Jurnal
Teknologi

Full Paper

OPTIMIZATION OF NON THERMAL PLASMA REACTOR PERFORMANCE FOR THE DECOMPOSITION OF XYLENE

Article history
Received
28 February 2016
Received in revised form
3 April 2016
Accepted
15 July 2016

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Abstract

Non Thermal Plasma (NTP) is an emerging method used for the decomposition of volatile organic compounds (VOCs). This research focuses on the optimization of NTP reactor performance for decomposition of xylene from wastewater using response surface methodology (RSM) by operating the NTP reactor at applied voltage of 12-15 kV, discharge gap of 2.0-3.0 cm and gas flow rate of 2.0-5.0 L/min. An optimum xylene removal efficiency of 81.98% was obtained at applied voltage 15kV, discharge gap 2.09cm and gas flow rate at 2.36 L/min. The experimental removal efficiencies and model predictions were in close agreement with an error of 0.63%.

Keywords: Xylene, non-thermal plasma, optimization, response surface methodology, wastewater, SS (Sums of Square), MS (Mean square), DF (Degree of Freedom)

Abstrak

Plasma Bukan Terma (NTP) adalah kaedah baru yang digunakan untuk menyingkirkan sebatian organik mudah meruap (VOCs). Kajian ini menumpukan kepada keadaan optimum tindak balas Reaktor Plasma Bukan Terma untuk menyingkirkan xylene daripada air sisa dengan menggunakan kaedah gerak balas permukaan (RSM) dengan mengendalikan Reaktor Plasma Bukan Terma pada voltan diantara 12-15 kV, jurang pelepasan 2.0-3.0 cm dan kadar aliran gas pada 2.0-5.0 L/min. Keadaan optimum bagi penyingkiran xylene yang effisien pada tahap 81.98% diperolehi pada voltan 15kV, jurang pelepasan 2.09cm dan kadar aliran gas 2.36 L/min. Eksperimen keberkesanan penyingkiran dan model ramalan adalah hampir tepat dengan ralat sebanyak 0.63%.

Kata kunci: Xylene, Plasma Bukan Terma, kaedah gerak balas permukaan, air sisa

1.0 INTRODUCTION

The treatment of wastewater contaminated with volatile organic compounds (VOCs) poses a major challenge to the chemical industries [1]. Using Non thermal plasma (NTP) for gas-phase pollution control shows much promise and offer several advantages compared to the other traditional method such as adsorption, absorption and incineration. These include moderate capital cost, compact system, ease of operation and high removal efficiency [2]. NTP is an excellent source of gas phase free radicals that are useful for destroying pollutants. Several studies have previously investigated the decomposition of VOCs using non thermal plasma of different reactor configurations operated under different conditions [3]. NTP has the ability to simultaneously destroy organic compound such as benzene, toluene, xylene as well as inorganic pollutants such as NO, SO_2 [4]. NTP has been reported as an efficient technique for the removal of pollutants in air due to its low energy requirement and the potentiality to produce highly reactive chemical species under ambient conditions such as hydroxyl (OH^-), atomic hydrogen (H), atomic oxygen (O) and hydroperoxyl (HO_2) radicals that promote the destruction of the target pollutants [5, 6]. NTP has also shown potential to treat large volume emissions from low to high concentration of gaseous pollutants [7]. NTP can be produced through electrical *discharge* by applying an electric potential between two electrodes placed in a glass tube filled with various gases or by use of laser, electron beam, microwave and Radio frequency (RF) generator [8, 3]. It has been reported that the efficiency of gaseous pollutants removed by using packed-bed reactors were slightly higher compared to non-packed reactor at specific energy density and energy efficiency for gaseous pollutant abatement [9]. This is due to the presence of packing material such as barium titanate ($BaTiO_{3)}$ that acts as catalysts that induces extra performance enhancement mechanisms and effectively generate active species useful for VOC destruction and better energy efficiency [10]. However, there is need to optimize NTP reactor performance by controlling the various operating conditions which include applied voltage, discharge gap and gas flow rate. A better alternative method of optimization is the use of response surface methodology (RSM) because it includes the influences of individual factors and their interactions as well as achieving the optimum condition for desirable responses with limited number of experimental runs [11]. However, report on the study of the optimization of NTP reactor performance in the removal of xylene from industrial wastewater is still not available. Therefore, this study is focused on investigating the influence of applied voltage, gas flow rate and discharge gap on the removal efficiency of xylene using NTP reactor and optimizing these parameters using RSM to obtain the optimum degree of removal.

2.0 METHODOLOGY

2.1 Materials

Xylene was obtained from Merck Sdn. Bhd Malaysia with greater than 99.5% purity. Synthetic wastewater containing 1,500 ppm of xylene was prepared for the experiment.

The experiment was carried out by using the ferroelectric packed bed NTP reactor consists of a Pyrex glass tube of 1-inch internal diameter and standard length and 3-mm diameter barium Titanate (BaTiO3) pellets. The packed-bed was constructed by placing the dielectric pellets at the discharge region of NTP reactor. The geometry of packed-bed reactors in this experiment consist of two coaxial electrodes packed with the ferroelectric pellet layer which is the spherical $BaTiO_3$ beads as the packing pellets that fill the plasma formation region.

2.2 Experimental Setup

The experimental set up is shown in Figure 1. To remove the xylene from the wastewater using the air stripper, the air flow rate (7.08 L/min) was set using a rota meter and the wastewater inlet was also set to 0.12 L/min by adjusting the rota meter while the wastewater and air heaters were set to 50 ^{0}C. The air and contaminated water were then pumped into the air stripper in counter-current operation. The treated wastewater was collected at the bottom while VOC rich air which comes out at the top of the column was sent to the non-thermal plasma reactor. The decomposition of xylene in ferroelectric packed bed NTP was conducted at applied voltages of 12.32, 14 and 15.68 kV and fixed discharge gap and flow rate of 25 mm and 2.36 L/min respectively. A high voltage probe was used to measure the applied voltage and the current was measured using digital Pico scope.

The concentrations of xylene in the outlet gas from air stripper were first determined with a frontier FT-IR spectrometer (Perkin Elmer) coupled with cyclone gas cell accessory. The nitrogen gas was used as background gas for the measurement [19]. The gas streams were passed into multiple optical gas cell (Specac Ltd) and the sample spectrum for each operating condition was captured at room temperature and atmospheric pressure at a spectra resolution of 1cm^{-1}. This was then followed by the determination of removal efficiency of xylene and by-products from non-thermal plasma treatment by passing gas streams from non-thermal plasma through the FTIR as explained earlier. Concentrations of xylene in each spectrum were determined by integrating the area under the peaks using Perkin Elmer spectrum standard v10.4.0 software and then comparing with standard spectra produced by Pacific Northwest National Laboratories. Three spectra were recorded in order to obtain the average concentration for the xylene gas that come out from the air stripper and entering the non-thermal plasma as inlet gas, the concentration was calculated by using the formula [18]:

$$N(ppm) = \frac{INT exp \text{ X } Nstd \text{ X } Lstd}{INT \text{ std X } Lmeas} \tag{1}$$

In this formula, Nstd, Lstd (given as 1 m), and Lmeas represent the concentration of the standard gas produced by Infrared Analysis, Inc. and Pacific Northwest National Laboratories, optical path length of the standard gas measurement, and optical path length of the measured spectrum, respectively. The Nstd xylene were given as 100 ppm [18,19].

The removal efficiency of xylene was calculated as follows.

$$\eta(\%) = \frac{[xylene]_{inlet\ (ppm)} - [xylene]_{outlet(ppm)}}{[xylene]_{inlet(ppm)}} \text{ x } 100\% \qquad (2)$$

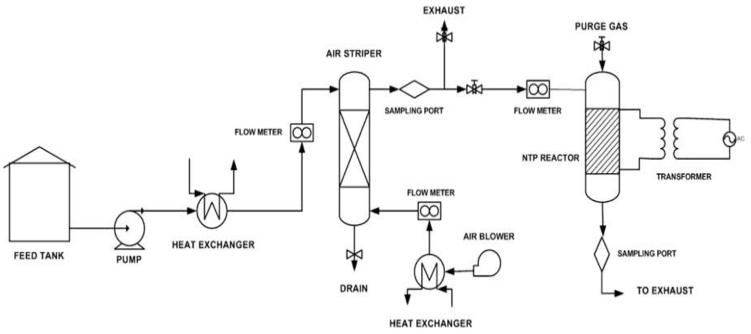

Figure 1 Schematic Diagram for the setup of the Ferroelectric Packed Bed NTP reactor

2.3 Design of Experiment

In order to optimize NTP reactor performance, the experimental design of xylene decomposition in a packed bed NTP reactor was done using Design expert 7.0 (Stat-Ease, Inc., Minneapolis, MN, USA) software [12]. In this optimization session, the Central Composite Design (CCD) under RSM was applied. Twenty numbers of experimental runs were determined by RSM with three variables factors which include applied voltage (X_1), discharge gap (X_2) and gas flow rate (X_3) as stated in Table 1 were considered for the experiments in order to allow modeling of quadratic effects as well as main effects, and their interactions on the response variable.

Table 1 Selected experimental domain for each of the variables

Independent variables	Symbols	Levels	
		-1	1
Applied Voltage (KV)	X_1	13	15
Discharge gap (mm)	X_2	20	30
Gas flow rate (L/min)	X_3	2.36	4.72

The propose model for the response (removal efficiency of xylene) is stated as Equation 3.

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{33} X_3^2 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3 \qquad (3)$$

Where Y is the predicted response, X_1, X_2 and X_3 represent the three variables respectively, and β_0, β_1, β_2, β_3, β_{11}, β_{22}, β_{33}, β_{12}, β_{13} and β_{23} are the model coefficients calculated from the experimental data. Design Expert 7.0 software was used to analyze the responses and to obtain the values of the coefficients of Equation 3. The regression model was checked by coefficient of determination R^2, F value and its associated probability P value to determine the overall model significance.

3.0 RESULTS AND DISCUSSION

The results of the experimental design are shown in Table 2.

3.1 Model Fitting

The results of experimental design as stated in Table 2 were used to run ANOVA and Multiple Regression Analysis using Design Expert 7 software. This allows the optimum degree of removal efficiency of xylene and others variable to be predicted. The coefficients of the model equation which are used to predict the optimum degree for percentage removal of xylene were determined by multiple regression analysis and are shown as stated in Equation 4.

$$Y = +66.21 + 7.01\ X_1 - 5.39\ X_2 - 4.59\ X_3 + 0.076\ X_1X_2 - 1.39\ X_1X_3 + 0.18\ X_2X_3 + 2.77\ X_1^2 - 4.63\ X_2^2 - 3.15\ X_3^2 + 2.16\ X_1X_2X_3 \qquad (4)$$

Table 2 Experimental design and predicted responses

Experimental Design			Predicted Reponses
Applied voltage (KV)	Discharge Gap (cm)	Gas flow rate (L/min)	Percentage Removal (%)
X_1	X_2	X_3	Y
13.00	2.00	2.36	64.21
15.00	2.00	2.36	82.61
13.00	3.00	2.36	55.58
15.00	3.00	2.36	65.64
13.00	2.00	4.72	62.17
15.00	2.00	4.72	66.37
13.00	3.00	4.72	45.62
15.00	3.00	4.72	58.77
12.32	2.50	3.54	57.20
15.68	2.50	3.54	86.89
14.00	1.66	3.54	58.21
14.00	3.34	3.54	44.00
14.00	2.50	1.56	63.67
14.00	2.50	5.52	47.10
14.00	2.50	3.54	66.32
14.00	2.50	3.54	66.32
14.00	2.50	3.54	66.32
14.00	2.50	3.54	66.32
14.00	2.50	3.54	66.32
14.00	2.50	3.54	66.32

Table 3 Analysis of Variance (ANOVA)

Sources	SS	Df	MS	F-value	P value prob >F
Xylene removal					
Regression	2000.41	10	200.04	20.88	<0.0001
Residual	86.22	9	9.58		
Lack of Fit	86.22	4	21.56		
Pure Error	0.00	5	0.00		

The significance of each variable are shown in Table 3 with the confidence interval of the model set at 95%. The P value is determined from the F-ratio used as a tool to check the significance of each coefficient that in turn indicated the pattern of interactions between the variables. Table 3 showed that the F test statistic value is 20.88 with 10 and 9 degrees of freedom F (10, 9) = 20.88. The F ratio is < 0.0001 which implied that the model was significant with the probability > F less than 0.05. The value for sum of square (SS) and mean square (MS) were used to decide the good fit between statistical model and the data itself. The SS value at 2000.41 are larger than residual sum of square value at 86.22 showed that, the large amount of variability in response can be explained by the model. The smaller residual sum of squares value, indicated the model was fits with the data [16, 17].

The graph of predicted versus observed variable in Figure 2 gave a straight line with coefficient of determination at 0.9128. The model showed a good relationship between observed and predicted results with coefficient (R^2) for the response close to 1, indicated that 95% of the variability in the response could be explained by the model and only less than 5% of the response could not be explained by the model. The model is reasonably reproducible to predict [11, 12] the percentage removal of xylene in this experiment.

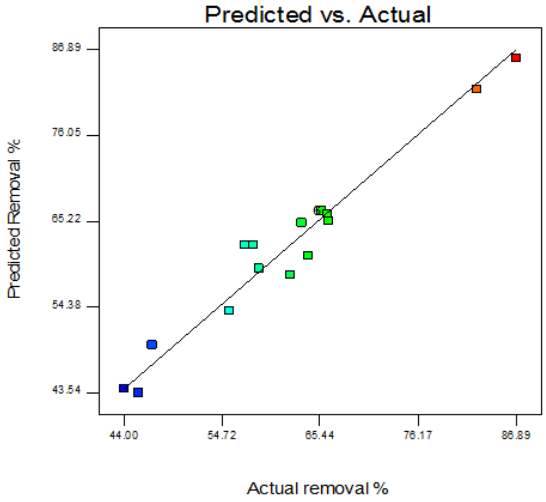

Figure 2 Graph of predicted values versus actual values for percentage removal of xylene

3.2 Pareto Chart of Percentage Removal of Xylene

The effects of the independent variables on the dependent variable were elaborated by using response surface plots and Pareto charts. Figure 3 shows the significance level for each variable on percentage removal of xylene. It shows that the applied voltage has the most significant effect on removal of xylene by NTP reactor with P value of 0.0042 followed by discharge gap with P value 0.0183 and flow rate with P value 0.0383. P values less than 0.05 indicate that all individual variables are statistically significant. These imply that the percentage removal of xylene by NTP reactor is most dependent on the applied voltage and moderately on discharge gap and flow rate. Figure 3 also showed that the combined effect for each individual variable such as applied voltage, discharge gap and air flow rate (ABC); applied voltage and air flow rate (AC); discharge gap and air flow rate (BC); applied voltage and discharge gap (AB) have the P value above 0.05 which are 0.4142, 0.5965, 0.9445 and 0.9766 that respectively indicated the least significant effects on the removal efficiency of xylene by NTP reactor.

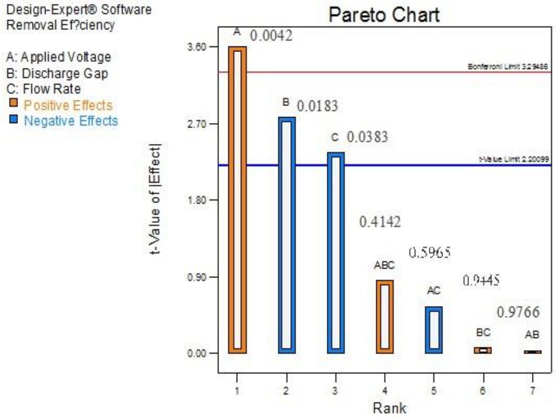

Figure 3 Graph of predicted values versus actual values for percentage removal of xylene

3.3 Effect of Applied Voltage on xylene Removal Efficiency

The effect of applied voltage on xylene removal efficiency was studied at voltage values between 12-15 kV. The result shows that increase in the applied voltage enhances the removal efficiency of xylene as shown in Table 2. Increased applied voltage enhances the reaction activity between high energy electron and xylene molecules. The increased intensity of the plasma discharge observed as the applied voltage is increased. The effective collisions that occur between high energy electrons and xylene molecules decomposes the molecular structure of xylene and convert it into smaller molecules like carbon dioxide (CO_2), carbon monoxide and water (H_2O) [2]. It is reported that the electron density increases with discharge current and voltage [20, 21].

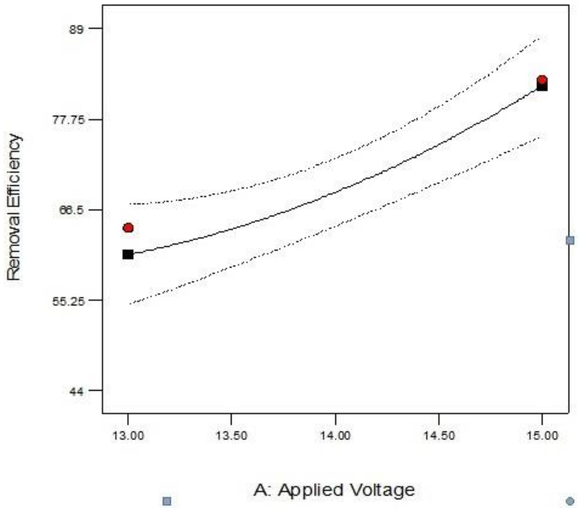

Figure 4 The effect of applied voltage on removal efficiency of xylene

3.4 Effect of Discharge Gap on xylene Removal Efficiency

Figure 5 shows that the removal efficiency of xylene tends to decrease as the discharge gap increases. This result can be attributed to the requirement of higher energy to decompose xylene molecules at large discharge gap [4]. The energy efficiency is also higher when small discharge gap is used [13].

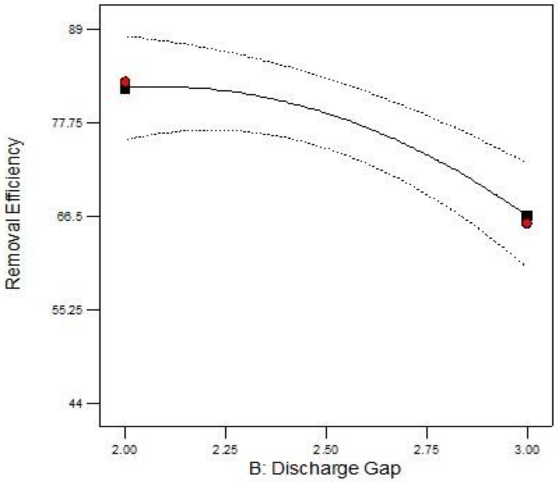

Figure 5 The effect of discharge gap on removal efficiency of xylene

3.5 Effect of gas flow rate on xylene Removal Efficiency

Figure 6 shows that the removal efficiency was higher at lower flow rate compared to the high flow rate. This result is similar to that of the study on the abatement of Gas-phase p-xylene via Dielectric Barrier Discharges by Lee and Chang [14]. This due to the higher residence time the gas spends the reactor at low flow rate which allows for more probability of collision between xylene and high energetic electrons to occur [4].

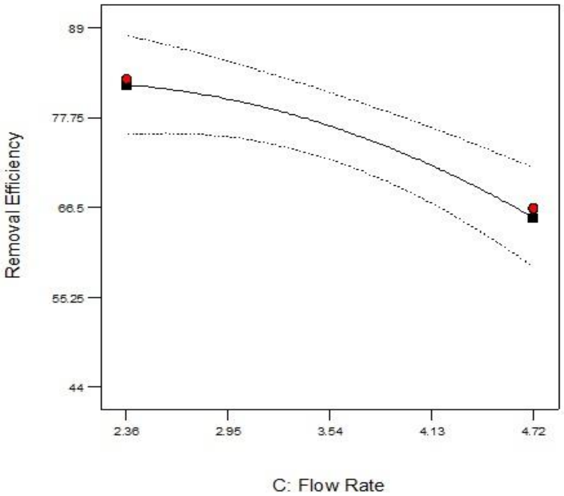

Figure 6 The effect of gas flow rate on removal efficiency of xylene

3.6 The Combine Effects of the Variables on the Response

The combined effect for applied voltage and discharge gap (AB); applied voltage and air flow rate (AC); and discharge gap and air flow rate (BC) are shown as stated in Figures 7, 8 and 9 respectively. Figure 7 shows that as the applied voltage increase, the discharge gap decrease, and the removal efficiency of xylene increases. This result is in line with the previous research which states that deposited power in the plasma increases by increasing the input voltage to the NTP reactor [3], while decrease in the discharge gap changes the electron distribution in the discharge region because energy efficiency increases as discharge gap decreases [13].

Figure 8 shows that as the applied voltage and flow rate decrease, the removal efficiency of xylene increases. This result implies that the combine effect of applied voltage and flow rate can give significant impact on the performance of NTP reactor to decompose xylene. This is because the amount of high energy electron produced in NTP reactor increases with increasing applied voltage. The decrease in flow rate helps to enhance the contact time of the electron with the xylene molecule thus enhancing decomposition [15].

Figure 9 shows the combined effects of discharge gap and air flow rate. It reveals that as the discharge gap decrease and the flow rate decrease, the removal efficiency of xylene increases significantly. This is because at small discharge gap and low flow rate, the energy required to decompose xylene is much lower compare with the large discharge gap [4].

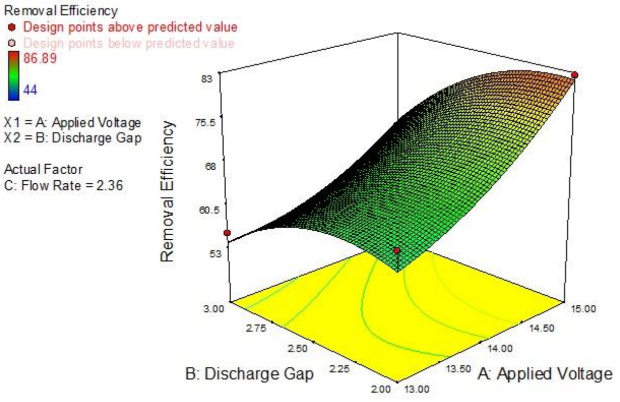

Figure 7 3D Response Surface Plots of Percentage Removal of xylene versus Discharge Gap and Applied Voltage

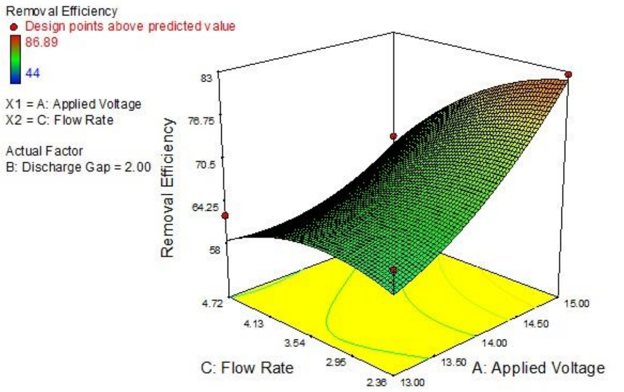

Figure 8 3D Response Surface Plots of Percentage Removal of xylene versus Applied Voltage and Flow Rate

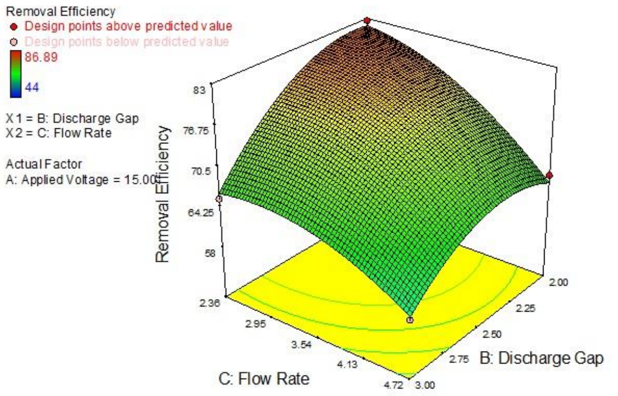

Figure 9 3D Response Surface Plots of Percentage Removal of xylene versus Discharge Gap and Flow Rate

3.7 Optimization

Numerical optimization was used to determine the optimum operating conditions for the NTP reactor performance in the decomposition of xylene. Optimum applied voltage, discharge gap and air flow rate were found to be 15.0 kV, 2.09cm and 2.36 L/min respectively. This translates to 81.98% of percentage removal of xylene by NTP reactor as shown in Table 4. The percentage removal of xylene for the observed value was 82.61% under the same condition of variables. This result showed 0.63% error between observed and predicted value which is within the 5% of allowable limit

Table 4 Predict and observed values on percentage removal of xylene at optimum process condition

Response	**Predicted value**	**Observed value**	**Error (%)**
Percentage Removal (%) of xylene	81.98	82.61	0.63

4.0 CONCLUSION

The optimization of NTP reactor performance for decomposition of xylene from wastewater using response surface methodology (RSM) by operating the NTP reactor at applied voltage of 12-15 kV, discharge gap of 2.0-3.0 cm and gas flow rate of 2.0-5.0 L/min. Xylene was removed from wastewater using air stripper and a model was obtained where Y is the percentage of removal (%), X_1 the Applied voltage (KV), X_2 Discharge Gap (cm) and X_3 Gas flow rate (L/min).

$$Y = +66.21 + 7.01\, X_1 - 5.39\, X_2 - 4.59\, X_3 + 0.076\, X_1X_2 - 1.39\, X_1X_3 + 0.18\, X_2X_3 + 2.77\, X_1^2 - 4.63\, X_2^2 - 3.15\, X_3^2 + 2.16\, X_1X_2X_3 \quad (4)$$

The coefficient of determination (R^2) of 0.9578 shows that the model used in predicting the removal efficiency of xylene by NTP reactor has a good fit. The result showed the applied voltage, discharge gap and flow rate has the significant effect on the removal efficiency of xylene by NTP reactor with P values showed less than 0.05 indicated that the variable is significant. Based on the ANOVA table, the most significant effect would be applied voltage with P value at 0.0042, followed by discharge gap with P value 0.0183 and applied voltage at P value of 0.0383. The result shows the optimum xylene removal to be 81.98% at optimum variables of applied voltage 15kV, discharge gap 2.09cm and flow rate 2.36 L/min. The experimental removal efficiencies and model predictions were in close agreement with an error of 0.63%.

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