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Original Article

# Process modeling of NH<sub>3</sub> contaminated waste air treatment in photocatalytic reactor using TiO<sub>2</sub> coated glass tubes

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#### Abstract

Ammonia (NH<sub>3</sub>) is a noxious gas that can cause serious damage to human health and to the environment. Thus, elimination of NH<sub>3</sub> in waste air by treating before releasing to the atmosphere is necessary. In this study, titanium dioxide (TiO<sub>2</sub>) nanoparticle Degussa P25 coating on glass cylindrical tube supporters was synthesized and used as photocatalytic in a continuous photoreactor device. UV light source was vertically installed around the photoreactor in a stainless steel chamber. Experiments were designed and conducted based on central composite design (CCD) and analyzed using response surface methodology (RSM). This RSM was used to evaluate the effects of process variables and their interactions towards attainment of their optimum conditions. Three significant variables, light intensity (23-114W/m<sup>2</sup>), waste air flow rate (1-5 l/min), and TiO<sub>2</sub> loading (1.18-5.90 g) were fitted in a quadratic model to the response of NH<sub>3</sub> treatment efficiency. R<sup>2</sup> of 0.9783 was satisfactorily evaluated for correlation coefficient of the quadratic model on the photocatalytic reactor. Based on statistical analysis, the NH<sub>3</sub> treatment model has been proven to be highly significant with very low probability values (<0.0001). The optimum conditions obtained were 114 W/m<sup>2</sup> in light intensity, 1 l/min waste air flow rate, and 5.90 g TiO<sub>2</sub> loading. This resulted in 91.45% treatment of NH<sub>3</sub> at 300 ppmv as obtained from the predicted model, which fitted well with the laboratory results (90.02%). According to the study, TiO<sub>2</sub> coated on glass tube supporter can effectively be used for the treatment of NH<sub>3</sub> from waste air and could possibly be applied at an industrial scale.

Keywords: RSM, ammonia, photocatalytic, TiO<sub>2</sub>, UV light

### 1. Introduction

Ammonia (NH<sub>3</sub>) is a caustic volatile compound and it is toxic. It is produced in nature by a variety of organisms, human, industrial, agricultural, and social activities. Although NH<sub>3</sub> contributes significantly to the nutritional needs of plants and biological systems, the gas itself can create odors and have negative impacts to animal and human health (Wu *et al.*, 2008). The Occupational Safety and Health Administration (OSHA) has set a 15 min exposure limit for gaseous NH<sub>3</sub>

\*Corresponding author. Email address: juntima.c@psu.ac.th of 35 ppmv in environmental air and an 8 h- exposure limit of 25 ppmv. Many chemical industries use  $NH_3$  as reactants and/or produce  $NH_3$  from various processes. The ' $NH_3$  slip' is now a significant problem to be solved urgently to reduce  $NH_3$  pollution (Yamazoe *et al.*, 2007). Indirect  $NH_3$  emissions into the atmosphere as nitrous oxide ( $N_2O$ ) contribute to the destruction of the ozone layer which is a cause of global warming (Philippe *et al.*, 2011).

Many technologies have been used to eliminate  $NH_3$ in emission air, such as catalytic oxidation, biological filtration, liquid absorption, and solid adsorption (Guo *et al.*, 2009). Photocatalytic oxidation of  $NH_3$  into nitrogen ( $N_2$ ), carbon dioxide ( $CO_2$ ) and water ( $H_2O$ ) is a potentially available method to reduce  $NH_3$  pollution proceeds at room temperature and atmospheric pressure as shown in Equation 1- 6. A photocatalytic system consisting of catalyst with photo-active properties and light source in the UV light wavelength has been used for UV induced photocatalysis of pollutants. Titanium dioxide (TiO<sub>2</sub>) has drawn much attention from the industry as a good candidate in the form of a large band-gap semiconducting oxide or optical semiconductor material for pollutant treatment. TiO, powder has favorable physical/chemical properties, low cost, ease of availability, and high stability to be coated as thin films on media. TiO<sub>2</sub>, however, is only photoactive under UV irradiation (< 405 nm), which accounts for less than 95% of solar light energy (Kuo et al., 2011). Glass is an interesting medium to be used as a supporter of TiO<sub>2</sub> catalytic species since it is economical and corrosion resistant. Glass tube substrate can be coated with TiO<sub>2</sub> powder suspension using dip coating method and applied to increase the contact area with gas emission stream. The photocatalytic oxidation has been prevalently applied as techniques for degradation of toxic organic and inorganic compounds in air streams due to its non-toxic, inexpensive, and highly reactive characteristics.

$$\operatorname{TiO}_2 + h\nu \rightarrow e^- + h^+$$
 (1)

 $h^+ + OH^- \rightarrow OH^-$  (2)

$$e^{-}+O_2 \rightarrow O_2^{-}$$
 (3)

$$NH_3 + OH' \rightarrow NH_2' + H_2O$$
 (4)

$$NH_{2}^{+}+OH^{-} \rightarrow NH+H_{2}O$$
 (5)

$$NH+OH' \rightarrow \frac{1}{2}N_2 + H_2O \tag{6}$$

Response surface methodologies (RSM) have long been used to design and evaluate physical, chemical, and biological experiments in which the outcomes are influenced by various input factors or process variables (Shahsavani et al., 2009). RSM has been applied to optimize and evaluate interactive effects of independent factors in numerous chemical and biochemical processes. This methodology arises from experiments which include interactive effects among the variables and, eventually, it depicts the overall effects of the parameters on the process (Amini et al., 2008). The responses of the experiment can be simply related to the chosen independent factors by quadratic models. The model is fitted using multiple regressions and used to plot 3Dresponse surfaces. This method could effectively be used to form a process model and prediction for achieving the optimal NH, treatment efficiency of the photocatalytic reactor performance. Analysis of variance (ANOVA) has been employed to further justify the significance of the models.

The aim of this research is to treat  $NH_3$  in waste air using  $TiO_2$  thin film photocatalyst on glass tube supporters synthesized from  $TiO_2$  nano-powders with polyethylene glycol binder in a solvent suspension. The laboratory scale of

the continuous up-flow photocatalyst fix-bedded column system was constructed together with UV light irradiation installation. Waste gas stream was generated and forced to flow through the continuous photoreactor to contact with  $TiO_2$  photocatalyst. The system was set to study the operating parameters of light intensity, waste air flow rate, and  $TiO_2$ loading for the response of NH<sub>3</sub> treatment efficiency. RSM for experimental design and ANOVA analysis were used to respectively perform surface plots and mathematical model formations. A quadratic equation was obtained from the regression analysis in the design-expert program for the NH<sub>3</sub> treatment system and solved to seek the optimization performance. The optimum condition was validated by testing the NH<sub>3</sub> treatment efficiency in the photocatalytic reactor.

#### 2. Materials and Method

#### 2.1 Chemicals and apparatus

Commercial-grade titanium dioxide powder (TiO<sub>2</sub>, Degussa P25) consisting of 80% anatase and 20% rutile phase was used as the starting material. Ethanol (99.9%; Merck Germany) was used as a solvent in TiO<sub>2</sub> suspension preparation. Nitric acid (HNO<sub>3</sub>) (J. T. Baker, Thailand) was employed for pH adjustment and polyethylene glycol (PEG) was chosen as binder. UV fluorescence light lamps (8W, F20T12-BLB, USA) with wavelengths of 315-400 nm were used as a light source for photocatalytic performance in this work. 25% NH<sub>3</sub> analytical-grade solution was obtained from Merck. NH<sub>3</sub> solution was applied to generate NH<sub>3</sub> vapor to produce simulated NH<sub>3</sub> gas contamination by mixing with the air stream from the air compressor. Cylindrical glass tube 0.8x2.0 cm in size was used as medium supporter for catalyst coating.

#### 2.2 TiO, photocatalyst preparation

TiO<sub>2</sub> suspension was prepared by dissolving 6 g of TiO<sub>2</sub> nanoparticles in 150 ml ethanol solvent and vigorously stirred for 1 hr at room temperature. Polyethylene glycol was used as catalytic binder by adding to the prepared TiO, suspension and served as the precursor (Yu, 2000). The suspension was kept in a refrigerator (4°C) for 24 hrs until the suspension turned into a clear low viscosity solution. The suspension was then coated on the 0.8x2.0 cm cylindrical glass tube by dip coating method. The coated substrates were dried at temperature of 100°C for 1 hr and calcined at 600°C for 2 hrs. The calcined procedure was performed in flowing air at a heat rate increase of 10°C/min until the final temperature was reached. The obtained TiO, thin film catalyst on the cylindrical glass media is as shown in Figure 1. Morphology of the synthesized TiO, coating glass tube was characterized by scanning electron microscope (SEM).

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Figure 1. Synthesized TiO, on cylindrical glass tubes photocatalysts.

#### 2.3 Photocatalytic reactor and experimental set-up

The photocatalytic system with TiO<sub>2</sub> on glass tube medium supporter was set up in a laboratory scale. Continuous up-flow fixed-bed glass columns (5 cm internal diameter and 30 cm column height) were packed by the raschigring typed catalytic packing media. The columns were installed in a cylindrical stainless steel chamber ( $\pi \times 20^2 \times 50$ cm<sup>3</sup>) with vertically fixed UV lights at the chamber wall. UV lights (23-114 W/m<sup>2</sup>) at 315-400 nm wavelengths (power consumption of 8 W per lamp) were chosen as the light source to create photocatalytic reaction. Air stream flowed through the reactor chamber to regulate a constant temperature in the system. The diagram of NH<sub>3</sub> contaminated waste air flow and the photocatalytic system is depicted in Figure 2.

The NH<sub>3</sub> contaminated air stream was continuously generated for the NH<sub>3</sub> treatment testing by photocatalytic system. Air flow from the air compressor was passed through a NH<sub>3</sub> tank (25% w/w) to pick up vaporized NH<sub>3</sub> gas and moisture at room temperature (28-29°C). The mixed-gas stream was forced to the NH<sub>3</sub> mixing tank under steady flow to the photocatalytic reactor. The waste air contaminated with NH<sub>3</sub> at concentration of 300 ppmv was constantly used to investigate the system capacity. NH<sub>3</sub> concentrations of the inlet waste air stream were measured every 15 min to monitor and regulate a constant concentration throughout the experiment.



Figure 2. Schematic diagram of NH<sub>3</sub> contaminated waste air generation process and the photocatalytic reactor treatment system.

#### 2.4 System performance analytical methods

NH<sub>3</sub> treatment efficiency (% *eff*) by photocatalytic continuous system performance was calculated employing Equation 7 through concentration of NH<sub>3</sub> in the inlet and outlet waste air streams. The ammonia gaseous sample from the treatment system was bubbled for 3 min through a glass bottle containing 50 ml boric acid solution. NH<sub>3</sub> concentrations in the waste air were determined by titration method (Rodrigues *et al.*, 2007).

$$\% eff = \frac{C_i - C_f}{C_i} \times 100 \tag{7}$$

where %*eff* is the percentage of NH<sub>3</sub> treatment efficiency,  $C_i$  and  $C_f$  are NH<sub>3</sub> concentrations (ppmv) in the waste air at the inlet and outlet points of the photocatalytic reactor, respectively.

#### 2.5 Experimental design and analysis

Process experimental design for model formation of NH, in the waste air treatment was carried out using response surface methodology. RSM encompasses mathematical and statistical techniques that are useful for optimization performing and experimental designs (Ye et al., 2009; Ling et al., 2010). In this study, RSM was used to assess the relationship between the response (NH, treatment efficiency, % eff) and the independent variables, as well as to optimize the relevant conditions of variables in order to predict the best value of the responses. Central composite design (CCD), the most widely used approach of RSM, was employed to determine the effect of operational variables on the NH, treatment efficiencies. CCD allows reasonable amount of information to test lack of fit when a sufficient number of experimental values exist (Guven et al., 2008). CCD and RSM were established with the help of the Design Expert software program. The three significant independent variables considered in this study were light intensity  $(X_i)$  in the range of 23-114 W/m<sup>2</sup>, waste air flow rate (X<sub>2</sub>) in the range of 1–5  $1/\min$ , and TiO<sub>2</sub> loading (X<sub>2</sub>) in the range of 1.18-5.90 g. The responses can be simply related to the independent factors by a quadratic model consisting of linear, two factorial, and quadratic terms as given in Equation 8.

$$y = \beta_0 + \sum_{i=1}^k \beta_i x_i + \sum_{i=1}^k \beta_{ii} x_i^2 + \left(\sum_{i=1}^{k-1} \sum_{j=i+1}^k \beta_{ij} x_i x_j\right)_{i < j}$$
(8)

where y is the predicted response evaluated;  $x_i$  and  $x_j$  are the independent variables;  $\beta_0$  is the constant coefficient;  $\beta_j$ ,  $\beta_{jj}$ , and  $\beta_{ij}$  are the interaction coefficients of linear, quadratic and second-order terms, respectively; and k is the number of studied factors.

Analysis of variance was used for graphical analyses of the data. The quality of the fit was expressed by the value of correlation coefficient ( $R^2$ ), and its statistical significance was checked by F-value (Fisher variation ratio). Model terms were evaluated by the P-value (probability) with 95% confidence level (Sahoo *et al.*, 2010). Three-dimensional plots and their respective contour plots were obtained for  $NH_3$  treatment based on the effects of the three variables at five levels.

#### 3. Results and Discussions

#### 3.1 Scanning electron micrographs of TiO, films

A dip coating method on the small glass tubes described in Section 2.2 was employed to effectively perform the catalytic preparation with suspension of pure  $\text{TiO}_2$  nano powder, ethanol solvent, and polyethylene glycol as binder. Figure 3 shows the scanning electron microscope of the surface of the synthesized  $\text{TiO}_2$  on one of the glass tubes. It is observed that  $\text{TiO}_2$  nanoparticles were well dispersed on the surface of the glass tube substrate.  $\text{TiO}_2$  surface is rough and large size particles are formed on the surface.

#### 3.2 Stability of TiO, photocatalyst on NH, treatment

 $\text{TiO}_2$  photocatalyst coated on glass tubes was used for the treatment of NH<sub>3</sub> in contaminated waste air at NH<sub>3</sub> concentration of 300 ppmv. The glass tubes were packed in a raschig-ring typed packing media in a series of three glass columns to increase the contact area among the catalyst, UV light, and the waste gas. The efficiency and stability of TiO<sub>2</sub> catalyst on NH<sub>3</sub> treatment from the waste air using photocatalytic reactor were investigated by plotting between %NH<sub>3</sub> treatment efficiency and irradiation time, as shown in Figure 4. Experimental conditions of 3 l/min waste air flow rate and TiO<sub>2</sub> loading of 3.54 g were set. Tests were performed three times under the same testing conditions to verify stability of the synthesized catalyst and the system. Efficiency of the NH<sub>3</sub> treatment was monitored for consistency of the system every 15 minutes for 2 hours.

The results show that  $TiO_2$  catalyst on the raschigring typed glass packing media performed effectively in the NH<sub>3</sub> treatment with constant efficiency throughout the operating time of 2 hours. After repeated operations, similar performance of the system with 60% efficiency was obtained. This indicates stability of the TiO<sub>2</sub> coated catalysts packed in the photoreactor column exposing to waste air flow and UV light irradiation. Also the TiO<sub>2</sub> catalyst, under constantly and continuously high activity UV light exposure, can effectively be regenerated after reacting with NH<sub>3</sub> in the waste air.

#### 3.3 Model fitting and statistical analysis

Process and parameters of the NH<sub>3</sub> treatment in the photocatalytic reactor were designed and investigated by RSM and CCD design. Table 1 shows the coded value ranges of the three operating variables  $(X_i, X_j, \text{ and } X_3)$  that were set at five coded levels:  $-\alpha$ , -1, 0, +1,  $+\alpha$  (Sahoo *et al.*, 2010). The coded values for the variable parameters were used to facilitate regression with  $-\alpha$  as the minimum level and  $+\alpha$  as the maximum level. The center runs (0, 0, and 0) were repeated three times, leading to 20 experimental runs as they contribute to the estimation of the quadratic terms in the process model.

Experimental design parameters by CCD and the response of NH<sub>3</sub> treatment efficiency in the TiO<sub>2</sub> photocatalytic reactor are given in Table 2. Relationships between the three variables (light intensity, waste air flow rate and TiO<sub>2</sub>)



Figure 3. SEM image of the surface of  $\text{TiO}_2$  on glass tube substrate of  $\text{TiO}_2$  suspension.



Figure 4. Stability of  $TiO_2$  photocatalyst, indicated by slight changes under repeating irradiation.

Table 1. Experimental range and level of the independent variable.

Variable	Range and level					
variable	-α	-1	0	+1	$+\alpha$	
$X_i$ : light intensity (W/m <sup>2</sup> )	23	45	68	92	114	
$X_{2}$ : waste air flow rate (l/min)	1	2	3	4	5	
$X_{3}^{2}$ : TiO <sub>2</sub> loading (g)	1.18	2.36	3.54	4.72	5.90	

_		Variable				
Run no. $X_1$ Light intensity (W/m <sup>2</sup> )	$\begin{array}{c} X_2 \\ \text{Waste air flow rate} \\ (l/min) \end{array}$	$\operatorname{TiO}_2^{X_3}$ loading (g)	NH <sub>3</sub> treatment eff. (%)			
1	23	3	3.54	47.40		
2	45	4	2.36	35.00		
3	45	2	2.36	46.20		
4	45	2	4.72	75.30		
5	45	4	4.72	60.10		
6	68	3	3.54	56.55		
7	68	3	3.54	55.77		
8	68	1	3.54	63.20		
9	68	3	3.54	56.10		
10	68	2	3.54	58.90		
11	68	3	4.72	68.80		
12	68	5	3.54	49.78		
13	68	3	1.18	49.63		
14	68	3	5.9	87.00		
15	92	2	4.72	75.60		
16	92	4	4.72	73.50		
17	92	2	2.36	62.99		
18	92	4	2.36	53.00		
19	92	3	3.54	58.70		
20	114	3	3.54	70.20		

Table 2. Experimental condition of CCD and response of NH<sub>3</sub> in waste air treatment efficiency by the photocatalytic system.

Table 3. ANOVA for adequacy of the quadratic model of NH, treatment in TiO, photocatalytic reactor.

Source	Sum of squares	DF	Mean Square	F-value	Prob > F	Remarks
Model	2,809.44	6	468.24	97.66	< 0.0001	significant
$X_{i}$	542.19	1	542.19	113.08	< 0.0001	significant
$X_{2}$	273.33	1	273.33	57.01	< 0.0001	significant
X,	1,709.47	1	1,709.47	356.55	< 0.0001	significant
$X X_{2}$	25.33	1	25.33	5.28	0.0388	significant
$X_{1}X_{2}$	54.83	1	54.83	11.44	0.0049	significant
$X_{2}^{2}$	243.49	1	243.49	50.78	< 0.0001	significant
Residual	62.33	13	4.79			-
Lack of Fit	62.02	11	5.64	36.78	0.0268	significant
Pure Error	0.31	2	0.15			-
Correlation Total	2,871.77	19				
Standard deviation	2.09					
R <sup>2</sup>	0.9783		Adjusted R <sup>2</sup>	0.9683		
Predicted R <sup>2</sup>	0.9306		Adequate Precision	41.48		

loading) and the responses were analyzed using regression analysis of the Design Expert software.

Fitting of the process data and the response to the quadratic models were performed. Their subsequent ANOVA analyses showed that the photocatalytic of NH<sub>3</sub> treatment was most suitably described by a quadratic model as shown

in Table 3. Regression coefficients of the model describing the NH<sub>3</sub> treatment were determined by F-value and P-value. Corresponding P-values suggest that, among the test variables used in this study,  $X_1$ ,  $X_2$ ,  $X_3$ ,  $X_1X_2$ ,  $X_1X_3$ , and  $X_3^2$ are significant model terms. Other model terms are not significant (with a probability value less than 0.05). The effects of the terms on the response could be easily seen from the normalized coefficients. Based on the results, the response surface constructed in this study for predicting NH<sub>3</sub> treatment efficiency was considered reasonable. The final regression model, in terms of their coded factors, is expressed by the following reduced quadratic Equation 9.

. . . . . .

$$y = 38.33 + 0.39X_1 - 8.04X_2 - 0.44X_3 + 0.06X_1X_2 - 0.09X_1X_3 + 2.18X_3^2$$
(9)

. . . . .

ANOVA results of the reduced quadratic model in Table 3 indicated that the model could adequately be used to describe the NH, treatment under a range of operating conditions. Data given in this Table demonstrate that the quadratic model was significant at 5% confidence level since P-values (0.0001) were less than 0.05. Values of P>F less than 0.0500 indicate that model terms are significant, while values greater than 0.1000 indicate that the model terms are not significant (Korbahti et al., 2008). The value of correlation coefficient  $(R^2=0.9783)$  obtained in the present study for NH<sub>3</sub> treatment indicated that only 1.66% of the total dissimilarity might not be explained by the empirical model. The quadratic equation obtained by multiple variables can be used to predict the NH, treatment efficiency. Figure 5 plots between predicted responses and actual values reveal that the predicted responses of the reduced quadratic model agree well with the actual ones in the range of the operating variables.

# 3.4 Effect of independent variables on NH<sub>3</sub> in waste air treatment

Individual and interaction effects of the three factors consisting of light intensity, waste air flow rate, and  $\text{TiO}_2$  loading on the NH<sub>3</sub> treatment efficiency were presented by the quadratic model (Equation 3). Three-dimensional surface plot of the NH<sub>3</sub> treatment by the fitting quadratic model from the regression analysis are given in Figure 6-8. These plots illustrate the response of different experimental variables of the photocatalytic system. All 3D plots can be used to explain and identify the effect of each variables and major interactions between the variables. The variables: light intensity, waste air flow rate and TiO<sub>2</sub> loading, were tested on the treatment of NH<sub>3</sub> individually. It was also important to check the interaction effects.

#### 3.4.1 Effect of light intensity and waste air flow rate

The 3D plot in Figure 6 indicates significant interaction between light intensity  $(X_1)$  and waste air flow rate  $(X_2)$ on the NH<sub>3</sub> treatment efficiency. It is observed that treatment efficiency significantly increases with increasing light intensity and decreasing waste air flow rate. As can be seen in Figure 6 at NH<sub>3</sub> concentration of 300 ppmv, the highest % of treatment efficiency is reached at 114 W/m<sup>2</sup> of light intensity and waste air flow rate of 1 l/min. Interaction of light intensity at 23  $W/m^2$  and the same waste air flow rate, however, presented very low efficiency of NH<sub>3</sub> treatment (%). The photonic nature of the photocatalysis reaction has emphasized the dependency of the overall photocatalytic rate on the light source used. Light intensity is major factor that affect the degree of photocatalytic reaction on organic substrates because electron-hole pairs can be produced more by light energy (Li *et al.*, 2008). There is more photon activation per unit time and unit area at higher light intensity.

# 3.4.2 Effect of light intensity and TiO<sub>2</sub> loading

In Figure 7, the two-factor interaction of light intensity and TiO<sub>2</sub> loading was investigated when NH<sub>3</sub> concentration was fixed at 300 ppmv. This interaction ( $X_1X_3$ ) exemplifies the real importance of TiO<sub>2</sub> loading on the NH<sub>3</sub> treatment. As shown in this figure, the highest value of treatment (83.5%) was observed at the highest light intensity (114 W/m<sup>2</sup>) with highest TiO<sub>2</sub> loading (5.9 g). However, there is a significant decrease on the treatment efficiency in the presence of low







Figure 6. 3D-surface plot on the effect of light intensity  $(X_1)$  and waste air flow rate  $(X_2)$  on the NH<sub>3</sub> treatment efficiency under 3.54 g TiO<sub>2</sub> loading at 300 ppmv NH<sub>3</sub> concentration.



Figure 7. 3D surface plot on the effect of light intensity  $(X_1)$  and TiO<sub>2</sub> loading  $(X_3)$  on the NH<sub>3</sub> treatment efficiency under 3 l/min waste air ow rate at 300 ppmv NH<sub>2</sub> concentration.

light intensity, especially at low TiO<sub>2</sub> loading (1.18 g). Higher TiO<sub>2</sub> loadings in the photocatalytic reactor generally lead to higher efficiency of NH<sub>3</sub> removal from the waste air. The TiO<sub>2</sub> photocatalyst generates a lot of active sites on the surface which leads to •OH radicals generation. The increase in the amount of active sites in accordance with the increase in TiO<sub>2</sub> photocatalyst loading consequently lead to enhancement of the production of •OH radicals (Abdollahi *et al.*, 2012). The figure clearly shows that higher NH<sub>3</sub> treatment efficiency depends on the higher amount of TiO<sub>2</sub>, as well as higher light intensity. The strong interaction between the light intensity and the TiO<sub>2</sub> loading ( $X_TX_3$ ) indicates an important interdependence in the response of treatment.

#### 3.5 Optimization of NH, treatment conditions

An optimization process was carried out to determine the optimum value of  $NH_3$  treatment efficiency using the Design Expert software. Parameters used in the software for each operational condition (light intensity, waste air flow rate, and TiO<sub>2</sub> loading) were set as constraints within the upper and lower limit ranges. The NH<sub>3</sub> treatment model (Equation 9) was employed as objective function for the response of NH<sub>3</sub> treatment efficiency at "maximum" to achieve the highest performance. The program combines the constraints into the objective function, and then searches for the best combination to maximize this function. Accordingly, the optimum working condition and respective percent treatment efficiencies were established. As presented in Table 4, a maximum 91.45% NH<sub>3</sub> treatment efficiency was predicted according to the objective function and constraints under optimum operational conditions at light intensity of 114 W/  $m^2$ ; 1 l/min of waste air flow rate; and TiO<sub>2</sub> loading of 5.9 g.

An experiment was then performed to validate and confirm the optimum results of the optimization performance. Efficiency of the system was monitored for consistency and stability of the system every hour for six hours. Figure 8 shows the NH<sub>3</sub> treatment efficiency along with irradiation time using the optimum conditions derived earlier. An average NH<sub>3</sub> treatment efficiency of 90.02%, shown also in Table 4, was obtained from the experiment which agrees well with the predicted response value (91.45%). The effectiveness of TiO<sub>2</sub> photocatalytic supporting on glass cylindrical tubes for continuous NH<sub>3</sub> pollutant treatment at laboratory scale is certain, as shown here, with a great potential to be proven in an industrial scale waste air treatment system.

Compared to literature data, reasonably successful results were obtained in this study under conditions from process modeling of  $NH_3$  contaminated waste air treatment in photocatalytic reactor using TiO<sub>2</sub> coated glass tubes. This treatment system could be regenerated by UV light and continuously run with more efficiency than other traditional waste air treatment methods (Rodrigues *et al.*, 2007; Guo *et al.*, 2009). A simple and efficient design of catalyst preparation and photo reactor system was applied to accommodate and support for high waste gas flow treatment.

#### 4. Conclusions

In the present study, NH<sub>3</sub> contaminated waste air treatment using TiO, nanoparticle coating on glass tubes





Table 4. Model validity of response surface for NH, treatment under optimal condition.

Light intensity $(W/m^2)$	Waste air flow rate	low rate $\text{TiO}_2$ loading $\text{NH}_3$ treatment efficient		
$(\mathbf{W}_{l})$	$(X_2)$	(g) $(X_3)$	Predicted	Experimental
114	1	5.9	91.45	90.02

inside a continuous photocatalytic reactor was investigated. The treatment process focused on the interaction effects of operating variables such as light intensity, waste air flow rate and TiO, loading using RSM with CCD. The multiple correlation coefficient of determination  $R^2$  obtained was 0.9783, indicating that the acquired data fitted well with the predicted data from the quadratic model. Regression analysis, ANOVA, and response surface were carried out and plotted using Design Expert software for the prediction of experiment responses. The optimum result derived from the model indicated that 114 W/m<sup>2</sup> of UV light intensity together with a waste air flow rate of 1 l/min and a TiO<sub>2</sub> loading of 5.9 g was most suitable to achieve 91.45% of NH<sub>3</sub> treatment at 300 ppmv, and was verified by an actual experimental result of 90.02%. Thus, TiO, coated on glass tubes can be effectively used for efficient treatment of NH, from waste air with high potential to be applied in industrial-scaled NH, contamination remedies.

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