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Research Article

Optimization of Process Parameters by Taguchi Method: Catalytic degradation of polypropylene to liquid fuel

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Abstract

In this study, Taguchi method is used to identify the factors and their interactions that may affect the thermo-catalytic degradation of waste polypropylene to liquid fuel in a batch reactor. The yield of liquid fuel in this process was greatly influenced by factors such as temperature, catalyst concentration and acidity of catalyst. By using orthogonal experimental design and analysis technique, the performance of this process can be analyzed with more objective conclusion through only a small number of simulation experiments. Analysis of variance (ANOVA) was carried out to identify the significant factors affecting the response and the best possible factor level combination was determined through. Finally, a regression model for yield of liquid fuel from catalytic degradation of waste polypropylene has been developed, as a function of process parameters. It was found that yield of liquid fuel in this process were highly dependent on temperature followed by acidity of catalyst and catalyst concentration.

Keywords: *Taguchi method, ANOVA, thermo-catalytic degradation, waste polypropylene, liquid fuel, batch reactor*

1. Introduction

The Taguchi method involves reducing the variation in a process through robust design of experiments. The overall objective of the method is to produce high quality product at low cost to the manufacturer. Therefore, poor quality in a process affects not only the manufacturer but also society. This is a method for designing experiments to investigate how different parameters affect the mean and variance of a process performance characteristic that defines how well the process is functioning. The experimental design proposed by Taguchi involves using orthogonal arrays to organize the parameters affecting the process and the levels at which they should be varied; it allows for the collection of the necessary data to determine which factors most affect product quality with a minimum amount of experimentation, thus saving time and resources. Analysis of variance on the collected data from the Taguchi design of experiments can be used to select new parameter values to optimize the performance characteristic [1].

Thermal and catalytic pyrolysis of polypropylene was reported in the literature. The performances of different catalysts are studied using different reactors in various reaction conditions in order to obtain a suitable process for the conversion of polypropylene to liquid fuel. In the present work different parameters such as temperature, catalyst concentration and catalyst type, have been identified which influence the decomposition of waste polypropylene in a batch process and Taguchi method was used to optimize the process parameters for the production of liquid fuel from waste polypropylene. With the help of regression modeling an equation has been developed for yield of liquid fuel as a function of temperature, catalyst concentration and catalyst type.

2. **Materials and methods**

In this experiment waste polypropylene (disposable glass) was degraded using different kaolin clay based catalysts of different acidity values which mostly affect the process.
The waste polypropylene is identified by the DSC of the sample that gives the melting point of the waste sample to be 171 °C, which ensures the samples to be polypropylene.
Commercial grade kaolin clay procured from Chemtex Corporation, Kolkata, India was used as catalyst in the pyrolysis reaction. The chemical composition of the kaolin sample was found to be SiO_2 43.12 %, Al_2O_3 46.07 %, Fe_2O_3 nil, MgO 0.027 %, CaO 0.030 %, ZnO 0.0064%, K_2O 0.01%, TiO_2 0.74, LOI at 1000°C 9.9%. The chemical activation/modification of kaolin was carried out by adding 50g of the clay to 500ml of sulphuric acid solution of different concentrations (1M, 3M, 5M) and refluxing at 110°C under the atmospheric pressure in a round bottomed flask equipped with a reflux condenser for 4 hours. The resulting clay suspension was then rapidly quenched by adding 500ml ice cold water. The content was then filtered, repeatedly washed with distilled water to remove any unspent acid, dried in an oven, calcined at 500°C for one hour and ground in a mortar pastel to powder form. The untreated sample is referred to as KC and treated samples are referred to as KC1M, KC3M, and KC5M in the subsequent text where the numbers refers to the different concentration of acid used. The detail characterization of the different clay materials are described in our previous work [2]. The acidity of the different clay materials used is mentioned in Table 1.

Table 1 Acidity of different clay

Catalyst	Acid centers (mmol/g)
KC	0.049
KC1M	0.108
KC3M	0.116
KC5M	0.210

The experimental setup used in this work consists of a batch reactor made of stainless steel (SS) tube (length- 145 mm, internal diameter- 37 mm and outer diameter- 41 mm) sealed at one end and an outlet tube at other end and is as shown in previous study [3]. The SS tube is heated externally by an electric furnace, with the temperature being measured by a Cr-Al: K type thermocouple fixed inside the reactor and temperature is controlled by external PID controller. Shimaden PID controller SR1 was used to control the temperature of the furnace. The accuracy of this PID controller is ± 0.3% FS (FS =1200ºC). So the temperature can be measured with ±3.6ºC. 20g. of waste polypropylene samples were loaded in each pyrolysis reaction. In the catalytic pyrolysis, a mixture of catalyst and the plastics samples in different catalyst to plastics proportion was subjected to pyrolysis in the reactor set up and heated at a rate of 20 °C/min. up to the desired temperature. The condensable liquid products were collected through the condenser and weighed. After pyrolysis, the solid residue left out inside the reactor was weighed. Then the weight of gaseous product was calculated from the material balance. Reactions were carried out at different temperatures ranging from 400-550 °C.
The statistical analysis has been done using MINITAB 14 software with Taguchi method.

3. **Design of experiments in present experiment**

3.1 Taguchi approach to parameter design

In the present work, experimental work has been designed in a sequence of steps to insure that data is obtained in a way that its analysis will lead immediately to valid statistical inferences. This research methodology is termed as DESIGN OF EXPERIMENT (DOE) methodology. DOE using Taguchi approach attempts to extract maximum important information with minimum number of experiments [4]. Taguchi techniques are experimental design optimization techniques which use standard Orthogonal Arrays (OA) for forming a matrix of experiments. Using an OA to design the experiment helps the designer to study the influence of multiple controllable factors on the

average of quality characteristics and the variations in a fast and economic way. OA's allow screening out few important main effects from the many less important ones. Also it allows us to estimate interaction effects if any and determine their significance [1].
In the present reaction system three operating parameters, each at three levels, are selected to evaluate yield of liquid fuel. The factors to be studied are mentioned in Table 2. Based on Taguchi method, the L_{27}-OA was constructed. The reason for using L_{27}-OA is to evaluate the significance of interaction terms. Interaction means the influence of an operating variable on the effect of other operating variable [5].

Table 2 Factors and their levels in the experimental design

Level	Temperature [T] (ºC)	Catalyst type defined by their acidity [A] (mmol/g)	Plastics to catalyst ratio [C]
1	450	0.049	3
2	500	0.108	6
3	550	0.210	10

In the present work only second order interaction terms have been considered viz., (T*A), (T*C), (A*C) but not the third order viz., (T*A*C) where [T = temperature, A = acidity of different catalysts, C = plastic to catalyst ratio].
The experiments were carried out according to the L_{27}-OA. The yield of liquid fuel (in wt.%) was considered as Taguchi array response. The L_{27}-OA and response values for yield of liquid fuel are shown in Table 3.
Table 3 L_{27}-OA response values and S/N ratio for yield of liquid fuel

Run no.	Temperature (T)	Catalyst type in terms of acidity(A)	Plastics to catalyst ratio (C)	yield of liquid product in wt.% (Y)	S/N Ratio
1	1	1	1	84.5	38.5371
2	1	1	2	83.5	38.4337
3	1	1	3	80.5	38.1159
4	1	2	1	87.8	38.8699
5	1	2	2	86.5	38.7403
6	1	2	3	85.5	38.6393
7	1	3	1	88.1	38.8995
8	1	3	2	87.2	38.8103
9	1	3	3	86.1	38.7001
10	2	1	1	87.5	38.8402
11	2	1	2	84	38.4856
12	2	1	3	83	38.3816
13	2	2	1	88.5	38.9389
14	2	2	2	87.6	38.8501
15	2	2	3	86.8	38.7704
16	2	3	1	92	39.2189
17	2	3	2	90.6	39.1426
18	2	3	3	89.2	39.0073
19	3	1	1	91.4	37.0861
20	3	1	2	73.55	37.3317

21	3	1	3	72.15	37.1647
22	3	2	1	71	37.0252
23	3	2	2	70.3	36.9391
24	3	2	3	70	36.902
25	3	3	1	79.5	38.0073
26	3	3	2	78.3	37.8752
27	3	3	3	76.3	37.6505

3.2 Analysis of data

After conducting the experiment, the results were converted into S/N ratio values. The final L_{27}-OA displaying response values and their corresponding S/N ratio values for yield of liquid fuel are shown in Table 3.

3.2.1 Main Effect Plot

Main effects plot for the main effect terms viz. factors T, A and C are shown in Figure 1. From the main effect plots, it has been observed that yield of liquid fuel increases with increase in temperature from 450ºC to 500ºC and decreases with further increase in temperature from 500ºC to 550ºC due to the formation of more non condensable gaseous/volatile fractions by rigorous cracking at higher temperature. Yield of liquid fuel increases with use of different catalyst of increasing acidity and increase in plastic to catalyst ratio owing to the increase in the acid centers which is mainly responsible for cracking process.

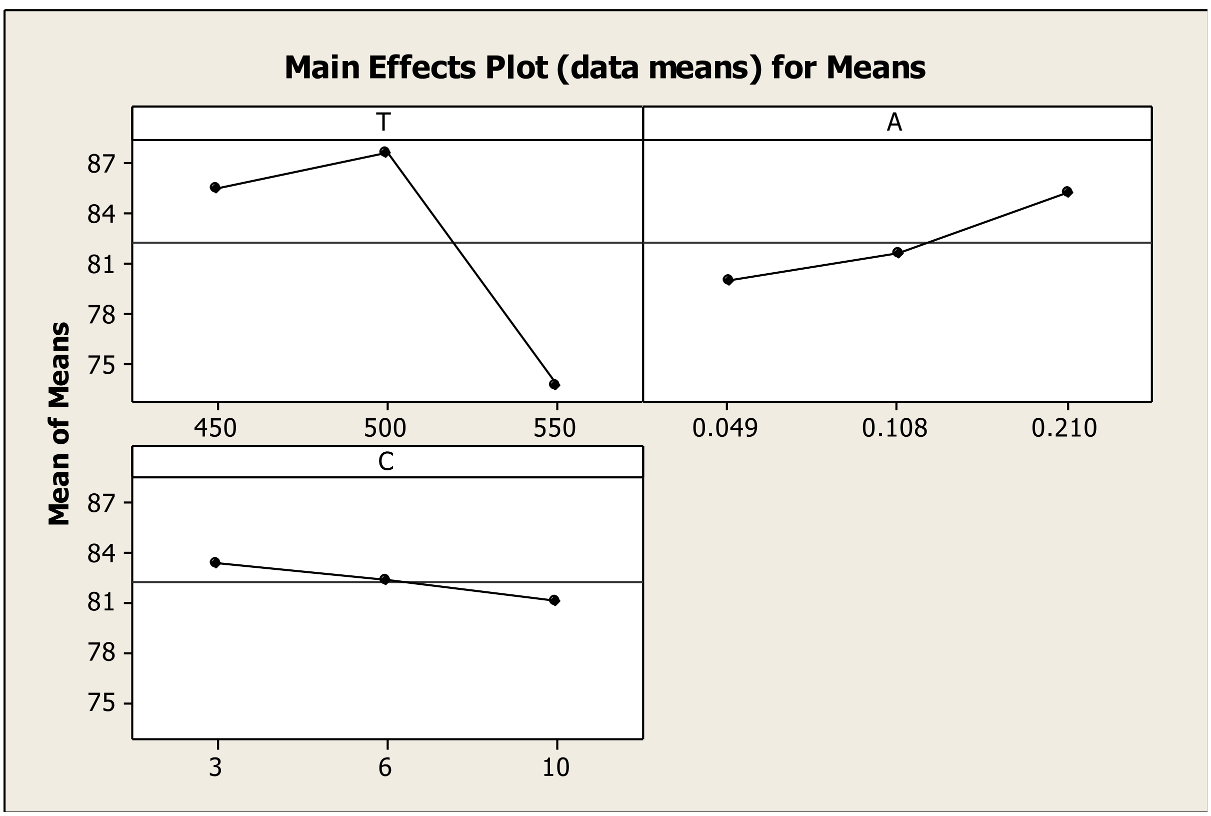

Figure 1 Main effect plots

3.2.2 Interaction Plot

Whether interactions between factors exist or not can be shown by plotting a matrix of interaction plot. Parallel lines in an interaction plot indicate no interaction. However, the interaction plot doesn't tell if the interaction is statistically significant [6].

Interaction plots are most often used to visualize interactions during DOE. Matrix of interaction plot for yield of liquid fuel is shown in Figure 2. It can be seen visually that there are non-parallel lines between temperature and acidity of the catalyst (T and A).The plastic to polymer ratio (C) does not show any significant interaction (parallel lines) for yield of liquid fuel.

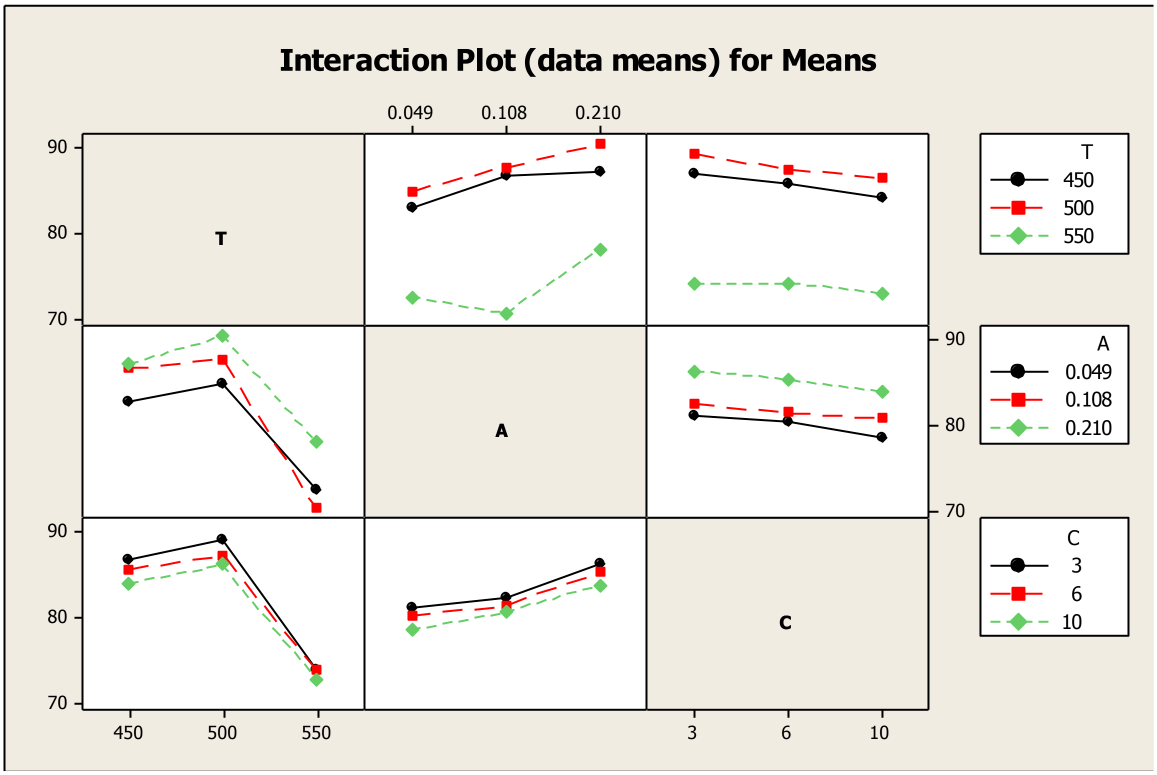

Figure 2 Interaction plot

4. Result evaluation

There are two different methodologies in carrying out the complete OA analysis. A common approach is to analyze the average result of repetitive runs, or a single run, through ANOVA analysis [4]. Since in the present work there is only one replication, further analysis of OA is done through ANOVA. Once the data for a designed experiment have been collected, the sample information is used to make inferences about the population means associated with the various treatments. The method used to compare the treatment means is known as analysis of variance, or ANOVA [[W. Mendenhall et al., 1989]].

An ANOVA Table breaks down the effect of each factor and the experimental error. In addition, it will also break down all of the possible interactions of the factors. In the present work ANOVA Table for yield of liquid fuel from waste polypropylene has been shown in Table 4 using MINITAB 14 software. In this process, it is seen that the significant factors are temperature (T), catalyst type in terms of acidity (A), interaction between temperature and acidity (T*A) and plastic to catalyst ratio (C), written in the decreasing order of significance.

Table 4 Analysis of Variance for Means (ANOVA).

Source	DF	SS	MS	P
T	2	1026.06	513.030	0.000
A	2	126.74	63.370	0.000
C	2	23.04	11.521	0.005
T*A	4	46.07	0.887	0.003
T*C	4	3.55	0.296	0.545
A*C	4	1.18	1.075	0.886
Residual error	8	8.60	8.60	1.075
Total	26	1235.24		

It is seen that the (T*A*C) term has completely vanished. The reasoning for this would be that since all three factors and four interactions use up the 26 degree of freedom in this experiment, there was nothing left over with which to measure the error. The solution to this predicament was to pool the effect of non-significant factors and interactions. Interaction (T*A*C) had very small variance. Therefore its degree of freedom and their sum of squares had been pooled together. These pooled figures are removed from their places on the ANOVA Table and an error factor is created [6].
The final ANOVA Table for percentage contribution (exact value in percent) of each of the significant factors in affecting the response value, which is calculated on the basis of ANOVA Table, is shown in Table 5. The significant main effect terms and interaction terms in the ANOVA Table 5 are computed using the F-ratio as a test statistic [5].

Table 5 Final ANOVA table

Source	DF	SS	MS	F-Ratio	Percentage contribution [SS/SS(Total)*100]
T	2	1026.06	513.030	615.64	83.065
A	2	126.74	63.370	76.03	10.26
C	2	23.04	11.521	13.82	1.865
T*A	4	46.07	0.887	13.82	3.72
Total	26	1235.24			
Pooled error	16	13.33	0.833		1.079

In the process of catalytic decomposition of waste polypropylene in a batch reactor the ranks indicate that temperature has the greatest influence followed by acidity of catalyst and then plastic to catalyst ratio which can be observed in Table 6. The optimum condition in this case where in the percentage degradation of waste polypropylene is maximum is at temperature: 500ºC, catalyst type in terms of acidity: 0.21mmol/g and plastic to catalyst ratio: 3.

Table 6 Response table for mean (Larger is better)

Level	T	A	C
1	85.52	80.02	83.31
2	87.62	81.56	82.39
3	73.62	85.19	81.06
Rank	1	2	3

5. Mathematical modeling

A general regression for the complete model was performed using MINITAB 14 software. Regression in MINITAB 14 uses the ordinary least squares method which derives the equation by minimizing the sum of the squared residuals. In the present study, there are three operational factors each at three levels. The complete model for the degradation of pollutants is shown below. The number of terms in the model depends on the degree of freedom of the main effect terms and the corresponding interactions.

$y = \beta_0+\beta_1x_1 + \beta_2x_1^2 +\beta 3x_2 + \beta_4x_2^2 + \beta_5x_3 + \beta_6x_3^2 + \beta_7x_1x_2 + \beta_8x_1x_2^2 + \beta_9x_1^2x_2 + \beta_{10}x_1^2x_2^2 + \beta_{11}x_1x_3 + \beta_{12}x_1x_3^2 + \beta_{13}x_12x_3 + \beta_{14}x_1^2x_3^2 + \beta_{15}x_2x_3 + \beta_{16}x_2x_3^2 + \beta_{17}x_2^2x_3 + \beta_{18}x_2^2x_3^2$---- (1)

y = yield percentage of liquid fuel

b = model coefficients

x_1, x_2, x_3, = dimensionless coded factors for temperature, catalyst type defined by their acidity, plastic to catalyst ratio respectively.

A simpler reduced model of lower order which provides sufficient information for the prediction of y was found using the F-test statistic [W. Mendenhall et al., 1989]. If $F>F\alpha$, $\alpha=0.05$, then the reduced model does not provide sufficient information and is rejected. A t-test of individual parameters in the regression model was further performed. The t value for each parameter is enlisted in the MINITAB 14 software program [Table 7].

Table 7 Estimated Model Coefficients for Means

Term	Coefficient	SE coefficient	T	P
Constant	82.2556	0.1996	412.153	0.000
x_1	3.2667	0.2822	11.574	0.000
x_1^2	5.3667	0.2822	19.014	0.000
X_2	-2.2333	0.2822	-7.913	0.000
x_2^2	-0.7000	0.2822	-2.480	0.038
x_3	1.0556	0.2822	3.740	0.006
$x_1x_2^2$	1.7778	0.3992	4.454	0.002

To test the null hypothesis H_0: $\beta i=0$ against the alternative hypothesis $\beta i\neq 0$, a two tailed t-test was performed and H_0 was rejected if $t>t\ \alpha/2$ or $t< t\ \alpha/2$ ($\alpha=0.05$). The level of confidence is 95 % i.e. p- value should be less than 0.05. If p-value is less than 0.05 it shows doubt on null hypothesis, which says all sample means equal and effect of that factor on the result is statistically significant [W. Mendenhall et al., 1989]. The final prediction equation obtained from MINITAB 14 software for the yield of liquid fuel by the catalytic decomposition of waste polypropylene is as per equation (1).

$y = 82.2556 + 3.2667x_1 + 5.3667x_1^2 – 2.2333x_2 – 0.7x_2^2 + 1.0556x_3 + 1.7778x_1x_2^2$---(2)

The coefficients of model for means are shown in Table 7.

S = 1.037 R^2 = 99.3% R^2(adj) = 97.7%, the parameter R^2 describes the amount of variation observed in yield is explained by the input factors. R^2 = 99.3 % indicate that the model is able to predict the response with high accuracy. Adjusted R^2 is a modified R^2 that has been adjusted for the number of terms in the model. If unnecessary terms are included in the model, R^2 can be artificially high, but adjusted R^2 (=97.7 %.) may get smaller. The standard deviation of errors in the modeling, S= 1.037. This indicates that model can explain the variation in yield of liquid fuel to the extent of 99.3% which makes the model adequate to represent the process.

The residual plot for means of this process is shown in Figure 3. This layout is useful to determine whether the model meets the assumptions of the analysis. The residuals are the deviations of the observed data values from the predicted value ŷ and estimate the error terms (e_i) in the model [5]. The e_i are assumed to be random and normally distributed with mean equal to zero and constant standard deviation. If the error terms follow a normal distribution, they will fall on a straight line on the normal probability plot. Because they are estimates of the error terms, the residuals should exhibit similar properties. If the assumptions are valid, plots of the residuals versus run sequence, predicted values, and other independent variables should be random and structure less. If structure remains in the residuals, residual plots may suggest modifications to the model that will remove the structure. Investigation of residuals has been used to evaluate the model adequacy. Residuals are found to be scattered and without any definite pattern which prove the adequacy of the model.

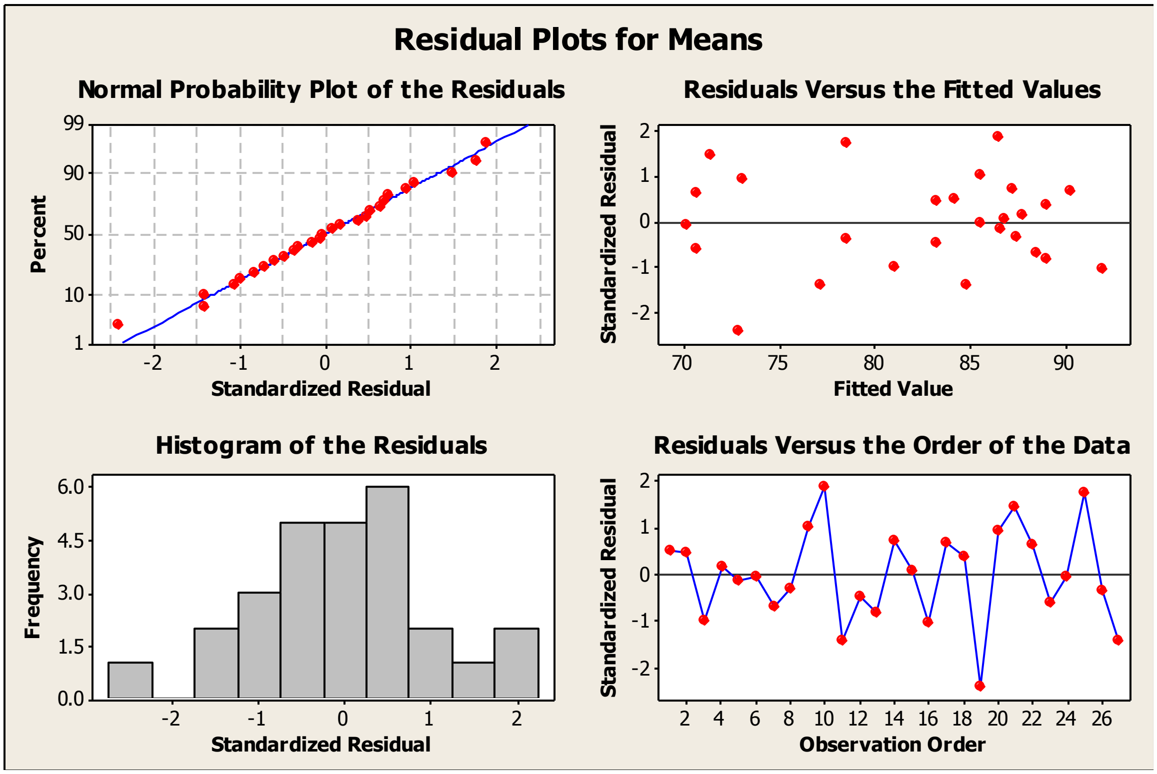

Figure 3 Different residual plots for testing the adequacy of the proposed model.

The residual plots in the Figure 3 and the interpretation of each residual plot for the present experiment is given below:

a. Normal probability plot indicates the data are normally distributed and the variables are influencing the response. Outliers don't exist in the data

b. Residuals versus fitted values indicate the variance is constant and a non-linear relationship exists.

c. Histogram proves the data are not skewed and no outliers exist.

d. Residuals versus order of the data indicate that there are systematic effects in the data due to time or data collection order.

6. Conclusion

With these experiments we upgraded our existing knowledge about the influence of the different process parameters on the yield of liquid fuel in a batch process and thus contributed to improving the process's reliability. This study has shown the application of Taguchi method on the performance evaluation of a chemical process for the production of liquid fuel from waste plastics in a batch reactor. The level of importance of the process's parameters is determined by using ANOVA. Moreover, regression modeling has helped us generate an equation to describe the statistical relationship between the process's parameters and the response variable (yield of liquid fuel) and to predict new observations. The simulation experiment was successful in terms of achieving the objective of experiment, which was to quantify the main effects as well as interactions of potentially influential factors on the degradation of pollutants.

Appendix A: Nomenclature

DF Degree of freedom
SS Sum of squares
MS Mean of squares
S/N Signal to noise ratio
s Standard deviation
R-sq Regression squared
R-sq(adj) Adjusted regression

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