

Retraction

Retracted: Research on Ethanol Coupling to Prepare C4 Olefins Based on BP Neural Network and Cluster Analysis

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This article has been retracted by Hindawi following an investigation undertaken by the publisher [1]. This investigation has uncovered evidence of one or more of the following indicators of systematic manipulation of the publication process:

- (1) Discrepancies in scope
- (2) Discrepancies in the description of the research reported
- (3) Discrepancies between the availability of data and the research described
- (4) Inappropriate citations
- (5) Incoherent, meaningless and/or irrelevant content included in the article
- (6) Peer-review manipulation

The presence of these indicators undermines our confidence in the integrity of the article's content and we cannot, therefore, vouch for its reliability. Please note that this notice is intended solely to alert readers that the content of this article is unreliable. We have not investigated whether authors were aware of or involved in the systematic manipulation of the publication process.

Wiley and Hindawi regrets that the usual quality checks did not identify these issues before publication and have since put additional measures in place to safeguard research integrity.

We wish to credit our own Research Integrity and Research Publishing teams and anonymous and named external researchers and research integrity experts for contributing to this investigation.

The corresponding author, as the representative of all authors, has been given the opportunity to register their agreement or disagreement to this retraction. We have kept a record of any response received.

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- [1] S. Zhang, W. Zhan, H. Hu, Y. Liu, and J. Zhu, "Research on Ethanol Coupling to Prepare C4 Olefins Based on BP Neural Network and Cluster Analysis," *Journal of Chemistry*, vol. 2022, Article ID 5324336, 10 pages, 2022.

Research Article

Research on Ethanol Coupling to Prepare C₄ Olefins Based on BP Neural Network and Cluster Analysis

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Ethanol, as a clean energy source, is an ideal raw material for the preparation of C₄ olefins, but there are few studies on the preparation of C₄ olefins by the coupling of ethanol. This research is based on the data from Question B of the 2021 Chinese Contemporary Undergraduate Mathematical Contest in Modeling. The researchers firstly perform data interpolation and visualization processing on the original data. Secondly, based on the two-dimensional visualization analysis, we cluster the different catalyst combinations; divide the influencing factors into temperature, Co/SiO₂ and HAP loading ratio, Co loading, and ethanol concentration; and construct a quaternary linear regression equation that affects ethanol conversion rate and C₄ olefin selectivity. Finally, according to the three-dimensional spatial visualization analysis and using the BP neural network model training data, we obtain that under the conditions of using loading method I, catalyst combination type 200 mg 0.5 wt% Co/SiO₂-200 mg HAP-ethanol concentration 0.9 ml/min and temperature 450°C, the yield of C₄ olefins can reach the maximum. This study provides new research ideas and methods for the preparation of C₄ olefins from ethanol.

1. Introduction

On September 11, 2020, General Secretary Xi Jinping mentioned at the symposium of scientists that China still has practical problems such as insufficient development of new energy technologies and excessive oil dependence. In order to implement General Secretary Xi Jinping's strategic thinking on sustainable development and respond to the "14th Five-Year Plan" strategic policy, China has changed the raw material for preparing C₄ olefins from traditionally used petroleum to ethanol. C₄ olefins are widely used in Chinese medicine and industry and are a very important chemical industry production material. In the process of preparing C₄ olefins from ethanol, studying the types of catalyst combinations, temperature changes, and other conditions is of vital significance and value for reducing energy waste and improving reaction effects and productivity.

In recent years, Chinese scholars have conducted various research and explorations on the preparation of C₄ olefins. Peng [1] adopted mixed C₄ alkane dehydrogenation technology to prepare C₄ olefins and used high-temperature solid phase diffusion method and impregnation method to prepare spent FCC catalyst and VO_x/Al₂O₃ catalyst, respectively. It is concluded that the best catalytic performance can be obtained when the VO_x loading in the VO_x/Al₂O₃ catalyst is 12%. Hu [2] used the impregnation method to prepare the supported catalysts and used characterization methods to evaluate certain properties of the prepared catalysts by constructing an evaluation model. He also analyzed the effects of loading and carrier properties on the catalytic process and explored the dehydrogenation effect and catalytic stability of the catalyst under different activity distribution ratios. Li [3] constructed two catalytic systems and analyzed the acid-base performance, surface characteristics, structure, and other aspects of the catalyst using various

technical means which reveals that the configuration of the *tert*-butyl carbocation intermediate determines the anti-*cis* ratio of the product 2-butene in the double-bond isomerization reaction of 1-butene.

At present, Chinese research on the preparation of C₄ olefins is mainly focused on the catalytic dehydrogenation of low-carbon alkanes, while the research on the preparation of C₄ olefins by ethanol coupling mostly stays at the level of experimental research. Based on the above content, this paper further applies mathematical methods to closely link chemical principles and experimental data and studies the reaction of ethanol as a raw material to prepare C₄ olefins from different angles and levels.

2. Basic Assumptions and Index Selection

2.1. Basic Assumptions. In order to facilitate the study of the problem, the following hypotheses are put forward:

- (i) The data given conforms to the laws of chemical experiments and is true and effective
- (ii) The influence of external factors such as indoor temperature changes and experimental equipment on the experimental data is not considered
- (iii) Ignore the influence of factors other than the type of catalyst combination and temperature on the reaction [4]

2.2. Index Selection and Description. The reaction of ethanol coupling to prepare C₄ olefins mainly involves the reactant ethanol, the main product C₄ olefins, the catalyst, and the temperature that has a significant impact on the experiment. After consulting a large amount of data and drawing on previous experience, we finally selected five types of indicators, including catalyst combination, temperature, ethanol conversion rate, C₄ olefin selectivity, and C₄ olefin yield [5], and made the following explanations:

- (i) Catalyst combination: including catalyst and reactant. A total of 21 types of catalyst combinations, of which A_{*i*} (*i* = 1, 2, ..., 14) catalyst combinations use charging method I and catalyst combinations B_{*j*} (*j* = 1, 2, ..., 7) use charging method II. The following are sorted by A_{*i*} = *i* and B_{*j*} = 14 + *j* from the smallest to the largest [6]
- (ii) Temperature: refers to the reaction temperature. The temperature data in the text refers to 250°C, 275°C, 300°C, 325°C, 350°C, 400°C, and 450°C
- (iii) Ethanol conversion rate: unit: %. Ethanol conversion rate = (ethanol intake - remaining amount of ethanol)/ethanol take × 100% [7]
- (iv) C₄ olefin selectivity: unit: %. The greater the selectivity is, the better the experimental results are
- (v) C₄ olefin yield: equal to ethanol conversion rate × C₄ olefin selectivity, combined with reactants and products to measure the experimental results [8]

- (vi) Loading method: comparing the catalyst combination data numbered A12 and B1, only the charging method is different where the experimental result of A12 is slightly better than that of B1. But the difference is not significant. Therefore, we ignore the influence of this factor and choose the loading method I first [9]

3. Data Preprocessing and Visualization

3.1. Interpolation of Data. There are differences in the data given. The maximum value of the temperature in the experimental data of catalyst combination numbers A1 and A2 is 350°C, while the maximum value of the temperature in the data of the rest of the experimental groups is all higher than 350°C. Among them, the maximum temperature of 18 groups is 400°C, and the maximum value of catalyst combination number A4 is 450°C [10]. For the convenience of follow-up research, spline interpolation [11] is used to fill in missing values, and the repaired interpolation results are shown in Table 1.

In Table 1, the data listed in italic are the repaired missing value, in which the ethanol conversion rate is reserved to 1 decimal place, and the C₄ olefin is selectively reserved to 2 decimal places. The rest are the original data, which serve as a comparison. It can be compared with the data of other temperatures under the same catalyst combination to intuitively judge the accuracy of the interpolation [12].

3.2. Data Visualization

3.2.1. Two-Dimensional Visualization. After interpolation, observe the numerical distribution of ethanol conversion rate and C₄ olefin selectivity for different catalyst combinations at the same temperature in which the temperature is selected as 250°C, 275°C, 300°C, 325°C, 350°C, and 400°C, as shown in Figures 1 and 2, respectively [13].

When the temperature is constant, the greater the value of the ethanol conversion rate and the C₄ olefin selectivity, the better the coupling reaction result, indicating that the catalyst combination effect used in the reaction is better [14]. It can be seen from Figure 1 that the catalysts numbered A1 to A7 have a significant combined effect. From Figure 2, it can be seen that the catalysts numbered A1 and A2 have a significant combined effect. Based on Figures 1 and 2, the reaction results at temperatures of 300°C, 325°C, 350°C, and 400°C are generally better than those at temperatures of 250°C, 275°C, and 300°C [15]. Further, study the reaction results of each catalyst combination at different temperatures. The reaction results are reflected by the ethanol conversion rate and C₄ olefin selectivity. Since there are 21 combinations in total, the representative catalyst combination numbered A7 is selected [16]. And the result is shown in Figure 3.

According to Figure 3, when the temperature is between 250°C and 400°C, both the ethanol conversion rate and the C₄ olefin selectivity are positively correlated with the temperature [17]. As the temperature rises, the values of ethanol conversion rate and C₄ olefin selectivity continue to increase.

TABLE 1: Spline interpolation to patch missing data.

Catalyst combination type	A1		A2		A6		A7		
Temperature (°C)	350	400	350	400	300	325	350	300	325
Ethanol conversion rate (%)	36.8	51.6	67.9	62.7	25.5	43.4	55.8	40	48.4
C ₄ olefin selectivity (%)	47.21	62.68	39.1	51.46	7.18	11.54	10.65	8.84	13.90
Catalyst combination type	A8		A9		A10				
Temperature (°C)	300	325	350	300	325	350	300	325	350
Ethanol conversion rate (%)	13.2	20.8	31.7	4.7	9.3	13.4	1.7	3.9	9.0
C ₄ olefin selectivity (%)	13.82	17.99	25.89	16.1	23.39	31.04	2.17	3.69	3.3
Catalyst combination type	A11		A12		A13				
Temperature (°C)	300	325	350	300	325	350	300	325	350
Ethanol conversion rate (%)	1.6	3.3	8.2	6.9	10.9	19.9	4.1	8.2	14.6
C ₄ olefin selectivity (%)	1.82	3.57	4.35	11.22	18.93	22.26	12.74	17.05	23.46
Catalyst combination type	A14		B1		B2				
Temperature (°C)	300	325	350	300	325	350	300	325	350
Ethanol conversion rate (%)	10.2	14.9	24	6.7	10.6	19.3	6.2	11.6	16.2
C ₄ olefin selectivity (%)	3.61	6.82	10.83	12.28	17.76	25.97	9.32	13.58	22.88

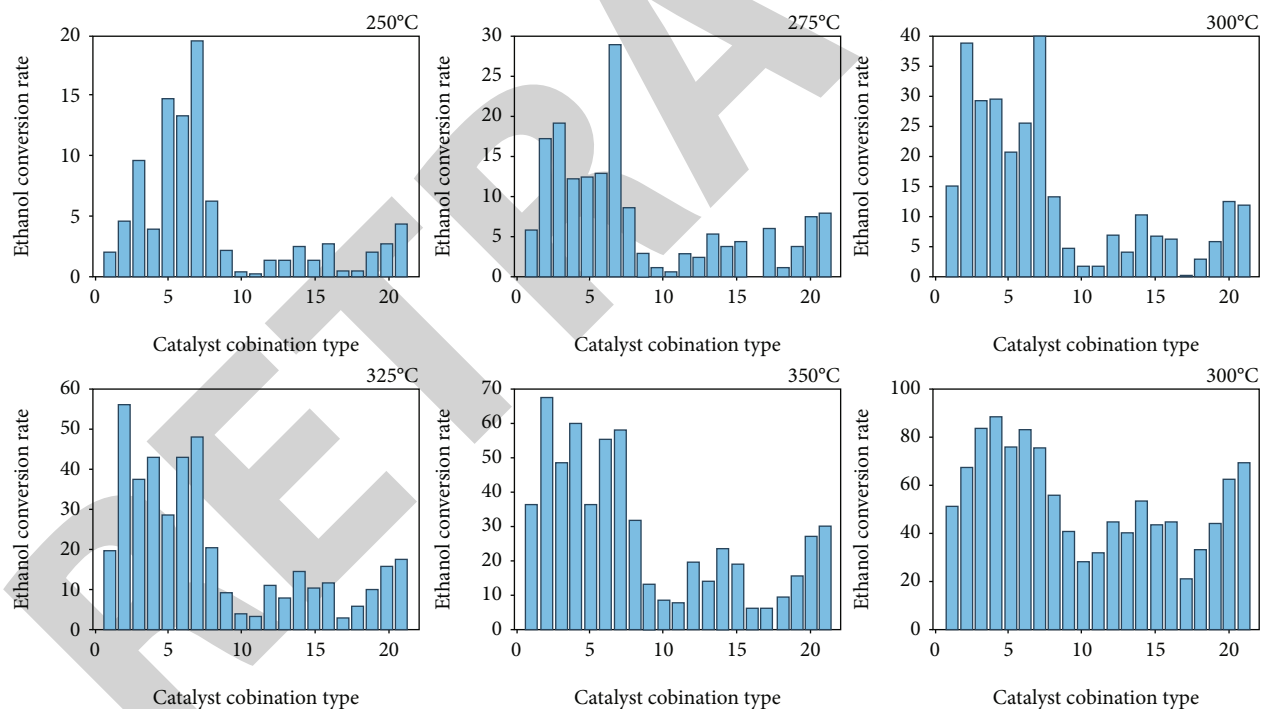


FIGURE 1: Ethanol conversion rate histogram.

That is, the catalyst group is controlled to remain unchanged, and within a certain temperature range, the reaction result becomes better as the temperature rises [18].

3.2.2. Three-Dimensional Visualization. The spatial distribution of ethanol conversion rate and C₄ olefin selectivity under different catalyst combinations and temperatures [19] are shown in Figures 4 and 5, respectively. It can be seen from the spatial distribution map that there are significant

differences in the spatial distribution of ethanol conversion rate and C₄ olefin selectivity [20].

4. Multiple Linear Regression Model Based on Systematic Clustering

According to the two-dimensional visualization processing, when the temperature is between 250°C and 400°C, both the ethanol conversion rate and the C₄ olefin selectivity

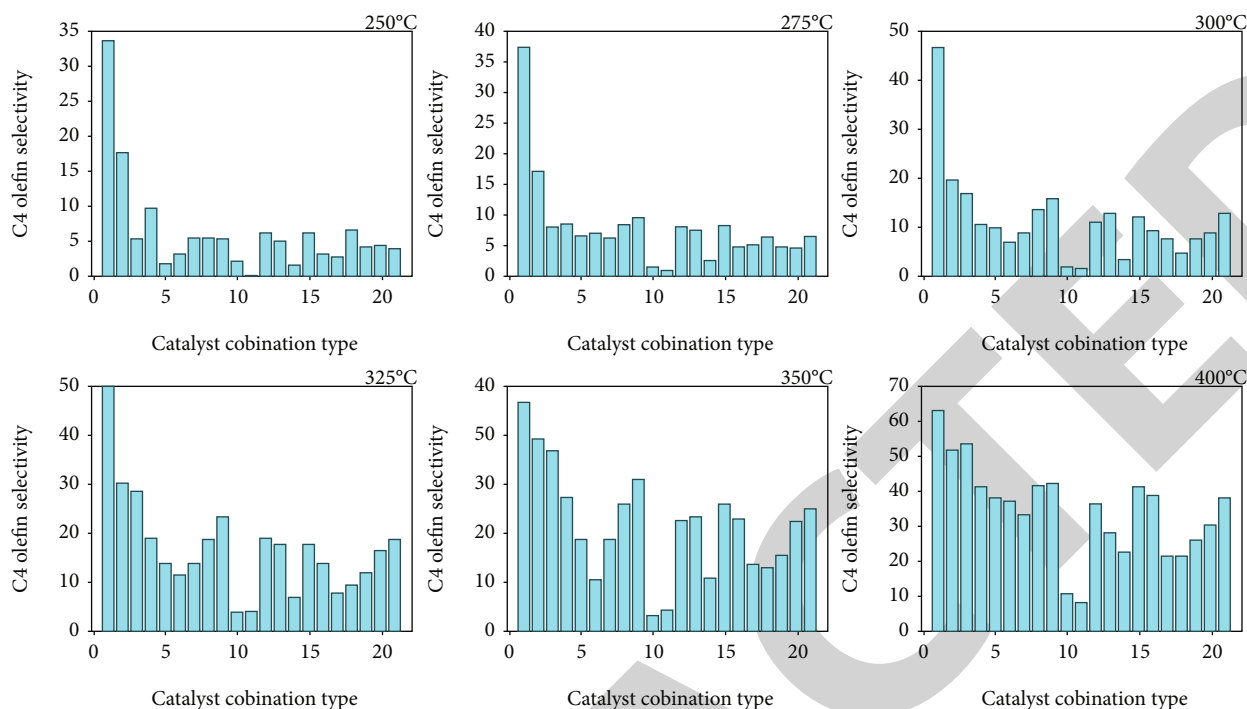


FIGURE 2: C_4 olefin selectivity histogram.

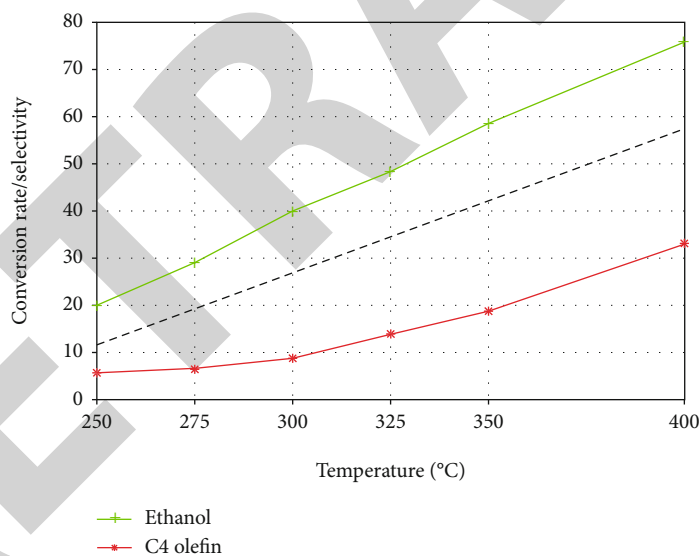


FIGURE 3: Spatial distribution of ethanol conversion rate.

are positively correlated with the temperature. As the temperature rises, the values of ethanol conversion rate and C_4 olefin selectivity continue to increase. That is, the catalyst group is controlled to remain unchanged, and within a certain temperature range, the reaction result becomes better as the temperature rises [21].

4.1. Model Establishment. Based on the above analysis, a cluster analysis was performed on the catalyst combination, and the reaction results were divided into four categories: “signifi-

cant,” “good,” “fair,” and “poor.” Hierarchical clustering method is used here. According to the distance between the two types of data, the systematic clustering method combines the closest two types of data and repeats the above operations repeatedly until all types of data are clustered into one type [22]. The specific process is as follows:

- (i) Divide the catalyst combinations into 21 categories, calculate the distance between each two categories, and find the smallest distance among them

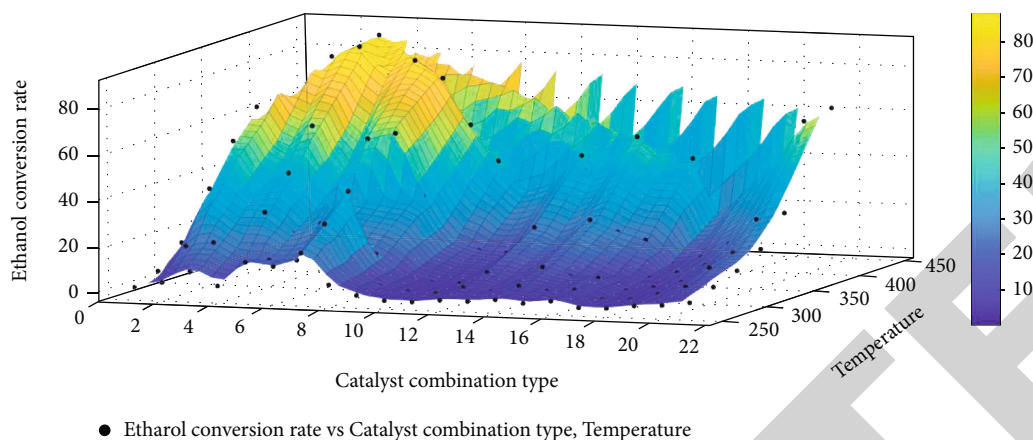
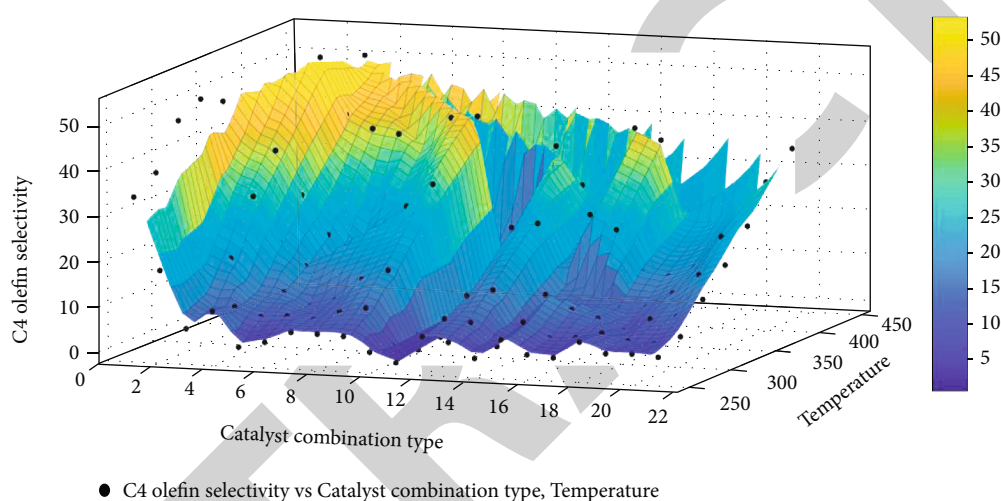


FIGURE 4: Spatial distribution of ethanol conversion rate.

FIGURE 5: Spatial distribution of C₄ olefin selectivity.

- (ii) Put the two catalyst combinations with the smallest distance into a new category, and let this new category and other catalyst combinations that are not in the new category form a new category group
- (iii) Calculate the distance between every two categories in the new classification group, and find the minimum distance
- (iv) Repeat steps (ii) and (iii), until finally, there is only one category left [23]

The ethanol conversion rate and C₄ olefin selectivity were clustered, respectively, and the reaction results of each catalyst combination were classified according to the obtained clustering pedigree [24]. Using the data under the catalyst group with “significant” reaction results, establish multiple linear regression equations that affect ethanol conversion and C₄ olefin selectivity:

$$y = a_1x_1 + a_2x_2 + \dots + a_nx_n + b. \quad (1)$$

Take the ethanol conversion rate as an example, where y is the ethanol conversion rate. x_i is the factor that affects the ethanol conversion rate ($i = 1, 2, \dots, n$). a_i is the influence coefficient of x_i on y . The larger the coefficient is, the greater the effect ($i = 1, 2, \dots, n$) is. b is a constant term, which is jointly determined by y , x_i , and a_i . n is the number of influencing factors [25].

4.2. Model Solution and Analysis. Using SPSS 25 software, the 21 types of catalyst combinations were systematically clustered according to the ethanol conversion rate, and the obtained results are shown in Figure 6.

It can be seen from the pedigree diagram of ethanol conversion rate that

- (i) the catalyst combinations that have “significant” effects on the ethanol conversion rate are A2, A4, A5, and A7
- (ii) the catalyst combinations that affect “good” are A1, A3, A6, A8, A14, B6, and B7

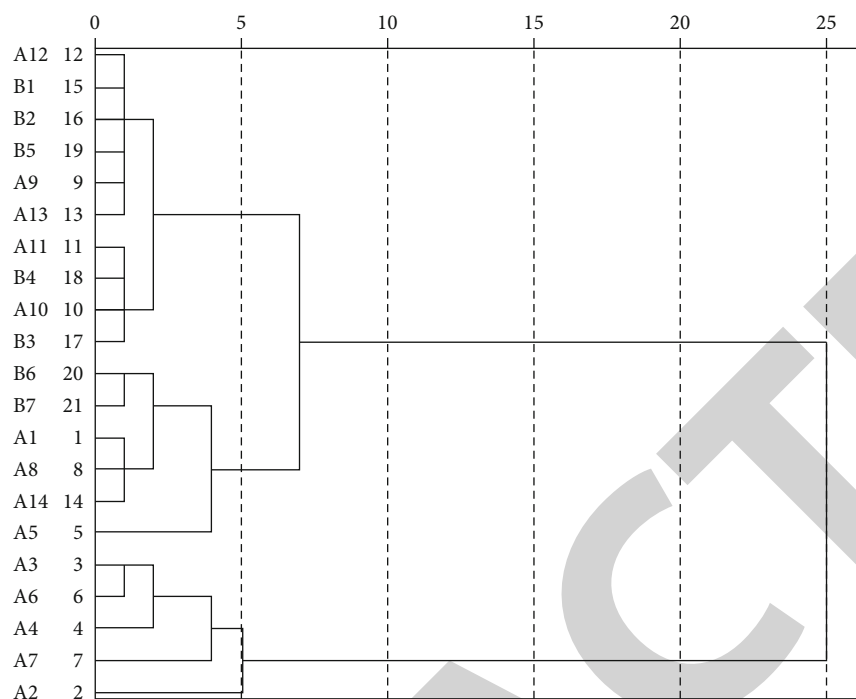


FIGURE 6: A pedigree of ethanol conversion rate using average linkage (among groups).

- (iii) the catalyst combinations that affect “fair” are numbered A9, A12, A13, B1, B2, and B5
- (iv) the catalyst combinations that affect “poor” are numbered A10, A11, B3, and B4 [26]

Similarly, using SPSS 25 software to do the same treatment for the C_4 olefin selectivity, the result is shown in Figure 7.

According to the C_4 olefin selectivity clustering pedigree diagram, it can be seen that

- (i) the catalyst combinations that “significantly” affect the C_4 olefin selectivity are A1 and A2
- (ii) the catalyst combinations that affect “good” are A3 and A9
- (iii) the catalyst combinations that affect “fair” are numbered A4, A5, A6, A7, A8, A12, A13, B1, B2, B6, and B7
- (iv) the catalyst combinations that affect the “poor” are numbered A10, A11, A14, B3, B4, and B5 [27]

In summary, for the ethanol conversion rate, the catalyst combination numbers A2, A4, A5, and A7 have “significant” effects. For C_4 olefin selectivity, the catalyst combination numbers A1 and A2 have “significant” effects. Analyze the composition of each catalyst combination, and finally, select temperature, Co/SiO₂ and HAP charging ratio (hereinafter referred to as “charge ratio”), Co loading, and ethanol concentration as the main influencing factors and establish a multiple regression equation [28].

Regarding the catalytic efficiency of ethanol, the catalyst combinations numbered A2, A4, A5, and A7 were selected to perform a significant test on the four parameters of temperature, charging ratio, Co loading, and ethanol concentration. Similarly, for the selectivity of C_4 olefins, the catalyst combinations numbered A1 and A2 were selected to test the significance of the four parameters [29]. The test results are shown in Table 2.

It can be seen from Table 2 that for the ethanol conversion rate, the temperature and the charging ratio are very significant, while the Co loading and the dripping rate are more significant. For C_4 olefin selectivity, temperature, charging ratio, and drip rate are very significant, while Co loading is more significant. Temperature, charging ratio, Co loading, and dripping rate can all be used as influencing factors for the study of ethanol conversion rate and C_4 olefin selectivity.

Using Stata software [30], select the catalyst combination data numbered A2, A4, A5, and A7 to perform multiple linear regression on the ethanol conversion rate to obtain the regression equation

$$y_1 = 0.4271x_1 + 0.1020x_2 + 0.6048x_3 - 1.2097x_4 - 118.5026. \quad (2)$$

Similarly, select the catalyst combination data numbered A1 and A2 to perform multiple linear regression on the selectivity of C_4 olefins to obtain the regression equation

$$y_2 = 0.2432x_1 + 0.2259x_2 - 18.132x_3 + 29.8192x_4 - 113.0406, \quad (3)$$

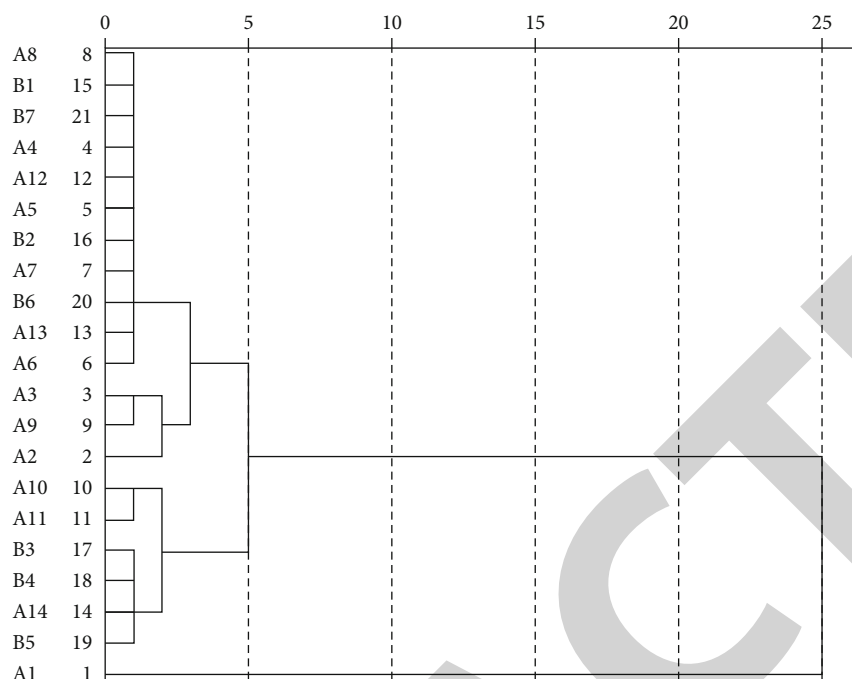


FIGURE 7: A pedigree of C_4 olefin selectivity using average linkage (among groups).

TABLE 2: The results of the significance test of each parameter.

Parameter	Temperature	Charge ratio	Co loading	Ethanol concentration
Ethanol conversion rate	$\leq 0.001^{***}$	$\leq 0.001^{***}$	0.008**	0.007**
C_4 olefin selectivity	$\leq 0.001^{***}$	$\leq 0.001^{***}$	0.002**	$\leq 0.001^{***}$

Note: *, **, and *** represent significant, relatively significant, and very significant, respectively.

where x_1 is the temperature, and the temperature range is 250°C-450°C. x_2 is the charging ratio, and the charging ratio range is 10 mg-200 mg/10 mg-200 mg. x_3 is the Co loading, and the Co loading range is 0.5 wt and 5 wt. x_4 is the concentration of ethanol, and the range of ethanol concentration is 0.3 ml/min-2.1 ml/min [31].

5. Exploration on the Optimal Yield of C_4 Olefins Based on BP Neural Network

According to the three-dimensional visualization processing, there are obvious differences in the ethanol conversion rate and the spatial distribution of the C_4 olefin selectivity under the catalyst combination and temperature [32]. Within a given catalyst group and temperature range, we can find a certain catalyst combination and temperature that maximizes the product of the corresponding ethanol conversion rate and C_4 olefin selectivity. Because the yield of C_4 olefins is determined by the conversion of ethanol and the selectivity of C_4 olefins, the catalyst combination and temperature found above are the conditions for maximizing the yield of C_4 olefins [33].

5.1. Research Ideas. The BP neural network is trained according to the error back propagation algorithm and is mainly used in function approximation, pattern recognition,

classification, data compression, etc. and is currently one of the most widely used neural network models [34].

The catalyst group in the form of charging ratio, Co loading, ethanol concentration, temperature, ethanol conversion rate, and C_4 olefin selectivity were used as indicators to study the yield of C_4 olefins [35]. For the processed data, select 70% of it as the training set and input it into the neural network model to train the model. Use 15% of the data as a validation set to test the accuracy of the model. Select the remaining 15% of the data as the test set, and constantly adjust the number of hidden layers to achieve the best learning effect [36].

Use the constructed BP neural network model to complete the missing temperature data under the existing catalyst combination. For example, the catalyst combination temperature numbered A1 is only 250°C, 275°C, 300°C, 325°C, and 350°C, and the filling temperature is 251°C, 252°C, ..., 349°C. Reuse the existing charging ratio, Co loading, and ethanol concentration to quantify to construct a new catalyst combination. The lowest temperature of the control experiment is 250°C, and the highest temperature is 450°C [37]. The catalyst combination and temperature with the largest product of ethanol conversion rate and C_4 olefin selectivity were selected as the optimal solution of the model [38].

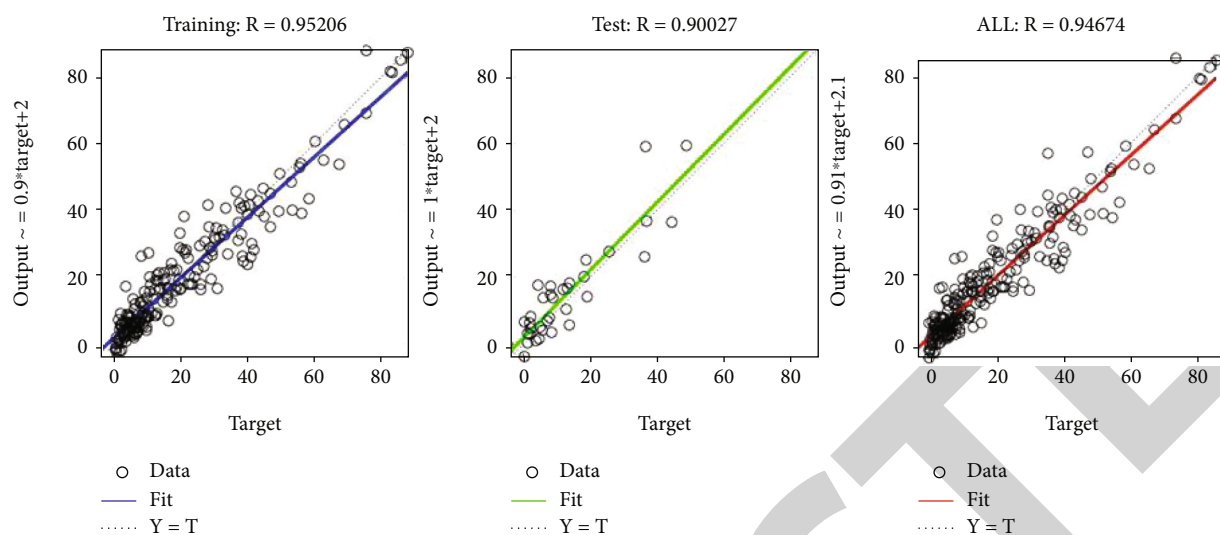


FIGURE 8: BP neural network fitting effect display.

5.2. *Empirical Analysis.* After repeated debugging, when the hidden layer of neurons is 15 layers, the fitting effect is better, and the result is shown in Figure 8.

It can be seen from Figure 8 that R^2 of each set is very close to 1 [39]. Under these conditions, the conditions for achieving the maximum yield of C_4 olefins are to use the loading method I, the catalyst type is 200 mg 0.5 wt% Co/SiO₂-200 mg HAP-ethanol concentration 0.9 ml/min, the temperature is 450°C, and the C_4 olefin yield is 48% [40].

6. Conclusion

Based on software such as SPSS 25 and Stata, this article selects Annex 1 of the 2021 Chinese Contemporary Undergraduate Mathematical Contest in Modeling as the research data [41]. According to the reaction process of ethanol coupling to produce C_4 olefins, the research content is divided into two parts. One is to study the factors affecting the ethanol conversion rate and the selectivity of C_4 olefins, and the other is to study the experimental conditions for obtaining the maximum yield of C_4 olefins [42].

For the former, we study the ethanol conversion rate and C_4 olefin selectivity, respectively, and use the ethanol conversion rate as an example to illustrate. First, through cluster analysis, class, and process all catalyst combinations, select the groups with “significant” impact on the ethanol conversion rate [43]. Then, on the basis of the experimental data of the “significant” catalyst groups, select temperature, charging ratio, Co loading, and ethanol concentration as the main influencing factors; carry out the significance test of these four parameters; and screen for significant factors. Finally, with the ethanol conversion rate as the dependent variable and the significant factor as the independent variable, we establish a multiple linear regression model based on cluster analysis [44].

For the latter, we fill in the missing temperature data, add new catalyst groups, and build an optimal model for the yield of C_4 olefins based on BP neural network. Then,

we select the group with the largest product of ethanol conversion rate and C_4 olefin as the condition with the largest C_4 olefin yield [45].

Data Availability

The data in this paper come from Question B of the 2021 Chinese Contemporary Undergraduate Mathematical Contest in Modeling.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

Authors' Contributions

Sheng-Mei Zhang was responsible for the methodology, conceptualization, supervision, and leadership. Wen-Li Zhan was responsible for the conceptualization, visualization, software, validation; and writing the manuscript. Han Hu was responsible for the software, method design, validation, and data analysis. Yu-Shu Liu was responsible for the data collation, visualization, verification, and investigation. Jia-Ming Zhu was responsible for the verification, supervision, writing, reviewing, and editing.

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