Pulp-mill integrated biorefineries: a framework for assessing net CO_2 emission consequences

E. Andersson[*] and S. Harvey. Heat and Power Technology Group, Department of Chemical Engineering and Environmental Science, Chalmers, Sweden

Abstract

There is currently much interest in producing biofuel-based transportation fuels. However, since biofuel is a limited renewable resource, it is important to assess whether such fuels are both produced and used as efficiently as possible. Efficient production can be achieved in the future by integrated biorefinery operations at pulp mill sites, co-producing pulp and biofuel-based energy products. This paper compares production of transportation fuel with other biorefinery options for future pulp mills. The comparison is based on net CO_2 emissions, i.e. accounting for off-site consequences associated with changes in the net flows of electricity, biofuel and biofuel-based transportation fuel entering or leaving the mill. The most important conclusion is that system variables (e.g. assumptions regarding the reference fuel and engine efficiency for future transportation systems) are of decisive importance for the net CO_2 emissions associated with pulp mill biorefinery operations. This paper illustrates such aspects and underlines the importance of a system perspective in process engineering research.

1 Introduction

1.1 Background

There is currently much interest in identifying new ways to produce non-fossil transportation fuels. This is due mainly to the transportation sector's almost total dependency on fossil fuels, the rapidly increasing energy usage in this sector, and concerns about global warming, depletion of non-renewable resources and national security of supply. Besides reducing the need for transportation and promotion of fuel-efficient vehicles, increased usage of biofuel-based transportation fuels is currently considered to be part of the solution to these problems. The European Union has set targets for renewable transportation fuels: at least 2 % in 2005 and 5.75% by 2010 (EU, 2003a). Many studies show that hydrogen production from woody biofuel is more efficient compared to ethanol from wood or hydrogen produced by electrolysis with renewable electricity (EU, 2003b, Hekkert et al, 2004). However, since biofuel is a limited renewable resource, it is important to assess whether transportation fuel produced from biofuel is both produced and used in the most efficient way.

Future pulp mills can be designed to incorporate biorefinery operations, co-producing pulp and biofuel-based energy products. Black liquor is an energy rich biofuel byproduct that is the residue from pulp production. Black liquor also contains chemicals that must be recovered to be reused in the pulping process. In most pulp mills today energy and chemicals are recovered in a Tomlinson boiler where black liquor is burnt to produce steam. Black liquor can also be gasified, generating a syngas

[*]Corresponding author. Tel.: +46 31 7723018, Fax: +46 31 821928

E-mail address: eva.andersson@chemeng.chalmers.se

that can be used to fuel a combined cycle co-generation unit or used as a feedstock for synthesis of transportation fuels such as DME, methanol or hydrogen. It is also possible to extract and export solid biofuel, lignin, from black liquor.

1.2 Objective

The objective of this paper is to evaluate production of transportation fuel, hydrogen and methanol, compared to other future pulp mill biorefinery concepts. The comparison is based on net CO_2 emissions. A framework for evaluation of such emissions for biorefinery concepts is proposed.

1.3 Method

Four biorefinery concepts are evaluated in the paper (see 2.1). Energy and material flows are based on calculated process data for the four biorefinery concepts. All processes considered are assumed to be integrated with the same size and type of pulp mill, i.e. raw material input, pulp output and pulp process electricity and steam demands are identical.

The different concepts are compared on the basis of net CO_2 emissions i.e. accounting for CO_2 consequences on the reference energy system (see Schlamadinger et al., 1997) associated with the net flows of different energy and material streams entering or leaving the mill (i.e. bark, wood fuel, electric power, and transportation fuel). Biofuel-based flows are assumed to substitute fossil fuel based flows on the basis of relevant functional units, i.e. electricity, and unit distance of travel. Consequently, the assumptions regarding the technologies included in the reference energy system clearly influence the results of the calculations. A set of energy market future scenarios, developed by Harvey and Ådahl (2004) was used to define the reference energy system (see Section 2.3).

2 System analysis

2.1 Alternative biorefinery concepts

The different pulp mill biorefinery concepts are all based on a model mill producing 2000 ADt (air dried tonnes) pulp per day. The different concepts considered are:

- Tomlinson recovery boiler (RB) (KAM report, 2001)
- Black liquor gasification with electric power generation (BLGCC) (Berglin et al, 1999)
- Black liquor gasification with methanol production (MeOH) (Berglin et al, 2002)
- Black liquor gasification with hydrogen production (H2) (Andersson and Harvey, 2004)

The pulp mill used for the calculations is KAM2 (**K**retslopps **A**npassad **M**assafabrik, in English: Eco-Cyclic Pulp Mill), a reference mill developed within a Swedish research program. The mill is assumed to generate a surplus of bark and electricity, see Figure 1 (KAM report, 2001).

Performance estimations for the concepts incorporating black liquor gasification are based on a gasification model developed by Berglin et al (2002) that assumes a pressurized oxygen-blown entrained flow gasifier reactor operating at 32 bar, 950 ^oC. The process downstream from the gasifier was modeled using a general chemical process simulation program, HYSYS. The mill energy system and the fuel synthesis unit were assumed to be integrated to optimize energy usage.

Both the Tomlinson boiler and black liquor gasification combined cycle (BLGCC) recovery systems have an excess of electricity and biofuel (bark). When transportation fuel is produced (MeOH and H2 concepts), thermal energy and electric power must be imported to the plant, see Figure 1. We assume that biofuel will be used in a biofuel CHP unit to produce the thermal energy needed and co-generate electric power. Additional electric power must be imported from the grid. **Table 1** summarizes the energy flows for the four biorefinery concepts.

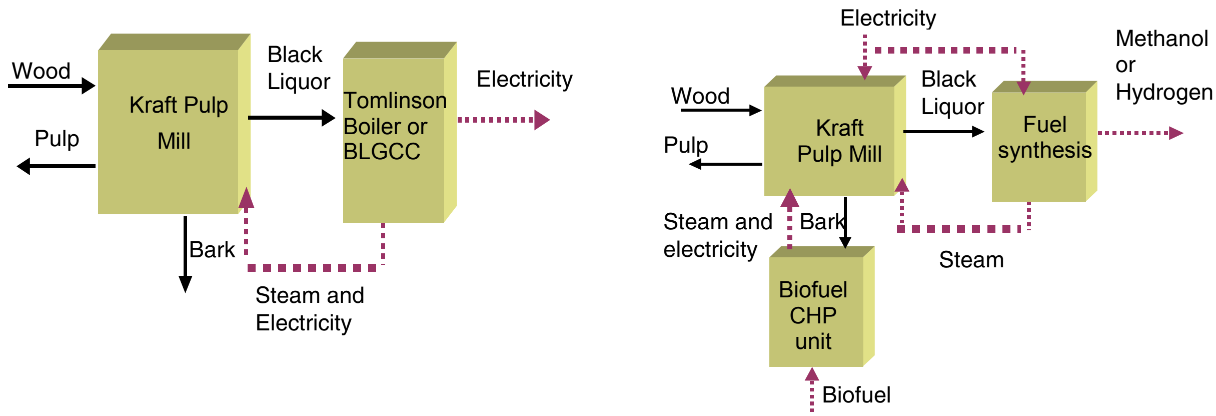

Figure 1 Energy and material flows for different pulp mill biorefinery concepts. The solid arrows represent flows that are the same in the four alternative configurations.

Table 1 Energy flows for the four alternative biorefinery concepts

		RB	BLGCC	MeOH	H2
Power to grid[1]	[MW]	45	86	-55	-57
Transportation fuel	[MW (LHV)]	0	0	271	261
Biofuel export[1]	[MW (LHV)]	32	21	-117	-123

1 A negative sign indicates import to the system

2.2 Assumptions

We assume that the regional demand for electricity and transportation is constant and not affected by the change of supply. This means that electricity or transportation fuel produced at the mill will substitute the same amount of electricity or transportation distance in the reference energy system. We also assume that biofuel is a limited resource, i.e. that the demand for biofuel exceeds supply at reasonable costs. The possible use of low-value excess process heat, for example for district heating, was not considered in this study.

The efficiencies for the different transportation fuel production processes and vehicles used to deliver the functional unit of transportation service in the reference system have, with a few exceptions, been taken from "Well to wheels", a study of a range of transportation fuels and driver trains (EU, 2003b). The CO_2 emission figures are based on the same study. CO_2 emissions are included for production and transportation of the biofuel that is imported and used at the pulp mill, 2.2 g CO_2/MJ biofuel (ÅF Energikonsult,1995). No CO_2 emissions are allocated to the black liquor or bark from the pulp mill process, since these are considered as byproducts. Distribution losses have been included for the transportation fuels produced in the bio-refinery, 3 % for methanol and 18 % for hydrogen (EU, 2003b).

2.3 Net CO_2 emissions evaluation

Energy market future scenarios, developed by Harvey and Ådahl (2004), were used to define the reference energy system for the study. Based on the proposed scenarios, five different reference energy systems were considered in this study, denoted Cases I, II, III, IVa and IVb. Reference values for CO_2 emissions associated with different technologies for production of one functional unit of electricity may be found directly in Harvey and Ådahl (2004). Reference values for transportation services are not included in these scenarios, and corresponding assumptions regarding possible options for providing transportation services in the future were used to compute the reference CO_2 emissions for transportation shown in **Table 2**. For the cases where biofuel is exchanged between the mill and the reference energy system, it is important to define a reference use for biofuel. The values listed in **Table 2** are based on results presented by Ådahl (2004).

Table 2 Reference systems used for calculation of net CO_2 emissions

	I Near future	II Business as usual	III Moderate change	IVa Sustainable A	IVb Sustainable B
Electric power production (g CO_2 /MJ)	Coal (216)	Advanced coal (202)	NGCC (97)	Adv. coal with CO_2–sep (25)	Adv. coal with CO_2–sep (25)
Transportation (g CO_2/km)	Gasoline in today's cars (196)	Gasoline in a hybrid vehicle (140)	Gasoline in a hybrid vehicle (140)	NG-hydrogen in fuel cell vehicles (83)	NG-hydrogen in fuel cell vehicles (83)
Biofuel use (g CO_2 /MJ biofuel)	Oil substitution (68)	Oil substitution (68)	Co-firing in coal power plant (91)	BIGCC (12)	Transportation fuel (44)

Table 2 shows the net CO_2 emissions reduction figures for the different electricity, transportation services and biofuel usages for the different reference energy system cases considered. Case I is assumed to represent the Nordic energy market in the near future. The reference electric power plant technology is a conventional coal-fired unit. Biofuel is assumed to substitute oil in heating applications. Transportation fuel (methanol and hydrogen) produced in the biorefinery is assumed in to be used in fuel-cell vehicles for all reference energy system cases, substituting gasoline in today's cars (Case I). The Case II (Business-as-usual) reference energy system is assumed to correspond to an economy with focus on high economic growth. The efficiency of coal power plants is slightly higher than in Case I. Gasoline is used in hybrid cars with higher efficiency. Biofuel substitutes oil for heating applications (a 90% substitution factor is assumed to account for different boiler efficiencies). Case III (Moderate change) assumes a balance between economic growth and reduction of CO_2 emissions. Compared to Case II, natural gas combined cycle (NGCC) is the reference technology for electricity generation, and the reference usage for biofuel is co-firing in the boiler of a conventional coal-fired power plant (assuming a 100% biofuel to coal substitution factor). Cases IVa and IVb (Sustainable) represent a sustainable society with low CO_2 emissions from the reference energy system. Electricity production is assumed to be CO_2 lean, e.g. produced from coal with CO_2 capture and storage. Transportation services are provided by fuel cell vehicles fuelled with hydrogen produced from natural gas. The reference usage for biofuel is electricity generation in an advanced biofuel integrated gasification combined cycle (BIGCC) unit in case IVa. In Case IVb biofuel is instead assumed to be used for production of hydrogen transportation fuel in a stand-alone gasification and fuel synthesis plant. Results achieved for the H2 pulp mill biorefinery concept with the Case IVb reference energy system are therefore of particular interest, since they can be interpreted as the gain in terms of net total CO_2 emissions for production of biofuel-based hydrogen in a pulp mill biorefinery compared to stand-alone production. It is also important to note the chain effects embedded in Cases IVa and IVb. For example, increased export of biofuel from the mill to the reference energy system in Case IVa results in increased electricity production from biofuel. This in turn results in decreased electricity production from the reference (fossil) electricity generation technology. A similar analysis is valid for increased production of transportation fuel in Case IVb.

3 Results

The results are illustrated in Figure 2 as the net annual reduction of CO_2 emissions assuming full-time full-load operation of the biorefinery. The largest CO_2 reduction corresponds to the Case I reference energy system, since that reference case has the highest CO_2 emissions.

Examining the two biorefinery configurations with a net electricity production (RB and BLGCC) we note that the BLGCC concept, that generates the most electricity, will always achieve the largest reduction of CO_2. Hydrogen production achieves a larger net reduction of CO_2 than methanol production for all the cases evaluated. This is due to more efficient use of energy in fuel cell vehicles resulting in increased gasoline substitution.

Electricity generation at the biorefinery is more valuable from a CO_2 point of view when electricity in the reference system is generated with coal as fuel, without CO_2 separation and storage. However, in the near future case (Case I), with transportation still assumed to be with today's cars, hydrogen production will achieve the largest net reduction of CO_2. If more efficient cars are developed and coal is assumed as the reference electricity generation fuel, without CO_2 separation and storage (Case II), the BLGCC biorefinery concept will achieve the largest reduction. With a more CO_2 lean electricity generation technology, as assumed in Cases III and IV a&b, production of electricity at the biorefinery will not result in so large decreases of CO_2 emissions. Instead production of transportation fuel at the biorefinery (Case III-only hydrogen, Case IV a&b methanol or hydrogen) will achieve the largest CO_2 reduction.

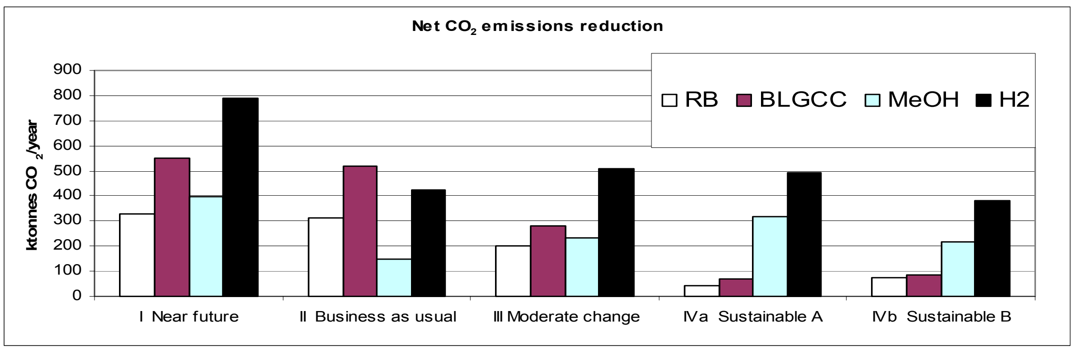

Figure 2 Reduction of CO_2 [ktonnes/year] for the different biorefinery concepts, assuming the five different reference energy systems described in Table 2.

4 Discussion

The net CO_2 emission consequences of the different biorefinery concepts considered depend on the reference generation of electricity and transportation fuel and the use of biofuel. The calculations are based on future efficiencies of different energy conversion technologies and vehicles. Unexpected development of these techniques could significantly change the results. CO_2 emissions can be reduced further if CO_2 is captured and stored in the biorefinery processes. The largest reduction can be achieved if applied to hydrogen production process. This option was not included in this study but should clearly be considered in further work.

Hydrogen and fuel cell vehicles are often assumed to be the transportation system of the future. The results in this paper depend on assumptions regarding the development of fuel cell vehicles and infrastructure for hydrogen distribution. Production of hydrogen in biorefineries is shown to consistently lead to better net CO_2 reduction results than production of methanol. It is however important to note that methanol can be used in today's vehicles and distribution systems with only minor modifications.

It is assumed in this work that the change in supply of electricity, biofuel or transportation fuel does not alter the level of consumption of these services. If the total Swedish production of chemical pulp of 7.5 M ADt/year were to be produced as efficiently as in the KAM mill, the potential hydrogen production from black liquor in Sweden would be able to replace all the gasoline used in the country today. However this would require 11 TWh/year of external biofuel to be imported to the pulp mills. This should be compared to the 64 TWh/year of biofuel (black liquor excluded) used in Sweden's energy system today. Such substantial changes would most probably impact prices and consumption levels substantially and thereby the net CO_2 emission consequences. Such market response effects were however beyond the scope of this study.

5 Conclusions

Hydrogen production from gasified black liquor has the greatest potential for net CO_2 reduction in four of the five biorefinery cases evaluated. This indicates that for many possible future scenarios, hydrogen production from gasified black liquor will be an efficient way to use biofuel. The net impact of the different pulp mill biorefinery options investigated on net CO_2 emissions is extremely dependent on assumptions regarding the reference energy system and its development over time. If improvements (resulting in lower CO_2 emissions) for transportation fuels and vehicles in the reference system are assumed to precede improvements for power generation technologies, then power generation is the most favorable biorefinery option. The most important conclusion is that input variables other than the technical performance of the biorefinery process are of decisive importance for the net CO_2 emissions associated with future biorefineries. The importance of a system perspective in this type of energy research is evident.

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