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Study on the preparation of C4 olefins by ethanol coupling based on multiple regression analysis

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Abstract: *In this paper, the optimal catalyst combination and temperature were obtained by using multiple linear regressions using polarization, curve fitting, and regression analysis to prepare C4 olefins with optimal yields. Firstly, the investigation direction was outlined to explore different catalyst combinations and study the relationship with ethanol conversion, C4 olefin selectivity and temperature. In addition to this, the temperature linkage with different catalysts was added to analyze the catalyst components that have the greatest effect at different temperatures and the catalyst components that have the least effect. In addition, the optimal catalyst combination and temperature were further selected to optimize the yield of C4 olefins under the same experimental conditions, and the solved multiple regression equations were used as the constraint variables to establish the particle swarm algorithm model, and finally, MATLAB software was used to program the optimal catalyst composition parameters and temperature data.*

Keywords: *Curve fitting; Polar difference method; Chemical safety; Gray correlation analysis; Multiple regression model; Particle swarm*

1. Introduction

C4 olefins are widely used in chemical production and other important fields of national economy worldwide, and have become an important chemical raw material. At present, the traditional process of steam cracking is used to produce C4 using methane mixture and light petroleum as the raw material, but there are many shortcomings in the traditional preparation method[1]. C4 olefin is becoming an emerging technology for the preparation of C4 olefin in the world due to its low cost and low additional products, therefore, the study of catalyst combination design for the catalytic coupling process and the control scheme for variables such as temperature is of great practical application to optimize the coupled preparation of C4 olefin[2].

Therefore, this paper specifically investigates the relationship between ethanol conversion and C4 olefin selectivity in the reaction products of each catalyst and temperature for different catalyst combinations composed of three catalysts, and analyzes the test results of certain catalyst combinations at 350 degrees for different times in one experiment[3]. Then, considering the effect of different catalyst combinations and temperature conditions on ethanol conversion and C4 olefin selectivity under different combinations of experimental variables, different catalyst combinations were set up with all temperature matching experimental groups to further find the optimal value of C4 olefin yield under the same experimental conditions, and the important experimental limit of temperature was studied when the value was lower than 350 degrees to find the catalyst and temperature that could optimize the C4 olefin yield. The experimental combination of catalyst and temperature that optimizes the yield of C4 olefins.

2. Assumptions and notations

We use the following assumptions.

- The amount and concentration of ethanol will not change due to volatilization during the preparation experiments.
- The reactants are well mixed with the catalyst during the experiment.
- The coupling process does not produce additional products other than the products given in the

ISSN 2522-3488 Vol. 6, Issue 2: 56-62, DOI: 10.25236/IJNDES.2022.060210

question.

● The reaction process is strictly controlled in terms of temperature without additional chemical exotherm.

The primary notations used in this paper are listed as Table 1.

Table 1: Notations

Symbols	Description
t	Temperature
R_y	Ethanol conversion rate
C_s	Yield of C4 olefins
x_{11}	Charging method
x_{12}	Co/SiO2 content
x_{13}	Co load capacity
x_{14}	HAP content
x_{15}	Quartz sand content
x_{16}	Co/SiO2 and HAP charge ratio
x_{17}	Ethanol concentration
R^2	Goodness of fit
β_i	Decision Variables $(i=1,2,3,4,5,6)$
F	Statistical quantities
y	Total estimated level of entitlement
$x_{1,2,3,4}$	Corresponding to contribution, demand, other, and fair rule levels, respectively
$w_{1,2,3,4}$	Weights corresponding to contribution, demand, other, and fair rule levels, respectively
MBF	Material system fraction
S	Spiritual coefficient
MBFmax	The largest fraction of material systems in the world
β	The weights corresponding to the degree of materiality
α	Spiritual coefficients corresponding to the weights
z_i	The i-th

3. Model construction and solving

3.1 Relationship between the conversion of ethanol and C4 olefin selectivity without catalyst combination

3.1.1 Preparation of the model

In this problem objective data on ethanol conversion and C4 olefin selectivity under different catalysts are used to get a macroscopic view of the relationship between the two and temperature, correlation analysis is performed, a scatter plot is drawn, and curve fitting is performed on the basis of the scatter plot to find the best-fit curve[4].

(1) Data pre-processing

Firstly, the data were visualized, and the scatter plots of each catalyst combination at different temperatures regarding the conversion of ethanol and the selectivity of C4 olefins are shown in Figure 1.

Observation of the scatter plot revealed only one data at the temperature of 400 degrees C. The values of ethanol conversion at other temperatures were not abnormal. Therefore, the five classifications of 250, 275, 300, 325, and 350 are mainly considered in the subsequent classification of temperatures in this paper.

ISSN 2522-3488 Vol. 6, Issue 2: 56-62, DOI: 10.25236/IJNDES.2022.060210

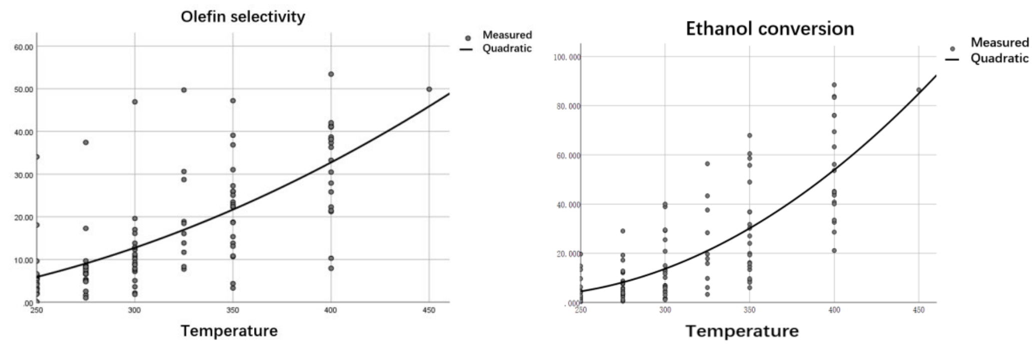

Figure 1: Scatter plot of C4 olefin selectivity and ethanol conversion at different temperatures

(2) Plotting scatter plots

Taking A1 as an example, the scatter plots of ethanol conversion versus temperature and C4 olefin selectivity versus temperature were plotted with the help of SPSS software based on the underlying data in Figure 2 and Figure 3.

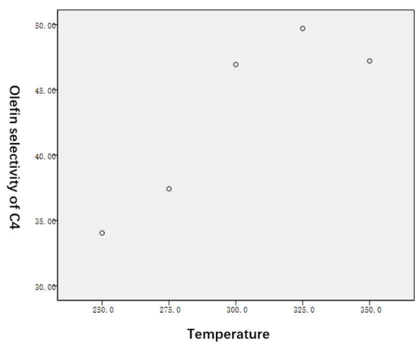

Figure 2: A1 Scatter plot of ethanol conversion versus temperature

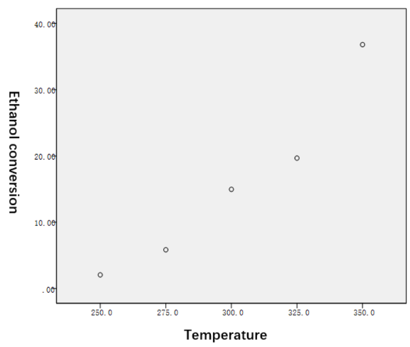

Figure 3: Scatter plot of A1C4 olefin selectivity versus temperature

Therefore, there is a significant correlation between ethanol conversion and C4 olefin selectivity, and the regression model can be determined by correlation analysis of the samples.

3.1.2 Model building

Due to the small sample, this paper adopts direct curve fitting and uses SPSS software to analyze the ethanol conversion of A1 and the selectivity of C4 olefins with temperature and row correlation analysis respectively, and obtain the Pearson correlation coefficient matrix of ethanol conversion with temperature and the Pearson correlation coefficient matrix of selectivity of C4 olefins with temperature.

3.1.3 Model solving

(1) Solution of the first part

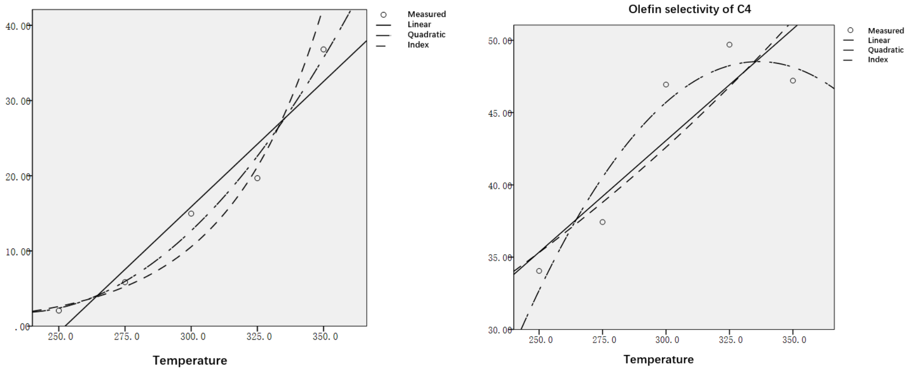

Figure 4: Comparison of fitted curves of ethanol conversion and C4 olefin selectivity

ISSN 2522-3488 Vol. 6, Issue 2: 56-62, DOI: 10.25236/IJNDES.2022.060210

Using C programming language, importing the basic data and analyzing the above scatter plot of ethanol conversion at different temperatures and C4 olefin conversion at different temperatures, we can get.

Comparison of fitted curves of ethanol conversion and C4 olefin selectivity is shown in Figure 4. The equation of each fitted curve and the degree of fit can be obtained from the programming results.

Linear equation.

Ethanol conversion. $$y = 0.333t - 84.083 \tag{1}$$

Selectivity of C4 olefins. $$y = 0.154t - 3.242 \tag{2}$$

According to the fitted results, it can be seen that the fitted linear regression equations are good, with correlation coefficients of 0.932 and 0.787, respectively, and the number of curve parameters of 41.230 and 11.079, respectively, with most of the scattered points regularly scattered on the curve or on the upper and lower sides.

Quadratic equation.

Ethanol conversion. $$y = 0.003t^2 - 1.194t - 141.753 \tag{3}$$

Selectivity of C4 olefins. $$y = -0.02t^2 + 1.422t - 190.783 \tag{4}$$

The results showed that the regression equations were significant, with goodness-of-fit coefficients R^2 of 0.980 and 0.916, respectively, and the number of curve parameters F of 48.377 and 10.899, respectively, indicating a good fit of the model to the data.

Exponential equation.

Ethanol conversion. $$y = 0.002e^{0.28t} \tag{5}$$

Selectivity of C4 olefins. $$y = 13.839e^{0.004t} \tag{6}$$

The results showed that the regression equations were significant, with correlation coefficients R^2 of 0.957 and 0.794, and the number of curve parameters were 67.522 and 11.531, respectively, indicating that the fitted equations were in good agreement with the data.

After comparing the above regression equation parameters, it was concluded that the quadratic equation had a better fit and was more suitable for the fitting of this scatter plot. Thus, the corresponding models of ethanol conversion versus temperature and C4 olefin selectivity versus temperature were established, and the goodness-of-fit R^2 and significance values of the sample determination coefficients were solved by SPSS.

Among them.

$$R^2 = 1 - \frac{\sum (y_i - \widehat{y_i})^2}{\sum (y_i - \overline{y_i})^2} \tag{7}$$

$$F = \frac{\sum (\widehat{y_i} - \overline{y_i})^2}{m \bullet s^2} \tag{8}$$

Some of the regression equations on temperature and ethanol conversion are shown in Table 2.

Analysis of the curve fitting equations obtained from the solution, the relevant fitting parameters from the equations obtained, only a few fitting effect is relatively poor, the results are less reliable, the error is larger, the significance of the remaining equations are, through the test, indicating that the error of the model is relatively small, the results are more reliable, so the model is more reasonable.

Table 2: Table of regression equation of partial temperature and ethanol conversion and related parameters

	A1	A2	…	B7
Sample coefficient of determination R^2	0.980	0.991		0.997
F	48.377	111.107		439.470
P	0.020	0.000		0.000
A1	$y_{A1}=0.003t^2-1.194t-141.753$			
A2	$y_{A2}=-0.001t^2+1.106t-227.393$			
…				
B7	$y_{B7}=0.031t^2-1.711t+228.711$			

(2) The second part of the solution

For the test results of a catalyst at 350 degrees Celsius, the same analysis method as in the first part was first used to correlate the data. Again, due to the small sample size, a direct fit is used in this paper. The curve was fitted using time as the horizontal coordinate and ethanol conversion and the percentage of each product among the total products as the vertical coordinate.

- Data visualization

The data were visualized using EXCEL software.

The approximate trend of different generators over time was analyzed precisely by using curve fitting, fitting a quadratic function, and analyzing the amount of unit change of each variable over time with MATLAB software.

- Curve Fitting

Due to the small sample drop, a direct curve fit between time and ethanol conversion and its generators was performed using SPSS software, and the goodness of fit $R^2=0.988$, $F=162.007$, significance, was statistically significant, the fit was good, and the quadratic regression equation was significant. Figure 5 shows a partial fit between time and variables as follows.

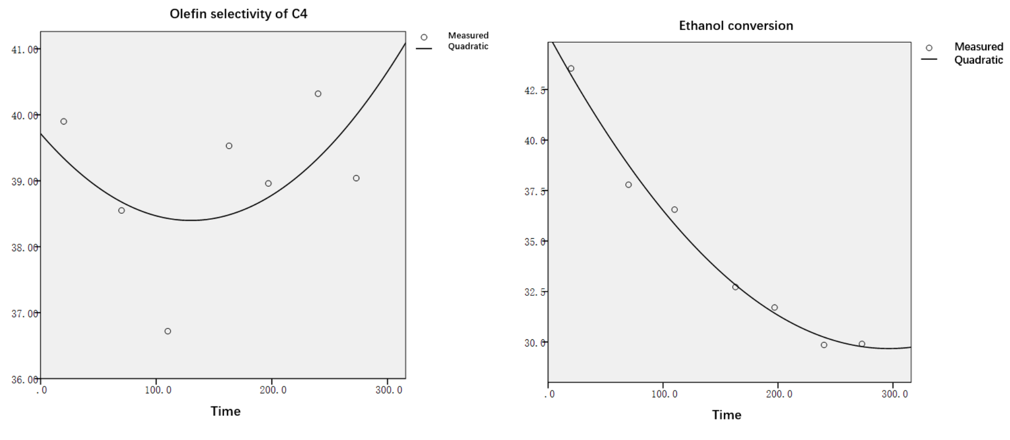

Figure 5: Plot of partial product vs. time curve fit

The resulting fitted equations are shown in Table 3.

(3) Analysis of results

The MATLAB software was used to analyze the fitted equations and compare the fitted equations with those in Annex I. Based on the reaction generation results, the catalyst combinations in Annex II were derived as A3 and A4. The ethanol conversion per unit time and the rate of change of each product per unit time derived from the image analysis and the distance between two points calculated for the sample data, were:0.114, 0.001, 0.027, 0.0086, 0.0468, 0.034, and 0.0302. It can be concluded that the conversion of ethanol has a greater effect with time, followed by , and the selectivity of ethylene has a smaller effect with . According to the fitted curve of the data it can also be concluded that the selectivity of C4 olefins with time is decreasing and then increasing, and there exists a minimum point of content

ISSN 2522-3488 Vol. 6, Issue 2: 56-62, DOI: 10.25236/IJNDES.2022.060210

percentage. This point should be circumvented in the subsequent analysis for the highest yield of C4 olefins.

Table 3: Curve fitting of generators versus time

Ethanol conversion rate	$y=-0.105t+45.249$
Ethylene selectivity	$y=-5.930*10^{-6}t^2+0.004t+4.112$
C4 olefin selectivity	$y=-1.7*10^{-6}t^3+0.001t^2+41.937$
Acetaldehyde selectivity	$y=-2.12*10^{-5}t^2+0.022t+4.499$
Carbon number of 4-12 fatty alcohols	$y=-0.098t+41.838$
Methylbenzaldehyde and methylbenzyl alcohol selectivity	$y=-8.936*10^{-5}t^2+0.03t+2.252$
Selectivity of other generators	$y=0.063t+7.583$

3.2 Experimental combination of catalyst and temperature for optimizing the yield of C4 olefins

The effect of temperature change on ethanol conversion was the highest for ethanol concentration, followed by Co/SiO2 content, and the lowest for quartz sand content. The effect on C4 olefin selectivity was more obvious at 400 degrees C. The decisive effects were Co/SiO and HAP charge ratio below 300 degrees C. The main effect was HAP content between 300 and 400 degree C. The secondary and least influential factors were Co/SiO2 content and quartz sand content[5].

In the limit of space, the optimization model developed for this problem is.

$$\begin{cases} y_{\text{Olefin yield}}=\sum_{i=0,j=1}^{6}\beta_i x_{1j} \\ s.t \\ \min=\sum_{j=1}^{6}(\bar{w}_m\hat{x}_{1j}+\bar{w}_k\hat{x}_{1j}) \\ \dfrac{\bar{w}_m\hat{x}_{12}+\bar{w}_k\hat{x}_{22}+\bar{w}_n\hat{x}_{32}}{x\bar{t}}>0 \\ x_{1j}+x_{2j}+x_{3j}+.....+x_{6j}=100 \qquad (j=1,2,3......6) \end{cases} \tag{9}$$

Then solve the model.

Step1: According to the data given in the question, denote the regression variable and the dependent variable with the corresponding letters

Step2: Apply the least squares estimation method and use MATLAB for programming to calculate the regression coefficients of the multiple regression equation.

Step3: Construct the objective function, and use the change functions of ethanol conversion and C4 olefin selectivity caused by the change of different components in the catalyst combination and the change of temperature solved from the first and second questions as the constraint variables.

Step4: A particle swarm model is established, and the APSO fast particle swarm finding algorithm is used to achieve the optimal solution of the objective function.

Step5: Add the temperature constraint to solve the catalyst combination with temperature below 350 degrees Celsius.

The results of the regression equation coefficient estimation are shown in Table 4.

ISSN 2522-3488 Vol. 6, Issue 2: 56-62, DOI: 10.25236/IJNDES.2022.060210

Table 4: Regression coefficient equation results

Regression variables	Regression coefficient estimates
β_0	-0.0124
β_1	0.4523
β_2	0.3358
β_3	-0.00024
β_4	-0.06
β_5	1.325
Statistical quantities: $R^2 = 0.325$、 F=24.325、 P<0. 0001、 $s^2 = 1.1137$	

4. Conclusion

In this paper, a multiple regression analysis equation was established to try to obtain the relationship between each subvariable (temperature) and the parent variable (C4 olefin selectivity, ethanol conversion) by studying the correlation between the subvariables (catalyst combination and temperature) and the parent variable (C4 olefin selectivity, ethanol conversion) separately, and with the help of this multiple regression model analysis, we found which subvariables are really correlated with the parent variable to achieve the variable screening The ultimate goal of variable screening was achieved. Secondly, this paper innovatively eliminates the irrelevant subvariables, leaving the subvariables with strong correlation, and obtains reliable regression coefficients through regression, revealing the degree of influence of the subvariables on the parent variables and the influence mechanism.

This paper establishes multiple regression models and particle swarm algorithm models based on the characteristic data and relevant indicators of the composite variables, and finds the local extreme values by iteration, and the problem solving scheme is more accurate.

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