Effect of Reaction Time and Air Flow Rate on the Yield of Hydrogen Obtained from the Gasification of Tobacco Waste

Serkan Karadeniz and Nezihe Ayas

***Abstract*—In order to meet the ever growing energy demand of the world and to keep the environment clean, new studies on the topic of utilization of biomass are being carried out. Using different methods such as gasification to convert waste biomass into more valuable forms are constantly being studied.**

In this study, gasification of tobacco waste was carried out using Na_2CO_3 as a catalyst and dry air as a gasifying agent. Effects of reaction time (10, 15 and 20 min.) and dry air flow rate (2, 3 and 4 L/h) on the product gas composition have been studied. Highest Hydrogen yield of 5.65 mole gas/kg biomass was obtained under 4 L/h dry air flow rate and 15 min. reaction time conditions.

***Index Terms*—Biomass, gasification, hydrogen, tobacco.**

I. INTRODUCTION

Petroleum is the most used energy source on world and it is followed by coal (25%), natural gas (20%), biomass (14%) nuclear power (7%) [1]. Negative effects of using fossil based fuels as a main energy source is the main driving force for the search of new, renewable and environmentally friendly energy sources [2]. Hydrogen fits all these needs and it is considered to be the power source of the future.

Hydrogen is obtained from biomass by using three main methods: pyrolysis, catalytic steam reforming and gasification [3].

Gasification is a process where organic, carbonaceous materials are converted into hydrogen, carbon monoxide and carbon dioxide. This conversion is achieved by reacting the material with oxygen at high temperatures. The amount of oxygen supply is limited below the amount required for total combustion of biomass. The resulting mixture is called syngas or product gas, which can be used as a fuel for various applications. If the product gas is obtained from biomass by gasification, it is considered as a source of renewable energy. Gasification process strips carbon away from biomass and adds hydrogen to it, therefore producing gases that have higher hydrogen-to-carbon ratio.

Gasifying agent is one of the main factors that effect the composition of product gas. In addition to gasifying agent, type of the gasifier also effects the product gas composition. Fixed bed gasifiers are one of the examples that are commonly used. Fixed bed gasifiers can operate as updraft and downdraft gasifiers. Updraft gasifier is thermally more efficient than downdraft gasification but the process results in gaseous product that has high tar content [4].

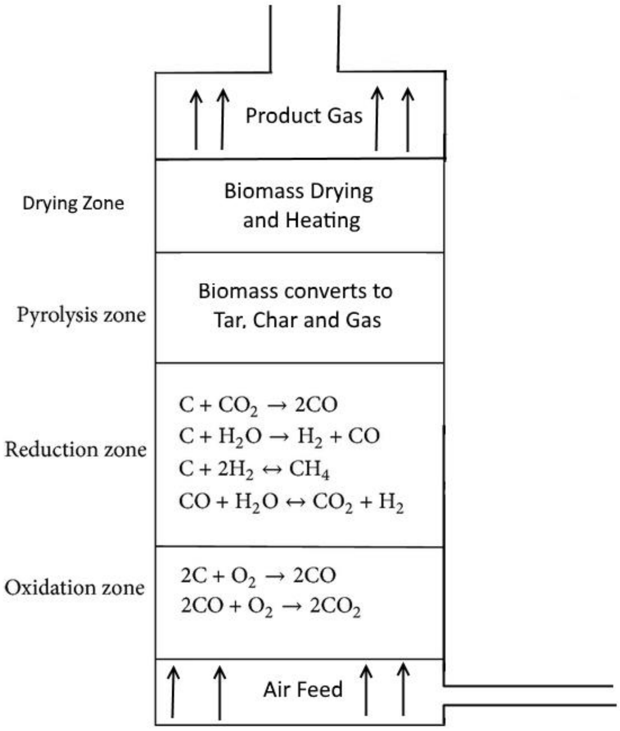

Fig. 1. Different types of reactions that occur during gasification process in an updraft gasifier and their relative zones.

Fig. 1 shows the reactions and their relative zones inside the gasifier during gasification of biomass.

Product gas obtained from gasification has many practical uses. Having a fuel in gaseous from rather than solid form provides more flexibility when it comes to controlling power levels. It is also cleaner than using solid fuels where there is usually a solid or liquid residue that needs to be disposed.

A number of practical applications include heating, electricity generation, combined heat and power and transport fuel. Product gas obtained from gasification process can be used as a replacement to fossil fuels used in ovens, furnaces and boilers. It can also be used in industrial scale heat generation applications where fossil fuels are burned in order to generate steam for steam turbines. Replacing the fossil fuel with a renewable fuel that is obtained from biomass reduces carbon emissions and provides an environmentally friendly alternative.

For combined heat and power, plants can be installed

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locally or individually to provide means of heating and power in places where there is a sustainable supply of biomass. Despite that, many of the agricultural waste is simply used as fuel for heating purposes. Suggesting alternative methods to this application benefits the industry and improves the general life quality of the local area.

Turkey has an important place in the tobacco industry and it occupies the 6th place in the world tobacco production. Between the years of 2002-2011 average tobacco production in Turkey was 98,674 tons per year where 7,000 to 10,000 tons of tobacco waste was destroyed annually during this period without recycling which is significant enough to have an effect on environment and the life quality of the local population [5].

There are several works on utilization of tobacco waste. These studies include reaction kinetics of tobacco under nitrogen atmosphere [6], fast and slow pyrolysis of tobacco residue [7], effect of temperature and inorganic additives on gas yield [8], determination of activation energy of tobacco waste and its parameters [9], hydrogen extraction under subcritical and supercritical conditions [10], investigation of pyrolysis of tobacco stem [11], kinetics and reaction chemistry of waste [12], hydrogen production by steam reforming of char derived from tobacco [13].

Özyuğuran, A. and Yaman, S. studied the heating values of tobacco waste and various biomass types. They determined that out of twenty-seven different biomass types, tobacco waste had lowest value of fixed carbon (11.78%), lowest higher heating value (14.51 MJ/kg) and highest ash yield (15.36%) [14].

Liu, H., E, J., Deng, Y., Xie, C. and Zhu, H. studied the microwave pyrolysis characteristics and the influences on pyrolysis of the tobacco stem. They determined that optimum dose of activated carbon was 10% for high bio-oil yield [15].

Most of the research in the literature is based on pyrolysis, supercritical or subcritical reactions. In our previous research, effects of reaction temperature and catalyst to biomass ratio were studied [16]. This paper focuses on effects of reaction time and gasifying agent flow rate’s effect on hydrogen yield.

Further research on gasification of biomass using commonly available gasifying agents under conditions that are suitable for scale up operations are required.

In this study thermochemical gasification of tobacco waste has been performed in a fixed-bed updraft gasifier by using dry air as gasification agent in order to investigate the effects of gasifying agent flow rate and reaction time on the hydrogen yield.

II. MATERIALS AND METHODS

A. Material

Same tobacco waste from previous study was used [16]. Na_2CO_3 was used as catalyst during gasification process.

B. Equipment

An updraft gasifier which is 900 mm in height, 10 mm wide in diameter and made of 316 stainless steel was used. Reactor itself acts as a resistor and it is capable of reaching 850 °C in under 60 seconds when voltage is applied by the power supply. A hollow, cylindrical, ceramic insulator is placed around the reactor to minimize the heat loss. Gases produced during the reaction are passed through a gas-liquid separator in order to cool the gas and recover the liquid product. Cooled gas is passed through two stage filtering system to filter out suspended particles and moisture before being collected in the gas sampling bag. Then the gas product is analyzed by using a SRA Technologies Micro Gas Chromatograph (μ-GC)(T-3000 series) equipped with MS5A (Molecular sieves 5 Å) and PPQ (PolarPlot Q) columns coupled to thermal conductivity detector (TCD). Helium and Argon are used as carrier gases in the μ-GC.

A schematic of reactor system for this study is shown in Fig. 2.

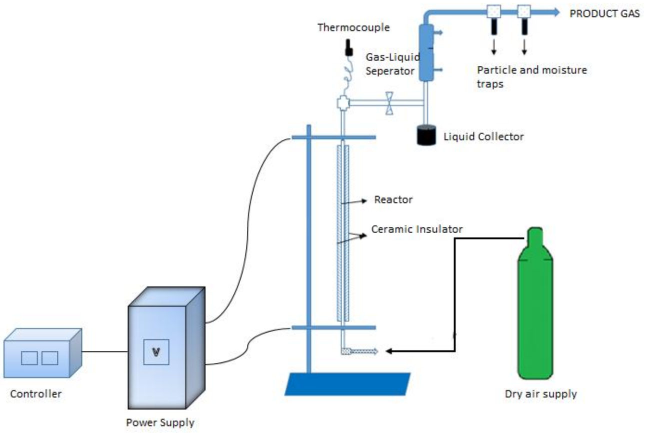

Fig. 2. Reactor system for the gasification of tobacco waste.

C. Experimental Procedure

Tobacco waste sample was milled in a rotary-cutter mill at 8000 RPM (Revolutions Per Minute) using 1.5 mm screen. Biomass samples were dried in oven at 105 °C for 24 h in order to remove any moisture content. Following procedures were carried out after sample preparation.

1) Gasification of the biomass

Tobacco waste was gasified in an updraft reactor in order to determine the effects of dry air flow rate and reaction time. 3 g of tobacco waste was mixed with appropriate amount of Na_2CO_3 catalyst an then placed in the reactor using supports that are made from inert materials. Usage of supports ensured that the reaction temperature was controlled at the same place where the tobacco waste and catalyst mixture was placed inside the reactor.

Reaction temperature was controlled by a controller unit connected directly to the power supply.

Dry air was introduced into the reactor as soon as the heating began, and was kept constant for the duration of the reactions. All experiments reached the desired temperature of 750 °C under 60 seconds. Flow rate of dry air was controlled by a dry air flowmeter.

Product gas is cooled by a gas-liquid separator which is a double-pipe heat exchanger, using water at ambient temperature as cooling fluid.

Cooled product gas is mostly free of tar content but the gas still needs to be cleaned in order to remove any remaining moisture, suspended solid particles and tar. This is done by passing the gas through a particle filter and moisture trap, in order to ensuring that analysis of the gas can be carried out correctly using the μ-GC.

Connection of a gas sampling bag where the product gas is collected after being filtrated for moisture and suspended particles ensured that the reactions were carried out under atmospheric pressure with very little fluctuations that usually occur during the initial heating of the reactor. Product gas was collected in a 4 L gas sampling bag.

Effect of dry air flow rate on the product gas composition was determined by keeping other parameters (Reaction temperature (750 ^{o}C), catalyst to biomass ratio (20%), catalyst type (Na_2CO_3) and reaction time (15 min.)) constant. Using the same method, effects of reaction time on product gas composition was also determined while keeping dry air flow rate (3 L/h) constant.

III. RESULTS AND DISCUSSION

A. Effect of Dry Air Flow Rate

To determine the different flow rates' effect on product gas composition, experiments were carried out at various dry air flow rates (2, 3 and 4 L/h) under the following conditions; reactor temperature of 750 ^{o}C, biomass-to-catalyst ratio of 20% by weight, Na_2CO_3 as catalyst and 15 min. reaction time. Hydrogen and Methane yields are plotted against the dry air flow rate in Fig. 3.

From Fig. 3 it can be seen that increase in flow rate of dry air resulted in an increase in Hydrogen yield which is about 53%. Despite having a higher Hydrogen yield, introducing more dry air to reaction resulted in diminished Hydrogen concentration in product gas due to Nitrogen in dry air being mostly inert during this reaction. In addition to that, having a product gas that is dilute in combustible gases has low heat value and requires more expensive purification systems [17]. Hydrogen and Methane show nearly same levels of increase in yield, which are 53% and 57%, respectively.

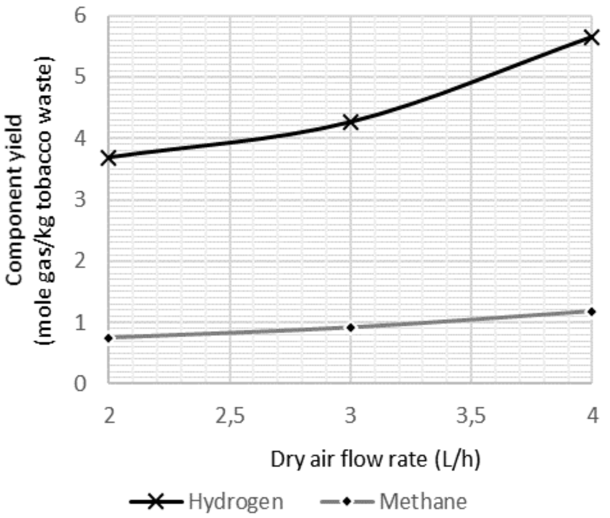

Fig. 3. Effect of dry air flow rate on Hydrogen and Methane yields (Na_2CO_3 as catalyst, %20 wt./wt. catalyst to biomass ratio, 750 ^{o}C, 15 min. reaction time).

Fig. 4 shows that having dry air flow rate of 2 L/h favors the production of CO due to oxygen amount delivered to the reaction being lower than stoichiometrically calculated amount, 3 L/h, therefore favoring partial oxidation of C. While CO and CO_2 yields are similar at 3 L/h dry air flow rate, further increase in the dry air flow rate nearly doubles the CO yield and decreases the CO_2 yield. Change in the yields can be explained by reduced residence time of product gases inside the reactor therefore not allowing enough time for complete oxidation reaction. Similar effects on CO yield caused by increased air flow rate has also been reported in other studies [17].

At 3L/h dry air flow rate, CO yields shows a small increase in yields rather than the expected near-linear increase that is obtained for Hydrogen and Methane. This behavior of CO results in increased Ethylene, Ethane, Propylene and Propane yields whereas CO_2 yields peaks before dropping again with the increased dry air flow rate. Drop in CO_2 yield combined with the drop in Ethylene, Ethane, Propylene and Propane yields result in increased CO yield.

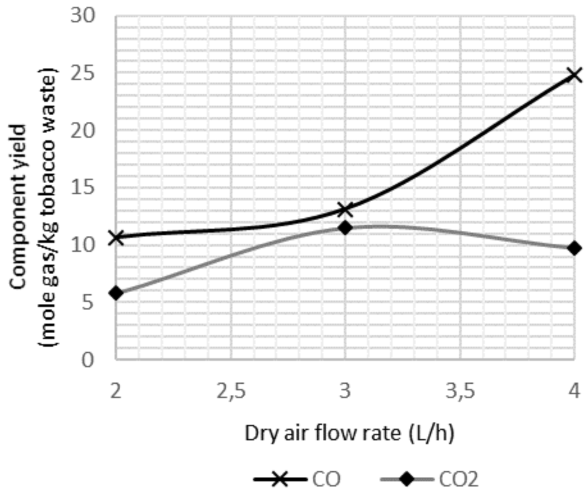

Fig. 4. Effect of dry air flow rate on CO and CO_2 yields (Na_2CO_3 as catalyst, 20% wt./wt. catalyst to biomass ratio, 750 ^{o}C, 15 min. reaction time).

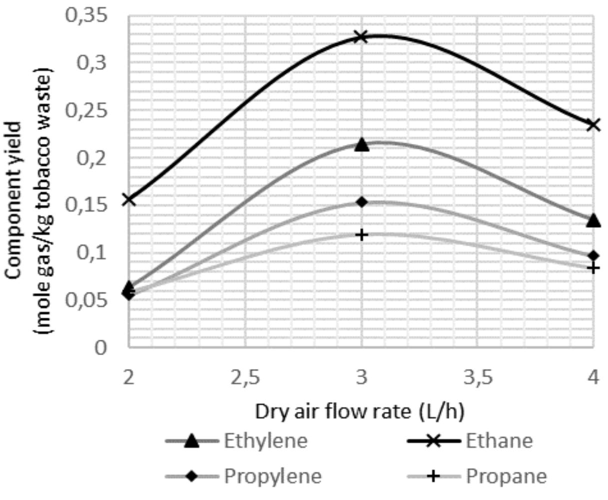

Fig. 5. Effect of dry air flow rate on Ethylene, Ethane, Propylene and Propane yields (Na_2CO_3 as catalyst, 20% wt./wt. catalyst to biomass ratio, 750 ^{o}C, 15 min. reaction time).

From Fig. 5 it can be seen that maximum yields for Ethylene, Ethane, Propylene and Propane were obtained at 3L/h dry air flow rate.

Ethylene, Ethane, Propylene and Propane yields reach their peak yields at 3 L/h dry air flow rate. At 4 L/h dry air flow rate their yields decrease where CO yield in Fig. 4 nearly doubles, showing that 3 L/h dry air flow rate favors the formation of Ethylene, Ethane, Propylene and Propane where 4 L/h dry air flow rate favors the formation of CO.

B. Effect of Reaction Time

In order to determine the effect of different reaction temperatures on product gas composition, experiments were carried out at various (10, 15 and 20 min.) reaction durations under the following conditions; reactor temperature of 750 ^{o}C, biomass-to-catalyst ratio of 20% by weight, Na_2CO_3 as catalyst and 3 L/h dry air flow rate. Hydrogen and Methane yields obtained from these experiments are given in the Fig. 6.

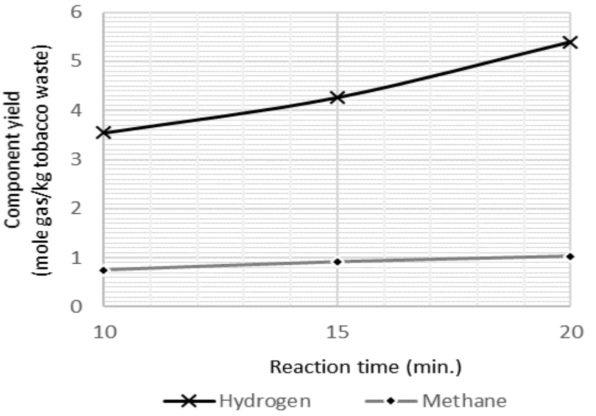

Fig. 6. Effect of reaction time on hydrogen and methane yields (Na_2CO_3 catalyst, %20 wt./wt. catalyst to biomass ratio, 750 ^{o}C, 3L/h dry air flow rate).

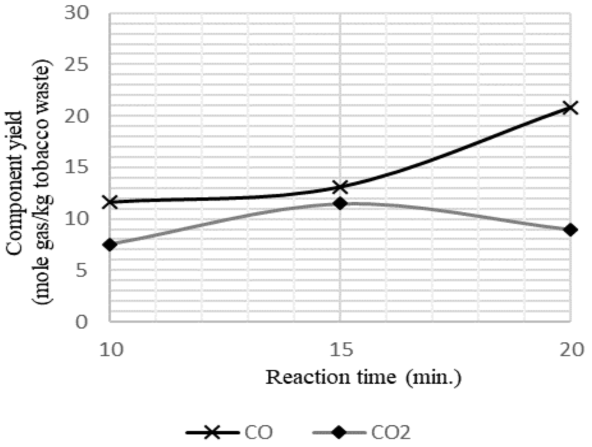

Fig. 7. Effect of reaction time on CO and CO_2 yields (Na_2CO_3 as catalyst, %20 wt./wt. catalyst to biomass ratio, 750 ^{o}C, 3 L/h dry air flow rate).

Hydrogen and Methane yields show a very similar trend when compared to Fig. 3 given in the previous section. Comparing the minimum yields (3.54-3.69 mole gas/kg tobacco waste) and maximum yields (5.39-5.65 mole gas/kg tobacco waste) of Hydrogen given in Fig. 5 and Fig. 3 respectively, it can be seen that having high dry air flow rate rather than longer reaction time increases the Hydrogen yield very slightly. Fig. 7 shows the effect of reaction time on yields of CO and CO_2, Fig. 8 shows the yields of Ethylene, Ethane, Propylene and Propane which show a very similar trend to Fig. 4 and 5 respectively.

From Fig. 7, increment in CO yield with time is higher than with dry air flow rate (Fig. 4). Minimum yields of CO and CO_2 are higher than the yields given in the on Fig. 4. As the reaction time increases, yields of CO and CO_2 also increase, but the change is not as big as the change in yields that are shown in Fig. 4.

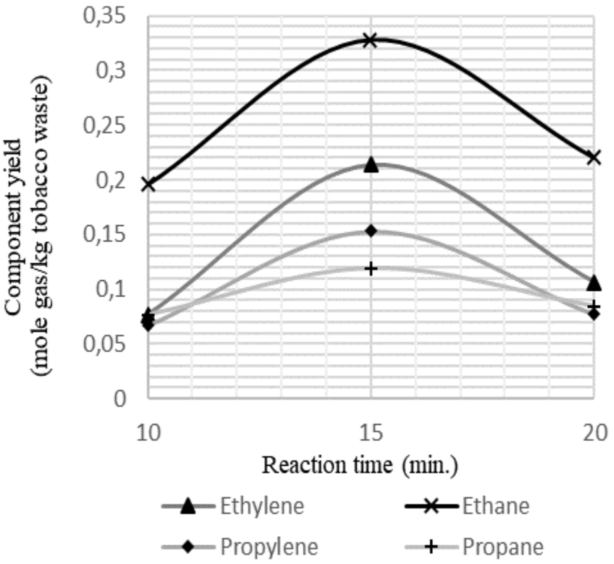

Fig. 8. Effect of reaction time on Ethylene, Ethane, Propylene and Propane yields (Na_2CO_3 as catalyst, %20 wt./wt. catalyst to biomass ratio, 750 ^{o}C, 3 L/h dry air flow rate).

Similar to the behavior in Fig. 7, average Ethylene, Ethane, Propylene and Propane yields are higher than the average yields shown in Fig. 5. This is also in line with the data obtained from CO and CO_2 yields.

IV. CONCLUSION

Effects of reaction temperature and gasifying agent flow rate on product gas composition obtained by gasification of tobacco waste were investigated. Highest Hydrogen yield was obtained as 5.65 mole H_2/kg tobacco waste under following conditions; Na_2CO_3 as catalyst, 750 ^{o}C reaction temperature, 20% wt./wt. catalyst to biomass ratio and 4 L/h dry air flow rate. It was observed that when same amount of gasifying agent is introduced into the reactor, having a higher flow rate for gasifying agent results in slightly higher Hydrogen yield when compared to having longer reaction time.

Studies of Özyuğuran, A. and Yaman, S. [15] show that out of 27 different biomass samples, tobacco waste has lowest amount of fixed carbon, lowest heating value and highest ash yield. These properties make tobacco waste most unsuitable biomass to be used in direct combustion applications. Considering these properties, alternative methods for utilization of tobacco waste should be considered.

Study of additional parameters like effect of different gasifying agents, particle size and catalyst type are also ongoing for this work.

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