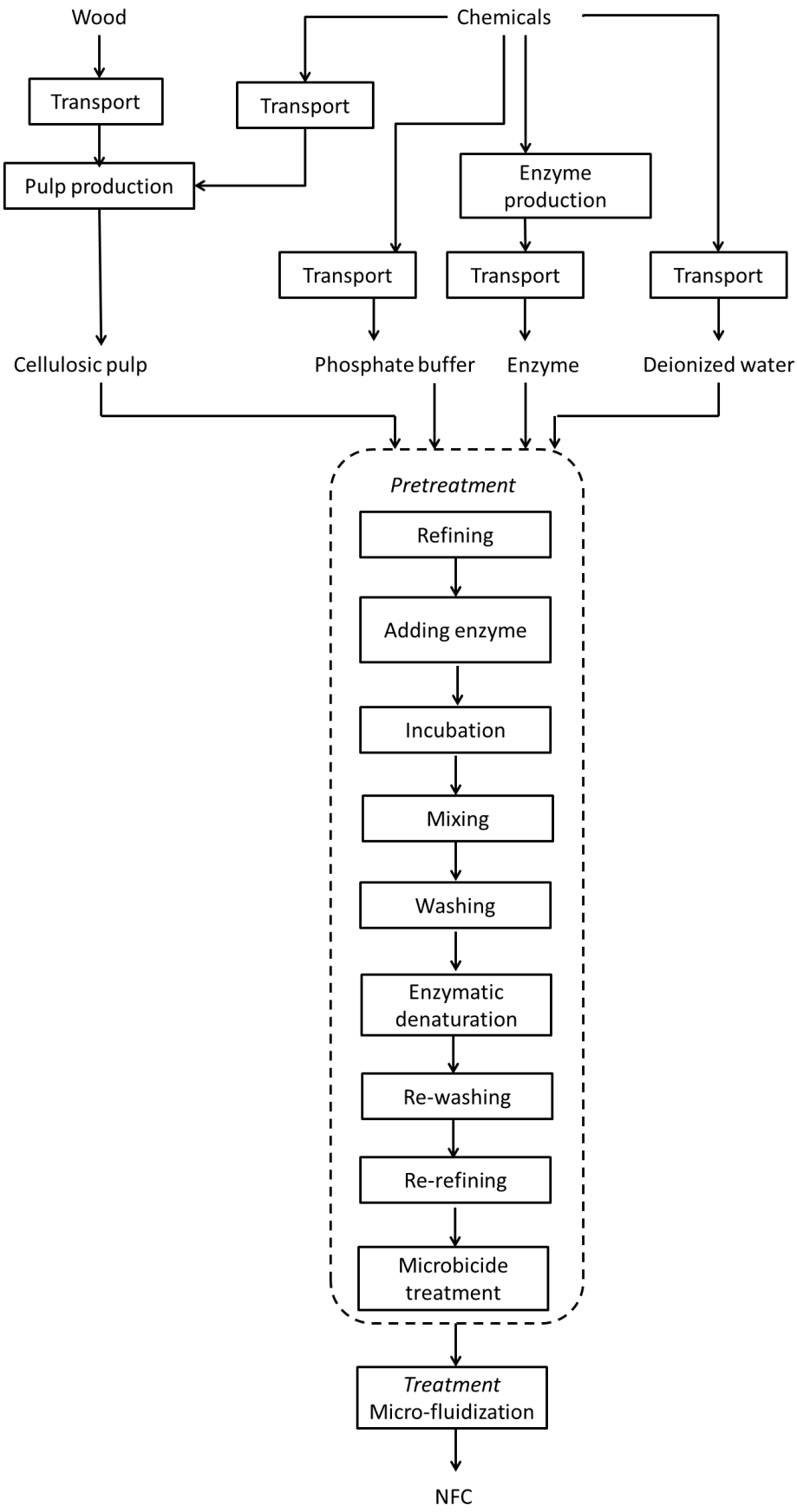

Figure 8. Flow chart describing the enzymatic route.

Carboxymethylation route

To produce 1 kg of dry NFC required 1 kg pulp, 7.17 kg ethanol (100% pure), 90 g monochloroacetic acid, 17.86 kg isopropanol (100% pure), 0.15 kg sodium hydroxide (50% pure), 3.6 kg methanol (100% pure), 0.1 kg acetic acid, 0.76 kg sodium carbonate and 475.76 kg deionized water (calculated for 1 kg NFC, based on the experiment by Wågberg et al., 2008 and assumptions – details are in the section: Key assumptions). Cellulosic pulp was treated in a nine-step process (Wågberg et al., 2008) (see figure 9):

1. Cellulosic pulp was dispersed in deionized water.
2. Pulp, then, was washed in ethanol 4 times with an intermediate filtration step.
3. A solution of monochloroacetic acid in isopropanol was used to impregnate treated pulp.
4. After that, pulp was added in portions to a solution of sodium hydroxide in methanol mixed with heated isopropanol. It is needed to heat isopropanol from room temperature (21°C) to just below its boiling temperature (82.6°C) in a reaction vessel fitted with a condenser.
5. Carboxymethylation reaction occurred for 1 h.
6. Deionized water was used to filter and wash the sample.
7. Then, the pulp was impregnated with sodium bicarbonate (4 wt % solution), for 60 min in order to convert the carboxyl groups to the sodium form.
8. The sample was re-washed with deionized water.
9. Finally, washed pulp was drained on a Büchner funnel.

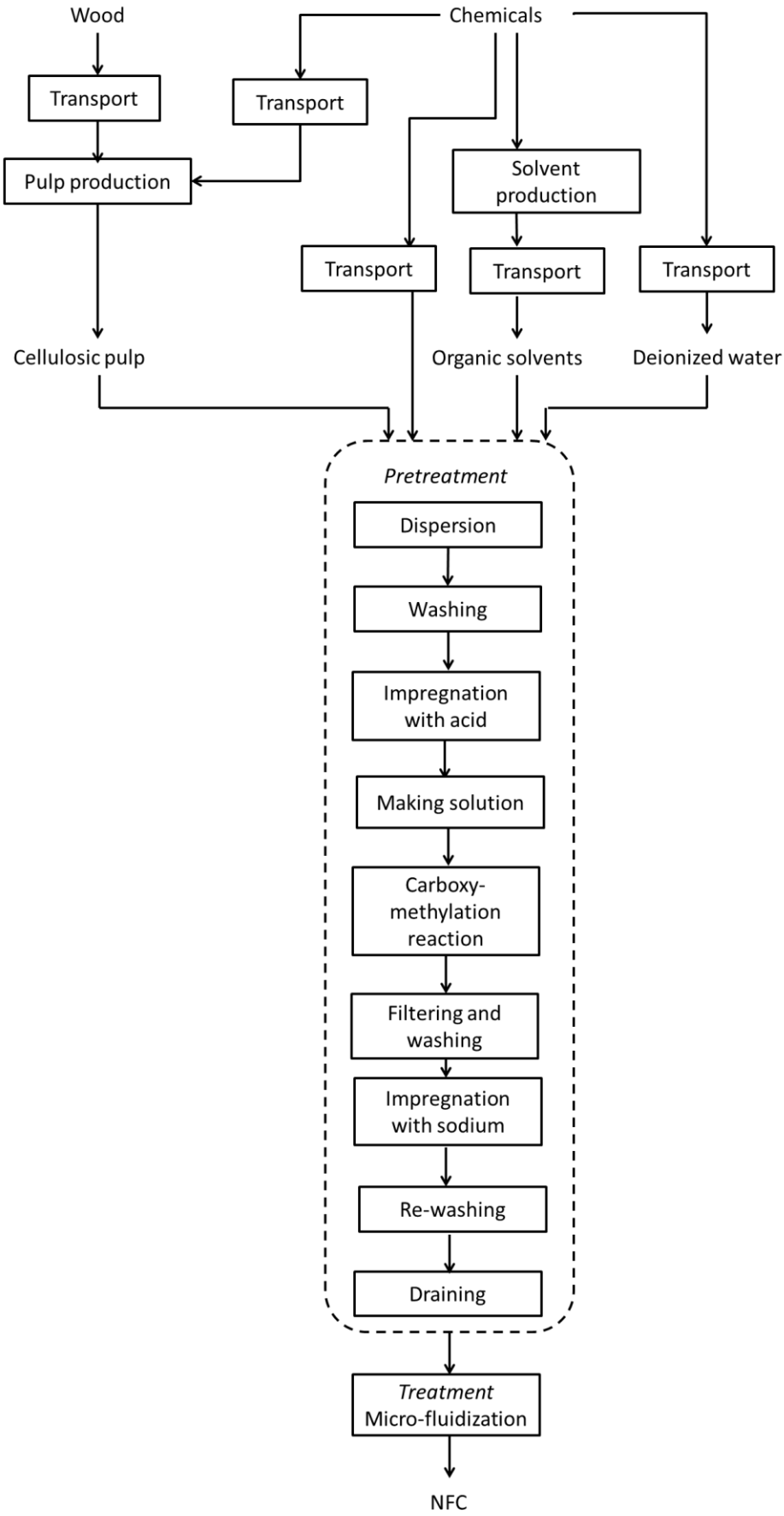

Figure 9. Flow chart describing the carboxymethylation route.

3.3. Key assumptions

Considering the limited energy data on nanocellulose production processes, several assumptions were made to enable the LCEA. Scenarios were made in the sensitivity analysis to test the influence of some of the assumptions.

Pulp making phase

The carboxymethylation processing model in this study is based on experiments by Wågberg (2008), in which the input material was sulfite softwood dissolving pulp with a hemicellulose content of 4.5 % and a lignin content of 0.6 %. The enzymatic route is based on the experiment by Pääkkö et al. (2007), using bleached sulfite softwood with a hemicellulose content of 13.8 % and a lignin content of 1 %. However, to create common models that can be applied in various manufacturing routes now and in the future, LCI of pulp in this calculation is taken from the Ecoinvent database. LCI data from Ecoinvent are for pulp from a mixture of softwood and hardwood, from Scandinavia and other parts of the world. Due to the various input pulps in other experiments, such as delignified sulfate pulp (Li et al., 2013), never-dried TCF bleached softwood sulfite dissolving pulp (Ankerfors, 2012) and unbleached pulp (Spence, 2011), four types of pulps (sulfite, ECF sulfate, TCF sulfate and unbleached) were chosen as scenarios in this study. These four different pulps were assumed to be possible to use in both processes, despite the specific pulps used in the experimental studies. Pulp loss in the process is below 3% (Li et al., 2013), and is therefore disregarded in this study.

Pretreatment phase

Each pretreatment leads to slight changes in chemical structure that likely affects the energy use. However, the major changes related to the modification step to make NFC application products are not included in the system. Therefore, in this report, these differences are assumed to be negligible.

In the enzymatic route, the enzyme used is monocomponent endoglucanases (Novozym 476, Novozym A/S) (Pääkkö et al., 2007, Ankerfors, 2012). Due to the lack of available LCI data for this specific enzyme, this enzyme is substituted with another type of cellulase which is described in the study by Liptow et al. (2013). The total energy use of this cellulase is categorized as heat energy in this study due to lack of data about the amount of energy in each type in the enzyme production. It is also assumed that pure enzyme (100% concentration) was used. Considering the small amount of enzyme in the process (170 µL/kg NFC), the energy for producing enzyme contributes only a small part to the total energy demand of the whole process. Differences in enzyme concentration thus lead to very small changes in the total energy use. Another substitution is phosphate buffer. It was used at a ratio of 11 KH_2PO_4:9 Na_2HPO_4 in the enzymatic pretreatment (Ankerfors, 2012). In this study, it is substituted by Na_3PO_4, for which data is available in the Ecoinvent database.

In the carboxylation route, organic solvents used are assumed to have 100% concentration. Solvent evaporation was considered negligible. In the study, Na_2CO_3 is

substituted for $NaHCO_3$ due to lack of LCI data in the Ecoinvent database. This study does not evaluate environmental impacts, only energy use, and the energy use of producing these chemicals is assumed to be equivalent. Some researchers listed polyelectrolytes and silicon wafers as input materials (Wågberg et al., 2008, Ankerfors, 2012). However, these materials are only included in the modification phase, which is not covered within the system boundary of this study.

Treatment phase

The homogenizer is considered being easily scaled up to industrial production and operated continuously despite its energy intensity and clogging problems. This study assumed that a micro-fluidizer can be commonly used at industrial scale in the period of ten years from now.

Transportation

Different transport scenarios are constructed to account for a range of options for transportation. Since the production of NFC is still at the laboratorial stage, all the distances of trade are assumptions.

Because numbers of data are taken from Ankerfors' experiments at Innventia, the NFC factory in this study is assumed to be located in Stockholm, Sweden (where Innventia is located). The study focuses only on NFC made from wood pulp; hence, it is assumed that NFC is produced from pulp in existing pulp mills.

To construct scenarios for transportation, the most feasible locations (of pulp mills, chemical factories, wood extraction points and ports) are tentatively selected based on currently available information. The transport was carried out by ship, train and truck. Based on the current services of some companies in Russia and Sweden, it is assumed that Russian locomotives are propelled to 50% by diesel and 50% electricity, while these in Sweden are 100% electric locomotives. Deionized water in both processes is assumed to be transported by pipes which contributes a small amount of energy in comparison with other types of transport, thus being neglected in this calculation. Details of the transport scenario assumptions are described in section 3.2.1.

Whole study

- Some common processes such as energy for washing, mixing, producing microbicide, and draining are assumed to be negligible and are excluded in every route. Therefore, it does not affect the comparison. The NFC models in this study are flexible enough to implement these data if and when they become available.
- This study neglects energy for producing and cooling water which is not expected to be a considerable energy use in comparison with, for example, chemical production. However, energy for manufacturing deionized water is included.
- Energy from waste incineration in pulp production is neglected because of lack of data. Energy for NFC storage is not considered since the system's end point is at the formation of 2% w/w NFC.
- NFC produced in each chemical modification process has different surface charge properties. The enzymatic process produces microfibrillated cellulose (Pääkkö et al.,

2007) while the carboxymethylation method creates carboxymethylated cellulose (Wågberg et al., 2008, Li et al., 2013). However, these NFC products are still comparable due to their same fiber consistency of 2% w/w and the lateral dimension of fibers that range from 5 to 20 nm (Pääkkö et al., 2007, Wågberg et al., 2008). After further modification steps, the NFC application products will likely not be comparable depending on their particular properties. This cradle-to-gate analysis stops before the modification steps, therefore the NFC materials of the two processes can be considered similar enough to allow for a comparison.

- The production yield of NFC reaches nearly 100% (Meyer et al., 2012). It is assumed that this efficiency would stay the same at industrial scale. The up-scaling efficiency factor is uncertain at the moment.
- Policy and technology change rates are not considered. Because of the novelty of nanocellulose, all manufacturing methods mentioned in this study are still new and under testing. It is assumed that they will be used at large scale in the next decade.

4. RESULTS

This chapter demonstrates findings of the research: The NFC energy use for two life cycle routes in various scenarios. The energy use of the two different routes in the baseline scenario is analyzed from different approaches, such as energy divided into phases, energy divided into productions and energy divided into types. Other scenarios contain alternatives of input pulp, ethanol sources and transport options.

4.1. Energy calculation

The energy use of NFC production in two different routes is the sum of the energy needed for each input material production, the manufacturing energy in each step of the process and the transport within processes and between processes. Table 9 and 10 show the cradle-to-gate energy use in each step of the two processes. Because the energy needed for transport between processes depends on each scenario, it will be presented later in the transport scenarios (section 4.3.3).

Table 9. Energy use for the enzymatic route

Input materials			
	Energy use (MJ/kg)	**Mass (kg/ kgNFC)**	**Energy use (MJ/kg NFC)**
Bleached sulfite pulp	69.1	1	69.1
Bleached sulfate pulp, ECF	59.2	1	59.2
Bleached sulfate pulp, TCF	58.5	1	58.5
Unbleached sulfate pulp	52.1	1	52.1
Trisodium phosphate	21.4	0.042	0.9
Enzyme cellulase	120	0.00017	0.02
Deionized water	0.0096	133	1.27
Manufacturing process			
Refining			0.34
Incubation			3.15
Enzyme denaturation			6.41
Re-refining			0.92
Micro-fluidization			24.0
Transports within processes			
Bleached sulfite pulp			0.64
Bleached sulfate pulp, ECF			0.59
Bleached sulfate pulp, TCF			0.60
Unbleached sulfate pulp			0.27
Trisodium phosphate			0.002
Enzyme cellulase			0.03
Deionized water			0.64

Table 10. Energy use for the carboxymethylation route

Input materials			
	Energy use (MJ/kg)	**Mass (kg/kg NFC)**	**Energy use (MJ/kg NFC)**
Bleached sulfite pulp	69.1	1	69.1
Bleached sulfate pulp, ECF	59.2	1	59.2
Bleached sulfate pulp, TCF	58.5	1	58.5
Unbleached sulfate pulp	52.05	1	52.1
Deionized water	0.0096	476	4.54
Ethanol from ethylene	42.2	7.17	303
Ethanol from corn	66.0	7.17	474
Monochloroacetic acid	44	0.09	4
Isopropanol	32.7	17.9	584
Sodium hydroxide	18.7	0.15	2.75
Methanol	33.2	3.6	120
Acetic acid	43.1	0.11	4.7
Sodium carbonate	13.8	0.76	10.5
Manufacturing process			
Heat			2.31
Micro-fluidization			22.8
Transport within processes			
Bleached sulfite pulp			0.64
Bleached sulfate pulp, ECF			0.59
Bleached sulfate pulp, TCF			0.60
Unbleached sulfate pulp			0.27
Deionized water			0.09
Ethanol from ethylene			1.12
Ethanol from corn			3.39
Chloroacetic acid			0.063
Isopropanol			4.35
Sodium hydroxide			0.02
Methanol			0.0006
Acetic acid			0.03
Sodium carbonate			0.46

4.2. Baseline scenario

To compare the two routes, one baseline scenario was chosen. In this case, the cellulose source was bleached sulfite pulp which was actually used in Innventia's experiments (Ankerfors, 2012) and in the studies by Pääkkö et al., (2007) and Wågberg et al., (2008). In the carboxymethylation route, ethanol from corn, one of the most common ethanol products on the market, was used. Fermentation ethanol was much more common than ethanol made from petroleum products, according to US Department of Energy's webpage. Transport in this baseline scenario contained the transport of wood from Swedish forest or plantation (local scenario), the import of enzyme from Denmark (in the enzymatic route), and the import of chemicals from Germany (in both routes). Merchandise was transported by truck and ship. Energy in the two life cycle routes was analyzed from three different points of view: energy

divided into main phases, energy divided into production processes, and energy divided into different energy types.

4.2.1. Energy divided into main phases

The total energy use of the whole life cycle was divided into three main life cycle phases: pulp making, pretreatment and treatment. Each phase contains the production and transportation of input materials.

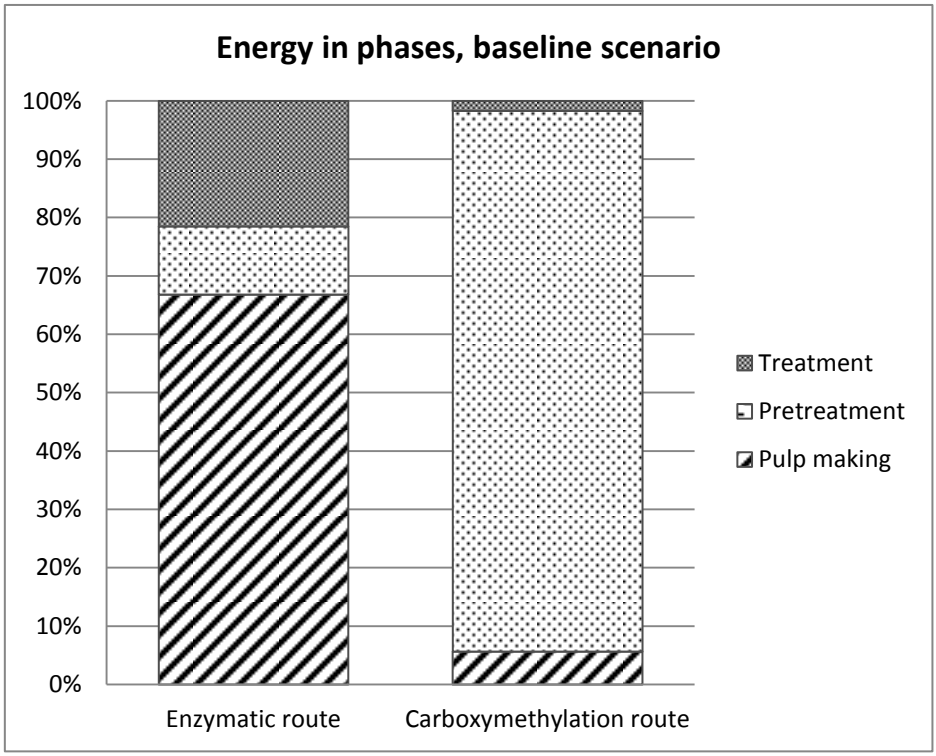

Figure 10. Energy use divided into main life cycle phases for the two compared routes, baseline scenario

The chart in figure 10 indicates the shares of each main phase in the two processes in terms of energy use. The pulp making phase, which includes both the pulping process and the transport of wood, was about 67% of the total energy use, becoming the largest phase in the enzymatic route. In that route, treatment and pretreatment phases contributed approximately 21% and 12%, respectively. Transport of chemicals was included in the pretreatment phase. A completely different energy use distribution is shown for the carboxymethylation route in figure 10, with nearly 93% energy use belonging to the pretreatment phase. The pulp making phase and the mechanical treatment phase contributed much less to the energy use.

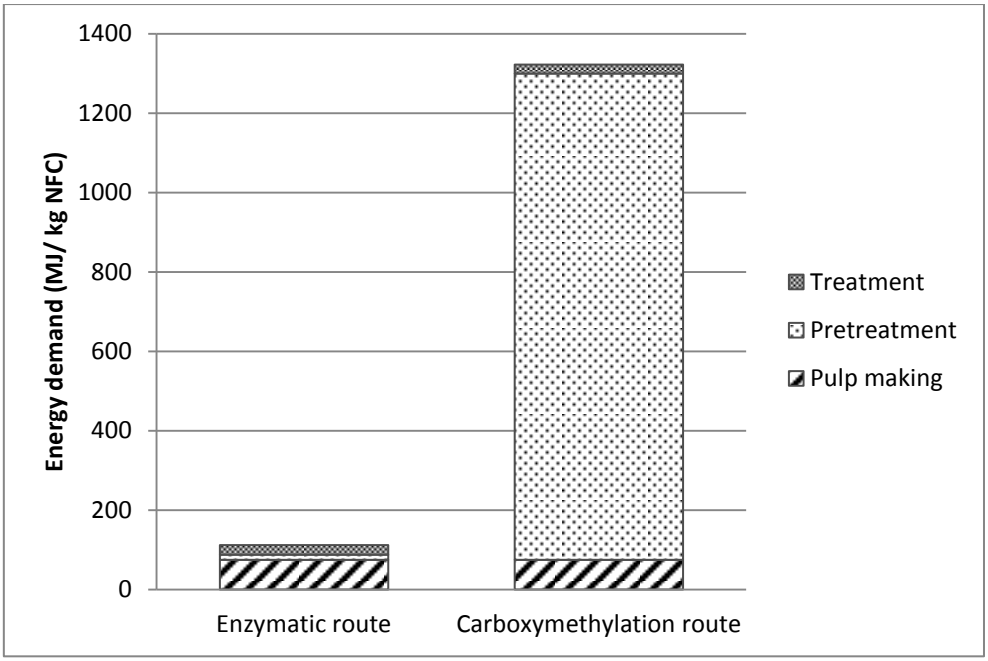

Figure 11. Energy use in absolute terms for the two routes, baseline scenario

Another chart type reveals the absolute amount of life cycle energy use in the two routes (figure 11). In total, using the enzymatic route required about 112 MJ per kg of NFC, while the carboxymethylation route required more than ten times more, reaching nearly 1,323 MJ per kg of NFC. Since this baseline scenario focused only on one type of input pulp, sulfite pulp, and the two methods use the same treatment process, the differences in energy came from the pretreatment phase, which is clearly shown in figure 11.

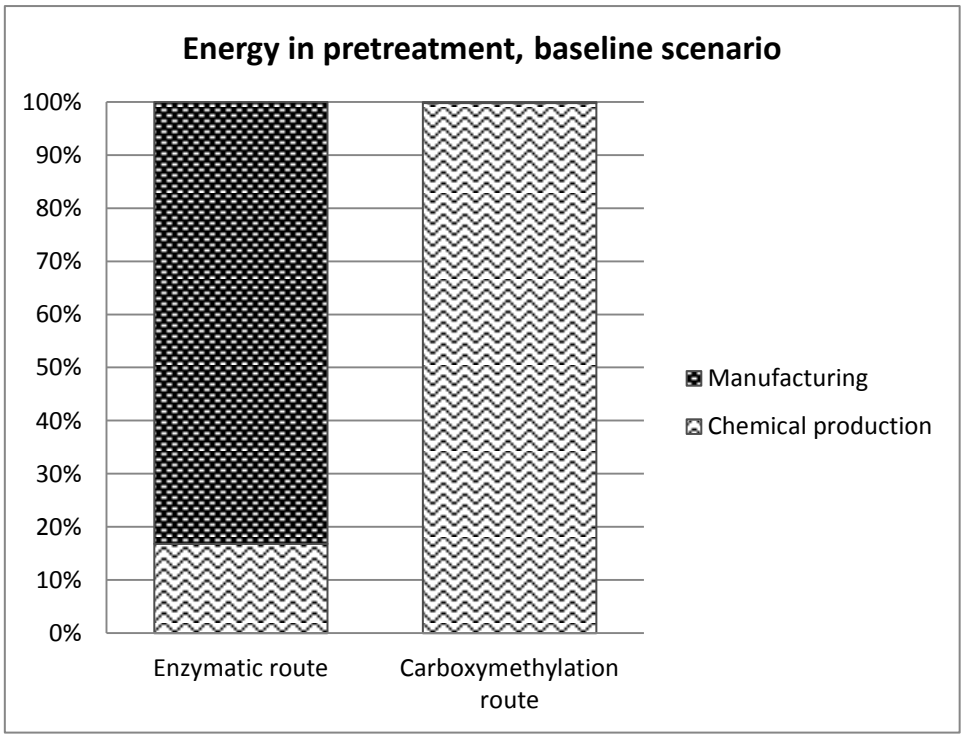

Figure 12. Energy in pretreatment for the two investigated routes, baseline scenario

Figure 12 demonstrates energy use in the pretreatment phase of the two methods, divided into two separate processes: direct energy use from manufacturing of NFC and energy for producing input materials used in this phase. It can be seen that most energy use in the enzymatic pretreatment (83%) comes from mechanical processes, including machine operation and heating. An opposite picture was observed for the carboxymethylation process, with over 99% energy use in chemicals production, such as the making of organic solvents and inorganic compounds. Specifically, about 40% of energy came from ethanol production, more than 48% from isopropanol production, and more than 10% from the making of methanol. The rest that accounted for only below 2% was the energy for making inorganic input materials (see figure 13). The pie chart in figure 13 does not include the transports of all chemicals which were assumed the same; therefore this does not affect the comparison.

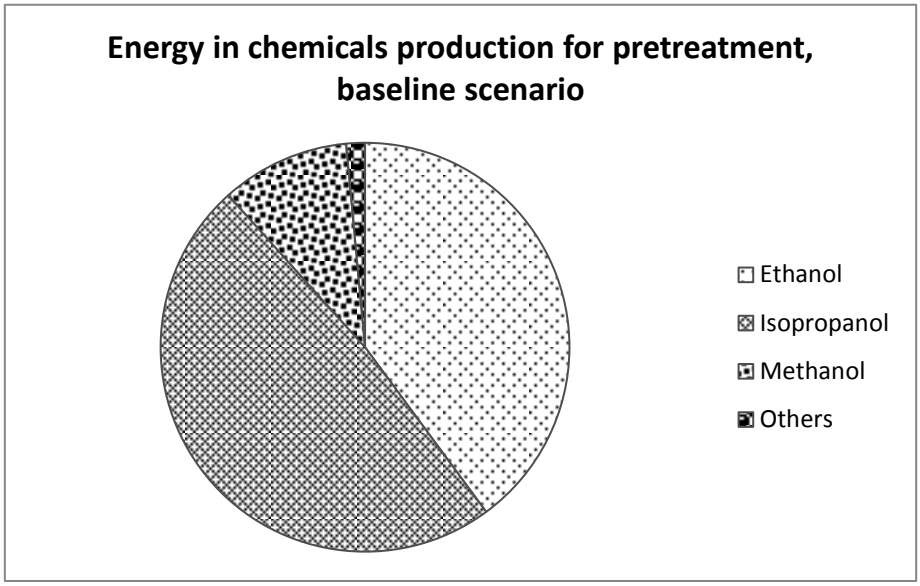

Figure 13. Energy use of the chemicals production for the pretreatment phase in the carboxymethylation route, baseline scenario

4.2.2. Energy divided into production processes

This section concerns a division into separate production processes. In this division, because of its large share, energy use for making input materials was categorized into two smaller groups: pulp production and chemicals production. Consequently, there are four production processes involved in this assessment. The bar chart in figure 14 shows the energy use of each of these processes.

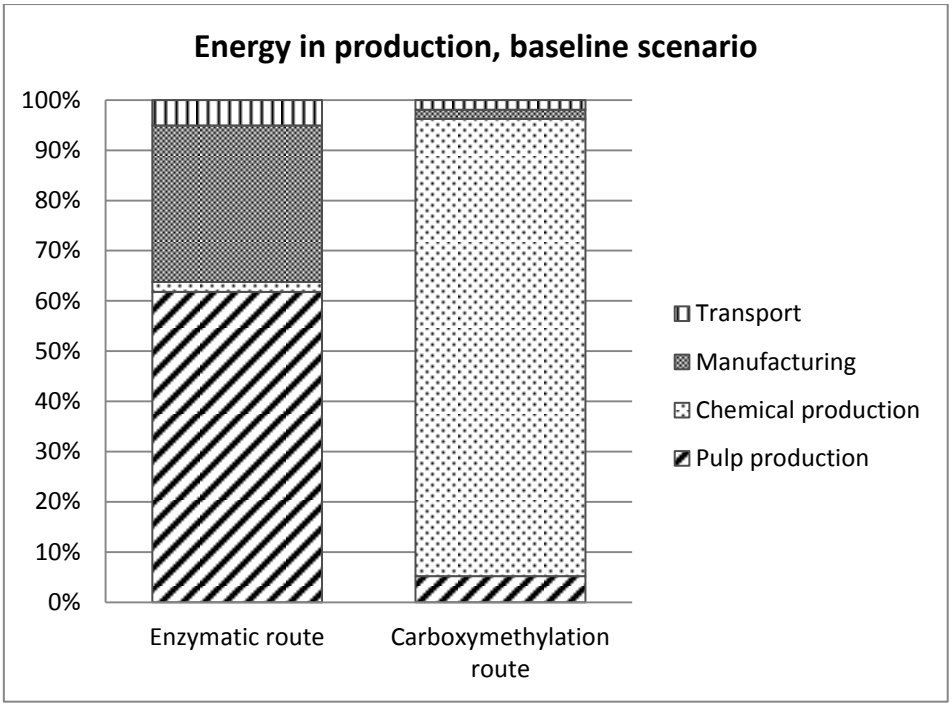

Figure 14. Energy use divided into production processes for the two investigated routes, baseline scenario

For the enzymatic route, energy for pulp production had the largest part (about 62%), followed by the energy for NFC manufacturing (about 31%). For the carboxymethylation route, input chemicals in NFC production had the highest energy demand (approximately 91%). This result was consistent with the previous main phase analysis because the production processes correspond to the main phases.

4.2.3. Energy divided into different types

This section compares the share of five types of energy (biomass, heat, resource, electricity and fuel) in the two production routes.

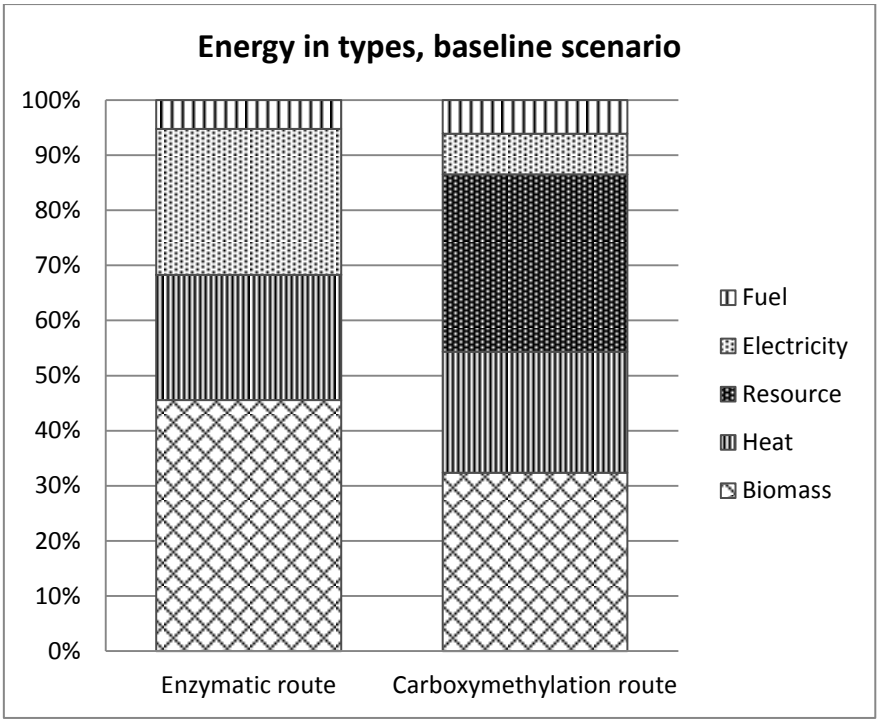

Figure 15. Energy use divided into energy types
for the two investigated routes, baseline scenario

Energy in biomass has a big part in both processes; contributing about 46% in the enzymatic route and around 32% in the carboxymethylation route (see figure 15). In the enzymatic route, most energy in biomass is from pulp. The carboxymethylation route contains the same amount of pulp's biomass, but the biomass energy from the corn-based ethanol is more than 7 times higher than the biomass from pulp shown in table 11.

The bar chart in figure 15 indicates around 22% of heat in the total energy use in the both two routes. Table 11 shows that in the enzymatic route, heating during the pulp production and heating required for the manufacturing steps in the NFC production (incubation and enzyme denaturation) make the big parts in the total heat. Differently, a large amount of energy refers to heating in the productions of the organic solvents in the carboxymethylation route.

In carboxymethylation route, above 32% of the total energy is resource energy while this type of energy contributes a very small part in the other route, nearly 0.009% (figure 15). Most resource energy in the carboxymethylation route comes from isopropanol, an organic solvent in the NFC production (see table 11).

Electricity accounts for nearly 27% in the enzymatic route, but it has only above 7% of the total energy in carboxymethylation route (figure 15). Manufacturing processes, especially the micro-fluidization in the treatment phase, have a large share in total electricity use. Micro-fluidizing step requires approximately 81% of the total electricity use in the enzymatic route, and about 23% of that in the carboxymethylation route (table 11). Together with the micro-fluidization, the productions of isopropanol and ethanol also use much more electricity than other processes in the carboxymethylation route (table 11).

Fuel, in both routes, is seen as a small part, contributing around 5% of the total energy in the enzymatic route and about 6% of the total energy in the carboxymethylation route (figure 15). Fuel energy includes both the input fuel in each material's production and the fuel for transports. Table 11 indicates that the transport between processes requires more energy than the transport within processes in the both two routes. It also shows a high amount of fuel energy in the production of the organic solvents (ethanol and methanol) in the carboxymethylation route.

Table 11: Details of the energy use divided into energy types for the two investigated routes, baseline scenario

	Enzymatic route (MJ/kg NFC)		Carboxymethylation route (MJ/kg NFC)	
Biomass	Bleached sulfite pulp	50.94	Bleached sulfite pulp	50.94
	Phosphate buffer	0.002	Ethanol from corn	376.12
			Isopropanol	0.8
			Sodium carbonate	0.0001
Heat	Bleached sulfite pulp	15.32	Bleached sulfite pulp	15.32
	Phosphate buffer	0.39	Deionized water	0.12
	Cellulase	0.02	Ethanol from corn	61.04
	Deionized water	0.03	Chloroacetic acid	0.98
	Incubation	3.15	Isopropanol	121.5
	Enzyme denaturation	6.41	Sodium hydroxide	0.02
			Methanol	85.66
			Acetic acid	1.47
			Sodium carbonate	2.16
			Heating isopropanol	2.31
Resource	Bleached sulfite pulp	9.61E-05	Bleached sulfite pulp	9.61E-05
	Phosphate buffer	0.009	Isopropanol	425.35
			Sodium carbonate	0.06
Electricity	Bleached sulfite pulp	2.82	Bleached sulfite pulp	2.82
	Phosphate buffer	0.25	Deionized water	4.43
	Deionized water	1.24	Ethanol from corn	18.65
	Refining	0.34	Chloroacetic acid	1.98
	Re-refining	0.92	Isopropanol	35.95
	Micro-fluidization	24.02	Sodium hydroxide	2.72
			Methanol	2.76
			Acetic acid	1.34
			Sodium carbonate	4.15
			Micro-fluidization	22.76
Fuel	Bleached sulfite pulp	0.02	Bleached sulfite pulp	0.02
	Phosphate buffer	0.25	Ethanol from corn	17.79
	Transport within processes	0.67	Chloroacetic acid	1.04
	Transport between processes	4.96	Methanol	31.14
			Acetic acid	1.89
			Sodium carbonate	4.1
			Transport within processes	9.05
			Transport between processes	16.13

In brief, the pulp making phase and the related pulp production contributed the most to the energy use for the enzymatic route. The pretreatment phase and the production of input materials, especially organic solvents in this step, had the largest contribution to the energy use in the carboxymethylation route. When it comes to types of energy, biomass and heat presented a significant share in both processes. Electricity was quite high in the enzymatic process but stayed small in the other route. Both routes required a small part of fuel in

comparison with other types of energy. Above all, the results in figure 11 show that the enzymatic route has a much lower energy use than the carboxymethylation route.

4.3. Scenario analysis

In addition to the baseline scenario, which is considered most likely, possible options of input pulps, ethanol production and transport routes will be discussed and assessed in this section. These factors were selected due to not only their high energy use based on the results for the baseline scenario, but also the potential for substitution. In each scenario, only one factor was changed, while other factors were kept the same as in the baseline scenario.

4.3.1. Pulp scenarios

The previous section illustrated that pulp production has the biggest energy demand in the baseline scenario of the enzymatic route. It can be seen from figure 16 that sulfate pulp requires less energy than sulfite pulp. In fact, sulfate pulp (Kraft pulp) is more commonly used on the market than sulfite pulp. The global production in 2000 of Kraft pulp was 117,000,000 tonnewhile that of sulfite pulp was 7,000,000 tonne (Sixta, 2006). In this study, the energy requirement when using bleached sulfite pulp was the highest (about 75 MJ/kg NFC), followed by that of bleached sulfate (around 64 MJ/kg NFC). Unbleached pulp required the least energy in total (about 57 MJ/kg NFC).

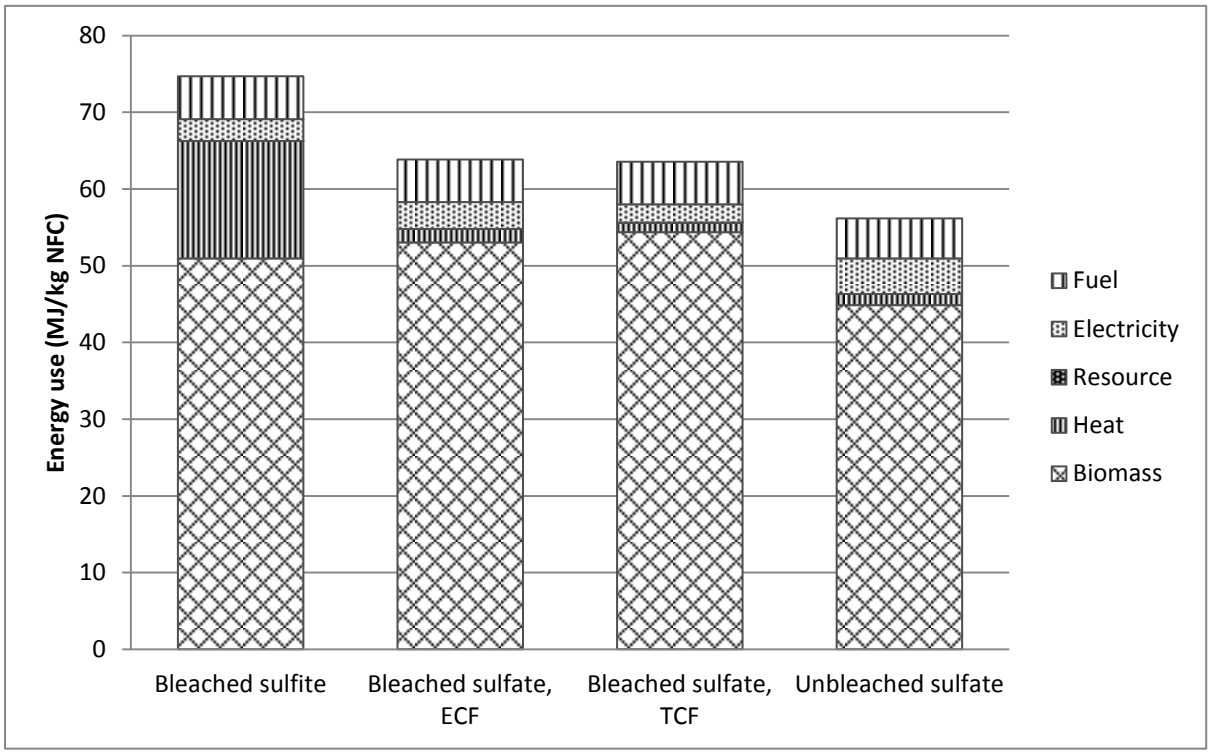

Figure 16. Energy use for the pulp production part of the NFC production, pulp scenarios

Bleaching pulp is the whitening process, sometimes causing environmental concern by the release of chlorinated organic compounds into the environment. The harmful conventional bleaching process, then, has often been abandoned and replaced by chlorine-free processes, ECF and TCF. Figure 16 showed the similarity in energy use of ECF and TCF sulfate pulps, which were 64.68 MJ and 64.03 MJ per kg of pulp, respectively. TCF, for

which all chlorine is removed from the pulp, is considered more environmentally friendly than ECF which does not fully eliminate chlorinated organic compounds from the effluent. Interestingly, in this case, TCF production requires even less energy than the ECF one, thereby being environmentally preferable also in that respect.

Regarding the types of energy, biomass contributed the most in every case, contributing between 68% and 85% of the energy use of pulp production. Heat was high for sulfite production only, as a result of the wood heating process. Electricity values varied from above 2 to below 5 MJ, playing a small part in the energy use. Fuel added approximately 5 or 6 MJ to the total energy, also constituting a small part in the four cases.

4.3.2. Ethanol scenarios

According to the baseline scenario results, chemicals production, especially the production of organic solvents, contributed with the most energy use for the carboxymethylation route. This can be explained by the large amount of organic solvents used in the pretreatment phase, as well as the energy content of such organic compounds.

Ethanol production is responsible for 39% of energy in the pretreatment phase (excluding transportation). Ethanol can be made from different sources. This study looks into the two common processes of making ethanol from corn fermentation and fossil ethylene hydration.

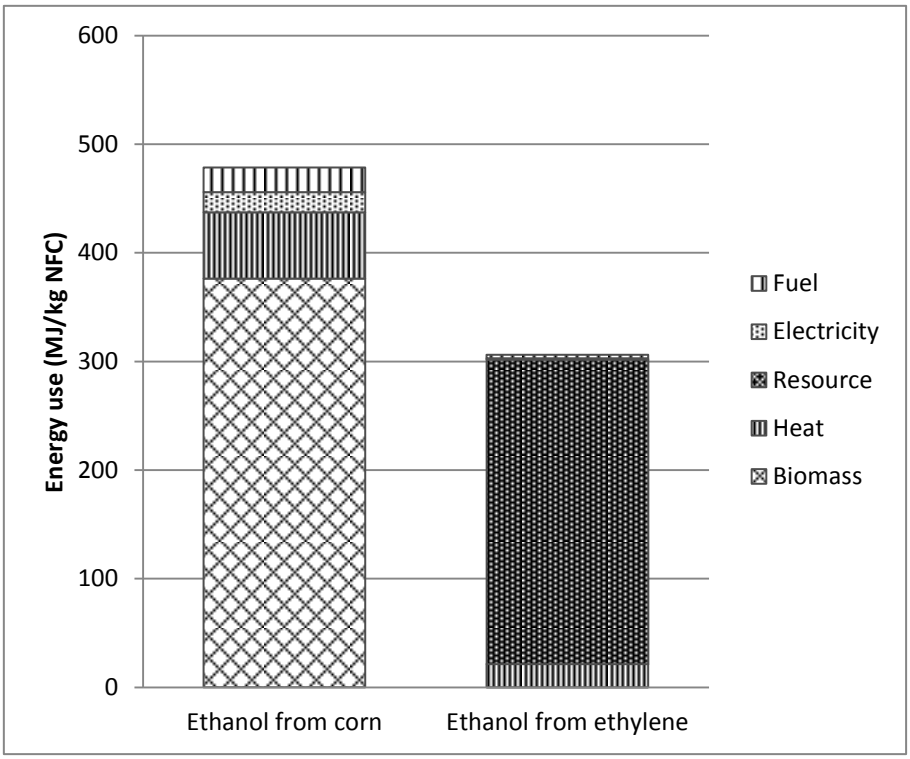

Figure 17. Energy use for the ethanol production part of the NFC production, carboxymethylation route, ethanol scenarios

The differences in energy use when applying the two ethanol production processes are shown in figure 17. In total, the energy use of producing ethanol from corn is about 478 MJ/kg NFC. In comparison, approximately 306 MJ/kg NFC is needed to make ethanol from

ethylene. The biomass-based ethanol process required about 1.5 times more than the fossil-based one. Feedstock energy is the biggest part in the energy use of both ethanol from corn and ethanol from ethylene. Nearly 79% of energy in the corn-ethanol production was biomass, while most energy in ethylene-ethanol (92%) was resource energy. Big parts of this resource energy are crude oil (64%) and natural gas (34%). Although a transition to fossil-based ethanol would reduce the energy use to some extent, a larger share of the energy use would be of fossil origin. The reduction is also not large enough to make the carboxymethylation route better than the enzymatic route from an energy use point of view.

4.3.3. Transport scenarios

Eight transport scenarios were constructed, concerning local transport and import, and the use of three types of vehicles (truck, train and ship). The total energy for transport includes the energy use for transports within production processes and between production processes. Energy in transportation of each life cycle route is illustrated in the graphs in figures 18 and 19. Both routes were analyzed with eight transport scenarios. Transport scenarios were outlined in table 7 and 8; 8 combinations illustrating 8 scenarios were described under table 7, in section 3.2.1.

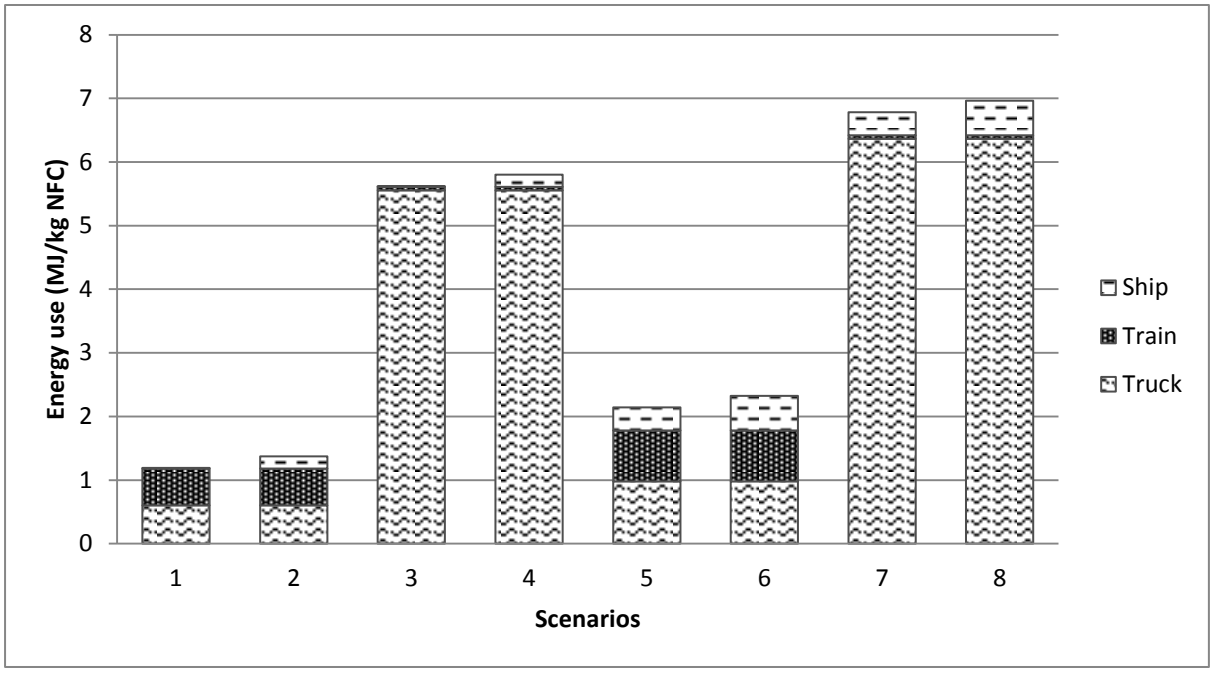

Figure 18. Energy use in the transport part of the NFC production, enzymatic route; transport scenarios

In the enzymatic route (figure 18), fuel for trucks contributed the most in most cases. However, it was more obvious in scenarios 3–4 and scenarios 7–8. These results indicate high energy use of transporting by trucks for a long distance. These cases included very long distances (around 1,500 – 1,700 km) from the forest or plantation to the factory. Truck transport in the other cases covered shorter distances between the factory and the port (below 100 km). Moreover, double distances applied for the transport by truck made it more energy-intensive. Figure 18 also indicates that transport by train requires much less energy than transport by truck.

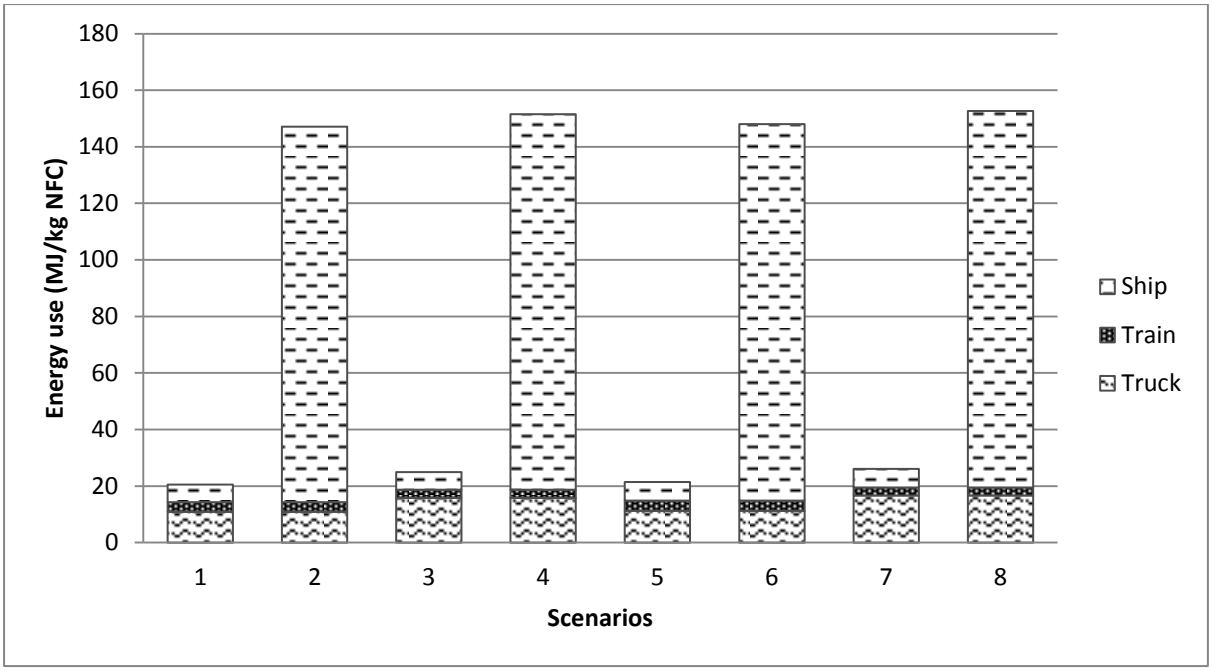

Figure 19. Energy use in the transport part of the NFC production, carboxymethylation route, transport scenarios

Figure 19 points out the ship transport as the most energy demanding factor in the carboxymethylation route in four scenarios: 2, 4, 6, 8. All of these referred to chemicals import from China. The energy use of large ships is 0.216 MJ/tkm, being the second lowest among selected vehicles in the study (the lowest is electric train with 0.151 MJ/tkm). Despite the low energy use per tkm of the vehicle, energy in ship transport in this case is still high resulting from a very long distance (over 20,000 km). Furthermore, the significant amount (in weight) of chemicals needed for the production of NFC for this route (nearly 30 kg/kg NFC) was much higher than that in the enzymatic route (0.042 kg/kg NFC).

4.3.4. Overview of scenario analysis

Concerning every option mentioned in the study including types of pulp, types of ethanol, methods of pretreatment and routes in transportation, there are 32 possible combinations in the enzymatic route and 64 in the carboxymethylation (see figure 20).

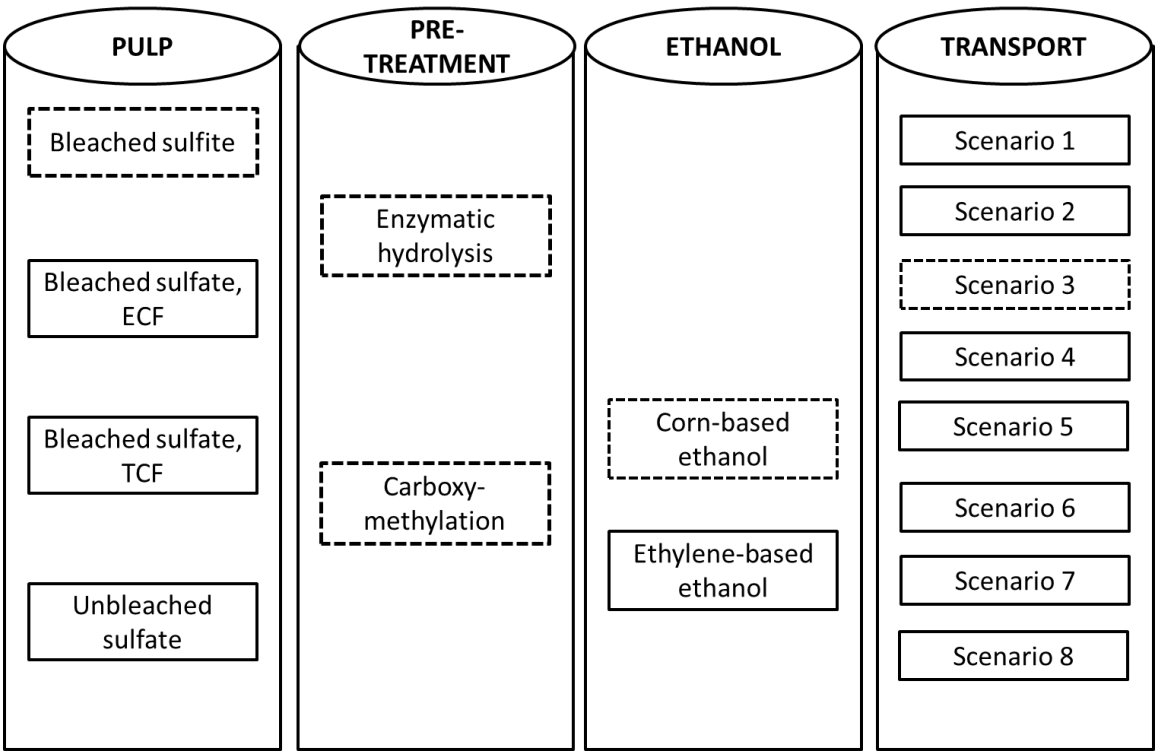

Figure 20. Summary of scenarios.
The options inside dashed line frames indicate the baseline scenario

Among all these 96 cases, the lowest energy demand scenario refers to unbleached pulp, enzymatic process and transport scenario 1. The most energy-intensive scenario corresponds to sulfite pulp, carboxymethylation process using ethanol from corn and transport scenario 8. In this study, NFC's life cycle energy therefore ranges from 90 MJ/kg NFC for the best case (which was for the enzymatic route) to 1,450 MJ/kg NFC for the worst case (which was for the carboxymethylation route).

5. DISCUSSION

In this chapter, the results in the previous chapter are discussed and put into a larger context that includes results from other related studies. The first research question about the energy use of the two routes is largely answered in the previous chapter, and the second question about factors influencing the energy use will be answered in this chapter.

5.1. Comparing energy use in the treatment phase

In recent years, energy use has been an issue of high concern to NFC producers. While outstanding properties of this new material have been proven, it is a question of whether it is worth the cost to commercialize such a material. To find the answer, a lot of studies calculating the energy use of NFC production have been conducted. However, most studies focused only on the producer's main interest: the manufacturing energy, because it directly affects the cost when scaling up to industrial production.

For manufacturing energy, the treatment process has been considered the most energy-intensive step. Siró (2010), Klemm (2011) and Lavoine (2012) suggested that the major impediment for commercial success has been the very high energy use as a result of the

numerous passes to obtain well-defibrillated fiber suspensions in homogenization. Khalil also mentioned the high energy demand regarding the mechanical fibrillation in his study in 2014.

The amount of energy needed for the treatment phase was reported to be high, equivalent to 97.2 MJ/kg NFC in Turbak's experiment (1983). In other research, it was reported similarly high, at the level of 72-108 MJ/kg NFC (Siró et al., 2010) and 43.2-252 MJ/kg NFC (Klemm et al., 2011).

Efforts to solve this problem has achieved energy savings when introducing different pretreatment methods. Physical pretreatment using a beater reduced the energy in treatment to 8.23–12.81 MJ/kg NFC (Spence et al., 2011). Applying enzymatic hydrolysis and refining in pretreatment saved a large amount of energy in the treatment phase, resulting in 6.44 MJ/kg NFC and 8.44 MJ/kg NFC (Ankerfors, 2012) and 5 MJ/kg NFC (Klemm et al., 2011). Carboxymethylation as pretreatment decreased drastically the energy in treatment to 8 MJ/kg NFC (Ankerfors, 2012) and 2 MJ/kg NFC (Klemme et al., 2011, Ankerfors, 2012). The energy demand for the treatment process reached 3.6–14.76 MJ/kg NFC (SUNPAP, 2012) due to the introduction of TEMPO (oxidation) pretreatment. Table 12 lists pulp-based NFC studies in the field of energy use in the treatment process.

Table 12. Energy use in the treatment phase

Input material (Pulp)	Pretreatment	Treatment	Treatment description	Output material (NFC)	Energy in treatment (MJ/kg NFC)	Sources
Bleached sulfite	None	Homo-genizer	5 - 10 passes	gel-like	97.2	Turbak et al., 1983
Bleached sulfate	None	Homo-genizer	5 - 10 passes	gel-like	43.2 – 252	Klemm et al., 2011
-	None	-	-	-	72 - 108	Siró et al., 2010
Softwood dissolving pulp, 2%	TEMPO	Homo-genizer	2 passes, 150 MPa	2%	14.76	Meyer et al., 2012
Softwood dissolving pulp, 4%	TEMPO	Homo-genizer	2 passes, 150 MPa	4%	5.2	Meyer et al., 2012
Bleached sulfate/sulfite	Carboxy-methylation	Homo-genizer	-	gel-like	2	Klemm et al., 2011
Bleached sulfate/sulfite	Enzymatic hydrolysis	Homo-genizer	-	gel-like	5	Klemm et al., 2011
Bleached softwood sulfite, TCF, 2%	Enzymatic hydrolysis	Micro-fluidization	1 pass, 160 MPa	2%	8.44	Ankerfors, 2012
Bleached softwood sulfite, TCF, 2.5% - 3%	Enzymatic hydrolysis	Micro-fluidization	1 pass, 150 MPa	2.5% - 3%	6.44	Ankerfors, 2012
Bleached softwood sulfite, TCF, 2%	Carboxy-methylation	Micro-fluidization	1 pass, 160 MPa	2%	8	Ankerfors, 2012
Bleached softwood sulfite, TCF, 8%	Carboxy-methylation	Micro-fluidization	1 pass, 160 MPa	8%	2	Ankerfors, 2012
Hardwood, bleached/unbleached	Beater	Micro-fluidization	1 - 20 passes, 69 MPa	-	8.23 – 11.23	Spence, 2012
Hardwood, bleached/unbleached	Beater	Micro-fluidization	5 passes, 138 MPa	-	9.18	Spence, 2012
Hardwood, bleached/unbleached	Beater	Micro-fluidization	5 passes, 207 MPa	-	10.38	Spence, 2012
Hardwood, bleached/unbleached, 3%	Beater	Micro-grinder	9 passes	3%	12.81	Spence, 2012
Softwood dissolving pulp	TEMPO	Rotor/ Stator	-	-	3.6	Meyer et al., 2012

The baseline scenario in this study applied the results from Ankerfors' experiments at Innventia, with 8.44 MJ/kg NFC and 8 MJ/kg NFC in the treatment phase of the enzymatic route and the carboxymethylation route, respectively. According to Ankerfors, in comparison with Turbak's conventional mechanical process, the introduction of enzymatic pretreatment saved 91% energy use in the treatment phase. Moreover, it is expected to save even more energy (93%) by the increase of the material's concentration and the decrease of pressure. Similarly, nearly 92% reduction of treatment energy was reported in the carboxymethylation process. The promising potential of 98% saving may be achieved when using pulps with a significantly higher concentration. In conclusion, it seems that adding pretreatment reduces the energy use of the treatment process by approximately one order of magnitude. However, as shown in figure 10, this comes at a high energy use in the pre-treatment process, particularly for the carboxymethylation route.

5.2. Comparing energy use in the life cycle process

Despite the impressive energy use savings thanks to the technical development of pre-treatment, direct manufacturing energy in the treatment phase is only the tip of the iceberg from a life cycle perspective. It contributes 7.6% of the total energy in the enzymatic route and only 0.6% of the total energy in the carboxymethylation route. Currently, few other LCA studies of nanocellulose exist, mainly the nanocellulose LCA by Li et al. (2013) and the LCA of cellulose nanowhiskers by Figueirêdo et al. (2012).

The total energy requirement for the NFC production using chloroacetic acid etherification in combination with homogenization was reported as 64.9 MJ per 10 g micro-fibrillated cellulose by Li et al. (2013). This process is similar to the carboxymethylation route in this paper, except there are some differences in the amount of input chemicals needed in the pretreatment phase, as well as the machine used in the treatment phase. Moreover, particular transport distances should be different considering that Li's experimental lab is located in the US and this study's NFC factory is assumed to be located in Sweden. When comparing results, the uncertain efficiency factor related to industrial scale used by Li et al. (2013) is ignored in this study. To calculate the energy use in industrial scale, Li et al. only used 8.3% of the direct electricity inputs from the baseline model (lab scale), and inputted and outputted only 20% of the ethanol and isopropanol solvent volumes (Li et al., 2013). Due to the uncertainty of these assumptions, this study excluded the efficiency factor.

By doing that, the corresponding total energy in the study by Li et al. (2013) reaches 6,490 MJ/kg NFC. The number is much higher than the life cycle energy of NFC produced by carboxymethylation in this study (around 1,323 MJ/kg NFC) because (1) Li et al. produced NFC from delignified kraft pulp which differs from the bleached sulfite pulp assumed in the baseline scenario, (2) the method by Li et al. requires a two to four times higher amount (in weight) of some energy-intensive chemicals than in this study, (3) the homogenizer (in the experiment of Li et al.) may require a different amount of energy in comparison with the micro-fluidizer (in this study), (4) different transport routes may result in different numbers, and 5) US electricity production in the work by Li et al. is not the same as the European grid electricity assumed in this study. The differences in energy use for the treatment step and transports in the study of Li et al. and this study are not clear since Li et al. (2013) shows only the cradle to gate energy, without detailed energy use in each type. It is difficult to

compare and contrast the work of Li et al. with this study, even if the production processes are similar.

Besides the carboxymethylation route, three other NFC fabrication routes are mentioned in the paper by Li et al. (2013), with the total energy use ranges from 34.7 MJ to 176.1 MJ per 10 g NFC. Without the uncertain up-scaling efficiency factor mentioned on the paper of Li et al., these numbers can be recalculated to an energy use of NFC ranging from 3,470 to 17,610 MJ per kg NFC (this study does not include the efficiency factors, see Key assumptions section). Another study reported an energy use at 1,800 kJ and 15,944 kJ per g cellulose nanowhiskers for two different production systems using different input fibers (Figueirêdo et al., 2012). Disregarding the efficiency factor of commercial production, the results correspond to 1,800–15,944 MJ/kg NCC. The maximum life cycle energy in this study was 1,450 MJ/kg NFC. Even for the carboxymethylation route, and particularly for the enzymatic route, the results of this study indicate lower energy use for NFC than previous studies.

Despite the high energy intensity reported in the studies by Li et al. (2013) and Figueirêdo et al. (2012), nanocellulose (NFC and NCC) still is at a low level compared with the energy demand of other nanomaterials, as shown in a report of Kim and Fthenakis (2012). In this study, NFC, even in the worst case (1,450 MJ/kg NFC), apparently needs much less energy than typical carbon nanotubes, which can have an energy demand as high as 1,000,000 MJ/kg (Kim and Fthenakis 2012). NFC produced with the enzymatic route, which has an energy use of 112 MJ/kg in the baseline scenario, has a very low energy use compared to other nanomaterials, approximately the same energy use as polyprolylene (Kim and Fthenakis 2012).

5.3. Factors affecting NFC's energy use

This section points out main aspects that can help developers and decision-makers in reducing energy use. The energy demand of wood-based NFC depends on many factors during the whole production route, from input materials, through the manufacturing stage, to output products with different characteristics. Figure 21 illustrates five main factors that significantly affects the total energy of NFC production. The scenarios of alternative pulps, pretreatment methods and transport routes in the previous chapter prove the importance of these factors (see section 4.3).

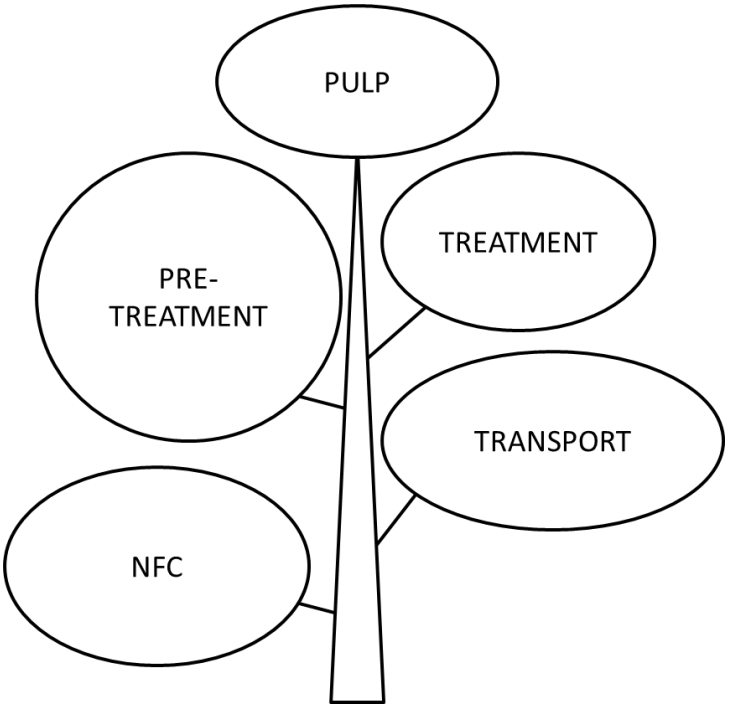

Figure 21. Main factors affecting NFC energy use

Regarding pulp options, a number of aspects should be considered, including whether the sources are pulp wood or waste paper, softwood or hardwood, and whether the type of pulp is sulfite or sulfate, ECF or TCF, bleached or unbleached. In addition to this, characteristics of pulp, such as concentration, lignin fraction and hemicellulose fraction may have an impact (see figure 22). Characteristics and impacts of various types of input pulp, such as form, dimensions, moisture content, Kappa Number and hemicelluloses content were analyzed in a study of Zimmerman et al. (2010). The study showed that proper pre-treatment and choice of starting cellulose materials can reduce the energy use, a key issue for industrial up-scaling (Zimmerman et al., 2010). Spence et al., in 2011, mentioned the possibility to use unbleached pulp to produce NFC with the potential benefits of better strength and barrier properties. The study also indicated the potential of recycled paper as a fiber source. Moreover, Takagi and colleagues also proposed a process to extract cellulose nanofibers from waste newspaper in 2013. Besides, percentages of pulp concentration and the ratio of lignin and hemicellulose in fibers decide the energy level needed in pretreatment and treatment phases, as shown in experiments by Ankerfors in 2012 (see table 12).

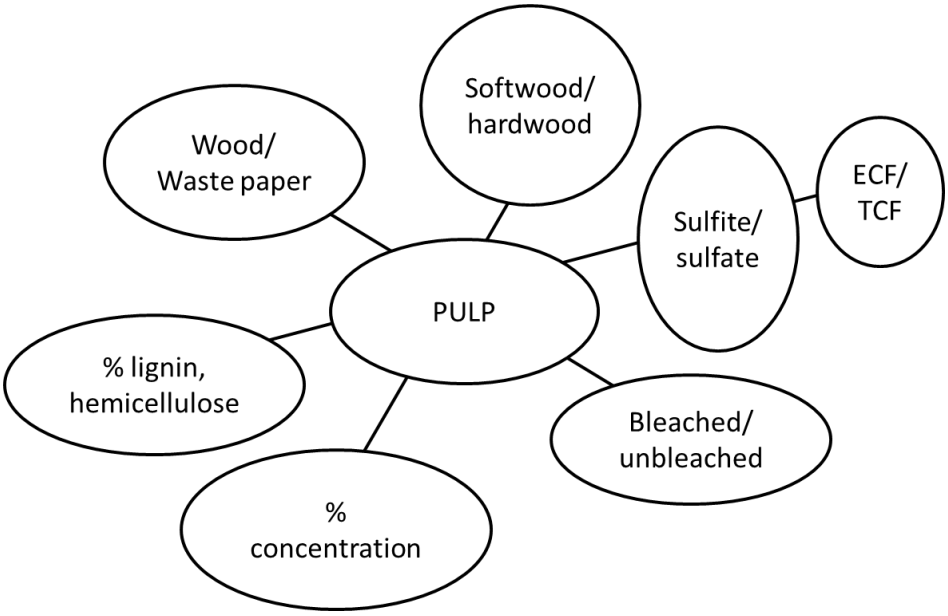

Figure 22. Sub-factors of pulp affecting NFC energy use

The pretreatment process, in combination with the treatment process, is a very important factor that has a large effect on the energy use of NFC, as shown in figure 11. Figure 23 lists several alternatives of pretreatment mentioned in this study. After choosing the suitable pretreatment, NFC producers still need to consider options of input chemicals, especially organic solvents for the carboxymethylation route, which contribute to a large amount of the energy use (figure 13). To solve the problem, recycling the solvent is a feasible solution. In this study, ethanol from corn was shown to require more energy than ethanol from ethylene. However, environmental impacts in terms of, for example, global warming potential should be also considered. This study only focuses on energy use, but it is possible to expand to make the LCA more comprehensive with regards to other environmental impacts. Another important factor is the share of energy for chemicals production and manufacturing (direct heat and electricity) in the pretreatment phase (figure 12). When up-scaling, a different share may lead to different costs because of the particular costs of chemicals and electricity. Not only the choice of treatment process (such as homogenization, micro-fluidization, sonication, etc.) but also the pressure and numbers of passes in the treatment process influence the total energy use, as shown in table 12 and figure 24.

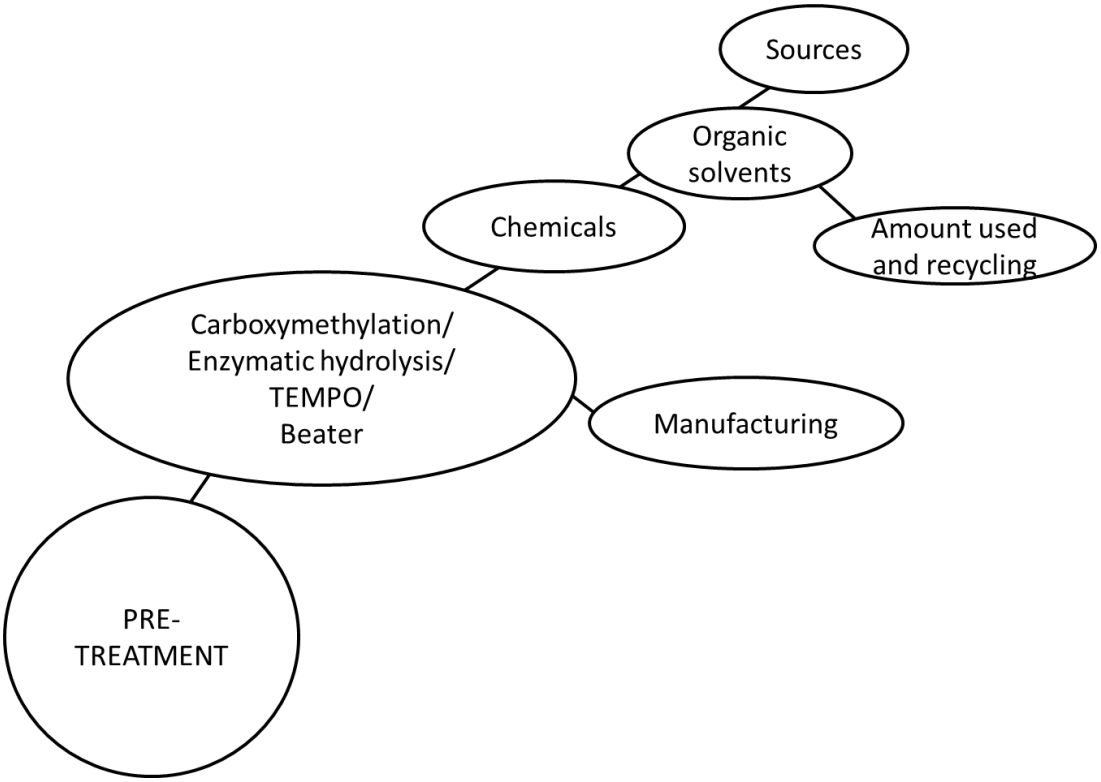

Figure 23. Sub-factors of pretreatment affecting NFC energy use

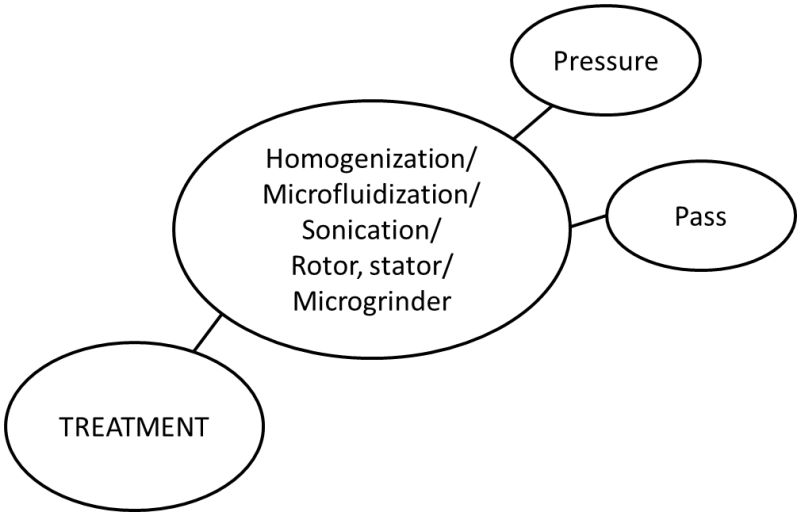

Figure 24. Sub-factors of treatment affecting NFC energy use

Transport plays a minor part in the total energy use for NFC (figure 14). Transport distances, or the choice of transport routes, including the options of local trade or import, has an effect on energy use of the transports themselves. Types of vehicles are also important because of the difference in energy use of different types. The scenario analysis indicates that load capacity and type of fuel used should be considered (table 6). Results from transport scenarios imply the advantages of using large ship for oversea transport and electric train for transport on land (figure 18). However, transport by truck is still probably necessary in many cases. Another important factor is the mass of products, for instance to transport a large amount of chemicals oversea requires a large amount of energy even if the ship is large and

requires a low amount of energy per tkm (figure 19). Therefore, NFC producers should consider using locally available raw materials first. Figure 25 points out the main transport factors that affect the energy use.

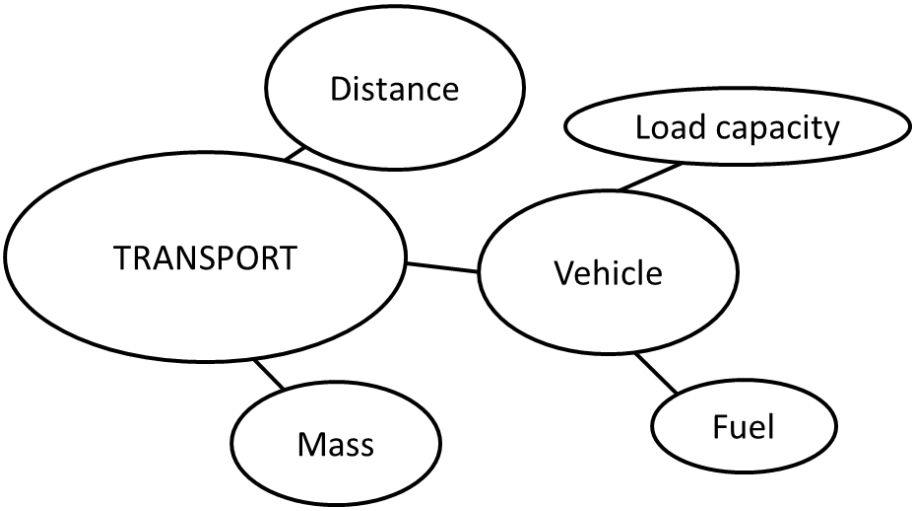

Figure 25. Sub-factors of transport affecting NFC energy use

A vital factor that affects the calculations is related to the NFC product itself. Concentration and specific properties decide which production process should be used (see figure 26). For example, although the carboxymethylation route requires much more energy than the enzymatic hydrolysis process, the carboxymethylated product likely supports other processes in the later modification phase in order to make certain application products from NFC. Therefore, it may reduce energy use in the next stages. However, this is a question of the system boundary, and this cradle-to-gate analysis does not include the modification process.

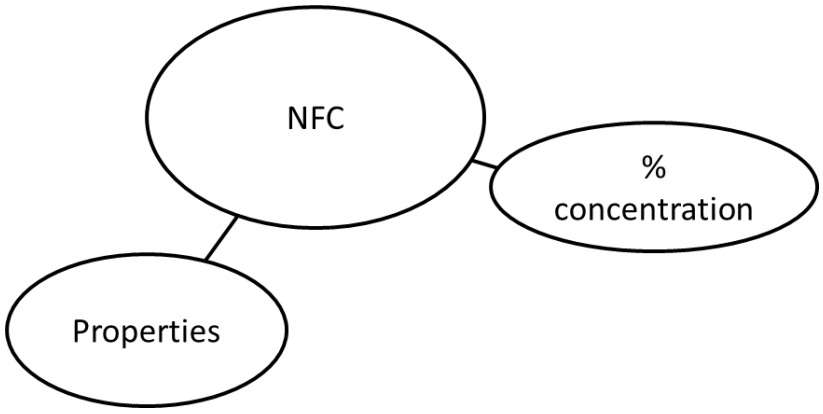

Figure 26. Sub-factors of NFC final product affecting NFC energy use

6. CONCLUSION

This chapter highlights the main findings and comments in the result and discussion chapters. Suggestions for future work are also mentioned at the end of the chapter.

In this cradle-to-gate LCA, the total energy use of wood-based NFC production was calculated for two different production routes. The carboxymethylation route had a much higher energy use than the enzymatic method, approximately ten times higher. This higher energy use is mostly due to the production of input chemicals, especially the organic solvents. Considering that this energy use is typically not the focus of NFC producers, this shows the ability of LCEA to highlight “hidden” energy use along the life cycle.

In the scenario analysis, the applied pulp scenarios show that sulfite pulp has a higher energy use than sulfate pulp, and bleached pulp requires more energy than unbleached pulp. Results from comparing the two types of bleached sulfate pulp indicate a similar energy use of ECF and TCF.

Scenarios of ethanol sources indicate a higher energy use of making ethanol from corn compared with ethanol from fossil-based ethylene. Findings in transport scenarios imply the significant effects of truck and ship transport for long distances.

When putting all the results together to form an overall picture, the contribution of direct energy for manufacturing is small compared to the total energy use. The life cycle energy use results for NFC in this study, together with other nanocellulose LCA studies, indicate that nanocellulose’s energy use is low (for the carboxymethylate route) or very low (for the enzymatic route) compared to the energy use of other nanomaterials.

However, there are still reasons to improve the energy use of NFC production. This study points out five main factors that affect NFC’s energy use, namely pulp properties, pretreatment, treatment, transport and NFC product’s properties.

This study only focuses on energy use, but it is possible to expand to make the LCA more comprehensive with additional environmental impacts induced. Another possible expansion could be to a cradle-to-grave LCA or LCEA of NFC application products.

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