AN OVERVIEW OF WASTEWATER TREATMENT PROCESSES OPTIMIZATION USING RESPONSE SURFACE METHODOLOGY (RSM)

Mohammed J.K. Bashir[1,*], Hamidi Abdul Aziz[2], Shuokr Qarani Aziz[3], Salim Abu Amr[4]

1: Assistant Professor. Environ. Eng. Dep., Faculty of Eng. Green Tech. Universiti Tunku Abdul Rahman, Perak, MALAYSIA. jkbashir@utar.edu.my; modbashir@gmail.com

2: Professor, School of Civil Eng., Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, MALAYSIA. cehamidi@eng.usm.my

3: Assistant Professor, Dep. of Civil Eng., College of Eng., University of Salahaddin–Erbil, Erbil, IRAQ. shoker71@yahoo.com

4: PhD Candidate, School of Civil Eng., Universiti Sains Malaysia, 14300 Nibong Tebal, Penang, MALAYSIA. sabuamr@hotmail.com

ABSTRACT: Optimization plays a key role in environmental engineering parameters since the best system performance mainly is on optimum point or optimum range. The majority of wastewater treatment processes are multi-variable and optimization through the classical method is inflexible, unreliable and time-consuming. Thus, response surface methodology (RSM), as a very efficient design and widely used technique, can be adapted for parameters optimization of various wastewater treatment processes. RSM is a practical mathematical and statistical tool that can be employed for analyzing the effects of several independent factors on the treatment process in order to obtain the maximum benefit from the process. Recently, several water and wastewater treatment processes have been optimized for treatment different type of wastewaters via RSM including; textile dye wastewater, tannery wastewater, industrial paint wastewater, landfill leachate, olive oil wastewater, and palm oil mill effluent. The present study focuses on the usability and effectiveness of RSM for process parameters modeling and optimization in wastewater treatment studies. In this paper, some of the RSM studies published recently were reviewed in order to verify the usability of RSM and its limitations.

Keywords: Wastewater, Treatment Processes, Optimization, Design of experiments, Response Surface Methodology.

I. INTRODUCTION

Increasing the efficiency of the processes without increasing the cost is very essential to get better performance of the operations. The method used for this purpose is called optimization. Optimization refers to choosing the best element from some set of available alternatives (independent variables). In most of process there is appropriate value of variable to reach to maximum yield. The traditional method used for determining the optimum operation conditions is by monitoring the influence of each variable individually on the response. Where, just one variable is changed and the others remain at a constant level. The interactive effects between the variables are not considered in this method. As a result, this method does not show the whole effects of the variables on the response. Another negative aspect of this technique is the increase in the total number of trials that are required for conducting the research, resulting in an increase of time, cost and materials combustions [1-4]. Thus, response surface methodology (RSM) can be used to address this problem. According to Myers and Montgomery [5], RSM is a statistical technique was useful for the optimization of chemical reactions and/or industrial processes and commonly used for experimental design. Nowadays, RSM has been widely and effectively applied for optimization of water and wastewater treatment processes in order to obtain the maximum benefit from the process. For example, several water and wastewater treatment processes have been optimized for treatment different type of wastewaters via RSM including; textile dye wastewater, tannery wastewater, industrial paint wastewater, landfill leachate, olive oil wastewater, and palm oil mill effluent [6-10]. Currently, RSM has been widely valid for different purposes in wastewater treatment processes. Although, RSM has some limitations, several studies were carried out without considering these limitations. The present study focuses on the use of RSM for process parameters optimization in wastewater treatment studies. In this paper, some of the RSM studies published recently were reviewed in order to verify the usability of RSM and its limitations.

II. DESIGN EXPERT (DoE)

Design Expert is a program for design of experiments, statistical analysis, modeling and optimization. It offers a range of programs including full factorial and fractional factorial designs, response surface method, mixing and D-optimal designs. The Design-Expert Software was used to develop the experimental plan for RSM [11]. The same software was also used to analyze the data collected. A regression is performed on the data collected where the observed variable (response) is approximated based on a functional relationship between the estimated input variables. In general, experimental design can be defined as a specific set of experiments defined by a matrix composed by the different level combinations of the variables studied. Factors or independent variables are experimental variables that can be changed independently of each other. Levels of a variable are different values of a variable at which the experiments must be carried out. Responses or dependent variables are the measured values of the results from experiments. Residual is the difference between the calculated and experimental result for a determinate set of conditions. A good mathematical model fitted to experimental data should present low residuals values.

III. RESPONSE SURFACE METHODOLOGY (RSM)

RSM is a collection of mathematical and statistical techniques which are useful for modeling and analysis of problems in which a response of interest is influenced by numerous variables and the aim is to optimize this response [12-13] According to Bas and Boyaci. [2] and Bezerra et al. [3], RSM is an efficient statistical method for the modelling and optimization of various variables to predict the best performance conditions with a minimum number of experiments. RSM was introduced in 1951 by Box and Wilson [14] and they suggested using a second-degree polynomial model. Recently RSM have been employed for optimization of process parameters especially in food science and technology, material engineering, chemistry and chemical engineering.

For application of RSM as an optimization technique, some stages should be followed such as: (1) selection of the most important independent variables and their level on the system through screening studies; (2) the choice of the experimental design and carrying out the experiments according to the selected experimental matrix; (3) the mathematic–statistical treatment of the obtained experimental data through the fit of a polynomial function; (4) the evaluation of the model's fitness; (5) the verification of the necessity and possibility of performing a displacement in direction to the optimal region; and (6) obtaining the optimum values for each variable [15].

The least square technique is used to fit a model equation containing the input variables by minimizing the residual error measured by the sum of square deviations between the actual and the estimated responses. This involves the calculation of estimates for the regression coefficients. However, the calculated coefficients of the model equation need to be tested for statistical significance. In this respect, three tests are Performed-test for significance of the regression model, test for significance on individual model coefficients and test for lack-of-fit [16]. Additionally, checks need to be made in order to determine whether the model actually describes the experimental data [16]. The checks performed here include determining the various coefficients of determination (R^2). In addition to this, the adequacy of the model is also investigated by the examination of residuals [12]. The residuals are the differences between the observed and the predicted responses and these are examined using the normal probability plots of the residuals and the plots of the residuals versus the predicted response. If the model is adequate, the points on the normal probability plots of the residuals should form a straight line. On the other hand, the plots of the residuals versus the predicted response should be structure less, that is, they should contain no obvious patterns.

A. Theory of RSM

Besides analyzing the independent variables effects, this experimental methodology also generates a mathematical model. The graphical viewpoint of the mathematical model has led to the term RSM. The relationship between the responses and the inputs is given in Eq. (1):

$$Y = f(x_1, x_2, x_3 .... x_n) \pm \varepsilon \qquad (1)$$

where Y is the response, *f* is the unknown function of response, x_1, x_2, x_3,… x_n are the input variables which can affect the response, n is the number of the independent variables and ε is the statistical error that represents other sources of variability not accounted for by *f*. After selection of the design, the model equation is defined and coefficients of the model equation are predicted.

As the results of Sequential model sum of squares suggestion of software and software suggestion, the quadratic model was selected. In the case that total number of experiments is n; the response surface can be expressed as follows using matrix notation of the model.

$$Y = X\beta \pm \varepsilon$$

Where

$$\underbrace{\begin{Bmatrix} y_1 \\ y_2 \\ \vdots \\ y_n \end{Bmatrix}}_{\mathbf{Y}} = \underbrace{\begin{Bmatrix} 1\, x_{11} & x_{12} & \cdots & x_{1k} \\ 1\, x_{21} & x_{22} & \cdots & x_{2k} \\ \vdots\ \ \vdots & \vdots & \ddots & \vdots \\ 1\, x_{n1} & x_{n2} & \cdots & x_{nk} \end{Bmatrix}}_{\boldsymbol{X}} \underbrace{\begin{Bmatrix} \beta_1 \\ \beta_2 \\ \vdots \\ \beta_n \end{Bmatrix}}_{\boldsymbol{\beta}} + \underbrace{\begin{Bmatrix} \varepsilon_1 \\ \varepsilon_2 \\ \vdots \\ \varepsilon_n \end{Bmatrix}}_{\boldsymbol{\varepsilon}} \qquad (2)$$

where ε is random error.

The following Equation is for calculating the total number of experiments

$$N = n^3 + 2n + n_c \qquad (3)$$

where N is the total number of experiments and n is the number of factors.

The selected independent variables were coded according to equation (4):

$$X_i = \frac{X_i - X_0}{\Delta X} \qquad i = 1, 2, ..., \quad k \qquad (4)$$

where X_i refers to coded value of the i th independent variable, X_0 is the value of X_i at the center point and ΔX is the step change value [12] The response function (Y) is measured as the yield in processes. The model used in RSM is commonly a full quadratic equation or the diminished form of this equation. The second order model can be written as follows:

$$Y = \beta_0 + \sum_{i=1}^{n} \beta_i x_i + \sum_{i=1}^{n} \beta_{ii} x_i^2 + \sum_{i \neq j=1}^{n} \beta_{ij} x_i x_{ij} + \varepsilon \qquad (5)$$

where β_0 is the value of the fixed response at the center point of the design; β_i, β_{ii}, and β_{ij} are the linear, quadratic and interaction effect regression terms, respectively; x_i denotes the level of the independent variable; n is the number of independent variables; and ε is random error. Normally, a second order polynomial equation was derived as:

$$Y = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_{12} x_1 x_2 + \beta_{13} x_1 x_3 + \beta_{23} x_2 x_3 + \beta_{11} x_1^2 + \beta_{22} x_2^2 + \beta_{33} x_3^2 + \varepsilon \qquad (6)$$

B. Statistical analysis by RSM

A wide range of statistical and diagnostics studies are obtainable from RSM. The Analysis of variance (ANOVA) including sequential F-test, lack-of-fit test and other adequacy measures is offered by RSM. Residual analysis and diagnostics case statistics is checked to ensure adequacy of models. The results of ANOVA are reported to check sufficiency of models as well. Diagnostics plot are showing the relaibilty of model , they are include: predicted versus actual plot, normal plot of residual , residuals versus predicted, residual versus the run, residual versus factors, Box-Cox Plot for Power Transforms. Cook's Distance,Outlier t, Residual, Leverage,Residual ae well as peridicted and actual values are reported by RSM [11].

In the RSM the quality of the fit of the polynomial model is expressed by the coefficient of determination R^2 in Equation (7), adjusted coefficient (R^2_{Adj}) in Equation (8) and predicted coefficient (R^2_{Pred}) in Equation (9).

$$R^2 = 1 - \frac{SS_{Residual}}{SS_{Model} + SS_{Residual}} \qquad (7)$$

$$R^2_{Adj} = 1 - \frac{SS_{Residual} / DF_{Residual}}{(SS_{Model} + SS_{Residual}) / (DF_{Model} + DF_{Residual})} \qquad (8)$$

$$R^2_{Pred} = 1 - \frac{PRESS}{(SS_{Model} + SS_{Residual})} \tag{9}$$

where the terms SS and DF are sum of squares and degrees of freedom, respectively, and PRESS denotes Predicted Residual Sum of Squares. The residual sum of squares (SS_E) is expresed in Equationss (10-12).

$$SS_E = SS_{PE} + SS_{LOF} \tag{10}$$

$$SS_{PE} = \sum_{i=1}^{m}\sum_{j=1}^{n}(y_{ij} - \bar{y}_i)^2 \tag{11}$$

$$SS_{LOF} = \sum_{i=1}^{m} n_i(\bar{y}_i - \hat{y}_i)^2 \tag{12}$$

where SS_{PE} is the sum of squares due to pure error and SS_{LOF} is the sum of sequences due to lack of fit [12]. Equation (13) is stamited Predicted Residual Sum of Squares (PRESS).

$$PRESS = \sum_{i=1}^{n} e^2_{(i)} = \sum_{i=1}^{n}[y_i - \hat{y}_{(i)}]^2 \tag{13}$$

Adequate Precision (AP) is determined the signal to noise ratio according to equations (14) and (15).

$$Adequate \quad precision = \frac{\max(\hat{Y}) - \min(\hat{Y})}{\sqrt{\bar{V}(\hat{Y})}} \tag{14}$$

$$\bar{V}(\hat{Y}) = \frac{1}{n}\sum_{1=1}^{n} V(\hat{Y}) = \frac{p\sigma^2}{n} \tag{15}$$

Where, p is the number of model parameters, σ^2 the residual mean square, and n is the number of experiments).

C. Optimization

In RSM Factors are optimized through a desirability function (D) for multiple responses according to Equation (16).

$$D = \left[\prod_{i=1}^{N} d_i^{r_i} \right]^{1/\sum r_i} \quad (16)$$

Where N is the number of responses, r_i and d_i are the importance of particular response and partial desirability function respectively.

IV. RSM APPLICATION IN OPTIMIZATION OF WASTEWATER TREATMENT PROCESSES

In this section, several published RSM studies in the last few years were reviewed focusing on the usability of RSM for optimization of various types of wastewater treatment processes. Table 1 summarizes different applications of RSM in optimizing various types of wastewater treatment techniques.

Table 1: Various application of RSM in wastewater treatment process optimization

Wastewater type	Treatment technique	Independent variables	Responses	Ref.
Textile wastewater	Electrochemical Oxidation	pollution load, applied potential, electrolyte concentration, temperature, and reaction time	COD, color turbidity removals	[6]
Textile dyes	Adsorption	Initial dye concentration, initial solution pH and temperature	the amount of dye adsorbed at equilibrium	[17]
Landfill leachate	Coagulation–Flocculation	coagulant dosage and pH	COD, turbidity, color, and TSS removals	[9]
Landfill leachate	Fenton Oxidation	pH, Reaction time, Initial concentrations of H_2O_2, and Ferrous ion concentration	COD, color and iron removals	[18]
Landfill Leachate	Electrochemical oxidation	electrolyte concentrations, current density and reaction time	COD and color removals	[19]
Landfill leachate	Ion Exchange (cation resin)	Cation resin dosage, contact time, shaking speed	Ammonia removal	[20]

Landfill leachate	Ion Exchange (anion resin)	anion resin dosage, contact time, shaking speed, pH	COD, color, turbidity, and SS removals	[21]
Pulp mill wastewater	Coagulation-Flocculation	coagulant dosage, flocculant dosage and pH	turbidity removal, lignin removal and clean water recovery	[22]
Slaughterhouse Wastewater	Electrochemical oxidation	current density, reaction time and influent COD	COD, BOD, and color, removal efficiencies and effluent pH	[23]
Simulated industrial wastewater	Photo-Fenton process	initial pH values, concentration of iron catalyst and the concentration of H2O2 type of UV irradiation	Mineralization rate	[24]
Dairy wastewater	Electrochemical oxidation	current density, dosage of sodium chloride (NaCl), electrolysis time and pH,	COD removal	[25]
Petroleum refinery effluent	Upflow anaerobic sludge blanket (UASB) bioreactor	hydraulic retention time (HRT) influent COD, up flow velocity (Vup)	COD removal, and rate of biogas production	[26]
Oily wastewater emulsion	Electro-coagulation	Current density, pH, and Electrocoagulation time	turbidity and COD removals	[27]
Drinking water	Coagulation-flocculation	coagulant dose and pH	removal efficiency of turbidity and dissolved organic carbon (DOC)	[28]
Olive oil mill wastewater	Fenton’s peroxidation	he ratio of hydrogen peroxide-to-Fe(II) . Fe(II) concentration , and H_2O_2 concentration	COD, total phenolics (TP), color and aromatocity removal	[29]

According to the above mentioned literatures, RSM has several advantages compared to the traditional method that used for experimental optimization in which one variable at a time technique is implemented. The advantages can be summarized as below [2];

i. RSM gives a large amount of knowledge from a small number of experimental runs. However, traditional methods are time consuming and a large number of experimental runs are required to describe the behavior of a process.
ii. The interaction effect of the independent parameters on the response can be observed and investigated via RSM. The model equation easily clarifies these effects for binary combination of the independent parameters.
iii. Moreover, the empirical model that related the response to the independent variables is utilized to obtain information about the process.

With respect to these, it can be concluded that RSM is a useful and helpful tool for the optimization of wastewater treatment processes. On the other hand, the major drawback of RSM is to fit the data to a second order polynomial. We cannot say that all systems containing curvature are well accommodated by the second order polynomial.

In most of the reported studies that use RSM for optimization of wastewater treatment process, there is not enough or adequate preliminary work in regards to the range of independent variables. Thus, in most of the optimization studies, there is no optimum point due to unsuitable range of independent variables. The max or min value of the response without stationary point has been given as the optimum point. It is not relay correct to say these kinds of studies as optimization. If RSM studies were carried out without estimation of stationary point, the aim of the study and selection of the range of independent parameters should be explained clearly. The verification of the predicted equation should be made. The physical mean of the estimated results should be discussed and suitability of the estimated response to the expected result should be evaluated [2].

V. CONCLUSION

Response surface methodology (RSM) is a widely used technique which can be adapted for parameters optimization. Statistical and diagnostics analysis indicated that RSM is a reliable tool to optimize of process parameters. RSM is successfully applied for optimization of several wastewater treatment processes. In the current study, s RSM method and some of its applications that published recently were reviewed in order to verify the usability of RSM and its limitations. It was observed that RSM has several advantages in wastewater treatment process optimization. However, In most of the reviewed articles that implement RSM for optimization of wastewater treatment processes, there is not sufficient or adequate preliminary work in regards to the range of process variables. This could lead to inaccurate outcome.

VI. REFERENCE

[1] Thomas, M.; Beaucbamp, Y.; Youssef, Y.A.; Masounave, I.: An experimental design for surface roughness and built-up edge formation in lathe dry turning. *Int. J. Qual. Sci.* 2 (1997), 167–180.

[2] Bas, D.; Boyaci, I.H: Modeling and optimization I: Usability of response surface methodology, *J. Food Eng.*78 (2007) 836–845.

[3] Bezerra, M.A.; Santelli, R.E.; Oliveira, E. P.; Villar, L.S. Escaleira, L.A.: Response surface methodology (RSM) as a tool for optimization in analytical chemistry, *Talanta* 76 (2008) 965–977.

[4] Arslan-Alaton, I.; Tureli, G.; Olmez-Hanci, T: Treatment of azo dye production wastewaters using Photo-Fenton-like advanced oxidation processes: Optimization by response surface methodology, *J. Photochem. Photobiol. A:* 202 (2009) 142-153.

[5] Myers, R.H.; Montgomery, D.C: Response Surface Methodology: Process and Product Optimization using Designed Experiments, 2nd ed., John Wiley & Sons, USA. 2002.

[6] Korbahti B.K., Tanyolac, A: Electrochemical treatment of simulated textile wastewater with industrial components and Levafix Blue CA reactive dye: Optimization throughresponse surface methodology, *Journal of Hazardous Materials* 151(2008) 422–431.

[7] Korbahti, B.K.; Aktas, N.; Tanyolac, A: Optimization of electrochemical treatment of industrial paint wastewater with response surface methodology. *Journal of Hazardous Materials,* 148 (2007) 83-90.

[8] Bashir, M.J.K.; Isa, M.H.; Awang, Z.B.; Aziz, H.A.; Kutty, S.R.M.; Mohajeri, S.; Farooqi, I.H: (2009) Landfill leachate treatment by electrochemical oxidation. *Waste Management*, 29 (2009) 2534-2541.

[9] Ghafari, S.; Aziz, H.A.; Isa, M. H; Zinatizadeh, A.A: Application of response surface methodology (RSM) to optimize coagulation-flocculation treatment of leachate using poly- aluminum chloride (PAC) and alum, *J. Hazard. Mater.* 163 (2009) 650-656.

[10] Zinatizadeh, A.A.L.; Mohamed, A.R.; Abdullah, A.Z.; Mashitah, M.D.; Isa, M.H.; Najafpour, G.: Process modeling and analysis of palm oil mill effluent treatment in an up-flow anaerobic sludge fixed film bioreactor using response surface methodology (RSM), *Water Res.* 40 (2006) 3193-3208.

[11] Design-Expert Software, Version 7, *User's Guide, Technical Manual. Stat-Ease Inc*., Minneapolis, MN, 2006.

[12] Montgomery DC: *Design and Analysis of Experiments,* 7th ed., John Wiley and Sons, Inc., New York, 2008.

[13]Mason, R.L.; Gunst, R.F.; Hess,J.L: *Statistical Design and Analysis of Experiments, Eighth Applications to Engineering and Science*, 2nd ed., Wiley, New York, 2003.

[14] Box, G.E.P.; Wilson, K.B.: On the experimental attainment of optimum conditions, *Journal of the Royal Statistical Society Series*, 13(1951) 1–45.

[15] Steppan, D.D.; Werner, I.; Yeater, R.P: Essential regressionand experimental design for chemists and engineers.1998, *http://llgeocities.corn/SiliconValley/Network/l032/CGPagel.html.*

[16] Aggarwal, A; Singh, H; Kumar, P; Singh, M: Optimizing power consumption for CNC turned parts using response surface methodology and Taguchi's technique—A

comparative analysis, *Journal of Materials Processing Technology* 2 0 0 (2 0 0 8) 373–384.
[17] Santos,S.C.R.; Boaventura R.A.R.: Adsorption modelling of textile dyes by sepiolite, *Applied Clay Science* 42 (2008) 137–145.
[18] Mohajeri, S.; Aziz, H.A.; Zahed, M.A.; Mohajeri, L.; Bashir, M.J.K., Aziz, S.Q.; Adlan, M.N.; Isa, M.H: Multiple responses analysis and modeling of Fenton process for treatment of high strength landfill leachate, *Water Science & Technology*, 64 (2011) 1652-1660.
[19] Mohajeri, S.; Aziz, H.A.; Isa, M.H.; Zahed, M.A.; Bashir, M.J.K.; Adlan, M.N: Application of the central composite design for condition optimization for semi-aerobic landfill leachate treatment using electrochemical oxidation. *Water Science & Technology*, 61 (2010)1257-1266.
[20] Bashir, M.J.K.; Aziz, H.A.; Yusoff, M.S.; Adlan, M.N: Application of response surface methodology (RSM) for optimization of ammoniacal nitrogen removal from semi-aerobic landfill leachate using ion exchange resin, *Desalination* 254 (2010)154-161.
[21] Bashir, M.J.K.; Aziz, H.A.; Yusoff, M.S.; Aziz, S.Q.; Mohajeri, S:Stabilized Sanitary Landfill Leachate Treatment Using Anionic Resin: Treatment optimization by Response Surface Methodology. *Journal of Hazardous Materials*, 182 (2010)115–122.
[22] Wang, J.-P.; Chen,Y.-Z.; Wang, Y.; Yuan, S.-J., Yu, H.Q: Optimization of the coagulation-flocculation process for pulp mill wastewater treatment using a combination of uniform design and response surface methodology, *Water Research*, 45 (2011) 5633–5640.
[23] Awang, Z.B.; Bashir, M.J.K., Kutty, S.R.M.; Isa, M.H. Post-Treatment of Slaughterhouse Wastewater using Electrochemical Oxidation. *Research Journal of Chemistry and Environment*, 15 (2011) 229-237.
[24] Dopar, M.; Kusic. H.; Koprivanac, N.; Treatment of simulated industrial wastewater by photo-Fenton process.Part I: The optimization of process parameters using design of experiments (DOE). *Chemical Engineering Journal*, 173 (2011) 267– 279.
[25] Kushwaha, J.P.; Srivastava,V.C.; Mall. I.D.: Organics removal from dairy wastewater by electrochemical treatment andresidue disposal, *Separation and Purification Technology* 76 (2010) 198–205.
[26]Rastegar, S.O.; Mousavi, S.M.; Shojaosadati, S.A.; Sheibani, S: Optimization of petroleum refinery effluent treatment in a UASB reactor using response surface methodology. *Journal of Hazardous Materials* 197 (2011) 26– 32
[27] Tir, M. Mostefa, N.M. Optimization of oil removal from oily wastewater by electrocoagulation using response surface method, *Journal of Hazardous Materials* 158 (2008) 107–115.
[28] Trinh, T.K.; Kang, L.S: Response surface methodological approach to optimize the coagulation–flocculation process in drinking water Treatment. *Chemical Engineering Research and Design* 8 9 (2 0 1 1) 1126–1135
[29] Ahmadi, M.; Vahabzadeh, F.; Bonakdarpour, B.; Mofarrah, E.; Mehranian, M: Application of the central composite design and response surface methodology to the advanced treatment of olive oil processing wastewater using Fenton's peroxidation, *Journal of Hazardous Materials* 123 (2005) 187–195.