ALLOTHERMAL STEAM GASIFICATION OF ALTERNATIVE BIOMASS FUELS FOR HYDROGEN PRODUCTION

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ABSTRACT: Considering the rapid increase in the energy demand for power generation and industrial processes over the last years, the potential offered by biomass and solid wastes for solving some of the world's energy and environmental problems is widely recognized. This research work reports the results of the experimental investigation of alternative biomass crops regarding their applicability as fuels in an allothermal steam gasification process.

The examined biomass fuel, the Jatropha Curcas nut, grows in South America and is not commercially available yet. The ash melting behaviour as well as the proximate and the ultimate analysis of Jatropha Curcas are reported. The nuts are gasified in pelletized form, as well as in complete seeds, in an allothermal bubbling fluidized bed reactor, which uses the technology of high temperature heatpipes to transfer the required heat to the gasification zone, using steam as gasification medium. The experimental measurements show that the producer gas compositions for both forms of Jatropha (pellets and seeds) are similar compared to results obtained from gasification of wood pellets with even higher hydrogen content. This supports the fact that alternative biomass fuels could be successfully used for H_2-rich producer gas synthesis.

Keywords: allothermal gasification, hydrogen, heatpipes, Jatropha Curcas.

1 INTRODUCTION

The shortage of oil and natural gas as well as environmental problems due to CO_2 emissions, make it necessary to exploit new sources for energy production. Biomass is already contributing to the global energy supply, but without further conversion to a synthetic energy carrier, its application for electricity production is limited. Several conversion routes, which require an additional consuming energy process step, have been tested during the last years demonstrating the use of biomass fuels in a large number of applications for the production of energy and second generation biofuels.

The objective of this paper is to present a new promising biomass fuel (Jatropha Curcas) regarding its applicability in the gasification process for the production of biogenous gases.

2 BIOMASS APPLICATION FOR ENERGY AND HEAT PRODUCTION

2.1 Solid biomass

Biomass is traditionally used for combustion in residential heating systems all over the world. Biomass is burnt as combustion fuel providing heat at relative low temperature levels.

As far as electricity production out of solid biomass is concerned, it is usually realized in Combined Heat and Power plants (CHP), where the waste heat of the power generation is used for production of warm water or steam, increasing the overall process efficiency. Another possibility for gaining electrical energy from biomass is the retrofitting of existing coal-fired boilers to biomass co-fired ones, which has been considered to be a prospective commercial technology. Thereby, the high efficiencies of large coal fired power plants can be used to generate considerable amounts of electricity [1]. Nevertheless problems such as biomass milling, ash melting and corrosion, limit the share of biomass that can be co-fired with coal [2], [3].

2.2 Gasified biomass

Although biomass has to be converted in an additional process step in order to produce a gaseous mixture as fuel, more applications instead of using solid biomass directly as a fuel exist. This conversion step can be realized through biological processes like fermentation or by applying thermo-chemical processes e.g. gasification. Through the gasification process, biomass is converted into a gas consisting mainly of H_2, CO, CO_2, CH_4 and undesired components like aromatic hydrocarbons known as tar.

By using a gas mixture as fuel, gas turbines or gas engines can be utilized for the production of power, which offers the possibility for a Biomass Integrated Gasification Combined Cycle (BIGCC), where the waste heat of a gas-turbine is used in a steam-process for further power generation [3]. Gas engines can be used in block-type thermal power stations with variable size and power output for CHP production. The producer gas can be also co-fired with coal, but without the above mentioned problems [4].

In addition, researchers have reported the further downstream use of biomass gasification producer gas directly in high temperature fuel cells (Solid Oxide Fuel Cells) without prior gas cleaning steps [5], [6].

Concerning the applicability of biomass in the transportation sector, the producer gas (with an adequate H_2/CO ratio) can be further converted to liquid biofuels via Fischer Tropsch synthesis [7]. If therefore the hydrogen content in the gas should be enhanced to manipulate the H_2/CO ratio, this can be achieved with an in situ CO_2 absorbent like CaO [8] or by steam reforming of methane and water-gas shift in a downstream process step.

The most effective way of using the appropriate form of biomass can also depend on economical issues. According to Grahn et al. [9], biomass is most cost effectively used for the production of heat if there are low CO_2 taxes. At higher CO_2 taxes this changes and biomass is better utilized for the production of hydrogen or liquid fuels.

3 ALTERNATIVE BIOMASS FUELS

Biomass is intended to be one of the most promising renewable sources for supplying the world with sustainable energy. The potential of the energetic use of biomass is rather high, but since some fuels can be also used in other applications, the decision where to use renewable but nevertheless limited resources has to be made. Wood for instance, the most important fuel in biomass gasification process, is also used in other industrial sectors like construction or the paper industry. Another example of different possibilities to use biomass is the production of first generation biofuels out of biomass seeds that contain a high percentage of oil, which can lead to a competition between energy and food production.

An ideal biomass feedstock for the production of energy could therefore be a residual from other processes or biomass that has no significant use as food or material supply source.

An example concerning residuals is sewage sludge, which was investigated by Sánchez et al. [10] by pyrolysing it at different temperatures. Since sewage sludge is a potential risk for human health, the disposal problem could be also solved.

Yin et al. [11] reported the use of rice husk for gasification in China. Residuals from rice mills are thereby gasified in a circulating fluidized bed and the producer gas is used in gas engines to provide the required energy for the mills.

Boateng et al. [12] examined the gasification of straw in a small scale gasifier. The foreseeable application is the energetic use of straw in decentralized gasifiers located at farms.

In Greece, olive kernels are one of the basic and most important agricultural residues. Skoulou et al. [13] used olive kernels for the production of H_2-rich gas in a bubbling fluidized bed gasifier.

The above references demonstrate that hydrogen production via thermo-chemical conversion could be realized using a large number of different biomass fuels, which is a promising way to gain energy from sources that cannot be used for other applications.

4 BIOMASS GASIFICATION

4.1 Overview

In order to transform solid biomass into a combustible producer gas, a gasifying agent and the necessary heat for the endothermic gasification reactions have to be provided. The gasifying agent can thereby be air, pure oxygen, steam or CO_2. The main types of existing gasifiers can be classified into fixed bed, moving bed, fluidized bed and entrained flow reactors. The fact that fluidized beds can deal with various shapes and sizes of biomass particles is of high importance in the gasification process and the main advantage in using this type of reactors [3].

In principle, there are two ways to provide the required heat for the gasification process: autothermal gasification and allothermal gasification.

In an autothermal gasifier, the gasifying agent is either air or oxygen in an under-stoichiometric composition. A part of the biomass is oxidized and supplies the gasification reactions with energy, through the exothermic combustion reaction. The drawback is a dilution of the producer gas with nitrogen when air is used, resulting in a low heating value producer gas. When pure oxygen is used for autothermal gasification the dilution with nitrogen is avoided, but the oxygen has to be produced in an additional process step, resulting in more complex facilities and higher costs.

In case of allothermal gasification, the heat has to be provided from an external source. Consequently the problem of heat transfer into the gasification zone, which operates at high temperature level, arises. A simple approach is to use the hot flue gas from the combustion chamber, which results in large heat exchangers due to low heat transfer coefficients [14]. Another possibility is to circulate a solid heat carrier between combustion and gasification zone. This is possible when both combustion chamber and gasifier are designed as fluidized beds. The hot bed material is thereby used as heat carrier and circulates between both zones. Various possibilities exist for designing the circulation of the bed material between gasification zone and combustion chamber, an overview of which (dual fluidized bed gasifiers) is given by Corella et al. [15].

4.2 Gasification facility

The gasifier is designed as an allothermal bubbling fluidized bed, where the required heat for the endothermic gasification reactions is provided through heatpipes. There are two different configurations of the facility, one in laboratory scale (the heatpipes are heated electrically, 20 kW fuel input) and one full scale (the heat is transferred through heatpipes from an external combustion chamber, 100 kW fuel input).

For this experimental research the heatpipes are electrically heated. The gasifier is illustrated in the following Figure 1.

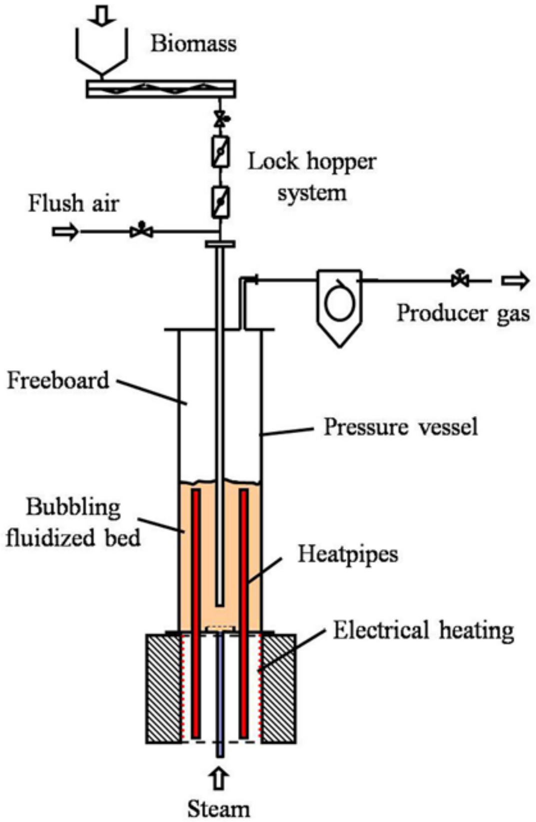

Figure 1: Scheme of the gasifier (laboratory scale).

The experimental facility consists of the following parts:

- gasifier and pressure vessel
- electrical heating
- high temperature heatpipes
- lock hopper system

The biomass is fed through a lock hopper system into the bubbling fluidized bed gasifier. The bed is fluidized with steam, which is used as gasification medium. The producer gas exits the reactor and after its way through a cyclone, it is driven to a filter for further purification. In the laboratory scale gasifier, the remaining chars leave the gasifier together with the producer gas and are separated in the cyclone. In the full scale application, the char is fed over a siphon system into the combustion chamber, where it is burnt to provide the required heat for the gasification reactions. A small amount of air is used to flush the biomass feeding system to avoid producer gas and steam to exit the reactor over the lock hopper system.

The heat is transferred via high temperature heatpipes, which work nearly isothermal over the entire length, into the gasification zone. The heatpipes have an evaporation zone (in the laboratory scale gasifier placed in the electrical heating, otherwise in the combustion chamber) and a condensation zone which is placed into the gasifier. The working fluid (sodium) is evaporated by heat input, flows to the gasification zone, where it condensates by heat emission providing the heat for the endothermic gasification reactions.

Since the char combustion zone and the gasification zone are not connected directly, the gasification zone can be pressurized up to 5 bar letting the combustion zone operate at atmospheric pressure.

The facility and its characteristics are described in detail by Metz [14].

5 INVESTIGATED FUEL

5.1 Jatropha Curcas

The nut has its origin in South America and today can be found in all temperate zones (30° N and 35° S). The seeds have an oil content of 32 %wt [16]. The leaves are toxic and the plant is traditionally used as a protecting hedge, for medical use or for soap production [17]. A competition between food and energy production is therefore excluded. The oil of the nut can be used for the production of biodiesel. Due to its favorable combustion characteristics, it is suitable as an alternative to regular diesel according to Sahoo et al. [18].

In this research work the applicability of Jatropha Curcas for gasification in an allothermal fluidized bed gasifier is investigated.

The Jatropha fuel is therefore investigated in two separate forms. In the first series of measurements the seeds of the nut are analyzed and gasified. Due to their adequate size they can be fed to the gasifier without further preparation. To investigate the whole nuts including the shell, they have to be milled and pressed to pellets. A picture of the seeds, the nuts and the pellets is displayed in Figure 2.

Figure 2: Jatropha nut, seeds and pellets

5.2 Preliminary fuel analysis

Prior to gasification the Jatropha nuts and their ash are analyzed. The values are compared to values of spruce pellets, one of the most widely used biomass fuel in the area of biomass gasification.

Table I: Ultimate analysis, %wt (db)

	spruce [19]	seeds	whole nut
C	51.9	56.6	46.4
H	6.10	7.58	5.79
N	0.3	2.67	4.11
S	-	0.17	1.9
O	40.9	28.41	26.38
HHV (MJ/kg)	20.1	23.6	18.6

The elemental analysis is shown in Table I on dry basis (db). The content of oxygen could not be measured directly (together with small amounts of chlorine the mass balance is closed to 100 %).

The elemental composition of Jatropha seeds shows a higher carbon, hydrogen, nitrogen and sulphur content in comparison to spruce. The whole Jatropha nut has a lower carbon and hydrogen content than spruce and a remarkable higher content of nitrogen and sulphur. It can be assumed that large amounts of nitrogen and sulphur can be found in the nut-shell. In general, gasification of Jatropha leads to high sulphur content in the producer gas, which is a drawback for possible downstream applications like high temperature fuel cells that have a limited SO_x tolerance.

The higher heating value (HHV) of the seeds and the whole nut is determined using a bomb calorimeter and is also presented in Table I on dry basis (db). The HHV of the seeds is higher than the value of spruce pellets due to the high oil content. The HHV of the whole nut is ~20% lower than the value of the seeds.

Table II: Proximate analysis, %wt (db)

	spruce [19]	seeds	whole nut
moisture	7.6	5.49	7.18
ash	1.5	4.57	15.4
volatiles	70.2	80.5	73.55
fixed carbon	28.3	14.93	11.05

In Table II the ash content, volatiles and fixed carbon are presented on dry basis (db). Both Jatropha seeds and the whole nuts have significantly higher ash contents compared to spruce pellets. The highest ash content can be found in the whole nuts, which explains their lower HHV compared to the seeds. Since the ash has to be removed in the gas cleaning step, this affects the downstream gas cleaning devices (cyclone and filter).

Table III: Ash fusion temperatures

	spruce	seeds	whole nut
°C			
IDT	1370	1183	1350
ST	n.o.	1197	n.o.
HT	1428	1238	1380
FT	1435	1314	n.o.

The ash melting behavior is analyzed according to DIN 51730 using a hot stage microscope. A cylindrical test specimen of ash, produced after CEN TC335/WG4 N43, is heated up gradually while continuously observing the shape via the microscope. With rising temperature one can identify four characteristic points: the initial deformation temperature (IDT), the softening temperature (ST), the hemispherical temperature (HT) and the flow temperature (FT).

In Table III the observed temperatures are listed and compared to measurements from spruce pellets. The IDT is considerably higher than the operating temperature in a biomass gasifier, therefore agglomeration was neither expected nor observed during the measurements.

The composition of the ash of Jatropha seeds and nuts is presented in Table IV.

Table IV: ash composition

component	seeds	whole nut
%wt		
SiO_2	7.87	8.89
Fe_2O_3	1.09	3.45
Al_2O_3	0.66	1.3
CaO	22.9	17
MgO	14.5	4.68
Na_2O	2.54	4.61
K_2O	27.1	27.8
SO_3	3.52	23
MnO	0.15	<0.10
BaO	<0.10	<0.10
TiO_2	0.12	0.2
P_2O_5	19.6	8.78

6 RESULTS AND DISCUSSION

6.1 Gas composition

In the main series of measurements, spruce pellets, Jatropha seeds and pelletized nuts are gasified in the laboratory scale facility**.** Olivine was used as bed material in the gasifier. Its properties are given in Table V.

Table V: Bed material properties

bed material	particle size	bulk density	porosity
Olivine	0.1-0.3 mm	1.45 kg/l	0.52

All measurements are performed under similar conditions in the gasifier (presented in Table VI).

Table VI: Gasification parameters

massflow (kg/h)	S/B ratio	Temperature (°C)	pressure
2	1	800	slight overpressure

The pressure during the experiments was between 1.1 and 1.3 bar absolute pressure. The temperature was approximately 800°C, which is far below the IDT temperature of the Jatropha seeds- and nuts-ash.

The main gas components are analyzed by means of gas chromatography (HP6890A) using a thermal conductivity detector. In the following figures (Figures 1-3), the gas composition of each experiment is shown after the gasifier has reached its steady state. A relatively stable gas composition over the measuring time (1-2 ½ hours) has been observed.

During the experiments, the Jatropha seeds showed favourable gasifying characteristics, while a steady state condition in the reactor was reached faster compared to the pelletized nut. The reason lies in the content of fines and dust in the nut pellets, which are gasified rapidly when fed to the reactor, resulting in pressure fluctuations in the beginning of the gasification process.

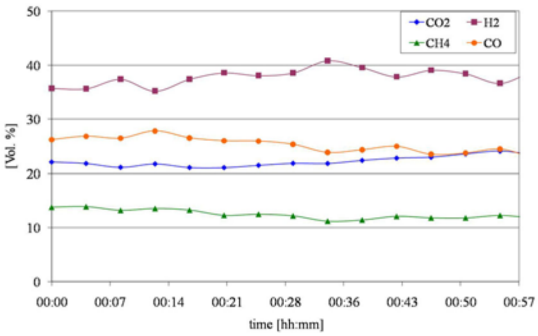

Figure 1: Gas composition spruce pellets

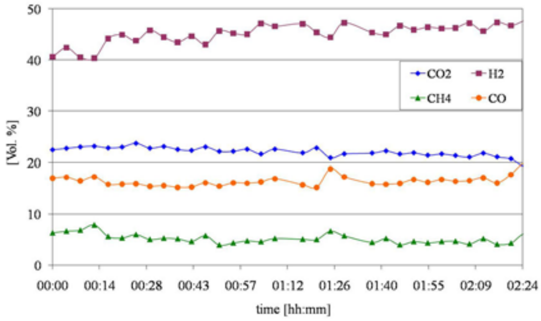

Figure 2: Gas composition Jatropha seeds

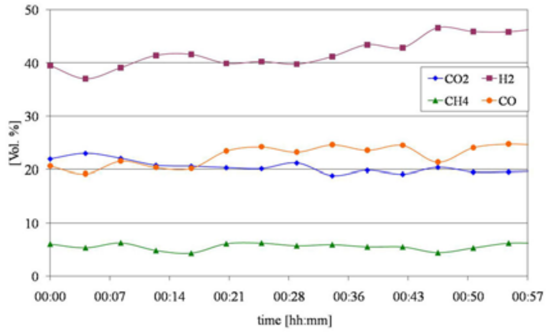

Figure 3: Gas composition Jatropha nut pellets

In Figure 4 the average gas composition of spruce, Jatropha seeds and Jatropha pellets are compared.

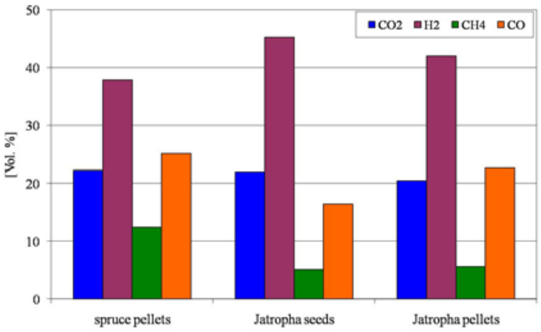

Figure 4: Comparison of gas composition

The most outstanding difference between gasified Jatropha and spruce can be found in the hydrogen and methane content. For both types of Jatropha, seeds and pelletized nuts, the hydrogen content in the producer gas is significantly higher than the value in the producer gas of spruce, whereas the highest value of hydrogen is found in the producer gas of the seeds. The methane content however is lower for both Jatropha fuels compared to spruce. A possible reason could be the high amount of oil in the Jatropha nut that is easily cracked to hydrogen and carbon monoxide.

Another difference in the gas compositions can be found in the content of carbon monoxide and carbon dioxide. For nut pellets the ratio between carbon dioxide and carbon monoxide is comparable to spruce, with more CO than CO_2, whereas in the producer gas of the seeds it is vice versa. The reason could lie in a greater amount of fine char particles in the gasifier bed. Due to the fluidized bed movement the char is partly milled to very fine particles, which occurs easier for the pellets (spruce or nut) than for the seeds. The fine char particles have a greater surface and the assumption that these particles are easier converted through the Boudouard reaction, which enhances carbon monoxide formation and carbon dioxide decomposition, can be made.

6.2 Gravimetric tar content

The gravimetric tar content in the producer gas is analysed by means of impinger sampling according to CEN/BT/TF 143 [20]. Tars are collected by solving them in isopropanol, while the volume of the tar containing producer gas is measured with the help of a gas flow meter. After evaporating the isopropanol, the mass of the remaining tars is determined and the gravimetric tar content (m_{tar}/V_{gas}) can be calculated.

A measurement is performed for the Jatropha seeds and the result is presented together with a value for spruce pellets [4] in Table VII.

Table VII: Gravimetric tar Jatropha seeds, spruce

Fuel	tar content g/ Nm^3
Jatropha seeds	24.1
spruce pellets [5]	3

The amount of tars in the producer gas of Jatropha seeds is eight times higher compared to values measured in the gas of gasified spruce pellets. Regarding the further use of the gas, this result is of significant importance since cooling will lead to tar condensation, which would affect tubes and downstream facilities.

Consequently, tars have to be removed in the gas cleaning step by either cracking or by scrubbing, when applications where the producer gas is cooled down (e.g. gas engines in block type thermal power stations) are foreseen.

Tar condensation can be also avoided when the gas is used directly at high temperatures. It is possible to inject the producer gas in the anode membrane of a Solid Oxide Fuel Cell (SOFC), which can tolerate tars in large amounts according to Saule et al. [5], or utilize it in a micro-gasturbine downstream to a pressurized gasifier.

7 CONCLUSIONS

In this research the applicability of the Jatropha Curcas nut for gasification in a bubbling fluidized bed gasifier is investigated. Jatropha seeds and pellets (produced out of the entire nut including the shell) are analyzed in two series of measurements.

In preliminary analysis the elemental and approximate analysis of the fuel, the HHV, the ash fusion temperatures and the ash composition are examined.

In the main experiments both forms of Jatropha (seeds and pellets) are gasified and the gas composition is compared to the gas composition of spruce pellets. The producer gas obtained from the gasification of Jatropha seeds and pelletized nuts has a higher value for hydrogen compared to the gas produced from the gasification of spruce pellets.

The Jatropha seeds show favourable gasifying characteristics during the experiments (e.g. stable gas composition is reached rapidly), whereas the fines and dust contained in the Jatropha pellets lead to prompt gasification and pressure fluctuations in the beginning of the gasification process.

The gravimetric tar content in the producer gas of the Jatropha seeds is measured and is significantly higher than values from measurements with spruce pellets. Therefore, either tar removal is required in the gas cleaning step prior to downstream applications, or the producer gas has to be used at temperatures where tar condensation does not occur.

As previously mentioned, the Jatropha nuts can be used for the production of biodiesel, which results in an oilseed press cake residue. The investigation of the applicability of the press cake for gasification could be interesting besides the gasification of the whole nut. This enhances the possibility to use the Jatropha Curcas nut more efficiently for energy production.

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