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Research on Preparation of C4 Olefins by Ethanol Coupling

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Abstract: The preparation of C4 olefins by ethanol coupling is a very important process in chemical production. In this paper, the Hermite interpolation method is used to complete the missing data when the temperature is 400 ℃, and a grey correlation model is established to calculate the correlation degree between CO loading, Co / SiO2 and HAP loading ratio, ethanol concentration and temperature and ethanol conversion and the selectivity of C4 olefins in different catalyst combinations. Then, the weights of different catalyst combinations and temperatures were obtained by using each correlation degree. Finally, the BP neural network algorithm model is used to obtain the optimal combination of temperature 402 ℃, catalyst combination 200mg 1wt% Co / SiO2 - 200mg HAP ethanol concentration 0.9ml/min, adding the constraint condition that the temperature is less than 350 ℃, and the temperature is 343 ℃, catalyst combination 200mg 2wt% Co / SiO2 - 200mg HAP ethanol concentration 1.68ml/min.

1. Introduction

Ethanol is the raw material for the production of C4 olefins, which are widely used in the production of chemical products and medicine. In the preparation process, different catalyst combinations and temperatures will have an impact on the selectivity and yield of C4 olefins. It is of great significance and value to explore the process conditions for the preparation of C4 olefins by ethanol catalytic coupling by adjusting the ratio of different catalysts. Experiments were carried out at different catalyst ratios and temperatures in a chemical laboratory.

2. Grey System Model

2.1 Model introduction

Grey system theory is a system science theory initiated by Professor Deng Julong, a famous scholar. The grey correlation analysis is grey correlation analysis (GRA), which is a multi factor statistical analysis method. In short, in a grey system, we want to know the relative strength of a project we are concerned about affected by other factors. To be more straightforward, that is, if we assume and know that an indicator may be related to several other factors, we want to know which other factors are more related to this indicator, And which factors have a weaker relative

relationship, and so on. Arrange these factors in order to get an analysis result, so that we can know which of the factors we pay attention to are more relevant.

2.2 Data preprocessing

After observing the temperature data in Annex 1, it was found that the experimental results at 400℃ were missing only in the two catalyst combinations A1 (200mg 1wt% Co / SiO2 - 200mg HAP ethanol concentration 1.68ml/min) and A2 (200mg 2wt% Co / SiO2 - 200mg HAP ethanol concentration 1.68ml/min), so we used Hermite three times The interpolation pair is preprocessed to complete the data of 400℃ data group.

The ethylene conversion (%) of A1 catalyst combination (200mg 1wt% Co / SiO2 - 200mg HAP ethanol concentration 1.68ml/min) is 65.94%, the C4 olefin selectivity (%) is 24.86%, and the ethylene conversion (%) of A2 catalyst combination (200mg 2wt% Co / SiO2 - 200mg HAP ethanol concentration 1.68ml/min) is 65.64% and the C4 olefin selectivity (%) is 46.45% at 400℃.

2.3 Establishment of model

According to the catalyst combinations with different ratios given in Annex I, the analysis series of ethanol conversion rate, C4 olefin selectivity and various factors of the catalyst are determined respectively. The results are as follows:

Table 1: Analysis number list of ethanol conversion and various factors of catalyst.

Parent sequence	Subsequence 1	Subsequence 2	Subsequence 3	Subsequence 4
Ethanol conversion (%)	charge ratio	Co load	concentration	temperature

Table 2: Analysis number list of C4 olefin selectivity and various factors of catalyst.

Parent sequence	Subsequence 1	Subsequence 2	Subsequence 3	Subsequence 4
C4 olefin selectivity (%)	charge ratio	Co load	concentration	temperature

According to the above table, calculate the maximum value a of the minimum difference and the maximum value B of the maximum difference, then:

$$a = min_{(i)}min_{(k)}|x_0(k), x_i(k)| \tag{1}$$

$$\mathrm{b} = max_{(i)}max_{(k)}|x_0(k), x_i(k)| \tag{2}$$

At this time, the resolution coefficient is taken ρ= 0.5, the incidence matrix gamma is:

$$y(x_0(\mathrm{k}), x_i(\mathrm{k})) = \frac{\mathrm{a} + \rho\mathrm{b}}{|x_0(k) - x_i(k)| + \rho b} \tag{3}$$

Calculate correlation

$$z = \frac{\sum_{k=1}^{3} y(x_0\,(k), x_i(k)\,)}{\sum_{k=1}^{4} y(x_0\,(k), x_i(k)\,)} \tag{4}$$

$$T_k = \frac{y_4(k)(k), x_4(k)\,)}{\sum_{k=1}^{4} y(x_0\,(k), x_i(k)\,)} \tag{5}$$

2.4 Model solving

Table 3: Maximum value table

Parent sequence	a	b
Ethanol conversion (%)	0.0023	3.5658
C4 olefin selectivity (%)	0.0072	3.3747

Table 4: Correlation data color scale table

Parent sequence	Subsequence 1	Subsequence 2	Subsequence 3	Subsequence 4
ethanol conversion (%)	charge ratio	Co load	concentration	temperature
Correlation degree	0.7138	0.7177	0.6829	0.7407
C4 olefin selectivity (%)	charge ratio	Co load	concentration	temperature
Correlation degree	0.7438	0.7255	0.7258	0.7766

According to the above table, the correlation degree between temperature and ethanol conversion is 0.741, and the correlation is the most significant. At the same time, the correlation degree between temperature and C4 olefin conversion was 0.777, and the correlation was the most significant.

According to the above formula,

Table 5: Weight table.

Parent sequence \ weight	z	T_k
Ethanol conversion (%)	0.7406	0.2594
C4 olefin selectivity (%)	0.7387	0.2613

Finally,

$$K_{\text{ethanol}} = 0.7406X_0 + 0.2594T \tag{6}$$

$$K_{C4} = 0.7406X_0 + 0.2594T \tag{7}$$

The effect of temperature on ethanol conversion is $T_k = 0.2594$ increased, and the selectivity to C4 olefins increased with $z = 0.2613$; The effect of catalyst combination on ethanol conversion increased with $z = 0.7406$, and the selectivity for C4 olefins increased with $z = 0.7387$.

3. BP Neural Network Model

3.1 Introduction of BP neural network

BP (back propagation) neural network is the most traditional neural network. It is a multilayer feedforward network trained according to the error back propagation algorithm. It collects the errors generated by the system in the simulation process, returns these errors to the output value, and then uses these errors to adjust the weight of neurons, thus, an artificial neural network which can simulate the original problem is generated.

The topology of BP neural network model includes input layer, hidden layer and output layer.

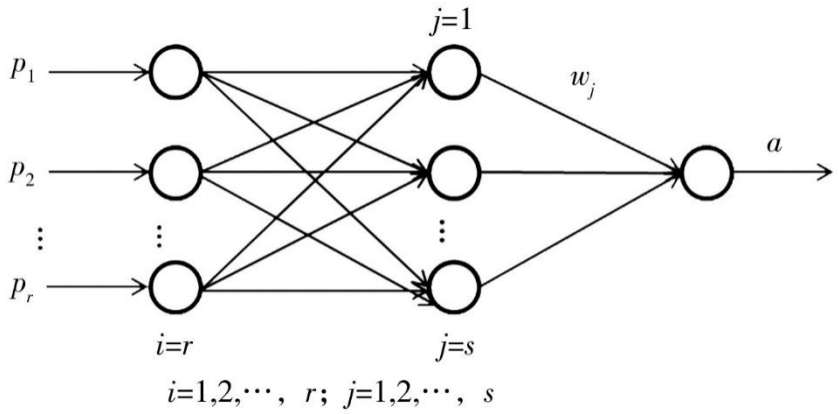

Figure 1: BP neural network topology.

Principle of BP neural network: gradient descent is used to search the hypothesis space of possible weight vector, and the weight vector of the best fitting sample has been found. Specifically, according to the loss function, the weight of the loss function about each layer and the partial derivative of the bias term, also known as gradient, are calculated, and the initial weight and bias term are updated with this value, It is updated until the loss function gets the minimum value or the set number of iterations is completed. So as to calculate the best parameters in the neural network.

3.2 Model establishment

1. Data sets and validation sets

We normalized and disorderly matched the temperature and catalyst combinations with different ratios in Annex I data, and there were 228 data values, including 7 catalyst combinations with different temperatures and 21 catalyst combinations with different ratios. The first 80% of the data were used as the samples for learning and training, and the last 20% of the data were used as the samples for verification set.

2. BP neural network structure

BP neural network has 2 input nodes, 1 output node and 10 hidden nodes. The weight from input layer to hidden layer is w and the paranoid term is b_1. The activation function is g_1. The weight value from the hidden layer to the output layer is V and the offset value is b_2. The activation function is g_2. The relationship between network input and output is as follows:

$$A = g_2(V^T g_1(W^T X + b_1) + b_2) \tag{8}$$

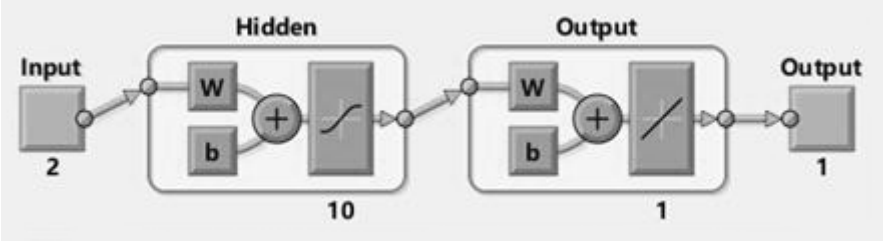

Figure 2: Schematic diagram of BP neural network.

3. Parameters of BP neural network

The main parameters in the BP neural network model include: interval steps of training results

net.trainparam.show, maximum training times net.trainparam.epochs, network learning rate net.trainparam.lr and minimum error of training target net.trainparam.goal. The numerical values of the parameters in the BP neural network prediction model set in this paper are shown in the table below:

Table 6: Parameter table in BP neural network prediction model

Training result interval steps	Maximum training times	Learning rate of network	Minimum error of training target
3	5000	0.001	0.01

3.3 Model solution

Matlab is used to input the combination of our treated temperature and catalyst combinations with different ratios into the neural network as the input layer for training to obtain the trained neural network, and then the combination of new temperature and catalyst combinations with different ratios is input into the network as the input layer for prediction, In the same figure, the actual C4 olefin yield is compared with the C4 olefin yield predicted by neural network.

The correlation coefficient reached 0.99682, indicating that the traditional BP neural network is more accurate in predicting C4 yield.

New temperature data are designed and combined with different catalyst combinations. The new combination is input into the trained neural network as an input layer to obtain new prediction results.

It is easy to see that when the temperature is 402℃ and the catalyst combination is A3 (200mg 1wt% Co / SiO2 - 200mg HAP ethanol concentration 0.9ml/min), the yield of C4 olefins is the highest.

When the temperature is lower than 350℃, the temperature is 343℃, and the catalyst combination is A2 (200mg 2wt% Co / SiO2 - 200mg HAP ethanol concentration 1.68ml/min), the yield of C4 olefins is the highest.

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