

WEIGHTED OPTIMAL COMBINATION OF NEURAL NETWORKS FOR CATALYST DESIGN AND EVALUATION

Viswanathan Arcotumapathy and Adesoji A. Adesina*
Reactor Engineering and Technology Group, School of Chemical Sciences and
Engineering, University of New South Wales, NSW 2052 Australia

*Corresponding author. Tel.: +61 2 9385 5268; Fax: +61 2 9385 5966
Email: a.adesina@unsw.edu.au

Summary

Catalyst design and evaluation is a multi-factorial multi-objective optimization problem and the absence of well-defined mechanistic relationships between wide ranging input-output variables has stimulated interest in the application of artificial neural network for the analysis of the large body of empirical data available. However, single ANN models generally have limited predictive capability and insufficient to capture the broad range of features inherent in voluminous but dispersed data sources. In the present study, we have employed a strategy involving weighted optimal combination of statistically-derived single ANN models weighted in a multi-error sense. Data from 200 cases for catalytic methane steam reforming have been used to demonstrate the veracity and robustness of the integrated ANN modeling technique.

Keywords

Steam Reforming, Weighted Optimal Combination, Artificial Neural Network, Catalyst Design

1. Introduction

Steam reforming is the most important route for the production of bulk hydrogen to meet energy needs and manufacturing of industrial chemicals. Whilst a large knowledge-base exists for the design of steam reforming catalyst, there is a distinct capacity for improvement in the catalyst technology to meet the requirements of new process intensification strategies. The development of highly efficient steam reforming catalyst is a multi-input-multi-output problem which can be handled most appropriately via ANN analysis of existing information library¹⁻⁶. Factors influencing steam reforming and corresponding hydrocarbon conversion can be used as input and output respectively in an ANN to map the input-output relationship. The input variables comprised of Ni loading; catalyst support type - SBA-15, SiO₂, α -Al₂O₃, γ -Al₂O₃; promoters - B, Mo, ZrO₂, CeO₂; catalyst reduction temperature and reactor operating variables, viz; reforming temperature, pressure, S:C ratio and time-on-stream. Evidence-based optimization of multifactor catalyst design using ANN approach has been reported by our group⁷. However, the prediction ability of a single trained ANN is significantly less than that of a combination of several trained ANNs⁸ especially, if, as it frequently happens, more than one ANN network satisfy the performance criteria. While an ensemble of such networks has been used to secure satisfactory performance⁷, a more efficient global ANN may be

derived from a weighted combination of single-ANNs with higher model accuracy. This combination is termed, multiple-error based weighted optimal combination (ME-WOC) of ANNs. The ANNs used for ME-WOC are selected on the basis of performance errors of the trained networks.

2. Methodology

This study utilizes a multilayer feedforward network which works on Widrow-Hoff learning rule by applying Levenberg-Marquardt backpropagation algorithm, with a conjugate gradient approach⁹. The first step in the modeling exercise involves eleven single hidden layer networks. The number of neurons in the hidden layer in each network was increased to obtain a different network by increasing the number of neurons following a Fibonacci sequence. The data from several steam reforming studies were collected and rescaled to an interval [0, 1] for various input/output variables to minimize numerical errors. Over 1100 single hidden layer networks containing different number of neurons in the hidden layer were trained. The numerical procedure was carried out in MATLAB Neural Network ToolboxTM version 7.8.0.347 (R2009a). A two-way ANOVA test of mean squared error (MSE), root mean squared error (RMSE), sum of squared errors (SSE), mean absolute error (MAE), mean absolute percentage error (MAPE),

* To whom all correspondence should be addressed

validation error and testing error of the network performance against the number of neurons revealed that only SSE, RMSE and MAE were significant in explaining the network performance with respect to variation in the number of neurons in the hidden layer. Therefore, trained networks were ranked on the basis of SSE, RMSE and MAE. The ranking revealed that single hidden layer network with 55 neurons as best network. Then single hidden layer networks with neurons ranging from 34 to 89 (56 networks) were trained hundred times each. Thus, from a total of 5600 trained networks, the optimum number of neurons was selected with the aid of a two-way ANOVA as explained earlier. The optimum number of neurons which is 83 was then used to form one, two and three hidden layer networks. The resulting 3404 networks were trained and the best networks based on all the errors were combined to form a WOC. The minimum and maximum local weights were calculated based on minimum and maximum error obtained for each of the error indices respectively.

$$w_{I_{\min}} = \frac{I_{\min}}{\sum I} \quad (1)$$

$$w_{I_{\max}} = \frac{I_{\max}}{\sum I} \quad (2)$$

The minimum and maximum global weights were ($gw_{I_{\min}}$ and $gw_{I_{\max}}$) obtained by dividing the minimum and maximum local weights by the sum of all the minimum and maximum local weights respectively. Then the weighted best output is obtained using the following equation:

$$y_w^{-best} = \sum_I gw_{I_{\min}} y_I^{best} \quad (3)$$

Where I = SSE or MSE or RMSE or MAE or MAPE of the respective networks. Similarly weighted worst output is calculated using equation (3).

$$\alpha = \left(\frac{\left| \overline{y_w^{-best}} - \overline{y_w^{-worst}} \right|}{\left| \overline{y_I^{-best}} - \overline{y_I^{-worst}} \right|} \right) \quad (4)$$

The α value is calculated using equation (4) and if it is greater than unity then that value times the local weight is used to obtain the new global weight for the relevant error index and in turn the new weighted best output is found using equation (3). This procedure is continued until the MAPE of the WOC reaches a minimum.

3. Results and Discussion

The best single hidden layer neural network (83 neurons) has a MAPE of 0.01611, whereas the best network among multi-hidden layer networks was the network with three hidden layers with 83 neurons (Layer1: 14 neurons, Layer 2: 23 neurons and Layer 3: 46 neurons) and this network has a MAPE of 0.00685. The WOC gave the least MAPE

of 0.0059 and the error in fact decreased exponentially with respect to number of iterations (i.e. number of times α value changed). The WOC has higher model accuracy when compared to other single best networks and was able to predict the targets for any given set of input data as evidenced in the parity plot shown in Figure 1. The associated linear relation between 'y', the output of the network and 't', the target vector is given as:

$$y = 1.009t - 0.0058 \quad (5)$$

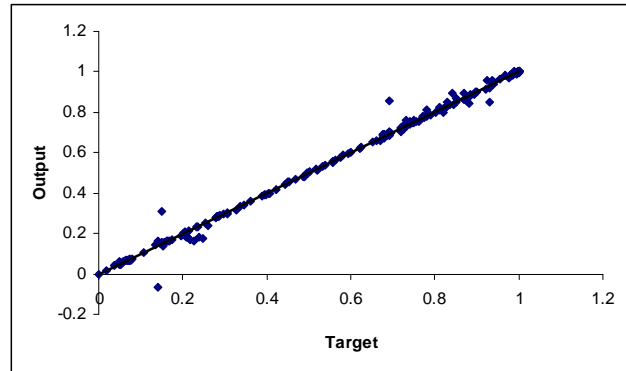


Figure 1. Parity plot for the ME-WOC neural network.

4. Conclusions

This study shows that even for a highly nonlinear process like steam reforming, an optimal linear combination of neural networks can be used for choosing optimal catalyst design and evaluation parameters from existing body of empirical data.

References

- (1) Wan, H.; Li, X.; Ji, S.; Huang, B.; Wang, K.; Li, C. Effect of Ni Loading and CexZr1-xO2 Promoter on Ni-Based SBA-15 Catalysts for Steam Reforming of Methane. *J. Nat. Gas Chem.* **2007**, *16*, 139.
- (2) Xu, J.; Chen, L.; Tan, K. F.; Borgna, A.; Saeys, M. Effect of boron on the stability of Ni catalysts during steam methane reforming. *J. Catal.* **2009**, *261*, 158.
- (3) Numaguchi, T.; Shoji, K.; Yoshida, S. Hydrogen effect on α -Al2O3 supported Ni catalyst for steam methane reforming reaction. *Appl. Catal., A.* **1995**, *133*, 241.
- (4) Maluf, S. S.; Assaf, E. M. Ni catalysts with Mo promoter for methane steam reforming. *Fuel.* **2009**, *88*, 1547.
- (5) Matsumura, Y.; Nakamori, T. Steam reforming of methane over nickel catalysts at low reaction temperature. *Appl. Catal., A.* **2004**, *258*, 107.
- (6) Hou, K.; Hughes, R. The kinetics of methane steam reforming over a Ni/ α -Al2O3 catalyst. *Chem. Eng. J.* **2001**, *82*, 311.
- (7) Chesterfield, D.; Adesina, A. A. Evidence-based design and optimization of titania photocatalyst via artificial neural network analysis. *Journal of Chemical Engineering of Japan.* **2009**, *42*, s185.
- (8) Hashem, S. Optimal linear combinations of neural networks. *Neural Networks.* **1997**, *10*, 599.

(9) Demuth, H.; Beale, M.; Hagan, M. Neural Network Toolbox™ 6. *User's Guide*. **2009**, 180.