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Experimental Model of Olefin Preparation from Ethanol Based on Multiple Nonlinear Regression

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Abstract: C4 olefins are widely used in the production of chemical products and medicine. The effects of different catalyst combinations and temperatures on ethanol conversion and C4 olefin selectivity were investigated. Firstly, according to the given data, the three-dimensional curve fitting for ethanol conversion, temperature and catalyst combination is made respectively, and the qualitative analysis based on data visualization is carried out. Then, the control variable method is used to compare and analyze the data of each group, and the effects of loading mode, addition of hap, CO loading and ethanol concentration on C4 olefin yield are analyzed respectively. Finally, based on the results of the above two qualitative analysis, the multiple nonlinear regression equations for ethanol conversion and C4 olefin selectivity are obtained, and the quantitative impact analysis is obtained.

1. Introduction

C4 olefins are widely used in social life and are closely related to people's daily life. China is a large olefin consumer with high demand for olefins. The traditional olefin production raw materials mainly rely on oil, which not only keeps the production cost of olefins high, but also seriously endangers China's energy security. Therefore, it is urgent to develop a route to prepare olefins from renewable clean material energy. Nowadays, China has created a new shortcut of "coal to olefin" and adopted the road of "coal instead of oil" to produce olefins, which has greatly reduced China's dependence on oil, reduced the pressure of crude oil supply and improved the national energy security strategic reserve. It is of great significance and value to explore the catalytic conditions for ethanol coupling to C4 olefins by controlling variables and combinatorial design experiments. In this paper, the effects of different catalyst combinations and temperatures on ethanol conversion and C4 olefin selectivity were analyzed qualitatively and quantitatively.

2. Model Establishment and Solution

2.1 Qualitative analysis based on data fitting given

A total of 21 groups including a and B, as well as several groups of data on temperature, ethanol conversion and C4 olefin selectivity, have been provided. Taking ethanol conversion and C4 olefin selectivity as two groups of dependent variables, corresponding to the common independent variable temperature and catalyst group, the three-dimensional curve fitting diagram corresponding to these data is made by using Matlab's curve fitting toolbox cftool, as shown in Figures 1 and 2.

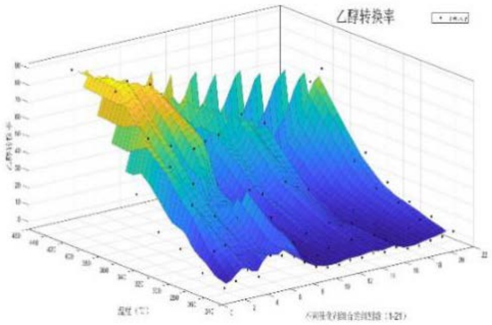

Figure 1: Relationship between ethanol conversion

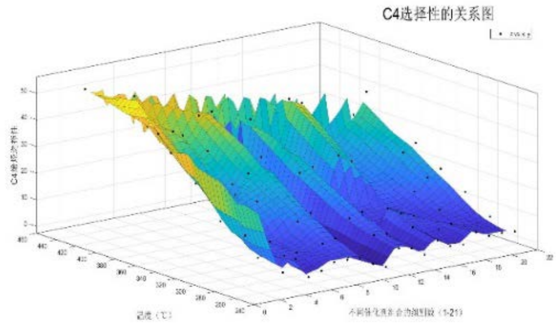

Figure 2: Relationship between selectivity of C4 olefins

(1) The higher the ethanol conversion, the darker the color becomes. It can be seen that with the increase of temperature, the ethanol conversion under the influence of different groups of catalysts also shows an upward trend, that is, the temperature between 250℃ and 400℃ is positively correlated with the ethanol conversion of this reaction;

(2) The ethanol conversion of different groups of catalysts at the same temperature has a certain degree of discrimination, showing a certain curvature and concavity in the fitting curve. The results show that the group of catalyst also has a certain relationship with the conversion of ethanol; The first 14 groups numbered A1 ~ A14 used loading mode I, while the latter seven groups numbered B1 ~ B7 used loading mode II. However, from the fitting surface, there was no obvious differentiation sign such as stratification or fault, and it was still a smooth surface. It can be boldly speculated that the loading mode had little or no impact on the ethanol conversion.

(3) From the three-dimensional fitting curve in Fig 2, it can be seen that the relationship is roughly similar to that of ethanol conversion rate, and the selectivity of C4 olefins is positively correlated with the reaction temperature; The catalyst group also has a certain influence on the selectivity of C4.

2.2 Qualitative analysis based on control variable method

(1) To explore the effect of loading mode on C4 yield

Through the observation data, it is not difficult to find that there is only the change of loading mode between A12 group and B1 group. Obviously, by comparing the left and right groups of data, it can be seen that the loading mode has no significant effect on C4 yield, ethanol conversion and C4 olefin selectivity. Therefore, the two groups are not discussed separately in the regression below.

(2) To explore the effect of HAP on C4 yield

In this paper, A11 and A12 group were selected as the research objects. It was found that the C4 yield without AHP was much lower than that with AHP. Therefore, it was concluded that AHP had a great impact on promoting C4 yield.

(3) To explore the effect of CO loading on C4 yield

A1 and A2 groups were selected as the research objects. Through data comparison, it was found that the loading amount of CO had an obvious growth effect on C4 yield.

(4) To explore the effect of ethanol concentration on C4 yield

A1 and A2 groups were selected as the research objects. Through data comparison, ethanol concentration had an obvious effect on C4 yield in a certain temperature range.

2.3 Quantitative analysis based on multiple nonlinear regression model

2.3.1 Model establishment

According to the experimental data, the effects of different catalyst combinations and temperatures on ethanol conversion and C4 olefin selectivity were discussed. According to the variables of different catalyst combinations, this paper selects three quantitative indexes: CO loading, ethanol addition per minute and loading ratio to analyze the effects of different catalyst combinations, and also quantitatively analyzes the effects of the same temperature on ethanol conversion and C4 olefin selectivity under different catalyst combinations, Therefore, a multiple regression model is established for analysis. The modeling process is as follows:

1. Extract relevant variable data:

Firstly, the relevant variable data are extracted, and the catalyst combination number, catalyst combination, temperature, ethanol conversion and C4 olefin selectivity data are analyzed. Other influencing factors are not considered temporarily, and finally the influence relationship is obtained.

2. Data preprocessing:

The extracted values are processed with outliers and normalized according to Matlab. In the A11 catalyst combination, the variable data we need to study is small, and the acetaldehyde selectivity is large, which may cause large deviation to the results. Therefore, we delete the A11 catalyst combination value, and finally leave 20 groups of catalyst combination data. Then normalize the 20 groups of catalyst combination data.

3. Establishment of nonlinear regression model:

The final data obtained after data preprocessing is analyzed by SPSS, and it is observed that there is no linear relationship between multiple variable factors. Therefore, this paper establishes a multiple nonlinear regression model to explore the influence between these variables.

2.3.2 Model solution

The nonlinear regression model is established by SPSS, and the normalized data values are input for regression analysis to solve the values of each parameter. According to the relevant data analysis operation and data access, the error of the estimated value of the loading ratio parameter has a great impact, so only the quadratic and cubic regression coefficient analysis of the loading ratio variable is considered in this nonlinear regression. The regression curves of ethanol conversion and C4 olefin selectivity are customized as follows:

It is assumed that the regression curve of ethanol conversion is defined as follows:

$$y_1 = ax_1 + b{x_1}^2 + c{x_1}^3 + dx_2 + e{x_2}^2 + f{x_2}^3 + g{x_3}^2 + h{x_3}^3 + ix_4 + j{x_4}^2 + k{x_4}^3 + l \quad (1.)$$

Where, a, b, c,..., k are the parameters of various variables of the regression function of ethanol conversion under different regression times, l is the error term constants, the variable x_1 is the co loading amount, the variable x_2 is the amount of ethanol added per minute (the reaction is affected by changing the concentration of ethanol reactants), the variable x_3 is the loading ratio and the variable x_4 is temperature, The values of various parameters in the definition curve are calculated through the nonlinear regression model in SPSS.

Table 1: Estimated parameters of ethanol conversion curve

parameter	Estimated value	Standard error	confidence interval	
			lower limit	upper limit
a	-3.548	0.727	-4.991	-2.104
b	12.751	2.461	7.866	17.635
c	-9.340	1.821	-12.954	-5.726
d	0.006	0.361	-0.710	0.722
e	-0.271	0.794	-1.847	1.306
f	0.011	0.491	-0.963	0.985
g	-0.165	0.786	-1.725	1.396
h	0.089	0.738	-1.376	1.554
i	0.177	0.297	-0.412	0.766
j	0.942	0.869	-0.783	2.667
k	-0.232	0.691	-1.604	1.139
l	0.413	0.097	0.222	0.605

According to the parameter data calculated in Table 1, the ethanol conversion curve is as follows:

$$y_1 = -3.548x_1 + 12.751{x_1}^2 - 9.34{x_1}^3 + 0.006x_2 - 0.271{x_2}^2 + \ldots + 0.413 \quad (2.)$$

Similarly, the selectivity curve of C4 olefins can be obtained:

$$y = 2.861x_1 - 8.499{x_1}^2 + 5.593{x_1}^3 + 0.828x_2 - 0.775{x_2}^2 \ldots - 0.631 \quad (3.)$$

It is further explained that different catalyst combinations and temperatures have an effect on the conversion of ethanol and the selectivity of C4 olefins. For ethanol conversion, CO loading variable has the greatest influence on it, while other factors such as loading ratio have little influence on it; For the selectivity of C4 olefins, the loading amount of CO, loading ratio and temperature have a great influence on it, and the amount of ethanol added per minute has little influence on the selectivity of C4 olefins.

3. Model Evaluation

In this paper, the three-dimensional curve fitting is used to present the changes of ethanol conversion and C4 olefin selectivity with temperature and catalyst group. Combined with the statutory analysis of the commonly used control variables in the experiment, the multiple non-linear regression equations of the two dependent variables are made on this basis. It is justified and meets the needs of real experiments. It can be applied to the scene of designing the best experimental scheme in the real experimental design.

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