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Artificial neural network modeling of forced cycling operation between propane steam reforming and CO_2 carbon gasifier

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ABSTRACT

This investigation has employed artificial neural network (ANN) modeling to describe the complex relationship between the forced cycling parameters and the reactor performance during periodic operation between propane steam reforming and CO_2—carbon gasifying agent. Experimental data from our laboratory were assessed against different ANNs and based on a 2-way ANOVA treatment of various error indices, a two-hidden layer network with 5 neurons emerged as the best model for both descriptive and predictive purposes. Cycle split has the most significant (85%) positive effect on the improvement in H_2 and CO production and the appearance of resonant peaks while cycle period appeared to have detrimental effect on product yield.

1. Introduction

Hydrocarbon steam reforming over Ni-based catalyst for H_2 or syngas production is industrially carried out using excess steam to avoid unwanted coke deposition [1]. The latter ultimately leads to poor reactor performance due to catalyst deactivation. In order to minimize energy costs (associated with steam generation) and improve reactor operation, CO_2 may be used as a carbon gasifier [2]. This has the added advantage of reducing overall greenhouse gas (GHG) emissions from the reforming plant. In a recent study [3], we demonstrated that periodic composition cycling as illustrated in Fig. 1, between the CO_2 gasifying agent (C_1) and propane steam reforming feed (C_2) has significant benefits in terms of increased syngas yield, tailored H_2:CO ratio suitable for downstream conversion in a gas-to-liquid (GTL) fuels plant and superior catalyst stability and longevity.

The time-average rate behaviour with respect to cycle frequency was characterized by resonant peaks depending on the cycle split (symmetry) employed. Modeling of periodically operated reactors is a multifarious exercise strongly reliant on the choice of reaction mechanism. Mihail and Paul proposed adsorption/desorption models [4] which Jain et al. [5,6] latter rejected as inadequate to explain the resonance effects seen in their experimental studies for SO_2 oxidation. Other attempts at providing mechanistically based models to describe the complex phenomena observed during forced cycling of catalytic reactors have also been met with mixed success. Indeed, Silveston et al. [7] have indicated that adsorption/desorption type models cannot accurately capture the nonlinear behaviour often implicated in periodically operated catalytic processes. However, optimal control of the forced cycling reactor requires a reliable model of its performance in order to harness the benefits (improved product yield and selectivity) associated with this dynamic operation. In this work, we have taken an artificial neural network (ANN) approach which does not require detailed understanding of the reaction mechanism. In particular, ANN permits the utilization of fewer latent variables which contain inherent information about the process. Indeed, ANN models are significantly more potent in capturing the process attributes with higher predictability and better description than multivariate data analysis as espoused by Bulsari [8].

Although a detailed mechanistic basis for this nonlinear behaviour is presently unavailable, a parsimonious model for process optimization may be secured via ANN analysis of existing data. ANN is a mathematical analogue of how the human brain recognizes and reproduces cause-effect relational patterns upon training (multiple-input–multiple-output systems). The ANN modeling of complex nonlinear behaviour or pattern in chemical engineering systems has been reported [8–13]. Even though nonlinear estimators like Holographic Research Strategy (HRS) and Genetic Algorithm (GA) are available, Tompos et al. [14] reported the robustness of these methods only after securing the relationship between the composition–activity of the catalysts via ANNs in their study. Moreover, they found that the predictive ability of the ANNs assisted in the enhancement of both HRS and GA methods to identify the optimized catalyst composition. The aim of the present work is to procure an ANN model to describe and predict the non-

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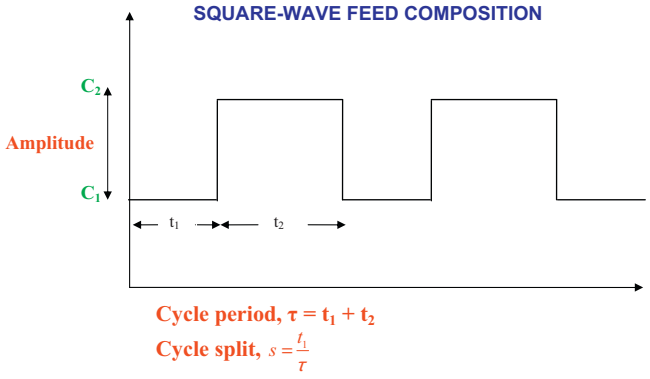

Fig. 1. Operating variables during forced cycling of a reactor.

linear dependency of key reaction metrics on cycle parameters (frequency and split) during periodic composition cycling between propane steam reforming feed and CO_2, a carbon gasifier without recourse to formal mechanistic details. In particular, since ANN is a mathematical imitation of the biological neural data processing operation, optimization of the network architecture used in this study was patterned after the Fibonacci search strategy which has meaningfully captured the behaviour in many natural ecosystems [15–17]. Moreover, the integer nature of optimum number of neurons in an ANN lends itself better to a Fibonacci-type algorithm rather than a continuous function-dependent method.

2. Theoretical basis and numerical procedure

2.1. Data scaling

The input variables were taken as the cycle period and split (fraction of the period spent under CO_2 feed for carbon gasification). In view of the difference in units, magnitude and range of the input variables, each variable needs to be rescaled so that values fall within the interval [0.01, 0.99]. The transformation was carried out using:

$$\hat{x} = \frac{x - m}{M - m} \tag{1a}$$

where $\hat{x}$ is the rescaled variable, with M and m chosen such that:

$$\frac{x_{\min} - m}{M - m} = 0.01 \quad \text{and} \quad \frac{x_{\max} - m}{M - m} = 0.99 \tag{1b}$$

where $x_{\min}$ and $x_{\max}$ are the minimum and maximum values of the original variable, x respectively. The variables in the output vector, H_2, CH_4 and CO were also similarly rescaled.

2.2. Methodology to select optimum neurons and architecture

The performance of an ANN is strongly dependent on the number of neurons employed and the network architecture. For instance, a simple feed-forward network with supervised learning algorithms such as back-propagation algorithm is competent in learning the relationship between the given inputs and targets for a nonlinear process data set [8–13]. Thus, a systematic procedure to optimize the number of neurons for building the multilayer feed-forward neural network to map the input–target relationship of the given data set is critical to the overall modeling exercise. The number of weights required to capture a reliable relationship between a given input and output is directly proportional to the number of neurons. ANN being a mimicry of the biological nervous system would be more appropriately optimized if the number of neurons were chosen in a manner that is patterned after natural evolutionary systems. Interestingly many naturally occurring networks (e.g. in forestry, microbiology and river systems—delta formation) containing optimum nodal points are known to be described by the Fibonacci sequence [15–17]. Consequently, it seems logical to derive the number of neurons in our ANN simulation of the optimal catalyst design using members of the Fibonacci series [18]. To the best of our knowledge, this is the first appropriation of the Fibonacci search strategy in ANN simulation of either chemically reactive systems or artificially contrived proceesses. The numerical procedure was carried out in MATLAB Neural Network Toolbox™ version 7.8.0.347 (R2009a) and may be summarized as:

Step 1: The MATLAB Neural Network Toolbox random data division function was used to partition the 34 data cases into 24, 5 and 5 for training, validation and testing of the networks respectively. A three-layer architecture (input, hidden and output layers) was considered with different artificial neural networks (ANNs) arising from variation in the number of neurons in the hidden layer. A tangent sigmoid transfer function was used for the neurons in the hidden layer. The number of neurons in the hidden layer for each ANN was chosen as terms of the Fibonacci series to introduce optimality in both the number of ANNs and computational effort. The total number of neurons required to obtain excellent model adequacy of an empirically fitted approximation and the ratio of number of neurons in the hidden layer to the number of data cases has to satisfy two opposing criteria detailed in Haykin [19] and Barron [20], namely:

(a) *Accuracy of best approximation*—the size of the hidden layer, N_{hl}, must be as large as possible in accordance with the universal approximation theorem and
(b) *Accuracy of the empirical fit to the approximation*—the ratio of the hidden layer size to number of training data cases, N_t, i.e. N_{hl}/N_t must be small.

In order to accommodate both criteria, we adopted the rule of thumb, $1 \leq N_{hl}/N_t \leq 10$ [9]. Given that N_t is 24 for this study, ANNs with 0, 1, 1, 2, 3, 5, 8, 13, 21, 34, 55, 89, 144 neurons in the hidden layer were chosen (total of 11 ANNs since the 2nd and 3rd ANNs with 1 neuron each, are mere repetitions while the 1st ANN with 0 neurons is physically inadmissible).

Levenberg–Marquardt algorithm was used to train each network based on the randomly selected 24 data cases. The random data division function was called 1000 times (cycles) to pick different combinations of the training, validation and testing data sets from the total data pool in each cycle. Thus, each of the 11 ANNs was trained 1000 times with different data set combinations resulting in a total of 11,000 trained, validated and tested ANNs. In this study after each cycle, i.e. for every ANN trained, after the entire training procedure was completed (R-square value ≥ 0.92 in nearly all cases), the weights and biases were used to simulate the network using all inputs (34 data cases) from the data set while the resulting output from the network after simulation and the targets from the data set were used to calculate the performance of the networks which was evaluated by different error indices, namely:

$$\text{Sum-of-squared error (SSE)} = \sum_{s=1}^{N} (y_s^o - t_s)^2 \tag{2}$$

$$\text{Mean squared error (MSE)} = \frac{1}{N} \sum_{s=1}^{N} (y_s^o - t_s)^2 \tag{3}$$

$$\text{Root-mean-squared error (RMSE)} = \sqrt{\frac{1}{N} \sum_{s=1}^{N} (y_s^o - t_s)^2} \tag{4}$$