

The BTL2 Process of Biomass Utilization Entrained-Flow Gasification of Pyrolyzed Biomass Slurries

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Abstract

Forschungszentrum Karlsruhe has developed a concept for the utilization of cereal straw and other thin-walled biomass with high ash content. The concept consists of a regional step (drying, chopping, flash-pyrolysis, and mixing) and a central one (pressurized entrained-flow gasification, gas cleaning, synthesis of fuel, and production of byproducts). The purpose of the regional plant is to prepare the biomass by minimizing its volume and producing a stable and safe storage and transport form. In the central gasifier, the pyrolysis products are converted into syngas. The syngas is tar-free and can be used for Fischer-Tropsch synthesis after gas cleaning.

Index Entries: Straw; flash-pyrolysis; slurry; entrained-flow gasification; synthesis gas.

Preliminary Considerations Regarding the Potential

The use of biomass is one important element toward a sustainable energy production in the future. In a former study, it was estimated that biomass will be able to contribute about 20% of the global primary energy in the next century (1). Biomass is unique among the sustainable energy sources, because it is the only renewable carbon source, whereas energy from wind, sea, geothermal sources, and sun can only supply heat and electricity. Carbon-based liquids will probably remain the most important fuels in the mobility sector in this century. Biomass conversion into liquids therefore makes economic sense, even if combustion or cocombustion is cheaper in the short term. Regarding the German energy biomass market, residual wood from forestry and waste wood from industry make up 41% of all organic residues (ash- and water-free mass) (2), but the market of wood residues is tight at the same time and a considerable increase in woody waste is not expected. Nineteen percent of organic residues originate

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from household and garden wastes, 19% from sewage sludge and liquid manure (both ash- and water-free). These two classes of wastes can be used in garbage incineration, composting or biogas production, for example. Twenty-one percent of organic residues are thin-walled lignocellulose substances like straw and hay. When calculating this percentage, it is taken into account that only half of the straw may be considered waste, whereas other half is used for keeping animals and improving soil quality. The so far not utilized part of straw and hay in Germany corresponds to 6 Mtoe (250 PJ or 70 TWh) and is worth being considered an energy source for the future. Unfortunately, straw is difficult to manage: Its high content of alkalis, chlorine, and other inorganic compounds leads to problems with corrosion and sticking ash (4). Although combustion plants for straw have already been established in Denmark (3), it is not trivial to satisfy the strict emission guidelines in the European Unit. Research on direct gasification of straw is also known (4,5). Gasification and syngas use is a sophisticated technology which is only economically efficient when used on a large scale. The gasifier should reach a power in the order of >1 GW(th). Considering a 3 GW(th) gasifier with 8000 h of annual operation, it should be fed with biomass with an annual energy of 86.4 PJ (24 TWh or 2.06 Mtoe), this would be 6,000,000 t of air-dry straw. In Germany, an average of 720 t of wheat was harvested per km² in the year 2000 (6). The grain : straw ratio is variable and considered to be 1:1. Half of the straw is needed for animal keeping and soil improvement, the rest is available for conversion into energy. A 3 GW(th) gasifier can therefore be supplied with straw from an area of 17000 km² under cultivation of cereals. Of the total German area, 19.7% is used for cultivation of wheat, rye, barley, and oats all over the country (7). Areas with cereal fields alternate with forests, areas covered by buildings (towns and villages), areas of other agricultural use, and so on. This means that a gasifier may be supplied with straw from an area of 85,000 km² on an average and that the distance of transportation is up to 170 km, provided that the gasifier is situated in the center of this area. Leible et al. (8) estimated transport costs for straw in Germany. For a distance of 170 km, costs of 63 € (train) and 76 € (truck) per t straw result, this is 4.4 € (train) and 5.3 € (truck) per GJ heat content (lower heating value [LHV]). Because of the high oxygen content of biomass, only 41% of the heat content in the biomass can be converted into oxygen-free diesel or gasoline, the remaining heat content is lost by various reactions like the formation of CO₂ by various equilibrium reactions (e.g., water-gas shift reaction) and partial combustion. The transport costs of straw feed for 1 GJ of Fischer-Tropsch (FT)-diesel from biomass would therefore be 10.7 € (train) and 12.9 € (truck). The actual price of fossil fuels is about the same (tax-free approx 10 €/GJ in August 2005) (9). If the transport costs of straw are as high as the all-inclusive production costs for fossil fuels, FT-diesel from direct gasification of straw cannot compete with fossil fuel, regardless of conversion technology details.

Table 1
Properties of Straw (15% Water Content) and Its Pyrolysis Products

	LHV GJ/t	Density kg/m ³	Energy density GJ/m ³	Costs for transport 250 km by train
Straw	14.4	50–200	0.7–2.6	100 €/t
Pyrolysis liquids	7–22	1100	8–24	
Char	31	200–700	6–22	
Slurry with 30% solids	14–25	1300	17–33	14 €/t

Table 2
Product Yields of Flash Pyrolysis in a Twin-Screw
Reactor at 500°C and Atmospheric Pressure

Mass (%)	Ash	Moisture	Char	Gas	Condensates
Wood sawdust	1	5–10	14–18	~15	~70
Wheat straw chops	5	5–10	25–30	~20	50–55
Rice straw chops	~16	5–10	25–30	~20	50–55

Sand/Biomass Mixing Ratio = 6:1 to 20:1.

Why BTL2?

The abbreviation means “Biomass to Liquid in Two Steps,” a regional flash-pyrolysis and a central gasification and synthesis. The concept was developed to solve the transport problem explained earlier. The products of flash-pyrolysis are pyrolysis liquids, char, and gas. Tar and char can be mixed to pumpable slurry with a much higher energy density than a straw bale, which results in lower transportation costs per tonne. Although the distance between the pyrolysis plant and the central gasifier may be >170 km, the energy consumption by the transportation of slurry is negligible (<1 %). The data of the energy content, energy density, and transport costs are given in Table 1.

Flash-Pyrolysis

The input material has to be air-dry (15% water content or less) and thin-walled. In an inert gas atmosphere (1 bar or low pressure), the small chopped biomass particles are heated quickly up to 500°C in the reactor with a residence time of the gaseous molecules of up to 1 s and of the solids of 10 s or more. These reaction conditions are chosen to maximize the yield of liquid (*see* Table 2). Typically, the biomass is converted into 53–78% condensed liquid (tar or so-called “bio-oil”), 12–34% char (with high-carbon content), and 8–20% gas (mainly H₂, CO₂, CO, CH₄, and little C₂H_x).

For flash-pyrolysis, a process development unit (*see* Fig. 1) was built for a throughput of up to 15 kg/h with the twin-screw mixing reactor

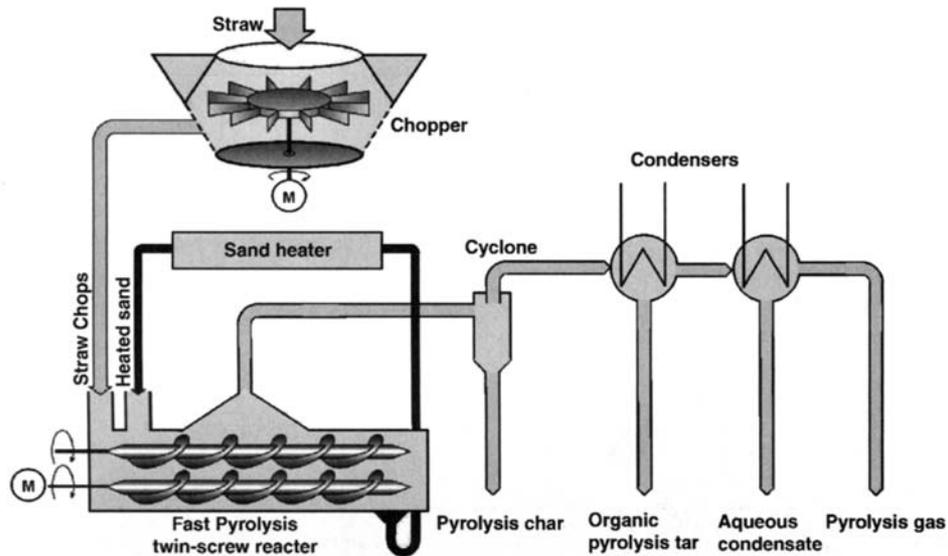


Fig. 1. Schematic representation of the process demonstration unit for flash pyrolysis at Forschungszentrum Karlsruhe.

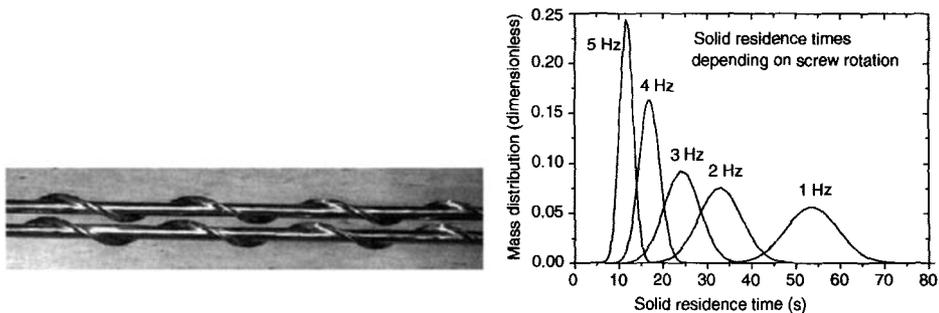


Fig. 2. Twin screws from the reactor, 1.5 m long (top). Solid residence times in the reactor depending on screw rotation (right).

developed by the LURGI company, Frankfurt (Germany) some decades ago. Although many different reactor types are conceivable, mainly two advantages lead to the decision to use this LURGI technology: (1) Feed and heat carrier are fluidized mechanically. Inert gas which dilutes the generated vapor is not needed. (2) Experience from the use of the twin-screw reactor in commercial-sized plants of oil refinery products was gathered over many decades (10,11), and can now be transferred easily to a similar reactor for biomass-pyrolysis. The screws of the process demonstration unit in Karlsruhe have a length of 1.5 m and an inner and outer diameter of 20 and 40 mm, respectively (see Fig. 2). They are intermeshing and self-cleaning. Hot sand (500°C) falls onto the straw and both are fluidized mechanically with an excellent heat transfer. The mass ratio of sand and straw was 20:1 in the first experiments and has been lowered to 6:1 so far.

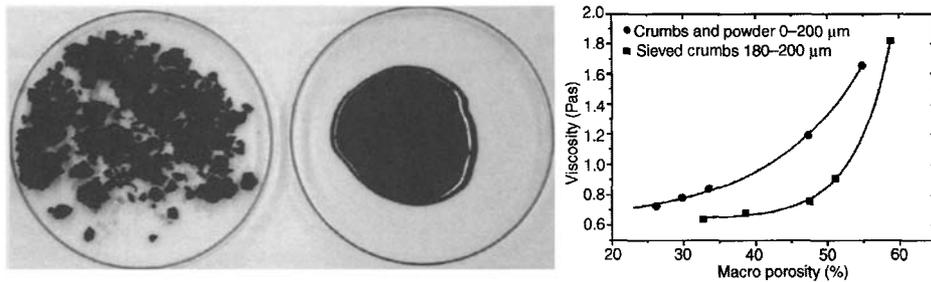


Fig. 3. Mixture of 22% straw char from flash pyrolysis and 78% pyrolysis tar. Left glass: Particle size 1–100 μm , directly from the cyclones of the reactor. Right glass: Same mixture after application of a mortar. Diagram: Viscosity of slurries with 20% char coal and different porosities, initial particle size up to 200 μm , two lines for two different char fractions. Porosity reduction via pressure.

Literature data regarding the energy of heat for flash-pyrolysis vary between 0.8 and 3.5 MJ/kg (12). The minimum mass of sand required for a sufficient heat supply has only been estimated roughly so far and needs to be investigated in future experiments. While the straw is pyrolyzed, the gaseous phase (mostly tar vapor) is sucked off by low pressure the coarse char is transported to the outlet at the end of the reactor and simultaneously premilled by abrasion. The fine-sized char particles leave the reactor with the vapors and are separated from the gaseous phase by two cyclones. The vapors are cooled in two condensers in which a highly organic tar is collected (about 10% water) as well as an aqueous solution of water-soluble molecules (about 70% water). The residual gas has a low-heating value only and consists of 45–65% CO_2 , 27–45% CO , 5–9% hydrocarbons, and <0.3% H_2 . In a commercial plant, this mixture of gases will be combusted for improving the energy balance. Its flammability is low. Consequently, it must be preheated first using low-temperature waste heat from the process. Downstream of the twin screws, the heat carrier is transported upwards by a bucket elevator and falls down again onto fresh straw passing a heat exchanger. A certain part of coarse char in the heat carrier can be accepted, because it will be milled during the following runs in the twin screws. No char accumulation is observed inside the heat carrier.

Slurry Preparation

Dry char is highly reactive and its handling is dangerous as far as powder explosions are concerned. For storage and transportation, char and tar are mixed to a highly viscous suspension (slurry) which has a much higher density than the biomass itself. A mixture of about 20% of straw char and 80% pyrolysis liquid at first represents a crumbly, thick, and wet bulk material (see Fig. 3). At surprisingly low-solid concentrations, flowability is lost. Because of its high porosity and wetting ability, the char absorbs the pyrolysis liquid like a sponge. This crumbly material has favorable safety properties and is suitable for long-term storage, but can neither be pumped through the

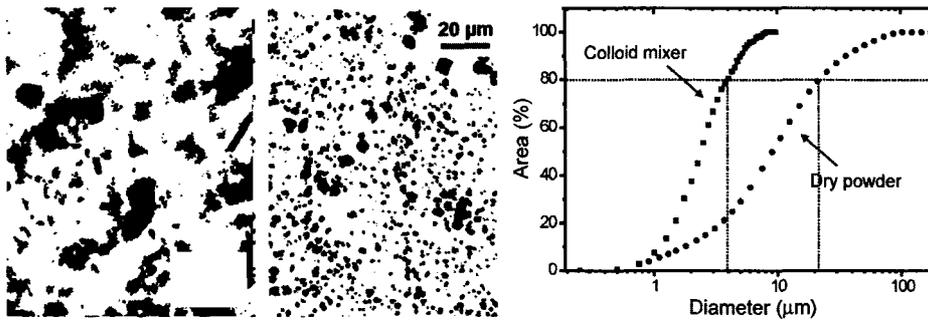


Fig. 4. Agglomerates of dry char particles (left) and single particles in the mixed slurry (center).

pipes of the gasifier nor can it be atomized which is necessary for complete carbon conversion at a low-reaction time. The crumbly substance becomes liquid by means of a colloid mixer (13). The porosity of char is very high because of its tendency of forming stable mechanical agglomerates (Fig. 4) which are destroyed by the high shear forces of colloid mixing. After forming single particles, the liquid absorbed by the agglomerates can contribute to the lubrication of the solid particles inside of the slurry, a highly viscous, pumpable slurry is formed (density approx 1300 kg/m^3). Particle size reduction caused by the milling processes has an additional large effect (Fig. 3), because not only the agglomerates, but also the single particles have a high porosity. Especially the macropores are destroyed by milling, because they act like breaking points. So far, 30 metric t of slurries have been mixed for the gasification experiments, which will be described in this next section.

Entrained-Flow Gasification

The gasification experiments are carried out in cooperation with Future Energy in Freiberg, Germany (14). The pilot-scale entrained flow facility of 3–5 MW(th) has a throughput of 350–500 kg/h and works under pressure (26 bar) (Fig. 5, center). It was designed as part of the Noell waste conversion process (15). An equivalent, but upscaled 130 MW(th) gasifier has been run successfully with various feedstocks at the Sekundaerverwertungszentrum (center of waste recycling) Schwarze Pumpe, Germany, for more than 15 yr (Fig. 5, right). The feed is atomized with technical oxygen and gasified with variable λ -values in order to achieve the desired temperature. High-ash content is needed, because the molten slag settles onto the cooling screen inside. A thin layer of slag near the cooling screen turns into a glass-like solid and protects the reactor from corrosion, whereas the main part of the slag flows down and is discharged via a lock. The sintering point of straw ash was measured to be 770°C , the flow point was 1320°C . In practice, flowability of slag is sufficient at 1250°C . Carbon conversion also depends on temperature and additionally on parameters like residence

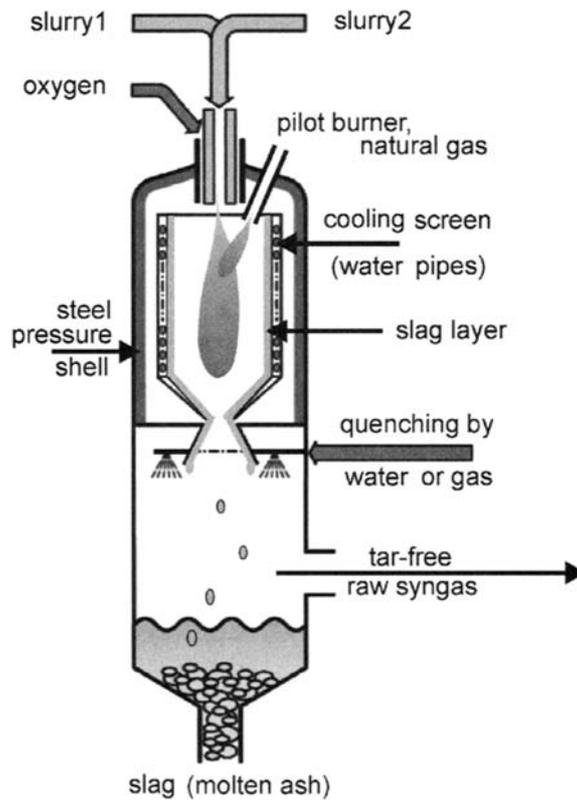
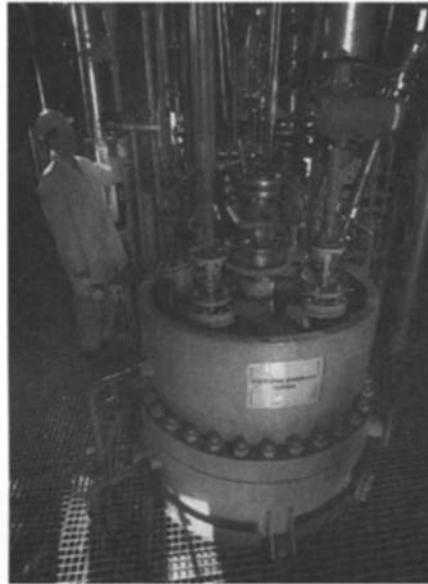


Fig. 5. Pressurized entrained flow gasifier: Diagram (bottom) of the reactor and photo (top) of the reactor head at FUTURE-ENERGY in Freiburg, Germany.

time (proportional to throughput⁻¹), atomization quality, energy content of the feed, and elemental composition. Carbon conversion rates of approx 99% (1250°C) and >99.5% (1400°C) were estimated roughly (residual carbon in the quench water). Differences caused by the char particle size were not observed. The x_{80} -value of particle size describes that all particles with an equivalent circle diameter smaller than x_{80} contribute 80% of the total volume. In these experiments particle sizes of $x_{80} = 6, 20, \text{ and } 50 \mu\text{m}$, respectively were used. The hot syngas is practically tar-free because of the high-gasification temperature. So far, we have not carried out our own experiments with gas cleaning (alkali metals, chlorine, and sulfur) and FT synthesis, but these processes are commercially available and state of the art (16–18).

Experience Gained From Gasification

Feedstocks used in the test runs in the 3–5 MW(th) gasifier were slurries of commercial pyrolysis products from Chemviron, Bodenfelde, Germany (19). Four different qualities of pyrolysis liquids were used and three chars with identical chemical compositions, but different particle size distributions. Viscosities were up to 2 Pa·s with solid contents of up to 39%. In the mixing unit, the slurry was heated up to 50°C and an even higher temperature (up to 80°C) before atomization with pressurized technical oxygen. By this, the viscosity of the slurry was reduced by the factor of 20–30 as compared with the initial viscosity at 20°C. There were no problems observed like the plugging of nozzles or sedimentation in pipes, which might be caused by slurry properties. So far, three experimental gasification campaigns have been performed, during which 20 separate stationary operating conditions were studied at various temperatures, solid concentrations, particle sizes of solid, slurry liquids, and residence times in the reactor. During the campaigns, about 30 t of different slurries were gasified. In the first experiments, a 0.6 MW(th) pilot burner (50 Nm³/h) with natural gas was kept for safety reasons. Later, the pilot flame was used during start-up and shut-down procedures only. Atomization was considered to be good when stationary states (gas composition, temperature) were achieved, which turned out to be impossible once only: After reducing the oxygen supply to measure a stationary state at a very low temperature, the O₂ pressure was too low for atomization of a highly viscous slurry. The composition of the dry syngas varied in the ranges of 13–31% CO₂, 21–32% H₂, 27–47% CO, 9–17% N₂, and 0–0.5% CH₄. Gasification efficiencies ranged between 50 and 71%. This will be improved drastically in a large >1 GW gasifier, because the heat loss via the radiation screen will be decreased as will the O₂ consumption. Furthermore, the inert gas flows (now 9–17% N₂) can also be reduced to a few percents. Tables 3–5 show input and output data of five stationary states, each of them maintained for 3 h. Because of the stoichiometry of CO and H₂ formation, the theoretically lowest λ -value for complete gasification of lignocellulose

Table 3
Elemental Composition of the Gasification Feedstock

%	H ₂ O	Ash	C	H	N	O
Pyrolysis liquid 1	6.4	–	58.6	6.2	0.3	28.4
Pyrolysis liquid 2	13.6	–	51.8	6.3	0.4	27.8
Pyrolysis liquid 3	49.7	–	24.3	3.8	0.3	21.9
Pyrolysis liquid 4	67.4	–	10	2.9	0.1	19.6
Char ^a	4.2	5.5 ^b	81.1	2.5	0.4	6.2

^aChar from commercial beech wood pyrolysis (straw char not available on a large scale).

^bAsh from beech wood char + additional straw ash.

Table 4
Stationary Reaction Conditions in the 3–5 MW(th) Entrained Flow Gasifier

No.	Char size x_{80} in μm	Solid [%] in slurry	Pyrolysis liquid	Oxygen λ	Feed in t/h	Temp. $^{\circ}\text{C}$
1	94	25	1	0.48	0.50	~1350
2	56	23	2	0.50	0.50	~1250
3	10	23	2	0.46	0.35	~1350
4	10	39	3	0.52	0.50	*
5	10	29	4	0.64	0.35	~1350

*=Temperature not measured.

Table 5
Gas Composition of Raw Syngas Produced in the 3–5 MW(th) Entrained Flow Gasifier at the Stationary States Characterized in Table 4

No.	CO	H ₂	CO ₂	N ₂ ^a	CH ₄
1	46.7	23.5	14.5	15.1	0.15
2	47.1	21.6	16.0	15.3	<0.1
3	47.0	30.2	13.1	9.6	<0.1
4	42.7	32.0	15.4	9.9	0.12
5	26.8	24.6	30.7	17.4	0.5

^aPurge gas for not used pipes.

biomass is 0.16 (Fig. 6). (λ is defined as the ratio between actual oxygen consumption and theoretical oxygen consumption for stoichiometric combustion). In reality, more oxygen than $\lambda = 0.16$ is needed, because the reaction temperature is achieved by partial combustion. The heat loss in a >3 GW gasifier is much lower than in the pilot-scale gasifier of 3 MW. Therefore, the λ -values measured on the pilot scale will be significantly higher than on the large scale. Furthermore, the water–gas shift reaction lowers CO and increases H₂ by forming CO₂. All in all, $\lambda = 0.32$ can be

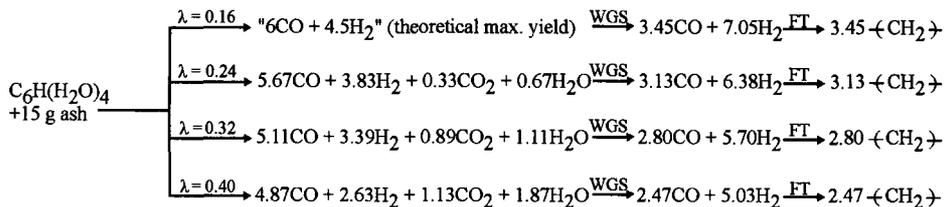


Fig. 6. The elemental composition of lignocellulose biomass can be described approx by $\text{C}_6\text{H}_9\text{O}_4 + 15 \text{ g}$ inorganics. When λ is decreased, the theoretical yield of the Fischer-Tropsch product increases. At a practically achievable λ -value of 0.32, the mass ratio between biomass input and synfuel product is 4.1:1. *Abbreviations:* WGS, water-gas shift reaction; FT, Fischer-Tropsch synthesis.

expected for a large-scale entrained-flow gasification of biomass. Raw syngas needs a thorough gas cleaning and little additional water which is still present after quenching the reaction heat by atomized water. Before FT synthesis, a H_2/CO ratio of approx 2.04 must be adjusted by means of the catalyzed water-gas-shift reaction. The FT product needs further refining like hydrocracking before it may be used as diesel fuel.

Efficiency and Economy of BTL2

The fuel yield of the total process can be calculated on the basis of Fig. 6. The elemental composition of biomass can be described empirically as " $\text{C}_6\text{H}_9\text{O}_4 + 15 \text{ g}$ inorganics + $18 \text{ g H}_2\text{O}$ " with a mass of 178 g which could theoretically be converted into 2.8 units CH_2 and 39.2 g FT fuel ($\lambda = 0.32$). Slurry production from biomass results in 15% mass loss (gas yield of flash-pyrolysis), carbon conversion of gasification is assumed to be 98%. FT yield of C_{5+} (hydrocarbons with five or more carbon atoms) in the once-through-then-out mode is 72% (20). If these realistic efficiencies are taken into account, 23.5 g fuel can be produced from the input of Fig. 6. In other words, for 1 t of synfuel, 7.5 t of biomass are needed. During entrained-flow gasification, about 15% of the energy content become high-temperature heat with $>1000^\circ\text{C}$ in the hot raw gas. By burning nonconverted residual syngas and nonmarketable secondary products, electricity can be obtained by gas turbines. In the case of 33% efficiency, 14.1 GJ(el) result/1 t fuel. On the other hand, electricity is consumed for generation and compression of O_2 , for the chopping, milling, and mixing of the feed and for numerous other activities. Therefore, only 4.2 GJ of electricity from the 14.1 GJ is left for sale. The principal dependent costs were simplified by assuming 25% of the unit original value per year. Costs of new installations were taken from literature (21,22), estimated for the desired unit size using a gradual decrease exponent of 0.7, and calculated with customary market rates and payout times. In Table 6, the substantial single contributions are given as production costs for 1 t FT product. Contributions for straw therefore refer to 7.5 t, for slurries to 6.4 t. So-called work overhead costs were

Table 6
Costs for the Production of 1 t Fischer-Tropsch-Diesel
Regarding the German Price Level 2005

Regional flash pyrolysis for 10 t/h slurry production		
Straw bale on field, air-dry (8% H ₂ O)	7.5 t × 45 €/t →	338 €
Straw transport with tractor	7.5 t × 25 €/t →	187 €
Slurry production	6.4 t × 47€/t →	302 €
Personnel (25/plant, 60 k€/ yr each person)	→	96 €
Central gasification and synthesis, 1 Mt Fischer-Tropsch product per year		
Slurry transport, 200 km by train	6.4 t × 12.2 €/t →	78 €
O ₂ - (1200 Nm ³ with a price of 6.25 c€/Nm ³ , without current)	→	75 €
Production of fuel	→	125 €
Personnel (300/plant, 60 k €/yr each person)	→	18 €
Sum of all costs		Σ 1219 €
Subtraction of credit for electricity (70 €)		1149 €

considered by 2% of fresh capital expenditures and 10% higher personnel expenditure. The production costs for 1 t fuel with the BTL2 process are 1149 €. This is 97 c€/L or 26.9 €/GJ, which is much too high to be competitive with production costs of fossil fuel (10 €/GJ). On the other hand, this cost estimation is calculated for production in Germany, and many different locations can be chosen. Hoogzaad, for example, investigated thoroughly the Ukrainian market situation and production costs (23). Comparing the cost estimation above with his results, it is appropriate to estimate a massive cost reduction for feed, transport, and personnel (90%) and for technical facilities (30%). Then, it would be possible to produce 1 t of diesel at 377 €, which is 32 c€/L or 8.8 €/GJ. This is a competitive price even in a country where biomass-based fuel is not promoted by tax reduction. BTL2 is an interesting option for the large-scale production of bio-synfuels. The last example shows that even now it can be an economic process in countries at the stage of economic take-off, if long-term contracts guarantee the large investments to be safe.

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