Research and development challenges for Swedish biofuel actors – three illustrative examples

Improvement potential discussed in the context of Well-to-Tank analyses

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Table of Content

Sammanfattning

I dagsläget är det politiska stödet för biodrivmedel mycket stort i både Sverige och EU. EU har, till exempel, satt upp ett mål för användningen av förnybara drivmedel som år 2020 ska uppgå till 10% av den totala inhemska användningen av energi för transport. När alla länder i EU ska försöka uppfylla detta mål kommer det under det närmsta årtiondet att uppstå en ökad efterfrågan på biodrivmedel. För att undvika utökad produktion av jordbruksbaserade biodrivmedel, som kräver stora jordbruksarealer, ligger nu fokus på att utveckla mer avancerade andra generationens (2G) biodrivmedel som kan produceras från råvaror som förknippas med en effektivare markanvändning.

Klimatnyttan av att använda biodrivmedel är något som ofta diskuteras. De totala utsläppen av växthusgaser vid användning av biodrivmedel kommer från hela produktionskedjan, mestadels från jordbruket/skogsbruket och drivmedelsproduktionsanläggningen. För att kunna jämföra olika biodrivmedelsalternativ är det nödvändigt att bedöma de olika drivmedlen genom att använda en metod som inkluderar alla utsläpp från källan till användningen. Denna metod kallas på engelska well-to-wheel (WTW) och well-to-tank (WTT) när analysen inte inkluderar fordonstekniken.

I Sverige är förutsättningar för att producera biomassabaserade drivmedel mycket stora. Vi har också flera forskningsanläggningar där produktionstekniker för lovande andra generationens drivmedel demonstreras och utvecklas. I den här studien har vi valt att fokusera på cellulosabaserad etanol, metan via förgasning av biomassa i fast form samt DME via förgasning av svartlut, med syfte att identifiera forsknings- och utvecklingspotential som kan leda till förbättrade utsläppsvärden i en WTT-analys. Vi studerar också utmaningar och möjlig utvecklingspotential för jordbruket/skogsbruket. *Syftet* med den här studien är alltså att, från litteraturstudier och diskussion med forskarna själva, identifiera utmaningar inom forskning och utveckling som berör svenska biodrivmedelsaktörer. *Målet* med den här studien är att, i ett WTT sammanhang, (i) öka kunskapen om biodrivmedelsproduktionens komplexitet, (ii) identifiera och diskutera potential för förbättrade energibalanser och minskade växthusgasutsläpp för de tre valda drivmedelsalternativen, samt (iii) identifiera och diskutera potential för ökat utbyte av bioråvara från jord- och skogsbruk. Vi har valt att fokusera på att diskutera teknik, systemaspekter och klimatpåverkan som förknippas med produktionen av drivmedel. Drivmedlens påverkan på t.ex. biodiversitet, försurning, övergödning och sociala aspekter ryms inte inom ramen för denna studie.

Resultat från *jordbruk/skogsbruks*-analysen visar att det finns potential för minska klimatpåverkande utsläpp i WTT kedjans första del, genom att minska användningen av fossila bränslen i arbetsmaskiner och traktorer, effektivisera odlingen liksom produktionen av konstgödsel (anläggningar som producerar konstgödsel kan också installera rening av dikväveoxid) och genom förbättrade gödslingsmetoder. Dessutom kan utsläppen minskas genom att man undviker att odla på kolrik mark (t.ex. torvmark) och nya jordbrukssystem skulle kunna införas där behovet av att plöja och harva minskas. Övriga möjligheter inkluderar att introducera nya framavlade grödor som till exempel vete med ökat innehåll av stärkelse eller salix som innehåller mer cellulosa.

Från studien om *cellulosabaserad etanol* ser vi att 2G etanol, i samproduktion med biogas, el, värme och/eller träpellets, kan spela en viktig roll i utvecklandet av hållbara biodrivmedelssystem. Beroende på råvara, efterfrågan på de olika produkterna samt hur förutsättningarna ser ut för integrering med anläggningar som producerar 1G etanol, kan 2G-etanolproduktionssystemen designas på olika sätt för att maximera lönsamheten. Optimeringen för att hitta den mest lönsamma kombinationen, i ett så komplext produktionssystem, kräver ökad kunskap och ökat samarbete mellan många olika aktörer inom flera olika kompetensområden. Detta nödvändiga samarbete skulle kunna innebära en barriär i sökandet efter den optimala lösningen. Tre viktiga

resultat från denna delstudie är: (i) produktionssystemen skulle kunna vara mycket mer komplext och intelligent designade än vad tidigare studier har visat, (ii) utvecklingspotentialen består av en mängd olika processintegrationskombinationer som delvis beror på de lokala förutsättningarna, och (iii) miljöpåverkan från varje unikt system kan variera stort beroende på produktionssystemets design och lokala förutsättningar.

Från studien om *metanproduktion via förgasning av biomassa* i fast form ser vi att en fördel med den här tekniken är att den kan ge ett högt utbyte av drivmedel från bioråvaran. Förbättringspotential finns framför allt inom rening och behandling av syntesgasen där processen kan bli än mer effektiv. En stor utmaning är att avlägsna tjära innan metaniseringssteget. Tre viktiga resultat från denna delstudie är: (i) det är viktigt att inte förstöra de metanmolekyler som produceras i förgasningssteget vilket indikerar behov av förbättrade, selektiva, katalysatorer i metaniseringssteget, (ii) det finns ett behov av ny gasseparationsteknik för att underlätta användandet av luft istället för syrgas i förgasaren och (iii) tekniken behöver skalas upp och testas i realistiska förhållanden.

Från studien om *DME-produktion via svartlutsförgasning* ser vi att processens fördelar jämfört med andra biodrivmedelsalternativ bland annat är att träråvaran i form av svartlut redan är förprocessad och i en flytande pumpbar form samt att processen redan är trycksatt i och med att förgasningen är väl integrerad i massabruket, vilket förbättrar omvandlingseffektiviteten. Några utmaningar kvarstår ändå innan denna biodrivmedelsproduktionsteknik kan anta kommersiell status, bland annat att visa att material och utrustning kan möta de höga kraven på tillgänglighet när tekniken skalas upp, liksom att visa att anläggningen kan drivas enligt de värme- och materialbalanser som beräknats. Tre viktiga resultat från denna delstudie är: (i) att moderna kemiska massabruk kan bli viktiga leverantörer av förnybara drivmedel, (ii) att det finns ett behov av att visa att DME/metanol-produktionen fungerar i stor skala och (iii) att det fortfarande finns utrymme för tekniska förbättringar och utökad energiintegration.

Trots att det finns kvantitativa förbättringspotentialer angivna i de tre drivmedelsstudierna är det inte uppenbart hur dessa verkligen skulle påverka WTT-värdena. Det beror framför allt på att drivmedelsprocesserna är komplexa vilket gör att förändring av en parameter leder till att andra parametrar ändras. Förbättringspotentialerna diskuteras därför kvalitativt. Från studien i helhet kommer vi fram till följande *gemensamma slutsatser*: (i) i Sverige är forsknings- och utvecklingsarbetet kring de tre studerade biodrivmedelsalternativen mycket stort, (ii) överlag fungerar de tre drivmedelsteknikerna mycket bra i pilot- och demonstrationsskala och det är nu dags att möta utmaningarna med att skala upp till kommersiella anläggningar, (iii) det finns potential för att ytterligare förbättra energibalanser och minska utsläpp av växthusgaser, (iv) eftersom biodrivmedelsproduktionssystemen är komplexa och beror på lokala förutsättningar behövs ett övergripande systemperspektiv för att hitta optimala lösningar (både inom produktionsprocessen och i ett WTT-sammanhang), och (v) de tre studerade biodrivmedelsalternativen kompletterar varandra.

Ett ytterligare resultat från den här studien är att processen med att ta fram den här rapporten har inneburit ett nära samarbete mellan industri och forskare (forskare från olika discipliner och olika universitet/forskningsinstitut). Att samarbetet har gått smidigt och att alla har visat stort intresse för att bidra till studien bådar gott för eventuella framtida samarbeten inom f[3], svenskt kunskapscentrum för förnybara drivmedel.

Till sist, eftersom den politiska ambitionen tydligt indikerar en ökad marknad för förnybara drivmedel i hela Europa, kan samtliga av de tre studerade drivmedelsalternativen parallellt bidra till att nå EUs mål.

Summary

Currently biofuels have strong political support, both in the EU and Sweden. The EU has, for example, set a target for the use of renewable fuels in the transportation sector stating that all EU member states should use 10% renewable fuels for transport by 2020. Fulfilling this ambition will lead to an enormous market for biofuels during the coming decade. To avoid increasing production of biofuels based on agriculture crops that require considerable use of arable area, focus is now to move towards more advanced second generation (2G) biofuels that can be produced from biomass feedstocks associated with a more efficient land use.

Climate benefits and greenhouse gas (GHG) balances are aspects often discussed in conjunction with sustainability and biofuels. The total GHG emissions associated with production and usage of biofuels depend on the entire fuel production chain, mainly the agriculture or forestry feedstock systems and the manufacturing process. To compare different biofuel production pathways it is essential to conduct an environmental assessment using the well-to-tank (WTT) analysis methodology.

In Sweden the conditions for biomass production are favourable and we have promising second generation biofuels technologies that are currently in the demonstration phase. In this study we have chosen to focus on cellulose based ethanol, methane from gasification of solid wood as well as DME from gasification of black liquor, with the purpose of identifying research and development potentials that may result in improvements in the WTT emission values. *The main objective* of this study is thus to identify research and development challenges for Swedish biofuel actors based on literature studies as well as discussions with the the researchers themselves. We have also discussed improvement potentials for the agriculture and forestry part of the WTT chain. *The aim* of this study is to, in the context of WTT analyses, (i) increase knowledge about the complexity of biofuel production, (ii) identify and discuss improvement potentials, regarding energy efficiency and GHG emissions, for three biofuel production cases, as well as (iii) identify and discuss improvement potentials regarding biomass supply, including agriculture/forestry. The scope of the study is limited to discussing the technologies, system aspects and climate impacts associated with the production stage. Aspects such as the influence on biodiversity and other environmental and social parameters fall beyond the scope of this study.

We find that improvement potentials for emissions reductions within the *agriculture/forestry* part of the WTT chain include changing the use of diesel to low-CO_2-emitting fuels, changing to more fuel-efficient tractors, more efficient cultivation and manufacture of fertilizers (commercial nitrogen fertilizer can be produced in plants which have nitrous oxide gas cleaning) as well as improved fertilization strategies (more precise nitrogen application during the cropping season). Furthermore, the cultivation of annual feedstock crops could be avoided on land rich in carbon, such as peat soils and new agriculture systems could be introduced that lower the demand for ploughing and harrowing. Other options for improving the WTT emission values includes introducing new types of crops, such as wheat with higher content of starch or willow with a higher content of cellulose.

From the case study on *lignocellulosic ethanol* we find that 2G ethanol, with co-production of biogas, electricity, heat and/or wood pellet, has a promising role to play in the development of sustainable biofuel production systems. Depending on available raw materials, heat sinks, demand for biogas as vehicle fuel and existing 1G ethanol plants suitable for integration, 2G ethanol production systems may be designed differently to optimize the economic conditions and maximize profitability. However, the complexity connected to the development of the most optimal production systems require improved knowledge and involvement of several actors from different competence areas, such as chemical and biochemical engineering, process design and integration and energy and environmental systems analysis, which may be a potential barrier.

Three important results from the lignocellulosic ethanol study are: (i) the production systems could be far more complex and intelligently designed than previous studies show, (ii) the potential improvements consist of a large number of combinations of process integration options wich partly depends on specific local conditions, (iii) the environmental performance of individual systems may vary significantly due to systems design and local conditons.

From the case study on *gasification of solid biomass for the production of biomethane* we find that one of the main advantages of this technology is its high efficiency in respect to converting biomass into fuels for transport. For future research we see a need for improvements within the gas up-grading section, including gas cleaning and gas conditioning, to obtain a more efficient process. A major challenge is to remove the tar before the methanation reaction. Three important results from the biomethane study are: (i) it is important not to crack the methane already produced in the syngas, which indicates a need for improved catalysts for selective tar cracking, (ii) there is a need for new gas separation techniques to facilitate the use of air oxidation agent instead of oxygen in the gasifier, and (iii) there is a need for testing the integrated process under realistic conditions, both at atmospheric and pressurized conditions.

From the case study on *black liquor gasification for the production of DME* we find that the process has many advantages compared to other biofuel production options, such as the fact that black liquor is already partially processed and exists in a pumpable, liquid form, and that the process is pressurised and tightly integrated with the pulp mill, which enhances fuel production efficiency. However, to achieve commercial status, some challenges still remain, such as demonstrating that materials and plant equipment meet the high availability required when scaling up to industrial size in the pulp mill, and also proving that the plant can operate according to calculated heat and material balances. Three important results from the DME study are: (i) that modern chemical pulp mills, having a potential surplus of energy, could become important suppliers of renewable fuels for transport, (ii) there is a need to demonstrate that renewable DME/methanol will be proven to function in large scale, and (iii) there is still potential for technology improvements and enhanced energy integration.

Although quantitative improvement potentials are given in the three biofuel production cases, it is not obvious how these potentials would affect WTT values, since the biofuel production processes are complex and changing one parameter impacts other parameters. The improvement potentials are therefore discussed qualitatively. From the entire study we have come to agree on the following *common conclusions*: (i) research and development in Sweden within the three studied 2G biofuel production technologies is extensive, (ii) in general, the processes, within the three cases, work well at pilot and demonstration scale and are now in a phase to be proven in large scale, (iii) there is still room for improvement although some processes have been known for decades, (iv) the biofuel production processes are complex and site specific and process improvements need to be seen and judged from a broad systems perspective (both within the production plant as well as in the entire well-to-tank perspective), and (v) the three studied biofuel production systems are complementary technologies. Futher, the process of conducting this study is worth mentioning as a result itself, i.e. that many different actors within the field have proven their ability and willingness to contribute to a common report, and that the cooperation climate was very positive and bodes well for possible future collaboration within the framework of the f3 center.

Finally, judging from the political ambitions it is clear that the demand for renewable fuels will significantly increase during the coming decade. This will most likely result in opportunities for a range of biofuel options. The studied biofuel options all represent 2G biofuels and they can all be part of the solution to meet the increased renewable fuel demand.

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Acronyms and Definitions

Energy carriers

Biofuels	Biomass-based fuels in the transportation sector
BTL	synthetic fuels derived from biomass
CTL	synthetic fuels derived from coal
DME	Dimethyl ether
ETOH	Ethanol
FT	Fischer-Tropsch products, e.g., synthetic diesel, gasoline, kerosene
GTL	synthetic fuels derived from natural gas
H_2	Hydrogen
Hythane	A mixture of hydrogen and methane (biogas and/or natural gas)
MEOH	Methanol
NG	Natural (fossil) Gas
SNG	Synthetic natural gas (here methane via gasification of solid biomass)
Synfuels	Fuels synthesized from syngas (via gasification), e.g., MeOH, DME, FT

Chemical formulas

Al_2O_3	Aluminium oxide
CH_3OCH_3	Dimethyl ether
CH_3OH	Methanol
CH_4	Methane
CO	Carbon oxide
CO_2	Carbon dioxide
COS	Carbonyl sulfide
CR_2O_3	Chromium oxide
Cu	Copper
H_2	Hydrogen
H_2O	Water
H_2S	Hydrogen sulfide
HCl	Hydrochloric acid
N_2O	Nitrous oxide
NH_3	Ammonia
S	Sulphur
SO_2	Sulphur dioxide
ZnO	Zinc oxide
ZnS	Zinc sulfide

Units

GJ	Giga Joule, 10^9 Joules
kW	kilo Watt, 10^3 Watts
kWh	kilo Watt hour, 3.6 MJ
MJ	Mega Joule, 10^6 Joules
PPM	parts per million
TW	Tera Watt, 10^{12} Watts

Miscellaneous

1G	First generation biofuels
2G	Second generation biofuels
AD	Anaerobic digestion
ADt	Air dry tonne (used for pulp as delivered at 90% dry content)
BIGCC	Biomass integrated gasification combined cycle
BLG	Black liquor gasification
BLGMF	Black liquor gasification with motor fuel production
BMG	(Solid) biomass gasification
CCS	Carbon capture and storage
CHP	Combined heat and power
GHG	Greenhouse gases, (e.g., CO_2, CH_4, N_2O, and chlorofluorocarbons CFCs)
IGCC	Integrated gasification combined cycles
LCA	Life Cycle Analysis
NGCC	Natural gas combined cycle (technology for electricity production)
Salix	The genus "Salix" hold more than 350 species of fast growing woody crops, mostly different kinds of willow
tDS	Tonne dry solids
TRI	ThermoChem Recovery International
TTW	Tank-to-wheel
WGS	Water gas shift reaction
WTT	Well-to-tank
WTW	Well-to-wheel

1. Introduction

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The energy use for Swedish transportation is currently dominated by petroleum-based fuels, mainly gasoline and diesel. Because of climate and energy security of supply issues the transport sector is now facing major changes.

The EU has set a target for the use of renewable fuels in the transportation sector stating that all EU member states should use 10% renewable fuels for transport by 2020 (European Parliament and Council, 2009). This can be achieved by moving towards biofuels, renewable electricity and hydrogen as well as by (or in combination with) increased use of more energy efficient vehicles in the fleet. Further, the Swedish Government's long term ambition is that Sweden by 2030 should have a fleet of vehicles that are independent of fossil fuels (Regeringskansliet, 2009). A significant increase in the production and use of renewable fuels represents a considerable challenge.

Within EU biofuels constituted 3.3% of the total amount of fuels used the transportation sector in 2008 (Eurobserv´er, 2009). This corresponds to approximately 117 TWh of which 92 TWh was biobiesel, 20 TWh ethanol and 5 TWh pure vegetable oils. 25% of the ethanol and 10% of the biodiesel was imported to Europe.

Over the last years, the use of biofuels for transport has been promoted by a range of policy instruments. In 2003, the European Commission proposed an increased use of biofuels in the transportation sector in a directive which states that biofuels should constitute 2% of the total amount of transportation fuels sold in 2005 (estimated as energy content) at the national level, and 5.75% in the year 2010 (European Council, 2003). Other policies promoting bioenergy and in particular biofuels for transport have also been implemented both in the EU and in Sweden (see e.g. AEBIOM, 2006).

Currently biofuels have strong political support, both in the EU and Sweden. If increasing the use of renewable fuels in Europe to 10% of all fuels for transport, which correspond to approximately 350 TWh, there will be an enormous market for biofuels during the coming decade. This will most likely lead to increased biofuels production and the building of more biofuels production plants. To avoid increasing production of biofuels based on agriculture crops, that require considerable use of arable area, there is a current focus to move towards more advanced second generation (2G) biofuels that can be produced from biomass feedstocks associated with a more efficient land use, read more in Section 1.2.

Climate benefits and greenhouse gas (GHG) balances are aspects often discussed in conjunction with sustainability and biofuels. Every now and then voices are heard in media claiming that biofuels have worse environmental impact compared to diesel and gasoline. This is true for a fraction of the biofuels on the market but not for the majority of the biofuels. The total GHG emissions depend on the entire fuel production chain, mainly from the agriculture or forestry feedstock systems and the manufacturing process. To compare different biofuel production pathways it is essential to conduct an environmental assessment using a well to wheel (WTW) analysis methodology, read more in Section 2.

1.1 Method, aim and objective

In this study we want to describe the well-to-tank (WTT) method and discuss that results from such analyses might differ from study to study depending on assumptions made, read more in Section 2. As an illustration of what might be included in a WTT analysis and how the result may change we have included an example in Section 2.3.

The main objective of this study is to *identify research and development challenges* for Swedish biofuel actors. Changes in the biofuel production regarding fuel yield, energy and materials savings or process integration will have an impact on the GHG emissions and thus the WTT emission result. From literature studies as well as from asking the researchers themselves we want to learn about ongoing research and find improvement potentials, dilemmas between different improvement options as well as if there are barriers to overcome or technology that need to be proven in large scale before the fuel production can achieve commercial status.

We have chosen to focus on three biofuel production technology options that are currently in the demonstration phase: cellulose based ethanol, methane from gasification of solid wood as well as DME from gasification of black liquor. We have also chosen to discuss improvement potentials for the agriculture and forestry part of the WTT chain.

The aim of this study is to
  - increase knowledge about the complexity of biofuel production.
  - identify improvement potentials, regarding energy efficiency and GHG emissions, for three biofuel production cases.
  - discuss improvement potentials, including agriculture/forestry, in the context of WTT analyses.

This study is limited to discuss the technology, system aspects and climate impacts associated with the production stage. Aspects such as the influence on biodiversity and other environmental and social parameters fall beyond the scope of this study.

The report is structured as follows: in Section 2, we describe and discuss the WTT method and difficulties. In section 3, we present challenges and improvement potentials for the agriculture and forestry part of the WTT chain. In Sections 4-6 we present challenges and improvement potentials for the three chosen biofuels production technologies. Finally, in Section 7, we discuss the results and offer some conclusions and suggestions for further work.

1.2 Renewable fuels

Current commercial alternative transportation fuels, as well as promising future options, which can be used in both conventional internal combustion engines and in new more efficient engines, are presented in Figure 1.1.

Primary energy sources   **Energy carriers**   **Vehicle options**
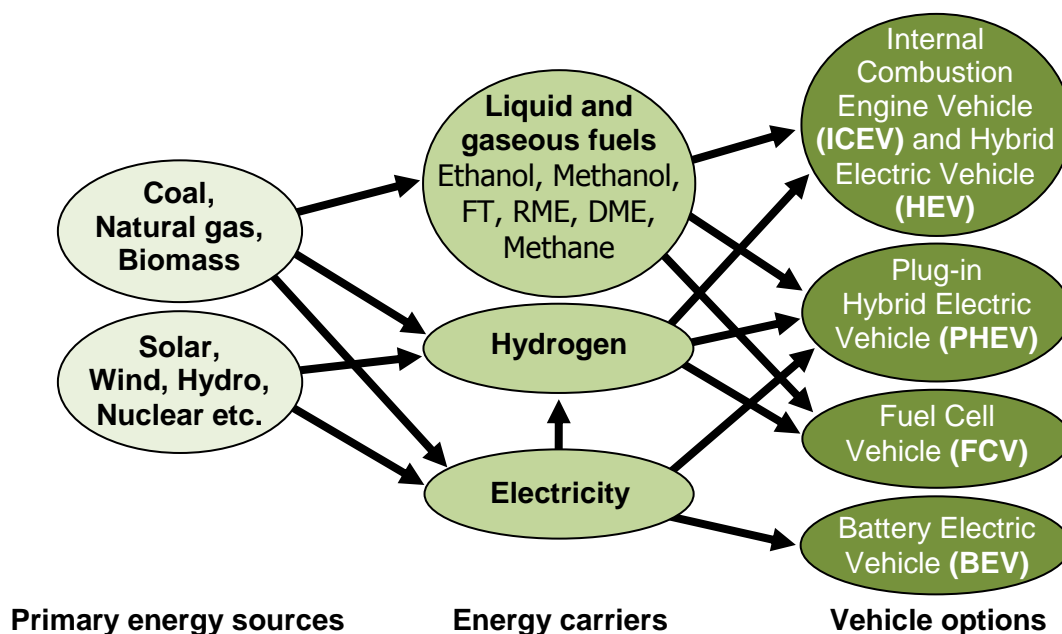
Figure 1.1. Alternative transportation fuels can be produced from solid, liquid and gaseous primary energy sources as well as from primary energy sources generating electricity. *Current* commercial alternative transportation fuels are ethanol, methane (biogas and natural gas), biodiesel here represented by rapeseed methyl ester (RME) and fossil-based Fischer-Tropsch (FT) gasoline and diesel. Promising *future* low CO_2 emitting energy carriers are electricity, hydrogen, and biomass-based so called second generation biofuels, e.g., methanol, FT fuels, dimethyl ether (DME), and biomethane.

Biomass is a useful primary energy source, which can be converted into transportation fuels in several ways, e.g., via anaerobic digestion into biogas, fermented into ethanol, gasified and synthesized into synfuels, (e.g., Fischer-Tropsch diesel, dimethyl ether (DME), methanol, methane, hydrogen), or vegetable oils can be transesterified into biodiesel (e.g., RME), see Figure 1.2.

Conventional or *first generation biofuels* are the biomass derived transportation fuels that are available today, including for example, ethanol from sugar or starch crops and biodiesel from esterified vegetable oil. A number of LCA and WTW studies have been made of first generation biofuels, and the results regarding possible GHG emissions reduction and energy efficiency are far from harmonious (see e.g,. Larson, 2006; Delucchi, 2006). Despite the wide range of results it can be concluded that the total potential for GHG emissions reduction from first generation biofuels in the long term is low, due to high land requirements and low cost efficiency (Larson, 2006; Hamelinck, 2006).

Advanced or *second generation biofuels* are transportation fuels based on lignocellulosic feedstock. The two main production routes are gasification of solid biomass or black liquor followed by synthesis into, for example, methanol, dimethyl ether (DME), synthetic natural gas (SNG) or Fischer-Tropsch diesel (FTD), and lignocellulosic ethanol. None of these technologies is yet commercial. Nevertheless, hopes are high that second generation biofuels will reach high energy and cost efficiency and that they will be able to contribute substantially to the reduction of GHG emissions (see e.g. IPCC, 2007, COM 2006:34). Potential lignocellulosic feedstocks include forest residues, waste wood, black liquor and farmed wood. What feedstock will come to predominate in a country or region will very much depend on local conditions.

Biofuels for transport
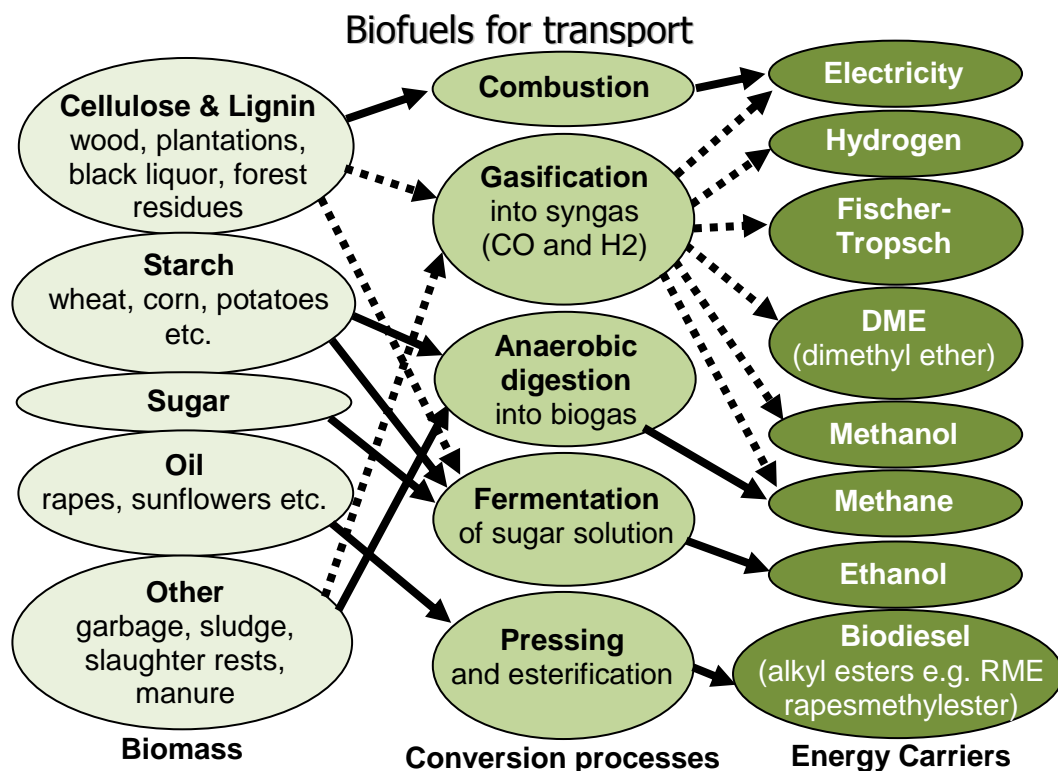
Figure 1.2. Biomass can be divided into groups depending on chemical composition of the biomass. Different elements are better suited for different processes that convert the biomass into energy carriers useful for the transportation sector. Commercially available options are marked with solid lines, while processes still on demonstration plant level are marked with dotted lines.

There are four main parts that need to be fulfilled before a biofuel can be said being totally CO_2-neutral:

- the carbon dioxide emissions to the atmosphere, originating from the biofuel combustion, must be absorbed in growing biomass,
- the emissions of non-CO_2 greenhouse gases due to the use of the fuel, must end or be compensated for,
- the soil carbon, connected to the biomass production, needs to be constant and
- all input energy for agriculture/forestry and fuel production need to be CO_2-neutral.

In the foreseeable future no fuels for transport are likely to become totally CO_2-neutral. Note that improved energy efficiency is also an important tool for reducing GHG emissions.

In this study we have no intention of trying to determine which options are to prefer among the alternative fuels. However, in Appendix 1, an overview of advantages and disadvantages for a range of fuel options can be found.

2. Well to tank (WTT) analyses

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The evaluation of energy efficiency and climate impact of transportation options is usually done from a well-to-wheel (WTW) perspective. A WTW study is a form of life cycle analysis (LCA) that is normally limited to the fuel cycle, from feedstock to tank, and the vehicle operation, and that typically focuses on air emissions and energy efficiency (Edwards et al., 2007; MacLean and Lave, 2003). A WTW analysis generally does not consider the energy or the emissions involved in building facilities and vehicles, or end of life aspects. It neither attempts to estimate the overall "cost to society" such as health, social or other cost areas.

The main reason for this simplified life cycle analysis is that the fuel cycle and vehicle operation stages are the life cycle stages with the greatest differences in energy use and greenhouse gas (GHG) emissions compared to conventional fuels. In this report focus is on biomass based transportation fuels, referred to as biofuels. Note also that the tank-to-wheel (TTW) part of the WTW analysis lies outside the scope of this study, i.e. this study is focusing on the well-to-tank (WTT) part of the WTW analysis. Figure 2.1 illustrates the main steps in a WTW analysis of biofuels.
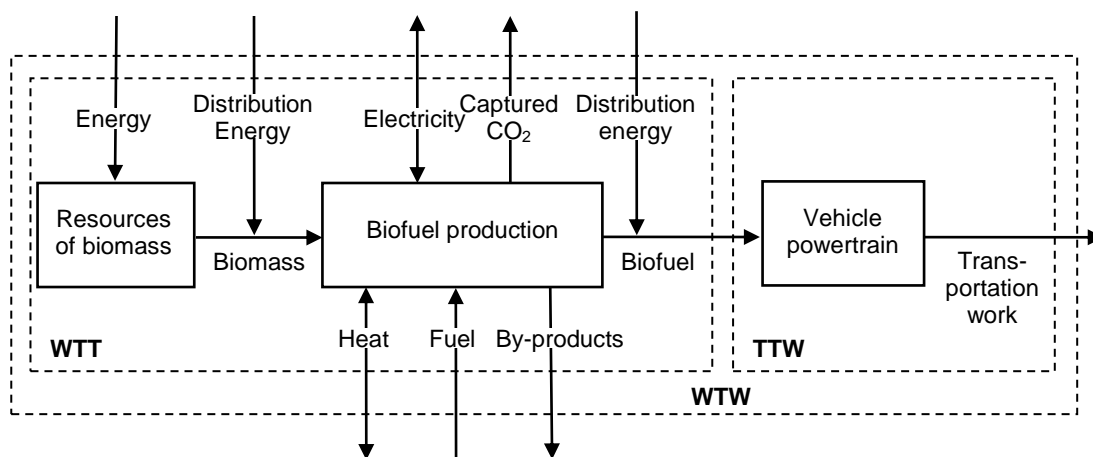
Figure 2.1. Simplified illustration of the main energy and material flows in the main steps of a well-to-wheel (WTW) analysis of biofuels where also the well-to-tank (WTT) and tank-to-wheel (TTW) parts are illustrated.

The first step includes operations required to extract, capture or cultivate the primary energy source, in this case some kind of biomass feedstock. Then, the biomass needs to be distributed to the biofuel production plant. At the biofuel production plant, the biomass is processed into a biofuel and possibly also other products such as electricity, heat or other by-products[1].

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[1] In some cases the term by-products is used to describe products of a more undesired nature that are produced together with the main product (biofuel) and the term co-product is used when a more desired product are co-produced with the main product. Here, the term by-product is used for all products produced with the main product.

The biofuel production plant could also have a deficit of electricity and/or heat. In order to cover a heat deficit, external fuel could be used at the biofuel production plant. It could further be possible to capture CO_2 in the process[2]. The produced biofuel is then distributed to refueling stations. The final step includes the vehicle operation where the biofuel is transformed into transportation work. A WTT analysis includes the steps from feedstock to tank and thus does not include the vehicle operation stage.

In this study the main focus is on second generation biofuels. The general methodological issues are the same for first and second generation biofuels. In connection to second generation biofuels, mainly produced from woody feedstock, one of the main issues is on potential for integration of the heat surplus from these processes with other industries or district heating systems.

2.1 Method difficulties

Comparison of the results from different WTW or LCA studies of biofuels is often problematic, with the results showing a remarkably wide range of GHG emissions and energy use for a given fuel and biomass source (see for example Börjesson, 2006; Larson, 2006; Delucchi, 2006; Fleming et al., 2006; Gnansounou et al., 2009). Besides the many sources of uncertainty in the data itself, there are also different accepted methods on how to handle the data in existing WTW analysis[3].

Parameters identified as responsible for introducing the largest variations and uncertainties are to a large part connected to system related assumptions, for example system boundaries, reference system, allocation methods, time frame, functional unit and what GHG species to include in the analysis. In order to give the reader an improved understanding of the complexity of WTT studies of biofuels, a selection of these general issues is thoroughly discussed in Appendix 2.

There are challenges in how to make reasonable assumptions in each step of the WTW chain. Emissions from the feedstock part of the WTT chain come from fossil fuel used in tractors and other machines, the production and usages of fertilizers and pesticides as well as from the land itself. Challenges, when making assumptions for the biomass feedstock part in WTT analyses, are presented in Appendix 3.

Emissions from the biofuel production part of the WTT chain come from the use of fossil fuels, chemicals, electricity and depend on what kind of by-products that are produced, how the process is integrated within the plant as well as with other industries, and whether CCS is applied to the plant or not. Challenges, when making assumptions for the biofuel production part in WTT analyses, are presented in Appendix 4.

In a full WTW analysis also the emissions from distribution, dispensing and usage of the biofuels are included. Although the TTW part is not included in this study it is worth

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[2] The amount of captured carbon depends on what energy carrier is produced, e.g., DME (CH_3-O-CH_3) contains carbon (i.e., only carbon losses can be captured) while hydrogen (H_2) does not (i.e., theoretically 100% of the carbon can be captured).

[3] Börjesson (2006) lists several reasons why the energy balances differ between different studies, even where the feedstock and by-products are identical. He also conclude that depending on the systems boundaries, and the allocation method, the energy balance may differ by a factor of five.

mentioning that also the last part of the WTW chain are drawn with challenges in how to make assumptions, read more in Appendix 5.

The degree of sensitivity analysis varies widely between different studies. Some studies only present one result, while other studies vary a large number of parameters to show the impact of these on the results. Especially for non-commercial technologies, to be implemented in the future, there are large uncertainties both when it comes to the biofuel conversion system (fuel production, usage in vehicles etc) and surrounding systems (electricity, transportation, district heating etc).

2.2 Illustration of how WTT results may change with different assumptions

To illustrate how different assumptions in a WTT analysis may affect the result an example of cellulose-based ethanol is presented in this section. In Table 1.1, the different parts included in three cellulose-based ethanol options, taken from Edwards et al. (2007), are presented.

Table 1.1. WTT-values on gCO_2/MJ for cellulose-based ethanol where the by-product lignin is used for electricity production (Edwards et al., 2007).

	Farmed wood	**Waste wood**	**Straw**
N fertilizer	4.40	-	-
Diesel for cultivation	1.55	1.02	-
Collecting straw	-	-	3.35
Diesel for chipping	1.02	-	-
Losses during chipping and storage	0.17	-	-
Wood chips road transport	0.99	0.99	-
Wood chips coastal ship transport	-	3.76	-
Straw road transport	-	-	0.22
Diesel	3.13	3.13	-
H_2SO_4	0.69	0.69	0.81
NH_3	6.31	6.31	2.39
$(NH_4)_2 SO_4$	0.55	0.55	0.21
Antifoam	0.43	0.43	0.16
Corn steep liquor	0.03	0.03	0.03
CaO	1.45	1.45	1.70
Debit for additional P fertilizer	-	-	0.31
Debit for additional K fertilizer	-	-	0.92
Credit, lignin used for electricity production	-0.13	-0.13	-0.07
Total	**20.60**	**18.24**	**10.03**

From Table 1.1 it can be seen that the major CO_2 emission posts in the WTT analysis comes from agriculture (in the case of farmed wood) and from the biofuel production part, where ammonia (NH_3) stands for the largest contribution. Emissions from road transportation are minor posts in the WTT analysis. When wheat straw is not used for energy purposes it is assumed to be ploughed back into field giving nutrients to the soil. The two "debit-posts" in Table 1.1 represent the need for additional fertilizer inputs to compensate for soil nutrient losses, when straw is used as feedstock.

Since this WTT analysis, carried out by Edwards et al., use a substitution method for the allocating of emissions all emissions generated by the process are allocated to the main product and the by-products are given an emission credit equal to the energy and emissions

saved by not producing the product that the by-product is likely to replace. In this case the by-product lignin is assumed to replace feedstock for electricity production.

As an exercise to show how the WTT values can change, we have in Figure 2.2 illustrated the result of assuming that all transportation posts (the fuels needed for tractors, trucks, ships) could be done using fuels without CO_2 emissions, i.e. all posts including diesel, or transport, are set to zero. As comparison we have also included the WTT results from Edwards et al. (2007), which were presented in Table 1.1 (in Figure 1.3 denoted the "base case").
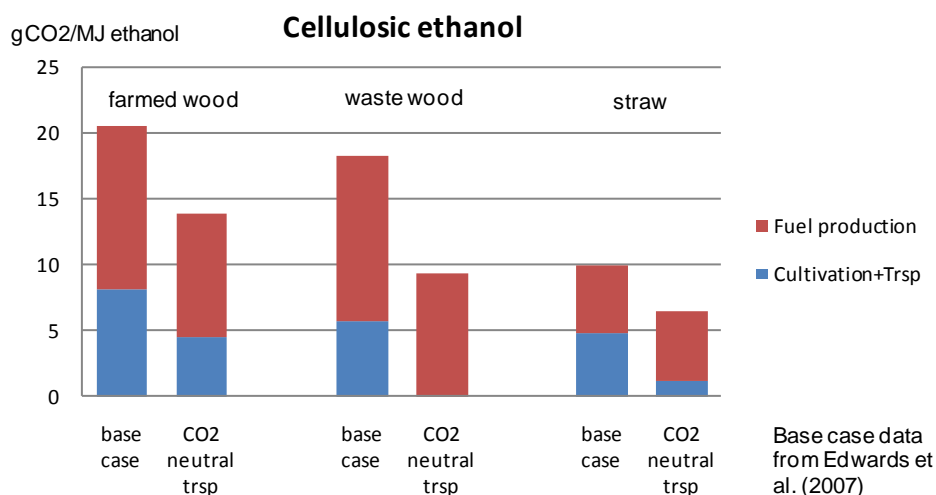
Figure 2.2. An illustration of how the WTT values will change if assuming that tractors and trucks will run on CO_2-neutral fuels in future.

2.3 Illustration of how WTT analyses differ in literature

In this section we want to present that WTT analyses can focus on different aspects and that the biofuels can be ranked in different order depending on the WTT method. We have chosen to present results from four different WTW or WTT studies. The chosen studies are focusing on (i) the combination of GHG emissions and expended energy (Edwards et al., 2007), (ii) land use efficiency (Börjesson, 2007), (iii) systems expansion (Wetterlund et al., 2009b), and (iv) seven different criteria judged by score (Volvo, 2007).

2.3.1 Greenhouse gas emissions and expended energy

Most of the WTT analyses found in literature base their environmental assessment on climate impacts and expended energy. This is also the case for this study made by Joint Research Center, EUcar and Concawe (Edwards et al., 2007).

In the criteria of climate impact Edwards et al. (2007) take the three main GHG emissions: carbon dioxide, methane and nitrous oxide into account. For the allocating of energy and emissions between biofuel and by-products they use the substitution method, following the principals described in the international standard ISO 14041. That is, all energy and emissions generated by the process are allocated to the main product (fuels for transport) and the by-products generate an energy and emission credit equal to the energy and emissions saved by not producing the product that the by-product is likely to replace.

A basic flowchart of energy flows and GHG emissions in this WTW analysis is illustrated in Figure 2.3.
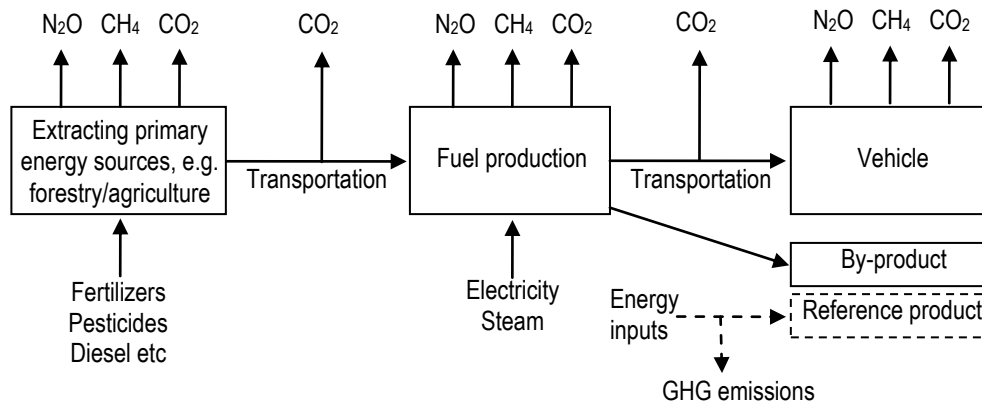
Figure 2.3. Outline of energy inputs and greenhouse gas emissions in Edwards et al. (2007). The credits from by-products are calculated with the substitution method, i.e. the by-products generate an energy and emission credit equal to the energy and emissions saved by not producing the material that the by-product is likely to replace.

Details about the main assumptions made in Edwards et al (2007), regarding for example the electricity mix, biomass sources, expended energy, greenhouse gas emissions, are presented in Appendix 6.

Results for both the energy balance and the greenhouse gas emissions in the entire WTW chain are presented in Figure 2.4.
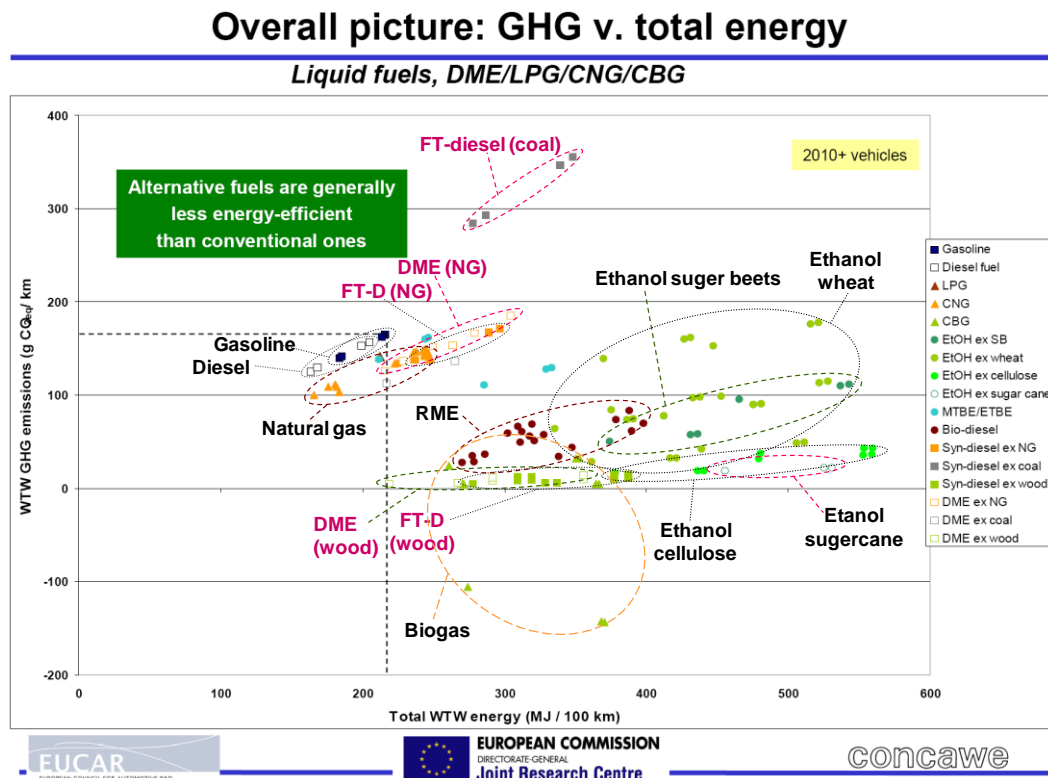
Figure 2.4. Results on WTW greenhouse gas emissions (gCO$_2$/km) and total energy expended (MJ/100 km) calculated in Edwards et al., (2007). The lower and the more left a fuel is placed, the better. Note that the horizontal line for zero emission is one third up in the chart.

From Figure 2.4 it can be seen that it is very seldom an alternative fuel can be produced using less energy compared to conventional fossil fuels. Most biofuels have, however, lower GHG-

21

emissions than fossil fuels, except for wheat-based ethanol produced using coal in the manufacturing process. Wood based fuels generally present emissions very close to zero followed by sugarcane ethanol. Sugarcane ethanol is, however, found among the biofuels options that has the highest expended energy which is a result of that the CONCAWE/EUCAR/JRC study includes the energy content in the bio-feedstock (i.e. the energy content in the sugarcane itself) when calculating the expended energy[4]. When excluding the energy content in the feedstock the ranking change, within the biofuels, gaining sugarcane-based ethanol.

It is interesting to note that the environmental performance differ a lot for the same fuel (especially wheat-based ethanol, biogas and RME) depending on assumptions made on which primary energy source is used in the manufacturing process and how the by-products are used.

2.3.2 Focus on land use efficiency

In the part "Conversion and utilisation of biomass from Swedish agriculture", included in Börjesson (2007), a comparison between different agriculture-based biofuels can be found. The biofuels are compared from the amount of biofuels that can be produced from one hectare average arable land in eight different Swedish climate zones. The gross production indicate how much biofuels are actually produced whereas the net production show the biofuels production minus the amount of energy used for the transportation as well as in the agriculture and energy conversion processes.

Results from one of the eight different Swedish climate zones analyzed in Börjesson (2007) are presented in Figure 2.5.
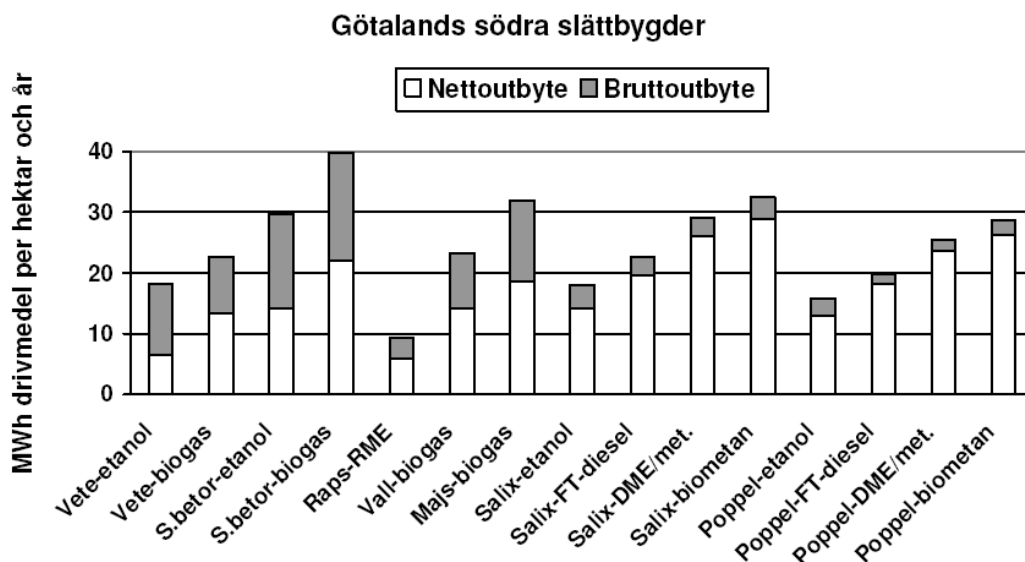
Figure 2.5. Average net (netto) and gross (brutto) biofuels produced per hectare and year, on average arable land, in southern Sweden (Götalands södra slättbyhgder). Translation of the analyzed biofuel option from the left: wheat-ethanol, wheat-biogas, sugarbeet-ethanol, sugarbeet-biogas, rapeseed methyl esther, pasture-biogas, corn-biogas, willow-ethanol, willow-Fischer-Tropsch-diesel, willow-DME/methanol, willow-biomethane, poplar-ethanol, poplar-Fischer-Tropsch-diesel, poplar-DME/methanol, poplar-biomethane.

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[4] Including the energy content in the bio-feedstock itself leads to that biomass-based fuels always will receive an expended energy higher than the energy content in the final biofuel. Other WTW-analysis might only include input energy (e.g. diesel, fertilizers, pesticides, electricity, steam).

The results show that biomethane and DME/methanol from willow or poplar[5] generate the highest net biofuel yield per hectare in southern Sweden (25-30 MWh). Highest net production is shown for biogas produced from sugarbeets (40 MWh). The energy expended is generally much higher for the fuels produced from annual crops compared to fuels produced from willow and poplar.

For the other climate zones it can be mentioned that for the area around Gothenburg the same results are shown as in southern Sweden with the difference that the yield is approximately 15-25 % lower per hectare and year, and that sugarbeets no longer is an option. In the forest dominating area in southern Sweden (Småland) the highest net fuel yield is shown for hybride aspen (hybridasp) 16-17 MWh per hectare followed by fertilized pine tree plantations. In the forest areas in the middle of Sweden the results follow the results from the southern forest area but with a 10-30% lower net yield. In the northern part of Sweden fertilized pine shows the highest net yield (10 TWh per hectare).

When producing wheat-, sugarbeet- and cellulose-based ethanol as well as rapeseed methyl esther it is possible to extract by-products which in energy terms might correspond to one third of the initial biomass' energy content. The total energy (gross) yield from including the by-products would increase to approximately 60% for the wheat- and sugarbeet-based ethanol if the draff and pulp can be used for heat and electricity production or as animal fodder. For the RME production the total gross yield could increase to approximately 75% and for cellulose-based ethanol utilizing the lignin the total gross yield could be 90% (Börjesson, 2007).

2.3.3 Focus on system expansion

Wetterlund et al. (2009b) analyze the effects of expanding the system (to include the systems surrounding to the biomass conversion system) when evaluating well-to-wheel CO_2 emissions for some biofuels options.

Four different cases are considered, all four are biomass conversion technologies currently in focus in Sweden. The cases are: DME via black liquor gasification (BLG:DME), methanol via gasification of solid biomass (BMG:MeOH), lignocellulosic ethanol (EtOH) and electricity from a biomass integrated gasification combined cycle (BIGCC) used in a battery-powered electric vehicle (BEV). All four cases are considered with as well as without carbon capture and storage (CCS). System expansion is used consistently for all flows. The results are compared with results from the European JRC/EUCAR/CONCAWE study by Edwards et al. (2007), which is a conventional well-to-wheel study that only uses system expansion for certain co-product flows. All biofuels are assumed to be used in hybrid vehicles.

To show the impact of surrounding systems on the WTW CO_2 emissions from the biofuel options, the system is expanded to include a reference system, illustrated in Figure 2.6.

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[5] The entire tree (except for the stump) is assumed when using Poplar.
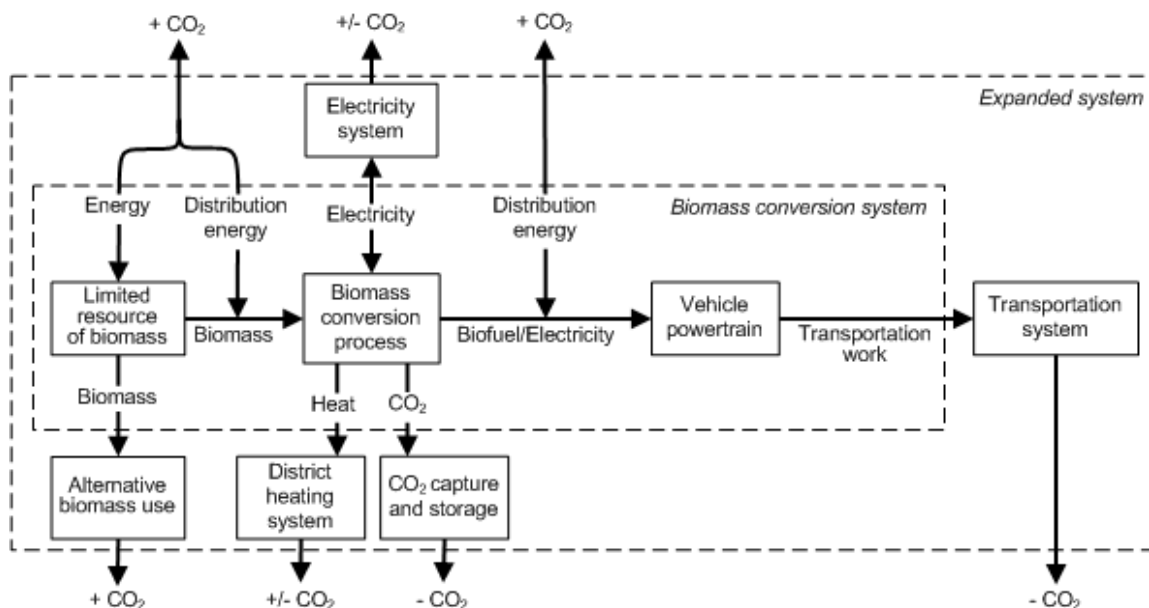
Figure 2.6. Schematic representation of a biomass conversion system and surrounding systems that need to be considered when evaluating the climate impact of biomass based transportation. The CO_2 effect of each flow is indicated with +/-, where + means an increase and - means a decrease in CO_2 emissions. For electricity production, the CO_2 emissions can either increase or decrease depending on whether the biofuel production plant is a net importer or net exporter of electricity.

As can be seen in Figure 2.6, flows of energy or material entering or leaving the biomass conversion system are assumed to cause a change in the surrounding system, an approach often used in WTW studies to avoid by- or co-product allocation problems. In Wetterlund et al. (2009b), the method is taken one step further and used to evaluate the CO_2 effect of not only co-product flows, but of all flows of energy or material entering or leaving the biomass conversion system[6].

A number of studies acknowledge that the supply of bioenergy is limited and that efficient use is essential if the CO_2 benefit of substituting biomass for fossil fuels is to be maximized. However, few studies consider the marginal CO_2 effects of biomass. With forceful policies promoting bioenergy use, certain biomass assortments may be fully exploited. Additional demand, for example from power plants co-firing biomass and coal, would then have to be met by fossil fuel. In Wetterlund et al. (2009b) the biomass system is expanded to include alternative biomass use, by assuming that biomass used for transportation reduces the amount of biomass available for other applications in the system, thus increasing the CO_2 emissions from those applications.

The reference system determines the emission baseline, which is defined as an estimation of what would have occurred in a project's absence. To highlight the influence of the surrounding system on the results, the reference system[7] and the emission baseline are varied systematically, thus covering a large number of possible future energy systems. Details about the assumptions in Wetterlund et al. (2009b) can be found in Appendix 7.

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[6] For example, if the plant has a surplus of electricity, this causes a decrease in grid electricity production, and vice versa for an electricity deficit (it should be noted that this is not applicable for the BIGCC case, since all the electricity produced is assumed to be used for transportation and thus affects the transportation system rather than the electricity system).

[7] Alternative biomass use, marginal electricity production and marginal transportation fuels.

The results from analyzing the effect on CO_2 emission reduction from system expansion are presented in Figures 2.7 and 2.8. Hatched columns indicate combinations that are considered to be less probable[8].

Figure 2.7. Net CO_2 effect for the studied cases when the alternative biomass use is assumed to be co-firing with coal, for four different reference electricity technologies over a range representing the three marginal transportation technologies. Hatched columns indicate combinations that are considered less probable. Recalculated results from the EU study (Edward et al., 2007) are included for comparison. BIGCC for use in BEV has been recalculated from results in the EU study.

Figure 2.7 shows the net CO_2 effect compared to reference systems when assuming that the biomass supply potential is limited and that the alternative biomass use is to be co-firing with coal in power plants. The figure shows the CO_2 effect for each of the four different marginal electricity production technologies considered, over a range representing the three marginal transportation technologies. When the reference electricity production and transportation are varied, the potential for CO_2 emissions reduction fluctuates, with several cases. Lignocellulosic ethanol and solid biomass gasification to methanol show little or no potential for CO_2 reduction. The electricity production technology affects the results and differs

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[8] For example, if CCS is implemented in the biofuel and BIGCC plants, it will probably also be implemented when transportation fuels are produced via coal. Similarly it can be assumed that if CCS is implemented in coal power plants it will probably also be implemented in the production of transportation fuels from coal, where CO_2 is separated as part of the process. An electricity system with a CO_2-neutral build margin will probably be an indication of strong policy instruments promoting reduction of greenhouse gases in the atmosphere. Hence, if the marginal electricity production is CO_2-neutral, a marginal transportation technology based on coal (without CCS) is considered less probable. Implementation of CCS for coal based electricity and/or transportation fuel production, in combination with biofuel production without CCS, could also be regarded as less probable. It has however not been defined as such here, since CCS is not yet as established for biomass systems as for fossil systems.

between the technologies studied. The difference is mainly due to the fact that the electricity production technologies have very different energy balances (see Table A6.1). BLG, with a substantial electricity deficit, shows the largest variation (largest vertical separation between columns) and benefits from a low CO_2 emitting electricity production technology (coal with CCS or CO_2-neutral electricity). EtOH, with a relatively high surplus of electricity, benefits from a high CO_2 emitting electricity production technology (coal) [9]. The BMG process has a fairly low dependency on the electricity production technology, and thus shows similar results for all four electricity production technologies. For BIGCC, it was assumed that all the electricity produced is used in the transport sector[10]. In the same way as the BLG process, the BIGCC benefits from low CO_2 emitting marginal electricity production. As can be seen, Edwards et al. (2007) in general shows a significantly higher potential for CO_2 reduction[11].

As the figure also shows, variation of the reference transportation system introduces a larger degree of uncertainty than variation of the reference electricity production. The reason for this is that the biofuel production (electricity production in the BIGCC case) is higher than the electricity deficit or surplus. However, a number of the possible reference system combinations can be considered less probable (hatched columns)[12].

Unsurprisingly, the cases with CCS show a considerably larger potential for CO_2 reduction than the cases without, in particular for black liquor gasification, solid biomass gasification and biomass integrated gasification combined cycle, where the sequestrable amount of CO_2 is high.

Figure 2.8 shows the net CO_2 effect when it is assumed that biomass use has no marginal effect. As can be seen, all the technologies investigated now show a considerable potential to reduce CO_2 emissions, in line with the potential shown in Edwards et al. (2007).

When the alternative use of biomass is excluded, the main differences between Wetterlund et al. (2009b) and Edwards et al. (2007) are that marginal electricity production is used instead of recalculating electricity to biomass, and that surplus heat is assumed to be used for district heating. Again the black liquor gasification process with its large electricity deficit shows the largest variation (and the greatest potential for CO_2 reduction) when the reference electricity production is varied, followed by the biomass integrated gasification combined cycle process, due to its large heat delivery.

The results show that failure to expand the system to take into alternative biomass use into account may result in overestimation of the potential of biomass-based transportation to contribute to reduced CO_2 emissions. Evaluations of biomass-based transportation should therefore reflect that biomass and land for biomass production are limited resources. This becomes particularly important when evaluating technologies that are expected to use a substantial amount of available biomass in future, as is the case with many biofuel technologies.

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[9] However, as the large district heating delivery from the EtOH plant replaces biomass CHP heat which decreases the CHP electricity production, the electricity surplus from the EtOH plant is effectively almost cancelled out.

[10] Similarly to the EtOH process the BIGCC however delivers a large amount of district heating, which affects the alternative biomass CHP electricity production.

[11] Results from Edwards et al. (2007) have been recalculated from CO_2 emissions per vehicle km, to net CO_2 effect per MWh of biomass for each of the three reference transportation technologies without CCS.

[12] It is primarily combinations that contain coal as margin transportation technology that are regarded as less probable, which has the effect that the probable CO_2 range decreases considerably for all studied technologies.

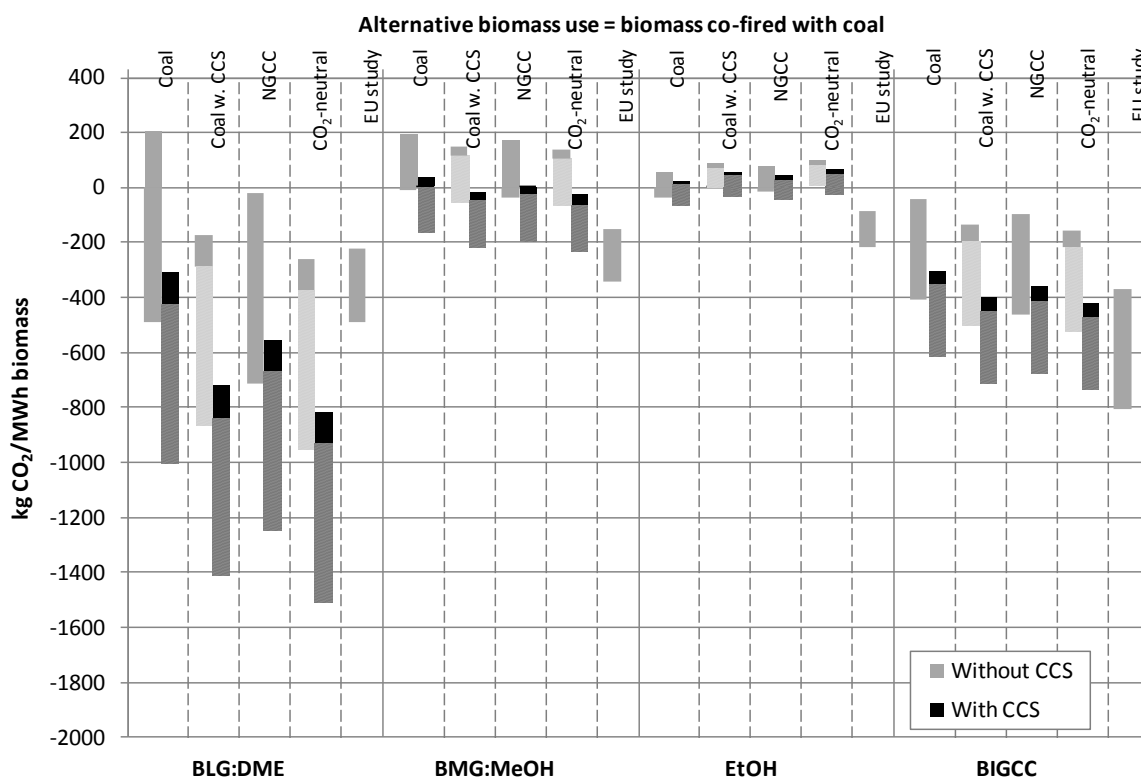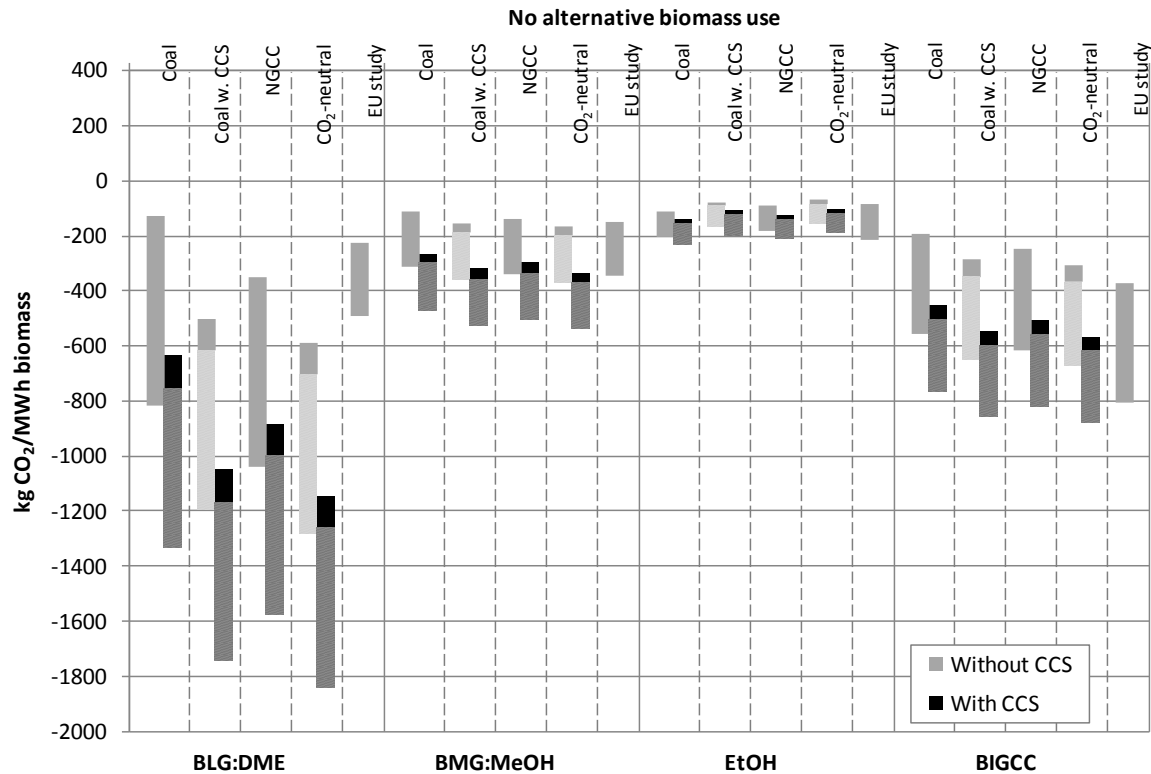**Figure 2.8.** Net CO_2 effect for the studied cases when no alternative biomass use is assumed, for four different reference electricity technologies over a range representing the three marginal transportation technologies. Hatched columns indicate combinations that are considered less probable. Recalculated results from the EU study (Edward et al., 2007) are included for comparison. BIGCC for use in BEV has been recalculated from results in the EU study.

2.3.4 Different criteria judged by score

"Well to wheel" analysis can also be done by comparing and judging a wide range of criteria. Such comparison can be presented in a matrix where the reader can get an overview of the advantages and disadvantages of each fuel pathway. Such study on alternative fuels was made by AB Volvo in 2007 as a part of the presentation of seven climate neutral trucks to the public (Volvo, 2007).

It is possible to build vehicles that run on almost any type of fuel however the environmental performance and technical requirements are depending on the choice of fuel. The Volvo study was made in order to identify the barriers and possibilities for the most discussed renewable fuels for commercial vehicles. The methodology and reasoning in the study is described here.

The Volvo study includes analysis of seven different criteria where some of them can be measured quantitatively while others are qualitative. The study is based on fuel used for commercial vehicles such as trucks and buses with a European Perspective. The final result is a matrix where the fuels were given scores for each criteria.

The following criteria are included: (i) climate impact, (ii) energy efficiency, (iii) land use efficiency, (iv) feedstock availability/fuel potential, (v) vehicle adaptation, (vi) fuel cost, and (vii) fuel infrastructure.

27

The fuel pathways analyzed in the Volvo study are illustrated in Figure 2.9. These pathways were judged to be the most relevant ones that can be produced in EU.
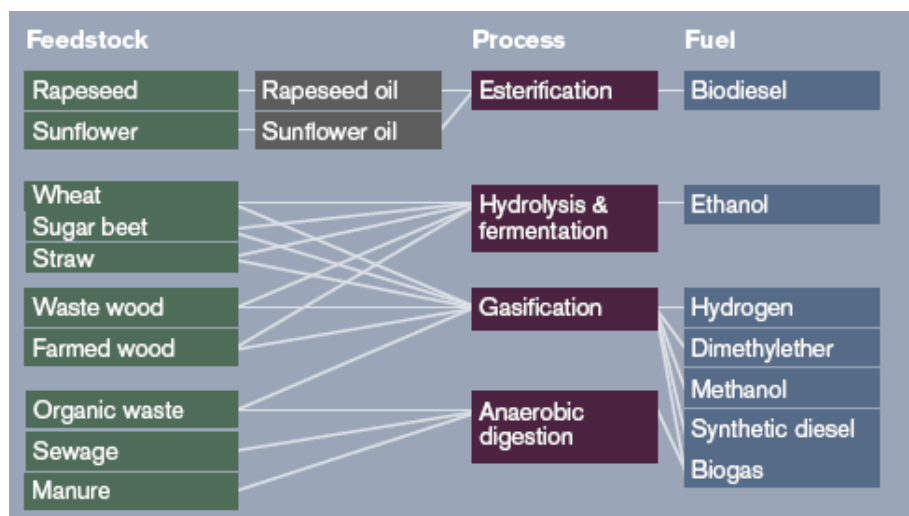
Figure 2.9. Fuel pathways judged to be the most relevant ones that can be produced in EU.

For the Climate impact the CO_2 reduction on a WTW basis compared to conventional diesel fuel is judged. The WTT CO_2 equivalent emissions are following Edwards et al., (2007), but modified in order to take into account ignition improvers for methanol and ethanol. The TTW data was taken from Volvo internal material on engine efficiency with different fuels. The engine's energy efficiency is used to calculate the CO_2 emissions. The score for the criteria *climate impact* is distributed in a falling scale as: 5 circles (91-100% reduction), 4 circles (76-90% reduction), 3 circles (51-75% reduction), 2 circles (26-50% reduction), and 1 circle (0-25% reduction). Some of the alternatives may end up in several intervals since there are different processes producing the same fuel. Results are presented in Appendix 8.

The energy efficiency is calculated using WTT background data from Edwards et al., (2007) but modified in order to take into account ignition improvers for methanol and ethanol. The tank to wheel data was taken from Volvo internal material. Data for biomethane production via gasification was estimated since it is not included in Edwards et al., (2007). The score for *energy efficiency* is set in a falling range as, 5 circles (over 22%), 4 circles (20-22%), 3 circles (17-19%), 2 circles (14-16%), and 1 circle (under 14%). Some fuels may end up in different scores since there are several processes that produce the same fuel with different overall energy efficiency. Results are presented in Appendix 8.

Driving distance per hectare per year is a measure of land use efficiency. The yield per hectare for each crop has been calculated using figures for Swedish average yields from good quality land. The quantity of fuel/energy used in harvesting, production, transport etc. is subtracted from the quantity produced. The distance which a heavy duty vehicle can travel per year and hectare is calculated using data from Edwards et al., (2007), RENEW[13] and discussions with Lund University. The score for *land use efficiency* is defined as distance travelled per year and hectare in a falling range as, 5 circles (over 10 000 km), 4 circles (7501–10 000 km), 3 circles (5001–7500 km), 2 circles (2500–5000 km), and 1 circle (under

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[13] Renewable fuels for advanced powertrains. A European project, supported under the European Commission's 6th Framework Programme, see http://www.renew-fuel.com/home.php.

2500 km). Results for the same fuel may vary depending on the production process used. Results as well as one calculation example for salix to DME, can be found in Appendix 8.

The availability of raw material and the choice of production process determine the amount of fuel that can be produced. The amount of fossil fuel that can be replaced by biomass varies depending on the efficiency of the fuel production process and the end use efficiency. Today the biomass potential in EU is limited and other sectors such as heat and power are already using large parts of the supply potential. To increase the European biofuel production the largest part will most likely be derived from energy crops such as Salix Willow. The supply potential for the different fuel alternatives was multiplied with the relative end use efficiency of different fuels. The score for *feedstock availability/fuel potential*, in EU, is set in a falling range as, 5 circles (350–420 TWh), 4 circles (280–349 TWh), 3 circles (210–279 TWh), 2 circles (140–209 TWh), and 1 circle (70–139 TWh). Results on the criteria "feedstock availability/fuel potential" are presented in Appendix 8.

All fuels do not suit all engines or transport applications. Some fuel adaptations are also more expensive and complex to develop and produce than others. The score for *vehicle adaptation* is set in a falling range as, 5 circles (suitable for all heavy applications; no special vehicle adaptation required), 4 circles (suitable for most heavy applications; no expensive or extensive vehicle adaptation required), 3 circles (suitable for most heavy applications; expensive and extensive vehicle adaptation required), 2 circles (suitable for up to half of all applications; complex, expensive and extensive vehicle adaptation required), and 1 circle (suitable for only a limited number of applications; major expensive and extensive vehicle adaptation required). Results on the criteria "vehicle adaptation" are presented in Appendix 8.

Future costs are difficult to predict and in many cases the cost of producing a fuel is only a small element of the fuel price, due to taxes and other charges. Here the cost of the particular fuel is compared with that of conventional diesel oil, assuming a crude oil price of USD 70 per barrel (excluding taxes). Comparison is made on energy equivalent basis using data from Edwards et al. (2007) but updated with the latest information available on energy and raw material cost. The results for the same fuel may vary depending on the feedstock/process used. The score for *fuel cost* is set in a falling range as, 5 circles (cheaper than diesel from crude oil), 4 circles (0-19% more expensive than diesel from crude oil), 3 circles (20-59% more expensive than diesel from crude oil), 2 circles (60-99% more expensive than diesel from crude oil), and 1 circle (100-140% more expensive than diesel from crude oil). Results on the criteria "fuel cost" are presented in Appendix 8.

Infrastructure issues determine how quickly and easily a new fuel can be introduced and integrated with existing systems. This evaluation also takes into account the safety and environmental aspects of handling the fuel within the infrastructure. The complexity of building new infrastructure is compared to a reference diesel infrastructure. The score for *fuel infrastructure* is set in a falling range as, 5 circles (no changes, liquid fuel), 4 circles (minor changes, liquid fuel), 3 circles (major changes, liquid fuel), 2 circles (gas handled in liquid form at low pressure), and 1 circle (gas handled under high pressure or in liquid form at low temperature). Results on the criteria "fuel infrastructure" are presented in Appendix 8.

The part results of the scores for the seven criteria were organized in a matrix in order to get an overview, see Figure 2.10. The ratings for biogas, biogas+biodiesel (methane-diesel[14]) and biogas/hydrogen mixture are based on production by gasification and anaerobic digestion.

Figure 2.10. A summary of the results from scoring the seven criteria.

One might be tempted to summarize the score in order to identify the "winner" or "winners". However, it is important to remember that in order to do this, weighting factors must be applied. These weighting factors depend on the time scale and other subjective factors. Criteria such as vehicle adaptation and fuel infrastructure are of "one time cost" character and therefore good performance in other criteria yields continues benefits. One general conclusion that can be drawn from the matrix is that there are many alternative fuel options, for heavy traffic, and that none of the options are obviously better than another.

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[14] The biogas+biodiesel concept is not a "bi-fuel" solution where the driver switch between gaseous or liquid fuel but instead a concept of mixing the two fuels at the same time in the combustion, read more in Volvo (2010).

3. Improvement potential in agriculture and forestry

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The first part of WTT analyses includes emissions from the agriculture or forestry. Greenhouse gases are emitted during cultivation of feedstock used to produce biofuels. This section describes and discusses the potentials for reduction of greenhouse gas emissions as well as the potential problems and uncertainties that may arise when calculating the greenhouse gas emissions from agriculture and forestry.

3.1 Biofuels from agricultural crops

The improvement potential and the uncertainty of the amount of greenhouse gas emissions from growing and producing biomass feedstock in agriculture and forestry may depend on the harvest method, external factors such as soil type and from differences in calculation methodology; each described in separate sections below.

3.1.1 Potentials and uncertainties related to cultivation and harvest of agricultural crops for biofuel production

Greenhouse gas emissions during crop cultivation are made up of:
- carbon dioxide (CO_2) from working machines, mainly tractors, and the fertilizer production plants, and
- nitrous oxide (N_2O) which is emitted from arable land as well as from the manufacture of nitrogenous fertilizer.

It is often the case that emissions of nitrous oxide contribute more to the global warming potential than emissions of carbon dioxide (Börjesson, 2009). There is, however, a great uncertainty about how much nitrous oxide is emitted by arable land, and such emissions may vary to a great extent depending on local conditions (Börjesson, 2009).

The issue of nitrous oxide emissions during cultivation of biofuel feedstock crops have been discussed intensively in the research community during the last years (see e.g., Crutzen et al., 2006; Rauh, 2007; Ammann, 2007). Emissions of nitrous oxide from land may be estimated from an IPCC model (IPCC, 2006). This method assumes a linear relationship between N_2O emissions and the amount of nitrogen from fertilizer, etc., that is applied to the field (Ahlgren et al., 2009).

More fuel-efficient tractors, more efficient cultivation and manufacture of fertilizers, etc., can somewhat reduce emissions of carbon dioxide per tonne of biomass, possibly by up to 20% (Börjesson, 2009). In addition, nitrous oxide emissions may be reduced during the manufacture of nitrogenous fertilizer thanks to catalytic nitrous oxide gas cleaning, which is starting to be implemented in Western Europe. In this way, nitrous oxide emissions can be reduced by approximately 75% (IPCC, 2006).

More efficient nitrogen utilization during cultivation can also reduce the risk of nitrous oxide formation in the ground. The amount of nitrogen available for nitrous oxide formation will be

reduced by improved fertilization strategies, such as more precise nitrogen application during the cropping season (Börjesson, 2009).

A joint study made by JRC, CONCAWE and EUCAR have described the emissions from and energy efficiency of a number of fuels and processes (Edwards et al., 2007). Emissions of greenhouse gases from agricultural land were, in Edwards et al. (2007), calculated through a DeNitrification-DeComposition (DNDC) model (Li et al., 2000). The model firstly calculates the emissions of N_2O per tonne of crop; see Table 3.1.

Table 3.1. Emissions of N_2O calculated for agricultural land use for different crops to be used for biofuel production. Adapted from Rydberg (2007), with data from Edwards et al. (2007).

	Wheat	Sugar beet	Rape	Sunflower
	g N_2O/tonnes moist crop			
Direct N_2O emissions	0.206	0.041	0.892	0.568
N_2O from N leaching	0.072	0.004	0.138	0.056
Total N_2O	0.278	0.046	1.03	0.625
+/- interval	0.185	0.014	0.407	0.186

The study thereafter calculates the indirect emissions of greenhouse gases from different crops from a life cycle perspective; see Table 3.1 and Table 3.2.

Table 3.2. Emissions of greenhouse gases from different part of the process from agriculture to fuel. Adapted from Rydberg (2007), with data from Edwards et al. (2007).

	g CO_2-eq /MJ ethanol		g CO_2-eq /MJ FAME	
	Wheat	Sugar beet	Rape	Sunflower
Agricultural cropping	31.92 (whereof 14.3 g CO_2)	20.83 (whereof 10.5 g CO_2)	51.26 (whereof 18.2 g CO_2)	28.03 (whereof 12.0 g CO_2)
Transport	0.54	2.12	0.66	0.9
Fuel production*	-17.82/58.58 (25.17)	5.20/33.00 (33.00)	1.9/7.4 (1.9)	3.3/8.9 (3.3)
Distribution (150 km incl. refueling station)	1.54	1.54	1.2	1.2

* The numbers correspond to a low/high estimate with difference assumptions regarding the fuel production system.

The tables show that the amount of greenhouse gas emissions from the agricultural cropping systems varies significantly. Also worth noting is the large difference in emissions caused by fuel production depending on the type of crop as well as the assumptions made during the modeling regarding type of energy system feeding the fuel production process with process energy.

3.1.2 Potentials and uncertainty related to type of agricultural crop

Using different agricultural crops for biofuel production may have different greenhouse gas profiles depending on the yield per hectar as shown by Table 3.3.

Table 3.3. Ethanol yield per ha from different crops. Source: Recalculated from Brown (2006).

Crop	Country	Yield (litres/ha)
Sugar beet	France	2700
Sugarcane	Brazil	2510
Cassava	Nigeria	1550
Sweet sorghum	India	1420
Corn	United States	1340
Wheat	France	1050

For the same type of crop, the exchange of biofuels per kg of biomass may also differ. Börjesson (2007) writes about the starch content of ethanol, which often is around 70% of the dry matter content. With new ethanol wheat varieties this may be increased to up to about 75%, which may increase the exchange of ethanol from 55% up to approximately 58% (Börjessson, 2007). However, at the same time, the exchange of the by-product draff will be slightly reduced (Börjesson, 2009).

3.1.3 Potentials and uncertainty related to soil type used for agriculture

Another factor which may be of great significance for the greenhouse gas balance of agricultural crops used for biofuels (and thus contributes to the uncertainties) is the difference in soil type.

The most important difference in this respect may be between mineral soil and peat soil. The definitions of peatlands vary depending on the source, but generally peat soils are land areas in which dead organic material has been naturally accumulated over a long time to reach a certain depth of peat (Montanella et al., 2006). Currently, this type of land is a significant store of soil carbon, but it also serves as a sink or a source of various greenhouse gases (Strack, 2008). As indicated in Table 3.4, the carbon content per area is likely much higher in the average peatland than in other types of ecosystems (Parish et al., 2008).

Table 3.4. Some examples of average carbon stock for different pre-anthropogenic ecosystems Unit: tonne C per ha. Source: Extract from Parish et al. (2008).

Ecosystem type	Vegetation, litter and soils	Peat	Total
Peatland (average)	75	1375	1450
Giant confier forest	726	*not applicable*	726
Warm temperate forest	371	*not applicable*	371
Cool temperate forest	325	*Unknown*	325
Tropical rainforest	320	*Unknown*	320

Peatlands are found on several continents and climatic zones with the largest occurrences in the Northern Hemisphere – North America, Russia and Europe (Strack, 2008). However, the tropical peatlands is much discussed due to its relatively large contribution to global emissions of greenhouse gases. The fastest peatland degradation currently occurs in South-East Asia where forested peatlands are cleared for activities such as food or biofuel production (Hooijer et al., 2006). If tropical peatlands are cleared, drained or if the land is used for an alternative purpose, the carbon stored in the peat may be oxidized and released to the atmosphere as CO_2 (Hooijer et al., 2006). Hooijer et al., (2006) concludes that 58% of the global emissions from peatlands currently occur in Indonesia (fires excluded), where it is a main contributor to making Indonesia one of the countries with the highest emissions of greenhouse gases.

An important driver of deforestation of peatlands is palm oil and timber plantations, which require drainage of the land (Rieley et al., 2006). Deforestation on peatlands accounted for one quarter of all deforestation in South-East Asia[15] in 2005 (Hooijer et al., 2006). The soil type may thus have a large impact on calculations on the greenhouse gas calculations, as shown by Fargione et al. (2008) (refered to by Möllersten, 2008). They estimate that the "carbon debt" of palm biodiesel from tropical peatlands is about five times higher than that of the same fuel from tropical rainforests (3,000 tonnes CO_2/ha compared to 610 tonnes CO_2/ha). Other numbers that feature in the literature on direct land-use effects are, for example, when straw is harvested which slightly reduces the content of soil carbon (approximately 150 kg C/ha and year), or when crops are cultivated on former grassland leading to soil carbon losses of approximately 500 kg C/ha and year (Börjesson & Tufvesson, 2010). If cultivation of grain starts on peat land the losses of soil carbon may amount to 7 ton C/ha and year (Börjesson, 2009).

The difference in emissions from cultivation on mineral and organic soil does not only depend on the mineralization of organic material. Ahlgren et al. (2009) present calculations of the greenhouse gas emissions from cultivation of various agricultural crops for biofuels in Sweden. In their sensitivity analysis, they investigate the greenhouse gas emissions related to cultivation of spring barley in Örebro County. The conclusion is that the extra nitrous oxide emissions from cultivation on organic soil cause three to four times higher emission values. This number is only related to N_2O emissions, since it does not include emissions related to mineralization of organic soil.

Besides the direct effects on land use, indirect land-use changes may occur such as when an expanded production of biofuels leads to displacement of food and feed production into new cropping land previously not cultivated, as indicated in Table 3.5 for a smal sample of countries (e.g. natural pasture land, natural forests, etc.). Such indirect land use change could lead to even higher losses of carbon than the direct land-use changes (Searchinger et al., 2008; Fargione et al., 2008). Regarding both direct and indirect land-use impacts, changes to the land's carbon store in mineral soils are reduced over time, and after approximately 30–50 years a new state of equilibrium can often be attained. How long emissions of carbon dioxide take place from cultivated peat land depends on – among other things – the thickness of the peat layer (Börjesson, 2009).

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[15] In Hooijer et al (2006), South-East Asia is defined as Indonesia, Malaysia, Brunei and Papua New Guinea

Table 3.5. The importance of land use change on carbon balance. Data are compiled from PAS 2050 (2008).

Country	Current land use	Previous land use	GHG emissions (t CO$_2$e / ha/yr)
Germany	Annual cropland	Forest land	21
	Annual cropland	Grassland	7.0
	Perennial cropland	Forest land	14
	Perennial cropland	Grassland	6.7
Indonesia	Annual cropland	Forest land	33
	Annual cropland	Grassland	19.5
	Perennial cropland	Forest land	31
	Perennial cropland	Grassland	17.7
USA	Annual cropland	Forest land	17
	Annual cropland	Grassland	1.9
	Perennial cropland	Forest land	16
	Perennial cropland	Grassland	1.5

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3.1.4 Uncertainties due to methodological choices: time period

Besides differences in agricultural practices and external factors such as the type of soil, a parameter that may affect the greenhouse gas balance of a wheel to tank analysis is the methodological choices. One such choice is the choice of time horizon of the calculations.

Fargione et al. (2008) used a 50 year time period during which the emissions from decomposition, etc., was calculated. As the decomposition may continue beyond this point, their calculated values should be conservative (Fargione et al., 2008).

3.2 Forestry-based biofuels

Forestry-based biofuels include ethanol and synthetic diesel from wood waste, farmed wood and wheat straw. Table 3.6 lists emissions of greenhouse gases for various cellulose-based fuels (Rydberg, 2007).

Table 3.6. Greenhouse gases from cellulose-based fuels. Adapted from Rydberg (2007), with data from Edwards et al. (2007).

	g CO$_2$ eq. / MJ produced ethanol (whereof g CO$_2$)			g CO$_2$ eq. / MJ produced synthetic -diesel		
	Waste wood	Farmed wood	Wheat straw	Wood waste	Farmed wood	Black liquor
Cultivation/ collection	0.95 (0.9)	6.96 (3.1)	3.35 (3.3)	0.8 (0.7)	5.5 (2.5)	0.7 (0.6)
Transport	3.18 (3.0)	0.88 (0.9)	0.62 (0.6)	2.9 (2.7)	0.7 (0.7)	0.6 (0.6)
Fuel production	12.31 (12.6)	12.31 (12.6)	3.42 (3.3)	0 (0)	0 (0)	0 (0)
Distribution, 150 km incl refueling station	1.54 (1.5)	1.54 (1.5)	1.54 (1.5)	1.1 (1.1)	1.1 (1.1)	1.1 (1.1)

As the table shows, there are differences between the different fuels and feedstock fuels, especially in the fuel production phase, but also in the cultivation/collection phase.

3.2.1 Potentials and uncertainties related to forest management and harvest of biomass used for biofuels

Today, different forestry management practices exist, which may have an effect on the different greenhouse gas profiles. Eriksson et al. (2007) calculated the effect of different management practices together with different methodology choices regarding substituted fossil fuel. Their conclusion was that the greatest net reduction of greenhouse gases occurred when "the forest was fertilized, slash and stumps were harvested, wood was used as construction material and the reference fossil fuel was coal." The effect on the results of different forest management practices was however smaller than that of the assumption regarding the use of the product (Eriksson et al., 2007).

3.2.2 Potentials and uncertainty related to type of biomass from forest

Using different forestry-based sources of for biofuel production may have different greenhouse gas profiles, due to the different characteristics of the crops, as shown by Table 3.7.

Table 3.7. Yield per hectare for some selected biofuel crops. Source: extract from Groom et al. (2008).

Biofuel crop	Biofuel	Yield (litres/ha)
Poplar & willow	Ethanol/synfuel	5,000–9,000
Fisher-Tropsch	Ethanol/synfuel	30,000–50,000
Wood residues	Biodiesel/ethanol	1,150–2,000

3.2.3 Potentials and uncertainties due to external factors

As with biofuels from agricultural products, the soil type may be crucial for the greenhouse gas balance of biofuels from forests.

3.3 Conclusions

The potential effect of using biofuels and the uncertainty of the amount of greenhouse gas emissions from agriculture and forestry depend on the harvest method, external factors such as soil type and from differences in calculation methodology. As this section has shown, there are large uncertainties in the greenhouse gas emissions related to agriculture and forestry used to produce biofuels. Local and specific characteristics such as soil type, (agricultural method) and N_2O emissions may have a large impact on the final result.

The agriculture/forestry part of the WTT chain is in general significant, sometimes dominating the emissons from biofuels. Improvement potentials for reducing the agriculture/forestry share of GHG emissions includes changing the use of diesel to low-CO_2-emitting fuels, changing to more fuel-efficient tractors, more efficient cultivation and manufacture of fertilizers and pesticides (commercial nitrogen fertilizer can be produced in plants which have nitrous oxide gas cleaning) as well as improved fertilization strategies (more precise nitrogen application during the cropping season). The cultivation of annual feedstock crops should, further, be avoided on land rich in carbon, such as peat soils.

The emisions can also be reduced from introducing new agriculture systems. Recent research indicates that farmers could produce more, and reduce the loss of soil carbon, by abandoning current land ploughing and harrowing practices in favour of "zero tillage". By using simple techniques of drilling seed into the soil, with little or no prior land preparation, the soil carbon

will be protected by the blanket of leaves, stems and stalks from the previous crops. Zero tillage systems provide higher yields at less cost and less emissions, both from less direct CO_2 emissions (from the soil) and less use of tractor fuels (FAO, 2001).

Most WTT analyses assume a certain biomass yield per hectare. However, discussions on how to improve the yield from energy crops and the conversion efficiency are ongoing. Craig Venter argues: "The ability to construct synthetic genomes may lead to extraordinary advances in our ability to engineer microorganisms for many vital energy and environmental purposes" (Venter, 2003). Synthetically produced organisms (cell level bio-factories) will enable new direct methods of bio-engineered industrial production, such as the production of bioenergy, including ethanol and hydrogen as alternative fuels or substitutes for petrochemicals (Synthetic Genomics, 2009).

Researchers within forest biotechnology have used poplar to reveal key genes in the wood forming process. Recent work has focused on the genes and proteins involved in wood cell expansion (Mellerowicz and Sundberg, 2008), with the goal of increasing the cellulose content in energy crops. In real world, a plant sometimes spontaneously produces 75% cellulose instead of normally approximately 45%. When this gene is identified, it may be possible to use biotechnology on energy crops, increase the crop's cellulose biosynthesis, and thereby improve the area efficiency of for instance cellulosic ethanol (Sundberg, 2002).

Further, the starch content is often around 70% of the dry matter content, but with new ethanol wheat varieties this may be increased to up to about 75%, which may increase the exchange of ethanol from 55% up to approximately 58% (Börjesson, 2007).

Greenhouse gas emissions from agricluture/forestry can, thus, be reduced by introducing new types of crops, maybe developed through advanced agricultural engineering sciences. Radical developments, in biomass and biofuels production, may occur in the future and consequensly reduce the greenhouse gas emissions from the agriculture/forestry part of WTT analyses.

4. Case study: Cellulose based ethanol

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4.1 Background

Ethanol has already been introduced in large scale in Brazil, the US and some European countries and will most probably be one of the major renewable biofuels in the transport sector within the coming 20 years. Ethanol can be blended with petrol or used in a concentrated form in dedicated engines, taking advantage of the higher octane number and higher heat of vaporization, and it is an excellent fuel for future advanced flexi-fuel hybrid vehicles. Currently, ethanol for the fuel market is produced from sugar, e.g. cane sugar or sugar-beet, or starch, e.g. corn, wheat etc. The main production is in the US (from corn starch) and in Brazil (from cane sugar). However, these raw materials which also has to be used for animal feed and human needs, will not be sufficient to meet the increasing demand for fuel ethanol and the reduction of green house gases resulting from use of sugar- or starch-based ethanol is not as high as desirable (Farrell et al. 2006). Both these factors call for the exploitation of lignocellulose feed-stocks such as agricultural residues (e.g. wheat straw, sugar cane bagasse, corn stover) and forest residues (e.g. sawdust, thinning rests) as well as dedicated crops, for production of ethanol. Production based on these raw materials generates lower net greenhouse gas emissions, thus reducing environmental impacts. The raw materials are sufficiently abundant and also available world-wide.

The composition of lignocellulosic materials differs from one species to another. However, the main constituents are of the same type: about 50–60% carbohydrates in the form of cellulose (made up of glucose) and hemicellulose (various pentose and hexose sugars), which also can be fermented to ethanol, and some 20–35% lignin (see Table 4.1). Agricultural crops and hardwood contain more pentose sugars than does softwood. There are also valuable components such as extractives and fatty acids, which preferably should be separated prior to ethanol production. Lignin is a valuable co-product which can be used to generate heat or a solid fuel, which help improving the overall energy efficiency and process economics. In a longer perspective lignin may be used for production of chemicals; lignin is a very complex molecule, which contains numerous aromatic compounds of which some have interesting properties. In contrast to the co-product from starch-based ethanol production (DDGS, which is used as animal feed) there is no real limitation for the use of the lignin-rich residue.

Table 4.1. Composition of some lignocellulosic raw materials (% of dry matter) (from Olofsson et al., 2008b)

Raw material	Glucan	Mannan	Galactan	Xylan	Arabinan	Lignin
Agricultural residues						
Corn stover	36.4	0.6	1.0	18.0	3.0	16.6
Rice straw	34.2	-	-	24.5	-	11.9
Sugar cane bagasse	40.2	0.5	1.4	22.5	2.0	25.2
Wheat straw	38.2	0.3	0.7	21.2	2.5	23.4
Switch grass	31.0	0.3	0.9	20.4	2.8	17.6
Hardwood						
Salix	41.5	3.0	2.1	15.0	1.8	25.2
Softwood						
Pine	46.4	11.7	-	8.8	2.4	29.4
Spruce	49.9	12.3	2.3	5.3	1.7	28.7

The challenges in biomass based ethanol production are mainly related to the conversion steps as cellulosic materials are much more difficult to break down to monomer sugars compared with starch. One of the major challenges is to improve the yield of sugars from hemicelluloses and cellulose in a cost effective way (Hahn-Hägerdal et al., 2006). This requires improved pretreatment methods, cheaper and more efficient cellulose degrading enzymes and novel technology to do this at high solids concentration. Another challenge is to develop robust fermenting organisms, which are more tolerant to inhibitors and ferment all sugars in the raw material, i.e. both hexoses and pentoses in concentrated hydrolysates at high productivity and with high ethanol concentration. Increased process integration, in order to reduce the number of process steps, decrease the energy demand and reduce the amount of fresh water and waste streams by re-using process streams, is also a challenge that has high research priority today (Wingren et al., 2008).

Softwoods, e.g. pine and spruce, are more difficult to hydrolyse due to higher lignin content but also due to the structure of the material. However, the advantage of these species is that the sugars are mainly hexoses, which can be fermented by normal baker´s yeast with a theoretical yield of ethanol higher than 400 liter per ton dry matter. The small amount of pentoses present can then be converted to biogas in an anaerobic digestion (AD) step, which is used to convert the remaining organic substances present in the stillage stream from the ethanol production.

Other crops, e.g. agricultural residues like corn stover, wheat straw and sugar cane bagasse as well as hardwoods like aspen and salix, are more easy to hydrolyse to monomer sugars, than soft wood, but on the other side the hemicelluloses consist mainly of pentoses (xylose and arabinose), which are more difficult to ferment to ethanol (Öhgren et al., 2006; Sassner et al., 2008). This requires either genetically modified baker´s yeast or another type of micro organism.

During the last years several pilot and demo scale plants have been constructed around the world. In Sweden there is the pilot plant in Örnsköldsvik that has been in operation since 2005. This is one of the few plants that can process wood materials. Most of the other plants are focused on agricultural residues, like corn stover and different types of straw. The demo plant that has been in operation the longest time is the Iogen plant in Ottawa, which has a capacity to produce around 2000 m^3 of ethanol per year from straw. This cellulosic ethanol is sold at a Shell filling station as Shell is a main owner of the plant. Other demo plants based on straw

have also been constructed in Europe, one in Salamanca Spain by the company Abengoa and one in Kalundborg, Denmark owned by Inbicon which is a subsidiary of Dong Energy. They have rather recently been taken into operation.

4.2 Improvement potential: Integration with heat and power plant

Integration of cellulose-based ethanol production with a combined heat and power plant (Figure 4.1) was investigated for conditions prevailing in Sweden and it is the main strategy pursued in the Swedish cellulose-to-ethanol effort (Sassner and Zacchi, 2008; Sassner et al., 2007).

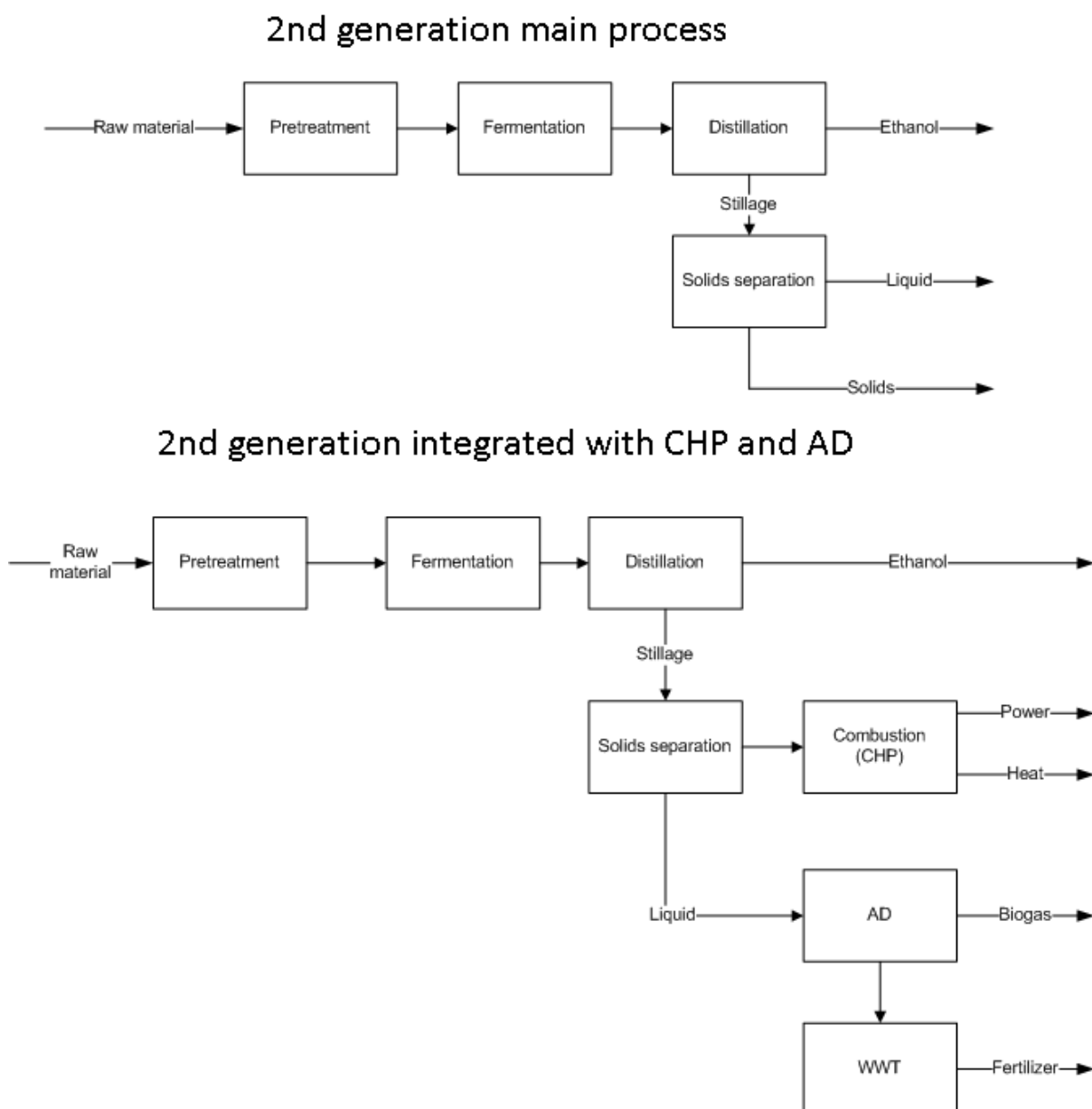**Figure 4.1.** Integration between ethanol production, combined heat and power production (CHP) and biogas production (anaerobic digestion, AD).

Various process configurations for integration of the ethanol production from spruce with a heat and power plant were studied using the flowsheeting program Aspen Plus based on experimental data from a mini pilot at LTH. Various options for the utilization of the solid

residue formed during ethanol production from spruce, such as the production of pellets, electricity and heat for district heating, were compared in terms of overall energy efficiency, exergy efficiency and production cost. Great importance was also given to utilisation of secondary steam from the process, either for internal purposes or for district heating. The effects of changes in the process performance, such as variations in the ethanol yield and/or the energy demand, were also studied. The process was based on SO_2-catalysed steam pretreatment, which was followed by simultaneous saccharification and fermentation. The capacity of the plant was 200 000 ton raw material (spruce) per year (based on dry matter).

For the five base case scenarios investigated, which is shown in detail in Section 4.6, the overall energy efficiency ranged from 53 to 92% (based on the lower heating values), the exergy efficiency, ranged from 41 to 60 % and a minimum ethanol selling price from 3.9 to 4.7 Swedish kronor per litre. The production cost should be seen more as a relative figure rather than absolute values as several assumptions are made in the economical evaluation. However, it was shown that the production cost could be decreased by 20% in the best scenario where district heating was utilized. From an energy point of view the district heating alternative is most attractive as it utilizes a large part of the low temperature waste heat from the process. However, this option restricts the location to the vicinity of larger cities of the plant as there must be a demand for the surplus heat.

Highly realistic improvements in the ethanol yield and reductions in the energy demand resulted in significantly lower production costs for all scenarios while the influence on the energy and exergy efficiency was less and the improvements did not always lead to increased efficiency.

Although ethanol was shown to be the main product, i.e. yielding the major part of the income, the co-product revenue had a considerable effect on the process economics. With the assumed prices of the co-products, utilization of the excess solid residue for heat and power production was highly economically favorable. This was also the scenario that resulted in the highest energy efficiency while the scenario with only pellets as co-product, i.e. no excess production of electricity and heat, resulted in the highest exergy efficiency.

In a recent study on ethanol from spruce, performed within the EU project NILE (data not yet published), several configurations where the evaporation of the liquid fraction of the stillage stream from the distillation was replaced by anaerobic digestion (AD) followed by aerobic waste water treatment were investigated. In the best case the energy efficiency, based on LHV, for the combined production of ethanol and biogas was 59% and the overall efficiency, including the contributions from electricity and district heat, was 92%. This process configuration resulted also in the lowest ethanol production cost (see Section 4.6 for further discussion about potential integration with AD).

4.3 Improvement potential: Integration with first generation ethanol

One promising alternative is to integrate second generation (2G) cellulosic ethanol production with first generation (1G) starch-based or sugar based ethanol production to use the whole agricultural crop. Examples of agricultural residues are corn stover, wheat straw and sugar cane bagasse and trash.

Taking it further the two methods could be integrated at some suitable point in a plant to share some common process equipment. Figure 4.2 shows some possible integration schemes for a

starch-based 1G plant. Due to the similarities in the two processes several points for process integration exist. The easiest point would be after fermentation and solid residue separation before the distillation as the two processes would have separate and dedicated equipments for pretreatment, hydrolysis and fermentation. However, by combining material streams further upstream the equipment cost for adding a second-generation technology into an existing first-generation plant could be lower and the energy demand could be decreased.
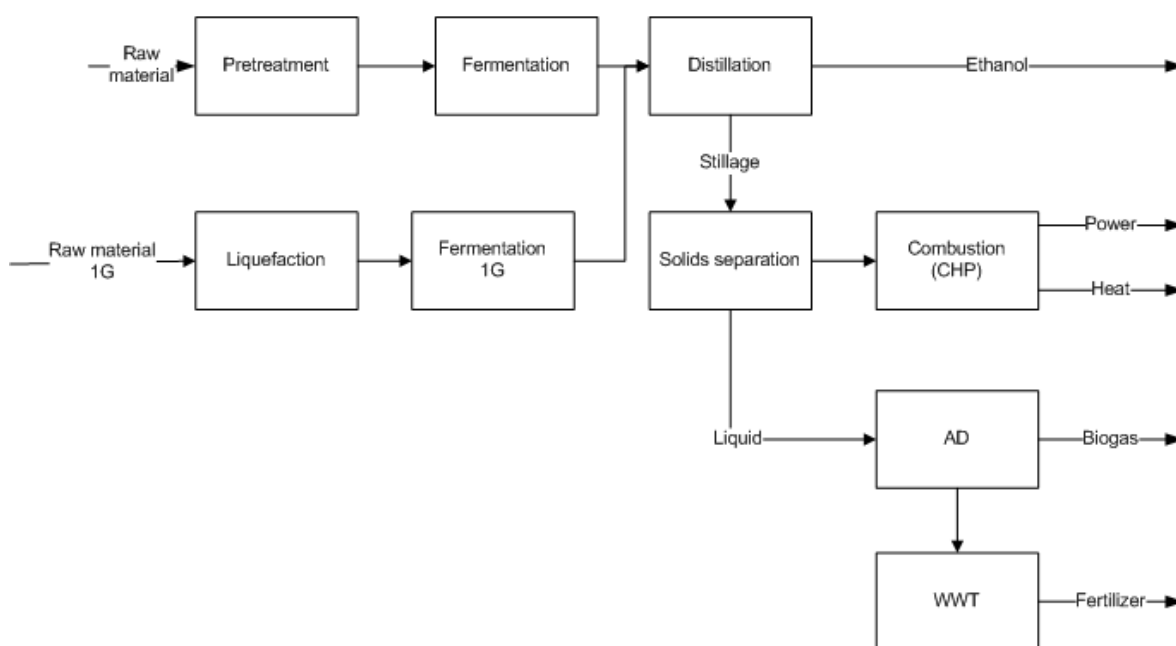

Figure 4.2. Integration between the production of the second and first generation ethanol.

Integration of the two concepts can be beneficial for both processes. As an example, the 2G ethanol production has an energy surplus in the form of lignin, which can be used in the whole plant. It is also usually difficult to reach high sugar and ethanol concentrations in the 2G ethanol production while starch- or molasses-based ethanol production require dilution of the sugar. By combining the process flows at some point in the plant, the energy situation in the distillation can be improved compared to two stand-alone plants. Also, the energy demand for evaporation of the stillage stream when applicable, (not shown in Figure 4.2) can be diminished for some of the process configurations. The demand for addition of nutrients will also be lower, since the 1G raw material contains sufficient levels of many of the required elements.

Integration may also alleviate some of the inhibitory effects occurring from formation of toxic compounds in the pretreatment step. If the process streams are mixed prior to fermentation the lignocellulosic streams will be diluted by the starch-based streams (Erdei et al., 2010).

Integration of first- and second-generation bioethanol production combined with CHP and biogas production (see Figure 4.2) will most probably result in higher ethanol yield, lower energy demand and lower production cost than by using a stand-alone second-generation ethanol production. To define the most optimal way of integration requires detailed studies, e.g., by flowsheeting calculations based on reliable experimental data.

4.4 Improvement potential: Choice of catalyst in pretreatment

The choice of catalyst in the pretreatment step is really dependant on how the various parts of the biomass are supposed to be used (Figure 4.3) (Galbe and Zacchi 2007).

One option is low pH methods, i.e. addition of acids, e.g. dilute acid hydrolysis and steam treatment with addition of acids. Most of the hemicellulose is usually hydrolysed to monomer sugars and to some extent oligomer sugars available in the liquid fraction after pretreatment. Depending on the severity, i.e. temperature, acid concentration and residence time, also a part of the cellulose may be hydrolysed. Also, a minor part of the lignin is solubilized as phenolic compounds, but the major part remains in the solid fraction although redistributed. These pretreatment methods usually also result in production of sugar degradation products, like the furans 2-furaldehyde (commonly called furfural) and 5-hydroxymethyl 2-furaldehyde (abbreviated HMF). A low pH method method is probably the best option if both hemicelluloses and cellulose sugars are to be converted to ethanol.

A second option is high pH methods, e.g. alkaline pretreatment and wet oxidation with addition of alkali. These methods result in partial solubilization of hemicellulose and solubilization of the major fraction of the lignin. The hemicellulose sugars that are solubilized are however obtained mainly as oligomer sugars. This makes it possible to utilize a part of the hemicelluloses with high molecular weight as starting materials for polymers, e.g. for barrier materials in food packaging. The liquid could also be used for biogas production as this can be performed without hydrolyzing the oligomers to monomer sugars. In case the hemicellulose sugars are to be converted to ethanol hemicellulases acting both on solid and dissolved hemicelluloses are required.

A third option is methods working close to neutral conditions at the start of the pretreatment e.g. steam pretreatment and hydrothermolysis. Most of the hemicellulose is solubilized due to the acids released from the hemicellulose, e.g. acetic acid. However, the sugars are obtained as a mixture of monomer and oligomer sugars. Also in this case hemicellulases are required acting on soluble oligomer fractions of the hemicelluloses if ethanol is produced. However, this is not needed if the hemicelluloses sugars are used for biogas production.
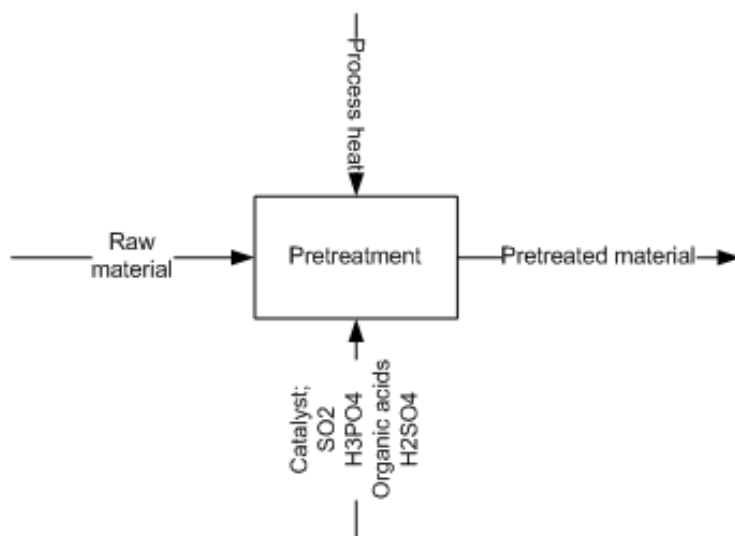

Figure 4.3. Pretreatment in ethanol production.

The catalyst may be chosen to be useful in the downstream processing. If biogas is to be produced from either the hemicelluloses sugars and/or the stillage stream an organic acid may be used in the acid catalyzed pretreatment. This acid may then be converted to biogas in the anaerobic digestion (AD). However, the liquid after pretreatment should in such case go directly to biogas production and not ethanol fermentation as this could be inhibited by the organic acid. The organic acid may be produced from the biomass in a separate fermentation step. Another acid that could be used is phosphoric acid which could then be neutralized with ammonia before fermentation resulting in ammonium phosphate which provides the AD with both nitrogen and phosphor. It would also be beneficial when the sludge from the AD is used as fertilizer for example for cultivation of energy crops.

4.5 Improvement potential: Expanded fermentation

The pretreatment provides a slurry in which the liquid fraction will contain solubilized hemicellulose – either in the form of oligomers or simple sugars depending on the catalyst used in the pretreatment – and in the case of alkaline pretreatment/wet oxidation solubilized lignin. The solid fraction will in all cases contain most of the cellulose present in the original raw material, some hemicellulose and – for the case of acid pretreatment – also most of the lignin.

The solid and liquid fractions may be separated – in which case the liquid fraction may be directly used in fermentation, whereas the solid part will have to be enzymatically hydrolyzed before fermentation. This process option is called SHF, for Separate Hydrolysis and Fermentation. The cellulolytic enzymes to be used will consist of a mixture comprising several endoglucanases (endo-1,4- β-glucanases, EGs), cellobiohydrolases (1,4-β-D-glucan cellobiohydrolases, CBH) and β-glucosidase. The former two act on the polymeric substrate and usually have descreet cellulose-binding modules that are essential for optimal hydrolysis of the insoluble substrate, whereas the β-glucosidase further cleaves the produced soluble saccharides (e.g. cellobiose) into glucose units. Early strain development of the industrially important filamentous fungus *Trichoderma reesei* gave at hand strains that secreets high levels of efficient cellulase mixtures. However, the cost of the enzymes in the hydrolysis has been identified as a major bottleneck in the process. For this reason the US Department of Energy (DOE) has been supporting industrial programmes for several years e.g. through grants to Novozymes A/S and Genencor (Danisco), aimed to lower the cost of cellulases (reviewed by Wilson 2009). The approach taken was, for example, to lower the production cost of enzymes by simplifying the medium and also try to increase the enzyme titer in the production e.g. by strain improvements. On a molecular level the issue of end-product inhibition has also been a target. The basic problem here is particularly cellobiose which inhibits some cellulases. For this reason it is difficult to obtain a full conversion of the cellulose or at least the hydrolysis process slows down. Approaches to optimize individual enzymes are e.g. rational engineering and directed evolution (Zhang et al., 2006) – or a combination of both these strategies.

A different problem concerns optimizing the enzyme mixture to be used. The synergistic action of cellulases has been extentisvely studied and it is also clear that other proteins can enhance the hydrolysis (Wilson, 2009). Although further strain improvements have been done, the enzyme mixture produced by some of most widely used strains of *T. reesei*, is typically deficient in β-glucosidase activity. This enzyme therefore has to be added to the mixture.

However, depending on the substrate used also other enzyme components, in particular hemicellulases but also various ligninases may be give improved hydrolysis. The remaining hemicellulose after pretreatment may block the cellulose, potentially circumvented by the addition of hemicellulases such as xylanse or mannanase which are the main endo-acting hemicellulases (Gilbert et al. 2008). A support for this strategy is the indicated synergy between xylanases and cellulases (Öhgren et al. 2007). The development of strategies where "helper" enzymes, such as hemicellulases and ligninases, are added can be expected to be rather tailored and directed to specific feedstocks, whereas improvement of cellulase mixtures will be more generic. It will be important to use realistic complex substrates to identify limiting factors. Discovery of new enzymes or proteins and optimal mixtures will be an important task. Here, post-genomic strategies as well as metagenomic strategies may prove to be important. An advantage of metagenomic approaches is that it allows cloning of genes from organisms that can not be cultivated *in vitro*.

An alternative to treating the solid and liquid fractions together, is to use both fractions simultaneously in a so-called SSF process (simultaneous saccharification and fermentation – see Fig 4.4). In this case, enzymes and yeast are both added and the enzymatic hydrolysis takes place together with the fermentation. This gives several advantages; the end-product inhibition caused by hydrolysis products is removed, one separation step is removed, and the hydrolysis reactor is no longer needed (for a review see e.g. Olofsson et al., 2008).

Expanded fermentation
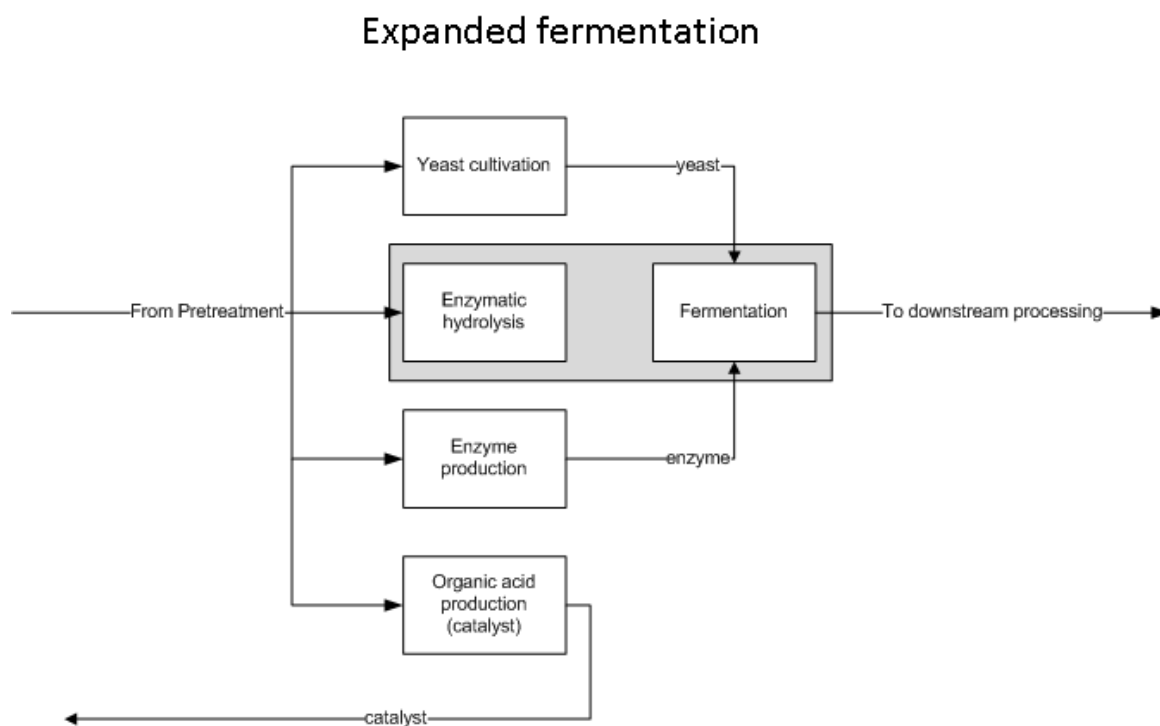
Figure 4.4. Expanded fermentation in ethanol production. The enzymatic hydrolysis may take place before the fermentation in a separate process step, or together with the fermentation, so-called SSF (simultaneous saccharification and fermentation).

However, the drawbacks are that the process flexibility is decreased in the sense that the conditions (temperature, pH) for the enzymatic hydrolysis have to be the same as those of the fermentation. The upper limit for the fermentation is typically around 36-38 C, depending on the yeast strain used, whereas the optimum temperature for enzymatic hydrolysis usually are

around 40-50 ^{o}C. Lower temperature for hydrolysis also increase the risk for contamination and potentially put additional demands on the enzyme preparation.

Another drawback relates to the large difficulties in separating the yeast after the SSF. Currently, reuse of the yeast is not possible in an SSF process. This necessitates the use of a low yeast loading in the fermentation, which in turn requires very tolerant yeast (reviewed by Almedia et al., 2007a,b).

Yeast tolerance is increased substantially by producing the yeast on the actual hydrolyzate to be fermented, and one can therefore see process integration advantages by on-site production of the yeast (as shown in Fig 4.4). Thereby, the necessary yeast concentration can be decreased by approximately 20-30%, which in turn may increase the overall ethanol yield by 3-5%. On site production also of enzymes, allows flexibility with respect to the usage of specific carbohydrate fractions from the pretreatment.

The pretreatment liquid in particular, consists of a mixture of different sugars. This relates to the fact that the hemicellulose composition – in obvious contrast to the cellulose – is strongly dependent on the raw material. From a fermentation point of view, there is an important difference between pentose rich materials, which include grasses, straw, hardwoods, and materials which do not contain very much pentoses, such as softwood (see Table 4.1). In the pentose rich materials, such as wheat straw or sugar cane bagasse, clearly the pentoses need to be utilized efficiently. The pentoses are typically – to a large extent – hydrolyzed already in the pretreatment and will be found in the liquid fractions. Options for use of the hemicellulose-derived sugar rich liquid stream include; production of enzymes; production of yeast, production of co-products, biogas production and/or fermentation. For the last option, efficient xylose fermenting organisms – preferably yeasts – will be needed (reviewed by e.g. Hahn-Hägerdal et al., 2007). Typically, the xylose conversion when fermenting at relatively high solid concentrations is still below 50% (Olofsson et al., 2008). A complete conversion of pentoses, may therefore improve the overall ethanol yield by another 20% in pentose-rich materials.

The SSF step as such can also be improved in terms of fermentation control, in which enzyme and substrate feed to the process are optimized. In this way, a substantial increase in the conversion of xylose can be obtained (Olofsson et al., 2008b).

A central issue for further development is to obtain higher ethanol titers (to decrease downstream costs). Final ethanol titers reported in lignocellulose conversion is normally below 50 g/L. Reaching higher values will require work at higher solid (WIS) contents – regardless of whether SHF or SSF concepts are used. The degree of conversion in enzymatic hydrolysis – in both SHF and SSF – decreases with increasing WIS contents at the same specific enzyme loading. Furthermore, the inhibition on the yeast metabolism also increases with increasing WIS content – primarily associated to the fact that the pretreatment slurry will be more concentrated with respect to the most important inhibitors. In addition, the mixing aspects in the highly viscous fiber solution during enzymatic hydrolysis must be solved.

Current development - on both enzymes and yeast strains – address the issue of attaining a more complete hydrolysis of the cellulose, a more complete fermentation of primarily the pentoses, an increased yeast tolerance towards inhibitors, and a decreased end-product inhibition of enzymes.

4.6 Improvement potential: Co-production with biogas

Both in 1G and 2G ethanol production, hexose fermentation to ethanol is the main production pathway. In agricultural residues, crops and hardwood, much of the organic carbon is bound in pentoses. Efforts are made to identify/develop organisms that can convert also these sugars to ethanol with high productivity, and yet another challenge is to make these organisms tolerant to the inhibitors that can occur in the fermentation broth.

An alternative is to explore the possibility of biomass fractionation that lignocellulose pre-treatment gives. Removal of the liquid after acid catalysed steam pre-treatment leaves a solid residue which retains the majority of the hexoses and lignin, while pentoses and other carbon sources, including potential fermentation inhibitors, can be transferred to anaerobic digestion (AD) with biogas production. In anaerobic digestion not only hexose and pentose sugars can be degraded, but also proteins, organic acids, lipids and other organic compounds. The development of integrated processes for ethanol production and AD is part of the ongoing research in collaboration between Lund University and the Swedish Agricultural University (Kreuger et al., 2010; Sipos et al., 2010), Figure 4.5.

This removal of the liquid fraction before further enzymatic hydrolysis and conversion and fermentation of the solid fraction has been shown to increase the fermentation yield of hexose based ethanol (Sipos et al.., 2010). Further, apart from yielding biogas, anaerobic digestion of this liquid fraction has been shown to remove potential inhibitors such as HMF, furfural, acetic acid, lactic acid and lignin derived products. This liquid detoxification opens up for recirculation of process liquid, potentially reducing both environmental impact and costs (Torry-Smith et al., 2003).

After the ethanol fermentation and distillation, the remaining stillage contains residual carbon sources. AD of this process stream (Fig. 4.5) can further increase the yield of biogas from the process. The organic compounds not converted to ethanol will here be converted to biogas, and an additional advantage is that the yeast and the enzymes added upstream will also act as carbon sources, and will significantly contribute to the output of biogas.
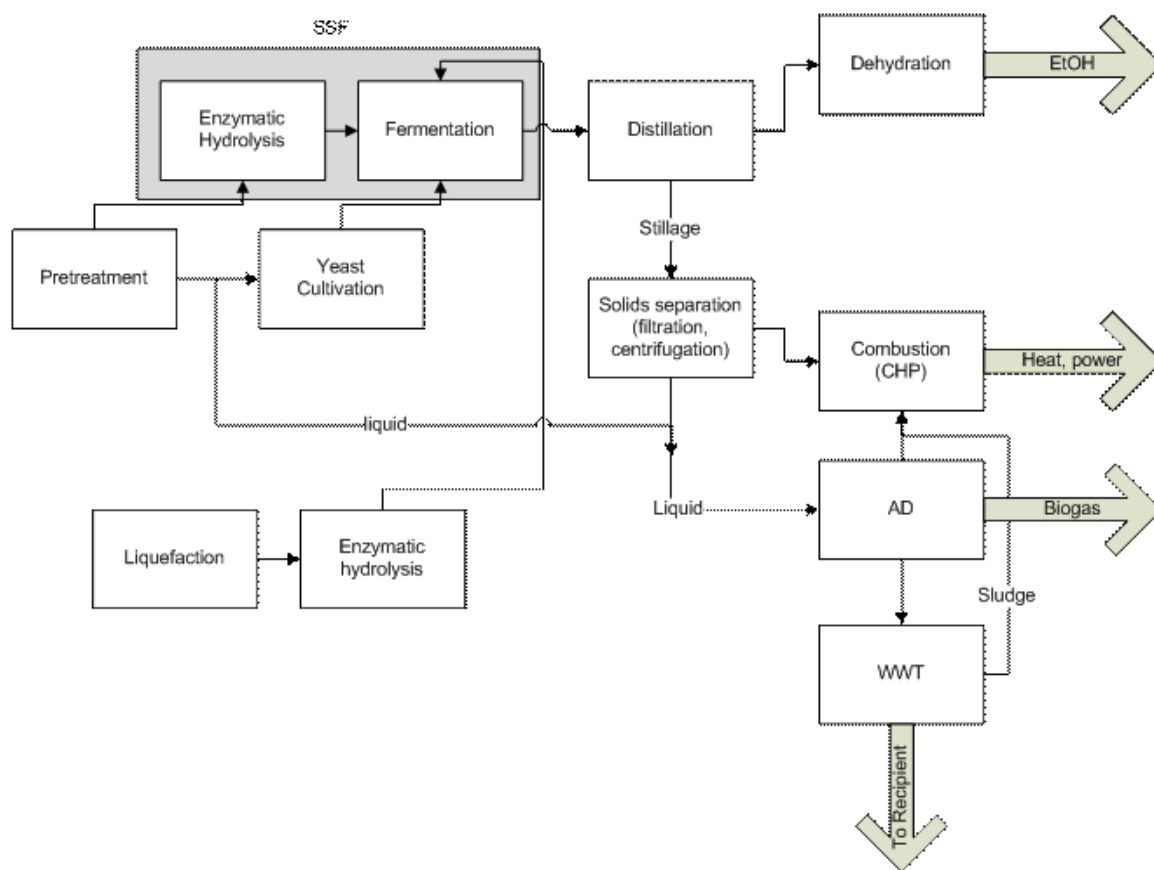
Figure 4.5. First and second generation technology for biogas production combined in first generation fermentation. Liquid from pretreatment, containing pentose sugars, are withdrawn and treated in the anaerobic digester instead of passing as dead load through the fermentation.

The concept of ethanol and biogas co-production has been investigated for several types of lignocellulosic biomass, like wheat straw, winter rye, hemp (Linde et al., 2007; Fan et al., 2006; Petersson et al.., 2007; Kreuger et al., 2010). In Figure 4.6, an example of the outcome of combined ethanol/biogas production from hemp is shown (modified from Kreuger et al., 2010). In scenario F, the pretreated biomass goes to ethanol production, and the stillage goes to AD. In scenario G, only the solid fraction after pre-treatment goes to ethanol production which increases the ethanol yield. In scenario H, both the liquid after pre-treatment and the stillage goes to AD, as shown in Figure 4.5. The latter increases both the ethanol and the biogas yield from the hemp. In addition, biogas is produced from the added enzymes and yeast.
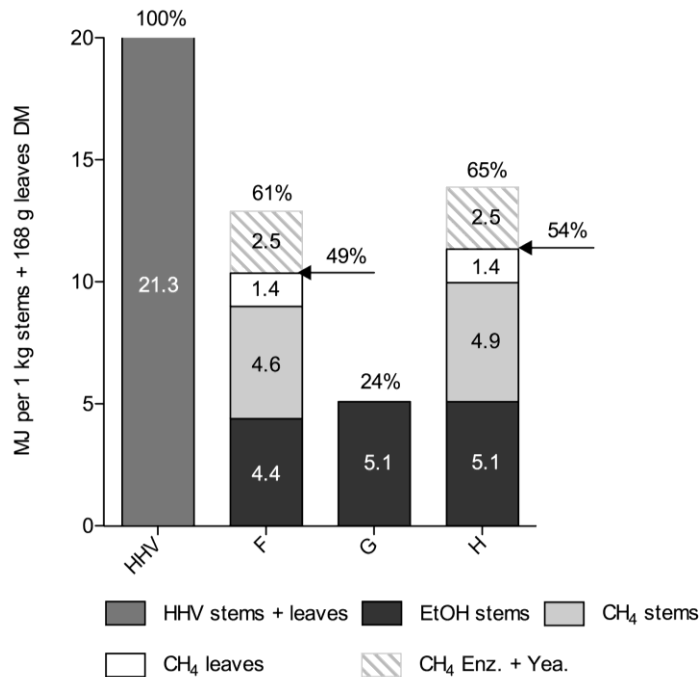
Figure 4.6. Energy yield for 1 kg hemp stems plus 168 g hemp leaves dry matter (DM) expressed as higher heating value (HHV) of ethanol and methane and compared to the HHV of hemp biomass. 'CH$_4$ Enz. + Yea.' designates biogas production from the degradation of enzymes and yeast added during SSF.

Integrated production of ethanol and biogas gives high output of fuels. In Figure 4.7 (modified from Kreuger et al., 2010) bar H shows the same co-production of biogas and ethanol as in Figure 4.6 above, where the total fuel yields are high partly because other carbon sources added in the system are fermented to biogas. In comparison, the theoretical ethanol yields from combined hexose and pentose fermentation of hemp stems, corn stover and *Salix* (dark grey bars) is shown. The co-production of gaseous and liquid fuels is a very promising alternative, which presently is being evaluated also from a techno-economic perspective.
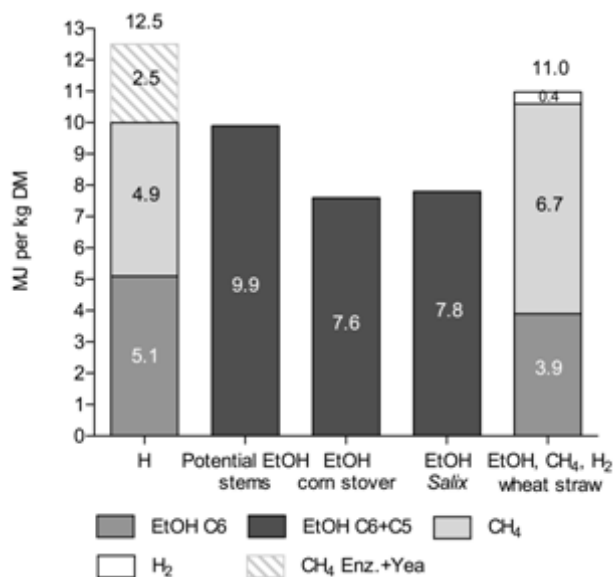
Figure 4.7. Biofuel yields after bioconversion of different lignocellulosic materials. The energy yield from 1 kg stem dry matter (DM) for scenario H in Figure B compared to: 90% of the theoretical ethanol yield from hexose and pentose sugars in hemp stems (Potential EtOH stems); the ethanol yield from hexoses and pentoses in corn stover (EtOH corn stover) from Öhgren et al. (2005); and Salix (EtOH Salix) from Sassner et al. (2006). All values based on the HHV of the biofuels.

4.7 Improvement potential: The value of co-products

The production of several co-products beside ethanol, e.g. electricity, biogas, heat and chemicals/materials is that it is easier to utilize all parts of the lignocellulosic material (see Figure 4.1) (Sassner and Zacchi, 2008). This gives both higher energy efficiency and a lower production cost (Figures 4.8 and 4.9). Many different process configurations are possible utilizing more or less of a fraction for a specific co-product. This can also influence the rest of the process in a beneficial way. Biogas, for instance can be produced from the stillage, which may replace evaporation as waste water treatment and reduce the energy demand in the process. Biogas may also be produced from the whole liquid stream after pretreatment, including the hemicelluloses sugars (see Section 4.6). This makes the ethanol fermentation easier as most of the inhibitors are removed, which makes it possible to perform the SSF at higher consistency and thereby decrease the distillation energy demand and the capital cost for SSF and distillation. Other examples are to utilize the solid fraction for production of heat and power and in the future to utilize e.g. lignin and hemicelluloses for chemicals/materials.

In Figure 4.8, the energy efficiency in various ethanol production systems design is shown. Case A represent stand-alone ethanol production process and residue streams used for generation of electricity, Case B pellets produced from the excess solids, Case C pellet production was supplemented with back-pressure power production, Case D residue streams utilized for heat and power production, and Case E represent residue streams utilized for heat and power production but the pressure of the evaporation condenser was raised so that the heat removed could be used for district heating.

In Figure 4.9, the production cost of ethanol in various production systems design is shown. Category Y represent a system where the enzyme dosage in SSF is reduced by 50% with a 6.8% reduction in ethanol yield and the required SSF residence time is increased from 72 to 96 hours. Category Y+ represents a system where the pretreatment is improved so that the sugar losses are reduced by 50%. It was also assumed that yeast able to grow on both hexose and pentose sugars is employed, and that 60% of the xylose and arabinose present in SSF are converted to ethanol together with the hexoses. The overall ethanol yield is increased by 12.1%. Category Q- represent a system where the SSF is performed at 12.5% WIS. The ethanol yield, SSF residence time and amounts of yeast and enzymes were maintained. A smaller amount of water is used in the process resulting in a reduced energy demand.

Category Y+Q- represent a system including a combination of the Y+ and Q- cases to study whether the effects of improved yield and reduced energy demand are additive.

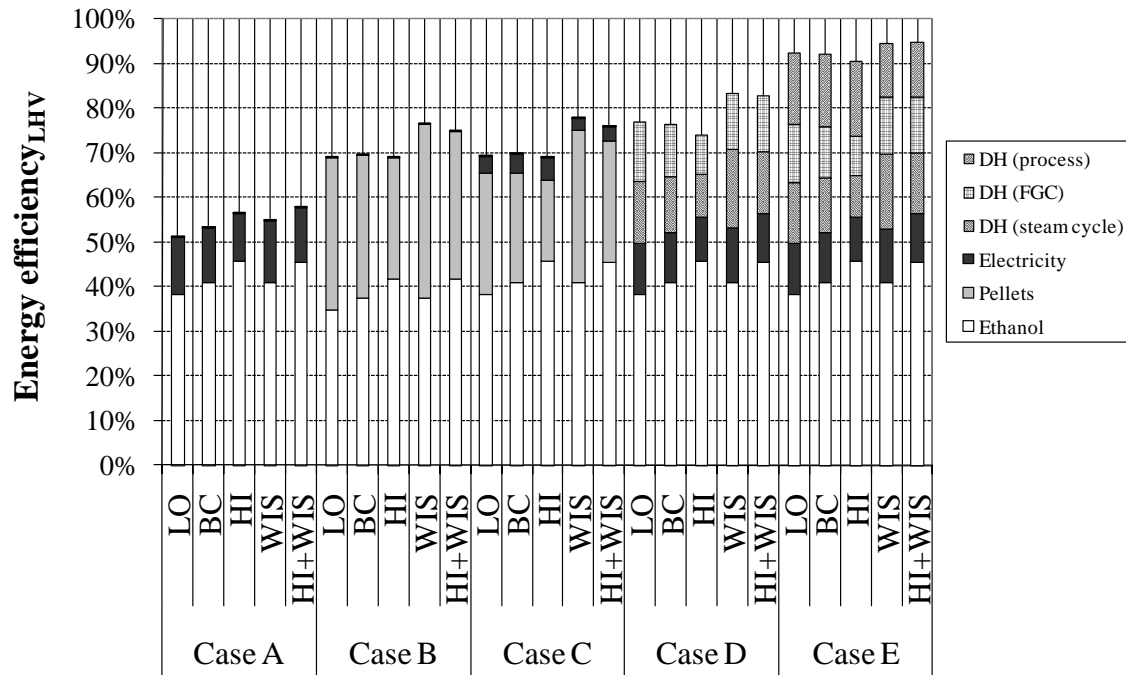**Figure 4.8.** Energy efficiency in ethanol production depending on various production systems design (see text for details).
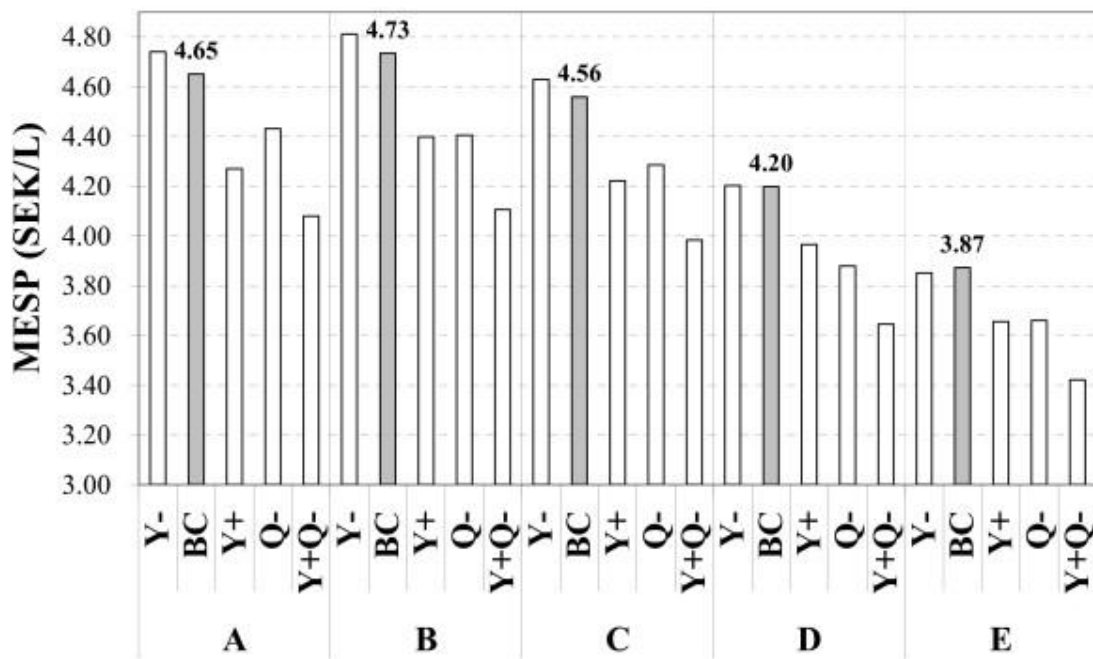

Figure 4.9. Cost efficiency in ethanol production depending on various production systems design (see text for details).

4.8 Improvement by integration with wood pulp production

There are various processes used for pulp manufacturing. Most of these are wet processes involving chemicals to separate the wood fibres, by dissolving the lignin and the hemicellulose, all or in part.

These processes are more or less the same as those used for pretreatment of wood as discussed in Section 4.2, with the difference that the pulp-making processes have been developed and refined over the years to provide pulp with optimum quality and yield to serve the purpose of the subsequent paper-making. Nevertheless, the liquors coming out of these processes all contain components from hemicellulose and lignin, some of which, especially the monomer hexoses, are directly suitable for fermentation to ethanol.

Already today, there is production of wood-based ethanol in Sweden, making use of the liquor from the sulphite process operated in Örnsköldsvik. That process is one of its kind it present day pulp-making, as it is the only plant in Sweden using Sodium sulphite. The production capacity of ethanol is 15000 tonnes, at a raw material use of 1.5 million m3 wood, also producing around 250000 tonnes of pulp and 55000 tonnes of lignosulphonate.

The total production of pulp in Sweden in all chemically supported process types summarized is in the order of 9 million tonnes pulp per year. If all this would be fermentable in the same way as in the existing plant, the production capacity for ethanol would be around 600 000 tonnes of ethanol. Unfortunately, this number is completely hypothetical.

The dominating part of pulp production is kraft pulp, around 80 % of the total. Kraft pulp is produced in an alkaline process, and the liquor cannot be fermented as easily as sulphite liquor. One reason has been described already in Section 4.4 on alkaline pre-treatment methods: The sugars are mainly obtained as oligomer sugars. Another point of concern is the sulphide in the kraft liquor, as this will "poison" the fermentation process.

The remaining chemically supported pulp production divides on a) sulphite pulp (magnefit pulp, ca 300 000 tonnes), b) CTMP, Chemo-thermo-mechanical pulp (about 700 000 tonnes), and c) Semi.chemical pulp (NSSC, about 200 000 tonnes). These potentially interesting sources of fermentable liquor to produce ethanol have not been explored to any significant extent, if at all.

4.9 WTT values presented in previous studies

There exist a large number of life cycle assessments (LCA's) of biofuels in general, and of bioethanol in particular (see e.g. Börjesson and Tufvesson, 2010). The main focus in these LCA's has been on the 1G ethanol based on sugar- and starch crops, and only to a limited degree on lignocellulose-based ethanol. The overall conclusion from these systems studies are that the environmental performance and energy balances are greatly affected by (i) the type of biomass raw material utilised, (ii) the specific design of the individual production systems, (iii) the geographical location, and (iii) the calculation methods utilised (see also Börjesson, 2009). Another conclusion is that ethanol from lignocellulosic materials normally scores better than ethanol from food crops from a WTT perspective. This is illustrated in Fig. 4.10 which shows a summary of the results from twenty five international studies regarding the energy balance of bioethanol systems.
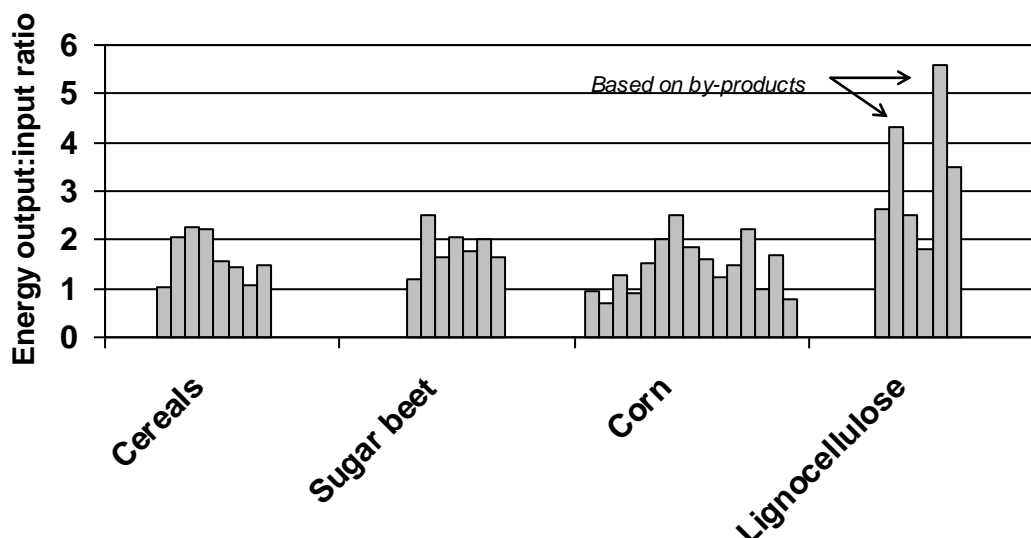
Figure 4.10. Summary of energy balances for various bioethanol systems published in 25 different international publications (Börjesson, 2006).

Since ethanol from lignocellulosic materials generate a significant amount of lignin as a co-product, the utilisation of this co-product will have a considerable impact on the energy and environmental performance of these systems. In previous systems studies, the lignin is normally assumed to be used to generate heat and electricity. The results in these studies often show a significant difference in the output of ethanol, electricity and heat due to the estimated system design. This is illustrated in Fig. 4.11 which show a summary of four different studies where the overall energy efficiency vary from 53% to 73%.

Figure 4.11 indicate that high overall energy efficiency will involve a high heat production and thus require a high potential heat sink. In Sweden potential heat sinks are, for example, district heating systems (DHS's) which may be integrated with ethanol production in the future. The number of DHS's that are large enough to utilise and distribute the excess heat from ethanol production is limited and depend on the size of the ethanol plant. A study by Börjesson (2007) show that if large-scale ethanol plant generating 600 GWh excess heat per year is developed in Sweden, the number of potential DHS's is limited to approximately 10. The total heat delivery per year in the DHS's is assumed to be twice as high, thus the heat production from ethanol plants is utilised as base-load all year around. However, if the excess heat from the ethanol plant is reduced to 100 GWh per year, the number of potential DHS's is increased to approximately 50. A conclusion from this is that a lower production of excess heat in ethanol plants will increase the practical integration potential with existing DHS's in Sweden.
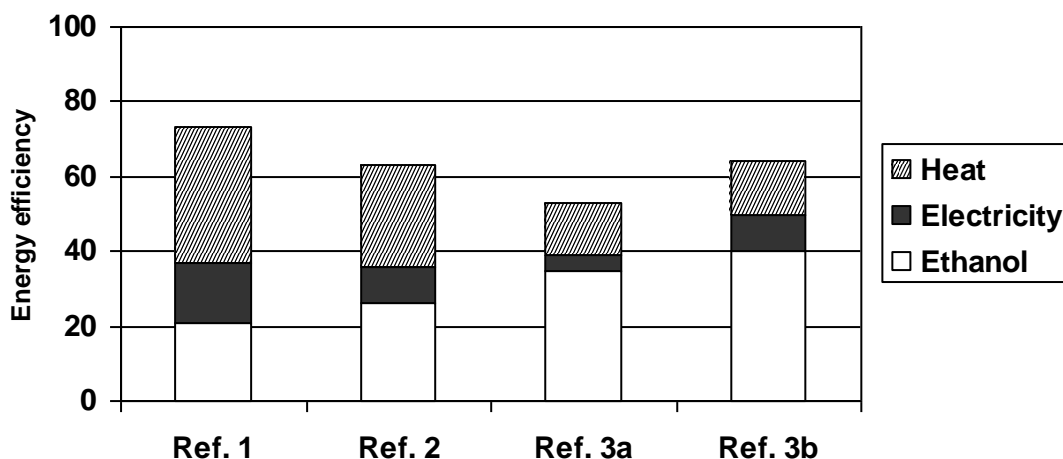
Figure 4.11. The energy efficiency in four previous system studies of lignocellulosic ethanol (Ref 1: BAFF, 2006; Ref 2: Goldschmidt, 2005; Ref 3: Hamelinck et al., 2005).

The greenhouse gas (GHG) balance of lignocellulosic ethanol is here exemplified by two studies; Concawe et al. (2007) and Börjesson and Tufvesson (2010), both assuming farm wood (short rotation coppice) as feed stock, Fig. 4.12. The GHG balances is in Concawe et al. (2007) shown to be, on average, 43 g CO_2-equivalents per km, having an interval from 35 to 55 depending on systems design. This is equivalent to a reduction of, on average, 74% compared with petrol (67-79%). In these calculations, excess electricity is assumed to replace stand-alone biomass-based electricity, thus not affecting the GHG balance. No excess heat is assumed to be utlised. In Börjesson and Tufvesson (2010), the comparable reduction is estimated to, on average, 85% when lignocellulosic ethanol is replacing petrol. These calculations are based on the assumption that lignin is dried and used as fuel pellets replacing wood pellets produced from fresh woody biomass. Today, dry by-products from saw mills (sawdust etc) are fully utilised for pellet production and other applications, thus an expanded pellet production normally requires new, undried biomass resources (Börjesson, 2007). This will lead to a somewhat improved GHG balance for the ethanol production system. The future market of wood pellets is estimated to increase significantly, e.g. in individual heating systems and within the European electricity sector replacing coal in condensing plants to full fill GHG reduction targets (see e.g. Hansson et al., 2009). Thus, the limitations are estimated to be much lower for excess lignin pellets from ethanol production than for excess heat.

An alternative to include co-products by calculating their indirect benefit when alternative products and energy carriers are replaced, e.g. electricity, pellets, heat etc by systems expansion, is to allocate the overall environmental impact of the production system between the ethanol and the co-products. This can be done either by their energy content (energy allocation) or their economic value (economic allocation), which is shown in Fig. 4.12 regarding the study by Börjesson and Tufvesson (2010). In the EU's Renewable Energy Directive (RED), it is suggested that biofuels GHG performance should be calculated using energy allocation including co-products generated in the biofuel process (thus excluding potential crop and harvest residues). According to Fig. 4.12, this calculation method should lead to a GHG reduction of lignocellulosic ethanol of approximately 80%.
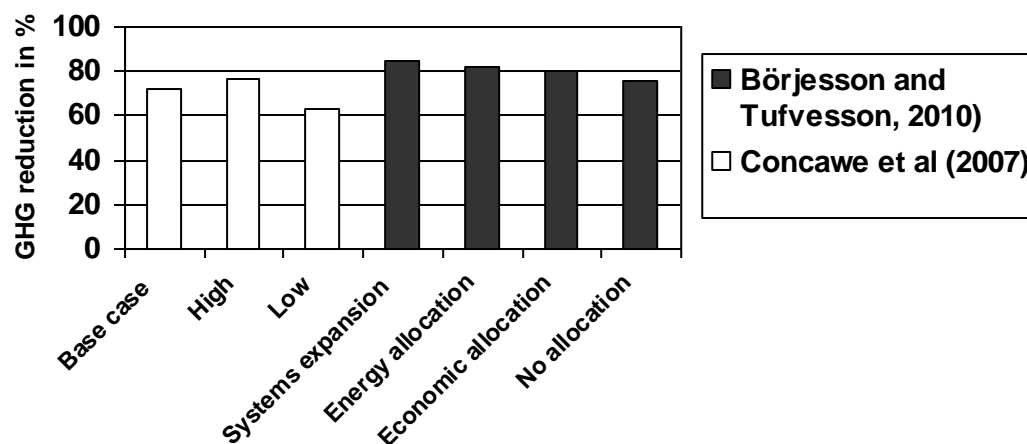
Figure 4.12. The GHG reductions of lignocellulosic ethanol when replacing petrol presented in two previous WTT studies.

A conclusion from the review of previous WTT studies of 2G ethanol is that the input data utilised are often old and not up to date, as well as the assumptions of production systems design. A large part of the data is five to ten years old (or more) and thus do not include the improvements achieved during the last years due to the research and development that has been carried out. As shown in Sections 4.2 to 4.8, there exist significant potentials of improvements both regarding individual measures and systems integrations. Production systems of bioethanol could be far more developed and complex than previous WTT studies normally describe, and the trend is that future production systems will include a much more diverse combination of feed stocks utilised and energy carriers produced. This is illustrated in the previous sections regarding, for example, integrated production of 1G and 2G bioethanol together with biogas, optimized utilisation of lignin for electricity, heat and pellet generation etc. Both improved systems integrations and individual measures will have an impact on future WTT values which are discussed in the following sections.

4.10 Improved potentials of systems integrations

Previous WTT studies (see Section 4.9.1) normally show an overall energy efficiency of approximately 65%, varying from 53% to 73%, in ethanol production systems also generating excess electricity and heat for external use (Fig. 4.11). These figures are relatively low compared with the results presented in Section 4.7 (Fig. 4.8), showing a potential improved energy efficiency in future developed systems. For example, the overall energy efficiency in developed systems is typically around 80%, varying from 75% to 95%. The generation of excess heat represent 18% up to 43%, thus the practical implementation of these developed systems will depend on existing heat sinks in e.g. district heating systems.

When heat sinks are limited, the overall energy efficiency could still be kept high by generating lignin pellets and electricity in addition to ethanol. Here, the overall energy efficiency may be, on average, 75%, varying from 70% to 80%. As discussed in Section 4.9.1, the future pellets market is estimated to increase significantly, especially within EU27. Thus, the practical limitations of these kinds of ethanol production systems are assumed to be lower than for ethanol systems generating large amounts of heat for external use.

Improved overall energy efficiency in ethanol production systems will normally also lead to improved GHG balances. The magnitude of these improvements will, however, depend on several factors, such as what kind of alternative energy carriers that are replaced by the co-products generated in developed ethanol systems. If, for example, lignin pellets is used to replace coal in European stand-alone condense power plants (typically 10-20% of the coal could be replaced in existing plants), the GHG performance of the ethanol production systems will be significantly improved. On the other hand, if excess heat is utilised in Swedish DHS's, the GHG improvements will be much lower if the alternative fuel is, for example, forest fuels. From a broader systems perspective, however, this amount of forest fuels will then be available for other purposes, such as producing transportation fuels by thermal gasification which could replace fossil vehicle fuels and thereby reduce GHG emissions. Thus, the impact on the GHG performance will to a large extent depend on the assumptions made of how the co-products are utilised and how the systems boundaries are set.

Despite these variations in performance due to assumptions made, a general recommendation could be concluded. Systems integration in ethanol production should be optimized in such a way that i) the overall energy efficiency is maximized based on the existing local conditions, such as available heat sinks in DHS's etc, and ii) the generation of high quality co-products (normally energy carrier) is maximized, consequently maximize the exergy efficiency (see Section 4.2). These strategies will also have implications on the economic conditions and profitability in ethanol production, which is discussed in Section 4.2 and 4.7.

Another example of how to improve the exergy efficiency in ethanol production is by co-production with biogas. Biogas is a high quality energy carrier which can be upgraded and utilised as vehicle fuel. Another option is to utilise biogas for electricity and heat production using gas turbines but this will normally lead to a somewhat lower environmental benefit than if the biogas is utilised as vehicle fuel (see e.g. Börjesson and Berglund, 2007). A conclusion from Section 4.6 is that an integrated production of ethanol and biogas from cellulosic feed stock could lead to a high over all energy efficiency. Examples are ethanol and biogas from hemp and straw having a total energy efficiency of approximately 60-65%, based on higher heating value (HHV), which is equivalent to roughly 65-75% based on lower heating value (LHV). Thus, if a maximized output of vehicle fuels is prioritised from cellulosic feedstock, the concept of integrated production of ethanol and biogas is promising for the future.

Biogas production could also improve the exergy efficiency in process integration concepts including heat and power generation (see Figure 4.1). In a recent study on ethanol from spruce, the best case the energy efficiency, based on LHV, for the combined production of ethanol and biogas was 59% and the overall efficiency, including the contributions from electricity and district heat, was 92%. This process configuration resulted also in the lowest ethanol production cost. Thus, the already high over all energy efficiency in ethanol production systems integrated with heat and power generation could be even higher if the liquid residue fraction is utilised for biogas production. The technology of utilising liquid organic waste streams for biogas production has been utilised in some pulp mills historically and are now developed further within the forest industry sector.

Biogas production may also be of importance in future systems where 1G and 2G ethanol production is integrated. These systems will generate digestion residues with high nutrient content, hence a valuable fertiliser product. This in return will lead to significant indirect GHG benefits when commercial fertilisers are replaced, since production of nitrogen fertiliser

cause emissions of fossil CO_2 and nitrous oxide (see e.g. Börjesson and Tufvesson, 2010). From a WTT perspective, this indirect GHG benefit may be equivalent to several percent.

Integration of the 1G and 2G ethanol processes is a most interesting concept for the future. As discussed in Section 4.3, this integration can be beneficial for both processes leading to synergetic effects. The lignin can be used as fuel in the whole plant, the distillation can be more efficient and the biological conversion may be improved due to a more optimal nutrient content when a mixture of raw materials is utilized. Besides leading to GHG benefits from a WTT perspective, this integration concept could be one efficient strategy to implement the 2G ethanol production in existing infrastructure. Today there is an increasing interest in the US and Brazil to also utilize corn stover in ethanol production from corn and bagasse in ethanol production from sugar cane, respectively.

There is a significant potential of integrating lignocellulosic ethanol production with existing pulp mills in Sweden, which is discussed in Section 4.8. Suitable types of pulp are sulphite pulp, CTMP, and semi chemical pulp, altogether representing approximately 20% of the total pulp production in Sweden. Utilising by-products in pulp mills as raw material in ethanol production leads to a high overall energy efficient which is shown in Figure 4.10. Furthermore, the WTT value is also favourable for these production systems based on waste streams. A value found in literature is 58 kg CO_2-eq/m_3 ethanol (Jonasson, 2007), which is equivalent to a GHG reduction of approximately 97% compared with petrol.

4.11 Improved potentials of individual measures

A second strategy to improve WTT values of 2G ethanol, in addition to process integration, is by individual measures. As shown in Sections 4.4 to 4.6, individual measures could be improved catalysts in pre-treatment, expanded fermentation and anaeraobic digestion of liquid residue streams.

In previous WTT studies, the estimated ethanol yield may vary from 20% up to 40% (see e.g. Fig. 4.10). An increase in ethanol yield is normally favourable from an exergy perspective, especially when the alternative co-product is heat, and thereby also from a GHG and WTT perspective. One way to increase the ethanol yield from lignocellulosic feed stock is by improved catalysts in pre-treatment. However, the pre-treatment method utilised will also affect the conditions of different potential process integration concepts. It is therefore necessary to define the design of the system and process integration when selecting the most optimal pre-treatment method.

Low pH methods are probably the best option if both hemicelluloses and cellulose sugars are to be converted to ethanol. The major part of the lignin will in these systems remains in the solid fraction. Thus, if process integrations aim at co-produce ethanol with pellets, and/or combined heat and power, low pH methods may be a suitable option.

High pH methods result in partial solubilization of hemicellulose and solubilization of the major fraction of the lignin. The hemicellulose sugars that are solubilized are obtained mainly as oligomer sugars which can be utilized as starting materials for polymers. The liquid could also be used for biogas production as this can be performed without hydrolyzing the oligomers to monomer sugars. Thus, high pH methods may be a suitable option when ethanol production is integrated with production of bioproducts, or with biogas production.

The ethanol production process includes two concepts: i) separate hydrolysis and fermentation (SHF) and ii) simultaneous saccharification and fermentation (SSF). As discusses in Section 4.5, there exist several potential measures to improve both these production options. Examples of measures in SHF are optimized individual enzymes and mixture of enzymes to increase the ethanol yield and lowering the costs of enzymes. Applying SSF leads to benefits, such as removing end-product inhibition, one separation step is removed and a hydrolysis reactor is not needed, but SFF also include some draw backs that needs to be overcome.

A central issue for further development by expanded fermentation is to obtain higher ethanol yields and thereby to decrease downstream costs. This will require work at higher solid contents – regardless of whether SHF or SSF concepts are used. Current development - on both enzymes and yeast strains – address the issue of attaining a more complete hydrolysis of the cellulose, a more complete fermentation of primarily the pentoses, an increased yeast tolerance towards inhibitors, a decreased end-product inhibition of enzymes. All these measures will have an impact on further WTT values leading to potential improvements in the GHG performance of ethanol production.

The WTT values of an integrated ethanol and biogas production system could be optimized by chose an organic acid in the acid catalyzed pre-treatment, which can afterward be converted to biogas. Another option is phosphoric acid which could then be neutralized with ammonia before fermentation resulting in ammonium phosphate which provides the AD with both nitrogen and phosphor. This will also improve the value of the digestion residues as a fertilizer and thereby the indirect GHG benefit when commercial fertilizers are replaced.

Another individual measure involving co-production of ethanol and biogas is removal of the liquid fraction after pre-treatment but before further enzymatic hydrolysis and conversion and fermentation. This result in two benefits from a WTT perspective: i) the yield of hexose based ethanol is increased, and ii) the process liquid is detoxified which opens up for recirculation of the liquid.

4.12 Conclusions and discussion

A general conclusion from this section about lignocellulosic ethanol is that there exist a significant potential of improvements and developments. Ethanol production systems could be far more complex and intelligently designed than previous studies show. The potential improvements consist of a huge number of combinations of process integration options and individual measures. This makes 2G ethanol production systems unique in some way and not directly comparable with other 2G biofuels. This makes it also impossible to draw general conclusions regarding WTT values and GHG balances of 2G ethanol, since the environmental performance of individual systems may vary significantly.

This complexity and large number of design options could be both positive and negative from a practical implementation perspective. A positive aspect is that the production system could be designed in an optimal way based on the specific local conditions. Thus, depending on available raw materials, heat sinks in DHS's, demand for biogas as vehicle fuel, existing 1G ethanol plants suitable for integration etc, 2G ethanol production systems may be designed quite differently to optimize the economical conditions and maximize profitability.

A negative aspect of the complexity may be that the amount of knowledge required to develop the most optimal production systems will be very large. Furthermore, several actors from

different competence areas are needed and have to be involved in the commercialisation of intelligent 2G ethanol production systems. This may be a potential barrier with similar importance as potential barriers of technical nature.

To conclude, 2G ethanol has a promising role to play in the development of intelligent and sustainable biofuel production systems in the future, but this requires improved knowledge and competence among all the different actors involved in this process. Furthermore, a more integrated work is needed between different research fields, such as process development and configuration and energy and environmental systems analysis (e.g. life cycle assessments) to optimize future production systems in a most efficient way regarding an economical, energy efficiency and environmental perspective.

5. Case study: Methane via gasification of solid biomass

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5.1 Background

Gasification is many times pointed out as the key conversion technology in all processes for the production of energy, fuels, and/or other products from biomass. In electricity production biomass is thermally converted by gasification into a combustible gas that can be used in a gas engine or turbine with electricity as well as heat as products. The advantage of the gasification route is the higher overall electric efficiency compared to the alternative of direct biomass combustion. Typical efficiencies of power generation in steam turbines are about 25%, while power generation via integrated gasification combined cycles (IGCC) offers first generation efficiencies greater than 35%, with up to 45-50% as a reasonable goal. In the production of fuels and chemicals the energy rich gasification gas is used in a catalytic process to synthesize a desired product with a high total efficiency. In this section methane, often called synthetic natural gas (SNG), will be discussed as the main product via gasification.

Concurrently with increasing prices of fossil natural gas and declining natural gas reserves, a raising interest of SNG has occurred. In particular, the interest for SNG from biomass has escalated during the last decade due to its high market value and theoretical high total efficiency of the process, up to 70% Mozaffarian and Zwart (2003). That is because SNG is produced from renewable energy sources the production can be seen as carbon dioxide neutral. In addition, if the carbon dioxide is captured and stored in a Carbon Capture and Sequestration (CCS) process the overall biomass-based SNG manufacturing has the potential to be carbon negative. Another incentive to produce SNG is when there are possibilities for gaining the excess heat which is a result from the strongly exothermic methanation reaction in a district heating system (Nemanova, 2010).

In the overall discussion on the utilization of gases from biomass gasification it is important to understand that gas specifications are different for the various gas applications, shown in Figure 5.1. The composition of the gasification gas is very dependent on the type of gasifier and very much on the gasification temperature.
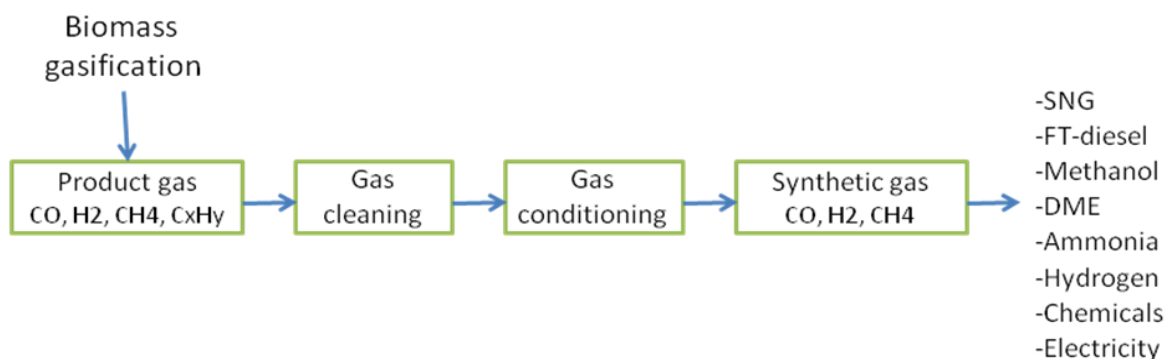
Figure 5.1 Synthetic gas production from biomass gasification and typical applications.

Gasification is the complete thermal breakdown of biomass into a combustible gas, volatiles, char, and ash in an enclosed reactor or gasifier (Knoef, 2005). Gasification can be divided into a two step endothermic process. It starts with pyrolysis at temperatures below 600 ˚C, the volatile components of the fuel are vaporized by a set of complex reactions. Included in the volatile vapors are hydrocarbon gases, carbon monoxide, carbon dioxide, methane, tar and steam. Char and ash are the by-products of the pyrolysis step. In the second step char is gasified through reactions with oxygen, steam and CO_2 producing CO and H_2. The heat needed for the endothermic gasification reactions is generated by partial combustion of the fuel, char, or gases – all depending on the gasification technology and reactor design.

In the second step, that is the actual gasification step, depending on the temperature the process yields a product gas with different components. The choice of the gasification agent determines the amount of nitrogen within the product gas (air, oxygen or steam). In this study methane is desired as the main gasification product and therefore low temperature gasification is the choice of gasification technology. For many other applications instead a minimum of methane in the syngas is desired, normally with high temperature gasification up to 1200 ˚C. The main reactor type is the entrained flow gasifier, further described in Section 6, DME production from black liquor.

Methane as product gas is generated by low-temperature gasification (around or slightly above 800˚C). The gasification processes can be divided into direct and indirect gasification processes. In case of air or oxygen as gasification agent a direct (autothermal) gasification is realized. Autothermal means that the necessary heat for gasification is produced by partial combustion of the biomass in the gasifier. A mixture of oxygen and steam is proposed to obtain a nitrogen free product gas to avoid a costly air separation. Another way to avoid an air separation unit has been establish, mainly for small and medium scale biomass gasification. This process is based on indirect gasification. Indirect implies that the heat is supplied indirectly via heat exchangers or a circulating heat carrier. Both types are currently studied/demonstrated, indirect gasification in Güssing/GOBIGAS, and direct gasification in the Värnamo plant.

Product gas composition
The composition of the product gas is mainly dependent on the type and operation conditions of the gasifier. The main parameters are the gasification agent, temperature of operation, pressure, and the type of gasification reactor. In Table 5.1 below gives an impression about the typical ranges of the two most interesting reactor types for SNG production. The indirect atmospheric steam blown gasifier lead to an almost N_2-free product gas with a high hydrogen and CO content. The methane concentration is relative high. For the direct oxygen blown

pressurized fluidized bed gasifier the product gas contains of a lower content of hydrogen and CO. The methane concentration is higher which can be beneficial for SNG production (Hofbauer et al., 2009).

Table 5.1. Typical ranges of product gas from low-temperature indirect and direct gasification (Hofbauer et al., 2009).

Component (%)	Low temp atm. Steam blown fluidized bed gasifier (data from Güssing)	Low temp direct pressurized oxygen blown fluidized bed gasifier (data from Värnamo)
H_2	35-40	23-28
CO	25-30	16-19
CO_2	20-25	33-38
CH_4	9-11	10-13
N_2	<1	<5

During the last years 2007-2009, a demonstration of a SNG poly-generation plant in an indirect atmospheric steam blown gasifier was carried out supported by the EU, the project was called BioSNG. A simplified process flow sheet of the SNG-process is shown in Figure 5.2. The demonstration plant is located in Güssing.
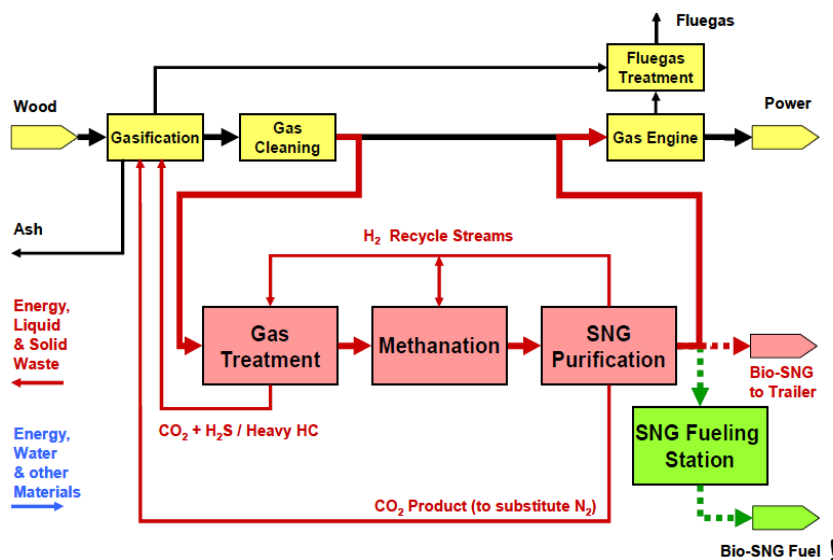
Figure 5.2 Flow sheet of the demonstration plant for BioSNG (Hofbauer, 2007).

A slip stream of product gas (H_2 and CO) is taken from the existing 8 MWth CHP-gasification plant in Güssing, running on wood-chips. The slip stream is already partly cleaned from particles and tars, however sulfur and chlorine have to be removed additionally. Thereafter a methanation reactor follows which is a fluid bed type reactor with Ni-based catalyst as bed material (Hofbauer, 2007). The gases coming out from the methanation reactor consists mainly of methane and carbon dioxide. Further cleaning of the SNG is necessary before feeding the SNG into a natural gas grid or a fueling station. (e.g. CO_2 separation and H_2). In Figure 5.3, the overall efficiency is presented for wood to SNG in the Güssing plant concept:
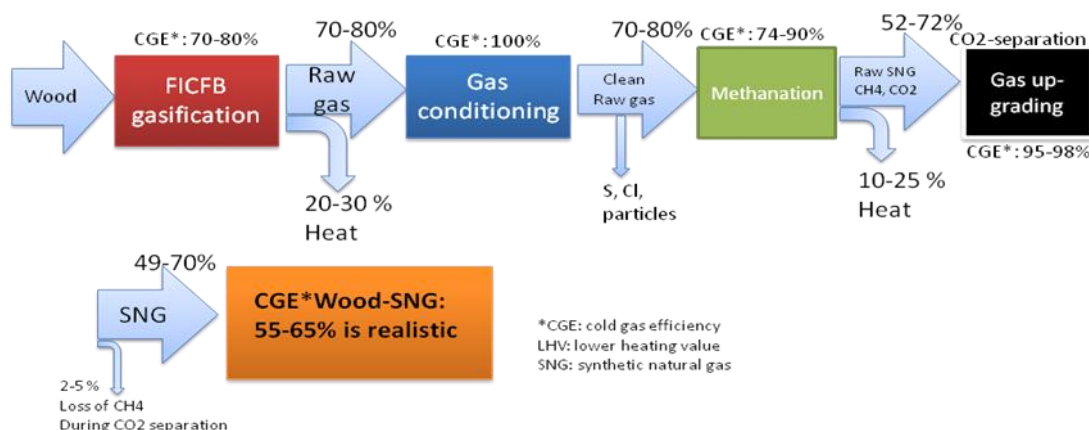
Figure 5.3 Overall Wood to SNG in the Güssing plant (Paul Scherrer Institute, 2009)

The Chalmers gasification unit is the same size and type as in Güssing, an indirect atmospheric steam blown gasifier, (2-4 MW of fuel, possibility with up to 8 MW of fuel in the gasifier). The Chalmers unit is flexible and can easily be switch between air and oxygen blowing conditions. The research is primarily focused on a concept solution and the gasification process for the production of methane. An interesting innovative concept solution is to design how to connect an indirect gasifier unit to an existing CHP boiler, and to show the flexible advantages with such a process (Heyne et al., 2008).

5.2 Improvement potential: Increased fuel yield, energy and material savings

In the discussion on the utilization of gases from biomass gasification it is important to understand that gas specifications are different for the various gas applications. Furthermore, the composition of the gasification gas is very dependent on the type of gasification process, gasification agent and the gasification temperature. However, all gases need additional gas cleaning and conditioning to afford a gas with the correct composition and specifications for the final application, e.g. synthesis.

For the production of SNG, a nitrogen-free product gas is desired. A nitrogen-free product gas can be produced by oxygen blown gasification or alternative by indirect processes. The indirect process is an economically attractive way for producing a nitrogen-free gas as no oxygen is required for the gasification. The conversion is generally complete whereas, direct gasification processes afford carbon containing ashes due to incomplete conversion (i.e. typically 90-95%) (Hofbauer et al., 2009). Direct pressurized gasification has the big economical advantage that the downstream processes (methanation) do not need to be pressurized. Concerning the scale indirect gasification processes have its merits at smaller scale (below 100 MW). Pressurized oxygen blown technologies can be applied at larger scale, 100-1000 MW, since the costs for building a oxygen separation plant is considerable. Therefore new innovative techniques for gas separation are of special interest with the aim of lowering the cost for an oxygen plant.

Mozaffarian and Zwart (2003) have done modeling based on comparable assumptions; a technical, economic, and ecological assessment has been performed for several biomass/waste-related SNG production technologies, with the objective to make a selection for future implementation of the most promising SNG options. The modeling results showed, that the upstream atmospheric steam-blown indirect gasification or direct pressurized oxygen-

blown gasification with downstream methanation routes are the most promising options for SNG production from biomass. In combination with downstream methanation, SNG production efficiencies up to approximately 70% (on LHV basis) can be achieved. The up-scaling potential of the indirect gasification technology was expected to be less than the direct pressurized oxygen-blown gasification, due to the complicated heat exchange between the gasifier and the combustor that is difficult to scale-up. This makes the technology mainly suitable for decentralized SNG plants (< 100 MWth). The fact that this technology does not require an oxygen plant is another positive aspect of this technology for decentralized applications. In contrary, the direct pressurized oxygen-blown gasification will be more suitable for centralized applications (> 100 MWth)

The specific investment costs of a system with a thermal biomass input of 100 MW were also modeled, and are higher for direct pressurized oxygen-blown gasification compared to indirect steam blown gasification, mainly due to the requirement of an oxygen plant.

In Figure 5.4 a simplified process of an SNG production route by a combined biomass gasification and a methanation step is showed. The gasification step is either oxygen-blown or an indirect gasifier. The product gas out from the gasifier passes a gas clean-up step for removal of the contaminants. Thereafter the clean gas enters the methanation process. Upgrading of the product gas in the gas conditioning step results in SNG as the final product.
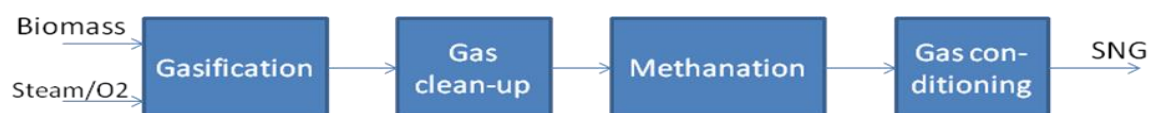
Figure 5.4 SNG production in a simplified combined gasification and methanation system (Boerrigter and Rauch, 2006).

Each synthesis gas production plant requires gas treatment facilities to purify product gases and also, in many cases to condition them, depending on the required gas quality. Whereas the gas purification system eliminates the components that would affect downstream processing or utilization of these gases, the purpose of a conditioning system is to remove undesired main gas compounds and to adjust gas components to the appropriate ratio. Especially, the ratio of hydrogen to carbon monoxide must be matched to the requirements of downstream synthesis. Depending on the type and composition of feedstock and the type of gasification process used, the gas purification and conditioning must handle the following impurities and undesired compounds:
- Particulates
- Tars/hydrocarbons
- Nitrogen compounds, i.e. NH_3
- Halogens, e.g. HCl
- Sulphur compounds, e.g. H_2S
- Volatile metals, e.g. alkali and earth alkali compounds
- Carbon formation

5.2.1 Gas treatment

Gas treatment is a great area of many different technologies, many are conventional, however many are currently being developed, since many of the existing technologies are too expensive. It is important to understand that gas treatment is more than gas cleaning.

Conditioning of the gas is the key for achieving a desired product gas as SNG. The main focus has for a long time been on different gasification technologies, but now the focus is on gas treatment and gas conditioning. The gas treatment is central for the efficiency of the process from biomass to end product and is also a considerable part of the cost of a plant. Therefore, improvements and research in the gas treatment technologies are very important for the development of a market for gasification.

The need for gas treatment after gasification is very different depending on the purpose for the gas. For simple gas firing a cyclone separator can be sufficient in many cases, while for gas engines and especially gas turbines further particle removal in a bag-house filter or a high temperature filter and also alkali metals in the gas has to be removed. For synthetic gases to be used in synthesis of SNG, FT-diesel etc. applications probably will require levels of sulfur and chlorine below ppm-levels and specially designed processes for removal of these substances are required, which are shown in Figure 5.5.
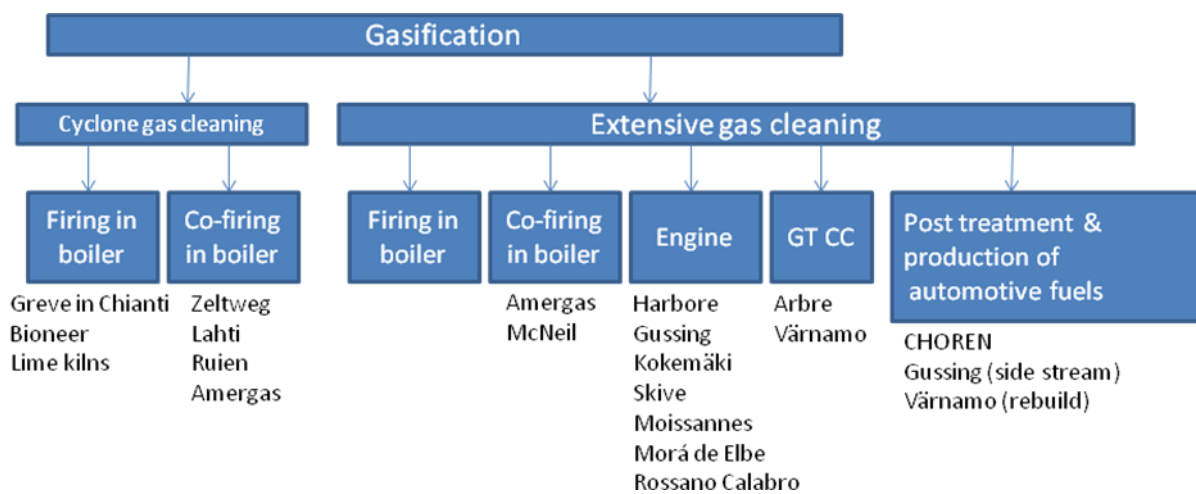
igure 5.5. Different end uses for biomass gasification and examples of plants (Larsson and Knoef, 2009).

5.2.2 Gas cleaning

Gas cleaning systems can be divided into four different areas (Karlsson, 2009):
- Removal of particles/dust
- Removal of water soluble gases as HCl, SO_2, NH_3 and HF
- Removal of NO_x
- Removal of very toxic substances as dioxin and mercury

There are many gasification systems developed to produce synthesis gas from biomass, and can be classified according to reactor design, gasification agent and the heat transfer or operating temperature. When it is desired to use the product gas as a synthesis gas directly from the gasification, only oxygen blown or allothermal gasification system can be used. Using air which contains nitrogen for gasification is not an option since the nitrogen has to be removed later on in the process when methane is desired as a product. Synthesis gas can be used to produce different products. The two main products today from synthesis gas are ammonia and methanol. Other possible future product gases are Fischer-Tropsch liquids, DME or chemicals like aldehyds and alcohols produced from biomass via gasification techniques. In this study the focus is on methane as a product (Hofbauer et al., 2009).

The syngas reactions have different requirements on the gas composition and gas quality coming out from the gasifier. For some it is important to have a specific hydrogen/carbon monoxide ratio, like the methanol synthesis, others not, like Fischer-Tropsch synthesis with iron catalysts. For SNG production it is important to have a low nitrogen content in the gas. The product gas from the gasifier can be changed or adjusted to the desired synthesis reaction by specific conditioning of the gas.

In general synthesis catalysts are very sensitive to poisoning, especially towards sulfur and chlorine compounds which have to be removed completely.

5.2.3 Gas clean-up requirements for the methanation process

It is important that the product gas out from the gasifier is thoroughly cleaned before entering the sensitive methanation process. Furthermore, the produced SNG must also meet the required standards before it can be injected into the natural gas infrastructure or delivered to fuel stations (Boerrigter and Rauch, 2006).

Particles: Particles can deposit on the methanation catalysts and make them inactive. In biomass gasification cyclones are normally used for the primary removal of particles, which 99% is usually separated. To achieve higher degrees of particle removal, for particles with a diameter of less than 1 μm, filters, scrubbers or guard beds would be necessary.

Tars/Hydrocarbons: Hydrocarbons and tars are a big issue for biomass gasification at low temperatures in general, not only they should be turned into product gas (H_2, CO, CH_4) and increase the total efficiency of the process, they may cause problems to the guard beds for removing particles before the methanation step and therefore has to be removed. Light hydrocarbons do not seem to affect catalyst activity and they do reform into methane, the same for higher hydrocarbons that are still present in the product gas after the clean-up step. However aromatic compounds have to be removed for SNG production, since polymerization reactions are likely to occur on the methanation catalyst.

Thermal cracking takes place at temperatures above 1000 ˚C. For SNG production catalytic cracking is more interesting, using catalysts such as dolomite or nickel. The tar reduction with catalysts take place at lower temperatures of about 750-900 ˚C. According to TPS, dolomite as a tar cracking catalyst after the gasifier can lead to tar reductions down to 100 mg/Nm^3, these levels could be acceptable depending on the process. Filters, scrubbers, activated carbon and zeoltic filters can also partly remove tars. At ECN a tar removal process called OLGA has been developed, which is a scrubbing system using special solvents. Overall it is vital to be able to selectively crack the tar components, including aromatic compounds, without cracking the methane that is desired in the total SNG process. Therefore, catalysts for selective tar cracking are an important area of research to cut the overall costs.

Nitrogen compounds: The nitrogen containing compounds as NH_3 and HCN are removed in the conventional clean-up down to 1 ppm and are not considered as an additional problem.

Halogen compounds: HCl is a strong poison to the nickel methanation catalyst. Most of the HCl and HF content of the gas is removed by scrubbing. However, an additional clean-up step to remove the residual HCl/HF to the desired concentration with a guard bed is necessary before methanation.

Sulfur compounds: Hydrogen sulfide, H_2S, is present in the feed gas. It can also be formed from any sulfur bearing compound by hydrogenation or hydrolysis over the nickel catalyst. Sulfur poisoning can occur at very low levels, i.e. below ppm. Most of the sulfur components in the gas will be removed by scrubbing, as for HCl and HF. The residual H_2S can be removed with a guard bed to the desired concentration and is also necessary before methanation.

Volotile metals: Metals like alkali and earth alkali compounds can be assumed to be poisons for the catalyst. However, the guard beds used for HCl and sulfur removal will also remove any particles or metals still present in the cleaned gas.

Carbon formation: Carbon formation can be predicted by thermodynamic calculations, however a number of other chemical reactions occur at the same time with unknown reaction rates. However it is useful to know if a specific mixture of synthetic gas and steam would have the thermodynamic potential for carbon formation when it is at equilibrium.

5.2.4 Gas conditioning

Gas conditioning includes all gas treatment steps to adjust the main gas composition of the desired synthetic gas, in this study methane (SNG). Production of methane requires carbon monoxide and hydrogen in the presence of a catalyst. The H_2/CO ratio prior to methanation should be at 3 for stoichiometric methanation. There are no gasifiers that produce a gas with a H_2/CO ratio of 3, therefore a Water-Gas Shift (WGS) reactor is necessary, see reaction (5.1). For the production of SNG the carbon monoxide content is higher than required and can be shifted into hydrogen through the WSG reaction. The main issue in the WGS reaction is the adjustment of the ratio between hydrogen and carbon monoxide and to remove carbon dioxide from the process, since it pushes the reaction backwards. The different gasification techniques produce mixtures of hydrogen, carbon dioxide and methane, and varying amounts of carbon monoxide.

Water gas shift reaction
To increase the hydrogen to carbon monoxide ratio or to completely convert CO into H_2 the WSG reaction is used:

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad (5.1)$$
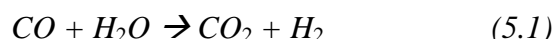
The WSG reaction depends on temperature, but is almost independent on pressure. Above 950 ˚C to 1000 ˚C, the equilibrium is rapidly established without a catalyst. However the WGS reaction is normally carried out at lower temperatures with a specific catalyst since the equilibrium towards hydrogen is favored at lower temperatures (Hofbauer et al., 2009).

Temperature ranges used in commercial WGS catalysts are:
- High temp WGS at about 300-500 ˚C (Cu-based catalysts)
- Low temp WGS at about 180-270 ˚C (Cu/Zn/Al catalysts)

Depending on the required hydrogen to carbon monoxide ratio the WGS reaction is carried out in two steps. In the first high temperature WGS reaction the bulk of the CO is converted. The second step is normally required if hydrogen production is aimed at, this second WGS step reduce the CO content to less than 0.5 vol%.

Carbon dioxide removal

The presence of CO_2 is undesired in most processes and also for SNG production and this compound should be removed. CO_2 can be removed from the synthetic gas by chemical and physical absorption with washing liquid or by adsorption with solid absorbents. For removal of CO_2 conventional technique can be applied as Pressure Swing adsorption (PSA) or Thermal Swing Adsorption (TSA).

5.2.5 SNG from methanation

Whereas high-temperature gasification processes yield synthetic gases with high concentrations of carbon monoxide and little methane, interest in SNG production is concentrated on gasification processes that yield product gases with high methane contents. SNG is a gas with similar content as natural gas but produced by methanation of H_2 and CO in gasification product gas. Methanation is the catalytic reaction *(5.2)* of carbon monoxide with hydrogen, forming methane and water. The reaction is normally operated at temperatures around 300-500 ˚C and at pressures of 1-5 bars:

$$CO + 3H_2 \rightarrow CH_4 + H_2O \qquad (5.2)$$
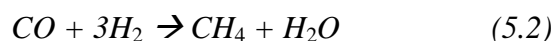
Consecutive and side reactions (shift conversion, Boudouard equilibrium, hydrogenation of carbon) make the calculation of equilibrium conditions very complex. The methanation reactions of both carbon monoxide and carbon dioxide are highly exothermic. Such high heat releases strongly affect the process design of the methanation plant since it is necessary to prevent excessively high temperatures in order to avoid catalyst deactivation and carbon deposition. The highly exothermic reaction generally creates a problem for the design of methane synthesis plants: either the temperature increase must be limited by recycling of reacted gas or steam dilution, or special techniques such as isothermal reactors or fluidized beds, each with indirect cooling by evaporating water, must be used.

5.3 Improvement potential: Co-production of SNG and FT liquids

There is a possible potential to co-produce SNG with Fischer-Tropsch (FT) liquids. Zwart and Boerrigter (2004) have evaluated different co-production systems. In the co-production concepts part of the SNG is produced by methanation of the FT off-gas, which already contains significant amounts of C1-C4 SNG compounds, see Figure 5.6. The additional required SNG is produced by dedicated methanation of parts of the gasification product gas. Their results show that co-production could results in higher biomass-to-fuel efficiencies than for separately production of SNG. The main overall conclusion of the study is that the co-production of FT transportation fuels and SNG from biomass is economically more feasible than the production of energy carriers in separate processes. Co-production of FT transportation fuels and SNG could become an economic feasible process.
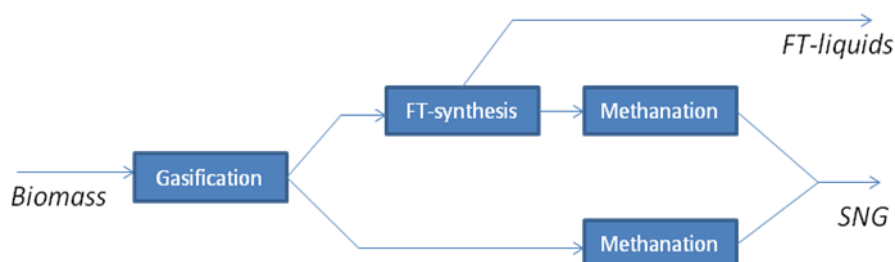

Figure 5.6 Integrated co-production of FT liquids and SNG (Zwart and Boerrigter, 2004).

67

The issue of co-production is an interesting possibility and has to be more investigated in system studies before drawing any conclusions.

5.4 Improvement potential: Process plants and integration

For the total economy of a plant the processes need to be integrated. As for the co-production of SNG and FT liquids polygeneration means that several products (heat, electricity, synthetic fuel) are produced together in one plant and sold at the market. System studies for the specific conditions in a desired plant has to be carried out in order to optimize the overall economics. A good example is the GOBIGAS-project that will demonstrate the feasibility of producing high yields of methane and the possibility to produce electricity while keeping total efficiency in the process over 90 %, by using the waste heat for district heating. The GOBIGAS project will use the described gasification technique as in Güssing and at Chalmers. Chalmers is also developing a concept solution designed to connect an indirect gasifier unit to an existing CHP boiler, providing flexibility to the process. This could be applied with great advantage at a district heating plant in which the demand for heat changes with the seasons. Having the gasifier attached to (and not integrated in) existing conventional units also reduces the risk of interruptions in the conventional production.

5.5 Discussion and Conclusions

Comparing production of methane via gasification with the production of other synthetic fuels, methane has a great advantage of having a high efficiency in respect to biomass converted into methane. At the moment the production of methane from biomass through gasification is of major importance, high yield, good WTT values, and most important a high expected price for SNG and total energy efficiency. Both the upstream atmospheric steam-blown indirect gasification (Güssing, GOBIGAS) and the direct pressurized oxygen-blown gasification (Värnamo plant) with downstream methanation routes are the most promising options for SNG production from biomass. In combination with downstream methanation, SNG production efficiencies up to approximately 70% (on LHV basis) can be achieved. The up-scaling potential of the indirect gasification technology is less than the direct pressurized oxygen-blown gasification, due to the complicated heat exchange between the gasifier and the combustor. This makes the technology mainly suitable for decentralized SNG plants (< 100 MWth). The fact that the indirect technology does not require an oxygen plant is a positive aspect of this technology for decentralized applications. In contrary, the direct pressurized oxygen-blown gasification will be more suitable for centralized applications (> 100 MWth).

For future research within the SNG area the main focus should be on the gas up-grading, including gas cleaning and gas conditioning to obtain high efficient processes were catalysis will play an important role. Therefore it is of high importance to remove the tar before the methanation reaction in the total SNG process.

- Catalyst development for selective tar cracking, it is important not to crack the methane already produced in the syngas.
- Aromatic compounds have to be removed, since polymerization reactions can occur on the methanation catalyst.
- High importance to have the possibility to test the integrated SNG process under realistic conditions, both at atmospheric and pressurized conditions in pilot-scale.
- Development of innovative separation techniques for gases for the possibility to use air as oxidation agent instead of oxygen in the gasifier.

6. Case study: DME via gasification of black liquor

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6.1 Background

A modern chemical pulp mill has a considerable surplus of energy, corresponding to about one third of the biomass available in by-products (electricity, bark, tall oil) from the process (KAM, 2003). In addition, the pulp and paper industry has the infrastructure and know-how necessary for the logistics of handling very large amounts of biomass. For comparison, a large mill in Sweden and Finland typically processes approximately 1000 MW of biomass. The pulp and paper industry could thus become an important supplier of renewable fuels and energy carriers of high value, e.g. by integration with the processes described in Section 4 and 5 or other routes from biomass to liquid fuels. In the present section we will focus on Black Liquor Gasification (BLG), which has some unique features in the integration with the pulp mill.

In this section the focus will be on the production of DME (dimethyl ether) but in a transition period, before all DME produced can be sold on the market, it may be beneficial to produce methanol instead. Methanol is an intermediate in the production of DME and the process routes are very similar. The gasification and gas cleaning processes described in this section applies to both DME and methanol production.

Moreover, it cannot be ruled out that also methanol returns as a blend (in gasoline) and neat fuel in Europe as well as in the rest of the world. The signals from China are very strong. China has legislated standards for M100 and M85 2009 and are expected to launch a national standard for M15 this year, 2010. Car manufacturing is global which means that brands which are sold in China must be methanol compatible in the near future.

6.1.1 Pulp mill overview

The most common pulping process, known as *kraft pulping*, is outlined in Figure 6.1. The process starts in the *wood yard* where logs are debarked and cut into wood chips, which are fed to the *digester*. The main objective is to dissolve as much of the lignin as possible while minimising the simultaneous dissolution of the cellulose. If high brightness is desired in the final product, the pulp also goes to a *bleach plant*.

In the recovery cycle, energy is recovered from the dissolved organic material, and the cooking chemicals are regenerated. Without the recovery cycle, the process would be both economically and environmentally impossible. The raw material for the recovery cycle is the spent cooking liquor that has been displaced during the washing of the pulp. Due to its colour, it is called *black liquor*. It contains approximately half of the organic material (mainly lignin) and almost all of the inorganic chemicals that were used for delignification.

After evaporation, the black liquor is burned in the *recovery boiler*. The sodium and sulphur is recovered as molten sodium sulphide and sodium carbonate – called *smelt* – that is tapped from the bottom of the boiler. Meanwhile, the organic material is completely oxidised in the upper parts of the furnace to provide heat for high pressure steam generation.
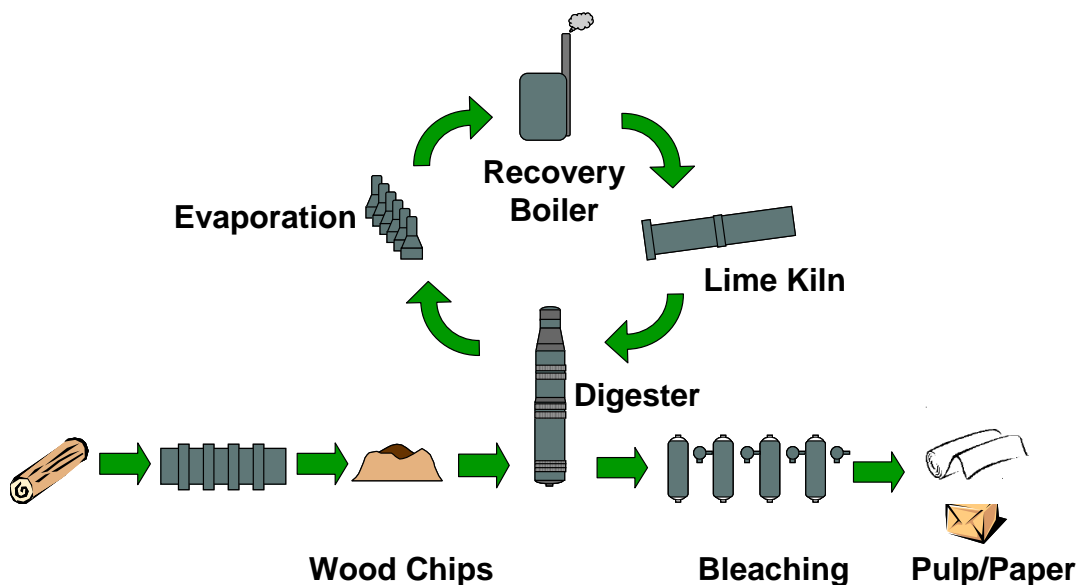

Figure 6.1. Pulp mill overview

After the smelt has been dissolved in *weak wash* it is known as *green liquor*. Before it can be reused in the cooking process, the carbonate ions in the liquor need to be replaced by hydroxide ions. This is done through a process called *causticizing* where the green liquor reacts with quick lime (produced in the lime kiln) to produce calcium carbonate and sodium hydroxide. After further processing, the liquor is turned into a mix of sodium sulphite and sodium hydroxide called *white liquor*, which is the cooking liquor needed to start the delignification process again.

6.1.2 Black liquor gasification as an alternative recovery technology

In a mill with black liquor gasification the conventional recovery boiler would be replaced by a gasifier, or operated with a gasifier in parallel. One important idea behind BLG is to take advantage of the fact that the recovery of the cooking chemicals in the recovery boiler is favoured by reducing conditions. Since gasification takes place under partial oxidation, the inorganic chemicals can be regenerated in the gasifier and directly processed into green liquor, while the main fraction of the organic material is converted into syngas.

Black liquor gasification can and must be well integrated into a pulp mill. The energy rich synthesis gas can be upgraded to, for example, FT-products, methanol or DME as well as chemicals. This system is called BLGMF, black liquor gasification with motor fuel production. Heat and other by-products generated in the gasification and synthesis steps can be utilized in the pulp mill.

The difference in energy balance for the two processes is illustrated below (Ekbom et al., 2005): a conventional pulp mill, where black liquor is combusted in the recovery boiler and falling bark is sold[16] (Figure 6.2), and a BLGMF plant where the black liquor is gasified and a

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[16] A part of the falling bark is gasified and used as fuel in the lime kiln.

bark/biomass boiler is used for steam and electricity production (Figure 6.3). Both processes regenerate the cooking chemicals used in the digester.

The overall energy balance of the mill will change radically if the recovery boiler is replaced entirely by a gasifier, since the black liquor will be used mainly for production of fuels that are sold instead of being used for steam and power production. The basic idea is to use low-grade biofuels (bark, forest residues, etc.) in an H&P boiler at site to make up for the deficit, according to the scheme outlined in Figure 6.3. The boiler should be designed to use a variety of low grade fuels. Motor fuels are thus indirectly produced from low-cost raw material.
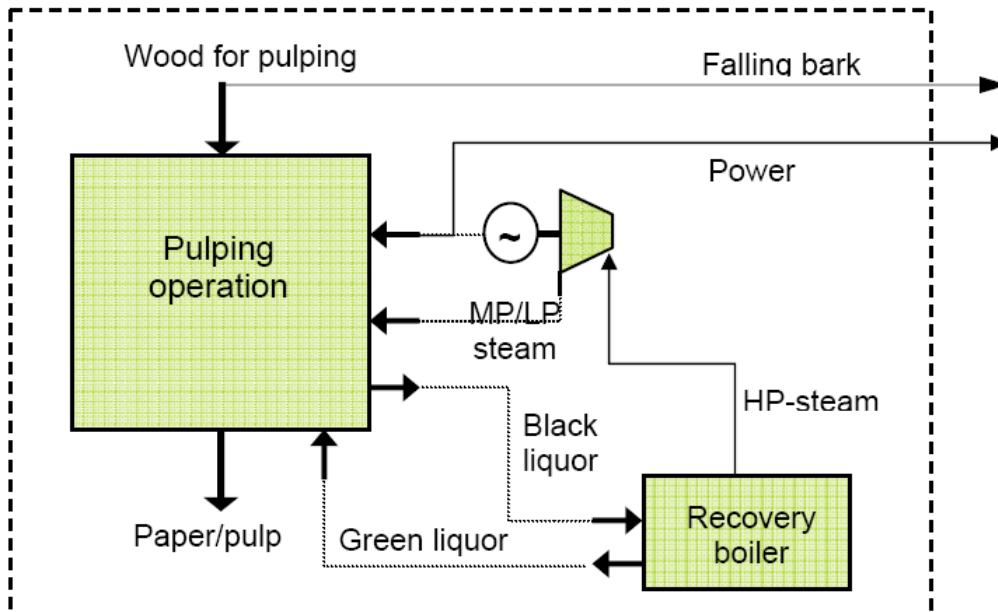

Figure 6.2. A conventional pulp mill with a recovery boiler and a bark boiler (Ekbom et al. 2005). Acronyms used in the figure are: HP= high pressure, MP= medium pressure, LP= low pressure.
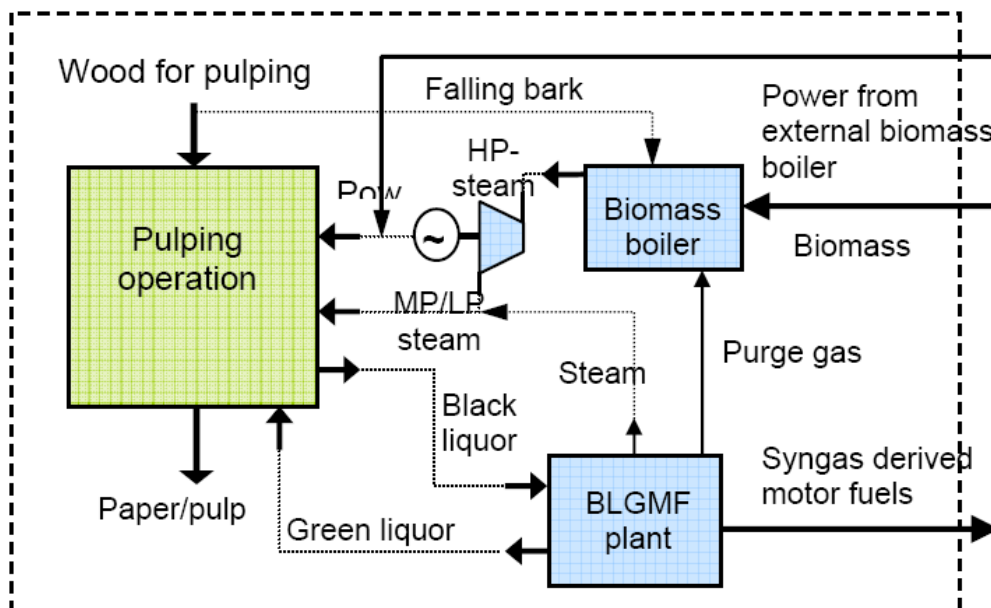

Figure 6.3. A pulp mill with a BLGMF plant and a new power boiler (Ekbom et al. 2005). Acronyms used in the figure are: HP= high pressure, MP= medium pressure, LP= low pressure, Pow= power.

71

6.1.3 Black liquor gasification with DME production

Quite a large number of process options for BLG have been proposed and developed to bench and pilot scale (Whitty et al., 2004). Two processes have reached commercial or near-commercial status; TRI (ThermoChem Recovery International), which is based on low-temperature steam reforming of the black liquor and Chemrec, which uses high-temperature partial oxidation to gasify the black liquor. At the time of writing, TRI is not pursuing this route for kraft black liquor and the following discussion therefore focuses on the Chemrec technology[17].

The Chemrec process is based on entrained flow gasification of the black liquor at temperatures above the melting point of the inorganic chemicals. The evaporated black liquor is gasified in a pressurized reactor and a synthesis gas is generated. The gasifier is operated at approximately 30 bar and 1000 °C. The black liquor consists of the inorganic cooking chemicals and organics in form of dissolved biomass. The gas and smelt, containing the cooking chemicals, are separated in the quench zone below the gasifier. The gas exits the gasifier and is further cooled in a condenser where the condensed water is used to generate steam. H_2S is then removed in an absorption stage, resulting in a sulphur-free synthesis gas (< 0.1 ppm sulphur components) mainly consisting of CO, H_2 and CO_2. Chemrec operates a black liquor gasifier pilot plant in Piteå, Sweden, since 2005 and have accumulated about 12 000 hours of operating experience to date.

The catalytic synthesis unit for production of synthetic fuels requires a high-purity syngas with a specific composition. The raw gas from the gasification process contains large amounts of CO_2 and H_2S, some COS and condensable tars, mostly benzene and also some small amounts of naphthalene. A commercial process which fulfils these different demands is the Rectisol unit which uses cold methanol as absorber liquid. Any small amount of tars are washed out prior to the Rectisol unit. Read more about the Rectisol process in Section 6.3.2.

A black liquor gasification plant with methanol/DME production is presented in Figure 6.4. The black liquor path via syngas to DME includes the following steps:

- Black liquor gasifier
- Quench – where the green liquor is produced
- Raw syngas cooler
- Tar wash
- CO_2 absorber
- Rectisol unit (sulphur removal)
- CO-shift reactor
- Syngas compressor
- DME synthesis unit
- Product distillation tower

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[17] Chemrec is planning to build a black liquor gasification plant with motor fuel production of the type that is described in this Section. The financial support (500 MSEK) from the Swedish Energy Agency is currently in an evaluation process, DG Competition, in Brussels. The decision from Brussels is expected during summer 2010.
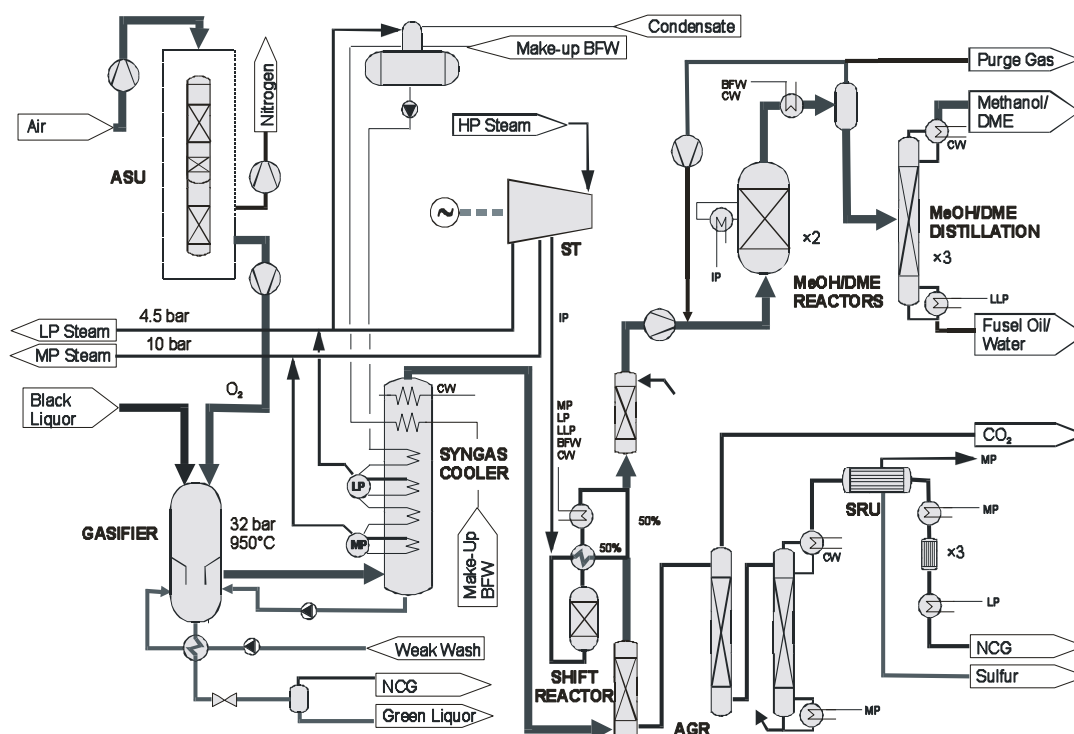
Figure 6.4. Black liquor gasification with DME or methanol production

6.1.4 Black liquor gasification benefits

In comparison with other potential biomass sources for chemical production, black liquor has the great advantage that it is already partially processed and exists in a pumpable, liquid form. Using black liquor as a raw material for DME or methanol production would have the following advantages:

- Biomass logistics are extremely simplified as the raw material for fuel making is handled within the ordinary operations of pulp and paper plant
- The process is pressurised, which enhances fuel production efficiency
- The produced syngas has a low methane content, which optimizes fuel yield
- Pulp mill economics becomes less sensitive to pulp prices as the economics are diversified to another product

In a well-to-wheel study (Larson et al., 2003), DME from biomass via black liquor gasification ranks the highest, among the biofuels, regarding energy efficiency and the lowest regarding contribution to the greenhouse effect by net CO_2 emissions. Today, all methanol and DME available on the market are of fossil origin.

6.1.5 Economics

The investment and operating costs for the BLGMF case with DME production have been calculated by comparing it with a base case, which is a mill that replaces its recovery boiler with a new recovery boiler. This is then compared with a mill where a black liquor gasifier with a DME production unit is installed instead of the new recovery boiler.

The investment cost and operating costs for the BLGMF unit is calculated as an incremental cost compared with the base case, defined above. Results have been presented in studies by Ekbom et al. (2003, 2005) and Larson et al. (2006), see Table 6.1.

Table 6.1. Investment cost for a BLGMF plant. Costs calculated in 2005 (Ekbom et al., 2005) have been adjusted to 2010 level by using the chemical engineering plant cost index. All investment and operating costs are based on a pulp mill capacity of 2000 ADt of pulp per day, which is a normal size for a modern mill.

Investment cost estimate		Recovery boiler	BLGMF - DME
Total investment cost	M EUR	204	430
Incremental BLGMF investment	M EUR	0	226
Operating benefits			
Incremental BLGMF operating benefits	M EUR	0	82.3[1]

1 The netback/selling price is set to 0.62 euro/liter

The operating costs are influenced by the extra income from DME. The additional costs are the biomass that needs to be imported to compensate the lack of steam and produce the required electricity[18]. There are also some extra chemical, water and maintenance costs. Sensitivity analysis has been performed and yielded a modest sensitivity on the production cost for all parameter changes (purchased biomass, purchased electricity cost and incremental investment cost) except availability.

The investment cost for the first BLGMF plant is higher than the numbers mentioned above. The investment calculations done assume that the concept is an "N[th] plant" assuming that all units are technically proven and reliable.

6.1.6 Potential for black liquor gasification

A modern sized pulp mill produces about 2000 ADt/day of pulp and generates 3400–3600 tonnes of black liquor (measured as dry content), or 1.7–1.8 tonnes of black liquor per tonne of pulp. Black liquor thus represents a potential energy source of 250–500 MW per mill and in 2005 the total world production of black liquor was 670 TWh.

A modern size pulp mill equipped with a black liquor gasifier can produce large amounts of DME. Table 6.2 below shows that a mill with a capacity of 2000 tonnes of pulp per day can produce 286 000 tonnes DME per year. An additional biomass flow to the new power boiler will be required at a size of 408 MW.

Table 6.2. Example of efficiencies and production in a modern pulp mill producing 2000 ADt/pulp per day converted to produce DME via black liquor gasification

Fuel options	DME
Biomass consumption (to biomass/bark boiler) [1)	408 MW
Black liquor consumption	487 MW
Fuel production, total	275 MW
Fuel production, total	286 000 t/year
Energy efficiency (LHV). Black liquor to fuel	56%
Biomass to fuel (biomass/bark to boiler)	67%

1) includes additional biomass for power generation in external boiler, see Figure 6.3

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18 Electricity production costs are based on a biomass-fired stand-alone plant with a conversion efficiency of 40%.

6.1.7 Technology which need to be proven before achieving commercial status

There is a need to demonstrate that renewable DME/methanol is available in practice in large scale and to show that the technology exists and will be proven to function. Some of the functions which need to be proven that they work in a large scale, oxygen blown plant are listed below:

- Material problems of the linings of reaction vessels. Ceramic material has proven to work for two years operation in air-blown, atmospheric service and for about a year under pressurized, oxygen-blown service but under pilot plant operating conditions (a lot of stops and starts). At least 1.5 year life length is expected for the ceramics in a large pressurized plant but it still has to be verified.

- Additional causticizing load, which is due to the sulphur split in the gasifier, and to co-absorption of carbon dioxide, e.g. in the quench. This item needs further attention and development in this area has high priority in DP1. This is however not an issue in the planned demonstration plant at the Domsjö mill. CO_2 absorption is positive in the sodium sulphite based process.

- Demonstrate reliability of major plant equipment under long term test. The Chemrec DP1 gasifier in Piteå has now been in operation for over 10 000 hours and lots of experience has been gathered. To reach availability numbers in line with recovery boilers Chemrec will install a spare gasifier train in the Domsjö plant. The gasification unit will be built as 3 x 50% capacity trains.

- Feed flexibility. The DP1 plant has gasified sulphite thick liquor for 27h (May 2009) and about 100h (March 2010). Tests went smooth and the sulphite liquor showed very good gasification characteristics and was more easy to gasify to full carbon conversion than kraft liquor. For kraft liquor the DP1 unit has shown to perform well with fast changes in solids content (drop without pre-warning from 73 to about 65% ds content). The process is also capable of fully (100% reduction) reduce sodium sulphate when the host mill recycle all precipitator ash to the evaporator section. No sulphate ends up in the green liquor.

- The investment cost for the first demonstration in industrial scale plant is high, and it is hard to motivate someone to invest in the first plant. Combination of investment grant and green incentives on the produced fuel is however enough to generate an attractive economy.

6.2 Improvement potential: Increased fuel yield, energy and material savings

A pulp mill and a BLGMF plant will be tightly integrated. In particular good heat integration can improve the economic performance of a plant. Figure 6.5 shows what integration between a pulp mill and a BLG plant with DME production may look like. The figure shows the energy flows in GJ per tonne produced pulp. Further improvements in fuel yield (measured as motor fuels produced divided by incremental biomass used) must be obtained by better utilization of the streams that are purged or where heat is lost. Some interesting streams from this point of view are marked with a red circle in Figure 6.5.
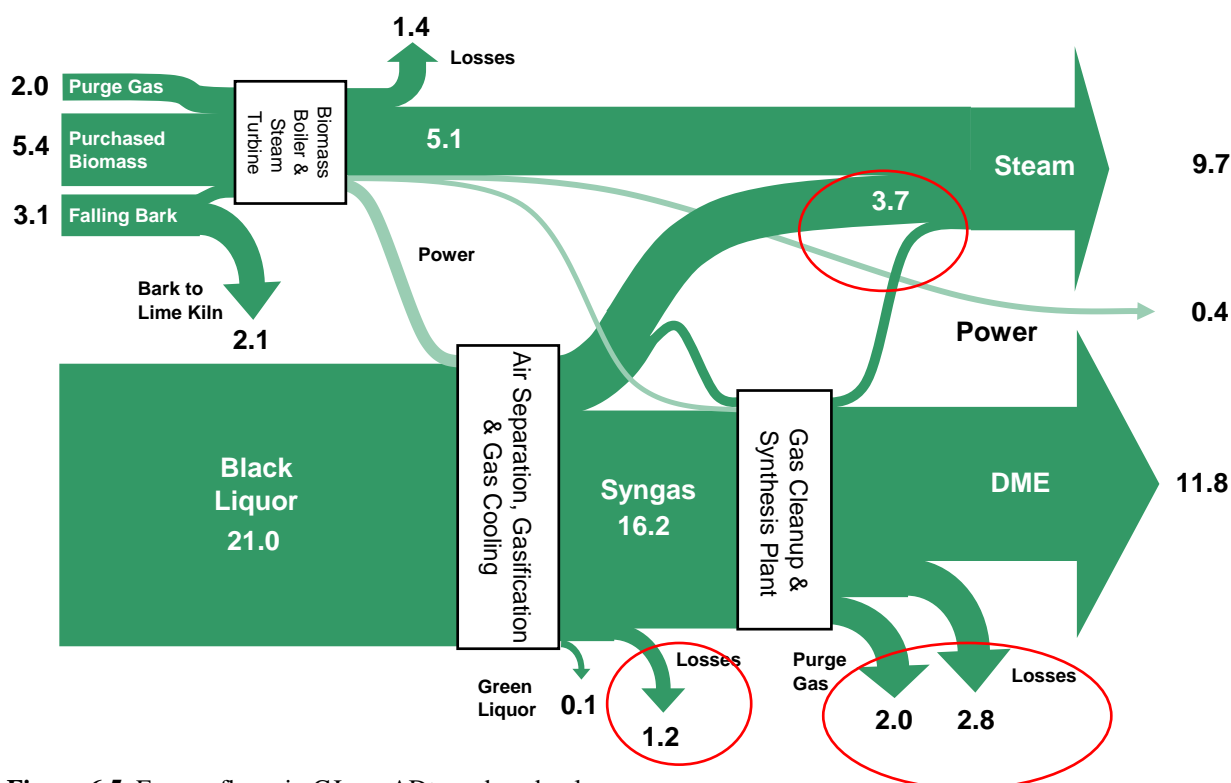
Figure 6.5. Energy flows in GJ per ADt produced pulp.

Increased steam production in the gasification and gas cooling system can be achieved by increasing the process pressure. Since the gas is saturated with water from the quench, the process pressure determines at what temperature the heat can be recovered. For example, there is almost no heat recovered as steam when the process pressure is below 10 bar, while at 30 bar approximately 80% can be recovered as steam. Higher pressures will not greatly affect the total amount of steam produced but will maximize medium pressure steam generation in relation to low pressure steam (Berglin and Stigsson, 1999). Increased export of steam to the mill will reduce the need for purchasing external fuel while the DME production remains constant. However, reduced production of steam in the power boiler will also reduce the electricity production and the influence on the DME yield will depend on the ratio in which electricity is recalculated into biomass[19].

The purge gas can be combusted in the power boiler and produce steam and electricity. Lower methane content in the syngas would reduce the amount of purge gas and thus increase the fuel yield.

Other minor energy losses (however roughly zero in an energy balance) from the system are tars (benzene and small amounts of naphthalene) from the gasifier and energy that leaves with the off-gases and waste water.

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[19] If electricity production is based on a biomass-fired stand-alone plant with a conversion efficiency of 40%, as above, the yield increases.

6.2.1 Synthesis gas

The main parameters that affect the yield in the gasifier and how much of the energy in the black liquor that is to be found in the synthesis gas, are the temperature and pressure in the gasifier and the dry solids content of the black liquor.

A lower gasifier temperature increases the amount of energy found in the synthesis gas (higher cold gas efficiency). Higher dry solids content in the black liquor entering the gasifier have the same effect as lower temperature (higher cold gas efficiency). A higher gasifier pressure increases the amount of heat that can be recovered as steam, thus the imported fuel will be lower. In Figure 6.6 the cold gas efficiency is shown as a function of dry solids content and gasifier temperature.
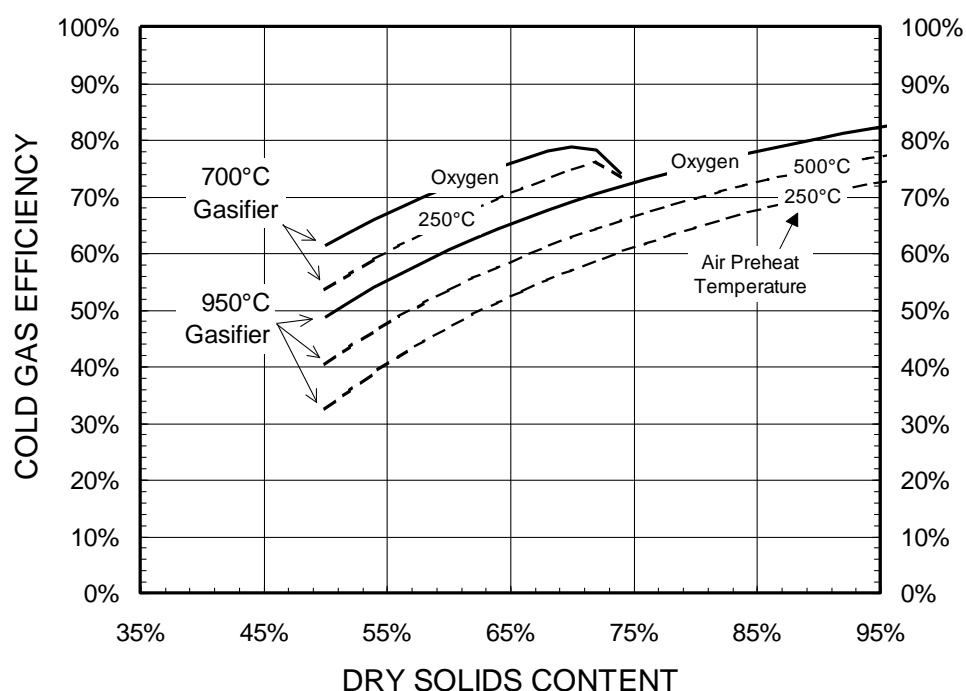
Figure 6.6. Cold gas efficiency dependency on gasifier temperature and black liquor dry solids (Berglin and Berntsson, 1999).

As an example, at a normal dry solids content of about 70% there is a ten percentage point difference in cold gas efficiency between a gasifier operating at 700 ^{o}C and a gasifier operating at 950 ^{o}C. Gasifier temperatures up to and above 1050 ^{o}C have also been used. There is thus a 5–15% efficiency and fuel yield potential in optimising the gasifier temperature [20].

The produced syngas contains methane, which is not a desirable product when producing synthetic fuels. Analysis performed on the syngas from black liquor gasification has shown that it contains low amounts of methane, 0.5 – 1%. With sodium sulphite based liquor the methane content drops to less than 0.5%. To lower it even more and increase the fuel yield in the DME production, the temperature in the gasifier must be increased. This would on the

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[20] The "normal" values pointed out here refer to conditions that have been tested in the pilot plant. The case described in Section 6.1 is based on 80% dry solids content and a gasifier temperature at 950 ^{o}C and is, thus, already assuming improvements in this matter.

other hand decrease the cold gas efficiency, and the net increase in fuel yield would be very small or even negative, depending on how the methane purged from the synthesis loop is used.

Maximising the dry solids content would increase the DME yield. Again, increasing the dry solids from 70% to the current maximum of about 85% would increase the cold gas efficiency by about ten percentage points. However, a dry solids content of 85% is technically challenging since the black liquor still needs to be possible to atomize into fine droplets. The risk for process problems will be higher at very high solids concentrations.

6.2.2 Gas purification

Many scientists and reports claim that the main challenges with gasification of biomass and conversion to methanol (or any other syngas based biofuel) is gas cleaning and conditioning of the syngas, scale up and process integration (*Faaij, 2006*). It is also reported that very large scale production plants are necessary for economical reasons. Impurities in synthesis gas needs to be removed in order not to reduce the lifetime of downstream catalysts and optimize the synthesis gas composition.

There are several techniques to remove sulphur compounds from synthesis gas. The most used are Rectisol, Selexol and amine gas treating processes. These processes use physical or chemical absorption to remove the impurities. Physical or chemical adsorption on fixed beds can also be used. Activated carbon and zeolites are common physical adsorbents used in gas clean up processes. Large organic molecules are easily adsorbed on activated carbon whereas smaller molecules are adsorbed to lesser extent. Activated carbon is used for non polar compounds and zeolites can be used for polar compounds. Regeneration of the physical adsorbents can be carried out thermally or by changing the pressure. ZnO is used as chemical adsorbent for adsorption of H_2S forming ZnS. The ZnS is very difficult to regenerate and is therefore used for Zn production or sent to waste. Chemical adsorption processes are thus mainly suitable for traces of sulphur.

Physical solvents are used mainly in high pressure applications to remove acid gas. At standard temperature and pressure, the solubility of H_2S is generally 10 times higher than the solubility of CO_2 in organic solvents, thus a selective H_2S removal is possible. This is of special importance for a Claus sulphur recovery unit downstream. Mercaptans, COS and C_2S are soluble in the organic solvents and can therefore also most often be removed together with the acid gas. Hydrocarbons with 3 carbons and higher (>ethane) are also removed by the organic solvents although large quantities make is less economical. Aromatics need a special design of the process since they are strongly absorbed in the organic solvent and tend to accumulate in the solvent if not treated appropriate even at trace amounts.

The Rectisol wash is a physical acid gas wash licensed by Lurgi and Linde. It uses methanol at low temperature as solvent. The process is used to selectively absorb gases such as H_2S, COS and CO_2 from the synthesis gas. The CO_2 can be withdrawn in a separate stream for further use. The methanol needs to be cooled down to about –40 to –50 °C and the pressure should be higher than 25 bars. The sulphur content after a Rectisol wash can be below 0.1 ppm which is necessary for the downstream catalysts. CO_2 with a very low concentration of sulphur (5 ppm) is also produced and this can be used for urea production, beverage industry or even fuel (methanol/DME/gasoline) synthesis if large amounts of H_2 can be provided. It is also a usable source of CO_2 for CCS. Rectisol removes not only sulphur and carbon dioxide. The process is also used to remove trace components such as tars, cyanide, ammonia, mercury,

all sulphur types and metal carbonyls. No further clean up is needed after a Rectisol unit. The possibility for the Rectisol wash to selectively perform 5 tasks in one unit (trace contaminants removal, deep desulphurization, bulk CO_2 removal, CO_2 purification and acid gas enrichment) makes it an attractive option. However, the process is also considered expensive, thus the use of it in small scale is limited. 75% of the synthesis gas worldwide produced from coal, oil and waste gasification is purified in Rectisol units.

The Selexol process developed over 30 years ago is licensed by UOP and is carried out at 20–70 bars. The Selexol solvent physically absorbs the acid gases and may be suitable for synthesis gas upgrading (only one reference) from gasification but is common for treatment of natural gas when the requirement on sulphur reduction is less stringent than for syngas. The Selexol process uses a mixture of dimethylethers of polyethylene glycol as solvent and is able to separate H_2S and CO_2 in separate streams. The Selexol process also removes COS, mercaptans, ammonia, HCN and metal carbonyls but need special precautions to do so. Its ability to remove traces of organics has not been demonstrated. For physical absorption the regeneration can be partially carried out by pressure reduction which requires less energy than regeneration for chemical solvents.

An amine solution is used to absorb the acid gases by chemical reaction, in contrast to Rectisol and Selexol which uses physical solvents. Amine gas treatment is used for CO_2 and H_2S removal in various processes in refineries, petrochemical industry etc. Chemical solvents may cause formation of heat-stable salts that plague the amine systems. Chemical solvents are thus favourable for low concentration acid gases and physical solvents are favourable at higher concentrations. The amine solution rich in sour gases is fed to a regenerator where it is separated into fresh amine solution and an acid rich gas. Monoethanolamine (MEA), Diethanolamine (DEA), Methyldiethanolamine (MDEA), Diisopropylamine (DIPA) and Diglycolamine (DGA) can all be used. Amine based systems are not found in syngas treatment services when the goal is to purify the syngas to ammonia and methanol synthesis quality demand.

The sulphur containing streams from the above processes can be fed to a Claus unit for elemental sulphur production. The Claus process was developed over 100 years ago. Some of the H_2S is first oxidized to sulphur dioxide by air and later converted to elemental and liquid sulphur by the following overall reaction

$$2\ H_2S + SO_2 \rightarrow 3S + 2\ H_2O. \hspace{3cm} (6.1)$$
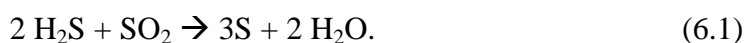
The reactions are carried out both without and with a catalyst. First and at high temperature the reaction proceeds without catalysts and secondly and at lower temperatures an alumina catalyst is used. This sulphur is later used for production of sulphuric acid, medicine, cosmetics, fertilizers and rubber products.

In summary, it is important to remember that the gas purification plant is important for the overall investment in a biomass to liquids plant. The gas purification plant can be a relatively large investment. Many different parameters will determine the actual purification plant set up. The present available purification techniques were not developed for cleanup of synthesis gas from a biomass gasifier. It may in fact be possible or even needed in certain cases to develop special technique for removal of impurities that are found in biomass derived synthesis gas that previously not were present in traditional synthesis gas.

6.2.3 Methanol synthesis – room for improvement despite almost 90 years of experience

Methanol (CH_3OH) has very good features as a future transport fuel in low-blend or as neat motor fuel. It may also be used as an energy carrier in fuel cells in the future. Today, nearly half of the methanol produced is used for formaldehyde production and other chemicals[21]. Today large quantities of methanol are blended into the gasoline pool in China. Methanol can also be used as a feedstock to produce gasoline (Stöcker, 1999) in a so called MTG (methanol to gasoline) process. In this Section we present challenges and improvement potentials for both methanol and DME production from biomass-based synthesis gas.

In 1923 the chemical company BASF engineers converted a mixture of CO and H_2 called synthesis gas (or syngas) catalytically into methanol. Previously, methanol was obtained by wood distillation which is an ineffective method. The BASF engineers developed the so-called "High pressure process". Here, the methanol synthesis is carried out at 250–350 bar and 320–450 °C. This process was dominating for about 45 years and a ZnO/Cr_2O_3 catalyst was used. This type of catalyst is poison resistant and can tolerate relatively high sulphur contents in the synthesis gas. However, there was need for a more active catalyst at lower temperatures and pressures and in the 1960s ICI developed the "low pressure process". The synthesis is carried out at 35–55 bar and at 200–300 °C. The catalyst is $Cu/ZnO/Al_2O_3$. However, this catalyst is more easily deactivated by sulphur and the syngas thus needs to be of higher quality. Methanol is produced from reaction of hydrogen and carbon oxides:

$$2H_2 + CO \Leftrightarrow CH_3OH \qquad\qquad (6.2)$$
$$3H_2 + CO_2 \Leftrightarrow CH_3OH + H_2O \qquad\qquad (6.3)$$
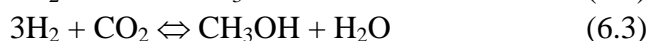
However, in the reactor where the synthesis gas is converted into methanol the water gas shift reaction (see below) also occurs to some extent depending on operating conditions and choice of catalyst.

$$CO_2 + H_2 \Leftrightarrow CO + H_2O \qquad\qquad (6.4)$$

Methanol synthesis at lower temperature and pressure use less energy in e.g. a compressor, and a low pressure process has therefore potential for a lower cost. On the other hand, synthesis gas conversion into methanol is favoured by higher pressure and only a low fraction of the gas is therefore converted in each pass due to the restrictions in the methanol reaction equilibrium. It is therefore common to recycle the unreacted synthesis gas. This is carried out in combination with a small purge to remove impurities that otherwise would accumulate over time. The use of selective membranes, such as zeolite membranes, could open up the possibility for other designs. In a membrane reactor configuration, instead of recycle the unreacted gas, certain gas compounds, e.g. methanol, could be removed from the gas leaving the methanol reactor and thereby increase the conversion per pass. This is an area of research that is currently ongoing at Luleå University of Technology.

A modern methanol catalyst has a lifetime of a few years depending on type and manufacturer. The catalyst become less active due to poisoning, fouling or thermal sintering over time and needs to be replaced finally. Sulphur, chlorine and metal carbonyls are the most common poisons for the methanol catalyst. The most common deactivation cause is thermal sintering.

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[21] World production: 38% formaldehyde, 20% methyl tertiary butyl ether, 11% acetic acid and 31% various chemicals/solvents or fuel additives (Olah et al., 2006).

The methanol reaction is highly exothermic which means that the reactors and catalysts must be properly designed to avoid higher temperatures caused by the exothermic reaction itself.

Although the synthesis of methanol[22] has been known for soon 100 years there is still research and developments ongoing in the design of methanol catalysts and reactor configurations. Several catalyst manufacturers have put a lot of effort to add certain promoters to the catalyst to hinder sintering and increase the specific surface area of the catalyst. In for example Synetix catalyst 51-7, MgO is used in the catalyst to hinder sintering. Some reports say that high CO_2 levels in the synthesis gas can be harmful for the catalyst. This could also be a result from H_2O formation as a by-product in the methanol reaction from H_2 and CO_2. Gasification of biomass normally results in high CO_2 concentrations, compared to coal gasification or by reforming/partial oxidation of natural gas. Development of a catalyst suitable for the synthesis gas composition (H_2/CO/CO_2) from biomass gasification would thus be of interest.

The synthesis can be carried out both in gas and liquid phase (packed bed and slurry reactor[23]), and many different reactor configurations exist (Lange et al., 2001; Tijm et al., 2001). Optimum gas phase methanol synthesis is carried out at a stoichiometric number (H_2–CO_2) / (CO+CO_2) = 2. However, gas phase (packed bed reactor) normally occur at H_2/CO ratios up to 5 (thus hydrogen excess) whereas in liquid phase (slurry reactor) a lower H_2/CO ratio is used, from 1 to 2 (Tijm et al., 2001). An advantage of using liquid phase, or slurry reactors, is better heat removal and that lower H_2/CO ratios are tolerable. The produced methanol needs to be separated from other compounds by e.g. distillation.

Several researchers throughout the world have during the last 10–20 years been more and more interested in catalytic distillation with structured catalysts. In catalytic distillation the catalytic conversion and distillation is carried out in the same unit, which could improve the overall energy efficiency and reduce the capital cost significantly.

In summary, despite the fact that methanol has been produced from synthesis gas for almost 100 years there is a lot of investment in catalyst and reactor design ongoing.

Below a few areas where improvement is desired is listed:
- Catalyst design
    - Poison resistant
    - High surface area
    - Temperature resistant (avoid sintering)
    - CO_2 and H_2O tolerant
    - Increase lifetime
- Reactor design to improve the conversion per pass
    - Catalytic distillation
    - Membrane reactors
    - Liquid phase reactors
- Low pressure processes
    - Reduce cost for e.g. compression

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[22] It is worth noticing that methanol production is carried out in the same way, no matter if the synthesis gas is derived from the gasification of coal or biomass. Coal to methanol has been demonstrated in Germany in the 80s and 90s and about 10 coal to methanol plants have been built in China since 2000.

[23] Curently only one process in the world uses the slurry option. This is a demonstration plant in Kingsport, USA, partly financed by DOE.

6.2.4 DME synthesis – new potential as a worldwide chemical

Dimethylether, DME (CH_3OCH_3), a sweet ether-like odour gas can be prepared either by dehydration of methanol or directly from synthesis gas (Ogawa et al., 2003; Voss et al., 1996; Semelsberger et al., 2006). The following overall reactions may occur during DME synthesis from synthesis gas[24]. Methanol is formed as an intermediate product in these reactions according to reactions (6.2–6.4).

$$3\ CO + 3\ H_2 \Leftrightarrow CH_3OCH_3 + CO_2 \qquad (6.5)$$

$$2\ CO + 4\ H_2 \Leftrightarrow CH_3OCH_3 + H_2O \qquad (6.6)$$
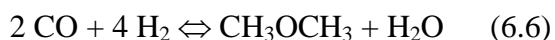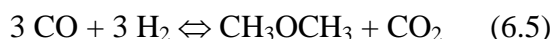
The overall reaction is strongly exothermic. The reactor and catalyst thus needs advanced reactor configurations to achieve proper cooling in order to avoid catalyst deactivation and other problems. Cu/ZnO catalyst can be used for MeOH synthesis and an acid catalyst such as gamma alumina or zeolite is used for the dehydration reaction. Depending on the engineering technology and catalyst a H_2/CO ratio from 1 to 2 is optimum. Coal gasification generates a syngas with H_2/CO = 0.5–1 and catalysts and reactor configuration that operates with reaction (6.5) is thus best choice, whereas syngas with a higher H_2 content a configuration with reaction (6.6) should be targeted.

Direct synthesis as it is called, although it occurs via methanol as intermediate, of DME from syngas can be done in fixed bed or in slurry rectors. JFE Holdings of Japan uses a liquid phase slurry reactor in which reaction (6.5) is achieved. A bifunctional (mixed) catalyst is used to achieve methanol synthesis and dehydration to DME. The operating temperature for the slurry reactor is 240–280 °C at 30–70 bar (Ogawa et al., 2003) and H_2/CO = ~ 1.0. JFE Holdings have also tested DME synthesis in demonstration scale where they produced 100 ton per day of DME (*JFE*). The methanol synthesis reaction is strongly controlled by equilibrium. However, in the case for direct DME synthesis the syngas conversion increases dramatically due to consumption of methanol that forms DME.

Haldor Topsoe A/S of Denmark synthesizes DME directly from syngas by reaction (6.6) in fixed beds (Ogawa et al. 2003; Haldor Topsoe). In the technology used by for example Haldor Topsoe methanol is first synthesized from syngas in a cooled fixed bed reactor at optimum methanol synthesis conditions. Secondly, the dehydrogenation of methanol, which is less exothermic, takes place in a second adiabatic fixed bed reactor at a different temperature and loaded with a dual function catalyst (Haldor Topsoe). The dual-function of the catalyst makes it interesting. The catalyst is active for both the production of methanol from synthesis gas and the formation of DME from the intermediate methanol. Simultaneously, the dual-function catalyst equilibrates the shift reaction, which according to Topsoe makes it a "truly flexible catalyst".

The ability to equilibrate the shift reaction makes it versatile with respect to the possibility to change the H_2 and CO_2 concentrations in the synthesis gas. Thereby both reactions can occur at optimum conditions as opposed to reactions in single reactors. This configuration used by Topsoe does not require an expensive and space consuming slurry to handle the catalyst and

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[24] It is, however, worth noticing that reaction 6.5 is used in a so called direct DME process which has been demonstrated in Japan. The absolutely dominating process when producing DME is through methanol as in reaction 6.2-6.4.

dissipate the reaction heat. An additional advantage is that large scale production can be produced in a single-line layout. On the other hand, water is not accumulated (reacts with CO for the slurry configuration) in the slurry configuration which otherwise may lead to catalyst deactivation and it also requires less energy to separate CO_2 from DME in a slurry reactor than DME/H_2O in fixed bed reactors (Ogawa et al., 2003). The slurry reactor has an advantage of easy temperature control whereas the fixed bed for example needs to be divided into multistages to remove the generated heat.

6.3 Improvement potential: By-products and CCS

Installing a black liquor gasifier has some positive effects on the pulp mill. The extracted hydrogen sulphide increases the flexibility of sulphur usage. For example, it is easier to divide the sulphur between different white liquor streams for modified cooking. It is also easy to produce elemental sulphur from the H_2S gas if the plant is integrated with a Claus unit[25]. Sulphur can then be mixed with cooking liquors to produce polysulfide. Polysulfide improves yield by retaining more hemicellulose. Pulp rich in hemicellulose sometimes exhibit lower tear strength than conventional kraft pulps.

It deserves to be pointed out that the main "by-product" is the green liquor itself. From the pulp mill's point-of-view the green liquor is really the main product from the process and any large variation in the quality of the green liquor or in the availability of the black liquor gasifier would drastically affect the economics of the concept.

6.3.1 Methane

The produced syngas contains methane that is not converted in the synthetic fuel process and needs to be purged. By installing a gas burner in the power boiler, the methane can be fired to produce steam and electricity, but methane could in principle be a sellable by-product as well, as shown in Section 5.

6.3.2 Carbon Capture and Storage (CCS)

In the BLGMF/DME plant CO_2 is removed as part of the process, both after the gasifier and after the water gas shift. The two removed CO_2 streams are however combined and emitted as one stream. Alternatively the water gas shift can be put first and then the CO_2 will be removed in one place. Table 6.3 presents the net usage of biomass and electricity, together with DME production and captured CO_2, for the BLGMF/DME plant with CCS (see Wetterlund et al., 2009b) in comparison to the BLGMF/DME case from Ekbom et al. (2003). In the CCS case it is assumed that all separated CO_2 can be captured and stored[26]. The amount of CO_2 possible to capture is calculated based on process stream data from Ekbom et al. (2003). The captured CO_2 is assumed to be compressed to 80 bar and transported by pipeline for geological storage.

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[25] The Claus process uses a combination of thermal and catalytic treatment to convert hydrogen sulphide into elemental sulphur in the overall reaction: $2 H_2S + O_2 \rightarrow S_2 + 2 H_2O$.

[26] It should be noted that CO_2 could also be captured from the flue gases in the mill as well as from the combustion of DME/methanol (see e.g Hektor and Berntsson, 2007). These options are not taken into account in Table 6.3.

Table 6.3. Net usage of biomass and electricity, together with the DME production and captured CO_2, for the BLGMF/DME plant with CCS in comparison to the BLGMF/DME case from Ekbom et al. (2003).

		BLGMF/DME no CCS	BLGMF/DME CCS
Bark/other wood fuels	[MW]	157	157
Electricity	[MW]	101	111
DME production	[MW]	275	275
Captured CO_2	[kg/h]	0	87,500

Unsurprisingly, the case with CCS show a considerably larger potential for CO_2 reduction than the case without, since the sequestrable amount of CO_2 is high. Jönsson et al. (2010) show that inclusion of CCS can significantly improve both the economic performance and the CO_2 emissions effect of a BLGMF/DME and BLGCC plant, since CO_2 could be separated at a low cost (both in terms of economy and energy).

Also note that methanol can be produced from CO_2 and H_2, see section 6.2.3. Thus, if the plant would be located close to a hydrogen source it would also be possible to use the CO_2 captured and react it with hydrogen to produce methanol/DME. The hydrogen could potentially come from some other process (steel plant etc) or from electrolysis of water.

6.4 Important systems analysis issues: Process integration

The environmental performance of black liquor gasification is not only dependent on the technical characteristics of the gasification process and assumptions about systems surrounding the mill. Other parameters such as characteristics of the host mill and level of process integration also affect the performance of black liquor gasification. Sweden is at the forefront of knowledge regarding this type of systems analysis for black liquor gasification processes; hence we have chosen to devote significant space in this report to illustrate some of the results that researchers have generated in this area.

In Pettersson and Harvey (2010b) the effect of the mill steam demand on the net biomass and electricity usage for a BLGMF/DME plant is illustrated. Different mill steam demand levels are considered; ranging from approximately 7 GJ/ADt (representative of a future highly energy-efficient market pulp mill with a low steam demand) to 19 GJ/ADt (representative of a host mill with relatively high steam demand, e.g. future integrated pulp and paper mill). See Section 6.4.1 for a more detailed description.

For most future mills the steam surplus from a BLGMF plant will not be sufficient to cover the total mill steam demand and it will be necessary to e.g. have a biomass fired CHP plant to cover the rest of the mill steam demand. Pettersson and Harvey (2009) shows that it is economically profitable to consider increased heat integration when a BLGMF/DME plant is integrated to a pulp mill. A reduced steam demand leads to a decreased need for external biomass but increased need for external electricity. However, the results show that the CHP plant should not be over-dimensioned, i.e. despite the increased need for external electricity resulting from the steam saving measures they are still profitable even when the electricity prices are high. See Section 6.4.2 for a more detailed description.

It should be noted that in these studies a steam deficit was handled by firing bark (and other wood fuels) in a boiler (connected to a steam turbine) and a steam surplus was used for additional electricity production in a condensing steam turbine. Another way to handle a

steam deficit is for example integration of solid biomass gasification[27] connected to production of electricity or biofuels and steam (see e.g. Larson et al., 2009). Another way to utilize a steam surplus is to extract lignin from the black liquor to use as a fuel or as a feedstock for production of materials or chemicals. But Jönsson et al. (2010) illustrate the importance of not only comparing black liquor gasification with investment options based on conventional technologies, but also other emerging technologies such as lignin extraction. An important factor is also the recovery boiler steam data (see e.g. Berglin et al., 1999)

6.4.1 Effect of mill steam demand

In order to illustrate the effect of the mill steam demand on the net biomass and electricity usage of a BLGMF/DME plant different mill steam demand levels are considered; ranging from approximately 7 GJ/ADt (representative of a future highly energy-efficient market pulp mill with a low steam demand[28]) to 19 GJ/ADt (representative of a host mill with relatively high steam demand, e.g. future integrated pulp and paper mill).

The mill considered produces 2000 ADt/day of bleached kraft pulp from softwood. Table 6.4 presents some key data for the mill.

Table 6.4. Key mill data.

Pulp production	[ADt/d]	2000
Black liquor	[MW]	487
HP[1] steam data	[bar/°C]	81/490
MP[2] steam data	[bar/°C]	10/200
LP[3] steam data	[bar/°C]	4.5/150

[1] High pressure
[2] Medium pressure
[3] Low pressure

The mill is assumed to be considering replacement of the recovery boiler (and steam turbine/s). Two different options are to be considered: (1) invest in a new recovery boiler (the reference case) or (2) invest in a BLGMF (black liquor gasification with motor fuel production) plant producing DME (the BLGMF case). Figure 6.7 illustrates the main energy and material streams for the reference case with a recovery boiler.

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[27] It might, however, take several of years until pulp mill owners would feel comfortable with installing two gasification technologies.
[28] A market kraft pulp mill based on best available Scandinavian technology has a steam use of 11 GJ/ADt (KAM, 2003; Delin et al., 2005b). With state-of-the-art equipment and a higher degree of heat integration, it is possible to substantially further decrease the steam use (see e.g. KAM, 2003; Algehed, 2002).
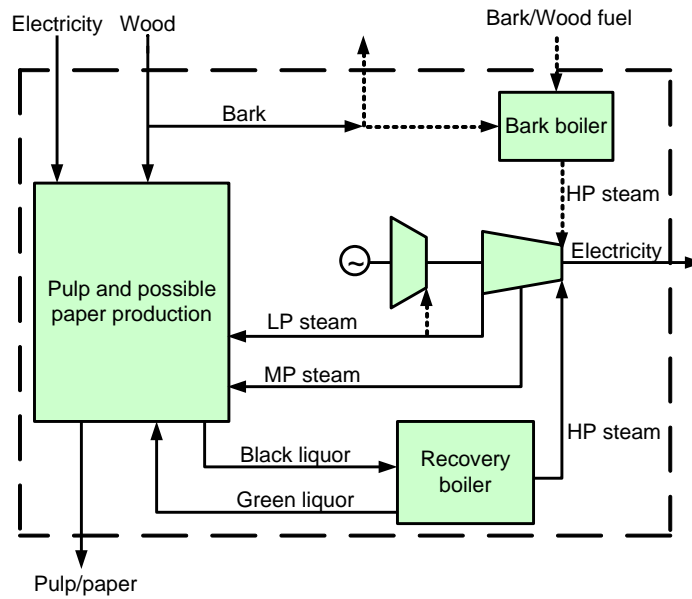
Figure 6.7. Main energy and material streams for the reference case with a recovery boiler. Solid lines represent flows that are relevant for all steam demand levels whereas dotted lines represent possible flows. Acronyms used in the figure are: HP= high pressure, MP= medium pressure, LP= low pressure.

The produced pulp is either dried and transported to a paper mill or refined at the mill. Here both cases are addressed by considering different mill steam demand levels. Whether the final product is pulp or paper is not relevant since this is not affected by what kind of energy and chemical recovery system that is considered and since black liquor gasification is evaluated in comparison to a recovery boiler based system, the pulp/paper production stream will be cancelled out.

Figure 6.8 illustrates the main energy and material streams in the BLGMF case. Excess heat (at suitable temperature levels) in the BLGMF plant is used to produce steam. Some steam is used internally at the BLGMF plant, but there remains a large steam surplus that can be used at the mill. However, it should be noted that less steam is produced compared to the recovery boiler case since DME is produced in the BLGMF case. Purge gas from the BLGMF plant is fired in the bark boiler. No condensing steam turbine is included in Figure 6.8, since the mill steam demand has to be extremely low in order for a steam surplus to exist in this case. Export of falling bark is not indicated for the same reason.
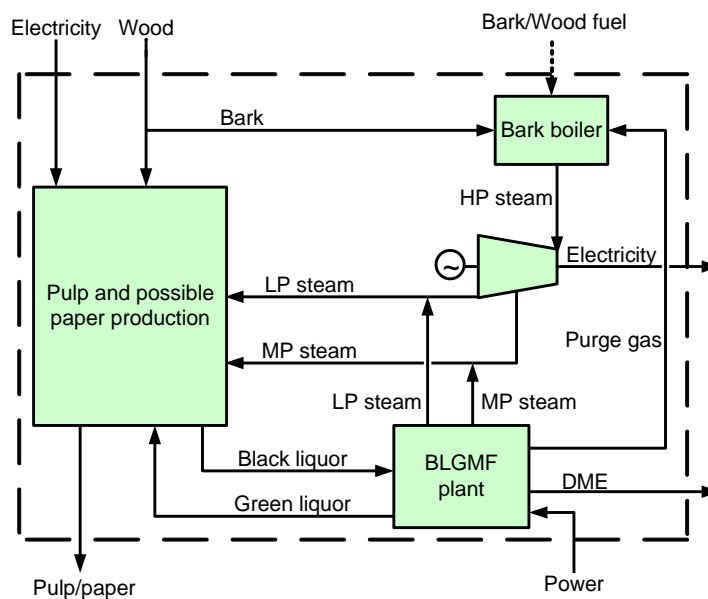
Figure 6.8. Main energy and material streams in the case of black liquor gasification. Solid lines represent flows that are relevant for all steam demand levels whereas dotted lines represent possible flows. Acronyms used in the figure are: HP= high pressure, MP= medium pressure, LP= low pressure.

A detailed presentation of the calculated energy balances, from the two cases, is found in Appendix 9. Results of interest from the two cases' energy balance calculations are e.g., that at 7 GJ/ADt the kraft pulp mill integrated with a BLGMF/DME plant can satisfy its steam demand using only internal fuel (no need to purchase any external fuel), see Table A9.2). At 15 GJ/ADt the mill reference case has a neutral steam balance (i.e. the steam produced by the recovery boiler is equal to the mill steam demand, see Table A9.1). At 16 GJ/ADt the mill reference case can satisfy its steam demand using internal fuel only (see Table A9.1).

By comparing the mass and energy balances derived for the mill reference case (see Table A9.1) with the mass and energy balances for the mill with a BLGMF/DME plant (see Table A9.2), the net usage of biomass (bark or other wood fuels) and electricity can be determined. Table 6.5 presents the net usage of biomass and electricity, together with the DME production, for the BLGMF/DME case considering the five different mill steam demand levels.

Table 6.5. Net usage of biomass and electricity, together with the DME production, for the BLGMF case.

Mill steam energy use[1]	[GJ/ADt]	7	11	15	16	19
Bark/other wood fuels[2]	[MW]	32	170	310	309	309
Electricity	[MW]	154	109	64	64	64
DME production	[MW]	275	275	275	275	275

[1] In mill production process (excluding steam energy conversion to electricity in turbine).
[2] LHV

As can be seen from Table 6.5 the net usage of biomass and electricity, for production of a certain amount of DME, vary significantly depending on the mill steam use. The balance for 11 GJ/ADt is rather close to the one in Ekbom et al. (2003), since the steam usage of the KAM2 mill considered in that study is close to 11 GJ/ADt.

In Pettersson and Harvey (2010b), the resulting mass and energy balances are used to investigate the net annual profit for DME production via BLG under different future energy market scenarios. The results show that the net annual profit from DME production via BLG is strongly dependent on future energy market conditions and the steam demand level of the

mill with which the BLGMF plant is integrated. In Pettersson and Harvey (2010a), CO_2 emission balances for different black liquor gasification cases, including DME, were investigated considering different types of mills with different steam demands. The results show e.g. a significant difference in CO_2 emission effect from DME production via black liquor gasification depending on the host mill's steam demand[29].

6.4.2 Effect of increased heat integration

From the results presented in the previous section, it can be seen that even for mills with a very low steam use it would be necessary to have additional steam production in e.g. a biomass fired CHP plant when a BLGMF/DME plant is introduced to a mill. The possibilities for increased heat integration and other type of steam saving measures could be considered, thereby decreasing the need for additional steam production. It could be increased heat integration within the mill processes, within the BLGMF processes and between the mill and BLGMF processes. Further, more energy efficient equipment could be used. The possibilities for this will of course vary depending on several factors, above all how well integrated the processes are and what type of equipment that is considered. If, for example, the steam demand at a mill is 7 GJ/ADt, one can presume that it is very well integrated and that it uses highly energy efficient equipment. In this case focus could be on steam saving measures in the BLGMF processes, if possible, and/or optimizing the integration between the mill and BLGMF processes. A mill with a steam demand of 15 GJ/ADt could either be rather energy efficient, if it is an integrated pulp and paper mill, or rather energy inefficient, if it is a market pulp mill (see previous section). Thus, the potential for steam saving measures could vary significantly for mills with the same steam use.

If a steam deficit is covered by a biomass fired CHP plant, a reduction of the steam use will result in a decreased need for biomass but an increased need for electricity since the internal electricity generation is decreased. The consequences, both economic and environmental, of implementing steam saving measures will then be determined by how biomass and electricity are valued.

Pettersson and Harvey (2009) investigated the effect of increased heat integration when a BLGMF/DME plant is introduced to a kraft market pulp mill. The study is based on the study by Ekbom et al. (2003). High temperature excess heat in the BLG plant is used to produce steam, partly used internally at the BLG plant, partly used at the mill. Additional steam for the mill is produced in a biomass fired CHP plant. Low temperature excess heat from the BLG plant is used in the mill's secondary heat system, together with low temperature excess heat from the mill. In Pettersson and Harvey (2009), the secondary heat system is redesigned in order to make more heat available at sufficient temperature levels to replace steam in the evaporation plant of the mill. As a result of lower mill steam demand, the size of the CHP plant is reduced and consequently the biomass use decreases. At the same time, however, the need for external electricity increases.

Table 6.6 shows the mass and energy balances for the KAM2 mill with BLGMF with increased heat integration in comparison with the BLGMF base case (from Ekbom et al., 2003) and the mill reference case with a recovery boiler. The use of wood fuels in the CHP

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[29] Swedish black liquor gasification industry, however, declare that it is fundamental that all energy, needed to compensate for the energy losses when introducing the BLGMF concept, is renewable. It is a central argument that the DME/methanol is a fully renewable fuel.

plant is decreased by 77 MW, whereas the electricity production is decreased by 18 MW. This corresponds to a decrease of the external need for biomass by 62% and an increase of the external need for electricity by 35%.

Table 6.6. Mass and energy balances for the KAM2 mill with BLGMF/DME with increased heat integration in comparison to the BLGMF/DME base case and the mill reference case with a recovery boiler (RB).

		RB reference case	BLGMF/DME base case	BLGMF/DME with increased heat integration
Black liquor	MW	487	487	487
DME	**MW**	-	**275**	**275**
Electricity				
Production	MW	104	43	25
Consumption	MW	59	99	99
Surplus/Deficit	MW	45	-55	-74
Incremental use	**MW**	-	**101**	**119**
Bark/other wood fuels				
Mill excess	MW	32	18[1]	18[1]
Consumption in CHP plant	MW	-	143	66
Surplus/Deficit	MW	32	-125	-48
Incremental use	**MW**	-	**157**	**80**

[1]: Larger bark use in the lime kiln due to different green liquor composition in the BLG case compared to the recovery boiler case.

The derived mass and energy balances are used to investigate the effect on the DME production cost. The results show that the steam saving measures are profitable, even in case of a high electricity price in relation to biomass price. See Pettersson and Harvey (2009) for further details about the integration and the economic calculations.

6.5 Conclusions and discussion

Modern chemical pulp mills, having a developed infrastructure for handling large amounts of biomass and having a potential surplus of energy, could become important suppliers of renewable fuels for transport. If black liquor is gasified and the pulp mill is compensated for the energy loss by a (larger) biomass boiler for steam and electricity production, motor fuels are indirectly produced from low-cost solid biomass. Using black liquor as a raw material for DME or methanol production would have many advantages compared to other biofuel production options, such as that black liquor is already partially processed and exists in a pumpable, liquid form, and that the process is pressurised, which enhances fuel production efficiency. However, to achieve commercial status, of the production of DME or methanol from a syngas via gasification of black liquor, some challenges still remain. The key ones to overcome is (i) the risk which connected to scale up to industrial size of key equipment and then (ii) in large scale show that the plant can operate according to calculated heat and material balances and (iii) achieve high availability.

A general conclusion from this section is that there is potential of improvements in the entire process. We have presented how the DME/methanol yield can be improved, how material and energy can be saved and how CO_2 emissions can be reduced. However, since the processes are complex most of the improvement potentials presented also come with technical or economical dilemmas.

One example is that the fuel yield depends on the gasifier temperature. Lower gasifier temperature increases the energy content of the synthesis gas (leading to an increased DME

yield). On the other hand, higher gasifier temperature leads to that less methane is produced in the synthesis gas which also would benefit the DME yield. In reality (in the Chemrec process) the gasifier temperature is kept as low as possible to get good carbon conversion and green liquor of high quality resulting in no control of the methane content[30]. The yield of the synthesis gas also depends on the dry solids content of the black liquor. Increasing the dry solids content would benefit the DME yield, but high dry solids content, at about 85%, is technically challenging since the black liquor still needs to be fluid. The risk of process problems will increase.

Another example is that the heat recovery depends on the process pressure. Higher gasifier pressure increases the amount of heat that can be recovered as steam leading to that less imported fuel is needed. Almost no heat is recovered as steam when the process pressure is below 10 bar, while at 30 bar approximately 80% can be recovered as steam. It is, however, technically challenging to increasing the pressure[31].

Materials in general in the entire process might have potential for improvement. Some steps in the gasification and synthesis processes are initially not developed for the gasification of black liquor or biomass but for the gasification of fossil fuels. It might be possible to find improvement potentials if developing special techniques for removal of impurities that are found in biomass derived synthesis gas that were not identified in traditional synthesis gas. It should, however, be noted that suppliers of gas purification technologies do not see any problems with the biomass-based synthesis gas from the Chemrec process[32].

Improvements leading to increased environmental performance of black liquor gasification are not only dependent on technical characteristics of the gasification process. Other parameters such as characteristics of the host mill and level of process integration also affect the performance of black liquor gasification.

Previous studies indicate generally both a better economic performance and reduced global CO_2 emissions for black liquor gasification in future market pulp mills compared to future integrated pulp and paper mills if the technology is compared with reference investment options based on conventional technologies. It would of course also be interesting to compare black liquor gasification with other emerging technologies such as lignin extraction from the black liquor for use as a feedstock for production of materials or chemicals. Increased heat integration, thereby decreasing the need for external biomass but increasing the need for external electricity, is profitable for a BLGMF/DME plant even when the electricity price is high in relation to the biomass price.

Although two black liquor gasification processes have reached commercial or near-commercial status, it is difficult to predict if, or when, a full scale black liquor gasifier with motor fuel production will replace a conventional recovery boiler. Some challenges remain but in general the technology has been proved working. However, so far the pulp and paper industry has only shown a moderate interest. The investment cost for the first full scale plant is high, and it is hard to motivate someone to invest in the first plant.

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[30] The methane content is actually used as an indicator that the gasification process works fine.
[31] The reason for not increasing the pressure is in principle that materials are not tested for that.
[32] Chemrec has measured their biomass-based synthesis gas down to the level of ppb. The suppliers of Rectisol (Linde and Lurgi) have not found see any unwanted components.

7. Overall discussion and conclusions

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In this study we have described the WTT method and discussed that results depend on assumptions made. We have focused on three biofuel production technology options, which currently are in demonstration phase (cellulose based ethanol, methane from gasification of solid wood as well as DME from gasification of black liquor), to identify research and development potentials that may result in improvements in the WTT values. We have also discussed improvement potentials for the agriculture and forestry part of the WTT chain.

7.1 Difficulties when interpreting improvement potentials in the context of WTT

Although quantitative improvement potentials are given in the three biofuel production cases, presented in Section 4-6, it is not obvious how these potentials would affect WTT values. Difficulties are first and foremost related to that the biofuel production processes are complex as well as that there is no one alone standard method for WTT analysis.

As an illustration of how difficult it can be to calculate new WTT values from identified improvement potentials it is in Section 6 found that there is a 5–15% efficiency and fuel yield improvement potential by optimising the gasifier temperature. Since there is no linear relation between the gasifier temperature and the overall CO_2 emissions from the process (i.e., the process is complex and changing one parameter impacts other parameters) we cannot just assume that the CO_2 emissions from the process also decrease by 5-15%.

It is further challenging how to combine the different improvement potentials, since improvements in one area of the process might increase or decrease the improvement potentials in other areas of the process.

Moreover, the improvement potentials depend on which base case you compare with. For example, the WTT-values presented in Edwards et al. (2007) are generally based on best available technology and thus already assume some of the improvement potentials discussed in this study.

To be able to compare how the improvement potentials affect the WTT-values, completely new WTT-values, taken the entire process into account, needs to be calculated which lies outside the scope of this study.

7.2 Common conclusions

From the entire study we have come to agree on the following common conclusions:
- There is a lot of research and development going on in Sweden within (and Swedish biofuels actors show a great interest in) the three studied second generation biofuel production technologies.
- In general, recently developed technology processes, within the three cases, work well at pilot and demonstration scale and are now in a phase to be proven in large scale before the fuel production can achieve commercial status.
- There is still room for improvement although some processes have been known for decades.

- The biofuel production processes are complex and detailed improvements need to be seen and judged from a wider systems perspective (both within the production plant as well as in the entire well-to-tank perspective).
- Enhanced environmental performance can be achieved from energy integration, both within the process as well as from integration with other industries. Such solutions might be unique for each biofuel production plant.
- From the political ambitions (both within EU and Sweden) it is clear that the demand for renewable fuels will significantly increase during the coming decade. This will most likely result in opportunities for a range of parallel biofuel options.
- The studied biofuel options all represent second generation biofuels and all three can be part of the solution to meet the increased renewable fuel demand.
- The process of conducting this study is worth mentioning as a result itself, i.e. that many different actors within the field have proven their ability and willingness to contribute to a common report, and that the cooperation climate was very positive and bodes well for possible future collaboration within the framework of the f3 center.

7.3 Further work

Over the past years there has been a discussion whether biofuels should be viewed as an environmental threat or opportunity. From the situation where biofuels were considered to be one of several vital solutions to the climate problem, the view has now shifted into a more complex picture where the use of biofuels also imposes threats of different kinds. All kinds of renewable fuels, biofuels as well as renewable electricity and hydrogen can be produced in many different ways. To bring about a more varied discussion, as well as providing better basic data for various organizations decision-making, more knowledge needs to be developed and disseminated.

This study may serve as an example of research that can lead to such improved basic data for decision-making, here coming from refined knowledge about research and development challenges within the complex biofuels production.

An interesting idea for further work would be to calculate completely new WTT-values, for the three biofuels production technologies discussed in this study, to be able to compare how the identified improvement potentials affect the WTT-values.

Another idea for further work could be to expand the study to also include other renewable fuels. By analyzing a wide range of fuel options it would lead to a completely new WTT-study focusing on the effect from improvement potentials.

In order to to improve basic data for decision-making one might suggest a wide range of further work. Examples of questions, that would be interesting to further study in order to get improved knowledge within the subject of renewable fuels, could be:
- How can current barriers and technology challenges be reduced?
- How can identified improvement potentials be implemented at lowest cost?
- What parts of research and developments are Sweden currently focusing on? Can any synergy effects be identified (between refineries, petro chemical industry, agriculture and forestry, energy companies, renewable fuel industry etc)?
- Under what circumstances could Swedish biofuels become a large export product? Is that a cost-effective scenario?

- Can an increased use of biofuels be motivated from an energy security perspective? Is that a cost-effective scenario?
- Under what circumstances is it a cost-effective solution to use biofuels for the reduction of greenhouse gas emission? Is the result sensitive to different kinds of biofuels? To different kinds of biofuel production systems e.g., the use of by-products and level of integration? To different time perspectives? To different future transportation demands? To different goegraphic perspectives?
- What can we learn from increased communication between research groups, industry and authorites?

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Appendix 1. Advantages and disadvantages of some fuels

To get an overview of the different renewable fuel options and their performance, we have listed some fuel options' advantages and disadvantages in Table A1.1.

Table A1.1. Summary of advantages and disadvantages for some renewable fuel options inclusive hydrogen and electricity.

	Advantages	Disadvantages
Ethanol	• Can be blended in conventional gasoline. • When used as low blend in gasoline there is no need of new vehicles or infrastructure. • Well-tried as vehicle fuel • High octane rating, can replace other chemical additives used for increasing octane in gasoline.	• Lower energy content per volume unit, compared to gasoline. Larger fuel storage tanks are needed for keeping the same driving distance as gasoline. • Somewhat corrosive, can impact various metals like aluminum, plastics and rubber. • Pure ethanol and high blends exhibit poor cold start performance. An engine heater might be needed. • Large scale ethanol production, based on agricul-ture crops, requires considerable use of arable area.
Biodiesel FAME	• Can be blended in conventional diesel. • When used as low blend in gasoline there is no need of new vehicles or infrastructure. • Almost the same energy content per volume unit as conventional diesel. • It has a higher cetane and lubricity rating than conventional diesel, which improves engine efficiency.	• At low temperatures, diesel fuel forms wax crystals, which can clog fuel lines and filters in a vehicle's fuel system. • Somewhat corrosive, can impact various metals, plastics and rubber. • Switching to biodiesel on a large scale requires considerable use of arable area.
Biogas and bio-methane via gasi-fication	• Can be blended in fossil natural gas. • Combustion give raise to significant lower emissions of NOx, CO and soot compared to diesel. • Anearobic digestion of manure, sludge and waste can be seen as a benefit to society.	• Need to be upgraded (increase the CH_4 concentration) to be able to be used as fuel for transport. • Need for new infrastructure. • Large and bulky fuel storage tank. • Gas engines have in general lower performance compared to diesel engines, especially the torque. • Methane leakages can lead to failure of climate benefits (CH_4 is a 23 times stronger GHG compared to CO_2).
Methanol	• Can be blended in conventional gasoline. • When used as low blend in gasoline there is no need of new vehicles or infrastructure. • Well-tried as vehicle fuel (used in motor sports) • High octane rating, can replace other chemical additives used for increasing octane in gasoline.	• Lower energy content per volume unit (half compared to gasoline). Larger fuel storage tanks are needed for keeping the same driving distance. • Corrosive, can impact metals, plastics and rubber. • Strongly toxic and water soluble. Fuel spillage can destroy ground water. • Low acceptance from vehicle industry.
DME	• Combustion give raise to significant lower emissions of NOx, CO and soot compared to diesel. • No need for particle filters (as diesel engines often have for exhaust emission control). • Has the potential to reduce engine noise.	• Need for new infrastructure. • Lower energy content per volume unit compared to diesel.
Fischer-Tropsch Diesel	• Can be blended in conventional diesel. • No need for new vehicles or infrastructure. • Same energy content per volume unit as conventional diesel.	• The FT synthesis generates carbon chains of different lengths (as in a conventional fossil refinery). That is also very long carbon chains which are difficult to use as fuels for transport.
Hydrogen	• Can be used in internal combustion engines but are more efficient used in fuel cells. • Local emissions are only water vapor when hydrogen is used in fuel cells. • Approximately 10% hydrogen can be blended in natural gas without changing equipment • Flexible feedstock, e.g. biomass, natural gas coal (with or without CCS). H_2 can also be produced via electrolysis of water.	• CO_2-neutral H2 is currently expensive. • Need for new infrastructure. • Storage technologies are expensive, bulky and technicallly challenging. • Large and bulky fuel storage tank. • Hydrogen vehicles currently very expensive.
Electri-city	• Significantly more energy efficient powertrain compared to internal combustion engines. • Flexible feedstock • Few mobile parts. Electric vehicles have the potential for reduced service need. • Quiet engine.	• Current batteries are very expensive (need improvements in e.g. capacity, safety, and lifetime), • Electric vehicles have lower driving range compared to internal combustion engines. • Less useful for long-distance heavy vehicles, ships and aviation.

Appendix 2. General WTW methodological choices

A deepened discussion on General WTT methodological choices

Comparison of the results from different WTW, WTT or LCA studies of biofuels is often problematic. Parameters identified as responsible for introducing the largest variations and uncertainties are discussed below.

System boundaries and time frame

The first step of any life cycle oriented study is to identify and define the system boundaries, where the system boundaries act as cut-off points beyond which the system under study is assumed to not have any significant effect. Today it is widely accepted that to be able to analyze CO_2 emissions from or net energy use for a bioenergy system, the general full chain, from plant growth to end use, needs to be included. There is, however, no consensus of where to draw the system boundary.

In theory, any activities expected to be affected by the change in the life-cycle analyzed should be included. In practice, this can be hard to achieve, especially since the environmental consequences of a product or process are more than just the direct impact resulting from production, use and waste handling of the product. Sandén and Karlström (2007) use fuel cell buses as an illustrative example of the effects of including indirect effects when calculating possible CO_2 emissions reduction attributed to the introduction of fuel cell buses and corresponding fuel. They argue that not only direct effects resulting from marginal change of the current system should be included when making life cycle related analyses of emerging technologies but also indirect effects, where investments today can constitute marginal contributions to radical system change. For the case of the fuel cell buses the indirect CO_2 effects were shown to be several orders of magnitude larger than the direct effects.

As discussed by Gnansounou et al. (2009) the appropriate system boundaries are highly dependent on the purpose of the study. If the purpose is to, for example, compare different production pathways for one specific biofuel a WTT study is sufficient, as the vehicle combustion properties will be the same for all studied pathways. If on the other hand the purpose is to compare different fuels with each other and/or with fossil fuels it is, according to Gnansounou et al. (2009) crucial to always include the vehicle efficiency in a full WTW chain, even in studies intended for a simplified implementation, or serious inconsistencies may be introduced. In this study we will, however, not compare any fuel paths with each other. We will instead problematise the WTT method and discuss the uncertainties associated with calculating WTT values.

When making life cycle studies of bioenergy systems the time-frame of the study must be explicitly considered. In a general sense bioenergy is often viewed as carbon neutral, under the implicit assumption that the CO_2 emissions associated with the combustion of for example a tree will be fully balanced by the sequestration over the tree's growth period. However, as discussed by for example Schlamadinger et al. (1997), Reijnders and Huijbregts (2003) and Holmgren et al. (2007) bioenergy production systems are highly dynamic systems, with a number of CO_2 effects not directly related to the combustion process, but rather to land-use changes. The CO_2 effects from land-use change can effectively offset the emission decrease from substituting fossil fuels, if land with a high carbon stock (for example forest land or peat land) is cultivated (Reijnders and Huijbregts, 2003; Börjesson, 2009).

Time frame is also very important to consider when it comes to for example the reference system, as will be discussed in the next section.

Reference system

In system analyses of the potential of biofuels to decrease global fossil GHG emissions or fossil energy use, a baseline or reference system must be defined, where the reference system constitutes an estimation of what would have occurred in the biofuel project's absence. The reference system should include alternative pathways for the transportation fuel as well as for electricity, heat, and by-products. If the biofuel feedstock production results in land-use change, an alternative land-use must also be included in the reference system. Similarly, when the same feedstock is in demand for other purposes an alternative biomass use should be included, as the increased use of a resource with constrained production volume results in less of that resource being available for other parts of the system, which can cause important indirect effects that may significantly affect the results (Ekvall, 1999; Ekvall and Weidema, 2004; Merrild et al., 2008).

The choice of reference system depends largely on the aim and time frame of the study. In general the reference system should constitute a close alternative to the studied system, using the same technology level. If, for example, the study includes technology of which commercialisation is not imminent, such as large-scale black liquor gasification with biofuel synthesis, the reference system should incorporate projected best available technology for the same time frame rather than present average technology.

Another concern is the choice between average and marginal technologies for the reference system. A number of LCA related publications recommend the use of a marginal approach for change-oriented studies of possible future systems, particularly for comparison between different systems (see for example Weidema et al., 1999; Tillman, 2000; Ekvall and Weidema, 2004; Sandén and Karlström, 2007).

Several studies concerning the influence of the reference system have been published recently. The impact of assumptions regarding reference land-use has been discussed by for example Börjesson (2009) and Gnansounou et al. (2009), both showing a very strong influence of the land-use considered. Wetterlund et al. (2009b) and Hillman and Sandén (2008) vary the entire reference system. Both studies show that the assumed reference system brings a large degree of variation in the WTT CO_2 emissions and that it may have consequences on the ranking order of the studied biofuels. This makes it reasonable to include several different reference systems (scenarios) in biofuel WTT studies, in particular when studying biofuels for a future situation (see also Finnveden et al., 2009).

By-product allocation

By-product allocation is the distribution of environmental burdens between the different outputs of a process producing more than one product. This has been one of the most controversial and heavily debated issues of LCA methodology, as it can have significant impact on the results (see for example Finnveden et al., 2009). Several reviews of WTT studies of various biofuels show that by-product allocation is one of the key issues that influence the GHG and energy efficiency results (Larson, 2006; Delucchi, 2006; Fleming et al., 2006; Gnansounou et al., 2009, Börjesson, 2009).

Allocation can be done on the basis of physical properties (mass, energy content, volume etc.) or on the basis of economic value. Allocation can also be avoided through system expansion or substitution, i.e. expansion of the system's boundaries to include the additional functions of all by-products. For example, in the production of ethanol from grain, the protein-rich by-product is likely to be used as animal fodder substituting soy meal, the production of which will then need to be included in the reference system.

As discussed by Börjesson (2009) one advantage of energy allocation is that the relations between different by-products remain constant over time, while economic allocation is based on data that fluctuates over time. The results of economic allocation can however be more rational in systems with large amounts of low-value co-products, such as straw from grain based ethanol or low grade heat. Gnansounou et al. (2009) argue that price variations, subsidies and other market effects could imply practical problems when using economic allocation.

The ISO standard of life cycle assessment (ISO, 2006) recommends that allocation should be avoided by using system expansion when possible. This is however not without limitations. Examples are the issue of selecting the correct substitute, the need for accurate life cycle data on the alternative product or products, and that system expansion itself might cause new allocation problems, as discussed by for example Finnveden et al. (2009) and Gnansounou et al. (2009). Further, as discussed by Hillman and Sandén (2008) and Börjesson (2009) the size and saturation level of by-product markets should be considered when using system expansion.

The previously mentioned JRC/EUCAR/CONCAWE study (Edwards et al., 2007) uses system expansion for by-products from the biofuel systems, with the exception of electricity. Instead, for biofuel production processes with a non-neutral electricity balance, it is assumed that the electricity is produced in a biomass fired power plant. For production processes with a deficit of electricity the calculated amount of biomass for electricity production is added to the amount of biomass feedstock, and vice versa for processes with a surplus of electricity. The reason for this is reportedly that the large credit that could be the case if the electricity was deemed to replace marginal or average electricity, would distort the results. This reasoning only occurs with regard to electricity. For different biofuel production routes, different electricity production technologies are considered[33]. In Wetterlund et al. (2009b) system expansion is used consistently for all flows, also for electricity, which means that a non-neutral electricity balance affects marginal electricity production. System expansion is also used to make allowances for biomass scarcity by considering alternative uses for the available biomass. The results are compared to the results of the JRC/EUCAR/CONCAWE study, showing an in general considerably lower potential for reduced CO_2 emissions, mainly due to the expansion of the system to include alternative biomass use.

The choice of allocation method for biofuel WTT assessments is not trivial, not even for relatively simple systems where the main product can be easily defined, reducing other flows of energy or material from the process to by-products. In Börjesson (2009) and Gnansounou et al. (2009) the choice of allocation method is shown to have a substantial impact on the GHG emission results for such a system; in these studies ethanol from wheat with straw and

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[33] For the gasification of solid biomass, gasification based electricity production with an efficiency of 42.5% is used. For cellulosic ethanol, a wood-fired steam turbine condensing power station with an efficiency of 32% is used. The same type of electricity production is considered for black liquor gasification, but with an efficiency of 40%.

draff as by-products. Processes involving the co-production of several products, which is the case with for example many biorefinery concepts, can affect a large number of processes outside the biofuel life cycle investigated, and may not even have the biofuel as a clearly defined main product. If the biofuel production process is integrated with other industries or district heating systems, further problems arise. For example, if system expansion is used for a system with a relatively low biofuel output and a large output of a particular by-product, such as low-grade heat, a high CO_2 emissions reduction potential may be erroneously attributed to the properties of the biofuel when it is really an effect of the large heat output. This issue is further discussed in the following section.

Functional unit

In studies where different systems are compared the functional unit must be carefully selected and defined. When biofuels are compared to each other and/or to fossil fuels the service provided, i.e. the distance travelled, can be chosen as the functional unit, as argued by for example Edwards et al. (2007) and Gnansounou et al. (2009). This of course requires that the full WTW chain is considered in the study.

If biofuels are to be compared with other bioenergy applications, such as heat and/or electricity production, another functional unit must be chosen. Several studies, for example by Schlamadinger et al. (1997) and Gustavsson et al. (2007), emphasise the importance of considering the resource that will be delimiting, for example for the GHG reduction potential. For bioenergy systems this will typically be the available amount of biomass or the available land for biomass production. This is supported by Larson (2006), who however acknowledges that this will bring additional site-specific dimensions into the analysis, typically biomass crop yield. If the feedstock is the same in all considered cases, for example forest residues, the relative order of the results will of course be the same when reporting per ha and year as when reporting per unit biomass. When different feedstocks are compared however, land use efficiency becomes increasingly important, since the land area available for biomass production is limited. Schlamadinger et al. (2005) broaden the discussion to also include economic limitations. As the studies mentioned above, they argue that for studies comparing for example the GHG mitigation potential of various systems, the functional unit should reflect the limiting resource. In addition to the already mentioned factors of available biomass or land area, the limiting factor can be available monetary resources or bioenergy output (for example heat, electricity or biofuels) that can be absorbed by the market.

The choice of functional unit is associated with several methodological considerations. If, for example, the results are presented as driving distance per ha adjustments of included processes need to be made by recalculation into the considered type of biomass. This may lead to the inclusion of unlikely components to the system studied.
As discussed in the previous section methodological choices concerning allocation method when studying biofuels, in particular when studying systems co-producing several energy carriers or products, may lead to misleading results. This has been previously discussed by for example Schlamadinger et al. (1997) and Johansson (1996). For instance, a biofuel process co-producing large amounts of low-grade heat may give the largest GHG reduction compared to other biofuel options if evaluated on a fuel input or land use basis. This should not be interpreted as that the biofuel in question is a better option from a GHG point of view than other fuels, but rather be taken as an indication that biofuel production may not be an efficient way to reduce GHG emissions, compared to other biomass usages.

To counter this problem Schlamadinger et al. (1997) propose a method where the reference entity is expanded so that all studied systems produce the same output. A similar approach is used by Gustavsson and Karlsson (2006) who propose to expand the functional unit to include all energy carriers or products produced. Using the method of an expanded functional unit, however, may lead to the inclusion of unlikely components to the system studied, such as biomass fired condensing plants. Further, when comparing very different systems or systems of a very complex nature there is a risk for losing transparency in the calculations.

Summary of general approaches for WTT studies
Most studies are focused on direct effects from physical flows in the WTT chain, but some studies also include an estimation of contributions to radical system change. The time perspective is dependent on for example the considered technology and the aim of the study. When it comes to methodological issues such as choice of reference system and handling of by-products, one can discern a tendency for some different approaches.

A common approach in WTT studies is for example to use system expansion or substitution for crediting by-products, as recommended by for example the ISO standard. This is sometimes compared with other ways to handle by-products such as allocation based on energy content or economic value. The reference system for example concerning electricity generation are based on average, or sometimes marginal technology, or for example a system using the same raw material as for the motor fuel production and then the amount of raw material used for the electricity generation are added to/subtracted from the usage of raw material for production of the fuel.

Another approach is to use system expansion for all flows involved in the WTT chain. The flows entering or leaving the biomass conversion system is assumed to cause a change in the surrounding system. Since all flows are handled by system expansion, allocation of by-products need not be considered.

One approach is to expand the reference entity so that all studied systems produce the same output. For example if a biofuel is produced together with electricity and district heating, it is compared to a reference system where the same amounts of transportation fuel/transportation work, electricity and district heating are produced. This method also avoids allocation of by-products.

To be able to, for example, present the results as driving distance per ha, all flows have to be recalculated into the considered type of biomass. Thus, technologies in the reference system for example concerning electricity and district heating have to be biomass based.

As is apparent from these descriptions, there are no sharp dividing lines between the different approaches. There are similarities, as well as clear differences, between the described methods. The first one is more a general description of conventional WTT studies, mixing elements from the other described approaches/methods. Whichever method is used, it is important that the reference system constitute a close alternative to the studied system, using the same technology level. If, for example, the study includes technologies of which commercialisation is not imminent, the reference system should incorporate projected best available technology for the same time frame rather than present average technology. Inclusion of alternative land-use and/or alternative usage of the biomass feedstock may significantly affect the results, as shown by several studies.

Appendix 3. WTW challenges, biomass feedstock

Challenges when describing biomass feedstock in WTT analyses

Land-use changes
Land-use and land-use changes have a substantial impact on GHG from biofuel production, since soils and plant biomass constitute the largest carbon stores on earth (Fargione et al., 2008). Unless fallow land or waste biomass is used, the land-use change to biofuel production will create a rapid release of CO_2 into the atmosphere. This "carbon debt" of land conversion can in extreme cases (conversion of rainforest for, for example, palm oil production) takes several centuries to repay by fossil fuel substitution (Fargione et al., 2008).

Direct land-use change, where the production of biofuel feedstock displaces previous land-use which may lead to changes in the land carbon stock, is considered in several recent studies, for example in the widely cited JRC/EUCAR/CONCAWE study (Edwards et al., 2007). The effects of indirect land-use change that would occur if increased bioenergy production displaces for example existing food production to new agricultural land are more complex to determine, but may have large impact (Searchinger et al., 2008; Gnansounou et al., 2009).

The land-use change effects have different impact depending on whether first or second generation biofuels are considered. As discussed in for example Searchinger et al. (2008) and Holmgren et al. (2007), first generation fuels, such as ethanol from corn, will give a higher total net effect of GHG due to land-use change. In general, bioenergy production on arable land, e.g. corn, grass and SRF, has a higher impact mainly due to the indirect effects mentioned above, but there is a large variation in how high, as showed by for instance Gnansounou et al. (2009), who give example of wheat ethanol CO_2 emissions ranging from −80% to +5%, compared to gasoline. For waste biomass, e.g. forest residues, soil carbon dynamics can have a substantial impact. When logging residues are removed from the forest, the soil carbon stock will in general be lower than if the residues were left in the forest to decompose, particularly if looked at over a short time period. The magnitude of the impact of the soil carbon decrease is, however, uncertain, with estimated numbers ranging from around 10 to almost 50 kg CO_2/MWh (Holmgren et al., 2007).

Biomass supply
The supply of biomass is very much dependant on local conditions and what kind is available will vary for different regions. Another important issue when it comes to the future supply is that not only the theoretical potential will matter but also technical, ecological and economic factors have an impact. A number of studies have been made, trying to estimate the future potential of biomass, and for a forest rich country like Sweden the total potential may be somewhere between 560 and 800 PJ in 2025 (Lindfeldt et al., 2010).

The fact that the supply of biomass is limited is acknowledged by several studies (Wetterlund et al., 2009b; Lindfeldt et al., 2010; Hoogwijk, 2004; Berndes et al., 2003), however, other uses of biomass than for biofuels, such as e.g. for heat and power production or in the pulp and paper industry, is not always stressed. An increased use of a resource with constrained production volume results in less of that resource being available for other parts of the system, therefore efficient use is essential if the CO_2 benefit of substituting biomass for fossil fuels is to be maximized.

Since biomass is a limited resource, it is not possible to solve the whole climate problem by substituting biomass for fossil fuels. To be able to give credits for biomass released for other use when implementing efficiency measures, for example, it is of crucial importance to take the marginal effects of limited resources into consideration when evaluating GHG emissions.

Handling, transportation and distribution of the biomass
Another issue concerning the biomass feedstock is the handling, transportation and distribution of the biomass. The energy needed for this, as well as the emissions because of it, is of course dependant on the type of biomass. For instance, solid biofuels originating from the forest industry used in biofuel fuel production has a clear advantage over energy crops grown explicitly for biofuel production, since the solid biofuel is seen as a by-product from the forest industry and the emissions associated with it is allocated to the main product of the forest industry (Holmgren et al., 2007).

How large emissions are and how much energy is needed for the transportation, handling and distribution, will also be depending on the size of the biofuel production plant, and if it is possible to supply the plant with biomass from the local region, or if biomass have to be transported from a larger area or even imported from another country.

Appendix 4. WTW challenges, biofuel production

Challenges when describing biofuel production in WTT analyses

For emerging technologies, such as second generation biofuels, factors that add uncertainty to the WTT analyses are the development status of the biofuel production technologies, the variance in process performance between different concepts, and the availability of reliable input data concerning the processes. In general, publicly available data is scarce and it is usually difficult to evaluate whether available data is based on actual achieved results, extrapolated data from laboratory or pilot scale, or estimations based on simulation models.

For some emerging technologies, the uncertainty regarding for example the maximum achievable fuel yield or the degree of energy efficiency of the process that will be economic is significant. It is important to state what kind of technology level that is considered in a study and make comparison between different technologies adopting similar technology levels. The overall energy efficiency of a process can be improved by considering for example increased heat integration, both internally and with external processes (see e.g. Fornell et al., 2009). If a biofuel production process is integrated with for example another industrial process, the reference system needs to include a relevant reference case (or several) for the industrial process non-integrated with the biofuel production plant (see e.g. Pettersson and Harvey, 2010a). It is also important to discuss the integration potential, for example if it is even relevant in all cases, as well as how a different integration possibility affects the results of the WTT study (see e.g. Wetterlund et al., 2009a).

Electricity

As well as raw material, electricity is needed in biofuel production processes for compression and pumping of flows and production of oxygen, to mention some examples. If a steam/heat deficit is present in the process, the usage of a CHP (combined heat and power) plant fuelled by either external fuel or some kind of by-product fuel from the production process is appropriate. In gasification processes, for example, electricity could be generated using excess steam and/or off gas. All in all, the plant could either have a net deficit or surplus of electricity.

The net deficit or surplus of electricity can be handled in different ways. In the case of electricity grids, one can use the average GHG or energy intensity of the entire system, the build margin or the operating margin (see e.g. Kartha et al., 2004; Ådahl and Harvey, 2007; Schlamadinger et al., 2005). What is a relevant grid electricity mix or margin technology to use is dependent on for example the time frame and the system boundaries of the study. In Wetterlund et al. (2009b) the influence of different future build margin technologies on the WTT CO_2 emissions for different second generation biofuel routes are shown.

An electricity deficit or surplus can also be handled by assuming that the electricity is produced in a biomass fired power plant. For production processes with a deficit of electricity the calculated amount of biomass for electricity production is added to the amount of biomass feedstock, and vice versa for processes with a surplus of electricity (see e.g. Edwards et al., 2007). Especially for processes with a large surplus or deficit of electricity, the assumptions regarding the reference electricity system affects the results of a WTT study significantly.

Heat

Heat at different temperature levels, warm and hot water, steam at different pressure levels etc., are to different extent needed in biofuel production processes. Some processes, e.g. gasification processes, also have large cooling demands used to generate useful steam/heat and these processes can have a net steam/heat surplus. For processes with a net steam/heat deficit, a heat only boiler or a CHP plant fired by external fuel or a fuel by-product could be used. It is of course possible for a process to have, for example, a deficit of steam and a surplus of low grade heat.

Processes with a net steam deficit could benefit from integration with an industrial process with a net steam surplus and vice versa for a process with a net steam surplus. For example, the integration between a lignocellulosic ethanol process with a net steam deficit and a market pulp mill with a net steam surplus has been investigated in KAM (2003). Wetterlund et al. (2009a) investigated the integration between a solid biomass gasifier connected to production of DME, having a net steam surplus, and an integrated pulp and paper mill with a net steam deficit. As previously discussed, if a biofuel production process is integrated with another industrial process, the reference system needs to include a relevant reference case for the industrial process non-integrated with the biofuel production plant.

For processes with excess heat, e.g. gasification processes, integration could be of crucial importance for the profitability of the process. Except for integration with another industrial process, delivering heat to a district heating system could be an option. In the case of district heating, the size of the market is determined by local conditions. Choosing the location for co-production of biofuel and district heating needs careful consideration, since it is necessary to find a district heating network with a base load large enough to be able to accept all excess heat from the plant. The alternative district heating production is very much dependent on local conditions, such as the heat demand and availability of different fuels. For example, in a Swedish perspective a biomass CHP is often considered as a competing technique to industrial excess heat (see e.g. Jönsson et al., 2008; Wetterlund et al., 2009b).

When heat from the biofuel process substitutes CHP heat, biomass is released for other uses. Thus, it is important to be able to give a CO_2 emission credit for the, in this case, indirect contribution to a decreased use of biomass. Note also that utilisation of heat from the biofuel process decreases the system's electricity generation, since the CHP plant is replaced. This could actually lead to a negative CO_2 effect from district heating production from industrial excess heat, even if a CO_2 credit for the release of biomass is given (see e.g. Wetterlund et al., 2009b).

By-products

A surplus of electricity, as well as heat, is of course a kind of by-product and can be handled not only by system expansion but also by allocation on the basis of physical properties (mass, energy content, volume etc.) or economic value. Examples of by-products from biofuel processes, except for electricity and heat, are different kinds of fuels, animal feed and chemicals.

When expanding the system to include the additional functions of all by-products, the size of by-product markets must be taken into consideration. Hillman and Sandén (2008) point out that the size of by-product markets should be considered and different by-product credits could be given depending on the degree of market penetration of the studied biofuel.

Carbon capture and storage (CCS)

An often proposed strategy for reducing CO_2 emissions is by using carbon capture and storage (CCS). This is commonly discussed for fossil based systems, such as coal power plants, but could also easily be applicable to a biofuel production system, thus lowering the total CO_2 impact from the biofuel. In gasification processes (connected to fuel synthesis) as well as in ethanol fermentation, for example, CO_2 is removed as a part of the process. By capturing and storing the CO_2, substantial emission reductions could be achieved (Lindfeldt and Westermark, 2008; Möllersten et al., 2003; Azar et al., 2006; Wetterlund et al., 2009b). The amount of captured carbon depends on what energy carrier is produced, e.g., DME (CH_3-O-CH_3) contains carbon while hydrogen (H_2) does not.

The possibility for CCS could affect the WTT CO_2 emissions of a biofuel system both directly, if CO_2 capture is possible in the biofuel production process, and indirectly if for example CCS is implemented in coal power plants (lowers CO_2 emissions from grid electricity). Factors such as time perspective, the amount of removed CO_2 at the biofuel production plant and nearby industrial processes or power plants and possibilities for transportation and storage of the CO_2 determines if CCS is a realistic option.

Appendix 5. WTW challenges, biofuel usage

Challenges when describing biofuel usage in WTT analyses

The energy used in the Swedish road sector was approximately 88 TWh in 2008 of which 4.9% was biofuels, i.e., ethanol, biodiesel and biogas (Energimyndigheten, 2009). The main part of this is used in low-volume blends with fossil fuels[34]. By using blends the need for technical adaptations of for example infrastructure and vehicles, is minimal. If, however, higher targets for renewable energy in transportation (10% for 2020, (Dir 2009/28/EC)) are to be met biofuels will need to be used pure or in higher volume blends, which will put demands on new infrastructure and vehicles.

While the biofuels already on the market have properties similar to petrol and diesel, some of the emerging biofuels require completely new technology, for example DME (dimethyl ether) which is gaseous at normal pressure and temperature, making it impossible to blend with conventional liquid fuels. Fuels used as blends have implications for CO_2 emissions related to distribution, dispensing and usage.

The type of vehicle used will also affect the WTW value. Fuels that are used in diesel engines have some advantages over fuels used in otto engines. Fuels that can be used in even more energy efficient power trains, e.g., fuel cells and plug-in hybrids will also benefit in the WTW comparison.

Today oil based fuels, above all gasoline and diesel, are the total dominating in the transport sector and different biofuels are likely to replace these fuels. However, since crude oil is a considerably limited resource the dominating transportation fuels of the future could be coal based. For example, FTD produced via gasification of coal, with as well as without CCS, could be considered for the future reference transportation system.

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[34] Low-volume blends of ethanol in gasoline was 1.5% (1.34 TWh) and biodiesel blended in diesel 1.7% (1.47 TWh) compared to ethanol in E85 1.1% (1.01 TWh), ethanol to buses 0.2% (0.14 TWh), pure biodiesel 0.1% (0.04 TWh) and biogas 0.4% (0.33 TWh) (SPI, 2009).

Appendix 6. Assumptions in Edwards et al. (2007)

Details about the assumptions in the CONCAWE/EUCAR/JRC study

In this Appendix we have gathered more information about the assumptions made in Edwards et al. (2007), which results are presented in Section 2.3.1.

Regarding the biomass Edwards et al. (2007) consider sources that have the potential to substitute a significant amount of fuels for transport in the EU i.e. farmed crops such as sugar beet, wheat, oil seeds but also woody biomass either in the form of waste wood or farmed wood. Wood farming incorporates also perennial grasses such as miscanthus, switch grass or reed canary grass (Swedish: rörflen). In total 38 fuel routes are considered, which are illustrated in Figure A6.1. Note that the coal-based fuel options have been excluded from the illustration.
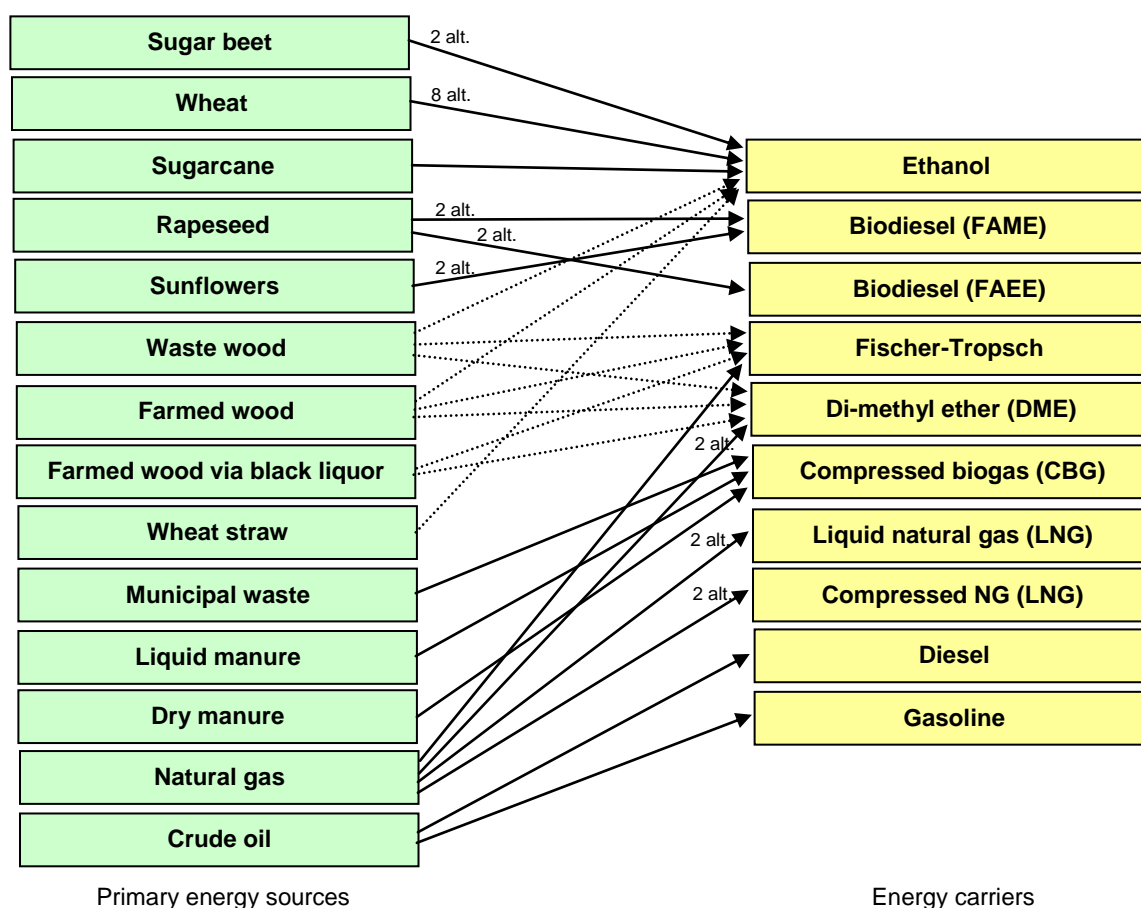

Figure A6.1. An illustration of the 38 fuel routes considered in the Edwards et al. (2007). Dotted line represents the fuel routes not yet available on the market. The study also includes coal-based fuels but since all of them emit more CO_2 than conventional oil-based fuels they are not presented here.

The notional time horizon for the WTW-study is the next decade 2010-2020. The technologies considered are those that are expected to become commercially available in that time frame. The values for fuels not yet on the market are engineering estimates.

When system expansion is applied in Edwards et al. (2007), draff is assumed to replace imported soy bean meal from Brazil and straw is assumed to replace forest chips as fuel. Dried draff is currently used as animal protein feed in primarily milk production which is normally using soy protein feed. This replacement is estimated to result in a GHG reduction equivalent to approximately 430 kg CO_2-equiv./GJ ethanol (Börjesson & Tufvesson, 2010; Flysjö, Cederberg and Strid, 2008).

The total WTW energy and greenhouse gas emissions combine the well-to-tank (WTT) total expended energy, (MJ_{xt}) per unit of energy in the fuel (MJ_f), with the tank-to-wheel (TTW) energy consumed by the vehicle per unit of distance covered, as

Total WtW energy (MJ/100 km) = TtW energy (MJ_f/100 km) * [1 + WtT total expended energy (MJ_{xt}/MJ_f)].

Each biofuel plant uses steam in the fuel production plant. Currently most common in Europe is steam originating from natural gas conventional boilers. As comparison Edwards et al. (2007) performed figures for alternative steam sources, e.g. natural gas CCGT, coal fired CHP and straw fired CHP.

In addition to steam, conversion plants also use electricity. Edwards et al. (2007) bases the greenhouse gas emissions from electricity on the typical European electricity supply mix, as listed in Table A6.1. The result may therefore change if Nordic electricity mix or assumptions on any other future electricity mix were chosen. On the other hand European electricity production is covered by the CO_2 emissions cap and trade system, meaning that the electricity emissions theoretically can be omitted from the WTT analyses.

Table A6.1. EU-mix electricity sources assumed in Edwards et al. (2007)

Source	Share (%)
Nuclear	37.5
Coal	22.5
Oil	9.6
Natural gas	15.5
Hydro	12.4
Wind	0.4
Waste	1.8
Other renewables	0.3

Including the distribution losses at the medium voltage level the overall energy efficiency is around 35% and the corresponding GHG emissions 430 gCO_{2eq}/kWh_e. This number is (as stated in the study) coincidently quite close to the global figure for generation of electricity with natural gas in a state-of-the-art combined cycle gas turbine, a route often viewed as the most likely marginal electricity source in Europe for the foreseeable future (Edwards et al., 2007).

Energy balance

The energy balance is presented as total primary energy expended (MJ_{xt}), regardless of its origin, to produce one MJ_f of the final fuel (LHV basis[35]), i.e. 2 MJ_{xt}/MJ_f means that twice as much energy (both fossil and renewable) is required to produce the fuel, as is available to the final user. As such they describe the energy efficiency of the pathway.

Greenhouse gas emission

Values on greenhouse gas emissions represent the total grams of CO_2 equivalent emitted in the process of obtaining 1 MJ_f of the finished fuel. The CO_2 equivalence is applied to methane (CH_4) and nitrous oxide (N_2O) (other greenhouse gases are not emitted in significant quantities in any of the processes considered) according to the conversion coefficients recommended by the third assessment report of the Intergovernmental Panel on Climate Change (IPCC, 2001), see Table A6.2.

Table A6.2. IPCC factors for calculating the total impact of greenhouse gases

Greenhouse gas	tCO_{2eq}/t
Carbon dioxide (CO_2)	1
Methane (CH_4)	23
Nitrous oxide (N_2O)	296

Most liquid biofuels rely on traditional food crops, typically produced through intensive farming which is responsible for a large portion of the greenhouse gases. Nitrous oxide is a very powerful greenhouse gas (almost 300 times that of CO_2), leading to even relatively small emissions having a significant impact on the overall GHG balance. There are essentially two sources for N_2O emissions: nitrogen fertilizer production and emissions of nitrous oxide from the field. N_2O emissions from different fields vary by more than two orders of magnitude, depending on a complex combination of soil composition, climate, crop and farming practices (Edwards et al., 2006).

Most of the European crops for biofuel production have so far been produced in replacement of cereals exports, i.e., not much new land areas have been needed. The most common scenario for growing additional biofuel crops is to grow on set-aside land. The land use changes occurring when farming on set-aside land will impact the GHG emission. That is, anything planted on grazing or forest land in the short and medium term will lead to increased GHG emissions, which then is counter-productive with regards to the GHG reductions from biofuel use.

Due to the assumption of crop expansion on set-aside land, unfertilized, unharvested grass is used as baseline, having negligible energy inputs but a significant N_2O emission, which is subtracted from the N_2O calculation (Edwards et al., 2006).

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[35] All energy contents referred to in this report are on LHV basis i.e. excluding the heat generated after the combustion process by the condensation of water vapour.

Appendix 7. Assumptions in Wetterlund et al. (2009b)

Details about the WTW study assuming system expansion

In this Appendix we have gathered more information about the assumptions made in Wetterlund et al. (2009), which results are presented in Section 2.3.3. Table A7.1 presents input data for the different technology cases considered.

Table A7.1. Input data for the plant configurations studied. Negative values indicate import to plant.

		BLG:DME		BMG:MeOH		EtOH		BIGCC	
		no CCS	CCS	no CCS	CCS	no CCS	CCS	no CCS	CCS
Biomass feedstock	MW	-157[a]	-157[a]	-229	-229	-222	-222	-140	-140
Biofuel/electricity for use in transp.	MW	275	275	123	123	58	58	60	52
Electricity	MW	-101[a]	-111[a]	-13	-17	46	45	–	–
District heating	MW	–	–	13	13	88	88	60	61
Captured CO_2	kg/h	0	87,500	0	38,600	0	7,300	0	42,300
Biomass conditioning emissions	kg CO_2/ MWh$_{biomass}$	7.13		7.13		7.13		7.13	
Biofuel distribution emissions	kg CO_2/ MWh$_{biofuel}$	3.74		3.96		3.35		–[b]	
Vehicle energy use	MWh/ 1000 km	0.39		0.41		0.45		0.17	

[a] The incremental biomass and electricity use compared to the pulp mill reference case, i.e. for biomass the 125 MW forest residues imported to the plant plus the 32 MW bark that could be exported in the reference case, and for electricity the 56 MW (or 66 MW when CCS is considered) imported electricity plus the 45 MW electricity that could be exported in the reference case.
[b] Distribution efficiency is assumed to be 93%.

For biomass, it is assumed that the high volume user with the highest willingness to pay will be price setting and thus constitute the alternative use. One potential marginal biofuel user, considered in this study, is coal power plants where biomass is co-combusted with coal. Other biomass users, such as boiler fuel substitution (oil) and industrial CHP, often have higher willingness to pay for biomass compared to coal power plants. However, due to these users' limited demand, they are assumed to not constitute marginal high volume users. The alternative where biomass is not subject to competition, and thus the use can be viewed as CO_2-neutral, is also considered in order to illustrate the marginal effect of biomass usage.

For each of the alternative biomass options, different possible future margin electricity technologies are considered. Since the timeframe for the biomass conversion projects studied here is relatively long, base load build margin rather than operating margin is used. The base load build margin is here defined as the type of electricity production, the building of which is affected by the biomass conversion project in question. Three state-of-the-art fossil electricity production technologies are considered as build margin technologies: coal power, coal power with CCS, and NGCC (natural gas combined cycle). In addition to these, fossil free CO_2-neutral electricity production (for example, but not limited to, wind power) is also included as a possible future build margin technology.

Table A7.2 shows the reference system matrix with CO_2 emission values for the different reference technologies considered.

Table A7.2. Reference system matrix with CO_2 emission values.

Biomass use[a] kg CO_2/MWh$_{biomass}$		Electricity[a] kg CO_2/MWh$_{el}$		District heating[b] kg CO_2/MWh$_{heat}$		Transportation[c] kg CO_2/1000 km	
Co-firing with coal	329	Coal	723	Biomass CHP	142	Oil	134
						Coal	289
						Coal w. CCS	160
		Coal with CCS	136	Biomass CHP	359	Oil	134
						Coal	289
						Coal w. CCS	160
		NGCC	374	Biomass CHP	271	Oil	134
						Coal	289
						Coal w. CCS	160
		CO_2-neutral	0	Biomass CHP	410	Oil	134
						Coal	289
						Coal w. CCS	160
No alternative use	0	Coal	723	Biomass CHP	-268	Oil	134
						Coal	289
						Coal w. CCS	160
		Coal with CCS	136	Biomass CHP	-50	Oil	134
						Coal	289
						Coal w. CCS	160
		NGCC	374	Biomass CHP	-138	Oil	134
						Coal	289
						Coal w. CCS	160
		CO_2-neutral	0	Biomass CHP	0	Oil	134
						Coal	289
						Coal w. CCS	160

[a] Well-to-gate values.
[b] Biomass CHP. Well-to-gate values.
[c] Use in hybrid vehicles. Well-to-wheel values.

For district heating, the alternative production is very much dependent on local conditions, such as the heat demand, which makes determination of the margin technology difficult. In order to keep the number of reference systems considered manageable, Wetterlund et al. (2009b) only comprises one alternative district heating technology; biomass CHP based on a conventional steam cycle. The reason for choosing biomass CHP is that it frequently constitutes a competing technique to industrial excess heat. When heat from the biomass conversion processes substitutes CHP heat, biomass is released for other uses, in this case the alternative biomass usage described above. Note also that utilisation of heat from the biomass conversion processes decreases the system's electricity generation, since the CHP plant is replaced.

Since hybrid vehicles are assumed for all analyzed biofuels, the reference transportation technology is assumed to be hybrid fossil fuelled vehicles. Today fossil oil is the dominating fuel in the transportation sector. However, since crude oil is a considerably limited resource the marginal transportation fuels of the future could be coal based. For this reason, in addition to oil based transportation fuels[36], synthetic fuels from coal (FTD), with as well as without CCS, are considered for the reference transportation system.

The only feedstock considered is forest residues, why the change in CO_2 emissions is evaluated per unit of biomass. An alternative approach could be to report on a per hectare and

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[36] Average value between gasoline and diesel.

year basis, an approach that is commonly used for agricultural feedstocks, but that could be useful also for forest residues.When the feedstock is the same in all considered cases, the relative order of the results will of course be the same when reporting per hectare and year as when reporting per unit biomass.

As mentioned, the results are compared with the results from Edwards et al. (2007) to show the effects of expanding the system. The results from Edwards et al. (2007) are here considered in reference to all three marginal transportation alternatives included in this study. Edwards et al. (2007) takes CCS into account for fossil based systems but not for biomass based systems. For this reason only the results for the studied technologies without CCS are compared with the results from Edwards et al. (2007). A major methodological difference between Edwards et al. (2007) and Wetterlund et al. (2009b) is the handling of electricity. In Wetterlund et al. (2009b), electricity is managed by system expansion, which means that a non-neutral electricity balance affects marginal electricity production.

Edwards et al. (2007) uses a substitution method for co-product allocation, which corresponds to system expansion. Electricity, however, is not considered a co-product[37]. Instead, for biofuel production processes with a non-neutral electricity balance, it is assumed that the electricity is produced in a biomass fired power plant. For production processes with a deficit of electricity the calculated amount of biomass for electricity production is added to the amount of biomass feedstock, and vice versa for processes with a surplus of electricity. Different electricity production processes with different efficiencies, are considered for different biofuel routes[38].

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[37] The reason for this is reportedly that the large credit that could be the case if the electricity was deemed to replace marginal or average electricity, would distort the results. This reasoning only occurs with regard to electricity.

[38] For the gasification of solid biomass, gasification based electricity production with an efficiency of 42.5% is used. For cellulosic ethanol, a wood-fired steam turbine condensing power station with an efficiency of 32% is used. The same type of electricity production is considered for black liquor gasification, but with an efficiency of 40%.

Appendix 8. Assumptions and part results in Volvo (2007)

Details about the WTW study judging seven criteria by score

In this Appendix we have gathered more information about the assumptions made and part results found in Volvo (2007). The main results are presented in Section 2.3.4.

Climate impact

Although the calculations refer to fully renewable raw materials, fossil fuels are currently used in the cultivation and production of the fuels. In the future, it will be possible to replace this fossil energy with renewable energy.

The CO_2 emissions are calculated for the complete fuel chain "Well to Wheel" as CO_2 equivalent emissions. This means that any emission that contributes to global warming such as N_2O, CH_4 etc. is converted into CO_2 equivalents according to IPCC conversion factors. The non fossil CO_2 is not included since these emissions do not lead to a net increase of GHG in the atmosphere. The data on GHG emissions of fuels for the well to tank part was taken from Edwards et al. (2007). This data was modified in order to take into account ignition improvers for methanol and ethanol. The tank to wheel data was taken from Volvo internal material on engine efficiency with different fuels. The energy efficiency of the engines is used to calculate the CO_2 emissions.

Results

Five of the fuel pathway options reduce the impact on the climate by over 90 percent. In the case of methanol, gasification of black liquor is required to achieve the highest rating. In the case of biogas and biogas/hydrogen mixture, biomass gasification is required to achieve the highest rating. The lower rating applies if the biogas is produced by anaerobic digestion of household waste. Ethanol offers a reduction of 0 to 75 percent depending on the production method. Production and use of ignition improvers somewhat decrease the emission savings from vehicles using ethanol and methanol. Results on climate impact are presented in Figures A8.1 and A8.2.
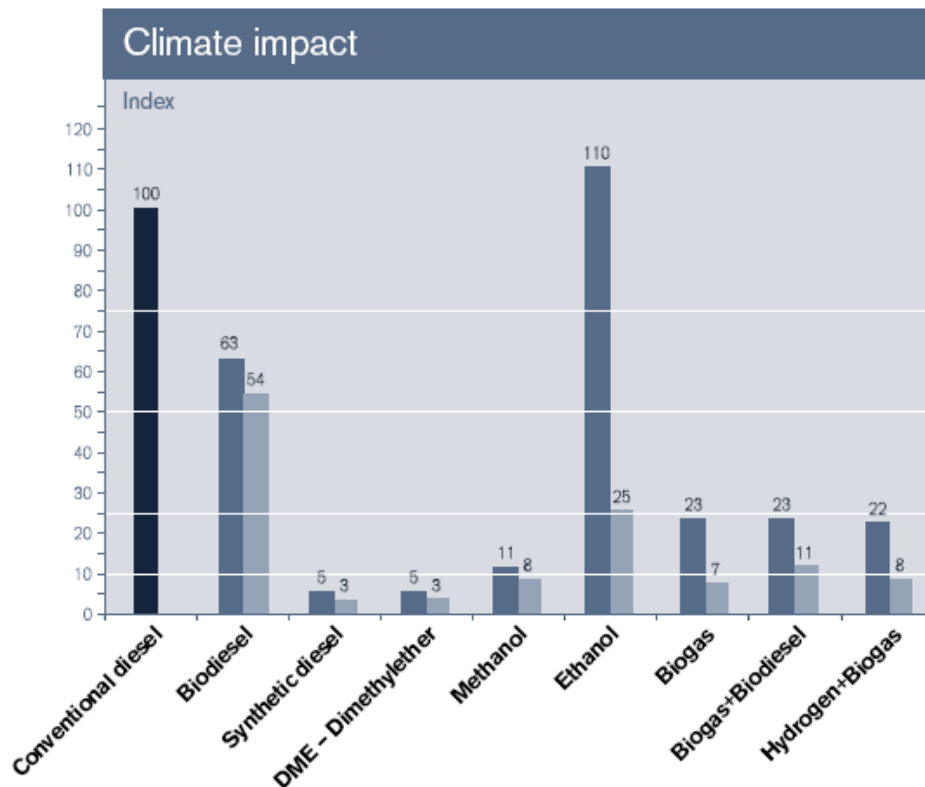
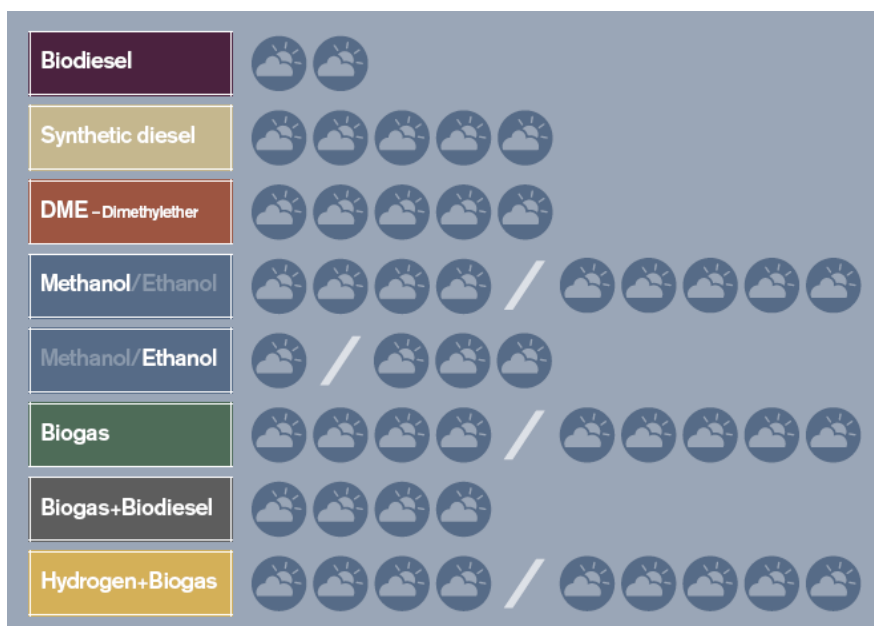**Figure A8.1.** Quantitatively results on the criteria "climate impact".

Figure A8.2. Score results on the criteria "climate impact". Note that some of fuel alternatives may end up in several intervals since different processes producing the same fuel are analyzed.

Energy efficiency

The energy efficiency is defined as the total energy use of a fuel pathway from well to wheel. The WTT energy efficiency is calculated using background data from Edwards et al., (2007) for all relevant steps of the process including the following: cultivation of the feedstock, use

of fertilizers, harvest of feedstock, transportation to fuel production plant, production of fuel, distribution of fuel to filling station. The data was modified in order to take into account ignition improvers for methanol and ethanol. The TTW data was taken from Volvo internal material. Data for biomethane production via gasification was estimated since it is not included in Edwards et al. (2007). Allocation methods applied is consistent with Edwards et al. (2007).

Results

DME and methanol are rated highest when produced from black liquor gasification within the pulp industry. The higher rating for synthetic diesel is also based on the gasification of black liquor. The ratings for biogas, biogas+biodiesel (methane-diesel) and biogas/hydrogen mixture are based on production by gasification and anaerobic digestion. However, the production of biogas (biomethane) via black liquor gasification is not included due to lack of data. The low rating of ethanol is due to the high energy use in the cultivation and fuel production processes. Results are presented in Figures A8.3 and A8.4.
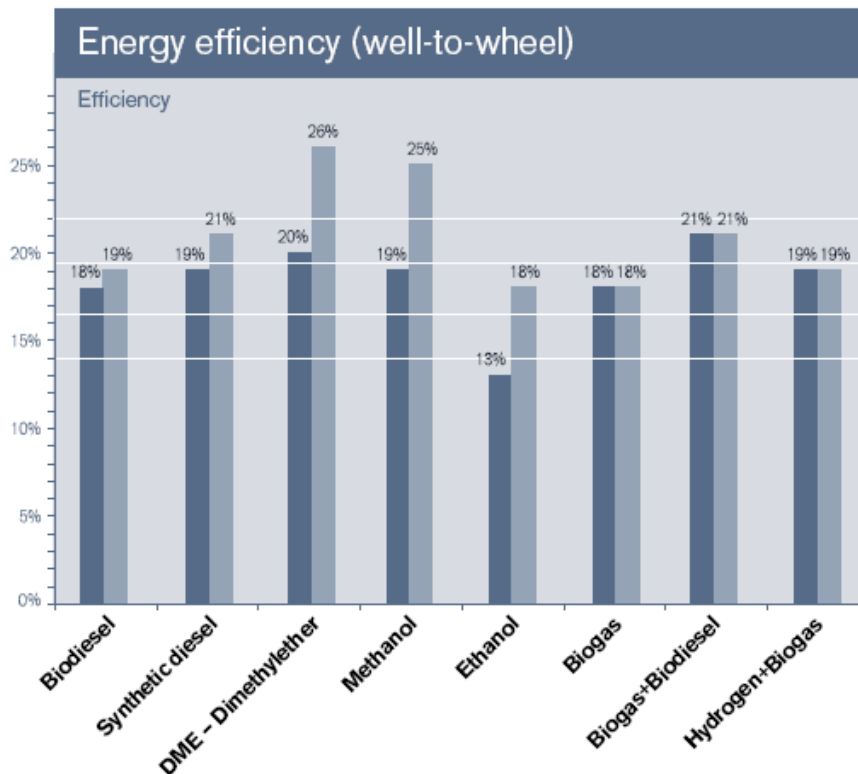

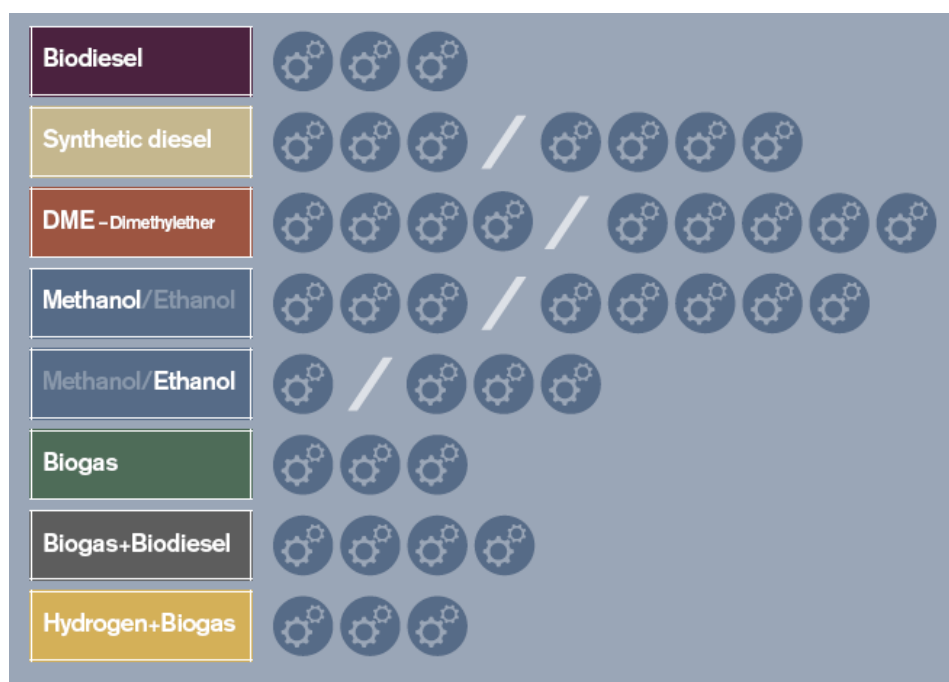**Figure A8.3.** Quantitatively results on the criteria "energy efficiency".

Figure A8.4. Scores results on the criteria "energy efficiency". Note that some of fuel alternatives may end up in several intervals since different processes producing the same fuel are analyzed.

Land use efficiency

Efficient land use will be an increasingly important factor in meeting the world's ever-growing demand for food and fuel. Driving distance per hectare per year is a measure of the performance of biofuel. The yield per hectare for each crop has been calculated using figures for average yields from good quality land. Growth is based on Swedish conditions. Although crop cultivation in other locations may yield different results, the relativities are more or less the same. The quantity of fuel/energy used in harvesting, production, transport etc. is subtracted from the quantity produced. Results for the same fuel may vary depending on the production process used.

The distance which a heavy duty vehicle can travel per year and hectare is calculated using the following data: (i) yield of the selected crop from the field in dry tons per hectare and year, (ii) input of fossil fuel in order to evaluate the net yield from the field, (iii) energy efficiency of producing the biofuel from the selected crop "Well to Tank", and (iv) fuel consumption per km in a commercial truck (3 litre diesel equivalents per 10 km).

A low yield together with high fossil energy input and low energy efficiency will give poor results on distance per hectare. The data used for the calculations was taken from Edwards et al. (2007), RENEW and discussions with Lund University.

Calculation example for Salix to DME

Salix is a woody crop (e.g., willow) which can be harvested every 3-4 years. If the yield is allocated to each year an amount of approximately 10 tons of biomass can be achieved per hectare and year. This figure is for a well managed salix farm. In the future the yields are expected to increase since there still is a potential for improvement. Salix is not a rotation crop.

123

The energy content of salix is 19 MJ/kg. The total yield of salix is therefore 10 tons multiplied by 19 which equal 190 GJ/ha and year.

DME can be produced via gasification of salix with an energy efficiency of roughly 54%. The total amount of DME is therefore the total salix yield multiplied with 0.54. The yield of DME is 102.6 GJ/ha and year.

The fossil energy input for this process is 0.059 MJ/MJ of DME produced.

The net yield of DME is 96.5 GJ/ha and year. If DME is produced via the black liquor route the net yield is increased to 116 GJ/ha and year.

The transport distance is calculated using a fuel consumption of 30 liters of diesel equivalents per 100 km. This equals 0.0105 GJ/km. Thus a DME vehicle can travel approximately between 9200-11000 km per year and hectare.

Results
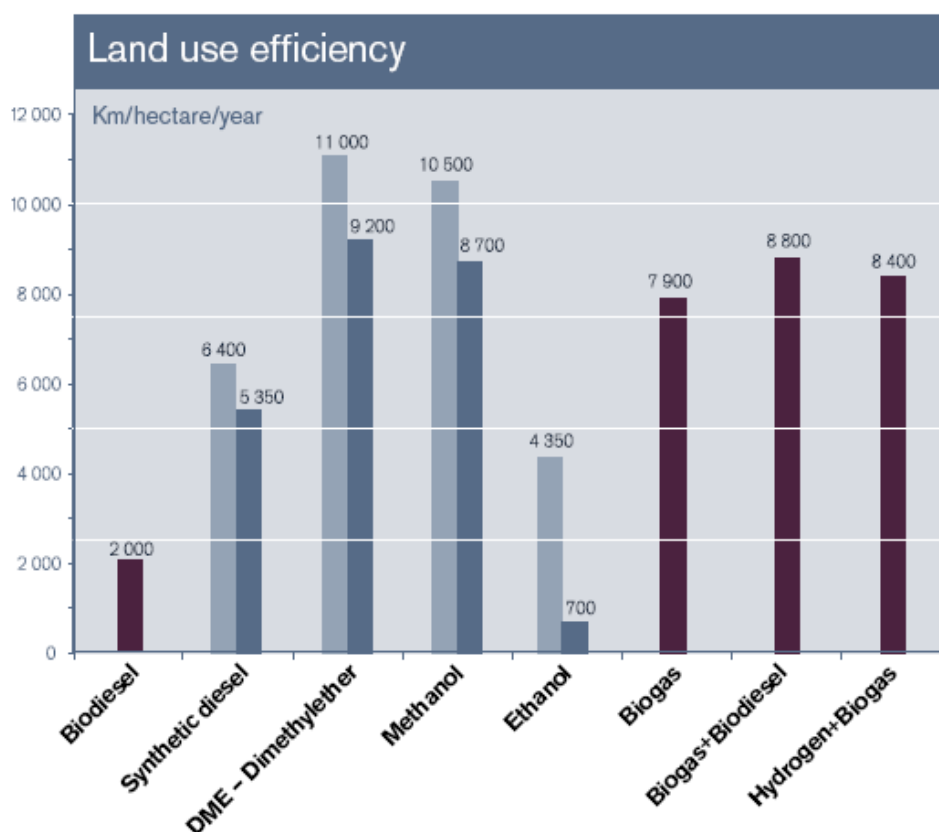Results on the criteria "land use efficiency" are presented in Figures A8.5 and A8.6

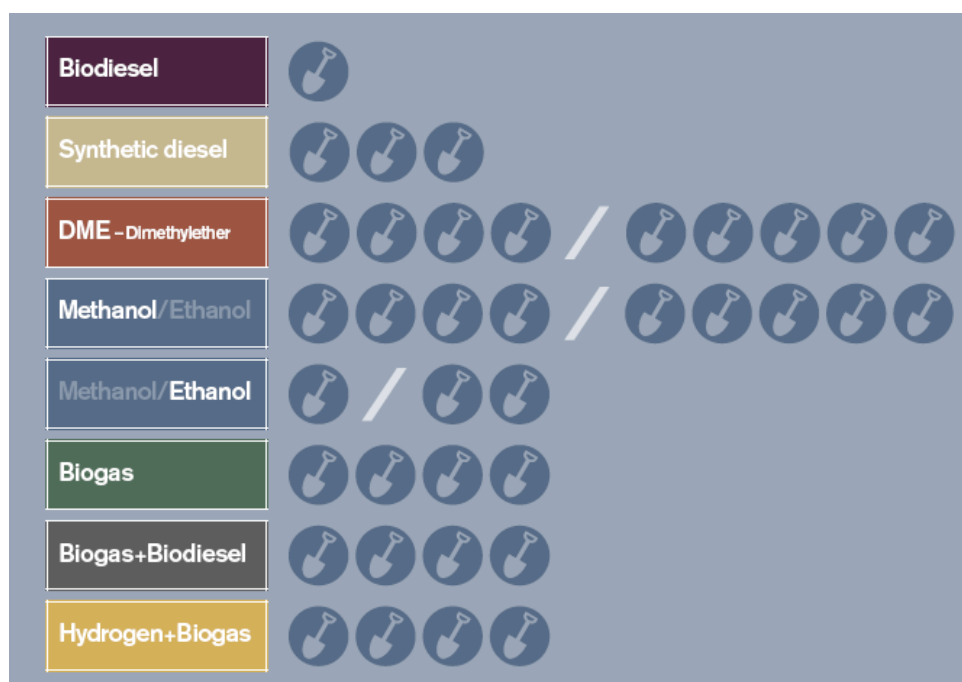**Figure A8.5.** Quantitatively results on the criteria "land use efficiency".

Figure A8.6. Score results on the criteria "land use efficiency". Note that some of fuel alternatives may end up in several intervals since different processes producing the same fuel are analyzed.

DME and methanol based on black liquor gasification receive the highest rating. Harvest yields are high, only small quantities of fossil fuels are required and the fuels have high energy efficiency. Synthetic diesel also benefits from high harvest yields and low fossil fuel consumption; however, its energy efficiency is lower and the selectivity in production is limited. Ethanol receives a low rating because of its limited energy efficiency and in certain instances, high fossil energy requirement. Biodiesel is rated lowest due to low average harvest yields and very high fossil energy use. Biomethane produced via black liquor gasification is not included due to lack of data.

Feedstock availability/fuel potential
The availability of raw material and the choice of production process determine the amount of fuel that can be produced. Some processes can use many different feedstock sources and complete crops, while others are limited to parts of individual crops. According to Edwards et al. (2007), the potential availability of waste wood, farmed wood, and straw in the EU in 2012 will be approximately 700 TWh per year, while that of sunflower oil and rapeseed oil are estimated to 80 TWh per year. The amount of fossil fuel that can be replaced by biomass varies depending on the efficiency of the fuel production process and the end use efficiency. Since biomass potential in the EU in 2012 will not be sufficient to replace all fossil fuels, further initiatives and dedicated measures will be required to increase the proportion. In the longer term, it will be possible to replace fossil fuels in significant quantities provided that the right options are chosen. Importing biomass from regions with better cultivation conditions is a further possibility.

There is no standardized method for measuring feedstock potential since the word "potential" can be defined in several ways. The sustainable potential is the most relevant compared to technical or economical potential. A sustainable supply of biomass produced in a sustainable manner is the ultimate goal.

125

Today the biomass potential in EU is limited and other sectors such as heat and power are already using large parts of the potential. If the EU wants to increase the biofuel production the largest part will be derived from energy crops. The specific crop will depend on the geographical location and the local soil properties. The Nordic region is for example judged to have the largest potential in short rotation coppice such as Salix Willow.

The potential production of biofuels was multiplied with the relative end use efficiency of different fuels. Therefore the actual amount of biogas is higher but the lower energy efficiency of a biogas engine reduces the potential somewhat.

Results
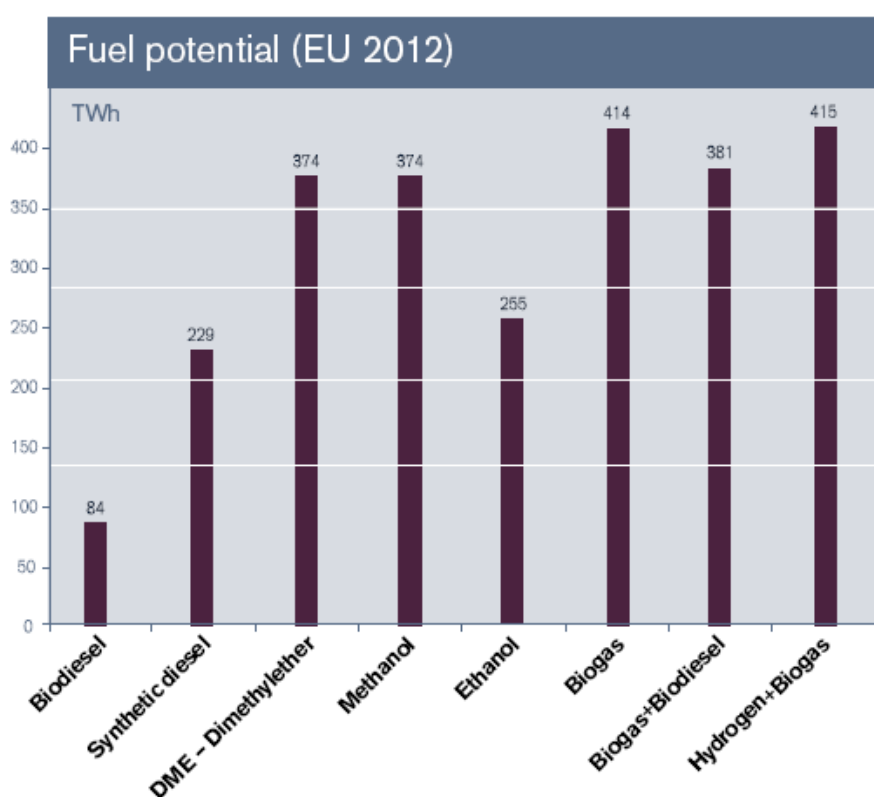Results on the criteria "feedstock availability/fuel potential" are presented in Figures A8.7 and A8.8.

Figure A8.7. Quantitatively results on the criteria "feedstock availability/fuel potential".
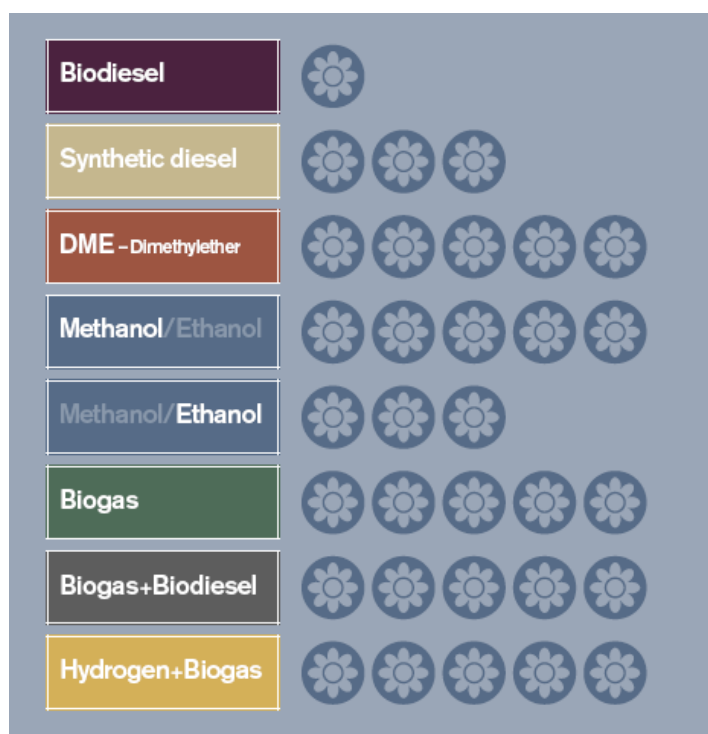
Figure A8.8. Score results on the criteria "feedstock availability/fuel potential".

The 350-420 TWh range is equivalent to approximately 10-12 percent of the predicted demand for petrol and diesel in the EU in 2015. DME, methanol, biogas, biogas+biodiesel and biogas/hydrogen mixture receive the highest rating. Synthetic diesel, DME, methanol, and biogas can all be produced from complete crops, wood feedstocks or other biological materials; however, synthetic diesel has lower energy efficiency and yields a lower proportion of fuel for vehicle use. Household refuse and sewage can also be used in the production of biogas.

Ethanol can be produced from a number of feedstock sources, including waste wood and other biological materials containing cellulose, although at a relatively low efficiency. Biodiesel, which has the lowest rating, is produced from vegetable oils, such as rapeseed oil and sunflower oil. Its availability is limited since rapeseed can only be grown on the same land every fourth or sixth year, while only the oil in the seeds can be used as fuel.

Vehicle adaptation
All fuels do not suit all transport applications due to that the fuels characteristic sometimes causes limitations in the engine performance and/or action range (distance/fuel fill). Some fuel adaptations are also more expensive and complex to develop and produce than others. The assessment includes the effects of various parameters – such as maximum engine performance, increased weight and range between refuelling – on vehicle efficiency. The last of these, for example, may affect vehicle payload. The complexity of adaptation includes factors that necessitate additional fuel storage capacity, and require new and more expensive components, as well as the technology needed to meet future emission standards. As an example, some fuels require more advanced emission control systems than others.

Results
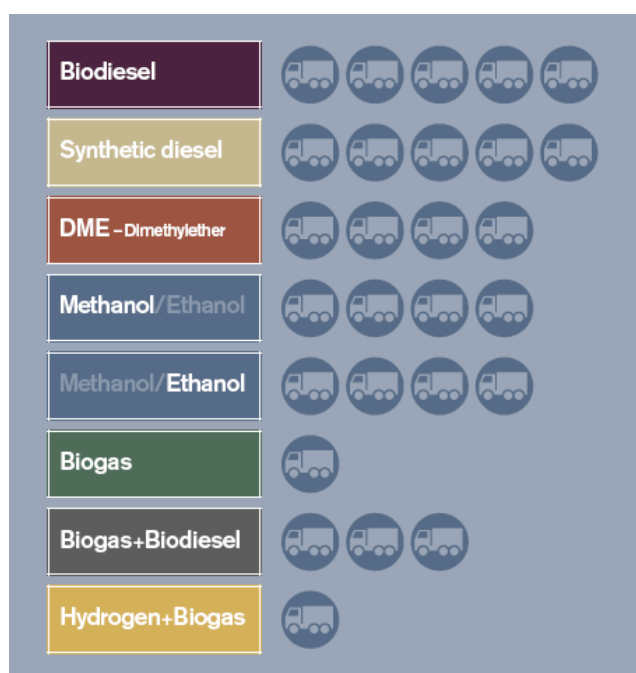Results on the criteria "vehicle adaptation" are presented in Figure A8.9.

Figure A8.9. Score results on the criteria "vehicle adaptation".

Biodiesel and synthetic diesel receive the highest rating. Vehicles powered by these fuels are essentially comparable with conventional diesel fuelled vehicles. However, biodiesel necessitates more service and generates higher nitrogen oxide emissions.

Although the lower energy content of DME reduces vehicle range by approximately 50 percent, the fuel can still be used for long-haul transport. While it requires a unique and advanced fuel system, DME also offers savings in terms of the cost and weight of exhaust silencing and post-treatment systems.

The lower energy content of ethanol reduces the range of the vehicle by 30 percent per tank of fuel. The overall vehicle changes are not very costly or complex.

The lower energy content of methanol reduces the range of the vehicle by 50 percent per tank of fuel.

Although biogas+biodiesel offer maximum engine performance, vehicle range is cut by half if the gas is in liquid form. In addition, two separate fuel systems are required.

Biogas and biogas/hydrogen mixture require an Otto engine, which has a lower energy efficiency compared to diesel engines and power output is limited. The low energy density of the compressed gas limits the range of the vehicle to approximately 20 percent of that of a diesel. Cost and weight are increased by a complex fuel tank system.

Fuel cost
Future costs are difficult to predict due to fluctuations in raw material prices and the rapid pace of technological development. In many cases, the cost of producing a fuel is only a small element of the price to the end user, due to taxes and other charges. Here, the cost of the

particular fuel is compared with that of conventional diesel oil, assuming a crude oil price of USD 70 per barrel (excluding taxes). Comparison is made on energy equivalent basis. This means that over a litre of fuel is required in some cases to obtain the same energy content as a litre of diesel. The results for the same fuel may vary depending on the feedstock/process used. The production cost of alternative fuels from "well to tank" includes the following components: raw material cost, capital expenditure cost (CAPEX) in production plant, operational expenditure cost (OPEX) in production plant, cost for transportation and infrastructure, cost for energy consumption in the distribution chain, and distribution infrastructure investments.

All of these costs are derived from the background data presented in Edwards et al. (2007). The data was updated with the latest information available on energy and raw material cost.

Results
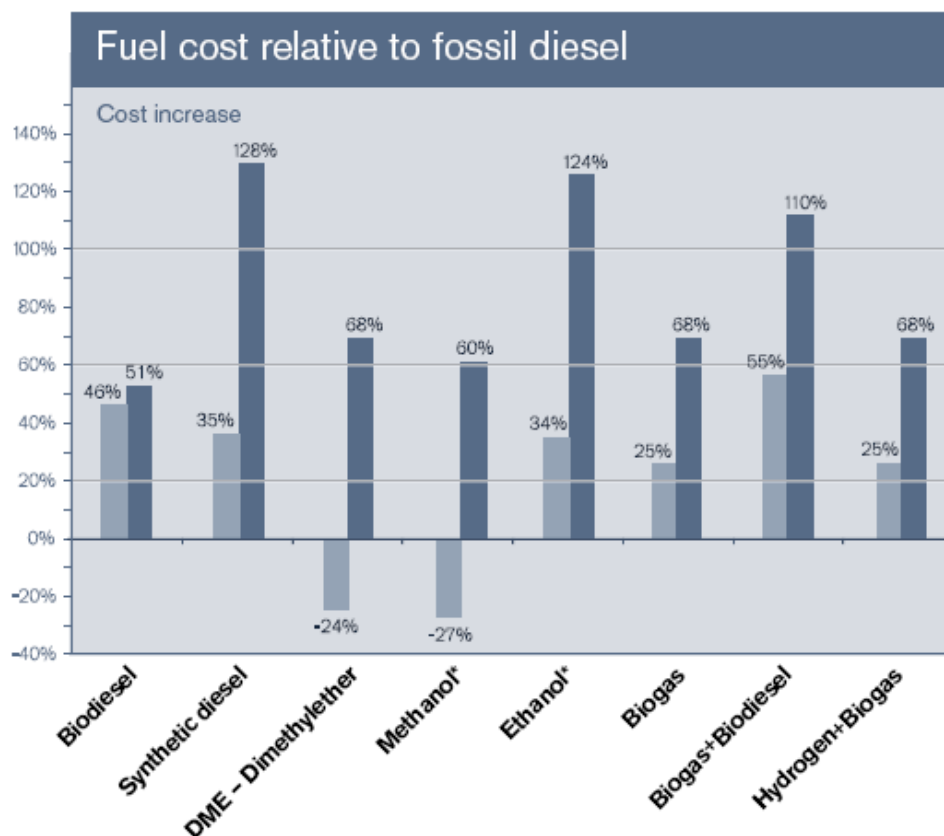Results on the criteria "fuel cost" are presented in Figures A8.10 and A8.11.

Figure A8.10. Quantitatively results on the criteria "fuel cost".

Figure A8.11. Score results on the criteria "fuel cost". Note that some of fuel alternatives may end up in several intervals since different processes producing the same fuel are analyzed.

DME and methanol receive the highest rating. These fuels are already cost-competitive when produced from black liquor; however, production by gasification of forest products or farmed wood is more expensive. Biodiesel is about 60 percent more expensive than conventional diesel. In the case of biogas and biogas/hydrogen mixture, biogas based on waste materials is the most cost effective, mainly due to low feedstock cost.

In the case of biogas+biodiesel, biogas in liquid form is approximately 25 percent more expensive than compressed biogas. Biogas produced by gasification of black liquor is not included in the summary due to lack of data. Synthetic diesel is the most expensive fuel due to high capital costs and the relatively low energy efficiency of production. Ethanol is generally more expensive to produce. Ethanol production from cellulose is the most expensive process.

Fuel infrastructure
Infrastructure is an important criterion in terms of how quickly and easily a new fuel can be introduced and integrated with existing systems. As such, it is often regarded as the greatest challenge to the introduction of an alternative fuel. However, it should be noted that since the infrastructure for conventional fuels is also in need of major investment, infrastructure is a secondary issue in the longer term. This evaluation also takes into account the safety and environmental aspects of handling the fuel within the infrastructure.

Results
Results on the criteria "fuel infrastructure" are presented in Figure A8.12.
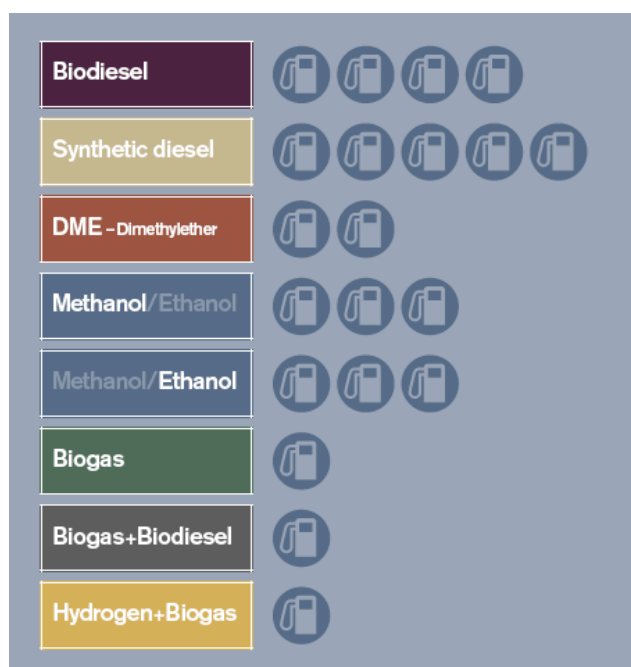
Figure A8.12. Score results on the criteria "fuel infrastructure".

Synthetic diesel falls into the "no changes" category since large amounts of this fuel can be blended into existing diesel fuel while still meeting the current fuel standards/specifications. Biodiesel falls into the "minor changes" category. This is due to the fact that some gaskets have to be replaced in some cases and there is some attention needed in order to deal with stability issues etc.

Ethanol and methanol falls into the category "major changes". Ethanol and methanol needs corrosive resistant materials, flammability precautions and separate infrastructure if used as pure fuels. Alcohols should not be blended into diesel fuel due to safety issues. Methanol and ethanol requires ignition improving additives when used in diesel engines.

DME falls into the category "gas handled in liquid form at low pressure" since it is a gaseous fuel at room temperature and atmospheric pressure. DME is pressurized to approximately 5 bars at room temperature and then it is handled in liquid state. DME requires dedicated infrastructure which is technically similar to LPG (propane and butane).

Biogas and hydrogen falls into the category "gas handled under high pressure or in liquid form at low temperature". Biogas is stored at either 200 bars in gaseous state or at -162 °C as a liquid. Hydrogen is stored at even higher pressures but in the biogas/hydrogen mixture considered in this project it is stored at 200 bars. Hydrogen is a liquid at -253 °C.

These part results are summarized in Section 2.3.4.

Appendix 9. Two cases of mill investments

Results from calculated energy balances in two cases of mill investments

Two cases are presented in Section 6.4.1 for a mill considering replacement of the recovery boiler (1) reference case (invest in a new recovery boiler) and (2) BLGMF case (invest in a black liquor gasification with DME production plant).

The levels of the mill steam demand reflect different possible host mill types, including an integrated pulp and paper mill with a high steam use to a market pulp mill with a low steam use. Within the national Swedish research programmes KAM (Eco-cyclic pulp mill) and FRAM (Future Resource adapted pulp mill), model mills representing both average Scandinavian mills and mills based on best available Scandinavian technology (technology in use at mills today) have been developed (KAM, 2003; FRAM, 2005).

An average Scandinavian integrated fine paper mill has a steam use of 19 GJ/ADt (Delin et al., 2005). Implementation of BLG on a large scale is unlikely to occur before around year 2020. Considering the increasing focus on energy efficiency, we expect current average steam use for a mill will be considered as high steam use a decade into the future. A market kraft pulp mill based on best available Scandinavian technology has a steam use of 11 GJ/ADt (KAM, 2003; Delin et al., 2005b). With state-of-the-art equipment and a higher degree of heat integration, it is possible to substantially further decrease the steam use (see e.g. KAM, 2003; Algehed, 2002). 7 GJ/ADt is chosen to represent a future kraft market pulp mill with a (very) low steam use. This is the steam use at which a kraft pulp mill integrated with a BLGMF/DME plant can satisfy its steam demand using only internal fuel (no need to purchase any external fuel, see Table A9.2). 15 GJ/ADt represents the steam use level at which the mill reference case has a neutral steam balance (i.e. the steam produced by the recovery boiler is equal to the mill steam demand, see Table A9.1). 16 GJ/ADt represents the steam use level at which the mill reference case can satisfy its steam demand using internal fuel only (see Table A9.1). These levels are close to the current levels of an average Scandinavian market kraft pulp mill (between 14 and 15 GJ/ADt, see Delin et al., 2005b) and an integrated fine paper mill based on best available Scandinavian technology (between 15 and 16 GJ/ADt, see Delin et al., 2005a). The mill steam use levels discussed above are valid for mills using softwood as raw material.

Table A9.1presents the calculated energy balances for the mill reference case with a recovery boiler considering five different mill steam demand levels. If the steam produced by the recovery boiler cannot cover the mill's steam demand, additional steam is produced in a boiler fired by falling bark and additional purchased bark or other wood fuels, if required. If more steam is produced by the recovery boiler than is needed at the mill, additional electricity is produced in a condensing steam turbine. As shown in Table A9.1, two of the reference cases considered (16 and 19 GJ/ADt) have a deficit of steam and require a bark boiler whereas two cases (7 and 11 GJ/ADt) have a surplus of steam and produces additional electricity in a condensing steam turbine[39]. In the case where the mill steam demand is 16 GJ/ADt, no purchased fuel is required in the bark boiler. In the case of 15 GJ/ADt steam use, the steam production in the recovery boiler is equal to the mill steam use (and thus no bark boiler is

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[39] Note that the *steam* surplus/deficit is not shown in the table. A steam deficit is indicated by a usage of bark/other wood fuels in the bark boiler and a steam surplus is indicated by electricity production in the condensing turbine.

used and no condensing mode electricity is produced). Gasified bark is assumed to be used as a fuel in the lime kiln.

Table A9.1. Energy balances for the mill reference case.

Mill steam energy use[1]	[GJ/ADt]	7	11	15	16	19
Bark/other wood fuels[2]	*[MW]*					
From debarking		67	67	67	67	67
Usage in lime kiln		35	35	35	35	35
Usage in bark boiler		0	0	0	32	134
Surplus/deficit		**32**	**32**	**32**	**0**	**-102**
Electricity production	*[MW]*					
Back-pressure turbine		89	88	86	91	109
Condensing turbine		38	19	0	0	0
Total production		**127**	**107**	**86**	**91**	**109**

[1] In mill production process (excluding steam energy conversion to electricity in turbine/s).
[2] LHV

Table A9.2 presents the calculated energy balances for the BLGMF cases considering five different mill steam demand levels. Input data for the BLGMF plant, based on Chemrec's pressurised, oxygen blown, high-temperature entrained flow gasification, is taken from Ekbom et al. (2003). A steam deficit is handled by producing additional steam in a boiler fired by falling bark, purge gas and additional purchased bark or other wood fuels, if required. As shown in Table A9.2, there is a need for a bark boiler for all of the considered mill steam demands. When considering a mill steam demand of 7 GJ/ADt, no purchased wood fuel is required. Note that no electricity consumption for the mill is given in Table A9.1. This is because, as previously explained in connection to the pulp/paper production, since this is not affected by what kind of energy and chemical recovery system that is considered and since black liquor gasification is evaluated in comparison to a recovery boiler based system, the mill electricity consumption will be cancelled out. Hence, only the incremental electricity consumption for the BLGMF case (which is the electricity consumed in the BLGMF plant) is given (see Table A9.2). The load of the lime kiln is higher due to different green liquor composition in the BLG case compared to the mill reference case with a recovery boiler Ekbom et al. (2003). Except for the difference in green liquor composition, affecting the load of the chemical recovery cycle, no other changes to the pulping processes are assumed.

Table A9.2. Energy balances for the BLGMF case.

Mill steam energy use[1]	[GJ/ADt]	7	11	15	16	19
Bark/other wood fuels[2]	*[MW]*					
From debarking		67	67	67	67	67
Usage in lime kiln		49	49	49	49	49
Usage in bark boiler		18	156	295	327	429
Surplus/deficit		**0**	**-137**	**-277**	**-309**	**-411**
Electricity production	*[MW]*	*13*	*37*	*62*	*67*	*85*
Incremental electricity consumption	*[MW]*	*40*	*40*	*40*	*40*	*40*
DME production	*[MW]*	*275*	*275*	*275*	*275*	*275*

[1] In mill production process (excluding steam energy conversion to electricity in turbine).
[2] LHV
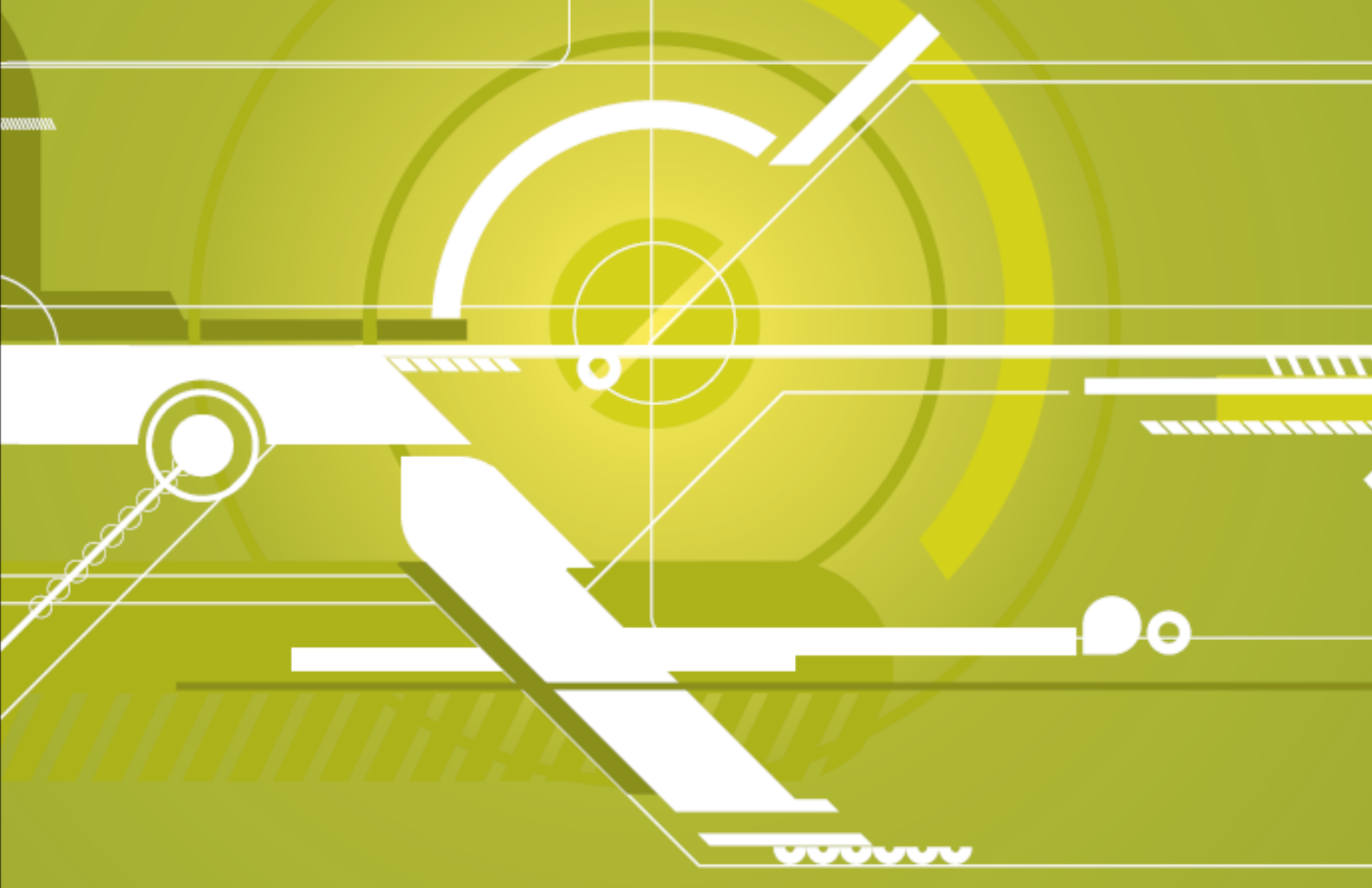
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