



Optimization of Microwave Power, CO₂/CH₄ Ratio and Total Feed Flow Rate for the Plasma Dry Reforming of Methane

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Abstract

In this work, microwave (MW) plasma reactor was used for dry reforming of methane (DRM) reaction to produce syngas (H₂ and CO). The Box-Behnken algorithm based on the response surface methodology (RSM) was utilized to optimize plasma DRM process. The effects of process variables including MW power, CO₂/CH₄ ratio and total feed flow rate on produced syngas selectivities and syngas ratio (H₂/CO) were studied by the Analysis of variance (ANOVA) method using three different models based on quadratic polynomial regression. Both experimental and optimized results confirmed the important paramount role of MW power on syngas selectivity compared to other investigated parameters. The CO selectivity and H₂/CO ratio was considerably influenced by CO₂/CH₄ ratio while the effects of total feed flowrate on plasma DRM performance was insignificant. The interactions between the different variables had a weak effect on the H₂ selectivity and ratio of H₂/CO. At the maximum desirable value of 0.95, the optimum H₂ and CO selectivities were 53.57% and 57.35 % with a H₂/CO ratio of 0.9.

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1. Introduction

The rapid increase in population and high-energy consumption has created environmental issues by the production of greenhouse gases (GHG). Methane and carbon dioxide constitute significant part of GHG and are prime contributors to global warming and climate change [1]. Various technologies have been developed to produce synthesis gas (syngas) from CH₄ and CO₂ [2]. Syngas plays an important role in chemical engineering since it is an intermediate for synthesising various chemicals and fuels, such as methanol, dimethyl and diesel fuel [3]. Plasma technology offers a unique way to induce gas phase reaction. Several plasmas dry reforming methods of methane have been employed to convert methane and carbon dioxide into syngas, which include: non-thermal plasma discharge and thermal plasma [4, 5]. Syngas can be produced by steam reforming of methane (CH₄ + H₂O → CO + 3H₂) [6], dry reforming of methane (CH₄ + CO₂ → 2CO + 2H₂) [7], partial oxidation of methane (2CH₄ + O₂ → 2CO + 4H₂) [8]. Among reforming of methane process, DRM exhibited its superiority in using greenhouse gas to produce syngas instead of releasing those directly to the atmosphere [9]. Plasma DRM process is considered the effective way to achieve a high conversion for CO₂, CH₄ and syngas selectivity [5]. Two main methods used in the dry reforming, which are cold plasma discharge and thermal plasma. Methods for CH₄-

CO₂ reforming by the cold plasma which includes dielectric barrier discharge (DBD), corona discharge (CD), atmospheric pressure glow discharge (APGD), gliding arc discharge (GAD), MW discharge (MWD) and spark discharge. While the thermal method includes direct current (DC), alternating current (AC) arc torch and radio frequency (RF) [10].

In the mean times, design of experimental (DoE) is a powerful tool for process optimization since it allows multiple input factors to be manipulated, determining their individual and combined effects on the process performance in the form of one or more output responses, while significantly reducing the number of experiments compared to the conventional experiments with one factor at a time [11]. Response surface methodology (RSM) is one of the most useful experimental designing methodologies for building the relationship between the multiple input parameters and output responses, which enables us to get a better understanding of the effect of individual factors and their interactions on the responses by three-dimensional and contour interpretations. Plasma DRM including many variables affecting the H₂ and CO selectivity as well as H₂/CO ratio such as feed gas flow rate, CO₂/CH₄ ratio, residence time, and discharge power [12]. These parameters are independent on each other, and therefore their interactions must be considered to optimise the plasma DRM process. Identifying the optimum performance of the plasma process using standard experiments is time-consuming and costly due to the need for multiple experiments under different test conditions [13]. In the previous study, we successfully developed an algorithm [14]. It was found that the chemical model is useful in determining the optimum value for output responses. This model requires a significantly lower number of experiments compared to using traditional methods [12]. Therefore, this work aims to study and optimise the effects of the MW power, CO₂/CH₄ ratio, and total feed flow rate. Moreover, the influences of different process parameters and their interactions on the DRM reaction performance are also attempted.

2. Experimental Procedure

The experimental details were presented elsewhere [14]. Response surface is as a function of independent variables where the response surface can be expressed as follows in Eq. (1):

$$y = f(x_1, x_2, x_3, \dots, x_n) \quad (1)$$

Where y is the answer of the system, and x_i the variables of action called factors. The goal is to optimize the response variable Y and x_i , the variables of action called factors. An important assumption is that the operating variables are continuous and controllable by experiments with negligible errors. It is required to find a suitable approximation for the true functional relationship between independent variables and the response surface [15-17].

In this work, three factors in the three-level Box-Behnken design (BBD) were utilised to investigate the interaction impact among these factors on the performance process of H₂ and CO selectivities and H₂/CO ratio. The input MW power (x_1), CO₂/CH₄ ratio (x_2), and total feed flow rate (x_3) have been identified as the three independent variables affecting the conversions of the selectivities of H₂, CO and ratio of H₂/CO. Therefore, they were selected as the input parameters for the BBD, while the selectivities of H₂ (Y_1), CO (Y_2) and H₂/CO ratio (Y_3) are identified as responses. Either independent process variable contains three different levels, which are coded as low (-1), centre (0) and high (+1), as shown in Table 1.

Table 1: Experimental range and levels of the independent input variables in the Box-Behnken design

Independent Variables	Symbols	Level and Range		
		Low [-1]	Centre [0]	High [+1]
Microwave Power [W]	x_1	600	700	800
CO ₂ /CH ₄ Ratio [-]	x_2	1	2	3
Total Flow Rate [L min ⁻¹]	x_3	1.9	2.1	2.3

A total of 15 experimental runs worked at random in BBD, including three duplicated experimental runs, as shown in Table 2. Response surfaces were generated by JMP statistical discoveryTM software from SAS (version 13.1.0). The analysis of variance (ANOVA) is used to evaluate the adequacy and fitness of the models. The statistical significance of the models and each term in the models can be identified by the *F*-test and adequacy measures such as the coefficient of determination R^2 . The effect of the process parameters was studied by plotting 3-dimensional surface plots and the projected contour plots.

3. Results and Discussion

3.1 DoE Analysis

The real relationships between the input and output values are described in four equations based on the DoE analysis (see Table 2). The H_2 and CO selectivities (Y_1 , Y_2), and the ratio of H_2/CO (Y_3) are presented below (see Eqs. (2-4)).

$$Y_1 = 48.78 + 4.40x_1 - 0.67x_2 - 18.57x_3 + 0.87x_1x_2 - 3.11x_1x_3 + 0.67x_2x_3 - 7.45x_{12} - 6.90x_{22} - 23.03x_{32} \quad (2)$$

$$Y_2 = 57.35 + 2.80x_1 - 1.69x_2 - 22.14x_3 + 0.59x_1x_2 - 0.81x_1x_3 + 3.00x_2x_3 - 7.53x_{12} - 8.70x_{22} - 27.08x_{32} \quad (3)$$

$$Y_3 = 0.84 + 0.05x_1 - 0.0075x_2 - 0.31x_3 - 0.02x_1x_2 - 0.03x_1x_3 - 0.02x_2x_3 - 0.14x_{12} - 0.09x_{22} - 0.40x_{32} \quad (4)$$

ANOVAs were used to determine the significance and adequacy of the quadratic models (Tables 3-5). The coefficient of determination (R^2) of the regression equations for H_2 and CO selectivities and H_2/CO ratio were 0.99, 0.98 and 0.99, respectively. The relationship between the variables and responses described by the second order equation and the agreement between experimental and predicted values were acceptable because R^2 closes to 1, as shown in Figure 1 (a-c).

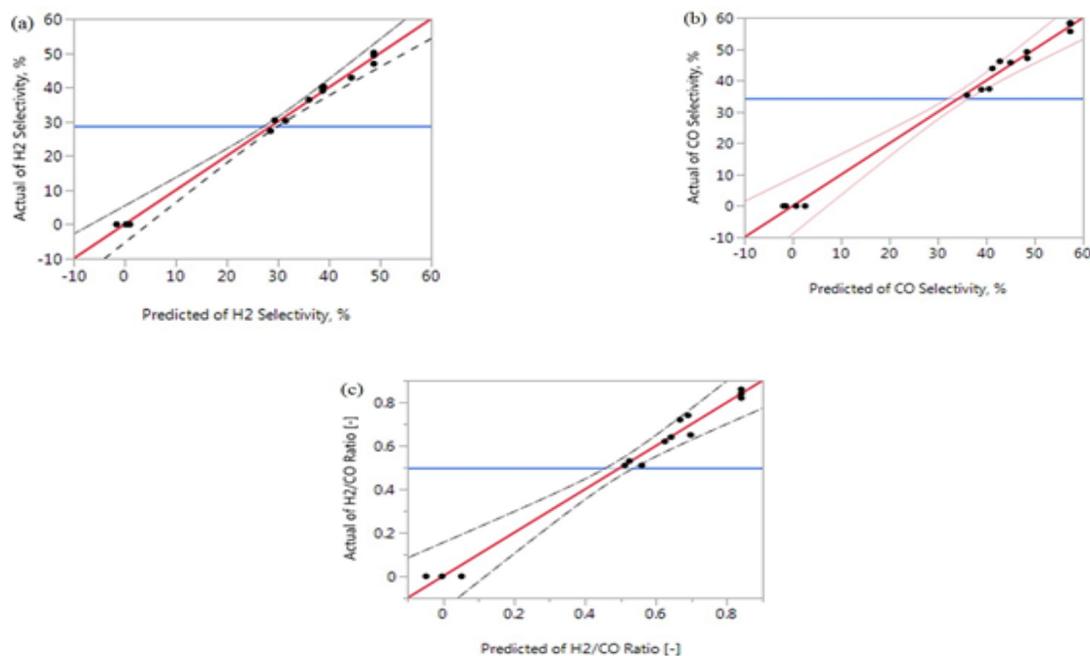


Figure 1: Comparison between actual and predicted values; (a) Selectivity of H_2 ; (b) Selectivity of CO; (c) Ratio of H_2/CO [(•) experimental points, (···) confidence bands > (95%), (—) fit line, Equations (5-7), (—) mean of the Y leverage residuals].

3.2 Effects of Plasma Process Parameters

3.2.1 H₂ and CO Selectivity

The analysis of variance (ANOVA) is used to estimate the indication of adequacy and modelling fitting. The coefficient (β), standard error (ST), the squares sum (SS), the degree of freedom (DF), f -values and p -values are created by ANOVA, as presented in Tables 3 and 4. Furthermore, x_1 , x_2 , and x_3 means first order effects, x_1x_2 , x_1x_3 , and x_2x_3 means second order effects and x_1^2 , x_2^2 , and x_3^2 means interaction effects. The effects of all process variables and their interaction were not observed on the H₂ selectivity, as shown in Table 3. The increase of the MW power and flow rate lead to decreasing the selectivity of H₂. The reason for this could be related to the residence time of the gases in the MW discharge zone, with increase the total flow rates, the resident time of the gas mixture molecules reduced in the discharge zone. The increment in the total flow rates reduces the possibility of collisions between C-H bond in CH₄ molecules and C-O bond in CO₂ molecules resulting in the decrease in the H₂ and CO production [8, 18], as depicted in Figures 3 and 4 (b, d and f). In the meanwhile, the increase the CO₂/CH₄ ratio leads to decrease the H₂ selectivity and yield. This is because the increasing amount of CO₂ in the reacting gas can facilitate the formation of CO from CH₄, as shown in reaction, as shown in reaction (CH₄ + CO₂ → 2CO + 2H₂ [19].

In the CO selectivity and yield, x_1 and x_2 are identified as significant effect, while the terms x_3 is not significant effect, as shown in Table 4. Moreover, the x_1^2 and x_2^2 are a significant term of H₂/CO ratio, whereas the effects of x_3^2 has less significant influenced. The term of x_1x_2 is identified as the significant factor as their p -value is below the critical value of 0.05. According to these results, MW power and CO₂/CH₄ ratio are more significant compared with the other variable. As Tables 3 and 4, respectively indicate, the F -value for the regression model of CO selectivity are 373.97, and 299.18 respectively, which suggests that the model is statistically significant and represent the correlation between the input process parameters and the performance of the plasma process. These results show that the regression model is an adequate for the prediction and optimization of the plasma H₂ selectivity and CO selectivity. A maximum H₂ and CO selectivities of 53.57% and 57.35% respectively are achieved at the highest of input MW power, CO₂/CH₄ ratio, and total flow rate of 726 W, 1.97 and 2.02 L min⁻¹, respectively. The three-dimension response surface and two-dimension contour lines are based on Equations 2 and 3, plots in Figures 2 and 3 (a-f), respectively.

The results show that the interaction between two variables are not significantly effect on the H₂ and CO selectivities, as shown with high p -values (0.25, 0.9907, 0.6080, 0.5490 and 0.9159) for H₂ and (0.41, 0.6192, 0.7360, 0.2963 and 0.6326) for CO, of the terms x_1 , x_3 , x_1x_2 , x_1x_3 and x_2x_3 , respectively, as listed in Tables 3 and 4, respectively. The both effects of the reverse RWGS (Eq. (5)) and reverse Boudouard reactions (Eq. (6)) might contribute to the production of syngas (selectivities of H₂ and CO), as is evident from the present results.

Reverse Water-gas Shift Reaction (WGSR)



Disproportionation Reaction (Boudouard Reaction)



Table 2: Actual Values of the Independent Variables with the Experiment and Predicted Values in the Box-Behnken Design

Run order	Actual Values			Response Values, H ₂ Selectivity [%]		Response Values, CO Selectivity [%]		Response Values, H ₂ /CO Ratio [-]	
	X ₁	X ₂	X ₃	^d Experimental of H ₂ Selec.	Predicted of H ₂ Selec.	^d Experimental of CO Selec.	Predicted of CO Selec.	^d Experimental of H ₂ /CO Ratio	Predicted of H ₂ /CO Ratio
1 ^a	700	2	2.1	50.12	48.78	58.42	57.35	0.86	0.84
2	800	2	1.9	42.83	44.39	47.08	48.48	0.65	0.69
3	700	3	2.3	0.00	0.27	0.00	0.72	0.00	-0.00
4	600	2	2.3	0.00	-1.56	0.00	-1.40	0.00	-0.04
5	600	1	2.1	30.21	31.5	37.3	40.59	0.53	0.52
6	800	2	2.3	0.00	1.01	0.00	2.575	0.00	-0.00
7 ^b	700	2	2.1	46.89	48.78	55.65	57.35	0.82	0.84
8	700	1	2.3	0.00	0.27	0.00	-1.89	0.00	0.05
9 ^c	700	2	2.1	49.34	48.78	57.98	57.35	0.84	0.84
10	600	2	1.9	30.38	29.36	43.84	41.26	0.51	0.51
11	700	1	1.9	39.04	38.76	49.13	48.40	0.64	0.64
12	600	3	2.1	27.27	28.55	35.34	36.02	0.51	0.56
13	800	3	2.1	40.25	38.96	46.11	42.811	0.62	0.62
14	700	3	1.9	36.35	36.07	37.12	39.01	0.72	0.66
15	800	1	2.1	39.99	38.70	45.7	45.01	0.74	0.69

Table 3: Anova Result for the Quadratic Regression Model of H₂ Selectivity

Model Terms	B ^a	SE ^b	SS ^c	DF ^d	F-Value	P-Value
Intercept	48.7833	1.1419	-	-	-	-
X ₁	4.4012	0.6993	154.9680	1	39.6088	0.3116
X ₂	-0.6712	0.6993	3.6046	1	0.9213	0.5876
X ₃	-18.5752	0.6993	27.2450	1	70.4996	0.2906
X ₁ X ₂	0.8765	0.9889	2.5600	1	0.6543	0.4553
X ₁ X ₃	-3.1125	0.9889	38.7506	1	9.9044	0.3255
X ₂ X ₃	0.6725	0.9889	1.8090	1	0.4624	0.1764
X ₁ ²	-7.4491	1.0293	20.8865	1	52.3676	0.8645
X ₂ ²	-6.9041	1.0293	176.0031	1	44.9852	0.7453
X ₃ ²	-23.0316	1.0293	195.6129	1	50.6080	0.4698

R², 0.99; ^aCoefficient; ^bStandard error; ^cSum of Squares; ^dDegrees of freedom; f-values and p-values

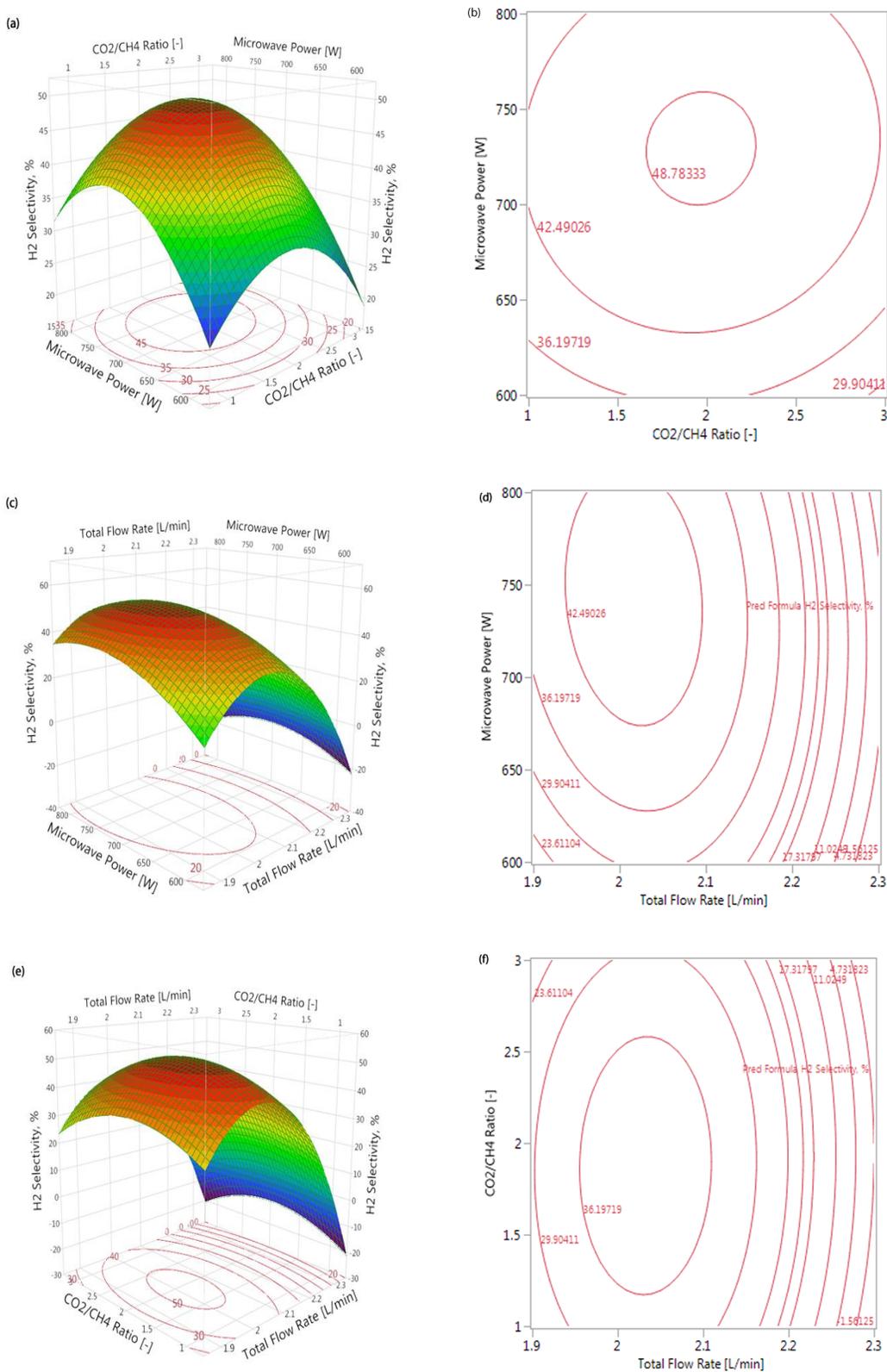
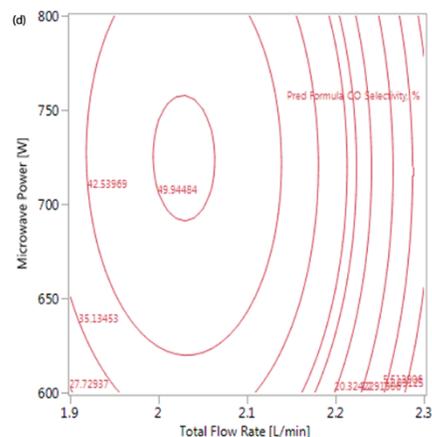
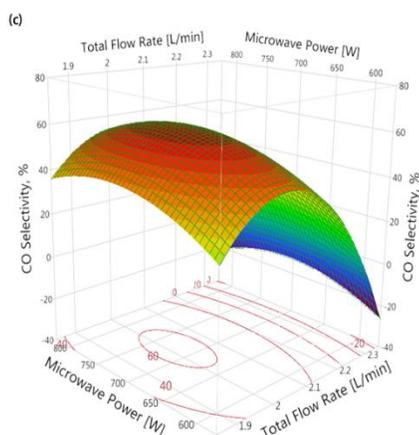
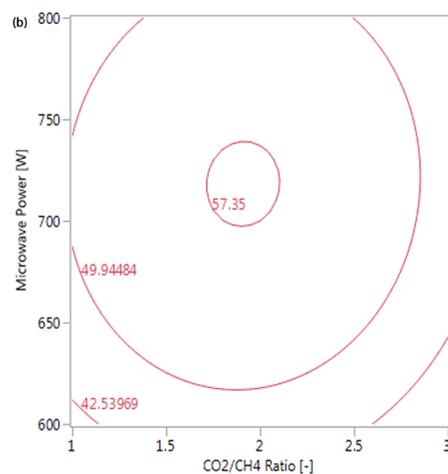
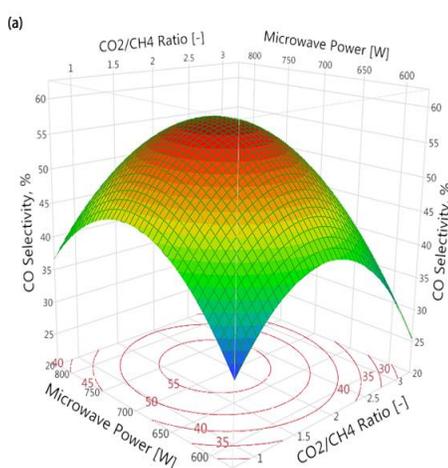


Figure 2: Effect of feed gas flow rates and their interaction on H₂ selectivity at a CO₂:CH₄ ratio of 2:1 and MW plasma of 700 W (a, c, and e) 3-dimensional surface plot; (b, d, and f) projected contour plot

Table 4: Anova Result for the Quadratic Regression Model of CO Selectivity

Model Terms	B ^a	SE ^b	SS ^c	DF ^d	F-Value	P-Value
Intercept	57.35	1.871808	-	-	-	-
X ₁	2.8012	1.1462	4962.7760	1	373.9724	<.0001*
X ₂	-1.6953	1.1462	3322.9842	1	299.1867	<.0001*
X ₃	-22.1462	1.1462	192.6511	1	5.2902	<.0001*
X ₁ X ₂	0.5925	1.6210	2156.4042	1	257.1336	0.0097*
X ₁ X ₃	-0.8142	1.6210	2.6244	1	0.2497	0.6385
X ₂ X ₃	3.0025	1.6210	36.0600	1	3.4307	0.2679
X ₁ ²	-7.5352	1.6872	2209.6353	1	89.9444	0.0032*
X ₂ ²	-8.7025	1.6872	1279.6314	1	76.6037	<.0001*
X ₃ ²	-27.0857	1.6872	270.6667	1	27.6984	0.4365

R², 0.99; ^aCoefficient; ^bStandard error; ^cSum of Squares; ^dDegrees of freedom; f-values and p-values



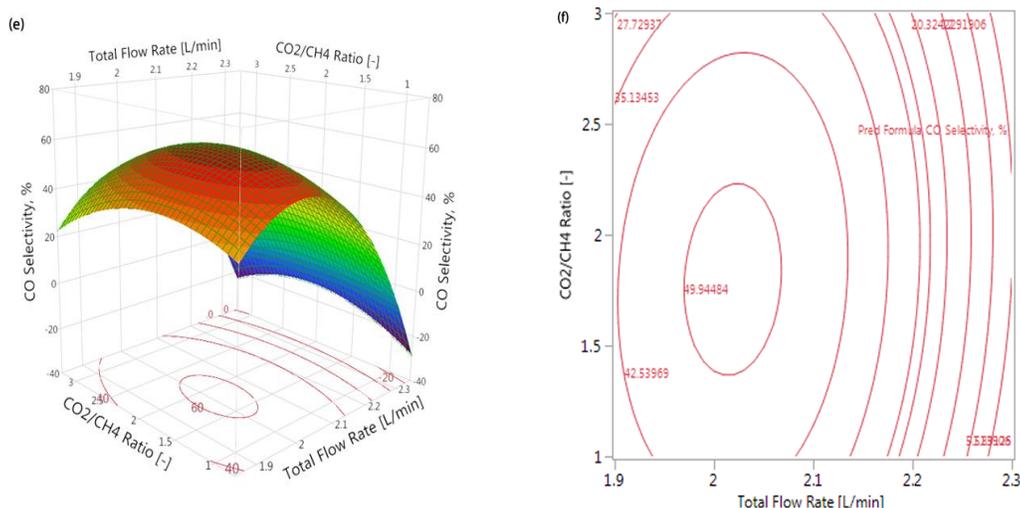


Figure 3: Effect of feed gas flow rates and their interaction on CO selectivity at a CO₂:CH₄ ratio of 2:1 and MW plasma of 700 W (a, c and e) three-dimensional surface plot; (b, d and f) projected contour plot

3.2.1 H₂/CO Ratio

The results of ANOVA analysis for the quadratic regression model of H₂/CO (syngas) ratio are showed in Table 5. The x_3 and is identified as the significant terms ($P < 0.05$) on H₂/CO ratio, while x_1 and x_2 are the less significant term ($P > 0.05$), are given in Table 5. The x_3^2 is a significant term of H₂/CO ratio, whereas the effects of x_1^2 and x_2^2 have less significant influenced, as illustrated in Table 5. It appears from Table 5 that, all the interactions (x_1x_2 , x_1x_3 , and x_2x_3) are weak significant terms on H₂/CO ratio. The F-value is 249.62 for H₂/CO molar ratio and the high F-value gives the most significant parameter that effect the ratio of H₂/CO, as shown in Table 5. The influence of feed gas flow rate parameters and their interactions on H₂/CO ratio is presented in Figure 4 (a-f) by 3D response surface plots and 2D contour lines (based on Equation 4). The contour lines plotted in Figure 4 show that the x_2 strongly interact on H₂/CO ratio due to the elliptical contour produced by the equation model. The optimum H₂/CO ratio of 0.7 was achieved at 0.19, 0.38 1.49 L min⁻¹ of CH₄, CO₂ and N₂, respectively. In the Figure 4 (b, d and f), we find that the H₂/CO ratio decreased slightly with increasing the feed gas flow rates.

As shown in Table 5, the interaction between two parameters have not significant effect on H₂/CO ratio according to p-values (0.4494, 0.6080, 0.4743, 0.9025 and 0.9025) of x_1 , x_3 , x_1x_2 , x_1x_3 and x_2x_3 , respectively. According to the results, the CO₂ feed flow rate (x_2) is considered have the most significant impact on H₂/CO ratio due to it has the highest F-value (Table 5).

At these parameters, the results mentioned above were continuously stabilized for more than four hours with plasma flame remained stable. In this work, the amount of water (according to Eq. (5)) and the amount of solid carbon powder (according to Eqs. (6-8)) was formed out the quartz tube of the MW plasma reactor. The small amount of carbon power could be produced because CO₂ is not completely converted to syngas.

Methane Cracking Reaction



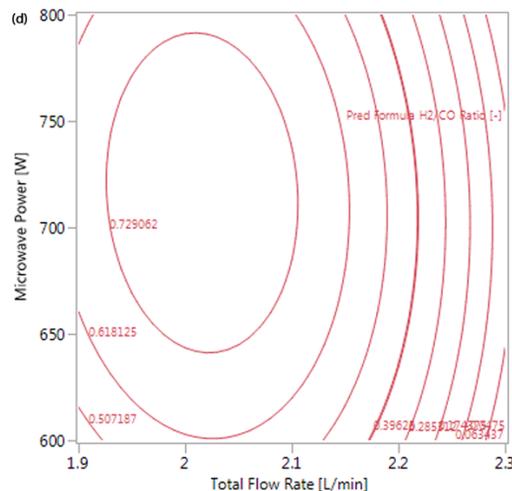
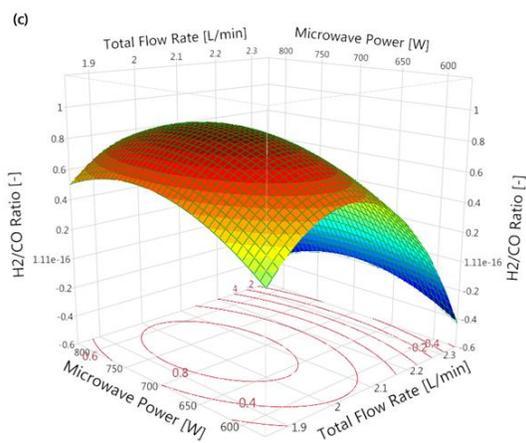
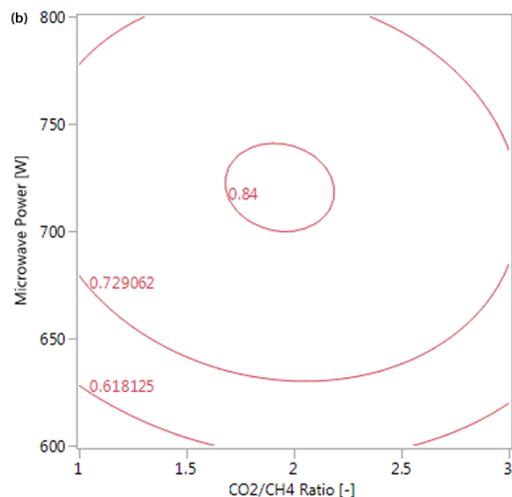
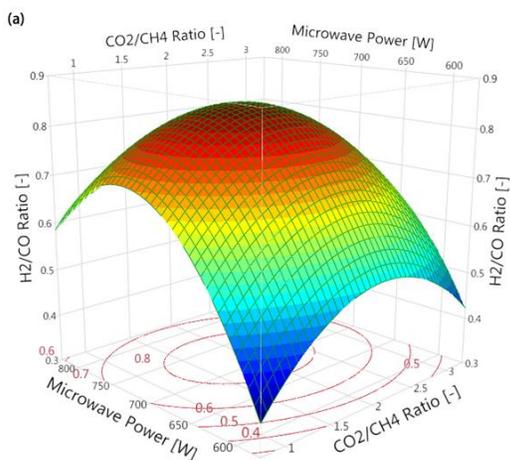
Carbon Gasification



Table 5: Anova Result for the Quadratic Regression Model of H₂/CO Ratio

Model Terms	B ^a	SE ^b	SS ^c	DF ^d	F-Value	P-Value
Intercept	0.8421	0.0325	-	-	-	-
X ₁	0.0575	0.0199	0.0264	1	8.3176	0.1344
X ₂	-0.0075	0.0199	0.0004	1	0.1415	0.6854
X ₃	-0.3152	0.0199	1230.7938	1	249.6226	<.0001*
X ₁ X ₂	-0.0253	0.0281	0.0025	1	0.7862	0.4159
X ₁ X ₃	-0.0352	0.0281	0.0049	1	1.5409	0.2695
X ₂ X ₃	-0.0255	0.0281	0.0016	1	0.5031	0.3873
X ₁ ²	-0.1452	0.0293	0.0776	1	24.4122	0.5367
X ₂ ²	-0.0954	0.0293	0.0333	1	10.4790	0.7543
X ₃ ²	-0.4051	0.0293	1092.6056	1	190.4499	<.0001*

R², 0.99; ^aCoefficient; ^bStandard error; ^cSum of Squares; ^dDegrees of freedom; f-values and p-values



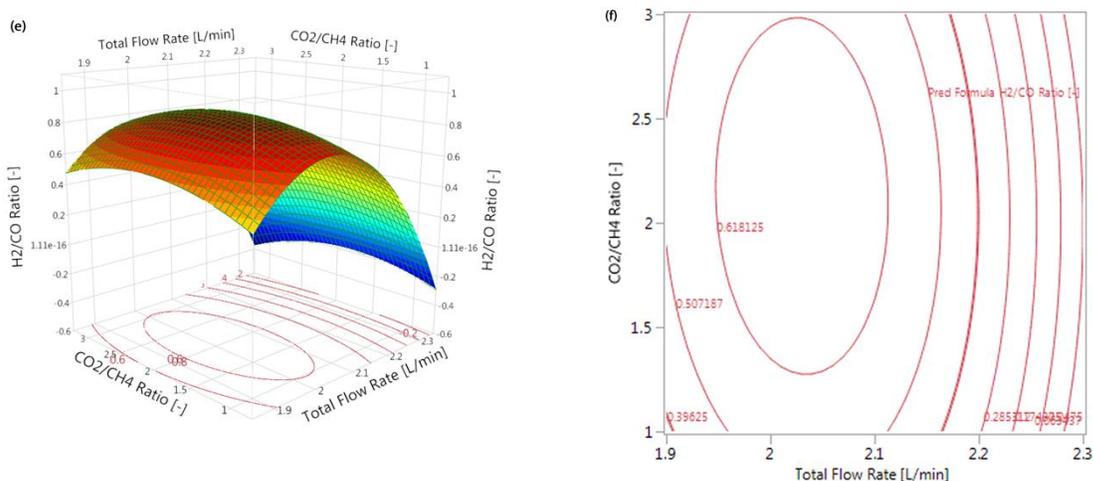


Figure 4: Effect of feed gas flow rates and their interaction on H₂/CO ratio at a CO₂:CH₄ ratio of 2:1 and microwave plasma of 700 W (a, c and e) three-dimensional surface plot; (b, d and f) projected contour plot

3.3 Desirability and Maximum Conditions

The optimum operating conditions were determined for several input variables, which led to obtaining the desirable output response values. Desirability Function (DF) method is used to prove the optimal approaches of multiple responses. Also, the values of DF are dimensionless and are ranged from zero to one (zero means the unacceptable response value while one represents gaining the goal) Ehrigott, 2005 #382}. In this study, the maximum desirability for MW power, CO₂/CH₄ ratio and total feed flow rate is 0.95. This value of the desirability gives strong support to the fitting model. The optimum experimental data was achieved at MW power; CO₂/CH₄ ratio and total flow rate were 726, 1.97 and 2.02 L min⁻¹, respectively. The validity of the equations of the model (Eqs. (2-4)) is good and with a reasonable error, as shown in Table 6.

Table 5: Comparison between the Experimental and Predicted Data at Optimum Conditions

Parameters [L/min]	Response [%]	Experimental Data [%]	Predicted Data [%] (Eqs. (2-4))	Error [%]
MW Power = 726	H ₂ Selectivity	50.12	49.79	0.65
CO ₂ /CH ₄ Ratio = 1.97	CO Selectivity	58.42	59.68	2.11
Total Flow Rate = 2.02	H ₂ /CO Ratio	0.86	0.87	1.15

Table 7 summarises the results of H₂ and CO yields in the previous studies compared with those in this work using different plasma forms at a different flow rates of CH₄, CO₂ and N₂. All previous reports were done at different operating conditions that are higher than those used in this study including the flow rate, CO₂/CH₄ ratio and MW power. In this research, the total feed flow rate of 2.02 L min⁻¹, CO₂/CH₄ ratio of 2/1 and MW power of 726 W were used for producing MW plasma with a good performance and a good stable to plasma flame. The selectivities of H₂, CO and ratio of H₂/CO were 53.57%, 57.35% and 0.9, respectively. Hwang, et al. [20] reported a maximum selectivities of H₂, CO and ratio of H₂/CO were 80.98%, 78.23% and 1.1, respectively in an Arc Jet Plasma (AJP) at a discharge power of 1000 W, a total flow rate of 4 L min⁻¹ and a CO₂/CH₄ ratio of 1/1. They claimed that the highest conversions and yields could be achieved at the high flow rate and high power. Chung and Chang [21] found that the selectivities of H₂ and CO change with increasing flow rate in an Spark Discharge Plasma (SDP). The selectivities of H₂ and CO were 79.27% and 61.78%, respectively obtained at a low total flow rate of 0.2 and the low power of 26.6 W. Zhu, et al. [22] investigated that the high discharge power affects the selectivities of H₂ and CO. They noticed that the maximum H₂ and CO selectivities of 82.19% and 70.83%, respectively can be

obtained at lowest total flow rate and the highest input power 1,344 W in a KHZ Spark discharge plasma. Therefore, the balance between selectivities H₂, CO and molar ratio of H₂/CO is important to the development of an active plasma process. Thus, the performance plasma process generally depends on a wide range of operating conditions and especially on a MW power, CO₂/CH₄ ratio and total feed flow rate. It is necessary and fundamental for optimising the performance plasma process with multiple inputs and multiple responses. This study aims to optimise the process to find the plasma process variables (various parameters) that jointly optimise the yields of H₂, CO and ratio of H₂/CO (various responses).

Table 7: Comparison between Previous Studies with Current Study

Plasma Form	Feed Gas Flow Rate [L min ⁻¹]			CO ₂ /CH ₄ Ratio	Total Flow Rate [L min ⁻¹]	Microwave Power [W]	Yield [%]		H ₂ /CO Ratio [-]	Refs
	CH ₄	CO ₂	N ₂				H ₂	CO		
									Gliding Arc Discharge (GAD)	
Arc Jet Plasma (AJP)	2	2	16	1/1	4	1000	80.98	78.23	1.1	[20]*
Spark Discharge Plasma (SDP)	0.075	0.075	0.05	1/1	0.2	26.6	79.27	61.78	N/A	[21]
AC Spark Discharge Plasma (SDP)	N/A	N/A	N/A	1.5/1	0.15	45	62.22	87.52	2	[24]*
KHZ Spark Discharge	N/A	N/A	N/A	2/3	0.15	1,344	82.19	70.83	N/A	[22]*
Microwave Discharge Plasma	0.18	0.36	1.48	2/1	2.02	726	53.57	57.35	0.9	This study

NA, Not available

4. Conclusions

The influences of the MW power, CO₂/CH₄ ratio and total feed flow rate and their interactions on H₂, CO selectivities and syngas ratio for plasma DRM were successfully studied and optimised in this work. Behnken-Box design and RSM based on multi-objective optimization. Regression models were developed to describe the relationships between the plasma process variables and reaction performance. ANOVA was also applied to estimate the significance and adequacy of the models for each response (H₂, CO selectivities and H₂/CO ratio). The results showed that the selectivity of CO increase with increasing MW power and CO₂/CH₄ ratio, while the H₂/CO molar ratio increase with increasing of total feed flow rate. The MW power was found to be the most important variables driving the CO selectivity and yield while the CO₂/CH₄ ratio has the significant effect on CO selectivity. The interactions of all plasma process variables had negligible effects on the selectivity of H₂ and the syngas ratio.

There was a significant interaction of the input MW power with the CO₂/CH₄ ratio on the CO selectivity. The optimum coefficients of determination (R²) of the regression equations for H₂ and CO selectivities and H₂/CO ratio were 0.99, 0.98 and 0.99, respectively. The optimal selectivities of H₂, CO and ratio of H₂/CO were 53.57%, 57.35% and 0.9, respectively for the plasma process achieved at a MW power, CO₂/CH₄ ratio and total flow rate of 726 W, 1.97 and 2.02 L min⁻¹, respectively. The experimental results under the theoretical optimal conditions well simulated the effects of process variables and their interaction on the process parameters and performances.

Conflict of Interest: The authors declare that they have no conflict of interest.

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