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Optimization of Saponification Reaction in a Continuous Stirred Tank Reactor (CSTR) Using Design of Experiments

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Abstract

The objective of this study was to maximize the conversion of saponification reaction in a continuous stirred tank reactor (CSTR). Full two-level factorial design and response surface methodology (RSM) were used to determine the optimum values of significant factors. The effect of five factors (sodium hydroxide and ethylacetate concentrations, feed ratio, agitation rate and temperature) was studied on the fractional conversion of sodium hydroxide (X_{NaOH}). As a result of screening experiments, two factors (sodium hydroxide and ethylacetate concentrations) and their combined effect were found to be significant operating parameters for the saponification reaction in continuous stirred tank reactor (CSTR). The optimum values of these significant factors were also determined using response surface methodology (RSM). For maximum conversion of sodium hydroxide (X_{NaOH}), i.e., 96.71%, the optimum values of sodium hydroxide and ethylacetate concentrations were found to be 0.01mol/L and 0.1 mol/L, respectively. Acorrelation was developed to show the relationship between different significant factors and response. The validity of the model was checked using analysis of variance (ANOVA). The experimental results are believed to be within reasonable accuracy and may be applicable for the improvement of such processes on industrial scale.

Key Words: Saponification; Design of Experiments; Optimization

1. Introduction

Statistical experimental designs are widely employed to improve the selection, development and optimization of a process. The design of experiments (DOE) reduces experimental costs because it requires only a small set of experiments. In design of experiments a set of experiments is performed, in which all the factors are varied systematically. Experimental results are analyzed and the factors that most influence and those that do not affect the target value, termed as the response, of the process are determined. Thus the process achieves the optimal conditions[1].

Ahmad *et al.,* [2] employed statistical experimental design for optimization of cartridge filtration of brine at oil fields. Rocak*et al.*, [3] used factorial design of experiments for optimization of ceramic suspension. Chen *et al.*, [4] presented different examples to show “clear effect concept” for the blocked fractional factorial designs. The authors also proposed a method to increase the number of clear effects. Zhang *et al.*, [5] introduced the concept of optimal blocking of two-level fractional factorial designs. Different blocked fractional factorial designs were suggested and their results were compared.

Various techniques based on design of experiments have been developed and are available in open literature. For example, Berkum *et al.*, [6] presented a method for employing two-level fractional factorial designs for two-step production processes. Response surface methodology (RSM) is a useful statistical method to investigate the effect of different factors on the response developed by Box and co-workers in the 1950s [1]. Response surface methodology is widely applied in production processes of important chemicals and fuels. Zinatizadeh *et al.*, [7] studied the effect of different factors on the performance of a bioreactor using response surface methodology. Bidin *et al.*, [8] optimized the synthesis of palm amino acid surfactant. Mu *et al.*, [9] applied response surface methodology for production of hydrogen from glucose. Ferella *et al.*, [10] optimized the transesterification reaction of bio-diesel production using response surface methodology. Response surface methodology has also been adopted in food industry [11, 12], analytical chemistry [13], hydrometallurgy [14] and analysis of saponification reaction [15-17].

However, it may be observed that the application of design of experiments for process improvement, as in the case of saponification reaction, is not extensively available in open literature. Bursali *et al.*, [18] applied statistical experimental design for the improvement of saponification process in a batch reactor through investigating the effect of various reaction and process parameters on saponification reaction of sodium hydroxide (NaOH) and ethyl acetate (EtOAc, $CH_3COOC_2H_5$). Bursali *et al.*, [18] proposed second order polynomial model which correlates the significant factors to the response, i.e. fraction conversion of the reaction. Similarly, Ahmad and co-workers [19] investigated the alkaline hydrolysis of ethyl acetate in a batch and plug flow reactor and compared the performance of batch and continuous systems.

The alkaline hydrolysis of EtOAc is generally carried out in batch reactor [18]. Nonetheless, the process improvement stage of this process in continuous stirred tank reactor needs to be explored further. Moreover, the application of design of experiments' techniques seems suitable in order to minimize the optimization cost. The aim of this work is to determine the optimum operating conditions for saponification reaction in a continuous stirred tank reactor. Experiments were carried out to find the influence of different reaction and operating parameters on the product quality. The significant factors were determined and a correlation has been proposed using the Design-Expert 8.0.4 Trial version (Stat-Ease Inc., Minneapolis, USA) software.

2. Theory

Saponification is the hydrolysis of ethyl acetate (EtOAc) to produce sodium acetate (CH_3COONa) and ethyl alcohol (C_2H_5OH) using NaOH. The stoichiometric representation of saponification reaction between ethyl acetate and sodium hydroxide is given by Eq. 1 [18]:

$$NaOH + CH_3COOC_2H_5 \rightarrow CH_3COONa + C_2H_5OH \quad (1)$$

This is an irreversible reaction with overall second-order and first order with respect to each reactant. The rate expression is represented by Eq. 2 [18]:

$$-r_{NaOH} = -r_{EtOAc} = k.C_{NaOH} \,.\, C_{EtOAc} \quad (2)$$

In this reaction, hydroxyl ions are consumed and acetate ions are produced. Since hydroxyl ions are more conductive than the acetate ions, a decrease in the conductivity is observed as the reaction progresses. Thus the change in conductivity is used to monitor the alkaline hydrolysis of ethyl acetate. Based on this principle, a relation between the conductivity of the reaction mixture and NaOH concentration is obtained as shown in Eq. 3 [18]:

$$\frac{C - C_\infty}{C_0 - C_\infty} = \frac{C_t^{NaOH} - C_{t->\infty}^{NaOH}}{C_{t->0}^{NaOH} - C_{t->\infty}^{NaOH}} \quad (3)$$

Since,

$C_{t->\infty}^{NaOH} = 0$, Eq. 3 is rearranged and is given as in Eq. 4 [18]:

$$\frac{C - C_\infty}{C_0 - C_\infty} = 1 - X_A = \frac{C_t^{NaOH}}{C_{t->0}^{NaOH}} \quad (4)$$

where, C, C_∞, and C_0represent the specific conductivity at any instant, at the completion of reaction, and at the beginning of the reaction respectively.

The relation between reaction rate and X_A is given by Eq. 5 [18]:

$$-r_{NaOH} = k.C_{NaOH}.C_{EtOAc} = k.\left(C_{t->0}^{NaOH}\right)^2.\left(1 - X_A\right).\left(\frac{C_{t->0}^{EtOAc}}{C_{t->0}^{NaOH}}.X_A\right) \quad (5)$$

3. Materials and Method

3.1 Experimental Set up and Procedure

A laboratory unit of the continuous stirred tank reactor, model CEM Mk II, Armfield Ltd, UK was used to run the experiments. The schematic diagram of the experimental set up is shown in Figure 1.

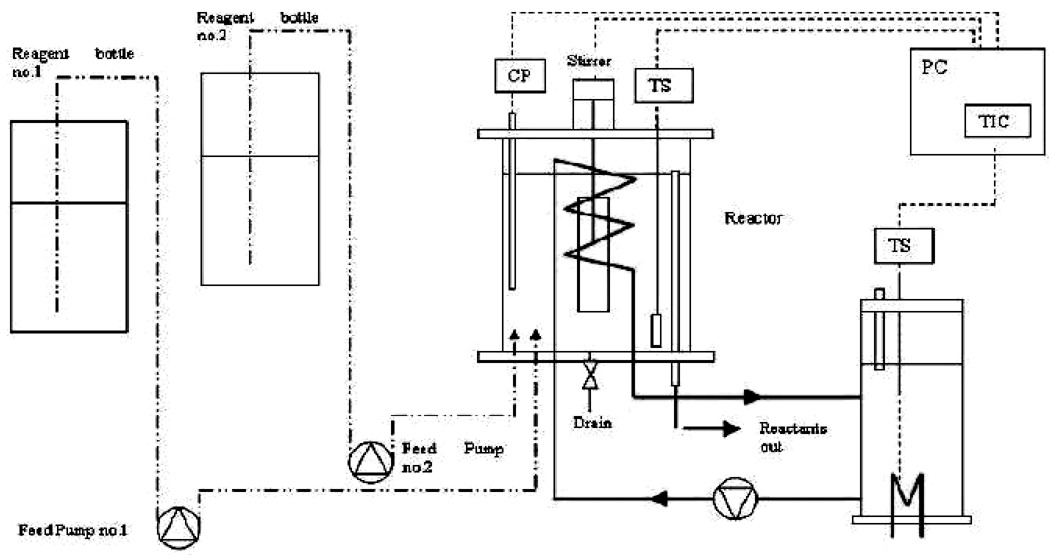

Fig.1 Schematic of experimental setup

It can be seen from Figure 1 that the unit is placed and fixed on bench type service unit on a table. Temperature inside the reactor is controlled by a stainless steel coil which works on continuous recirculation of hot water. Heating coil placed in the hot water storage tank is operated by a PID controller and logged through the computer. A turbine baffled agitator, driven by electric motor works for mixing and to provide uniform heat transfer to the reaction mixture inside the reactor. Conductivity and temperature probes are installed within the reactor to measure the conductivity of the reaction mixture and temperature of heating fluid in hot water storage tank. Peristaltic pumps are used to pump the feed components and hot water from the feed tanks, and hot water storage tank, respectively.

Analytical grade chemical reagents, $CH_3COOC_2H_5$ and NaOH, were used as reactants. A volume of 300 mL of each reactant of known concentration was prepared and stored in respective storage tanks. Both the reagents were fed to the reactor by the respective feed pumps at 60 $mL.min^{-1}$ each at the same time. The mixture was continuously agitated with the help of mixer. Reaction time was kept constant at five minutes and therefore outlet valve was opened after five minutes of reaction time. The hold-up volume of the reactor thus remained constant at 600mL. The conductivity was noted at fixed regular time intervals.

Similarly, experiments were repeated for various samples of different feed concentration, and operated at different temperature, feed flow rate and agitation speed. Each experiment was repeated three times and means values was calculated and employed in the analysis. The conversion of NaOH was calculated using Eq. 3 through Eq.5.

3.2 Screening Experiment

Five factors were considered in this work. These are the concentration of sodium hydroxide and ethyl acetate, flow rate of each reactant, speed of the stirrer and the temperature of the recirculation hot water, representative of the reaction temperature. All the factors were studied at specified high(maximum) and low (minimum) levels. The number of experiments was then calculated as $2^5 = 32$ according to full two-level factorial experimental design method [1].

Maximum, minimum and average levels of the factors are codified as (+1), (−1) and (0), respectively as shown in Table 1. Experimental design matrix was constructed according to standard order rule [1]. The response of the system, i.e. fractional conversion of the reaction along with the values of various parameters is shown in Table 2.

3.3 Determination of Significant Factors

Significant factors are the factors causing a variation in the response. Fisher's *F-test* is a statistical technique used to determine significant factors affecting response of a process [1]. It can also be used to determine the combined effect of variables on the response. F-value for different factors and/or combination of factors is measured with the help of Eq. 6 through Eq. 8 [18]:

Table 1 Natural and codified values of factors

Factors	Neutral			Codified		
	Max.	**Centre**	**Min.**	**Max.**	**Centre**	**Min.**
Agitation rate (rpm), X_4	230	185	140	+1	0	-1
Temperature (°C), X_3	28	34	40	+1	0	-1
NaOH Concentration (mol/L), X_1	0.1	0.055	0.01	+1	0	-1
Ethyl acetate Concentration (mol/L), X_2	0.1	0.055	0.01	+1	0	-1
Feed Ratio, X_5	2	1.5	1	+1	0	-1

Table 2 Results for screening experiment

N	X_1	X_2	X_3	X_4	X_5	X_{NaOH}
1	0.01	0.01	140	1	28	0.863636
2	0.01	0.01	140	1	40	0.948276
3	0.01	0.01	230	1	28	0.944444
4	0.01	0.01	230	1	40	0.884615
5	0.01	0.01	140	2	28	0.93617
6	0.01	0.01	140	2	40	0.962264
7	0.01	0.01	230	2	28	0.897436
8	0.01	0.01	230	2	40	0.854167
9	0.1	0.1	140	1	28	0.826802
10	0.1	0.1	140	1	40	0.515982
11	0.1	0.1	230	1	28	0.949541
12	0.1	0.1	230	1	40	0.983264
13	0.1	0.1	140	2	28	0.801075
14	0.1	0.1	140	2	40	0.886076
15	0.1	0.1	230	2	28	0.933333
16	0.1	0.1	230	2	40	0.929947
17	0.1	0.01	140	1	28	0.095
18	0.1	0.01	140	1	40	0.098
19	0.1	0.01	230	1	28	0.048
20	0.1	0.01	230	1	40	0.045
21	0.1	0.01	140	2	28	0.094
22	0.1	0.01	140	2	40	0.094
23	0.1	0.01	230	2	28	0.05
24	0.1	0.01	230	2	40	0.049
25	0.01	0.1	140	1	28	0.916667
26	0.01	0.1	140	1	40	0.979167
27	0.01	0.1	230	1	28	0.956522
28	0.01	0.1	230	1	40	0.972973
29	0.01	0.1	140	2	28	0.972222
30	0.01	0.1	140	2	40	0.967742
31	0.01	0.1	230	2	28	1
32	0.01	0.1	230	2	40	0.973684

$$F = \frac{MSS_{effect}}{MSS_{error}} \quad (6)$$

$$MSS_{effect} = \frac{SS_{effect}}{d.f.1} \quad (7)$$

$$MSS_{error} = \frac{SS_{effect}}{d.f.2} \quad (8)$$

where

SS_{effect} = sum of square of factor effect
SS_{error} = sum of square of error
SS_{effect} = values may be determined using Yates' Algorithm[1] as shown in Table 3, while SS_{error} may be determined form the center point runs, as shown in Table 4, using Eq. 9 [18]:

$$SS_{error} = \sum (y_{c,i} - y_{c,avg})^2 \quad (9)$$

where,

$y_{c,I}$ = response of center point runs
$y_{c,avg}$ = average of the response of center point runs.

4. Results and Discussion

4.1 Experimental Study of Effects

The experiments on saponification reaction were carried out in a CSTR to investigate the effect of initial concentration of reactants, reactor temperature, agitation rate, and feed ratio of the reactants on the fractional conversion of the reaction (X_{NaOH}).

It may be observed from the results shown in Table 2 that:

1. By increasing sodium hydroxide concentration from 0.01 mol/L to 0.1 mol/L,X_{NaOH} decreases.
2. By decreasing ethyl acetate concentration from 0.1 mol/L to 0.01 mol/L,X_{NaOH} decreases.
3. Increasing reactor temperature from 28°C to 40°C, X_{NaOH} increases slightly.
4. Increasing agitation rate from 140 rpm to 230 rpm, X_{NaOH} increases.
5. Increasing feed ratio from 1 to 2, X_{NaOH} increases.

The F-value of different factors is shown in Table 5. It may be observed from the Table 5, that the calculated F-value for two variables (X_1 and X_2) and their combined effect (X_1. X_2) are greater than the critical F-value. Therefore, the initial concentration of sodium hydroxide and ethyl acetate are the significant factors. Similar results have been reported previously by Bursali *et al.*, [18] for the same reaction system operated in a batch reactor, and by Ahmad and co-workers [19] for batch and plug flow reactor. The F-value for remaining three factors (X_3, X_4 and X_5) and their combined effects is less than the critical F-value indicating that the influence of other operating parameters on the fractional conversion of the reaction is not profound as the significant factors.

4.2 Response Surface Methodology (RSM)

Response surface methodology (RSM) and face centered central composite (FCCC) design were used to develop a correlation relating significant factors to the response of the process. FCCC design matrix is shown in Table 6. As it may be observed from Table 5 that three variables, viz. temperature, agitation rate, and feed ratio are not significant, so only the significant variables: initial concentrations of sodium hydroxide and ethyl acetate are included in Table 6.

Design-Expert 8.0.4 Trial version (Stat-Ease Inc., Minneapolis, USA) was employed in this work to develop the correlation. Using the software a polynomial model was selected to correlate significant factors and response of the process as given in Eq. 10:

$$Y = b_0 + b_1X_1 + b_2X_2 + b_{12}\, X_1.X_2 \qquad (10)$$

The correlation obtained in terms of codified values of the factors is given by Eq. 11:

Table 3 Application of Yates' Algorithm to determine sum of squares of effects

Treatment combination	*Y*	(1)	(2)	(3)	(4)	(5)	$(5)\div2^{5-1}$ Estimate of effect	$(5)^2$	$(5)^2 \div 2^5$ SS_{effect}
1	0.86364	0.95864	2.70211	5.24353	11.0279	22.3775	1.39859	500.75	15.6484459
X_1	0.095	1.74347	2.54143	5.78436	11.3496	7.57941	0.47371	57.4475	1.795233
X_2	0.91667	1.04628	2.89851	5.66199	3.90471	-6.7525	-0.422	45.5969	1.42490158
X_1X_2	0.8268	1.49515	2.88585	5.68757	3.6747	5.7535	0.35959	33.1027	1.03445988
X_3	0.94828	0.99244	2.75191	2.17196	-3.1739	0.08914	0.00557	0.00795	0.00024829
X_1X_3	0.098	1.90606	2.91008	1.73275	-3.5786	-0.3038	-0.019	0.09231	0.00288473
X_2X_3	0.97917	0.92962	2.88077	1.91169	2.80523	-0.2055	-0.0128	0.04224	0.00131993
$X_1X_2X_3$	0.51598	1.95624	2.8068	1.76301	2.94827	0.18345	0.01147	0.03365	0.00105164
X_4	0.94444	0.97862	0.8585	-1.2337	0.17334	-0.5664	-0.0354	0.32081	0.01002535
X_1X_4	0.048	1.7733	1.31346	-1.9402	-0.0842	0.5879	0.03674	0.34562	0.01080075
X_2X_4	0.95652	1.05626	0.90343	-1.5922	-0.3809	1.10066	0.06879	1.21145	0.03785789
$X_1X_2X_4$	0.94954	1.85382	0.82932	-1.9864	0.07703	-0.8096	-0.0506	0.65552	0.0204849
X_3X_4	0.88462	0.94744	0.96176	1.06586	-0.223	-0.0841	-0.0053	0.00708	0.00022111
$X_1X_3X_4$	0.045	1.93333	0.94993	1.73937	0.01744	-0.5824	-0.0364	0.33922	0.0106007
$X_2X_3X_4$	0.97297	0.90317	0.9141	1.40607	0.33124	-0.4607	-0.0288	0.21221	0.00663143
$X_1X_2X_3X_4$	0.98326	1.90363	0.8489	1.5422	-0.1478	0.06565	0.0041	0.00431	0.0001347
X_5	0.88462	0.76864	-0.7848	0.16068	-0.5408	-0.3217	-0.0201	0.10347	0.00323353
X_1X_5	0.094	0.08987	-0.4489	0.01266	-0.0256	0.23001	0.01438	0.05291	0.0016533
X_2X_5	0.97222	0.85028	-0.9136	-0.1582	0.43921	0.40465	0.02529	0.16374	0.00511693
$X_1X_2X_5$	0.80108	0.46319	-1.0266	0.07397	0.14869	-0.143	-0.0089	0.02046	0.00063934
X_3X_5	0.96226	0.89644	-0.7947	-0.455	0.70654	0.25753	0.0161	0.06632	0.00207262
$X_1X_3X_5$	0.094	0.00698	-0.7976	0.0741	0.39413	-0.4579	-0.0286	0.20966	0.00655198
$X_2X_3X_5$	0.96774	0.83962	-0.9859	0.01183	-0.6735	-0.2404	-0.015	0.05779	0.00180594
$X_1X_2X_3X_5$	0.88608	-0.0103	-1.0005	0.0652	-0.1361	0.47903	0.02994	0.22947	0.00717087
X_4X_5	0.89744	0.79062	0.67877	-0.336	0.14803	-0.5153	-0.0322	0.26549	0.00829652
$X_1X_4X_5$	0.05	0.17115	0.38709	0.113	-0.2321	0.29053	0.01816	0.08441	0.0026377
$X_2X_4X_5$	1	0.86826	0.88946	0.00287	-0.5291	0.31241	0.01953	0.0976	0.00305
$X_1X_2X_4X_5$	0.93333	0.08167	0.84991	0.01457	-0.0534	-0.5374	-0.0336	0.28877	0.00902409
$X_3X_4X_5$	0.85417	0.84744	0.61947	0.29168	-0.449	0.38017	0.02376	0.14453	0.00451644
$X_1X_3X_4X_5$	0.049	0.06667	0.7866	0.03956	-0.0117	-0.4757	-0.0297	0.22628	0.0070714
$X_2X_3X_4X_5$	0.97368	0.80517	0.78077	-0.1671	0.25212	-0.4373	-0.0273	0.1912	0.0059751
$X_1X_2X_3X_4X_5$	0.92995	0.04374	0.76143	0.01934	-0.1865	0.43859	0.02741	0.19236	0.00601134

Table 4 Results of centre point runs

S.No	X_1	X_2	Temperature (°C)	Feed Ratio (-)	Agitation Rate(rpm)	X_{NaOH}
1	0.005	0.005	34	1.5	185	0.998
2	0.005	0.005	34	1.5	185	0.998
3	0.005	0.005	34	1.5	185	0.971

Table 5 Analysis of variance of factor effects

Source of Varation	SS_{effect}	d.f	MSS_{effect}	F	Critical F
X_1	1.795232998	1	1.795232998	73.87790117	8.52
X_2	1.424901577	1	1.424901577	58.63792499	
X_1X_2	1.034459882	1	1.034459882	42.57036551	
X_3	0.000248288	1	0.000248288	0.010217627	
X_1X_3	0.002884733	1	0.002884733	0.118713289	
X_2X_3	0.001319927	1	0.001319927	0.054317963	
$X_1X_2X_3$	0.001051639	1	0.001051639	0.043277308	
X_4	0.010025351	1	0.010025351	0.412565877	
X_1X_4	0.010800752	1	0.010800752	0.444475384	
X_2X_4	0.037857889	1	0.037857889	1.557937803	
$X_1X_2X_4$	0.020484904	1	0.020484904	0.843000167	
X_3X_4	0.000221109	1	0.000221109	0.009099153	
$X_1X_3X_4$	0.010600699	1	0.010600699	0.436242766	
$X_2X_3X_4$	0.006631431	1	0.006631431	0.272898396	
$X_1X_2X_3X_4$	0.000134701	1	0.000134701	0.005543271	
X_5	0.003233527	1	0.003233527	0.13306697	
X_1X_5	0.001653298	1	0.001653298	0.068036934	
X_2X_5	0.005116926	1	0.005116926	0.210573074	
$X_1X_2X_5$	0.000639335	1	0.000639335	0.026310089	
X_3X_5	0.002072618	1	0.002072618	0.085292903	
$X_1X_3X_5$	0.006551977	1	0.006551977	0.269628668	
$X_2X_3X_5$	0.001805945	1	0.001805945	0.07431872	
$X_1X_2X_3X_5$	0.00717087	1	0.00717087	0.295097511	
X_4X_5	0.008296523	1	0.008296523	0.341420712	
$X_1X_4X_5$	0.002637704	1	0.002637704	0.108547478	
$X_2X_4X_5$	0.00305	1	0.00305	0.125514414	
$X_1X_2X_4X_5$	0.009024088	1	0.009024088	0.371361646	
$X_3X_4X_5$	0.004516443	1	0.004516443	0.185861867	
$X_1X_3X_4X_5$	0.007071399	1	0.007071399	0.291004092	
$X_2X_3X_4X_5$	0.005975103	1	0.005975103	0.245889022	
$X_1X_2X_3X_4X_5$	0.006011342	1	0.006011342	0.247380327	
Curvature	1.639522871	1	1.639522871	67.47007699	
Error	0.0486	**2**	0.0243		
Total	4.321376006	34			

Table 6 FCCC experimental design matrix

N	X_1	X_2	X_{NaOH}
1	-1	-1	0.863636
2	-1	-1	0.948276
3	-1	-1	0.944444
4	-1	-1	0.884615
5	-1	-1	0.93617
6	-1	-1	0.962264
7	-1	-1	0.897436
8	-1	-1	0.854167
9	1	1	0.826802
10	1	1	0.515982
11	1	1	0.949541
12	1	1	0.983264
13	1	1	0.801075
14	1	1	0.886076
15	1	1	0.933333
16	1	1	0.929947
17	1	-1	0.095
18	1	-1	0.098
19	1	-1	0.048
20	1	-1	0.045
21	1	-1	0.094
22	1	-1	0.094
23	1	-1	0.05
24	1	-1	0.049
25	-1	1	0.916667
26	-1	1	0.979167
27	-1	1	0.956522
28	-1	1	0.972973
29	-1	1	0.972222
30	-1	1	0.967742
31	-1	1	1
32	-1	1	0.973684
33	0	0	0.998
34	0	0	0.998
35	0	0	0.971
36	-1(α)	0	0.9375
37	+1(α)	0	0.895487
38	0	-1(α)	0.922481
39	0	+1(α)	0.96875
40	0	0	0.971

$$Y = 0.72 - 0.22\, C_{NaOH} + 0.22 * C_{EtOAc} + 0.19 * C_{NaOH}\, C_{EtOA} \quad (11)$$

Eq. 11 was found to represent the experimental results adequately with calculated 0.9885 R^2 value. Using the least squares method for parameter estimation, the correlation developed in terms of actual values of significant factors is given by Eq. 12:

$$Y = 1.041 + 10.9 * C_{NaOH} - 0.631 * C_{EtOAc} + 104.1 * C_{NaOH} * C_{EtOAc} \quad (12)$$

4.3 Graphical Residuals Analysis

In order to test the validity of the proposed correlation, graphical residual analysis was applied using Design-Expert 8.0.4 Trial version (Stat-Ease Inc., Minneapolis, USA).The purpose of using the software was to see whether or not:

1. Errors are distributed normally with constant variance and mean zero.
2. Errors are distributed independently and are random.

Figure 2 shows normal graph of residuals. It may be observed from Figure 2 that most of the residuals lie close to the diagonal line indicating that the residuals are distributed almost normally. If the residuals lie close to the diagonal line, as shown in Figure 2, then experimental results may be relied upon with reasonable accuracy and confidence.

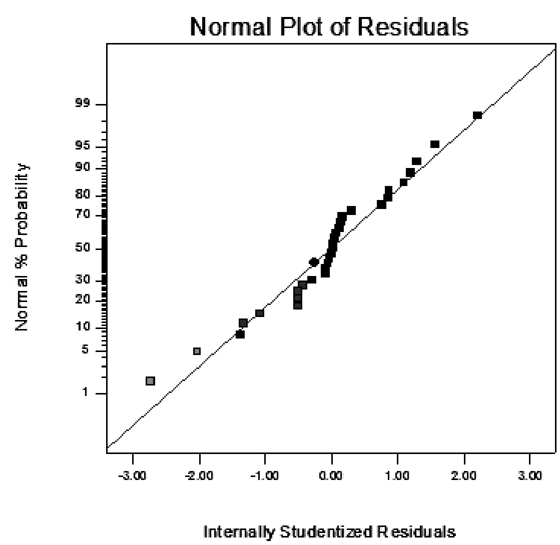

Fig. 2 Normal graph of residuals

Response surface contours were obtained by using Eq. 11 to represent conversion of sodium hydroxide at various levels of initial concentrations of sodium hydroxide and ethyl acetate. The response surface contours are shown in Figure 3.

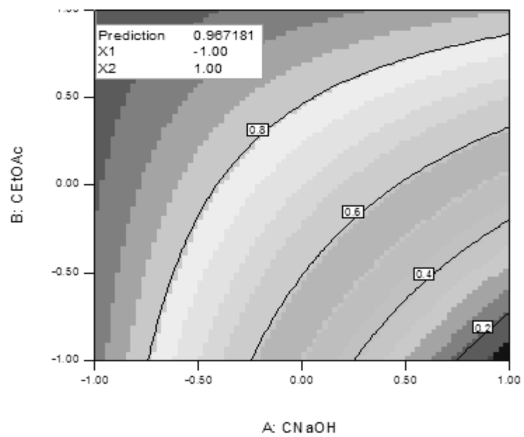

Fig. 3 Response surface contours

5. Conclusions

In this work hydrolysis of ethyl acetate was investigated experimentally in a continuous stirred tank reactor to determine the significant process parameters using statistical experimental design. In the screening experiment, two variables: NaOH initial concentration and ethyl acetate initial concentration were found to be significant operating parameters for the hydrolysis of ethyl acetate. While the effect of agitation rate, feed ratio and temperature were found to be less significant. To obtain maximum conversion, optimum values of the initial concentrations of sodium hydroxide and ethyl acetate were determined to be0.01 and 0.1 mol/L respectively by using response surface methodology.

Two types of experimental design methods were employed in this work. First screening experiment was performed to determine the significant factors. In the screening experiment, Yates' Algorithm was employed for the determination of factor effects. It was found that the feed ratio, reaction temperature and agitation rate are less significant factors compared to the initial concentrations of sodium hydroxide and ethyl acetate. After screening experiment, response surface methodology (RSM) was employed for developing a correlation using face-centered central composite (FCCC) design. In this way a polynomial correlation was proposed. Later on graphical residual analysis, including normal graph of residuals, was used to check the accuracy of the proposed correlation. Comparison of the results of proposed correlation with experimental results indicates reasonable accuracy, and the results may be suitable for improvement of saponification reactions in CSTRs on large scale.

6. Nomenclature

ANOVA	analysis of variance
C_t^{NaOH}	sodium hydroxide concentration at time$t$ (mol/L)
$C_{t->0}^{NaOH}$	sodium hydroxide concentration at time$t = 0$ (mol/L)
$C_{t->\infty}^{NaOH}$	sodium hydroxide concentration at time$t = \infty$ (mol/L)
C_{EtOAc}	ethyl acetate concentration (mol/L)
CSTR	continuous stirred tank reactor
$d.f._1$	degrees of freedom for effect
$d.f._2$	degrees of freedom for error
DOE	design of experiments
F	Fisher's F-value
FCCC	face centered central composite
Feed Ratio	ratio of NaOHto EtOAc
k	second-order rate constant (L/mol.s)
MSS_{effect}	mean sum of square of factor effect
MSS_{error}	mean sum of square of error
N	number of experiment
$-r_{\text{EtOAc}}$	ethyl acetate consumption rate (mol/L.s)
$-r_{\text{NaOH}}$	sodium hydroxide consumption rate (mol/L.s)
RSM	response surface methodology
$SS_{curvature}$	sum of square of curvature
SS_{effect}	sum of square of factor effect
SS_{error}	sum of square of error
X_{NaOH}	conversion of NaOH
X_1	codified value of sodium hydroxide concentration
X_2	codified value of ethyl acetate concentration
X_3	codified value of temperature
X_4	codified value of agitation rate
X_5	codified value of feed ratio
Y	process response, i.e. fractional conversion of sodium hydroxide

$y_{c,avg}$	average of the response of centre point runs
$y_{c,i}$	response of centre point runs

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