APPENDICES

APPENDIX A

Calculations Examples

A.1 Raw Data for Calculation Examples

The calculation examples were used raw data of waste paper as follow;

Table A.1 Material properties for calculation examples

Properties	Waste paper	
	Raw	Char
Proximate analysis (% wt)		
Moisture	3.19	1.96
Volatile	83.19	14.43
Fixed carbon	4.53	54.96
Ash	9.09	28.65
Ultimate analysis (% wt)		
Carbon	43.54	83.55
Hydrogen	6.24	1.62
Oxygen	50.16	14.81
Nitrogen	0.06	0.01
Sulphur	0	0

Table A.2 Waste paper experimental data

Flow rate (lpm)	Weight (g)		Product gas (liter)
	Feedstock	Char	
0.50	5.08	1.12	5.25
0.75	5.12	0.98	6.50
1.00	5.04	1.08	7.00
1.25	5.11	1.09	7.25

Table A.3 Product gas composition

Feed stock	Gas composition (%mol)	Argon flow rate (lpm)				
		0.50	0.75	1.00	1.25	Average
Waste paper	H2	22.05	23.81	23.52	21.38	22.69
	CO	57.91	58.38	58.39	57.65	58.08
	CO2	14.71	12.15	12.70	15.72	13.82
	CH4	3.38	4.36	3.98	3.05	3.69
	O2	1.94	1.29	1.41	2.20	1.71
	Sum	100	100	100	100	100
	Combustible fraction	83.35	86.56	85.88	82.09	84.47

A.2 Calculation Examples

A.2.1 Product Gas Analysis and Conversion

Gas mixture standard was analyzed by GC and collected the chromatogram area for the mixture gases. The area was the average value from five experiments. The conversion ratio can be calculated as follow;

$$Conversion\ ratio = Gas\ standard\ (\%mol)/GC\ area \tag{A.1}$$

The example gas standard composition and conversion ratio were shown in Table A.4.

Table A.4 Gas standard composition and conversion ratio

Gas composition	Gas standard (%mol)	Gas standard GC area	Conversion ratio
H2	4	159	0.025157233
O2	5	8808	0.000567666
N2	5	11696	0.000427497
CO	5	8937	0.000559472
CH4	4	6298	0.000635122
CO2	5	9893	0.000505408
He	72	-	-
Total	100	-	-

A.2.2 Lower Heating Values

Lower heating values (LHV) of the product gas was defined as the summation of corresponding heating values of H_2, CO and CH_4 (Tendero et al., 2005);

$$LHV = [(25.7 \times H_2) + (30 \times CO) + (85.4 \times CH_4)] \times 4.2 \tag{A.2}$$

For waste paper, the LHV of product gas at argon flow rate of 0.75 lpm can be calculated as follow;

$$LHV = [(25.7 \times 23.81) + (30 \times 58.38) + (85.4 \times 4.36)] \times 4.2$$

$$LHV = 11489.78 \frac{kJ}{m^3} \; or \; 11.49 \; \frac{MJ}{m^3}$$

A.2.3 Gas Yield

Gas yield was calculated from the volume of total gas generated and reacted original mass of feedstock as (Lv et al., 2004);

$$Y_{gas} = Q_{total}/m_f \tag{A.3}$$

where, Q_{tatal} is volume of total gas generated (Nm^3) and m_f is original mass of feedstock (kg).

For waste paper, the gas yield at argon flow rate of 0.75 lpm can be calculated as follow;

$$Y_{gas} = 6.50/5.12$$

$$Y_{gas} = 1.27 \ \frac{Nm^3}{kg}$$

A.2.4 Char Yield

Char yield is defined as the ratio of the mass of obtained char and original mass of feedstock;

$$Y_{char} = \left(m_{char}/m_f\right) \times 100\% \tag{A.4}$$

where, m_{char} is mass of obtained char (kg) and m_f is original mass of feedstock (kg).

For waste paper, the char yield at argon flow rate of 0.75 lpm can be calculated as follow;

$$Y_{char} = (0.98/5.12) \times 100\%$$

$$Y_{char} = 19.14\%$$

A.2.5 Higher Heating Value

The higher heating value (HHV) of the solid material was calculated from its contents as (Moreno et al., 2012);

$$HHV = 0.491(C) + 0.261(H) - 2.907 \tag{A.5}$$

where, C and H are respective concentrations of carbon and hydrogen (% w/w) in material from ultimate analyses.

The HHV of waste paper can be calculated as follow;

$$HHV = 0.491(43.54) + 0.261(6.24) - 2.907$$

$$HHV = 20.1\ \frac{MJ}{kg}$$

A.2.6 Carbon Conversion Efficiency

Carbon conversion efficiency is defined as a ratio between carbon in gaseous fuel and carbon in feedstock;

$$\eta_C = \frac{Q_{gas}.[(CO+CO_2+CH_4)/100].(12/22.4)}{m_f.[1-(A_f/100)].(C_f/100)} \times 100\% \qquad \text{(A.6)}$$

where, Q_{gas} is volume of product gas generated (Nm^3), m_f is original mass of feedstock (kg), m_{char} is mass of obtained char (kg), A_f is ash content in feedstock (% w/w) and C_f is carbon content in feedstock (% w/w).

For waste paper, the carbon conversion efficiency at argon flow rate of 0.75 lpm can be calculated as follow;

$$\eta_C = \frac{4.25 \times [(58.39 + 12.16 + 4.36)/100] \times (12/22.4)}{5.12 \times [1 - (9.09/100)] \times (43.54/100)} \times 100\%$$

$$\eta_C = 84.14\%$$

A.2.7 Energy Efficiency

In this thesis, energy efficiency was defined as Output energy divided by microwave input energy. Microwave input energy was calculated from microwave input power multiplied by reaction time. Output energy was the energy from gas product. It was calculated from the mass of feedstock (m_f), yield (Y_{gas}) and calorific value (LHV) of product gas. The energy efficiency of plasmochemical conversion was calculated from the following equation;

$$\eta_{energy} = \frac{m_{feed}\,(kg) \times Y_{gas}(m^3/kg) \times LHV(J/m^3)}{microwave\ input\ power\ (W) \times time(s)} \times 100\% \qquad \text{(A.7)}$$

$$\eta_{energy} = \frac{0.00512 \times 1.27 \times 11489780}{800 \times 3 \times 60} \times 100\% = 51.68\%$$

APPENDIX B

Thermodynamic Equilibrium Modeling

B.1 Model Calculation

B.1.1 Formula and calculation data

The equilibrium model in this thesis is formed as;

$$CH_pO_qN_r + wH_2O = n_{CO}CO + n_{CO_2}CO_2 + n_{CH_4}CH_4 + n_{H_2}H_2 + n_{H_2O}H_2O + \frac{r}{2}N_2 \quad \text{(B.1)}$$

- **Given;** $n_{CO} = x_1, n_{CO_2} = x_2, n_{CH_4} = x_3, n_{H_2} = x_4, n_{H_2O} = x_5$

Hence, equation (B.1) can be written as;

$$CH_pO_qN_r + wH_2O = x_1CO + x_2CO_2 + x_3CH_4 + x_4H_2 + x_5H_2O + \frac{r}{2}N_2 \quad \text{(B.2)}$$

The water content (w in equation (B.2)) of raw material is defined as;

$$w = M \times mc/18.015 \times (1 - mc) \quad \text{(B.3)}$$

where, mc is moisture content in raw material (%) and M is calculated from following equation

$$M = (12.0107 \times nC) + (1.0079 \times nH) + (15.994 \times nO) \qquad (B.4)$$

- **Mass Balance;**

Carbon balance; $$n_{CO} + n_{CO_2} + n_{CH_4} - 1 = 0 \qquad (B.5)$$

Hence; $$f_1 = x_1 + x_2 + x_3 - 1 \qquad (B.6)$$

Hydrogen balance; $$4n_{CH_4} + 2n_{H_2} + 2n_{H_2O} - 2w - p = 0 \qquad (B.7)$$

Hence; $$f_2 = 4x_3 + 2x_4 + 2x_5 - 2w - p \qquad (B.8)$$

Oxygen balance; $$n_{CO} + 2n_{CO_2} + n_{H_2O} - w - q = 0 \qquad (B.9)$$

Hence; $$f_3 = x_1 + 2x_2 + x_5 - w - q \qquad (B.10)$$

- **Thermodynamics Equilibrium;**

Modified boudouard reaction can be written as;

$$f_4 = K_1.\left(n_{CO_2}\right)(n_{total}) - (n_{CO})^2 = 0 \qquad (B.11)$$

Hence; $$f_4 = K_1.(x_2)(x_{total}) - (x_1)^2 = 0 \qquad (B.12)$$

Modified water-gas shift reaction can be written as;

$$f_5 = K_2(n_{CO})(n_{H_2O}) - (n_{CO_2})(n_{H_2}) \quad \text{(B.13)}$$

Hence; $$f_5 = K_2(x_1)(x_5) - (x_2)(x_4) \quad \text{(B.14)}$$

The value of equilibrium constants K_1 and K_2 are found at constant temperature and pressure using the standard state Gibbs function of change (ΔG_T^O).

$$\ln(K_i) = -\frac{\Delta G_T^O}{RT} \quad \text{(B.15)}$$

$$\Delta G_T^O = \sum_i v_i \, . \, \Delta g_{f,T,i}^O \quad \text{(B.16)}$$

A standard Gibbs function $(\Delta g_{f,T,i}^O)$ of formation of CO, CO_2, CH_4, and H_2O at any temperature can be calculated using the following equation;

$$\Delta g_{f,T}^O = h_f^O - a'T\ln(T) - b'T^2 - \left(\frac{c'}{2}\right)T^3 - \left(\frac{d'}{3}\right)T^4 + \left(\frac{e'}{2T}\right) + f' + g'T \quad \text{(B.17)}$$

where, $a', b', c', d', e', f' \, and \, g'$ are the coefficients of empirical equation, and h_f^O is the enthalpy of formation of gases, are presented in Table. B.1.

Table B.1 The value of $\boldsymbol{h_f^O}$ (kJ/kmol) and coefficients of empirical equation for $\Delta \boldsymbol{g_{f,T}^O}$

Compound	h_f^O	a' (x10^{-3})	b' (x10^{-6})	c' (x10^{-9})	d' (x10^{-12})	e' (x10^{2})	f' (x10^{-1})	g' (x10^{-2})
CO	-110.5	5.619	-11.900	6.383	-1.846	-4.891	8.684	-6.131
CO_2	-393.5	-19.490	31.220	-24.480	6.946	-4.891	52.700	-12.070
CH_4	-74.8	-46.200	11.300	13.190	-6.647	-4.891	141.100	-22.340
H_2O	-241.8	-8.950	-3.672	5.209	-1.478	0.000	28.680	-1.722

(Source; Syed et al., 2012)

The value of ΔG_T^O is calculated by taken the species present in the relevant elementary reactions. The value of $\Delta G_T^O$for boudouard and methanation reaction are formulated as;

- ΔG_T^O for boudouard reaction (Syed et al., 2012);

$$\Delta G_T^O = 2\Delta g_{f,CO}^O - \Delta g_{f,CO_2}^O \tag{B.18}$$

- ΔG_T^O for water-gas shift reaction (Wongsiriamnuay, 2012);

$$\Delta G_T^O = \Delta g_{f,CO_2}^O - \Delta g_{f,H_2O}^O - \Delta g_{f,CO}^O \tag{B.19}$$

B.1.2 Calculation Sample

Input data as following;

T = 1536 K, x0 = [0.1;0.1;0.1;0.1;0.1], mc = 3.19/100, C = 43.54, H = 6.24,

O = 50.16, N = 0.06, S = 0 and, m = 0

Calculation mole of fuel per number of carbon.

- **Given :** nC = 1

nH = (H/1.00794)/(C/12.0107)

nH = (6.24/1.00794)/(43.54/12.0107) = 1.7078 mole/ mole carbon

nO = (O/15.9994)/(C/12.0107)

nO = (50.16/15.9994)/(43.54/12.0107) = 0.8648 mole/ mole carbon

nN = (N/14.0074)/(C/12.0107)

nN = (0.06/14.0074)/(43.54/12.0107) = 0.00118 mole/ mole carbon

nS = (S/32.066)/(C/12.0107)

nS = (0/32.066)/(43.54/12.0107) = 0 mole/ mole carbon

M = 12.0107(nC) + 1.00794(nH) + 15.9994(nO)

M = 12.0107(1) + 1.00794(1.7078) + 15.9994(0.8648) = 27.5683 mole/ mole carbon

W = (M)(mc)/[18.015x(1-mc)]

W = (27.5683)(0.0319)/[18.015x(1-0.0319)] = 0.0504 mole/ mole carbon

Testing condition error < 0.1. If condition is true, program will proceed Gibbs function to calculate $\Delta g^{O}_{f,T}$ (Source; Dow Chemical Co., 1971)

Table B.2 The calculated values of $\Delta \boldsymbol{g^{O}_{f,T}}$ compared with Dow Chemical Co.

$\Delta g^{O}_{f,T}$	Calculate (kJ/kmol)	Dow Chemical Co. (kJ/kmol)	Error (%)
CO	-137,330	-137,150	0.009987
CO_2	-394,420	-394,360	0.009998
CH_4	-50,847	-50,790	0.009989
H_2O	-228,620	-228,590	0.009999

B.2 Programming Code

B.2.1 Main Programming Code

This function was fixed reaction temperature for calculation of product gas composition. The main programming code can be written as follow;

```
%Finding product gas, input temperature in Kelvin.

function f= main_input_temp

%------T=input('enter->T=');%input temperature in Kelvin.
%------x0=input('enter->x0=');
%------mc=input('enter->mc=');%input moisture/100
%------C=input('enter->C=');%input carbon(wt%)
%------H=input('enter->H=');%input hydrogen(wt%)
%------O=input('enter->O=');%input oxygen(wt%)
%------N=input('enter->N=');%input nitrogen(wt%)
%------S=input('enter->S=');%input sulfur(wt%)

T=input('enter->T=');   %input temperature in Kelvin.

%mc=input('enter->mc='); %input moisture/100
```

```
mc=3.19/100; %input moisture/100

x0= [0.1;0.1;0.1;0.1;0.1];

%------m=input('enter->m=');%m=(air*M*0.21)/(22.4*1.187)or input kmol of oxygen
per kmol
m=0;


%------input constants to adjust the model results
c1=1; c2=1; c3=1; c4=1; c5=1;


C = 43.54;      %input carbon(wt%)
H = 6.24;       %input hydrogen(wt%)
O = 50.16;      %input oxygen(wt%)
N = 0.06;       %input nitrogen(wt%)
S = 0;          %input sulfur(wt%)

nC =1;
nH =(H/1.00794)/(C/12.0107);
nO =(O/15.9994)/(C/12.0107);
nN =(N/14.0074)/(C/12.0107);
nS =(S/32.066)/(C/12.0107);

M=(12.0107*nC)+(1.00794*nH)+(15.9994*nO);

w=(M*mc)/(18.015*(1-mc));


save('main_input_temp.mat','x0','mc','C','H','O','N','S','nC','nH','nO','nN','nS','M','w','m',
'c1','c2','c3','c4','c5');

% Gibbs function of reaction
   eval('[Gibb_CO,Gibb_CO2,Gibb_CH4,Gibb_H2O] = Gibb(T)') ;

% The standard Gibbs function of reaction
   eval('[SG_K1,SG_K2] = SG_K(Gibb_CO,Gibb_CO2,Gibb_H2O)');

% The standard Gibbs function of reaction
   eval('[K1,K2] = K (SG_K1,SG_K2,T)');

% modified from example in Numerical Methods in Chemical Engineering
   eval('[solution] = Newton_Raphson(@f,@Jacobain,x0)');

total = solution(1,1)+solution(2,1)+solution(3,1)+solution(4,1)+solution(5,1);
```

```
disp(['total = ' num2str(total)])

CO_percen = c1*(solution(1,1)*100)/total ;
CO2_percen = c2*(solution(2,1)*100)/total ;
CH4_percen = c3*(solution(3,1)*100)/total ;
H2_percen = c4*(solution(4,1)*100)/total ;
H2O_percen = c5*(solution(5,1)*100)/total ;

disp(['nH = ' num2str(nH)])
disp(['nO = ' num2str(nO)])
disp(['nN = ' num2str(nN)])
disp(['nS = ' num2str(nS)])


disp(['nCO = ' num2str(solution(1,1))])
disp(['nCO2 = ' num2str(solution(2,1))])
disp(['nCH4 = ' num2str(solution(3,1))])
disp(['nH2 = ' num2str(solution(4,1))])
disp(['nH2O = ' num2str(solution(5,1))])

disp(['CO_percen = ' num2str(CO_percen)])
disp(['CO2_percen = ' num2str(CO2_percen)])
disp(['CH4_percen = ' num2str(CH4_percen)])
disp(['H2_percen = ' num2str(H2_percen)])
disp(['H2O_percen = ' num2str(H2O_percen)])
save('main_input_temp.mat','CO_percen','CO2_percen','CH4_percen',
'H2_percen','H2O_percen');
```

B.2.2 Mass and Energy Balance Functions Programming Code

```
function [f] =  f(x)
load K.mat;
load('main_input_temp.mat','x0','mc','C','H','O','N','S','nC','nH','nO','nN','nS','M','w','m')
;

% 5 unknowns; CO, CO2, CH4, H2, H2O

%Function balancing

%CH_n_O_m -->nCO+nCO2+nCH4+nH2+nH2O

%nCO->x(1),nCO2->x(2),nCH4->x(3),nH2->x(4), nH2O->x(5)

% Carbon balance
f1 = x(1)+x(2)+x(3)-1;
```

```
% hydrogen balance
f2 = 4*x(3)+2*x(4)+2*x(5)-(2*w)-nH;
% oxygen balance
f3 = x(1)+2*x(2)+x(5)-w-nO;

% equilibrium constant balance 1; Modified boudouard reaction
f4 = (K1*x(2)*(x(1)+x(2)+x(3)+x(4)+x(5)))-((x(1))^2);

% equilibrium constant balance 2; Modified water-gas shift reaction
f5 = (K2*x(1)*x(5))-(x(2)*x(4));

f = [f1;f2;f3;f4;f5];
% end function
```

B.2.3 Standard Gibbs Function of Reaction Programming Code

```
function [SG_K1,SG_K2] =  SG_K( Gibb_CO,Gibb_CO2,Gibb_H2O)
% The standard Gibbs function of reaction

% boudouard reaction
SG_K1 = (2*Gibb_CO) - Gibb_CO2;

% water-gas shift reaction
SG_K2 = Gibb_CO2-Gibb_H2O-Gibb_CO ;

save ('SG_K.mat','SG_K1','SG_K2');
```

B.2.4 Gibbs Function of Reaction Programming Code

```
function [Gibb_CO,Gibb_CO2,Gibb_CH4,Gibb_H2O] = Gibb (T)

% Gibbs function of reaction

Gibb_CO = (-110530) -(((5619)*(10^(-3)))*T*log(T))-(((-1190)*(10^(-5)))*(T^2))...
   -((((6383)*(10^(-9)))/(2))*(T^3))-((((-1846)*(10^(-12)))/(3))*(T^4))...
   +(((-4891)*(10^2))/(2*T))+((8684)*(10^(-1)))+(((-6131)*(10^(-2)))*(T));

Gibb_CO2 = (-393520) -(((-1949)*(10^(-2)))*T*log(T))-(((3122)*(10^(-5)))...
    *(T^2))-((((-2448)*(10^(-8)))/(2))*(T^3))-((((6946)*(10^(-12)))/(3))...
    *(T^4))+(((-4891)*(10^2))/(2*T))+(5270)+(((-1207)*(10^(-1)))*(T));

Gibb_CH4 = (-74850) -(((-4620)*(10^(-2)))*T*log(T))-(((1130)*(10^(-5)))...
   *(T^2))-((((1319)*(10^(-8)))/(2))*(T^3))-((((-6647)*(10^(-12)))/(3))...
   *(T^4))+(((-4891)*(10^2))/(2*T))+((1411)*(10^1))+(((-2234)*(10^(-1)))*(T));
```

```
Gibb_H2O = (-241820) - (((-8950)*(10^(-3)))*T*log(T))-(((-3672)*(10^(-
6)))*(T^2))...
    -((((5209)*(10^(-9)))/(2))*(T^3))-((((-1478)*(10^(-12)))/(3))...
    *(T^4))+((0)/(2*T))+(2868)+(((-1722)*(10^(-2)))*(T));

save ('Gibb.mat','Gibb_CO','Gibb_CO2','Gibb_CH4','Gibb_H2O');
```

B.2.5 Equilibrium Constants Programming Code

```
function [K1,K2] =  K (SG_K1,SG_K2,T)

% Modified boudouard reaction
K1 =(exp(-((SG_K1)/(8.314*T))));

% Modified water-gas shift reaction
K2 =(exp(-((SG_K2)/(8.314*T))));

save ('K.mat','K1','K2');
```

B.2.6 Jacobain Function Programming Code

```
function [J] = Jacobain (x)
load K.mat;
load main_input_temp.mat;

% 5 unknowns, CO, CO2, CH4, H2, H2O;
% 5 equations;

%f1 = x(1)+x(2)+x(3)-1; % Carbon balance

J(1,1) = 1; J(1,2) = 1; J(1,3) = 1; J(1,4) = 0; J(1,5) = 0;

%f2 = 4*x(3)+2*x(4)+2*x(5)-2*w-nH; % hydrogen balance

J(2,1) = 0; J(2,2) = 0; J(2,3) = 4; J(2,4) = 2; J(2,5) = 2;

%f3 = x(1)+2*x(2)+x(5)-w-nO; % oxygen balance

J(3,1) = 1; J(3,2) = 2; J(3,3) = 0; J(3,4) = 0; J(3,5) = 1;

%f4 = K1*x(2)*(x(1)+x(2)+x(3)+x(4)+x(5))-((x(1))^2);
% equilibrium constant balance 1; Modified boudouard reaction
```

```
J(4,1) = K1*x(2)-2*x(1); J(4,2) = K1*(x(1)+2*x(2)+x(3)+x(4)+x(5)); J(4,3) =
K1*x(2); J(4,4) = K1*x(2); J(4,5) = K1*x(2);

%f5 = K2*x(1)*x(5)-x(2)*x(4);
% equilibrium constant balance 2; Modified water-gas shift reaction

J(5,1) = K2*x(5); J(5,2) = (-x(4)); J(5,3) = 0; J(5,4) = (-x(2)); J(5,5) = K2*x(1);

% end function
```

B.2.7 Newton Raphson Method Programming Code

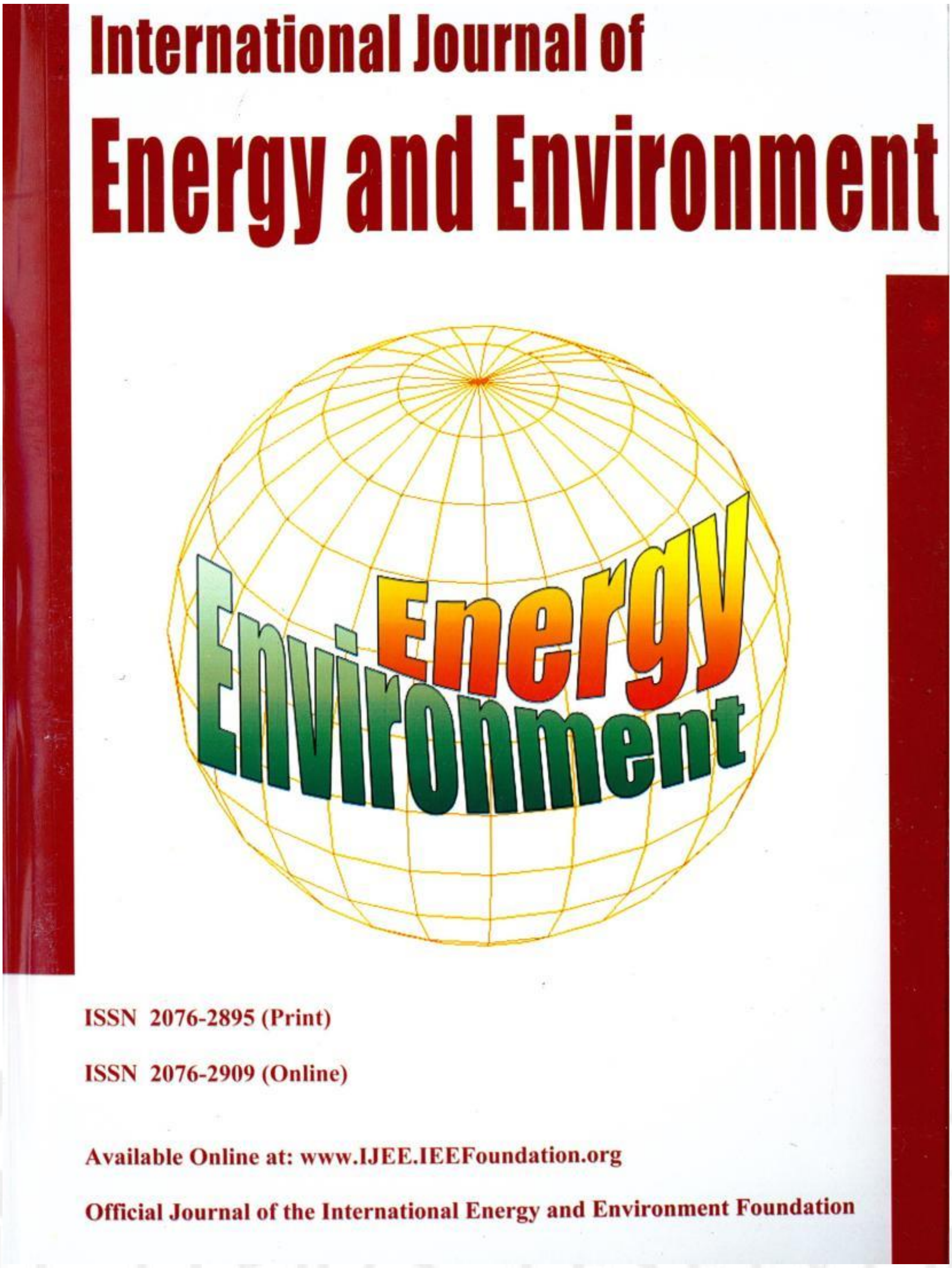
```
% Newton Raphson Method;
% modified from example in Numerical Methods in Chemical Engineering;

function [solution] = Newton_Raphson(f,Jacobain,x0)
x=x0;
error = 2*1e-6;
iter = 0;
while error > 1e-6

   P = feval(f,x);
   P = double(P);
   error1 = max(abs(P(:)));
   J = feval(Jacobain,x);
   dx = J\(-P);
   error2 = max(abs(P(:)));
   m = 1;
   while error2 >=error1||~isreal(P)
      xnew = x+(dx*0.5^m);
      P = feval(f,xnew);
      P = double(P);
      error2 = max(abs(P(:)));
      m = m+1;
   end
   x=xnew;
   error = error2;
   iter = iter+1;
   %disp(['error = ' num2str(error)])
   %disp(['iter= ' num2str(iter)])
end
solution = x ;
save ('Newton_Raphson.mat','solution');
```

APPENDIX C

Publication

C.1 Papers in International Journals

1) Khongkrapan, P., Thanompongchart, P., Tippayawong, N., and Kiatsiriroat, T., (2013). "Fuel gas and char from pyrolysis of waste paper in a microwave plasma reactor", *International Journal of Energy and Environment*, 4(6), 969-974.
2) Khongkrapan, P., Tippayawong, N., and Kiatsiriroat, T., (2013). "Thermochemical Conversion of Waste Papers to Fuel Gas in a Microwave Plasma Reactor", *Journal of Clean Energy Technologies*, 1(2), 80-83.
3) Khongkrapan, P., Thanompongchart, P., Tippayawong, N., and Kiatsiriroat, T., (2013). "Microwave plasma assisted pyrolysis of refuse derived fuels", *Central European Journal of Engineering*, Accepted for publication.
4) Thanompongchart, P., Khongkrapan, P., and Tippayawong, N., (2013). "Partial oxidation reforming of simulated biogas in gliding arc discharge system", *Periodica Polytechnica Chemical Engineering*, Accepted for publication.



International Journal of Energy and Environment

ISSN 2076-2895 (Print)

ISSN 2076-2909 (Online)

Available Online at: www.IJEE.IEEFoundation.org

Official Journal of the International Energy and Environment Foundation

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

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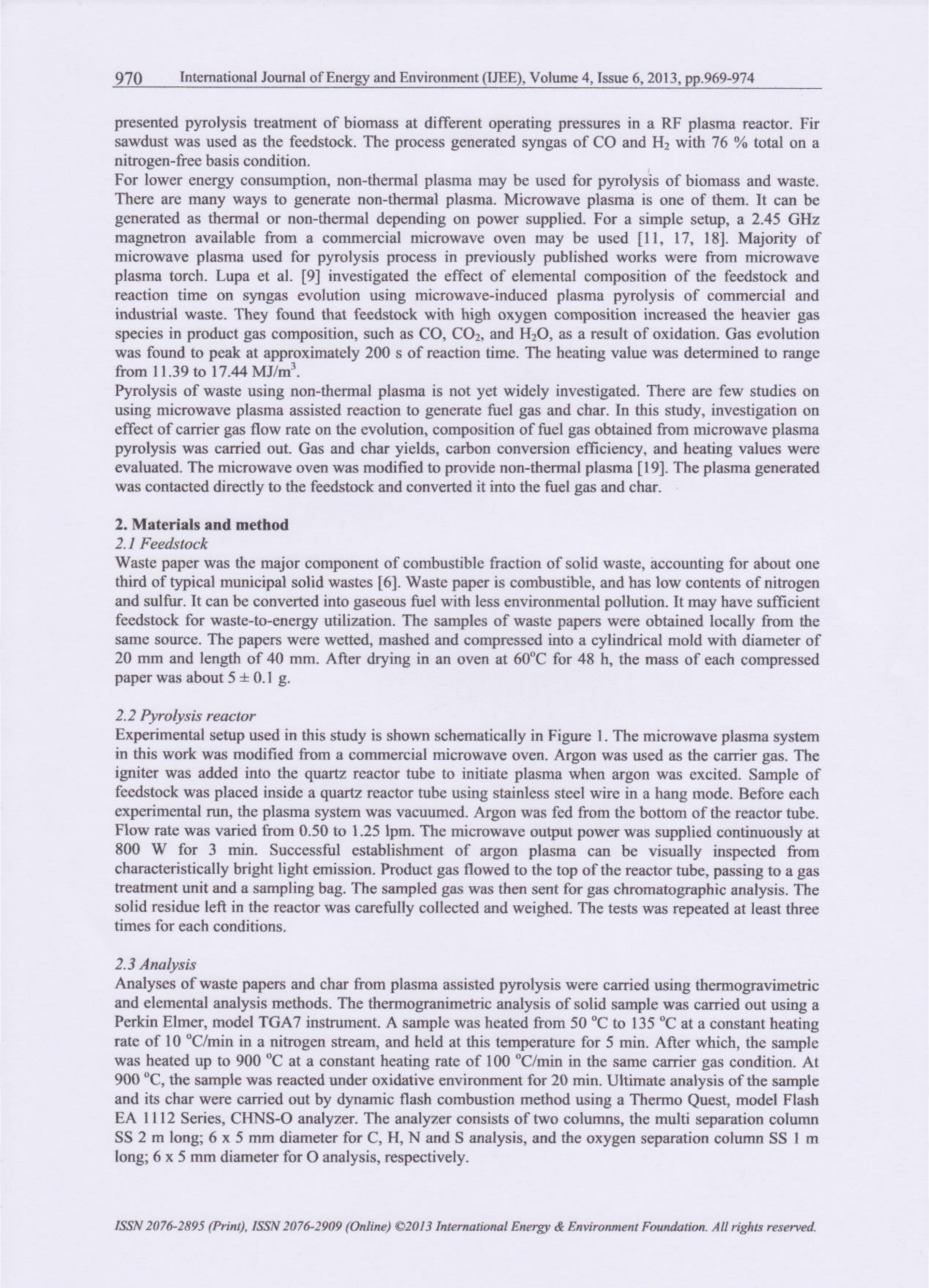
Abstract
In this study, a microwave plasma reactor was used for pyrolysis of waste papers. The effects of different argon flow rates on char and gas generation were investigated. Changes in carbon and oxygen contents from those in paper to char were significant. Char yield of over 25 % was obtained with 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°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 °C to 135 °C at a constant heating rate of 10 °C/min in a nitrogen stream, and held at this temperature for 5 min. After which, the sample was heated up to 900 °C at a constant heating rate of 100 °C/min in the same carrier gas condition. At 900 °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

$$HHV = 0.491(C) + 0.261(H) - 2.907 \tag{1}$$

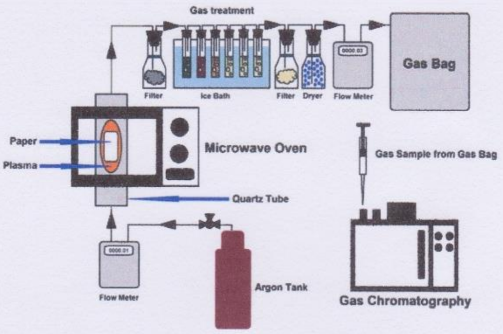


Figure 1. Experimental setup of the microwave plasma reactor for pyrolysis of waste paper

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

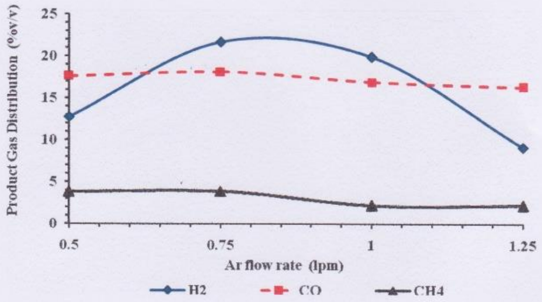


Figure 2. Variation in concentrations of product gas generated with argon flow rates

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.

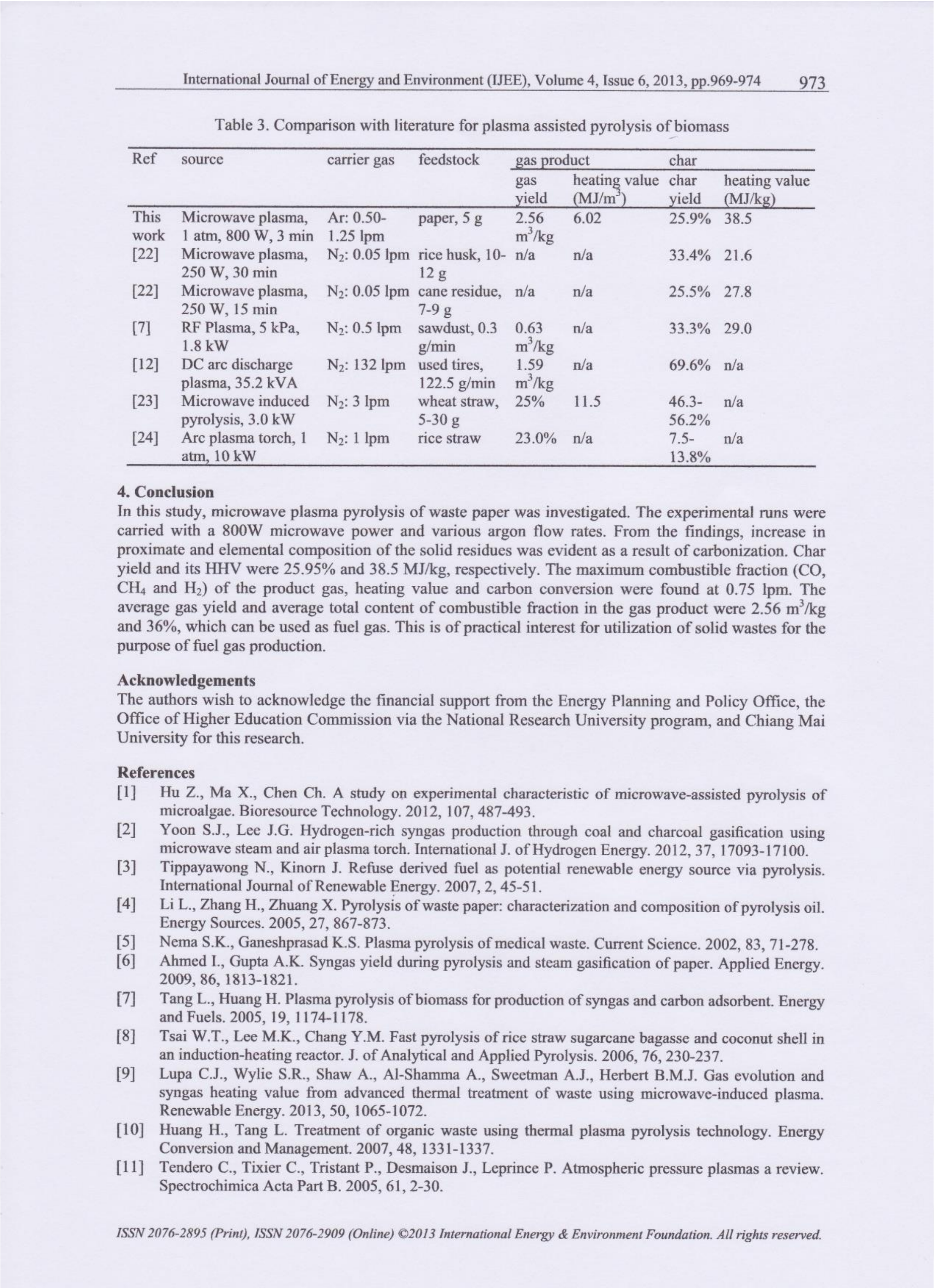


Table 3. Comparison with literature for plasma assisted pyrolysis of biomass

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.

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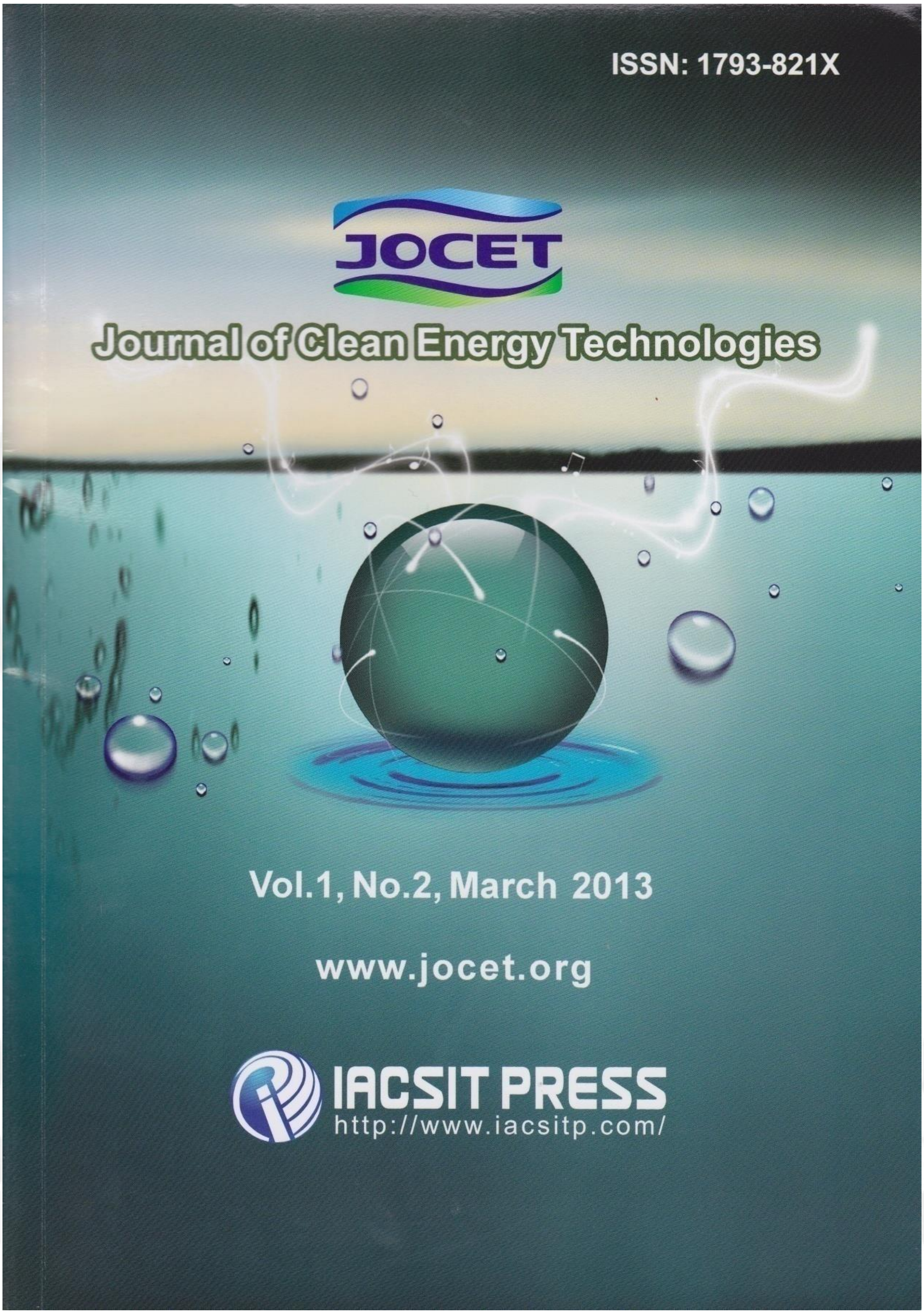

ISSN: 1793-821X

JOCET

Journal of Clean Energy Technologies

Vol.1, No.2, March 2013

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Journal of Clean Energy Technologies

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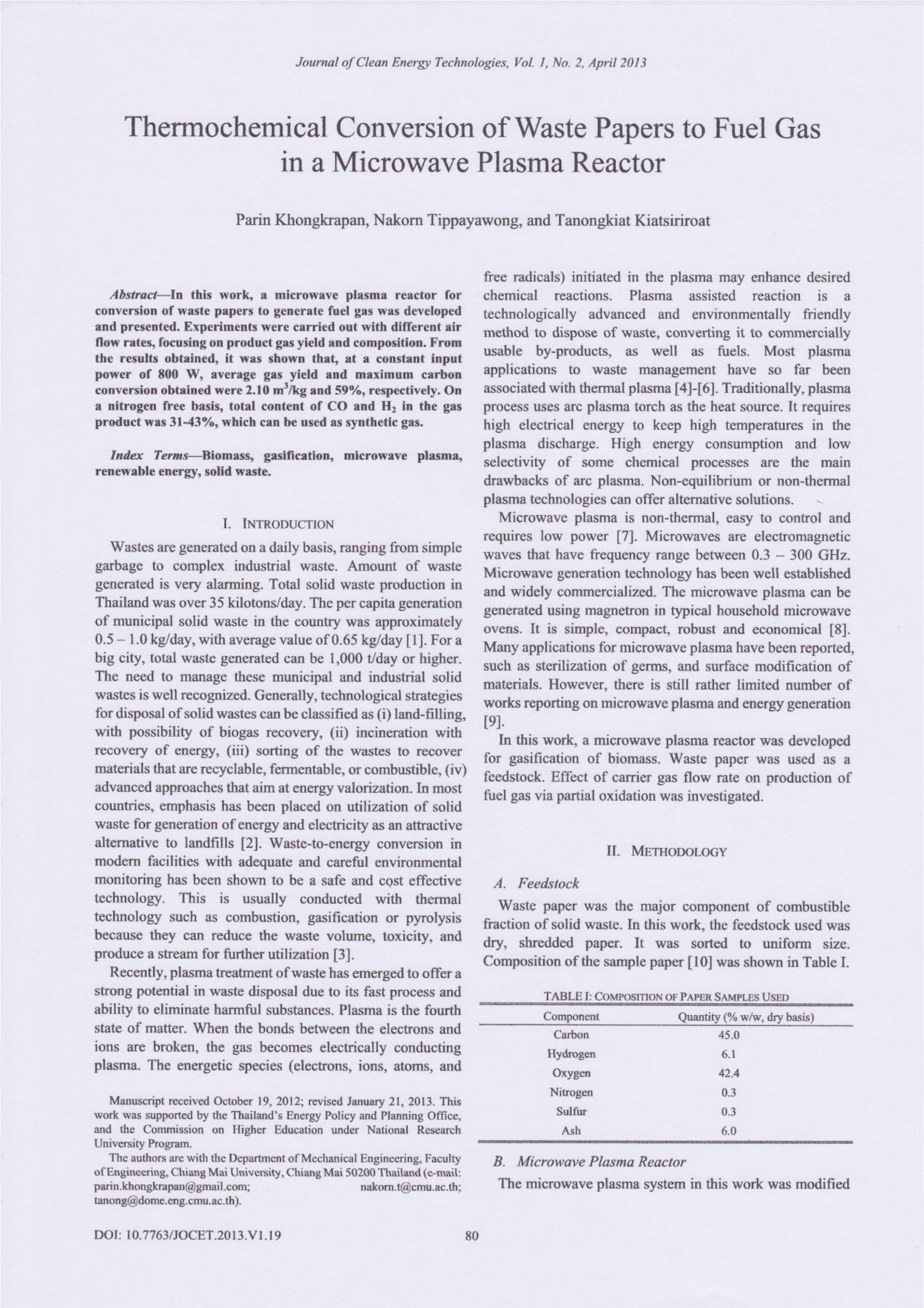

Journal of Clean Energy Technologies, Vol. 1, No. 2, April 2013

Thermochemical Conversion of Waste Papers to Fuel Gas in a Microwave Plasma Reactor

Parin Khongkrapan, Nakorn Tippayawong, and Tanongkiat Kiatsiriroat

***Abstract*—In this work, a microwave plasma reactor for conversion of waste papers to generate fuel gas was developed and presented. Experiments were carried out with different air flow rates, focusing on product gas yield and composition. From the results obtained, it was shown that, at a constant input power of 800 W, average gas yield and maximum carbon conversion obtained were 2.10 m^3/kg and 59%, respectively. On a nitrogen free basis, total content of CO and H_2 in the gas product was 31-43%, which can be used as synthetic gas.**

***Index Terms*—Biomass, gasification, microwave plasma, renewable energy, solid waste.**

I. INTRODUCTION

Wastes are generated on a daily basis, ranging from simple garbage to complex industrial waste. Amount of waste generated is very alarming. Total solid waste production in Thailand was over 35 kilotons/day. The per capita generation of municipal solid waste in the country was approximately 0.5 – 1.0 kg/day, with average value of 0.65 kg/day [1]. For a big city, total waste generated can be 1,000 t/day or higher. The need to manage these municipal and industrial solid wastes is well recognized. Generally, technological strategies for disposal of solid wastes can be classified as (i) land-filling, with possibility of biogas recovery, (ii) incineration with recovery of energy, (iii) sorting of the wastes to recover materials that are recyclable, fermentable, or combustible, (iv) advanced approaches that aim at energy valorization. In most countries, emphasis has been placed on utilization of solid waste for generation of energy and electricity as an attractive alternative to landfills [2]. Waste-to-energy conversion in modern facilities with adequate and careful environmental monitoring has been shown to be a safe and cost effective technology. This is usually conducted with thermal technology such as combustion, gasification or pyrolysis because they can reduce the waste volume, toxicity, and produce a stream for further utilization [3].

Recently, plasma treatment of waste has emerged to offer a strong potential in waste disposal due to its fast process and ability to eliminate harmful substances. Plasma is the fourth state of matter. When the bonds between the electrons and ions are broken, the gas becomes electrically conducting plasma. The energetic species (electrons, ions, atoms, and free radicals) initiated in the plasma may enhance desired chemical reactions. Plasma assisted reaction is a technologically advanced and environmentally friendly method to dispose of waste, converting it to commercially usable by-products, as well as fuels. Most plasma applications to waste management have so far been associated with thermal plasma [4]-[6]. Traditionally, plasma process uses arc plasma torch as the heat source. It requires high electrical energy to keep high temperatures in the plasma discharge. High energy consumption and low selectivity of some chemical processes are the main drawbacks of arc plasma. Non-equilibrium or non-thermal plasma technologies can offer alternative solutions.

Microwave plasma is non-thermal, easy to control and requires low power [7]. Microwaves are electromagnetic waves that have frequency range between 0.3 – 300 GHz. Microwave generation technology has been well established and widely commercialized. The microwave plasma can be generated using magnetron in typical household microwave ovens. It is simple, compact, robust and economical [8]. Many applications for microwave plasma have been reported, such as sterilization of germs, and surface modification of materials. However, there is still rather limited number of works reporting on microwave plasma and energy generation [9].

In this work, a microwave plasma reactor was developed for gasification of biomass. Waste paper was used as a feedstock. Effect of carrier gas flow rate on production of fuel gas via partial oxidation was investigated.

II. METHODOLOGY

A. Feedstock

Waste paper was the major component of combustible fraction of solid waste. In this work, the feedstock used was dry, shredded paper. It was sorted to uniform size. Composition of the sample paper [10] was shown in Table I.

TABLE I: COMPOSITION OF PAPER SAMPLES USED

Component	Quantity (% w/w, dry basis)
Carbon	45.0
Hydrogen	6.1
Oxygen	42.4
Nitrogen	0.3
Sulfur	0.3
Ash	6.0

B. Microwave Plasma Reactor

The microwave plasma system in this work was modified

Manuscript received October 19, 2012; revised January 21, 2013. This work was supported by the Thailand's Energy Policy and Planning Office, and the Commission on Higher Education under National Research University Program.

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DOI: 10.7763/JOCET.2013.V1.19

from a commercial microwave oven. It consists of the 800 W, 2.45 GHz microwave generator, a cylindrical tube reactor, and auxiliary electrical equipment. The microwave radiation generated from the magnetron passed and guided through the oven cavity, and entered the discharge quartz tube centrally located inside the oven cavity. The plasma generated inside the tube was stabilized by injecting a carrier gas, which entered the tube from the bottom as a turbulent flow.

C. Experimental Setup and Procedure

The experimental setup is shown schematically in Fig. 1, equipped with gas cylinders for air and argon, connected to gas flow regulators.

A gas collection module was used for fuel gas conditioning and tar capture. It consists of a series of filter, impinger bottles containing a solvent for tar absorption placed in cold baths, and a moisture trap. Each impinger bottle was filled with approximately 100 ml of isopropanol. The gas flow rates were measured with flow meters. The cool, dry, clean gas was sampled using gas bags and analyzed on a Shimadzu model GC-8A gas chromatography fitted with a ShinCarbon ST Micropacked column and a thermal conductivity detector, for measuring volumetric concentration of H_2, O_2, N_2, CH_4, CO, CO_2. Standard gas mixtures were used for quantitative calibration.

Test and operating conditions were summarized in Table II. Initially, the quartz tube was loaded with shredded paper mass of 5 g, before mounted inside the oven. Carrier gas was fed from the bottom of the reactor, varying between 1 - 4 lpm. The microwave generator was then switched on to start the reaction, and run for 4 min. Gas collection was continually carried out. Solid residues were collected and weighed after each run. Each test was repeated in triplicate. Average values were reported.

D. Data Analysis

The following parameters are calculated [11];
Specific gas yield:

$$SGY = V_{gas} / W_B \tag{1}$$

Carbon conversion efficiency:

$$n_c = \frac{V_{gas} \times \sum x_i \times (12/22.4)}{W_B \times (1 - C_A) \times C_c} \times 100\% \tag{2}$$

Lower heating value:

$$LHV = \sum x_i LHV_i \tag{3}$$

where V_{gas} is total volume of product gas generated, W_B is converted mass of solid material, x_i is volume fraction of fuel component of product gas, C_A is carbon content in residue, C_C is carbon content in biomass, and LHV_i is the corresponding heating values of the gas component.

III. RESULTS AND DISCUSSIONS

A. Reactor Operation

The microwave plasma reactor was operated at atmospheric pressure. The microwave was powered by a 2.45 GHz generator, whose continuous output power was constant at 800 W. The discharge takes place inside a quartz tube with internal/external diameters of 27/30 mm and length of 250 mm, inserted vertically and perpendicularly to the oven cavity. The plasma can be observed in the quartz reactor tube.

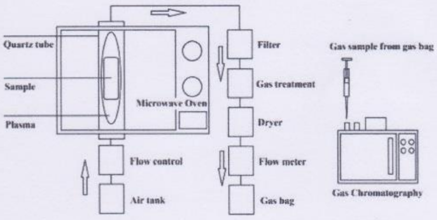

Fig. 1. Schematic of the microwave plasma reactor setup for gasification of waste papers.

TABLE II: OPERATING CONDITIONS OF MICROWAVE PLASMA TESTS

Parameter	Condition
Input power	800 W
Frequency	2.45 GHz
Pressure	101 kPa
Mass of paper	5 g
Air flow rate	1, 2, 3 and 4 lpm
Reaction time	4 min

TABLE III: PRODUCT GAS GENERATED

Air flow rate (lpm)	Gas yields (m^3/kg paper)	Carbon conversion (%)	LHV (MJ/m^3)
1	0.87	19.6	1.86
2	1.71	38.8	1.93
3	2.53	58.6	2.29
4	3.29	51.1	1.56

TABLE IV: PERFORMANCE COMPARISON WITH LITERATURES

Reference	Plasma source	power	Feedstock	Carrier gas	H_2 (%)	CO (%)	CO_2 (%)	CH_4 (%)
This work	Microwave	800 W	Paper, 5 g	Air, 1-4 lpm	7.7	8.5	8.7	1.1
[12]	Microwave	4500 W	Coal, 3-50 kg/h	Air, 0-150 kg/h	2.4	8.1	9.6	1.2
[13]	Microwave	3000 W	Wheat straw, 5-30 g	N_2, 3 lpm	22.1	34.7	33.8	7.9
[14]	Microwave	600 W	Polyethylene, 1 g	0-20% steam/Ar, 3.5 lpm	14	26	12	6
[15]	Microwave	1000 W	Waste wood, 10 g	Ar, 1 lpm	-	56.9	33.8	0.5
[16]	Gliding arc	1140 W	Waste oil, 10 g	10-30% O_2/Ar, 5-16 lpm	-	0.5	2.0	-
[17]	RF, 13.6 MHz	2000 W	Sawdust, 0.3 g/min	N_2, 0.5 lpm	8.5	11	4.0	1.5

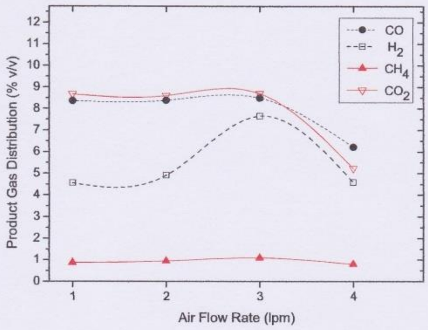

Fig. 2. Variation in concentrations of product gas generated with air flows.

The test run was performed for the microwave plasma system to confirm that the generated plasma can be achieved with our reactor. The plasma was characterized by high intensity light emission. It appeared to occupy most volume of the quartz tube inside the oven cavity.

B. Fuel Gas Production

Table III lists the gas yields, carbon conversion from those in solid material to those present in product gas, and corresponding heating values under different experimental conditions. For all test runs, there was about 1 g of solid residue left in the tube reactor. The specific gas yields obtained were found to increase with air supplied, as expected. Average gas yield was 2.10 m^3/kg paper converted. However, carbon conversion and energy content of the product gas showed initial increase with increasing flow rate, reaching maximum at 3 lpm. Further increase in air supply led to reduction in conversion efficiency and LHV of the product gas. This was contributed to the fact that combustible gas components (CO, H_2 and CH_4) were found to peak at this flow rate, and drop at higher supply rate of carrier gas, as shown in Fig. 2. The observed decline in combustible fractions at higher flow rate may be due to the fact that the flow was too fast inside the reactor, hence, less likely for biomass material and the plasma to react with each other more completely.

C. Comparison with Literature

Product gas obtained from plasmochemical conversion of waste paper in this study was compared against those obtained from other types of biomass and carrier gas in microwave plasma reactors. Results are summarized in Table IV. With air plasma reaction, gas products obtained in this work were comparable to those in the literature.

IV. CONCLUSION AND FUTURE WORK

In this paper, a laboratory scale, microwave plasma reactor has been described. This type of plasma was highly reactive, enabling conversion of solid materials into gas. Under oxidative environment, microwave plasma was able to generate combustible gas from waste paper. This is of practical interest for utilization of solid wastes for the purpose of fuel gas production. However, tests with regards to characterization of microwave plasma generated, as well as parametric investigation of operating conditions are still required. They are planned for the next stage of this research.

ACKNOWLEDGMENT

We wish to thank technical and secretarial assistance from Mr N. Kunnang, P. Thanompongchart, and T. Hanisch.

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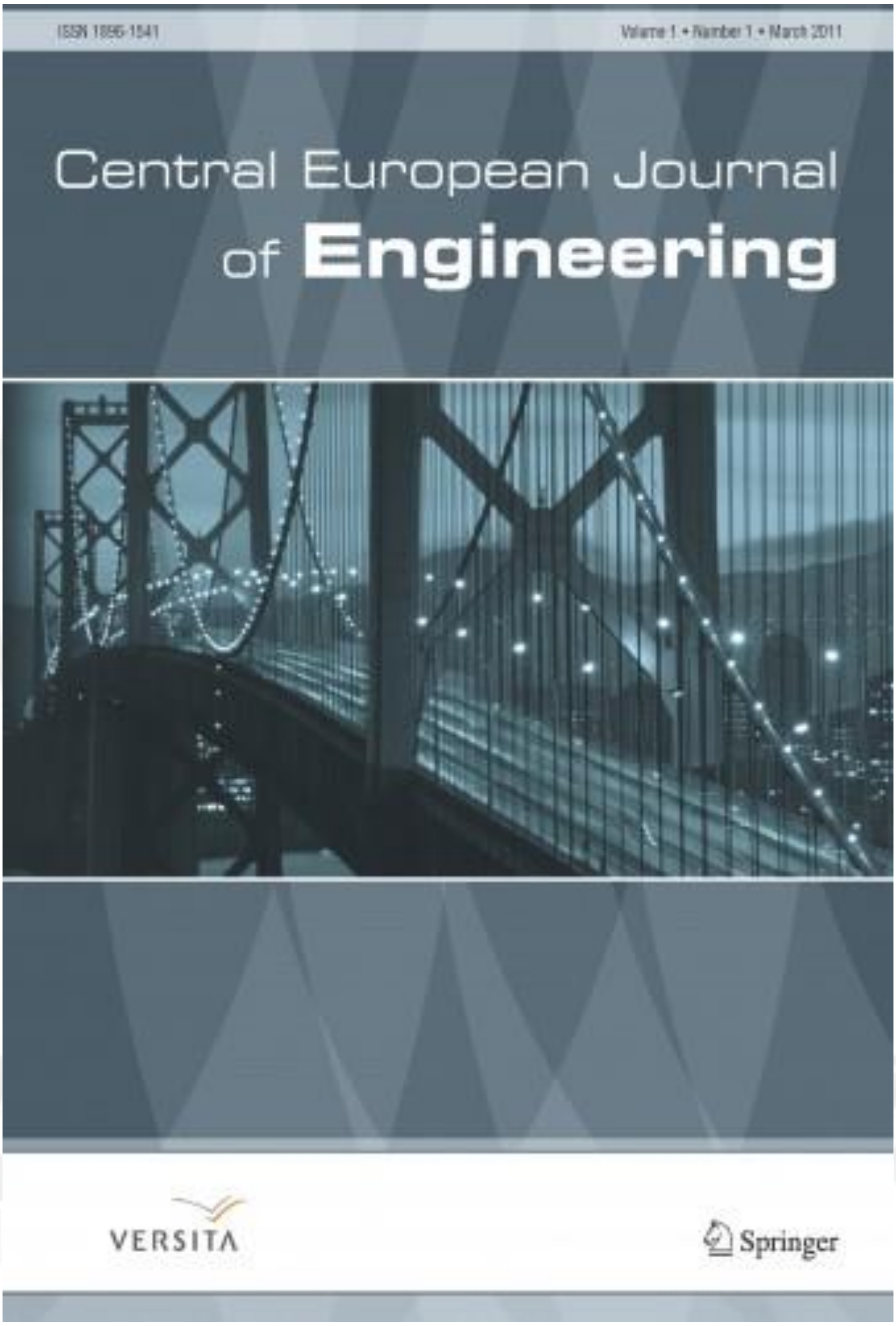

ISSN 1896-1541

Volume 1 • Number 1 • March 2011

Central European Journal of Engineering

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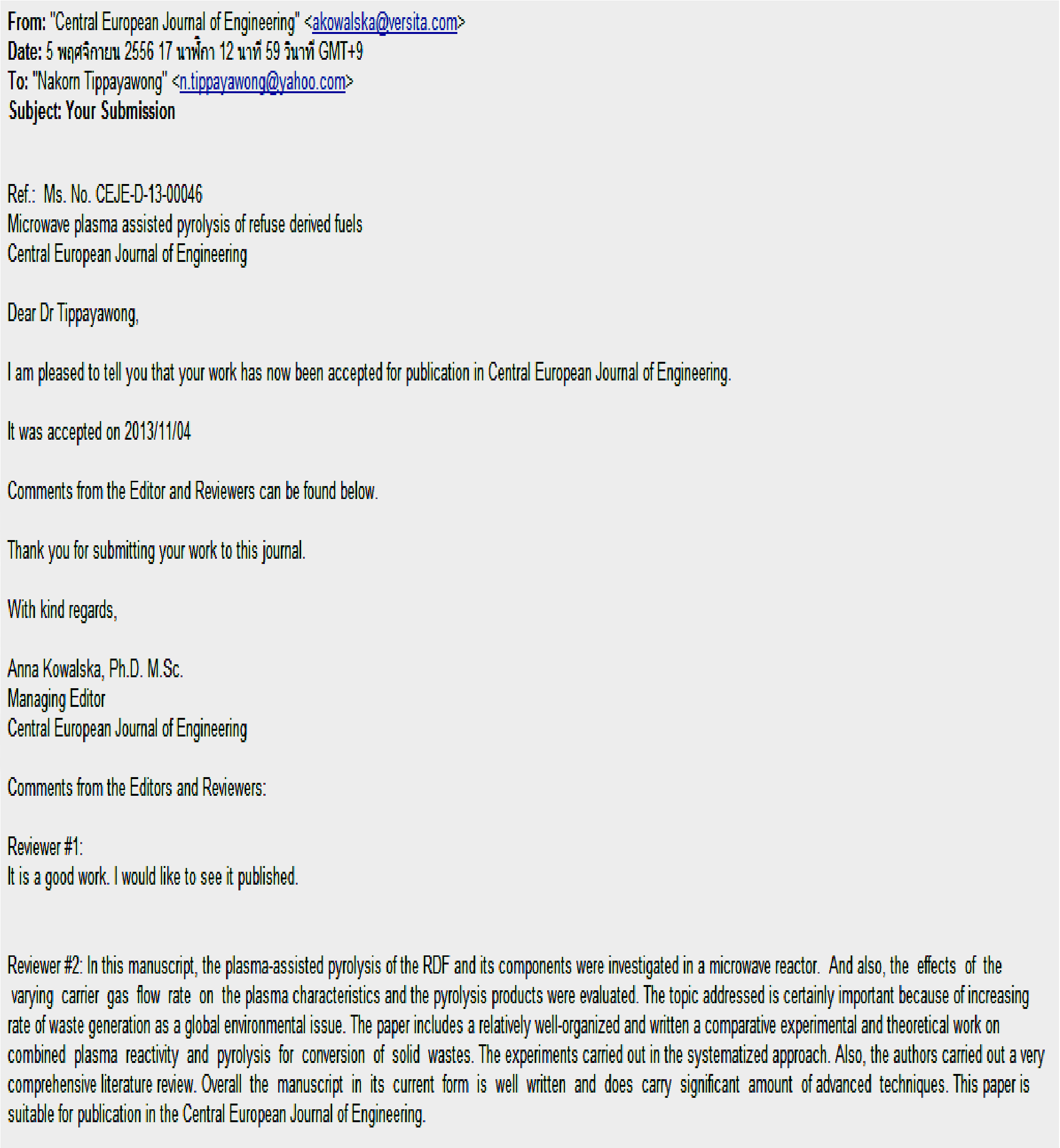

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Reviewer #1:
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Reviewer #2: In this manuscript, the plasma-assisted pyrolysis of the RDF and its components were investigated in a microwave reactor. And also, the effects of the varying carrier gas flow rate on the plasma characteristics and the pyrolysis products were evaluated. The topic addressed is certainly important because of increasing rate of waste generation as a global environmental issue. The paper includes a relatively well-organized and written a comparative experimental and theoretical work on combined plasma reactivity and pyrolysis for conversion of solid wastes. The experiments carried out in the systematized approach. Also, the authors carried out a very comprehensive literature review. Overall the manuscript in its current form is well written and does carry significant amount of advanced techniques. This paper is suitable for publication in the Central European Journal of Engineering.

Microwave plasma assisted pyrolysis of refuse derived fuels

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Abstract

This work combined plasma reactivity and pyrolysis for conversion of solid wastes. Decomposition of refuse derived fuel (RDF) and its combustible components (paper, biomass, and plastic) in an 800 W microwave plasma reactor was investigated at varying argon flow rates of 0.50 to 1.25 lpm for 3 minutes. The characteristic bright light emission of plasma was observed with calculated maximum power density of about 35 W/cm^3. The RDF and its components were successfully converted into char and combustible gas. The average char yield was found to be 12–21% of the original mass, with a gross calorific value of around 39 MJ/kg. The yield of the product gas was in the range 1.0–1.7 m^3/kg. The combustible gas generated from the pyrolysis of the RDF contained about 14% H_2, 66% CO, and 4% CH_4 of the detected gas mass, with a heating value of 11 MJ/m^3. These products are potentially marketable forms of clean energy.

Key words: biomass, RDF, renewable energy, thermochemical conversion, waste-to-energy

1. Introduction

Increased generation of waste is a global environmental issue. Different kinds of waste are generated on a daily basis. The total volumes of solid waste production in the low-, middle-, and high-income countries were approximately 0.4–0.6, 0.5–1.0, and 1.1–5.0 kg/person/day, respectively [1]. The World Bank estimated that the quantity of municipal solid waste (MSW) from the urban areas of Asia would rise from 760 Gg/day in 1999 to 1.8 million Gg/day in 2025. MSW includes wastes produced from commercial, domestic, industrial, institutional, demolition, construction, and municipal services. In Thailand, the total MSW production is over 35 Gg/day [2]. The need to manage MSW properly is well recognized. Generally, technological strategies for the disposal of solid wastes in the developing countries can be classified as (i) land filling, (ii) recycling, (iii) incineration, and (iv) waste-to-energy (WTE) conversion. Land filling is considered the simplest and cheapest of disposal methods. Recycling is the sorting of the wastes to recover materials that are recyclable, fermentable, or combustible. Incineration is described as thermal treatment for reducing the mass and volume of the wastes. A high temperature is employed for the combustion process. The energy utilized may be recovered from the process. WTE conversion is the utilization of solid waste for generation of energy and electricity [2].

The WTE conversion is an extremely attractive option. It refers to any waste treatment that generates energy in the form of electricity or heat from a waste source as well as energy byproducts, such as synthesis gas, liquid fuel, and char. At present, the WTE technologies consist of many conversion methods, such as physical, thermochemical, and biochemical. Physical conversion is a basic technology involving various processes to improve the physical properties of waste. Hazardous waste, metal, and other incombustible matters can be removed. The remaining combustible fraction of the waste is subsequently dried, size-reduced, and compacted into fuel pellets. Refuse-derived fuel (RDF) is the main product from the physical conversion process. In the low- and middle-income countries, the composition of the mechanically separated MSW consists of 30% paper, 50% biomass, and 20% plastic [1]. It has low moisture content, improved calorific value, and uniform size.

Thermochemical conversion is a technology of choice for the utilization of RDF. It includes combustion, gasification, and pyrolysis. Direct combustion of waste with energy recovery is the most common process of WTE implementation. Nevertheless, this method normally gives low energy efficiency. Pyrolysis/gasification is a thermochemical process in an oxygen-starved environment at relatively high temperatures (> 500ºC), transforming wastes into liquid and gaseous fuels as well as char. The fact that the product gas contains H_2 and CO is a key advantage as these can be further used in a variety of applications such as combustion in a turbine or an engine, heat and power generation, as well as for use in fuel cells [3]. Plasmochemical, or plasma-processing, method is relatively new. It offers advantages over the conventional thermochemical processes such as fast heating, ease to control, and low power consumption. The thermal and chemical properties of plasma are synergized with the pyrolysis and gasification process. The process was claimed to interrupt the formation of dioxins, kill bacteria, and have organic materials converted to gases at more than 99% conversion efficiency [4-6].

There is a large body of research work on the pyrolysis of solid wastes and RDF [7-12]. Increasing attention has recently been paid to the plasma process [13-21].

Plasma is the fourth state of matter, comprising energetic electrons, ions, and neutral particles. The degree of ionization of plasma is proportional to atoms that have lost or gained electrons. Two basic types of plasma are employed for industrial processes: thermal and non-thermal [21, 22]. Plasma-assisted conversion process can generate high product gas quality as compared to conventional thermochemical conversion methods such as pyrolysis and gasification. There are many ways to generate plasma. Most published reports employed high-powered thermal plasma to convert biomass materials. The equipment required can be prohibitively expensive to set up and operate. Microwave plasma is an interesting alternative in that it can be generated using a 2.45 GHz magnetron from commercial microwave ovens. It is simple, economical, easy to control, and requires relatively low power input [22-24]. So far, the studies on plasmochemical conversion using microwave plasma have been few and far between [14, 19, 20, 25-28]. Those that focused on solid waste were very few. Among the notable works is the study conducted by Lupa et al. [14, 26], investigating the microwave plasma pyrolysis of waste. The gas evolution was found to peak at approximately 200 s of the reaction time. CO was the most abundant gas species in the product gas. It was reported that feedstock with high oxygen composition increased the heavier gas species in the product gas, such as CO, CO_2, and H_2O, as a result of oxidation.

It is clear that plasma-assisted thermal conversion is a potential alternative technology to utilize solid wastes for energy application. However, there remains a big knowledge gap on the effect of microwave plasma on fuel gas and char production. To date, literature on plasmochemical conversion of RDF has been rare. In this study, three major combustible fractions of the MSW were investigated, both as single component and in the form of RDF. A microwave oven was modified for use as the direct-contact plasma pyrolysis reactor [19]. The plasma generated was used to convert the feedstock into the fuel gas and char. Investigation on the effect of the carrier gas flow rate on the plasma characteristics, such as plasma temperature, discharge length, and power density, was carried out. The evolution and composition of the char and fuel gas obtained from the pyrolysis of each feedstock were investigated. Product yields, carbon conversion efficiency, and calorific value were calculated and compared with relevant literature.

2. Materials and Methods

2.1 Feedstock

In this study, the main combustible fractions of the MSW (paper, biomass, and plastic) were investigated, as single component and in the form of RDF. Raw materials were obtained locally from the same source: (i) office paper was chosen as a representative of the paper fraction, (ii) bamboo was chosen as a representative of the biomass fraction, and (iii) polyethylene (PE) taken from transparent plastic bottles was chosen to represent the plastic fraction. While data for PE was taken from a published report [12], the proximate analyses of the RDF, its other components, and chars were carried out in this work by the thermogravimetric method using a Perkin Elmer, model TGA7 instrument. The ultimate analyses were carried out by dynamic flash combustion method using a ThermoQuest, model Flash EA 1112 CHNS-O analyzer. The results of the compositional analyses of the RDF and its main components are summarized in Table 1.

As for the pyrolysis experiments, bamboo and PE were made into small pieces with a diameter of 2 mm and a length of 20 mm. A number of bamboo and PE pieces were banded using wires. Paper was mashed, and compacted into a cylindrical mold of diameter 20 mm and length 40 mm, after drying in an oven at 90^{o}C for 48 h. The simulated RDF consisted of 30% paper, 20% PE, and 50% bamboo. All of the raw materials were mashed, mixed, and compacted into a cylindrical mold 20 mm in diameter and 40 mm in length, after drying in an oven at 90^{o}C for 48 h. Each sample mass was about 5 ± 0.1 g.

Table 1. Fuel Properties of Raw Materials Used

	Paper	Biomass	Plastic	RDF
Proximate analysis (% w/w)				
Moisture	3.2	5.7	-	3.3
Volatile	83.2	74.7	99.97	83.4
Fixed carbon	4.5	14.0	-	11.7
Ash	9.1	5.6	0.03	2.3
Ultimate analysis (% w/w)				
C	43.5	45.6	86.0	48.1
H	6.2	4.3	14.0	6.3
O	50.2	49.7	-	45.4
N	0.1	0.2	-	0.2
S	0.0	0.0	-	0.0
H/C molar ratio	1.71	1.12	-	1.6
O/C molar ratio	0.87	0.82	-	0.7
HHV (MJ/kg)	20.1	20.6	45.0	22.3

2.2 Experimental apparatus and procedure

The experimental setup is shown schematically in Figure 1. It consists of a plasma reactor, and a gas cooling and cleaning unit. A commercial microwave oven was modified into a plasma generator system. A quartz reactor tube with internal and external diameters of 27 and 30 mm, and length of 250 mm was installed vertically at the central cavity of the oven. The unit was cooled using a water cooling system. Microwave power of 800 W was supplied by a 2.45 GHz magnetron. A stainless steel wire was used as a hanger for the feedstock tested. The plasma generated inside the reactor tube was initiated from the electromagnetic stress concentration at the wire tip, inducing the carrier gas into the ionized phase.

Prior to each test run, the reactor system was vacuumed to ensure absence of air. Argon was used as the carrier gas. It is a relatively cheap, stable, and long-living ion stabilizer. It has been reported that it enlarges electron density [29]. The carrier gas was fed at the bottom of the reactor to purge oxygen in all the systems, including the reactor tube, pipeline, and the gas treatment unit. The argon flow rate was kept varying between 0.50 and 1.25 lpm. It was fed tangentially as a swirling flow to confine the plasma flame and increase the residence time within the reactor. The feedstock was placed into the center of the quartz tube, direct to the aperture of the wave guide in the oven cavity. Once the power was switched on, the discharge took place inside the reaction tube, engulfed the feedstock, and converted it into pyrolysis products. The product gas flowed out at the top of the reactor tube and through the gas treatment unit. After the treatment, the product gas was sampled using a Restek multi-layer foil gas sampling bag for the subsequent gas chromatographic analysis. Tar in the gas product was collected in a wet type tar trap using a series of isopropanal tubes placed in an ice bath. Moisture was removed in a silica gel chamber. After 3 minutes of reaction, the solid residue was carefully collected, weighed, and sent for analyses.

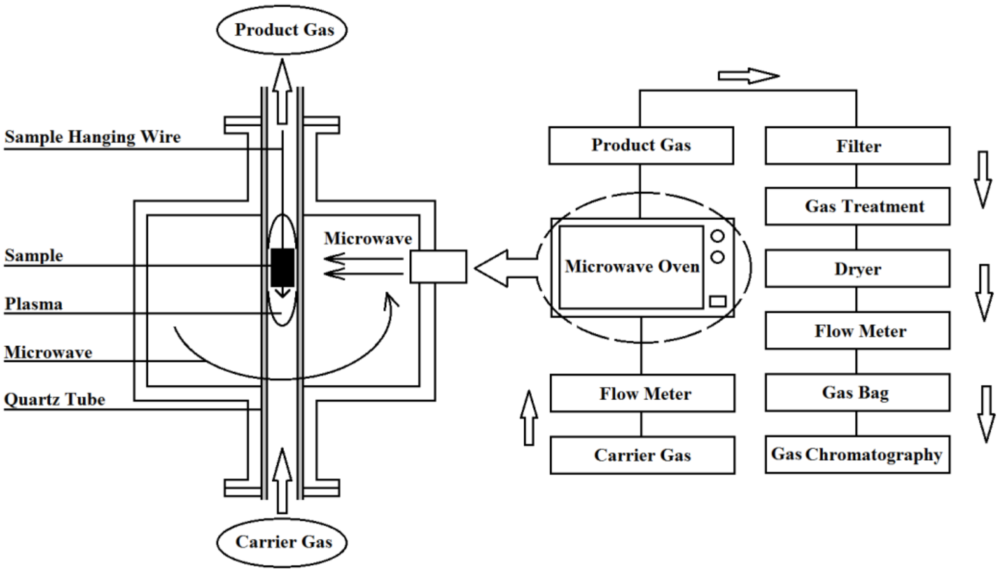

Figure 1. The experimental setup of the microwave plasma reactor for the pyrolysis of the RDF.

2.3 Analysis

The plasma characteristics, including temperature, discharge length, and volume, as well as power density, were evaluated. The plasma temperature was measured indirectly using a thermocouple placed at a distance of 20 mm downstream of the discharge zone inside the quartz tube. The discharge length was measured from the discharge images captured by a digital camera. The discharge volume was calculated from its length multiplied by a cross section area of the reaction tube. The power density was defined as the ratio between the input microwave power and the discharge volume.

The product gas was analyzed for its composition using a Shimadzu model GC-8A gas chromatography, fitted with a Micropacked column model ShinCarbon ST 80/100 mesh and a TCD. The analyzer was able to measure the molar fractions of H_2, O_2, N_2, CH_4, CO, and CO_2. An operational column temperature of 40°C and a detector temperature of 70°C were used. The results were processed via a C-R8A chromatopac data processor. The Restek pure gases and mixtures standard model Scotty 14 were used as standard gas for quantitative calibration.

As for the data processing, the higher heating value (HHV) of the solid material was calculated from its contents [30] as

$$HHV = 0.491(C) + 0.261(H) - 2.907, \tag{1}$$

where, *C* and *H* are the concentrations of carbon and hydrogen in the material (% w/w).

Char yield was defined as the ratio of the mass of the obtained char to the original mass of the feedstock:

$$Y_{char} = m_{char}/m_f. \tag{2}$$

The product gas yield was calculated from the volume of gas generated and the reacted original mass of feedstock [22] as

$$Y_{gas} = Q_{gas}/m_f. \tag{3}$$

The lower heating value (LHV) of the product gas was defined as the summation of the corresponding heating values of H_2, CO, and CH_4 [22]:

$$LHV = [(25.7 \times H_2) + (30 \times CO) + (85.4 \times CH_4)] \times 4.2 \qquad (4)$$

Carbon conversion efficiency was defined as the ratio between the carbon in the gaseous fuel and the carbon in the feedstock:

$$\eta_C = \frac{Q_{gas}.[(CO+CO_2+CH_4)/100].(12/22.4)}{m_f.[1-(A_f/100)].(C_f/100)} \qquad (5)$$

where, Q_{gas} is the volume of the product gas generated (Nm^3), m_f is the original mass of the feedstock (kg), m_{char} is the mass of the obtained char (kg), A_f is the ash content in the feedstock (% w/w), and C_f is the carbon content in the feedstock (% w/w).

3. Results and Discussion

3.1. Plasma characteristics

With power on and an absence of the feedstock, a bright light emission in the form of pinkish plasma flame was observed in the quartz tube. For different carrier gas flow rates, the post plasma temperature 20 mm away from the top of discharge zone inside the quartz tube was found to be about 600 to 1100°C. The plasma stream was established and stabilized inside the reaction tube with a cross section area of 5.7 cm^2. The discharge length was estimated to be in the range of 45–70 mm. For the range of flow rates considered, the residence time, defined as the plasma volume divided by the respective flow rate, was between 1.9 and 2.8 s. The calculated power densities were in the range 19.5–34.9 W/cm^3. A summary of the plasma characteristics under variable flow rates is given in Table 1. The post plasma temperature showed an initial rise with increasing carrier gas flow rate, reaching the maximum at about 1100^{o}C for 0.75 lpm of the argon flow rate. At the lowest flow rate, the plasma with the short discharge length was generated. Hence, its volume, rate of electron collision, and energetic particle concentration were small. As the flow increased, more molecules were available for excitation; hence, evidently, there were more energetic particles, until the optimum point was reached. Further increases in the argon supply led to a drop in the temperature. The observed decline in the post plasma temperature may be attributed to the fact that, at higher flow rates, the discharge length and volume were increased, leading to reduction in the power density and residence time. At very high flow rates, the plasma generated may become non-stabilized. Breakdown of the electromagnetic field was occasionally observed as a result of the plasma temperature changes. With the presence of the feedstock, a bluish yellow flame was observed at the start of the conversion process. This may be due to partial oxidation and release of volatile matter from the feedstock.

Table 2. Microwave Plasma Characteristics

Flow rate (lpm)	Temperature (^{o}C)	Length (mm)	Residence time (s)	Power density (W/cm^3)
0.50	783 ± 26	45 ± 5	2.75	34.9
0.75	1084 ± 23	52 ± 5	2.29	27.9
1.00	873 ± 18	63 ± 5	2.06	23.3
1.25	593± 13	70 ± 5	1.93	20.0

3.2 Pyrolysis product characteristics

The details regarding the yields of char and gas products as well as gas calorific value and carbon conversion efficiency from the pyrolysis of paper, biomass, plastic, and RDF are listed in Table 3. The char characteristics and yield are important in evaluating the performance of the reactor. It was found that the average char yields were about 21, 12, 18, and 16% for paper, biomass, plastic, and RDF, respectively. Even through plastic showed the highest remaining solid residue, a close examination of the inner residue revealed that a major bulk of its mass had remained unreacted. Longer reaction time may be needed for the plastic sample. Nonetheless, successful carbonization appeared to have occurred for the other feed materials. A compositional analyses of the chars from the paper, biomass, and RDF (Table 4) showed significant increases in the carbon and energetic content, compared to their original proximate and ultimate composition. This was a clear indication of the high degree of carbonization that had taken place. A marked decrease in volatile matter was indicative of the high conversion of the organic materials into the gaseous phase. From Table 3, it can be observed that the average gas yields were in the range between 1.0 and 1.7 m^3/kg. Bamboo was found to give the highest gas yield, while PE the lowest. This observation was true for the carbon conversion efficiency, as well. Nonetheless, the calorific values of these gas products were of similar magnitude, ranging between 10.1 and 11.1 MJ/m^3. Within the range of the argon flow rates considered, a change in the carrier gas supply did not affect the yields of the pyrolysis products significantly. Nonetheless, the optimum flow rate was 0.75 lpm, where the highest gas calorific value and carbon conversion efficiency were obtained. The post plasma temperature was also observed to be the maximum at this flow rate.

With respect to the product gas distribution, Figures 2 to 5 show the effect of the carrier gas flow rate on the evolution of H_2, CO, CH_4, and CO_2, respectively. The mean values are shown with error bars indicating the standard deviation. From the results obtained, it can be seen that H_2 and CO were by far the largest components. The volume of CH_4 was about 2–4% only. In this study, H_2 was produced from the use of paper, biomass, plastic, and RDF in volumes of about 24, 22, 10, and 14%, respectively. CO was the most abundant combustible fraction of the product gas. It was found to be in the range of 56–73%. Within the range of the flow rates considered, the average total content of the combustible fractions in the product gas of all feedstock was about 80%, showing similar patterns of variation. Changes in the combustible gas components with the argon flow rate were more noticeable for the paper and biomass samples. As for the plastic sample, CO was found to be the major component, more than all the other components combined. Meanwhile, the evolution of the product gas from the RDF pyrolysis was observed to fall between these two groups. For all feed materials, H_2 and CO appeared to initially increase with increasing argon flow, reaching the maximum value at the flow rate of 0.75 lpm, after which their contents dropped at higher gas supply rates. The patterns were similar to those observed for the post plasma temperature. The reasons for this may be related to the concentration of the energetic particles in the discharge zone and the resulting temperature evolution. However, it is not yet conclusive as to how the presence of each material and its property might affect the generation of plasma from the carrier gas.

3.3 Comparison with literature

The details regarding the char and product gas obtained from the plasma-assisted pyrolysis of paper, biomass, plastic, and RDF in this analysis were compared with those obtained from the other types of feedstock and plasma sources and conditions. The comparison is given in Tables 5 and 6 for the char and the product gas, respectively. When compared to the microwave plasma pyrolysis of sawdust [6], rice husk, and cane [26], the char yields obtained in this work were smaller, but with higher calorific value. With the DC arc plasma, the quantity of the reported char yields varied markedly between the raw materials used. Regarding the gas product, the gas heating values were similar in magnitude to those reported in the literature.

4. Conclusion

In this work, the plasma-assisted pyrolysis of the RDF and its components were investigated in a microwave reactor. The effects of the varying carrier gas flow rate on the plasma characteristics and the pyrolysis products were evaluated. Apart from the plastic sample, significant degrees of volatile release and carbonization appeared to take place for all raw materials under the plasma environment. The average char yield of the RDF and its calorific value were 16% and 39 MJ/kg, respectively. The average total content of the combustible fractions in the product gas for all feed materials was about 80%. The major components of the product gas generated were H_2 and CO. They were found to initially increase with increasing argon flow, reaching the maximum value at the optimum flow rate of 0.75 lpm. At this flow rate, the use of RDF generated about 14% of H_2, 56% of CO, and 4% of CH_4. The heating value and yield of the product gas from the pyrolysis of the RDF were 11.1 MJ/m^3 and 1.0 m^3/kg, respectively. The calorific value obtained was consistent with the other pyrolysis studies. The process offered interesting alternatives for the utilization of solid wastes for the purpose of char and fuel gas production.

Acknowledgment

Supports from the Office of Higher Education Commission via the National Research University program, and the Energy Policy and Planning Office, Ministry of Energy, Thailand, are acknowledged.

Table 3. Yields of Char and Gas Products

Flow rate (lpm)		Char yield (%)	Gas yield (m^3/kg)	Gas LHV (MJ/m^3)	Carbon conversion (%)
Paper	0.50	22.40	1.05	10.89	77.15
	0.75	19.22	1.27	11.49	84.47
	1.00	21.18	1.37	11.32	79.69
	1.25	21.37	1.42	10.67	70.99
	average	21.04	1.28	11.09	78.08
Biomass	0.50	10.32	1.29	9.96	84.17
	0.75	11.94	1.61	10.75	89.77
	1.00	12.00	1.83	10.35	85.88
	1.25	13.79	1.98	9.28	71.81
	average	12.01	1.68	10.08	82.91
Plastic	0.50	16.77	1.13	9.70	76.65
	0.75	16.13	1.45	10.81	84.78
	1.00	18.33	1.67	10.41	77.91
	1.25	20.69	1.90	9.45	70.81
	average	17.98	1.54	10.09	77.54
RDF	0.50	15.70	0.85	10.46	77.12
	0.75	15.57	1.01	11.17	82.43
	1.00	15.68	1.08	10.88	79.99
	1.25	15.43	1.11	10.13	75.31
	average	15.59	1.01	10.66	78.71

Table 4. Fuel Properties of Char Products

	Moisture	Volatile	Fixed carbon	Ash	C	H	O	N	S	HHV (MJ/kg)
Paper	2.0	14.4	54.9	28.6	83.6	1.6	14.8	0.01	0.0	38.5
Biomass	3.1	12.2	72.0	9.7	87.2	1.4	10.4	0.01	0.0	40.5
RDF	1.6	11.6	60.7	24.5	84.3	1.7	14.0	0.01	0.0	38.9

Table 5. Char from Plasma-assisted Pyrolysis Process

Reference	Method	Feedstock	Char	
			Yield (%)	HHV (MJ/kg)
This work	Microwave plasma	RDF	15.6	38.9
		bamboo	12.0	40.5
		paper	21.0	38.5
		PE	18.0	-
[6]	RF plasma	sawdust	33.3	29.0
[17]	DC arc plasma	used tires	69.6	-
[27]	Microwave plasma	rice husk	33.4	21.6
[27]	Microwave plasma	cane residue	25.5	27.8
[31]	DC arc plasma	rice straw	7.5-13.8	-

Table 6. Product Gas from Plasma-assisted Pyrolysis Process

Reference	Method	Feedstock	Product gas content				LHV
			H_2	CO	CH_4	CO_2	(MJ/kg)
This work	Microwave plasma	RDF	13.8	65.5	4.0	14.2	11.1
		bamboo	22.4	55.6	3.7	15.5	10.8
		paper	23.8	58.4	4.4	12.2	11.5
		PE	9.5	72.6	1.8	11.9	10.8
[6]	RF plasma	sawdust	8.5	11.0	1.5	4.0	2.8
[13]	DC arc plasma	MSW	43.5	34.5	0.01	0.03	9.0
[14]	Microwave plasma	mixed waste	-	55.7	0.7	26.9	15.0
[20]	Microwave plasma	PE	14.0	26.0	6.0	12.0	6.9
[26]	Microwave plasma	waste wood	0.0	56.9	0.5	33.8	7.3

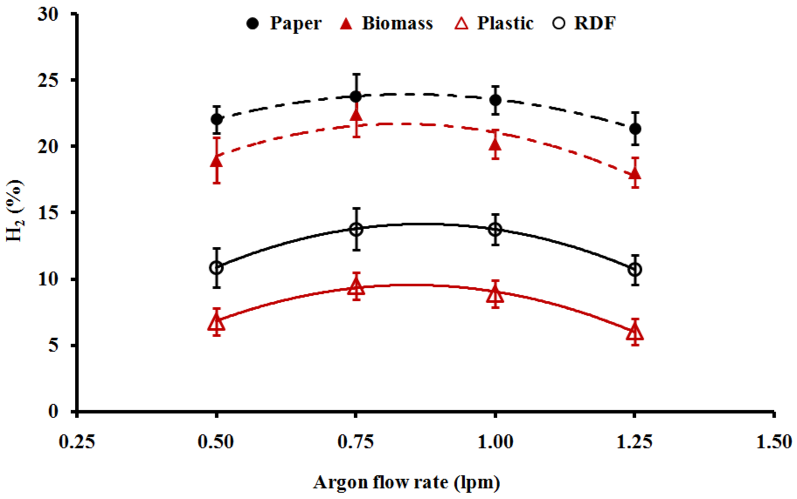

Figure 2. The variation in the H_2 evolution with the carrier gas flow rate and the type of feedstock.

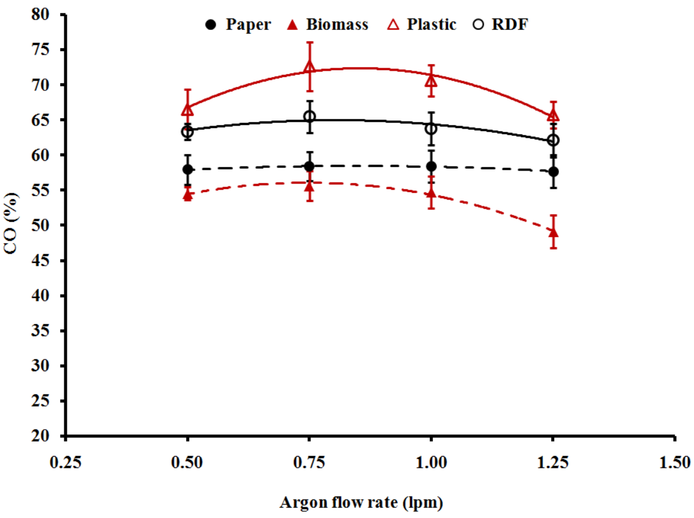

Figure 3. The variation in the CO evolution with the carrier gas flow rate and the type of feedstock.

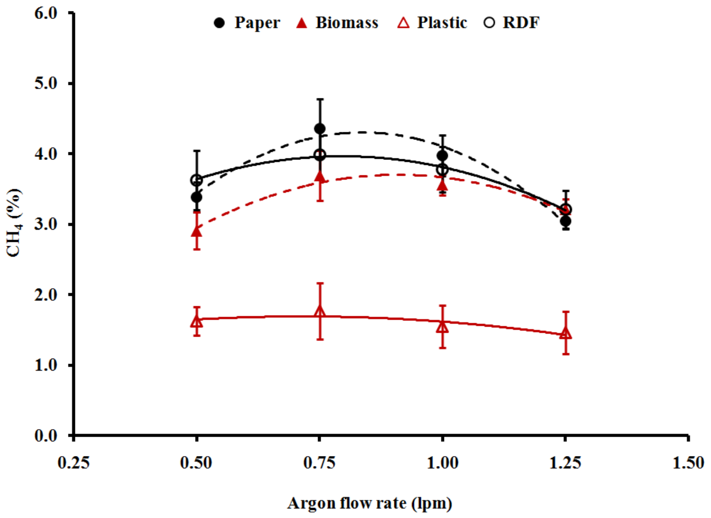

Figure 4. The variation in the CH_4 evolution with the carrier gas flow rate and the type of feedstock.

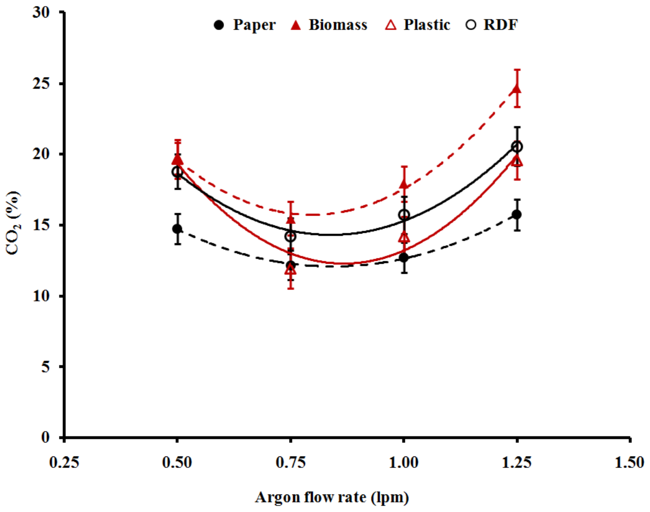

Figure 5. The variation in the CO_2 evolution with the carrier gas flow rate and the type of feedstock.

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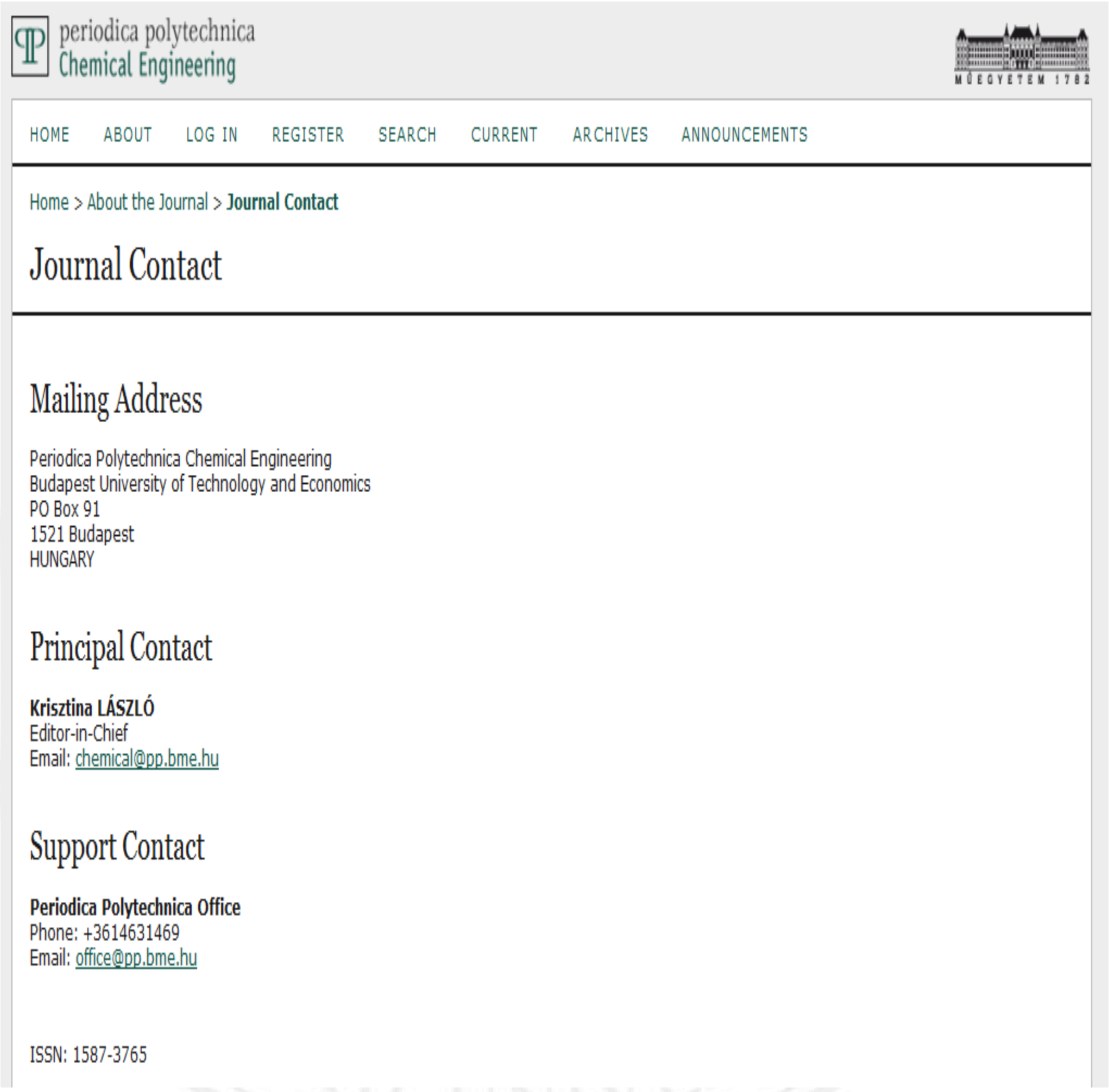

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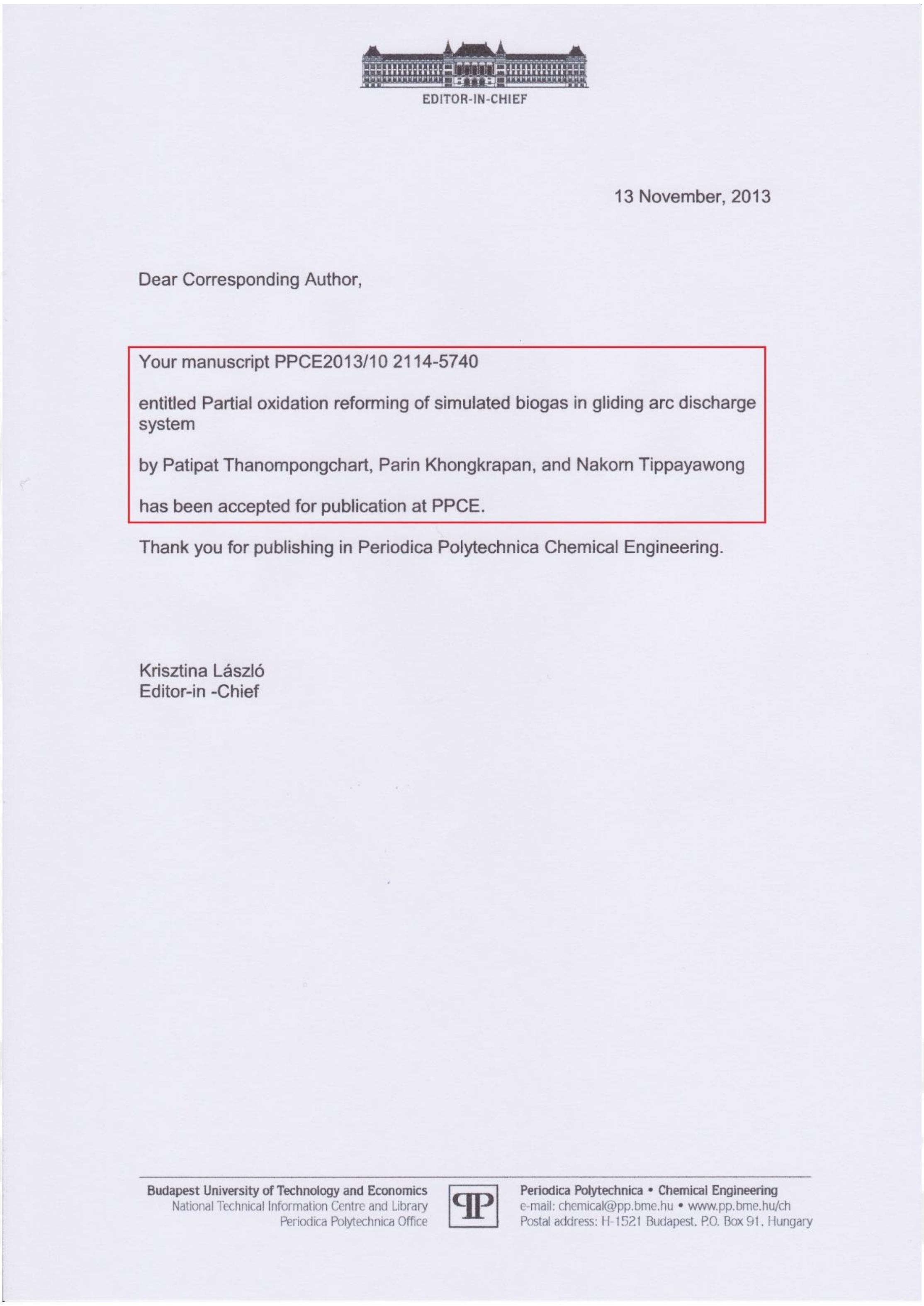
ISSN: 1587-3765



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Partial oxidation reforming of simulated biogas in gliding arc discharge system

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Abstract

Plasma assisted, partial oxidation reforming of biogas is considered to be a promising technology to produce synthesis gas. In this work, a 0.1 kW gliding arc plasma reformer was employed to investigate the effects of biogas composition and oxygen availability on CH_4 and CO_2 conversions, as well as the product distribution. Air was used in the partial oxidation of biogas. The results showed that at low CH_4/O_2 ratio or high oxygen availability, increasing CH_4 content appeared to show higher H_2 yield and CH_4 conversion. Increasing CH_4/O_2 ratio adversely affected H_2 and CO yields, and CH_4 conversion. Optimum condition was found at CH_4/CO_2 of 90:10 and CH_4/O_2 of 1.2 for the maximum CH_4 conversion and H_2 yield of 45.7 and 25.3%, respectively.

Keywords: biomass, methane reforming, non-thermal plasma, renewable energy, synthesis gas

1. Introduction

Energy is recently becoming one of the most pressing issues in many societies and countries. It affects wellbeing of the people, economic development, national security, and civilization. The energy demand remains high and growing, while conventional supply from crude oils is increasingly fragile. Concerns over climate change and recent incidents at Fukushima have presently made coal and nuclear powers rather unfashionable. To tackle a major threat of energy crisis, strong energy conservation and efficiency improvement program must be adopted. At the same time, alternative energy sources must be explored and utilized. Renewable energy is, if properly integrated, able to cover all energy needs. Change from fossil fuels to renewables is happening and relevant to many nations around the World.

Among various types of renewable energy resources, biogas appeared to be one of the most promising options. Thailand has the potential to produce over one billion m^3 of biogas a year from its agricultural industry alone [1]. Normally, biogas contains 45-70% CH_4, 30-45% CO_2, and a trace amount of other gases. Composition of biogas depends on raw biomass materials and conditions of anaerobic digestion [2, 3]. The biogas produced is generally utilized at farm levels for heating, mechanical shaft works, and electricity generation. To further harness this renewable energy source, biogas may be upgraded to more attractive and marketable gaseous fuels such as compressed biogas, biomethane, or synthesis gas [1, 4, 5].

Synthesis gas (H_2 and CO) production is of great interest because it can be used as starting feed to generate synthetic chemicals and liquid fuels [6, 7]. There are several technologies available for synthesis gas production through CH_4 rich gas, namely, steam or wet reforming, CO_2 or dry reforming, and partial oxidation reforming, shown in eqs. (1) to (3) below;

$$CH_4 + H_2O \rightarrow CO + 3H_2 \qquad \Delta H = 206 \text{ kJ/mol} \qquad (1)$$

$$CH_4 + CO_2 \rightarrow 2CO + 2H_2 \qquad \Delta H = 247 \text{ kJ/mol} \qquad (2)$$

$$CH_4 + 1/2O_2 \rightarrow CO + 2H_2 \qquad \Delta H = -38 \text{ kJ/mol} \qquad (3)$$

Because the former two routes are highly endothermic, hence require heat input and consume large amount of energy, considerable attention has been drawn to focus on the exothermic partial oxidation. For non-catalytic partial oxidation of hydrocarbons, it usually occurs at high temperature (1300-1500°C) to complete conversion. In biogas reforming, methane decomposition and boundary reactions (4 and 5) are possible,

$$CH_4 \leftrightarrow 2H_2 + C \qquad \Delta H = 75 \text{ kJ/mol} \qquad (4)$$

$$2CO \rightarrow CO_2 + C \qquad \Delta H = -171 \text{ kJ/mol} \qquad (5)$$

For biogas, combination of carbon dioxide and partial oxidation of methane can reduce coke deposition on electrodes’ surface. Furthermore, application of plasma technology may enable high conversion at reduced energy consumption of the chemical process [8].

Plasma is ionized gas that can be generated from combustion, electric discharges (arc, spark, plasma jet, microwave discharge, corona, glow and radio frequency), and shocks (electrically, magnetically and chemically driven) [9]. It is effective in generating active species such as electrons, ions, and radicals. It can be classified into non-thermal and thermal plasma. Non-thermal plasma has low electron density ($< 10^{19}$ m^{-3}) and dissimilarity between electron and heavy particle temperatures. Inelastic collisions between electrodes and heavy particles create the plasma reaction species whereas elastic collisions heat the heavy particle [8, 10, 11]. Advantage of using non-thermal plasma is associated with lower temperatures, lower energy consumption and electrode erosion [12]. Gliding arc discharge is one of non-thermal plasma, associated with low reaction temperature, high selectivity, and compact equipment [12–14]. It has at least two diverging knife shaped electrodes. When a high voltage is applied, a relatively low current arc discharge is formed repeatedly at the narrowest gap across the electrodes, and spreads along the edges, and eventually disappears downstream.

Gliding arc plasma has been utilized in reforming of CH_4 rich gas to produce synthesis gas [10, 13]. But, there have been relatively few studies investigating plasma assisted partial oxidation of CH_4/CO_2 system. Reported works using gliding arc discharge reactor were even more limited. Notable article on gliding arc plasma reforming of biogas include Sreethawong et al. [15], Yang et al. [16], Rafiq and Hustad [17, 18], and Rafiq et al. [19]. It was noted that none of these works focused on non-catalytic effect at high CH_4/O_2 ratios. In this work, biogas reforming via partial oxidation under gliding arc plasma was carried out. The main objective is to explore partial oxidation of biogas to produce synthesis gas. Effects of biogas composition and rich fuel-to-air mixing on process performance were investigated.

2. Materials and Methods

Fig. 1 shows schematic diagram of the experimental setup for plasma reforming of biogas. It consists of a gliding arc reactor, a power supply, gas feeding line, measurement and analysis instrumentation. Simulated biogas was generated from mixing CH_4 and CO_2. Air was used as source of oxygen in partial oxidation. These gases were of research grade obtained from Thai Industry Gas Plc. The feed gas line was responsible for supplying CH_4, CO_2 and air. The main input gases were controlled by Dwyer VFA gas flow meters and regulators as well as Hewlett-Packard soap film flow meter. It was injected through a cylindrical tube with diameter of 1 mm with total flow rate of 1 L/min. The reactor was made of transparent acrylic and glass plates. Knife shaped electrodes were made from stainless steel (3 mm thick). The electrode gap was 4 mm. The input power was supplied from an AC high voltage (HV) Lecip Neon transformer was fixed at 7.5 kV and 100 W. The analysis system was divided into electrical characterization, temperature measurement, and gas analysis. The electrical measurement consists of Fluke 80K-40 HV probe, Gwinstek GOS-620 oscilloscope and Pro Elec PL09564 power meter to measure the supplied electric power. The temperature measurement was carried out using Digicon ND-400N type K thermocouples. The temperature was monitored in real time from the thermocouples installed in the electrode gap, avoiding contact with electrodes and discharge region. The gas analysis was done by a Shimadzu 8A gas chromatography equipped with a thermal conductivity detector and Shin carbon column), able to analyze H_2, O_2, N_2, CO, CH_4 and CO_2.

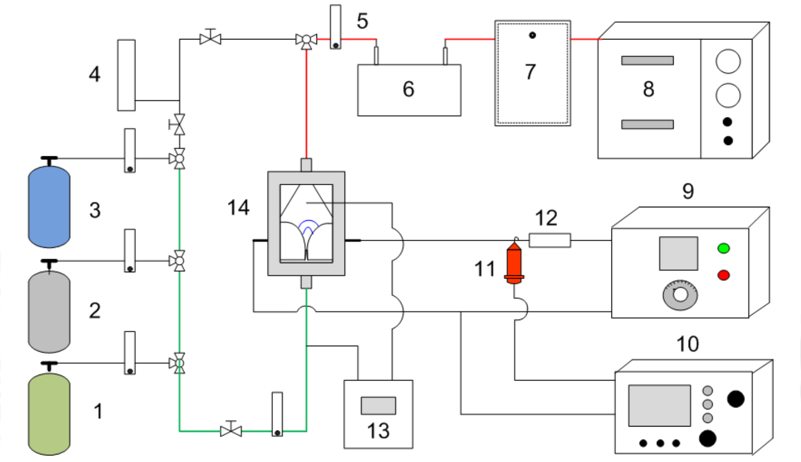


FIGURE 1: Schematic diagram of the experimental setup. (1. methane, 2. carbon dioxide, 3. air, 4. bubble flow meter, 5. flow meter, 6. filter and silica gel, 7. gas collection bag, 8. GC, 9. HV power supply, 10. oscilloscope, 11. HV probe, 12. electrical resistance, 13. digital thermometer, 14. gliding arc reactor)

Feed gas composition can be adjusted. Once constant composition of mixed gas passed into the gliding arc plasma reactor was established, the power supply was then switched on. The system was allowed to stabilize, then gas samples up- and downstream of the reactor were collected in Restek multilayer foil gas bags, and sent immediately for analysis. At least three experimental runs were carried out for each case, and average results were shown. Effect of the following parameters on reforming reaction were studied; biogas composition (CH_4/CO_2: 50/50, 70/30, 90/10) and oxygen content in partial oxidation process (CH_4/O_2: 0.5 – 20). To evaluate the performance of the process, equations (6) to (12) were used;

CH$_4$ conversion: $$C_{[CH_4]}(\%) = \frac{M_{[CH_4,in]} - M_{[CH_4,out]}}{M_{[CH_4,in]}} \times 100 \quad (6)$$

CO$_2$ conversion: $$C_{[CO_2]}(\%) = \frac{M_{[CO_2,in]} - M_{[CO_2,out]}}{M_{[CO_2,in]}} \times 100 \quad (7)$$

H$_2$ selectivity: $$S_{[H_2]}(\%) = \frac{M_{[H_2,produced]}}{2 \times M_{[CH_4,converted]}} \times 100 \quad (8)$$

CO selectivity: $$S_{[CO]}(\%) = \frac{M_{[CO,produced]}}{M_{[CH_4,converted]}} \times 100 \quad (9)$$

H$_2$ yield: $$Y_{[H_2]}(\%) = \frac{M_{[H_2,produced]}}{2 \times M_{[CH_4,in]}} \times 100 \quad (10)$$

CO yield: $$Y_{[CO]}(\%) = \frac{M_{[CO,produced]}}{2 \times (M_{[CH_4,in]} + M_{[CO,in]})} \times 100 \quad (11)$$

Applied energy density:

$$\text{AED (kJ/L)} = \frac{AP}{FR} \quad (12)$$

where, $M_{[CH_4,in]}$ is amount of CH$_4$ input, $M_{[CH_4,out]}$ is amount of CH$_4$ out, $M_{[CO_2,in]}$ is amount of CO$_2$ input, $M_{[CO_2,out]}$ is amount of CO$_2$ out, $M_{[H_2,produced]}$ is amount of H$_2$

produced, $M_{[CH_4,converted]}$ is $CH_{4[in]}$ - $CH_{4[out]}$, $M_{[CO,produced]}$ is amount of CO produced, AP is power or input electricity in kW, FR is feed gas flow rate in L/s, respectively.

3. Results and Discussion

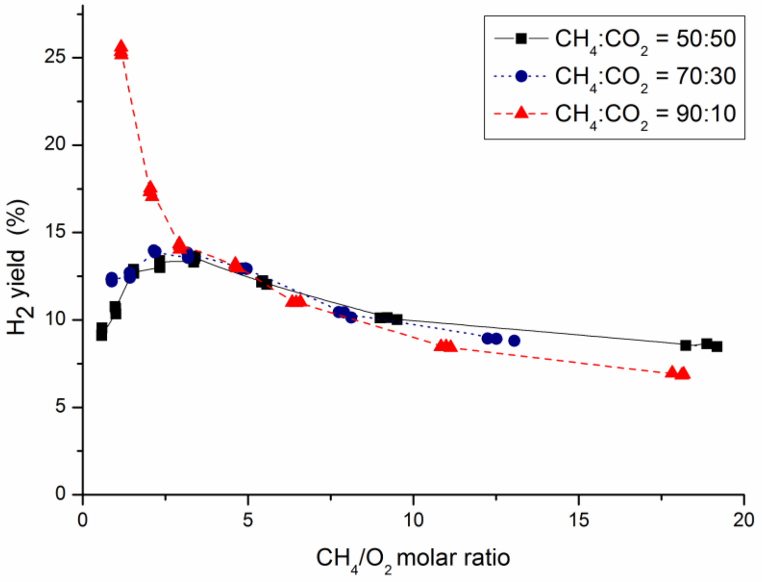
Effects of CH_4/CO_2 and CH_4/O_2 molar ratios in plasma assisted partial oxidation on H_2 and CO yields and selectivities, as well as CH_4 and CO_2 conversions were considered. They are shown in Figs. 2 to 4. Biogas composition was varied (CH_4/CO_2: 50/50, 70/30, 90/10), such that enrichment of methane was taken into account. In this work, large range of CH_4/O_2 molar ratios (0.5 - 20) was considered. For a fixed and very low value of CH_4/O_2 ratio, enriching CH_4 content or decreasing CO_2 content in biogas was found to produce higher yield and selectivity of H_2, while no clear pattern was emerged for yields and selectivity of CO, as well as conversion of CH_4 and CO_2. Maximum H_2 yield of over 25% was obtained at CH_4/CO_2 = 90/10, whereas at CH_4/CO_2 = 50/50, around 10% was observed. There were no significant changes in H_2 yields and CH_4 conversions with increasing CH_4 content for the CH_4/O_2 ratios beyond two. It may be noted that greater presence of CO_2 in biogas seemed to encourage higher CO yields and selectivity as well as higher CH_4 conversion. Possible reaction of biogas reforming at high concentration of CO_2 is [20, 21];

$$CH_4 + 2CO_2 \rightarrow 3CO + 2H_2 + H_2O \qquad \Delta H = 288 \quad \text{kJ/mol} \qquad (13)$$

Increased CO yields were observed with decreasing content of O_2. However, in this work, the observed changes were relatively small.

Increasing the CH_4/O_2 ratio was found to affect the performance of the plasma reformer significantly. This was contributed to the fact that an increase in CH_4/O_2 ratio resulted in having less O_2 available to react with the fuel molecules, leading to lower conversion of CH_4, and yields of H_2 and CO. For the biogas with CH_4/CO_2 = 90/10, changing the CH_4/O_2 ratios from about 1 to 10 led to declines in CH_4 conversion and H_2 yield from 45 to 15%, and 25 to 10%, respectively. The highest H_2 yield and CH_4 conversion were obtained at the lowest CH_4/O_2 ratio considered. Optimum condition was found at CH_4/CO_2 of 90:10 and CH_4/O_2 of 1.2 for the maximum CH_4 conversion and H_2 yield of 45.7 and 25.3%, respectively.

With respect to the effect on the selectivities of H_2 and CO, it was found that CO selectivity decreased while H_2 selectivity increased with increasing CH_4/O_2 molar ratio. This was in line with Sreethawong et al. [15]'s observation. The plasma system appeared to promote two spontaneous reactions; partial oxidation of CH_4 and the coupling reaction. The former reaction was favorable at very low CH_4/O_2 ratios, while the coupling reaction and the hydrogenation become more pronounced with increasing CH_4/O_2 ratios [15]. At high CH_4/O_2 ratios (> 10), change in oxygen availability did not significantly affect partial oxidation of CH_4. At the extremely rich fuel-to-air mixture where oxygen is in short supply, the reaction was expected to behave like dry CO_2 reforming of CH_4 [10]. It should be noted that small traces of moisture as well as H_2S are normally found in biogas, especially from animal farms. Their influences on reforming reaction should not be overlooked. These impurities may affect the performance of the process considered. However, this is outside the scope of the present investigation.



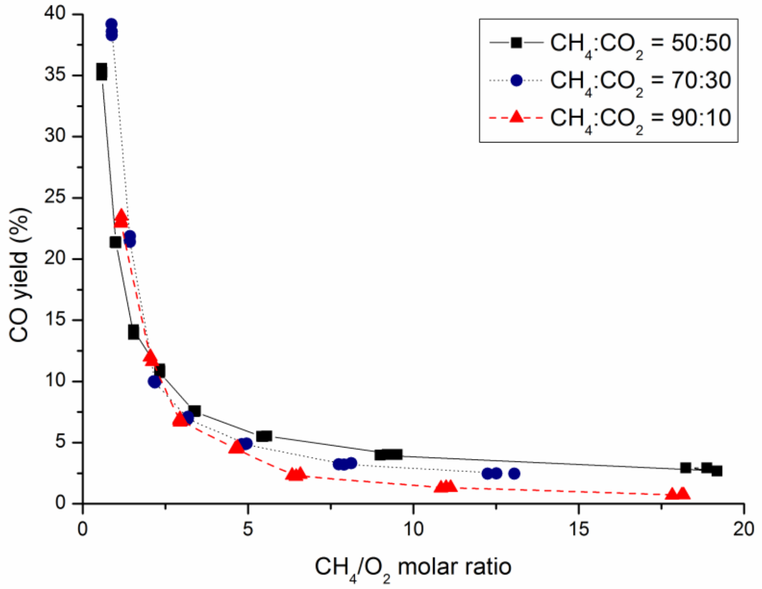
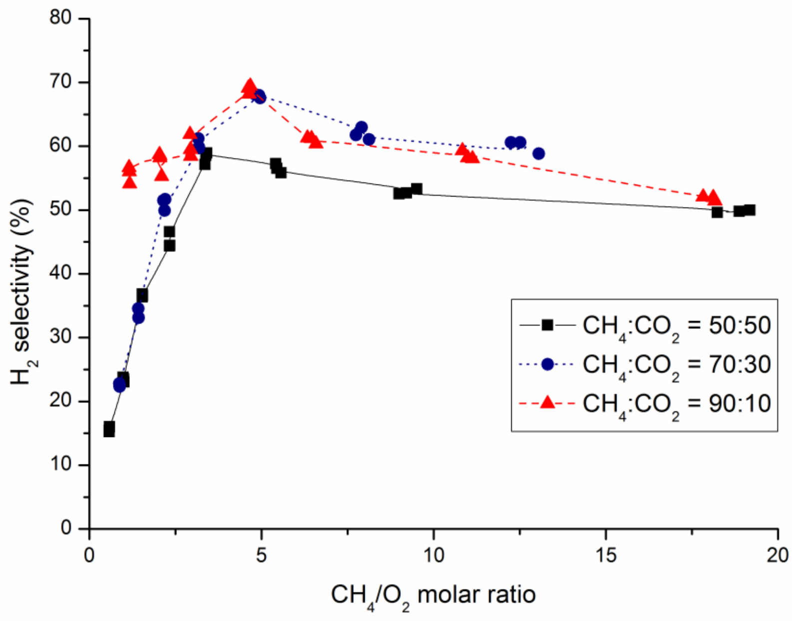


FIGURE 2: Effects of CH_4/CO_2 and CH_4/O_2 on H_2 and CO yields.



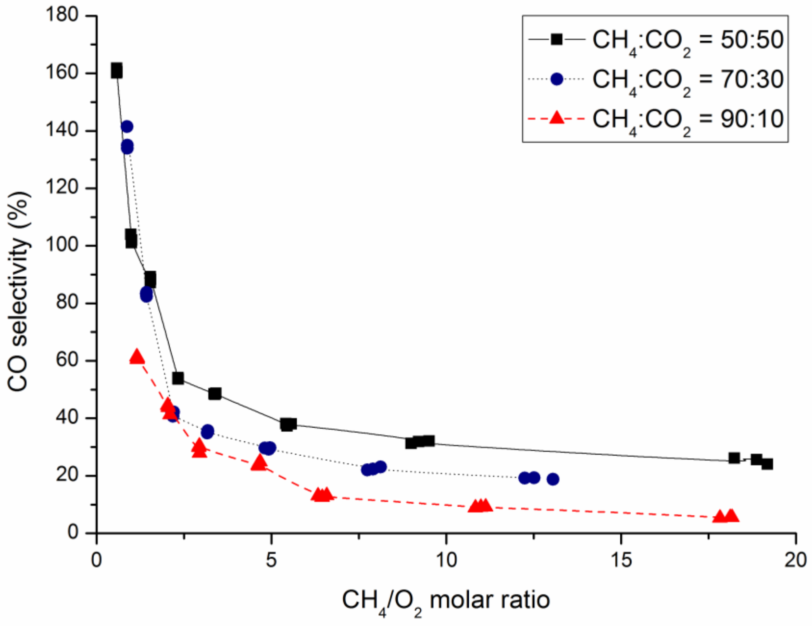


FIGURE 3: Effects of CH_4/CO_2 and CH_4/O_2 on H_2 and CO selectivities.

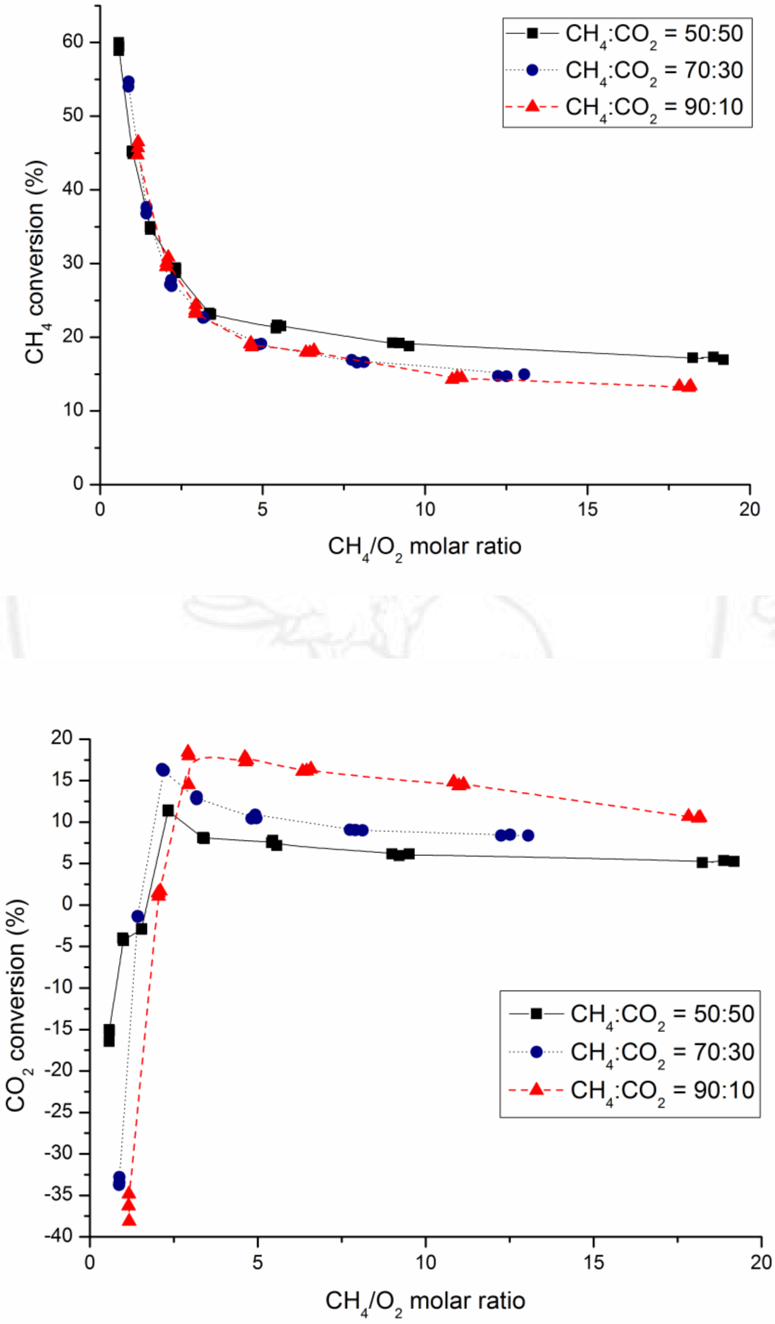


FIGURE 4: Effects of CH_4/CO_2 and CH_4/O_2 on CH_4 and CO_2 conversions.

Table 1 shows the comparison of the performance for partial oxidation of CH_4 at the optimum conditions between the gliding arc, dielectric barrier, and corona discharge reactors. The gliding arc plasma systems appeared to show similar ranges of CH_4 conversion, H_2 and CO selectivities. Against other types of discharge, the gliding arc consumed much lower applied energy per unit flow rate than the dielectric barrier discharge system [22], but exhibited higher CH_4 conversion. Corona discharge [23] gave highest H_2/CO ratios of 2.5 and 3.4, compared to 18 – 2.0 found in this work.

TABLE 1: Comparison with literature on plasma assisted, partial oxidative reforming of methane

Parameters	**This study**			**[15]**		**[22]**		**[23]**	
discharge	Gliding arc			Gliding arc		Dielectric barrier		Corona	
feed, CH_4/CO_2	1	2.33	9	3	3	pure CH_4	pure CH_4	pure CH_4	pure CH_4
CH_4/O_2	3.4	2.2	1.2	3	3	2	10	3	5
flow rate (L/min)	1	1	1	0.15	0.15	0.02	0.02	n/a	n/a
AED (kJ/L)	6	6	6	n/a	n/a	15	15	n/a	n/a
power input (kW)	0.1	0.1	0.1	n/a	n/a	0.005	0.005	0.016	0.015
number of reactors	1	1	1	1	4	1	1	1	1
exit temperature (K)	523	543	573	473	473	n/a	n/a	373	373
H_2 yield (%)	13.6	13.9	25.3	n/a	n/a	4	5	n/a	n/a
CO yield (%)	7.6	10.0	23.1	n/a	n/a	6	6	n/a	13
H_2/CO	1.8	2.0	2.0	1.3	1.4	n/a	n/a	2.5	3.4
H_2 selectivity (%)	58.9	51.2	56.7	46	37	n/a	n/a	62	n/a
CO selectivity (%)	48.7	41.6	56.0	32	28	n/a	n/a	n/a	n/a
CH_4 conversion (%)	23.1	27.1	45.7	12	45	19	16	38	24

As far as energy consumption is concerned, the energy utilized in this work was calculated to be 6.0 kJ/L or MJ/m^3 of feed gas. For a biomethane with CH_4/CO_2 of 90:10, its calorific value is estimated about 38.3 MJ/kg or 28 MJ/m^3, assuming biomethane density of 0.73 kg/m^3. Hence, the energy utilized was approximately 20% of the energy contained in the feed gas. The current CH_4 conversion was high, around 45%, in comparison with other plasma sources reported in the literature. Nonetheless, to achieve higher conversion of methane in this plasma reactor setup, the following modifications may be needed; (i) using higher applied energy to generate more energetic active species to encourage higher conversion, (ii) combining the plasma reactor with a catalytic reformer, (iii) passing the reactants into many stages of the reactor in cascade, and (iv) increasing the feed gas injector size. The latter two modifications would increase total residence time of the feed gas within the reactor, which will result in higher conversion. So far, reactor development with regards to the gliding arc plasma reactor remains at laboratory level. Scaling up of the plasma reactor is not yet achieved in realization of industrial plants. Works on the previously mentioned modifications, investigation on other critical factors associated with fluid dynamics and transport properties, as well as process modeling and analysis should be further carried out, prior to development into pilot and industrial units.

4. Conclusion

Gliding arc plasma is a promising technology for biogas reforming into useful products. In this work, a gliding arc discharge system was utilized in partial oxidation of biogas with air to generate synthesis gas. Effects of varying composition and CH_4/O_2 molar ratio on performance of the plasma reformer were investigated. For very low CH_4/O_2 molar ratio, CH_4 conversion and H_2 yield were high. At higher CH_4/O_2 molar ratios, CH_4 conversion and synthesis gas yields decreased. The optimum condition of the gliding arc plasma system was found at CH_4/CO_2 of 90:10 and CH_4/O_2 of 1.2 for the maximum CH_4 conversion. The gliding arc plasma system proved to successfully generate high synthesis gas yields at low energy consumption.

Ackno wledgement

This work is financially supported by the Commission on Higher Education under National Research University project. Support from the Thailand Research Fund for PT under a Royal Golden Jubilee PhD program is acknowledged.

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C.2 Papers in International Conferences

1) Khongkrapan, P., Tippayawong, N., and Kiatsiriroat, T., (2012). "Design and Simulation of a Microwave Plasma Reactor for Synthetic Gas Production from Biomass", International Conference on Green and Sustainable Innovation (ICGSI), May 24 – 26, 2012, Chiang Mai, Thailand**.**

2) Khongkrapan, P., Tippayawong, N., and Kiatsiriroat, T., (2013). "Thermochemical Conversion of Waste Papers to Fuel Gas in a Microwave Plasma Reactor", International Conference on Future Environment and Energy (ICFEE), February 24-25, Rome, Italy.

3) Chaiya, E., Khongkrapan, P., and Tippayawong, N., "Plasma Assisted Dry Reforming of Biomethane into Synthetic Gas", Joint Symposium on Mechanical - Industrial Engineering, and Robotics (MIER), November 14-17, 2013, Chiang Mai, Thailand.

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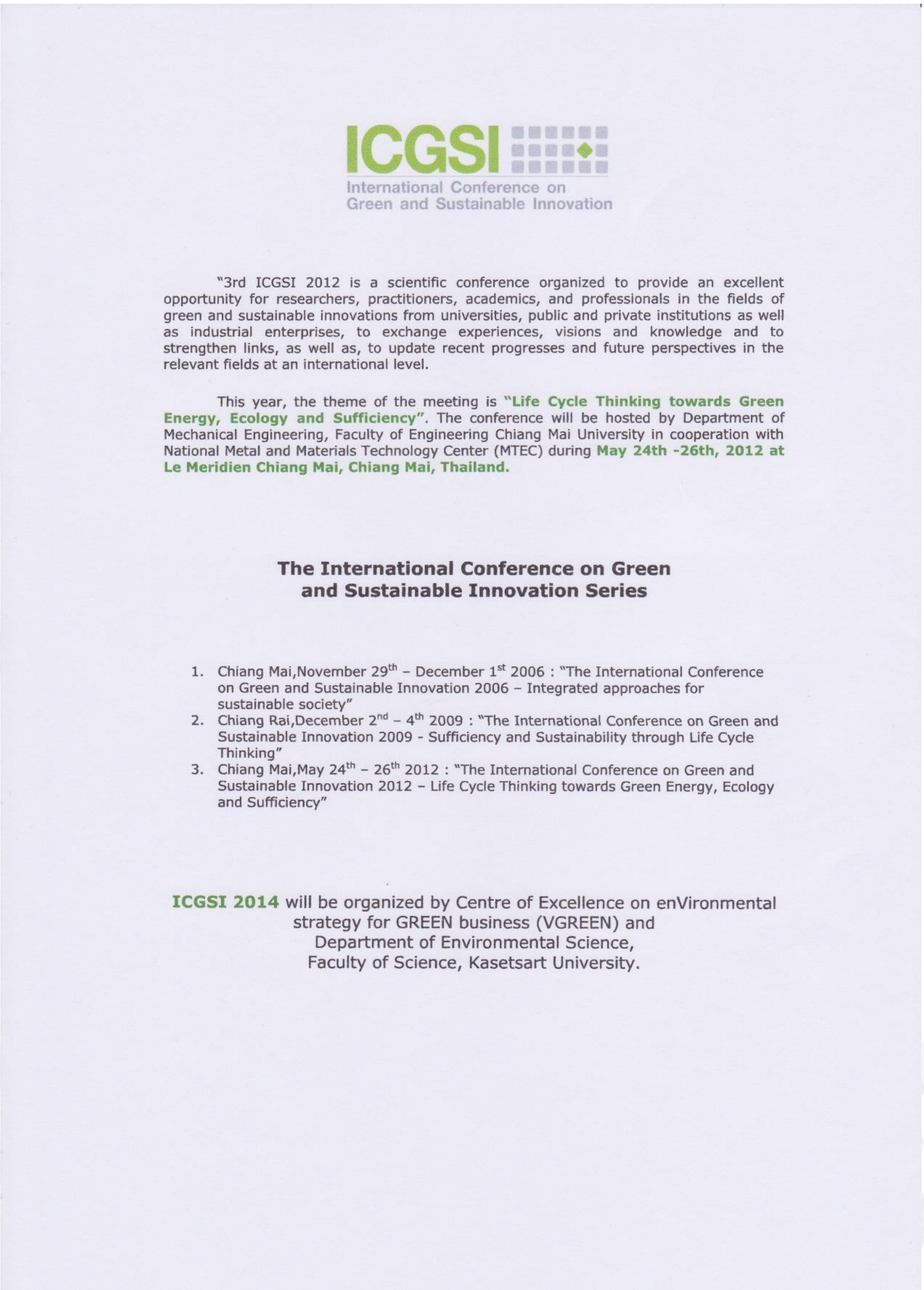

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This year, the theme of the meeting is **"Life Cycle Thinking towards Green Energy, Ecology and Sufficiency"**. The conference will be hosted by Department of Mechanical Engineering, Faculty of Engineering Chiang Mai University in cooperation with National Metal and Materials Technology Center (MTEC) during **May 24th -26th, 2012 at Le Meridien Chiang Mai, Chiang Mai, Thailand.**

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1. Chiang Mai,November 29th – December 1st 2006 : "The International Conference on Green and Sustainable Innovation 2006 – Integrated approaches for sustainable society"
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ICGSI 2014 will be organized by Centre of Excellence on enVironmental strategy for GREEN business (VGREEN) and
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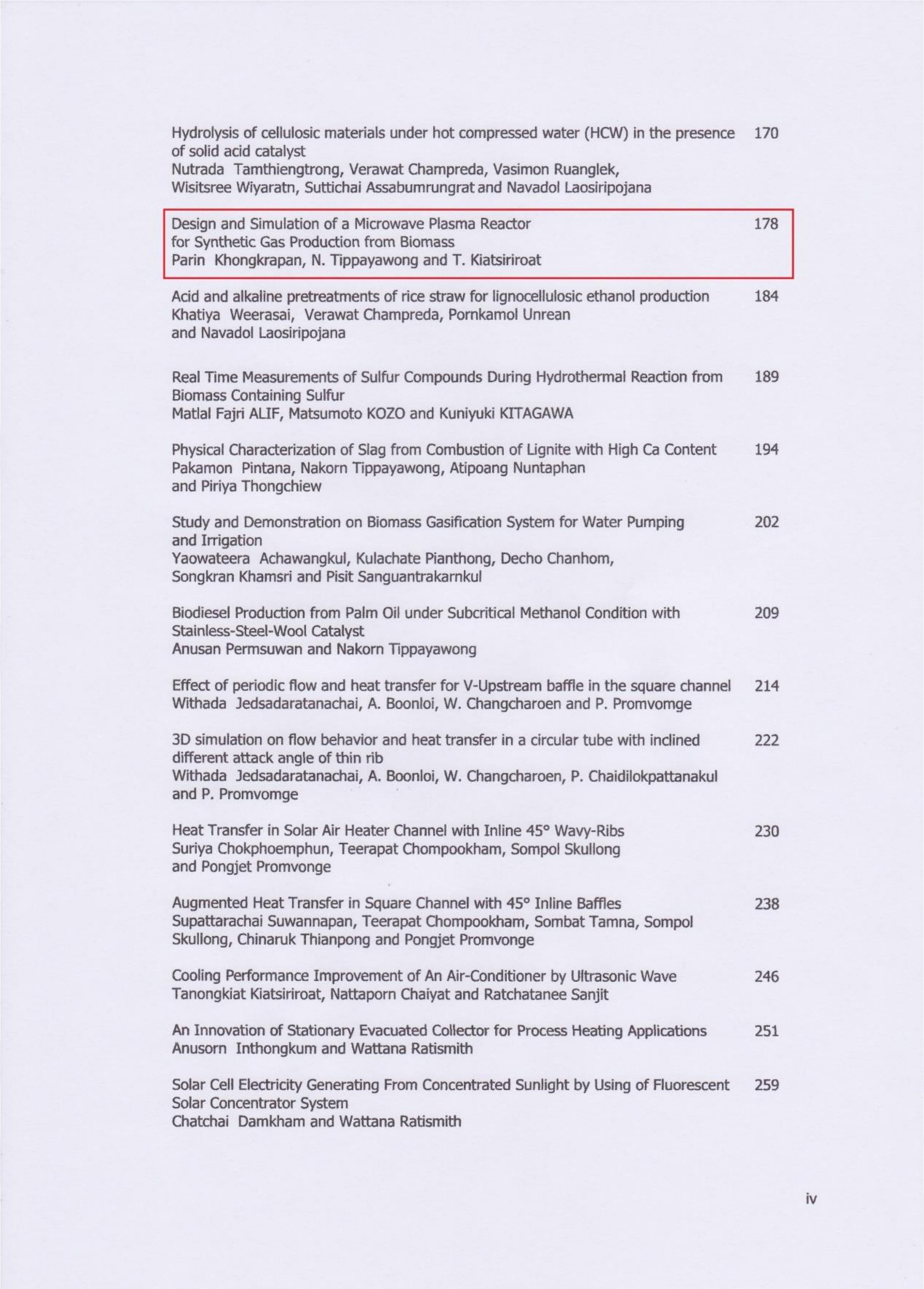

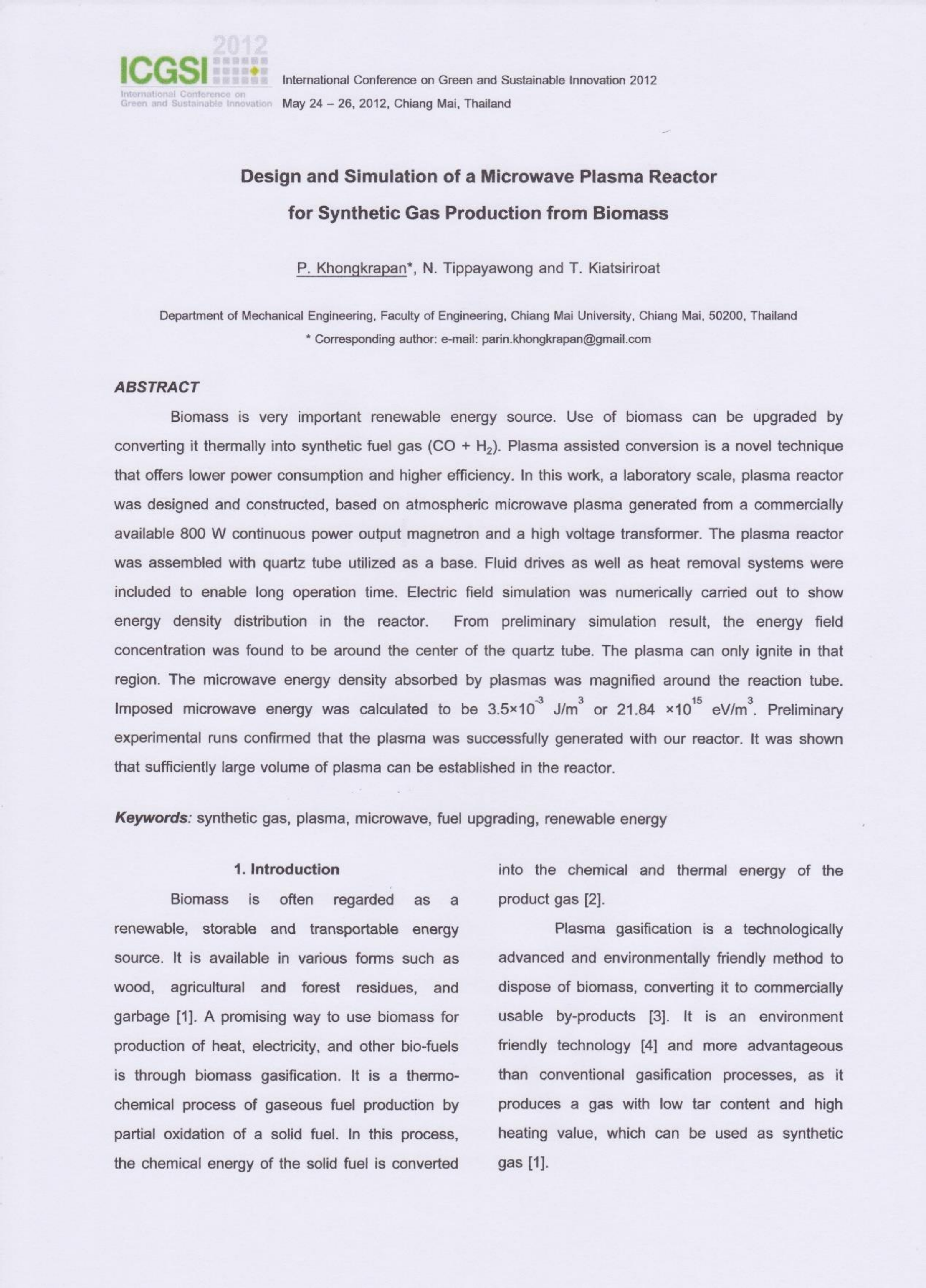

International Conference on Green and Sustainable Innovation 2012
May 24 – 26, 2012, Chiang Mai, Thailand

Design and Simulation of a Microwave Plasma Reactor for Synthetic Gas Production from Biomass

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ABSTRACT

Biomass is very important renewable energy source. Use of biomass can be upgraded by converting it thermally into synthetic fuel gas ($CO + H_2$). Plasma assisted conversion is a novel technique that offers lower power consumption and higher efficiency. In this work, a laboratory scale, plasma reactor was designed and constructed, based on atmospheric microwave plasma generated from a commercially available 800 W continuous power output magnetron and a high voltage transformer. The plasma reactor was assembled with quartz tube utilized as a base. Fluid drives as well as heat removal systems were included to enable long operation time. Electric field simulation was numerically carried out to show energy density distribution in the reactor. From preliminary simulation result, the energy field concentration was found to be around the center of the quartz tube. The plasma can only ignite in that region. The microwave energy density absorbed by plasmas was magnified around the reaction tube. Imposed microwave energy was calculated to be 3.5×10^{-3} J/m^3 or 21.84 $\times10^{15}$ eV/m^3. Preliminary experimental runs confirmed that the plasma was successfully generated with our reactor. It was shown that sufficiently large volume of plasma can be established in the reactor.

Keywords: synthetic gas, plasma, microwave, fuel upgrading, renewable energy

1. Introduction

Biomass is often regarded as a renewable, storable and transportable energy source. It is available in various forms such as wood, agricultural and forest residues, and garbage [1]. A promising way to use biomass for production of heat, electricity, and other bio-fuels is through biomass gasification. It is a thermo-chemical process of gaseous fuel production by partial oxidation of a solid fuel. In this process, the chemical energy of the solid fuel is converted into the chemical and thermal energy of the product gas [2].

Plasma gasification is a technologically advanced and environmentally friendly method to dispose of biomass, converting it to commercially usable by-products [3]. It is an environment friendly technology [4] and more advantageous than conventional gasification processes, as it produces a gas with low tar content and high heating value, which can be used as synthetic gas [1].

Traditionally, plasma gasification process uses arc plasma torch as the heat source. It requires high electrical energy to keep high temperatures in the plasma discharge. The high energy consumption is the main drawback of arc plasma. Microwave plasma is non-thermal, easy to control and requires low power [5]. It can be applied in many different ways by changing the design of power transmission and reactor [6]. Hydrocarbon reforming using microwave plasma is an effective way to solve these disadvantages [7-10].

In this work, an atmospheric laboratory scale microwave plasma reactor was designed and simulated. The microwave plasma can be generated using magnetron in typical household microwave ovens. It is simple, compact and economical [11]. The majority of the design of a microwave plasma reactor for synthetic gas production from biomass focused on the microwave concentration point and plasma temperature in the reactor. The model, validated using simulation and experimental data, was used to identify the optimal operating conditions of the microwave plasma gasification system.

2. Design of the Microwave Plasma System

The microwave plasma system in this work is the waveguide discharge type [12]. It was assembled from a microwave source, waveguide, reactor and gas treatment equipment. The main objective of the plasma system is to generate the plasma in the reaction zone of the reactor.

Fig. 1 shows a schematic view of the microwave plasma reactor system in this work. It consists of the 800 W, 2.45 GHz microwave generator with the air cooling system. The D-band frequency of 2.20 to 3.30 GHz WR-340 rectangular 43.2x86.4 mm waveguide components included a resonator, a reactor and the water cooling system.

A rectangular waveguide was connected to reactor. The microwave source was placed at the end of the waveguide. The reactor was inserted with the discharge tube vertically perpendicular to the top wall. The center axis of discharge tube made of quartz with an internal diameter 27 mm was located at one-quarter wavelength away from the end of the reactor. Distance between the discharge tube and the end plate of the reactor was fixed at 3 cm (122 mm wavelength). This location was calculated from high frequency structure simulator (HFSS). Microwave power was either absorbed by the plasma or confined within a waveguide and a reactor. There was no safety problem with radiated power [11].

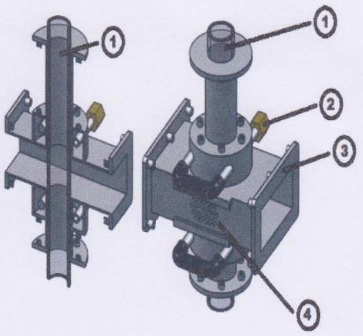
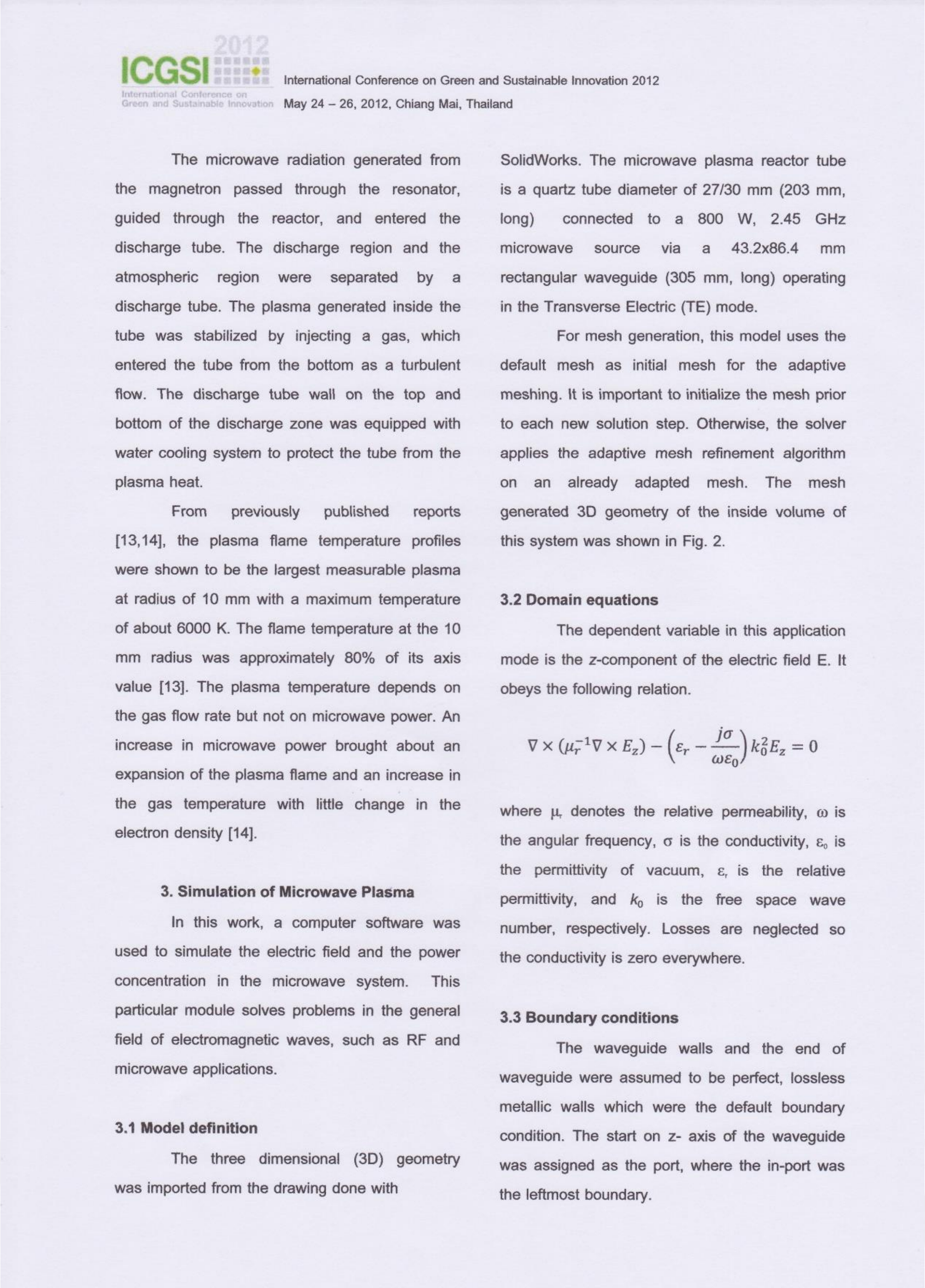


Fig. 1 Schematic view of the microwave plasma reactor system (1) quartz tube (2) cooling water system (3) waveguide connector and (4) reaction observer channel



The microwave radiation generated from the magnetron passed through the resonator, guided through the reactor, and entered the discharge tube. The discharge region and the atmospheric region were separated by a discharge tube. The plasma generated inside the tube was stabilized by injecting a gas, which entered the tube from the bottom as a turbulent flow. The discharge tube wall on the top and bottom of the discharge zone was equipped with water cooling system to protect the tube from the plasma heat.

From previously published reports [13,14], the plasma flame temperature profiles were shown to be the largest measurable plasma at radius of 10 mm with a maximum temperature of about 6000 K. The flame temperature at the 10 mm radius was approximately 80% of its axis value [13]. The plasma temperature depends on the gas flow rate but not on microwave power. An increase in microwave power brought about an expansion of the plasma flame and an increase in the gas temperature with little change in the electron density [14].

3. Simulation of Microwave Plasma

In this work, a computer software was used to simulate the electric field and the power concentration in the microwave system. This particular module solves problems in the general field of electromagnetic waves, such as RF and microwave applications.

3.1 Model definition

The three dimensional (3D) geometry was imported from the drawing done with SolidWorks. The microwave plasma reactor tube is a quartz tube diameter of 27/30 mm (203 mm, long) connected to a 800 W, 2.45 GHz microwave source via a 43.2x86.4 mm rectangular waveguide (305 mm, long) operating in the Transverse Electric (TE) mode.

For mesh generation, this model uses the default mesh as initial mesh for the adaptive meshing. It is important to initialize the mesh prior to each new solution step. Otherwise, the solver applies the adaptive mesh refinement algorithm on an already adapted mesh. The mesh generated 3D geometry of the inside volume of this system was shown in Fig. 2.

3.2 Domain equations

The dependent variable in this application mode is the *z*-component of the electric field E. It obeys the following relation.

$$\nabla \times (\mu_r^{-1} \nabla \times E_z) - \left(\varepsilon_r - \frac{j\sigma}{\omega \varepsilon_0}\right) k_0^2 E_z = 0$$

where μ_r denotes the relative permeability, ω is the angular frequency, σ is the conductivity, ε_0 is the permittivity of vacuum, ε_r is the relative permittivity, and k_0 is the free space wave number, respectively. Losses are neglected so the conductivity is zero everywhere.

3.3 Boundary conditions

The waveguide walls and the end of waveguide were assumed to be perfect, lossless metallic walls which were the default boundary condition. The start on z- axis of the waveguide was assigned as the port, where the in-port was the leftmost boundary.

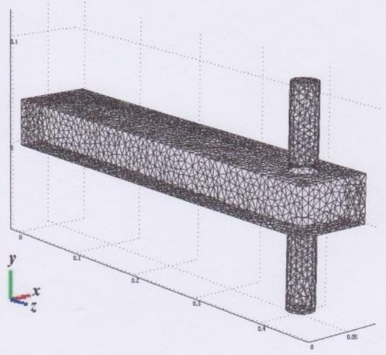


Fig. 2 Geometry with mesh generated used in the simulations

The two ends of the reactor tube were assigned as ports, where the out-port were the topmost and the bottommost boundary.

4. Simulation Result

The energy density and the electric field concentration are the key in this model as they are main parameters of the plasma system, designed in this work. The symmetry cut was applied vertically through the quartz tube, waveguide and the end plate. Fig. 3 shows contours of the electric fields. The electric field was found to concentrate at the reaction zone in the quartz tube that located at one-quarter wavelength away from the end of the reactor. This simulation results indicated the best location of the reaction tube for this design.

Fig. 4 illustrates contours of the energy density, which only the cross section of the waveguide and the quartz tube are shown. Energy field concentration was found to be around the center of the quartz tube. It should be noted that the plasma can only ignite in this region. The microwave energy density that absorbed by plasmas was magnified around the reactor tube. Imposed microwave energy in this case was calculated to be 3.5×10^{-3} J/m^3 or 21.84×10^{15} eV/m^3. It seemed to be enough for the first ionization of air that requires at least 10–15 eV [15].

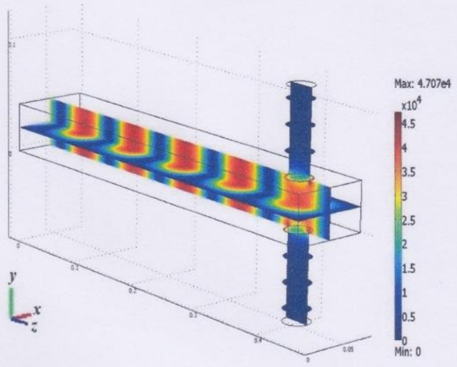


Fig. 3 Contours of the electric field

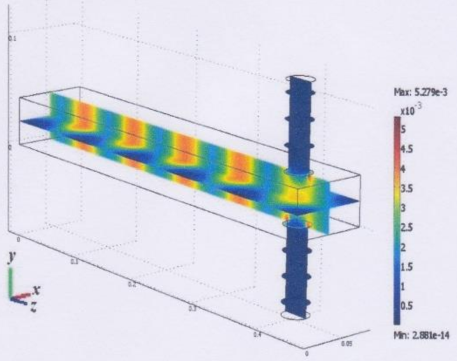


Fig. 4 Contours of the energy density

5. Preliminary Experimental Runs

The microwave plasma reactor was operated at atmospheric pressure. The microwave was powered by a 2.45 GHz generator, whose continuous output power is constant at 800 W. The discharge takes place inside a quartz tube with internal/external diameter of 27/30 mm, which is inserted vertically and perpendicularly to the waveguide wider wall. To ignite the plasma in the reactor, the igniter was used. After ignition, the plasma can be observed in the quartz reactor tube.

For preliminary test run, there was no carrier gas and biomass fed to the reactor. The test run was performed for the microwave plasma system to confirm that the generated plasma can be achieved with our reactor. Fig. 5 shows the air plasma emission in the reactor tube.

The plasma was characterized by high intensity light emission. The plasma flame temperature is expected to be around 1000-2000 K [15] and will be confirmed by the optical emission spectroscopy method in the future study.

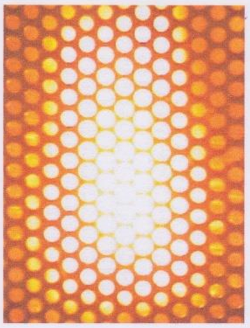


Fig. 5 Plasma emission in the reactor tube

6. Concluding Remarks

In this work, a microwave plasma reactor was designed, constructed and tested. Electric field and energy density in the reactor tube were also simulated. From preliminary study, it was demonstrated that the high volume plasma can be generated.

Next step, we are going to run the reactor to generate synthetic gas from biomass. A carrier gas will be injected into the reactor at various flow rates. Plasmochemical reaction in the reactor is expected to enable synthetic gas production.

7. Acknowledgement

The authors would to acknowledge the financial support provided by the Office of the Higher Education Commission via the National Research University program. Assistance from Thermal Systems Research Unit, Department of Mechanical Engineering, Faculty of Engineering, Chiang Mai University was appreciated. Many thanks also go to colleagues of Sustainable Renewable Energy Laboratory for technical assistance.

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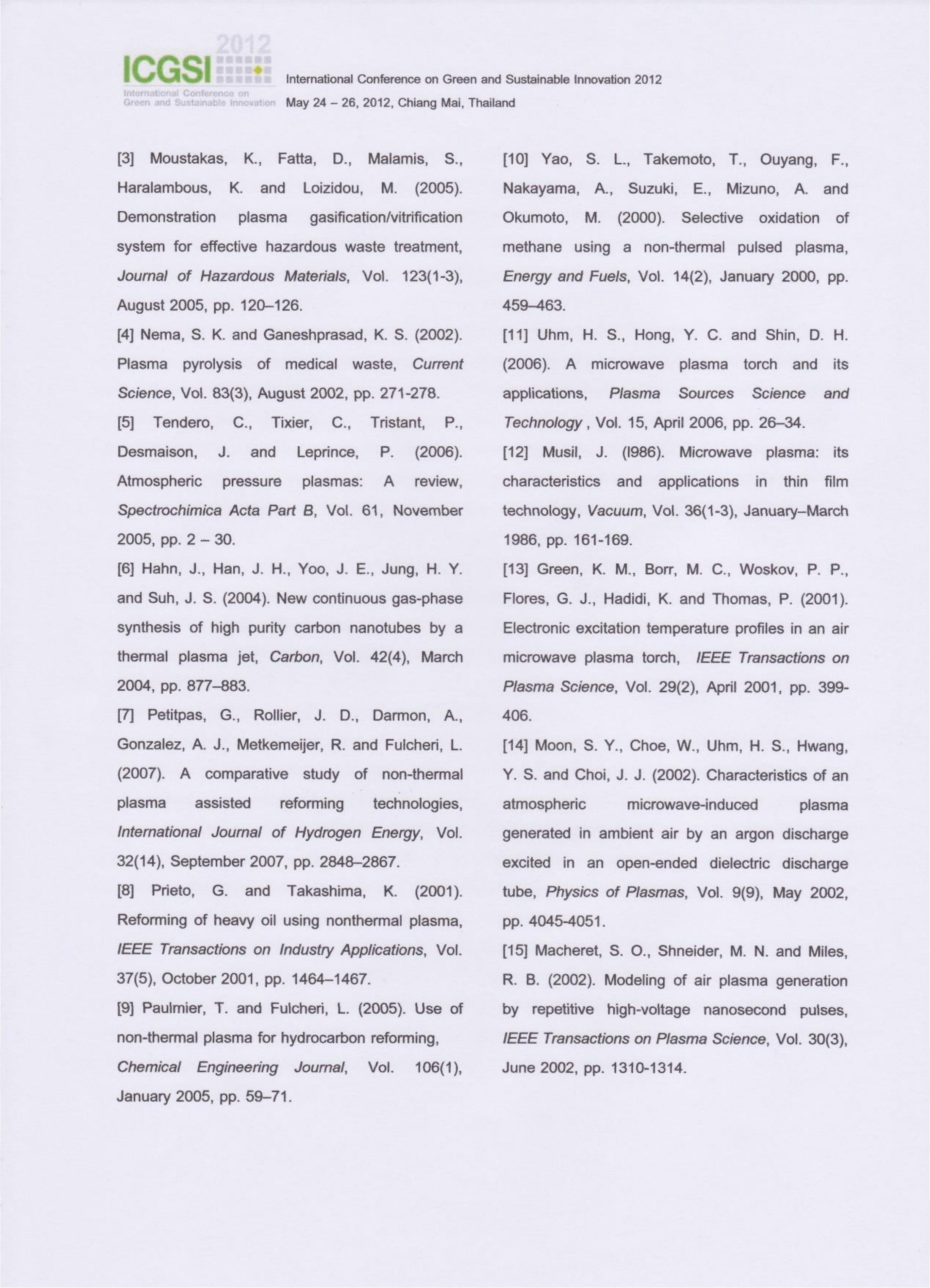

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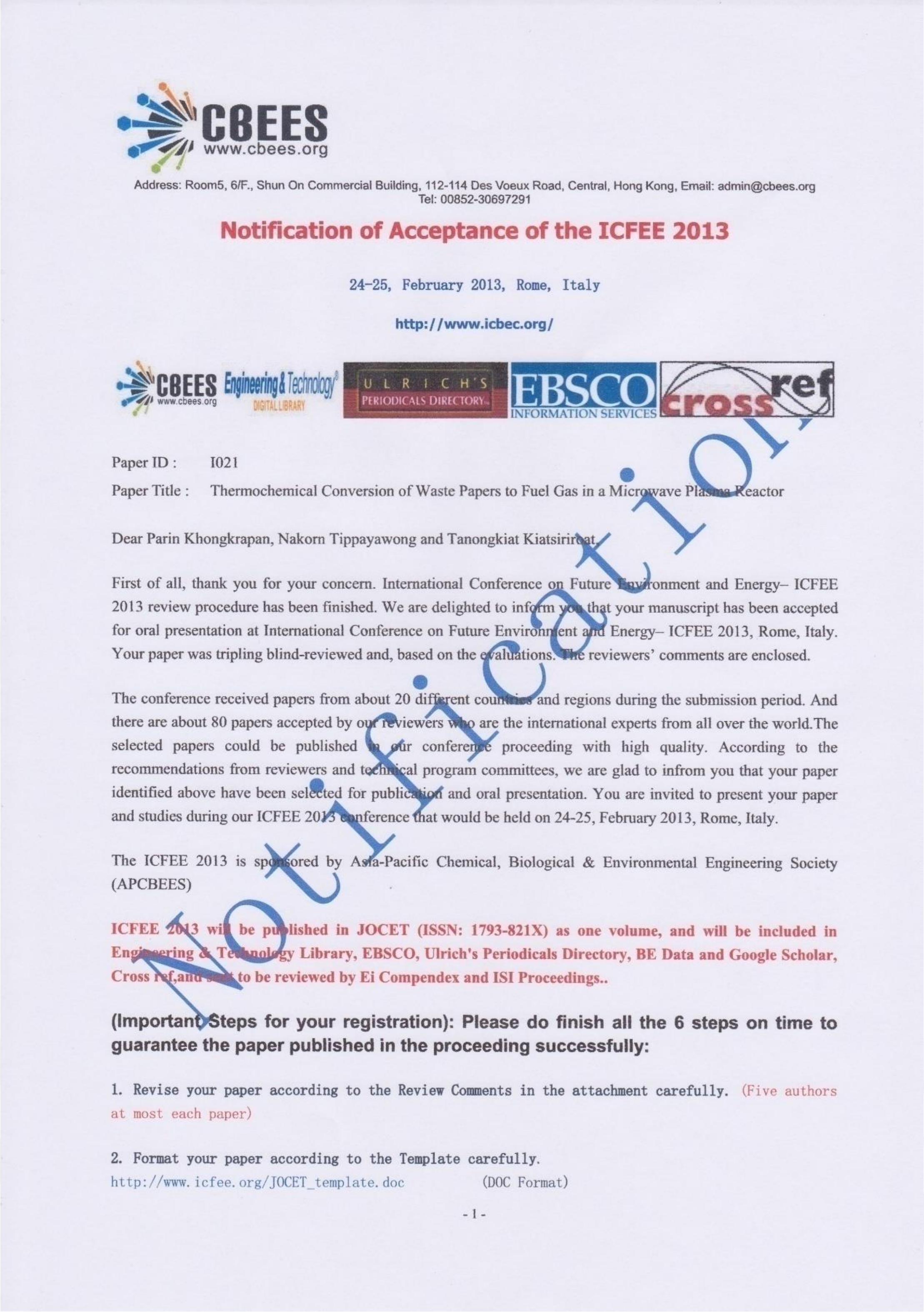

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24-25, February 2013, Rome, Italy

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Paper ID : I021
Paper Title : Thermochemical Conversion of Waste Papers to Fuel Gas in a Microwave Plasma Reactor

Dear Parin Khongkrapan, Nakorn Tippayawong and Tanongkiat Kiatsiriroat,

First of all, thank you for your concern. International Conference on Future Environment and Energy– ICFEE 2013 review procedure has been finished. We are delighted to inform you that your manuscript has been accepted for oral presentation at International Conference on Future Environment and Energy– ICFEE 2013, Rome, Italy. Your paper was tripling blind-reviewed and, based on the evaluations. The reviewers' comments are enclosed.

The conference received papers from about 20 different countries and regions during the submission period. And there are about 80 papers accepted by our reviewers who are the international experts from all over the world.The selected papers could be published in our conference proceeding with high quality. According to the recommendations from reviewers and technical program committees, we are glad to infrom you that your paper identified above have been selected for publication and oral presentation. You are invited to present your paper and studies during our ICFEE 2013 conference that would be held on 24-25, February 2013, Rome, Italy.

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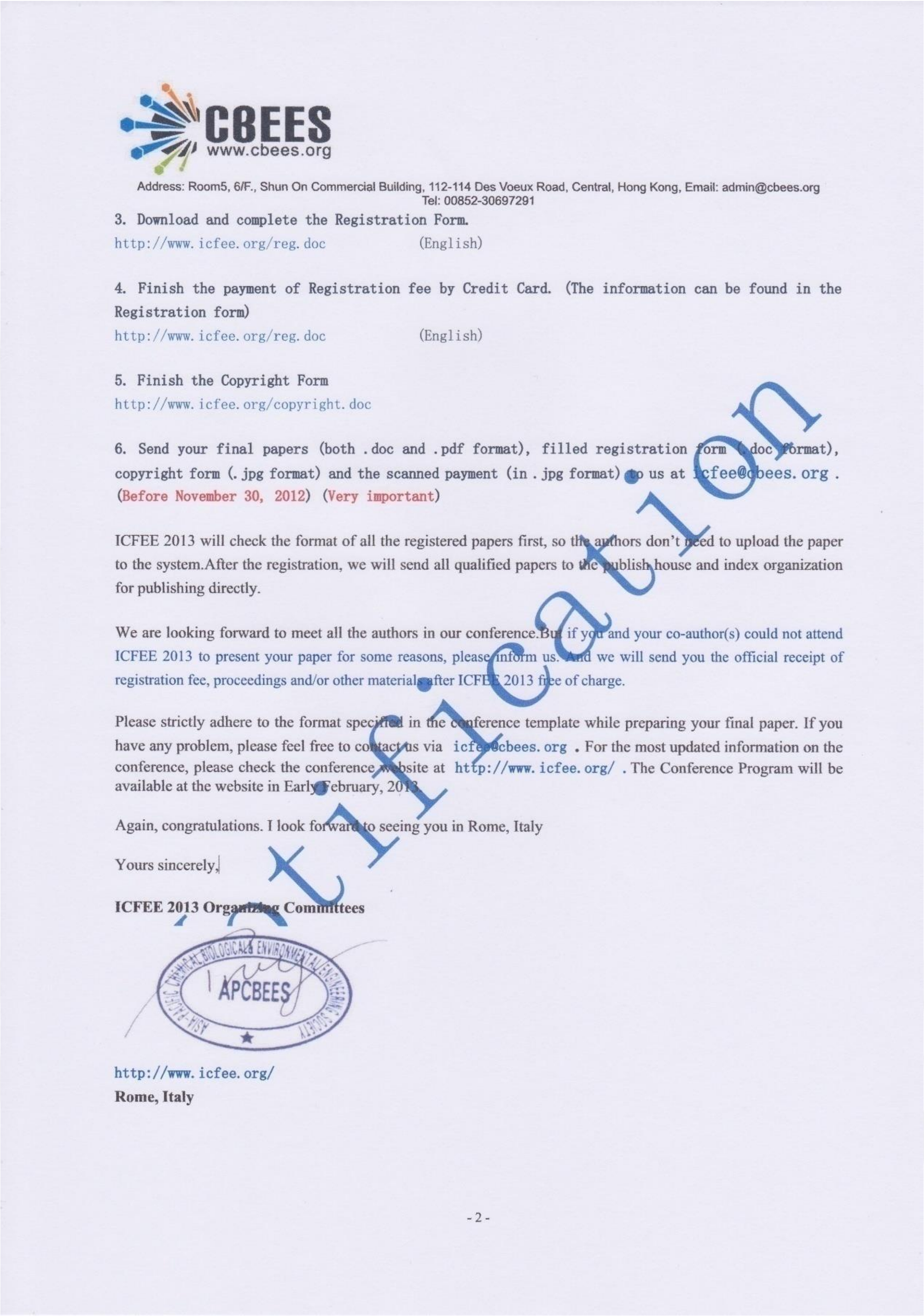

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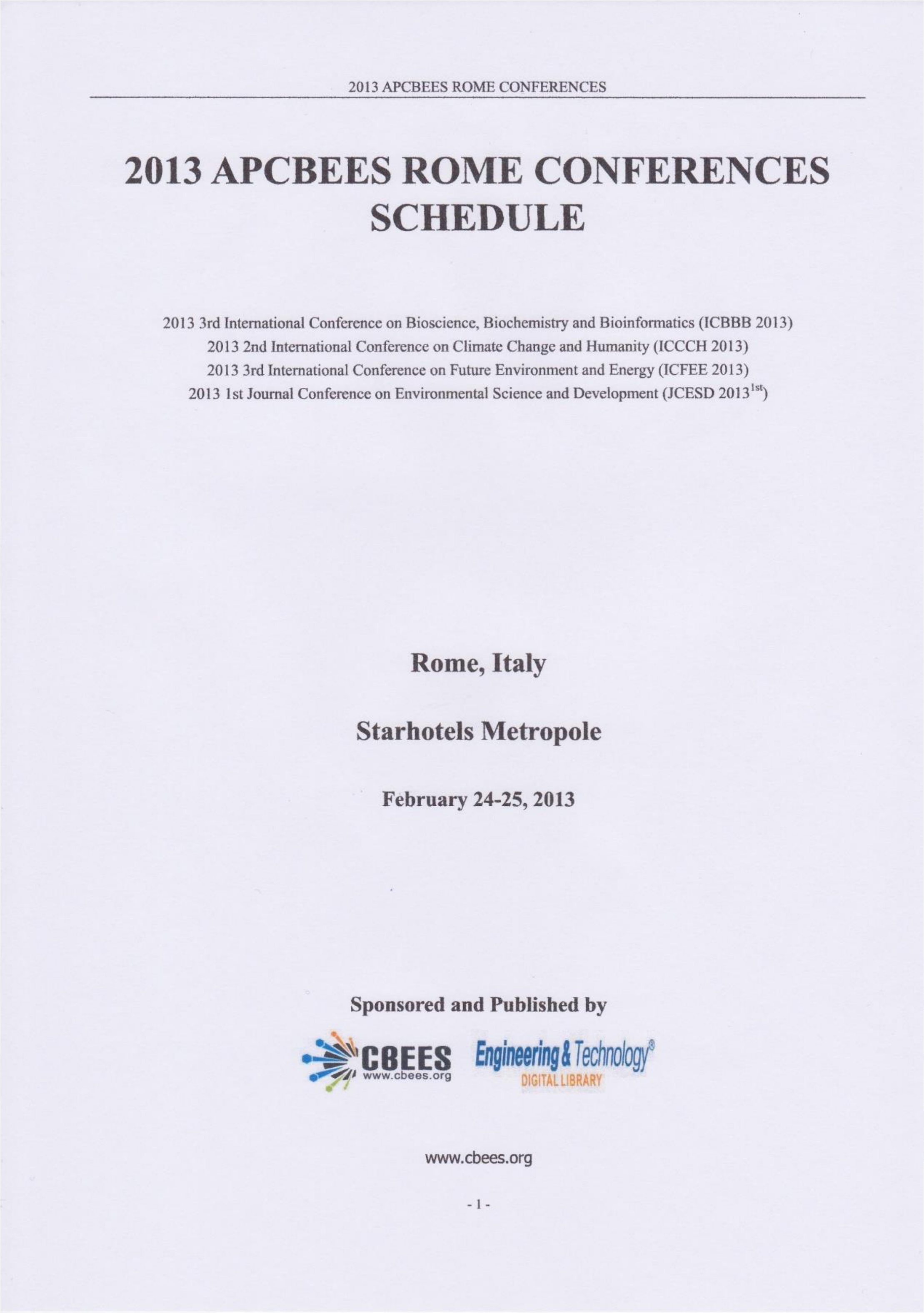

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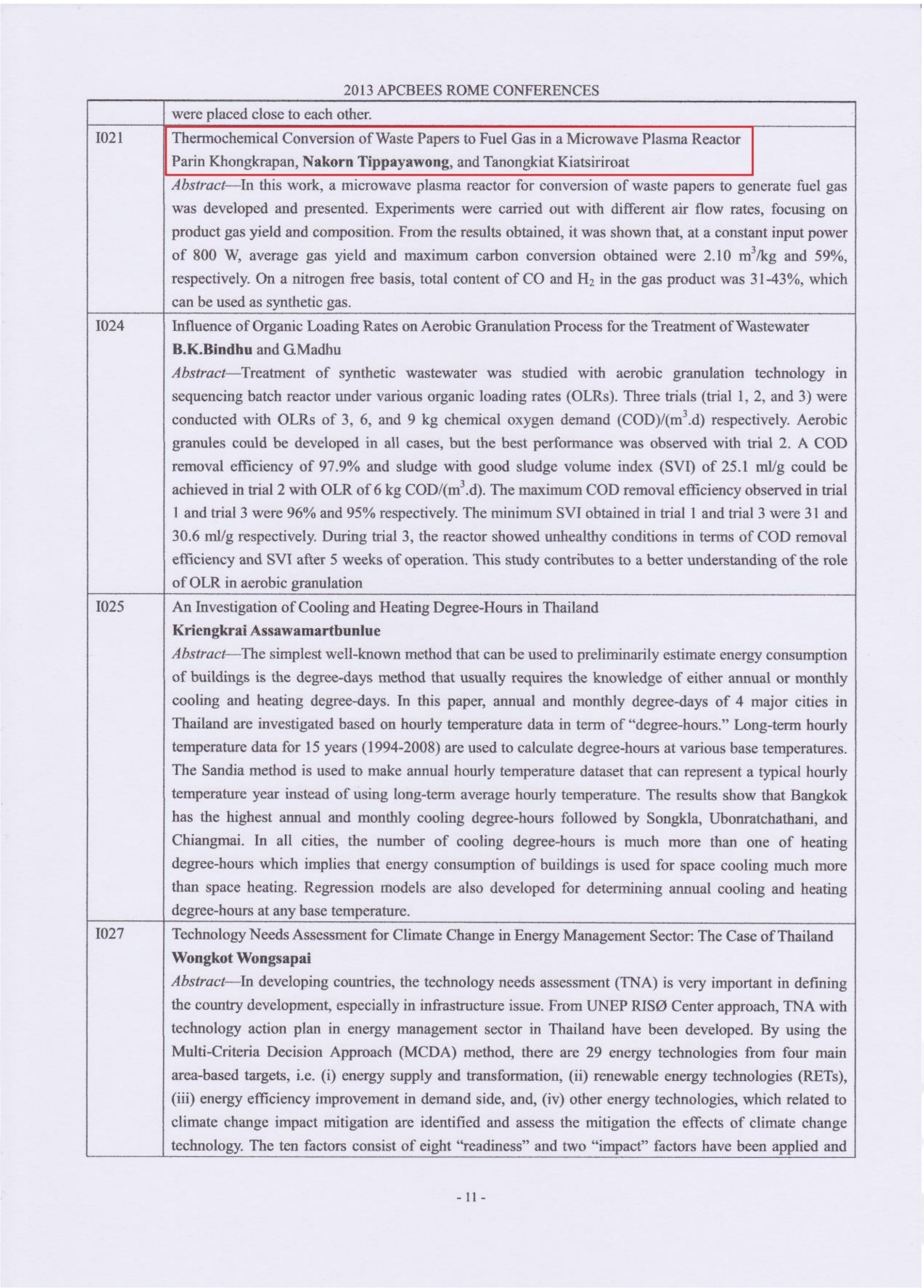
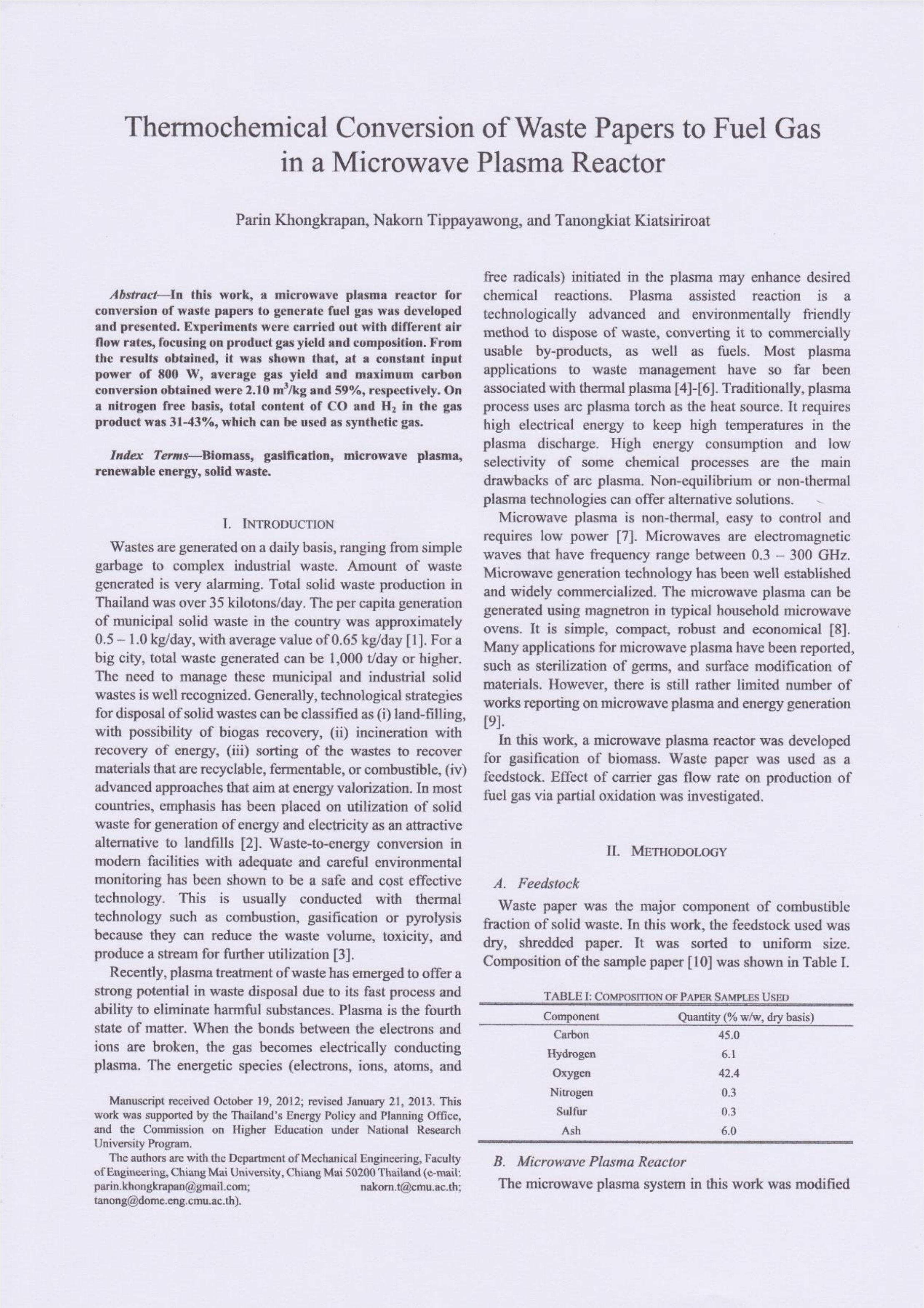


	were placed close to each other.
I021	Thermochemical Conversion of Waste Papers to Fuel Gas in a Microwave Plasma Reactor Parin Khongkrapan, **Nakorn Tippayawong**, and Tanongkiat Kiatsiriroat *Abstract*—In this work, a microwave plasma reactor for conversion of waste papers to generate fuel gas was developed and presented. Experiments were carried out with different air flow rates, focusing on product gas yield and composition. From the results obtained, it was shown that, at a constant input power of 800 W, average gas yield and maximum carbon conversion obtained were 2.10 m^3/kg and 59%, respectively. On a nitrogen free basis, total content of CO and H_2 in the gas product was 31-43%, which can be used as synthetic gas.
I024	Influence of Organic Loading Rates on Aerobic Granulation Process for the Treatment of Wastewater **B.K.Bindhu** and G.Madhu *Abstract*—Treatment of synthetic wastewater was studied with aerobic granulation technology in sequencing batch reactor under various organic loading rates (OLRs). Three trials (trial 1, 2, and 3) were conducted with OLRs of 3, 6, and 9 kg chemical oxygen demand (COD)/(m^3.d) respectively. Aerobic granules could be developed in all cases, but the best performance was observed with trial 2. A COD removal efficiency of 97.9% and sludge with good sludge volume index (SVI) of 25.1 ml/g could be achieved in trial 2 with OLR of 6 kg COD/(m^3.d). The maximum COD removal efficiency observed in trial 1 and trial 3 were 96% and 95% respectively. The minimum SVI obtained in trial 1 and trial 3 were 31 and 30.6 ml/g respectively. During trial 3, the reactor showed unhealthy conditions in terms of COD removal efficiency and SVI after 5 weeks of operation. This study contributes to a better understanding of the role of OLR in aerobic granulation
I025	An Investigation of Cooling and Heating Degree-Hours in Thailand **Kriengkrai Assawamartbunlue** *Abstract*—The simplest well-known method that can be used to preliminarily estimate energy consumption of buildings is the degree-days method that usually requires the knowledge of either annual or monthly cooling and heating degree-days. In this paper, annual and monthly degree-days of 4 major cities in Thailand are investigated based on hourly temperature data in term of "degree-hours." Long-term hourly temperature data for 15 years (1994-2008) are used to calculate degree-hours at various base temperatures. The Sandia method is used to make annual hourly temperature dataset that can represent a typical hourly temperature year instead of using long-term average hourly temperature. The results show that Bangkok has the highest annual and monthly cooling degree-hours followed by Songkla, Ubonratchathani, and Chiangmai. In all cities, the number of cooling degree-hours is much more than one of heating degree-hours which implies that energy consumption of buildings is used for space cooling much more than space heating. Regression models are also developed for determining annual cooling and heating degree-hours at any base temperature.
I027	Technology Needs Assessment for Climate Change in Energy Management Sector: The Case of Thailand **Wongkot Wongsapai** *Abstract*—In developing countries, the technology needs assessment (TNA) is very important in defining the country development, especially in infrastructure issue. From UNEP RISØ Center approach, TNA with technology action plan in energy management sector in Thailand have been developed. By using the Multi-Criteria Decision Approach (MCDA) method, there are 29 energy technologies from four main area-based targets, i.e. (i) energy supply and transformation, (ii) renewable energy technologies (RETs), (iii) energy efficiency improvement in demand side, and, (iv) other energy technologies, which related to climate change impact mitigation are identified and assess the mitigation the effects of climate change technology. The ten factors consist of eight "readiness" and two "impact" factors have been applied and



Thermochemical Conversion of Waste Papers to Fuel Gas in a Microwave Plasma Reactor

Parin Khongkrapan, Nakorn Tippayawong, and Tanongkiat Kiatsiriroat

***Abstract*—In this work, a microwave plasma reactor for conversion of waste papers to generate fuel gas was developed and presented. Experiments were carried out with different air flow rates, focusing on product gas yield and composition. From the results obtained, it was shown that, at a constant input power of 800 W, average gas yield and maximum carbon conversion obtained were 2.10 m^3/kg and 59%, respectively. On a nitrogen free basis, total content of CO and H_2 in the gas product was 31-43%, which can be used as synthetic gas.**

***Index Terms*—Biomass, gasification, microwave plasma, renewable energy, solid waste.**

I. INTRODUCTION

Wastes are generated on a daily basis, ranging from simple garbage to complex industrial waste. Amount of waste generated is very alarming. Total solid waste production in Thailand was over 35 kilotons/day. The per capita generation of municipal solid waste in the country was approximately 0.5 – 1.0 kg/day, with average value of 0.65 kg/day [1]. For a big city, total waste generated can be 1,000 t/day or higher. The need to manage these municipal and industrial solid wastes is well recognized. Generally, technological strategies for disposal of solid wastes can be classified as (i) land-filling, with possibility of biogas recovery, (ii) incineration with recovery of energy, (iii) sorting of the wastes to recover materials that are recyclable, fermentable, or combustible, (iv) advanced approaches that aim at energy valorization. In most countries, emphasis has been placed on utilization of solid waste for generation of energy and electricity as an attractive alternative to landfills [2]. Waste-to-energy conversion in modern facilities with adequate and careful environmental monitoring has been shown to be a safe and cost effective technology. This is usually conducted with thermal technology such as combustion, gasification or pyrolysis because they can reduce the waste volume, toxicity, and produce a stream for further utilization [3].

Recently, plasma treatment of waste has emerged to offer a strong potential in waste disposal due to its fast process and ability to eliminate harmful substances. Plasma is the fourth state of matter. When the bonds between the electrons and ions are broken, the gas becomes electrically conducting plasma. The energetic species (electrons, ions, atoms, and free radicals) initiated in the plasma may enhance desired chemical reactions. Plasma assisted reaction is a technologically advanced and environmentally friendly method to dispose of waste, converting it to commercially usable by-products, as well as fuels. Most plasma applications to waste management have so far been associated with thermal plasma [4]-[6]. Traditionally, plasma process uses arc plasma torch as the heat source. It requires high electrical energy to keep high temperatures in the plasma discharge. High energy consumption and low selectivity of some chemical processes are the main drawbacks of arc plasma. Non-equilibrium or non-thermal plasma technologies can offer alternative solutions.

Microwave plasma is non-thermal, easy to control and requires low power [7]. Microwaves are electromagnetic waves that have frequency range between 0.3 – 300 GHz. Microwave generation technology has been well established and widely commercialized. The microwave plasma can be generated using magnetron in typical household microwave ovens. It is simple, compact, robust and economical [8]. Many applications for microwave plasma have been reported, such as sterilization of germs, and surface modification of materials. However, there is still rather limited number of works reporting on microwave plasma and energy generation [9].

In this work, a microwave plasma reactor was developed for gasification of biomass. Waste paper was used as a feedstock. Effect of carrier gas flow rate on production of fuel gas via partial oxidation was investigated.

II. METHODOLOGY

A. Feedstock

Waste paper was the major component of combustible fraction of solid waste. In this work, the feedstock used was dry, shredded paper. It was sorted to uniform size. Composition of the sample paper [10] was shown in Table I.

TABLE I: COMPOSITION OF PAPER SAMPLES USED

Component	Quantity (% w/w, dry basis)
Carbon	45.0
Hydrogen	6.1
Oxygen	42.4
Nitrogen	0.3
Sulfur	0.3
Ash	6.0

B. Microwave Plasma Reactor

The microwave plasma system in this work was modified

Manuscript received October 19, 2012; revised January 21, 2013. This work was supported by the Thailand's Energy Policy and Planning Office, and the Commission on Higher Education under National Research University Program.

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from a commercial microwave oven. It consists of the 800 W, 2.45 GHz microwave generator, a cylindrical tube reactor, and auxiliary electrical equipment. The microwave radiation generated from the magnetron passed and guided through the oven cavity, and entered the discharge quartz tube centrally located inside the oven cavity. The plasma generated inside the tube was stabilized by injecting a carrier gas, which entered the tube from the bottom as a turbulent flow.

C. *Experimental Setup and Procedure*

The experimental setup is shown schematically in Fig. 1, equipped with gas cylinders for air and argon, connected to gas flow regulators.

A gas collection module was used for fuel gas conditioning and tar capture. It consists of a series of filter, impinger bottles containing a solvent for tar absorption placed in cold baths, and a moisture trap. Each impinger bottle was filled with approximately 100 ml of isopropanol. The gas flow rates were measured with flow meters. The cool, dry, clean gas was sampled using gas bags and analyzed on a Shimadzu model GC-8A gas chromatography fitted with a ShinCarbon ST Micropacked column and a thermal conductivity detector, for measuring volumetric concentration of H_2, O_2, N_2, CH_4, CO, CO_2. Standard gas mixtures were used for quantitative calibration.

Test and operating conditions were summarized in Table II. Initially, the quartz tube was loaded with shredded paper mass of 5 g, before mounted inside the oven. Carrier gas was fed from the bottom of the reactor, varying between 1 - 4 lpm. The microwave generator was then switched on to start the reaction, and run for 4 min. Gas collection was continually carried out. Solid residues were collected and weighed after each run. Each test was repeated in triplicate. Average values were reported.

D. *Data Analysis*

The following parameters are calculated [11];
Specific gas yield:

$$SGY = V_{gas} / W_B \qquad (1)$$

Carbon conversion efficiency:

$$n_c = \frac{V_{gas} \times \sum x_i \times (12/22.4)}{W_B \times (1 - C_A) \times C_c} \times 100\% \qquad (2)$$

Lower heating value:

$$LHV = \sum x_i LHV_i \qquad (3)$$

where V_{gas} is total volume of product gas generated, W_B is converted mass of solid material, x_i is volume fraction of fuel component of product gas, C_A is carbon content in residue, C_C is carbon content in biomass, and LHV_i is the corresponding heating values of the gas component.

III. RESULTS AND DISCUSSIONS

A. *Reactor Operation*

The microwave plasma reactor was operated at atmospheric pressure. The microwave was powered by a 2.45 GHz generator, whose continuous output power was constant at 800 W. The discharge takes place inside a quartz tube with internal/external diameters of 27/30 mm and length of 250 mm, inserted vertically and perpendicularly to the oven cavity. The plasma can be observed in the quartz reactor tube.

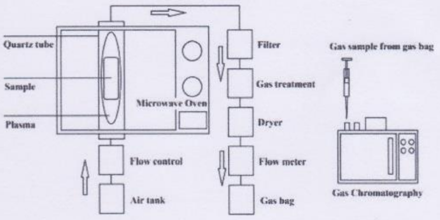

Fig. 1. Schematic of the microwave plasma reactor setup for gasification of waste papers.

TABLE II: OPERATING CONDITIONS OF MICROWAVE PLASMA TESTS

Parameter	Condition
Input power	800 W
Frequency	2.45 GHz
Pressure	101 kPa
Mass of paper	5 g
Air flow rate	1, 2, 3 and 4 lpm
Reaction time	4 min

TABLE III: PRODUCT GAS GENERATED

Air flow rate (lpm)	Gas yields (m^3/kg paper)	Carbon conversion (%)	LHV (MJ/m^3)
1	0.87	19.6	1.86
2	1.71	38.8	1.93
3	2.53	58.6	2.29
4	3.29	51.1	1.56

TABLE IV: PERFORMANCE COMPARISON WITH LITERATURES

Reference	Plasma source	power	Feedstock	Carrier gas	H_2 (%)	CO (%)	CO_2 (%)	CH_4 (%)
This work	Microwave	800 W	Paper, 5 g	Air, 1-4 lpm	7.7	8.5	8.7	1.1
[12]	Microwave	4500 W	Coal, 3-50 kg/h	Air, 0-150 kg/h	2.4	8.1	9.6	1.2
[13]	Microwave	3000 W	Wheat straw, 5-30 g	N_2, 3 lpm	22.1	34.7	33.8	7.9
[14]	Microwave	600 W	Polyethylene, 1 g	0-20% steam/Ar, 3.5 lpm	14	26	12	6
[15]	Microwave	1000 W	Waste wood, 10 g	Ar, 1 lpm	-	56.9	33.8	0.5
[16]	Gliding arc	1140 W	Waste oil, 10 g	10-30% O_2/Ar, 5-16 lpm	-	0.5	2.0	-
[17]	RF, 13.6 MHz	2000 W	Sawdust, 0.3 g/min	N_2, 0.5 lpm	8.5	11	4.0	1.5

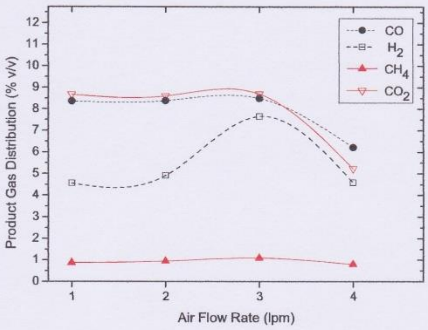

Fig. 2. Variation in concentrations of product gas generated with air flows.

The test run was performed for the microwave plasma system to confirm that the generated plasma can be achieved with our reactor. The plasma was characterized by high intensity light emission. It appeared to occupy most volume of the quartz tube inside the oven cavity.

B. Fuel Gas Production

Table III lists the gas yields, carbon conversion from those in solid material to those present in product gas, and corresponding heating values under different experimental conditions. For all test runs, there was about 1 g of solid residue left in the tube reactor. The specific gas yields obtained were found to increase with air supplied, as expected. Average gas yield was 2.10 m^3/kg paper converted. However, carbon conversion and energy content of the product gas showed initial increase with increasing flow rate, reaching maximum at 3 lpm. Further increase in air supply led to reduction in conversion efficiency and LHV of the product gas. This was contributed to the fact that combustible gas components (CO, H_2 and CH_4) were found to peak at this flow rate, and drop at higher supply rate of carrier gas, as shown in Fig. 2. The observed decline in combustible fractions at higher flow rate may be due to the fact that the flow was too fast inside the reactor, hence, less likely for biomass material and the plasma to react with each other more completely.

C. Comparison with Literature

Product gas obtained from plasmochemical conversion of waste paper in this study was compared against those obtained from other types of biomass and carrier gas in microwave plasma reactors. Results are summarized in Table IV. With air plasma reaction, gas products obtained in this work were comparable to those in the literature.

IV. CONCLUSION AND FUTURE WORK

In this paper, a laboratory scale, microwave plasma reactor has been described. This type of plasma was highly reactive, enabling conversion of solid materials into gas. Under oxidative environment, microwave plasma was able to generate combustible gas from waste paper. This is of practical interest for utilization of solid wastes for the purpose of fuel gas production. However, tests with regards to characterization of microwave plasma generated, as well as parametric investigation of operating conditions are still required. They are planned for the next stage of this research.

ACKNOWLEDGMENT

We wish to thank technical and secretarial assistance from Mr N. Kunnang, P. Thanompongchart, and T. Hanisch.

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MIER2013

Joint Symposium
on Mechanical-Industrial
Engineering and Robotics 2013

14 - 17 November 2013
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Chiang Mai, Thailand.

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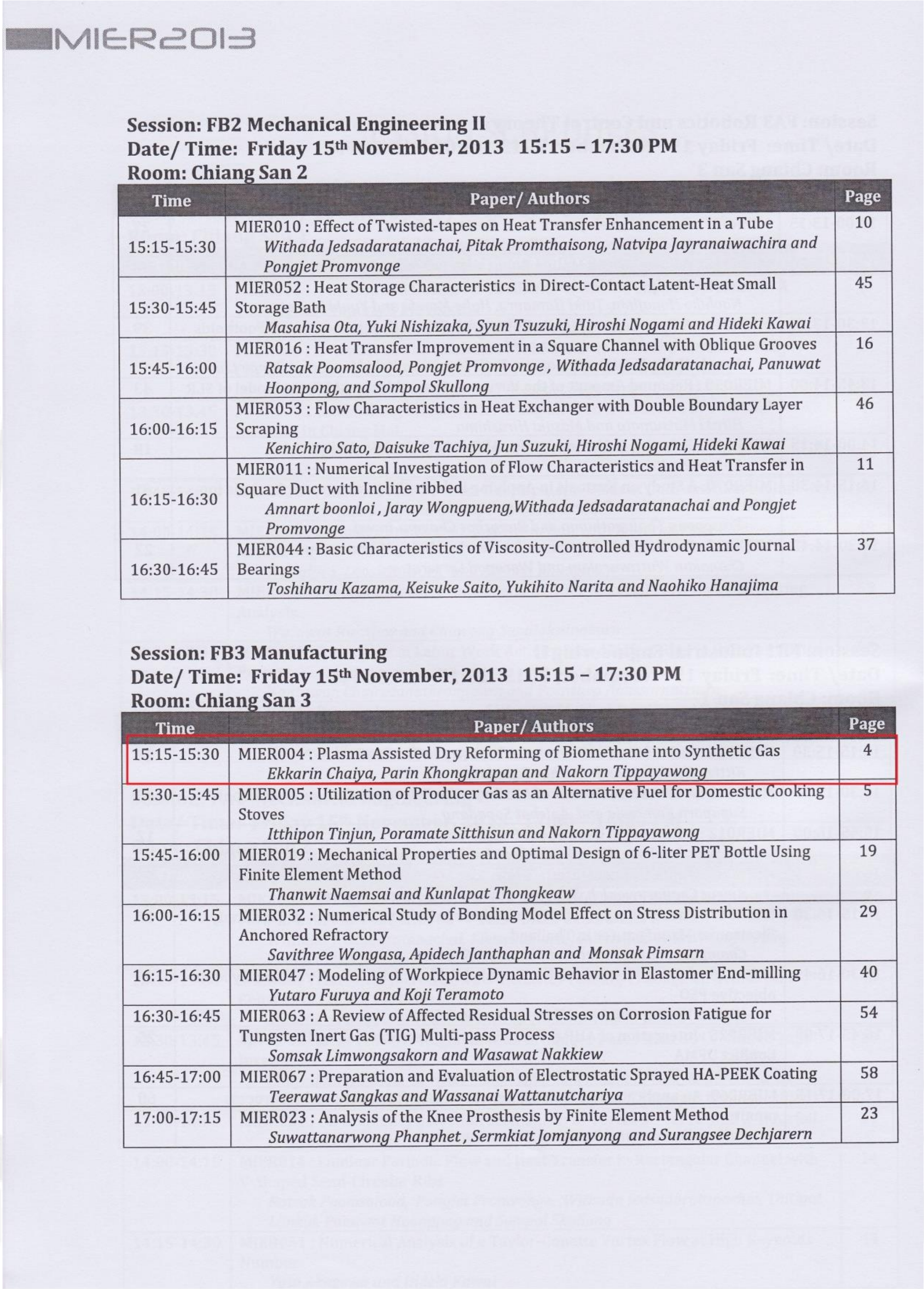

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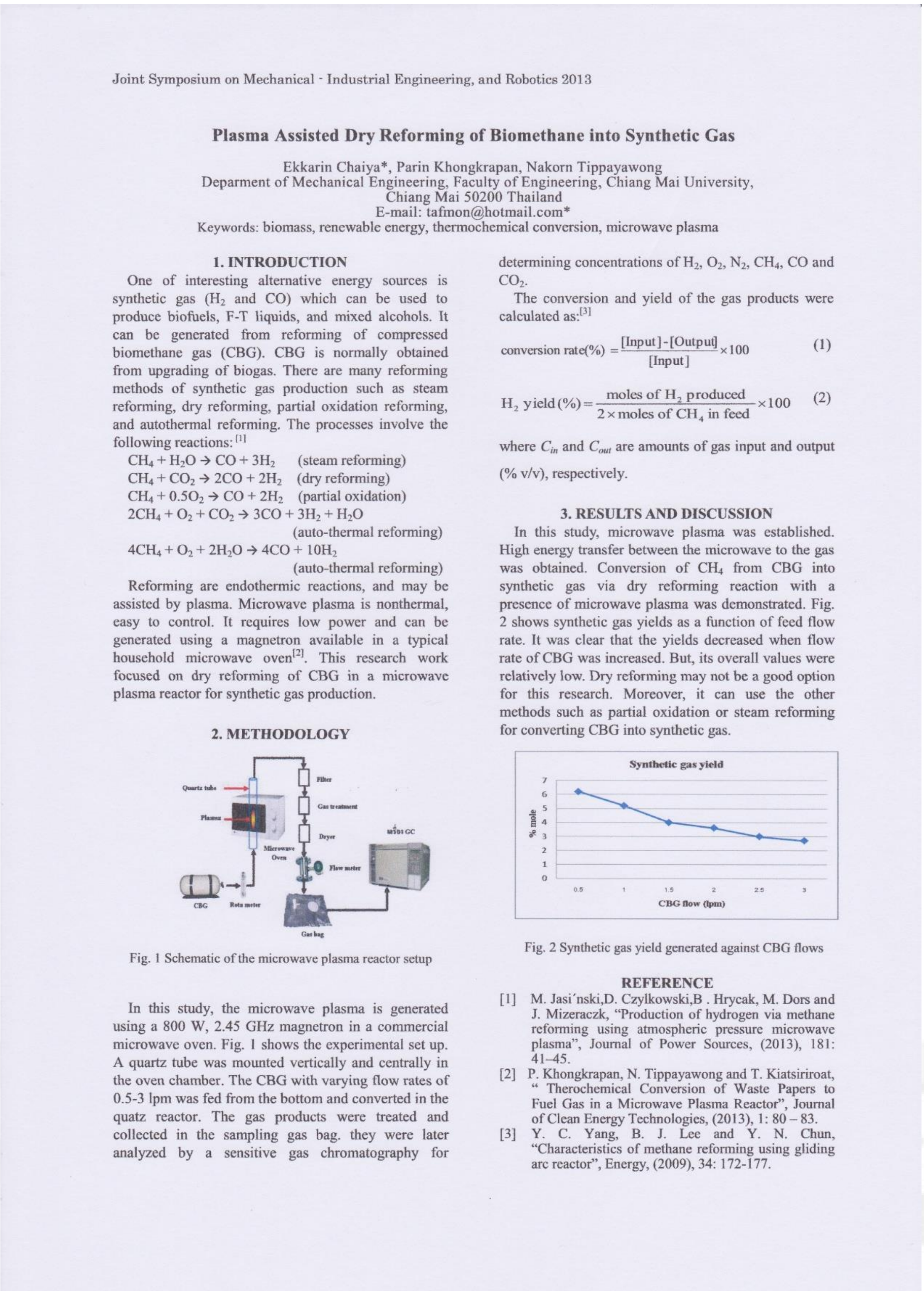
16:45-17:00	MIER067 : Preparation and Evaluation of Electrostatic Sprayed HA-PEEK Coating *Teerawat Sangkas and Wassanai Wattanutchariya*	58
17:00-17:15	MIER023 : Analysis of the Knee Prosthesis by Finite Element Method *Suwattanarwong Phanphet , Sermkiat Jomjanyong and Surangsee Dechjarern*	23



Joint Symposium on Mechanical - Industrial Engineering, and Robotics 2013

Plasma Assisted Dry Reforming of Biomethane into Synthetic Gas

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Keywords: biomass, renewable energy, thermochemical conversion, microwave plasma

1. INTRODUCTION

One of interesting alternative energy sources is synthetic gas (H_2 and CO) which can be used to produce biofuels, F-T liquids, and mixed alcohols. It can be generated from reforming of compressed biomethane gas (CBG). CBG is normally obtained from upgrading of biogas. There are many reforming methods of synthetic gas production such as steam reforming, dry reforming, partial oxidation reforming, and autothermal reforming. The processes involve the following reactions: [1]

$CH_4 + H_2O \rightarrow CO + 3H_2$ (steam reforming)
$CH_4 + CO_2 \rightarrow 2CO + 2H_2$ (dry reforming)
$CH_4 + 0.5O_2 \rightarrow CO + 2H_2$ (partial oxidation)
$2CH_4 + O_2 + CO_2 \rightarrow 3CO + 3H_2 + H_2O$
(auto-thermal reforming)
$4CH_4 + O_2 + 2H_2O \rightarrow 4CO + 10H_2$
(auto-thermal reforming)

Reforming are endothermic reactions, and may be assisted by plasma. Microwave plasma is nonthermal, easy to control. It requires low power and can be generated using a magnetron available in a typical household microwave oven[2]. This research work focused on dry reforming of CBG in a microwave plasma reactor for synthetic gas production.

2. METHODOLOGY

Fig. 1 Schematic of the microwave plasma reactor setup

In this study, the microwave plasma is generated using a 800 W, 2.45 GHz magnetron in a commercial microwave oven. Fig. 1 shows the experimental set up. A quartz tube was mounted vertically and centrally in the oven chamber. The CBG with varying flow rates of 0.5-3 lpm was fed from the bottom and converted in the quatz reactor. The gas products were treated and collected in the sampling gas bag. they were later analyzed by a sensitive gas chromatography for determining concentrations of H_2, O_2, N_2, CH_4, CO and CO_2.

The conversion and yield of the gas products were calculated as:[3]

$$\text{conversion rate}(\%) = \frac{[\text{Input}]-[\text{Output}]}{[\text{Input}]} \times 100 \qquad (1)$$

$$H_2 \text{ yield}(\%) = \frac{\text{moles of } H_2 \text{ produced}}{2 \times \text{moles of } CH_4 \text{ in feed}} \times 100 \qquad (2)$$

where C_{in} and C_{out} are amounts of gas input and output (% v/v), respectively.

3. RESULTS AND DISCUSSION

In this study, microwave plasma was established. High energy transfer between the microwave to the gas was obtained. Conversion of CH_4 from CBG into synthetic gas via dry reforming reaction with a presence of microwave plasma was demonstrated. Fig. 2 shows synthetic gas yields as a function of feed flow rate. It was clear that the yields decreased when flow rate of CBG was increased. But, its overall values were relatively low. Dry reforming may not be a good option for this research. Moreover, it can use the other methods such as partial oxidation or steam reforming for converting CBG into synthetic gas.

Fig. 2 Synthetic gas yield generated against CBG flows

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