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Neural network aided design of Pt-Co-Ce/Al_2O_3 catalyst for selective CO oxidation in hydrogen-rich streams

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Abstract

In this study, the design of Pt-Co-Ce/Al_2O_3 catalyst for the low temperature CO oxidation in hydrogen streams was modeled using artificial neural networks. The effects of five design parameters, namely Pt wt.%, Co wt.%, Ce wt.%, calcination temperature and calcination time, on CO conversion were investigated by modeling the experimental data obtained in our laboratory for 30 catalysts. Although 30 points data set can be considered as small for the neural network modeling, the results were quite satisfactory apparently due to the fact that the experimental data generated with response surface method were well balanced over the experimental region and it was very suitable for neural network modeling. The success of neural network modeling was more apparent when the number of data points was increased to 120 by using the time on stream as another input parameter. It was then concluded that the neural network modeling can be very helpful to improve the experimental works in catalyst design and it may be combined with the statistical experimental design techniques so that the successful models can be constructed using relatively small number of data points.

Keywords: Artificial neural networks; Response surface method; CO oxidation; Pt-Co-Ce/Al_2O_3 catalyst

1. Introduction

The number of researches on clean energy sources and conversion technologies increases everyday as a result of increasing global concerns for the environmental protection. The fuel cells seem to be one of the most promising energy conversion technologies of the future due to their high transformation efficiency and low emission and noise. However, the safe storage of required hydrogen is not technically feasible yet. Therefore on-site production of hydrogen from a conventional fuel such as natural gas, gasoline or ethanol using a fuel processor seems to be the most feasible choice for the near future for small size applications such as houses, offices and transportation vehicles [1].

On the other hand, the hydrogen stream from a fuel processor contains 0.5–1.0% CO which is harmful to the anode catalysts of the PEM fuel cell (which is the most suitable fuel cell type for the mobile and small-medium size stationary applications) even at the trace levels, and it must be eliminated [2]. One of the most appropriate ways to clean up hydrogen stream from CO is low temperature selective oxidation of carbon monoxide using a catalyst. A noble metal, especially Pt based catalysts in the absence or presence of a promoter such as Co, Ce, Fe, Sn oxides have been extensively studied for this purpose [3–7].

Catalyst design is a tedious and a complex process involving many steps, many variables and complex interactions among these variables making the experimental studies quite expensive and time consuming. Therefore, effective computational methods such as artificial neural networks can be used to interpret the findings of experimental studies, to feed the results to the future experiments, and therefore to increase the efficiency and the effectiveness of the experimental work. Artificial neural network modeling, which was inspired from the functioning of biological nervous systems, is proved to be a powerful technique for complex and nonlinear problems with a strong ability to learn and predict [8]. Several successful applications of artificial neural networks on catalyst studies were reported in the literature [8–12].

In this work, artificial neural network models were developed to analyze the effects of design parameters on CO oxidation over "Pt-Co-Ce/Al_2O_3" catalyst using the experimental data obtained in our laboratory and reported in a previous communication [7].

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Multiple regression models were also developed and compared with neural network models since the experimental data was produced using response surface method, which is generally used to perform multiple regression analysis.

2. Computational work

2.1. Experimental data used

The preferential CO oxidation over Pt-Co-Ce/Al_2O_3 in hydrogen-rich stream had been experimentally studied in our group. The catalysts were prepared using incipient to wetness impregnation technique and tested in a microflow reaction system. The effects of Pt wt.%, Co wt.%, Ce wt.%, calcination temperature and calcination time on the catalyst performance was investigated using response surface method [13]. The experimental CO conversions at various times on stream were given in Table 1 for 30 different catalysts, whereas the details of the experimental work were discussed elsewhere [7].

2.2. Computational details

The artificial neural networks used in this work were created by writing computer codes in MATLAB 7.2. The logistic sigmoid function, which is a good choice for many nonlinear functions, was employed as the activation function, while the delta rule was applied as the error correcting rule, and the backpropagation algorithm was constructed as the learning algorithm to adapt the weights [14]. The experimental data were iterated randomly using randomly generated initial weights in the interval of −0.1 to +0.1. Each network topology was trained at an average of three times giving similar results.

As it is known, a network having few number of neurons may fail to model the relation between the input and the output parameters, while a network with too many number of neurons may over-fit the experimental data. Molga stated that the error for the training data decreases with the increasing network size while the error for the test data increases. In order to find an optimum neural network structure, the number of input data points should be several times higher than the total number of weights [15]. Hence, in this study, the model best simulating and generalizing the experimental results was searched by starting from small networks (small number of weights) and then enlarging the networks until the best model without over-fitting was achieved. Enlarging a network was done by either adding more neurons to the existing hidden layer or adding one more hidden layer to the network. Model validation was carried out on the

Table 1
CO conversions for various catalyst design conditions. The reaction conditions were kept constant at $T = 90\,^{\circ}C$, F/W (inlet gas flowrate/catalyst weight) = 24,000 $cm^3/(g\,h)$, 1.0% CO, 1% O_2, 60% H_2, He as balance [7]

Exp.#	Design parameters					CO conversion (%) at			
	Pt (wt.%)	Ce (wt.%)	Co (wt.%)	Calcination temperature (°C)	Calcination time (h)	30 min	60 min	90 min	120 min
1	1	5	2.5	500	3	100.0	73.7	52.5	47.4
2	1	2.5	0	500	3	2.4	10.7	7.9	14.7
3	1	2.5	2.5	500	1	39.5	35.2	27.5	26.8
4	1	2.5	2.5	500	3	55.0	59.9	49.9	47.6
5	1	2.5	2.5	500	3	54.5	41.9	44.5	39.9
6	0.6	1.25	1.25	550	2	27.5	29.2	22.1	22.1
7	0.6	1.25	1.25	450	4	28.8	28.7	23.4	19.1
8	0.6	3.75	3.75	550	2	100.0	100.0	66.8	57.4
9	1	2.5	5	500	3	55.7	53.5	39.3	32.2
10	1	2.5	2.5	500	3	37.9	28.0	21.5	22.2
11	1.4	3.75	3.75	450	2	85.7	100.0	100.0	100.0
12	1.4	1.25	3.75	550	2	64.4	50.7	44.1	40.5
13	1	2.5	2.5	600	3	23.6	20.2	17.3	16.5
14	1	2.5	2.5	500	3	55.9	41.2	41.8	32.1
15	1	0	2.5	500	3	21.3	30.5	23.9	19.9
16	1.4	1.25	3.75	450	4	100.0	74.5	65.2	59.4
17	1.4	3.75	1.25	450	4	67.2	66.7	67.7	59.7
18	0.6	1.25	3.75	450	2	16.0	24.3	26.8	25.7
19	1.8	2.5	2.5	500	3	100.0	100.0	100.0	100.0
20	0.6	3.75	1.25	450	2	46.6	32.1	25.3	20.8
21	1.4	1.25	1.25	450	2	100.0	100.0	100.0	100.0
22	0.2	2.5	2.5	500	3	2.4	12.8	15.3	16.2
23	0.6	3.75	1.25	550	4	24.9	20.0	21.9	17.6
24	1	2.5	2.5	400	3	52.4	41.1	26.2	30.5
25	0.6	3.75	3.75	450	4	76.8	43.3	37.0	35.6
26	1.4	1.25	1.25	550	4	100.0	70.2	59.4	55.4
27	1.4	3.75	3.75	550	4	31.7	26.9	23.9	23.3
28	1	2.5	2.5	500	5	79.7	27.9	24.7	23.9
29	1.4	3.75	1.25	550	2	95.3	85.4	60.3	54.4
30	0.6	1.25	3.75	550	4	0.0	10.0	0.0	0.0

neural network models by using the residual and the k-fold cross validation analyses [16].

The coefficient of determination (R^2), the adjusted coefficient of determination (R^2_{adj}) and the root mean square error (RMSE) were used to compare the various neural network topologies with each other and also with multiple regression models [17]. The corresponding equations for R^2, R^2_{adj} and RMSE are given below.

$$R^2 = 1 - \frac{\sum_1^n (p_i - t_i)^2}{\sum_1^n (t_i - \bar{t})^2} \quad (1)$$

$$R^2_{adj} = 1 - \frac{(1 - R^2)(n - 1)}{(n - k - 1)} \quad (2)$$

$$\text{RMSE} = \sqrt{\frac{1}{n}\sum_1^n (p_i - t_i)^2} \quad (3)$$

where p_i is the predicted, t_i is the target (experimental) value of CO conversion and $\bar{t}$ is the mean of target values; n is the total number of experiments and k is the total number of independent variables (number of weights).

The test of input significance, which indicates the relative impacts of the design variables over the output variable (CO conversion), was also performed using the method of "change of root mean square error" [18].

3. Results and discussion

3.1. Modeling CO conversion

Neural network modeling of CO oxidation over Pt-Co-Ce/Al_2O_3 was performed in two steps: First, the CO conversion data obtained at 60 min time on stream over 30 different catalysts were modeled using five design parameters (Pt wt.%, Co wt.%, Ce wt.%, calcination temperature and calcination time) as input parameters of the network. Second, the larger network models were constructed by using the time on stream as another input parameter. This increased the number of data points from 30 to 120, since the experimental conversion values were measured at four different times on stream (30, 60, 90 and 120 min).

3.1.1. Modeling CO conversion using 60 min data

Several neural networks consisting five input parameters and one output parameter (CO conversion labeled as X_1) with varying number of neurons in one or two hidden layers were trained and tested. In each neural network, the neurons named as bias had the constant value of 1.

R^2 and RMSE values for various neural network topologies are given in Fig. 1a and b, respectively. The full quadratic multiple regression model (FMR) is also included in the figure for comparison. R^2 value increases while RMSE value decreases expectedly with increasing size of the network, which is defined based on the total number of weights rather than the number of neurons. However, the change in both R^2 and RMSE is not significant for the topologies larger than the network having one hidden layer with three neurons. Since larger topologies also have the risk of over fitting; the cross validation analysis, which indicates the generalization ability of the network, was done only for the networks 5-3-1 and 5-2-2-1, both of which have sufficiently higher R^2 and lower RMSE values. The notation 5-3-1 represents a network having one input layer with five input parameters, one hidden layer with three neurons and one output neuron. Similarly, the network 5-2-2-1 has one input layer with five input parameters, two hidden layers with two neurons in each and one output neuron.

Among the various cross validation techniques, k-fold cross validation technique was employed. The method is applied by first dividing the whole data into k subsets randomly, then training the network k times using the k-1 subsets as the training data and the remaining one subset as the test data in each run. Thus, all the data points are eventually used for both training and testing and the generalization accuracy of the network is examined in the entire experimental region [16]. After applying this procedure to several different neural network structures, the network having the minimum average RMSE for k test sets is chosen as the one that has the highest generalization accuracy, and that best represents the experimental data points. Since the total number of experiments in our case was relatively small, a high k value of 15 was chosen first to keep the number of training data points high (28 experiments out of 30). However, two experiments for training can be considered as small, and there is a risk that the test error calculated may not represent the true value of the validation error of the network. Thereby, 6-fold cross validation was also performed to provide more test data in

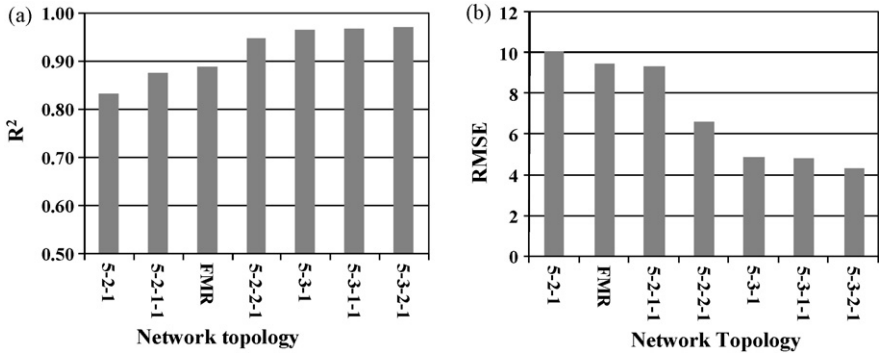

Fig. 1. R^2 (a) and RMSE (b) for different neural network topologies when all the data were used for training. FMR is the full quadratic multiple regression.

Table 2
15-fold cross validation results for 5-2-2-1 and 5-3-1 networks

Subset number	Experiments excluded	RMSE	
		5-2-2-1	5-3-1
1	12, 28	9.70	3.81
2	10, 14	9.48	7.58
3	4, 19	23.92	21.09
4	11, 18	1.38	0.14
5	3, 6	41.06	45.82
6	1, 27	18.08	18.64
7	7, 17	18.37	28.49
8	15, 16	28.94	20.21
9	5, 13	11.79	2.03
10	20, 24	10.90	32.82
11	21, 25	2.08	35.50
12	26, 29	34.05	37.10
13	9, 23	17.90	23.60
14	2, 30	21.11	25.00
15	8, 22	42.00	42.74
	Average	19.38	22.97

each subset (five experiments), though the number of training data slightly decreased (25 experiments).

Table 2 shows the comparison of the generalization ability of the 5-2-2-1 and 5-3-1 networks for 15-fold cross validation. RMSE in the table represents the root mean square errors calculated for the two experiments (test data) that had been excluded from the entire data. The average RMSE value of 5-2-2-1 network is fairly lower than that of the 5-3-1 network. The 6-fold cross validation results in Table 3 supports the same conclusion. Although, the average RMSE values are slightly higher for both networks in 6-fold validation, 5-2-2-1 network has still higher generalization capability. The structure of the 5-2-2-1 network is shown in Fig. 2.

The prediction accuracy of the 5-2-2-1 network was also fairly satisfactory as seen from the experimental versus predicted CO conversion plot in Fig. 3, when the entire data is used for training. This is also evident from the R^2, R^2_{adj} and RMSE shown in Table 4. Values of the same parameters of multiple regression are also presented for comparison. The R^2 value of 0.947 of the neural network is well above the value of 0.887 of the full quadratic model. The R^2_{adj} is also much closer to the R^2 for the neural network model indicating that over-fitting is less significant than the full quadratic regression model [17].

Table 3
6-fold cross validation results for 5-2-2-1 and 5-3-1 networks

Subset number	Experiments excluded	RMSE	
		5-2-2-1	5-3-1
1	10, 12, 14, 19, 28	11.07	18.87
2	3, 4, 6, 11, 18	13.73	18.82
3	1, 7, 16, 17, 27	12.33	20.83
4	5, 13, 15, 20, 24	33.16	29.33
5	21, 23, 25, 26, 29	39.78	44.96
6	2, 8, 9, 22, 30	20.37	13.73
	Average	21.74	24.42

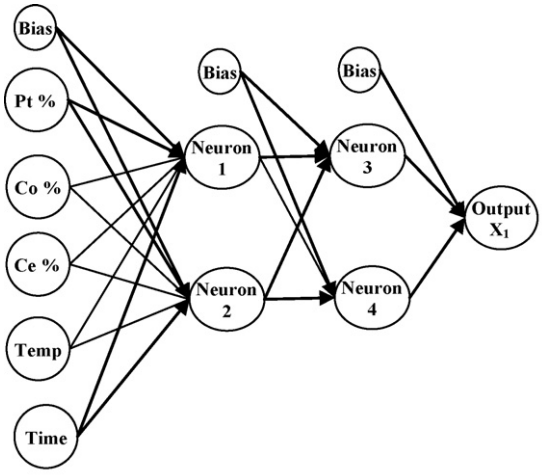

Fig. 2. Neural network architecture used for CO conversion (X_1) at 60 min.

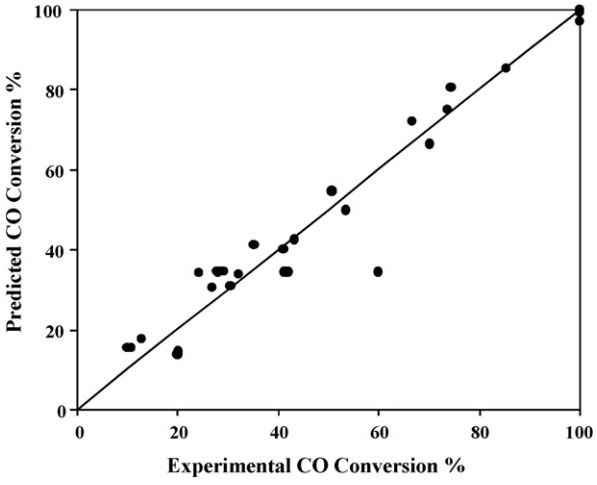

Fig. 3. Comparison of experimental and predicted CO conversions for 5-2-2-1 neural network model at 60 min when all the data were used for training.

Although 30 experimental data points were not large enough to construct a network with five input parameters, the results for the 5-2-2-1 network were considerably successful probably due to the fact that the data were generated using the response surface method. Apparently the well balanced distribution of the data points over the entire experimental region helped to create a network that represents the experimental results fairly well. However, when all the excluded experiments (test data) used in 15-fold cross validation were combined and a residual analysis was performed, the errors of some experiments can be considered as quite high (Fig. 4). This indicates that the success of the

Table 4
Measures of regression for CO conversion at 60 min when all the data were used for training

	5-2-2-1 Neural network results	Multiple regression results
R^2	0.947	0.887
R^2_{adj}	0.809	0.590
RMSE	6.599	9.454

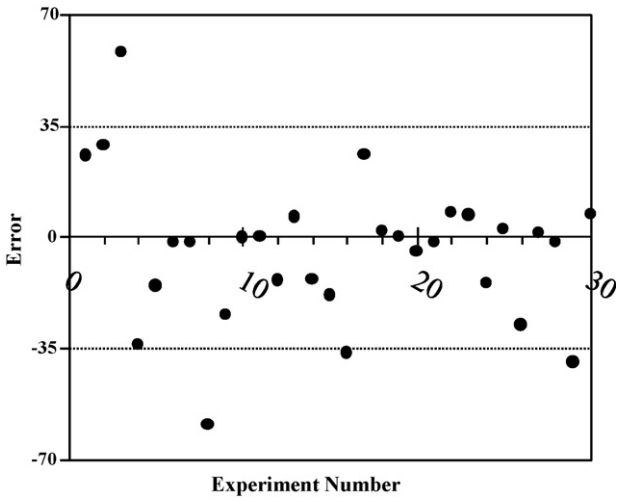

Fig. 4. Residual analysis of the test data for 5-2-2-1 neural network.

5-2-2-1 network is still limited and it cannot be improved further using the current number of data points; due to the fact that the smaller network structures have high training error while the larger ones cause over-fitting. Therefore, the time on stream was used as another input parameter to increase the experimental data points.

3.1.2. Modeling CO conversion by using the time on stream as another input parameter

Although the time on stream is not a design parameter, it can help us to see the effects of the design parameters better by increasing the number of data points from 30 to 120 since the experimental conversion values were measured at four different times on stream (30, 60, 90 and 120 min). Obviously, a neural network with six input parameters, trained by 120 points can represent the experimental data much better than a 5-parameter network trained by only 30 data points.

On the other hand, the change of time on stream does not change the main character of the catalyst like the other parameters. Hence, while doing cross validation analysis, all four data points obtained at four different times on stream for a specific catalyst should be excluded simultaneously. Otherwise, the exclusion of one data point will be compensated by the others and the cross validation results would seem much better than the true power of the model.

Labeling the experimental data set was done as follows: The first 30 experiments in this section represented the 30 min time on stream data in the order of appearance in Table 1. Likewise, numbers 31–60 were assigned to 60 min data, numbers 61–90 to 90 min and 91–120 to 120 min.

The performance of several neural networks together with full quadratic multiple regression is examined in Fig. 5. Similar to the 60 min models discussed in Section 3.1, each neuron added to any hidden layer contributes to the statistical success of the network as indicated by higher R^2 and lower RMSE.

15-fold cross validation method was again applied to 6-5-2-1 and 6-5-3-1 neural networks since the R^2 values for these models are significantly higher than those of any smaller networks and they are as high as that of the 6-5-4-1 neural network. The trainings were performed with 112 experiments and the network was forced to predict those eight experiments that had been excluded from the main data. The exclusion of the data was done by taking out the CO conversion values over the same catalyst at all time on stream values. For example the data points 12, 42, 72 and 102, all of which show the results of the 12th experiment at times 30, 60, 90 and 120 min were excluded at the same subset as seen in Table 5.

As it is seen in Table 5, the 6-5-2-1 neural network has slightly better generalization ability than the 6-5-3-1 neural network in predicting those points that were excluded from the main data. Although, RMSE values are very close for nearly all subsets, 6-5-3-1 network failed to validate the subset number 8.

Since the number of connection lines is too many, the schematic representation of the 6-5-2-1 network is shown in a simple form (Fig. 6). The additional neuron "sample" in the input layer represents the time on stream (minutes).

The high prediction accuracy of 6-5-2-1 network is seen from the predicted versus experimental CO conversion plot in Fig. 7 when all the data is used for training. Furthermore, the residual analysis using all the excluded experiments in 15-fold cross validation test (Fig. 8) indicates that the generalization ability of this network is also considerably better than that of the 5-2-2-1 network shown in Fig. 4.

Lastly, the superiority of the neural network against the full quadratic multiple regression model is more apparent in this

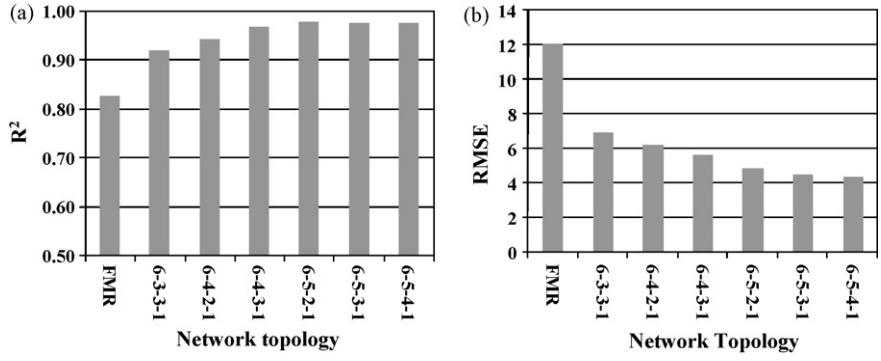

Fig. 5. R^2 (a) and RMSE (b) for different neural network topologies when all the data were used for training. The time on stream was also used as an input parameter.

Table 5
15-fold cross validation results for 6-5-2-1 and 6-5-3-1 networks (time on stream was also used as an input parameter)

Subset number	Experiments excluded	RMSE	
		6-5-2-1	6-5-3-1
1	12, 42, 72, 102, 28, 58, 88, 118	4.90	4.85
2	10, 40, 70, 100, 14, 44, 74, 104	10.25	11.30
3	19, 49, 79, 109, 4, 34, 64, 94	8.40	8.63
4	11, 41, 71, 101, 18, 48, 78, 108	5.33	5.50
5	3, 33, 63, 93, 6, 36, 6, 96	8.69	3.06
6	1, 31, 61, 91, 27, 57, 87, 117	3.51	6.31
7	17, 47, 77, 107, 7, 37, 67, 97	7.60	3.55
8	16, 46, 76, 106, 15, 45, 75, 105	6.76	34.20
9	13, 43, 73, 103, 5, 35, 65, 95	3.35	6.41
10	24, 54, 84, 114, 20, 50, 80, 110	7.63	4.24
11	21, 51, 81, 111, 25, 55, 85, 115	3.28	2.11
12	29, 59, 89, 119, 26, 56, 86, 116	6.69	5.51
13	23, 53, 83, 113, 9, 39, 69, 99	9.87	7.80
14	2, 32, 62, 92, 30, 60, 90, 120	5.55	6.30
15	8, 38, 68, 98, 22, 52, 82, 112	3.87	2.96
	Average	6.38	7.51

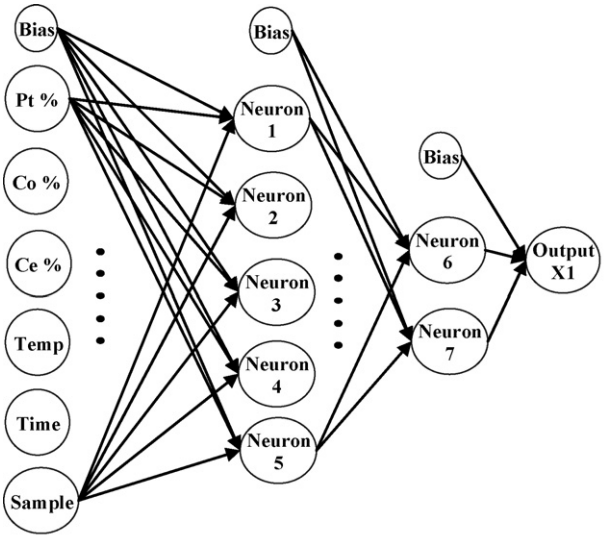

Fig. 6. Neural network architecture used for CO conversion (X_1) when the time on stream was also used as an input parameter.

case (Table 6). Although, multiple regression model was also improved with the addition of the time on stream as an input parameter (for example R^2_{adj} increased from 0.590 to 0.773), the improvement in neural network model is much more impressive.

Table 6
Measures of regression for CO conversion when the time on stream was also used as input parameter and all the data were used for training

	6-5-2-1 Neural network results	Multiple regression results
R^2	0.978	0.825
R^2_{adj}	0.961	0.773
RMSE	4.884	12.073

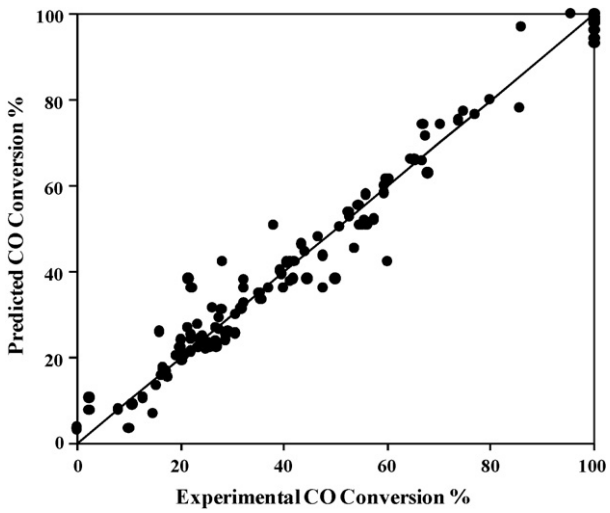

Fig. 7. Comparison of experimental and predicted CO conversions for 6-5-2-1 neural network model, when all the data were used for training.

3.2. Optimizing the catalyst design parameters

Normally, the catalyst preparation conditions, which maximize the CO conversion, should be found by optimizing neural network using an appropriate technique. However, the CO conversion in some experiments reached to 100% limits making this step unnecessary. Although, the reaction temperature could be decreased and/or F/W could be increased to lower the CO conversions well below 100% (so that numerical optimization would be possible), this was avoided to prevent the condensation of the water in the feed (due to low reaction temperature), and to be able to see the effects of design parameters on conversion as discussed in the following section. Instead, the catalyst number 21 in Table 1 was assumed to be the optimum one due to its lowest metal content among the other catalysts resulting 100% conversion, and used for the remaining part of the experimental work [7].

3.3. Effects of design parameters

The input significance analysis was performed first to see the relative importance of the design parameters on CO conversion for the 6-5-2-1 model using "the change of root mean square error" test [18]. The procedure for this test is to leave out each

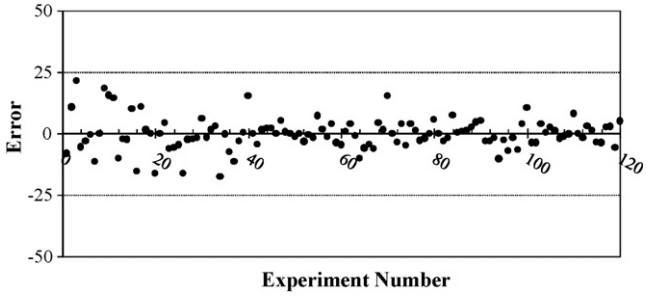

Fig. 8. Residual analysis for the test data of 6-5-2-1 neural network.

Table 7
Test of input significance using the change of RMSE method

Parameter excluded	RMSE	R^2	%Difference in RMSE
Pt	19.111	0.565	291.289
Co	7.949	0.925	62.759
Ce	8.725	0.884	78.635
Calc. temp.	6.389	0.924	30.807
Calc. time	5.941	0.926	21.635
Time on stream	10.252	0.906	109.913
No exclusion	4.884	0.978	

of the six input parameters one by one, then to check the level of decline of the RMSE of the training data caused by each of these exclusions.

The RMSE obtained after the exclusion of Pt is much higher than the values obtained excluding other parameters indicating that Pt wt.% is the most significant parameter (Table 7). This is an expected result and a further evidence for the success of the neural network modeling since Pt is the primary constituent of the catalyst and any change in its concentration is expected to have a significant change in the performance of the catalysts. Although Co and Ce weight percents seem to be more significant than calcination temperature conditions, the results are not as conclusive as that for Pt. The significance of time on stream should not be compared with the others since it is not a design parameter.

The effects of the five design parameters and the time on stream on CO conversion are given in Fig. 9. The solid lines represent the model prediction for the effect of each parameter while keeping the other design parameters at their mean values (Pt: 1 wt.%, Co and Ce: 2.5 wt.% each, calcination temperature: 500 °C, calcination time: 3 h) and the time on stream at 60 min. The available experimental data points are also presented in the same figure as circular symbols. The good agreement between the model prediction and the experimental data points is a further evidence for the success of the neural network modeling.

The CO conversion increases with increasing Pt wt.% significantly as expected (Fig. 9a). The effects of Co and Ce are also positive as seen from Fig. 9b and c, though they are not as effective as Pt. These are also expected results considering that these two metals were used to promote Pt [7]. Some intermediate levels of the calcination temperature and the calcination time, however, seem to be more suitable (Fig. 9d and e) for

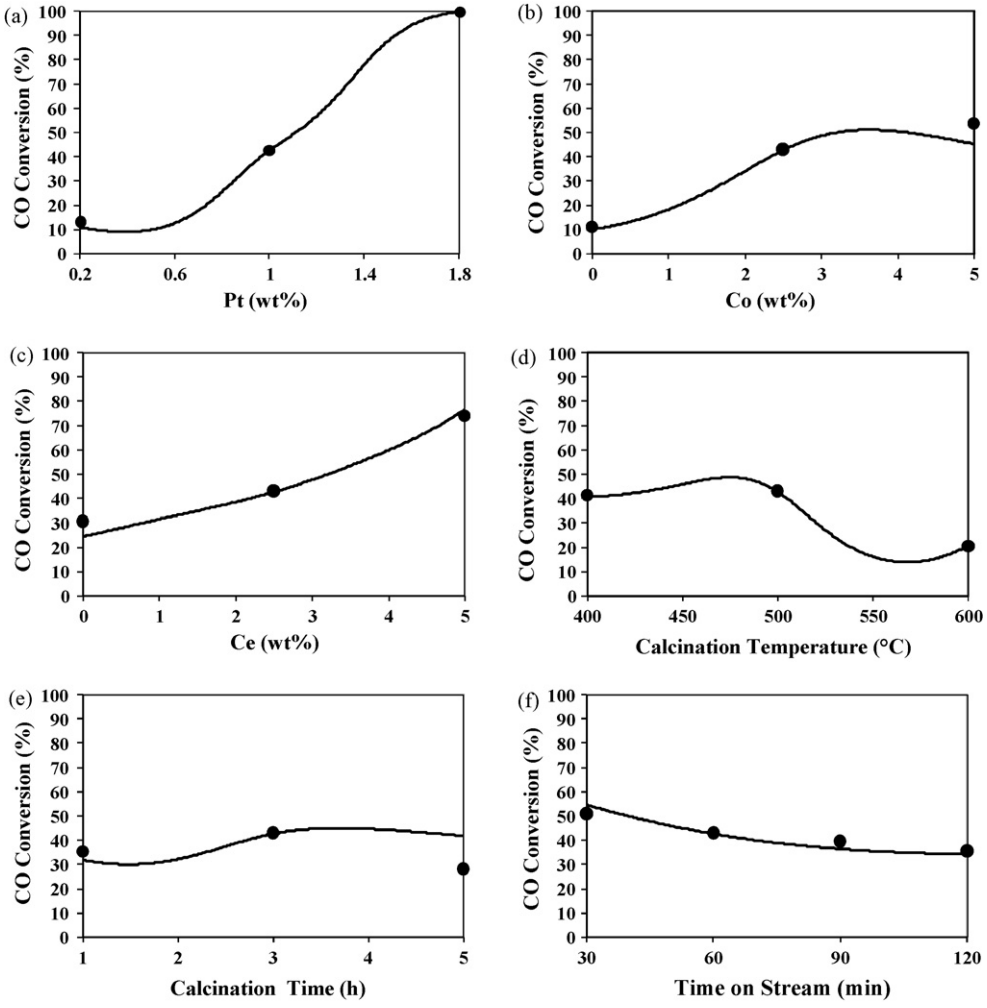

Fig. 9. Effects of input variables on CO conversion.

higher CO conversions. Finally the decrease of CO conversion with increasing time on stream in Fig. 9f may be attributed to the catalyst deactivation. Although the best catalysts (for example experiment 11, 19 and 21) keeps their activity up to 120 min, the most of the catalysts in Table 1 do lose their activity with increasing time on stream, resulting a decrease in CO conversion.

4. Conclusions

This paper examines the use of artificial neural network models for the design of Pt-Co-Ce/Al_2O_3 catalyst for the low temperature selective CO oxidation. The conclusions drawn can be summarized as follows:

- Among many neural networks topologies, the neural network model that best represented the 60 min CO conversion data was the 5-2-2-1 model. However, the model had some limitations in predicting the results of the data points that it had never encountered before.
- When the time on stream was treated as another input parameter, the data points increased from 30 to 120, giving way to enlarge the network without risk of over-fitting. The generalization power of the neural network (6-5-2-1) constructed this way increased dramatically.
- The most significant input parameter was found to be Pt wt.% as expected. The model predictions for the effects of input parameters on CO conversion were also in a good agreement with the experimental results as an evidence for the success of the neural network model.
- Although the number of data points was small, the neural network modeling has considerable success apparently due to the fact that the data is generated using statistically designed experiments.

To conclude; the neural network modeling can be very helpful to improve the experimental works in catalyst design and it may be combined with the statistical experimental design techniques so that highly successful models can be established using relatively small number of data points. The model best representing the data can be used to optimize the catalyst if the data points are suitable, to study the effects of the design parameters, and to predict the possible performances of the new catalysts without doing any new experiments. However, it should be noted again that the time on stream is not a design parameter, and the results should be treated accordingly.

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