

# THE VALUE CHAIN FOR BIOMETHANE FROM THE FOREST INDUSTRY

Report from an f3 R&D project

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## PREFACE

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## SUMMARY

This study explores integration of anaerobic digestion (AD) in pulp mills. The AD process converts waste streams into valuable fuel for the transport sector while increasing the overall resource efficiency in the wastewater treatment at the pulp mills. Simulation models and economic calculations are used to evaluate the potential for biogas production in mills producing bleached kraft pulp. This is complemented by a compilation of related studies to cover the total potential for biogas production in the pulp and paper industry.

Considering the available technology for AD and the available substrate streams in a kraft pulp mill, two different concepts have been evaluated:

*Case 1: High-rate anaerobic reactor (UASB) integrated with the external wastewater treatment prior to the activated sludge process.* The evaporator condensate and the alkaline streams from the bleach plant are treated. The activated sludge process is still in operation treating the streams that cannot be treated in the anaerobic reactor and polishing the streams that are treated with AD.

*Case 2: External stirred tank reactor with a retention time of about 20 days.* Primary and secondary sludge are treated together with the evaporator condensate. This concept will also reduce the sludge amount and produce biogas. Though, the substrate is less accessible in the form of microbial sludge compared to organic material in solution. Therefore it is necessary with longer retention times and pre-treatment to reach the potential biogas production.

By assuming a lifetime of 15 years and using an interest rate of 6%, an annuity factor of 0.1 was used to be able to calculate the overall production cost of liquefied biogas (LBG). Both cases look promising with production cost of 83-85 EUR/MWh LBG, i.e. shows a profit corresponding to 34-36 EUR/MWh LBG based on a selling price of 119 EUR/MWh LBG.

Also included in the study was an investigation on the integration possibility for a biomass gasification process to a pulp and/or paper mill. The drying and gasification step of biomass related to the Bio-SNG process was integrated to both a market kraft pulp mill and an integrated pulp and fine paper mill. The size of the Bio-SNG process used in the integration study corresponds to a wood input of 100 MW<sub>wood</sub> or approx. 18 ton dry wood/hr. After gasification and gas cleaning, a product gas is formed containing 68 % of the energy in wood or 68 MW. The composition of the gas is a mixture of CH<sub>4</sub>, CO, CO<sub>2</sub>, H<sub>2</sub> and C<sub>n</sub>H<sub>n</sub>. After further upgrading in a methanation and gas upgrading step, a Bio-SNG stream containing 63 MW is formed. The composition of the SNG product is mainly related to CH<sub>4</sub> (> 94 vol%) and minor amounts of CO<sub>2</sub>, CO, H<sub>2</sub> and N<sub>2</sub> (Heyne 2013 and Gunnarsson 2012).

For the integration possibility, HP-steam from the gasification step is available by cooling the produced gas and thus possible to be used in the turbine in the pulp – and/or integrated paper mill. The benefit from that would be a higher power production and a possibility to get more low pressure (LP) steam to the mill. Only part of the extra produced LP-steam will be used for drying of the biomass used prior to the gasification step. The surplus of energy from the HP-steam from the gas cooling step could therefore be used to cover energy needs in the mills.

The total excess of LP-steam in the kraft pulp mill will be slightly increased with about 0.4-2.0 GJ/ton of pulp as a consequence of the imported HP-steam from the gasification process. If

this extra steam surplus accounting to 0.4 GJ/ton of pulp was sent to a condensing turbine, approx. 24 kWh/ton of pulp or 1 MW more power could be produced.

An average Nordic pulp mill has a power deficit of about 148 kWh/ton of pulp, and by including the net power consumption for the gasification process it will increase to about 199 kWh/ton of pulp. However, if there is a condensing available at the mill, the power production could increase with 98 kWh/ton of pulp.

For the integrated pulp and fine paper mill, the integration with a gasification plant is of higher interest due to a high steam consumption in the paper machine leading to a steam deficit in the mill. By having extra HP-steam to the mill from the gasification process, the supplementary wood fuel used in the bark boiler could be reduced with approx. 10% or 23 ton dry wood fuel per day. However, an average integrated pulp- and paper mill has a power deficit of about 559 kWh/ton paper and by including the net power consumption for the gasification process it has been estimated that the power deficit will increase to about 601 kWh/ton paper.

Comparison between the potential for biogas production via anaerobic digestion (AD) versus gasification of biomass integrated to an average Nordic pulp mill.

Comparison	AD of waste streams	Gasification of biomass	Unit
Production of biogas (CH <sub>4</sub> )	26-27	~490	GWh/year
Additional biomass used	0	~780	GWh/year
Extra wood handling in the mill	0	~20	%

For gasification of biomass to produce Bio-SNG it is considered necessary to build a rather large plant to benefit from economy of scale. To integrate a gasification plant with a normal sized Nordic pulp mill will not reach sufficient high production of biogas but several mills could probably be suppliers of biomass to a larger gasification plant. Some pre-treatment methods for upgrading the biomass to higher energy density and better fuel quality might then be necessary to allow for longer transport distances. Examples of pre-treatment methods are torrefaction, pyrolysis and lignin extraction.

## SAMMANFATTNING

Denna studie undersöker integrering av rötning (anaerobic digestion, AD) i massafabriker. AD-processen kan omvandla avfallsströmmar till värdefull bränslegas för transportsektorn och samtidigt öka den totala resurseffektiviteten i avloppsrening vid massabruken. Simuleringsmodeller och ekonomiska beräkningar används för att utvärdera potentialen för biogasproduktion i bruk som tillverkar blekt sulfatmassa. Detta kompletteras med en sammanställning av relaterade studier för att täcka den totala potentialen för biogasproduktion inom massa- och pappersindustrin.

Med tanke på den tillgängliga tekniken för AD och de tillgängliga strömmarna i en sulfatfabrik, har två olika koncept utvärderats:

**Fall 1:** *High -rate anaerob reaktor (UASB) integrerad med den externa avloppsreningen* före den aktiva slamprocessen. Indunstningskondensat och de alkaliska avloppsströmmarna från blekeriet behandlas. Den aktiva slamprocessen är fortfarande i drift för att behandla de strömmar som inte kan behandlas i den anaeroba reaktorn och polering av strömmarna som behandlas med AD.

**Fall 2:** *Extern omrörd tankreaktor med en retentionstid av ca 20 dagar.* Primär- och sekundärslam behandlas tillsammans med indunstningskondensat. Detta koncept kommer också att minska slammängden och producera biogas. Substratet är dock mindre tillgängligt i dess form som slam jämfört med organiskt material i lösning. Det är därför nödvändigt med längre uppehållstider och förbehandling för att nå den potentiella biogasproduktionen.

Genom att anta en livslängd på 15 år och med en kalkylränta på 6%, används en annuitetsfaktor för att kunna beräkna den totala produktionskostnaden för förvätskad biogas (Liquified Biogas, LBG). Med en annuitetsfaktor på 0.1 ser båda fallen lovande ut med produktionskostnader på 83-85 EUR/MWh LBG, d.v.s. de visar en vinst motsvarande 34-36 EUR/MWh LBG (baserat på ett försäljningspris på 119 EUR/MWh LBG).

I denna studie ingår även att titta på integrationsmöjligheten för en biomassaför gasare med ett massa- och/eller pappersbruk. Torkning av biomassan och förgasning till Bio-SNG integrerades till både ett avsalumassabruk och ett integrerat massa- och finpappersbruk. Storleken på Bio-SNG-anläggningen som används i integrationsstudien motsvarar ett biomassaintag på 100 MW eller ca 18 ton torrtänkt ved/timme. Efter förgasning och gasrening bildas en produktgas som innehåller 68% av energin i ingående ved, eller 68MW. Sammansättningen av gasen är en blandning av CH<sub>4</sub>, CO, CO<sub>2</sub>, H<sub>2</sub> och C<sub>n</sub>H<sub>n</sub>. Efter ytterligare uppgradering i ett metaniserings- och uppgraderingssteg bildas en Bio-SNG-ström innehållande 63 MW.

SNG-produkten består främst av CH<sub>4</sub> (> 94 vol%) och mindre mängder av CO<sub>2</sub>, CO, H<sub>2</sub> och N<sub>2</sub> (Heyne 2013 och Gunnarsson 2012).

Högtrycksånga (HP-ånga) från förgasningssteget bildas genom kylning av den producerade gasen och kan således användas i turbinen i massa- och/eller det integrerade pappersbruket. Nyttan av denna integration blir en högre elproduktion och en möjlighet att få ut mer lågtrycksånga (LP-ånga) till bruket. Endast en del av den producerade extra LP-ångan kommer att användas för torkning av den biomassa som används före förgasningssteget. Energiöverskottet från HP-ångan skulle därför kunna användas för att täcka energibehovet i bruket.

Som en följd av den importerade HP-ången från förgasningsprocessen kommer det totala överskottet av LP-ånga i massabruket öka något, ca 0.4-2.0 GJ/ton massa. Om denna extra ånga (0.4 GJ/ton massa) sänds till en kondensator skulle ca 24 kWh/ton massa eller 1 MW mer el kunna produceras.

Ett genomsnittligt nordiskt massabruk har ett underskott på ca 148 kWh/ton massa. Om elförbrukningen för förgasningsprocessen inkluderas ökar underskottet till ca 199 kWh/ton massa. Om en kondensator finns tillgänglig på bruket kan dock elproduktionen öka med 98 kWh/ton massa.

För integrerade massa- och pappersbruk är integrationen med en förgasningsanläggning av större intresse än för ett avsalubruk då den höga ångkonsumtionen i pappersmaskinen leder till ett ångunderskott. Genom att ha extra HP-ånga till bruket från förgasningsprocessen kan det extra biobränsle som används i barkpannan reduceras med ca 10% eller 23 ton torrt bränsle/dygn. Ett genomsnittligt integrerat massa- och pappersbruk har dock ett elunderskott på ca 559 kWh/ton papper och genom den ökning av elkonsumtion som förgasningsprocessen medför ökar detta elunderskott till ca 601 kWh/ton.

Tabellen nedan visar en jämförelse mellan potentialen för biogasproduktion via anaerob rötning och förgasning av biomassa integrerat med ett nordiskt typbruk.

Parameter	AD-avfallsströmmar	Förgasning av biomassa	Enhet
Produktion av biogas (CH <sub>4</sub> )	26-27	~490	GWh/år
Extra biomassa	0	~780	GWh/år
Extra vedhantering	0	~20	%

För biomassaförgasning krävs en förhållandevis stor anläggning för produktion av Bio-SNG om man vill dra nytta av ekonomiska skaleffekter. Att integrera en förgasningsanläggning med ett massabruk ger inte tillräckligt stor förgasare, men flera bruk kan förmodligen vara leverantörer av biomassa till en större anläggning. Några förbehandlingsmetoder för uppgradering av biomassan som ger högre energidensitet samt bättre bränslekvalitet är då förmodligen nödvändiga för att längre transporter av biobränslet ska kunna accepteras. Exempel på förbehandlingsmetoder är torrefiering, pyrolys och ligninextraktion.

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# 1 INTRODUCTION

Substitution of petrol and diesel with biomethane from lignocellulose has been pointed out as one of the most efficient ways of reducing greenhouse gas emissions in the transportation sector. Biomethane in the form of biogas is already an available product and contributes significantly to the fuel pool in several cities in Sweden. Furthermore, the pulp and paper industry currently has an increased focus on finding new product niches. One opportunity is to develop technically and economically attractive processes for producing biofuels in an existing kraft pulp mill.

Large-scale economically and environmentally sustainable production of biofuels will be a necessary requirement to reach political targets that have been set for the coming ten to twenty years. A significant obstacle for the introduction of biofuels production is that the specific investment costs tend to be prohibitively large at a small-to-medium scale, while the economic risk at large scale tends to be equally prohibitive especially considering the remaining technical risk connected with many new technologies.

One important prerequisite for any large-scale production of biofuels is know-how related to biomass logistics, handling and processing of biomass, and maximizing the use of available by-products and side streams. Such know-how is of course already well-established in the forest industry, especially in the Nordic countries, which produce more than 60% of all pulp produced in the EU. Integrating large-scale production of biomethane within existing sites in the forest industry also promises energy efficiency savings when considering e.g. the use of waste heat.

Another prerequisite is that a biofuel plant needs to be able to utilize multiple feedstocks. A multitude of raw materials are or will be available from forest industry-related activities. With co-feeding of intermediate fuel products derived from different raw materials and pretreatment methods the risks of locking into one feedstock will be greatly reduced, and this will also allow for larger production plants that can benefit from economy of scale.

Anaerobic wastewater treatment is today a mature technology, but it is relatively rare at pulp mills. At a mill there can be many individual subflows of wastewater and the potentials for anaerobic treatment of these subflows vary substantially between them. These subflows normally aggregate into one or a few total wastewater streams, which are treated in wastewater treatment plants. Anaerobic wastewater treatment applied to total wastewater flows results in a reduction of total organic carbon by about 50%. The anaerobic treatment stage yields biogas and reduces the electricity demand of subsequent aerobic treatment stages.

The market for biomethane as a vehicle fuel in Sweden has grown considerably over the last decade, primarily near the largest cities. Development has progressed at a slower pace in less urban areas, including areas in Sweden with pulp and paper mills. Biomethane production at mills would support local initiatives to decarbonize public transport and other logistics. Local markets are nonetheless limiting for biomethane production at mills. Two methods remain in order to transport the product to market: distribution via the natural gas grid or via tank trucks in liquefied form.

## 2 TECHNICAL POTENTIAL FOR PRODUCTION OF BIOMETHANE VIA ANAEROBIC TREATMENT

This study explores integration of anaerobic digestion (AD) in pulp and paper mills. The AD process converts waste streams into valuable fuel for the transport sector while increasing the overall resource efficiency in the wastewater treatment at the pulp mills. Simulation models and economic calculations are used to evaluate the potential for biogas production in mills producing bleached sulphate pulp. This is complemented by a compilation of related studies to cover the total potential for biogas production in the pulp and paper industry.

AD is today applied for a range of industrial and societal waste streams. Integration of AD in the pulp and paper industry wastewater treatment has been investigated since around 1970 and several full-scale treatment processes have been built. The technology has not yet reached a widespread application in this industry, despite several advantages over aerobic treatment. An increasing demand for waste-based renewable fuels like biogas together with the availability of more robust and efficient technologies have, however, led to an increased interest in these solutions.

AD can be integrated as an intermediate step in the wastewater treatment or used for external treatment of waste streams from the wastewater treatment process. The integrated AD step requires a high-rate anaerobic reactor, for example upflow anaerobic sludge bed reactor (UASB), and allows direct treatment of effluent streams from the pulping process. Removal of large particles is necessary prior to this step, and the AD step is usually followed by an aerobic step for odour reduction and additional COD removal. The second alternative – external treatment in an external stirred tank reactor with longer retention times – can be used for primary and secondary sludge and easily separated streams like methanol condensates.

### 2.1 COMPARISON WITH AEROBIC WASTEWATER TREATMENT

The following advantages are commonly reported in the literature (IVL - Svenska miljöinstitutet 2012; Rintala & Puhakka 1994) when introducing anaerobic technology in the wastewater treatment:

- Production of biogas
- Reduced energy use for aeration in the activated sludge process
- Lower excess sludge production
- Less space needed for the installations
- Reduced use of nutrition additives and other chemicals in the wastewater treatment

The main disadvantages are the slow start-up of the process and the sensitivity of the anaerobic process (IVL - Svenska miljöinstitutet 2012; Rintala & Puhakka 1994). The process is depending on stable physical and chemical conditions and may be inhibited by toxic components. The risk of inhibition may, however, be reduced by the increasing knowledge of inhibition mechanisms and process control. It is also possible to store anaerobic sludge to enable a quick start-up after a process failure.

## 2.2 PULP AND PAPER MILLS IN SWEDEN

Pulp and paper is an important part of the Swedish industry and economy. 12 million tons of wood pulp and 11.4 million tons of paper and paperboard were produced in Sweden during 2012 (Swedish forest industry 2012). About 67% of the pulp is classified as chemical pulp; mainly kraft pulp and a few sulphite mills. The remaining pulp is produced by mechanical pulping processes: groundwood, thermomechanical and/or chemothermomechanical. The classifications chemical and mechanical pulp is based on the method for separating the cellulose fibers. In chemical pulping the fibers are separated by a cooking process with chemicals, which also removes the lignin. The mechanical pulping process uses energy intense grinding to tear the fibers apart and over 90% of the raw material remains in the final product, compared to about 40-50% in the chemical pulping. Kraft pulp is here described more in detail since it accounts for most of the pulp production in Sweden and it is the mill type chosen for the techno-economic study.

### 2.2.1 Sulphate/Kraft pulp

Kraft pulping shown in Figure 1 is the most frequently used wood pulping process in the world. The objective of the pulping process is to defibrate the wood by degrading the lignin in the wood. This is performed either as batch or as a continuous process using white liquor which contains the active cooking chemicals hydroxide and hydrogen sulfide ions.

During the cooking, the lignin is extracted from the wood chips, and together with the spent cooking chemicals it forms the so called black liquor. The color of the black liquor is intensely dark, giving the spent liquor its name.

Pulp and black liquor are separated after the cooking process. The pulp stream is usually transported to the bleach plant, and the black liquor is transported to the evaporation plant where the weak black liquor is concentrated. After the evaporation, the concentrated black liquor, (thick black liquor) is burned in the recovery boiler.

In the recovery boiler both chemical and energy recovery takes place. The inorganic chemicals in the black liquor are recycled back to the cooking process. The organics in the black liquor are combusted and high pressure steam is produced that can be used in the process and for power generation. A modern kraft pulp mill often has an energy surplus so that power can be sold to the grid after the process demand for steam and power has been fulfilled.

The regeneration of cooking chemicals is important for the kraft process both from an economical and environmental point of view. In the bottom of the recovery boiler a smelt is formed that dissolves in water to form green liquor. This green liquor leaves the dissolving tank and is transferred to a causticizer where the final cooking liquor (white liquor) is produced. Calcium oxide (CaO) is used in the causticizer which transforms to calciumcarbonate (CaCO<sub>3</sub>) in the causticizer producing white liquor. CaCO<sub>3</sub> (lime mud) leaves the causticizer and enters the lime kiln. In the lime kiln, the CaCO<sub>3</sub> is reburned at high temperatures and CaO(s) (lime) is regenerated during the release of CO<sub>2</sub>. The CaO leaves the lime kiln for reuse in the causticizer.

In order to remove the remaining lignin and increase the brightness of the pulp, the pulp is bleached after the cook. Several different bleaching technologies exist, and the most important involve the use of hydrogen peroxide, chloride dioxide and oxygen as bleaching agents.

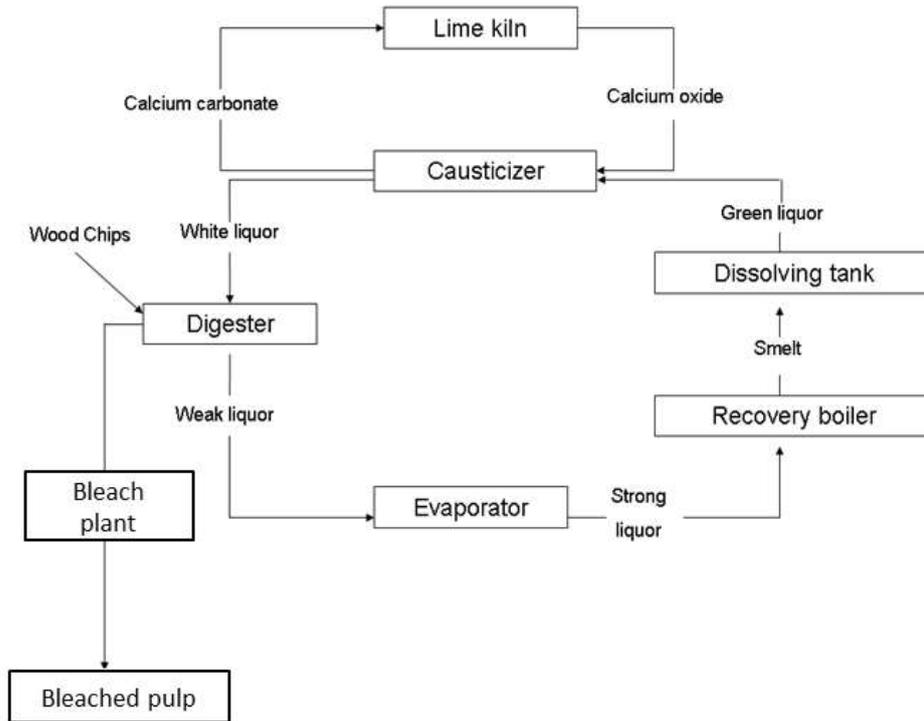


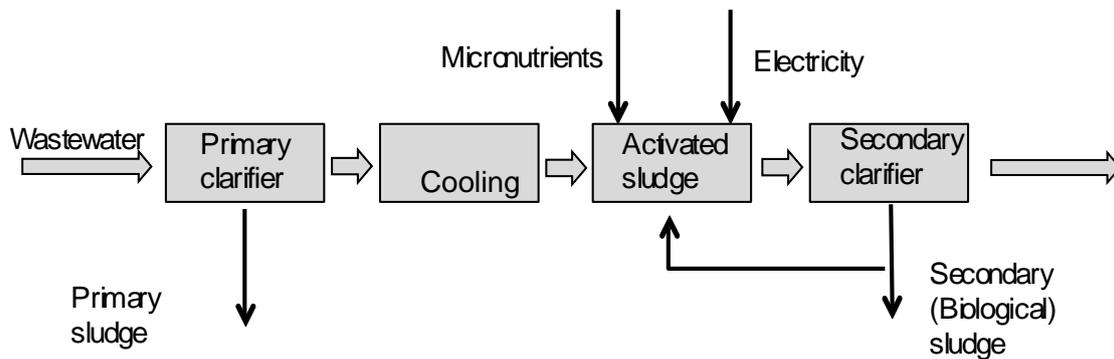
Figure 1. Process scheme for bleached kraft pulp production.

### 2.2.2 Wastewater treatment with activated sludge process

Pulping and especially chemical pulping generates high strength wastewaters that contain wood debris, soluble wood materials and toxic compounds. The wastewater contains large amounts of dissolved organic carbon, measured in terms of chemical oxygen demand (COD) and total organic carbon (TOC). The emissions from pulp and paper mills may bring severe effects on the environment including oxygen deficiency, toxicity and mutagenicity (Rintala & Puhakka 1994). Effluent streams from the pulping process are, therefore, collected and treated in an external wastewater treatment process, which in many cases includes aerobic activated sludge treatment (Figure 2).

A primary clarifier is commonly applied for removing suspended solids, including bark particles, fiber and coating materials. The removed material is called primary sludge, and is often dewatered and combusted.

The wastewater stream is cooled before it enters the activated sludge process. Here, a mix of aerobic microorganisms reduces the dissolved organic carbon. Toxic compounds like chlorinated phenolics and other halogenated organic compounds are also removed. Nitrogen and phosphorus are added to improve the growth and activity of the organisms. The microorganisms require oxygen and the aeration of the ponds consumes large amounts of electricity. The process generates a biological sludge that settles in the following after-sedimentation step. A fraction of the sludge is recycled to the sludge process and excess sludge is removed. The biological sludge is dewatered and deposited in a suitable way. A final flotation or sedimentation step is sometimes added to remove even more pollutants.



**Figure 2. External wastewater treatment in pulp and paper mills.**

### 2.3 ANAEROBIC DIGESTION

Biogas is produced in a process called anaerobic digestion (AD), where microorganisms degrade organic material and convert it to methane ( $\text{CH}_4$ ) and carbon dioxide ( $\text{CO}_2$ ). This process can handle a range of waste streams and reduce their climate impact while energy and nutrients are recycled. The material/waste used to produce biogas is called substrate.

The conversion process can be divided into several steps: hydrolysis, acidogenesis, acetogenesis and methanogenesis. The steps involve different consortia of microorganisms. It is crucial that there is a balance between all the microorganisms and that all steps are well functioning. The organisms in the methanogenesis are easily affected by changes in process conditions or the presence of toxic substances. Reduced methane production rate or failure of the process is called inhibition.

#### 2.3.1 Process conditions

The microorganisms are depending on suitable process conditions like pH, temperature, alkalinity, concentration of volatile fatty acids, and availability of nutrients (Chen et al. 2008; Yadavika et al. 2004). Optimal conditions vary between different microorganisms and the process can therefore be divided into different reactors to increase the efficiency. The process temperature can be around 35-38°C (mesophilic organisms) or around 49-55°C (thermophilic organisms). pH 7 is suitable for the methanogens, while a slightly lower pH suits the microorganisms in the initial step of the AD. The alkalinity is a measure of the buffering capacity of the AD system, which is important to keep a stable pH. The organic loading rate of substrate is also an important process parameter. A high loading rate can cause imbalance between the microorganisms, e.g. due to inability of the methanogens to process all the intermediates, leading to inhibition caused by high concentrations of the intermediates (Chen et al. 2008). This can be seen as an increasing concentration of volatile fatty acids. The loading rate is usually measured in kilograms of volatile solids per day, and reactor volume or chemical oxygen demand (COD) per day and reactor volume.

Toxic components can also cause inhibition (Chen et al. 2008). A range of toxic components are present in wastewater from the pulping process, especially in streams from the bleaching steps, e.g. chlorinated phenols. The possibility to treat different streams will be further discussed in section 2.5.

The microorganisms in the AD process can however adapt to different conditions, even toxic components, if they are given time. It is therefore important to find a suitable type of inoculum, a starting culture, for an AD process. The inoculum should preferably be adapted to the substrate/stream in the matter of a fast start-up process. Hopefully, the anaerobic microorganisms can be adapted to handle a range of streams and toxic components originating from the pulp and paper industry. They are already today adapted to some of them.

A well-balanced mixture of nutrients may enhance the process efficiency in AD (Nges & Björnsson 2012). The ratio between carbon and nitrogen is often mentioned and so is addition of crucial micronutrients. The nutrient mixture can be achieved by mixing different substrates, denoted co-digestion. This may be a challenge in wastewater streams from the pulp and paper industry since they contain a rather unbalanced mix of nutrients and there may not be any suitable substrates for co-digestion. Commercial nutrient mixtures can be used for balancing the process, though it comes with additional costs.

The performance of the AD process is measured in the rate of degradation of the substrate and the extent of degradation and conversion to biogas. This in turn decides how much substrate that can be treated in the biogas production facility and how much gas that can be produced. The rate and extent of the degradation depends both of the process performance and the substrate type. The substrate is never fully degraded in a commercial facility since this would require extremely long retention times. The retention time in the reactor for the microorganisms is denoted solids retention time and that for the wastestream is denoted hydraulic retention time. Easily degradable and diluted substrates can be treated in a high-rate AD reactor with high solids retention time and low hydraulic retention time. Less degradable or more complex substrates are treated in a stirred tank reactor with longer hydraulic retention times.

### **2.3.2 Pre-treatment**

Pre-treatment of the substrate before AD can increase the rate and extent of substrate degradation and ensure stable process conditions. Grinding, dilution and heating are commonly applied, but more technically advanced pre-treatments are sometimes used (Carlsson et al. 2012). Pre-treatment of waste streams from a pulping process could include: dilution, pH adjustment, cooling, addition of nutrients and removal of toxic components or suspended matter. Pre-treatment of primary or secondary sludge is often necessary since the microbial sludge structure is difficult to degrade. This includes a range of methods (Wood et al. 2009). Comprehensive pre-treatment is expensive and must of course be evaluated against the benefit.

### **2.3.3 Technologies**

The choice of anaerobic reactor technology depends on the type of waste stream. High-rate reactors are suitable for relatively diluted streams with easily accessible substrate while stirred tank reactors with longer retention times are suitable for more concentrated streams with more complex substrate material (e.g. sludge).

High-rate reactors enable treatment of large quantities of wastewater. They are designed to retain viable anaerobic sludge, and thus decoupling the hydraulic retention time from the solids retention time. It is important that the reactor design enables sufficient contact between the wastewater and the anaerobic microorganisms, and that metabolic end products can be effec-

iently removed. The up flow anaerobic sludge bed (UASB) reactor and modified versions of it are the most widely used high-rate reactors.

Sludge from the aerobic treatment may also be treated with AD. Biological sludge requires longer retention time since this type of substrate is more difficult to degrade. Stirred tank reactor is the most common reactor for this type of substrate.

#### *Stirred tank reactors*

A simple stirred tank reactor is the most common type of biogas reactor, basically a tank providing an anaerobic environment, temperature control and stirring. Substrate is semi-continuously fed in to the reactor and microorganisms and degraded substrate is removed, whereas the biogas is collected at the top of the tank. The treated material is retained in the reactor on average a few weeks. The retention time depends on how fast the substrate can be degraded and converted to biogas. In the most basic setups all the reaction steps in AD takes place in the same reactor. There are also process solutions with several reactors, each optimized for a certain step of the process.

#### *Continuously stirred tank reactors and contact reactors*

The continuously stirred tank reactor was one of the first versions of the high-rate reactors. The efficiency of this reactor type is compromised by washout of the microorganisms at high loading rates. A step for separation and recirculation of microorganisms was later added. This type of setup is called a contact reactor. None of these reactors can compete with the UASB reactor in terms of loading rates and degradation efficiency.

#### *Anaerobic filter*

The anaerobic filter reactor was created to increase the reaction speed and COD reduction by increasing the contact between microorganisms and substrate. The design was simple. The reactor was filled with a material that provided a surface that the microorganisms could attach to, e.g., gravel. This is also for most applications an outdated technology.

#### *Up flow anaerobic sludge bed reactor (Lettinga 2009)*

UASB reactors are commonly applied in wastewater treatment. The reactor design creates aggregation of microorganisms into granules, which are spherical particles with a size distribution between 0.5 and 2 mm. As the wastewater flows from the bottom to the top of the reactor, the bed is partly expanded. Some granules continue upwards together with the stream of wastewater and the produced biogas. The top of the reactor is designed to recirculate the solid granules back into the reactor, which is achieved by baffles. It is also the design of the recirculation system that allows formation of compact granules. Finally, the biogas is separated from the wastewater by a gas collection unit in the center of the reactor.

Compared with the aforementioned reactors, the UASB reactor can handle higher organic and hydraulic loading rates (Rajeshwari et al. 2000). The hydraulic retention time for UASB reactors vary between 7 and 30 hours (Pokhrel & Viraraghavan 2004). It is commonly applied for treatment of large wastewater flows. It is, however, necessary to remove suspended solids from the waste stream before the UASB reactor, which usually is the first treatment step of the wastewater treatment in the pulp and paper mill.

The energy use in the UASB reactor is low since no mechanical mixing is needed. The flow of the wastewater stream together with the rising gas bubbles provides mixing. Process control like pumping will however require electricity.

The UASB reactor concept has been modified into a range of different reactors. Expanded Granular Sludge Bed (EGSB) uses a high flow rate to achieve a slight expansion of the bed. The high flow rate comes from an increased height to diameter ratio in the reactor design and/or by recirculation of feed. EGSB is the most commonly used high-rate reactor besides the original UASB reactor. Compared to UASB, it can handle a wider range of wastewater concentrations and it is more resilient to toxic components (Tauseef et al. 2013). It is, however, more sensitive to suspended particles. Internal circulation (IC) reactor is yet another version of UASB reactor. The reactor includes two different stages of biogas separation and internal effluent circulation. This design is practically working like a regular UASB in series with a fluidized bed reactor, enabling even higher loading rates than both the original UASB and the EGSB reactors (Tauseef et al. 2013). The anaerobic migrating blanket reactor and anaerobic baffled reactor are compartmentalized by baffles. They function like a series of UASB reactors. This may create phase separation, i.e., that the different reaction steps takes place in different parts of the reactor. Phase separation often increases the process efficiency since the different anaerobic consortia have different optimal operation parameters. The phase separation effect may also be achieved by connecting several reactors in series.

#### *Fluidized bed reactors*

Fluidized bed reactors are similar to UASB reactors, but the microorganisms are attached to suspended carriers, e.g. sand or alumina. The carriers are kept in the reactor by some type of mechanical barrier and suspended by rapid flow of wastewater, gas flow and/or stirring. A fluidized bed reactor has a bed expansion of 25-300% while an expanded bed reactor only expands 15-25% (Tauseef et al. 2013). This process has been under development phase for some time and the method has to overcome several challenges before it is suitable for large scale application.

#### *Anaerobic membrane reactors (AnMBR)*

AnMBR's are mainly in a research phase where the reactors are being scaled up from lab to pilot scale (Stuckey 2012). AnMBR's use a membrane to retain the anaerobic microorganisms and also remove solid particles. The membrane can be external or placed within the reactor. Several combinations of AD reactors and membranes are being evaluated: pressure driven external cross-flow membrane, vacuum driven submerged membrane and sequential membrane reactors.

The technology has shown impressive results in lab scale trials, where COD removal up to 98% has been achieved at HRT:s down to 3 h (Stuckey 2012). According to Liao et al. (2006), membrane fouling and reduced flux are the main development issues for this technology. More research on the mechanisms for membrane fouling and the strategies to reduce this effect is needed to achieve stable operation. Moreover, the membrane separation technology has higher costs for investment and operation than granular sludge reactors (Dereli et al. 2012) and the technology is in need of further development before a widespread application of full-scale reactors is realistic.

### *Hybrid reactors and high-rate anaerobic/aerobic systems*

Hybrid reactors with elements from several reactor types have been created to overcome the difficulties in high-rate AD. Many hybrids are a combination of the UASB reactor and anaerobic filter or film, and the purpose is to increase the retention of microorganisms.

The high-rate anaerobic systems can also be combined with high-rate aerobic systems to achieve an overall compact and efficient process. This option will, however, not be covered here since it is assumed that the aerobic aftertreatment can be achieved in the aerobic activated sludge process that already is available in many wastewater treatment facilities.

#### **2.3.4 Upgrading**

The raw biogas consists of around 65% CH<sub>4</sub> and 35% CO<sub>2</sub> and some impurities, e.g. hydrogen sulphide. Raw gas can be combusted for production of heat and power. The gas can also be upgraded by removal of CO<sub>2</sub>, impurities and water. This is commonly achieved with physical absorption methods like water scrubbing or amine scrubbing, but a range of techniques are available (Ryckebosch et al. 2011). The upgraded biogas can be injected in the natural gas grid and/or used as vehicle gas.

Biogas can be liquefied by cooling to -162°C in order to increase the energy/volume ratio and allow more efficient transport and storage in vehicles. A cryogenic process where the CO<sub>2</sub> is separated during the cooling is under development. Biogas may also be upgraded in the traditional way and then liquefied.

## **2.4 TECHNICAL POTENTIAL FOR BIOMETHANE PRODUCTION**

Several studies of the potential to produce biogas in Swedish pulp and paper mills are available. In a study of the total Swedish potential for production of biogas, waste streams in the pulp and paper industry was estimated to yield 171 GWh of biogas per year, which was based on the amount of biosludge produced in Swedish pulp and paper industry (Avfall Sverige 2008). Ekstrand et al. (2013) performed lab scale of the biogas production potential from different streams and combined with production data for Swedish pulp and paper mills the total potential was estimated to around 70 MNm<sup>3</sup> CH<sub>4</sub>/year (≈700 GWh/year). Magnusson (2012) estimated a technical potential of up to 1 TWh of biogas per year, which was based on the amount of COD emissions that theoretically could be anaerobically treated in a high-rate reactors. The potential for each paper/pulp grade is shown in Table 1 and the highest potential lies within the bleached kraft pulp mills. In this study we therefore base our techno economic calculations on wastewater from bleached kraft pulp. These streams have traditionally been considered difficult to treat anaerobically, which is changing with evolving knowledge on anaerobic processes and technology. Ekstrand et al. (2013), for example, have shown that biogas can be produced from several streams in these types of mills.

**Table 1. Potential biogas production from Swedish pulp and paper mills (Magnusson 2012).**

Pulp/paper grade	Potential biogas production (GWh/year)
Kraft, unbleached	158
Kraft, bleached	198
Sulphite	89
NSSC	19
CTMP	91
TMP	156
GWP	44
RCF	94
Paper/board	109
Tissue	12
<b>Total</b>	<b>971</b>

An environmental and technoeconomic evaluation of integration of AD in the wastewater treatment process is available in a study on the pulp and paper mills Obbola and Rockhammar (IVL 2012). The results indicate that the environmental performance is improved; mainly the climate impact is reduced because the use of chemicals and electricity may be reduced with AD. It is also concluded that the cost for an AD (Table 2) process cannot be motivated by the achieved benefits when taking operating and investment cost into account. However, AD could be an advantageous alternative when the capacity of the wastewater treatment has to be expanded. The value for the produced biogas in Obbola and Rockhammar is based on a use in internal processes while in our study the value is calculated for upgraded biogas for vehicles. Upgraded gas for vehicles is more valuable though it also requires additional investments.

**Table 2. Technoeconomic results for integration of AD in the wastewater treatment of two Swedish pulp mills (IVL 2012).**

Location	Rockhammar	Obbola
Mill type	CTMP pulp	Liner; kraft pulp; recycled fiber pulp
Pulp production rate (kt /year)	78	396; 223; 174
Streams to AD	All streams	Streams from recycled fibre pulp
Flow of organic material to AD (tCOD/d)	14.9	5.9
Biogas production (MWh/year)	7,200	3,100
Reduced operating cost (MSEK/year)	2	1
Investment cost (MSEK)	53	50
Capital cost for 6 year depreciation and 6 % interest (MSEK/year)	10.04	10
Capital cost minus operating cost (MSEK/year)	8.04	9

## 2.5 STREAMS SUITABLE FOR ANAEROBIC DIGESTION

This section explores the possibility to treat wastewater streams from pulp and paper mills. Treatment of streams from TMP and CTMP are most suitable for anaerobic digestion and has been extensively tested and applied in full scale, whereas anaerobic treatment of streams from sulphate/kraft pulp is in a development phase.

The main part of the wastewater loads in pulp and paper mills originate from pulping and bleaching processes. The wastewater contains wood components and chemicals added in the pulping and bleaching process. The possibility to treat components anaerobically is decided by their degradability and toxicity to anaerobic microorganisms. The degradability decides the possible COD reduction and biogas production. Degradable components are e.g. methanol, cellulose and hemicellulose. Low molecular mass lignin is degradable while more complex lignin is not. Toxic components include wood extractives (lignin, resin acids, long chain fatty acids) added chemicals (e.g. sulphur) or a combination of these two (e.g. chlorinated lignin). The toxicity/inhibitory effect of components may vary from a reduced biogas production to complete process failure. The toxicity is increasing with concentration and may be handled by dilution with water, mixing with other streams or by applying pre-treatment methods. The anaerobic microorganisms may degrade toxic components containing chlorine or sulphur, though at critical levels they will inhibit the process.

Waste streams can be divided into streams that can be transported and streams suitable for internal wastewater treatment. The transportable streams are more concentrated and include methanol condensates, and primary and secondary sludge. They can be transported to another facility for treatment at reasonable cost and energy use. The diluted wastewater streams have to be treated at the site in high-rate anaerobic reactors, for example, debarking wastewaters and streams from the bleach plant.

Literature values for biogas production potential are given in different units, and it is important to differentiate  $Nm^3 CH_4/ton COD$  from  $Nm^3 CH_4/ton COD$  *reduced*, where the first refers to biogas production per total amount of COD in the stream, and the second refers to biogas production per kg COD reduced in the anaerobic treatment.

### 2.5.1 Debarking wastewaters

Debarking is necessary in all pulping processes. The debarking effluents contain wood components that inhibit the anaerobic microorganisms in sufficient concentrations: resin acids, LCFA, volatile terpenes and tannins (Ekstrand et al. 2013). Anaerobic treatment is possible though sometimes pre-treatment like dilution or autooxidation at high pH is necessary. Auto-oxidation followed by UASB reactor has been shown to reduce COD by 40% (Field 1999). According to Rajeshwari et al. (2000) the COD may be reduced by 44-78% with anaerobic treatment. The biogas potential in lab-scale is on average  $0.71 Nm^3 CH_4/kg COD$  (Ekstrand et al. 2013).

### 2.5.2 Sulphate/Kraft pulping

Streams from production of bleached sulphate pulp has up until recently been considered too difficult to treat anaerobically, i.a. due to the toxic sulphate and chlorine components. This is however under reconsideration since the knowledge of AD evolves. The possible COD reduction and biogas production for effluents from kraft pulp mills is given in Table 3.

**Table 3. Potential for COD reduction and biogas production when treated with anaerobic digestion (AD).**

Stream type	COD red.	Biogas Potential	Reference
Debarking wastewater	40-78 %	135 Nm <sup>3</sup> CH <sub>4</sub> /ton COD reduced*	(Rajeshwari et al. 2000; Ekstrand et al. 2013)
Kraft evaporator condensate	70-95 %	200-360 Nm <sup>3</sup> CH <sub>4</sub> /ton COD reduced	(Rintala & Puhakka 1994; Stuckey 2012; Värmeforsk 2011)
Alkaline bleaching effluents	30-50 %	300 Nm <sup>3</sup> CH <sub>4</sub> /ton COD reduced*	(Rintala & Puhakka 1994; Ekstrand et al. 2013)
Primary sludge	-	300 Nm <sup>3</sup> CH <sub>4</sub> /ton TS	(Värmeforsk 2011)
Secondary sludge	-	150 Nm <sup>3</sup> CH <sub>4</sub> /ton TS	(Värmeforsk 2011)

\*Recalculated from literature values given per ton COD assuming 60 % respectively 50 % COD reduction for debarking waste water and alkaline filtrate, respectively.

#### *Kraft evaporator condensate*

Kraft evaporator condensate (KEC) has been shown to be anaerobically degradable with very high biogas reduction potential, though in some cases inhibitory to methanogenic activity (Ekstrand et al. 2013). Sulphur compounds are one possible cause for the inhibition. The condensate has a high methanol content, which is easily degraded in AD.

The COD reduction of KEC streams is well studied in different reactor types. COD reductions from 70% to above 95% and biogas production between 200 and 360 Nm<sup>3</sup> CH<sub>4</sub>/ton COD reduced has been recorded for UASB and AnMBR reactors (Stuckey 2012; Rintala & Puhakka 1994; Badshah et al. 2012). The potential was measured to 0.25 Nm<sup>3</sup> CH<sub>4</sub>/ton COD in long-term biogas potential batch trials (Värmeforsk 2011). This corresponds to 294 Nm<sup>3</sup> CH<sub>4</sub>/ton COD reduced, assuming that there was about 85% COD reduction in the experiment. Ekstrand et al. (2012) showed a biogas potential of 184 CH<sub>4</sub>/ton COD, corresponding to 220 CH<sub>4</sub>/ton COD reduced with the aforementioned assumption.

#### *Streams from bleaching*

Effluent streams from the bleaching contain the main part of the COD. Effluents from chlorination and alkaline extraction stages are toxic to the methanogens, but still possible to treat anaerobically with appropriate pre-conditioning. Chlorinated organics and resin acids are the inhibitory components in these streams, even at low concentrations. The strategies for reducing the toxicity include dilution with water or mixing with condensates. Gradual adaption of the anaerobic microorganism may also reduce the inhibitory effects and increase the COD degradation. Anaerobic degradation of chlorophenols is possible and a combination of anaerobic and aerobic treatment can reduce the concentration of these compounds to acceptable levels (Savant et al. 2006; Schnell et al. 2000).

Ekstrand et al. (2013) showed that effluents from alkaline ECF bleaching steps may be suitable for anaerobic treatment while effluents from acid bleaching steps showed toxicity to the meth-

anaerobic activity, also when mixed with other streams. Anaerobic treatment of alkaline effluents from bleaching stages has also been reported by (Yu & Welander 1996). The study performed by Ekstrand et al. (2013) also indicated that alkaline effluents from hardwood processes are more suitable for AD than those from softwood processes. The proposed explanation is that hardwood effluents contain higher levels of easily degradable low molecular TOC and less inhibiting compounds like lignin and resin acids. The same difference between softwood and hardwood has been shown for effluents from sulphite and chemi-thermo-mechanical pulping process (Yang et al. 2010).

COD reductions between 30-50% have been achieved in lab scale trials (Rintala & Puhakka 1994). For the alkaline ECF effluents a biogas production of up to 150 Nm<sup>3</sup> CH<sub>4</sub>/ton COD has been achieved in lab scale trials (Ekstrand et al. 2013).

### *Primary and secondary sludge*

Primary sludge, also known as fibresludge, is removed in the primary clarifier and consists of fibers, bark and sand. The sludge is commonly dewatered to 50-70% dry matter and combusted for internal energy recycling. It can also be used for mixing into construction soil. Primary sludge has a biogas potential of about 320-330 Nm<sup>3</sup> CH<sub>4</sub>/ton wet substance, WS (Värmeforsk 2011). This can be converted to about 300 Nm<sup>3</sup> CH<sub>4</sub>/ton dry substance, DS, since the WS content is 70-95%.

Secondary sludge, also known as biological sludge is the surplus sludge produced in the activated sludge process. It consists of the aerobic microorganisms that degrade and consume the COD in this process step. Secondary sludge can be dewatered to only about 5-15% dry matter because the water is bound inside and around the cells. It can be used for construction soil or combustion. The sludge is usually not suitable as soil improvement since it contains heavy metals and other pollutions. Secondary sludge has a biogas potential of approx. 150-200 Nm<sup>3</sup> CH<sub>4</sub>/ton WS (Värmeforsk 2011). This can be converted into around 150 Nm<sup>3</sup> CH<sub>4</sub>/ton DS, with an assumed WS content of 65-80 %.

### **2.5.3 Thermomechanical pulp**

Waste streams from thermomechanical pulp (TMP) are suitable for AD and anaerobic treatment has been introduced in full scale in pulp and paper mills. The main part of the effluents is possible to treat using AD with COD reduction levels between 60-87% (Rintala & Puhakka 1994).

Secondary (biological) sludge from TMP is possible to treat in AD and has the same biogas potential of around 150 Nm<sup>3</sup> CH<sub>4</sub>/ton DS, same as that from chemical pulping. The primary sludge (fibresludge) is not suitable for anaerobic digestion due to high concentrations of lignin.

### **2.5.4 CTMP**

Effluents from chemithermomechanical pulping (CTMP) contain more toxic components than those from TMP. Though they are still possible to treat in AD and COD reductions up to 60% is possible (Rintala & Puhakka 1994). Several full-scale applications are in operation.

### **2.5.5 Recycled fibre pulp**

Effluents from recycled fibre pulp (RCF) can be treated with AD with 70% COD reduction (Magnusson 2012).

### **2.5.6 Paper and board**

Effluents from paper and board making are possible to treat in AD considering toxic effects. These streams are, however, often too diluted to be suitable for AD (Rintala & Puhakka 1994).

### **2.5.7 Societal waste streams for co-digestion**

Biogas production facilities at pulp and paper mills may also collect external waste streams from society, such as municipal sewage sludge or municipal solid waste. The purpose of co-digestion is to improve the mixture of nutrients available for the microorganisms. This, in turn, may improve the overall biogas production so that more biogas is produced when the substrates are mixed than when they are digested separately. Waste streams from pulp and paper are often poor in nutrients and could benefit from codigestion. Biogas production facilities are, like any other production, based on economies of scale. So by importing substrate the overall economy for investing in a biogas reactor might improve. The drawback of using external substrates is that the biogas yield from them is lower when co-digested with primary or secondary sludge from the pulp and paper (Värmeforsk 2011). Moreover, the possibility to use of the residue from AD as fertilizer may be compromised when the toxic components from pulp and paper is introduced in the biogas process. Despite these drawbacks the co-digestion option may be a way to improve the economy for AD in pulp and paper mills.

However, in many regions there is already competition for available substrates with high biogas potential, with increasing prices for substrates that used to be free. Anaerobic treatment of municipal sewage sludge is widespread and there is a limited amount of sludge available for use in AD in pulp mills.

## **2.6 EFFECTS ON THE WASTEWATER TREATMENT PROCESS**

This section covers the improvements in the external wastewater treatment that can be achieved by including a high-rate anaerobic reactor.

The commonly applied aerobic activated sludge process consumes a lot of electricity for aeration through pumping or stirring. An anaerobic reactor step before the activated sludge process will reduce the COD load in the activated sludge process and thus reduce the electricity use. According to Rintala & Puhakka (1994) the electricity consumption could be reduced up to 80% when switching from activated sludge process to a combined anaerobic/aerobic treatment. Estimated from the Rockhammar case in (IVL 2012) there could be up to 70% reduction of electricity use for the COD fraction reduced in the anaerobic reactor.

The total addition of chemicals and nutrients may be reduced when applying anaerobic technology instead of the activated sludge process. Phosphorus and nitrogen can be reduced up to 90% (Rintala & Puhakka 1994). Polymers, used in flotation steps and for dewatering sludge, may also be reduced. Additional chemicals may, however, be required for adjustment of pH before the

anaerobic reactor, which may bring additional costs. Though, the overall costs for chemical additions will likely decrease.

Excess sludge from the activated sludge process is difficult to dewater and put to any use. It is used for making construction soil, combusted after extensive dewatering or collected by a waste company. In either case there is a most likely a cost for handling the sludge (300 SEK per ton sludge with 20-25% DS (IVL 2012)) and it is beneficial to reduce the sludge amounts. This can be done with the anaerobic reactor, which only produces about 5-20% of the sludge amount produced in an activated sludge process (Rintala & Puhakka 1994). The reason for this is that the anaerobic microorganisms only use a fraction of the substrate for cell growth and the rest is used for producing biogas.

## 2.7 DESIGN AND MOTIVATION OF THE CASE STUDY

A bleached sulphate pulp mill was chosen for several reasons. A large part of the biogas production potential in the pulp and paper industry lies in sulphate pulp according to Magnusson (2012), see Table 1. Moreover, new findings suggest that it is possible to use AD for treating the streams from sulphate mills, which before was considered to be too difficult.

Considering the available technology for AD and the available substrate streams, two different concepts have been further evaluated.

*Case 1: High-rate anaerobic reactor (UASB) integrated with the external wastewater treatment prior to the activated sludge process.* The Kraft evaporator condensate and the alkaline bleaching streams are treated. The activated sludge process is still in operation treating the streams that cannot be treated in the anaerobic reactor and polishing the streams that are treated with AD. The advantage with the concept is that the COD load to the activated sludge process is reduced, which brings the aforementioned positive effects (see Section 2.6). In addition, the organic content is easier to degrade and convert to biogas when it is dissolved in the stream than when it is incorporated in the sludge as in Case 2. The disadvantage of this concept is the introduction and possible disturbances in the current wastewater treatment.

*Case 2: External stirred tank reactor with a retention time of about 20 days.* Primary and secondary sludge are treated together with the kraft evaporator condensate. This concept will also reduce the sludge amount, and produce biogas. Though, the substrate is less accessible in the form of microbial sludge compared to organic material in solution. Therefore it is necessary with longer retention times and pre-treatment to reach the potential biogas production.

The waste streams used in Case 2 could possibly also be transported to another facility since they are relatively concentrated. In this way, wastes from several pulp mills could be treated in one facility. It would also be possible to co-digest the waste with other types of waste, e.g. municipal waste as mentioned in section 2.5.7. The risk of process inhibition is likely higher for the pulp and paper mill waste than for the substrate it is mixed with. It is thus possible that the co-digestion of the substrates results in a lower total biogas production than separate digestion would. The option to transport the waste was therefore not evaluated in this case study.

## 2.8 TECHNOECONOMIC ASSESSMENT FOR TWO CASES

In accordance to Cases 1 and 2, an upscaled investigation with focus on the economical effects by the production of LBG (liquified biogas) integrated to a pulp mill was performed. The pulp mill used for the integration of the two cases is retained to an average Nordic pulp mill model, called the *Type Mill*, developed by ÅF and Innvenita during the FRAM (Future Resource-Adapted Pulp Mill) program (Delin 2005). Overall mass and energy balances for the cases were evaluated followed by a techno-economic assessment.

### 2.8.1 Background and general description of the *Type Mill* model

Innventia has developed theoretical full mill simulation models for different types of pulp and paper mills. The simulation models have been built in the program WinGEMS, designed for use in the pulp and paper industry. The simulation program calculates the steady-state distributions of fibre, water, energy and steam as well as the non-process elements in the process.

The constructed theoretical models may be adjusted to resemble a specific mill and can consequently be used to study an actual mill to a highly detailed degree. The models can also be used to study different process changes and/or introduction of new process concepts to a highly detailed degree. In this project, Cases 1 and 2 were evaluated by using available process streams in the *Type Mill*.

For the *Type Mill* the resource utilization is about the same as in an average pulp mill in Scandinavia. A summary of some process conditions for the *Type Mill* are:

- A pulp production of 1000 ADt (Air Dry ton)/24h
- A continuous digester with conventional 2-flash system
- Unscreened digester yield of 46.1%
- An alkali charge of wood as effective alkali of 20% NaOH
- A sulphidity in the white liquor of 35 mole-%
- One stage oxygen delignification
- Bleaching with conventional ECF bleaching
- Evaporation plant with 5.5 effect economy to 72% DS of the black liquor
- HP-steam data of 60 bar and 450°C in the recovery boiler
- Limekiln fired with oil
- No condensing turbine
- A runnability of 355 days per year and an availability of 92%

### 2.8.2 Liquefied biogas (LBG) production for case 1

Figure 3 shows a simplified mass balance of the most important and realistic flows related to the *Type mill* model for the production of LBG. For Case 1 these flows are retained to the alkaline filtrate and methanol condensate. The AD of the alkaline filtrate and the methanol condensate is assumed to be performed in a UASB reactor (see section 2.3). The produced raw biogas after AD treatment has then been assumed to be upgraded in a water scrubber followed by a polishing and liquefaction step to produce the LBG (Bauer 2013).

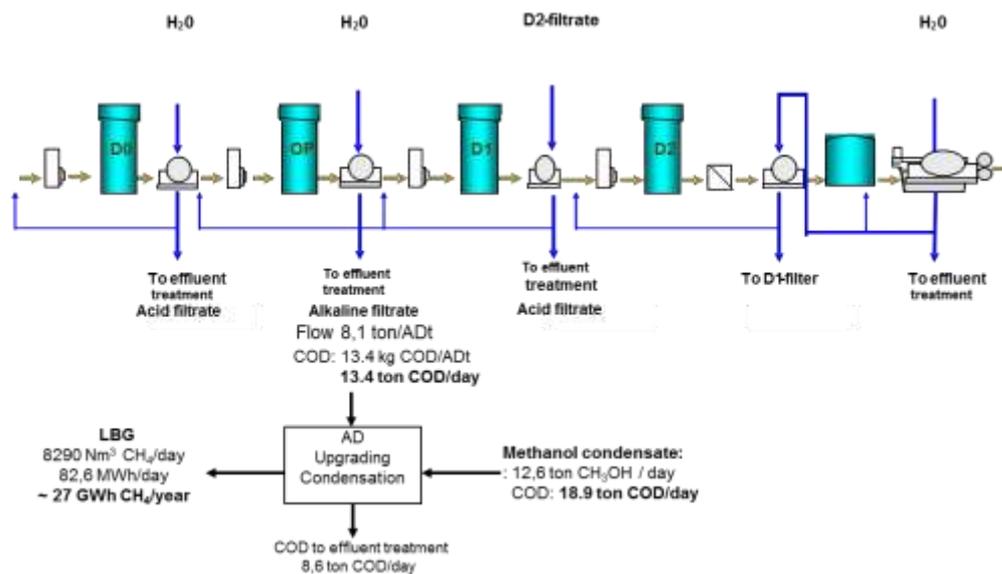
From the two present streams it has been estimated that about 27 GWh of LBG per year could be produced. Input data for the conversion of COD to biogas is showed in Table 4 and is in line

with data also showed in in Table 3. About 50% of the COD content in the alkaline filtrate, and about 10% of the COD content in the methanol condensate, will not be converted to biogas in the AD process. In total that accounts to 8.6 ton COD per day that still needs to be treated in the existing external water treatment system. However, that is a reduction with 4.8 ton COD/day, or ~1560 ton COD/year as a result of the addition of an AD step.

Not included in Figure 3, but of interest to point out, is that there are also significant amounts of COD in the acidic filtrates (~19 ton COD/day) that theoretically or potentially could be converted to biogas. However, due to the fact that acidic filtrates have showed toxicity to the methanogenic activity (see section 2.5), the potential to produce LBG from the acidic streams was excluded within this study.

**Table 4. Data used in the model for the production of LBG from the conversion of the COD in alkaline filtrate and methanol condensate, respectively.**

Data used in the estimation	Number	Unit
Biogas production alkaline filtrate	0.15	Nm <sup>3</sup> CH <sub>4</sub> /kg COD
Biogas production methanol condensate	0.34	Nm <sup>3</sup> CH <sub>4</sub> /kg COD
Composition of the raw biogas	65/35	Vol% CH <sub>4</sub> / vol% CO <sub>2</sub>
Losses of CH <sub>4</sub> in water scrubber	2	%
Composition of biogas after water scrubber	98/2	Vol% CH <sub>4</sub> / vol% CO <sub>2</sub>
Losses of CH <sub>4</sub> in condensation	0	%
Composition of biogas after condensation	100/0	Vol% CH <sub>4</sub> / vol% CO <sub>2</sub>



**Figure 3. Schematic overview of a bleach plant for a typical kraft pulp mill in Scandinavia with the addition of an upgrading process of the present COD in the alkaline filtrate and in the methanol condensate to produce LBG.**

### 2.8.1 Liquefied biogas (LBG) production for case 2

Figure 4 shows a schematic overview of LBG production from primary- and secondary sludge as well as from the present methanol condensate.

It has been estimated that about 26 GWh of LBG per year could be produced from the three substrates. The AD has been assumed to be performed in an external stirred tank reactor (see section 2.3). The produced raw biogas has then, in a similar way as for Case 1, also been assumed to be upgraded with a water scrubber followed by a polishing- and liquefaction step to produce the LBG (Bauer 2013).

Data used for the estimation is based on the flows showed in Figure 4 and from data in Table 5. It has roughly been assumed that the dry solid content (DS) in the primary- and secondary sludge will be reduced with about 50% as a consequence of the treatment in an external stirred tank reactor. For the methanol condensate the same number as for Case 1 was used i.e. 10% of the COD content in the methanol condensate will not be converted to biogas in the AD process. Of the remaining COD from unconverted methanol it is assumed that this amount needs to be treated in the existing external wastewater treatment and the amount accounts to approx. 1.8 ton COD/day. The COD from the unconverted methanol is added to the existing external wastewater treatment as press water occurring from dewatering of the remaining sludge formed after the external stirred tank reactor. The total production of sludge is reduced significantly for Case 2 from 10.3 ton DS/day to 5.4 ton DS/day, i.e. a reduction with about 5 ton DS/day or ~1,600 ton DS/year.

**Table 5. Data used in the model to calculate the production of LBG from the primary- and secondary sludge as well as from the present methanol condensate.**

Data used in the estimation	Number	Unit
Biogas production from primary sludge	300	Nm <sup>3</sup> CH <sub>4</sub> /ton DS
Biogas production from secondary sludge	150	Nm <sup>3</sup> CH <sub>4</sub> /ton DS
Biogas production methanol condensate	0.34	Nm <sup>3</sup> CH <sub>4</sub> /kg COD
Composition of the raw biogas	65/35	Vol% CH <sub>4</sub> / vol% CO <sub>2</sub>
Losses of CH <sub>4</sub> in water scrubber	2	%
Composition of biogas after water scrubber	98/2	Vol% CH <sub>4</sub> / vol% CO <sub>2</sub>
Losses of CH <sub>4</sub> in condensation	0	%
Composition of biogas after condensation	100/0	Vol% CH <sub>4</sub> / vol% CO <sub>2</sub>

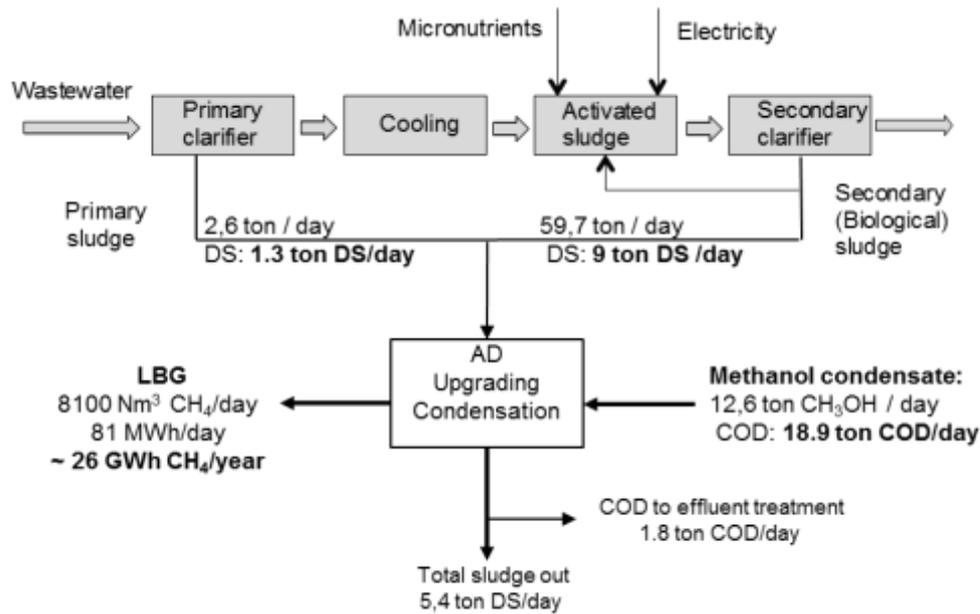


Figure 4. Schematic overview of the external wastewater treatment with the addition of an upgrading procedure to produce LBG from the DS in the primary- and secondary sludge and from the COD in the methanol condensate.

### 2.8.2 Effects in the mill with the production of LBG

Table 6 and Table 7 show used data in the calculations and main effects in the mill as a consequence of the production of LBG. After AD, either in an USAB reactor, as for Case 1, or with an external stirred tank reactor as for Case 2, similar upgrading procedures including a water scrubber followed by a polishing and liquefaction procedure were assumed in both cases. Water consumption and the regeneration of the water usage in the water scrubbers are related to information in Bauer (2013). Estimations point at a very small make-up water demand for Cases 1 and 2 (24-25 m<sup>3</sup> H<sub>2</sub>O/day), and the cost for that make-up water demand is therefore considered neglectable. The power consumption (3.4-3.5 MWh/day) on the other hand is significant for the water scrubber and of importance to include in the economical assessment. For the liquefaction step, the power consumption is also significant – 9.2-9.4 MWh/day – accounting for about 11% of the actual energy content of the produced LBG (81-83 MWh/day). The reason for such a high power demand in the liquefaction step is the need to cool the biogas to a temperature of -162°C to increase the energy/volume ratio. A liquefaction coefficient of 1.7 liters condensed CH<sub>4</sub>/Nm<sup>3</sup> CH<sub>4</sub> has been used in the study (Bauer 2013).

**Table 6. Data used in the model for the technoeconomic assessment for case 1 and 2.**

Data used in the estimation	Case 1	Case 2	Unit
<b>Water scrubber</b>			
Water consumption	0.19	0.19	m <sup>3</sup> H <sub>2</sub> O/Nm <sup>3</sup> raw biogas
Ditto	2,475	2,420	m <sup>3</sup> H <sub>2</sub> O/day
Regeneration of water	99	99	%
Water make-up demand	25	24	m <sup>3</sup> H <sub>2</sub> O/day
Power demand water scrubber	0.27	0.27	kWh/Nm <sup>3</sup> rawbiogas
Ditto	3.5	3.4	MWh/day
<b>Liquefaction</b>			
Liquefaction coefficient	1.7	1.7	liter cond. CH <sub>4</sub> / Nm <sup>3</sup> CH <sub>4</sub>
Power demand liquefaction	1.1	1.1	kWh/ Nm <sup>3</sup> CH <sub>4</sub>
Ditto	9.4	9.2	MWh/day

Effects in the *Type mill* model and mainly in the external water treatment system can be seen in Table 7. There are several differences between Cases 1 and 2. The main differences between the two cases compared to the *Type mill* model are:

- No reduction of primary sludge for Case 1 which is true for Case 2 because the primary sludge is used for biogas production in Case 2.
- Both cases produce less secondary sludge than the *Type mill*, Case 1 due to less COD to the external treatment and Case 2 due to the production of biogas from the sludge.
- The total COD content to the external treatment is reduced for Case 1 while it is slightly increased for Case 2. The reason for the differences is because biogas is produced from the alkaline filtrate in Case 1 while no alkaline filtrate is treated in Case 2. Additional COD from unconverted methanol condensate enters the external treatment as press water occurring from dewatering of the remaining sludge for Case 2.
- Within this study the chemicals used in the external treatment were assumed to be in line with Obbola pulp mill (IVL 2012). The estimated amounts of the added chemicals were based on the COD content to the external waste water treatment making it possible to estimate effects on the addition of chemicals between the *Type mill* model and the two evaluated cases. Due to lower COD content to the external treatment for Case 1 a slightly reduced chemical demand will occur. The opposite occurs for Case 2 and a similar result is also estimated for the power consumption related to the two cases.

**Table 7. Effects in the mill that relates to sludge formation, usage of chemicals and power consumption in external treatment.**

Effects by the LBG production	Type mill	Case 1	Case 2	Unit
Production of primary sludge	1.3	1.3	0.64	Ton DS/day
Red. (-) /addition (+) of prim. sludge	0	0	-0.64	Ton DS/day
Production of secondary sludge	9.0	7.9	4.8	Ton DS/day
Red. (-) /addition (+) of sec. sludge	0	-1.1	-4.2	Ton DS/day
COD content to extr. Treatment	34.5	29.6	36.3	Ton COD/day
Chemicals in extr. Treatment	5.9	5.3	6.4	Ton/day
Red. (-) /addition (+) of chemicals	0	-0.6	0.5	Ton/day
Power consumption extr. treatment	30	27	31	MWh/day
Red. (-) /addition (+) of power	0	-3	1	MWh/day

### 2.8.3 Economical effects in the mill with the production of LBG

This study is based on LBG production for two cases integrated to a typical pulp mill of Scandinavia. The investment cost for the equipment needed to produce the LBG has a big influence on the overall production cost and thus on the total benefit from selling the produced LBG. For Cases 1 and 2 a total investment cost of 18 and 15 MEUR, respectively was estimated, see Table 9. The main difference in the investment cost between the two cases is retained to the AD step. In Case 1 the investment cost for an UASB reactor was estimated with information related to IVL (2012) and in Case 2 the cost for an external stirred tank reactor was estimated with information found in Berg (2011). The cost for a water scrubber and for a liquefaction process was estimated with information in Bauer (2013). The investment cost for a tank station is estimated by Zinn (2013).

Table 8 shows the assumed prices that were used in the technoeconomic assessment, relating mainly to Innventia in-house data. Effects on the operating costs in the *Type mill* model and mainly in the external water treatment system and from implementation of new equipment for the two cases can be seen in Table 9.

**Table 8. Prices used in the assessment.**

Component	Unit	Price
Electricity	EUR/MWh	60
Sludge deposit	EUR/ton (25% DS)	36
Labor	EUR/year	60,000
Maintenance	% of investment/yr	2
Phosphoric acid	EUR/ton	670
Polymers to flotation	EUR/ton	3,000
Urea	EUR/ton	215
Polymers to sludge dewatering	EUR/ton	4,770
FeSO <sub>4</sub>	EUR/ton	200
Acid for pH adjustment	EUR/ton	50
Liquefied biogas (LBG)	EUR/MWh	119

Effects on the operating cost in the *Type mill* model will be different for Cases 1 and 2 and the main differences between the two cases are:

- In Case 1 the usage of chemicals and power in the external water treatment system could be reduced while the opposite occurs for Case 2.
- Power demand in the scrubbers and in the liquefaction step is significant for both cases. The reason the power consumption is slightly higher for Case 1 is only because the LBG production is slightly higher for Case 1 (27 GWh/yr) compared to Case 2 (26 GWh/yr).
- Cost savings due to reduced sludge formation is potentially higher for Case 2 than for Case 1. However, the reduction of the primary- and secondary sludge for Case 2 is only roughly estimated in this study and might be slightly overestimated, especially for the reduction of secondary sludge.
- In agreement with personnel contact (Zinn 2013) it is expected that two extra employee's are needed to run the equipment associated with the LBG production.
- In the calculation an annuity factor of 0.2 retained to a payback time of six years and an interest rate of 6% was used. That is in line with what has been used previously by IVL (2012). During the six years, the equipment is paid off with an interest rate of 6%. Both cases show increased operating costs corresponding to 951 and 612 kEUR/year for Case 1 and Case 2, respectively.
- After six years when the equipment has been paid off, the profit for Case 1 will be higher (~2.7 MEUR/year) than the profit for Case 2 (~2.5 MEUR/year), a difference that remains during the lifetime of the equipment. It can be pointed out that the income from sold LBG is set to 119 EUR/MWh which is a realistic selling price of transportation fuel after taxes of today (Zinn 2013).

**Table 9. Investment cost, effects on the operating costs, benefits and profit for case1 and 2 as a function of the annuity factor of 0.2.**

<b>Investment cost</b>	<b>Case 1</b>	<b>Case 2</b>	<b>Unit</b>
Anaerobic digestion (AD)	10	7.1	MEUR
Water scrubber	1.3	1.3	MEUR
Liquefaction	6.1	6.1	MEUR
Tank station	0.5	0.5	MEUR
<b>Total investment cost</b>	<b>18</b>	<b>15</b>	<b>MEUR</b>
<b>Effects on operating costs costs(+)/benefits(-)</b>			
Chemicals	-125	54	kEUR/year
Power change extr. Treatment	-58	22	kEUR/year
Power scrubber & liquefaction	252	246	kEUR/year
Less sludge	-50.9	-57	kEUR/year
Labor	119	119	kEUR/year
Maintenance 2 % of investment	361	303	kEUR/year
Benefit from LBG	-3,223	-3,154	kEUR/year
<b>Total benefit excl. investment</b>	<b>-2,724</b>	<b>-2,467</b>	<b>kEUR/year</b>
	0.2034	0.2034	
	6	6	Years
	6	6	%
<b>Cost per year during payback time (&lt; 6 yrs)</b>	<b>951</b>	<b>612</b>	<b>kEUR/year</b>
<b>Benefit after payback time (&gt; 6 yrs)</b>	<b>-2,724</b>	<b>-2,467</b>	<b>kEUR/year</b>

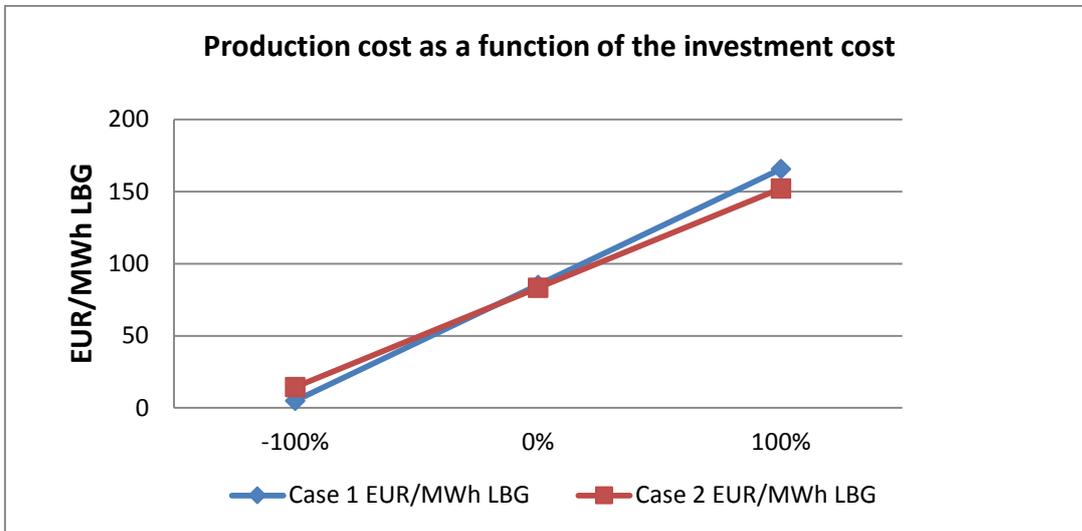
To be able to estimate an overall production cost of the LBG in relation to the whole lifetime of the equipment, an annuity factor of 0.1 were used. An annuity factor of 0.1 with an interest rate of 6% indicates a lifetime of 15 years. The number is considered relevant for an installation of a new process and estimations of the overall production cost of the LBG related to these assumptions can be seen in Table 10.

With the estimated investment cost as showed in Table 10 and with the usage of an annuity factor of 0.1 both cases looks promising with a profit corresponding to 34–36 EUR/MWh LBG. The profit from selling the LBG is based on a selling price of 119 EUR/MWh LBG.

**Table 10. Overall production cost of liquefied biogas (LBG) with an annuity factor of 0.1.**

<b>Overall production cost of the LBG</b>	<b>Case 1</b>	<b>Case 2</b>	<b>Unit</b>
Assumed lifetime of the equipment	15	15	Years
Annuity factor used	0.1	0.1	
Interest	6	6	%
Total investment cost	67	57	EUR/MWh LBG
Net operating costs	18	26	EUR/MWh LBG
<b>Total production cost of LBG</b>	<b>85</b>	<b>83</b>	<b>EUR/MWh LBG</b>
<i>Assumed selling price of LBG</i>	<i>119</i>	<i>119</i>	<i>EUR/MWh LBG</i>
<b>Estimated profit during lifetime</b>	<b>34</b>	<b>36</b>	<b>EUR/MWh LBG</b>

The cost for the equipment has a big influence of the overall production cost which is important to point out. Figure 5 therefore shows a sensitivity analysis of the overall production cost as a function of the total investment cost. The investment cost has been varied  $\pm 100\%$  from the estimated investment costs used in the study.



**Figure 5. Sensitivity analysis for the overall production cost of LBG as a function of the investment cost. The investment cost has been varied  $\pm 100\%$  from the estimated investment cost used in the study.**

## 3 TECHNICAL POTENTIAL FOR PRODUCTION OF BIOMETHANE VIA GASIFICATION OF WOODY BIOMASS

### 3.1 EFFICIENT VALUE CHAINS FROM FOREST TO FUELS

Low energy density and high moisture content represent typical disadvantages of lignocellulosic raw material. There are often long distances between the biomass and the end-users, e.g. district heating and transportation fuels, making it important to decrease transportation costs of the raw material. Pre-treatment techniques are therefore needed to increase the calorific value of the biomass, to remove contaminants and to make the biomass more homogenous. An important factor when it comes to transportation of the energy carriers themselves, i.e. feedstocks and fuel products, is the energy density. Some possible pre-treatment methods are direct milling, torrefaction and milling, pyrolysis and low temperature gasification. Extraction of lignin, which is the most energy rich component in wood, is also a potentially interesting method of preparing an intermediate fuel product.

The other benefit of an approach where materials are upgraded to higher energy density and better fuel quality in integrated value chains, is to allow for larger production plants that can benefit from economy of scale, such as biomass gasifiers, since more raw material can be transported to one plant when longer transport distances can be accepted. Biomass conversion processes to intermediate biomass products such as torrefaction, pyrolysis oil and lignin extraction are described below.

#### 3.1.1 *Torrefaction*

Torrefaction is a mild pre-treatment of biomass at temperatures of 200-300°C in the absence of oxygen. It removes moisture and low weight organic volatile components and depolymerises the long polysaccharide chains. Torrefied wood can replace fossil coal in a variety of applications. It gives advantages when the biomass must be transported longer distances, and lowers the costs related to the handling and feeding systems.

If the primary target is to produce fuel for heat and power generation, the thermal treatment is usually followed by pelletisation. Pelletisation of torrefied biomass increases the density to 750-800 kg/m<sup>3</sup>. The heating value is slightly higher than that for pellets from wood or bark: a lower heating value (LHV) of 18-23 MJ/kg or higher heating value (HHV) of 20-24 MJ/kg (5-6.4 and 5.5-6.7 MWh/ton, respectively) (Nordgren 2011). The process requires a separate plant and an input of heat. However, a torrefaction plant situated near a pulp mill gives synergy effects. The production of gaseous and volatile streams requires emission control, which entails capital and operating costs. Cooling water is also required which the pulp mill can supply and also cool or utilize the returning hot cooling water. The wood yard would also be shared and synergy effects could be achieved here as well.

#### 3.1.2 *Pyrolysis*

Pyrolysis oil is produced when biomass is rapidly heated to temperatures around 400-500°C in the absence of oxygen, converting it into a liquid oil with yields up to 60-80% (weight). Char, 5 - 20%, and gas, 15-30% yield is also produced. Pyrolysis oil is a mixture of oxygenated com-

pounds formed during the decomposition of lignin, cellulose and water (biomass moisture and formation products). Pyrolysis oil has a high acidity; the oxygen content is typically 45-50% and the water content 15-30%. Because of this the heating value is rather low compared with fossil oil and stainless steel must be used when handling the material because of the acidity which makes it high corrosive.

An example of a pyrolysis process, the Envergent process is presented in Figure 6. The pyrolysis process is often referred to as fast pyrolysis because of the short residence time, only a few seconds. A fluidized bed with sand is used as the heat transferring medium, circulating between the reactor and the reheater. The char produced in the reactor is sent to the reheater, which produces an exhaust gas and an ash fraction. The pyrolysis gas from the reactor is condensed and the product is pyrolysis oil.

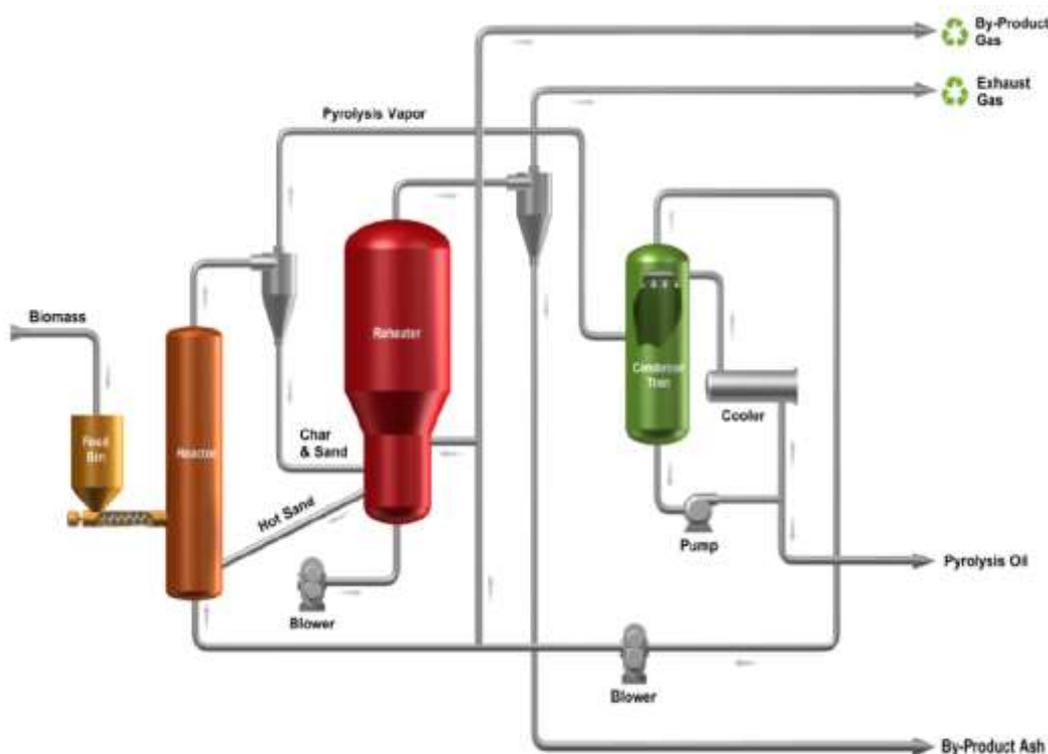


Figure 6. Process overview of pyrolysis of biomass (Envergent technologies).

### 3.1.3 Lignin extraction

LignoBoost, owned by Metso, is one of the technologies already commercially available and ready for use for lignin extraction. In the LignoBoost process a stream of black liquor is taken from the evaporation plant (Figure 7). The technology is based on addition of  $\text{CO}_2$ . The lignin is precipitated by acidification and filtered. The filter cake is re-dispersed and acidified (*Acidulation* in Figure 7). The resulting slurry is then filtered and washed using displacement washing (*Filtration and washing*). The filter cake is re-dispersed in a liquor, where the pH and temperature are controlled to approx. that of the final wash liquor.

It could be interesting to combine this technology with separation of  $\text{CO}_2$ , from the lime kiln or recovery boiler flue gases which potentially provide the required  $\text{CO}_2$  in the lignin separation

process. In a process with biomass gasification and methanation, CO<sub>2</sub> removed from the syngas could be used for the same purpose.

There is a limit to how much lignin can be extracted without affecting the combustion properties in the recovery boiler, normally around 60% of the lignin can be taken out.

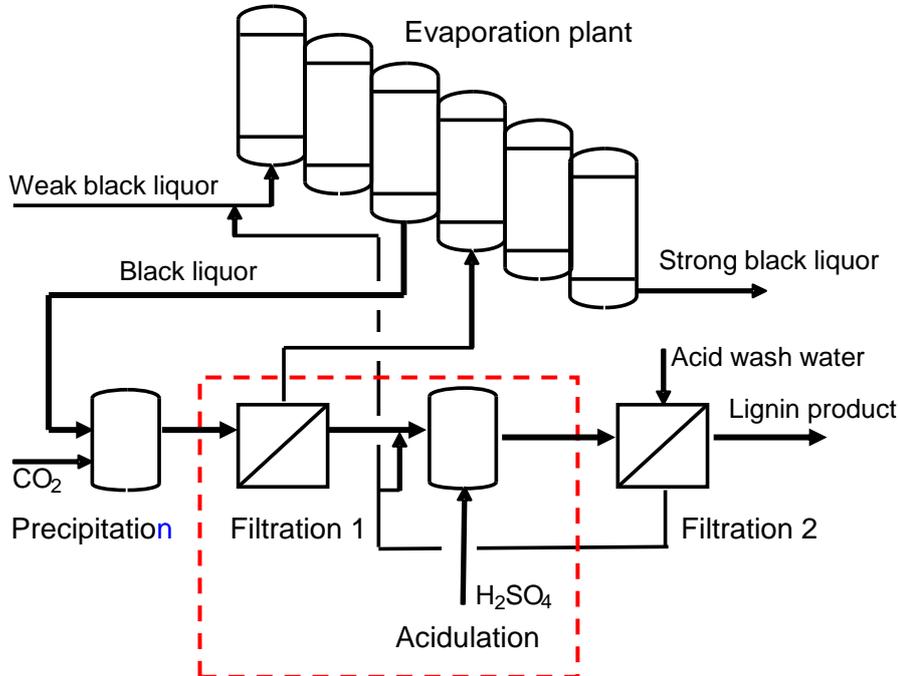


Figure 7. The LignoBoost process (Tomani et al. 2005).

### 3.2 INTEGRATION OF A BIOMASS GASIFICATION PROCESS WITH PULP AND PAPER PRODUCTION

In this study the production of synthetic natural gas from biomass (Bio-SNG) via an indirect gasification process was integrated to both the *Type Mill* model for market kraft pulp (see section 2.8.1), and to an integrated pulp- and fine paper mill model.

The integrated fine paper mill model exemplifies a typical Scandinavian mill as defined by ÅF and Innventia during the FRAM (Future Resource-Adapted Pulp Mill) program (Delin 2005). The paper production in the integrated fine paper mill is 1,570 ton paper/day with a dryness of 93%. The total dry paper production accounts to 1,460 ton dry paper/day and contains approx. 77% hardwood pulp (1,250 ADt/day) and 23% softwood pulp (370 ADt/day). For the paper production the corresponding hardwood pulp and softwood pulp is produced in campaigns and for this integration study the outfit of the mill during the hardwood campaign was used.

For the integration with the market pulp mill a pulp production of 1,000 ADt/day based on softwood was used.

For the Bio-SNG process to be integrated to the two mills, the set-up in accordance to the GoBiGas process has been used (Gunnarsson 2012). Figure 8 shows a simplified set-up of the biomass to SNG process and the marked boundary limit shows the parts that have been integrated with either a market pulp mill or with an integrated fine paper mill.

The size of the Bio-SNG process has been in line with the size presented by Heyne (2013) and is approx. three times the GoBiGas demonstration plant. The size of the Bio-SNG process corresponds to a wood input of 100 MW<sub>wood</sub> or approx. 18 ton dry wood/hr.

After the gasification and gas cleaning step a product gas containing 68 MW is formed with a typical dry gas composition of CH<sub>4</sub> (7-12 vol%), CO<sub>2</sub> (13-17 vol%), CO (17-32 vol%), H<sub>2</sub> (38-56 vol%) and C<sub>n</sub>H<sub>n</sub> (2 vol%).

After the methanation and final gas upgrading step, a Bio-SNG stream containing 63 MW is formed and corresponds to a production of about 490 GWh<sub>SNG</sub>/year. The composition of the SNG product is mainly related to CH<sub>4</sub> (>94 vol%) and minor amounts of CO<sub>2</sub>, CO, H<sub>2</sub> and N<sub>2</sub> (Heyne 2013; Gunnarsson 2012).

The integration with the two mills has been concentrated to the three first steps in the Bio-SNG process due to the following reasons:

- Drying of the biomass before the gasification is energy consuming because the biomass needs to be dried from about 50% DS to about 80% DS. Using a heat surplus probably available in a typical pulp mill for the drying of the biomass would be beneficial.
- The gasification step and mainly the cooling of the produced gas can be used for production of high pressure (HP) steam. The HP-steam could thereafter be used in an existing turbine in the mill. The benefit from that would be a higher power production and a possibility to get more low pressure (LP) steam in the mill by using already existing equipment. However, the integration opportunity requires that capacity is available in the present turbine in the mill. Importing steam to the mill could also result in that less steam needs to be produced in the existing power boiler and consequently less biomass needs to be burned in the bark boiler.
- The gas cleaning step for the indirect gasification process is described by Heyne (2013) and includes a cold gas cleaning chain consisting of a filter and a scrubber. Approx. 68% of the energy with the incoming wood will end up in the product gas leaving the gas cleaning step and about 8% could end up as HP-steam. The rest has been estimated to losses and mainly in the gasification process itself (Heyne 2013). The integration possibility of the gas cleaning step with a pulp- and/or paper mill is considered minor. However, the step is included in the boundary limit as Figure 8 shows. By including the step both realistic energy data related to the produced HP-steam as well as realistic energy content of the product gas leaving the gas cleaning step could be used (Heyne 2013).
- For the methanation step and gas upgrading step it has been difficult to find suitable integration possibilities with either a pulp and/or integrated paper mill. For the methanation step it is known that the process is exothermic and production of steam is possible. However, prior to the methanation step (not seen in Figure 8) a tar removal and a shift reaction step is necessary, and these two steps need energy. The energy surplus from the methanation process is therefore assumed to be enough to cover for the energy demand in the tar removal and in the shift reaction (Gunnarsson 2012). The net energy surplus from the methanation process is considered minor and thus not included as an integration possibility. In the Bio-SNG process there is also an energy surplus at low temperature, which might be used for production of district heating.

- There might be a possibility to integrate the last gas upgrading step with a pulp mill that separates lignin via the LignoBoost process. In the gas upgrading step separation of CO<sub>2</sub> is done which is a by-product in the Bio-SNG process. In the LignoBoost process (see section 3.1.3) CO<sub>2</sub> is used for the separation of lignin and represents one of the main operating costs. Integration with the gas upgrading step would therefore be interesting to look further into.

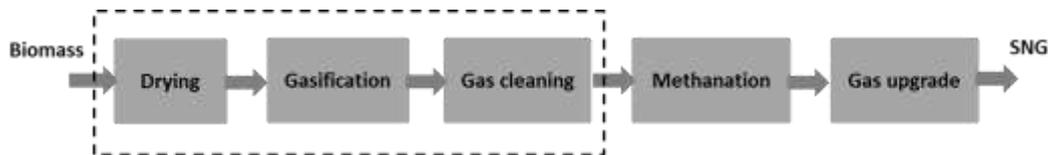


Figure 8. Simplified process layout of the synthetic natural gas from biomass (Bio-SNG) process.

### 3.2.1 Integration possibility with a market pulp mill

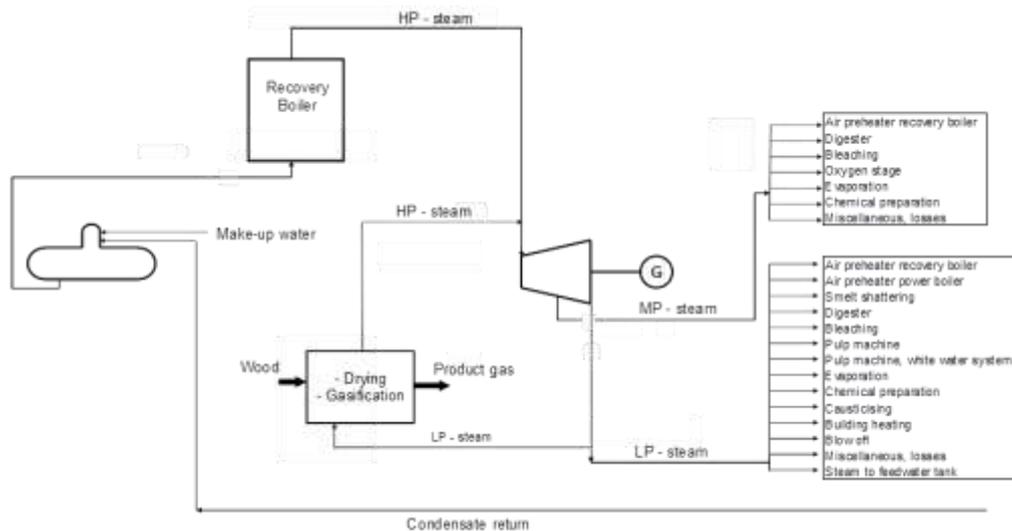
Figure 9 shows a simplified schematic overview of the steam distribution in the *Type mill* model with an integration possibility of a drying- and gasification step of biomass to produce a product gas related to the Bio-SNG process.

In the steam system for the softwood *Type mill* model a recovery boiler produces all the steam that is needed in the pulp mill. The produced HP-steam passes a back pressure turbine where medium pressure (MP) steam and low pressure (LP) steam is extracted for usage in the mill. The average softwood pulp mill also has a steam surplus of about 1.6 GJ/ADt or ~18 MW. The steam surplus is too low for investing in a condensing turbine and therefore blown to the atmosphere.

The integration possibility investigated within this study can be seen in Figure 9. HP-steam produced by cooling the gas in the Bio-SNG process could be mixed with the HP-steam from the recovery boiler and then used in the back pressure turbine. By adding more steam to the back pressure turbine also more power will be produced. However, to be pointed out is that the gasification step consumes power. The extra power produced from importing HP-steam will probably be lower than the power consumption of the gasification step. LP-steam needed for drying of the biomass to the gasification step could be done by exporting LP-steam from the pulp mill to the drying section of the Bio-SNG process.

Effects on the mill by the integration of a dryer and gasification unit in a Bio-SNG process can be seen in Table 11. The size of the Bio-SNG process has been set to 100 MW<sub>wood</sub> which is about 18 tons of dry biomass/hr and in line with the size presented by Heyne (2013). The size of the Bio-SNG process is also considered relevant to integrate with the size of a typical Nordic pulp mill. By the implementation approx. 20% more biomass will be added to the site, i.e. 100 MW or 18 ton dry biomass/hr. The number is considered possible to be handled within the mill.

It is also considered possible to import the HP-steam produced in the gasification step. The HP-steam to the back pressure turbine will be increased with about 8 MW or 10.5 ton HP-steam/hr which is a 4% increased steam flow.



**Figure 9. Suggested integration possibility of a drying- and gasification step into the steam balance of the market pulp mill.**

Table 11 shows the change in the energy balance for the pulp mill as a consequence of the integration with the drying- and gasification step related to the Bio-SNG process. The main effects in the mill are:

- Extra HP-steam of about 4% of the total amount produced in the recovery boiler or 0.7 GJ/ADt will be imported from the gasification process to the back pressure turbine.
- The drying of the biomass will increase the total steam consumption in the mill. The energy for the drying of the biomass prior to the gasification step was estimated to 0.3 GJ/ADt or approx. 40% of the energy imported from the gasification step. The energy consumption for a steam dryer has been set to ~800 MJ/ton water evaporated and is in line with a number presented in Heyne (2013).
- Of the HP-steam imported to the mill approx. 15% or 0.1 GJ/ADt is retained to more power produced. The number could also be presented as a power production increase with 29 kWh/ADt or 1.2 MW.
- It can be pointed out that the gasification process consumes power and the amount is approx. 80 kWh/ADt, or 3.3 MW. By importing HP-steam from the gasification step approx. 36% of the power needed in the gasification step could be covered by using the existing back pressure turbine and generator at the pulp mill.
- The total excess of LP-steam in the average pulp mill will be slightly increased with about 0.4-2.0 GJ/ADt as a consequence of the imported HP-steam from the gasification process. The steam surplus is still considered too small to invest in a condensing turbine to increase the power production and thus blown to the atmosphere. However if the extra steam surplus accounting to 0.4 GJ/ADt was sent to a condensing turbine approx. 24 kWh/ADt or 1 MW more power could be produced.
- An average Nordic pulp mill has a power deficit of about -148 kWh/ADt and by including the net power consumption for the gasification process it has been estimated that the power deficit will increase to about -199 kWh/ADt. If the mill would have had a condensing turbi-

ne the total steam surplus could have been used to increase the power production with an additional 98 kWh/ADt.

**Table 11. Overall energy balance for the average pulp mill producing 1000 ADt/day stand alone or with a drying- and gasification step related to the Bio-SNG process integrated.**

	<b>Pulp mill Stand alone</b>	<b>Gasification Integrated</b>	<b>Change</b>
<b>Energy balance</b>	<b>GJ/ADt</b>	<b>GJ/ADt</b>	<b>GJ/ADt</b>
Steam produced in recovery boiler	17.9	17.9	0
Steam produced in bark boiler	-	-	-
Steam produced from gasification	-	0.70	0.7
Secondary heat	0.53	0.56	0.03
<b>Total energy production</b>	<b>18.4</b>	<b>19.1</b>	<b>0.7</b>
Overall process consumption	14.3	14.3	0
Drying of fuel to gasification	-	0.3	0.3
Excess steam	1.6	2.0	0.4
Back-pressure turbine	2.4	2.5	0.1
<b>Total energy consumption</b>	<b>18.4</b>	<b>19.1</b>	<b>0.7</b>
<b>Power balance</b>	<b>kWh/ADt</b>	<b>kWh/ADt</b>	<b>kWh/ADt</b>
<b>Back-pressure turbine, generation</b>	<b>643</b>	<b>672</b>	<b>29</b>
Overall process consumption	791	791	0
Gasification, consumption	-	80	80
Power deficit	-148	-199	-51
<b>Sum</b>	<b>643</b>	<b>672</b>	<b>29</b>

### 3.2.2 Integration possibility with an integrated pulp- and paper mill

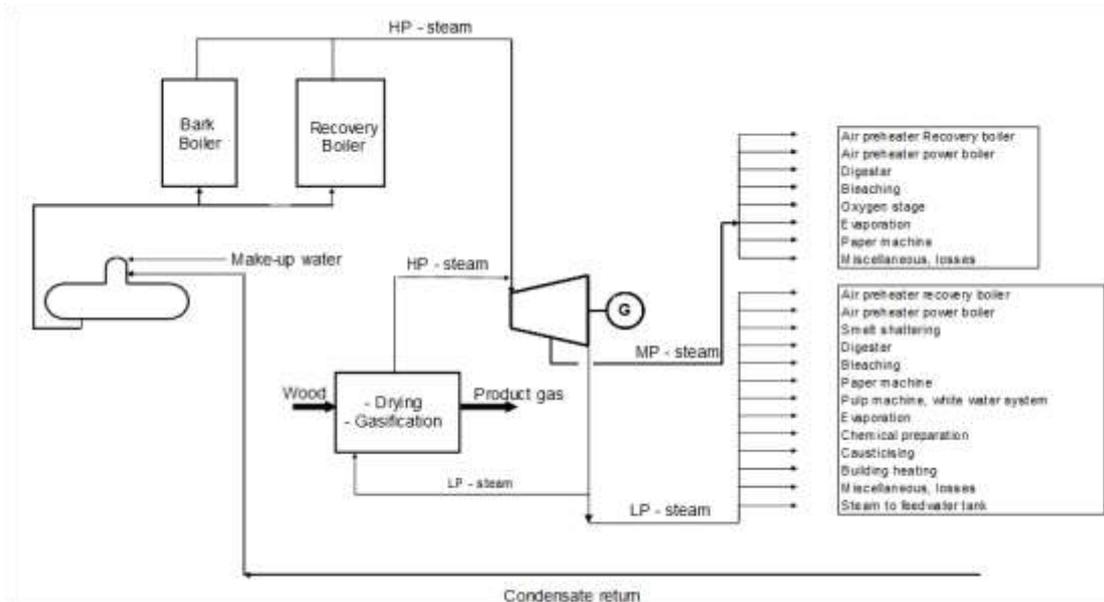
Figure 10 shows a simplified schematic overview of the steam distribution in an average typical integrated pulp- and paper mill model. The same figure also shows an integration possibility of the drying- and gasification step in relation to the Bio-SNG process.

In the steam system a recovery boiler and a bark boiler produces all the steam that is needed in the integrated pulp- and paper mill. The produced HP-steam passes a back pressure turbine where medium pressure (MP) steam and low pressure (LP) steam is extracted for usage in the mill. There is no steam surplus available in the integrated pulp and paper mill mainly due to a high steam demand in the paper machine. To balance the steam demand in the mill a bark boiler is highly necessary. In the bark boiler all bark of the incoming wood as well as some supplementary wood fuel is burned to sustain the steam demand in the mill.

By importing HP-steam from the gasification process more power could be produced in the existing back pressure turbine. Only part of the extra produced LP-steam is needed for drying of the biomass to the gasification step, and the remaining LP-steam could therefore be used in the pulp mill.

Effects on the integrated pulp- and paper mill by the implementation of a dryer and a gasification unit in relation to the Bio-SNG process can be seen in Table 12. The size of the Bio-SNG process is  $100 \text{ MW}_{\text{wood}}$  or about 18 tons of dry biomass/hr (Heyne 2013), i.e. in line with the size that was also integrated to the market pulp mill. From a mill perspective with a hardwood input of about 80 tons of dry wood/hr, it seems possible to handle an extra wood input of about 18 tons/hr.

For the import of HP-steam produced in the gasification step an increased steam flow to the back-pressure turbine of about 3% or 8 MW will be expected which probably can be handled. However, because only 15% of the energy in the HP-steam is converted to power in a back-pressure turbine and that only part of the energy that is left in the extracted LP-steam is used for drying of the biomass prior to gasification the surplus of energy could be beneficial for the mill and part of the supplementary wood fuel used in the bark boiler could be reduced. By doing this, the steam production from the bark boiler would be slightly reduced and the benefit from more produced power would only be slightly increased. That is because approximately the same amount of steam will pass the turbine. However the benefit from using less supplementary wood fuel in the bark boiler is greater and the case is also beneficial for a mill already running the turbine on max capacity.



**Figure 10. Suggested integration possibility of a drying- and gasification step into a typical steam balance of an average integrated pulp- and paper mill.**

Table 12 shows the change in the energy balance for the integrated pulp and paper mill model as a consequence of the integration with the drying and gasification step in relation to the Bio-SNG process. The main effects in the mill are:

- Extra HP-steam of about 4% of the total amount produced in the recovery boiler or  $0,45 \text{ GJ/ton}$  paper will be imported from the gasification process to the back pressure turbine.
- The drying of the biomass will increase the total steam consumption in the mill. The energy for the drying of the biomass prior to the gasification step was estimated to  $0,17 \text{ GJ/ton}$  paper or approx. 40% of the energy imported from the gasification step.

- Due to the import of HP-steam from the gasification step and due to the fact that only parts of that energy is converted to power and used for drying of biomass there is a surplus of 0.27 GJ/ton paper. The surplus could cover energy demands within the mill and less steam needs to be produced in the bark boiler. Approx. 10% of the fuel used in the bark boiler could be saved by importing HP-steam from the gasification step. A rough estimate shows a saving of about 23 ton dry woody biomass/day. That is about 5% of the biomass used in the gasification process (432 ton/day).
- Due to the saving of woody biomass in the bark boiler as a consequence of the imported HP-steam, only slightly less power production will occur in the mill approx. 0.03 GJ/ton paper. The number could also be presented as a power production increase with 9 kWh/ton paper or 0.6 MW.
- As mentioned before, the gasification procedure consumes power and the amount is approx. 51 kWh/ton paper or 3.3 MW. By importing HP-steam from the gasification step and reduce the woody biomass to the bark boiler only minor amounts of the power needed in the gasification step could be covered. The main benefit would be less biomass to the bark boiler.
- An average integrated pulp- and paper mill has a power deficit of about -559 kWh/ton paper and by including the net power consumption for the gasification process it has been estimated that the power deficit will increase to about -601 kWh/ton paper.

**Table 12. Overall energy balance for the integrated fine paper mill producing 1,570 ton paper/day with a dryness of 93%. The balances are for the mill stand alone and with a drying- and gasification step related to the Bio-SNG process integrated.**

	<b>Integrated fine paper Stand alone</b>	<b>Gasification Integrated</b>	<b>Change</b>
<b>Energy balance</b>	<b>GJ/ton paper</b>	<b>GJ/ton paper</b>	<b>GJ/ton paper</b>
Steam produced in recovery boiler	11.2	11.2	0
Steam produced in bark boiler	3.2	2.9	-0.27
Steam produced from gasification	-	0.45	0.45
Secondary heat	0.36	0.37	0.01
<b>Total energy production</b>	<b>14.7</b>	<b>14.9</b>	<b>0.2</b>
Overall process consumption	12.8	12.8	0
Drying of fuel to gasification	-	0.17	0.17
Excess steam	-	-	-
Back-pressure turbine	1.9	1.91	0.03
<b>Total energy consumption</b>	<b>14.7</b>	<b>14.9</b>	<b>0.2</b>
<b>Power balance</b>	<b>kWh/ton paper</b>	<b>kWh/ton paper</b>	<b>kWh/ADt</b>
<b>Back-pressure turbine</b>	<b>505</b>	<b>514</b>	<b>9</b>
Overall process	1,064	1,064	0
Gasification	-	51	51
Power deficit	-559	-601	-42
<b>Sum</b>	<b>505</b>	<b>514</b>	<b>9</b>

## 4 MARKET DEVELOPMENT

In order to be a marketable fuel the biomethane needs to meet specifications that are well defined and can be met regardless of the starting material and various ways of producing intermediate fuel mixtures or biogas. The possibility of transporting the biomethane over reasonably large distances will also depend on the density at which it can be distributed, where a liquefied product would be preferable. A summary of market activities indicating the future need of biogas in the transport sector will be given. The forecast will include the markets for CBG and LBG. The work will be based upon historic development and take into account strategy documents from public transport authorities (2) and information from transport gas distributors such as Fordonsgas Sverige AB. The value of biogas in the transport and petrochemical sectors in 2020 will be estimated. This work will be based on recent reports published by the IMF (3) and other recognized organizations.

### 4.1 CURRENT LBG PROJECTS IN SWEDEN AND NORWAY

#### 4.1.1 Lidköping Biogas, Sweden

Lidköping Biogas is one of the world's first plants for Liquefied Biogas (LBG). The new plant is a joint effort for the companies Swedish Biogas International Lidköping AB, Göteborg Energi AB and the Municipality of Lidköping. The plant supplies cars and trucks with renewable fuel.

The biogas production process is based on local vegetable waste products from grain trade and food production. The substrates are macerated, mixed and heated to 38°C before being pumped into the digestion chamber. New substrate material is continually pumped into the process which produces biogas and biofertilizer. The biofertilizer is pumped to a covered storage pool.

Plant production is designed at 7,5 MWth with an annual target of 60 GWhth. The production plant is designed by Swedish Biogas International AB. The biogas is upgraded in accordance with the Swedish standard for biogas as a vehicle fuel (SS 155428) in a water scrubber.

A majority of the biogas is liquefied in the condensation plant. The technology is supplied by Air Liquide. In order to liquefy the biogas, the majority of remaining CO<sub>2</sub> (<10 ppm) is purged by Pressure Swing Absorption (PSA) before the gas temperature is lowered using a nitrogen Brayton cycle. The technology allows for liquefaction in the span of -140°C (at 4 barg) to -161°C (at atmospheric pressure), depending on the developing requirements of the vehicle market. The design capacity is 12 ton LBG/day, but will initially be run at around 70%. The energy cost of liquefaction is in the vicinity of 1 kWh/Nm<sup>3</sup> upgraded biogas, which equals approx. 10% of the energy content of the biogas.

The liquefied biogas is stored in a 115 m<sup>3</sup>, 20 m tall insulated canister. The distributor, Fordonsgas Sverige AB (FGS), fills insulated 50 m<sup>3</sup> trailers every second day and transports the gas to filling stations in Göteborg with more to come.

A smaller portion (around 30%) of the biogas produced and upgraded to SS 155438 is delivered directly to FGS's two compressors, which fills mobile storage containers in one of six filling places. FGS distributes CBG to other filling stations in the vicinity using mobile storage containers. There is also a public filling station on site where vehicles are able to refill Compressed

Biogas (CBG) at 200 bar. As the demand for LBG increases in Sweden, CBG-production at Lidköping will decrease.

This project has been awarded grants from the Swedish government's Climate Investment Programme (KLIMP). The biogas produced in Lidköping is enough to fuel 6,000 cars that drive 17,000 kilometers per year. This results in 16,000 tons of reduced CO<sub>2</sub> emissions annually.

FGS is the only supplier of vehicle fuel approved by the Nordic Ecolabel and production at Lidköping must fulfill these high standards.

Liquefied methane gas, either from biogas or natural gas, is a new vehicle fuel in Sweden. Volvo Trucks has in 2013 started a serial production of their FM line using Methane-Diesel Engines. The target for the first year of production is 100 MDE-trucks for sale in the initial markets of Sweden, Holland and the UK.

At a national level, the biogas distributors FGS, E.ON and AGA aim at establishing a total of four LCMG-filling stations in Sweden, serving at least 100 Methane-Diesel trucks, in the next few years.

#### **4.1.2 Oslo, Norway**

The municipality of Oslo is currently constructing the Romerike anaerobic digestion plant, which will receive 50,000 tons of organic waste from households and industries in the region. The plant aims at annually delivering 4 million Nm<sup>3</sup> of LBG, starting in August 2013. The investment in the condensation plant is estimated at 45 million NOK. The condensation technology supplier is Wärtsilä. The technology used is expected to be more energy efficient than in Lidköping.

The wastewater treatment plant of Oslo currently produces approx. 2 million Nm<sup>3</sup> of biogas annually. The plant will expand to receive effluent from additional areas in 2016 and biogas production is expected to increase to 4 million Nm<sup>3</sup>. Further expansion is possible, which would lead to a total biogas production estimated at 10 million Nm<sup>3</sup>.

For more information on this project, contact the Waste-to-Energy Agency (EGE) of the municipality of Oslo.

#### **4.1.3 Biogas Sydöstra Skåne, Sweden**

A new biogas plant in south-east Skåne in Sweden promises to replace 11 million litres of diesel with liquefied methane from manure. The project aims at reducing the smell and methane releases from the current manure management as well as reducing nitrogen runoff to local recipients. In addition, the project will close to eradicate the CO<sub>2</sub> emissions currently released by the annual usage of 11 million litres of diesel.

Close to 100 farms are shareholders in the project and will deliver the manure commodity to the logistically well-placed facility. The project is also financially supported by the regional government (Region Skåne) and the County Administrative Board of Skåne (Länsstyrelsen).

Läckeby Water/Purac, a world leading biogas technology supplier, is expected to join the project and develop a suitable plant for the anaerobic digestion process. Foreign and domestic com-

panies such as Purac, Wertsilä, Air Liquide and Linde/AGA have expressed their interest in developing and possibly owning and running the condensation process of the plant.

Biogas Sydöstra Skåne has confirmed considerable interest amongst the larger trucking companies, who wish to reduce their emissions of CO<sub>2</sub>. Companies such as Malmö LBC, AKKA Frakt, DHL, Schenker and Posten have begun to use Volvo Trucks new motor technology for liquefied methane.

More information is available at [www.biogassydostraskane.se](http://www.biogassydostraskane.se).

#### **4.1.4 Trøndelag, Norway**

Farming communities in the Trøndelag region of Norway are currently aiming at establishing manure-based liquefied biogas production.

According to their estimate, the Ørland/Bjugn region can annually produce 4.5 million Nm<sup>3</sup> of methane, equivalent to 42 GWh of fuel. They have estimated the total investment for biogas production to 65 million NOK and believe that the project will be profitable.

The Rissa region is estimated to be able to produce 3 million Nm<sup>3</sup> of methane, corresponding to 28 GWh of fuel. They have estimated the total investment for biogas production to 45 million NOK and believe that the project will be profitable.

From the two biogas plants, the project estimates the cost of a common liquefaction plant to 110 million NOK. The plant would be able to produce 70 GWh of LBG.

The project is currently looking for a financially strong partner with experience in the field.

The project management can be reached through the Norwegian company Bioskiva AS.

## **4.2 THE MARKET FOR BIOMETHANE**

### **4.2.1 Historic development**

Methane as a vehicle fuel was introduced to Sweden in the early 1990's, initially using natural gas (SGC 1995). In 1992, upgraded biogas was used for the first time for transportation in Sweden. In many instances, natural gas and upgraded biogas is blended and marketed as *vehicle gas* (fordonsgas), which is the common name for methane used in vehicles.

Since 2000, the market for vehicle gas has on average grown 20% annually. Since 2006, the use of biogas has been equal to or in excess of the use of natural gas in vehicles.

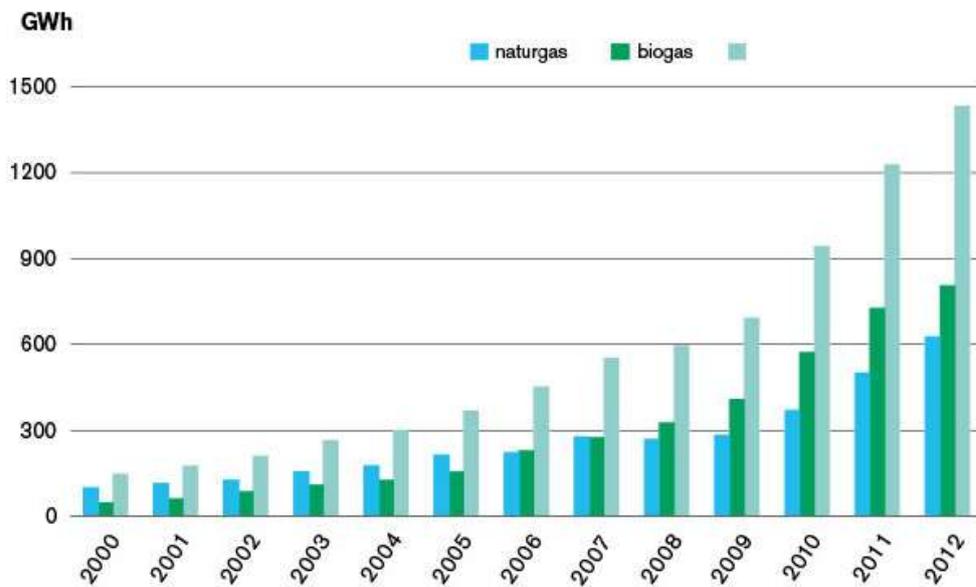


Figure 11. Methane usage in Swedish vehicles, 2000 – 2012. Adapted from [www.gasbil.se](http://www.gasbil.se).

The market for vehicle gas continues to grow in both the private and public sectors. Statistics for 2012 show that almost 140 million Nm<sup>3</sup> of vehicle gas were sold, corresponding to 1,426 GWh. The number of filling station in Sweden has continued to grow and now equals 138 public stations. The vehicle gas sold in 2012 has replaced petrol and diesel corresponding to 91,000 cars and reduced CO<sub>2</sub> emissions by 260,000 tons.

In Sweden, there are currently approx. 44,000 NGV's (Natural Gas Vehicles), which run on natural gas and biogas and often as well on petrol or diesel. The availability of vehicles on the Swedish market has grown steadily and now comprises over 20 cars and several truck models (Energimyndigheten, 2013).

#### 4.2.2 Current and future development

One of the most important drivers for the growth of renewable fuels in the transport sector in the EU target of 10 % by 2020. In Sweden, this target has also already been achieved (Energimyndigheten, 2013). The Swedish government has commissioned a study to develop a strategy of achieving a transportation sector without fossil fuels by 2030 (Biogas Väst, 2013). This study is expected to be published by the end of 2013.

A recent EU-proposal would limit the viability of crop-based biofuels for fulfilling these targets (EC, 2012). Only half of the 10 % may be constituted by crop-based alternatives if this proposal should be adopted. This would leave Sweden with the task of replacing crop-based ethanol and biodiesel currently used in the transport sector with non-crop-based alternatives, such as biogas.

Another recent EU-proposal, *Clean Power for Transport*, calls for an expansion of fuelling infrastructure for methane, hydrogen and electric vehicles (EC 2013). Fuelling station density would increase to a CNG-station every 150 kms and an LNG-station every 400 kms. This may

greatly increase the use of natural gas in vehicles and command an increased demand for biogas across Europe, in order to fulfil the 10% by 2020-target.

The public transport authority in the region of Västra Götaland, Västtrafik, has adopted the target proposed by the The Swedish Public Transport Association of using 90% renewable fuels by 2020. Västtrafik has also stated that biogas is a priority alternative, before other renewable fuels (Biogas Väst, 2011).

The similar authorities for the regions of Skåne (Skånetrafiken) and Östergötland (Östgötatrafiken) have even more ambitious target. Skånetrafiken aims at achieving 100% renewable fuel usage by 2020, of which mostly is anticipated to be biogas (Hanander & Rosqvist 2011). Östgötatrafiken aims at 100% by 2015, also mostly biogas (Östgötatrafiken 2011).

In 2012, busses in public transport used approx. 1.6 TWh of diesel in Sweden. This can be compared to the use of biogas in vehicles in Sweden in 2012: 0.8 TWh. Considering the anticipated increase of public transport in Sweden, the demand of biogas for this sector has the potential of doubling the current market by 2020.

Another development is the opening of a new segment for biogas as a fuel. Volvo Trucks has developed Methane-Diesel Engines (MDE), which allow for biogas and natural gas to be used in heavy-duty long-distance trucks. These trucks can replace up to 75% of their diesel consumption with methane. The methane is stored in liquefied form in insulated tanks on board the vehicle and re-gasified before being used in the engine. With the current support scheme in Sweden, MDE-trucks using a 50/50 blend of LBG and LNG can be competitive with conventional diesel alternatives.

The BiMe Trucks project assists companies in acquiring MDE trucks and aims at having 100 of these trucks on Swedish roads by 2014. The project also coordinates the development of infrastructure, which currently consists of filling stations in Göteborg, Järna, Älvsjö, Malmö and Jönköping. The next filling station is planned for in Örebro.

A realistic potential for biogas production in Sweden by 2030 has recently been assessed by the Swedish Energy Gas Association (WSP 2013). Their study shows that between 11 and 22 TWh of biogas from anaerobic digestion and gasification is reasonable, should adequate support programmes be introduced. The total usage of fuel in road transports in Sweden is approx. 90 TWh per year. Current and future efforts are likely to reduce the use of fuel in this sector, possibly as much as by 50%. Biogas can play an important part in fulfilling this remaining need with a climate-friendly, waste-based alternative.

## 5 DISCUSSION AND CONCLUSION

There is a large technical potential for production of upgraded biogas via anaerobic digestion (AD) in the Swedish pulp and paper industry, between 0.7 and 1 TWh/year according to reviewed studies. The economical potential, i.e. the fraction of the technical potential that would actually be profitable to invest in, is, however, not available in the literature or fully explored in this study. This study shows that a large part of the technical production potential lies within kraft pulp mills and that it, on the contrary to prior knowledge, seems possible to utilize this potential. It has so far only been shown in lab-scale studies and further studies are required before full-scale implementation.

In addition to production of valuable upgraded biogas, the integration of anaerobic treatment may also reduce electricity use in the activated sludge step, lower the use consumption of nutrition additions and chemicals in the wastewater treatment and reduce the excess sludge production.

This study further indicates that recent developments in high rate reactors may increase the production rate and biogas yield even further compared to the data used in this study.

In the study two cases for the production of liquefied biogas (LBG) via AD integrated to a typical Nordic pulp mill were evaluated. In case 1 methanol condensate and alkaline filtrate were used as substrate in an UASB reactor to produce raw biogas that was further upgraded in to LBG. In Case 2 the sludge from the waste water treatment and the methanol condensate were used as substrates. An external stirred tank reactor was used to produce the raw biogas that thereafter also was further upgraded in to LBG.

For comparison with a previous study IVL (2012) an annuity factor of 0.2 retained to a payback time of six years and an interest rate of 6% was used. During the six years the equipment is paid off both cases show increased operating costs corresponding to 951 and 612 kEUR/year for Cases 1 and 2, respectively. Once the equipment has been paid off the profit for Case 1 will be higher (~2.7 MEUR/year) than the profit for Case 2 (~2.5 MEUR/year) during the remaining life time of the equipment. The point out is that the income from sold LBG is based on a price of 119 EUR/MWh which is a realistic selling price of transportation fuel.

The study also investigates the integration possibility for a biomass gasification process to a pulp- and/or paper mill. The drying- and gasification step of biomass related to the Bio-SNG process was integrated to both a market kraft pulp mill and an integrated pulp- and fine paper mill. The size of the Bio-SNG process corresponds to an input of 100 MW<sub>wood</sub> from which it is possible to produce approx. 63 MW of Bio-SNG. The composition of the SNG product is mainly related to CH<sub>4</sub> (>94 vol%) and minor amounts of CO<sub>2</sub>, CO, H<sub>2</sub> and N<sub>2</sub> (Heyne 2013). The quality of the gas is enough to cover the quality demand for the natural gas grid (Gunnarsson 2012). The benefits by integrate a drying and gasification step in a Bio-SNG process with either a pulp- and/or paper mill is:

- Turbines at the mills could be used for additional power production by importing HP-steam from the gasification step.
- Steam for drying of biomass could be exported from the mills to the Bio-SNG process.

- A surplus of LP-steam originated from the import of HP-steam could be used in the mill which will reduce the need for supplementary wood fuel used in the bark boiler with 10% or 23 ton dry wood fuel/day.

Table 13 shows a comparison between realistic production potential of biogas via anaerobic digestion (AD) of available waste streams versus biogas production potential from gasification of biomass in an average Nordic pulp mill. The biogas production potential from gasification is approx. 20 times higher than the production potential via AD of the waste streams investigated within this study. Even though the production potential is about 20 times higher for gasification of biomass in comparison with AD of waste streams the size is probably still too small to get reasonable production costs of the Bio-SNG production via gasification. In Heyne (2013) overall production cost of about 120 €/MWh<sub>SNG</sub> is showed for a similar size, i.e. 490 GWh/year (no liquefaction step included). Most of the Swedish pulp mills are not connected to a gas grid and liquefaction of the biogas is therefore considered necessary which of course will increase the investment cost.

**Table 13. Comparison between the potential for biogas production via anaerobic digestion (AD) versus gasification of biomass integrated to an average Nordic pulp mill.**

Comparison	AD of waste streams	Gasification of biomass	Unit
Production of biogas (CH <sub>4</sub> )	26-27	~490	GWh/year
Additional biomass used	0	~780	GWh/year
Extra wood handling in the mill	0	~20	%

For gasification of biomass to produce Bio-SNG it is considered necessary to build a rather large plant to benefit from economy of scale. It is also considered necessary that both power and district heating could be exported from such a big gasification plant. To integrate a gasification plant with a normal sized pulp mill in Scandinavia will not reach sufficient high production of biogas but several mills could probably be suppliers of biomass to a larger gasification plant. Some pretreatment methods for upgrading the biomass to higher energy density and better fuel quality might then be necessary to allow for longer transport distances.

Biomass conversion processes to intermediate biomass products such as torrefaction, pyrolysis oil and lignin extraction are described in this study. The integrated value chains for untreated biomass to pretreated intermediate products that can be gasified to produce a biogas product which could be upgraded to vehicle fuel quality should be further studied.

Biogas is currently transported via the natural gas grid and sold according to the same principle as Green Electricity, i.e. the seller of biomethane can allocate sales to specific consumers along the natural gas grid who pay a premium. There are examples of mills in Sweden that are connected to or are in the vicinity of the natural gas grid that could exploit this possibility for efficient transport to market. The remaining alternative is liquefaction, an established technology from the liquefied natural gas (LNG) industry currently being further developed and down-sized to suit the biomethane industry. Sweden's first liquefied biomethane (LBG) plant opened in Lidköping in 2012. LNG and LBG are currently being introduced as new fuels for long-distance trucks and busses, where a blend of the two is expected to meet current market expectations and demands.

## 6 FUTURE WORK

- Further evaluation of anaerobic treatment of streams from sulphate/Kraft mills, including suitable pre-treatment methods, optimal mixing of streams and the use of different high rate reactors.
- A more detailed study on the geographical location of the available biogas potential in the pulp and paper industry, in order to decide how much of it that could be used in the transport sector. This can be combined with economic considerations to give a more accurate estimate of the actual potential.
- Integration of the power to gas concept in a gasification process or pulp mills. Renewable electricity from the mill is used to produce hydrogen. This is combined with the carbon dioxide stream from the biogas production to synthesize more methane.
- Integration of material flows between a pulp mill and thermal gasification process should be further studied, such as reject and bark available at the mill and CO<sub>2</sub> from gas cleaning that could be used in a LignoBoost process.
- Pre-treatment of the biomass to increase the calorific value, to remove contaminants and to make the biomass more compatible for gasification and to allow for longer transport distances which enables large scale facilities should be further evaluated.

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