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BTL TECHNOLOGIES

Ademar Hakuo Ushima

INTRODUCTION

The energy content of the main residues generated in the Brazilian ethanol and sugar mills, bagasse, trash and vinasse, are currently been wasted or used inefficiently and presents a huge economic potential for the production of fuels, power and products of higher value-added like chemicals products by using BTL technology – Biomass to Liquids. At the same time this technology can contribute to the solution of serious environmental problems, such as those caused by burnt trash in the field and contamination of the water table by vinasse dispersion in the soil.

In addition, the employment of biomass as power and energy supply does not compete with food production as the first generation technology does, based on production of ethanol or biodiesel from sugar, starch and vegetable oils, and does not stimulate agricultural area expansion in the country.

DESCRIPTION OF BTL TECHNOLOGY

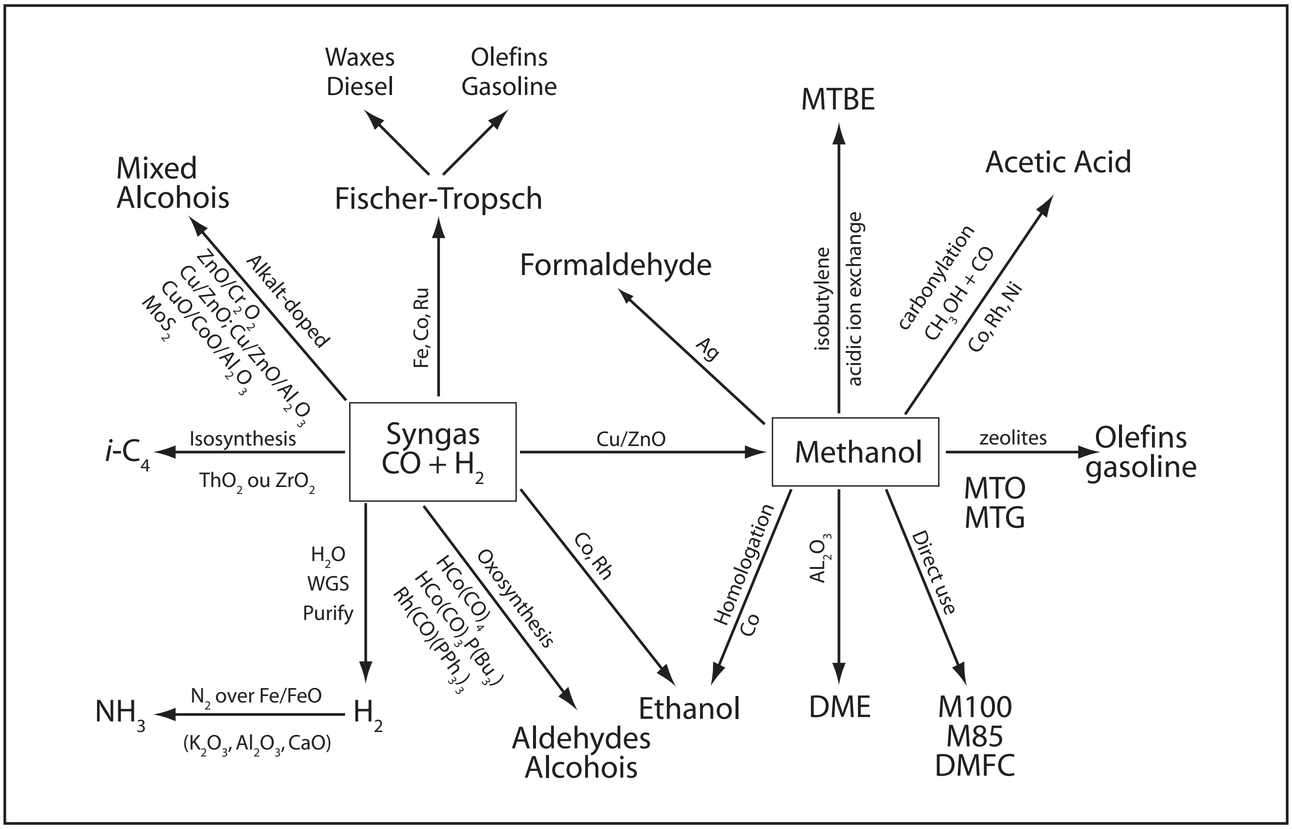
BTL technology, briefly, is the conversion of biomass into synthesis gas (gas with high concentration of CO and H_2), followed by conversion of the gas into liquid chemicals products via catalytic reactors. It is one of the main technological routes currently in development in the world for the utilization of the solar energy accumulated in the biomass, also called second generation technologies, able to convert the current sugar and ethanol mills/distilleries into petrochemicals refineries equivalent units, also known as biorefinaries.

Comparatively to biomass enzymatic hydrolysis technology, BTL technology offers the advantage to use all the components of biomass (lignin, cellulose and hemi-cellulose), and not only a fraction of it, as is the case of enzymatic route that can convert only cellulose into liquid fuels. It can also be a supplementary to the enzymatic route, converting fractions of lignin and hemi-cellulose into gaseous fuels or power.

CTL technology – coal to liquids, equivalent to the BTL in the carbochemical sector, uses coal or petrochemical wastes for the generation of synthetic gas, and is already commercial, with several units in operation or under construction in world, especially in South Africa, China, Europe and USA. The consumption of mineral coal in the US in 2007, only for the production of chemicals via gasification, reached 30.8 million t (DOE/EIA-0121-2008/01Q). In Brazil a fertilizer industry (450 MW_{th}), located in Araucaria city, Paraná, is producing ammonia from the gasification of asphaltic residue (RASF) with oxygen since 1983. RASF consumption unit is 1,000 t/d and ammonia production is 1,300 t/d.

Some of the products which can be obtained through BTL technology are presented in Figure 1. As can be seen, the range of products is very extensive, extrapolating liquid and gaseous fuel production.

Among the fuels that can be generated from BTL technology it can be highlight methanol, ethanol, gasoline, diesel oil, dimethyl ether – DME, hydrogen etc., all of them, with the exception of



Source: NREL/TP-510-34929, dec. 2003.

FIGURE 1 Products that can be obtained from synthesis gas.

ethanol, presently obtained in large scale from fossil fuels.

A chemical with great potential generation from biomass and of great economic importance in Brazil is ammonia (NH_3), consumed in large scale as fertilizer. It is obtained currently from fossil fuels (NG and petrochemicals waste) and a large part of the Brazilian consumption is imported, largely used in sugarcane cultivation.

A generic flowchart of BTL process is presented in Figure 2. Biomass, with humidity around 10% m. b., is gasified with oxygen in a pressurized gasifier, generating synthesis gas. The oxygen is often used in place of air due to the higher temperatures attained in the gasifier, which leads to an increased conversion rate of biomass into gas and lower tar content. Another important factor is that the nitrogen of the air generally acts as an inert agent in the synthesis reactor increasing the flow of gas in the reactors, power consumption and investment in equipment.

The gas generated in the gasifier must be cooled and cleaned before being sent to a catalytic reactor. Depending on the type of gasifier used (co-current, counter current, fluidized and entrained bed) the exit temperatures of gases and the levels of contaminants can vary significantly (150 °C to 1,400 °C and 1 to 100 g/Nm^3, respectively) as well the heat recovery and gas cleaning systems. In most of the gasification process gas leaves at high temperatures and its enthalpy can be used to generate steam, before its cleaning, which can be dry or wet. In the gas cleaning operation particulate material, tars and ammonia present in the gas must be removed and the water contaminated with them must be treated before their remanufacturing or disposal of.

In biomass gasification with oxygen, the molar ratio H_2/CO is usually just below 1 and for catalytic synthesis processes, depending on the product this relationship has to be higher. For example, in the case of methanol or aliphatic hydrocarbon production, the relationship must be slightly above 2. To do this, after gasification, usually it is introduced a catalytic conversion of CO to H_2, called “shift”.

In the “shift” reactor CO reacts with steam, generating H_2 and CO_2 that is removed from the gas by acid gas absorption via regenerative solutions as mono and diethanolamine compounds, potassium hydroxide, methanol at too low temperatures etc. In these systems can also occur separation of other contaminants as H_2S, COS, H_2O and hydrocarbons larger that methane. The elimination of these contaminants from synthesis gas is important because they cause the deactivation of catalytic converters and the levels permitted usually fall below 1 ppmv. The regeneration process of the absorbing solutions, which involves heating and decompression, usually demands significant quantities of energy, and in the evaluation of the process economical viabilities it must be considered.

Clean synthesis gas usually needs an additional compression step, since the synthesis reactors operates at pressures higher that can be achieved in gaseifiers, ranging from 60 to 100 bar.

The compressed gas is sent to a catalytic reactor where it is converted in product and by-products. The rate of conversion of the gas in the reactor never is complete, typically rated at 35%, which obliges the recycling of gases that is not converted. As some components of synthesis gas present in smaller quantities, such as CH_4 and N_2, act as inert in catalytic reactors synthesis, there is the need to promote a system purge of these gases, to avoid its accumulation in the internal circuit. To minimize the exhaust of reagent gases (mainly H_2) with the purge gas, PSA units (Pressure Swing

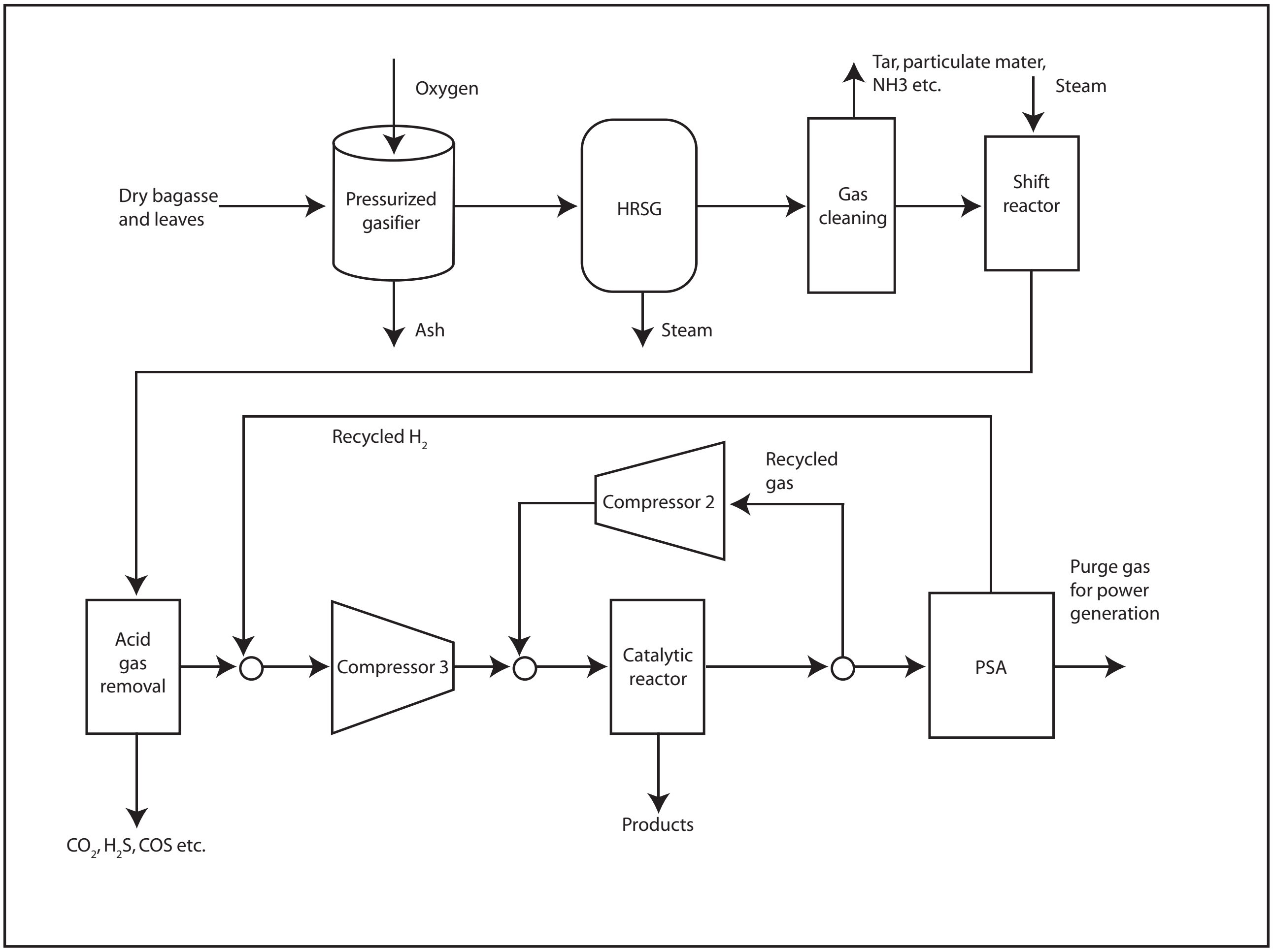


FIGURE 2 Generic flowchart of BTL process.

Adsorption), based on difference of diffusivity in porous media, promotes the remaining hydrogen separation of the gases, which is recycled to the process, as shown in Figure 2. The purge gas still contains high levels of fuel gases, such as CO and CH_4, and can be burned for steam generation and electrical power generation via Rankine cycle or gas turbines.

The product of the catalytic reactor is a mixture of components and often must pass by separation processes, typically distillation, for purification of the liquid fuel or chemical product.

In some situations in which electric energy presents higher profitability than liquid fuels (e.g. peak hours), the generation of synthetic products can be reduced and of electrical energy increased, simply reducing the recycle of gas in the compressor 2 and increasing the purge gas flow in the scheme presented at Figure 2. A similar situation already occurs in the Brazilian sugarcane mills, which can vary the production of sugar and ethanol, depending on their profitability.

An alternative route to the catalytic synthesis is the biological synthesis using bacteria cultures (generally Clostridium Ljungdahlii, as KASTEREN *et al*., 2005) able to synthesize liquid ethanol from synthesis gas. The advantages of this process is to enable the use of biomass gasification at pressures slightly above the atmospheric, air as gasification agent (since bacteria are not inhibited by the presence of nitrogen in the air) and the bacteria is tolerant to the presence of small quantities of contaminants in the gases, as tars. One of the main limitations of this process is the low solubility of H_2 and CO in water and low ethanol concentration in the solution.

STAGE OF BTL TECHNOLOGY IN THE WORLD AND IN BRAZIL

At moment there is no unity in the world producing liquid fuel from biomass in commercial scale via BTL route, but several pilot scale and demonstration units are in development.

One that is in the most advanced stage is CHOREN, located at Freiberg, Germany, was inaugurated in June 2008. That is a demonstration unit, with design capacity to process 68 000 t/year of dry wood chips and produce 18 million l/year of liquid fuel, that 21% of the total is diesel oil, should be operational in mid Jun 2010. The gasification system, developed initially in pilot scale, is composed of two stages. In the first, biomass is pyrolysed in a reactor at 400 °C to 500 °C, generating a stream of gases and vapors and other of charcoal. The gaseous phase is sent to a second reactor in which reacts with oxygen, reaching high temperatures and where tars are converted in gaseous components. The solid phase, after milling, enters in contact with the gases at high temperature in the middle of the second reactor region, suffering gasification and cooling the gases. The synthesis gas formed is passed through a heat recovery system for steam generation, a filter to remove the particulate matter entrained, a shift reactor to set the molar ratio of H_2/CO around 2, an acid gas absorber and a catalytic reactor, very similar to the flowchart shown in Figure 2. The unit of gasification described can also be used for fuel gas and electricity generation with, in this case, the replacement of oxygen by air. This development began in 1998 and the diesel oil generated at the pilot-scale unit was free of sulfur and aromatic compounds and presented high cetane number.

Another one in an advanced-stage development is CHEMREC, maintained by a Swedish consortium, consisting of black liquor gasification, a waste generated in the cellulose production, in place of biomass, with oxygen. Because black liquor is liquid, it dramatically simplifies the feeding system to pressurized gasifiers, eliminating one of the bottlenecks of biomass gasification. Black liquor gasifier is pressurized and of entrained flow type, similar to those used commercially in petrochemical refineries for petrochemical wastes gasification, with internal coatings resistant to chemical attack of reagents present in the black liquor. The gasifier also enables the recovery of the reagents, similar to the recovery boilers used presently in cellulose factories for steam generation. Once cooled and cleaned, the synthesis gas generated can be used for the production of liquid fuels or chemicals through routes already presented. In September 2008 the construction of a DME production unit with a capacity of 5 t/d was started and that will use the synthesis gas generated at a black liquor gasifier installed in Pitea, Sweden, with financial resources of order 28.4 M € (CHEMREC Press Release 08.Sept.09).

DME is a fuel that can replace diesel oil in diesel engines with minor modifications, and presents a high cetane number with very low emissions of air pollutants. It is not toxic and can even replace LPG in a proportion of up to 20% in LPG barrels used for domestic purposes. In addition, several direct production units of DME from synthesis gases has already been proven in demonstration scale, with high efficiency and reliability (OHNO *et al*., 2007; HEYDORNn *et al*., 2003). In these units, the molar ratio of H_2/CO required in the reactor entry was around 1, near the ratio obtained in biomass gasifiers employing oxygen, which can make unnecessary the “shift” reactor presented in Figure 2, reducing the investment and operational cost.

A third developing project that deserves highlight is of the National Renewable Energy Laboratory – NREL, of USA. The pilot scale gasifier is designed for wood chips with low humidity, and gasification with indirect heating and catalytic reform of tars formed, as described in PHILIPIS *et al*. (2007). The advantage of this process is that it doesn’t need an air separation unit – ASU, which can reduce significantly its investment and operational cost. The biomass heating and gasification occur in a bubbling fluidized bed sustained by heated steam, through the contact of biomass with the bed at around 850 °C, composed mainly of olivine (magnesium silicate and iron). The olivine is heated in an attached fluidized bed reactor, trough the combustion of the charcoal formed in the gasifier, with air. The olivine is maintained in circulation between the two fluidized beds and gases generated in the gasifier go through a fines collection system, and are sent after to a catalytic bed, indirectly heated to promote the reform of hydrocarbons present in the gases such as methane, ethylene, aromatic compounds etc. After the gas cleaning and the removal of all contaminants still present, the synthesis gas will be compressed and converted primarily to ethanol, in catalytic reactors with di-molybdenum sulfide (MoS_2).This catalyst has a number of advantages as greater selectivity for ethanol, more tolerance to the presence of H_2S and CO_2 than other catalysts and be able to promote the “shift” of CO to H_2, allowing the use of synthesis gas with a molar ratio H_2/CO near 1.

Brief information about other demonstration units using BTL technologies and their development stages, is presented in Table 1.

In Brazil some research institutes, universities and private companies have been developing biomass gasifiers, targeting mainly fuel gas production and not synthesis gas. To date, only IPT (Institute for Technological Research) has developed biomass gasifiers in pilot scale for synthesis gas generation using oxygen and steam as gasification agent, at pressures slightly higher than atmospheric pressures, as described in USHIMA (2008). From 1999 to 2004 tests for synthesis gas production were conducted in a bubbling fluidized bed, using sugarcane bagasse pellets, and a co-current gasifier employing eucalyptus pellets. The capacity of both gasifiers was around 100 kg pellets/h.

TABLE 1 Others project under development in the world using BTL technology.

Company	Biomass consumption (t/d)	Produced fuel	Capacity	Investment $(10^6\ \text{US}\$)$	Technology	Country
Range Fuels	1200	Ethanol	38.10^6 l/y	76	Indirect heating	USA
Alico, Inc.	700	Ethanol	27.10^6 l/y	33	Síntese biológica	USA
Enerkem	~45	Ethanol	5.10^6 l/y	***	Biological synthesis	Canada
Chrisgas	~86	Hydrogen	3,500 N, 3/h	~22	Entrained flow gasifier	Sweden

TECHNICAL AND ECONOMICAL FEASIBILITY STUDIES

Due to escalation of petroleum prices, the aggravation of global warming and growing concern with the energy dependency, various studies on technical and economic feasibility of BTL technologies have been submitted, as developed by HAMELINCK *et al*. (2002 and 2003), SPATH and DAUTON (2003) and EKBOM *et al*. (2005).

Because the current development stage of BTL technologies and the lack of a full demonstration-scale unit operation (from biomass treatment to liquid fuel produced), most feasibility studies cited are based only on a series of estimates costs and performance, mainly of equipments in development stages, as pressurized biomass feeding systems, pressurized gasifiers, gas cleaning systems and catalytic reactors.

Any case, in all of them, the required investment cost is the most significant and the factors impacting the fuel cost produced via BTL route were investment rate (40% to 60%) and biomass cost (around 30%) that in Brazil, by being one of the smallest of the World (~1.7 US$/GJ), can represent a huge differential for the deployment of this technology.

The majority of the previous studies cited – before to petroleum price peak in mid 2008 – indicates that the cost of liquid fuels obtained via BTL technology will be greater than that of equivalent fossils fuels, without taking into account the environmental benefits generated, and will be economically viable only with tax exemption.

R&D NEEDS FOR BTL TECHNOLOGY

Despite of the high cost of BTL technology development and the large fluctuations of petroleum and its derivates prices in recent times, there have been significant advances in the BTL technologies development abroad, as reported previously.

In Brazil, however, despite of the enormous potential of biomass generation with low cost (mainly sugarcane bagasse and trash), has done little progress on BTL technology development, as was evidenced in the BTL Workshop held in Institute for Technological Research – IPT, São Paulo, in February 2008.

The need for training a larger number of researchers in this area through scholarships and technical exchange with outside research centers was highlighted as very important in the Workshop. The settlement of fund lines to finance the cooperation with international institutions in gasification also should be fetched.

With the shortage of researchers in this area, dispersion of efforts and a great number of low cost minor works, it has not been possible get advances in this route and offer alternatives to the market.

Another scrappy factor for the development of BTL technology in Brazil, raised in the debate, was intellectual property. The private sector, in general, doesn’t has enough resources or lacks sufficient degree of confidence to perform the investments (million US$) required for the development of the BTL technology production chain. In this way, Brazilian industrial sector has only invested in the development of some of the chain links, in isolated and disorganized form, along with universities and research centers, which has hampered and even prevented the exchange of experiences of these centers and universities, on behalf of the industrial property issue. This way may also explain the occurrence of redundancy in various universities and research centers.

In the BTL technology route, many gaps have been identified, since the misunderstanding of important properties of biomass till applied technologies at the end of the production chain of liquid fuels.

As example, in the pretreatment of sugarcane bagasse and trash, the first used intensively for decades, some of their basic properties is unknown, such as the internal structure and composition of fibers, the difference between the structure of sugarcane bagasse and trash from other biomass as eucalyptus etc, that are essential for their recalcitrance reduction, which can enable their direct utilization as solid fuel or as raw material for the second generation technologies, as gasification or hydrolysis. Researches in the pretreatment of biomass, mainly sugarcane bagasse and trash, are very important for the development of industrial processes like combustion, gasification, pellet production, slow and fast pyrolysis (with bio-oil generation), acidic and enzymatic hydrolysis, focusing in the recalcitrance reduction of these materials and equipments wearing.

In the development of biomass feeding systems to pressurized reactors it was found that, for biomass with high densities and granular form (pellets, briquettes, chips) these systems already exist, simply adapting the existing systems for coal. For biomass of low density, as is the case of sugarcane bagasse and trash, it will be necessary to develop equipments capable of maintaining a regular feeding into pressurized environments. Abroad there is some equipment in the development stage, such as solid pump, that must be tested with sugarcane bagasse and trash.

For the development of biomass gasification systems, it must be necessary to provide funding lines for the development of atmospheric and pressurized circulating fluidized bed – CFD and gasifiers with separated pyrolysis and gasification stages, to increase the participation of new researchers in the projects, to arrange events and summer schools specialized in this subject (RENEW and SYNBIOS), to construct pilot scale plants for long-term tests (more than 1,000 h) and technical feasibility proof, and after that the building of a demonstration unit, always seeking the partnership of universities, research centers, governmental funding agencies and private companies.

In the gas treatment step, apparently the difficulty is more in the definition of synthesis gas composition and contaminants present in it, that depends on the biomass and gasification processes adopted, than in the existence of gas cleaning technologies. These systems are already in commercial scale for the treatment of synthesis gas generated from coal, natural gas and petrochemical wastes, and can be adapted for gases generated from biomass. Hot gas cleanup technology is not in commercial scale yet but it can't be necessary, as in cases where wet gas cleaning systems can be adopted. The gas processing industries only will invest in the definition and construction of a specific biomass synthesis gas treatment system when they will have a definition of its composition and contaminants.

In the development of gas catalytic synthesis very little has been done in Brazil. Specific catalysts for the production of ethanol, as has been developed in NREL, and even in commercial scale units for the production of methanol, paraffin etc. must be investigated and developed, in order to build pilot-scale reactors and connect then to biomass gasifiers, enabling the development and performance evaluation of all of the BTL process, not only of isolated parts, as has been done until now.

METHANOL PRODUCTION POTENTIAL IN A STANDARD SUGAR AND ETHANOL MILL BY USING BTL TECHNOLOGY

Most of the Brazilian sugar and ethanol mills present very low energy efficiency. In Figure 3 it is showed the energy framework of a typical sugar and ethanol mill, processing 1,3 million of sugarcane t/a, with a specific steam consumption of 530 kg/t sugarcane, as described in HASSUANI *et al.* (2005).

The energy content of 1 t. sugarcane is equivalent to 0.16 t. of petroleum – TEP and from this mass of sugarcane it has been produced sugar, ethanol and a surplus of bagasse in the quantities indicated in Figure 3. The trash, in general, is being burnt or left in the field, without any improvement of its energy content. The surplus of sugarcane bagasse generated at the mill has been commercialized, mostly as fuel, with improvement of its energy content.

In this way, the energy improvement of this mill has been only 21% of the sugarcane energy content. After its energy optimization, with the installation of equipments with higher energy efficiency like ethanol distillation and sugar concentration units already existing on the market, the mill steam specific consumption can be reduced to only 340 kg/t. sugarcane, as cited in HASSUANI *et al.* (2005). The mill's energy balance, in this new condition, is presented in Figure 4.

In this new situation there is an elevation of the bagasse surplus and 50% of the trash is

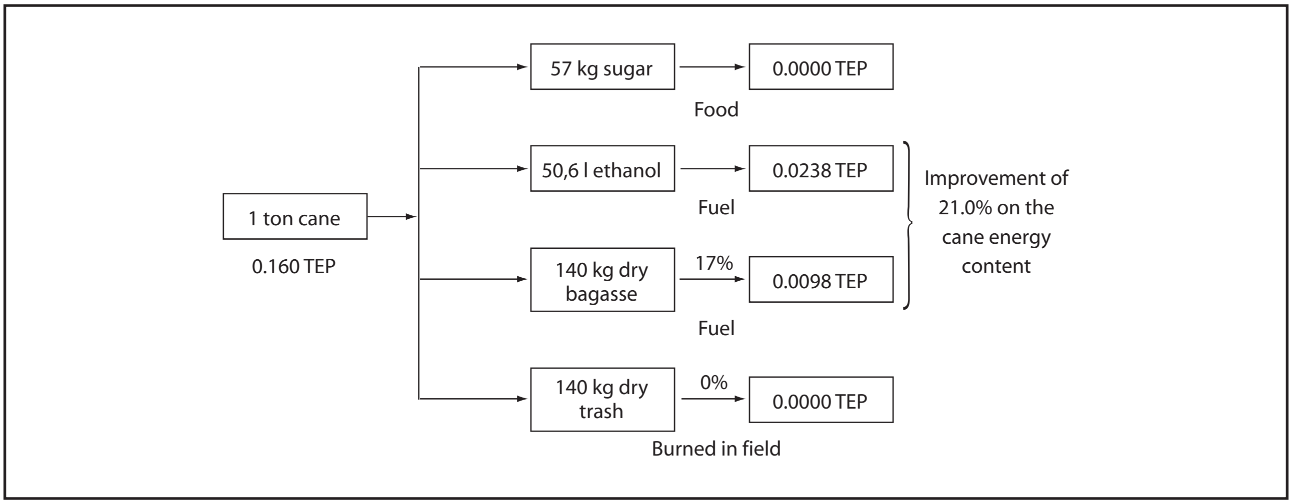


FIGURE 3 Energy improvement of a Brazilian standard sugar and ethanol mill.

brought from the field along with the sugarcane, and separated at the mill. The other half of trash is left in the field for soil protection and weeds control. In this frame, part of the organic material presented in the vinasse is converted to biogas for its energy improvement. After the mill's energy optimization, the potential energy improvement of the mill can reach 50.6%, without any change of sugar and ethanol production capacity.

With the installation of a BTL unit in this optimized mill, the bagasse surplus and trash brought from the field can be converted to methanol, as shown in Figure 5.

The conversion data of biomass to methanol presented in this Figure is based on gas composition and yields obtained on experimental biomass gasification tests performed at IPT (USHIMA, 2008) and on yields of demonstration catalytic reactors (HEYDORN, 2003).

From the gasification of the sugarcane bagasse surplus and the trash brought from the field, it can be produced 58.2 kg of methanol per ton of

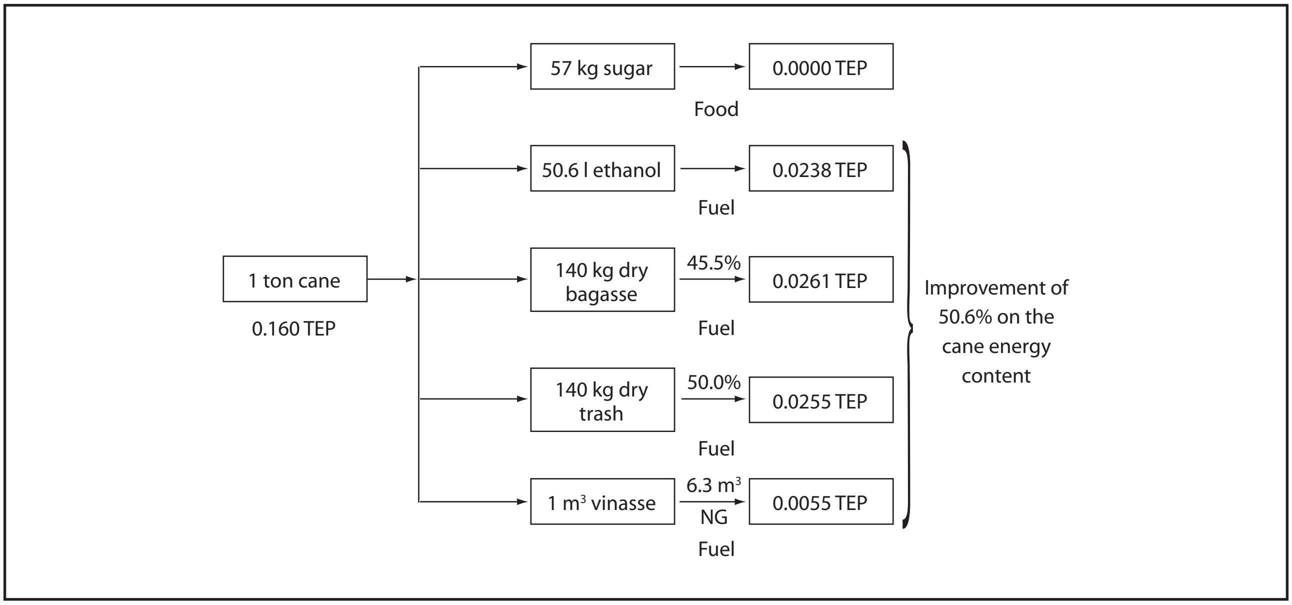


FIGURE 4 Energy profile of the standard mill after its energy optimization.

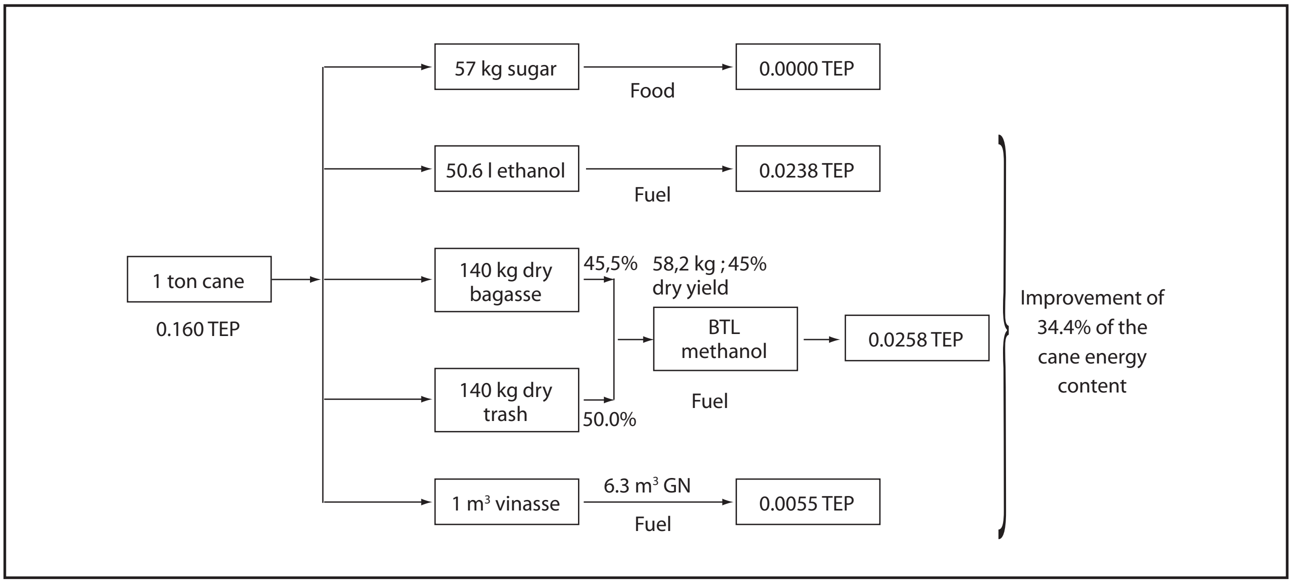


FIGURE 5 Mill's situation after energy optimization and BTL technology implantation for methanol production.

sugarcane, whose energy content is just a little bit more than of the ethanol produced from the sugar fermentation, i.e., it can be possible to double the net fuel production without the need to extend the planted area of sugarcane, only employing the biomass surplus currently wasted. In this evaluation, all energy inputs required for biomass treatment, gasification, gas cleaning, compression and synthesis were considered.

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