Gas yield variation in wood biomass co-current air gasification process – continuous operation

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Abstract. The purpose of this work was to study the gas yield variation resulted from the cherry wood gasification with air using a lab-scale rotary kiln gasifier. The feedstock was continuously fed into the preheated reactor at 600°C, in co-current configuration, using atmospheric air as a gasifying agent. The results indicate the importance of oxidation reaction control, through the feeding flow rates of biomass and air and the reactants mixing rate. From the experiment, the hydrogen yields were about 2-4%, while the carbon monoxide varied between 8-21%. Additionally, the paper provides process observations based on the continuous monitoring of gas composition. The specific flow rates of substances and installation operating conditions were linked to process run through syngas composition.

1 Introduction

Recent reports and former scientific studies, including the authors, have outlined the advantages and challenges of bioenergy usage: i) biomass is an important energy source but its structure could cause a chain of issues mainly associate at their phase change behavior affecting the overall process energy efficiency [1]; ii) globally, the bionergy usage could decrease the greenhouse gases with 65% by 2050 [2], iii) in 2017, bioenergy accounted for roughly 70% of the renewable energy consumption, worldwide [3]. Among the existing biomass thermochemical conversion technologies, gasification provides gas fuel (syngas) that can be used in furnaces, power machines and electrochemical fuel cells, for both heat and power generation [4]. The studies conducted on biomass waste gasification have highlighted the following: i) the syngas quality and overall process efficiency is influenced mainly by biomass composition and its particle size, gasifying agents, operating conditions, type of gasifier and their link with each other[5]; ii) its usage in a direct electrical heating system could offer low emissions [6]; iii) systems with capacities lower than 200 kWe are more preferable due to local direct usage, while higher than 2 MW are more favorably due to investment efficiency, although the feedstock collection implies high costs [7].

Previous studies on biomass gasification have used agricultural straws, crop residues, woody forestry and waste resulted from both mention activities or municipal solid stream

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[8,9]. Most of the experimental studies have been carried out on biomass air/oxygen gasification, at high temperatures typically ranging between of $800-1000^{\circ}$ C [10], more specifically agricultural waste 750-850 °C and woody biomass 850-950 °C [11].

According to Ren et al., 2019 review [5], the syngas heating values resulted from air biomass gasification ranges between 4 and 7 MJ/Nm³. Before its usage in internal combustion engines, a cleaning-cooling system is necessary, due to the moisture, tar, dust and temperature of the syngas. Hydrocarbons compounds with a molecular weight above benzene (tars) could appear at low temperatures, typically between $200^{\circ}C-500^{\circ}C[10]$, reaching an average level of 1 and 50 g/Nm³ [12]. Higher gasification operating temperatures ($\geq 800^{\circ}C$) is recommended for tar reduction, although, this can favor the formation of slag from ash agglomeration. [13]. The exit gas temperature from the gasifier can vary between $200-700^{\circ}C$ [10], while the ideal gas temperature for its usage in internal combustion engines should be around room temperature [14]. When operating on syngas as a substitute for petroleum fuels, the internal combustion engines are derated from about 15% to 40% (15% to 20% dual fuel diesel, 30% to 40% spark ignition)[15].

Studies have shown that perennial crops, such as pruned fruits and woody fruits, including cherry [14] could be explored as primary feedstock in the thermochemical conversion schemes. Same remark has been done for the Prunus avium tree family, commonly called sweet cherry, wild cherry and gean, due to its woody based structure [15] including also their stones [16] and their seeds [17, 18]. Apart from their role in the food supply chain, the Prunus avium trees could reach to their end life, becoming an agroforestry residue. Another cherry wood residue source is also the furniture manufacturing sector. In the current study the effects of cherry wood components on air gasification was investigated using a lab-scale rotary kiln gasifier at 600°C. The study findings are related to process set-up and installation operation influence on gasification reactions and products formation.

2 Material and Methods

The cherry wood waste originated from Prahova county, Romania. The schematic diagram of the experimental set-up is presented in the following figure. First, the feedstock was reduced in size by using a Fritsch cutting mill (Pulverisette 15). This step facilitated the feedstock waste characterization procedures, gasifier feeding system and degradation reactions. The primary analysis was carried out conforming to ASTM standard (E870 – 82). The high heating value (HHV) of the feedstock was determined using IKA C 200 calorimeter.



Fig. 1. Air gasification of cherry wood chips experimental set-up (adapted after [21]: 1. Rotary kiln reactor; 2. Reactor inclination degree; 3. Auger feeding system; 4. Air supply; 5. Syngas condensing system; 6. Syngas analysis (Testo 350XL). 7. Syngas sampling syringe; 8. Syngas analysis (Micro-GC Fusion gas analyser). 9. Ash.

The air gasification of cherry wood chips experiments were conducted in a rotary kiln gasifier lab-scale pilot plant. The rotary kiln gasifier (1) main characteristics have been presented previously by the authors [21]. In sum, the lab-scale pilot reactor allows the variation of the feedstock feeding rate and reactor rotation speed, its inclination degree and temperature. In this work, the cherry wood waste chips were continuously fed into the reactor (2), in co-current configuration, using atmospheric air as gasifying agent (3). The waste feeding rate was determined based on preliminary trials made on the hopper screw conveyor, considering also the reactor rotational speed and its 10°inclination (4). To assure the partial oxidation of the feedstock the Equivalent Ratio (ER) was calculated:

$$ER = \frac{Air introduced in the process}{Stoichiometric air}$$
(1) [22]

The calculations were done based on the elemental composition of the feedstock used for the air required for theoretically complete combustion (stoichiometric) determination. The cherry wood residues elemental composition expressed in percentage [%], used in the aforementioned determination was : C-49.52, H-5.81, O-42.97, N-0.31, S-0.02, Cl-0.02, Ash-1,35, Moisture-0 [23]. Cheery wood ultimate analysis is similar to other types of woody biomass, according to a study based on 95 different literature sources, regarding the ultimate analysis of woody biomass. According to this study woody biomass contains 41.78 - 57.00% Carbon, 4.85 - 8.11% Hydrogen, 35.85 - 52.26% Oxygen, 0.04 - 1.43% Nitrogen, 33ppm - 0.2% Sulphur, and 0.0004 - 0.08% Chlorine [24]. According to the formula presented above, the amount of air required for a complete combustion (stoichiometric condition) of cherry wood residues needs to be determined. To determined it, all combustible components of the material are considered. The gasification process temperature was set at 600°C. The syngas was passed through an ice water cooling system (5), aiming to retain the condensable fraction that could be formed during the process. The composition of the non-condensable fraction has been analyzed using two modalities presented in Figure 1., marked with 7-9. The Testo-350XL gas analyzer allowed real-time measurements, offering immediate information on CO, CO₂, O₂, H₂, SO₂, NO and NO₂ concentrations, hence, gasification process state and stability. Additionally, at fixed time intervals, syngas samples were extracted after gas cooling using a syringe (7) and injected to micro-gas chromatograph (Micro-GC Fusion, Inficon (8) immediately. Due to its features, the instrument is suitable for syngas fast and accurate composition analysis. Hence, the instrument allowed the detection of H₂, O₂, N₂, CH₄, CO, CO₂, and light hydrocarbons (C_1 to C_{12}).

Six gasification experiments were conducted at different reactor mechanic parameters and ERs in order to determine the optimal reactor settings for air cherry wood residues gasification. The process parameters modified were the reactor rotation speed, the biomass and air feeding rate (Table 1). Process duration may be controlled in this type of installation by reactor rotation rate and its inclination Since the most important gasification reactions are also the slowest, there must be ensured enough process time for all char oxidation reaction to take place [25]. As it is stated above, the gas composition was determined using two gas analyzers. Testo 350XL gas analyzer has the advantage of real time measurements, while the Micro-GC has the advantage of accurate measurements. The use of Testo 350XL provides real time information allowing users to observe and quantify the experiment results during process run. Gas-chromatography provides more accurate results and can determine far more gaseous species.

No. experiment	Gasifying agent (air) mass flow	Feedstock mass flow	ER	
	[kg/s]	[kg/s]	[-]	
1	0.00156	0.00036	0.74	
2	0.00104	0.00033	0.54	
3	0.00104	0.00033	0.54	
4	0.00031	0.00021	0.25	
5	0.00104	0.00031	0.57	
6	0.00021	0.00020	0.18	

Table 1. Rotary kiln reactor operating parameters for cherry wood residues air gasification

In the first experiment conducted, only the Testo gas analyzer was used, while for the experiments 2 to 6, both Testo and Micro-GC were used to determine the gas composition. During the second experiment, Testo measurements were interrupted to collect the gas samples for gas-chromatography analysis. For experiments 3 to 6 gas analysis by Testo and Micro-GC was conducted concomitant. To characterize the gasification experiments, oxygen, carbon dioxide, carbon monoxide and hydrogen concentrations in the produced gas were analyzed. Sulphur and nitrogen oxides registered very small concentration. Moreover, these gas species concentrations are very low and don't modify the process energy balance.

Since Testo350XL is a gas analyzer designed for combustion processes its results may not be perfectly accurate for gasification processes. Its use in gasification processes helps to characterize in real time the process run, and to determine if the gasification reactions occur.

3 Results and discussion

The physicochemical properties of cherry wood residues are presented in Table 2. The volatile matter (VM), fixed carbon (FC), ash content and HHV as received (a.r) values are closely with the ones reported by Telmo et.al., 2010 [26] on 84.9%, 15%, 0.1% and 17500 kJ/kg.

Property	Moisture	VM _{a.r.}	FC* _{a.r.}	Ash _{a.r.}	HHV _{a.r.}	HHV _{d.b.}
	[%]	[%]	[%]	[%]	[kJ/kg]	[kJ/kg]
Average	0.32	81.95	15.74	1.98	17323	18498

Table 2. Physicochemical properties of cherry wood residues average values

a.r.=as received

d.b.=dry basis

*=calculated by difference

Depending on process parameters the syngas flow rate varied between 0.00038 and 0.001887 kg/s. That corresponds to solid-gas conversion rate of about 93.31% - 98.23% [27]. Continuous operated systems provide process observation advanced over batched operated systems. The permanent monitoring of gas composition offers information about process status throughout the entire experiment. So, both specific flow rates of substances and installation operating conditions will be reflected in syngas composition. In the graphs

below is presented the variation of principal gas species concentration in time for the experiments conducted.



Fig. 2. Gas species concentration in time - experiment no. 1

According to first experiment results presented in Fig. 2, char gasification stage didn't occur, leading to an incomplete gasification process. High levels of CO_2 and the absence of H_2 and CO in the produced gas can be the result of short process duration. Both biomass feeding and the reactor rotation speed were set at high values, therefore the feedstock was discharged from the reactor at a faster rate than the char gasification minimum reaction time. Also, high O_2 concentration indicates an insufficient reactants residence time inside the reactor.

At lower gases flow rate (increase reaction time) both CO_2 and CO are formed (second experiment, Fig. 3). According to Testo measurements CO_2 maximum concentration increased with almost 8 percentage points, while the CO maximum concentration increased with more than 10 percentage points, when the air flow rate decreased. At lower air flow rates there is also a lower Nitrogen quota in the syngas. That will increase all other gas species yield in the syngas, including CO_2 and CO. Small H₂ concentration is also registered. Even though the H₂ concentration is low, comparing to usual biomass air gasification processes (6 – 22% according to Heidenreich et al. [28]), it is higher compared to the first experiment conducted.



Fig. 3. Gas species concentration in time – experiment no. 2

In Fig. 4 is presented the composition of syngas obtained from the third gasification experiment. According to the Micro-GC analysis, longer residence time of the reactants increases CO and H_2 concentrations, while CO_2 yield decreases.



Fig. 4. Gas species concentration in time - experiment no. 3

The decrease of feed-in substances flow rates along with the increase of reaction time (experiment no. 4), enhance the CO formation (14% - 15%), while the CO₂ concentration decreases to 28% (Fig.5). Hydrogen yield reaches 4%. During experiment no. 4 higher CO concentrations were obtained (about 20%) with an important decrease in CO₂ yield down to 17% while the Hydrogen quota maintained constant as in experiment no. 3.



Fig. 5. Gas species concentration in time - experiment no. 4

The control of oxidation reactions, through the feeding flow rates of biomass and air and the reactants mixing rate is very important. This can be best observed by comparing especially the results obtained from the experiments no. 3 and 5. So, by comparing these experiments can observe that for shorter residence time CO₂ tends to increase (to 30%), while CO decreases fast (to about 2%), because of limited C+CO₂ \rightarrow 2CO reaction. Although it is well known that at small process temperature, less than 700-800°C, CO₂ formation is favored to the detriment of the CO. However, the longer process duration can increase the CO formation. In the combustion stage of the gasification process CO₂ is produced. In the char gasification stage CO₂ reacts with the char to produce CO. Char-gas reaction rates are very slow, therefore in order to ensure a complete char conversion, process duration must be correlated to these reaction rates [28, 29]. The current results show a positive influence of a low reactor rotation speed on CO formation.



Fig. 6. Gas species concentration in time - experiment no. 5

Best gasification results were obtained for experiment no. 6 conducted at higher syngas retention time. Hydrogen concentrations determined by both gas analyzers records very small values, less than 5% with CO yield (20%) overtaken the CO_2 that decreased at about 15%.



Fig. 7. Gas species concentration in time - experiment no. 6

Even if the Hydrogen yields across all experiments are not typical for biomass air gasification processes [28, 30-32], their values are within expected range related to process temperature. Although H_2 production registered was so low, gas energy content was up to 3.50 MJ/Nm³ mainly because of CO relative high concentration [27]. As previously stated, the presence of Nitrogen from the air reactant also limits the syngas specific energy content.

Process energy efficiency was computed for all 6 gasification processes, according to process mass balance and reaction products properties, resulting a maximum process energy efficiency of 74.08% including the syngas sensitive heat calculated at 375°C (exhausted temperature) [27]. Therefore, we can assume that at higher process temperature, carbon monoxide and hydrogen production will increase, leading to a higher calorific value of the gas, and a higher process energy efficiency.

4 Conclusions

The core findings of the study are related to process set-up and installation operation influence on gasification reactions and products formation. The specific operating characteristics of the utilized installation were synthesized as common control parameters available in any gasification facility (feedstock residence time, gas flow rate, solid – gas mixing rate etc). The results indicate the importance of oxidation reactions control, through the feeding flow rates of biomass and air and the reactants mixing rate. Constant Hydrogen yield of about 2-4% was obtained for all experiments while the CO concentration varied from 8 - 21%. The lower heating value of the produced gas reached 3.5 MJ/Nm³ despite the possible false air infiltrations (because of installation construction characteristics) and high Nitrogen quota from the air used as reactant. As expected, the permanent monitoring of gas composition offers information about process status throughout the entire continuous operated system experiment providing process observation. Consequently, the specific flow rates of substances and installation operating conditions were linked to process run through syngas composition.

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