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RESEARCH ARTICLE /**ARAŞTIRMA MAKALESİ**

DETERMINATION OF OPTIMUM PURIFICATION CONDITIONS OF LABORATORY SCALE PACKED DISTILLATION COLUMN BY USING RESPONSE SURFACE METHOD

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ABSTRACT

The objectives of this study were to evaluate statistically the effects of input variables of the laboratory scale packed distillation on the mole fraction of the top product. Binary methanol-water mixture was distilled in the column. A statistical experimental design based on 2^3 full factorial central composite design method of The Response Surface Methodology (RSM) was planned. Reflux ratio (X_1), reboiler heat duty (X_2) and feed mole fraction (X_3) were selected as input variables. The parameters were designed at five levels (-2, -1, 0, 1 and 2). To determine the significance of the effect of the parameters, the analysis of variance (ANOVA) was investigated with 95% confidence limits. It was shown that reflux ratio and reboiler heat duty were the variables of highest significance for the top product mole fraction of the laboratory packed distillation column. The optimum operating conditions of the system were found as follows: reflux ratio, 4; reboiler heat duty, 1kW and feed mole fraction, 0.14. Under these conditions, the experimental mole fraction of the top product was close to the predicted value calculated from the polynomial response surface model equation.

Keywords: Central composite design, ANOVA, Optimization, Packed distillation column

LABORATUVAR ÖLÇEKLİ BİR DOLGULU DAMITMA KOLONUNUN OPTİMUM SAFLAŞTIRMA KOŞULLARININ CEVAP YÜZEY YÖNTEMİYLE BELİRLENMESİ

ÖZ

Bu çalışmanın amacı, laboratuar ölçekli bir dolgulu damıtma kolonunun giriş değişkenlerinin üst ürün mol kesrine olan etkisini istatistiksel olarak belirlemektir. Kolonda metanol-su ikili karışımı damıtılmıştır. Cevap Yüzey Metodu (RSM)'nin 2^3 tam faktöriyel merkezi karışık tasarım yöntemine dayalı bir istatistiksel deney tasarımı planlanmıştır. Giriş değişkenleri olarak geri akma oranı (X_1), kazan ısı yükü (X_2) ve besleme mol kesri (X_3) seçilmiştir. Parametreler beş seviyede (-2, -1, 0, 1 ve 2) tasarlanmıştır. Parametrelerin etkilerinin önemini belirlemek için %95 güven aralığında varyans analizi (ANOVA) uygulanmıştır. Elde edilen sonuçlardan laboratuar ölçekli dolgulu damıtma kolonunun üst ürün mol kesri için en önemli giriş parametrelerin geri akma oranı ve kazan ısı yükü olduğu gösterilmiştir. Sistemin optimum koşulları geri akma oranı = 4; kazan ısı yükü = 1 kW ve besleme mol kesri = 0.14 olarak bulunmuştur. Bu koşullarda deneysel olarak elde edilen üst ürün mol kesri değeri cevap yüzey model eşitliğinden hesaplanan tahmini değere oldukça yakın olduğu görülmüştür.

Anahtar Kelimeler: Merkezi karmaşık tasarım, ANOVA, Optimizasyon, Dolgulu damıtma kolonu

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1. INTRODUCTION

Distillation is one of the most frequently used unit operations in the chemistry industry. Distillation columns are of the two types-tray columns, in which vapor and liquid are contacted in discrete stages, and packed columns, in which contact occurs continuously through the column. There are several instances when the use of packed columns is preferred to that of plate columns. A large amount of work dealing with the modeling, simulation, and control of tray columns has been published. In contrast, relatively little work with packed columns has been reported. To design packed columns with a good degree of confidence, to predict their behaviour under various operating conditions accurately , and to optimise the system in an efficient manner mathematical models of good accuracy are necessary (Patwardhan et al., 1993 and Karacan et al., 1998).

Packed columns contain either random or structured packings. The past few years have seen an increase in the use of high efficiency structured packings, especially in the retrofitting of existing plants. Structured packings offers increased interfacial area, while reducing form drag. Structured packings are available in a variety of surface textures sheet metal with smooth, fluted, lanced, or perforated surfaces. Packings may also be made from gauze material. Knowledge of the response of packings with different surface treatment to different operating conditions is very important. A column containing efficient packing may exhibite behaviour that requires special consideration when designing a control system (Wagner et al., 1997, Iluta et al., 2004).

The optimal synthesis of distillation continues to be a major problem in the design of chemical processes due to the high investment and operating costs involved in these systems. The recent trends in this area have been to address models of increasing complexity through the use of mathematical programming. The high degree of nonlinearity and the difficulty of solving the corresponding optimisation models, however, have prevented methods with rigorous models from becoming tools that can be readily used by industry. Conventionally, the so called direct setting strategy has been used in the process industry: the values of control variables corresponding to the specified steady state are set to the columns for startup and one just waits for the column to reach to the steady state. A long time period is usually needed for startup by using this strategy. Total reflux (Ruiz et al., 1988) and zero reflux (Kruse et al., 1996) strategies together with a large reboiler heat duty have been proposed. Since a distillation column is influenced by many factors such as column structure, the type of trays and packings, component properties in the mixture as well as the top and bottom product specifications, reboiler heat duty and reflux ratio, these empirical strategies are suitable only for some specific cases. Therefore, systematic approaches concerning these influential factors are required to solve general optimisation problems for distillation columns. This calls for systematic methodologies of modeling and optimisation.

The conventional method of optimisation involves varying one parameter at a time and keeping the others constant. This often does not bring about the effect of interaction of various parameters as compared to factorial design (Cochran and Cox, 1992). It has some limitations for complete optimisation and cannot provide information about the interaction of different effective factors. Statistical experimental design techniques are very useful tools for this purpose, as they can provide statistical models that assist in understanding the interaction of different variables and predict the maximized top product mole fraction. The use of statistically designed experiments can allow the rapid and economical determination of the optimal conditions with fewer experiments and minimal resources. Response surface methodology (RSM) is a useful model for studying the effect of several factors influencing the response by varying them simultaneously and carrying out a limited number of experiments. RSM was successfully applied for especially many biochemistry and food processes, such as Linder et al.(2005), Soo et al. (2004); Faveri et al. (2004); Chu et al. (2003); Salah et al. (2005) ; Sonsuzer et al. (2004); and Ismail and Lai (2004). But no study focusing on the optimization of the packed distillation column by using RSM has been encountered in the literature.

In this work, the influences of reflux ratio, reboiler heat duty and feed mole fraction on distillate composition were investigated using a full-factorial experimental design in order to obtain a maximum yield. A quadratic model was evaluated by the F-test analysis of variance(ANOVA). The completely optimised parameter was determined by using numerical optimising technique.

2. EXPERIMENTAL PROCEDURES

Experiments were carried out in a laboratory scale packed column to distil the binary methanol-water mixture. Physical properties of the laboratory scale packed column are demonstrated in Table 1.

Table 1. Physical properties of packed distillation column

Packing height (mm)	1000
Inside diameter of packed column (mm)	85
Packing type	Raschng
Packing diameters (mm)	2 0 / 15
Feed tank volume (L)	5
Reboiler volume (L)	2
Total pressure (mmHg)	6 9 0

All experimental equipment’s were shown in Figure 1. In the experiments, overhead product composition and temperature changes with time were observed at steady-state and dynamic conditions.

The column used 1000 mm packing height. Packing type was Raschng ring with 20-15 mm diameter. The reboiler was made from 2 L glass container. To feed the relevant liquid into the column a peristaltic pump was utilized. The reflux ratio was adjusted by a computer . The system temperatures were measured with three thermocouples, placed in the rebolier at the top of the column and at the feed point. Each thermocouples was connected to a controller module and was transferred to the computer with a Digital/ Digital (D/D) converter. Temperature data measured at each second was recorded. Temperature profiles observed on the computer were recorded and samples were taken regularly from the top and bottom of the column. Refractive indices of the samples were measured. When the refractive index and temperatures were constant, the system is said to have reached the steady-state condition. After steady-state condition was maintained, step disturbances were given to the input variables.

Using the response surface methodology, it is possible to systematically study many processes. This is a single-factor-at a-time method which studies the phenomenon by varying one factor while keeping all other conditions constant. However, the effect of each factor is not necessarily additive. It is, therefore, necessary to take into account the influence of each factor and the interaction between these factors which may be synergistic or antagonistic (Box et al., 1978).

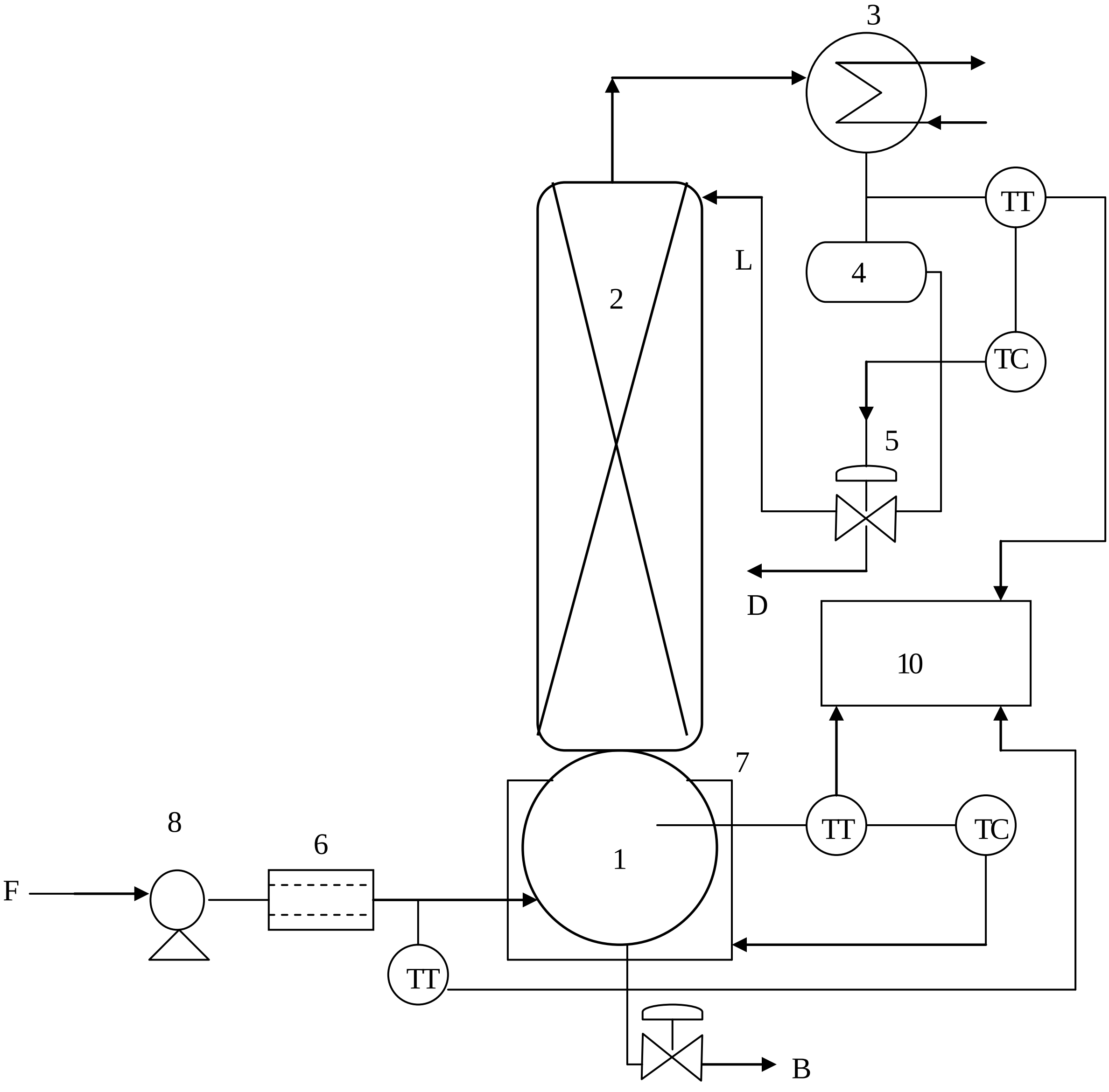

Figure 1. Experimental Equipment
1: Reboiler, 2: Packed Column, 3: Condenser, 4: Accumulator, 5: Reflux Valve, 6: Heat Exchanger, 7: Jacket Exchanger, 8:Peristaltic Pump, 9: Bottom Product Valve, 10: Computer

Prior experimental findings, the most influential factors on the mole fraction of the top product were expected to be the reflux ratio (X_1), the reboiler heat duty (X_2) and the feed mole fraction (X_3). A 2^3 full-factorial central composite design with five coded levels and three variable leading to 20 sets of experiments was adopted in this study (Cochran and Cox, 1992). The variables and their levels selected for the study are represented in Table 2. For statistical calculation, the variables were coded according to Equation (1):

$$X_i = \frac{U_i - U_i^0}{\Delta U_i} \qquad \textbf{(1)}$$

where X_i is the independent variable coded value; U_i, independent variable real value; U_i^0, independent variable real value on the central point; and ΔU_i, step change value.

Table 2. Coded and actual values of variables of design of experiment.

Variables	Range and levels				
	-2	**-1**	**0**	**1**	**2**
X_1 (R)	2	3	4	5	6
X_2 (Q_R)	0.8	1.0	1.2	1.4	1.6
X_3 (X_F)	0.08	0.12	0.16	0.20	0.24

The mole fraction of the top product was taken as response. The relationship between response and the variables can be represented by a quadratic mathematical model:

$$Y = \beta_0 + \sum_{i=1}^{3}\beta_i X_i + \sum_{i=1}^{3}\beta_{ii} X_i^2 + \sum_{i=1}^{2}\sum_{j=i+1}^{3}\beta_{ij} X_i X_j \qquad \textbf{(2)}$$

where Y is the response calculated by the model; β_0 is a constant ; β_i, β_{ii}, β_{ij} are linear, squared, cross-product coefficient, respectively. The applicability of the model was checked with significance, regression coefficient (R^2) and the coefficient of variation (CV) values. The 'Design Export' Software (version 6.0.4) from Stat-Ease Inc. was used for regression and graphical analyses of data obtained.

3. RESULTS AND DISCUSSION

Experiments were planned to obtain a quadratic model consisting of 2^3 trials plus a star configuration ($\alpha=\pm2$) and six trials at the central point. The design of this experiment is given in Table 3, together with the experimental results.

'Design Export Software' was used to find out the quadratic mathematical model. By applying multiple regression analysis on the experimental data (Table 3), the following polynomial equation was generated.

$$\begin{aligned} Y = {} & 0.92 - 0.095X_1 - 0.091X_2 + 1.875*10^{-3}X_3 \\ & -0.044X_1^2 - 0.022X_2^2 + 4.432*10^{-3}X_3^2 \\ & -0.094X_1X_2 + 4.0*10^{-3}X_1X_3 + 0.012X_2X_3 \end{aligned}$$

where Y represents mole fraction of top product as a function of , X_1, reflux ratio; X_2, reboiler heat duty and X_3, feed mole fraction.

The statistical significance of the second order model equation was evaluated by the F-test analysis of variance (ANOVA) which revealed that this regression is statistically significant (Prob>F) at 95% of confidence level. The model presented a high regression coefficient (R^2=0.974) explaining 97.4% of the variability in the response (Table 4). The coefficient of variation(CV) is low, 3.8, indicating a good precision and reliability for the experiments carried out. Also, a low value of coefficient of regression (R^2<0.8) was observed, lack of fit test designed to determine whether the selected model is adequate to describe the observed data, or whether another model should be used. If the model has a significant lack of fit, as indicated by Prob>F value less than 0.05 at the 95% confidence level, this model should not be used to predict the response, Although lack of fit was significant for regression model, Our regression model can be used to predict the response because coefficient of determination, R^2 was greater than 0.8.

Table 5 lists the coefficients calculated by the model. The significance of each coefficient was determined by F-value and Prob F values which are given in β_i, β_{ii} and β_{ij} along with significance levels of the terms. From Prob F values of terms in the Table 5, it can be seen that the linear term of feed mole fraction has least significance on mole fraction of the top production (Prob F = 0.873>0.05), the linear and square terms of reflux ratio and reboiler heat duty were highly significant (Prob F<0.05), and square term of reflux ratio is also one of the most influential factors. Interaction of reflux ratio and reboiler heat duty also had a major effect on mole fraction of the methanol of the top product.

Response surface methodology was used to investigate the effect of the three factors on mole fraction of methanol of top product, and three - dimensional plot was drawn. Figures 2-4 were the response surface and contour curves for three variables in the methanol mole fraction of the top product of laboratory scale packed distillation column. Figure 2 represents that interaction between the reflux ratio and reboiler heat duty on mole fraction of top product. The estimated response surfaces confirm that the reflux ratio and reboiler hear duty have positive effects on the response. The points giving the maximum mole fraction were found

Table 3. Coded level combination for a three-variable five level CCD

Test run	X_1(R)	$X_2(Q_R)$	$X_3(X_F)$	$Y(X_D)$
1	-1	-1	-1	1.000
2	1	-1	-1	0.993
3	-1	+1	-1	0.942
4	+1	+1	-1	0.539
5	-1	-1	+1	0,967
6	+1	-1	+1	0.957
7	-1	+1	+1	0.937
8	+1	+1	+1	0.573
9	-2	0	0	0.927
10	2	0	0	0.563
11	0	-2	0	0.967
12	0	+2	0	0.700
13	0	0	-2	0.947
14	0	0	+2	0.957
15	0	0	0	0.920
16	0	0	0	0.918
17	0	0	0	0.923
18	0	0	0	0.920
19	0	0	0	0.919
20	0	0	0	0.928

Table 4. ANOVA for the model regression representing mole fraction of top product

Source	**SS**	**DF**	**MS**	**F-value**	**Prob>F**
Model	0.41	9	0.0046	44.44	$<0.0001^{*}$
Residual	0.010	10	1.026E-003	-	
Lack of fit	0.010	5	2.038E-003	151.32	$<0.0001^{*}$
Pure error	6.733E-005	5	1.347E-005	-	
Cor Total	0.42				

SS,sum of squares; DF,degree of freedom; MS,mean square;
* Significant at "Prob>F"less than 0.05.

Table 5. The least fit and parameter estimates

Model terms	**Coefficient estimate**	**Sum of squares**	**DF**	**Standard error**	**F value**	**Prob F**
Intercept	0.920		1	$13.0^{*}10^{-3}$	44.44	<0.0001
X_1	-0.094	0.14	1	$8.0^{*}10^{-3}$	139.13	<0.0001
X_2	-0.091	0.13	1	$8.0^{*}10^{-3}$	129.72	<0.0001
X_3	$-1.312^{*}10^{-3}$	$5.6^{*}10^{-5}$	1	$8.0^{*}10^{-3}$	0.027	0.8730^{*}
X_1^2	-0.044	0.049	1	$6.3^{*}10^{-3}$	47.51	<0.0001
X_2^2	-0.022	0.012	1	$6.38^{*}10^{-3}$	11.76	0.0065
X_3^2	$7.727^{*}10^{-3}$	$4.9^{*}10^{-4}$	1	$6.38^{*}10^{-3}$	1.46	0.2541^{*}
X_1X_2	-0.094	0.071	1	$13.0^{*}10^{-3}$	68.37	<0.0001
X_1X_3	$4.375^{*}10^{-3}$	$1.3^{*}10^{-3}$	1	$12.0^{*}10^{-3}$	0.15	0.7073^{*}
X_2X_3	0.012	$1.1^{*}10^{-3}$	1	$12.0^{*}10^{-3}$	1.15	0.3094^{*}

DF, degree of freedom; * Values greater than 0.1000 indicate the model terms are not significant.

to be R= 4 and Q_R= 1.0 kW. In these conditions, the model predicted the maximum mole fraction of top product of 0.999. Figure 3 shows the effect of reflux ratio and feed mole fraction on the response. A quadratic increased was shown at R=(3-4) and X_F= (0.12-0.14) and then there was however decreased in the values of reflux ratio about R=(5-6). The maximum point of the mole fraction of methanol was found to be R=4 and X_F=0.13. The effect of reboiler heat duty and feed mole fraction on top product mole fraction at fixed point. Final response surface, Figure 4 depicts the reboiler heat duty, Q_R versus feed mole fraction, X_F profiles for the mole fraction of top product. An increase in mole fraction of top product with decrease in reboiler heat duty and increase in feed mole fraction was observed. The maximum methanol mole fraction of top product was obtained at optimum conditions of reboiler heat duty, 1.04 kW and feed mole fraction, 0.11. According to optimize mathematical model, the optimal parameters of three factors were: R= 4; Q_R= 1.0 kW and X_F= 0.14 and corresponding maximum methanol mole fraction was 0.999. It was clear that the optimal values obtained from response surface plots were almost consistent with that obtained from optimized mathematical equation. In order to verify the predicted results, an experiment was performed with optimized conditions, and indeed the experimental value of 0.99 for top product purity was obtained.

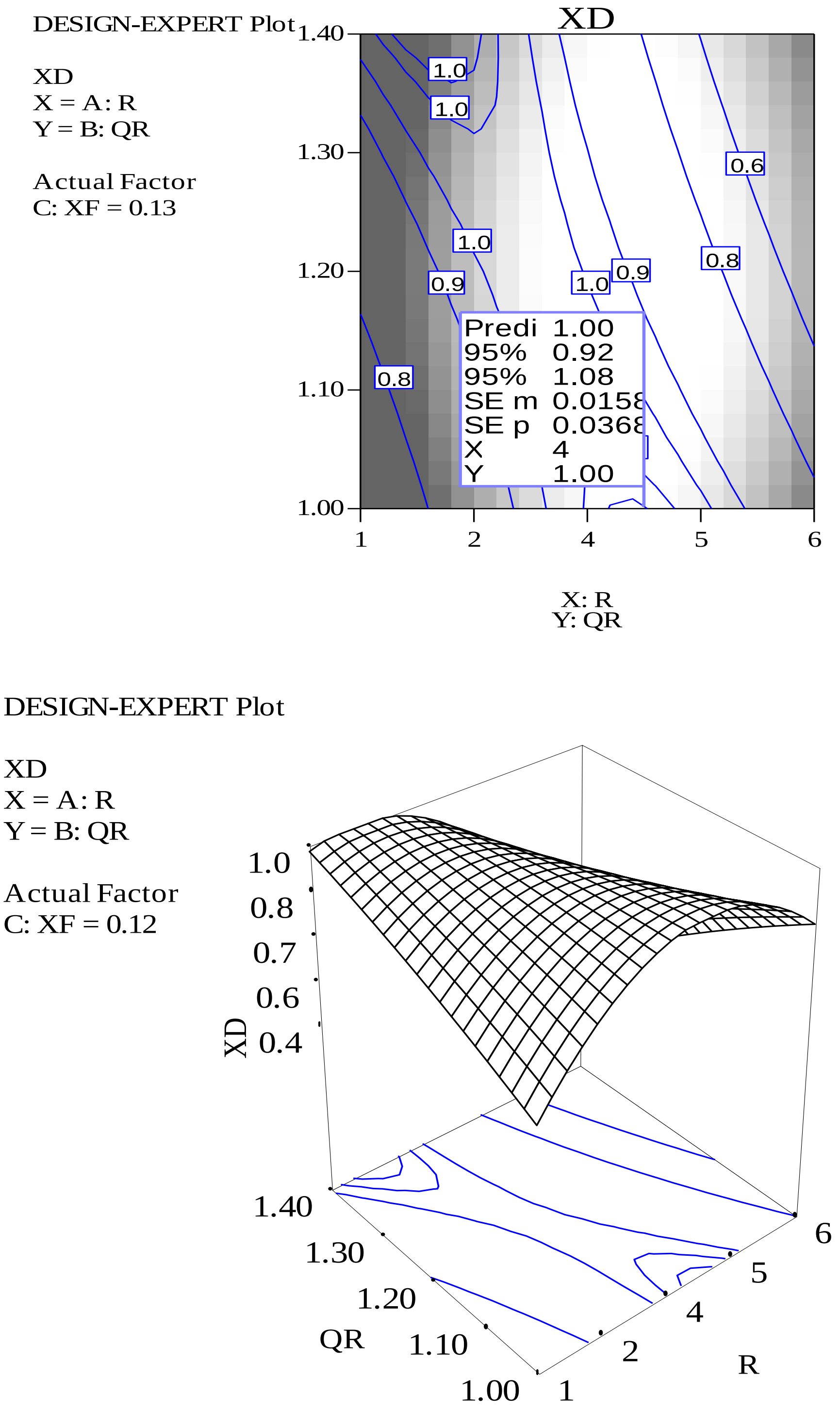

Figure 2. Response surface counter plots showing the effect of reflux ratio and reboiler heat duty , the third variable, X_F is fixed at “0” level

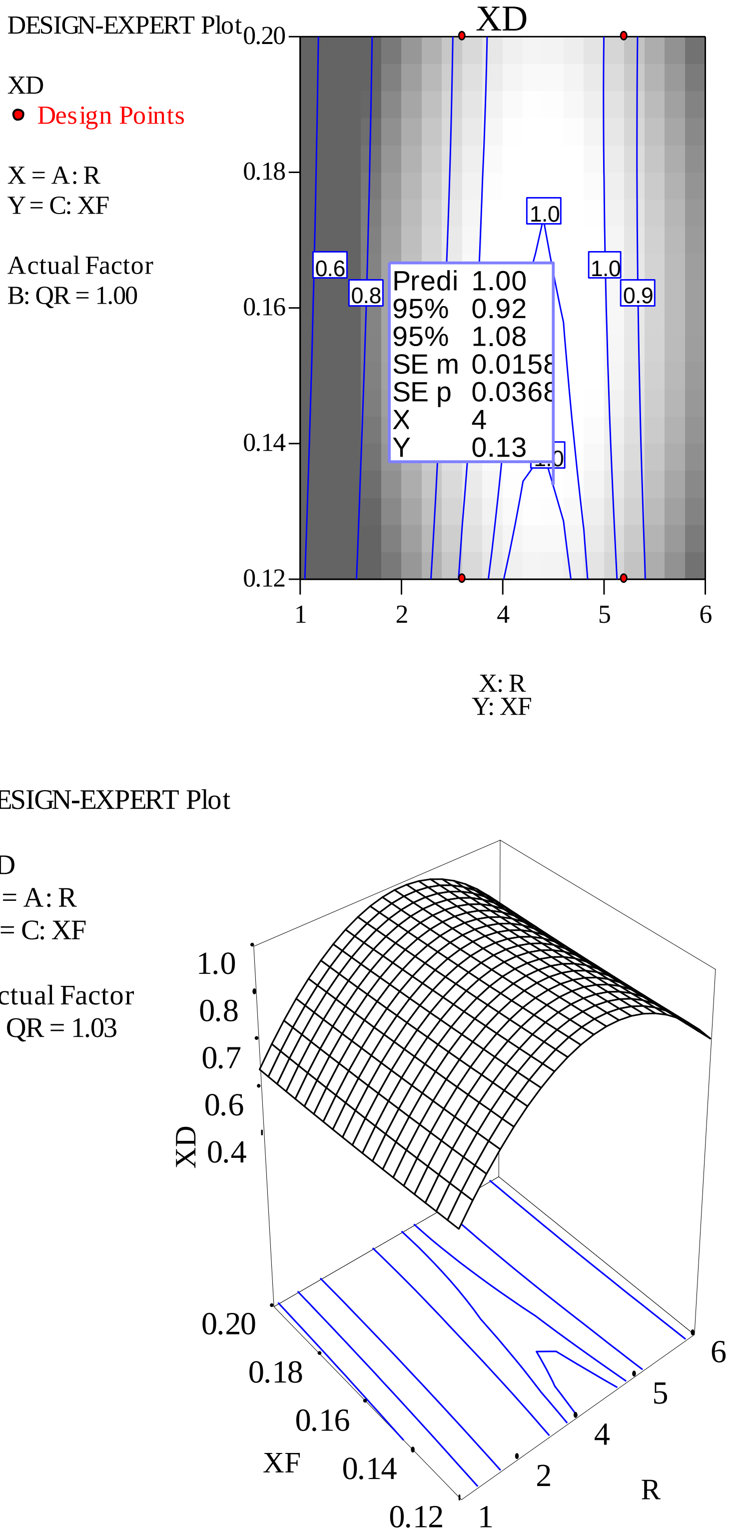

Figure 3. Response surface and counter plots showing the effect of reflux ratio and feed mole fraction , the second variable, Q_R is fixed at “0” level.

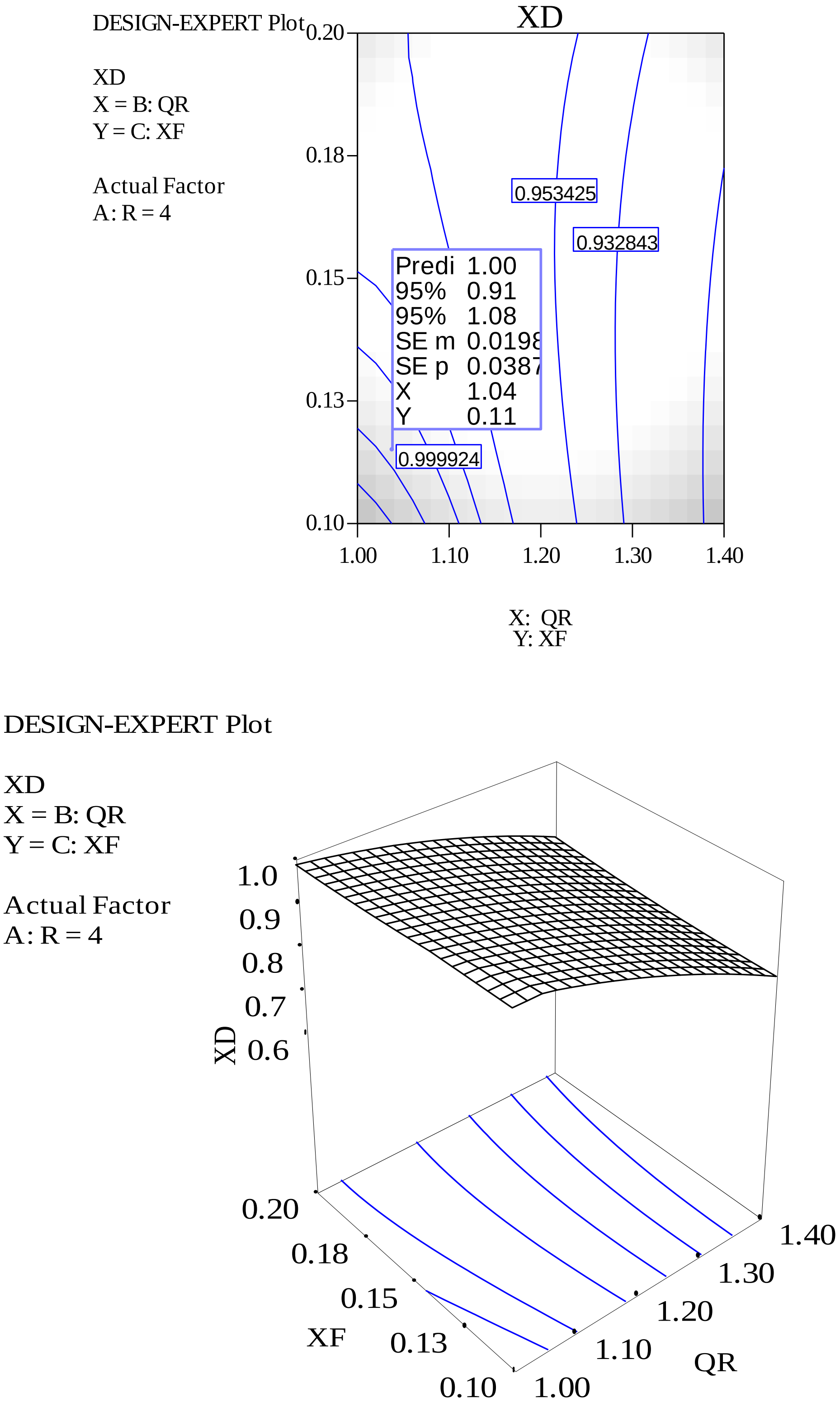

Figure 4. Response surface and counter plots showing the effect of reboiler heat duty and feed mole fraction , the first variable, R is fixed at “0” level

4. CONCLUSIONS

The response surface methodology was a useful tool to investigate the optimum conditions of a pilot packed distillation column. The R^2 value of the model showed a significantly good fit with the experimental data. To the best knowledge of the author, there are no reports of determination of the optimum conditions for the laboratory scale packed distillation column by using RSM approach in the literature. This work aims at indicating the practicability of statistical design to optimize methanol mole fraction of top product. The optimum conditions for a laboratory packed distillation column were satisfactorily determined by this method.

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