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Methanol Steam Reforming Performance Optimisation of Cylindrical Microreactor for Hydrogen Production Utilising Error backpropagation and Genetic Algorithm

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Abstract: To optimise methanol steam reforming performance of cylindrical microreactor for hydrogen production, an error backpropagation algorithm was used to build a mathematical model for reaction performance of different microreactors for hydrogen production. Additionally, a genetic algorithm (GA) was utilised to process the computational model to obtain the optimum reaction parameters. The reliability of the optimum reaction parameters of cylindrical microreactor for hydrogen production was verified by experiments. Firstly, take plate microreactor as an example, the porosity of porous copper fiber sintered sheet (PCFSS), reaction temperature of methanol steam reforming for hydrogen production, injection velocity of the methanol and water mixture, and catalyst loading of PCFSS were considered as input data, whereas methanol conversion was used as output data. The computational model for specific testing system was gained by utilising input and output data from specific testing system to train the mathematical model for different microreactors, combining with matrix laboratory (MATLAB) neural network toolbox and designed MATLAB program. The E_{max} of 5% for plate microreactor and E_{max} of 3.2% for cylindrical microreactor verified the good predictive ability and reliability of the computational model for plate and cylindrical microreactor, indicating the reliability and universal applicability of the mathematical model for different microreactors. Secondly, the effects and mechanisms of PPI, reaction temperature, injection velocity, and catalyst loading on methanol conversion were studied, relying on the computational model. Finally, the optimum reaction parameters were acquired using GA, MATLAB neural network toolbox and designed MATLAB program. The validity of the optimum reaction parameters of cylindrical microreactor for hydrogen production was confirmed by experiments. This study provides a reference method for methanol steam reforming performance optimisation for hydrogen production.

Keywords: Methanol steam reforming microreactor; Hydrogen production; Error backpropagation algorithm; Genetic algorithm; Methanol conversion

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1. Introduction

Compared with conventional reactor, owing to the characteristics of microchannel structure and small channel size, microreactor has the advantages, such as high surface-to-volume ratio, intensified heat and mass transfer, rapid and direct amplification, and high safety. Therefore, it has received considerable attention from researchers [1]. On the one hand, microreactor for hydrogen production has been received more attention because of its ability to provide reliable online hydrogen source for fuel cells [2]. On the other hand, methanol fuel has the advantages, such as liquid, sulphur-free, low reforming temperature, high hydrogen content, cheap, easy storage and transportation, renewable [3]. Hence, development of methanol microreactor for hydrogen production is an important direction in the research of mobile hydrogen source in vehicle [4-5].

In the previous work, structure design, process and manufacture of methanol microreactor for hydrogen production, and reaction support of methanol microreactor for hydrogen production are mainly studied in the research of methanol steam reforming technology [6-18]. In the structure design of the microreactor, microreactors such as a plate–fin microreactor, cube–post microreactor, annular microreactor, and cylindrical microreactor have been developed [6-10]. In the process and manufacture of a methanol microreactor, processing technologies such as milling, special process, and microelectromechanical systems (MEMSs) have been used to manufacture straight channels, serpentine channels, spiral channels, etc. [11-14]. In research on reaction support, porous metal materials used as the reaction support in microreactors have also been examined. Foam technology, solid-phase sintering technology, and liquid-phase sintering technology have been developed to fabricate the porous reaction support and have been successfully applied as catalyst support in ammonia decomposition and hydrogen production by methanol [15-18].

Nomenclature

Variables

b_h the thresholds of the hidden-layer neuron in the mathematical model, namely undetermined coefficients in the mathematical model

b_o the thresholds of the output-layer neuron in the mathematical model, namely undetermined coefficients in the mathematical model

E_{max} the maximum error rate for predicted methanol conversion of the mathematical model

$f(.)$ activation function in the mathematical model

h hidden-layer neuron in the mathematical model

K Kelvin environmental temperature of methanol steam reforming, K

m_c volume fraction of CO in reformate gas, %

n number of input layer neurons of the mathematical model, namely the number of factors affecting reaction performance

n_c volume fraction of CO_2 in reformate gas, %

N the number which is before the adjustment

$N+1$ the number which is after the adjustment

o output layer neuron in the mathematical model

q number of output-layer neurons

p number of hidden-layer neurons

s_o the o-th value of input vector of the output layer in the mathematical model

$\triangle t_o$ error of the o-th evaluation index value

t_o the o-th value of the output vector in the mathematical model, namely the o-th evaluation

index value in the *t*, the *o*-th reaction result in one experimental data

u_h the *h*-th value of input vector of the hidden layer in the mathematical model

v_h the *h*-th value of output vector of the hidden layer in the mathematical model

$V_{\text{injection}}$ injection velocity of methanol and water mixture, ml/h

$V_{\text{reformate gas}}$ injection velocity of reformate gas, ml/min

$w_{h,o}$ the weights between the hidden layer and the output layer in the mathematical model, namely undetermined coefficients in the mathematical model

$w_{i,h}$ the weights between the input layer and the hidden layer in the mathematical model, namely undetermined coefficients in the mathematical model

X_{CH3OH} methanol conversion, %

$X_{experiment}$ experimental methanol conversion

x_i value of the *i*-th factor affecting reaction performance, namely the *i*-th reaction parameter in one experimental data

X_{model} predicted methanol conversion of the mathematical model

y_o technical requirement value of the *o*-th evaluation index value

η learning rate which value is between (0, 1)

Abbreviations

GA genetic algorithm

MATLAB matrix laboratory

MEMSs microelectromechanical systems

PCFSS porous copper fiber sintered sheet

PPI pores per inch

Neural networks have strong feature extraction and abstraction capabilities, and they can integrate multisource information, process heterogeneous data, and capture change dynamics, thus playing an important role in parameter optimisation [19-20]. To date, some foreign scholars have used neural networks to optimise the reaction performance of microreactors. For example, Aghajani used an artificial neural network to research the size of synthesised nano-iodine in microreactors; it was found that the relationships between flow rate of solvent, flow rate of antisolvent, and size of the synthesised nano-iodine are in inverse relation [21]. Na researched the optimisation of catalyst loading in Fischer–Tropsch microchannel reactors, using the distribution of catalyst loading in microchannel reactors as a variable and considering C_{5+} productivity and temperature rise in microchannels as optimisation objects by using computational fluid dynamics, it was found that C_{5+} productivity was increased to 22% and ΔT_{max} was decreased to 63.2% by using a genetic algorithm (GA) [22]. Recently, Jung researched the structure optimisation of Fischer–Tropsch microchannel reactors, considering such structure parameters as the length, width, and height of microchannels in microreactor as variables, using reactor core volume and reaction temperature rise were used as optimisation objects by utilising the coupling method and artificial neural networks [23].

Although some research involving the design, processing, and manufacturing, as well as the methanol steam reforming performance optimisation of the microreactor for hydrogen production, has been conducted, the study of the reaction parameters optimisation of methanol steam reforming for hydrogen production has not been reported. Here, in order to obtain the optimum reaction parameters of cylindrical microreactor for hydrogen production, a mathematical model for the methanol steam reforming performance of different microreactors for hydrogen production was

created using the error backpropagation algorithm. The validity and universal applicability of the mathematical model for different microreactors were verified by experimental data from the plate and cylindrical microreactor. The predictive ability and reliability of the computational model for cylindrical microreactor were verified by experimental data. The relationships between reaction parameters and reaction results of methanol steam reforming were studied relying on the computational model for cylindrical microreactor. Subsequently, the optimum reaction parameters of cylindrical microreactor for hydrogen production were obtained using a GA, thereby optimising the reaction parameters.

2. Establishment of mathematical model for different microreactors

A mathematical model for methanol steam reforming performance for hydrogen production is established utilising an error backpropagation algorithm. Subsequently, the computational model for reaction performance of the specific testing system for hydrogen production is gained by training the mathematical model for different microreactors with several sets of experimental data from the specific testing system, combining with MATLAB neural network toolbox and the designed matrix laboratory (MATLAB) program.

2.1. Error backpropagation algorithm

Error backpropagation algorithm is a learning process of positive information dissemination and error backpropagation [24-26]. Input information passes to the output layer through a layer-by-layer process from the input layer to the hidden layer. The backpropagation algorithm uses the gradient search technology to minimise the mean square value of the error between the actual output value of the network and the desired output. When the output layer does not achieve the desired level, it runs into the backpropagation, by modifying the weights and thresholds of each layer of the neuron, it

gains the minimum error value between the actual output and desired output.

2.2. Establishment of mathematical model for different methanol steam reforming microreactors for hydrogen production

Eqs. (1)–(6) exhibit the mathematical model for different methanol steam reforming microreactors for hydrogen production. In this model, x_i is the value of the *i*-th factor affecting reaction performance, namely the *i*-th reaction parameter (input data) in one experimental data; t_o is the *o*-th value of the output vector, namely the *o*-th evaluation index value, the *o*-th reaction result (output data) in one experimental data; $w_{i,h}$, $w_{h,o}$, b_h, b_o are the undetermined coefficients in the mathematical model to be solved, can be obtained by using reaction parameters (input data) and reaction results (output data) from several experimental data from specific testing system to train the mathematical model for different microreactors. Methanol conversion is considered as the only one of the evaluation indexes for the methanol steam reforming performance for hydrogen production. Thus, the value of *q* in the mathematical model is 1.

The computational model for the various testing system can be built, when the mathematical model for different microreactors is applied in the various testing systems and several experimental data including input and output data from the various testing systems is used to train the mathematical model for different microreactors. The mathematical model for different microreactors has universal applicability, it can be applied in different methanol steam reforming microreactors for hydrogen production.

$$u_h = \sum_{i=1}^{n} w_{i,h} x_i - b_h, \qquad h=1,2,...,p \tag{1}$$

$$f(x) = \frac{1}{1+e^{-x}} \tag{2}$$

$$v_h = f(u_h), \quad h=1,2,...,p \tag{3}$$

$$s_o = \sum_{h}^{p} w_{h,o} v_h - b_o, \quad o=1,2,...,q \tag{4}$$

$$t_o = f(s_o), \quad o=1,2,...,q \tag{5}$$

$$\Delta t_o = t_o - y_o, \quad o=1,2,...,q \tag{6}$$

2.3. Generality verification of the mathematical model for different microreactors

The universal applicability procedure of the mathematical model for different microreactors is the setup procedure of the computational model for the specific testing system. The computational model for the specific testing system is established by using the mathematical model for different microreactors and several experimental data including input and output data from the specific testing system for specific methanol steam reforming microreactor for hydrogen production, combining with MATLAB neural network toolbox and the designed MATLAB program.

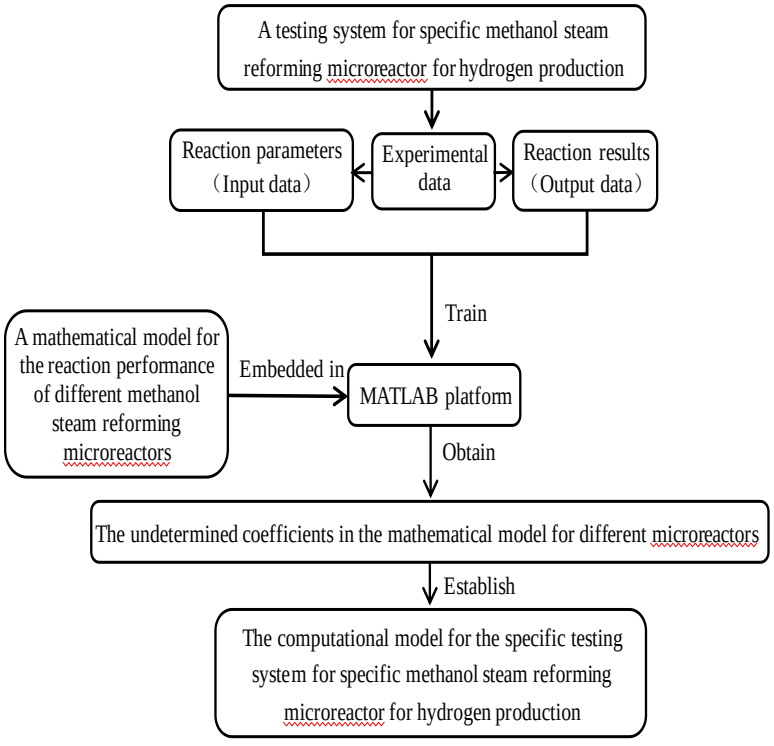

Fig. 1. Universal applicability procedure of the mathematical model for different microreactors

Fig.1 shows the universal applicability procedure of the mathematical model for different microreactors. Firstly, a testing system for specific methanol steam reforming microreactor for hydrogen production is decided. If the number of the reaction parameters in specific testing system is five, the value of n in the mathematical model for different microreactors is five. If the number of evaluation indexes for the reaction performance is two, the value of q in the mathematical model for different microreactors is two. The five reaction parameters in several sets of experimental data from the specific testing system for methanol steam reforming for hydrogen production are selected as input data, while two evaluation indexes are used as output data. Subsequently, the undetermined coefficients $w_{i,h}$, $w_{h,o}$, b_h, b_o in the mathematical model for different methanol steam reforming microreactors for hydrogen production are solved using the MATLAB neural network toolbox, combining the input and output data—building the computational model for the specific testing system.

Here, the reliability of the mathematical model for different microreactors was verified by taking a plate microreactor as an example, as shown in Fig.2. The computational model for the plate microreactor was established by using the mathematical model for different microreactors and 30 sets of experimental data including input and output data from the testing system for plate microreactor for methanol steam reforming for hydrogen production, as shown in Fig.3. The system mainly consisted of a hydrogen bottle, a nitrogen bottle, a mass flowmeter, a precise injection pump, thermostats, a condensing cube, a drying cube, an electric soaping flowmeter, a computer, and a gas chromatograph. The plate microreactor mainly consisted of an evaporation chamber, a reforming chamber, heating cartridges, thermocouples, and reaction support consisting of the porous copper fiber sintered sheet (PCFSS). Methanol and water were evaporated into gas in an evaporation

chamber. The PCFSS was coated with a Cu-based catalyst [12]. Then, methanol and steam were reacted in the PCFSS to produce H_2, CO and CO_2 in the reforming chamber.

Table 1 shows input data including the porosity of the porous copper fiber sintered sheet (PCFSS), the reaction temperature of methanol steam reforming for hydrogen production, the injection velocity of the methanol and water mixture, and the catalyst loading of the PCFSS. The output data including methanol conversion from 30 sets of experimental data of plate microreactor for methanol steam reforming for hydrogen production. Appendix A shows the main MATLAB program designed to solve the computational model for plate microreactor for methanol steam reforming for hydrogen production by utilising the MATLAB neural network toolbox.

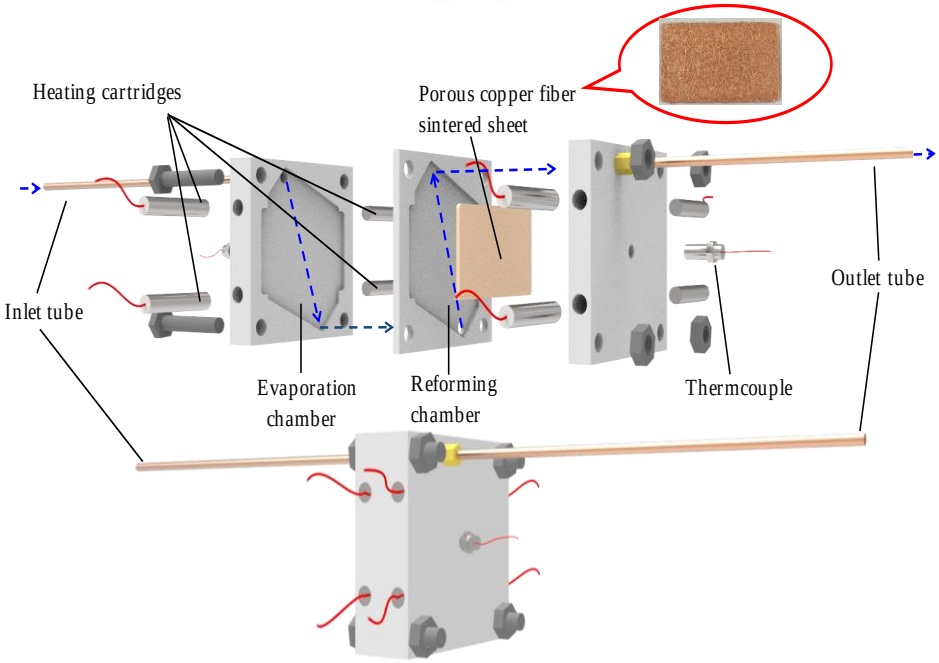

Fig.2. Plate microreactor for methanol steam reforming for hydrogen production

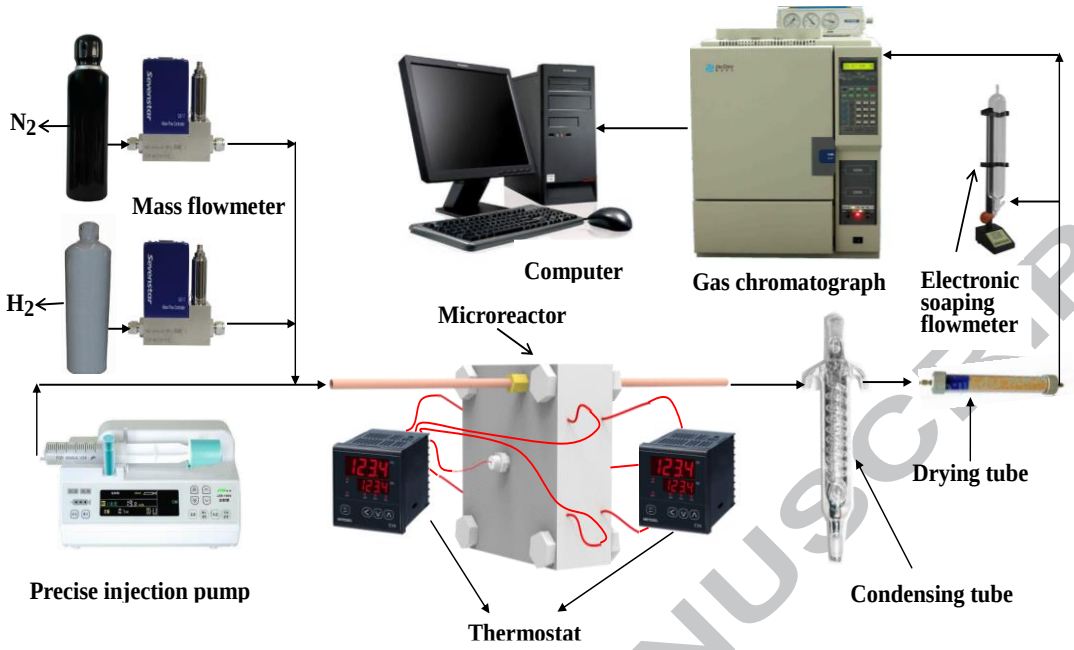

Fig.3. Testing system for plate microreactor

Eq. (7) exhibits the maximum error rate for predicted methanol conversion of the computational model, compared with the experimental value.

$$E_{\max} = \frac{\left| X_{model} - X_{experiment} \right|}{X_{experiment}} \times 100\% \tag{7}$$

Table 1. Thirty sets of input and output data of plate microreactor

Experimental number	Input data				Output data
	Porosity	Reaction temperature (℃)	Injection velocity（ml/h）	Catalyst loading (g)	Methanol conversion (%)
1	90	300	3.5	0.5	88.10
2	90	300	4.5	0.5	84.60
3	90	300	5.5	0.5	79.46
4	90	300	6.5	0.5	76.78
5	90	300	7.5	0.5	74.60
6	90	280	5.5	0.5	75.60
7	90	320	5.5	0.5	82.10
8	90	360	5.5	0.5	88.80
9	90	380	5.5	0.5	91.50
10	80	300	3.5	0.5	92.50

11	80	300	4.5	0.5	90.00
12	80	300	5.5	0.5	86.00
13	80	300	6.5	0.5	83.00
14	80	300	7.5	0.5	81.50
15	80	280	5.5	0.5	83.50
16	80	320	5.5	0.5	90.55
17	80	340	5.5	0.5	92.32
18	80	380	5.5	0.5	94.10
19	70	300	3.5	0.5	96.24
20	70	300	4.5	0.5	95.67
21	70	300	5.5	0.5	94.02
22	70	300	6.5	0.5	89.20
23	70	300	7.5	0.5	86.81
24	70	280	5.5	0.5	91.92
25	70	340	5.5	0.5	96.58
26	70	360	5.5	0.5	97.02
27	70	380	5.5	0.5	99.10
28	70	320	5.5	0.5	95.78
29	90	340	5.5	0.5	85.60
30	80	360	5.5	0.5	93.50

Fig.4 and Table 2 show methanol conversion comparison of plate microreactor between the computational model and experiment under different injection velocities of the methanol and water mixture, in the condition of 90 porosity, 320℃ reaction temperature, and 0.5-g catalyst loading. The predicted methanol conversion of plate microreactor is broadly in line with the experimental methanol conversion in the same reaction conditions, the partial deviation is not big, and the maximum $E_{\max}$ is 5.0%.

The above results reveal the good predictive capability and reliability of the computational model for methanol steam reforming performance of plate microreactor for hydrogen production, which indicate the reliability of the mathematical model for methanol steam reforming performance of different microreactors for hydrogen production.

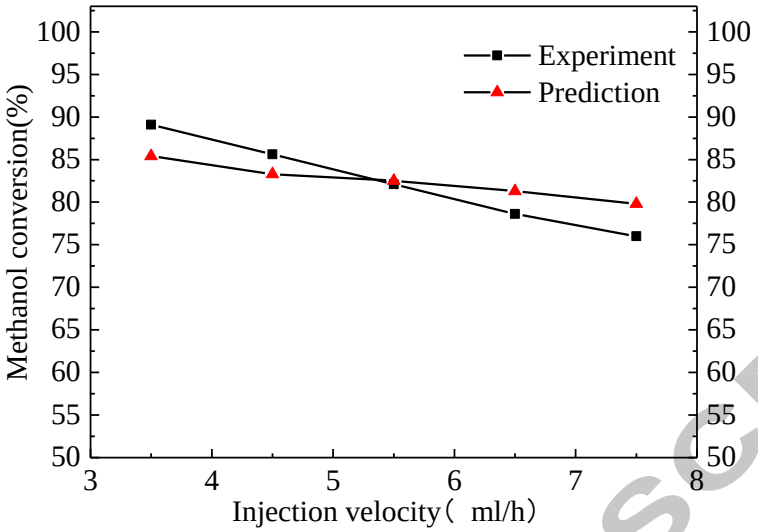

Fig. 4. Methanol conversion comparison of plate microreactor between the computational model and experiment under different injection velocities

Table 2. Maximum error rate for predicted methanol conversion of the computational model for plate microreactor under different injection velocities

Reaction parameters / Different injection velocities	90 Porosity	320℃ reaction temperature	0.5-g catalyst loading
E_{max}	5.0%		

3. Cylindrical microreactor and its testing system

Fig.5 shows a cylindrical microreactor for methanol steam reforming for hydrogen production. The cylindrical microreactor mainly consisted of an evaporation chamber, a reforming chamber, heating cartridges, thermocouples, and reaction support consisting of three round foam metals. Methanol and water were evaporated into gas in an evaporation chamber. The foam metal was coated with a Cu-based catalyst [12]. Then, methanol and steam were reacted in three round foam metals to produce H_2, CO and CO_2 in the reforming chamber. Fig.6 shows the testing system for cylindrical

microreactor. The system mainly consisted of a hydrogen bottle, a nitrogen bottle, a mass flowmeter, a precise injection pump, thermostats, a condensing cube, a drying cube, an electric soaping flowmeter, a computer, and a gas chromatograph.

The methanol and water mixture (at a mole ratio of 1:1.3) was injected into a cylindrical microreactor by means of a precise injection pump. The injection velocity of the methanol and water mixture was controlled by a precise injection pump. Three round foam metals were chosen as reaction support for the reaction of methanol and steam, and the reaction support of the foam metal was plated with the catalyst needed for methanol steam reforming. The mounts of the catalyst loading coated with the three foam metals were the same. The catalyst loading coated on foam metal in this study was the total amount of catalyst plated on three foam metals. The reaction temperature of the cylindrical microreactor was controlled by heating cartridges, thermocouples, and thermostats. Unreacted methanol and steam were separated from reacted hydrogen-rich gas by a condensing tube and drying tube. The flow speed of the reformate gas was measured by a soaping flowmeter. The volume fractions of CO, CO_2, and H_2 in reformate gas were analysed by a gas chromatograph.

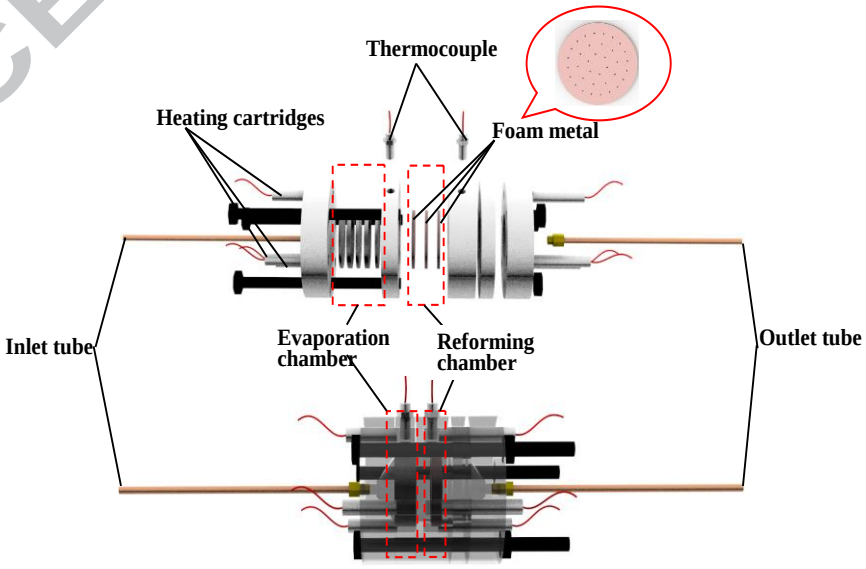

Fig.5. Cylindrical microreactor for methanol steam reforming for hydrogen production

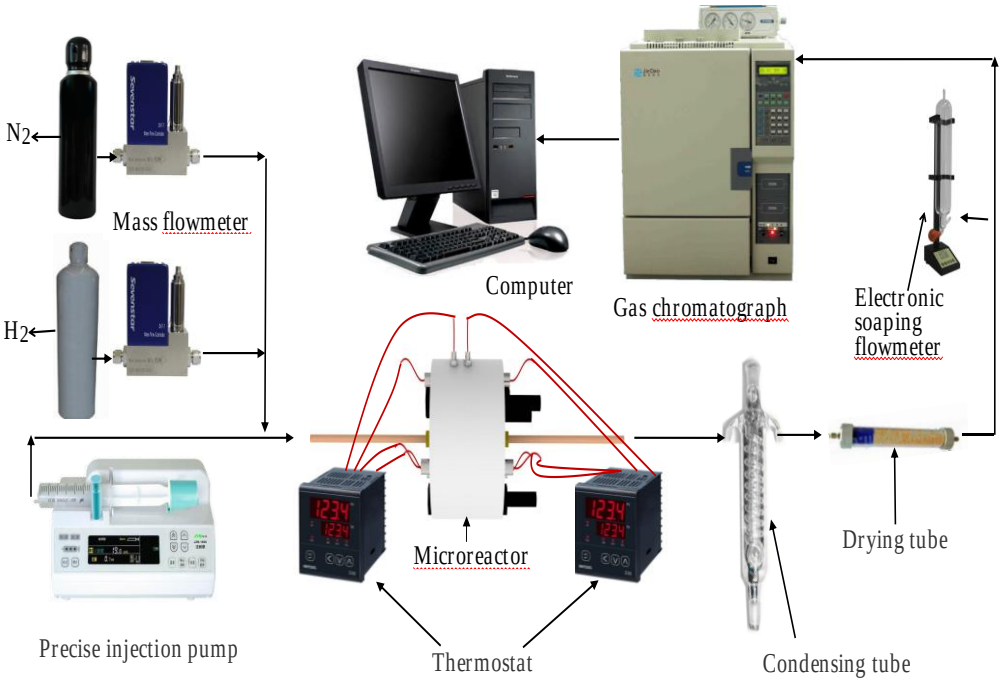

Fig.6. Testing system for cylindrical microreactor for hydrogen production

Methanol conversion was used as an index to evaluate the reaction performance of methanol steam reforming for hydrogen production. High methanol conversion reveals better reaction performance of methanol steam reforming, whereas low methanol conversion reveals poor reaction performance of methanol steam reforming. Eq. (8) exhibits the empirical formula for methanol conversion. In this formula, 273 represents Kelvin temperature (K) at 0℃, $\frac{1}{60}$ represents the conversion coefficient between $V_{injection}$ (ml/h) and $V_{reformate\ gas}$ (ml/min), 22400 represents volume (ml) of 1 mole gas at a temperature of 0℃ (273 K) and standard atmospheric pressure, and $\frac{1}{64}$ represents the mole quantity of methanol in 1 ml methanol and water mixture. In this study, the Kelvin environmental temperature of methanol steam reforming is 298 K.

$$X_{CH_3OH} = \frac{V_{\text{reformate gas}} * (m_c + n_c)}{V_{injection} * \frac{1}{60} * \frac{1}{64} * \frac{273}{K} * 22400} \quad (8)$$

The methanol and water mixture is vaporised in a vaporisation chamber; then, in the form of gas, it enters the reforming chamber and reacts by auxiliary action of the catalyst. Eqs. (9)–(11) exhibit the main reaction process [27-29].

$$CH_3OH + H_2O \rightarrow 3H_2 + CO_2, \quad \Delta H_{298^\circ C} = +49.4\ KJ/m\ o \tag{9}$$

$$CH_3OH \rightarrow CO + 2H_2, \quad \Delta H_{298^\circ C} = +92.0\ KJ/m\ o \tag{10}$$

$$CO + H_2O \rightarrow CO_2 + H_2, \quad \Delta H_{298^\circ C} = -41.1\ KJ/m\ o \tag{11}$$

Eq. (9) is the methanol steam reforming reaction directly for hydrogen production, Eq. (10) is the methanol decomposition reaction, and Eq. (11) is the conversion reaction of water and gas. The dominant products in reformate gas are H_2 and CO_2, while a small percentage of CO is also produced.

4. Establishment of the computational model for cylindrical microreactor

4.1. Solution of the computational model for cylindrical microreactor

Table 3 shows input data, including PPI of foam metal, reaction temperature of methanol steam reforming for hydrogen production, injection velocity of methanol and water mixture, and catalyst loading of foam metal; output data include methanol conversion from 76 sets of experimental data of cylindrical microreactor for methanol steam reforming for hydrogen production. Appendix B shows the main MATLAB program designed to establish the computational model for methanol steam reforming performance of cylindrical microreactor for hydrogen production by utilising the MATLAB neural network toolbox.

Table 3. Seventy-six sets of input and output data of cylindrical microreactor

Experimental number	Input data				Output data
	PPI	Reaction temperature (℃)	Injection velocity（ml/h）	Catalyst loading (g)	Methanol conversion (%)
1	50	380	6	1.2	90.00
2	50	380	10	1.2	89.00
3	50	380	18	1.2	85.00
4	50	380	14	0.9	83.80
5	50	380	14	1.2	88.00
6	50	380	6	0.3	84.00
7	50	360	14	0.9	82.23
8	50	360	14	1.2	87.50
9	50	360	6	0.3	83.92
10	50	360	6	1.2	90.00
11	50	340	10	0.6	75.26
12	50	340	10	0.9	86.00
13	50	340	10	1.2	88.00
14	50	340	6	1.2	90.00
15	50	320	6	0.3	80.95
16	50	320	10	0.3	71.57
17	50	320	10	0.6	73.91
18	50	320	6	1.2	89.00
19	50	300	6	1.2	88.80
20	70	380	6	1.2	95.00
21	70	380	6	0.9	88.00
22	70	380	18	0.9	86.00
23	70	380	6	1.2	90.00
24	70	380	10	1.2	89.50
25	70	380	14	1.2	88.80
26	70	380	18	1.2	88.00
27	70	380	10	0.3	82.00
28	70	380	6	0.6	92.00
29	70	380	18	0.6	84.00
30	70	380	14	0.6	86.00
31	70	360	18	0.3	75.73
32	70	340	10	0.6	88.00
33	70	340	6	1.2	93.50
34	70	320	6	1.2	93.00
35	70	320	18	0.9	84.50
36	70	320	6	0.3	83.00
37	70	320	10	0.9	88.00
38	70	300	6	1.2	92.00
39	90	380	10	0.3	86.00
40	90	380	6	0.6	90.00
41	90	380	6	1.2	100.00
42	90	360	6	0.6	89.84
43	90	360	10	0.3	86.00
44	90	360	14	1.2	97.80

45	90	360	10	1.2	98.20
46	90	360	18	1.2	96.00
47	90	360	6	1.2	100.00
48	90	340	18	0.9	95.00
49	90	340	10	0.9	96.30
50	90	340	6	1.2	100.00
51	90	320	14	1.2	97.00
52	90	320	18	0.3	81.00
53	90	320	14	0.3	83.00
54	90	320	10	0.3	84.00
55	90	320	6	0.3	86.00
56	90	300	10	0.3	84.00
57	90	300	6	1.2	100.00
58	110	380	6	0.9	100.00
59	110	380	18	1.2	100.00
60	110	380	14	0.6	94.00
61	110	380	6	1.2	100.00
62	110	360	10	1.2	100.00
63	110	360	10	0.9	98.00
64	110	360	6	0.6	95.00
65	110	360	14	1.2	100.00
66	110	360	18	1.2	100.00
67	110	360	10	1.2	100.00
68	110	360	6	1.2	100.00
69	110	340	14	0.3	90.00
70	110	320	18	0.6	93.00
71	110	320	6	0.3	92.00
72	110	320	18	0.3	88.00
73	110	320	14	0.3	89.80
74	110	320	10	0.3	90.50
75	110	300	6	1.2	100.00
76	110	300	10	1.2	100.00

4.2. Experimental verification of the computational model for cylindrical microreactor and universal applicability verification of the mathematical model for different microreactors

Fig.7 indicates methanol conversion comparison of cylindrical microreactor between the computational model and experiment under different injection velocities of the methanol and water mixture, in the condition of 50 PPI, 330℃ reaction temperature, and 1.2-g catalyst loading. Table 4 shows that the $E_{\max}$ is 3.2%.

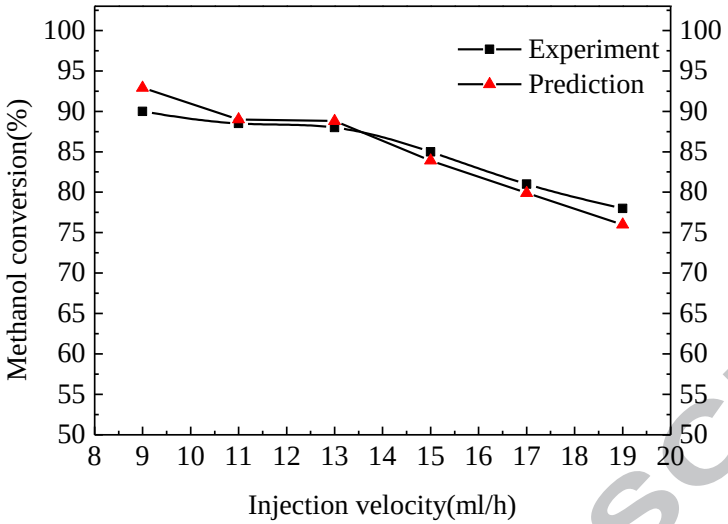

Fig. 7. Methanol conversion comparison of cylindrical microreactor between the computational model and experiment under different injection velocities

Fig.8 indicates the methanol conversion comparison of cylindrical microreactor between the computational model and experiment under different reaction temperatures of methanol steam reforming for hydrogen production, in the condition of 70 PPI, 10-ml/h injection velocity, and 0.9-g catalyst loading. Table 5 shows that the E_{max} is 3.3%.

Fig.9 shows the methanol conversion comparison of cylindrical microreactor between the computational model and the experiment under different injection velocities and reaction temperatures, in the condition of 90 PPI and 0.6-g catalyst loading. Table 6 shows that E_{max} is 2.9%.

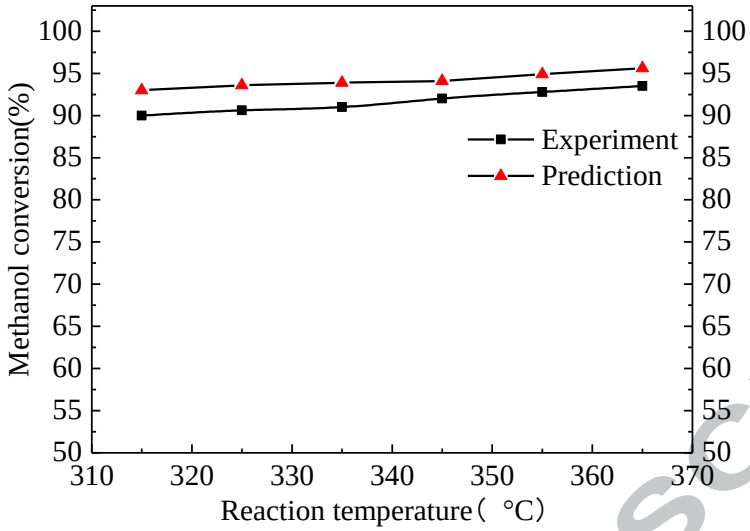

Fig.8. Methanol conversion comparison of cylindrical microreactor between the computational model and experiment under different reaction temperatures

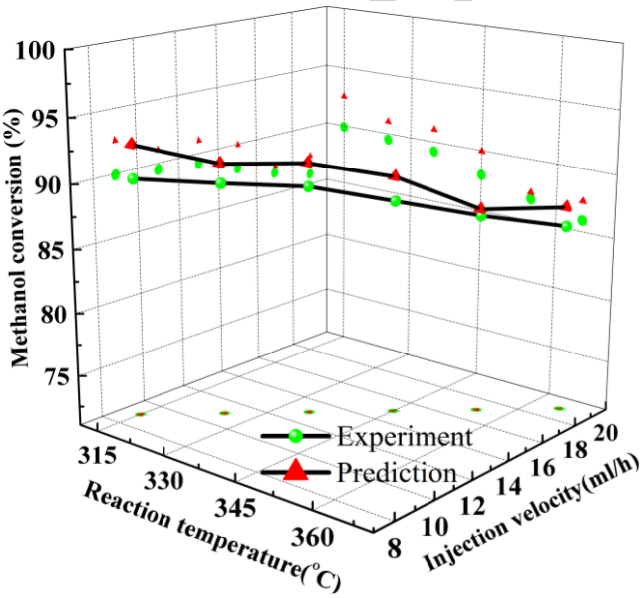

Fig. 9. Methanol conversion comparison of cylindrical microreactor between the computational model and experiment under different injection velocities and reaction temperatures (90 PPI)

Fig.10 shows the methanol conversion comparison of cylindrical microreactor between the computational model and the experiment under different injection velocities and reaction temperatures, in the condition of 110 PPI and 0.3-g catalyst loading. Table 7 shows that E_{max} is 3.2%.

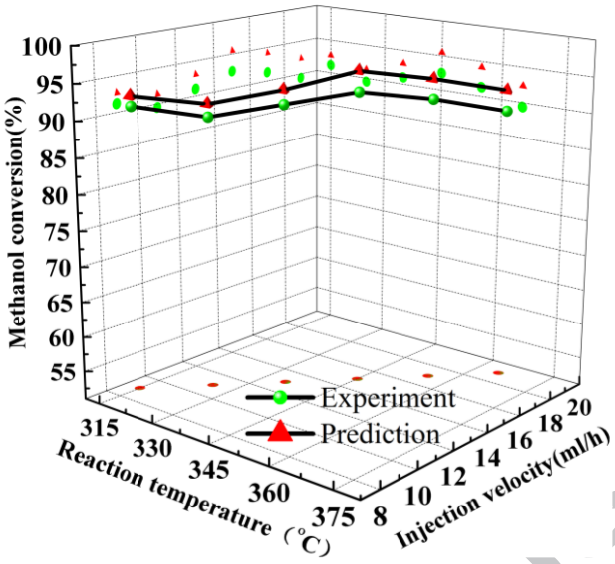

Fig.10. Methanol conversion comparison of cylindrical microreactor between the computational model and experiment under different injection velocities and reaction temperatures (110 PPI)

Table 4. Maximum error rate for predicted methanol conversion of the computational model for cylindrical microreactor under different injection velocities

Reaction parameters / Different injection velocities	50 PPI	330℃reaction temperature	1.2-g catalyst loading
E_{max}	3.2%		

Table 5. Maximum error rate for predicted methanol conversion of the computational model for cylindrical microreactor under different reaction temperatures

Reaction parameters / Different reaction temperatures	70 PPI	10-ml/h injection velocity	0.9-g catalyst loading
E_{max}	3.3%		

Table 6. Maximum error rate for predicted methanol conversion of the computational model for cylindrical microreactor under different injection velocities and reaction temperatures (90 PPI)

Reaction parameters / Different injection velocities and reaction temperatures	90 PPI	0.6-g catalyst loading
E_{max}	2.9%	

Table 7. Maximum error rate for predicted methanol conversion of the computational model cylindrical microreactor under different injection velocities and reaction temperatures (110 PPI)

Reaction parameters / Different injection velocities and reaction temperatures	110 PPI	0.3-g catalyst loading
E_{max}	3.2%	

The predicted methanol conversion of cylindrical microreactor is broadly in line with the experimental methanol conversion in the same reaction conditions, the partial deviation is not big, and the maximum $E_{\max}$ is only 3.3%. The results above reveal the good predictive capability and reliability of the computational model for cylindrical microreactor for reaction performance of methanol steam reforming for hydrogen production.

The successful application of the computational model in the plate microreactor and cylindrical microreactor indicates the reliability and universal applicability of the mathematical model for different microreactors.

Here, 30 sets of experimental data were used to obtain the computational model for plate microreactor, 76 sets of experimental data were used to establish the computational model for cylindrical microreactor. Compare with cylindrical microreactor, the E_{max} from plate microreactor is bigger. It can be drawn the conclusion that the more the experimental data, the better the reliability of the computational model, when the mathematical model for different microreactors is applied in the specific testing system for specific methanol steam reforming microreactor for hydrogen production.

5. Effects of reaction parameters on reaction performance for cylindrical microreactor

The effects of the reaction parameters of methanol steam reforming for hydrogen production on the methanol conversion are studied relying on the computational model for cylindrical microreactor for methanol steam reforming for hydrogen production. The reaction parameters consist of the PPI of foam metal, reaction temperature of methanol steam reforming for hydrogen production, injection velocity of the methanol and water mixture, and the catalyst loading of foam metal. Then, the mechanisms of the reaction parameters on reaction performance are studied, based on the influences of the reaction parameters of methanol steam reforming for hydrogen production on methanol conversion, combined with the structural characteristics of the reaction support of foam metal and the reaction principal of methanol steam reforming for hydrogen production.

The effect of PPI on methanol conversion is studied, relying on the computational model for cylindrical microreactor. As an example, in the condition of 380℃ reaction temperature, 5-ml/h injection velocity, and 0.3-g catalyst loading, as shown in Fig. 11, when PPI is between 50 and 150, methanol conversion enhances with an increase in PPI. When PPI is greater than 150, methanol conversion can steadily reach 100%. Increasing PPI leads to enhanced methanol conversion.

Nevertheless, for the higher PPI, methanol conversion steadily remains at 100%.

The mechanism of PPI on methanol steam reforming for hydrogen production is obtained by using the effect of PPI on methanol conversion, combined with the structural characteristics of the reaction support of foam metal and the principle of methanol steam reforming reaction. The bigger the PPI, the higher the surface-to-volume ratio of foam metal, the smaller the pore size of foam metal, the more dense the pore distribution, the more uniform the spatial distribution of the catalyst coated with foam metal, the more the amount of reaction gas contacting the catalyst in the condition of a certain amount of reaction gas, and the more the reacted percentage of reaction gas [30]. Hence, when PPI increases to a certain value, the distribution of the catalyst on foam metal is uniform enough, the reaction gas has fully reacted, and the methanol conversion remains 100%.

The effect of reaction temperature on methanol conversion is studied relying on the computational model for cylindrical microreactor. As an example, in the condition of 70 PPI, 10-ml/h injection velocity, and 0.9-g catalyst loading, as shown in Fig. 12, when the reaction temperature is between 335 and 385℃, methanol conversion is enhanced as reaction temperature increases. When the reaction temperature is greater than 385℃, methanol conversion reduces with increasing reaction temperature. Methanol conversion is enhanced as reaction temperature increases. For the higher reaction temperature, however, methanol conversion reaches a relative maximum and methanol conversion reduces with an increase in reaction temperature.

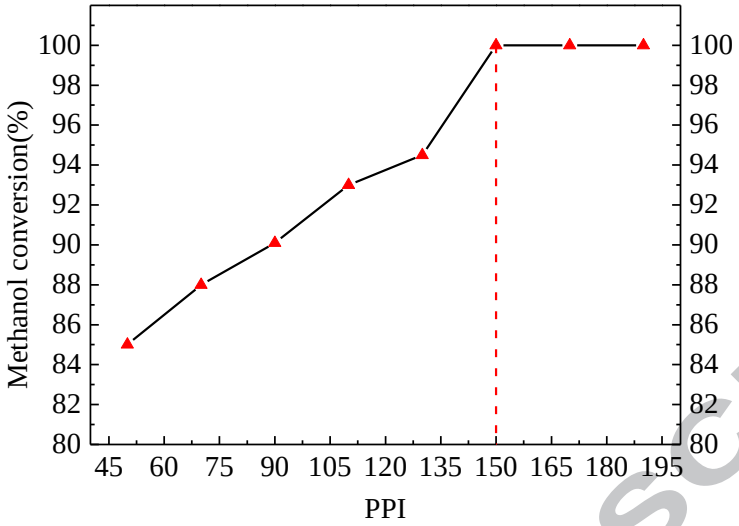

Fig.11. Relationship between PPI and methanol conversion

The mechanism of reaction temperature on methanol steam reforming for hydrogen production is obtained, based on the effect of the reaction temperature on methanol conversion, combined with the principle of the methanol steam reforming reaction. When the reaction temperature is lower than the reaction condition temperature of methanol steam reforming for hydrogen production, the reaction cannot be carried out [4]. The higher the reaction temperature, the more active the reaction gas, the greater the amount of reaction gas contacting the catalyst in the condition of a certain amount of reaction gas, and the greater the reacted percentage of the reaction gas [31]. If the reaction temperature is too high, the activity of the catalyst will be reduced, and the reacted percentage of the reaction gas will decrease [32]. Accordingly, when the reaction temperature is at a certain value, methanol conversion reaches the highest value.

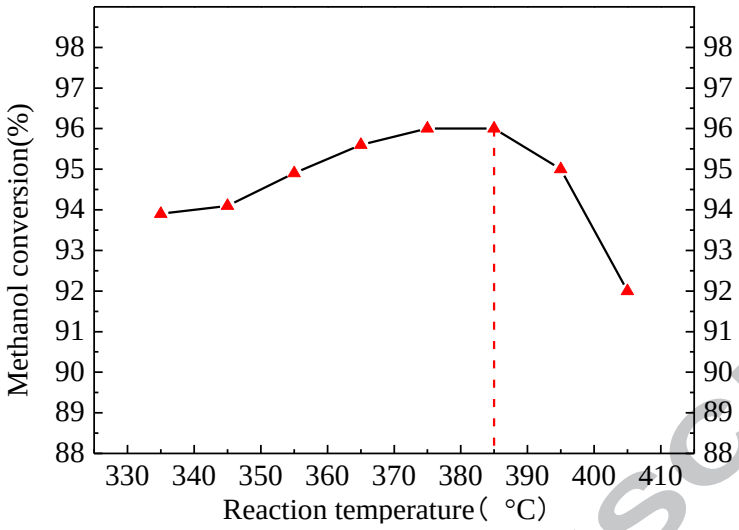

Fig.12. Relationship between reaction temperature and methanol conversion

The effect of injection velocity on methanol conversion is studied, based on the computational model for cylindrical microreactor. As shown in Fig. 13, when injection velocity is between 9 and 23 ml/h, methanol conversion reduces with injection velocity increasing, in the condition of 50 PPI, 330℃ reaction temperature, and 1.2-g catalyst loading. Methanol conversion decreases with the increase in injection velocity.

The mechanism of injection velocity on methanol steam reforming for hydrogen production is determined, relying on the effect of injection velocity on methanol conversion, combined with the principle of the methanol steam reforming reaction. The faster the injection velocity is, the shorter the residence time of the reaction gas in the reaction support of foam metal, the less the amount of reaction gas contacting the catalyst in the condition of a certain amount of reaction gas, and the less the reacted percentage of the reaction gas [33]. Thus, when the injection velocity of the methanol and water mixture increases, methanol conversion reduces.

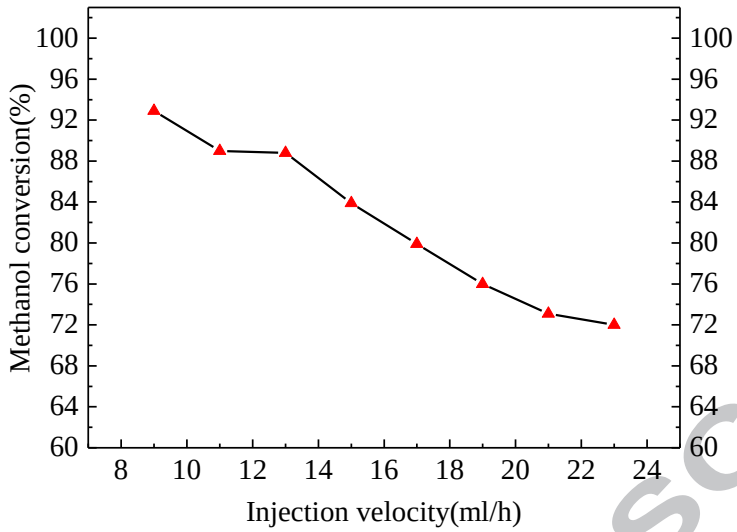

Fig.13. Relationship between injection velocity and methanol conversion

The effect of catalyst loading on methanol conversion is studied based on the computational model for cylindrical microreactor. As an example, in the condition of 90 PPI, 380℃ reaction temperature, and 5-ml/h injection velocity shown in Fig. 14, when catalyst loading is between 0.3 and 1.5 g, methanol conversion enhances as catalyst loading increases. When catalyst loading is greater than 1.5 g, methanol conversion can steadily reach 100%. Increasing catalyst loading leads to an enhancement in methanol conversion. However, for the higher catalyst loading, methanol conversion steadily remains at 100%.

The mechanism of catalyst loading on the methanol steam reforming for hydrogen production is obtained, based on the effect of catalyst loading on methanol conversion, combined with the principle of the methanol steam reforming reaction. The greater the amount of catalyst loading, the greater the amount of reaction gas contacting the catalyst in the condition of a certain amount of reaction gas and the more the reacted percentage of reaction gas [34]. However, when the amount of catalyst loading increases to a certain value, the reaction gas has been fully reacted, so methanol conversion remains 100%.

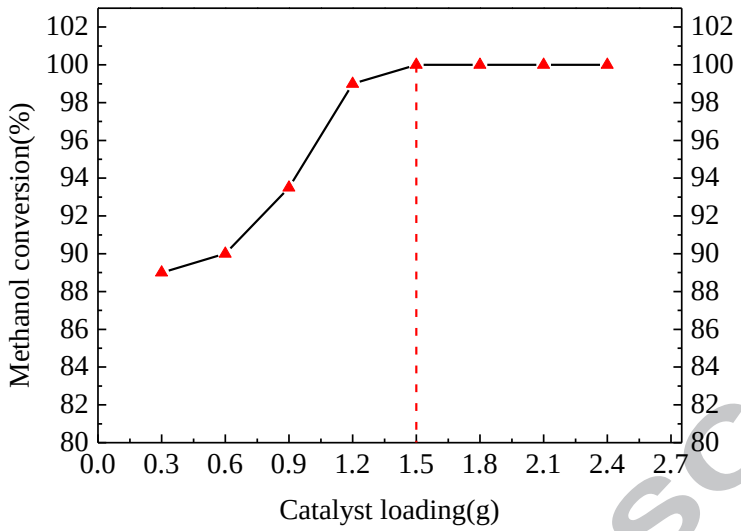

Fig.14. Relationship between catalyst loading and methanol conversion

6. Optimisation of reaction parameters for cylindrical microreactor

The optimum reaction parameters for cylindrical microreactor are obtained by invoking the MATLAB neural network toolbox, combined with the GA and the designed MATLAB program, and relying on the computational model for cylindrical microreactor. The reliability of the optimum reaction parameters is validated by experiments.

6.1. Genetic algorithm

The GA is an optimisation method to simulate natural selection and genetic mechanisms based on Darwin's theory of biological evolution and Mendel's genetic theory [35-38]. Individuals with good fitness are preserved, and a new group is formed by selecting individuals through genetic selection, crossover, and mutation, using the fitness function. The new group inherits the information from the previous-generation group, and it is more fit than the previous-generation group. Individual fitness in the group is continuously optimised until a certain condition is met, and the optimum individual fitness in the group is found.

6.2. Application of GA in reaction parameter optimisation

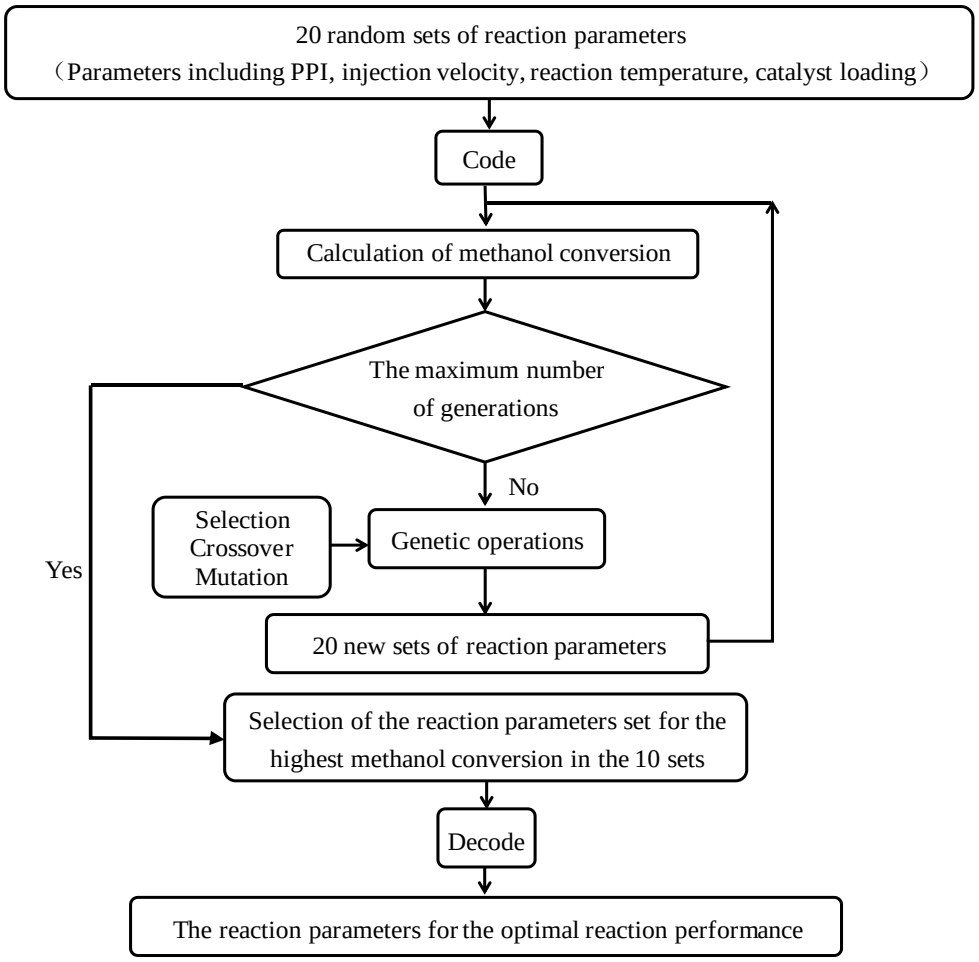

Fig.15. Application procedure of the GA in reaction parameter optimisation of methanol steam reforming for hydrogen production

Fig.15 exhibits the application procedure of the GA in reaction parameters optimisation of methanol steam reforming for hydrogen production. Firstly, 20 sets of reaction parameters are randomly generated, including PPI, injection velocity, reaction temperature, and catalyst loading. The 20 sets of reaction parameters are coded for computerised identification and processing. The fitness values of the 20 sets of reaction parameters, i.e. methanol conversions, are calculated, using the defined fitness function. Subsequently, the 20 sets of methanol conversions are processed to determine whether they meet the preset requirements. If they are satisfied, the reaction parameters

for the highest methanol conversion are selected from the 20 sets of reaction parameters. If they are not satisfied, the 20 sets of reaction parameters are inherited, and the 20 new sets of reaction parameters are generated. Then, the new sets of methanol conversions are processed to determine whether they meet the preset requirement of the maximum number of generations. If they are satisfied, the reaction parameters for the highest methanol conversion are selected from the 20 new sets of reaction parameters. If they are not satisfied, genetic operations are done continuously until the 20 sets of methanol conversions meeting the preset requirements are found. Finally, the reaction parameters for the highest methanol conversion are decoded, and the optimum reaction parameters are acquired.

6.3. Solution of optimum reaction parameters for cylindrical microreactor

Appendix C shows the main MATLAB program to obtain the optimum reaction parameters for methanol steam reforming for hydrogen production. The optimum reaction performance of 100% is gained, while the reaction parameters are 109.6 PPI, 311.6℃ reaction temperature, 6.06-ml/h injection velocity, and 0.89-g catalyst loading, by using the designed MATLAB program to process the computational model for the specific testing system. Because of the difficult of the manufacture for the foam metal of 109.6 PPI, the foam metal of 110 PPI in the experiment for verifying the reliability of optimum reaction parameters is used. Therefore, the reaction parameters of 110 PPI, 311.6℃ reaction temperature, 6.06-ml/h injection velocity, and 0.89-g catalyst loading are used in the experiment, the methanol conversion is 100%. Table 8 reveals the reliability of the optimum reaction parameters. The study on the computational model for methanol steam reforming performance for hydrogen production has excellent guiding significance for optimising the methanol steam reforming performance for hydrogen production.

Table 8. Methanol conversion comparison between the computational model and experiment under the optimum reaction parameters

Samples	PPI	Reaction temperature (℃)	Injection velocity (ml/h)	Catalyst loading (g)	Methanol conversion (%)
Computational model	109.6	311.6	6.06	0.89	100.0
Experiment	110	311.6	6.06	0.89	100.0

7. Conclusions

Methanol steam reforming performance optimisation for hydrogen production was studied using an error backpropagation and a genetic algorithm (GA). The main conclusions can be drawn as follows:

(1) The established mathematical model for different microreactors had reliability and universal applicability, which could be applied in different methanol steam reforming microreactors for hydrogen production. When the mathematical model for different microreactors is applied in the specific testing system for specific methanol steam reforming microreactor for hydrogen production, the better reliability of the computational model for the specific testing system can be obtained with the more experimental data used to train the mathematical model for different microreactors.

(2) The computational model for cylindrical microreactor for the methanol steam reforming performance for hydrogen production was founded. The computational model for cylindrical microreactor used reaction parameters, including the pores per inch (PPI) of foam metal, the reaction temperature of methanol steam reforming for hydrogen production, the injection velocity of the methanol and water mixture, and the catalyst loading of foam metal as input. The computational model considered reaction results only including the methanol conversion as output. The

computational model for cylindrical microreactor had good predictive ability and reliability.

(3) The mechanisms of reaction parameters on reaction performance for cylindrical microreactor were examined as below. The bigger the PPI, the higher the surface-to-volume ratio of foam metal, the smaller the pore size of foam metal, the denser the pore distribution, the more uniform the spatial distribution of the catalyst plated with foam metal, the greater the amount of reaction gas contacting the catalyst in the condition of a certain amount of reaction gas, and the more the reacted percentage of reaction gas. When the reaction temperature is lower than the reaction condition temperature of methanol steam reforming for hydrogen production, the reaction cannot be done. The higher the reaction temperature, the more active the reaction gas, the greater the amount of reaction gas contacting the catalyst in the condition of a certain amount of reaction gas, and the more the reacted percentage of reaction gas. If the reaction temperature is too high, the activity of catalyst will reduce, and the reacted percentage of reaction gas will decrease. The faster the injection velocity, the shorter the residence time of the reaction gas in the reaction support of foam metal, the less the reaction gas contacting the catalyst in the condition of a certain amount of reaction gas, and the less the reacted percentage of reaction gas. The more the amount of catalyst loading, the more the amount of reaction gas contacting the catalyst in the condition of a certain amount of reaction gas, and the more the reacted percentage of reaction gas.

(4) The optimum reaction performance of 100% for cylindrical microreactor was gained in the condition of 109.6 PPI, 311.6℃ reaction temperature, 6.06-ml/h injection velocity, and 0.89-g catalyst loading by using the GA based on the computational model for cylindrical microreactor. The reliability of the optimum reaction parameters was verified by the experiments.

This proposed mathematical model for different microreactors and analysis procedure provides

a guidance for methanol steam reforming performance optimisation for hydrogen production, which can be also applied on the wide range of microchannel reactor optimization considering reaction parameters and performance with various operation conditions.

Acknowledgments

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Appendix A

```
clc

clear

load data input output

inputnum=4;

hiddennum=20;

outputnum=1;

input_train=input(1:27,1:4)';

input_test=input(28:30,1:4)';

output_train=output(1:27,1:1)';

output_test=output(28:30,1:1)';

[inputn,inputps]=mapminmax(input_train);

[outputn,outputps]=mapminmax(output_train);

net=newff(inputn,outputn,hiddennum);

net.trainParam.epochs=100000;

net.trainParam.lr=0.1;

net.trainParam.goal=0.1;

net=train(net,inputn,outputn);

inputn_test=mapminmax('apply',input_test,inputps);

an=sim(net,inputn_test);

test_simu=mapminmax('reverse',an,outputps);
```

```
error=test_simu-output_test;

save data1 net inputps outputps
```

Appendix B

```
clc

clear

load data input output

inputnum=4;

hiddennum=20;

outputnum=1;

input_train=input(1:76,1:4)';

input_test=input(77:86,1:4)';

output_train=output(1:76,1:1)';

output_test=output(77:86,1:1)';

[inputn,inputps]=mapminmax(input_train);

[outputn,outputps]=mapminmax(output_train);

net=newff(inputn,outputn,hiddennum);

net.trainParam.epochs=100000;

net.trainParam.lr=0.1;

net.trainParam.goal=0.1;
```

```
net=train(net,inputn,outputn);

inputn_test=mapminmax('apply',input_test,inputps);

an=sim(net,inputn_test);

test_simu=mapminmax('reverse',an,outputps);

error=test_simu-output_test;

save data net inputps outputps
```

Appendix C

```
clc

clear

load data net inputps outputps

maxgen=100000;

sizepop=20;

pcross=[0.4];

pmutation=[0.2];

lenchrom=[1 1 1 1];

bound=[30 180;260 450;2 36;0.3 2.0];

individuals=struct('fitness',zeros(1,sizepop),'chrom',[]);

avgfitness=[];

bestfitness=[];
```

```
bestchrom=[];
for i=1:sizepop
individuals.chrom(i,:)=Code(lenchrom,bound);
x=individuals.chrom(i,:);
individuals.fitness(i)=fun(x);
end
[bestfitness bestindex]=max(individuals.fitness);
bestchrom=individuals.chrom(bestindex,:);
avgfitness=sum(individuals.fitness)/sizepop;
trace=[avgfitness bestfitness];
for i=1:maxgen
individuals=Select(individuals,sizepop);
avgfitness=sum(individuals.fitness)/sizepop;
individuals.chrom=Cross(pcross,lenchrom,individuals.chrom,sizepop,bound);
individuals.chrom=Mutation(pmutation,lenchrom,individuals.chrom,sizepop,[i maxgen],bound);
for j=1:sizepop
x=individuals.chrom(j,:);
individuals.fitness(j)=fun(x);
end
[newbestfitness,newbestindex]=max(individuals.fitness);
```

```
[worestfitness,worestindex]=min(individuals.fitness);

if bestfitness<newbestfitness

bestfitness=newbestfitness;

bestchrom=individuals.chrom(newbestindex,:);

end

individuals.chrom(worestindex,:)=bestchrom;

individuals.fitness(worestindex)=bestfitness;

avgfitness=sum(individuals.fitness)/sizepop;

trace=[trace;avgfitness bestfitness];

end

[r c]=size(trace);

plot([1:r]',trace(:,2),'r-');

title('fitness curve','fontsize',12);

xlabel('evolution population','fontsize',12);ylabel('fitness','fontsize',12);

axis([0,100000,0,150])

disp('fitness      variable');

x=bestchrom;

disp([bestfitness x])
```

Highlights

>Error backpropagation algorithm is used to built general mathematical model.>Computational model is established based on general mathematical model.>Computational model shows good reliability and predictive ability. >Relations between reaction parameters and performance are studied. > Optimum reaction parameters are obtained by using genetic algorithm.

Graphical abstract