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Biomethane production from starch and lignocellulosic crops: a comparative review

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Abstract: The methane produced from the anaerobic digestion of organic wastes and energy crops represents an elegant and economical means of generating renewable biofuel. Anaerobic digestion is a mature technology and is already used for the conversion of the organic fraction of municipal solid wastes and excess primary and secondary sludge from waste-water treatment plants. High methane yield up to 0.45 m³ STP CH4/kg volatile solids (VS) or 12 390 m³ STP CH₄/ ha can be achieved with sugar and starch crops, although these cultures are competing with food and feed crops for high-quality land. The cultivation of lignocellulosic crops on marginal and set-aside lands is a more environmentally sound and sustainable option for renewable energy production. The methane yield obtained from these crops is lower, 0.17–0.39 m³ STP CH4/kg VS or 5400 m³ STP CH₄/ha, as its conversion into methane is facing the same initial barrier as for the production of ethanol, for example, hydrolysis of the crops. Intensive research and development on efficient pre-treatments is ongoing to optimize the net energy production, which is potentially greater than for liquid biofuels, since the whole substrate excepted lignin is convertible into methane. © 2010 Crown in the right of Canada

Keywords: anaerobic; methane; lignocellulosic; crops; energy

Introduction

A naerobic digestion (AD) of biomass has an inherent advantage, as compared to typical pathways to biodiesel or bioethanol; it can be performed using many different input streams, carrying the majority of the 'electron freight' into methane independently of the chemistry of the substrate.¹ Besides, AD maximizes the resource recovery, as it releases a solid digestate that can be used as a peat-type organic amendment for soil as well as an effluent, that can be concentrated into a nutrient-rich liquid that is easy to spray as a fertilizer on agricultural fields. Another asset is that anaerobic digestion, as a microbial community-based process, requires neither substrate sterilization, nor special measures for culture inoculation. And, in contrast to liquid biofuel chain, a step for product separation is unnecessary, as the biogas distillates off by itself from the liquid. Biogas typically contains (v/v) 50–75% methane, 25–50% carbon dioxide, 1–5% water vapor, 0–5% nitrogen, smaller amounts of hydrogen sulfide (0–5,000 ppm) and ammonia (0–500 ppm), and trace concentrations of

Correspondence to: Serge R. Guiot, Biotechnology Research Institute, National Research Council of Canada, 6100 Royalmount, Montreal, Canada, H4P 2R2. E-mail: serge.guiot@cnrc-nrc.gc.ca hydrogen and carbon monoxide.² This means the energy yield can be high, in theory.

A theoretical or maximal methane yield (Y_{CH4} , m³ STP/ kg substrate converted) can be calculated from the elemental composition of a substrate, $C_cH_hO_xN_nS_s$, as shown in Eqn (1): with 22.4 as the molar volume of any ideal gas (L STP/mol).

$$Y_{CH4} = \frac{22.4 \left(\frac{c}{2} + \frac{h}{8} - \frac{x}{4} - \frac{3n}{8} - \frac{s}{4}\right)}{12c + h + 16x + 14n + 16s}$$
(1)

Application of Eqn (1) gives 0.37 m^3 STP/ kg carbohydrates (CH₂O), 0.51 m^3 STP/ kg proteins (C₁₀₆H₁₆₈O₃₄N₂₈S), 1 m³ STP/ kg fat (C₈H₁₅O), and 0.48 m³ STP/ kg biomass (based on the formula of Roels, ${}^3C_5H_9O_{2.5}NS_{0.025}$). In practice, however, the methane yield of biomass does not often exceed 60% of the theoretical value, 4 because it contains compounds that are poorly or not biodegradable (e.g., lignin, peptidoglycan, membrane-associated proteins...), or compounds the solubilization of which might be limited by a hydrolytic deficiency of the actual populations (cellulose, hemicellulose, proteins). As it will be shown later, this highlights the importance of hydrolytic pre-treatment or *in situ* hydrolysis to be applied to the crop in order to obtain the best net energy gain possible, regardless to the AD technology type.

Relevance of biomethane production from energy crops

'Escalating energy costs and energy shortages in recent years have become problems of national significance and have prompted the search for new sources of energy.' Although this quotation could fit very well in any newspaper from the past year, it is actually cited from the work of Clausen *et al.*⁵ These authors believed that the bioconversion of plant matter to methane gas was economically attractive at the fossil fuel prices of that time. However, multiple factors resulted in a sharp decrease in fossil fuel prices and increase in availability during the same years as some pionneering work on the anaerobic digestion of crops was being done.⁵⁻⁷ Nevertheless, the generation of biofuels from energy crops is being prompted in recent years in part for the same reasons as 30 years ago: an expected energy shortage, or at least significant fossil fuel prices increase. Today's incentive to produce biofuels is also derived from the need to reduce fossil fuel consumption, and the related increase in atmospheric CO_2 , in order to decrease its impact on global climate change.

The increase of interest in producing renewable energy from crops is acknowledged by the significant increase in publications on the subject in recent years. The production of bioethanol from starch crops (corn, wheat) and the production of biodiesel from crops such as soy, are established and operational, although under a heavily subsidized form at least in the United States. Moreover, the net energy balance for corn-based ethanol, 1.5 X 10¹⁰ L ethanol produced in the USA for 4.0 X 10¹⁰ kg of corn in 2005, represents only 25% net energy return, equivalent to only 0.3% of the fuel utilization.⁸ The increase in corn to ethanol production in the United States has also led to an increase in the cost of corn by over 50% in the past year.9 Because of this, approaches relying on the use of food crops for biofuel production are failing to meet renewable criteria since they compete with food production for high-grade arable land and their bioenergy output is limited.¹⁰ In effect, the food versus fuel debate is making it hard to justify the diversion of lands traditionally harvested for human feed and convert them into lands harvested for transportation. These first-generation biorefineries, operating from first-generation crops, were probably a necessary step in the evolution toward more sustainable practices for renewable energy production. A second generation of biorefineries is underway, with the production of biofuels from lignocellulosic material, crops, agricultural wastes, or forestry feedstocks.

Although the concept of biorefinery is mostly applied to the production of ethanol and biodiesel as of now, an anaerobic-digestion-based biorefinery would deserve to be more carefully evaluated for the conversion of crops, as it would generate potentially more renewable energy, as methane. For instance ca. 60% of the energy from sugarcane that has been used for the production of bioethanol for decades in Brazil, can be converted to biogas while only 38% of the cane energy is converted into alcohol.¹¹ With wheat or maize, up to three times more net energy yield can be obtained per hectare by making methane instead of biodiesel or bioethanol.^{12, 13} Anaerobic digesters can be built more locally, and a variety of feedstock can be used for biomethanation (more versatile). Also, there is a flexibility on the type of energy produced, where feedstock can be transformed into heat, combined heat and power (electricity), or purified and used as compressed natural gas for use as vehicle fuel, for example. In effect, anaerobic digestion is one of the most energy-efficient, as well as environmentally benign ways to produce vehicle biofuel.¹⁴ Biogas production from energy crops represents a more thermodynamically efficient option than converting plant matter into liquid fuels.¹⁵ Moreover, biomethane obtained from anaerobic digestion is the most efficient, clean burning biofuel available today.¹⁶ Although biogas may contain siloxanes (0–50 mg/m³) and dust particles, the combustion of methane reduces emissions of NOx, CO, particulate matter and unburned hydrocarbons, by 80, 50, 98, and 80%, respectively, as compared to petroleum-derived diesel.²

The use of anaerobic digestion has evolved in the past 20 years from a pollution control technology (agrofood and pulp and paper waste-waters treatment, sludge stabilization) to a renewable energy-producing technology. The anaerobic conversion of crops into methane is now viable in some countries, with the help of grants and guaranteed premium prices for the purchase of the generated electricity.⁸ The most spectacular example of this trend is the construction and operation of over 5000 anaerobic digesters in Germany during the past 20 years, following a series of financial incentives put in place by the government. The use of energy crops, mainly corn silage, is widely spread in the operation of the farm-scale anaerobic digesters treating manure, in order to increase the methane yield and the resulting net renewable energy production.^{17, 18}

Optimization of methane production from energy crops

Comparative methane yields from different energy crops

Different categories of energy crops have been studied for the past 30 years with regard to their methane potential after anaerobic digestion. Gunaseelan¹⁹ has made an extensive review of methane production from fruit and vegetables, grass, woody biomass, terrestrial weed, marine biomass, and freshwater biomass. An overview of different crops potential can also be found in Murphy and Power.²⁰ It is outside the scope of this paper to cover all crops, and the focus instead will be on two large categories of energy crops that are drawing most of the attention: starch crops for their high methane potential and lignocellulosic crops as the second generation of biofuel crops. The first group includes the sugar and starch crops, which are relatively efficient converters of solar energy that will produce either fermentable sugars (sugarcane, sugarbeet), or starch (corn, potatoes). The sugar and starch crops are the main energy crops currently used on a commercial scale for the production of biomethane. Although these crops generate high yields of methane, they also have other uses as food and/or feed, which may often compete with biofuel production. Cellulosic or lignocellulosic crops are represented by different grasses containing small percentage of lignin (hay, clover, reed canary grass), while other energy crops such as Miscanthus or switchgrass are containing higher levels of lignin (12–20%).

Methane yield from starch crops

Numerous studies have evaluated the methane potential obtained from starch crops such as sugarbeets, corn grain, and potatoes. Some examples will be presented here, to demonstrate the high biofuel yield achievable from anaerobic digestion. It has to be noted that the preparation of these crops prior to anaerobic digestion remains relatively simple and generally does not go further than a size reduction. Although it is not stated as such by the experimenters, the authors consider the ensiling of crops to constitute a pretreatment in itself.

A vast selection of crops have been evaluated for their potential in methane, including fafa bean,²¹ horse beans,²² Jerusalem artichoke,²³ oats,⁵ rye,^{21, 24} sorghum,^{25, 26} sugarcane,²⁶ sunflower,²⁴ triticale,^{2, 24} and wheat.²⁴ The sugar or starch crops that have been the most intensively researched are sugarbeets,^{23, 27} corn (grain)^{24, 28} and potatoes.^{29, 30}

It can be seen in Table 1 that the obtained methane yield was high, at around 400–445, 250–406, and 310–430 L STP CH₄ /kg VS added for sugarbeets, corn, and potatoes, respectively. There was quite a large variation in the obtained methane yield for corn, while results were more consistent for potatoes, although lower by around 15–20%. Sugarbeets showed consistent high methane yield in the available literature. The volatile solids degradation levels were in accordance with the methane produced from the

Table 1. Methane	potential from starch and sugar crops.		
Crops	Operating Conditions	Yield (m ³ STP CH ₄ / kg VS added) ¹	References
Corn (grain)	BMP	0.25–0.40	6, 24, 25, 28
Corn (grain)	CSTR, OLR 2.5 – 4.0 kg TS/L.d, HRT 10 - 20d	0.18–0.41	22, 30
Fafa beans	BMP	0.44	21
Jerusalem artichoke	BMP	0.37±0.06	39
Oats	BMP	0.254	6
Potatoes	BMP	0.31–0.33	29, 45, 78
Potatoes	CSTR, OLR 2.5 kg TS /L.d, HRT 20d	0.43	30
Rye (ensiled)	BMP	0.14–0.36	21, 24
Sugarbeets	BMP	0.25–0.45	6, 23
Ensiled sugarbeets	CSTR, OLR 14.3 kg VS /m3.d, HRT 6.5d	0.40 ²	27
Sugarcane	BMP	0.23–0.30	26
Wheat	BMP	0.14–0.34	24

BMP: biochemical methane potential (batch assays).

CSTR: continuously stirred tank reactors (fed-batch or continuous feeding).

¹ Values in references 6, 22, 30 are expressed in m^3 STP CH₄ / kg TS. These values should be similar when reported in kg VS since most of the solid fraction of these crops are organic (corn grain, potatoes, VS/TS ratio of 0.985 and 0.950).

² Based on 0.65-0.67 L biogas /gVS.d, assuming a methane content of 60%.

crops at 67–92, 40–65, and up to 96% for corn, potatoes and sugarbeets, respectively. Indeed, this is a confirmation that the conversion potential of such crops into methane can be very close to the maximal theoretical yield.

The vast majority of the literature is reporting experiments performed at mesophilic temperature, and very few experiments conducted at thermophilic temperature,²⁵ possibly since the methane production process is efficient enough at 35°C and little gain is made when increasing the operation temperature compared with the associated increased costs. It is worth mentioning that co-digestion of different crops can lead to improved methanization, as shown by Parawira et al.²⁹ when co-digesting potato wastes and sugarbeets, reaching methane yield of 0.42–0.52 L CH_4 /gVS compared with average yields of 0.32 for potatoes and 0.40 L CH_4 /gVS for sugarbeets only as reported in the literature (Table 1). The use of two-stage digestion can also improve the process, by uncoupling the hydrolysis of the crop and its fermentation, which can lead to a drastic fall in pH and possible inhibition.³¹ This is described in Lehtomäki and Bjornsson³² where a combination of a 7.6 m³ leach bed and a 2.6 m³ UASB generated 0.38 $-0.39 \text{ m}^3 \text{ CH}_4$ /kg VS added at a retention time of 55 days when digesting sugarbeets and grass silage, respectively. In this case, the VS reduction was nearly complete at 96%.

Although many large-scale anaerobic digesters include energy crops in their feeding, there is actually little literature on the use of dedicated sugar or starch crops for methane production as a biofuel. One example is a full-scale system generating 500 m³ per day of biogas and able to generate 100 kWh of electricity per day, at an OLR of 1.67 kg VS /m³.d and an HRT of 52.5 days from ensiled sugar beets reported by Scherer and Lehman.²⁷

Sugar and starch crops are showing the best methane yield per hectare, at 5 300–12 390, 6604 and 5400 m³ CH₄/ ha for corn, triticale and sugarbeets, respectively (Table 2). The higher yield obtained from sugar and starch crops, however, should be weighted against their use of quality land, their effect on the price of food and feed crops, and the more intensive care attached to these types of cultures (nutrients, pesticide, tillage...). Therefore, additional work should be directed toward the evaluation of biofuel dedicated crop that could be grown and harvested on marginal lands; for example, without displacing food and feed crops.

Methane yield from cellulosic and lignocellulosic crops

The cultivation and use of cellulosic and lignocellulosic crops is arguably a more environmentally sound and sustainable

production and crop field yield.					
Crops	Yield (m ³ STP CH ₄ / ha)	References			
Cocksfoot	2392	38			
Corn	5300-12 390	24, 28			
Festlolium	2806	38			
Giant knotweed	3800	39			
Hemp	2840	79			
Jerusalem artichoke	3100–5400	39			
Reed canary grass	3800–4200	39			
Rhubarb	800–1700	39			
Sugarbeets	5400	32			
Sunflower	4695	24			
Tall fescue	2749	38			
Timothy	1842–2335	38			
Timothy clover grass	2900–4000	39			
Triticale	1112–6604	2			

Table 2 Biomethane vield in function of methan

option for renewable energy production than using sugar and starch crops. These crops can generally be cultivated on marginal lands; for example, this will not displace the production of food or feed crops. Lignocellulosic material is composed of three types of polymers, namely cellulose, hemicellulose, and lignin.³³ In addition to these compounds, lignocellulosic crops contain non-structural carbohydrates such as glucose, fructose, sucrose and fructans, proteins, lipids, extractives, and pectins.³⁴ To our knowledge, there is no clear delimitation between cellulosic and lignocellulosic cultures and both terms are employed in the literature. One could presume that cellulosic feedstock would refer to plant material containing very little or no lignin, such as grass or alfalfa at 5–7% lignin content,³⁵ while lignocellulosic material would refer to plants containing a fairly high (over 15%) concentration of lignin.³⁶ For now, most of the reported literature is focused on cellulosic crop.

The use of energy crops, mainly corn silage, is already widely spread especially in Germany where crops are added to more than 90% of the on-farm digesters.³⁷ Nevertheless, a vast selection of cellulosic and lignocellulosic crops have been under study in the past recent years^{23, 38} and showed a wider diversity of potential candidates than the sugar and starch crops (Table 3). Lehtömaki²³ has performed a systematic study on a number of crops, at different harvest times,

fresh and ensiled. Most crop potential reached 0.3–0.4 m³ of methane per kg of VS added (Table 2). Feedstock with a higher percentage of lignin, such as straw, showed in general lower methane potential (0.18–0.32 m³ CH₄ / kg VS). Stewart *et al.*³⁰ obtained an exceptionally high yield (0.46–0.49 m³ CH₄ / kg TS) when digesting lucerne, ryegrass and clover in CSTR at a loading rate of 2.5 kg TS / L.d.

The methane yield on a VS basis is important to identify the promising crops, but solid content, and crop yield on the field are what will matter at the end when selecting a crop for biomethane production. In effect, some crops have high methane yield on a VS-added basis, such as rhubarb at 0.49 m³ CH₄ / kg VS (Table 2). However, its low solid content results in a low methane yield on a wet basis ($40 \pm 2 \text{ m}^3$ CH₄ / wet ton) that impacts on the global yield per hectare cultivated ($800-1700 \text{ m}^3 \text{ CH}_4$ / ha)³⁹ (Table 3). Also, Seppälä *et al.*³⁸ reported similar methane potential when comparing grasses such as timothy, festlolium, tall fescue or cocksfoot ($0.328-0.333 \text{ m}^3 \text{ CH}_4$ / kg VS). It was their yield per hectare that ended up making a difference, from 1842 m³ CH₄/ ha.yr for timothy to 2806 m³ CH₄ / ha.yr for festlolium.

Among the various lignocellulosic crops evaluated for biofuel production, Panicum vergatum or switchgrass would certainly represents a solid choice. Switchgrass is a C4 perennial tall grass that is high yielding (12–18 tonnes dry solids per hectare), is highly adaptable to poor soils and requires low fertilizer applications.⁴⁰⁻⁴² It was chosen as the model lignocellulosic crop by the US Department of Energy in the 1990s and is believed to return 540% more renewable energy than fossil fuel consumption,43 compared to 25% for maize.⁸ Currently switchgrass is mostly used for bedding and combustion in Canada¹⁵ and for the ethanol production in the USA.⁴³ Switchgrass is composed of around 12–19% lignin, 31-37% hemicellulose and 29-45% cellulose, 36, 44 hence suggesting a high potential conversion of the plant into biofuel, despite the fact that lignin is poorly degraded under anaerobic conditions. Preliminary data are showing a moderate potential for raw switchgrass, at 0.125 m³ CH₄/ kg VS.45 Its high solid content (400 kg VS/ wet ton) places it at an estimated 650 m³ CH₄/ ha, which is much lower than for the other crops, however there is a lot of room for improvement considering that this was obtained from less than 35% VS conversion.

Table 3. Methane pote	ential from lignocellulosic c	rops.	
Crops	Operating Conditions	Yield (m ³ STP CH ₄ / kg VS added) ¹	References
Alfalfa	BMP	0.24	22
Alfalfa, ensiled	CSTR	0.24–0.26	30
Clover	BMP	0.29–0.39	24
Cocksfoot	BMP	0.333–0.344	38
Corn silage	BMP	0.270–0.298	22
Festlolium	BMP	0.328–0.359	38
Giant knotweed	BMP	0.170	39
Grass, mixed	BMP	0.298–0.315	22
Sugarbeet leaves, alfalfa	CSTR	0.174–0.226	22
Grass, lawn	BMP	0.300±0.040	23
Grass, fresh	BMP	0.231±0.030	23
Grass, ensiled	BMP	0.128–0.392,	23, 28, 32
	Leach bed + UASB	0.39	32
Hemp	BMP	0.230–0.409	2, 79
Lucerne	BMP	0.247	6
	CSTR 2.5 kg TS/L.d	0.46±0.06	30
Lupine	BMP	0.360±0.040	39
Marrow kale	BMP	0.310±0.020	39
Napier grass	BMP	0.19–0.34	26
Nettle	BMP	0.210	39
Oats, straw	BMP	0.320±0.020	39
Rapeseed	BMP	0.240±0.020	39
Reed canary grass	BMP	0.340	39
Red clover	BMP	0.300±0.060	23
Rhubarb	BMP	0.490±0.030	39
Ryegrass and clover	CSTR 2.5 kg TS/L.d	0.498±0.056	30
Straw, barley	CSTR 2.5 kg TS/L.d	0.285±0.054	30
Straw, ryegrass	CSTR 2.5 kg TS/L.d	0.177±0.062	30
Sugarbeet leaves	BMP	0.294	22
Tall fescue	BMP	0.332–0.340	38
Timothy	BMP	0.333–0.385	38, 39
Vetch	BMP	0.323	22
Vetch - oat	BMP	0.410±0.020	23

BMP: biochemical methane potential (batch assays).

CSTR: continuously stirred tank reactors (fed-batch or continuous feeding).

¹Values in references 6, 22, 30 are expressed in m³ STP CH₄ / kg TS. These values should be similar when reported in kg VS since most of the solid fraction of these crops are organic (corn grain, VS/TS ratio of 0.985).

Large-scale dedicated anaerobic digesters using cellulosic crops as a substrate are not commonly seen nowadays, although high methane yields can be achieved, such as in a digester in Eugendorf, Austria, where 150 ha of grass silage is transformed into biomethane and used as transport fuel at a yield of 0.3 $\rm Nm^3$ / kg $\rm VS_{added}$ for a loading of 1.4 kg VS / $\rm m^3$ reactor.d. 46

In most of the aforementioned literature, there is no specific pre-treatment of the crop prior to its anaerobic digestion, excepted particle size reduction (chopping, shredding,

mulching...) and ensiling. Mechanical pre-treatment, such as milling, reduces the particle size of the lignocellulosic biomass and can increase the hydrolysis yield and reduce the digestion time.47,48 Although milling is not considered economically feasible due to the high energy requirements, 49,50 the reduction of the particle size still represents a mandatory first step for the preparation of lignocellulosic substrate reaching 2-4 meters high in their original state (corn stover, switchgrass, Miscanthus, for example). There is also the option of cutting instead of milling or grinding in order to minimize the energy input, as was performed by Jorgensen et al.⁵¹ Ensiling has been shown to have a positive impact most of the time, and may result in up to 31% more methane production from the crops.²³ The particle size reduction had a different effect on sugar and starch or cellulosic crops and lignocellulosic crops. In effect, the benefit of size reduction was mitigated for the sugar and starch as well as cellulosic crops (oat, sorghum, grass) and generally highly positive for the lignocellulosic crops (straw) with 0-19% and 21-65 % more methane produced, respectively.²³ However, it can be presumed that an ensiled lignocellulosic crop would still retain a structure that would require a more intense pretreatment in order to obtain methane production closer to the maximal theoretical yield. In this sense, some additional pre-treatment needs to be applied to the crop in order to obtain the best net energy gain possible.

The impact of pre-treatments on increased biomethane yield

Pre-treatments are seldom mentioned in the literature when digesting sugar and starch crops, possibly because these types of crops are already well digestible after a simple size reduction. The choice of pre-treatment should then be made carefully, as the main purpose should be hydrolysis, or liquefaction of the substrate, in order to maximize the methane potential from the target crop. In this regard, pre-treatment such as high temperature, microwave, and autoclave all result in a cooked version of corn grain or potatoes.⁴⁵ A particle size reduction was applied as a first step pre-treatment to potatoes. Then, an alkali pre-treatment resulted in a significant improvement of the amount of methane from potatoes, from 329 to 435 L STP $CH_4/$ kg VS after 9 days of incubation. Besides the 32% methane yield increase, an additional benefit was the reduction of the incubation time, from 4 weeks to 9 days, which would significantly impact the sizing of the digester processing the crop. Corn grain was ground and subjected to sonication after being slurried, which resulted in an increase by 22% of the methane generated, at 454 compared to 373 L STP CH_4/kg VS. A reduction in the incubation time required was also observed, with 70% of the total methane yield obtained in 48 h. Thus, pretreatments of sugar and starch crops can have a significant impact on the final methane yield, but also by reducing the time required to extract it from the crops.

The rate-limiting step in anaerobic digestion of solid feedstock such as lignocellulosic crops is the hydrolysis of complex polymeric substances⁵²⁻⁵⁵ and in particular, the cross-linking of lignin which is non-biodegradable with the cellulose and hemicellulose.³² Moreover, the crystalline structure of cellulose prevents penetration by micro-organisms or extracellular enzymes.⁵⁶ An ideal pre-treatment would then aim at the partial or complete decomposition of the feedstock into soluble fermentable products.

Abundant literature is available on lignocellulosic pretreatment technologies such as enzymatic liquefaction and saccharification, solvent-based, dilute acid, ammonia fiber explosion, ammonia recycle percolation, lime, steam explosion, and OrganoSolv pre-treatment, that are under intensive investigation on both laboratory scale and as pilot plants, mostly in order to generate ethanol in a biorefinery concept.44,51,57-60 Pre-treatments directed more specifically to enhanced methane production fom lignocellulosic biomass were reviewed recently by Hendrick and Zeeman,⁴⁹ who concluded that steam-, lime-, liquid-hot-water-, and ammonia-based pre-treatments showed high potentials. Other physical pre-treatments offering potential for improving methane yields from lignocellulosic materials are, for example, steam explosion, thermal hydrolysis, wet oxidation, pre-incubation in water, and treatment with ultrasound or radiation.^{44, 61-63} Energy-intensive pre-treatments including steam explosion, wet explosion and ammonia fiber explosion (AFEX) have the advantage of practically solubilizing the whole substrates and achieve very high yield of methane. However, their energy costs have also to be taken into consideration, and the net energy gain of using these pre-treatments has yet to be clearly demonstrated.

Although literature is abundant on the description of pretreaments for increasing methane yield, actual experimental assays are more scarce (Table 4). Thermochemical pre-treatments (alkalis, autoclaving) are promising^{64,65} but did not result in a significant increase in methane production for the energy crops that were tested. In effect, addition of 2% NaOH or 3% Ca(OH)₂ during 72 h only allowed for 9% and 17% more methane from sugarbeet tops and hay while showing no improvement on straw.⁶⁵ Biological treatments, either with micro-organisms, or with enzymes, are simple and do not require major capital investments, although the increase in biogas yield has, been low so far.²³

The increase in methane production should not be the only benefit evaluated when applying pre-treatment to lignocellulosic crops. In effect, as for the sugar and starch crops, pre-treatments can also allow for a reduction in the reaction time to obtain methane, from 30 to 18 days when digesting grass hay and wheatstraw.⁶

Energy-intensive pre-treatments can yield high amounts of methane from crops. For example, Petersson *et al.*²¹ performed wet oxidation (195°C, 15 min, 2g/L Na₂CO₃, 12 bars oxygen) on winter rye, oilseed rape and fafa bean, and obtained 96, 85 and 75% of the theoretical yield from the crop, at 342, 286 and 258 L STP CH₄/ kg TS added. An economical evaluation of wet oxidation was performed by Uellendahl *et al.*⁶⁶ who have calculated the energy gain that could be obtained from the pre-treatment of corn, *Miscanthus* and willow. The *Miscanthus* and willow methane yield increased from 200 to 360 L CH₄/kg VS after wet oxidation. This was converted into 39.6 and 35.4 MWh/ha, compared with 31.0 MWh/ha for corn. An economic balance concluded that the anaerobic digestion of untreated crops was not profitable, even for corn (- \in 5/ha), but that wet oxidation could make it profitable with \in 547 and \in 502/ha for *Miscanthus* and willow.

Concerns and future trends for the optimization of the net energy field

One important risk that our dependence on fossil fuels poses is from global climate change caused by the net increase in atmospheric CO_2 due to combustion of the fossil fuels. The establishment of a strong renewable energy economy, originating from biofuel production from lignocellulosic crops grown on dedicated non-food lands could certainly alleviate the risks described earlier.

Guidelines on optimum energy crop production, optimum harvesting time, optimum nutrient composition, optimum conservation, and pre-treatment technology must be developed.²⁴ Economic biogas production requires high biogas yield per hectare. Biogas production can become a key technology for the production of renewable energy source. The key final factor is methane yield per hectare. Amon *et al.* suggest that sustainable biogas production from energy crops must not be based on maximum yields from single crops, but on maximum methane yield from the whole system of sustainable and environmentally friendly crop rotation.²⁴ Other tools are created in parallel such as a model prepared by Gunnarsson *et al.*⁶⁷ to account for the costs related to transportation (transport system, distance to

Table 4. List of pre-treatments associated with improvement in methane yield for lignocellulosic crops.				
Crops	Pre-treatments	Energy, %CH4 Increase	Reference	
Sugarbeet tops Hay Straw	Alkalis 2% NaOH 24h or 2% NaOH 72h or 3% Ca(OH)2+4% Na2CO3 72h	+ 17%	65	
	xylanases, cellulases	0 %		
Winter harvested switchgrass Summer harvested switchgrass	Temperature (90°C, 3h) Alcalinization (NaOH 7 g/L, 3h) Microwave (1300 W, 20 min) Sonication (20 kHz, 120 min) Autoclave (121°C, 15 psi, 20 min) Combined alkalis – autoclave Enzymatic peroxidase	0 - + 32 %	45	
Miscanthus	Wet oxidation	+ 80 %	66	

storage, size of field), crop preparation (chopping, ensiling) in order to evaluate the profitability of anaerobic digestion of dedicated crops in Sweden.

Also, to engender a viable biobased energy system, bioenergy crops must compete successfully both as crops and as fuels. Owners of cropland will only produce these crops if they provide an economic return that is at least equivalent to returns from the most profitable conventional crops. This will probably require some form of subsidies or incentives, to allow for the energy crops to be competitive with other source of fuel. For example, the break-even cost for *Miscanthus* varied between US\$41 and US\$58/t compared with US\$20–22/t for coal, therefore needing incentives for the power plant to buy energy crops instead of coal for electricity production.⁶⁸

The best motivation for deciders and entrepreneurs to implement such large-scale biorefinery concepts for methane production from energy crops, is to see a strong potential for profits. For now, there are only a few large-scale systems with detailed costs analysis. Nordberg and Edström⁶⁹ have proceeded with such an analysis for a 1.6 MW plant, giving 14 GWh of biofuel annually from 6000 tons of crops (ensiled grass and clover) and 6000 tons of source-sorted municipal solid waste. The capital cost reached €8.2 million, split between the plant itself (€6.3 million) and the biogas purification and compression system (€1.8 million). The operational costs were €1.04, 0.14 and 0.40 million per year for the biogas plant, ley crop (farmer compensation of €0.034/kg TS) and upgrading of the biogas, respectively. In order to have revenues that match these costs, the selling price for the purified methane should be at least €0.078/kWh, for revenues of €0.37, 1.08 and 0.13 million for the gate fees, the vehicle fuel and the digestate, respectively. The gate fee was fixed at €47 per ton of waste, and the digestate was sold as fertilizer. Another example is the description of the first continuous full-scale dry anaerobic digester (1200 m³) for crops producing 500 kW, at a total investment of €3 million.¹³

Although the literature is reporting mainly on 'wet' anaerobic digestion of lignocellulosic crops, the use of high solids digesters or dry fermentation would be worth investigating as it would resolve some problems like the addition of process water and flotation of the crops on the top of the digester.^{13, 70} Furthermore to achieve a stable and

efficient process, in addition to the development and use of low-energy and cost-effective pre-treatments, research and development can be pursued to improve the production of methane from lignocellulosic crops, including:

- adjustment of the carbon to nitrogen ratio with nutrients addition or co-digestion;^{71, 72}
- addition of trace metals;⁷³⁻⁷⁶
- integration of cost-effective thermochemical or/and enzymatic pre-treatments;
- improvement of the hydrolytic functions of the *in situ* microbial populations, either by enrichment of naturally present hydrolytic micro-organisms, or by the addition and retention of *de novo* hydrolytic capabilities.

Future work should also focus on the use of a crop rotation system for sustainability and thus use an average methane yield from the whole crop rotation. Regarding this concern, a clear advantage of lignocellulosic crop cultivation is the use of marginal or set-aside lands and harvesting for more than 10-12 years without jeopardizing the soil quality, with minimal or no fertilizer supplementation. Amon et al.²⁴ have developed a Methane Energy Value Model (MEVM) to estimate the methane yield from substrate, from the content of crude protein, crude fat, crude fiber, and N-free extracts and their models for maize and cereal are in good agreement with experimental values. If rotation crops (specialized energy crop, feeding, food) was made with all arable land of the European Union as of 2007 (EU 25) (93 millions ha), the average yield would be 4000 m³ STP CH₄/ ha for 372 000 million m³ of methane per year, or 320 million tons COE (crude oil equivalents). This is equal to 96% of the energy demand of the road traffic in the European Union as of 2007 (EU 25). Similarly but at a country level, Singh et al.⁷⁷ provided data for the anaerobic digestion of grass silage cultivated on excess lands along with agricultural wastes (manure, slaughter waste), and the cumulative energy generated could theoretically power 91% of the private car fleet in Ireland by 2020.

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