Application of PSO-BP neural network in methane chemical looping reforming reaction

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Abstract. In order to overcome the disadvantage of BP neural network easy falling into local convergence, a BP neural network model with particle swarm optimization (PSO) is established to simulate and predict chemical looping reforming (CLR) of methane. The prediction results of PSO-BP model are analyzed, the results showed that the fitting degree of PSO-BP model training set and test set are both above 0.979, and the error of the network is finally stable at 0.013509. It is proved that this model is a good prediction model. The experimental results under a certain reaction condition are predicted by the PSO-BP model, and the relative errors between the predicted date and the actual data are 1.581% at the maximum and 0.118% at the minimum. Then the PSO-BP model is used to simulate and screen all the experimental data, and the optimal experimental conditions are obtained as follows: the ceria-iron composite oxygen carrier with a molar ratio of 7/3 prepared by co-precipitation method, calcinated at 800 °C for 6h, and the proper reaction temperature is 850 °C. Finally, the predicted reaction performance under optimized operating conditions were consistent with the actual experimental.

1. Introduction

Natural gas is an abundant, cheap, clean and convenient fossil energy, and an important part of the world energy system [1]. The main composition of natural gas is methane, so it makes sense to convert methane into high added value and easily transportable chemicals. Among the many conversion routes, methane to syngas is the most competitive [2]. Now, the CLR is a promising technology [3], it is a new type of methane producing syngas technology based on the chemical looping combustion [4]. CLR is a mild exothermic and get the H₂/CO ratio of 2, which can be directly used in the synthesis of methanol and F-T. So in recent years, scholars at home and abroad have carried out extensive studies on the CLR of methane to syngas [5].

The BP neural network with complex pattern classification ability and good ability of multidimensional function mapping, has been widely applied in various fields [6,7]. Yan et al. [8] proposed an optimized BP model to estimate nitrogen concentration, which the model had good prediction performance and high prediction accuracy, and for the other modeling problems in
ammonia synthesis production also had good generalization ability. Yin et al. [9] predicted the carbon content of burning coal using BP neural network, and found that BP model has better performance than emission factor method or capacity standard method. Karaci et al. [10] developed a model to predict the hydrogen rich gas (H-rG) produced by biomass waste pyrolysis under different catalysts. The results showed that the model could accurately predict the H-rG ratio of pyrolysis process without experiments. Yin et al. [11] predicted the optimal discharge pressure of the system COP by PSO-BP neural network, and the new model had higher accuracy compared with the existing relevant models.

In this paper, PSO-BP neural network simulation is introduced in methane to syngas reaction, and a reliable model is established based on appropriate experimental data, which can greatly reduce the workload of experimental research. The PSO-BP model can be used to find better reaction conditions based on experimental data, and provide valuable reference for system analysis, result prediction and process optimization.

2. Experimental data and methods

2.1. Experimental data
The relevant research data of the CLR in literature [12] is selected. 419 sets of experimental data were extracted from the literature. After data normalization, 80% were randomly assigned as training set and the rest as test set.

2.2. Establishment of BP model
According to the experimental data, nine variables including the composite metal oxide species, the molar ratio of ceria oxide, the molar ratio of composite oxide, the preparation method, the calcination temperature, the calcination time, the reaction temperature, the reaction time and the cycle numbers were selected as the input layer nodes, and three outputs were the CH4 conversion rate, the CO selectivity and the H2 selectivity. The optimal number of hidden layer nodes was determined by formula (1). We selected tansig function as the excitation function of BP neural network, and selected the adaptive learning rate momentum gradient descent algorithm (traingdx algorithm) as its learning algorithm.

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

(1)

Where \( k \), \( m \), and \( n \) are the number of hidden layer nodes, input layer nodes, and output layer nodes, respectively; and \( a \) is a constant between 0 and 10.

2.3. PSO algorithm optimization
The PSO algorithm was first proposed by Eberhart and Kennedy [13]. Its basic idea was to simulate the mutual cooperation between individuals in the process of random foraging of birds, so that the collective can reach the optimal global optimization algorithm. Every individual was called the particle represented a potential solution, with position, velocity and fitness value indicated that the particle characteristics. The velocity of the particle determined the direction and distance of the particle. The fitness value was calculated by the fitness function.

In this paper, the parameters of PSO algorithm were set as follows:

The population size and the number of evolutions were respectively 60 and 30. The learning factor \( C_1 \) and \( C_2 \) are both 1.49445. The position change of particles in the search space is set within the range of \([X_{\text{min}}, X_{\text{max}}]=[-5, 5]\). The speed change was set within the range of \([V_{\text{min}}, V_{\text{max}}]=[-1, 1]\).

3. Results and discussion

3.1 PSO-BP model
As can be seen from Figure 1, the node of hidden layer with the minimum training error is 11, and the error is 0.07198, meanwhile the correlation between the training set and the test set of the network is also relatively high. Hence, the network structure of PSO-BP model is determined as 9-11-3.
Figure 1. Performance of PSO-BP model with different hidden layers

Figure 2 shows that all the data are uniformly distributed around the oblique line, and the correlation coefficient between the target and training data and between the target and test data is close to 1, indicating the good fitting effect of the PSO-BP model. It can be seen from Figure 3 and Figure 4 that in the process of model iterations times from 0 to 250,000, the training mean square error of the network gradually decreased and finally stabilized at 0.013509. Figure 4 shows that the gradient of the model is 0.0030804, and the validation check is 0. This means that the output error obtained by the system after the input validation data not increase after successive iterations. This indicates that the PSO-BP model does not fall into the state of over-fitting, and the training error still has the possibility of decreasing, which also indicates that the training state of the PSO-BP model is good.

Figure 2. The fitting degree of PSO-BP model to training data (a) and prediction data (b)
3.2. PSO-BP model predicts the experimental results
In the methane CLR system, experimental results under certain conditions may be required. Although the experiment under this experimental condition has not been actually operated, it can still be predicted by the established PSO-BP model. In order to facilitate understanding, a set of experimental conditions are simulated. The ceria-iron composite oxygen carrier with a molar ratio of 7/3 prepared by co-precipitation method, calcinated at 700 °C for 6h, the proper reaction temperature is 850 °C, and the reaction time is 0.066667min, these data are input into the PSO-BP model to obtain the prediction results under the experimental conditions. Relevant data are summarized in table 1.

Table 1 Comparison of experimental results with predicted results

|                       | Experimental results (%) | Predicted results (%) | Relative error (%) |
|-----------------------|--------------------------|-----------------------|--------------------|
| CH₄ conversion rate   | 87                       | 86.236252             | 0.878              |
| CO selectivity        | 96.6                     | 96.713881             | 0.118              |
| H₂ selectivity        | 96.8                     | 95.269954             | 1.581              |

From Table 1, using the PSO-BP model to predict the experimental results under specific reaction conditions, the maximum relative error between the predicted data and the actual data is 1.581%. Therefore, using the PSO-BP model can obtain more accurate and reference-value data without actual experiments, saving a lot of manpower, material resources and financial resources, which is of great practical value.

3.3. Prediction of optimal reaction conditions for the CLR
We extract 419 groups of experimental data for the CLR. The experimental conditions that are worth exploring are listed and sorted out, and 123552 sets of experimental conditions are obtained. The PSO-BP neural network model is used to predict the processed data. In the predicted data set, it is easy to find the optimal preparation conditions of the compound oxygen carrier and the reactor reaction conditions when the CH₄ conversion rate, CO selectivity and H₂ selectivity are optimized. The best preparation and reaction conditions of oxygen carrier are obtained as follows: the ceria-iron composite oxygen carrier with a molar ratio of 7/3 prepared by co-precipitation method, calcinated at 800 °C for 6h, and the proper reaction temperature is 850 °C.

The optimal experimental conditions obtained by the PSO-BP model simulation are compared with the actual experimental conditions of the CLR, and the results are consistent. The forecast data of
123552 groups with great reference value are obtained from the 419 groups of actual data, and the data expansion rate is 293.87 times.

4. Conclusions
In this paper, the PSO-BP model is used to model and analyze the CLR of methane. By using PSO algorithm to explore the optimal initial network weight and threshold, the model's convergence ability, convergence speed and network performance are greatly improved. Secondly, the PSO-BP model is used to predict the experimental results under specific conditions, and the relative error between the predicted results and the actual results is within 1.1%, which has a good prediction effect. Finally, the optimal experimental conditions obtained by using the PSO-BP model to simulate the full set experimental data are consistent with the actual experimental conditions. It shows that the application of PSO-BP model in the CLR of methane is feasible, and it can provide a valuable advice for finding the best preparation conditions of oxygen carrier and reactor reaction conditions.

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