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Techno-economic study of NMMO pretreatment and biogas production from forest residues

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ABSTRACT

Biogas is nowadays getting more attention as a means for converting wastes and lignocelluloses to green fuels for cars and electricity production. The process of biogas production from N-methylmorpholine oxide (NMMO) pretreated forest residues used in a co-digestion process was economically evaluated. The co-digestion occurs together with the organic fraction of municipal solid waste (OFMSW). The process simulated the milling of the lignocelluloses, NMMO pretreatment unit, washing and filtration of the feedstock, followed by an anaerobic co-digestion, upgrading of the biogas and de-watering of the digestate. The process also took into consideration the utilization of 100,000 DW (dried weight) tons of forest residues and 200,000 DW tons of OFMSW per year. It resulted in an internal rate of return (IRR) of 24.14% prior to taxes, which might be attractive economically. The cost of the chemical NMMO treatment was regarded as the most challenging operating cost, followed by the evaporation of the washing water. Sensitivity analysis was performed on different plant size capacities, treating and digesting between 25,000 and 400,000 DW tons forest residues per year. It shows that the minimum plant capacity of 50,000 DW tons forest residues per year is financially viable. Moreover, different co-digestion scenarios were evaluated. The co-digestion of forest residues together with sewage sludge instead of OFMSW, and the digestion of forest residues only were shown to be non-feasible solutions with too low IRR. Furthermore, biogas production from forest residues was compared with the energy produced during combustion.

KEYWORDS: anaerobic digestion, NMMO pretreatment, lignocellulose, forest residues, economic analysis

HIGHLIGHTS

- Biogas from co-digestion with pretreated forest residues was simulated and evaluated
- Plant capacity of > 50,000 DW tons/year forest residues is financially feasible
- The cost of the NMMO was regarded as the largest operating expenditure
- Biogas production was compared with the energy produced during incineration

1. INTRODUCTION

The global market and demand for biogas as a vehicle fuel, electricity production, and even as a heating energy source has had a positive trend. The biogas is produced in household digesters to provide cooking or lightening energy to replace kerosene or LPG, while the larger plants burn it in gas engines to produce electricity or upgrade it to almost pure methane to inject in the gas grids or compress it to CBG (compressed biogas) and sell as car fuel. The traditional substrates utilized for biogas production are municipal solid waste, organic wastes from industrial and agricultural activities, as well as high strength wastewater are. However, these sources are limited, and there is a demand for the development of new technologies utilizing other substrates. Lignocellulosic-rich materials have a great potential as an alternative feedstock for anaerobic digestion, since they are found in high abundance globally.

The degradation of lignocelluloses into biogas is a complicated process, since lignocelluloses have a recalcitrant structure which is naturally designed to prevent enzymatic degradation. Lignocelluloses are formed in a compact and crystalline structure and often contain a high amount of lignin. In order to permit degradation of these materials in an anaerobic digester, the structure has to open up and/or the lignin has to be degraded or removed. This can be performed by using different pretreatment methods [1], such as mechanically, e.g., by milling; physically by steam explosion or radiation; chemically by acids, bases or solvents; and biologically by enzymes or fungi [1-3].

Solvent pretreatment on lignocelluloses was shown to be an effective method due to the low degradation of the carbohydrates in the material under the applied, relatively mild conditions. Furthermore, pretreatment with a solvent does not require neutralization, and almost a complete recirculation of the treating chemical is possible [4]. The pretreatment using the solvent N-methylmorpholine oxide (NMMO) has previously been studied on bagasse [5] and on spruce [6] for ethanol production, and on spruce, rice, and triticale straws [7] as well as pure cellulose [8] for biogas production. NMMO is an organic solvent that interrupts inter- and intra-molecular bonds [9] in the lignocelluloses, making the carbohydrates of the material more accessible and thereby facilitating the enzymatic degradation. NMMO is an environmentally friendly cellulose solvent, and used in industrial scale in the lyocell process [10, 11], where cellulose fibers are treated to produce textile. Since no toxic compounds are produced within the NMMO pretreatment and the recirculation of the solvent is possible [10, 12], this process can be regarded as environmentally friendly.

Techno-economic analysis is a useful tool to examine the profitability and performance of a proposed process. Recently, Shafiei et al [6] performed a techno-economical study on bioethanol production from NMMO pretreated wood. They found that the process is feasible when bioethanol production is combined with a subsequent biogas production utilizing the pentoses. Conversion of lignocellulosic pentoses to ethanol is one of the obstacles in the utilization of lignocelluloses to ethanol, since the ordinary industrial yeast species are unable to assimilate pentoses [13]. Furthermore, the production of biogas from lignocelluloses has several advantages compared to bioethanol production, since the overall energy efficiency is much higher in biogas production compared to that in ethanol production [14].

The focus of this study was therefore to develop a feasible industrial process for NMMO pretreatment and subsequent utilization of forest residues (branches, tops, barks, and needles) in anaerobic digestion. Forest residues were selected because they are the most abundant lignocellulosic waste stream in Sweden, and several other countries. In 2008, 1.6 Mtons total solids (TS) /year of the tree tops and branches were delivered from the forests in Sweden, and this is expected to increase to 3.5 Mtons total solids /year by 2018 [15]. Moreover, the total energy potential of bioenergy production from the forest is calculated as being 49 TWh [15]. An industrial scale process was designed and simulated using SuperPro Designer[®] 8.0 simulation software (Intelligen, Inc., NJ, USA) based on unpublished biomethane potential test (BMP) experimental data provided by Kabir et al. [16]. A process including an NMMO pretreatment step with filtration, evaporation and recirculation of NMMO and washing water together with a following co-digestion step was evaluated to determine economic feasibility and profitability, such as capital costs for the total plant, annual operating costs, and unit costs. Finally, sensitivity analyses were performed on different scenarios, where effects of the plant size, different co-digestion set-ups as well as the methane price and the water consumption were evaluated.

2. PROCESS DEVELOPMENT AND FINANCIAL ANALYSIS

2.1 Process description

A novel process of the NMMO pretreatment of forest residues prior to anaerobic digestion was developed. The process includes the feedstock handling, pretreatment by NMMO, anaerobic digestion, and upgrading of the biogas as well as the dewatering of the digestate. It is assumed that the plant is located close to a power plant, so that steam and electrical power are readily available. It is further assumed that the plant is situated in Sweden with a high availability of

forest residues. The type of forest residues investigated in this study includes the rejected tops and branches.

The base case is constructed for 100,000 tons DW (dry weight) forest residues per year. However, capacities ranging from 25,000 to 400,000 tons DW forest residues/year were also studied. The plant is in operation for 7,920 h/year, and the construction material was chosen to be stainless steel 304. The cost index was set at 2012.

2.2 Pretreatment unit

The forest residues arrive at the plant in truck trailers, where the price of the feedstock includes the handling all the way to the plant. The feedstock contains 42% carbohydrates, 44% lignin, 75% total solids (TS), and 64% volatile solids (VS) [17]. The forest residues have a C/N ratio of 325 [18]. The raw material is then placed into a grinder, which reduces the size of the biomass to 2 mm. After grinding, the biomass is conveyed to the pretreatment unit. The pretreatment is performed using 85% NMMO solution in water for 12 h at 90°C. During the pretreatment, the lignocellulosic structure is opened up, resulting in less intra-molecular linkages and less cellulosic crystallinity [9]. The pretreated biomass is then washed with water and filtered using a rotary vacuum filtration unit (Figures 1 and 2). The NMMO-solution is then evaporated back to 85% for reuse in the pretreatment unit. The recovery in the washing step is expected to be 99.5%. The use of the rotary vacuum filtration allows for a minimum usage of water during the washing, in order to save energy in the following evaporation unit. Previous experimental studies were performed with 500 mL washing water for 200 g NMMO/biomass mixture [16], where these conditions were applied in the base case of the simulation study. The evaporation unit was designed with a mechanical vapor design (MVR). The MVR design with two effects and two compressors was found to be the most energy efficient and an economically beneficial alternative for the evaporation of NMMO water solution in a previous investigation, focusing on NMMO pretreatment of spruce prior to ethanol production [6]. The same design for the evaporation step was applied in this study.

2.3 Biogas and digestate production

The washed and pretreated forest residues are mixed with the organic fraction of municipal waste (OFMSW, Figures 1 and 2), in order to achieve a C/N ratio of between 20-30 which is regarded as the optimum ratio [19]. Two-thirds of the OFMSW and one-third of the forest residues are used in the base case, which results in a C/N ratio of 30. The OFMSW in the simulation consists of 60% carbohydrates, 17% fats, 8% proteins based on the dried weight, and the water content was estimated to be 67 % water. The cost of OFMSW is set to zero. The methane yield of similar substrate mix was 0.470 Nm³/kg VS [20], which corresponds to a conversion rate of 86.7%. The methane production from forest residues is based on experimental results from lab scale BMP tests showing a yield of 0.137 Nm³ CH₄ per kg total solids of forest residues [16], which corresponds to a conversion rate of 73.4%. The two fractions are together passed through a screw press prior to the anaerobic digester, together with extra water in order to reach a TS of 12% in the incoming stream. The digester runs at thermophilic conditions (55°C) with a hydraulic retention time of 20 days. It is a fixed roof storage tank, which allows for mixing, constructed of stainless steel. The gas produced is a mixture of the main components methane and carbon dioxide, and trace amounts of some other components, such as hydrogen sulfide, nitrogen, and hydrogen, which are neglected in the study. The gas produced in the anaerobic digester is upgraded to 98% methane content, using the water scrubber technique. The water scrubber technique is regarded as a low cost technique [21], and is globally the most widespread upgrading technique [22]. This upgrading step consists of a gas compressor, an absorption tower where the carbon dioxide is absorbed in water, and a degasification tower, where the carbon dioxide and water are separated. The upgraded methane is then injected into the biogas/natural gas grid. The solid residuals remaining from the process, so called digestate residues, are dewatered in a centrifugal separator to 45% TS, together with 10 kg flocculating agent polyacrylamide per ton TS, in order to improve the dewatering process [23]. The solid fraction after the dewatering step is lignin-rich, which has a high heating value, and can be used as fuel for combustion in combined heat and power (CHP) plants [24]. In this study, the dewatered digestate is sold to CHP plants. However, due to the high nutrient value, the digestate residue can also be used as a fertilizer in agriculture or on forestland. Consequently, the dewatering process would then be unnecessary.

2.4 Process simulation and economic calculations

SuperPro Designer[®] 8.0 (Intelligen, Inc., NJ, USA, licensed to the University of Borås) was used for the simulation of the main steps of the process. The software performs the rigorous material and energy balance calculations. The purchase costs of the equipment were calculated with the built-in software calculations, except for the purchase cost of the tanks, which was calculated according to Turton et al [25]. Other than the purchase costs, SuperPro Designer estimates the cost for the installation, the process piping, instrumentation, insulation, electrical utilities, buildings, yard improvements, and auxiliary facilities. The total direct plant cost (DC) is a sum of these costs and was 329% of the equipment purchase cost at base conditions. The total indirect plant costs, such as engineering (25% of DC) and construction fee (35% of DC) was based on the equipment purchase cost, and was obtained by the above- mentioned software. The fixed capital investment (FCI) was calculated as a sum of the direct costs, the indirect costs, the contractor's fee, and the contingency. The contractor's fee and contingency were estimated to be 5% and 10%, respectively, of the sum of the direct cost and the indirect cost together [26]. The project is regarded as 100% equity financed. The project life is set to 20 years and the depreciation period to 10 years. The construction period is set to 30 months and a startup period of 4 months is used. The working capital was assumed to be 5% of the fixed capital investment [27], and the cost index for all calculations was set at 2012.

The annual operating cost was calculated as the sum of the expenses for raw materials, utilities, labor, waste management, and facility dependent cost and can together with the product prices be found in Table 1. The maintenance and insurance costs are regarded as facility dependent operating costs, and are together 1 and 2%, respectively, of the total plant capital costs [28-30]. The methane price used in the present study was the price of methane sold in the market in Sweden, minus the cost for the connection and distribution into the gas grid, including compression and cost for tank stations. The methane price used in this study was 1.895 euro/kg [31]. A value of 22% taxation rate is assumed, which is the current corporation tax in Sweden since 2013 [32].

Furthermore, the plant was divided into sections in order to determine the cost distribution for the different parts of the plant. These calculations were performed using the base case.

2.5 Sensitivity analysis

Different plant sizes were investigated in a sensitivity analysis in order to study the effect of the capacity on the construction and production costs. Plant sizes with the feed capacity of 25,

50, 100, 200, and 400% of the base case were studied. The cost prediction of the total investment costs, annual operating costs, and production cost per unit methane produced, as a function of the plant capacity was studied and simulated. Cash flow analysis was performed where the net present value (NPV) was set to zero and the process time was equal to 20 years. The internal rate of return (IRR) was calculated, and was regarded as being financially feasible at 15% rate of return (IRR) or higher, in order to cover the firms costs of raising funds and making a sufficient profit [33]. The IRR is the discount rate, when the NPV is set to zero and was calculated as [33]:

$$NPV = \sum_{t=0}^n \frac{A_t}{(1+r)^t} = 0$$

Where:

NPV , t , n , A_t and r are net present value, project year, total project lifetime, the cash flow in year t , and the discount rate, respectively.

The cash flow analysis was performed in order to study the effect of the methane price, the water consumption in the washing step following the NMMO pretreatment, and the price of the feedstock on the economic feasibility of the process under different scenarios. A co-digestion study where the forest residues were co-digested with sewage sludge instead of OFMSW was also performed, as well as a scenario where only forest residues were digested.

3. RESULTS

3.1 Process development and economic calculations

The plant was divided into five different sections (1) the NMMO pretreatment, (2) the filtration and evaporation following the NMMO pretreatment, which also includes the recirculation of water and NMMO, (3) the anaerobic digestion of both forest residues and OFMSW, (4) the upgrading of the biogas, and (5) the dewatering of the lignin-rich digestate. The fixed capital investment (FCI) for the different sections can be found in Figure 3. The most capital-intensive sections are the anaerobic digestion, followed by the filtration and evaporation, and the upgrading. Auxiliary capital investments, buildings, and yard improvements are excluded from the calculation.

A block flow diagram of the process is presented in Figure 1, which gives an overview of the process. The material composition of the streams in the block flow diagram is presented in Table 2. The developed process flow sheet, showing the equipment used in all processes, is presented in Figure 2. All process steps were run continuously, except the NMMO-pretreatment reactor, which was operated in batch mode. For this purpose, four staggered NMMO pretreatment reactors of each 970 m³ were used to perform 1975 batch pretreatments per year. The base case was considered to pretreat and utilize 100,000 DW tons forest residues/year, together with 200,000 DW tons OFMSW/year, and the plant was calculated to produce about 975 GWh (98 MNm³) methane per year. The produced amount of dewatered digestate and carbon dioxide are 290 and 133 kt/year, respectively. The consumption of electricity was 48 GWh per year, steam 355 GWh, and water 5,043 kt per year. In order to pretreat 100,000 DW tons forest residues per year, six batches of NMMO treatment per 24 h were performed. The fixed capital investment (FCI), is a sum of direct fixed capital, working capital, and startup cost, and was calculated for the base case as being 145,053,000 €. The annual operating cost is a sum of raw materials, labor costs, energy and power, waste management, as well as facility dependent costs. For the base case, this cost was calculated as being 103,810,000 €/year. The total revenue per year is a sum of the revenues of produced methane, carbon dioxide, and the dewatered lignin-rich digestate. The annual revenue for the base case was calculated as 136,179,000 €/year. This gives a net profit value (taxes and depreciation are included) of 181,333,000 €, at 7.0% interest rate over 20 years project lifetime. A cash flow analysis, with the net profit value set at zero resulted in an internal rate of return of 24.14% prior to taxes, and 20.39% after taxes, at a process time of 20 years.

The costs for the distribution of the upgraded methane into the distribution gas grid, were calculated according to as described by Benjaminsson and Linné [31]. The authors performed a techno-economic study of 300 GWh biogas plant in Sweden. For this size of plant, the cost of a gas pipeline for 40 km connected to the distribution gas grid was 0.001 €/kWh, the distribution cost 0.007 €/kWh, and the compression and tank station cost was 0.012 €/kWh, a total of 0.020 €/kWh. Calculating with an 8% price increase in Sweden between 2007 and 2012 [34], the price for gas grid distribution, compression, and tank stations are set to 0.285 €/kg methane.

The total annual operating costs divided into different cost items are presented in Figure 4. The costs of the raw materials have the highest share of operating costs, followed by facility dependent costs, which include maintenance, depreciation, insurance, and other factory

expenses. The cost for the NMMO corresponds to 80% of the material cost for the base case with 99.5% recirculation, and the cost for the forest residues corresponds to 15%. The annual operating cost divided into the different sections is presented in Figure 5, where the price of the materials is excluded. Filtration and evaporation represent the biggest part of the annual operating costs, followed by the anaerobic digestion, where the costs for materials are excluded.

3.2 Sensitivity analysis

Different plant sizes were investigated in a sensitivity analysis in order to study the effect of the plant capacity on the construction and production costs. Plants treating 25, 50, 100, 200, and 400 thousand DW tons forest residues per year were studied in co-digestion with 50, 100, 200, 400, and 800 thousand DW tons OFMSW per year, respectively. All the estimations of total investment costs, annual operating costs and production cost per unit methane produced, as a function of the plant capacity is presented in Figure 6. The revenue per unit was calculated as being 2.12 €/kg produced methane, which is higher than the production cost for all plant sizes. However, a cash flow analysis of the five different plant size scenarios show that only plant capacities of 50,000 tons per year and above are financially viable with an IRR over 15%. This is in contrast to the IRR of the plant size of 25,000 tons per year, which was 5.08% prior to taxes.

The economic feasibility of the process was further analyzed through different scenarios. The effect of water consumption in the washing process following the NMMO pretreatment was evaluated with 50% more and 50% less water consumption. The effect of 20% increase and 20% decrease on the methane price and the cost of feedstock was also calculated. Cash flow analysis was performed and the resulting IRR's were compared with the base case and are presented in Figure 7. The water volume during the washing step following the NMMO pretreatment has a large effect on the IRR. The use of more water during the washing step requires a larger and more expensive evaporation unit, which in turn results in a lower IRR. Furthermore, the price of the produced methane has a large impact on the IRR, while the cost of forest residues has a minor effect.

3.3 Co-digestion scenarios

In order to achieve a proper C/N ratio, forest residues can be co-digested with other nitrogen rich substrates. Sludge from wastewater treatment (sewage sludge) has been studied as an alternative co-digestion source. Due to the high nitrogen content, one part of sewage sludge together with two parts of forest residues result in an optimum C/N ratio of about 20, compared with two parts of OFMSW and one part of forest residues in the base case (Table 3). In Sweden, biogas plants get paid for the digestion of sewage sludge (Table 1), which will increase the unit revenue. However, our calculations showed that the co-digestion with sludge results in a unit production cost of 2.78 €/kg and a unit revenue of 2.75 €/kg. The IRR of the process was calculated as being 3.52% (Table 3), which is lower than the financially feasible limit of 15% and is therefore considered to be a non-feasible solution.

The process can be further designed to digest forest residues exclusively, which is not a real scenario, since it is unfavorable to digest forest residues by itself due to the low nitrogen content. However, the simulation of the pretreatment and anaerobic digestion of forest residues only can give us a better insight in the contribution of forest residues in the co-digestion process. With the exclusive digestion of forest residues, the IRR is negative (Table 3). The unit production cost has increased to 9.35 €/kg CH₄, while a higher unit revenue comes from the higher fraction of lignin in the digestate residue which was sold to a combustion plant.

Moreover, a sensitivity analysis has been performed in order to study the effect of different scenarios when only the forest residues are digested (Table 4). The effect of circulation of NMMO was evaluated, as well as the effect of the methane price. An increase in the recirculation of NMMO from 99.5% to 99.99% will decrease the unit production cost by a factor of three, while an increase in methane price increases the unit revenue. The unit revenue was the same as the unit production cost, after an increase of the NMMO recirculation to 99.99%, together with a methane price increase of 25%. However, none of the present scenarios reached the targeted IRR of 15% (Table 4).

3.4 Anaerobic digestion versus combustion

The energy produced from anaerobic digestion of NMMO-pretreated forest residues can be compared with the energy production of the same amount of forest residues when incinerated. Combustion of the feedstock in a combined heat and power plant (CHP) will produce 17 MJ/kg TS, with the assumption of 90% efficiency in the CHP [35]. On the other hand, when biogas

produced from the anaerobic digestion of only forest residues is utilized in a CHP with 90% efficiency [36], the energy generated can be calculated as being 12 MJ/kg TS. This is with the assumption that the lignin-rich residue from the anaerobic digestion is combusted separately, and the energy produced by this process is included in the above-mentioned calculation. Both processes are assumed to yield similar fractions of electricity and heat. It can, therefore, be concluded that the combustion of forest residues in CHP will yield about 1.5 times more energy compared with that in the anaerobic digestion.

There are another aspects that should also be considered when comparing anaerobic digestion or combustion of forest residues. Utilization of these materials for vehicle fuel production is only possible if they are converted to biogas. There is a large demand for alternative fuels produced from renewable resources worldwide, since a considerable part of the total greenhouse gas emissions originates from the transport sector [37]. Moreover, the organic nutrients cannot be retained and recycled back to soils after combustion, which in turn will result in the removal of structural material from the soil. On the other hand, the digested residue left after anaerobic digestion can be utilized as a sustainable fertilizer. Additionally, combustion is also connected with other serious problems as well, such as fly ash disposal and super heater corrosion.

4. DISCUSSION

The anaerobic digestion of NMMO pretreated forest residues, co-digested with household organic wastes in the base case is an economically viable process, with an IRR over 15%. The analysis of different sections of the process shows that the price of the raw material, i.e., NMMO, used for the pretreatment has the largest share of the costs. A challenge for the future is to increase the recirculation of the NMMO, in order to limit the consumption of the raw material, and thereby the costs. Furthermore, evaporation of the washing water is a costly process, and solving the technical challenge of using less washing water should further improve the economy of the process.

In order to reach a financially viable process for the digestion of pretreated forest residues, the methane price needs to be increased substantially. This could perhaps partly be reached by incentives in order to increase the fraction of renewable vehicle fuels production, together with increasing oil price. The European Commission has set the goal that by 2020, 20% of the energy consumed and 10% of the vehicle fuels should be renewable [38]. Furthermore, the cost of gas injection into the gas grid and the cost of the tank stations are probably reduced with larger plant sizes as is the case in the present study.

The use of the biogas produced from the anaerobic digestion of the NMMO pretreated forest residues in a CHP plant was shown to be a less attractive alternative compared with the combustion of the same amount of forest residues. These two processes, however, produce electricity and heat, while the anaerobic digestion process produces high-valued vehicle fuel. Another advantage of producing biogas from the forest residues, compared with combustion, is that the digestion of the feedstock results in a rich solid residue. In this study, this residue is calculated as being sold to combustion plants. As an alternative, it could also be used as a nutrient rich fertilizer. The use of the solid residue as a fertilizer is a sustainable way of recycling the nutrients back into the soil, and also structural material being placed back into the soil.

Compared with co-digestion of forest residues with OFMSW, the digestion of only pretreated forest residues has a negative IRR. The scenario of digesting only forest residues however, is a fictive scenario, since an optimal C/N ratio of 20-30 should be reached for a sufficient nutritional balance in the digester. Therefore, a co-digestion of nitrogen-rich substrates together with forest residues is required. Many digesters with e.g. sewage sludge or protein-rich substrates have problems with a too low C/N ratio, which means a lack of carbohydrate-rich

substrates. Addition of carbon-rich materials, such as lignocelluloses, was previously shown to both stabilize sensitive processes as well as result in good synergetic effects [39]. These synergetic effects have implied higher methane yields when a lignocellulosic-rich material (i.e., paper tube residuals) has been digested with nitrogen-rich substrate mixture compared to the expected methane production calculated from the methane potentials of the single substrate streams alone. The co-digestion of NMMO-pretreated forest residues with OFMSW has not yet been experimentally studied, but similar synergetic effects can be assumed, which can lead to higher methane yields and a more economically feasible process. The anaerobic co-digestion of pretreated lignocelluloses has not yet implemented commercially, but could emerge in the future.

5. CONCLUSIONS

The possible co-digestion of NMMO pretreated forest residues together with the organic fraction of municipal solid waste is an economically feasible process with an IRR over 15%. In order to avoid nitrogen deficiency, one-third of forest residues were co-digested with two-thirds of OFMSW. Technical improvements such as increased recycling rate of the NMMO solvent, as well as decreased water consumption in the washing step can further increase the economic viability of the process. The co-digestion with sewage sludge instead of OFMSW resulted in lower methane yields, which had a negative effect on the process economy. In general, the co-digestion circumstances, such as the type of feedstock used in the co-digestion and the relationships between the different feedstocks have large consequences on the methane yields and thereby the process economy.

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7. REFERENCES

[1] Johnson DK, Elander RT. Pretreatments for enhanced digestability of feedstocks. In: Himmel ME, editor. Biomass Recalcitrance: Blackwell Publishing; 2008. p. 436-53.

- [2] Taherzadeh MJ, Karimi K. Pretreatment of lignocellulosic wastes to improve ethanol and biogas production: a review. *International Journal of Molecular Sciences*. 2008;9:1621-51.
- [3] Hendriks ATWM, Zeeman G. Pretreatments to enhance the digestibility of lignocellulosic biomass. *Bioresource Technology*. 2009;100:10-8.
- [4] Shafiei M, Karimi K, Taherzadeh MJ. Pretreatment of spruce and oak by N-methylmorpholine-N-oxide (NMMO) for efficient conversion of their cellulose to ethanol. *Bioresource Technology*. 2010;101:4914-8.
- [5] Kuo C-H, Lee C-K. Enhanced enzymatic hydrolysis of sugarcane bagasse by N-methylmorpholine-N-oxide pretreatment. *Bioresource Technology*. 2009;100:866-71.
- [6] Shafiei M, Karimi K, Taherzadeh MJ. Techno-economical study of ethanol and biogas from spruce wood by NMMO-pretreatment and rapid fermentation and digestion. *Bioresource Technology*. 2011;102:7879-86.
- [7] Teghammar A, Karimi K, Sárvári Horváth I, Taherzadeh MJ. Enhanced biogas production from rice straw, triticale straw and softwood spruce by NMMO pretreatment. *Biomass and Bioenergy*. 2012;36:116-20.
- [8] Jeihanipour A, Karimi K, Taherzadeh MJ. Enhancement of ethanol and biogas production from high-crystalline cellulose by different modes of NMO pretreatment. *Biotechnology and Bioengineering*. 2009;105:469-76.
- [9] Cuissinat C, Navard P. Swelling and dissolution of cellulose part 1: free floating cotton and wood fibers in N-methylmorpholine-N-oxide-water mixtures. *Macromolecular Symposia* 2006. p. 1-18.
- [10] Adorjan I, Sjöberg J, Rosenau T, Hofinger A, Kosma P. Kinetic and chemical studies on the isomerization of monosaccharides in N-methylmorpholine-N-oxide (NMMO) under Lyocell conditions. *Carbohydrate Research*. 2004;339:1899-906.
- [11] Fink HP, Weigel P, Purz HJ, Ganster J. Structure formation of regenerated cellulose materials from NMMO-solutions. *Progress in Polymer Science*. 2001;26:1473-524.
- [12] Hall ME, Horrocks AR, Seddon H. The flammability of Lyocell. *Polymer Degradation and Stability*. 1999;64:505-10.
- [13] Karimi K, Zamani A. *Mucor indicus*: Biology and industrial applications perspectives: a review. *Biotechnology Advances*. 2013;31:466-481.
- [14] Murphy J, Power N. Technical and economic analysis of biogas production in Ireland utilising three different crop rotations. *Applied Energy*. 2009;86:25-36.
- [15] Thuresson T. Bioenergi från skog - uppdaterad bedömning av potentialer och förutsättningar för svenskt skogsbruk att producera främst primära skogsbränslen. *Skogsindustrierna*; 2010.
- [16] Kabir M, Sárvári Horváth I. NMMO pretreatment of straw and forest residues for enhanced methane production. Manuscript.
- [17] Kabir MM, del Pilar Castillo M, Taherzadeh MJ, Horváth IS. Effect of the N-Methylmorpholine-N-Oxide (NMMO) Pretreatment on Anaerobic Digestion of Forest Residues. *BioResources*. 2013;8:5409-23.
- [18] Båth B, Winter C. Bibliografiska uppgifter för växtnäringsstyrning i ekologisk odling i växthus. *VäxtEko, Jordbruksverket*; 2008.
- [19] Yadavika, Santosh, Sreekrishnan TR, Kohli S, Rana V. Enhancement of biogas production from solid substrates using different techniques—a review. *Bioresource Technology*. 2004;95:1-10.
- [20] Carlsson M, Uldal M. Substrathandbok för biogasproduktion. SGC, Swedish Gas Technology Centre; 2009.
- [21] de Hullu J, Maassen J, van Meel P, Shazad S, Vaessen J, Bini L, et al. Comparing different biogas upgrading techniques. Eindhoven, the Netherlands: Dirkse Milieutechniek, Eindhoven University of Technology; 2008.
- [22] Bauer F, Hulteberg C, Persson T, Tamm D. Biogas upgrading - Review of commercial technologies. SGC, Swedish Gas Technology Center; 2013.
- [23] Henriksson G, Del Pilar Castillo M, Jakubowicz I, Enocksson H, Ascue J, Lundgren P, et al. Environmental effects of the use of polymers in the biogas industry - Pre study. *Wasterefinery*; 2010.

- [24] Larsen J, Ostergaard Petersen M, Thirup L, Wen Li H, Krogh Iversen F. The IBUS process - lignocellulosic bioethanol close to a commercial reality. *Chemical Engineering & Technology*. 2008;31:765-72.
- [25] Turton R, Bailie RC, Whiting WB, Shaeiwitz JA. Analysis, synthesis and design of chemical processes. Upper Saddle River, New Jersey: Pearson Education; 2009.
- [26] Kumar D, Murthy GS. Impact of pretreatment and downstream processing technologies on economics and energy in cellulosic ethanol production. *Biotechnology for Biofuels*. 2011;4:27.
- [27] Aden A, Ruth M, Ibsen K, Jechura J, Neeves K, Sheehan J, et al. Lignocellulosic biomass to ethanol process design and economics utilizing co-current dilute acid prehydrolysis and enzymatic hydrolysis for corn stover. NREL National Renewable Energy Laboratory; 2002.
- [28] Sassner P, Galbe M, Zacchi G. Techno-economic evaluation of bioethanol production from three different lignocellulosic materials. *Biomass and Bioenergy*. 2008;32:422-30.
- [29] Forgács G, Niklasson C, Sárvári Horváth I, Taherzadeh MJ. Methane production from feather waste pretreated with Ca(OH)₂: process development and economical analysis. *Waste and Biomass Valorization*. March 2013.
- [30] Forgács G, Pourbafrani M, Niklasson C, Taherzadeh MJ, Hováth IS. Methane production from citrus wastes: process development and cost estimation. *Journal of Chemical Technology & Biotechnology*. 2012;87:250-5.
- [31] Benjaminsson J, Linné M. Biogasanläggningar med 300 GWh årsproduktion - system, teknik och ekonomi. SGC, Swedish Gas Center; 2007.
- [32] Gauthier Reberg K. Klart med sänkt bolagsskatt. Riksdag & Departement; 2012.
- [33] Dolan T, Cook MB, Angus AJ. Financial appraisal of wet mesophilic AD technology as a renewable energy and waste management technology. *Science of the Total Environment*. 2011;409:2460-6.
- [34] Inflation i Sverige 1831-2012, konsumentprisindex. <http://www.scb.se/Pages/TableAndChart33831.aspx> Statistiska centralbyrån; 2013.
- [35] Naturvårdsverket. Förbränningsanläggningar för energiproduktion inklusive rökgaskondensering. Naturvårdsverket; 2005.
- [36] Hagen M, Polman E, Jensen JK, Myken A, Jönsson O, Dahl A. Adding gas from biomass into the gas grid. GASTECH NV, Danish Gas Technology Center, Swedish Gas Center; 2001.
- [37] Skinner I, Essen Hv, Smokers R, Hill N. EU Transport GHG: Routes to 2050? Towards the decarbonisation of the EU's 2009.
- [38] Renewable Energy - Targets by 2020, European Commission. http://ec.europa.eu/energy/renewables/targets_en.htm 2011.
- [39] Teghammar A, Del Pilar Castillo M, Ascue J, Niklasson C, Sárvári Horváth I. Improved anaerobic digestion by the addition of paper tube residuals: pretreatment, stabilizing and synergetic effects. *Energy and Fuels*. 2013;27:277-84.
- [40] Wiberg S. Vatten- och avloppstaxa. Tibro kommun; 2012.
- [41] Dahlberg A, Ekander I. Wood fuel and peat prices No 4 2012. Swedish Energy Agency and SCB, Energy and transport statistics 2012.
- [42] Kastberg S. Fuel data. Swedish University of Agricultural Sciences, Unit of Biomass Technology and Chemistry: Länsstyrelsen i Västerbottens län; 1997.
- [43] Schnürer A, Jarvis Å. Mikrobiologisk handbok för biogasanläggningar. Rapport U2009:03. Avfall Sverige, Svenskt Gastekniskt Center; 2009.
- [44] Lindén B. Samkompostering av rötslam, halm och djupströgsödsel för framställning av ett lätthanterligt gödsel- och jordförbättringsmedel. SLU, Swedish University of Agricultural Sciences; 2000.

508 **TABLE LEGENDS**

509 Table 1. Prices for raw materials, products, and utilities.

510 Table 2. Stream components based on data obtained by batch NMMO pretreatment
511 experiments and expressed as ton/batch.

512 Table 3. Co-digestion scenarios with forest residues, OFMSW, and sewage sludge.

513 Table 4. Sensitivity analysis for the digestion of forest residues only.

514

515 **FIGURE LEGENDS**

516 Figure 1. Block flow diagram of the NMMO pretreatment and biogas production from forest
517 residues within a co-digestion with OFMSW.

518 Figure 2. Process flow diagram of the entire process.

519 Figure 3. FCI, Fixed capital investment per section, including equipment prices, installation,
520 instrumentation, electricity, piping, insulation, engineering and construction, contractor's fee,
521 and contingency. Auxiliary facilities, yard improvements, and buildings are excluded.

522 Figure 4. Annual operating costs for the base case divided into cost items.

523 Figure 5. Annual operating cost per section. Cost of materials is excluded.

524 Figure 6. Sensitivity analysis of total investment and annual operating costs, as well as
525 methane production costs, as a function of plant capacity of digested forest residues per year.

526 Figure 7. Result of cash flow analysis. Internal rate of return before taxes (IRR) of 50%
527 increased or decreased water consumption during washing, and of 20% increased or decreased
528 price of methane and forest residues, compared to base case, after taxes.

529

530

531 Table 1.

Raw materials	€/kg	Reference
Forest residues	0.057	Market price ¹
NMMO	4.0	[6]
OFMSW	-	-
Fresh water	6.7*10 ⁻⁵	[6]
Polyacrylamide	2,171	Market price ²
Sewage sludge	-3.28	³
Products		
Methane	1.895	Market price ⁴
Carbon dioxide	0.003	[6]
Lignin rich digestate	0.030	Market price ⁵
Utilities		
Electricity	0.0346 €/kWh	[28]
Steam	0.0084	SuperPro Designer [®]
Chilled water	2.28*10 ⁻⁴	SuperPro Designer [®]
Others		
Waste water treatment	9.79*10 ⁻⁴	[40]
Labor wage	70.000 €/employee/year	[28]

532 ¹Based on prices from the fourth quarter of 2011 [41], and the energy content of 1 kg
533 branches and tops [42], ²www.alibaba.com, ³personal communication with Moshe Habagil,
534 VIVAB, Vatten och miljö i Väst, 2013, ⁴methane price sold on the market (www.fordonsgas.se)
535 minus the cost for injection and distribution into the gas grid, together with the cost for tank
536 stations [31] and ⁵www.bioenergiportalen.se [41].

537

538 Table 2.

Stream component	1	2	3	4	5	6	7	8	9	10	11
Cellulose	14.3			14.3				4.1		4.1	
Hemicellulose	7.6			7.6				2.1		2.1	
Lignin	21.0			21.0				21.0		21.0	
Ash	7.6		15.2	7.6				22.7		22.7	
Water	16.8	3.3	205.1	133.8	2166.4	1815.1	117.0	1046.9	820.8	81.1	
NMMO		3.3		666.0	662.7		666.0	3.3		3.3	
Carbohydrates			60.7					8.1		8.1	
Proteins			8.0					1.1		1.1	
Fats			17.2					2.3		2.3	
Polyacrylamide										0.9	
Methane											32.6
Carbon dioxide											67.6
Total (ton/batch)	67.3	6.6	306.1	850.4	2829.1	1815.1	783.0	1111.6	820.8	146.8	100.2

539

540

541 Table 3.

Co-digestion substrates	C/N ratio	Forest residues	Unit prod. cost (€/kg CH ₄)	Unit revenue (€/kg CH ₄)	Total raw material (tons DW /year)	IRR % ⁴
Forest residues + OFMSW	29.5 ^{1,2}	33%	1.58	2.12	300,000	20.70
Forest residues + Sewage sludge	20.5 ^{1,3}	67%	2.78	2.75	300,000	3.52
Forest residues	325 ¹	100%	9.35	3.12	300,000	-100

542 ¹C/N ratio for forest residues is set as the middle value of a range between 150-500 according
543 to [18], ²C/N ratio for OFMSW is set as the middle value of a range between 15-32 according
544 to [43] and ³C/N ratio for sewage sludge is set as 5.98 according to [44], ⁴IRR is the internal
545 rate of return.

546

547 Table 4.

Methane price	NMMO recirculation	Unit production cost (€/kg CH ₄)	Unit revenue (€/kg CH ₄)	IRR ¹ %
+0%	99.5%	9.35	3.12	-100
+0%	99.99%	3.21	3.12	-100
+25%	99.99%	3.21	3.21	4.30
+50%	99.99%	3.21	3.69	11.0

548 ¹IRR is the internal rate of return.

549

550

Figure 2.

Figure 3.

Figure 4.

Figure 5.

Figure 6.

Figure 7.