INTERNATIONAL JOURNAL OF
ENERGY AND ENVIRONMENT

Volume 4, Issue 6, 2013 pp.969-974
Journal homepage: www.IJEE.IEEFoundation.org

Fuel gas and char from pyrolysis of waste paper in a microwave plasma reactor

Parin Khongkrapan, Patipat Thanompongchart, Nakorn Tippayawong, Tanongkiat Kiatsiriroat

Department of Mechanical Engineering, Faculty of Engineering, Chiang Mai University, Chiang Mai 50200, Thailand.

Abstract
In this study, a microwave plasma reactor was used for pyrolysis of waste papers. The effects of different argon flow rates on char and gas generation were investigated. Changes in carbon and oxygen contents from those in paper to char were significant. Char yield of over 25 % was obtained with the heating value of about 38 MJ/kg. Average gas yield and total content of combustible fraction (CO, CH_4 and H_2) in the gas product were 2.56 m^3/kg and 36 %, respectively. The heating value of gas product and carbon conversion efficiency of the process were maximum at 6.0 MJ/m^3 and 73 %, respectively.

Keywords: Biomass; Plasma; Pyrolysis; Renewable energy; Solid waste.

1. Introduction

Concerns over depleting fossil resources and the global climate change have motivated active research on clean energy. Waste-to-energy conversion is an attractive option. It is safe and cost effective. Many studies focus on finding a way to convert waste into energy economically, efficiently and friendly to the environment. Various methods have been proposed, such as thermochemical and biochemical conversions. Thermochemical conversion includes combustion, pyrolysis and gasification. Pyrolysis is a thermo process in an oxygen starved environment. It is important for transforming wastes to gaseous fuel as well as char. The product gas containing CO, CH_4 and H_2 is a key advantage as it may be utilized as fuel [1-6]. There are many types of pyrolysis reactors, including muffle furnace, tube furnace, fixed bed, fluidized bed, and entrained flow reactors [7, 8]. Typical pyrolysis reactors employ energy input from conventional sources of heat. Alternatively, plasma pyrolysis is becoming of interest because it is relatively easy to control, enables fast heating, and can work effectively at low power consumption. It has high potential for renewable energy generation.

In the past, plasma pyrolysis process usually uses high energy, plasma torch as external heat source. Many types of plasma torch have been used, such as arc plasma (transferred and non-transferred configurations) and radio-frequency (RF) plasma torches [9-16]. These devices generate thermal plasma. For example, Nema et al. [5] used the arc plasma torch to treat hazardous medical waste. It was reported that plasma interrupts the formation of dioxins and kills bacteria. The organic mass of waste can be converted to gaseous, at more than 99% conversion efficiency. Chang et al. [15] investigated pyrolysis of used tires using thermal plasma. The product gas heating value about 4 to 7 MJ/m^3. Many types of biomass were used as a substrate, such as sawdust, agricultural residue and wood. Tang et al. [7]

presented pyrolysis treatment of biomass at different operating pressures in a RF plasma reactor. Fir sawdust was used as the feedstock. The process generated syngas of CO and H_2 with 76 % total on a nitrogen-free basis condition.

For lower energy consumption, non-thermal plasma may be used for pyrolysis of biomass and waste. There are many ways to generate non-thermal plasma. Microwave plasma is one of them. It can be generated as thermal or non-thermal depending on power supplied. For a simple setup, a 2.45 GHz magnetron available from a commercial microwave oven may be used [11, 17, 18]. Majority of microwave plasma used for pyrolysis process in previously published works were from microwave plasma torch. Lupa et al. [9] investigated the effect of elemental composition of the feedstock and reaction time on syngas evolution using microwave-induced plasma pyrolysis of commercial and industrial waste. They found that feedstock with high oxygen composition increased the heavier gas species in product gas composition, such as CO, CO_2, and H_2O, as a result of oxidation. Gas evolution was found to peak at approximately 200 s of reaction time. The heating value was determined to range from 11.39 to 17.44 MJ/m^3.

Pyrolysis of waste using non-thermal plasma is not yet widely investigated. There are few studies on using microwave plasma assisted reaction to generate fuel gas and char. In this study, investigation on effect of carrier gas flow rate on the evolution, composition of fuel gas obtained from microwave plasma pyrolysis was carried out. Gas and char yields, carbon conversion efficiency, and heating values were evaluated. The microwave oven was modified to provide non-thermal plasma [19]. The plasma generated was contacted directly to the feedstock and converted it into the fuel gas and char.

2. Materials and method

2.1 Feedstock

Waste paper was the major component of combustible fraction of solid waste, accounting for about one third of typical municipal solid wastes [6]. Waste paper is combustible, and has low contents of nitrogen and sulfur. It can be converted into gaseous fuel with less environmental pollution. It may have sufficient feedstock for waste-to-energy utilization. The samples of waste papers were obtained locally from the same source. The papers were wetted, mashed and compressed into a cylindrical mold with diameter of 20 mm and length of 40 mm. After drying in an oven at 60^{o}C for 48 h, the mass of each compressed paper was about 5 ± 0.1 g.

2.2 Pyrolysis reactor

Experimental setup used in this study is shown schematically in Figure 1. The microwave plasma system in this work was modified from a commercial microwave oven. Argon was used as the carrier gas. The igniter was added into the quartz reactor tube to initiate plasma when argon was excited. Sample of feedstock was placed inside a quartz reactor tube using stainless steel wire in a hang mode. Before each experimental run, the plasma system was vacuumed. Argon was fed from the bottom of the reactor tube. Flow rate was varied from 0.50 to 1.25 lpm. The microwave output power was supplied continuously at 800 W for 3 min. Successful establishment of argon plasma can be visually inspected from characteristically bright light emission. Product gas flowed to the top of the reactor tube, passing to a gas treatment unit and a sampling bag. The sampled gas was then sent for gas chromatographic analysis. The solid residue left in the reactor was carefully collected and weighed. The tests was repeated at least three times for each conditions.

2.3 Analysis

Analyses of waste papers and char from plasma assisted pyrolysis were carried using thermogravimetric and elemental analysis methods. The thermogranimetric analysis of solid sample was carried out using a Perkin Elmer, model TGA7 instrument. A sample was heated from 50 ^{o}C to 135 ^{o}C at a constant heating rate of 10 ^{o}C/min in a nitrogen stream, and held at this temperature for 5 min. After which, the sample was heated up to 900 ^{o}C at a constant heating rate of 100 ^{o}C/min in the same carrier gas condition. At 900 ^{o}C, the sample was reacted under oxidative environment for 20 min. Ultimate analysis of the sample and its char were carried out by dynamic flash combustion method using a Thermo Quest, model Flash EA 1112 Series, CHNS-O analyzer. The analyzer consists of two columns, the multi separation column SS 2 m long; 6 x 5 mm diameter for C, H, N and S analysis, and the oxygen separation column SS 1 m long; 6 x 5 mm diameter for O analysis, respectively.

The sampled product gas was also analyzed for its composition using a Shimadzu model GC-8A gas chromatography fitted with a ShinCarbon ST Micropacked. The analyzer consists of a column and a thermal conductivity detector for measuring the molar fraction of H_2, O_2, N_2, CH_4, CO and CO_2, respectively. The Restek model 14 Scotty analyzed gases were used as standard gas for quantitative calibration. The char yield, gas yield, carbon conversion, and heating values were evaluated. The char yield is defined as mass of char obtained after pyrolysis divided by mass of feedstock. The gas yield is defined as a ratio between total volume of product gas generated from the biomass (m^3) and the reacted mass of original feedstock (kg). Carbon conversion efficiency is defined as a ratio between carbon produced and carbon supplied. Overall heating value of the product gas is a summation of corresponding heating values of the combustible fractions (CO, CH_4 and H_2). The higher heating value (HHV) of the char was calculated from carbon and hydrogen contents [20, 21] as

$$HHV = 0.491(C) + 0.261(H) - 2.907 \quad (1)$$

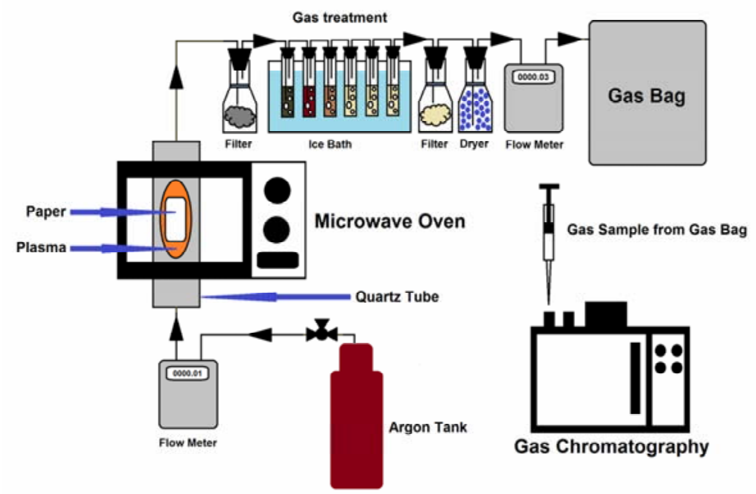

Figure 1. Experimental setup of the microwave plasma reactor for pyrolysis of waste paper

3. Results and discussion

3.1 Raw material and char

Proximate and ultimate analyses of the waste paper and char are shown in Table 1. The proximate analysis gives the properties of the sample paper in mass concentration of moisture, volatile matter, fixed carbon and ash. Release of volatile matter was found to be about 69% after microwave plasma pyrolysis process which was indicative of high conversion of biomass to gaseous fuel. The fixed carbon of the char was found to significantly increase, compared to that original in the starting material. Significant degree of carbonization appeared to take place under non-thermal plasma environment. The ultimate analysis gives the compositions in mass concentration of carbon, hydrogen, oxygen, nitrogen and sulphur. Increase in carbon content in the resulting char was inline with proximate analysis result. Hydrogen and oxygen contents were reduced. The reductions were about 80% and 77%, respectively. The volatile and char yields were listed in Table 2. They were in range between 73-74%, and 25-26%, respectively. Average char yield and its HHV were 25.95 % and 38.5 MJ/kg, respectively.

3.2 Gas evolution

The product gas obtained from the reaction was collected and measured. Major gas components generated were CO, H_2, CH_4, CO_2 and O_2. The most important gas species to consider for pyrolysis processes were CO, CH_4, and H_2. Figure 2 shows effect of carrier gas flow rate on combustible gas fractions of the product gas. Within the range of flow rates considered, CO and CH_4 were not found to vary significantly with argon flow. They remained relatively stable at 16-18% and 2-4%, respectively. H_2 appeared to exhibit more pronounced change with respect to carrier gas flow rate. It was found to initially increase with increasing argon flow, reaching maximum value of about 22% at flow rate of 0.75 lpm. After which, it was markedly reduced at higher carrier gas supply rate. The total content of combustible fractions in the product gas was between 28-44%. Table 2 also shows the gas yields, carbon conversion and heating values of product gas. The heating value was found to be in the range between 3.85 and 6.02 MJ/m^3, showing similar pattern to change in H_2 content with argon flow rate. Average gas yield and carbon conversion efficiency obtained were 2.56 m^3/kg and 70.3 %, respectively. Although present in the detected product gas, other detected fraction would not be taken into account due to their low contents. A majority was undesirable tars.

Table 1. Proximate and ultimate analyses of waste paper and its char

	Waste paper	Char
Proximate analysis (%wt)		
Moisture	3.19	1.96
Volatile matter	83.19	14.43
Fixed Carbon	4.53	54.96
Ash	9.09	28.65
Ultimate analysis (%wt)		
C	43.54	83.55
H	6.24	1.62
O	50.16	14.81
N	0.06	0.01
S	0.00	0.00
HHV (MJ/kg)	20.10	38.53

Table 2. Gas and char products

Argon flow rate (lpm)	Gas heating value (MJ/Nm^3)	Gas yield (Nm^3/kg)	Carbon conversion (%)	Char yield (%)
0.50	4.99	2.08	64.74	25.83
0.75	6.02	2.45	73.16	26.12
1.00	5.07	2.86	72.98	25.92
1.25	3.85	2.86	70.24	25.92
average	4.98	2.56	70.28	25.95

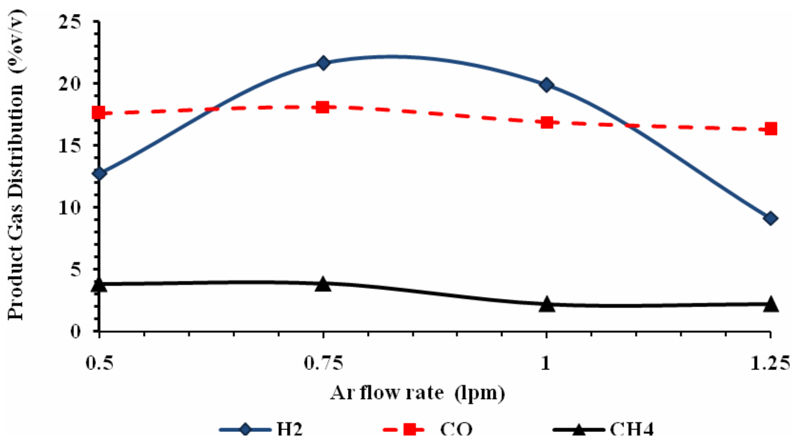

Figure 2. Variation in concentrations of product gas generated with argon flow rates

3.3 Comparison with literature

Product gas and char obtained from plasma pyrolysis of waste paper in this study were compared against those obtained from other types of feedstock and plasma sources and conditions. The comparison is summarized in Table 3. Against non-thermal plasma assisted torrefaction of rice husk and cane residue [22], the char products obtained in this work were in similar magnitude, but with higher energetic content. In comparison with thermal plasma assisted pyrolysis of sawdust [7] and used tires [12], the products obtained in this work showed higher gas yield, but lower char yield.

Table 3. Comparison with literature for plasma assisted pyrolysis of biomass

Ref	source	carrier gas	feedstock	gas product		char	
				gas yield	heating value (MJ/m^3)	char yield	heating value (MJ/kg)
This work	Microwave plasma, 1 atm, 800 W, 3 min	Ar: 0.50-1.25 lpm	paper, 5 g	2.56 m^3/kg	6.02	25.9%	38.5
[22]	Microwave plasma, 250 W, 30 min	N_2: 0.05 lpm	rice husk, 10-12 g	n/a	n/a	33.4%	21.6
[22]	Microwave plasma, 250 W, 15 min	N_2: 0.05 lpm	cane residue, 7-9 g	n/a	n/a	25.5%	27.8
[7]	RF Plasma, 5 kPa, 1.8 kW	N_2: 0.5 lpm	sawdust, 0.3 g/min	0.63 m^3/kg	n/a	33.3%	29.0
[12]	DC arc discharge plasma, 35.2 kVA	N_2: 132 lpm	used tires, 122.5 g/min	1.59 m^3/kg	n/a	69.6%	n/a
[23]	Microwave induced pyrolysis, 3.0 kW	N_2: 3 lpm	wheat straw, 5-30 g	25%	11.5	46.3-56.2%	n/a
[24]	Arc plasma torch, 1 atm, 10 kW	N_2: 1 lpm	rice straw	23.0%	n/a	7.5-13.8%	n/a

4. Conclusion

In this study, microwave plasma pyrolysis of waste paper was investigated. The experimental runs were carried with a 800W microwave power and various argon flow rates. From the findings, increase in proximate and elemental composition of the solid residues was evident as a result of carbonization. Char yield and its HHV were 25.95% and 38.5 MJ/kg, respectively. The maximum combustible fraction (CO, CH_4 and H_2) of the product gas, heating value and carbon conversion were found at 0.75 lpm. The average gas yield and average total content of combustible fraction in the gas product were 2.56 m^3/kg and 36%, which can be used as fuel gas. This is of practical interest for utilization of solid wastes for the purpose of fuel gas production.

Acknowledgements

The authors wish to acknowledge the financial support from the Energy Planning and Policy Office, the Office of Higher Education Commission via the National Research University program, and Chiang Mai University for this research.

References

[1] Hu Z., Ma X., Chen Ch. A study on experimental characteristic of microwave-assisted pyrolysis of microalgae. Bioresource Technology. 2012, 107, 487-493.

[2] Yoon S.J., Lee J.G. Hydrogen-rich syngas production through coal and charcoal gasification using microwave steam and air plasma torch. International J. of Hydrogen Energy. 2012, 37, 17093-17100.

[3] Tippayawong N., Kinorn J. Refuse derived fuel as potential renewable energy source via pyrolysis. International Journal of Renewable Energy. 2007, 2, 45-51.

[4] Li L., Zhang H., Zhuang X. Pyrolysis of waste paper: characterization and composition of pyrolysis oil. Energy Sources. 2005, 27, 867-873.

[5] Nema S.K., Ganeshprasad K.S. Plasma pyrolysis of medical waste. Current Science. 2002, 83, 71-278.

[6] Ahmed I., Gupta A.K. Syngas yield during pyrolysis and steam gasification of paper. Applied Energy. 2009, 86, 1813-1821.

[7] Tang L., Huang H. Plasma pyrolysis of biomass for production of syngas and carbon adsorbent. Energy and Fuels. 2005, 19, 1174-1178.

[8] Tsai W.T., Lee M.K., Chang Y.M. Fast pyrolysis of rice straw sugarcane bagasse and coconut shell in an induction-heating reactor. J. of Analytical and Applied Pyrolysis. 2006, 76, 230-237.

[9] Lupa C.J., Wylie S.R., Shaw A., Al-Shamma A., Sweetman A.J., Herbert B.M.J. Gas evolution and syngas heating value from advanced thermal treatment of waste using microwave-induced plasma. Renewable Energy. 2013, 50, 1065-1072.

[10] Huang H., Tang L. Treatment of organic waste using thermal plasma pyrolysis technology. Energy Conversion and Management. 2007, 48, 1331-1337.

[11] Tendero C., Tixier C., Tristant P., Desmaison J., Leprince P. Atmospheric pressure plasmas a review. Spectrochimica Acta Part B. 2005, 61, 2-30.

[12] Tang L., Huang H. An investigation of sulfur distribution during thermal plasma pyrolysis of used tires. Journal of Analytical and Applied Pyrolysis. 2004, 72, 35-40.
[13] Zhao Z., Huang H., Wu Ch., Li H., Chen Y. Biomass pyrolysis in an argon/hydrogen plasma reactor. Engineering in Life Science. 2001, 1, 197-199.
[14] Gomez E., Amutha Rani D., Cheeseman C.R., Deegan D., Wise M., Boccaccini A.R. Thermal plasma technology for the treatment of wastes: a critical review. Journal of Hazardous Materials. 2009, 161, 614-626.
[15] Chang J.S., Gu B.W., Looy P.C., Chu F.Y., Simpson C.J. Thermal plasma pyrolysis of used old tires for production of syngas. J. of Environmental Science and Health. 1996, 31, 1781-1799.
[16] Beck R.W. Inc. Review of plasma arc gasification and vitrification technology for waste disposal. City of Honolulu final report, January 23, 2003.
[17] Chaichumporn C., Ngamsirijit P., Boonklin N., Eaiprasetsak K., Fuangfoong M. Design and construction of 2.45 GHz microwave plasma source at atmospheric pressure. Procedia Engineering. 2011, 8, 94-100.
[18] Uhm H.S., Hong Y.C., Shin D.H.A microwave plasma torch and its applications. Plasma Sources Science and Technology. 2006, 15, 26-34.
[19] Khongkrapan P., Tippayawong N., Kiatsiriroat T. Thermochemical conversion of waste papers to fuel gas in a microwave plasma reactor. Journal of Clean Energy Technologies. 2013, 1, 80-83.
[20] Moreno J.M.V., Ferre A.J.C., Alonso J.P., Marti B.V. A review of the mathematical models for predicting the heating value of biomass materials. Renewable and Sustainable Energy Reviews. 2012, 16, 3065-3083.
[21] Ouensanga A., Largitte L., Arsene M. A. The dependence of char yield on the amounts of components in precursors for pyrolysed tropical fruit stones and seeds. Microporous and Mesoporous Materials. 2003, 59, 85-91.
[22] Wang M.J., Huang Y.F., Chiueh P.T., Kuan W.H., Lo S.L., Microwave-induced torrefaction of rice husk and sugarcane residues. Energy. 2012, 37, 177-184.
[23] Zhao X., Wang M., Liu H., Li L., Ma C., Song Z. A microwave reactor for characterization of pyrolyzed biomass. Bioresource Technology. 2012, 104, 673-678.
[24] Shie J.L., Tsou F.J., Lin K.L., Chang Ch.Y. Bioenergy and products from thermal pyrolysis of rice straw using plasma torch. Bioresource Technology. 2010, 101, 761-768.

Parin Khongkrapan graduated with B.Eng. degree in agricultural machinery engineering and M.Eng. degree in mechanical engineering from Chiang Mai University, Chiang Mai, Thailand in 2005, and 2008 respectively. He was a lecturer at Rajamangkala University of Technology Lanna. He is currently working towards his Ph.D. in sustainable energy at Chiang Mai University.
E-mail address: parin.khongkrapan@gmail.com

Patipat Thanompongchart graduated with B.Eng. degree in agricultural engineering from Maejo University Chiang Mai, Thailand in 2007 and M.Eng. degree in mechanical engineering from Chiang Mai University, Chiang Mai, Thailand in 2009 respectively. He is currently working towards his Ph.D. in sustainable energy at Chiang Mai University.
E-mail address: pakorn_t13@hotmail.com

Nakorn Tippayawong received his B.Eng. degree in mechanical engineering and Ph.D. degree in internal combustion engines from Imperial College London, UK in 1996 and 2000, respectively. He is currently an associate professor at the Department of Mechanical Engineering, Faculty of Engineering, Chiang Mai University, Chiang Mai, Thailand. His research interests include biomass utilization, energy efficiency improvement, and emission control. So far, he has published more than 70 papers in peer reviewed international journals. He also serves as an editor, an associate editor and an editorial board member of several leading international journals. E-mail address: n.tippayawong@yahoo.com

Tanongkiat Kiatsiriroat received his B.Eng. and M.Eng. degrees in mechanical engineering from Kasetsart University and King Mongkut University of Technology Thonburi, Thailand, and Ph.D. degree in energy technology from Asian Institute of Technology, Bangkok, Thailand, respectively. He is currently a professor at the Department of Mechanical Engineering, Chiang Mai University. His research interests include thermal engineering, energy technology, and life cycle assessment. He has over 300 papers in peer reviewed national and international publications. Prof. Kiatsiriroat serves as an associate regional editor of *Applied Thermal Engineering*, as well as an editorial board member of various leading international journals. E-mail address: tanong@dome.eng.cmu.ac.th