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# Optimization of Oxytetracycline Degradation Inside UV/$H_2O_2$ Reactor Using Box-Behnken Experimental Design

[1]A.U. Rahmah, [1,2]S. Harimurti, [1]A.A. Omar and [1]T. Murugesan
[1]Department of Chemical Engineering, Universiti Teknologi PETRONAS, Perak, Malaysia
[2]Department of Pharmacy, Faculty of Medicine and Health Sciences,
Muhammadiyah University, Yogyakarta, Indonesia

**Abstract:** Oxytetracycline degradation inside UV/$H_2O_2$ system was carried out through mediation of hydroxyl radical attack which oxidizes recalcitrant, toxic and non-biodegradable compounds to various by-products. The Box-Behnken design of experiment was applied to find the optimum condition of OTC degradation. Three levels fractional design which consist of a full $2^2$ factorial with three centre points resulted incomplete block design were resulted from the chosen design. Temperature at 30-50°C, pH range 3-11 and oxidant/pollutant ratio 0.5:1-1.5:1 were chosen as the experimental factors. Total organic carbon was measured for monitoring degradation efficiency. Percentage of total organic carbon removal was chosen as the response. Based on Pareto chart, the quadratic effect of pH was the most significant factor that minimizes percentage of total organic carbon removal. As suggested by the model, the maximum total organic carbon removal could be obtained at 30°C, pH 6.30 and 1.5 mole equivalent of oxidant/pollutant ratio. At its optimum condition, about 95.35% and percentage total organic carbon removal was achieved. At the optimum condition, the ratio of $BOD_5$/COD value was 1.19, which implied the biodegradability of the degradation product of oxytetracycline.

**Key words:** Optimization, oxytetracycline, UV/$H_2O_2$, Box-Behnken

## INTRODUCTION

Our society has consumed high amount of antibiotics for the treatment of bacterial diseases in humans and animal. After being consumed, antibiotics may leave the human or animal body through excretion, in metabolized or unmetabolized form and entered the water stream as contaminant (Li *et al*., 2008). Subsequent discharge from hospital or other medical facilities also may become the sources of antibiotics contamination (Xu *et al*., 2007). These contaminants have been detected in surface and ground water, drinking water, tap water, ocean water, sediments and soil (Klavarioti *et al*., 2009; Kummerer, 2009). Antibiotics accumulation in the natural ecosystem may produce antibiotic-resistant bacteria within the bacteria host or modification of the indigenous microbiota (Halling-Sorensen *et al*., 2002). It may also damage the liver and kidney organ of rainbow trout and lethal to brine shrimp (Martinez, 2009; Garofalo *et al*., 2007; Gulkowska *et al*., 2008). Antibiotics could also remain inside the animals tissue as food pollutant which might trigger the allergic reactions (Cabello, 2006).

Four classes of antibiotics were known, namely: β-lactams, tetracyclines, aminoglycosides, quinolones, macrolides, glycopeptides and sulfonamides. Oxytetracycline hydrochloride (OTC) is belongs to Tetracycline classes of antibiotics. OTC is the most widely used antibiotics in livestock productions (Arikan *et al*., 2008). Therefore, in this study OTC is chosen as the model source of antibiotics contaminant. A chemical structure of OTC is shown in Fig. 1.

One of important features of antibiotics is their resistance towards biological degradation. Therefore many researches have been investigated towards non-biological process for organic destruction. Advanced Oxidation Processes (AOPs) have emerged as an effective non-biological process that capable of transforming organic pollutant into non toxic substances. AOPs generally utilizes generation of hydroxyl radicals ($^{\bullet}OH$) or any other highly reactive species to attack the organic pollutant. After the radical attack, the organic pollutant will undergo a series of degradations oxidation reaction and lead to the formation of $CO_2$ and $H_2O$ as the final product.

Oxytetracycline degradation have already conducted in many AOPs systems, such as Fenton process, water hydrolysis, ozone process, UV photolysis, UV-$TiO_2$-zeolite photocatalytic system, enzymatic degradation and also γ and pulse irradiation (Doi and Stoskopf, 2000; Bautiz and Nogueira, 2007; Hassani *et al*., 2008; Jiao *et al*.,

**Corresponding Author:** A.U. Rahmah, Department of Chemical Engineering, Universiti Teknologi PETRONAS, Perak, Malaysia

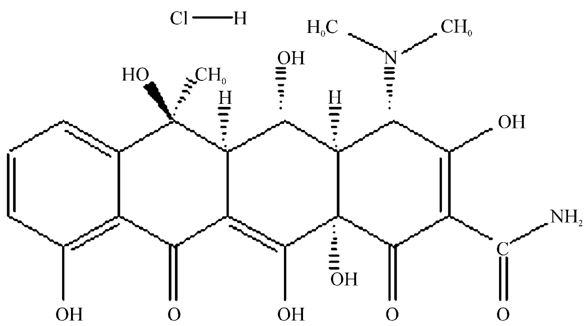

Fig. 1: Chemical structure of OTC

2008; Loftin *et al*., 2008; Uslu and Balcioglu, 2009; Zhao *et al*., 2010; Jeong *et al*., 2010). However, limited studied have been conducted in studying OTC degradation inside UV/$H_2O_2$ system (Lin *et al*., 2010). In this paper, OTC degradation inside an UV/$H_2O_2$ system was studied. Its efficiency was monitored trough Total Organic Carbon (TOC) value. Box-Behnken experimental design was applied to evaluate the percentage of TOC removal at different temperature, pH and oxidant/pollutant ratio. Statistical analyses using analysis of variance (ANOVA) were performed to evaluate the model obtained. Oxytetracycline degradation have already conducted in many AOPs systems, such as Fenton process, water hydrolysis, ozone process, UV photolysis, UV-$TiO_2$-zeolite photocatalytic system, enzymatic degradation and also γ and pulse irradiation (Doi and Stoskopf, 2000; Bautiz and Nogueira, 2007; Hassani *et al*., 2008; Jiao *et al*., 2008; Loftin *et al*., 2008; Uslu and Balcioglu, 2009; Zhao *et al*., 2010; Jeong *et al*., 2010). However, limited studied have been conducted in studying OTC degradation inside UV/$H_2O_2$ system (Lin *et al*., 2010). In this paper, OTC degradation inside an UV/$H_2O_2$ system was studied. Its efficiency was monitored trough Total Organic Carbon (TOC) value. Box-Behnken experimental design was applied to evaluate the percentage of TOC removal at different temperature, pH and oxidant/pollutant ratio. Statistical analyses using analysis of variance (ANOVA) were performed to evaluate the model obtained.

## MATERIALS AND METHODS

Oxytetracycline hydrochloride (Merck, Germany) was used as the contaminant model. Hydrogen peroxide 30% (Merck, Germany) was used as the source of hydroxyl radical. NaOH (Merck, Germany) and $H_2SO_4$ 98% (Merck, Germany) were added to adjust the pH of sample solution.

A glass-jacketed reactor equipped with low pressure UV lamp (8 W) was used throughout the experiment (Fig. 2). About 400 mL working volume was applied. Homogeneity of solution was maintained by continuously stirring the solution with magnetic stirrer. Its temperature was maintained using cooling water flowing inside the reactor jacket. The progress of degradation was monitored by means of TOC value. Before the reaction starts, antibiotic and $H_2O_2$ were added. About 5 mL of liquid samples were drawn from the reactor at 0 and 180 min. Hence, the volume taken was considered negligible to the total sample volume. About 180 min degradation times were applied for each experiment. Biodegradability of sample at its optimum condition was measured by analyzing $BOD_5$/COD ratio.

**Analyses:** TOC analyses were carried out using a TOC analyzer (TOC-VCSH, Shimadzu, Japan). Biodegradability test were performed at the optimum operating condition, as suggested by the model, by evaluating the value of $BOD_5$/COD. BODTrak™ (HACH, USA) was used to measure the value of Biological Oxygen Demand for five days ($BOD_5$). COD was measured by using COD Test'N Tube (HACH, USA). DR 5000 spectrophotometer (HACH, USA) was used to determine the value of COD reading at 420 nm (DR 5000, HACH, USA).

**Statistical analysis:** Optimization experiments were conducted inside a UV/$H_2O_2$ system by varying temperature (A), initial pH (B) and oxidant/pollutant mole ratio (C). Box-Behnken experimental design combined with Response Surface Modelling (RSM) was chosen. The experiment was constructed with three levels fractional designs which consist of a full $2^2$ factorial with three centre points resulted incomplete block design. OTC concentration was kept constant at 250 ppm. Three level factor were chosen and coded as -1 (low), 0 (middle or central point) and 1 (high). The factors included pH range 3, 7, 11, temperature 30, 40, 50°C and oxidant/pollutant ratio 0.5, 1.0, 1.5 and the optimize response is to maximize the percentage of TOC removal. The intervals were chosen based on our preliminary study (Rahmah *et al*., 2011). Table 1 shows the Box-Behnken operational matrix with coded factors. Statistical analyses were performed using Statgraphic Centurion 15 (Statpoint Technologies, Inc, USA). The Box-Behnken method was selected due to that fewer variable are required to estimate a potentially complex response function. Experimental data obtained were fitted to the second-order polynomial equation as described by Eq. 1 and optimized for the maximum percentage of TOC removal. True values of the unknown parameters are represented by $\beta_o$, $\beta_i$, $\beta_{ii}$ and $\beta_{ij}$ coefficients, with i and j symbolize the factors. Therefore, $x_i$, $x_i^2$ and $x_ix_j$ symbolize the single interaction, quadratic

Table 1: Box-Behnken design matrix with response value for OTC degradation

| | Operational parameter | | | Response % TOC Removal | |
|---|---|---|---|---|---|
| Experiment | Temp. (°C) | pH | Oxidant/pollutant ratio | Experimental value | Fitted value |
| 1 | -1 | -1 | 0 | 81.73 | 79.67 |
| 2 | 1 | 0 | -1 | 90.90 | 89.45 |
| 3 | 0 | -1 | 1 | 87.20 | 87.80 |
| 4 | -1 | 0 | -1 | 81.83 | 83.30 |
| 5 | 1 | 0 | 1 | 82.33 | 80.86 |
| 6 | 1 | -1 | 0 | 90.12 | 90.98 |
| 7 | 0 | 0 | 0 | 89.83 | 89.34 |
| 8 | 0 | 1 | -1 | 74.68 | 74.08 |
| 9 | 0 | 0 | 0 | 88.55 | 89.34 |
| 10 | 0 | 0 | 0 | 89.63 | 89.34 |
| 11 | -1 | 1 | 0 | 82.43 | 81.57 |
| 12 | 1 | 1 | 0 | 61.46 | 63.52 |
| 13 | -1 | 0 | 1 | 92.30 | 93.75 |
| 14 | 0 | 1 | 1 | 66.55 | 65.96 |
| 15 | 0 | -1 | -1 | 77.22 | 77.81 |

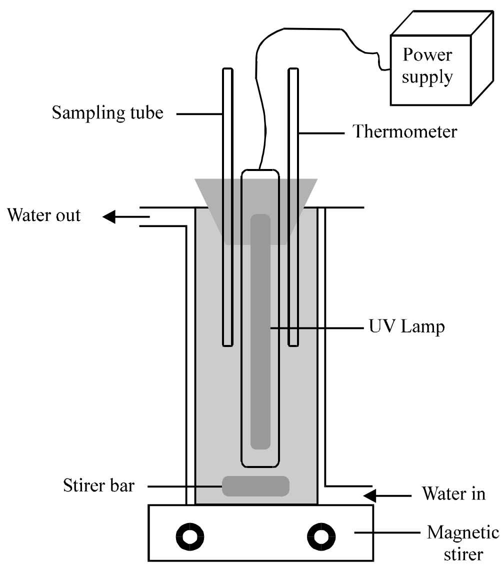


Fig. 2: Experimental setup for OTC degradation

interaction of one factor and two factors interacting with each other, respectively. ANOVA study was performed to identify the significant factors at p = 0.05.

$$y=\beta_0+\sum_{i=1}^{k}\beta_i x_i+\sum_{i=1}^{k}\beta_{ii}x_i^2+\sum_{i<j}\sum\beta_{ij}x_i x_j+\varepsilon \tag{1}$$

## RESULTS

Oxytetracycline degradation inside a UV/$H_2O_2$ was studied by varying temperature, initial pH and oxidant/pollutant mole ratio. The observed and fitted TOC removal (%) values were shown in Table 1. Percentage of TOC removal was ranging from 61.4 to 92.3 %. It shows

Table 2: Coefficient of fitted equation

| Coefficient Value | Symbols | Estimate |
|---|---|---|
| Constant | $\beta_0$ | -40.7043 |
| A: Temperature | $\beta_i$ | 2.05767 |
| B: pH | $\beta_j$ | 17.1181 |
| C: oxidant/pollutant mole ratio | $\beta_k$ | 74.9404 |
| AA | $\beta_{ii}$ | 0.000129167 |
| AB | $\beta_{ij}$ | -0.1835 |
| AC | $\beta_{ik}$ | -0.952 |
| BB | $\beta_{jj}$ | -0.650911 |
| BC | $\beta_{jk}$ | -2.26375 |
| CC | $\beta_{kk}$ | -10.0383 |

that the proposed fitted model is suitable for predicting percentage TOC removal, revealing a reasonably good agreement (Fig. 3). High value of $R^2$ and adjusted $R^2$ value at 0.9823 and 0.9503 were obtained. The correlation factor or $R^2$ describes the variability degree of the experimental value response and ranges between of 0 and 1. These high $R^2$ coefficients ensured a satisfactory adjustment of the second order polynomial models to the experimental data. Therefore, the response surface models could be satisfactory for predicting percentage variation of OTC degradation. The coefficients for fitted equation are shown in Table 2 while the resulted polynomial equation was shown in Eq. 2:

$$y=\beta_0+\beta_i A+\beta_j B+\beta_k C+\beta_{ii}A^2+\beta_{jj}B^2+\beta_{kk}C^2+\beta_{ij}AB+\beta_{ik}AC+\beta_{jk}BC \tag{2}$$

ANOVA analyses were presented in Table 3 which indicates the factor and interaction between the factors that significant to degradation of OTC. Significant response model ($p<0.05$). Pareto charts describe the relative importance of the factor and also the effect of factor setting adjustment, by displaying the most influencing factor followed by the least one. Figure 4

Table 3: ANOVA analyses of response

| Factors | Statistic<br>Sum of squares | df | Mean square | F-ratio | p-value |
|---|---|---|---|---|---|
| A: Temperature | 22.7138 | 1 | 22.7138 | 5.45 | 0.0669 |
| B: pH | 327.04 | 1 | 327.04 | 78.44 | 0.0003 |
| C: oxidant/pollutant mole ratio | 1.75781 | 1 | 1.75781 | 0.42 | 0.5448 |
| AA | 0.00061603 | 1 | 0.00061603 | 0.00 | 0.9908 |
| AB | 215.502 | 1 | 215.502 | 51.69 | 0.0008 |
| AC | 90.6304 | 1 | 90.6304 | 21.74 | 0.0055 |
| BB | 400.481 | 1 | 400.481 | 96.06 | 0.0002 |
| BC | 81.993 | 1 | 81.993 | 19.67 | 0.0068 |
| CC | 23.2542 | 1 | 23.2542 | 5.58 | 0.0646 |
| Total error | 20.8455 | 5 | 4.1691 | | |
| Total (corr) | 1175.08 | 14 | | | |

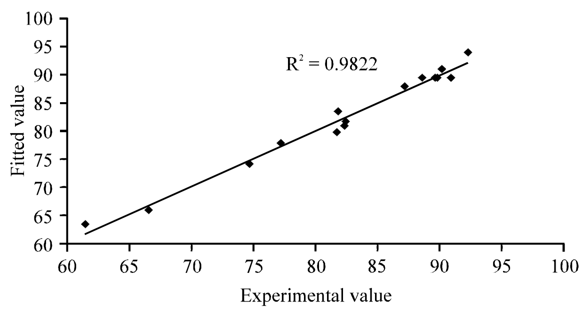


Fig. 3: Relation between experimental and fitted value of percentage TOC removal

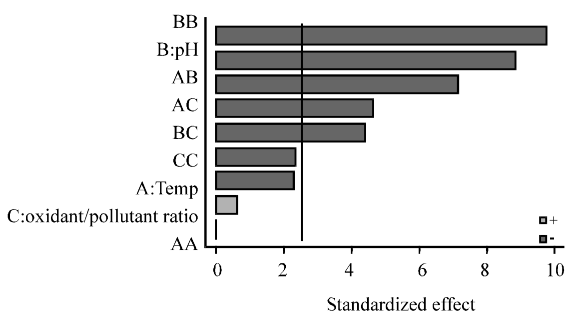


Fig. 4: Pareto chart for percentage TOC removal

shows the Pareto charts of the standardized effects of the interaction between each factor affecting the percentage of TOC removal. The most significant factors in minimizing percentage TOC removal is the quadratic effect of pH, followed by pH as a single factor, interaction between temperature and pH, interaction between temperature and oxidant/pollutant ratio and lastly pH and oxidant/pollutant interaction ratio. The quadratic effect of pH has the lowest p-value compared to any other interaction (Table 3), hence it implied that as the most significant interaction that minimize the percentage of TOC removal.

Table 4: Biodegradability test of OTC before and after degradation

| OTC sample | $BOD_5$ | COD | $BOD_5$/COD |
|---|---|---|---|
| Before degradation | 19.75 | 596 | 0.03 |
| After degradation | 61.75 | 52 | 1.19 |

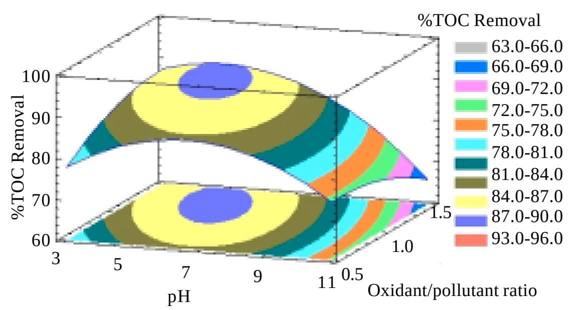


Fig. 5: Contour plots for percentage TOC removal

Location and curvature shape can be predicted by deriving the contour plots from the fitted data. The bull's eye of the curvature is the ideal position of the optimum point. Figure 5 suggests that the lowest percentage TOC removal was obtained at 30°C, pH 6.30 and oxidant/pollutant ratio at 1.5 mole equivalents. About 95.35% of TOC removal was obtained during the experimental work at the optimum condition suggested by the statistical model. Percentage difference of the experimental and fitted value for TOC removal is about 1.37%. Any points outside the interval tested in the experiment resulted in lower TOC removal, as suggested by the fitted model. Even though the best prediction of the model lies within the range applied. Biodegradability test was also performed on the final products of OTC degradation using BODTrak$^{TM}$, at its optimum condition. Table 4 shows significant increased in the ratio of $BOD_5$/COD, starting with ratio 0.03 to 1.19. This increment shows OTC-degraded product has higher biodegradability compared to OTC sample before degradation.

## DISCUSSION

Pareto chart suggested that the factor pH that mostly influenced the percentage of TOC removal, whereas the quadratic effect of pH, pH as single factor, interaction of pH and temperature and lastly the interaction between pH and oxidant/pollutant mole ratio. At higher pH, $H_2O_2$ will undergo faster self-decomposition rate (Eq. 3) which lead to reduced capabilities of attacking organic compound (Aleboyeh *et al*., 2005). Cleavage of $H_2O_2$ molecules into hydroxyl radical is the most commonly accepted mechanism for photolysis $H_2O_2$ (Eq. 4) which was found to be pH dependent (Legrini *et al*., 1993). Higher photolysis rate would increase at alkaline pH which might be mainly due to higher molar absorption coefficient (at 253.7 nm) of the hydroperoxy anion ($HO_2$ 240 $M^{-1}$ $cm^{-1}$) compared to $H_2O_2$ (18.6 $M^{-1}$ $cm^{-1}$). Deprotonation of $H_2O_2$ produced a conjugated base, mentioned as hydroperoxy anion (Eq. 5) which was regulated by acid-base reaction (White, 2010). However, high alkaline condition may also cause $H_2O_2$ to decompose by a dismutation reaction with a maximum rate at the pH of its $pK_a$ value (Eq. 6) which led to lower degradation rate (Legrini *et al*., 1993). Structural conformation of OTC may also changes based on pH which also influenced its stability towards degradation (Doi and Stoskopf, 2000; Jiao *et al*., 2008; Zhao *et al*., 2010). OTC is a zwitterionic molecule which could exist as positively charged (pH ~3), neutral (pH ~7) and negatively charged (pH ~10). At lower pH, OTC tends to has higher stability toward degradation due to its structural conformation, followed by neutral and the lowest at alkaline condition. Higher photolysis rate were found for OTC at higher pH (Jiao *et al*., 2008). Therefore, by affecting the amount of hydroxyl radical and structural stability of OTC, quadratic effect of pH become the most significant factor for percentage TOC removal. The optimum pH suggested by the model is 6.34 which were in accordance with the result from Lin and colleague (Lin *et al*., 2010). Based on their study on oxytetracycline degradation, better efficiency was achieved at acid (pH 5.5) and near neutral medium (pH 7.3) than in alkaline medium (pH 8.4):

$$H_2O_2 \rightarrow 2H_2O + O_2 \tag{3}$$

$$H_2O_2 \rightarrow 2{}^{\bullet}OH \tag{4}$$

$$H_2O_2 \rightarrow HO_2^- + H^+ \tag{5}$$

$$H_2O_2 + HO_2^- \rightarrow H_2O + O_2 + {}^{\bullet}OH \quad pK_a = 11.6 \tag{6}$$

$$H_2O_2 \; H_2O + \frac{1}{2}O_2 \tag{7}$$

Interaction between pH and temperature also found to have significant interaction that minimizes the percentage of TOC removal. At higher temperature, $H_2O_2$ tend to decompose producing water and oxygen (Eq. 7). Hence, it led to lower production of hydroxyl radical and percentage of TOC removal. Since, OTC is a thermolabile compound, faster disappearance of OTC molecule from the solution was observed at higher temperature (Doi and Stoskopf, 2000). However, the model suggested that temperature as single factor is not a significant factor that minimizes percentage of TOC removal. This insignificant may be due to the temperature interval were chosen in the experimental design that is 30, 40 and 50°C. It effects may be lesser in minimizing the percentage of TOC removal rather than the pH which were chosen at wider range, from 3-11. This temperature interval chosen was due to the reason that at higher temperature the solution evaporation would occur faster. The oxidant/pollutant mole ratio as single factor was also not significant factor that minimize percentage of TOC removal which was supported by the optimum ratio of 1.5 eq. OTC-degraded product has 1.19 of $BOD_5$/COD implying the biodegradability of the product. As mentioned earlier, antibiotics are recalcitrant molecules that prone to biodegradation. Therefore chemical degradation treatment, like AOPs, was chosen as the strategic treatment prior to biological process for final treatment. In this experiment, OTC treatment at the optimum condition suggested by the model has increased the value of $BOD_5$/COD significantly. The results then confirmed that this system is very suitable for an alternative process to degrade recalcitrant molecule, such as OTC.

## CONCLUSIONS

Optimization of OTC degradation inside a UV/$H_2O_2$ system was conducted. Based on the Pareto chart, quadratic effect of pH is the most significant factor that minimized percentage TOC removal. Statistical analyses utilizing the Box-Behnken experimental design suggested that the optimum percentage of TOC removal could be obtained at 30°C, pH 6.30 and 1.5 mole equivalent of oxidant/pollutant ratio. About 1.19 $BOD_5$/COD value was obtained for OTC-degraded final product which proved its biodegradability. Therefore, this system can be chosen as an alternative process for pre-treatment of wastewater contaminated with recalcitrant molecule such as OTC.

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