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Effect of temperature and steam on hydrogen production and producer gas quality from MSW steam gasification

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Abstract. Thermal conversions of municipal solid waste (MSW) are sustainable solution in waste treatment since the amount of MSW trends to increase every year. Compared to other biochemical and physiochemical processes, their main advantages are strong reduction of solid waste in mass and volume by rapid conversion. Hence, thermal treatment preserves landfill space and time including labour expenses. In 2015, approximately 26.9 million tons of MSW was generated in Thailand but just twenty percent of those amount was sanitary disposal whereas only 150k tons was utilized in heat and electricity by thermal conversion. This paper proposes a high efficiency steam gasification as a selected thermal technology to recover energy from carbonaceous waste into form of combustible gas which is mainly hydrogen. Small dropped tube fixed bed reactor was represented as experimental gasifier. The average compositions of Thailand MSW consisting of 55%wt. food & kitchen waste, 22.4%wt. plastic (70% PE & 30% PP), 13%wt. paper, 1.4%wt. rubber & leather, 3.1%wt. textile and 5.1%wt. yard waste & biomass were utilized for gasifying feedstock. The experiments were varied by the two steam flow rates with the reaction temperatures 700-850°C to investigate the effect of steam quantity and temperature on hydrogen production and related compositions e.g. methane, carbon monoxide, carbon dioxide and light hydrocarbons. The results indicated the increasing of steam and temperature improved hydrogen production. Hydrogen yield reached 30.0 and 33.0 g_{H₂}/kg_{MSW} when steam flow rate of 0.05 and 0.13 ml/min was supplied at highest reaction temperature, respectively. In addition, energy yield of hydrogen in steam gasification overcame pyrolysis which mostly containing of light hydrocarbon. The optimized condition performed energy output as 16,015 kJ/kg_{sample} and the energy conversion efficiency of the system was 65.2%.

1. Introduction

Municipal Solid Waste (MSW) management has become a crucial issue since the main waste disposal form still employ open dumped site. It is an essential objective, while the need for a complete sustainable energy solution is apparent. There is a pressing need to develop and deploy new alternatives in order to minimize the environmental impacts at all links in the MSW chain. The concept of Waste-To-Energy is

popular for handling disposal of MSW in many countries and it is more attractive when landfill space is limited.

To-date technologies have to deal with the MSW management problems, to be environmental friendly and economical attractive. Gasification is considered as an important sustainable solution in fast conversion of solid fuel with high energy recovery. Typically, air is fed as gasifying agent for partial combustion to produce producer gas e.g. H_2 , CO, CH_4 and C_xH_y . To improve gas quality, oxygen and/or steam are supplied instead or mixing with air into the gasifier to produce higher heating value gas. However, oxygen production unit is very high investment cost, hence, steam generated by waste heat recovery is more attractive. When steam is employed to be an agent, hydrogen-rich gas is produced. Not only higher heating value of gas but tar or aromatic hydrocarbon is also reformed by thermal and steam cracking. It provides one of the most competitive means of obtaining hydrogen-rich gas from renewable sources compared to other hydrogen productions such as methanol production by fermentation or methane reforming.

This paper presents experimental research conducted of gasification in a small dropped tube fixed bed. MSW was fed as feedstock considering only combustible materials. However, its compositions were not constant according to the different of waste management in each area. Therefore, constant fraction of surrogate MSW was adopted in the experiment. Steam was supplied as gasifying agent with two different steam flow rates and three different temperatures were varied. Pyrolysis were conducted parallel with the same temperatures of gasification for basis reference. The influent of steam quantity and varying of temperature on producer gas concentration, hydrogen yield, dry gas yield and energy conversion efficiency were investigated.

2. Material and Methods

2.1 Municipal Solid Waste

The data of waste compositions from several provinces in Thailand has been reviewed. Only combustible materials were considered to be calculated the constant components of solid waste which represented of Thailand MSW. The average wet percentage of surrogate MSW is presented in Table 1 and its proximate and ultimate analysis are presented in Table 2. Each composition was a model sample, for example plastic was totally 22.4% by weight. It comprised of polyethylene (PE) for 70% which came from plastic carrier bag and polypropylene (PP) for 30% which came from plastic glass and plastic packaging for hot food. They were separately prepared by drying and grinding processes. In the experiment, each component had been weighted by its dry percentage and mixed together before compressing into small tablets.

Table 1. Average composition of surrogate MSW (%wt.) [1-4]

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

Proximate analysis (dry basis)	(%wt.)	Ultimate analysis	(%wt.)
Moisture	0.94	C	54.30
Volatile matter	83.65	H	8.34
Fixed Carbon	6.45	N	0.70
Ash	9.91	S	0.48
Lower heating value (kJ/kg)		24.53	

2.2 Experimental procedure

The schematic diagram of test laboratory is demonstrated in Figure 1. Test rig included electric furnace with temperature controller, quartz tube reactor, sample holder, water supply pump, steam generator, flow controller and gas & tar sampling unit. Temperature controllers were installed to monitor temperature through the experiment at five different positions including of (1) steam generator, (2) gas inlet line, (3) gas & tar sampling unit, (4) electric furnace and (5) inside reactor. Steam generator was made from heat pipe tube operating at temperature 300°C. In gasification, water was pumped from reservoir into steam generator then water in liquid phase was totally converted into steam vapor. Nitrogen was inert to carry steam agent into reactor and all experiments were conducted in atmospheric pressure.

The reactor has internal diameter 13.5 mm and 50 cm length. 1.2 g of sample was mixed and compressed into small tablets and after that, they were put inside the sample holder. Flow of carrier gas was set up at 100 ml/min. When the reactor reached designed temperature and remain stable, steam was supplied into reactor at least 15 minutes before sample was dropped. Steam flow rates were set up as 0.05 and 0.13 ml/min with varying of three reaction temperatures as 700, 800 and 850°C. In parallel, pyrolysis experiments were conducted as the same temperatures of gasification. Without steam but only nitrogen was fed at the flow rate of 100 ml/min.

Since sample tablets was being dropped, producer was collected into gas bag. It was collected every 30 minutes until reaction time reached 120 minutes. There were two gas chromatography (GC) machines to analyze producer gas comprising of GC-2014 and GC-14B. H_2 was analyzed by TCD detector while CH_4 and C_2H_6 ($H = 2, 4, 6$) were analyzed by FID detector, Porapak-Q column of GC-2014. Besides, CO_2 was analyzed by TCD detector, Porapak-Q column and CO was analyzed by TCD detector, MS5A column of GC-14B.

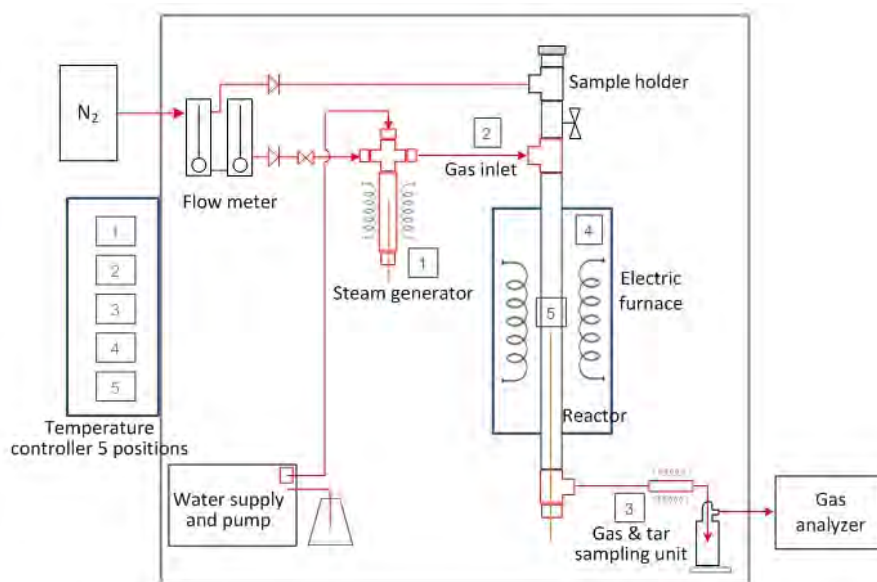


Figure 1. Schematic diagram of test laboratory

3. Results and Discussion

3.1 Producer gas yield and concentration

Gasification were conducted at temperature 700, 800 and 850°C with two steam flow rate at 0.05 and 0.13 ml/min including with no steam in pyrolysis at the same reaction temperatures. The experiment results of dry gas yield and hydrogen yield are demonstrated in Figure 2. The main principle of using steam instead of air or oxygen is to produce hydrogen-rich gas. H_2 yield was reported in term of gram

of hydrogen produced per one kilogram of feedstock and it could observe that H_2 increased twice in steam gasification comparing with pyrolysis [5]. Theoretically, when steam was added into gasification reaction, hydrogen would be more produced by water gas ($C + H_2O \rightarrow CO + H_2$) and water gas shift reaction ($CO + H_2O \rightarrow CO_2 + H_2$). At low temperature, H_2 was less produced compared to higher temperature with same steam amount by endothermic of water gas reaction [6-7]. In pyrolysis, H_2 yield was obtained as 6.4, 11.3 and 13.8 g_{H_2}/kg_{MSW} at reaction temperature 700, 800 and 850°C, respectively, whereas it increased to 28.8, 29.5 and 29.9 g_{H_2}/kg_{MSW} when minimal steam was fed at the same temperature. The maximum hydrogen yield could be observed at steam flow rate 0.13 ml/min with temperature 850°C as 33.0 g_{H_2}/kg_{MSW} . In addition, the same test condition also offered the highest dry gas yield as 0.98 Nm^3/kg_{MSW} while the highest dry gas yield of pyrolysis was 0.80 Nm^3/kg_{MSW} from the same reaction temperature.

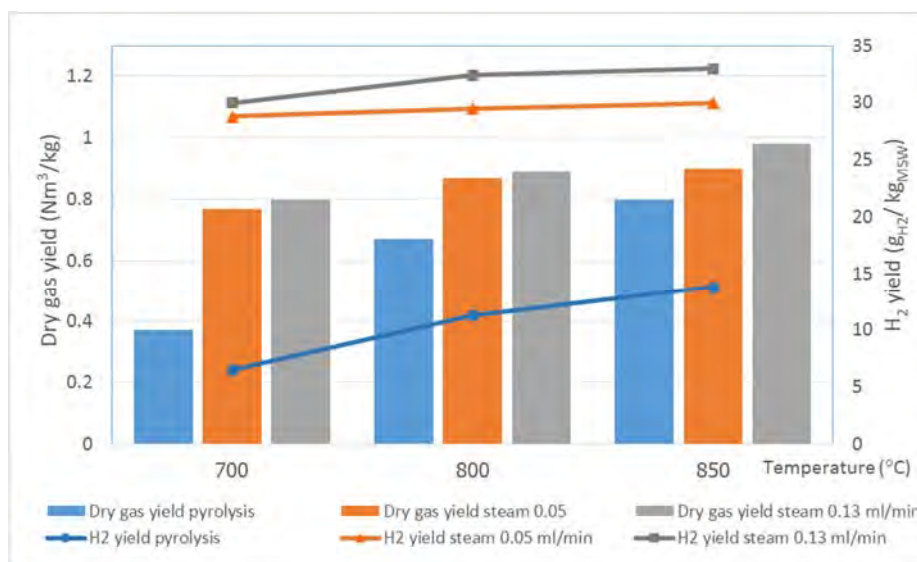


Figure 2. Dry gas yield and hydrogen yield of pyrolysis and gasification

Figure 3. presents concentration of carbon monoxide (a) and carbon dioxide (b) and Figure 4. presents the concentration of CH_4 (a) and C_2H_y (b) in millimole unit. In pyrolysis, 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. C_2H_y in pyrolysis was produced higher than that of gasification. The highest concentration of C_2H_y reached 6.8 mmol/1.2 g_{MSW} at temperature 850°C. In addition, at temperature above 830°C, CO could be produced by Boudouard reaction as can be seen that CO trended to increase when temperature was increased from 700 to 850°C. The optimized condition of pyrolysis offered highest CO concentration as 7.7 mmol/1.2 g_{MSW} . In gasification, light hydrocarbons were generated by thermal cracking of large aromatic hydrocarbon and it was consumed by steam reforming ($C_nH_m + nH_2O \leftrightarrow nCO + (n + m/2)H_2$), hence, trend of C_2H_y of steam gasification was lower than pyrolysis at the same reaction temperature. Besides, methane could be produced by methanation ($CO + 3H_2 \leftrightarrow CH_4 + H_2O$), however, it was reformed by methane steam reforming ($CH_4 + H_2O \leftrightarrow CO + 3H_2$) [8]. Therefore, increasing of steam flow rate resulted in increasing of H_2 and CO_2 but decreasing of CO. Nevertheless, reaction temperature takes more effect than steam quantity because most of related chemical reactions are endothermic. The exposition can be observed by Figure 3 (a) and (b). The concentration of CO_2 was increased from 7.9 mmol/1.2 g_{MSW} to 8.6

mmol/1.2 g_{MSW} when steam flow rate was increased from 0.05 to 0.13 ml/min at temperature 800°C. When the temperature increased to 850°C, CO₂ of the latter was still higher than the former but the trend of CO₂ of both steam flow rates were decreased. They were 6.6 and 7.8 mmol/1.2 g_{MSW} obtained from steam flow rate 0.05 and 0.13 ml/min, respectively. In contrary, concentration of CO of both steam flow rates rose up when the temperature was increased from 800 to 850°C. This caused by Boudouard reaction even though water gas-shift might took place in steam-rich gasification but the influent of high temperature induced endothermic reactions [9].

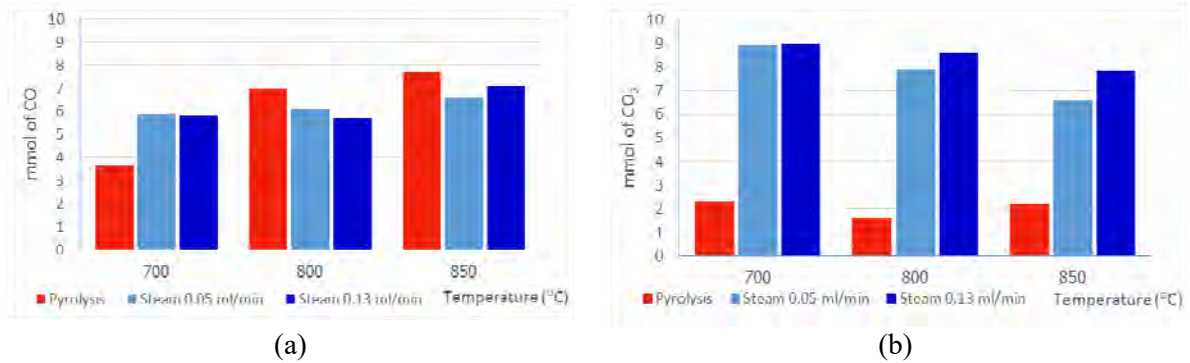


Figure 3. Mole concentration of CO (a) and CO₂ (b) of pyrolysis and gasification

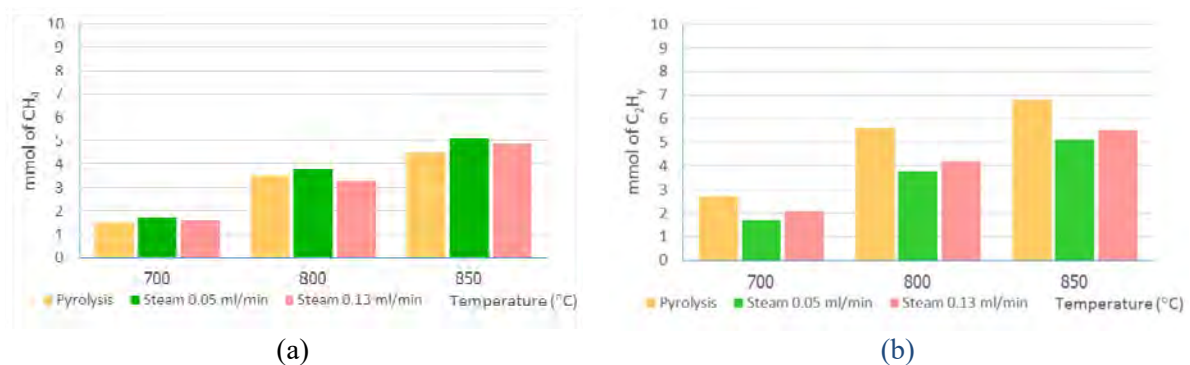


Figure 4. Mole concentration of CH₄ (a) and C₂H_y (b) of pyrolysis and gasification

3.2 Energy conversion efficiency

Energy output of each experiment condition of pyrolysis and gasification was calculated from

$$\text{Total energy output} = \sum \{ \text{volume of producer gas} \times \text{net heating value} \} \text{ Nm}^3/\text{kg}, \quad (1)$$

where producer gas was including of H₂, CO, CH₄, C₂H_y and the calculation was based on one kg gasification of sample.

Similarly, energy efficiency was calculated from

$$\text{Energy conversion efficiency} = \frac{\text{Total energy output per 1 kg of sample}}{\text{Lower heating value of dry basis}} \times 100\%. \quad (2)$$

Trend of energy output and conversion efficiency were plotted in Figure 5. The energy output was a function of temperature as same as mole concentration of producer gas. At temperature 700°C, the overall performance of gasification and pyrolysis was quite low but energy output of pyrolysis and gasification increased 2 times and 1.5 times, respectively, when reaction temperature reached 800°C. In pyrolysis, most of energy output was obtained from gas heating value of CH₄ and light hydrocarbon gas

while in gasification most of energy came from H_2 and CO which were more beneficial in secondarily continual process e.g. Fischer-tropsch synthesis. Pyrolysis performed high energy yield closed to gasification because thermal cracking of plastic produced C_2H_4 which contain higher energy per unit volume of gas 5 times more than H_2 . However, the energy yield of steam gasification overcame pyrolysis especially when more H_2 was produced in higher steam flow rate. The highest energy output of both pyrolysis and gasification were offered from temperature 850°C . The former was $15,186 \text{ kJ/kg}_{\text{MSW}}$ with energy conversion efficiency 61.9% whereas the latter performed energy output $16,025 \text{ kJ/kg}_{\text{MSW}}$ with energy conversion efficiency 65.2%.

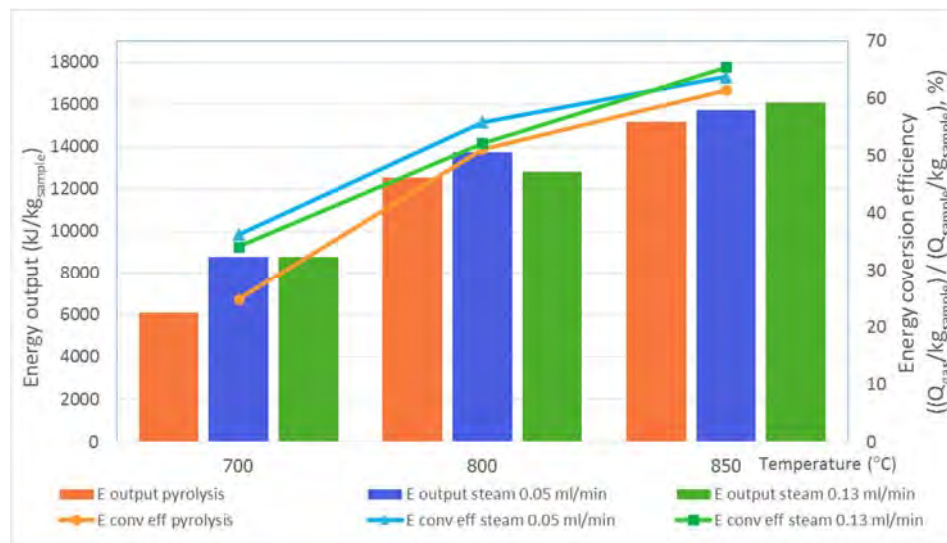


Figure 5. Energy output and energy conversion efficiency of pyrolysis and gasification

4. Conclusion

Steam gasification of municipal solid waste can provide hydrogen-rich gas. However, due to different consumptions and waste disposal managements in different areas, surrogate MSW was represented as combustible waste of Thailand. The experiment studied the effect of temperatures and steam quantities on producer gas quality. In pyrolysis, producer gas concentration trended to increase when temperature was increased by thermal decomposition. When steam gasification was conducted, yield of hydrogen increased drastically from pyrolysis. Hydrogen can be produced by increasing of steam flow rate and temperature since most of the related chemical steam reactions are endothermic. Hence, reaction temperature took more influential in producer gas quality. There were two steam flow rates of 0.05 and 0.13 ml/min conducted in this experiment. Hydrogen yield was optimized by steam flow rate 0.13 ml/min which can be observed 30.0, 32.4 and $33.0 \text{ g}_{H_2}/\text{kg}_{\text{MSW}}$ with reaction temperature 700, 800 and 850°C , respectively. Increasing of reaction temperature, increasing the producer gas concentration. When the temperature was increased from 800 to 850°C , mole of carbon monoxide, methane and light hydrocarbon increased because the related endothermic reactions took place while carbon dioxide decreased by reverse water gas shift reaction.

Pyrolysis gas mostly consisting of light hydrocarbons which contains higher energy per unit value than hydrogen. Although methane and light hydrocarbons were consumed by steam reforming in gasification to produce hydrogen but the energy efficiency yield of gasification overcame pyrolysis. At temperature 800°C , steam flow rate 0.05 ml/min offered the highest energy output as $13,727 \text{ kJ/kg}_{\text{MSW}}$ with energy conversion efficiency 55.9% while steam flow rate 0.13 ml/min offered highest energy output as $16,025 \text{ kJ/kg}_{\text{MSW}}$ with energy conversion efficiency 65.2% at temperature 850°C .

Acknowledgements

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