PAPER • OPEN ACCESS

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To cite this article: Nuth Sirirermrux and Somrat Kerdsuwan 2019 *IOP Conf. Ser.: Earth Environ. Sci.* **265** 012017

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Steam Gasification of Municipal Solid Waste in Drop Tube Fixed Bed Reactor

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Abstract. Waste-to-Energy (WTE) technology becomes crucial option for Municipal Solid Waste (MSW) disposal and recovery clean energy. Thermal conversion technology by steam gasification plays important key role for sustainable solution of WTE and enrich the production of Hydrogen. In this work, gasification experiment was conducted in small dropped tube fixed bed reactor by feeding surrogate MSW which including of food & kitchen waste, plastic (polyethylene & polypropylene), paper, rubber & leather, textile and biomass. The experimental conditions were varied at temperature 700, 800 and 900°C. Steam was supplied as gasifying agent with flow rate of 0.1, 0.2 and 0.3 ml/min. The main purpose was to produce hydrogen by water gas-shift reaction, nevertheless, other related producer gas e.g. carbon monoxide, methane, carbon dioxide and light hydrocarbon gas were also examined. The result showed reaction temperature 800°C with steam flow rate 0.2 ml/min offer the optimized hydrogen yield as 34.84 g_{H2}/kg_{MSW} whereas it trended to decrease when reaction temperature increase. In addition, the overall performance of experimental condition was evaluated by energy output and energy conversion efficiency which were calculated from volumetric of combustible gas. The minimum energy output and energy conversion efficiency were 7,638 kJ/kg_{sample} and 31.11%, respectively, obtained at reaction temperature 700°C with steam flow rate 0.2 ml/min while the maximum value was offered by reaction temperature 900°C with same steam flow rate as 17,756 kJ/kg_{sample} and 72.32%, respectively.

1. Introduction

Hydrogen energy is considered as one of the most attractive energy for future trend due to its high energy content and zero global warming emission. In present, main technology to produce hydrogen is steam reforming of fossil fuels. This causes the question of sustainable management; thereby other alternative sources of producing hydrogen have been studied. Other attractive sources of hydrogen production come from renewable source e.g. bioethanol, biodiesel, biogas, biomass and municipal solid waste (MSW). Although the cost of hydrogen production of those mentions are currently higher than that from conventional fossil method but it is postulated to reduce in the near future.

Since the increasing of MSW causes a lot of health and sanitation problems for human including of environmental impact, waste management becomes prior inquiry in many countries. A lot of land space is spend for waste disposal by landfill including cost of waste transportation especially in developing country, however, the huge number of unsanitary disposal of MSW is still remain. In Thailand, 27.37 Million tons of MSW was collected in 2017 while 8.51 Million tons was recycled but 7.17 Million tons was unsanitary disposal [1]. The unsanitary disposal does not only cause environmental harmful in soil and water contamination but also air pollution when MSW was burned in open atmosphere. Hence, gasification technology is one of the promising solutions in fast conversion of carbonaceous solid waste into the form of combustible gas and can minimize the cost of waste management. To produce hydrogen-rich gas, steam is selected gasifying agent. It is lower cost of installation compared to pure oxygen gasification which produces high producer gas heating value but the cost of investment is not attractive while air gasification is the cheapest investment but lowest efficiency [2]. The molar fraction of hydrogen was observed to more than 50% in producer gas in steam gasification without any catalyst involved [3]. The presence of steam in gasification reaction is not only increasing hydrogen concentration but also decreasing tar content. Tar causes the process's problems by blocking pipes and ruining syngas treatment system that reduces the overall efficiency of the process. Steam gasification is a technique to convert aromatic rings hydrocarbon in fuel into smaller molecule by steam reforming reactions. After diminishing of hydrocarbon aromatic rings, the smaller molecule hydrocarbon can be converted to carbon monoxide, hydrogen, methane and light hydrocarbon gas. From the studies of tar cracking of MSW by varying steam and temperature, with the combination of steam gasification and special design catalytic tar reforming reactor, tar-free producer gas can be presented [4, 5].

This paper presents gasification experiment in small drop-tube fixed bed reactor. Steam was supplied as gasifying agent for the main purpose of producing hydrogen-rich gas. To study the influence of temperature and steam amount on gas production, the reaction was conducted at 700, 800 and 900°C with different steam flow rate of 0.1, 0.2 and 0.3 ml/min. Besides, pyrolysis was conducted at the same reaction temperature to distinguish gas compositions among these two different methods. Surrogate MSW which was considered only combustible materials was experimental feedstock. Its compositions came from the average composition of Thailand's municipal solid waste to prevent the unstable waste composition from different provinces. The producer gas quality was reported in term of hydrogen yield and molar concentration of related gas e.g. carbon monoxide, methane, carbon dioxide and light hydrocarbons. In addition, energy output and energy conversion efficiency remarkably the overall efficiency of each test condition were investigated.

2. Materials and methods

2.1. Municipal solid waste
Since the different topographies, occupations and behavior of consumptions cause different compositions of municipal solid waste in Thailand. To find the constant fractions in the experiments, waste compositions from several provinces in Thailand has been reviewed. Only combustible materials were considered and had been averaged to the overall constant fraction called 'surrogate MSW'. Wet percentage the surrogate MSW is presented in table 1 and its proximate & ultimate analysis are presented in table 2. Each component was separately prepared by drying at temperature 105°C for 24 hours and grinding processes. The average size was 100-500 μm. Each component had been weighted by its dry percentage and mixed together, afterward; a mixed sample was compressed into small tablets.

Table 1. Average composition of surrogate MSW (%wt.) [6].

Food & kitchen waste	plastic (70% PE, 30% PP)	Paper	Rubber & leather	Textile	Yard waste & biomass
55.0	22.4	13.0	1.4	3.1	5.1

Table 2. Proximate and ultimate analysis of surrogate MSW [6].

Proximate analysis (dry basis)	(%wt.)
Moisture	0.94
Volatile matter	83.65
Fixed Carbon	6.45
Ash	9.91
Ultimate analysis	(%wt.)
C	54.30
H	8.338
N	0.695
S	0.483
Lower heating value (kJ/kg)	24.53

2.2. Experimental procedure

Test rig included of reactor, electric furnace with temperature controller, steam generator, temperature controller, water pump, sample holder, mass flow controller and gas & tar sampling unit. The operation unit was demonstrated in figure 1. Reactor was made from quartz tube with internal diameter 13.5 mm and length 500 mm installed vertically inside furnace with internal heating zone length 250 mm. Sample holder located on the top of reactor and they were separated by stainless steel gate valve. The experiments were conducted slightly over atmospheric pressure in isothermal condition. Before the experiment, temperature at center of reaction zone inside reactor was measured by thermocouple type K to confirm that the position of reaction occurred at surface of sample was exactly the same with the designed temperature. Afterward, it was removed and argon carrier gas had been supplied into the reactor to replace oxygen and nitrogen inside. Flow rate of carrier gas was set up at 90 ml/min. Steam generator was the heat pipe operating at temperature 300°C. Water was pumped from reservoir into steam generator then water in liquid phase was totally converted into steam vapor. It was supplied at least 20 minutes before conducting the gasification reaction.

In the experiment, 1.2 g of mixed sample was compressed into 7-8 tablets. Size of each was approximately 9 mm diameter and 4-5 mm thickness. Then they were put inside sample holder on the top of reactor. Reaction temperatures were set up at 700, 800 and 900°C and steam flow rate of each different temperature were 0.1, 0.2 and 0.3 ml/min. When the temperature inside reactor remained stable, steam was supplied into reactor at least 20 minutes before sample was dropped. To start experiment, since gate valve at sample holder was opened and sample was being dropped into the center of reactor, producer gas had been collected immediately into gas sampling bag. Besides, large molecule aromatic hydrocarbon was trapped by tar cold trap unit. The reaction was hold until 75 minutes later before producer gas was analyzed by gas chromatography with TCD detector. The analysis units can be separated into; Activated Carbon column *(SRI model 310C)* for hydrogen and methane analysis, Molecular Sieve 5A column *(Shimadzu model GC-8A)* for carbon monoxide analysis and Porapak N column *(Shimadzu model GC-8A)* for carbon dioxide and light hydrocarbons *(C_2H_2, C_2H_4, C_2H_6)* analysis.
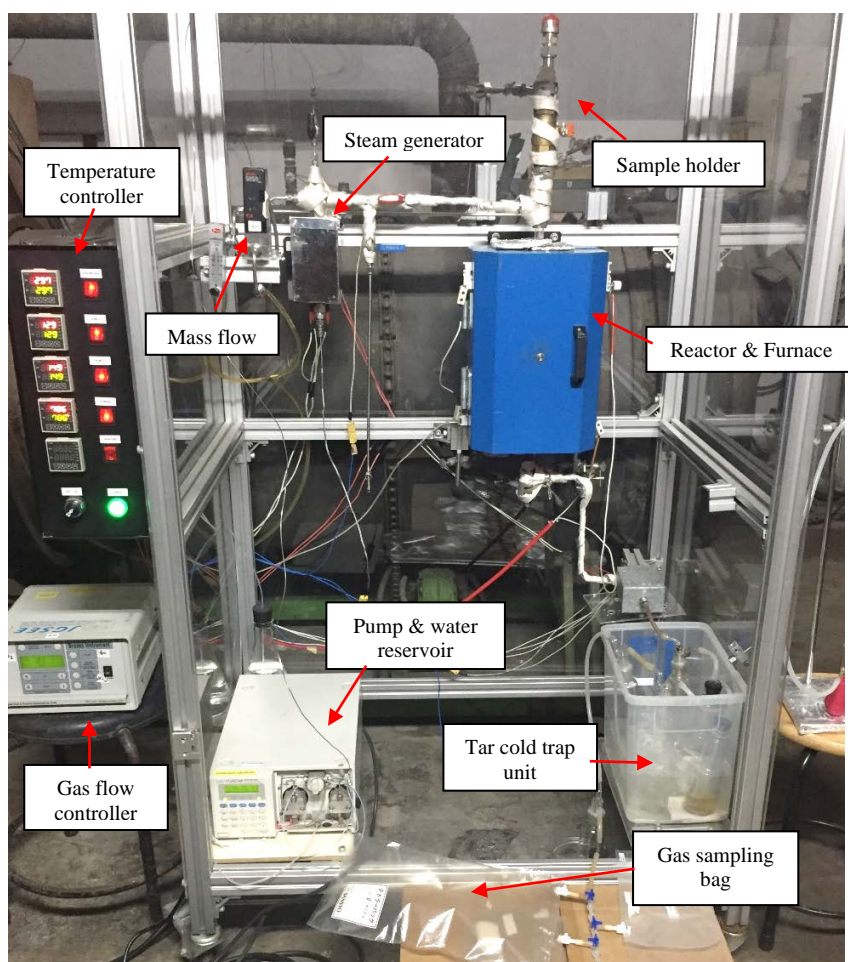
Figure 1. Operation unit of drop tube fixed bed reactor.

3. Results and discussion

3.1. Gas yield and hydrogen yield

The purpose of using steam agent in gasification is to produce hydrogen-rich gas including of tar cracking by steam and enhancing efficiency of the system. The experiments were conducted at temperature 700, 800 and 900°C with steam flow rate of 0.1, 0.2 and 0.3 ml/min. In addition, pyrolysis experiments were conducted as the same reaction temperature. The chemical mechanism might be described by the following reaction as shown in equation (1)-(9);

$$MSW \rightarrow Gas_{(H_2, CO, CO_2, C_nH_m,)} + H_2O + Tar + Char \tag{1}$$

$$Tar \rightarrow CO_2 + CO + H_2 + CH_4 + C_nH_m \tag{2}$$

$$C_nH_m + nH_2O \leftrightarrow nCO + (n + m/2)H_2 \tag{3}$$

$$C + H_2O \rightarrow CO + H_2 \tag{4}$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 \tag{5}$$

$$C + 2H_2 \rightarrow CH_4 \tag{6}$$

$$CO + 3H_2 \leftrightarrow CH_4 + H_2O \tag{7}$$

$$C + CO_2 \rightarrow 2CO \tag{8}$$

$$CH_4 + CO_2 \leftrightarrow 2CO + H_2O \tag{9}$$

The experiment result of hydrogen yield is demonstrated in figure 2. H_2 yield was reported in term of gram of hydrogen produced per one kilogram of feedstock. It could be observed that H_2 yield increased drastically in steam gasification compare to pyrolysis in the lowest reaction temperature and minimal steam flow rate. H_2 yield became higher both pyrolysis and gasification since temperature increased from 700 to 800°C. Higher temperature would probably lead to an increase in H_2 production due to tar thermal cracking, equation (1) and (2), as well as several steam reforming reactions, equation (3). Most of steam reactions are endothermic and as a result favours the H_2 production by water gas, equation (4), and afterward water gas shift, equation (5) [7]. When steam flow rate was increased at temperature above 800°C, H_2 slightly decreased. This came from two competing effects; first, the condensable hydrocarbons decomposed from sample got a shorter residence time to be reformed. Second, too much steam quantity in higher temperature accelerates the reaction [8]. In addition, at temperature 900°C, reversed water-gas shift might take place. The highest H_2 yield was 34.47 g_{H_2}/kg_{MSW}, offered by test condition of 800°C with steam flow rate 0.2 ml/min. Figure 3 demonstrates the dry gas yield. In pyrolysis, char and tar were rapidly decomposed in high temperature followed equation (1) and (2). The rate of reaction depended on reaction temperature significantly; therefore, the higher temperature resulted in larger gas yield. In contrary, gas yield of gasification was generated from steam reforming of tar and char. Hence, at the same experimental condition and reaction time, dry gas yield of gasification was higher while pyrolysis needed longer reaction time
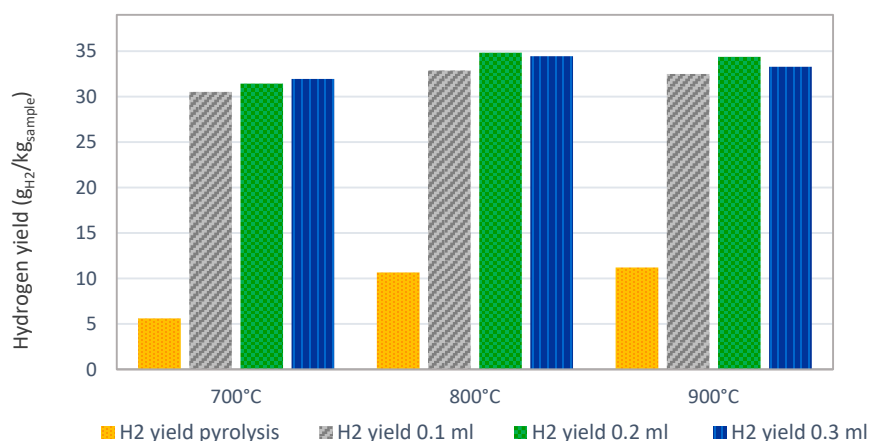

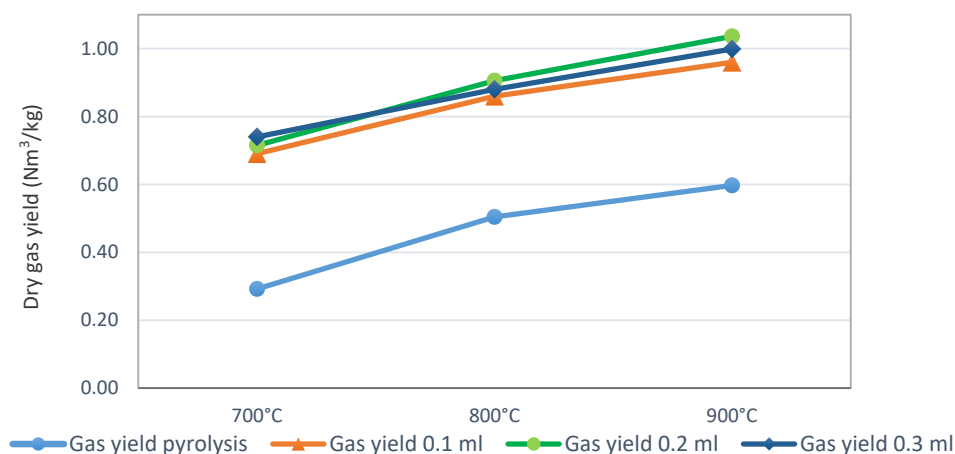**Figure 2.** Hydrogen yield of pyrolysis and gasification.

Figure 3. Dry gas yield of pyrolysis and gasification.

3.2. Effect of temperature and steam on CO, CO_2, CH_4 and C_2H_y concentration

The molar concentrations of methane, light hydrocarbons)C_2H_y(, carbon monoxide and carbon dioxide were presented in figure 4) .a (and)b (and figure 5) .a (and)b(, respectively .In pyrolysis condition, sample was rapidly thermal decomposed, hence, methane and light hydrocarbon gas including of acetylene, ethylene and ethane were produced in high rate due to decomposition of plastic fraction .From this experiment, ethylene fraction was higher than 50 %of C_2H_y .Thermal cracking of tar is depended on temperature so that the higher temperature caused more hydrocarbon gas produced . In gasification conditions, tar cracking by steam in high temperature was following equation)2 (and)3 (caused CH_4 and C_2H_y produced .CH_4 also was generated by hydrogasification of char, equation (6) and methanation by consuming of CO, equation (7). Although the reactions are exothermic and they could occur in both auto-thermal; means heat released drives the gasification itself, and allothermal; means the system requires heat supplied from external source, but the heat needed to initiate the reaction was sufficient in isothermally allothermal condition . In parallel, it was reformed by steam reforming of methane when higher steam content existed) reverse of equation (7) .(Hence, when temperature was changed from 800 to 900°C, trend of CH_4 obviously increased but it decreased when fixed temperature and steam flow rate was increased from 0.2 to 0.3 ml/min due to steam reforming of methane.
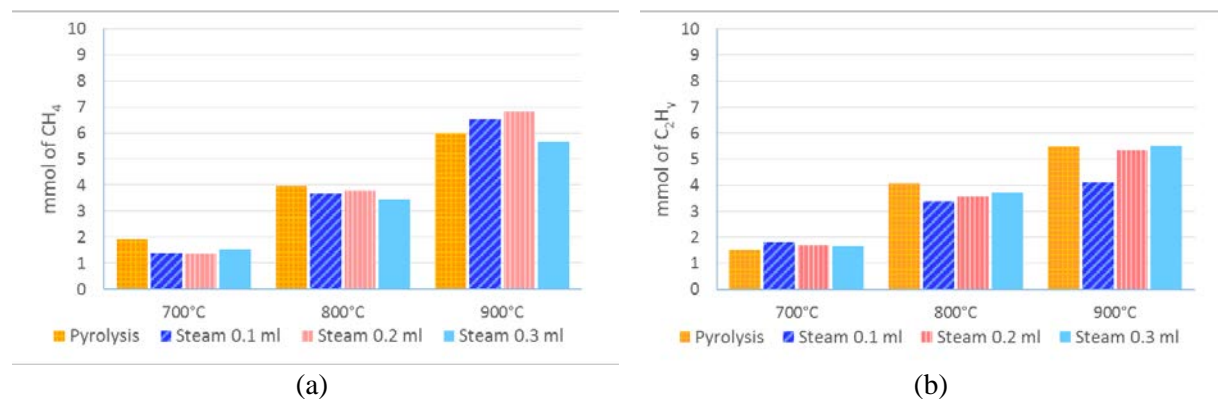

(a)   (b)

Figure 4. Molar concentration of CH_4 (a) and C_2H_y (b) from pyrolysis and gasification conditions.

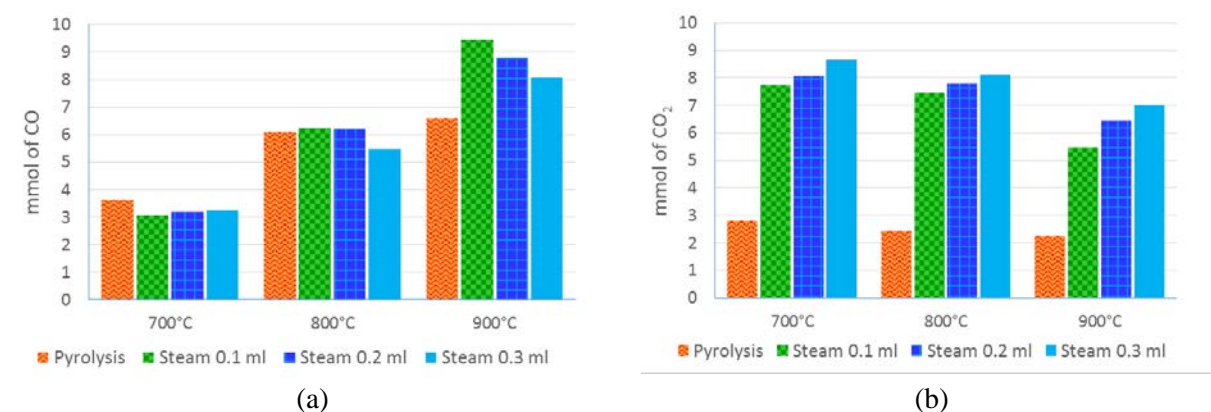(a)   (b)

Figure 5. Molar concentration of CO (a) and CO_2 (b) from pyrolysis and gasification conditions.

Theoretically, at temperature above 830°C, CO could be produced by Boudouard reaction, equation (8), as can be seen that, in pyrolysis condition, CO trended to increase when temperature raised from 700 to 900°C [9]. The same action remarkably occurred in gasification, at the same steam flow rate, CO trended to increase in higher temperature by reversed water gas-shift and methane reforming reaction. However, at temperature 800 and 900°C, when steam was increased from 0.1 to 0.3 ml/min, CO concentration was found to be decreased. The highest CO concentration was 9.42 mmol obtained

from temperature 900°C with steam flow rate 0.1ml/min. In contrary, CO_2 showed decreasing trend when increasing of temperature. Previous studies notified that in pyrolysis condition, CO_2 was less produced than that of CO especially in higher temperature [8, 10]. When steam existed in gasification condition, it drastically increased from pyrolysis condition as 2.81 mmol to 7.75 mmol at 700°C with minimal steam flow rate 0.1 ml/min. This came from water gas-shift reaction takes place when steam content exists, resulting in decreasing of CO but increasing of CO_2.

3.3. Energy conversion efficiency
Energy output of steam gasification and pyrolysis were calculated by;

$$Total\ energy\ output = \sum\{volume\ of\ producer\ gas\ \times net\ heating\ value\}\ Nm^3/kg, \qquad (10)$$

where producer gas include H_2, CO, CH_4, C_2H_2, C_2H_4, C_2H_6. Each gaseous possess different heating value based on Nm^3 per 1 kg of sample.

Similarly, energy conversion efficiency was calculated from;

$$Energy\ conversion\ efficiency = \frac{Total\ energy\ output\ per\ 1\ kg\ of\ sample}{Lower\ heating\ value\ of\ dry\ basis} \times 100\%. \qquad (11)$$
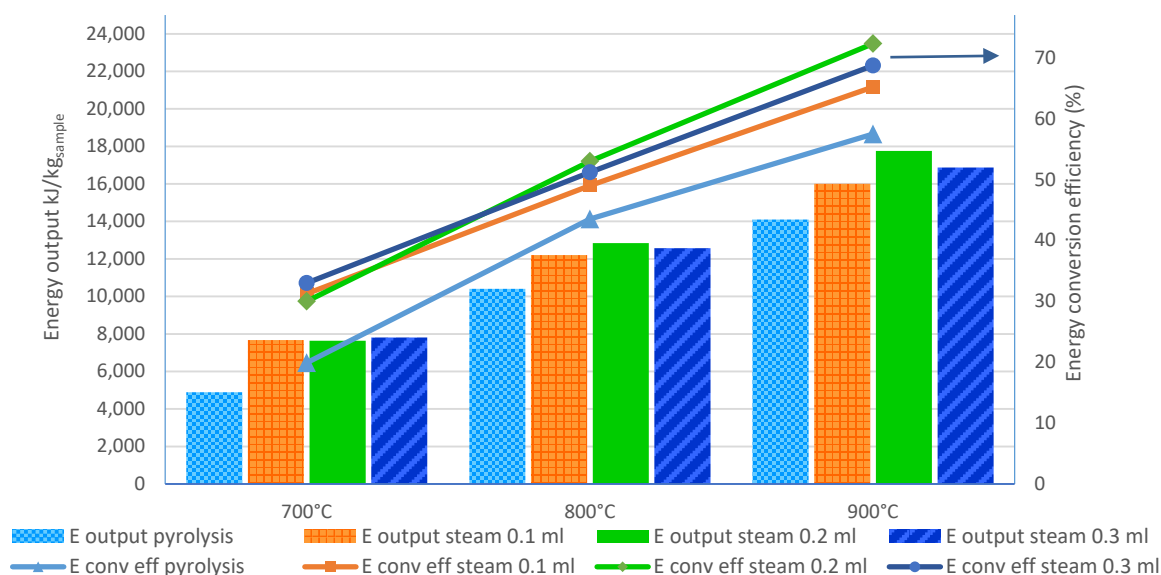

Figure 6. Energy output and energy conversion efficiency from gasification and pyrolysis.

Energy output and energy conversion efficiency were plotted in figure 6. Energy output was a function of temperature which related to dry gas yield. When the reaction temperature raised from 700 to 800 and 900°C, the energy output was significantly increased more than 1.5 and 2 times, respectively. In pyrolysis condition, most of the heating value of gas came from carbon monoxide, methane and light hydrocarbon gas whereas hydrogen and carbon monoxide was apparent for heating value of gasification. Although the heating value of C_2H_y from tar cracking of pyrolysis was 5 times larger than hydrogen but the energy yield of gasification overcame pyrolysis especially when more H_2 was produced at optimized condition. The produced gas produced in pyrolysis condition might be suitable for furfure thermal conversion process, however, high performance second stage of tar removal is required whereas the gas from steam gasification may be more beneficial in secondarily continual process e.g. Fischer-Tropsch synthesis. The smallest energy output of pyrolysis and gasification conditions were observed at temperature 700°C as 4,891 and 7,638 kJ/kg$_{MSW}$, respectively. The maximum energy output of both pyrolysis and gasification conditions were obtained from the reaction temperature 900°C. The former was 14,059 kJ/kg$_{MSW}$ with energy conversion efficiency

57.26% whereas the latter offered energy output 17,756 kJ/kg$_{MSW}$ with energy conversion efficiency 72.32%.

4. Conclusion

Surrogate MSW was represented as Thailand municipal solid waste to utilize as feedstock for gasification. Steam agent was supplied into reaction for the main purpose of producing hydrogen-rich gas. The experiment studied the effect of temperatures and steam quantities on producer gas quality. In pyrolysis condition, producer gas concentration trended to increase when temperature was increased by thermal decomposition. When steam gasification was conducted, H$_2$ yield drastically increased from pyrolysis condition although the reaction temperature was lowest with the minimal steam flow rate. The highest H$_2$ yield as 34.84 g$_{H_2}$/kg$_{MSW}$, was obtained from test reaction temperature 800°C with steam flow rate 0.2 ml/min. At temperature 900°C, H$_2$ slightly decrease since higher temperature and too much steam amount accelerated the residence time of condensable hydrocarbons. Since most of the related chemical reactions are endothermic, by the same steam flow rate, increasing of temperature resulted in higher concentration all gaseous species by steam reforming of tar except of CO$_2$ which found to be decreased by revered water gas-shift. However, when steam flow rate was increased in the same reaction temperature, CO and CH$_4$ decreased but CO$_2$ increased due to the performing of water gas-shift reaction and steam reforming of methane.

The overall performance of test condition was evaluated by energy output and energy conversion efficiency. Pyrolysis gas consisted mainly of CH$_4$ and C$_2$H$_y$ where gasification mostly consisted of H$_2$ and CO. Although C$_2$H$_y$ contains energy per unit volume 5 times higher than H$_2$ but the energy output of gasification condition offered higher heating value than pyrolysis condition starting from the lowest operation temperature. Producer gas of pyrolysis condition might be suitable for thermal conversion, however, high performance tar removal unit is required whereas producer gas from steam gasification can be applied in various purposes. The maximum energy output and energy conversion efficiency of pyrolysis condition was offed by temperature 900°C as 14,059 kJ/kg$_{sample}$ and 57.26%, respectively while gasification condition were offered by the same reaction temperature with steam flow rate 0.2 ml/min as 17,756 kJ/kg$_{sample}$ and 72.32%, respectively.

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