Academic Journal of Materials & Chemistry
ISSN 2616-5880 Vol. 3, Issue 1: 31-37, DOI: 10.25236/AJMC.2022.030106

Research and application of ethanol coupling to C_4 olefins based on BP neural network algorithm

Jianxin Liang*

School of Water Conservancy & Civil Engineering, Northeast Agricultural University, Heilongjiang, Harbin, 150000, China
a13190513@neau.edu.cn

Abstract: *In order to explore the influence of catalyst combination on the preparation of C_4 olefins by ethanol catalytic coupling, this paper first studied each catalyst by polynomial regression method to analyze the relationship between ethanol conversion, C_4 olefin selectivity and temperature. Secondly, BP neural network algorithm is used to select the best catalyst combination to obtain the highest C_4 olefin yield. The results show that: 1. Most ethanol conversion and C_4 olefin selectivity are directly proportional to temperature. 2. Without considering the temperature, when Co/SiO_2 is 3%, the temperature is 400 ℃, Co / SiO_2 and HAP are 200mg, The C_4 yield is the best (47.23%). When the set temperature is lower than 350 ℃ and the temperature is 325 ℃, Co/SiO_2 and HAP are 266mg and 133mg respectively, the maximum yield is 18.10%.*

Keywords: *ethanol preparation C_4 olefin; polynomial regression method; BP neural network algorithm*

1. Introduction

C_4 olefins are widely used in the production of chemical products and medicine. Ethanol is the raw material for production of C_4 olefins. In the preparation process, the catalyst combination (the combination of Co loading, Co / SiO_2 and HAP loading ratio, ethanol concentration) and temperature will have an impact on the selectivity and yield of C_4 olefins. Therefore, it is of great significance and value to explore the process conditions for the preparation of C_4 olefins by ethanol catalytic coupling through the combination design of catalysts.

Based on the experimental data given by the national college students mathematical modeling competition of China 2021 Higher Education Society cup [1], the relationship between ethanol conversion, C_4 olefin selectivity and temperature was studied by polynomial regression method with each catalyst combination. In addition, on this basis, BP neural network algorithm is used to select the catalyst combination and temperature to make the C_4 olefin yield as high as possible under the same experimental conditions.

2. Model establishment and solution

2.1 Relationship between ethanol conversion and temperature

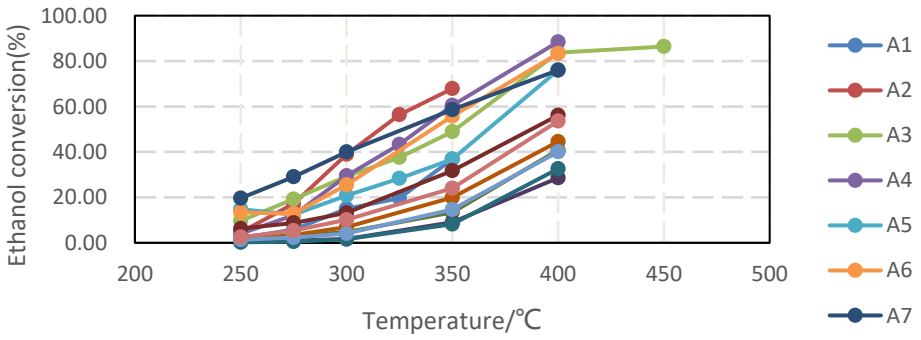


Figure 1: Broken line diagram of temperature-ethanol conversion under A loading mode

ISSN 2616-5880 Vol. 3, Issue 1: 31-37, DOI: 10.25236/AJMC.2022.030106

Visualize 21 groups of data of ethanol conversion and temperature in the given data [1], and connect the scatter diagram with broken lines, as shown in Fig. 1 and Fig. 2. Among them, A1-A14 in Fig. 1 represents 14 samples under type a loading mode. Similarly, B1-B7 in Fig. 2 represents the data under B loading modes. In terms of trend, most of the 21 groups of data show an "upper concave" shape. With the increase of temperature, the slope of ethanol conversion slows down, such as A2 and A3.

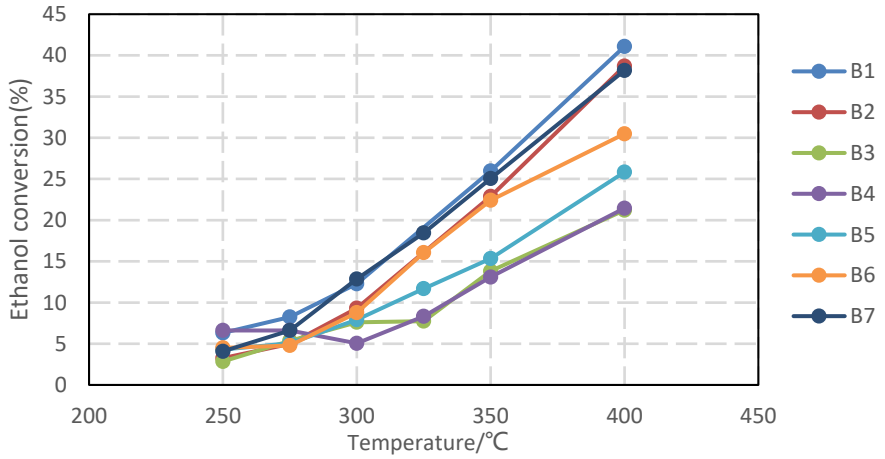
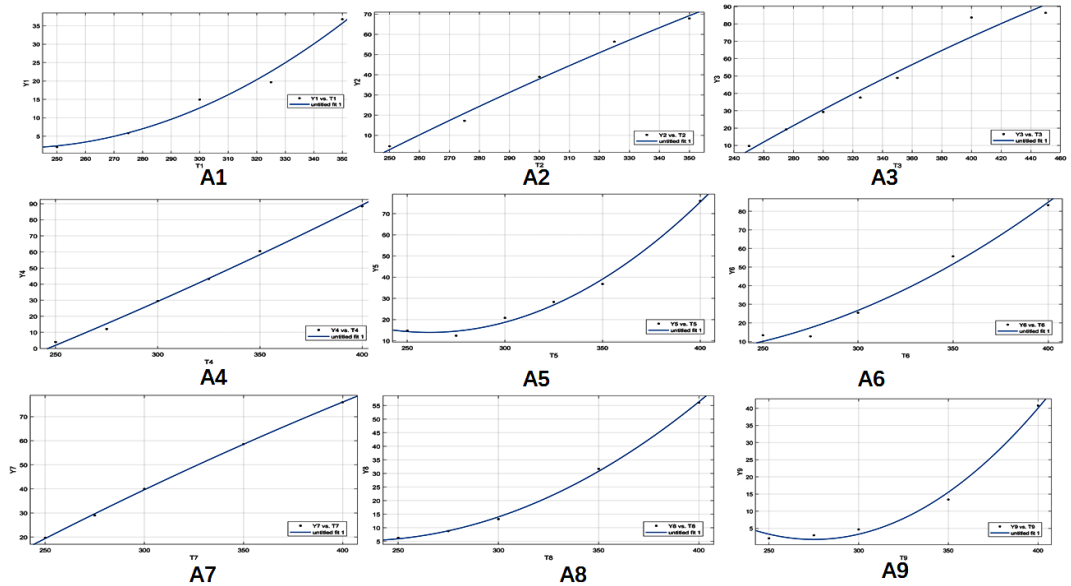


Figure 2: Broken line diagram of temperature-ethanol conversion under B loading mode

In view of the arrangement law of scattered points, this paper uses the least square method and polynomial fitting, with the order of 2. Its basic form is as follows [2]:

$$y = p_1 x^2 + p_2 x + p_3 + \varepsilon \tag{1}$$

21 groups of data are fitted by MATLAB. The fitting curve is shown in Fig. 3, and the fitting equation coefficients are shown in Table 1. The $\overline{R^2}$ of 21 groups of curves are greater than 0.95, indicating that the overall fitting representativeness of the equation is high and has excellent reliability. At the same time, observing 21 groups of images, it is found that the fitted images can be roughly divided into gentle rising type and 'upward concave' type with increased slope. The gently rising curve shows that the yield is not elastic by temperature change, such as A2, A3, A4 and A7. For the rising curve with increasing slope, the yield increases greatly with the increase of temperature, and obvious samples such as A9, A10, B3 and B4.



ISSN 2616-5880 Vol. 3, Issue 1: 31-37, DOI: 10.25236/AJMC.2022.030106

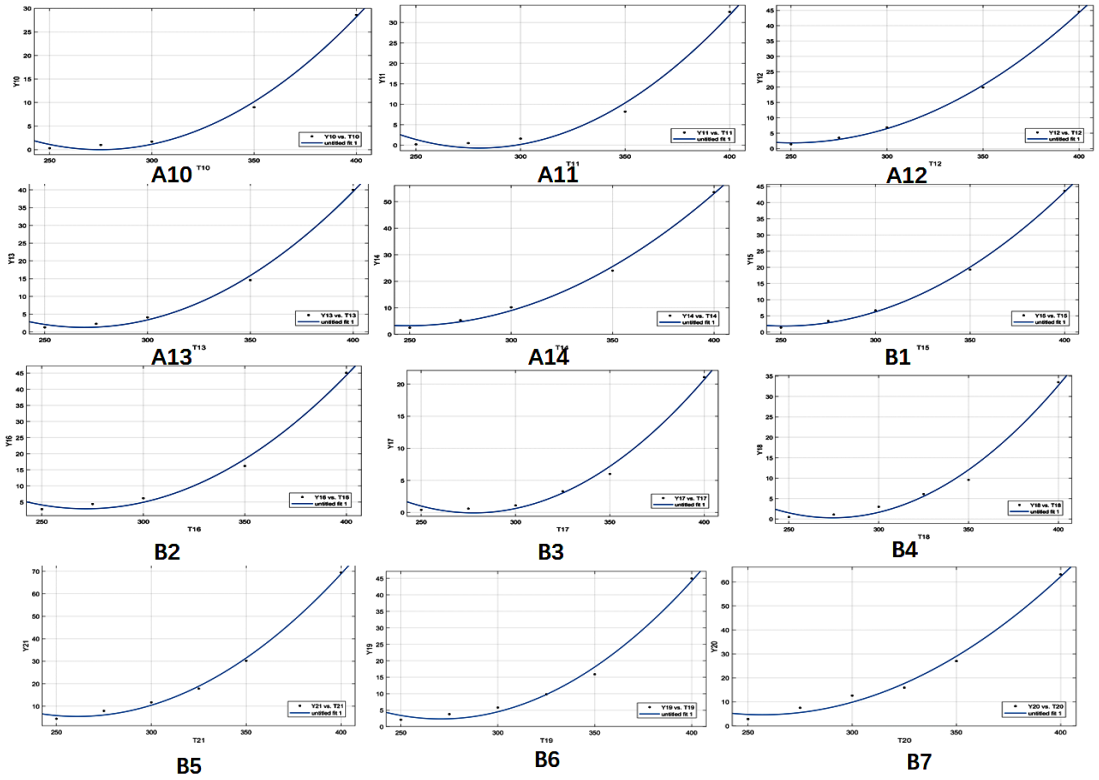


Figure 3: 21 groups of fitting graphs of ethanol conversion

Table 1: Equation coefficients of ethanol conversion and temperature fitting curve

Catalyzer	p1	p2	p3	Adjusted R-square
A1	0.002545	-1.194	141.8	0.9595
A2	-0.0007383	1.106	-227.4	0.9822
...	...	...	...	...
A14	0.002176	-1.083	137.9	0.9942
B1	0.001887	-0.9503	121.5	0.9978
B2	0.002514	-1.366	188.5	0.983
...	...	...	...	...
B7	0.003278	-1.711	228.7	0.9943

In order to further confirm the relationship between ethanol conversion and temperature, it can be roughly divided into the above two categories. In this paper, the quadratic term coefficient p1 and the primary term coefficient p2 which can reflect the shape of the graph line in 21 groups of data are K-means clustered by MATLAB. The main idea of K-means algorithm is as follows [3]:

$$J = \sum_{i=1}^{c} J_i = \sum_{i=1}^{c} \left(\sum_{k, x_k \in G_i} d(x_k - c_i)^2 \right) \tag{2}$$

$$c_i = \frac{1}{|G_i|} \sum_{k, x_k \in G_i} x_k \tag{3}$$

Iterate the adjusted clustering center to the objective function formula. If the objective function does not change, the iteration ends. The clustering results are shown in Fig. 4. Fig. 4 (a) shows the sample points arranged by the selected indicators, Fig. 4 (b) shows the clustering results, which can be divided into red and blue, and Fig. 4 (c) shows the iterative convergence process. Consistent with the above observations, samples A2, A3, A4 and A7 belong to category 1, and the rest can be divided into category 2.

ISSN 2616-5880 Vol. 3, Issue 1: 31-37, DOI: 10.25236/AJMC.2022.030106

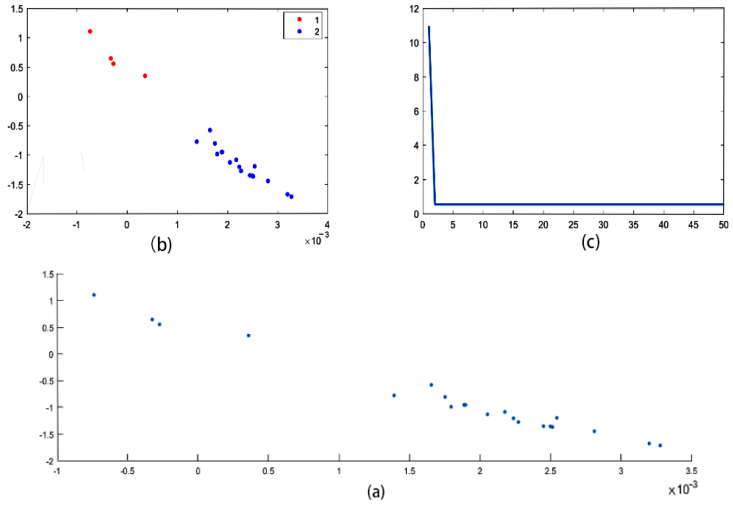


Figure 4: Clustering results of ethanol conversion

Table 2: Clustering results

Sample	A1	A2	A3	A4	A5	A6	A7
Category	2	1	1	1	2	2	1
Sample	A8	A9	A10	A11	A12	A13	A14
Category	2	2	2	2	2	2	2
Sample	B1	B2	B3	B4	B5	B6	B7
Category	2	2	2	2	2	2	2

2.2 Relationship between C4 olefin selectivity and temperature

As in subsection 2.1, Fig. 5 and Fig. 6 show the experimental broken line diagram of 21 groups of samples. Most samples show that the C_4 selection rate increases with the increase of temperature, but the selectivity of A1, A3, A5 and B6 decreases at the end of the experiment. B4 is the lowest point of C_4 selectivity at 300 ℃.

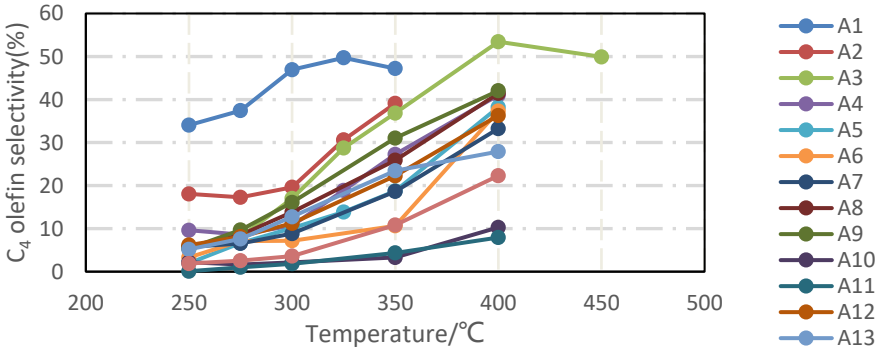


Figure 5: Broken line diagram of temperature—C4 olefin selectivity under A loading mode

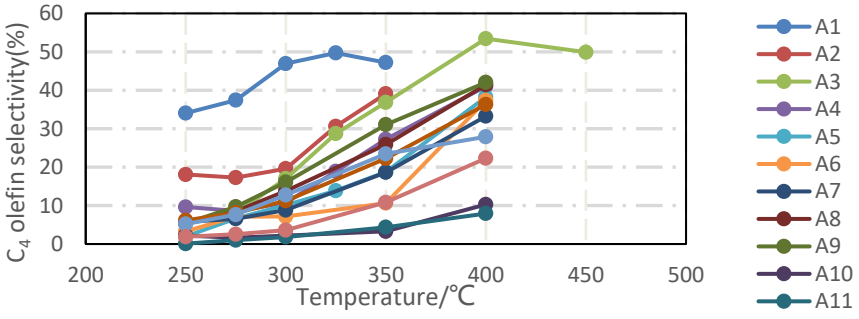


Figure 6: Broken line diagram of temperature—C4 olefin selectivity under B loading mode

ISSN 2616-5880 Vol. 3, Issue 1: 31-37, DOI: 10.25236/AJMC.2022.030106

Table 3: Selectivity and temperature equation coefficients of C4 olefins

Catalyzer	p1	p2	p3	Adjusted R-square
A1	-0.002113	1.422	-190.8	0.8319
A2	0.003113	-1.646	234.7	0.9606
...	...	...	...	...
A14	0.0009691	-0.494	64.86	0.9989
B1	0.0002179	-0.1184	16.19	0.9782
B2	0.0005404	-0.3021	42.27	0.9641
...	...	...	...	...
B7	0.00004438	-0.01145	0.05679	0.8708

Table 4: Clustering results

Sample	**A1**	**A2**	**A3**	**A4**	**A5**	**A6**	**A7**
Category	1	2	1	2	2	2	2
Sample	**A8**	**A9**	**A10**	**A11**	**A12**	**A13**	**A14**
Category	2	1	2	1	2	1	2
Sample	**B1**	**B2**	**B3**	**B4**	**B5**	**B6**	**B7**
Category	1	2	1	1	1	1	1

As shown in Fig. 7, the data of each group show obvious concave convex difference on the fitted image. Table 3 shows the coefficients of the fitting equation. The fitting curve can be divided into two categories according to the concavity and convexity. The selective rising speed of C_4 decreases with the increase of temperature, while the concave curve still increases after 400 ℃ with the increase of temperature.

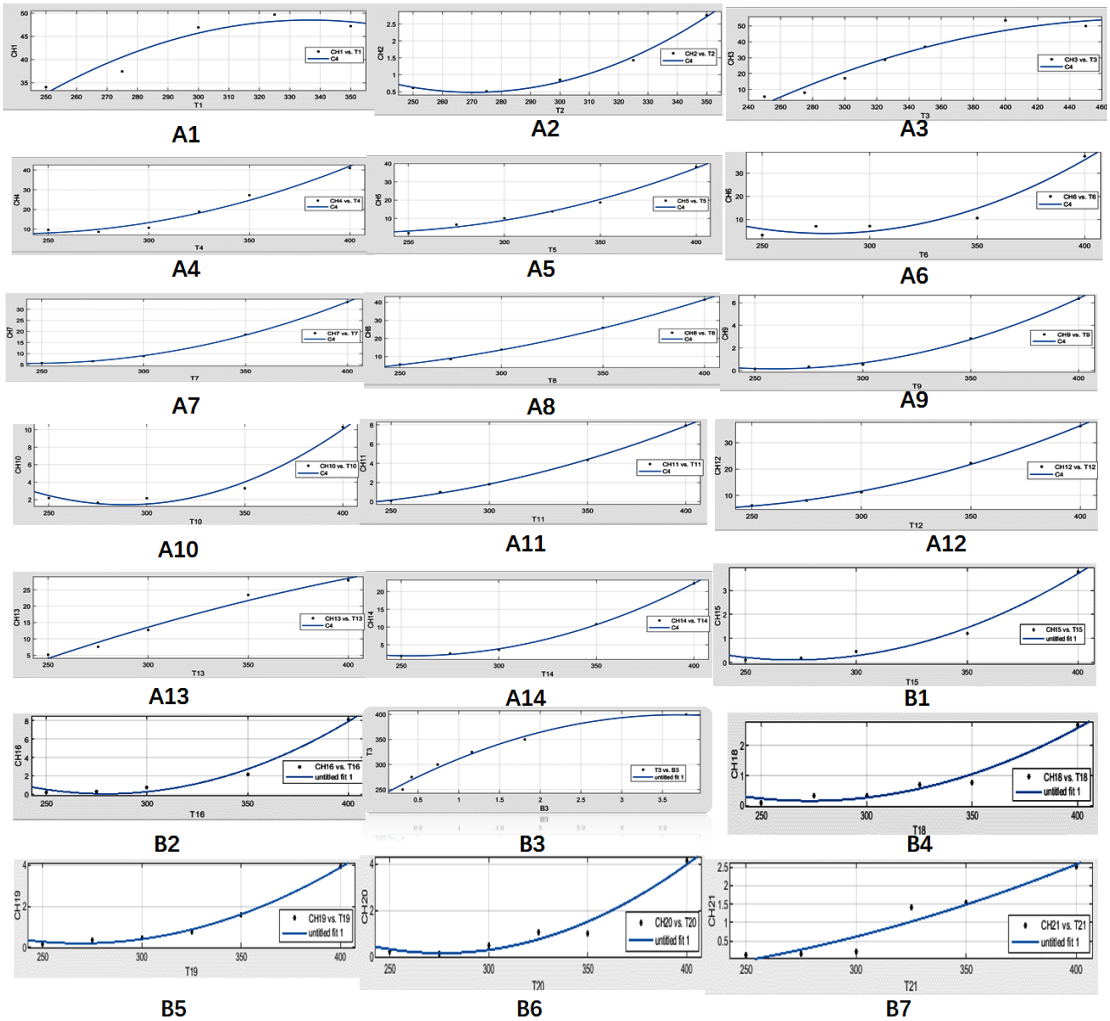


Figure 7: 21 groups of fitting graphs of C4 olefin selectivity

ISSN 2616-5880 Vol. 3, Issue 1: 31-37, DOI: 10.25236/AJMC.2022.030106

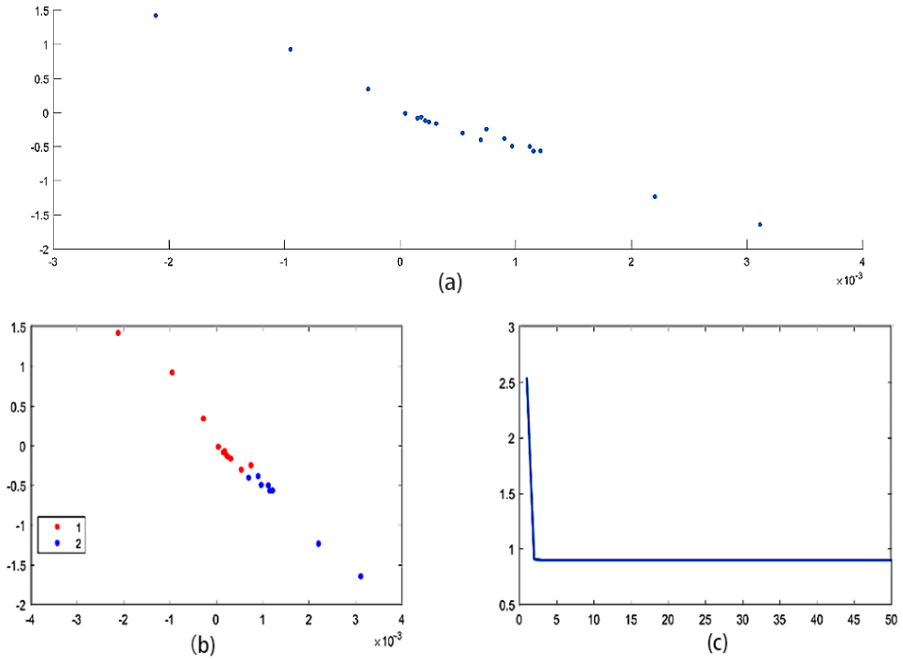


Figure 8: C4 olefin selectivity

2.3 Determination of the best catalyst combination based on BP neural network

2.3.1 Develop test sets

The ratio and quality of catalyst and the concentration of ethanol at different temperatures have different effects on the reaction products, which is difficult to study. In view of this, BP neural network is used to predict the experimental results. As for the optimum reaction conditions, Lv et al. points out through experiments that the common ratios of Co/SiO_2 and HAP are 1:1, 1:2 and 2:1[4]. Among them, the ratio of 1:1 can obtain the highest C_4 yield, while the effects of 1:2 and 2:1 on C_4 yield are similar, which can be regarded as a situation. Therefore, in the neural network test set, a total of 38 sets of test sets with a ratio of Co/SiO_2 to HAP of 1:1 and 1:2 and a total mass of no more than 400mg under various temperatures (250-400℃) are additionally formulated. In addition, in order to facilitate data processing, the ethanol concentration is taken as 1.68ml/min.

2.3.2 Forecast

BP neural network prediction formula [5]:

$$\begin{aligned} f_t &= relu\left((W_f * X) + b_0\right) \\ f_t &= relu\left((W_i * f_t) + b_0\right) \\ output &+ (W_{output} * i_t) + b_{output} \end{aligned} \tag{4}$$

f_t is the output of the first hidden layer; i_t is the output of the second hidden layer; Output is the output value; W_f is the weight matrix of the current layer; b_0 is the offset top of the current layer; $relu()$ is the relu activation function.

The BP neural network toolbox is used to train 149 groups of sample data. Without considering the temperature, when Co/SiO_2 = 3%, the temperature is 400 ℃, Co / SiO_2 and HAP are 200mg, the C_4 yield is the best (47.23%). When the set temperature is lower than 350 ℃ and the temperature is 325 ℃, Co/SiO_2 and HAP are 266mg and 133mg respectively, the maximum yield is 18.10%.

3. Promotion and evaluation of the model

The effect of polynomial regression is better, and the curve is classified by clustering algorithm, with clear hierarchy. Because the proportion and type of each catalyst are different at different temperatures,

ISSN 2616-5880 Vol. 3, Issue 1: 31-37, DOI: 10.25236/AJMC.2022.030106

it belongs to a nonlinear problem, and the three-layer neural network can approximate the nonlinear continuous function with arbitrary accuracy, which is suitable for such nonlinear and highly coupled data. However, polynomial regression belongs to the generalization of the least square method, and its use should be based on large number of samples ($n \geq 30$).

Through reasonable prediction and analysis, this model can also provide ideas and direction for the next test, and is of great significance to reduce the test cost and improve the purity of the product. If a new catalyst or catalyst support is found in the future, if you want to study its utility, you can apply this model to analyze the experimental results quickly.

References

[1] Chinese society of industrial and applied mathematics. 2021. Higher Education Society Cup National College Students' mathematical modeling competition [EB / OL]. (2021-09-09)[2021-10-05]. http://www.mcm.edu.cn/html_cn/node/4d73a36cc88b35bd4883c276afe39d89.html

[2] Gong Xinya. Study on estimation of landslide movement information based on polynomial fitting [J]. Construction technology development, 2020,47 (19): 155-157

[3] Li Cuiling, Li Yukang, Tan Haoran, Wang Xiu, Zhai Changchang. Detection and classification method of grape downy mildew based on K-means clustering and random forest [J / OL]. Journal of agricultural machinery: 1-14. [2022-04-20]

[4] Lv Shaopei. Preparation of butanol and C by ethanol coupling_ 4 olefins [D]. Tutor: Lu Anhui Dalian University of technology, 2018

[5] Lan Xueping, Chen Jinying, Jiang Youjun, Zou Xiao, Chen Meng. Construction of grain storage quality prediction model based on BP neural network algorithm [J]. Chinese Journal of grain and oil, 2020,35 (11): 147-151