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Optimization of C4 olefin preparation process based on control variables and simulated annealing algorithm

Renjie Hao[1,*], Qingyu Ge[1], Songyuan Huang[1]

[1]*College of Internet of Things Engineering, Jiangnan University, Wuxi, Jiangsu, 214122, China*
**Corresponding author*

Abstract: *The preparation of C4 olefins by catalytic coupling of ethanol is an important reaction in chemical production and pharmaceutical preparation industries. In the preparation process, the choice of catalyst combination and reaction temperature will affect the selectivity and yield of product C4 olefins. Under the principle of controlling variables, by classifying the experimental data, we studied the effects of various factors in the catalyst on ethanol conversion and C4 olefin selectivity, respectively. After completing the study of catalyst factors, it was found that 1wt% Co loading and 1:1 loading ratio were the most suitable conditions. From this quantitative relationship model, we established an optimization model with the C4 olefin yield as the objective function. During the solution process, due to the large amount of calculation and the complexity of the traditional solution method, we chose the simulated annealing algorithm. For this model, it is easier to find the global optimal solution. It was found that the yield of C4 olefins was the highest at 38.09% when the Co loading was 1wt%, the charging ratio was 1:1, the ethanol concentration was 0.76ml/min, and the temperature was 399.9214℃.*

Keywords: *Controlled Variable, Simulated Annealing Algorithm, Preparation of C4 Olefins*

1. Introduction

C4 olefin is an important chemical raw material, which can be prepared by ethanol catalytic coupling.[1] In the preparation process, the catalyst combination and reaction temperature will affect the selectivity and yield of C4 olefins.It is of great value to explore the process conditions for preparing C4 olefins from ethanol through catalyst combination design.[2]

2. Catalyst influence model based on control variables

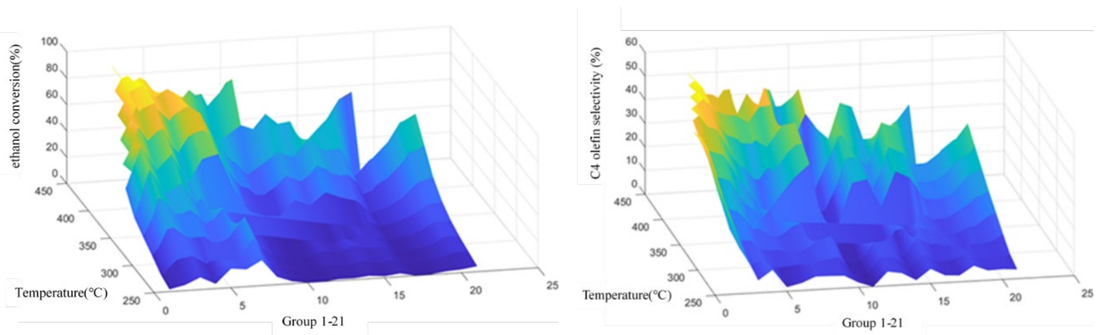

Figure 1: Overview of ethanol conversion and C4 olefin selectivity

In order to explore the effect of different catalyst combination and temperature on ethanol conversion and C4 olefin selectivity, according to 21 groups of different experimental conditions and experimental results in the data, the ethanol conversion rate as shown in Fig. 1 was plotted Y_1 And C4 olefin selectivity Y_2 The relationship between temperature and catalyst combination can be intuitively felt by the overview diagram corresponding to the temperature and catalyst combination.

According to the principle of single variable, 21 groups of data were selected, including A12, A13, A14 and B1 with 1wt of CO loading and 13.255 of ethanol relative concentration.Because the influence of loading mode is small, B1 and A12 can be combined. Finally, we only need to analyze the experimental data of a12-a14.

Taking the reaction conditions at 350 ℃ as an example, the curve is drawn with the charge ratio as the independent variable, the ethanol conversion and C4 olefin selectivity as the dependent variables, as shown in Figure 2. The curve is drawn with the charge ratio as the independent variable and the C4 olefin yield as the dependent variable, as shown in Figure 3.

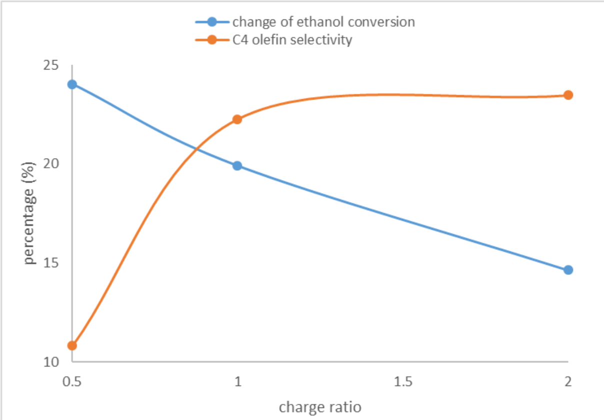

Figure 2: Change of ethanol conversion and C4 olefin selectivity with charge ratio

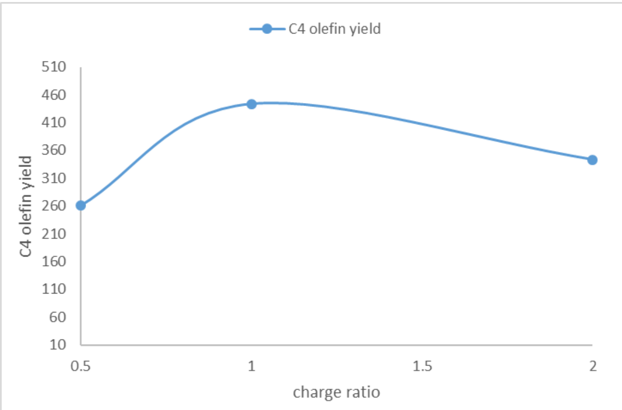

Figure 3: Variation of C4 olefin yield with charge ratio

The conversion of ethanol can be seen from Figure 2Y_1There is a negative linear correlation between C4 olefin selectivity and charge ratio KY_2It has a positive correlation with the charge ratio K, but the change is not obvious between k = 1 and K = 2.It can be seen from Figure 3 that the yield of C4 olefin reaches the maximum value near k = 1, and the difference is obvious compared with other charging ratios.A similar conclusion can be obtained when the temperature is other values.

In addition, it was noted that the a11 group did not add HAP, but replaced it with 90mg quartz sand, that is, SiO2. It is not difficult to observe this group of data, it is not difficult to find that the ethanol conversion and C4 olefin selectivity are very low, even at 350 ℃, only 8.2% and 4.35% respectively, while the acetaldehyde selectivity is as high as 85.83%.Compared with other groups, HAP is essential in the reaction, quartz sand can not be used as a substitute.

In conclusion, when the ratio of SiO2 to SiO2 is 1, the best yield is obtained.

ResearchY_1AndY_2Taking co loading and temperature as independent variables and C4 olefin yield as dependent variables, regression analysis was carried out by quadratic polynomial. The results showed that R^2= 0.95, P < 0.0001, and the fitting degree was high

It can be seen from the Figure 4 olefin yield that when the temperature reaches about 400 ℃, the predicted yield is about 39.9% when m = 1wt%, and about 40.5% when m = 2wt%. The latter is slightly higher, and the yield is much lower when m is other values.Therefore, it is considered that M = 1wt% and 2wt% have little effect on the yield at higher temperature.

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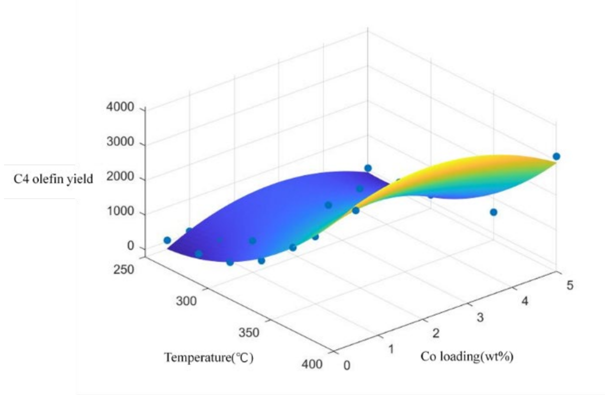

Figure 4: Variation of C4 olefin yield with temperature and co loading

Considering the other reaction products given in the data, when CO loading M = 2wt%, the conversion of ethanol is relatively high, but the selectivity of C4 olefins is low, and more ethanol is converted into 4-12 fatty alcohols.When the temperature is 350 ℃, the selectivity of 4-12 fatty alcohols with 2wt% Co loading in group A2 is as high as 36.92%, which is about four times of that in group A1, while the selectivity of ethylene and acetaldehyde is only 22% and 23% of that in group A1, respectively.According to the literature review, [3] ethylene and acetaldehyde are the intermediate products of ethanol coupling to produce C4 olefins. Although the C4 olefin yield of group A1 is slightly lower than that of group A2, a large number of intermediate products can still support the reaction to continue. Compared with the conditions of 2wt% Co loading, 1wt% Co loading conditions have greater advantages for the preparation of C4 olefins.

Based on the above analysis, it is considered that the reaction efficiency is relatively high and C4 olefin yield is better when the charge ratio of CO / SiO2 and HAP is 1:1 and the co loading is m = 1wt%.

3. Optimization of C4 olefin yield based on simulated annealing algorithm

In order to explore what kind of catalyst combination and temperature can improve the yield of C4 olefins as much as possible, the literature was searched and the data were analyzed.

With the reaction going on and the temperature rising, when the charge ratio is 1:1, the coupling of ethylene and acetaldehyde can be converted into C4 olefins, so C4 olefins have the highest selectivity. When the co loading is about 1wt%, the conversion of ethanol increases with the increase of temperature. Therefore, in order to maximize the yield of C4 olefins, a target optimization model based on simulated annealing algorithm is established.

A_1~ $a_{14}$and B_1~ B_7catalysts were selected, and the relative concentration of ethanol N and temperature T were determined when the loading ratio K was 1:1 and the loading amount of CO was 1 wt%Y_1And C4 olefin selectivityY_2Two binary cubic equations are obtained by fitting

$$Y_1 = A_1 + A_2T^3 + A_3N^3 + A_4T^2N + A_5N^2T + A_6T^2 + A_7N^2 + A_8TN + A_9T + A_{10}N \quad (1)$$

$$Y_2 = B_1 + B_2T^3 + B_3N^3 + B_4T^2N + B_5N^2T + B_6T^2 + B_7N^2 + B_8TN + B_9T + B_{10}N \quad (2)$$

Among them, a_1~ $a_{10}$and B_1~ B_{10}respectively represent the coefficients before the two equations, which can be solved by MATLAB.

It is known that the yield of C4 olefins is equal to the conversion rate of ethanol multiplied by the selectivity of C4 olefins. At the same time, combined with the constraints of temperature T and ethanol relative concentration n, the objective optimization model of yield is obtained

$$\begin{cases} f(T,N) = Y_1 * Y_2 \\ T \in [250,400] \\ N \in [1,26] \end{cases} \quad (3)$$

among$f(\mathrm{T},\mathrm{N})$The yield of C4 olefin is related to temperature and relative concentration of ethanol.

Considering that formula (1) and formula (2) are binary functions with the highest degree of cubic, the expression of the product yield will be a bivariate sixth power function. If we directly solve the maximum point, we need to determine all the extremum points, that is to find all the points whose first

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partial derivative is zero.For the complex equations which have not been solved by the above quadratic approximation formula, we can get the complex solution of the quadratic system. In addition, the calculation of this problem also involves the range of independent variables, and more equations need to be introduced to directly calculate the maximum value, which increases the difficulty of calculation. It can be seen that direct solution will bring huge amount of calculation and difficulty.

Therefore, this paper introduces simulated annealing algorithm to solve, in contrast, simulated annealing algorithm is a probability based algorithm, which is more suitable for solving the optimal solution problem. It comes from the principle of solid state annealing. Starting from a higher initial temperature, the temperature is controlled from high to low. At each temperature, n rounds of search are carried out. In each round of search, random disturbance is added to the old solution to generate a new solution, and the new solution is accepted according to certain rules.

The advantage of simulated annealing algorithm is that no matter how complex the function is, it is more likely to find the global optimal solution[5].For this problem, the bivariate sixth function must produce many maximum points and boundary value points, but in all the maximum points and boundary value points, the general algorithm is relatively difficult to do, simulated annealing algorithm can eliminate the local optimal solution, so as to find the best solution in the global, which is more consistent with the problem.[4]

According to the model in this question, assume that the initial temperature is 400 degrees, and the temperature attenuation coefficientα98, and the thermal equilibrium condition is that the temperature is less than 250 ℃, so the general steps of simulated annealing algorithm are as follows:

Step 1: according to the boundary conditions of the objective optimization model, the initial temperature of simulated annealing algorithm is set as 400 ℃, and the temperature attenuation coefficient is setα98, and the random perturbation function is calculated.[5]

Step2: get the equation by MATLABY_1、Y_2The coefficient matrix is obtained by the coefficient before each term. The coefficient matrix of C4 olefin yield equation in formula (3) is obtained by matrix transformation.

Step 3: the simulated annealing algorithm is used to solve one group of images, as shown in Fig. 5 and Fig. 6.In Fig. 5, the ordinate represents the temperature and the abscissa represents the relative concentration of ethanol. It can be seen from the figure that when the simulated annealing algorithm is iterated to 255.7432 degrees, the algorithm ends because the set boundary value condition is greater than or equal to 250 degrees.It can also be seen from the figure that there are many solutions when the temperature approaches 400 ℃ and the relative concentration of ethanol is about 1.5. Therefore, it can be judged that the optimal solution is in this range.In Figure 6, the ordinate represents the opposite number of the objective function value, and the abscissa represents the number of iterations. It can be seen from the figure that when the number of iterations is greater than 20, the simulated annealing algorithm gradually approaches the maximum value of the global objective function with the increase of the number of iterations.

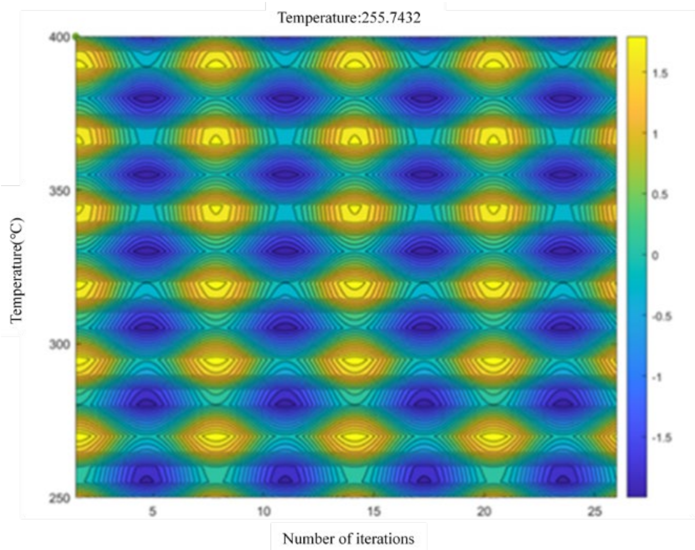

Figure 5: Diagram of simulated annealing algorithm seeking the optimal solution

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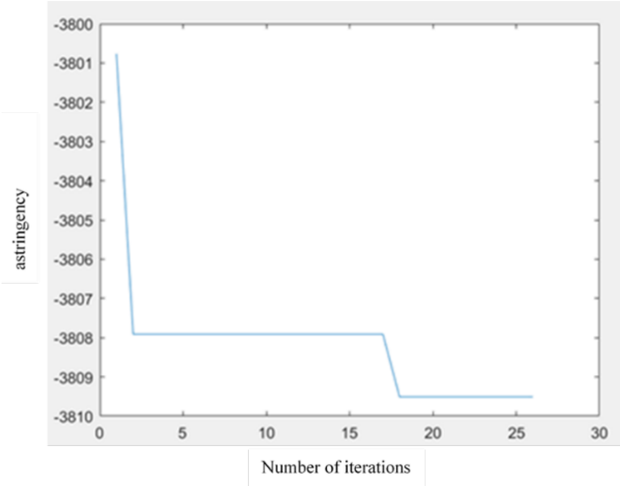

Figure 6: The number of iterations of simulated annealing algorithm

Based on the above analysis, the final simulated annealing algorithm was used to solve the target optimization model. The results showed that when the relative concentration of ethanol was 1.5052, the temperature was 399.9214 degrees, the loading amount of CO was 1wt%, and the loading ratio was 1:1, the maximum yield of C4 olefins was 38.09%. Therefore, the optimal catalyst combination was 200mg 1wt% Co / sio2-200mg HAP ethanol concentration of 0.76ml/min.

4. Conclusion

In order to optimize the preparation process of C4 olefins, the effects of various factors in catalyst on ethanol conversion and C4 olefin selectivity were studied by classifying the experimental data under the principle of variable control.Based on the model, an optimization model with C4 olefin yield as the objective function was established.In the process of solving, this paper chooses simulated annealing algorithm, which is easier to find the global optimal solution for this model.This model is not only used to explore the process conditions of ethanol catalytic coupling to produce C4 olefins, but also can be extended to the optimal process exploration model in the field of chemical production and automation.

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