Study on chemical looping reforming reaction of methane based on PSO-BP neural network

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Abstract. Through the study of methane chemical looping reforming reaction, the BP neural network model optimized by the PSO algorithm was established, and the network structure of the PSO-BP model was determined to be 9-11-3. After training the model, the mean square error of the network was finally stabilized at 0.013509, and the learning rate was finally fixed at 0.083453, while the fitting degree of the training sample and the test sample were both above 0.979, indicating that the network of the PSO-BP model had strong learning ability and generalization capability, and was a simulation prediction model with good performance. The PSO-BP model was used to simulate the full set of experimental condition data, and the most experimental condition was the cerium iron composite oxygen carrier prepared by co-precipitation method (No.2) with a molar ratio of 0.7/0.3, a calcination temperature of 800°C, a calcination time of 6h, a reaction temperature of 850°C, a reaction time of 13min, and a circulation number of 0, while it was consistent with the actual experimental.

1. Introduction

Natural gas is a clean and environmentally friendly resource that will play a key role in energy transformation[1]. Natural gas is mainly methane, so in the conversion and utilization of natural gas, the indirect technology of producing synthetic gas from methane and then producing high-value-added chemicals or liquid hydrocarbons has been a research hotspot[2]. Methane chemical looping reforming technology is a new synthetic gas preparation technology based on the concept of chemical looping combustion[3-4]. In this technology, methane catalytic partial oxidation reaction is exothermic and mild. The synthetic gas produced has a hydrocarbon ratio of 2, which can be directly used for methanol and Fischer-Tropsch synthesis. Moreover, it has the advantages of small reactor volume, high efficiency and low energy consumption, and has been widely valued at home and abroad[5].

The artificial neural network is a mathematical model which has been widely applied in recent years, with a strong adaptive and self-learning and nonlinear mapping ability. At present, the artificial neural network is mainly applied in fault diagnosis[6-7], process control and optimization[8-9] and physical property estimation[10]. Wang AL et al.[9] used particle swarm optimization (PSO) algorithm to optimize BP neural network, established foaming quality control model of asphalt and a parameter optimization model of asphalt, and improved the prediction accuracy and generalization capability.
GAO YM et al. [11] used the BP neural network model optimized by PSO and the normal BP neural network model to predict the diesel oil production, and concluded that the PSO-BP model has a higher prediction accuracy than the normal BP model.

The PSO algorithm has the advantages of easy realization, fast convergence speed, and high efficiency, and can make up for the disadvantages of BP neural network such as slow learning process speed and easy falling into local minimum points [9,11]. Therefore, BP neural network optimized by PSO algorithm was proposed in this paper to simulate methane chemical looping reforming reaction, and to use the model to predict the experimental result data under specific experimental conditions and to study the optimal oxygen carrier system preparation and reaction conditions, providing valuable reference for the prediction of results, system analysis, process optimization and other chemical processes.

2. PSO-BP neural network model

2.1. BP neural network model
In this paper, the relevant research data of methane chemical chain reforming reaction in literature [12] is selected. The experiment based on Ce(NO₃)₂·6H₂O, with Fe(NO₃)₃·9H₂O, Mn(NO₃)₃·6H₂O, Cu(NO₃)₂·3H₂O as doping phase, using two different co-precipitation preparation a series of cerium based composite metal oxides. 419 groups of experimental data were extracted from this experiment. After normalization, 80% of the data groups were randomly selected as training samples and the remaining 20% as test samples.

According to the extracted data, Selection of Compound metal oxide type, molar ratio of ceria oxide, molar ratio of composite oxide, preparation method, calcination temperature, calcination time, reaction temperature, reaction time and cycle numbers as input variables, CH₄ conversion rate, CO selectivity and H₂ selectivity were selected as the output variables. Therefore, the establishment of the BP neural network model of the input layer node number is 9, output layer node number is 3.

2.1.1 Selection of the number of hidden layer nodes
The number of nodes in the hidden layer of the BP neural network determines the network structure and has a great influence on fault tolerance, information processing ability and generalization ability of the network. In this paper, the approximate range of the number of nodes in the hidden layer is determined by the formula, and the number of nodes with the best comprehensive performance of the BP model is found out through the trial-and-error method, which is the optimal number of nodes in the hidden layer.

\[ k = \sqrt{m + n + a} \]

Where k is the number of hidden layer nodes, m is the number of input layer nodes, n is the number of output layer nodes, and a is a constant between 0 and 10.

2.1.2 Selection of activation function
In this paper, the tan sigmoid function that can accelerate the convergence rate is chosen as the activation function from the input layer to the hidden layer and from the hidden layer to the output layer of the BP neural network. Its function expression is

\[ f = \frac{2}{(1 + e^{-2n})} - 1 \]

2.1.3 Selection of learning algorithm
Adaptive learning rate refers to the timely adjustment of learning rate according to the objective function of the network. In the process of solving the network, the moving step size more in line with the current situation is adopted according to the actual situation, so as to make the convergence of the network more intelligent, which will improve the convergence speed and convergence ability of the network.

2.2. PSO algorithm optimization
The PSO algorithm optimizes the initial weight threshold. Based on the BP neural network model, the
weight threshold is treated as a particle. The PSO algorithm is used to globally optimize and find the relative optimal initial weight threshold. The neural network model is called For the PSO-BP model. In the PSO-BP model, the population size is 60, the number of evolution is 30, the learning factors $c_1$ and $c_2$ are both 1.49445, the position change of particles in the search space is set within $[X_{\text{min}}, X_{\text{max}}]=[-5, 5]$, and the velocity change is set within $[V_{\text{min}}, V_{\text{max}}]=[-1, 1]$.

3. Results and discussion

Figure 1 shows that when the number of hidden layer nodes is 11, the training error of the network is the smallest, and the network of correlation of training samples and testing samples was higher than other numbers of hidden layer nodes. Therefore, the optimal number of hidden layer nodes in this model is 11. Finally, the PSO-BP neural network structure was selected to be 9-11-3.

![Figure 1. Network performance of different hidden layer nodes](image1)

Figure 2 shows that the fitting degree of the PSO-BP model to the training sample data is 0.9847, and the fitting degree of the test sample data can reach 0.97926, indicating that the PSO-BP model has a good training effect.

![Figure 2. Fitting degree of PSO-BP model to sample data (a is training data, b is prediction data)](image2)

It can be concluded from figure 3 and figure 4 that in the first 8000 times of network training, the learning rate is between 1-2, the particles move in the solution space to search for solutions with big step size, and the mean square error of the network decreases rapidly. After about 8000 times of training, the learning rate of the network fluctuates between 0-1, the moving step size of particles in the solution space shrinks and global optimization continues to approach the optimal solution, the decrease rate of the mean square error of the network gradually decreases and eventually tends to be stable, the final training mean square error is 0.013509, and the learning rate is finally fixed at
0.083453. It shows that the PSO-BP neural network model has good convergence ability.

![Figure 3. The training mean square error curves of PSO-BP model](image1)

![Figure 4. Training gradient curve of PSO-BP model](image2)

After optimizing the initial weight threshold, the fitting degree of the PSO-BP model for training samples and test samples was above 0.979, and the learning ability and generalization capability of the network were strong. Therefore, PSO-BP model is a good simulation and prediction model for methane chemical looping reforming reaction.

3.1. Prediction of optimal reaction conditions for methane chemical looping reforming reaction

We extracted 419 groups of experimental conditions and corresponding experimental results from the literature. The experimental conditions worthy of exploration in this system were listed and sorted out, and 123,552 groups of experimental conditions were obtained. By using PSO-BP model to predict these experimental conditions, all the corresponding experimental results can be obtained. All the experimental results were optimized to obtain the optimal preparation conditions and fixed bed reaction conditions for the composite oxygen carrier system when the CH₄ conversion rate, CO selectivity, and H₂ selectivity were optimized. The relevant data are shown in table 1.

| Experimental condition | Parameters |
|------------------------|------------|
| Compound metal oxide type | Fe(NO₃)₃.9H₂O |
| Molar ratio of ceria oxide | 0.7 |
| Molar ratio of composite oxide | 0.3 |
| Preparation method | co-precipitation method (No.2) |
| Calcination temperature | 800°C |
| Calcination time | 6h |
| Reaction temperature | 850°C |
| Reaction time | 13min |
| Cycle numbers | 0 |

PSO-BP neural network simulation to find the best experimental conditions, compared with the experimental data and the related conditions set of data 419 is extracted methane chemical looping reforming reaction results are in agreement. From the 419 sets of actual data, 123,552 sets of highly predictive data were obtained, and the data expansion rate was 293.87 times. It can be seen that the PSO-BP model can provide a valuable reference for finding the optimal oxygen carrier preparation conditions and reactor reaction conditions.
4. Conclusions
Based on the experimental data of methane chemical looping reforming reaction, the PSO-BP model was established with compound metal oxide type, molar ratio of ceria oxide, molar ratio of composite oxide, preparation method, calcination temperature, calcination time, reaction temperature, reaction time and cycle numbers as input variables and CH$_4$ conversion rate, CO selectivity and H$_2$ selectivity as output variables. PSO algorithm which solves the instability of BP neural network initial weights and thresholds randomly selected causes problems predictions, improved stability and prediction accuracy of the network. In addition, the PSO-BP model was used to make a complete set prediction of the experimental data and select the optimal reaction conditions, and the results were consistent with the facts. It is shown that the model has good prediction ability and can provide a valuable reference for the chemical process such as result prediction, system analysis, and process optimization.

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