ATLANTIS PRESS
Advances in Social Science, Education and Humanities Research, volume 664
Proceedings of the 2022 8th International Conference on Humanities and Social Science Research (ICHSSR 2022)

Study on Optimization of Olefin Preparation by Ethanol Coupling Based on Polynomial Regression Model

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ABSTRACT

In this paper, aiming at the problem of ethanol coupling to prepare C4 olefin, the relationship between ethanol conversion, selectivity of C4 olefin and temperature was studied, and the effects of different combination of catalysts and temperature on ethanol conversion and selectivity of C4 olefin were discussed. In this paper, firstly, the polynomial regression model is established, the standard deviation of each order regression model is calculated, and the order with the best goodness of fit is determined. Then the linear regression model and regression coefficients are obtained. Finally, the multiple decision coefficients of each equation are calculated to evaluate the goodness of fit of each equation. At the same time, it is not difficult to find that under different catalyst combinations, the ethanol conversion and the selectivity of C4 olefins will increase with the increase of temperature, showing a positive correlation.

Then the binary quadratic polynomial regression model is established after the decision variables are determined by the method of control variables. The obtained model is transformed into linear form, and the linear regression model is obtained. Then the regression coefficients of each equation are calculated and the expressions of all equations are obtained. Finally, the multiple decision coefficients used to evaluate the goodness of fit of each equation are calculated. The visual analysis of the eight sets of regression equations shows that the selectivity of C4 olefins is better when the load of Co is near 1%wt, the content of Co/SiO2 is 200mg, the charge ratio of Co/SiO2 to HAP is about 1.2, and the intake rate of ethanol is near 1.2ml/min. When the Co loading is near 1%wt, the content of Co/SiO2 is 200mg, the loading ratio of Co/SiO2 to HAP is 0.5, and the ethanol intake rate is near 0.5ml/min, the ethanol conversion is better.

Keywords: *Polynomial regression model, Single objective optimization model, Multiple decision coefficient*

1. INTRODUCTION

With the development of the global olefin market, C4 olefin, as an important basic chemical raw material, is widely used in the production of chemical products and medicine, while ethanol, an organic compound, is the main raw material for the production and preparation of C4 olefin. In the process of coupling preparation, the combination of ethanol concentration, Co loading, Co/SiO2 and HAP ratio as catalysts has different effects on the yield and selectivity of C4 olefin, which affects the preparation of C4 olefin. In view of this, it is of great value and profound significance to further analyze and design the catalyst combination and study the technological conditions for the preparation of C4 olefin by ethanol catalytic coupling.

2. MODEL PREPARATION

In this paper, the linear regression model is used to fit 14 schemes in 1 charging mode and 2 charging mode, and it is found that the fitting effect is poor[1]. Therefore, there is a big error when the linear regression model is used to describe the relationship between ethanol conversion, C4 olefin selectivity and temperature. In this paper, the polynomial regression model is selected for fitting, and the polynomial regression equations can be divided into several categories according to different orders, in which the first order polynomial is the linear equation. For polynomial regression, the accuracy corresponding to different orders can be measured by the standard deviation of a single measured value, and the calculation formula is as follows:

$$S_r = \sqrt{\frac{\sum_{i=1}^{m}\left(y_i - \hat{y}\right)^2}{m-f}} \qquad (1)$$

M is the number of cases of temperature; *Sr* is standard deviation;*(m-f)* is the degree of freedom; *f=max+1*, *max* is the maximum degree in the polynomial. Through the running results of the program, it is found that the standard deviation of the second-order polynomial regression equation is the smallest. Therefore, the second-order polynomial regression model is used for fitting.

3. POLYNOMIAL REGRESSION MODEL

3.1. Second Level Heading

Ethanol conversion rate y_z , C4 olefin selectivity y_x and temperature *x* have non-linear correlation. After analysis, unary quadratic polynomial regression model with good fitting degree is selected. The specific functional relationship is as follows[2]:

$$\begin{aligned} y_z &= \beta_1 x^2 + \beta_2 x + \beta_0 + \varepsilon \\ y_x &= \mu_1 x^2 + \mu_2 x + \mu_0 + \varepsilon \end{aligned} \qquad (2)$$

The polynomial regression model is transformed into linear regression model Correlation matrix expression:

$$\begin{aligned} Y_z &= X\beta + E \\ Y_x &= X\mu + E \end{aligned} \qquad (3)$$

The calculation formula of regression coefficient:

$$\begin{aligned} \hat{\beta} &= \left(X^T X\right)^{-1} X^T Y \\ \hat{\mu} &= \left(X^T X\right)^{-1} X^T Y \end{aligned} \qquad (4)$$

Multiple linear regression equation is obtained:

$$\begin{aligned} \hat{y}_z &= \hat{\beta}_1 x_2 + \hat{\beta}_2 x_1 + \hat{\beta}_0 \\ \hat{y}_x &= \hat{\mu}_1 x_2 + \hat{\mu}_2 x_1 + \hat{\mu}_0 \end{aligned} \qquad (5)$$

3.2. Calculation of goodness of fit of model

In order to test whether there is a significant linear relationship between dependent variables and independent variables[3], it is necessary to conduct a significance test. Calculation of the sum of squares of regression deviations:

$$SSR = \sum_{i=1}^{n}\left(\hat{y}_i - \bar{y}\right)^2 \qquad (6)$$

Calculation of the sum of squares of residual deviations:

$$SST = \sum_{i=1}^{n}\left(y_i - \bar{y}\right)^2 \qquad (7)$$

Thus, multiple decision coefficient R^2 can be obtained:

$$R^2 = \frac{SSR}{SST} = \frac{\sum_{i=1}^{n}\left(\hat{y}_i - \bar{y}\right)^2}{\sum_{i=1}^{n}\left(y_i - \bar{y}\right)^2} \qquad (8)$$

Multiple decision coefficient R2 can reflect the fitting degree of regression line, and its value range is [0,1]. R2-1indicated good regression equation fitting, and 20R indicated poor regression equation fitting.

4. SOLUTION OF MODEL

4.1. The solution of regression coefficient and regression equation

Tab.1 Polynomial regression equation coefficient table

Ethanol conversion	Serial number	$\hat{\beta}_1$	$\hat{\beta}_2$	$\hat{\beta}_0$
	1	0.003	-1.194	141.753
	2	-0.001	1.106	-227393
	...	...	...	...
	21	0.003	-1.711	228.711
C4 olefin selectivity	Serial number	$\hat{\mu}_1$	$\hat{\mu}_2$	$\hat{\mu}_0$
	1	-0.002	1.422	-190.783
	2	0.003	-1.646	234.745
	...	...	...	...
	21	0.001	-0.060	-9.858

(numbers 1-14 represent catalyst combination numbers A1-A14, and numbers 15-21 represent catalyst combination numbers B1-B7)

The relationship between ethanol conversion, C4 olefins selectivity and temperature under different catalyst combinations can be obtained after obtaining various regression coefficients.

4.2. The solution of regression coefficient and regression equation

The multiple decision coefficients of the polynomial regression model for each catalyst combination scheme can be obtained by using MATLAB. The goodness of fit of the model can be described by the multiple decision coefficients.

Tab.2 Multiple decision coefficients of regression model under different catalyst combination schemes

	Serial number	Multiple decision coefficient
Ethanol conversion	1	0.980
	2	0.991
	…	…
	21	0.997
Selectivity of C olefin	Serial number	Multiple decision coefficient
	1	0.916
	2	0.980
	…	…
	21	0.997

The results showed that the minimum value of multiple decision coefficients of 42 polynomial regression models was $R_{\min}^2 = 0.916 > 0.9$. It indicated that the fitting effect of each model was good and could be used to study the relationship between ethanol conversion and C4 olefins selectivity and temperature.

4.3. Model result analysis

According to the research and calculation of each catalyst combination, the relations between ethanol conversion, C4 olefin selectivity and temperature were obtained respectively. Three catalyst combinations were randomly selected from 21 catalyst combinations for analysis. With the increase of temperature, ethanol conversion rate increased and increased rapidly. Because the catalytic performance of catalyst is different at different reaction temperatures, the dehydrogenation activity of ethanol at low temperature is weak, so the conversion rate of ethanol at low temperature is low. When the reaction temperature is 400 ℃, the ethanol conversion rate is at a high value. When the reaction temperature is higher than 400 ℃, the increase rate of ethanol conversion rate slows down obviously.

The selectivity of C-olefin also increased with the increase of experimental temperature, but the change rate was obviously lower than the change rate of ethanol conversion. When the reaction temperature reached 400 degrees, the selectivity of C4 olefins reached the peak, and then showed a downward trend, reflecting that 400 degrees is a more suitable temperature for the preparation of C4 olefins by ethanol coupling.

5. MULTIVARIATE POLYNOMIAL REGRESSION MODEL

5.1. The decision variables

One decision variable x1 is temperature, and the other is x2. Therefore, the method of control variable is used to determine[4]. According to the different combinations of Co loading, content of Co and SiO2, ratio of Co/SiO2 to HAP, ethanol inlet rate, the presence or absence of HAP, and charging mode, one factor was controlled as variable 2x, and the other factors were quantification, and 6 combinations of 1X and 2X were obtained. The effects of catalyst combination and temperature on ethanol conversion and selectivity of C4 olefins were analyzed.

5.2. Establishment of polynomial regression equation

The ethanol conversion rate y_z and the selectivity of C4 olefin y_x have a nonlinear correlation with temperature 1x and the selected variable x2. The specific functional relationship obtained by the binary quadratic polynomial regression model is as follows:

$$\begin{aligned} y_z &= \beta_0 + \beta_1 x_1 + \beta_2 x_1^2 + \beta_3 x_2 + \beta_4 x_2^2 + \beta_5 x_1 x_2 \\ y_x &= \mu_0 + \mu_1 x_1 + \mu_2 x_1^2 + \mu_3 x_2 + \mu_4 x_2^2 + \mu_5 x_1 x_2 \end{aligned} \quad (9)$$

Establish matrix representation:

$$\begin{aligned} Y_z &= X\beta + E \\ Y_x &= X\mu + E \end{aligned} \quad (10)$$

5.3. Calculation of regression coefficient

The calculation formula of regression coefficient is as follows:

$$
\begin{aligned}
\hat{\beta} &= \left(X^T X\right)^{-1} X^T Y \\
\hat{\mu} &= \left(X^T X\right)^{-1} X^T Y
\end{aligned} \tag{11}
$$

From this, the coefficients in the multiple linear model can be obtained, and the multiple linear regression equation can be obtained:

$$
\begin{aligned}
\hat{y}_z &= \hat{\beta}_0 + \hat{\beta}_1 v_1 + \hat{\beta}_2 v_2 + \hat{\beta}_3 v_3 + \hat{\beta}_4 v_4 + \hat{\beta}_5 v_5 \\
\hat{y}_x &= \hat{\mu}_0 + \hat{\mu}_1 v_1 + \hat{\mu}_2 v_2 + \hat{\mu}_3 v_3 + \hat{\mu}_4 v_4 + \hat{\mu}_5 v_5
\end{aligned} \tag{12}
$$

6. SOLUTION OF MULTIPLE REGRESSION MODEL

6.1. The solution of regression coefficient and regression equation

Tab.3 Table of regression coefficients for regression model of ethanol conversion

	wt	*mg*	*Ratio*	*Ethaol*
β_0	-51.893	184.454	131.291	93.994
β_1	0.151	-1.308	-1.022	-0.618
β_2	0.001	0.002	0.002	0.002
β_3	-4542.533	-0.299	3.591	-30.935
β_4	140841.503	-0.001	3.310	13.935
β_5	3.753	0.002	-0.052	-0.061

(wt is the Co load, mg is the content of Co and SiO2, Ratio is the charge Ratio of Co/SiO2 and HAP, Ethaol is the rate of ethanol intake)

The polynomial regression equation with different catalyst combinations and temperature as decision variables and ethanol conversion as dependent variables was obtained

$$
\begin{aligned}
y_{z1} &= -51.893 + 0.151x_1 + 0.001x_1^2 - 4542.533x_2 + 140841.503x_2^2 + 3.753x_1x_2 \\
y_{z2} &= 184.454 - 1.308x_1 + 0.002x_1^2 - 0.299x_2 - 0.001x_2^2 + 0.002x_1x_2 \\
y_{z3} &= 131.291 - 1.022x_1 + 0.002x_1^2 + 3.591x_2 - 0.001x_2^2 - 0.052x_1x_2 \\
y_{z4} &= 93.994 - 0.618x_1 + 0.002x_1^2 - 30.612x_2 + 13.935x_2^2 - 0.061x_1x_2
\end{aligned} \tag{13}
$$

Similarly, the regression model regression coefficient of C4 olefins selectivity can be obtained. The expression of polynomial regression equation with different catalyst combinations and temperature as decision variables and C4 olefins selectivity as dependent variable was obtained:

$$
\begin{aligned}
y_{x1} &= 56.232 - 0.606x_1 + 0.001x_1^2 + 8979.605x_2 - 392987.992x_2^2 + 0.242x_1x_2 \\
y_{x2} &= 60.944 - 0.457x_1 + 0.001x_1^2 - 0.228x_2 + 0.001x_2^2 + 0.001x_1x_2 \\
y_{x3} &= -1.656 - 0.188x_1 + 0.001x_1^2 + 33.100x_2 - 12.387x_2^2 + 0.008x_1x_2 \\
y_{x4} &= 29.189 - 0.348x_1 + 0.001x_1^2 + 24.646x_2 - 8.702x_2^2 - 0.016x_1x_2
\end{aligned} \tag{14}
$$

6.2. Model result

The multiple decision coefficients of the polynomial regression equation calculated by MATLAB are shown in the following table.

Tab.4 Regression coefficient of regression model under different catalyst combination and temperature

	Ethanol conversion	Selectivity of C4 olefin
wt	0.955	0.974
mg	0.963	0.958
Ratio	0.998	0.979
Ethaol	0.992	0.982

The regression coefficients of all regression models are above 0.95, which indicates that the binary quadratic polynomial regression model selected has a good fitting effect.

After visual processing of the data, the catalyst combination is divided into the following categories for discussion.

(a) The variables are Co load and temperature

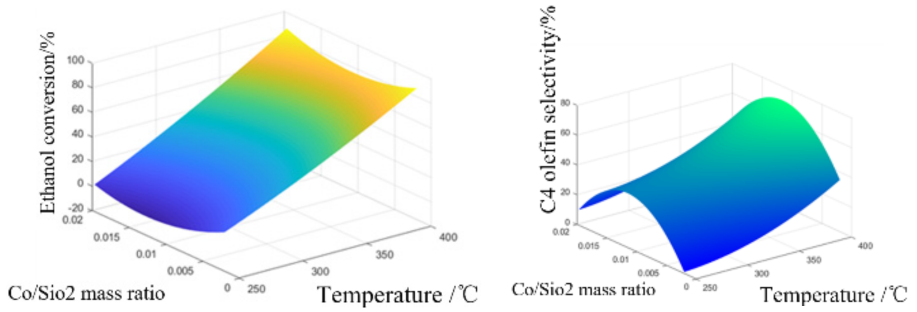

Figure 1 Three-dimensional diagrams of ethanol conversion, C4 olefins selectivity and decision variables

Both ethanol conversion and C4 olefins selectivity increase with increasing temperature. For ethanol conversion, when the Co load is small or large, the ethanol conversion is large; The minimum ethanol conversion rate was located near the Co loading of 1%wt. For the selectivity of C4 olefin, the selectivity of C4 olefin is the highest when the Co load is near wt1%, and the selectivity of C4 olefin decreases greatly when the Co load is too high.

(b) The variables are Co/SiO2 content and temperature

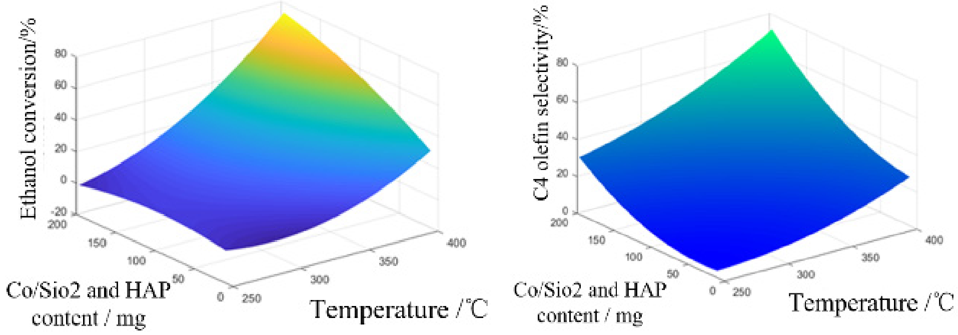

Figure 2 Three-dimensional diagrams of ethanol conversion, C4 olefins selectivity and decision variables

As for the ethanol conversion rate, when the content of Co/SiO2 is high, the ethanol conversion rate increases the fastest with the increase of temperature, and when the content of Co/SiO2 is 200mg and the temperature is 400 degrees, the ethanol conversion rate reaches the maximum. The selectivity of C4 olefin is similar to that of ethanol conversion[5]. When the content of Co/SiO2 is high, the selectivity of C4 olefin increases the fastest with the increase of temperature, and when the content of Co/SiO2 is 200mg and the temperature is 400 degrees, the ethanol conversion reaches the maximum.

(c) The variables are Co/SiO2 and HAP charging ratio and temperature:

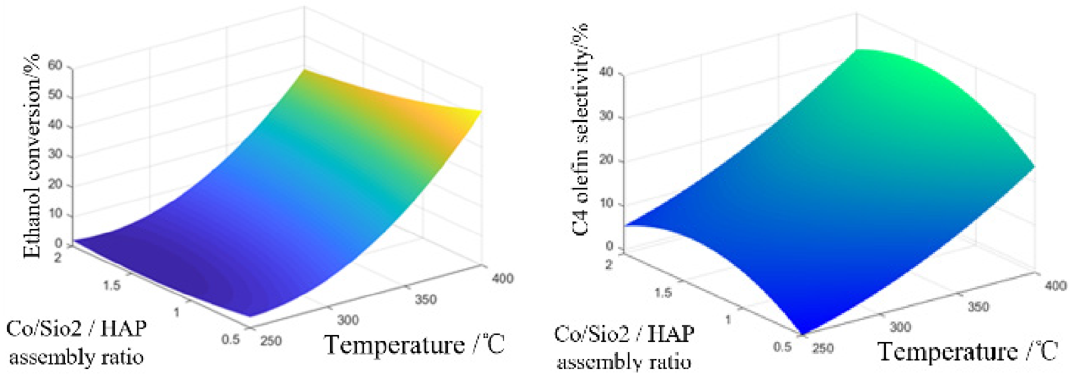

Figure 3 Three-dimensional diagrams of ethanol conversion, C4 olefins selectivity and decision variables

As for the ethanol conversion rate, the ethanol conversion rate increases the fastest with the increase of temperature when the Co/SiO2 to HAP charging ratio is low, and the ethanol conversion rate reaches the maximum when the Co/SiO2 to HAP charging ratio is 0.5 and the temperature is 400 degrees. For C4 olefin selectivity, the selectivity of C4 olefin is not ideal when the Co/SiO2 to HAP charging ratio is low or high, and the selectivity of C4 olefin reaches the maximum when the Co/SiO2 to HAP charging ratio is around 1.2 and the temperature reaches 400 degrees.

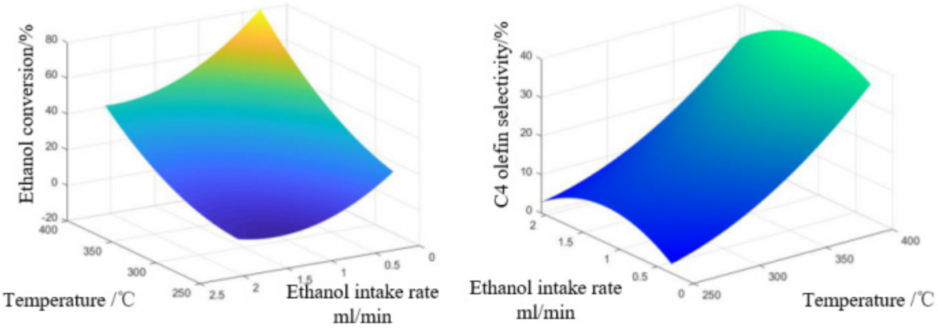

Figure 4 Three-dimensional diagrams of ethanol conversion, C4 olefins selectivity and decision variables

As for the ethanol conversion rate, the ethanol conversion rate when the ethanol inlet rate is slow is generally higher than that when the ethanol inlet rate is fast, and the ethanol conversion rate reaches the maximum[6] when the temperature reaches 400 degrees and the ethanol inlet rate is around 0.5 mL /min. For C4 olefin selectivity, the selectivity of C4 olefin is not ideal at a slower or faster ethanol intake rate, and the selectivity of C4 olefin reaches the maximum at the ethanol intake rate of 1.2 mL /min and the temperature of 400 degrees.

7. CONCLUSION

In this paper, aiming at the problem of ethanol coupling to prepare C4 olefins, polynomial regression models were established respectively. Based on the single objective optimization model of simulated annealing algorithm, the relationship between ethanol conversion, C4 olefin selectivity and temperature was studied. the effects of different combined catalysts and temperature on ethanol conversion and C4 olefin selectivity were discussed. In order to study the relationship between ethanol conversion, selectivity of C4 olefins and temperature under different catalyst combinations, a polynomial regression model was established to evaluate the goodness of fit of each equation. The 42 sets of regression equations obtained from the analysis show that under different catalyst combinations, the ethanol conversion and the selectivity of C4 olefins will increase with the increase of temperature, showing a positive correlation. Then the polynomial regression model is established, the regression coefficients of each equation are calculated, and the expressions of all equations are obtained. Finally, the multiple decision coefficients used to evaluate the goodness of fit of each equation are calculated. The visual analysis of the eight sets of regression equations shows that the selectivity of C4 olefins is better when the load of Co is near 1%wt, the content of Co/SiO2 is 200mg, the charge ratio of Co/SiO2 to HAP is about 1.2, and the intake rate of ethanol is near 1.2ml/min. When the Co loading is near 1%wt, the content of Co/SiO2 is 200mg, the loading ratio of Co/SiO2 to HAP is 0.5, and the ethanol intake rate is near 0.5ml/min, the ethanol conversion is better.

REFERENCES

[1] LV Shaopei. Preparation of butanol and C4 by ethanol coupling olefins [D]. Dalian University of technology, 2018.

[2] Zhang Xi. Application of polynomial fitting in mathematical modeling [J]. Peer, 2016 (14): 340.

[3] Liu Penghua, Zeng Lifang, Yuan Xianhong, Du Baolin, Su Weihang. Quality calibration of pycnometer for soil particle specific gravity test - Comparison of linear fitting and polynomial fitting curves [J]. Guangdong chemical industry, 2021,48 (14): 81-83.

[4] Xiong Qingru. Research on multi-objective programming model processing based on linear programming model [J]. Science and technology innovation, 2020 (28): 42-43.

[5] Liu Jianjun, Si Guangya, Wang Yanzheng, he Dachuan. Research on model-based multi-objective optimization problem method [J]. Journal of system simulation, 2020,32 (11): 2138-2145.

[6] You Jinfeng, an Ying. Air quality data calibration model based on multivariate polynomial regression [J]. Journal of Shandong Institute of Commerce and technology, 2020,20 (06): 91-99.