

# PRODUCT GAS MEASUREMENT IN FLUIDIZED BED STEAM GASIFICATION OF RICE HUSKS

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As the common agricultural feedstock, rice husks can be a sustainable biofuel option with significant calorific value (16-17 MJ/kg) and ash deforming temperature recorded above 1450°C. The research determines product gas on the 100 kW dual fluidized bed steam gasifier performance of rice husks pellets at temperatures between 760 and 810°C. Pure steam was used as a gasification agent at a ratio of 0.87 kg<sub>steam</sub>/kg<sub>fuel, db</sub>. Calcite with mainly CaCO<sub>3</sub> in compositions was used as bed material for the reactor. Significant H<sub>2</sub> content was determined with high quantity of CO<sub>2</sub> and CH<sub>4</sub> in product gas, while CO level was relatively low. Due to the considerable sulfur and nitrogen contents of this fuel, values of the impurities NH<sub>3</sub> and H<sub>2</sub>S in the producer gas was detected. It is also shown that the majority of sulfur was released in the gasification zone and, therefore, no further cleaning of the flue gas was necessary. Ethylene, ethane and propane were also formed but only in amounts below 2 vol.-% dry gas. Specific product gas yield reached 1.2 Nm<sup>3</sup><sub>db</sub>/kg<sub>fuel, db, af</sub>

Keywords: solid biofuel, rice husks, steam gasification, product gas

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## 1. Introduction

In the growing concern over global warming and limited supply of conventional energy sources, there is an increasing demand in the diversification of renewable energy options targeted to heat and electricity generation, gradually replacing fossil fuel. The most promising future energy source is biomass due to its widely availability and commodity [1]. Beside the wood, waste wood, and forest residues, agricultural residues, particularly rice husks and straws are important biomass source for energy production [2, 3]. In the world leading rice production countries, large amount of rice husks and straws have been main field based residues of rice cultivation for centuries. Recently, the use of rice husks and straws is highlighted as a potential non-woody solid biofuel [4, 5].

Previous studies [6, 7] have summarized that direct combustion for heat is the most common thermo-chemical conversion system for this solid biofuel, with low efficiency and inadequately controlled of gaseous emissions. Gasification enlarges the range of more effective applications, using the gases synthesized, mainly hydrogen, carbon monoxide, carbon dioxide, methane and nitrogen for combined heat and power generation in gas engine or gas turbine [8]. An appropriate gasification system particularly designed for rice husks provides an opportunity for electricity production and hence secure power supply [9]. In further, high calorific product gas, practically free of nitrogen, can be utilized for the Fischer-Tropsch synthesis of liquid biofuels and also for production of bio synthetic natural gas or other related gaseous products.

Several studies [10, 11] showed that conventional fluidized bed technology offers great advantages when used for gasifying low calorific value solid biofuel. The

investigation on the feasibility of gasifying rice husks using dual fluidized bed reactors and steam used as a gasification agent is essential. The study examines the suitability and potential of rice husks pellets in an existing 100kW pilot plant, which is capable and flexible to pelletized solid biofuel [12]. Since steam is used as the gasification medium, this gasifier produces a nearly nitrogen free, hydrogen rich produced gas.

## 2. Experimental setup

### 2.1. Dual fluidized bed steam gasifier

The fundamental principle of the novel dual fluidized bed gasification process is shown in Figure 1. This gasifier works on the basis of separating the endothermic gasification process from the exothermic combustion process. Gasification and combustion zones are separated by two fluidized bed reactors connected by loop seals.

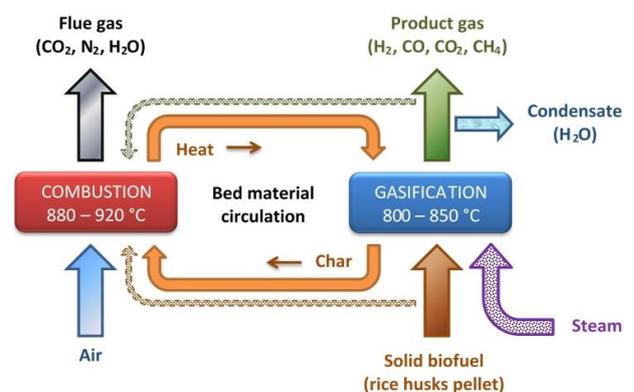


Figure 1. Schematic principle of the dual fluidized bed steam gasification process

The gasification zone placed in a bubbling fluidized bed reactor and the combustion zone placed in a transporting fluidized bed reactor. Both reactors were connected via two siphons, one connecting the lower part of the gasification zone with the lower part of the combustion zone, the other connecting the lower part of the separator in the combustor with the free board of the gasifier.

Hot bed material, circulating between these two reactors, carries the required heat for gasification from the combustion reactor, where residual char together with some other fuel for combustion, if required, was burned and heats up the bed material.

In a separator, the upwards-transported bed material was separated from the flue gas stream and led into the gasification zone, where the hot bed material was mixed with the fuel. The arrow inside the system indicates the global solids circulation rate of bed material.

At the top of the gasification zone the producer gas leaves the gasifier, while at the bottom the bed material together with the residual char was transported to the

combustion zone. Leakage of product gas from the gasification reactor to the combustion reactor, which would cause a loss of the valuable gas, can be effectively avoided by the loop seals. Also, any flow of air (the fluidization agent of the combustion reactor) or flue gas through the loop seals from the combustion reactor to the gasification reactor can be eliminated as it would cause a dilution of the product gas, mainly with nitrogen.

## 2.2. Measurement of the main product gas composition

A gas cooler is essential to cool the gas while stopping all gasification reactions within the gas stream. After travelling through the gas cooler, samples were taken at two points from the gas stream for online product gas measurement as well as for sampling of impurities (tars, ammonia, hydrogen sulfide, etc.). Figure 2 shows the actual pilot scale of sampling train with insulation-assembled components.



**Figure 2.** Actual pilot scale of online product gas measurement with insulation-assembled components

To avoid contamination and damage to the gas analyzer and the column of the online gas chromatograph, the product gas had to be cleaned in terms of particulate matter and condensable components like higher hydrocarbons (tars) and water, as both gas measurement devices require dry gas. To maintain this condition, the gas cleaning line was used. Particles like dust and char were removed by a glasswool-stuffed filter. After this particle removal the gas was led through six impinger bottles. Water and hydrocarbons that condense at temperatures higher than 4 °C were collected in the first two bottles. They were followed by three further bottles. Those were filled with rapeseed oil methyl ester where tars were

washed out of the gas. The last bottle ensures that no rapeseed oil methyl ester can leave the gas cleaning line accidentally and cause damage to the online gas analyzer or the online gas chromatograph.

The composition of the product and flue gases was measured after their exit from the reactors. As seen in Figure 3, main product gas components: H<sub>2</sub>, CO, CO<sub>2</sub>, CH<sub>4</sub>, were analyzed using a Rosemount NGA 2000. The components N<sub>2</sub>, C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> were measured with a gas chromatograph (PerkinElmer Clarus 500) every 15 minutes. The sum of main components with the impurities made up the difference to 100% by volume on dry basis.

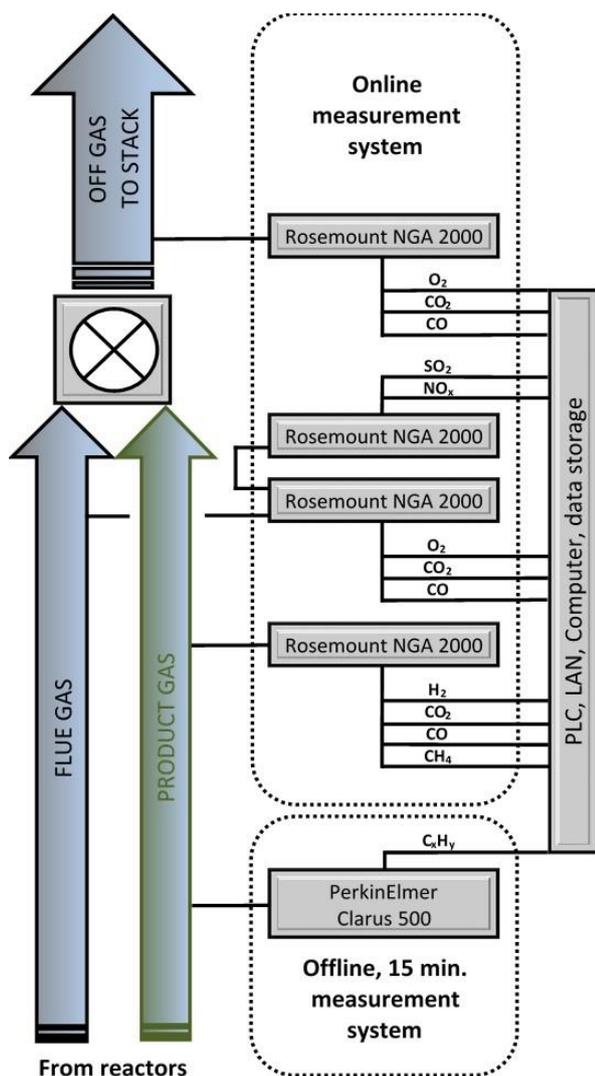


Figure 3. Online product gas measurement equipment

### 2.3. NH<sub>3</sub> and H<sub>2</sub>S measurement

For ammonia measurement, gas was sampled in a similar way to the sampling for the tar measurements, using impinger bottles. The solvent used in this procedure was diluted sulfuric acid at a temperature of about  $-2^{\circ}\text{C}$ . The impinger bottles were placed in a glycol/water mixture whose temperature was cooled down by a cryostatic temperature regulator. To avoid tar condensation in the pump, a bottle with toluene was added after the solvent for NH<sub>3</sub>.

After this procedure the concentration of ammonium ions in the sulfuric acid was detected by a photometric method according to DIN 38 406 Part 5 and ISO 7150. Hydrogen sulfide was sampled using impinger bottles filled with an aqueous potassium hydroxide solution at a temperature of about  $-2^{\circ}\text{C}$ . Subsequently, the H<sub>2</sub>S values were determined by potentiometer. It was not possible to measure NH<sub>3</sub> and H<sub>2</sub>S at the same time due to different solvents of the measurement procedure. The sampling equipment is shown in Figure 4.

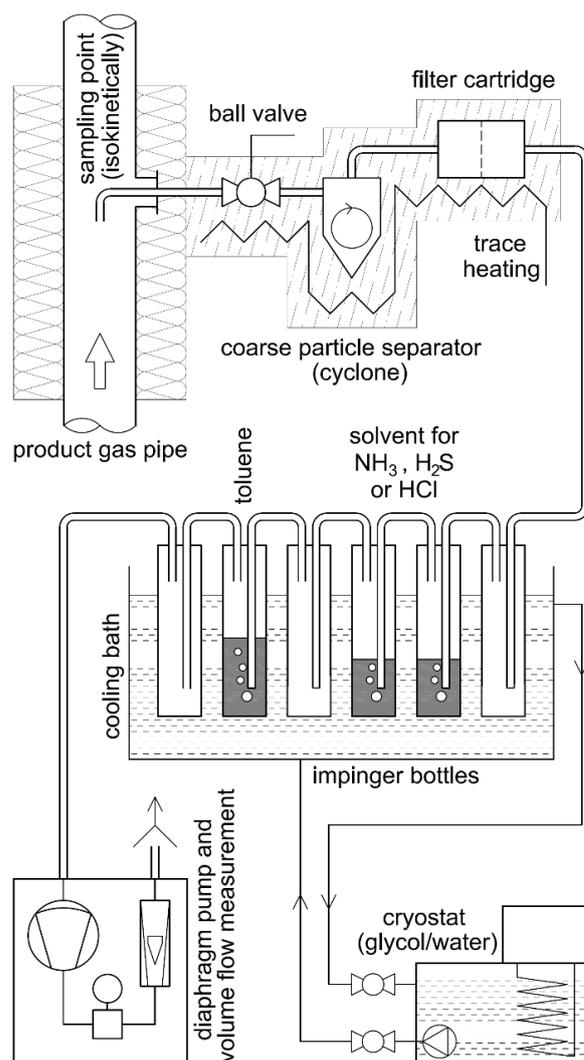


Figure 4. Schematic of NH<sub>3</sub> and H<sub>2</sub>S measurement

### 2.4. Bed material properties

Calcite (250–600  $\mu\text{m}$ ) and olivine sand (100–300  $\mu\text{m}$ ) were used as bed material for the fluidized bed reactors during the test runs of the novel pilot plant. In the dual fluidized bed concept, the circulating bed material served as a heat carrier to promote the endothermic gasification reactions. Previous studies indicated that olivine could be catalytically active in terms of tar degradation [13, 14]. Additionally, the bed material calcite was able to act as a CO<sub>2</sub> carrier.

The initial bed material mass typically 70kg contained mainly calcite (82 wt.%) and 18 wt.% olivine. Fresh charges (5kg) of bed material was regularly added during the runs.

### 2.5. Fuel characterization

Rice husks pellets were used as the fuel for the experimental investigations. The pellets were cylindrically shaped with a diameter of 8 mm and a mean particle length of 15–20 mm. The pellets were homogeneous in quality. Table 1 summarizes the important values of the investigated fuel.

**Table 1.** Fuel characteristics of the tested rice husks pellets

Parameter	Value	Unit
General		
Moisture	7.44	w%
Volatile	68.5	w% db
Residual char	31.6	w% db
Ash content	14.99	w% db
Higher heating value	16929	kJ/kg db
Lower heating value	15791	kJ/kg db
Elemental analysis		
Carbon (C)	43.16	w% db
Hydrogen (H)	5.18	w% db
Nitrogen (H)	0.47	w% db
Oxygen (O)	35.7	w% db
Sulphur (S)	0.06	w% db
Chlorine (Cl)	0.09	w% db
Ash melting behavior		
Deformation temperature	1450	°C

## 2.6. Steady states in operation

Electrical heating started at 06:00 and at 10:00 auxiliary fuel of heating oil was introduced into the combustion reactor to archive the required temperature in the plant. Fuel was introduced at 12:45 as auxiliary fuel into the gasification reactor to reach desired gasification temperatures (heat up - combustion modus). Steam was partially supplied to the gasification reactor at 13:30 and fluidization with pure steam started at 13:45 used as a gasi-

fication agent at a steam-to-fuel ratio of  $0.87 \text{ kg.kg}^{-1}$ . Operational gasification conditions for gasification test run were reached at 14:00. The first steady-state phase for the test run with over 100 kW fuel power was reached and  $\text{NH}_3$  in the product gas was measured. Directly next operation following by similar adjustments was reached and  $\text{H}_2\text{S}$  content in the product gas was measured. The last steady-state phase was reached at 18:10.

## 3. Results and discussion

### 3.1. Temperature

During the steady-state operation, mean temperature at the lower zone of gasification reactor was recorded  $762^\circ\text{C}$  at both GR1 (represented in red color) and GR6 (green line), as showed in Figure 5, indicating the uniform temperature distribution within the bed. This stability attested to good particle distribution within the bed. Meanwhile temperature reading at the upper part of gasification reactor was much higher at  $904^\circ\text{C}$  at the point GR15 (illustrated by purple spot on the reactor) as compared with the lower zone of reactor, and thus increased the rate of endothermic gasification reactions released energy and thus, raised temperature. Detailed temperature profiles were obtained at several measurement points distributed at different heights in the reactors, not only as a key control parameter of the process, but also on purpose of examining the influence of temperature on the gas composition.

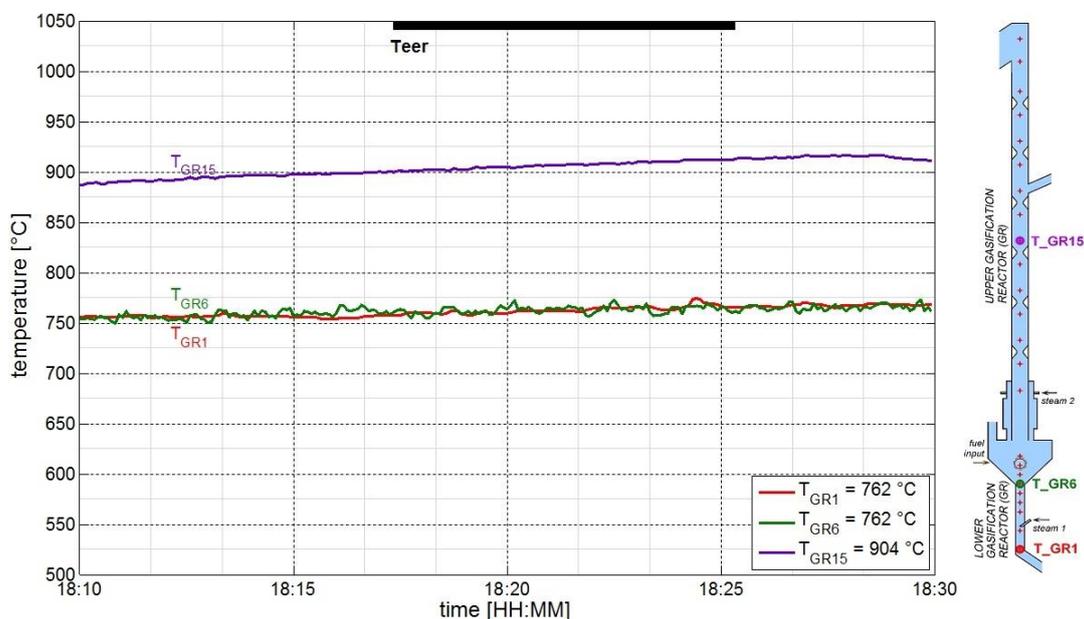
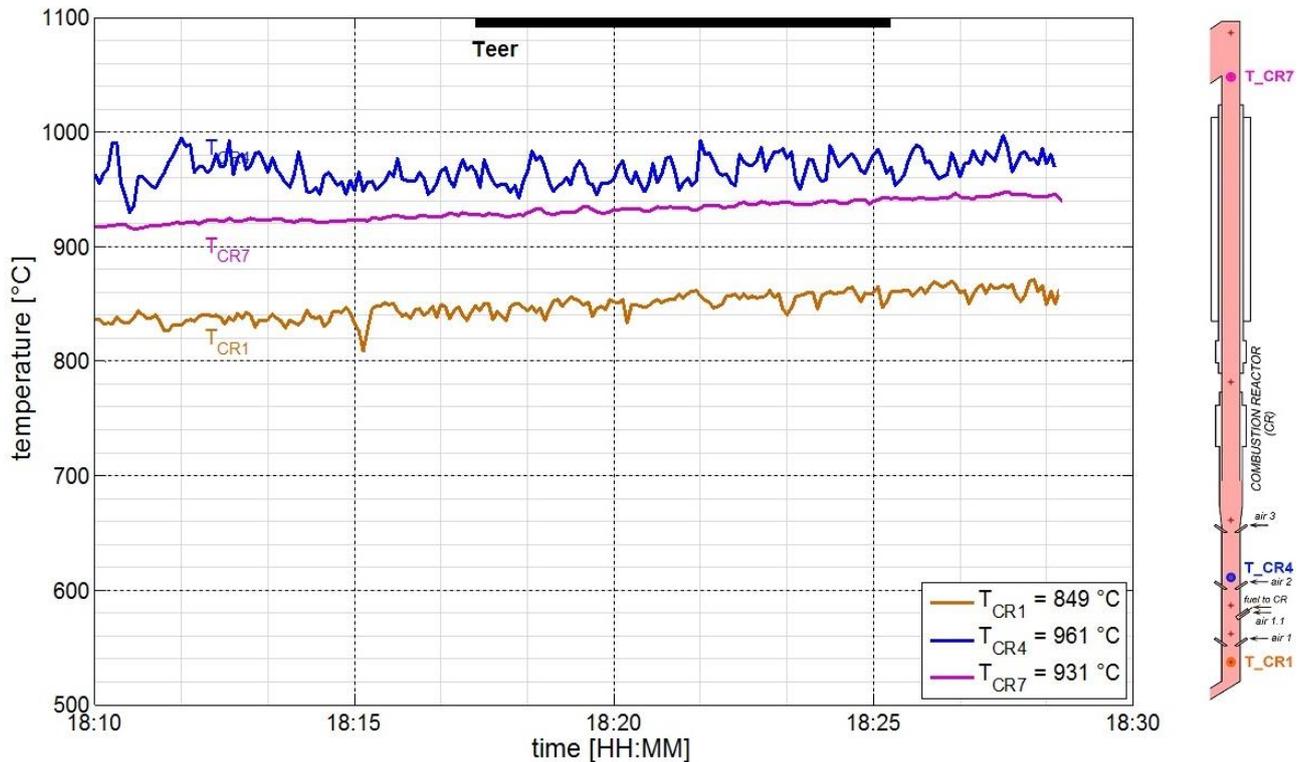
**Figure 5.** Overview on the temperature variation in different gasification zones

Figure 6 illustrates the mean temperature readings of combustion reactor. Obtained values at the lower zone reached  $849^\circ\text{C}$  and  $961^\circ\text{C}$  at CR1 and CR4 respectively are presented in orange and blue colors. The purple line shows mean temperature readings at  $931^\circ\text{C}$  at measuring

point CR7 on the upper zone of combustion reactor. Recorded values explained the fact that enhancing the exothermic carbon combustion reactions released heat, and thus gave rise to the combustion temperature.



**Figure 6.** Overview on the temperature variation in different combustion zones

### 3.2. Product gas composition

Two gas streams were obtained separately from the pilot dual fluidized bed gasification processes. The gasification reactor yields a product gas stream while the combustion reactor generates a conventional flue gas stream. Product gas stream primarily consists of hydrogen ( $H_2$ ), carbon dioxide ( $CO_2$ ), carbon monoxide ( $CO$ ), methane ( $CH_4$ ), ethylene ( $C_2H_4$ ), ethane ( $C_2H_6$ ), and unconverted water ( $H_2O$ ). Mean values of the main product gas components are summarized in Table 2 by volume on dry basis (vol.-%<sub>db</sub>). The sum of these main components with the impurities made up the difference to 100% of product gas composition. Nitrogen ( $N_2$ ) content of 0.93 vol.-%<sub>db</sub> was detected. This is typically presented in the product gas, as  $N_2$  was used for inertization of the fuel feeding system (purging/flushing feedstock hoppers), temperature & pressure measuring points with nitrogen.

Profiles of four major components in the product gas stream during a steady-state operation are shown in Figure 7. The results revealed that hydrogen (represented by the blue line) was in the highest amount (44.5 vol.-%<sub>db</sub>). The content of carbon dioxide was 24.3 vol.-%<sub>db</sub> as illustrated by the green line, followed by carbon monoxide (17.2 vol.-%<sub>db</sub>) and then methane at the reading value of 9.94 vol.-%<sub>db</sub>, as shown in red and purple lines, respectively. Those product gas components can be reasonably evaluated as quality product gas from gasification of non-woody solid biofuel.

The simulation work via the mass and energy balance software IPSEpro validated the reliable and representative

values of main product gas contents. Highest quantity of  $H_2$  was simulated of 43.1 vol.-%<sub>db</sub> followed by contents of  $CO_2$  (23.6 vol.-%<sub>db</sub>) and  $CO$  (18.0 vol.-%<sub>db</sub>), then  $CH_4$  content (11.0 vol.-%<sub>db</sub>), with errors to a minimum.

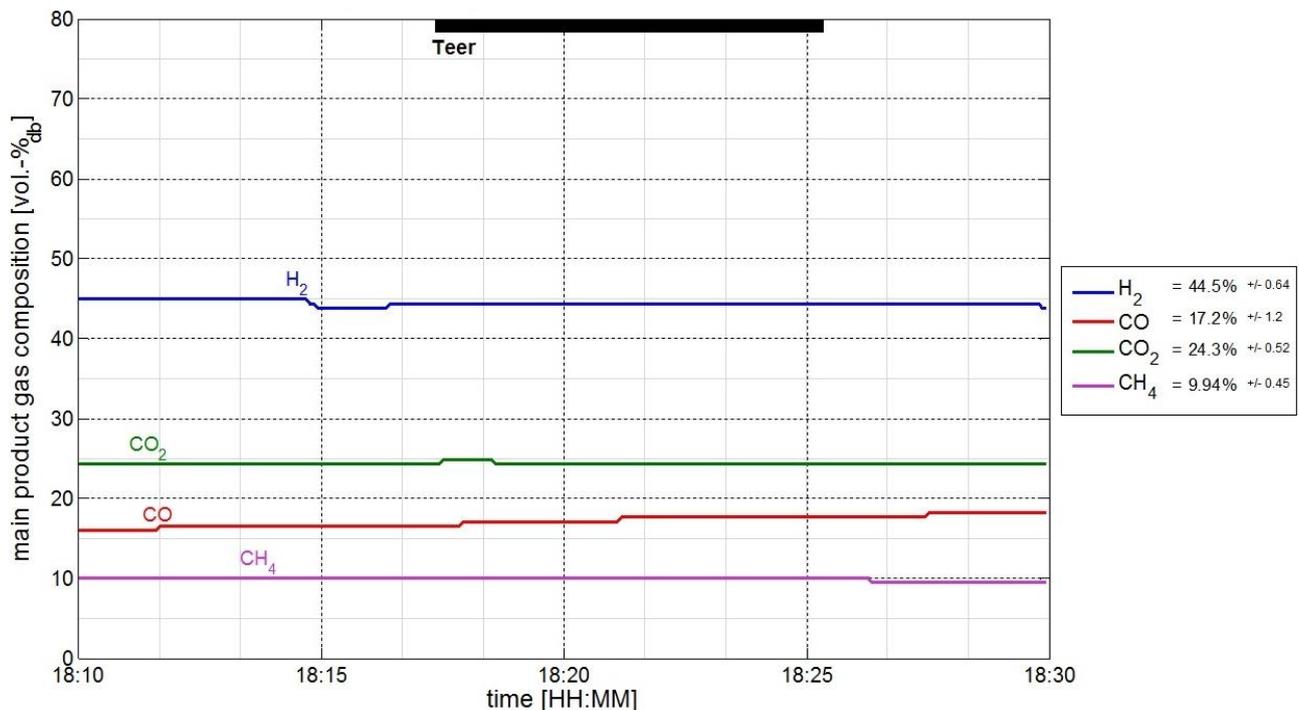
Gaseous  $C_2 - C_5$  hydrocarbons were detected by the measurement devices but only in quantities below 2 vol.-%<sub>db</sub> dry gas. Ethene ( $C_2H_4$ ) was stably presented by 2% vol.-%<sub>db</sub> while ethane ( $C_2H_6$ ) was roughly introduced at an amount of 0.48 vol.-%<sub>db</sub>, whereas propane ( $C_3H_8$ ) and higher  $C_4, C_5$  hydrocarbons contents show no significant value. In practice, as they are not easy to separate, all hydrocarbon above propane ought to be absorbed through rapeseed oil methyl ester in the impinger bottles.

The obtained values of impurities, particularly ammonia ( $NH_3$ ) was 7611 ppm<sub>db</sub> and hydrogen sulfide ( $H_2S$ ) was 462 ppm<sub>db</sub> in the product gas stream were correspondently derived from the nitrogen and sulfur elemental compositions of rice husks as a solid biofuel. Mostly introduced nitrogen was released in the gasification reactor mainly as ammonia, while the hydrogen sulfide value strongly depends on the sulfur content introduced in the fuel composition.

The rest of sulfur formed as sulfur dioxide ( $SO_2$ ) in the combustion reactor or captured in the ash. Due to the limitation of different used solvents for discontinuous measurements, values obtained in different time during the steady-state operation.

**Table 2.** Main gaseous components in the product gas stream

Parameter	Unit	NGA 2000	PE Clarus 500	IPSEpro
Product gas composition				
Hydrogen (H <sub>2</sub> )	vol.-%, db	44.5	-	43.1
Carbon monoxide (CO)	vol.-%, db	17.2	18.1	18.0
Carbon dioxide (CO <sub>2</sub> )	vol.-%, db	24.3	23.6	23.6
Methane (CH <sub>4</sub> )	vol.-%, db	9.94	10.4	11.0
Ethylene (C <sub>2</sub> H <sub>4</sub> )	vol.-%, db	-	1.96	2.00
Ethane (C <sub>2</sub> H <sub>6</sub> )	vol.-%, db	-	0.48	0.50
Propane (C <sub>3</sub> H <sub>8</sub> )	vol.-%, db	-	0.00	0.00
Minor gaseous components		Typical value	PE Clarus 500	
Nitrogen (N <sub>2</sub> )	vol.-%, db	0.5-2.0	0.93	
Sum of C <sub>4</sub> , C <sub>5</sub> hydrocarbons	vol.-%, db	1.0-4.0	-	
Additional components		Value		
Ammonia (NH <sub>3</sub> )	ppm,db	7611		
Hydrogen sulphide (H <sub>2</sub> S)	ppm <sub>db</sub>	462		

**Figure 7.** Major components in product gas stream during the steady-state operation

#### 4. Conclusion

The experiment on the 100kW pilot plant operated with a dual fluidized bed steam gasifier successfully confirmed the suitability of rice husks pellets. The study has shown that allothermal gasifying process is capable for the conversion of this carbonaceous solid biofuel into high quality product gas. Although values of the impurities ammonia and hydrogen sulfide in the producer gas was detected due to the sulfur and nitrogen contents of this fuel, rice husks gasification provide an opportunity

for the flexible utilization of non-woody biomass sources.

In practice, local availability of rice husks and dependence on the harvesting episodes are common issues. The results enable us to exercise various tests and research on co-gasification of rice husks with conventional fossil fuel and woody biofuel, since those are still significant parts of future energy. If so, the tar contents and levels of impurities such as NH<sub>3</sub> and H<sub>2</sub>S is the key issue influencing the product gas composition.

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