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PRE-STUDY OF BIOGAS PRODUCTION BY LOW-TEMPERATURE PYROLYSIS OF BIOMASS

Report from an f3 R&D project

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THE SWEDISH KNOWLEDGE CENTRE
FOR RENEWABLE TRANSPORTATION FUELS

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PREFACE

This report is the result of a cooperation project within the Swedish Knowledge Centre for Renewable Transportation Fuels (f3). The f3 Centre is a nationwide centre, which through cooperation and a systems approach contribute to the development of sustainable fossil free fuels for transportation. The centre is financed by the Swedish Energy Agency, the Region Västra Götaland and the f3 Partners, including universities, research institutes, and industry (see www.f3centre.se).

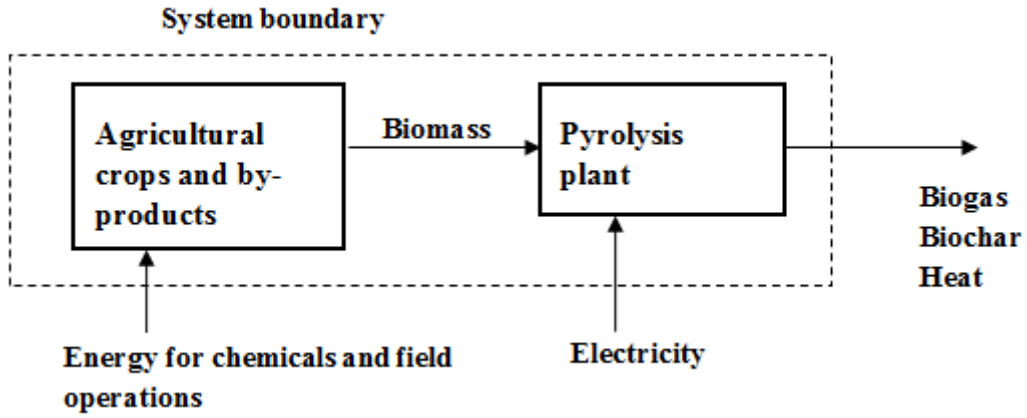
The aim of this study was to make a cross-disciplinary assessment of a novel route for biogas production via low-temperature pyrolysis using agricultural crops. Different aspects of biomass production and chemical conversion to biogas and other products were evaluated by cooperation between different experts at KTH and SLU.

The results revealed that the pyrolysis technology may provide high energy efficiency compared to other technologies, e.g. gasification, and that Sweden has a high potential for producing suitable biomass. A case scenario was used to evaluate the energy balance of the entire system from biomass production to the final products including biogas, biochar, and heat. It was concluded that about 2000 ha of *Salix* corresponding to approximately 14 000 ton dry matter biomass is needed to supply the case plant using 70 GWh_{LHV} of biomass per year. By using that biomass and 4 GWh electricity, the plant produces 49 GWh_{LHV} biogas and 13 GWh_{LHV} charcoal. The Swedish production potential, if using 250 000 ha fallow agricultural land, would be 5.8 TWh y^{-1} of biogas.

1. INTRODUCTION

Using biomass for fuel production is an alternative in the effort for replacing fossil fuels used in the transport sector. Agricultural crops cultivated on fallow land and by-products are considered as a potential feedstock that would not cause significant competition with food production nor with the supply of biomass to the forest industry. However, biomass originating from agricultural land usually has a low ash-melting point and a high content of alkali metals and ash. This will cause operational problems (e.g. poisoning of catalysts) in conventional gasification at high temperature.

Low-temperature pyrolysis of biomass is an interesting alternative to gasification, since alkali metals will remain in the charcoal (biochar) and the formation of persistent tar compounds is minimized. This enables the alternative of using feedstock with high alkali and ash content, such as agricultural crops and by-products, for fuel production. The aim of this F3 project was therefore to assess energy flows of a novel route of biogas[1] production from biomass via pyrolysis.



Figur 1 Principal system flow description including system boundaries.

A cross-disciplinary approach has been used to study the suggested production route. The different aspects of biomass production, chemical conversion to final products including biogas, biochar, and heat are evaluated by cooperation between different experts at KTH and SLU. The system displayed in Figure 1 has been used to exemplify the process in a case study. The study is divided into two work packages:

WP 1: "Assessment of suitable agricultural residues and energy crops for low temperature pyrolysis (Elham A. Moghaddam, SLU).

WP 2: "Simulation and system integration of a low temperature biogas production module" (Martin Bojler Görling and Mårten Larsson, KTH).

[1] Biogas is also referred to as bio-methane and SNG (Substitute Natural Gas).

1.1 WP1: "ASSESSMENT OF SUITABLE AGRICULTURAL RESIDUES AND ENERGY CROPS FOR LOW TEMPERATURE PYROLYSIS"

This section aimed to assess suitable and available agricultural feedstock for the designed low-temperature pyrolysis system. *Salix*, hemp (*Cannabis sativa* L.), Reed Canarygrass (*Phalaris arundinacea* L.), and straw were selected as potential biomass feedstock. Evaluation of the proposed feedstock were based on criteria, such as, chemical and physical characteristics of feedstock for adaptability to pyrolysis reactions, yield, climate conditions, experience of cultivation, and cost of production. Review of the selected feedstock is presented in the report "*Overview of suitable agricultural residues and energy crops for low-temperature pyrolysis"*, Elham Ahmadi Moghaddam (2012). *Salix* was assessed as the best option in terms of chemical and physical characteristics of feedstock, cost of production and long experience of cultivation including availability of machinery and technology. An energy balance of *Salix* production was performed for the integration with the energy balance of the pyrolysis process.

1.2 WP 2: "SIMULATION AND SYSTEM INTEGRATION OF A LOW TEMPERATURE BIOGAS PRODUCTION MODULE"

The aim of WP2 was to present an overall systems analysis of a poly-generation plant that produces biogas, bio-char and heat via pyrolysis. The hypothesis was that a production route via pyrolysis will give a higher total yield compared to gasification where a greater amount of the energy input is degraded into sensible heat. Methane can more efficiently be formed directly from the pyrolysis gas containing longer hydrocarbon chains compared to using syngas as the intermediate step. The pyrolysis production route has been suggested for several fuels, but to our knowledge never for methane. The process design has been simulated using the Aspen Plus® software using different pyrolysis rectors and integration alternatives.

In addition to this report, the main work performed in WP2 is presented in a paper:

Larsson, M., Görling, M., Grönkvist, S., Alvfors, P., 2013. "*Bio-methane upgrading of pyrolysis gas from charcoal production*", Submitted to Sustainable Energy Technologies and Assessments

The authors have also made a previous study related within the area:

Görling, M., Larsson, M., and Alvfors, P., 2013. "*Bio-Methane via Fast Pyrolysisof Biomass*". Applied Energy Journal (In press), 10.1016/j.apenergy.2013.01.002

2. BACKGROUND

Pyrolysis of biomass produces biochar, bio-oil, and gases including methane, hydrogen, carbon monoxide, and carbon dioxide. Biomass pyrolysis is an appealing technology mainly due to its high efficiency, high environmental performance characteristics, and opportunity for processing agricultural residues, wood wastes, etc. into renewable energy (Figure 2). Pyrolysis of biomass at relatively small scale and remote locations enhances the energy density of the biomass resource and reduces energy and costs related to handling and transport.

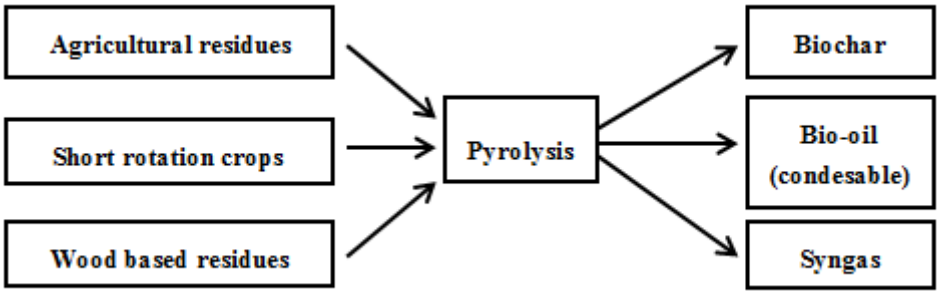


Figure 2 Feedstock and products from biomass pyrolysis.

The chemical structure, ash content, and water content of biomass are some of the major factors in process development and yield of biofuel production during pyrolysis. Biomass as hydrocarbon material contains carbon, hydrogen, oxygen, nitrogen, and a small proportion of sulphur. The major organic compounds of biomass are cellulose, hemicellulose, and lignin. Some biomass includes considerable amounts of inorganic elements. Ash concentration ranges from less than 1% in softwoods to 15% in herbaceous biomass and agricultural residues. Different inorganic elements have different effects on the pyrolysis process and for instance potassium (K) and calcium (Ca) catalyze biomass decomposition and char forming reactions.

2.1 FEEDSTOCK AND FEEDSTOCK SUPPLY

Reed canary grass (*Phalaris arundinacea* L.), straw, hemp (*Cannabis sativa* L.) and *Salix* were studied as potential pyrolysis biomass. Agricultural operation (production process), chemical composition, storage, yield (ton dry matter per hectare and year), local conditions and cost of production of the proposed crops is comprehensively described in the review report "*Overview of suitable agricultural residues and energy crops for low-temperature pyrolysis"* by Elham Ahmadi Moghaddam. Below follows an overview of the different biomass considered.

Reed canary grass (RCG) is a perennial forage crop with a high yield (6-8 tons dry matter $ha^{-1}y^{-1}$) compared to other grasses. RCG can maintain its yields at least up to 10-12 years and is a suitable crop for cold climates (Strömberg and Svärd, 2012). The harvesting technique for RCG to be used as a bioenergy feedstock has been developed, i.e. the delayed harvest system. This means that the grass is cut in late autumn and left on the field until the next spring (May-June) before collection (Finnan and Caslin, 2007) (Stromberg and Svärd, 2012) (Pahkala et al., 2005). The delayed harvesting favours crop fuel quality by reducing the alkali and other

problems associated to crop elements. Ash content of RCG ranges between 2-10% depending on fertilization patterns and soil type (Xiong et al., 2008) (Pahkala et al., 2005). RCG has a high ash content compared to woody fuels such as *Salix* (Encorp, 2009). However, the drawbacks of RCG are its susceptibility to various pests, prone to lodging and high fertiliser requirements, especially nitrogen.

Straw is a by-product to cereal production, which means that its use as a bioenergy feedstock does not have an impact on the land use for food and feed production. The energy yield is high in relation to harvesting and handling operations (Nilsson, 2010). However, the disadvantage of straw is its non-uniform composition and a low yield per hectare. Moreover, straw as fuel requires large volumes due to low bulk density (Stromberg and Svärd, 2012).

Industrial hemp (*Cannabis sativa* L.) belongs to the family of *Cannabaceae*, cultivated for fibre, pulp and paper, oil and energy applications. Average cultivation area of hemp in Sweden was only 829 hectare in 2007 with an average dry matter yield of 5.6 ton $ha^{-1}y^{-1}$ (Prade, 2011). Due to increasing drug problems, all commercial hemp production including cultivation and processing was banned in 1960s in Sweden. Advantages of hemp over available energy crops are its high biomass and energy yields, low pesticide requirements, good weed competition and suitability to fit into existing crop rotations. Drawbacks of hemp as feedstock are mainly its costly production (Kreuger, 2012) and lack of practical experience of hemp cultivation among farmers and advisors.

Salix is a deciduous woody perennial crop from the *Salicaceae* family with 400 species, of which 30 species exist in Sweden. *Salix* is a fast growing tree with long and straight shoots. Due to high yields of biomass and low production costs, *Salix* is regarded as a suitable bioenergy source. Osier (*Salix viminalis*) is mainly used for energy purposes and "vattenpil" (*Salix dasyclados*) to a much more limited extent. Crop breeding has resulted in new varieties resistant to diseases, pests and frost. Tora and Gudrun are examples of new varieties that currently are being used (Strömberg and Svärd, 2012). *Salix* is grown mainly in southern Sweden because it is frost sensitive, but there are ongoing attempts to develop more resistant varieties in northern regions (Söderström, 2008). In 2009, 13 700 ha of *Salix* were cultivated in Sweden. The calculated potential indicate that it would be possible to grow 100 000-300 000 ha, i.e. 3-10% of all agriculture land in Sweden (Baky et al., 2009). The life span of *Salix* is approximately 25 years and for energy purposes 3-5 year intervals between harvests are applied. Average yield of *Salix* is 8-10 tons dry matter $ha^{-1}\,y^{-1}$. Well-managed *Salix* cultivation can produce 12 tons dry matter $ha^{-1}y^{-1}$ (Strömberg, 2005).

Today, *Salix* is one of the best options for fuel production in Sweden due to the long experience of cultivation and well developed agricultural machinery. Furthermore, the chemical properties regarding alkali and ash content (Appendix A) will not likely cause any problems for low-temperature pyrolysis, and the production cost is competitive in comparison with the other feedstock considered (Appendix B). Due to the criteria mentioned above *Salix* is selected and extensively described in terms of production, supply and storage.

2.1.1 Salix

2.1.1.1 Agriculture operations

Salix is planted in spring as fully dormant cuttings in twin rows and special planting techniques and machinery are applied. Mechanical and chemical (glyphosate) weed control is applied until the root system is fully developed. If problems with fungal pathogens and leafeating insects occur they should also be managed. Foliage development of the plant helps shade out weeds (Monsanto International Sarl, 2010) (Encrop, 2009). Nutrients and water should be well supplied for a good harvest. *Salix* can be irrigated by wastewater in order to provide nutrients for growth and remove nitrate and other nutrients from the wastewater. Wastewater cleaning also referred to as "polishing" is thus a win-win situation providing a cheap alternative to conventional wastewater treatment and an ideal water and nutrient source for the energy crop (Rowe et al., 2009). Sewage sludge along with ash from biofuel combustion can also be used as a nutrient supply (ENCROP, 2009). However, mineral fertilisers are often used and the sequence and rate of fertilizer and pesticide applications are given in Appendix C. *Salix* is normally harvested between November and April, depending on the low water content (ca. 50%) of plant biomass and the high stability of frozen soil. *Salix* can be harvested as whole-stem (bundles and billets) [2] or chipped directly by the harvester. Direct chipping is the main commercial method for Salix harvest. However, biofuel production from biomass needs a continuous supply during the whole year, which disproves direct chipping due to poor storage properties of chips. Using bundles or billets instead of chips makes it possible to deliver *Salix* during the whole year (Baky et al., 2009). Based on the harvest method, different machinery has been developed. Reports show a loss of 1.2 and 3.4 ton dry matter per hectare during harvest (Meijden and Gigler, 1995).

2.1.1.2 Storage

The continuous production of biofuels requires secure and steady biomass supply during the whole year, while the harvest is made during a limited period of the year. Thus, long-term storage of the feedstock will be necessary. *Salix* moisture content ranges from 25 to 50 % (Strömberg and Svärd, 2012), while at harvest and at beginning of storage, moisture content is approximately 50%. If *Salix* is stored in piles as chips, loss of dry matter and energy occurs due to precipitation (loss of water soluble extractives), evaporation (loss of volatile non-structural cell wall component) and microbial degrading activity. According to Agblevor (1995) 14% of the dry matter is lost, while the higher heating value falls with 13% over a 12 month period during storage of *Salix* chips. Therefore, it is recommended to store *Salix* as whole timber (shoot) in long and wide piles well exposed to wind (air flows) and sun in order to achieve drying without substance and energy losses (Meijden and Gigler, 1995). Higher effective energy values are obtained when moisture content of biomass is reduced, and keeping the substance losses to a minimum. Studies show that a 10% decrease in moisture content results in a 5% increase ineffective achievable energy (Meijden and Gigler, 1995). Table 1 shows changes in energy content of *Salix* bundles and billets[2] during storage.

[2] Chips, billets and whole-stem are three different size ranges for *Salix* harvest. Chips (ca. 0-55mm) billets (50-250mm), whole-stem harvest (up to 8 meter long) (Forsberg, 2010). Bundle harvesting is a form of whole stem harvesting, where bundling is performed at harvesting stage.

Table 1: Specific energy content increase of Salix bundles and billets during storage (Johansson and Nilsson, 2009)

	Energy content – storage[a]	
	3 months storage	**6 months storage**
Bundled [MWh/ton]	2.9	4.0
Billet [MWh/ton]	2.9	3.8

[a] Energy content of newly harvested *Salix* is 2.2 MWh/ton

Agblevor (1995) reported the influence of storage on pyrolysis products. For biochar production no significant difference of char/ash yields between fresh and stored woody feedstock during storage were observed. However, in the case of biooil, losses were indicated by reduction in extractive contents. Woody biomass such as *Salix* contains polyphenolic components with higher higher heating value (HHV) than structural components of the cell wall. Loss of extractives in woody biomass will therefore account for a partly loss of HHV in the pyrolysis oils. Since lignin is highly aromatic, it has a higher HHV than carbohydrates. Loss of lignin through microbial activity during storage will therefore account for a reduced HHV of pyrolysis oil. However the study shows that in the case of very little microbial activity and weathering during storage there would be no significant changes in the HHV of pyrolysis oil. Storage of feedstock did not significantly affect gas yields (Agblevor, 1995). A study by Johnson (1994) show that small changes have been witnessed in the quantity of structural components such as lignin, cellulose and hemicellulose and substances extractable by ethanol 95 during storage or woody biomass.

2.1.1.3 Chemical composition and fuel quality

Besides the water content, the particle size of feedstock is of great importance for handling and preparation processes. *Salix* from young plantations have a higher proportion of bark partially enclosing the wood and makes the fuel dry considerably slower than older stands of willow. Bark proportion in wood fuels significantly affects fuel quality in terms of zink (Zn) concentration per ton harvestable shoot biomass. While the soil characteristic factor significantly affects the wood fuel quality in terms of phosphor (P) and potassium (K), as well as zink, nickel (Ni), cadmium (Cd) and copper (Cu) concentrations. Nitrogen content is a less important fuel characteristic compared with the concentrations of alkali metals and chlorine (Cl). Potassium and chlorine are involved in corrosion processes and potassium and sodium (Na) in slagging processes which cause technical complications during power plant operation. *Salix* has an acceptable concentration of such elements comparing to other crops (Appendix A). Calcium (Ca) and magnesium (Mg) usually increase the ash melting point, while potassium decreases it. High concentrations of zink and cadmium in biomass fuels can cause problems in ash recycling, since these metals accumulate in the ash during combustion or are emitted to the atmosphere as particulates (Adler, 2007).

2.1.1.4 Cost of production

Cost of *Salix* cultivation is higher for small fields. Establishment and cultivation costs of *Salix* include planting costs at the first year and fertiliser applications each year. Cost of harvest determines the economic efficiency of *Salix* cultivation which depends on harvest method, harvest machinery (capacity and intensity of utilization) and local conditions. Harvesting *Salix* as bundles is cost effective while the collection and transport (field and road) of bundles is costly mainly due to low volume weight of bundles (Baky et al., 2009). Storage costs are mainly related to unloading/loading before and after storage and interest costs for the invested capital (Appendix B)

2.2 PYROLYSIS AND BIOGAS UPGRADING DESCRIPTION

In the pyrolysis process, the biomass feedstock is heated to between 400°C and 800°C under anaerobic conditions, resulting in a decomposition that produces three products: bio-oil (condensable), gas (non-condensable under ambient conditions) and charcoal (solid char). Pyrolysis is currently used in several commercial plants and the purpose of these plants is often to produce one specific product, either bio-oil or charcoal. The allocation of the pyrolysis products yields depends on several process variables, e.g. heating rate, pre-treatment, final temperature, and residence time.

A lower heating rate (slow pyrolysis) favours the formation of charcoal while a high heating rate (fast and flash pyrolysis) increase the formation of bio-oil. Typical yields for some general thermal processes can be seen in Table 2. About 10% of the lower heating value (LHV) input is required to fulfil the heating demand in the process. The specific energy content in the products also varies with process and feedstock, e.g. dilution by water in the bio-oil and CO_2 in the gas. Water is also formed in the pyrolysis processes and this part results in about 8% water in the bio-oil on dry basis.

Table 2: Typical product yield for different thermal treatment (International Energy Agency, 2012).

Process	**Conditions**		**yield [weight basis, %]**		
	Temperature [°C]	Residence time	Bio-oil	Charcoal	Gas
Fast	~500	~1s	75	12	13
Intermediate	~500	~10-30s	50	25	25
Slow – Carbonisation	~400	hrs -> days	30	35	35
Gasification	~800-1,000		5	10	85

The largest markets for charcoal are the developing countries where it is used for cooking. During the 17th century, charcoal was used in the steel industry, but was later replaced by coke in most countries for economic reasons (Emrich, 1985). Charcoal has also been suggested as a soil improver and to act as a carbon sink when applied in the soil (Lehmann et al., 2006). Char from biomass is generally denoted “biochar” when intended to be used for soil

improvements and carbon storage while "charcoal" is the most common term for energy-related applications.

Today, the research is more focused on the development of the fast pyrolysis production route, where bio-oil is the main product. The advantage of bio-oil is that the high energy content enables transportation over greater distances. Even though bio-oil has similarities with fossil oil and about half of the heating value, it is a challenge to convert industrial processes to bio-oil use. The main problems are related to the inhomogeneous content, rapid ageing, and high viscosity. Both the ageing and viscosity problems can be solved by blending in methanol which is a requirement if the bio-oil is stored for a longer period (weeks) (Boucher et al., 2000). Several paths to upgrade the bio-oil to high quality fuels have therefore been suggested. The most discussed final product is hydrogen (Sarkarand Kumar, 2010; Iojoiuet al., 2007; Heracleous, 2011; Wang et al., 1997; Czerniket al., 2007), but there are proposals for upgrading to other transportation fuels: Fischer-Tropsch diesel (Ng and Sadhukhan, 2011a), petrol (Jones et al., 2009), and methanol (Ng and Sadhukhan, 2011b).

The suggested process is a poly-generation plant that produces biogas, bio-char, and heat via pyrolysis of biomass. Methane has not earlier been evaluated as a potential upgrading option (to our knowledge). A general process scheme for the biomass to biogas process can be seen in Figure 3. The process includes pre-treatment (drying and chipping), pyrolysis, fuel synthesis, and product upgrading. The pyrolysis gas[3] is immediately transferred to the fuel synthesis after char removal and sulphur cleaning, i.e. without intermediate condensation. The fuel synthesis consists of pre-reforming followed by water gas shift combined with methanation. The product is finally upgraded by removal of water and CO_2. This process have been evaluated for both fast pyrolysis (Larsson et al., 2013) and integrated in a charcoal production process using slow pyrolysis (Görling et al., 2013).

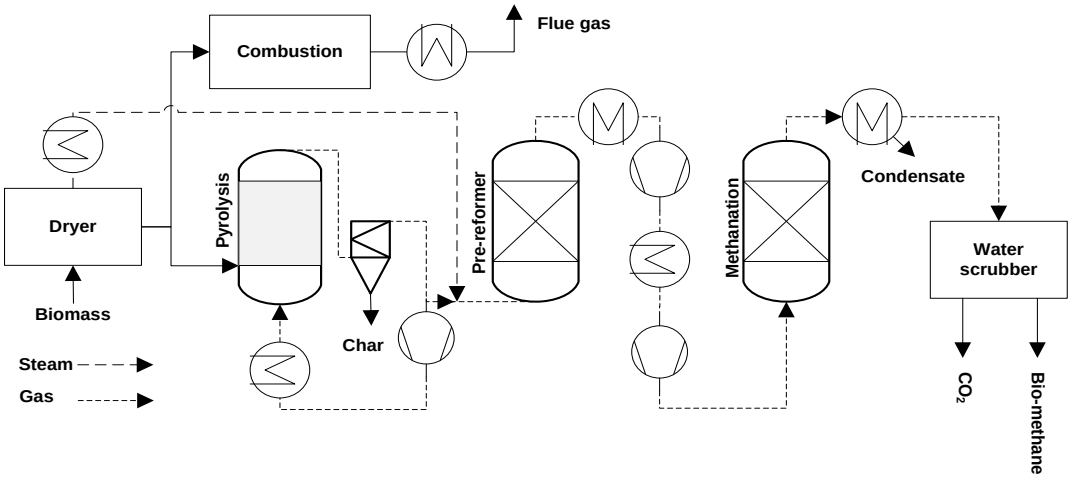


Figure 3: Flow sheet for bio-methane upgrading of pyrolysis gas (Larsson et al. 2013)

The first step in the fuel synthesis is the pre-reformer that crack long hydrocarbons into CH_4, CO_2, CO and H_2. When producing other final products than methane, complete reforming

[3] "Pyrolysis gas" refers to both the condensable fraction (bio-oil) as well as the non-condensable gas fraction.

(elimination of CH_4) is not desired as in several other processes. According to Wang et al. (1997), pre-reforming can be performed in an adiabatic fixed bed reactor filled with a nickel-based catalyst, operated at 500°C and atmospheric pressure. The operation and robustness of the process is affected by gradual deactivation of the catalyst in the pre-reformer as well as in the methanation reactor. The gradual deactivation of the catalyst is due to poisoning by alkali metals, compounds containing sulphur and/or nitrogen (Bulushev and Ross, 2011), and carbon deposition. However, alkali metals in the biomass are not as troublesome in pyrolysis as in gasification since they will remain in the char (van Rossum et al., 2007) and do not affect the catalyst in the downstream process steps. Carbon deposition on the catalyst in the pre-reformer and methanation reactor can be minimised by adding water vapour to the pyrolysis gas.

In the methanation reactor, CO_2, CO and H_2 are converted to CH_4. The water gas shift reaction also takes place in the methanation reactor, and it is necessary to avoid shortage of H_2 as well as attaining maximum conversion to CH_4. Methanation of biomass-derived syngas has been successfully tested in the Güssing demonstration plant; the tests resulted in low amounts of unreacted H_2 and CO (Schildhauer et al., 2007).

The produced gas after methanation contains CH_4, CO_2, H_2O and traces of CO and H_2. Up-grading by removing of CO_2 and condensation of water must be performed to attain the high methane content and quality required for vehicle gas or for injection into natural gas grids.

3. SMALL-SCALE METHANE PRODUCTION – CASE STUDY

A case study is used to illustrate the capabilities and the agricultural resources needed for a typical plant with $10MW_{LHV}$ biomass input. A general process flow sheet including the system boundaries can be seen in Figure 1. The plant is assumed to have an annual operation time of 7000 h, thus demanding biomass feedstock corresponding to 70 GWh/y. Assuming an average yield of 7 ton dry matter $ha^{-1}{\cdot}y^{-1}$ for *Salix* as feedstock, 13 263 ton dry matter biomass and 1 895 ha of land is required, based on a LHV of 18.6 MJ/kg ash free dry matter (0 % moisture in the biomass) and a yield of 6.86 tonnes of ash free dry matter per ha (2 % ash of dry matter). The *Salix* production process includes the cultivation and harvest phase, near-field storage, and transportation to the energy plant.

The main production from the system is biogas, but the process also co-generates bio-char and heat. The heat can be retrieved at different temperature levels and could, if possible, be sold as district heating or used for steam production.

3.1 ENERGY BALANCE FOR FEEDSTOCK PRODUCTION

The *Salix* cropping system is divided in three phases:

Phase 1: Establishment (soil preparation, planting and weed control)

Phase 2: Cutting

Phase 3: Field recovery

The lifespan of *Salix* is set to be 22 years with five cutting cycles and a four year harvest rotation. The establishment (phase 1) of *Salix* plantations consisted of ground preparation (harrowing, ploughing during autumn, and weed control (usually during the summer) in order to eradicate couch grass and perennial weeds. After field preparation, the planting is usually performed from mid-March to mid-June. Plantation density is 10 000–13 000 plants.ha^{-1} carried out in a twin row formation with 75–150 cm between rows and 59–65 cm between plants. Irrigation is not necessary and the plantation is properly weeded with herbicides. *Salix* easily fades out weeds by developing its canopy. In order to promote sprouting and ensure rapid canopy closure to reduce weed competition, the plants are cut back after the first year (Nilsson and Bernesson, 2008) (González-García et al., 2009; 2012).

During each cutting cycle (phase 2), crops will be fertilized, harvested and transported to the energy plant. For a whole year supply of material to the energy plant, a storage unit near the field has been considered. In the field recovery step (phase 3) mechanical and chemical weed control is performed and *Salix* stools are collected. All these activities take place in a 22 year rotation. Rate of fertilizer and pesticide applications are stated in the Appendix C. Average transportation distance is 10.5 km. Energy requirement by transportation from crop production units to the energy plant has been calculated based on Nilsson (1995).

The following tables present the total energy input for the *Salix* production process in order to supply 70 GWh/y of biomass feedstock. Table 3 shows energy inputs in the agricultural operations, consisting machinery implementation for soil preparation, plantation, mechanic weed

control, chemical spreaders, harvest and finally clipping and extraction of *Salix* stools including transportation to the pyrolysis plant. Energy input for chemicals include the primary energy for chemical production (pesticide and fertilizers). Energy for bundling harvested *Salix* and transportation to energy plant has been presented.

Table 3: Total energy input to Salix production based on different operations for a 22 year cycle.

Total energy input in *Salix* life cycle		**GWh**
Agriculture operations		36
	Machinery *11*	
	Chemicals *25*	
Transportation		1
	Total	**37**

Table 4 presents energy inputs into harvest cycles only (phase 2). As previously mentioned in the first cutting cycle in order to ensure rapid canopy closure and plants growth development, plants are cut back after the first year of plantation and harvest is done in year six. Cutting cycles 1-5 are similar in sequence of operations (Appendix C). Table 5 presents energy input to the triple-phase production process of *Salix*.

Table 4: Energy input in cycles 1 and cycle 2-5 (phase 2)

Total energy input for each cutting cycle	**GWh**
Cutting cycle 1	5.8
Cutting cycle (individual) 2, …5	6.8

Table 5: Total energy input in three phases of Salix production

Total energy input of phases		**GWh**
Phase 1	Establishment	2
Phase 2	Cutting	33
Phase 3	Field recovery	1

The main output of the sub-system is providing *Salix* feedstock (as an energy carrier) to the energy production unit (pyrolysis reactor). Statistically there is no significant energy loss during storage of whole stem *Salix* (Thörnqvist, 1982). Therefore, loss during storage is not included.

3.2 ENERGY BALANCE FOR BIOGAS AND CHARCOAL PRODUCTION

The results shown for the case plant is based on a fast pyrolysis process, which is more deeply described in Görling et al. (2013). Although, the production route was also considered using slow pyrolysis (Larsson et al., 2013), the fast pyrolysis reactor was chosen for this case study since sufficient input data made it possible to calculate the entire system.

The fuel synthesis and a part of the upgrading, including pre-reformer, WGS/methanation, and water removal, was modelled in Aspen Plus® (Aspen Technology, 2012), while the mass and energy balance for the other process steps were calculated from literature values. The simulations are indicative and not validated by experimental data. The total mass and energy balance was calculated taking the possibilities for process integration into consideration (minimum temperature difference 20°C). The process and how the calculations are performed are further explained in Larsson et al. (2013).

The results for the production process can be found in Table 6. The power demand is dominated by the compression work in order to increase the gas pressure before fuel synthesis and to re-circulate gas used to heat the pyrolysis reactor. The power consumption for final grinding is also included in the figures.

Table 6: *Overall results for biogas production (adopted from Larsson et al. 2013)*

Input:	**[MW_{LHV}]**	**[GWh_{LHV}/year]**	**GWh_{LHV}/cycle (22y)**
Biomass	10.0	70	1540
Electricity	0.6	4.2	92.4
Output:			
Bio-methane	7.0	49	1078
Bio-char	1.8	12.6	227
Heat >300 °C	0.6	4.2	92.4
Heat (80-300 °C)	0.6	4.2	92.4
Total	**10**	**70**	**1540**

3.3 TOTAL SYSTEM ENERGY PERFORMANCE

To supply the pyrolysis plant with sufficient amount of feedstock, approximately 2000 ha would be needed. The system performance in Table 7 is calculated both per year of operation and for the whole cycle for the *Salix* plantation (22 years). The annual energy usage for feedstock production and preparation is an average for the cycle.

Table 7: *Total system energy performance*.

	[GWh/year]	[GWh/cycle]
Feedstock preparation[4]		
Chemicals	1.1	25
Machinery	0.5	11
Transportation (farm to energy plant)	0.05	1
Pyrolysis production		
Electricity use	4.2	92
Total energy input	**5.9**	**150**
Energy output		
Biogas	49	1100
Biochar	13	280
Heat	8.4	190
Total energy output	**70**	**1600**

[4]The figures for GWh/year is an average for the 22 year cycle.

4. DISCUSSION AND CONCLUSIONS

The results from the simulations of methane production via pyrolysis can be compared to the corresponding processes using gasification. When comparing these two processes on a common basis (by allocating[5] the feed used for methane production) it is shown that the pyrolysis route have about 4%-points higher total efficiency (Görling, 2012). The higher efficiency is a result of lower losses due to lower process temperature and avoiding reaction losses when using syngas as an intermediate step. One issue evaluated in this work is which type of pyrolysis reactor (fast or slow) that is preferable. The major setback with fast pyrolysis is the power demand for pre-treatment and re-circulating of gas. However, the slow pyrolysis process yields a gas with lower specific energy content implying higher power consumption during the methanation steps. Consequently, the difference in power demand is smaller than first expected.

Small-scale bio-methane production is more suitable than large gasification plants for local markets with short transport distances of both feedstock and products. Even though the plant can be considered small in terms of biomass usage, the annual production is in line with the largest existing anaerobic digestion plants. The suggested annual production of about 50 GWh can be compared to the total Swedish production of upgraded biogas from digestion, which amounts to 734 GWh/y from sewage treatment and co-digestion plants (Swedish Energy Agency, 2011). The case plant can potentially supply about 10 000 light passenger cars or 200 city buses (based on the average distance and fuel consumption for compressed biogas vehicles).

The unused agricultural land in Sweden is between 300 000 and 400 000 ha (Baky et al. 2009).A large part of this is of low quality and consists of small fields within the range of 1-6 ha, which is a size that may be of great interest for *Salix* production. It is estimated that 250 000 ha of this land is possible to utilize for *Salix* cultivation by well-designed cultivation systems (Baky et al. 2009). If 250 000 ha of the unused agricultural land was used for biogas production, it could supply in the vicinity of 125 “case plants”, producing in total 5.8 TWh/y biogas. However, the main production costs are related to harvest and handling of *Salix*. Therefore, it is necessary to further develop whole stem harvest systems for relatively small fields in order to achieve a secure supply of storable biomass for pyrolysis plants.

Biochar, as a product of pyrolysis, is of great interest due to its stable structure and energy value which can maximize the energy efficiency of the pyrolysis facility. However, biochar can also be applied as a soil amendment with the aim to enhance the physical and biological properties of the soil. Studies have revealed that the addition of biochar to soils will improve the supply of nutrients to crops and increase plant growth (Glaser et al., 2002). According to Gaunt and Lehmann (2008), the net effect on emissions of greenhouse gases is between 2 and

[5] Inputs and outputs are recalculated on a common basis, biomass in and methane out. Used electricity and produced charcoal is converted to biomass using 33% and 85% conversion efficiency, respectively (input reduction/addition). The biomass to methane efficiency is calculated as follow:

$$\eta_{Biomass\ to\ Methane} = \frac{Q_{Methane}}{Q_{Biomass\ input} - \frac{Q_{Biochar}}{0.85} + \frac{P_{el}}{0.33}}$$

5 times higher when biochar is applied to agricultural land compared to when biochar is used solely for replacing fossil fuels.

4.1 RECOMMENDATION FOR FURTHER STUDIES

The promising energy balance of the suggested route for biogas production via low-temperature pyrolysis using *Salix*, and the possibility to implement the system in small-scale make additional studies interesting. Production economy is obviously crucial to the attractiveness of the process and this needs to be further investigated. A life cycle assessment is also recommendable in order to compare pyrolysis and *Salix* with other biofuel production routes and crops in terms of environmental performance. The LCA should also include the alternative use of biochar as an energy source or as a soil improver. Further studies of the long term effects and deactivation of catalyst for pyrolysis vapour pre-reforming are also necessary to continue the technical development, since this may be one of the most technically challenging steps in the process. Another important production step is the gas cleaning process which needs further attention and testing.

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APPENDIX A: CHEMICAL CHARACTERISTICS OF DIFFERENT FEEDSTOCK

(Adopted from (Strömberg and Svärd, 2012)

Elemental analysis (%dm ash free)[1]				
	Salix	RCG	hemp	straw
C	49.8	48.6	49.6	48.32
H	6.12	5.89	6.1	5.93
O	43.6	43.7	43.8	44.89
S	0.04	0.18	0.06	0.08
N	0.50	1.16	0.7	0.6
Cl	0.02	0.82	0.28	0.12
Ash analysis (% ash) [1]				
Al	0.5625	0.2805	0.8867	0.2434
P	3.8009	3.2292	2.0000	2.3565
As	0.0006	-	0.0012	
Ba	0.0367	-	0.0301	
Ca	24.4425	6.6466	22.3333	9.0766
Cd	0.0092	-	0.0005	
Co	0.0032	-	0.0011	
Cr	0.0150	-	0.0087	
Cu	0.0150	-	0.0241	
Fe	0.6000	0.3287	1.8182	0.4057
Hg	0	-	0	
K	12.6182	12.9503	4.4667	15.2747
Mg	2.2000	2.1710	1.4091	2.0505
Mn	0.2250	0.0387	0.2592	0.0620
Mo	0.0357	-	0.0016	
Na	0.7626	0.7047	0.3533	0.6677
Sb	0.0256	-	0	
Si	7.9469	21.8306	14.5625	21.7371
Ti	0.0563	-	0	
V	0.0013	-	0.0033	
Zn	0.3440	-	0.0900	
Other characteristics				
Lignin (% dm)	13.79[3]	14[4]	14-17	0.49-1[2]
Cellulose (% dm)	55.94[3]	28[4]	23-37	3.62-4.06 [2]
Hemicellulose (% dm)	13.96[3]	22[4]	-	2.18-3.6 [3]
Volatiles (% dry ash free)	81.6	-	-	-
Moisture (w %)	12.6	14.4	9.8	12.4
Ash (w % dm)	2.1	5.9	2.2	4.95
Density (kg/m^3)	200-350	-	-	
Heff (MJ/kg)(dry % ash free)	18.6	17.66	18.47	17.56
Heff (MJ/kg) (wet)	15.9	13.84	15.53	14.51

1. Mean amounts
2. Antongiovanni and Sargentini, 1991
3. Szczukowski et al., 2002
4. Reed canarygrass (online) available at: http://www.ienica.net/crops/reedcanarygrass.htm [assessed 1 june 2012]

APPENDIX B: COMPARABLE COSTS

Comparable cost of *Salix*, hemp, reed canary grass (RCG) and straw production in main agricultural areas of Sweden are presented in table 1. Figures show low production costs for *Salix* and straw in comparison to hemp and RCG. Main costs related to *Salix* are establishment and harvest cost. In table 2 storage of *Salix* has not been considered due to the direct use of Salix as chips. In case of a whole year supply to the energy unit, storage must be considered. At a *Salix* price of 200 SEK/ MWh the storage costs after 3 and 6 months are 4.5 SEK/ MWh and 10 SEK/ MWh for bundles (Baky 2009).

Table 1. Approximate figures of production cost in (SEK/MWh) for energy crops (Baky, 2009).

Production area	***Salix***	**Hemp**	**RCG**	**Straw**
Götalands south plains (Gss)	130	318	222	150
Götalands *mellan* plains (Gms)	160	318	225	
Götalands north plains (Gns)	140	325	228	
Svealands plains (Ss)	153	330	232	
Götalands woodland (Gsk)	168	345	236	-
Central Sweden woodlands (Ssk)	188	351	239	-
Lower Norrland (Nn)	-	362	239	-
Upper Norrland (Nö)	-	362	239	-

Table 2. Distribution of costs in percent for different crops at certain yields. Land costs are not included. The share of costs related to harvest and transport increase with an increase of the yield (Baky, 2009).

	Salix	Hemp	RCG	Straw
Harvest (ton dm/ha)	8	6	5	-
Costs				
Establishment	20	29	6	0
Fertilization	18	9	23	9
Harvest and chip transport	25	24	28	39
Storage	0	11	12	22
Road transport	16	12	10	18
Sales	6	3	4	6
Supervision and other	6	4	6	1
Overhead	10	7	12	5

APPENDIX C: SALIX PRODUCTION SCHEME

phase	Time (year)	Operation		Machinery implemented	Material inputs	amount of material input per ha
Phase 1	-1	Pre-planting	Weed control	spreader	Roundup	2,43 kg
			Disking	Harrow		
			Ploughing (*harvning*)	Plough		
	0	Planting	Planting	Planter	stems	
			Weed control	spreader	Cougar	0,6 kg
			fertilising	spreader	(P-K)	(40-120) kg
Phase 2	1	1st cutting cycle	cut back (*putsning*)	Betesputs		
	2		fertilising		N	80 kg
	3		fertilising		N	120kg
	4					
	5					
	6		Harvesting	harvester		
	6		transport to field corner	tractor and tippvagn		
	6		transport to storage			
	6	2 nd cutting cycle	fertilising	spreader	(N-P-K)	(80-40-120) kg
	7		fertilising	spreader	N	120 kg
	8					
	9					
	10		harvesting	harvester		
	10		transport to field corner	tractor and		
	10		transport to storage			
	10	3rd cutting cycle	fertilising	spreader	(N-P-K)	(80-40-120) kg
	11		fertilising	spreader	N	120 kg
	12					
	13					
	14		harvesting	harvester		
	14		transport to field corner	tractor and		
	14		transport to storage			
	14	4rd cutting cycle	fertilising	spreader	(N-P-K)	(80-40-120) kg
	15		fertilising	spreader	N	120 kg
	16					
	17					
	18		harvesting	harvester		
	18		transport to field corner	tractor and tippvagn		
	18		transport to storage			
	18	5th cutting cycle	fertilising	spreader	(N-P-K)	(80-40-120) kg
	19		fertilising	spreader	N	120 kg
	20					
	21					
	22		harvesting	harveter		
	22		transport to field corner	tractor and tippvagn		
	22		transport to storage			
phase 3	22	Field recovery	Weed control	spreader	Roundup	2,43 kg
			Trimming			
			Extraction of stools	disking		
			Collecting stools			

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