

A Review paper on Ethanol

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ABSTRACT: Ethanol produced from various lignocellulosic materials such as wood, agricultural and forest residues has the potential to be a valuable substitute for, or complement to, gasoline. One of the major resources in the Northern hemisphere is softwood. This paper reviews the current status of the technology for ethanol production from softwood, with focus on hemicellulose and cellulose hydrolysis, which is the major problem in the overall process. Other issues of importance, e.g. overall process configurations and process economics are also considered. The review deals with fuel ethanol production from plant-based lignocellulosic biomass as raw materials. In this article, the technologies for producing fuel ethanol with the main research prospects for improving them are discussed. The complexity in the biomass processing is identified by the analysis of various stages involved in the conversion of lignocellulosic biomass into fermentable sugars. Further, the fermentation processes with its important features are explained based on biomass conversion. Comparative index for different types of biomass for fuel ethanol production is listed. Finally, some concluding remarks on current research regarding the pre-treatment along with biological conversion of biomass into ethanol are presented. Ethanol produced from various lignocellulosic materials such as wood, agricultural and forest residues has the potential to be a valuable substitute for, or complement to, gasoline. One of the major resources in the Northern hemisphere is softwood. This paper reviews the current status of the technology for ethanol production from softwood, with focus on hemicellulose and cellulose hydrolysis, which is the major problem in the overall process. Other issues of importance, e.g. overall process configurations and process economics are also considered.

KEYWORDS: Conversion process, Development, Efficiency of Ethanol, Uses of ethanol, Pre-treatment.

1. INTRODUCTION

At the beginning of the 1970s, OPEC decreased the output of oil, which resulted in a dramatic increase in oil prices and the world's interest in alternative fuels increased significantly. The aim then was to become less dependent on oil and to reduce the cost of expensive oil imports. Still today, an interest in production of gasoline substituting fuels is maintained in many countries all over the world. However, the emphasis today is on reducing pollution and helping to satisfy the Kyoto protocol, established in 1997, by limiting the global net emission of carbon dioxide (CO₂) [1]. In December 1997, The European Commission adopted a white paper for a community strategy and action plan, "Energy for the Future", with the given objective of a contribution of renewable sources of energy to the European Union's gross inland energy consumption of 12% by 2010. One of the objectives is to reach 5 million metric tons of liquid biofuels by 2003. During the past 150 years, human activities have caused a dramatic increase in the emission of a number of greenhouse gases, e.g. CO₂, which has led to changes in the equilibrium of the earth's atmosphere. The content of CO₂ in the air has increased from 280 ppm to 365 ppm during this period.

The OECD countries contribute more than half of the world's total emission of CO₂. The United States is one of the countries with the highest rates of discharge, emitting more than 20,000 kg CO₂ per capita per year (The Swedish Energy Administration 1999). The corresponding CO₂ emission for Sweden for the same period was about 6,000 kg per capita (The Swedish Energy Administration 1999). The transport sector is responsible for the greatest proportion of CO₂ emission, and it is increasing from year to year. One way of reducing environmental effects and the dependence on fossil fuels is to use renewable bio ethanol [2]. Apart from a very low net emission of CO₂ to the atmosphere, the combustion of bioethanol in general results in the emission of low levels of non-combusted hydrocarbons, carbon monoxide (CO), nitrogen oxides and exhaust volatile organic compounds (Bailey 1996; Wyman 1996). However, of environmental concern regarding the increased use of ethanol fuels [3] is the increased exhaust emission of reactive aldehydes, such as acetaldehyde and formaldehyde. Thus, a key factor with respect to the possible effects of ethanol on urban air quality will be the durability and effectiveness of catalyst systems for aldehyde control. Today, all cars with a catalyst can be run on a mixture of 90% gasoline and 10% ethanol without adjusting the engine.

New cars can even use mixtures containing up to 20% ethanol. There are also new engines available that can run on pure ethanol, and so-called flexible fuel vehicles that are able to use mixtures of 0–85% ethanol in gasoline (E85). Ethanol can also replace diesel fuel in compression-ignition engines. However, to be able to mix diesel with ethanol an emulsifier is needed. Fuel ethanol is used in a variety of ways; however, the major use of ethanol today is as an oxygenated fuel additive (Wheals et al. 1999). Mixing ethanol and gasoline has several advantages. The higher octane number of ethanol (96–113) increases the octane number of the

mixture, reducing the need for toxic, octane-enhancing additives. Ethanol also provides oxygen for the fuel, which will lead to the reduced emission of CO and no combusted hydrocarbons. Bailey (1996) has summarised the pros and cons of replacing spark-ignition and compression-ignition engines with optimised ethanol engines.

His conclusions were that ethanol has about the same overall transport efficiency as diesel in compression-ignition engines, but is about 15% more efficient than gasoline in optimised spark-ignition engines (Bailey 1996). So, even though ethanol has only about two-thirds of the volumetric energy content of gasoline, it will still be possible to drive 75–80% of the distance on a given volume of ethanol (Wyman 1996). Despite the advantages of bioethanol, Brazil and the United States are still the only two countries that produce large quantities of fuel ethanol (from sugar cane and maize, respectively Wheals et al. 1999). Brazil produces 12 million m³ ethanol per year and the United States about half this amount. Fuel ethanol production is considerably more modest in the European Union, where France and Spain are the largest producers; each country producing about 200,000 m³/year, used for production of ethyl tertiary butyl ether. However, countries such as The Netherlands, Italy and Portugal have also shown interest in ethanol. In Sweden, an ethanol plant based on grain, with a capacity of 50,000 m³/year, came into operation in 2001. The ethanol is used as a 5% blend in gasoline.

1.1 Efficiency of Ethanol:

The efficiency of ethanol[4] production from biomass has steadily increased, but tax relief will be required to make fuel ethanol commercially viable compared to oil (Wheals et al. 1999). To attain an economically feasible process, the production cost must be reduced so as to approach the corresponding cost for fossil fuels. When using sugar- or starch-containing feedstocks, such as sugar cane or maize, the raw material accounts for 40–70% of the total ethanol production cost and the production cost is also dependent on animal feed as a by-product (Claassen et al. 1999). To achieve lower production costs, the supply of cheap raw materials is thus a necessity. When lignocellulosic raw materials are used, the main by-product is lignin, which can be used as an ash-free solid fuel for production of heat and/or electricity for which there are no foreseeable market limits. Accordingly, it will only be possible to produce large amounts of low-cost ethanol if lignocellulosic feedstocks such as fast-growing trees, grass, aquatic plants, waste products (including agricultural and forestry residues) and municipal and industrial waste are used (Wheals et al. 1999). The potential of using lignocellulosic biomass for energy production is even more apparent when one realises that it is the most abundant renewable organic component in the biosphere. It accounts for approximately 50% of the biomass in the world, with an estimated annual production of 10–50×10¹² kg (Claassen et al. 1999) [5].

2. DISCUSSION

2.1 Conversion Process Pathways

Bio-ethanol can be produced from a large variety of carbohydrates (mono-, di-, polysaccharides). Monosaccharides (glyceraldehydes, xylose, ribose, glucose, fructose) consist of single sugars bound together with a general formula of (CH₂O)_n, where n = 3-7. The most common monosaccharides in nature are pentoses (n = 5, xylose) and hexoses (n = 6, glucose). When glucose is the most widespread sugar transport form in animal organisms, in the plants, sugar is often transported in the form of disaccharides (sucrose, maltose and lactose). One molecule of disaccharide results from a chemical reaction (dehydration synthesis) in which a new bond is formed between two monosaccharides after removal of one molecule of water. Polysaccharides are often organised as chains of bonded monosaccharides molecules, which result from dehydration syntheses. Polysaccharides composed of similar subunits (monomers) are called polymers. Starch and cellulose are polymers composed of monomers of glucose. Polysaccharides must be decomposed in disaccharides and/or monosaccharides through hydrolysis before fermentation to ethanol.

Large-scale biomass-to-ethanol industry mostly uses sugarcane or sugar beet juice, corn or wheat. Ethanol is also commercially produced in the pulp and paper industry as a by-product. Lignocellulosic biomass is envisaged to provide a significant portion of the feedstocks for bio-ethanol production in the medium and long term due to their low cost and to their high availability. For a given production line, the comparison of feedstocks includes chemical composition of the biomass, cultivation practices, availability of land and land use, use of resources, energy balance, emission of greenhouse gases, acidifying gases, ozone depletion gases, emissions of minerals to water and soil, emission of pesticides, soil erosion, contribution to biodiversity and landscape value losses, farm-gate price of the biomass, logistic cost (transport and storage of the biomass),

direct economic value of the feedstocks taking into account the co-products, creation or maintain of employment. Different authors have proposed criteria and methodologies for assessing the sustainability of energy crops. Biomass-to-ethanol crops comprise:

- (i) Multipur- pose crops that are also devoted to food markets; and
- (ii) Dedicated ethanol crops.

While the latter are cultivated especially for ethanol production on non- agricultural lands[6] (fallow or undeveloped lands), the former provide almost all the feedstocks used to date for ethanol production (sugarcane in Brazil and corn in the United States). In most industrialized countries, the development of biomass-to-ethanol conversion emerged as alternative markets for sugar and grain surpluses. As the feedstock cost often represents more than 75 percent of the ethanol production in these cases, the economic viability of multipurpose crops- to-ethanol depends on the food markets situations (sugar and grain markets). This correlation between food and ethanol markets may generate a volatility of the ethanol prices. In developing countries, the possible competition with food is one of the risks when using agricultural crops for ethanol production. Thus this option should be limited to cases where actual and sustainable surplus of crops occurred. Finally, ethanol production cost is scale sensitive. Feedstock and investment cost affect economy of scale in different ways. Marginal costs of feedstock collection and transport increase with the size of the ethanol plant whereas marginal investment cost decreases. Optimal sizes (50-500 million l/y) of ethanol plants depend on the particular context under study (availability of feedstocks, demand of ethanol, cost of transport and storage).

2.2 Main Steps in Biomass-to-Ethanol Processes

Once the biomass is delivered to the ethanol plant, it is stored in the warehouse and conditioned to prevent from early fermentation and bacterial contamination (Fig. 1). Through pre-treatment, carbohydrates are extracted or made more accessible for further extraction. During this step, simple sugars may be made available in proportions depending on the biomass and the pre-treatment process. A large portion of fibers may remain for saccharification through hydrolysis reactions or other techniques, in order to obtain simple sugars, which are then fermented.

In the batch fermentation, the hydrolysate, the yeasts, nutrients and other ingredients are added from the beginning of the step. In case of a fed batch process, one or more inputs are added as fermentation progresses. Continuous processes, in which ingredients are constantly input and products removed from the fermentation vessels, are also used⁴. In efficient processes, the cell densities are made high by recycling or immobilising the yeasts in order to improve their activity and increase the fermentation productivity. The fermentation reactions occur at 25-30°C and last 6-72 h depending on the composition of the hydrolysate, the type, the density and activity of the yeasts. The broth typically contains 8-14 % v/v glucose and fructose. Fermentation of sucrose is performed using commercial yeast such as *Saccharomyces cerevisiae*. Chemical reaction is composed of enzymatic hydrolysis of sucrose followed by fermentation of simple sugars. First, invertase (an enzyme present in the yeast) catalyzes the hydrolysis of sucrose to convert it into glucose and fructose.



Then, another enzyme (zymase), also present in the yeast, converts the glucose and the fructose into ethanol and CO₂.

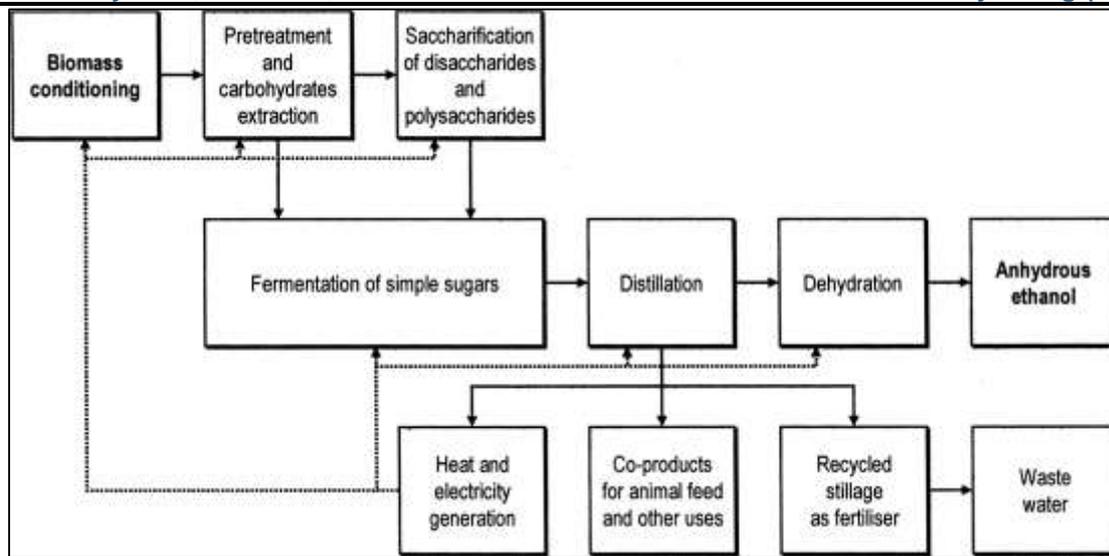


Figure 1: Biomass to Ethanol process

2.2.1 Starch-to-Ethanol Process

Starch stored in grains is long chains of α -glucose monomers, 1000 or more monomers for one amylose molecule and 1000 to 6000 or more monomers for amylopectin. To convert starch to ethanol, polymer of α -glucose is broken into glucose through a hydrolysis reaction with gluco-amylase enzyme. Hydrolysis is then followed by fermentation, distillation and dehydration to yield anhydrous ethanol. In the bio-ethanol fuel industry, grains (corn, wheat or barley) mainly provide starch. Corn (60-70% starch) is the dominant feedstock in starch-to-ethanol industry worldwide. In dry milling process, grain is grinded to a powder which is hydrolysed and the sugar contained in the hydrolysate is converted to ethanol while the remained flow containing fibre, oil and protein is dried and converted into a by-product known as Distillers Dried Grains (DDG) or DDGS when it is combined to process syrup[7].

2.3 Lignocelluloses-to-Ethanol Process

2.3.1 Structure of Lignocelluloses Materials

Lignocellulose, which is the principal component of the plant cell walls, is mainly composed of cellulose (40-60% of the total dry weight), hemicellulose (20-40%) and lignin (10-25%). Cellulose molecules consist in long chains of glucose monomers gathered into micro-fibril bundles. The hemicelluloses can be xyloglucans or xylans depending on the types of plants. The backbone of the former consists of chains of glucose monomers to which chains of xylose (5-C sugar) are attached. The latter are mainly composed of xylose linked to arabinose or/and other compounds that vary from one biomass source to the other. The hemicellulose molecules are linked to the micro-fibrils by hydrogen bonds. Lignins are phenolic compounds which are formed by polymerisation of three types of monomers (p-coumaryl, coniferyl and synapyl alcohols), the proportion of which differs significantly depending whether the plant is from gymnosperms, woody angiosperms or grasses. Lignin adds to the cell wall a compressive strength and stiffness.

Lignocellulose is abundant in nature and does not compete with food. Typical sources of lignocellulosic biomass are agricultural and forestry residues (biogases of sugarcane or sweet sorghum, corn stover, Grasses, woody biomass) industrial wastes and dedicated woody crops (poplar). Once the lignocellulosic biomass is pre-treated, and hydrolysed, the released sugars are fermented and the down-stream process is similar to that of sweet juice and starch.

2.3.2 Pre-treatment

Pre-treatment involves delignification of the feedstock in order to make cellulose more accessible in the hydrolysis step, using physical, physico-chemical, chemical and biological treatment. Performance of a few methods is assessed with regard to the yield of fermentable sugars, inhibitors, the recycling of chemicals, the production of wastes and the investments. It results from this comparison that carbonic acid and alkaline extraction have the best performance. However, the most common methods are steam explosion and dilute acid pre-hydrolysis, which are followed by enzymatic hydrolysis. In steam explosion method, the lignocellulosic materials are treated with high-pressure saturated steam (0.69-4.83 MPa) at high temperature (160-260 °C) for several seconds to a few minutes. Then the pressure is suddenly dropped to atmospheric

pressure causing the materials explosion. Most of the hemicellulose is solubilised during the process whose efficiency depends on the temperature and residence time. Lower temperature and longer residence time give a higher efficiency. Sulphuric acid or carbon dioxide is often added in order to reduce the production of inhibitors and improve the solubilisation of hemicellulose. Steam explosion has a few limitations: i) Lignin-carbohydrate matrix is not completely broken down; ii) Degradation products are generated that reduce the efficiency of hydrolysis and fermentation steps; and iii) A portion of xylan fraction is destroyed.

The use of dilute acid has the preference of the US National Renewable Energy Laboratory. In this method, the structure of the lignocellulosic material is attacked with a solution of sulphuric acid (0.5- 1.0%) at about 160-190 °C for approx 10 min. During this reaction, hemicellulose is largely hydrolysed releasing different simple sugars (xylose, arabinose, mannose and galactose) but also other compounds of the cellulosic matrix, a few of which can inhibit the enzymatic hydrolysis and fermentation. The stream is then cooled. Part of the acetic acid, much of the sulphuric acid and other inhibitors produced during the degradation of the materials are removed. Finally, neutralisation is performed and pH is set to 10 before hydrolysis and fermentation.

2.4 Development and Deployment

Contrary to the conversion of sweet juice and that of starch to ethanol which are mature technologies, modern lignocellulose-to-ethanol process [8] still is on pilot and demonstration stage. NREL (USA) has built a pilot plant based on SSCF method capable to process one ton of dry material per day. Iogen Corporation (Canada) built in 2003 a demonstration plant with an annual production of 320,000 litres of ethanol, using wheat straw as feedstock and a sequential steam explosion pre-hydrolysis – cellulose production – enzymatic hydrolysis of cellulose and co-fermentation of xylose and glucose. In 2004, a Sweden company, ETEK, has developed a pilot plant capable to produce 150,000 litres of ethanol per year using soft wood as feedstock.

2.5 Use of Bio-Ethanol in Spark Ignition Internal Combustion (IC) Engines

Ethanol has good properties for spark ignition IC engines. Its Motor Octane Number (MON) and Research Octane Number (RON) [9] are respectively 90 and 109 leading to an average octane number of 99 compared to 88 for regular gasoline. Lower heating value (LHV) [10] of ethanol (21.2 MJ/l) is two-thirds that of gasoline (30.1 MJ/l). Bio-ethanol fuel is used in IC engines as 5-26 percent anhydrous ethanol blends to gasoline (5% maximum in Europe and in India, 10% in USA, 22-26% mandatory blends in Brazil) or as pure fuel (100%) of hydrated ethanol in dedicated vehicles. When anhydrous bio-ethanol is blended to gasoline in small proportion (up to 15%), influence of lower LHV has no significant effect. For higher blend levels, fuel economy is reduced compared to that with conventional gasoline.

Ethanol dedicated vehicles are optimised so that the engine efficiency is improved by running at higher compression ratios to take advantage of the better octane number of ethanol compared to gasoline. Therefore, for pure hydrated ethanol used in optimised vehicles, ethanol can achieve about 75 percent or more of the range of gasoline on a volume basis. Finally, special vehicles known as Flexible Fuelled Vehicles (FFV) [11] are equipped with line sensors, which measure ethanol levels and adapt air-fuel ratio to maintain good combustion conditions. These vehicles can burn fuel containing ethanol (0-85%) in gasoline and are becoming more and more frequent in Brazil and Sweden with various manufacturers developing such vehicles on commercial scale. As compression ratio cannot be adapted in case of FFV, the reduction in fuel economy is somewhat proportional to the bio-ethanol content in the fuel blend. Apart from this issue, use of bio-ethanol in IC engines exhibits a few disadvantages: (i) Low levels of ethanol blended with gasoline increases vapour pressure and favour evaporative emissions that contribute to smog formation, and (ii) For higher ethanol blend levels, vapour pressure drops significantly leading to more difficulty in cold weather conditions.

2.5.1 Use of Ethanol in Compression Ignition (CI) Engines

Due to its low cetane number, ethanol does not burn efficiently by compression ignition. Moreover, ethanol is not easily miscible with diesel fuel. Three directions are followed to improve the use of ethanol in CI vehicles. The first that consists in direct blend of ethanol with diesel needs addition of an emulsifier in order to improve ethanol-diesel miscibility. Other additives are used such as ethylhexyl nitrate or di-tert-butyl peroxide in order to enhance the cetane number. Most of blends of ethanol to diesel (E-diesel) limit by up to 15 percent ethanol and up to 5 percent emulsifiers. The second way is a dual fuel operation in which ethanol and diesel are introduced separately into the cylinder¹⁵. Finally, modification of diesel engines has been experienced in order to adapt their characteristics of auto-ignition and make them capable to use high blends such as 95% ethanol.

2.6 Greenhouse Gas Balance

The average net CO₂ balance of bio-ethanol production is not presented here, because of the additional issues of system boundaries and variations of the incorporation rate of ethanol to gasoline. Indeed, if the common practice for comparing the net energy balance of ethanol to that of conventional fossil fuels does not generally include the utilization phase of the fuels (although it should be), it is more often the case as far as the CO₂ balance is concerned. In such a case, the incorporation rate of ethanol within gasoline (and thereby the performance of the blend) is of major importance, although often neglected, which makes it even more difficult to compare the results of different studies. The Laboratory of Energy Systems (LASEN) of the Swiss Federal Institute of Technology of Lausanne (EPFL), Switzerland, has conducted various studies concerned with the life cycle assessment of various pathways of ethanol production and use. With an incorporation rate of 5% anhydrous ethanol within gasoline and an equal performance with respect to conventional gasoline, the net savings of greenhouse gas (GHG) emissions were found to vary between 2.0 (agricultural feedstocks) and 2.5 (waste lignocellulosic biomass) kg of CO₂ eq. per litre of ethanol incorporated to gasoline. In these evaluations, the life cycle inventory was described in the context of Switzerland with economic allocation, and the reference vehicle was a recent standard 1.6 l light passenger vehicle.

2.7 Other Environmental Effects

As ethanol contains more oxygen than gasoline, its use favours more complete combustion and reduces the emissions of particulate matter (PM) and hydrocarbons (HC), which result from incomplete combustion of gasoline. Tailpipe emissions of carbon monoxide (CO) and sulphur dioxide (SO₂) are also improved. However, low levels blends of ethanol with gasoline can increase the emissions of volatile compounds (VOCs) and nitrogen oxides (NO_x). These emissions favour ozone formation. Emissions of aldehydes (mostly acetaldehydes) and peroxyacetyl nitrate (PAN) also increase to an extent that depends on weather conditions. The use of catalytic converters reduces the emissions of aldehydes. Reducing in refinery the vapour pressure of gasoline, which is blended with ethanol, can prevent VOCs emissions. Experiments about different percentages of ethanol- diesel blends show significant advantages concerning PM, NO_x and CO. However, no evidence is given for improvement of HC emissions. Ethanol is more corrosive than gasoline and diesel, and at high concentration, can damage fuel system components. For low-levels blends, these concerns are limited and E5 or E10 can operate on existing vehicles without violating most of manufacturers' warranties. For high concentrations of ethanol, compatible materials are used in dedicated designed vehicles.

2.8 Production of Bio-ethanol from Sweet Sorghum Juice

The present ethanol facility processes whole sorghum stems from which the juice is extracted and separated from the biogases. With a total project investment of 54.5 million US\$ and under the same economic conditions as described above, the capital cost amounts to 5.4 million US\$ per year. The price of sorghum stems was taken as 12.50 US\$/ton, while the main by-product (sorghum biogases) is sold at 5 US\$/ton. The stillage is returned to the farmers and used as a liquid fertilizer in agricultural fields but was considered to bring zero revenue (large volumes, little added value). The plant operates 100 days per year (due to sorghum storage limitations) and employs 400-450 people. The net production cost amounts to about 0.25 US\$/l and again is strongly dependent upon the cost the feedstock and the price at which the biogases (about 550,000 ton/y) is sold.

2.8.1 Production of Bio ethanol from Sweet Sorghum Biogases

Each ton of sweet sorghum generates (after extraction) approx 640 kg of sweet juice and 360 kg of biogases. The biogases (30-35% of the plant's fresh wt, moisture content 50% wt) consists mainly lignocellulosic residue left after extraction of the sugar. It has a lower heating value (15-18 MJ/kg). For every thousand litre of anhydrous ethanol produced quantity of biogases generated amounts to 4.6 ton. This corresponds to about 70-80 MJ (2.2-2.6 kg coal equivalent) per litre of anhydrous ethanol, which would be sufficient to cover all the energy needs (heat and power) of the ethanol production process. The biogases, however, also represents a significant potential for further alcohol production, with an ethanol yield of 157 l/ton of biogases (56 l/ton of sweet sorghum). In comparison, the ethanol yield from sweet juice reaches 121 l/ton (77 l/ton of sweet sorghum).

3. CONCLUSIONS

Due to its environmental merits, the share of bio- ethanol in the automotive fuel market will grow fast in the next decade. Having developed long experience with low-level ethanol blends (E10 to E26), as well as with nearly pure ethanol (E85), Brazil and the US benefit from learning curve and particularly favorable conditions with regards to agricultural feedstocks, sugarcane for Brazil and corn for the US. Especially,

Brazil exhibits the lowest production costs of bio-ethanol fuel worldwide and is in position to capture a large share of the international market in the future. However, the market price of bio-ethanol fuel will fluctuate as a result of the balance between demand and supply of bio-ethanol, oil and sugar. It is likely that the trend will be for an increase as a consequence of the fast growth of the world demand in the future. India is one of the first producers and consumers of sugar worldwide and it is not envisaged for this country to use sugarcane for producing part of its future requirement of bio-ethanol fuel. However, molasses of sugarcane can be used complementarily to sweet sorghum juice. It is assumed that a portion of grain sorghum is replaced by sweet sorghum without any additional agricultural land. Although the results show that India is not competitive with Brazil, opportunities for Indian production of bio-ethanol must be analyzed in more detail in the light of both future oil and ethanol markets. In a longer term, lignocellulose-to-ethanol is very promising due to its independence from food markets. It is expected that the production cost will decrease as a result of the research works undertaken in several countries. Finally, due to the higher potential of CO₂ emissions reduction of this pathway, use of Clean Development Mechanism (CDM) should be envisaged to reduce its production cost.

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