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Hydrogen conversion using gasification of tea factory wastes

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Abstract: In this study, gasification performance and importance of hydrogen production using waste of a tea factory were evaluated. A mathematical model was developed for the gasification system, which includes a water gas shift reactor used for hydrogen purification. The gasifier temperature was 877 °C for the developed model. The model has been validated against experimental data from an 80 kW t h cylindrical downdraft gasifier, given in the literature for syngas composition for three different air-to-fuel ratios. With the developed model, hydrogen production from tea wastes was achieved to yield a higher level by additionally using a water gas shift reactor. Tea waste (1000 kg) was gasified and after the hydrogen purification process, a total of 4.1 kmol hydrogen was achieved, whereas the amount would be 2.8 kmol gas hydrogen if a normal gasification method were used. The validity of the developed model was verified by comparing the experimental results obtained from the literature with the results of the model under the same conditions. After verification of the developed model, the effect of the moisture content of the biomass and the air/fuel ratio on the composition of the product gas were investigated. These investigations were also confirmed by experimental data. The results show that it is important to convert biomass waste into a clean energy source of hydrogen to minimize its environmental impact.

Keywords: biomass; environment; conversion; syngas; thermodynamic; equilibrium.

INTRODUCTION

Biomass will be one of the most significant energy sources in the near future owing to its immense accessibility and encouraging potential to decrease global

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warming problems.[1] Many researchers believe that hydrogen is a clean and efficient gas and hence, hydrogen-powered systems could be an important solution to the global energy problem.[2,3] Biomass is an important source for hydrogen production and could be the primary energy-supply system used for implementing hydrogen economy and energy production as a major fuel of the future.[3–6]

Thermo-chemical conversion processes have been of utmost importance in the field of scientific research and these processes have been widely and commercially employed all over the world. Gasification is one of the conventional thermo-chemical conversion processes that enables partial oxidation of biomass at a high temperature in the range of 800–1000 °C, resulting in the conversion of biomass into a flammable gas mixture. The gasification process is realized using air, oxygen and/or steam like gasifying agents. Product of gasification process is the producer gas that contains H_2, CO, CO_2, CH_4 and N_2.[7,8]

Mathematical modeling of biomass gasification could improve both its design and operation, reduce associated problems and facilitate the implantation of this technology.[9,10] The advantages of mathematical modeling are now widely acknowledged by researchers and have been performed by many researchers in the scientific studies.[8,11–13] There are three well-known modeling ways of biomass gasification, the computational fluid dynamic model (CFD), kinetic models and equilibrium models.[14] In the CFD model, the examination of fluid flow is conducted.[15] In order to virtually generate a solution for a physical phenomenon associated with fluid flow, without compromise on accuracy, fluid properties and some major inputs, such as detailed geometry, well mesh structure and boundary conditions have to be considered simultaneously.

Kinetic modeling can give precise results, in particular concerning the time evolution of the process.[16] Nonetheless, this kind of modeling is rather multifaceted. An advantage these models have is that they could be appropriate for studies that focus on reactor design and parameters of different processes (reaction rate, residence time, etc.).

Equilibrium models are known as being simple and speedy. It is almost impossible to reach the equilibrium conditions within the gasifier but these models can define gasification processes with successful approximations. The thermodynamic equilibrium model (TEM) is a simple and beneficial tool for the first estimate and initial assessment of the findings of the gasification process. Through TEM, the influence of fuel and process parameters are studied conveniently independent of gasifier design and thus it enables a reasonable prediction of the maximum achievable yield of a particular product useful for a designer.[17]

There are few advanced technologies to separate hydrogen from the product gas. To obtain high purity hydrogen, the product gas is treated through several steps. Mainly, there are four types of additional operations such as: 1) water gas shift (WGS), 2) scrubber for drying and cleaning, 3) membrane separation and 4) pressure swing adsorption. Very recently, a WGS process with gasification has gained more interest in the production of hydrogen from biomass.[18–20]

Turkey is one of the most important tea producers in the world, and comes immediately after China, India, Sri Lanka, Kenya, Indonesia and Vietnam. The Black Sea region of Turkey is where tea is planted and grown widely. Approximately 70.000 ha are covered with tea plantations and 150.000 t of dry black tea is produced each year. The tea factories in and around the Eastern Black Sea region produce about 30.000 t of waste every year.[21] Despite the fact that the production area remains almost the same, the amount of production increases. According to the production statistics for the year 2016, 1,350,000 t of wet tea were produced.[21] Especially in the Eastern Black Sea region where tea cultivated, solid tea leaves of organic origin, fiber and dust result during the transformation of wet tea leaves to black tea. During tea production, overgrown tea plants are not used in the production process and are directly collected as factory waste.[22] The increasing amount of tea waste creates environmental problems due to limited storage capacity. This massive amount of tea waste is not used for any purpose and hence, they have to be stored in depository areas.[21,22] Although interest is growing for the use of low cost materials and abundantly available lignocellulosic materials to produce liquid and gaseous products and activated carbon, there are very rare studies about tea factory waste treatments, such as pyrolysis, gasification, carbonization, etc. in the literature.[23] Uzun *et al.* pyrolysed tea waste and characterized the obtained bio-oil and bio-char fractions.[21] The effect of temperature (400–700 °C), heating rate (5–700°C/min) and N_2 flow rate (200–800 $cm^3 min^{-1}$) on the product yield was investigated. Maximum yield of the liquid product was determined as 30.4 % at 500 °C, 500 °C min^{-1} heating rate and 200 $cm^3 min^{-1}$ N_2 flow rate.[21] Mahmood *et al.* performed a three step process in order to produce biofuels from spent tea.[24] In the first step, spent tea was gasified in the presence of Co nanocatalyst at 300 °C and atmospheric pressure. Catalytic gasification of spent tea waste yielded 60 % liquid extract, 28 % gaseous product and 12 % charcoal. Gullu investigated the effect of Na_2CO_3 and K_2CO_3 catalysts on the yield of the liquid product obtained from flash pyrolysis of tea waste.[25] Catalytic pyrolysis experiments were performed in order to increase the yield of methanol, which is one of the most valuable fractions in the liquid product. Methanol yield was 8.65 % for non-catalytic run. The methanol yield increased from 8.65 to 10.93 % in the presence of Na_2CO_3.[26]

In this study, the potential of converting the tea factory wastes to renewable energy by gasification was examined. In this context, a mathematical modeling for a biomass gasifier was developed. In the model, the stoichiometric thermodynamic equilibrium biomass gasification model was used. Simulations of models were performed with the optimal rates, which are assumed to be 100 % for carbon conversion efficiency in the gasification process. The most important difference of the developed model from the studies in the literature is that the gasifier has a WGS unit and the model includes both gasifier and WGS reactor operations. The validity of the developed model was verified by comparing the experimental data obtained from the literature with the model results under the same conditions.

EXPERIMENTAL

Model

Achieving the best synthesis gas composition for production of biomass chemicals and power generation is a challenging problem. The correct understanding of the gasification phenomenon, reliable performance prediction through modeling can greatly avoid expensive upsets. Briefly, developing a model is very advantageous for the evaluation, design and analysis of the process. From this viewpoint, in this study, evaluation of tea factory residues gasification performance was simulated by a presently developed stoichiometric thermodynamic equilibrium model. For the gasification model, air and steam are utilized as the gasification agent, and model includes a WGS reactor for hydrogen purification. The concept of the thermodynamic equilibrium model is based on the second law of thermodynamics as applied to chemical reacting systems. A stoichiometric thermodynamic equilibrium model (STEM) based on specific chemical reactions is used here for the estimation of the composition of the product gas. Thermodynamic equilibrium models do not require any knowledge of the mechanisms of transformation. Moreover, they are independent of the reactor and are not limited to a specified range of operating conditions. A STEM was formulated to assess the hydrogen yield from different locally available biomasses at specified operating conditions to select the most appropriate one.[27] In this study, the assumptions of the biomass gasification model are as follows.

- The biomass is modeled considering the carbon, hydrogen and oxygen atoms only.
- The gasifier is a steady state system with uniform pressure and temperature.
- At equilibrium, the reaction system achieves the most stable composition.
- The gases H_2, CO, CO_2, CH_4, H_2O and N_2 considered in the reaction system behave ideally.
- Gasification reaction rate is fast enough and residence time is long enough for the equilibrium state.
- No tar leaves the reaction system at the end of the process.
- Carbon conversion efficiency in the gasification process is assumed as 100 %.

In this study, stoichiometric thermodynamic equilibrium model for biomass gasification is based on the work of Basu.[28] A denotes the air supply in kg dry air per kg dry fuel. F kg of dry fuel is required to obtain N m^3 of the gas, and X_c is the carbon content of the fuel (kg carbon per kg dry fuel). Carbon is split between CO, CO_2 and CH_4. For 1 N m^3 of gas produced, one can write the carbon molar balance between inflow and outflow streams as:

$$FX_C/12 = (V_{CO} + V_{CO_2} + V_{CH4})/22.4 \quad (1)$$

where V represents the volumetric fraction of a constituent of the gas. The molar balance of H_2 is as follows:

$$F(S/18 + X_H/2 + W/18) = (V_{H2} + V_{H2O} + V_{CH4})/22.4 \quad (2)$$

where X_H is hydrogen content of the fuel (kg hydrogen/kg dry fuel). S represents the total steam supplied as humidifier associated with air and added steam (kg steam/kg of dry fuel), and W represents the moisture content of fuel (kg water / kg dry fuel). If O_a represents the mass fraction of oxygen in air and X_O is the oxygen content of the fuel (kg oxygen/kg dry fuel), the molar balance of O_2 is as follows:

$$F(S/36 + X_O/32 + W/36 + AO_a/32) = (0.5V_{CO} + 0.5\ V_{H_2O} + V_{CO_2})/22.4 \tag{3}$$

where A is kg air / kg dry fuel. If X_N is the nitrogen content of the fuel (kg nitrogen/kg dry fuel) and N_a is the mass fraction of nitrogen in air, the molar balance of N_2 gives:

$$F(X_N/28 + AN_a/28) = V_{N_2}/22.4 \tag{4}$$

The volume fractions of all constituents of the product gas are equal to 1.0.

$$V_{CO} + V_{CO_2} + V_{H_2} + V_{CH_4} + V_{H_2O} + V_{N_2} = 1 \tag{5}$$

To estimate the values of the seven unknowns: V_{CO}, V_{CO2}, V_{H_2}, V_{H_2O}, V_{N_2}, V_{CH4} and F, seven equations are needed. The remaining two equations can be formed by assuming that all the reactions occurring in the gasification zone are in thermodynamic equilibrium. For the Boudouard reaction, the equilibrium constant is:

$$K_{pb} = P_{CO}^2/P_{CO_2} = (V_{CO}P)^2/V_{CO_2}P \tag{6}$$

where P_{CO} is the partial pressure of CO, which is equal to volume fraction of CO ($V_{CO}P$ – the pressure of the reactor, P). Similarly, for the water–gas reaction:

$$K_{pw} = (P_H P_{CO})/P_{H2O} = (V_{H2}P)(V_{CO}P)/(V_{H_2O}P) \tag{7}$$

Volume fractions of the product gas constituents can be obtained by simultaneously solving these equations using the combined relaxation Newton–Raphson method in the visual basic net platform, which uses the results of ultimate and proximate analysis of biomass as input data. It is assumed that all of the reactions in biomass gasifiers are thermodynamically balanced with each other in the equilibrium model in this work. The reactions used in the model are given in Table I. The equilibrium constants of the water–gas reaction (K_{pw}), Boudouard reaction (K_{pb}), and the WGS reaction (K_{ps}) are given in Table II.[28,29]

TABLE I. Chemical reactions involved in the gasification system[8]

No.	Reaction	Equals	ΔH / kJ mol^{-1}
1	Oxidation I	$C + O_2 = CO_2$	–394.5
2	Oxidation II	$C + ½O_2 = CO$	–111.5
3	Water–gas	$C + H_2O = CO + H_2$	131.4
4	Boudouard	$C + CO_2 = 2CO$	172.6
5	Methanation	$C + 2H_2 = CH_4$	–74.9
6	Steam reforming	$CH_4 + H_2O = CO + 3H_2$	206.2
7	WGS	$CO + H_2O = CO_2 + H_2$	–41.2

TABLE II. Equilibrium constants

Temperature, K	K_{pw}, Eq. (3)	K_{pb}, Eq. (4)	K_{ps}, Eq. (7)
400	7.7×10^{-11}	5.2×10^{-14}	4050.00
600	5.1×10^{-5}	1.9×10^{-6}	27.00
800	4.410^{-2}	1.1×10^{-2}	4.04
1000	2.62	1.90	1.38
1500	6.08×10^{2}	1.62×10^{3}	0.37

When the volume fractions are determined, the lower heating value of the product gas (*LHV*) can be calculated from gas composition and this value is stated in volume basis as shown in Eq. (8):[30]

$$LHV = 10.79V_{H_2} + 12.26V_{CO} + 35.81V_{CH_4} \tag{8}$$

The input parameters for the model are biomass-feed rate, biomass properties, gasification temperature and pressure, air-to-fuel (*A*/*F*) ratio and steam-to-fuel (*S*/*F*) ratio. The simulation model calculates the synthesis gas composition, H_2/CO and *LHV* for the *A*/*F* and *S*/*F* under the given operating conditions. The gasifier temperature has to be conserved within the range of 700–850 °C. At lower temperatures, the efficiency of gasification is lower, and the tar content of the gas is excessive.[8] The *A*/*F* rate and *S*/*F* rate are 0.2 due to the high content of O_2 in biomass residues. The gasifier temperature is 877 °C for the developed model. In this study, the developed biomass gasification model also includes a WGS reactor for hydrogen purification. In the WGS process, CO is converted to H_2 *via* the reaction between steam and CO at different conditions. In Fig. 1, the flow diagram of the model calculations is given. The input parameters for the WGS reactor are syngas composition from the gasifier outlet, the WGS reactor temperature and pressure, and steam feed. In WGS reactor, the model assumptions are as follows:

- The WGS reactor is a steady state system with uniform pressure (1 atm) and temperature (310 °C).
- At equilibrium, the reaction system achieves the most stable composition. The WGS reaction rate is fast enough and residence time long enough for the equilibrium state.

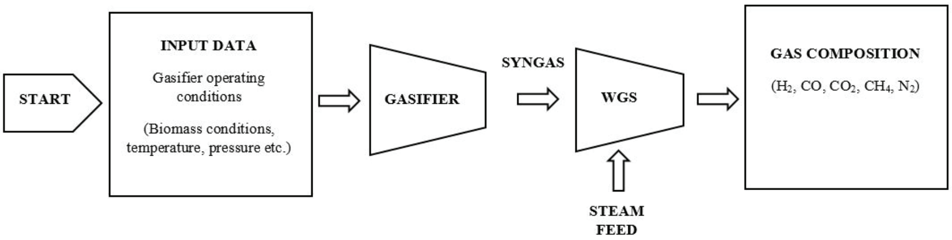

Fig. 1. Flowchart of the biomass gasification model.

In this study, a high temperature WGS reactor was used. It is clear from the model results that using the WGS reactor is important for H_2 production. However, results and discussion, the high temperature WGS reactor did not provide complete conversion of CO. It should be noted that the WGS reaction is exothermic and the hydrogen product is thermodynamically favored at low temperature, 200 °C.[31,32] However, at low temperatures, the reaction kinetics are slow, even using catalysts and excess steam needs to be recycled.[33] Typically, a high temperature shift, operating 300–350 °C maximizes the conversion[34] and on an industrial scale, a higher temperature WGS is usually realized.[35] Therefore, in this study, a WGS reactor operating at 310 °C was considered in the model.

Model validation

The predictions of the STEM model developed in this study were validated with the experimental data obtained from the literature for an 80 kWth cylindrical downdraft gasifier (inner reactor diameter is 0.92 m and reactor height is 1.15 m) fed with rubber wood.[36] The ultimate and proximate analysis of rubber wood is given in Table III. Three experimental data from

Jayah *et al.* for three different air-to-fuel ratios were used to compare with the model predictions in terms of the gas composition on the gasifier exit.[36] The comparison of the gas composition at the gasifier exit between experimental data and model predictions is given in Table IV. The model results have a good consistence with experimental ones at air-to-fuel ratios of 1.86, 1.96 and 2.03. The model predictions fit very closely with the experimental data in Table IV. The maximum error between the results of experiments and the model is less than 2.09 %.

TABLE III. Proximate and ultimate analysis of rubber wood[36]

Component	Value
Content (proximate analysis, dry basis), wt.%	
Volatile matter	80.10
Ash	0.70
Fixed carbon	19.20
Content (ultimate analysis, dry basis), wt.%	
C	50.60
H	6.50
N	0.20
O	42.00

TABLE IV. A comparison of model predictions with the experimental data from Jayah *et al.*[36]

Content[a] wt.%	Air/fuel ratio	Gas composition, dry basis, vol.%										Data source
		CO	Error	H_2	Error	CO_2	Error	CH_4	Error	N_2	Error	
18.5	2.03	19.6	2.061	17.2	1.872	9.9	1.909	1.4	2.071	51.9	2.025	Model
		20.8		15.0		9.0		1.5		53.2		Experiment
16.0	1.96	18.4	2.032	17.0	1.864	10.6	1.924	1.3	2.076	52.7	2.024	Model
		19.0		14.7		9.8		1.4		54.0		Experiment
14.7	1.86	19.1	1.952	15.5	1.864	11.4	1.929	1.1	2.090	52.9	2.039	Model
		18.2		13.4		10.6		1.2		55.0		Experiment

[a] Fuel moisture content

RESULTS AND DISCUSSION

In this study, hydrogen production from tea wastes by the gasification method was evaluated with the developed mathematical model. The composition of the tea wastes used in the study is given in Table V in detail.

TABLE V. Proximate and ultimate analysis of tea waste[37]

Component	Value
Content (proximate analysis, as received basis), wt.%	
Moisture	4.00
Ash	4.13
Fixed carbon	31.90
Content (ultimate analysis, dry basis), wt.%	
C	46.40
H	5.32
N	4.14
O	44.14

An overview of the hydrogen production process and the results of the gasification and WGS reactor are given in mol and percentages in Fig. 2. Besides, the mathematical model results for tea waste gasification also resulted in the observation of the higher heating value of fuel and hydrogen content. Fig. 2 illustrates the yield of H_2 is 2.8 mol H_2/kg tea waste and CO yield is 6.2 mol CO/kg tea waste at the gasifier outlet. However, CO_2 yield is relatively low under this condition. This can be associated with the Boudouard reaction, which occurs in the gasification of tea waste. Ergun reported that carbon has ability of retaining oxygen at sites on its surfaces by chemical bonding. CO_2 is reduced to CO on the carbon surface at temperatures as low as 600 °C. Oxygen retained on the carbon surface can be removed by CO. At equilibrium, most of the CO_2 produced in the gasification was converted back to CO by the Boudouard reaction.[37,38] These results are also in agreement with parametric study of Ayas and Esen in which tea waste was gasified at temperatures of 450, 650 and 850 °C and 1 atm in an updraft air gasifier.[37] By the gasification of tea wastes, it was found that the amount of CO is nearly two times higher than the amount H_2 in the syngas at the gasifier outlet. The O_2 and C ratios of tea wastes are quite high, as shown in the ultimate analysis in Table III. This causes the C in the tea wastes to turn into CO with the air used as the agent during the gasification. Therefore, a high amount of CO emerges from the tea waste in the gasification process. The WGS reaction is exothermic and hence, at higher reaction temperatures (300–400 °C) the reaction shifts to the reagents and reduces CO and favors H_2 production, as can also be seen from Fig. 2.

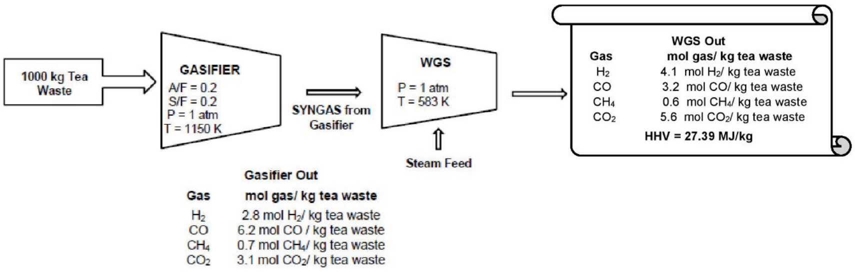

Fig. 2. Production of H_2 from tea waste.

CONCLUSIONS

Hydrogen plays a very important role in the relationship between sustainable environment and energy. Hydrogen is seen as the fuel of the future because it is clean and environmentally friendly. In this study, the gasification performance of the waste of a tea factory was investigated by a developed stoichiometric thermodynamic equilibrium biomass gasification model. This study proves that a WGS reactor system plays a very important role in production of hydrogen from the conversion of CO. With the development of hydrogen production technology, biomass will play a vital role in the development of hydrogen usage. In this study, it was revealed that tea wastes have a serious hydrogen potential and they should be reused with gasification technology. In the scope of the study, it was shown that the addition of WGS reactors in biomass gasifiers increases the potential of converting biomass fuels to hydrogen.

ИЗВОД

КОНВЕРЗИЈА ВОДОНИКА ГАСИФИКАЦИЈОМ ОТПАДА ФАБРИКЕ ЧАЈА

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У овој студији процењене су перформансе гасификације и значај производње водоника коришћењем отпада фабрике чаја. Израђен је математички модел за гасификациони систем, који укључује реактор за измену воде и гаса који се користи за пречишћавање водоника. Температура гасификатора је 877 °C за развијени модел. Модел је валидиран на основу експерименталних података цилиндричног гасификатора снаге 80 kW th датих у литератури за састав синтетског гаса за три различита односа ваздуха и горива. Са развијеним моделом, постигнута је већа производња водоника из отпадног чаја уз употребу реактора за измену вода–гас. Укупно 1000 kg чајног отпада је гасификвано и после процеса пречишћавања водоника добијено је укупно 4,1 kmol водоника. Употребом уобичајене методе гасификације, резултат би био 2,8 kmol водоника. Валидност развијеног модела проверена је упоређивањем експерименталних резултата из доступне литературе и резултата модела под истим условима. Након верификације развијеног модела, испитиван је утицај садржаја влаге биомасе и ваздух/гориво однос на састав произведеног гаса. Ова истраживања су потврђена и експерименталним подацима. Резултати указују да је важно претворити отпад из биомасе у чист извор енергије попут водоника како би се смањио утицај отпада на животну средину.

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