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Elucidating the non-linear effect of process parameters on hydrogen production by catalytic methane reforming: an artificial intelligence approach

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Abstract. This study focuses on the non-linear effect of gas hourly space velocity (GHSV), oxygen (O₂) concentration in the feed, the reaction temperature, and the CH₄/CO₂ ratio on hydrogen production by catalytic methane dry reforming using artificial neural networks (ANN). Ten different ANN models were configured by varying the hidden neurons from 1 to 10. The various ANN model architecture was tested using 30 datasets. The ANN model with the topology of 4-9-2 resulted in the best performance with the sum of square error (SSE) of 0.076 and coefficient of determination (R²) greater than 0.9. The predicted hydrogen yield and the CH₄ conversions by the optimized ANN model were in close agreement with the observed values obtained from the experimental runs. The level of importance analysis revealed that all the parameters significantly influenced the hydrogen yield and the CH₄ conversion. However, the reaction temperature with the highest level of importance was adjudged the parameter with the highest level of influence on the methane dry reforming. The study demonstrated that ANN is a robust tool that can be employed to investigate predictive modeling and determine the level of importance of parameters on methane dry reforming.

Keywords: Artificial Neural Network; Backpropagation; Methane dry reforming; Hydrogen yield; Greenhouse gases

1. Introduction

Carbon dioxide and methane are two prominent components of greenhouse gases that are responsible for the greenhouse effect [1]. These greenhouse gases can be sustainably utilized for the production of hydrogen or hydrogen-rich syngas [2–4]. The methane dry reforming has been reported as a potential technological pathway suitable to simultaneously utilize carbon dioxide and methane for hydrogen-rich syngas production and mitigate their effects on the environment [5–9]. However, the methane dry reforming reaction is often characterized by complex chemical reaction pathways that are yet to be fully understood despite a series of kinetic and mechanistic studies that have been reported in the literature [10]. One major complexity is the non-linear effects of the various process parameters on hydrogen production [11–13].

The artificial intelligent approach can be employed to decipher the effects of these non-linear parameters on hydrogen production during the reforming reaction [14,15]. The use of ANN for modeling various processes has been widely reported in the literature [16–18]. The use of artificial



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neural networks for dynamic modeling of dry reformers under catalyst sintering has been reported by Azzam et al. [19]. The results revealed that industrial suitable catalysts can be achieved via the optimization of pressure and temperature. Similarly, Ghasemzadeh et al. [20] employed ANN for modeling the performance of the silica-based membrane reactor during methanol steam reforming. The influence of gas hourly space velocity (GHSV), reaction temperature, membrane pressure, and steam/methanol molar ratio on the methanol conversion was investigated. The results revealed that the ANN model accurately predicted the methanol conversion with the reaction temperature having the most significant influence. In a similar study by Ghasemzadeh et al. [21], it was reported that the ANN model was robust in predicting hydrogen production from glycerol steam reforming by a Pd-Ag membrane reactor. The present study employed Artificial Neural Network (ANN) algorithms to investigate the effects of Gas Hourly Space Velocity (GHSV), O₂ concentration in the feed, reaction temperature, and the CO₂ to CH₄ ratio. Several ANN model architectures were configured. This will help to properly understand the effects of these parameters on hydrogen production. Thirty datasets generated using the design of the experiment were obtained from methane dry reforming reaction using the bimetallic Ni-Co catalyst. These datasets consist of the input parameters (Gas Hourly Space Velocity (GHSV), O₂ concentration in the feed, reaction temperature, and CO₂ to CH₄ ratio) and their corresponding responses (Hydrogen and methane conversion) were employed for training, testing and validating the ANN models. The hidden neuron of the Several models of the ANN were optimized using backpropagation algorithm. The representative structure of the ANN model architecture is depicted in Figure 1. It consists of the input layer, the hidden layer, and the output layer.

2. Material and Methods

2.1 Data Description

The datasets used in this study for the ANN modeling was obtained using a central composite experimental design (CCD). The variable consists of GHSV, O₂ concentration in the feed, reaction temperature, and the CO₂ to CH₄ ratio. Based on the CCD, 30 datasets comprise of the variables were employed to obtain the responses which include the hydrogen yield and CH₄ conversion. The detailed description of the experimental runs has been reported by Fan et al.[22].

2.2 ANN Model Configuration and Analysis

A multilayer perceptron artificial neural networks with backpropagation configuration were employed in this study [11,23]. The configuration, which is depicted in Figure 1, consists of an input layer, the hidden layer, and the output layer. The input layer consists of the input variables which are made of GHSV, O₂ concentration in the feed, reaction temperature, and the CO₂ to CH₄ ratio. The hidden layers are made of interconnected artificial neurons with associated weights. For the efficient performance of the neural networks, the hidden neurons need to be optimized. The output layer consists of the responses from the experimentation and these consist of hydrogen yield and the CH₄ conversion. In this study, 10 ANN models were configured by varying the hidden neurons. The datasets were employed to train the various models to determine the configuration with the best performance. The datasets were partition into two for training and testing at a ratio of 0.7 and 0.3 respectively. The model performance was measured using the sum of square error (SSE) and the coefficient of determination (R²). The modeling was performed using the Neural Network analysis tools in IBM-SPSS version 22. To set up each the ANN model, the Sigmoid function was employed for the hidden layer and the output layers. A 0.02 normalized correction for employed for the rescaling of the scale-dependent variable.

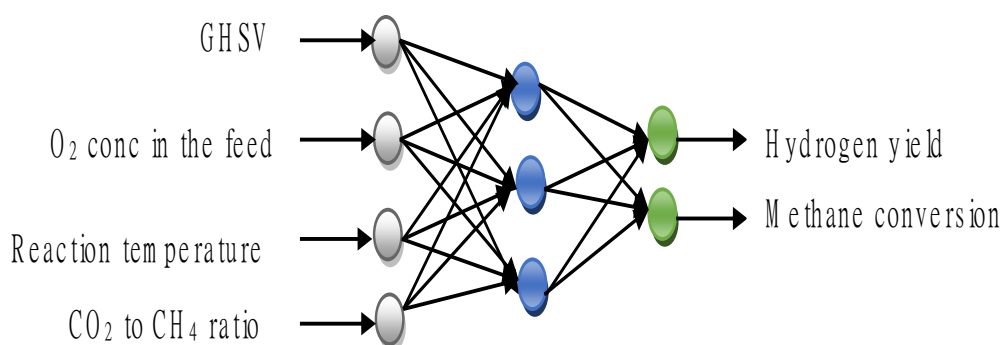


Figure 1. ANN architecture to investigate the effect of process parameters on hydrogen production

3. Results and Discussion

Figure 2 depicts the optimization of hidden neurons for ten ANN model architectures. The hidden neurons were varied from 1 to 20 and the errors from training each of the models were recorded in form of SSE. It can be seen that the SSE varies as the hidden neuron of each of the ANN model varies. The ANN model configuration of 4-9-2 recorded the least SSE of 0.076, hence was subsequently employed for modeling. The model performance as a function of the dispersion and regression plots are depicted in Figure 3. As shown in Figure 3 (a), the observed hydrogen yield for each of the experimental runs is in close agreement with the ANN predicted values. This can further be confirmed from the regression plot in Figure 3 (b) which shows a strong correlation between the observed and the predicted hydrogen yield as indicated by the high R^2 of 0.947. Similarly, the dispersion plots in Figure 3 (c) also show that the observed values of the CH_4 conversion for each of the experimental runs are consistent with the ANN model predicted values. A high R^2 of 0.960 obtained from the regression plot in Figure 3 (d) is a strong indication the ANN model is robust in predicting CH_4 conversions from the reforming process. The robustness of employing ANN as a predicting modeling tool has been reported by Hossain et al. [24]. Considering the effect of three different parameters namely feed ratio, reaction temperature, and metal loading, the authors reported that ANN accurately predicted CH_4 conversion, CO_2 conversion, H_2 , and CO yield with a high R^2 value > 0.9 . Also, Ayodele et al. [14] reported that an optimized ANN model efficiently predicted the rate of CO and H_2 production from methane dry reforming using reaction temperature, CH_4 partial pressure, and CO_2 partial pressure as the input variables. The use of ANN for the design of Co-MgO catalyst was reported by Omata et al. [25]. Using parameters such as Co contents, calcination temperature, citric acid equivalent, and the pellet pressure, the ANN model was efficient in predicting the effects of the parameters on the CO yield and carbon deposition on the catalyst.

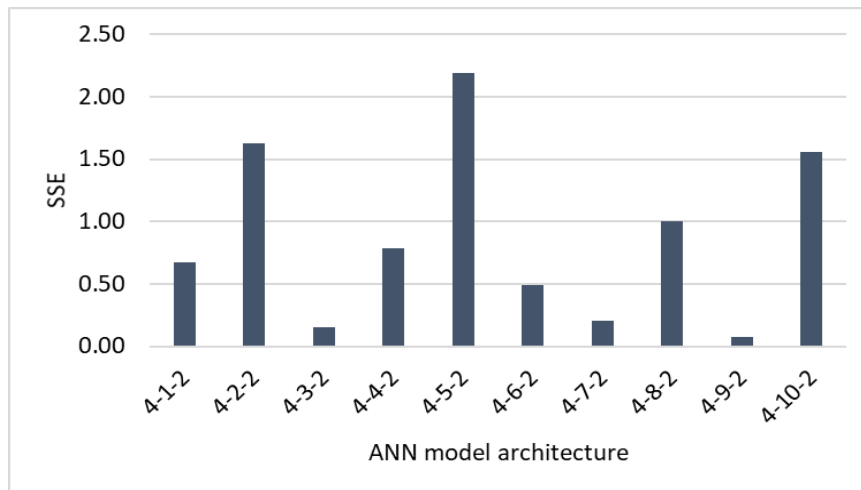


Figure 2. Optimization of the hidden neurons

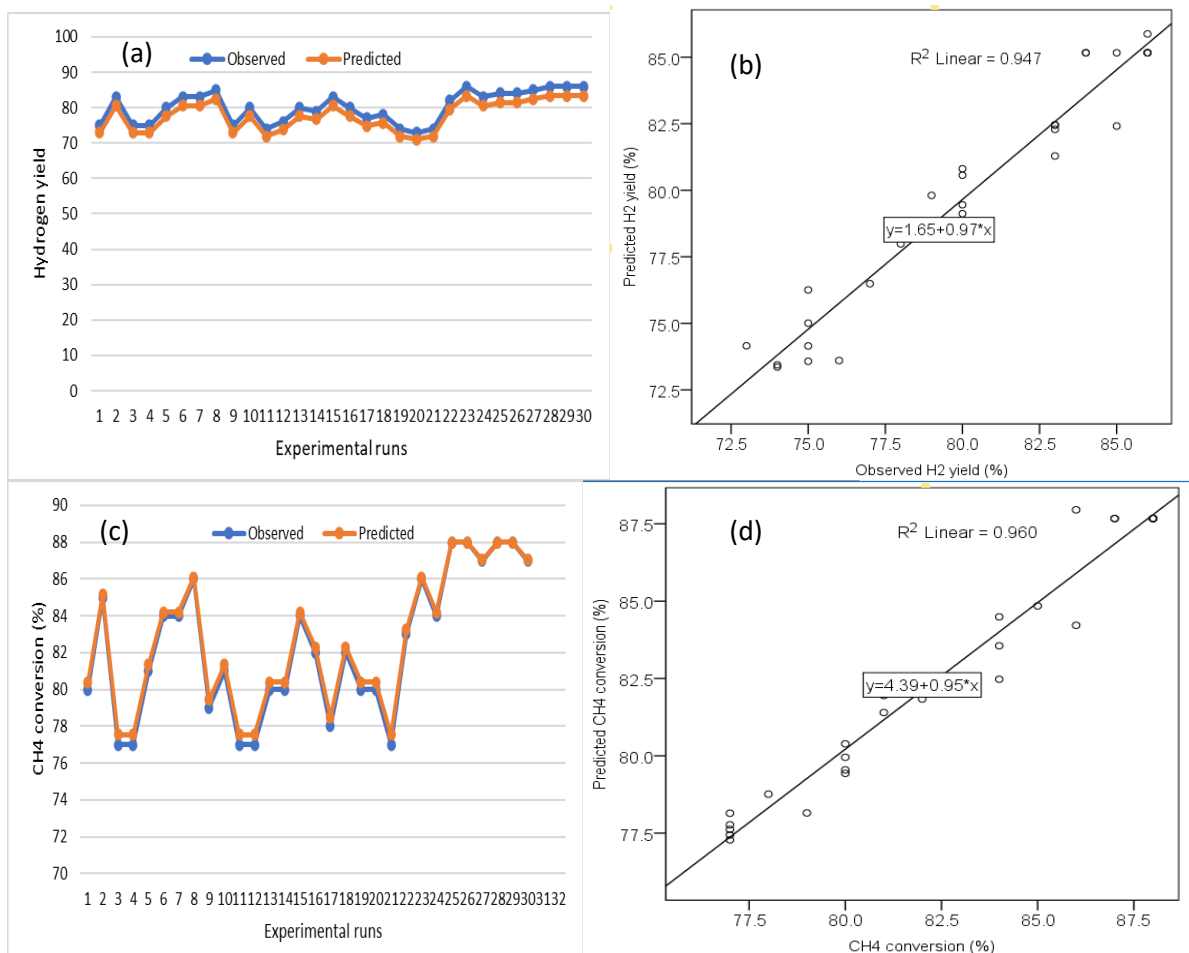


Figure 3. (a) Dispersion and (b) regression plots showing the observed and predicted hydrogen yield (c) Dispersion and (d) regression plots showing the observed and predicted CH₄ conversion

The level of importance of each of the parameters on the CH₄ conversion and hydrogen yield is depicted in Figure 4. The effect of five parameters namely the gas hourly space velocity, O₂ concentration in the feed, reaction temperature, and the CH₄: O₂ ratio based on the level of importance

of the hydrogen yield and CH₄ conversion were analyzed. The analysis shows that the five parameters had varying levels of significance on the hydrogen yield and CH₄ conversion. The level of significance of the gas hourly space velocity, O₂ concentration in the feed, reaction temperature, and the CH₄: O₂ ratio was estimated as 0.259, 0.260, 0.338, and 0.142, respectively. This is an indication that the methane dry reforming reaction was influenced by the four parameters. However, the most significant parameter was the reaction temperature which is consistent with that reported by Ghasemzadel et al. [21] for ANN modeling of methanol steam reforming. Omoregbe et al. reported that reaction temperature and CH₄/CO₂ ratio had a significant effect on methane dry reforming reaction over Ni/SBA-15 catalysts. A maximum of CH₄ and CO₂ conversions of 91% and 94% were obtained respectively, at a reaction temperature of 750 °C. Also, Ricks et al. [26] reported that the CH₄: CO₂ ratio and reaction temperature significantly influence the reactant conversions and the H₂: CO ratio in methane dry reforming over perovskite catalyst. The details effects of the parameters on the hydrogen yield and CH₄ conversion are summarized in Table 1. Both positive and negative influences were reported for each of the parameters as indicated by the negative and positive values of the synaptic weights of the parameters. It can be seen that the highest positive impact on the prediction of the hydrogen yield and CH₄ conversion by the ANN model was obtained at GHSV, O₂ concentration in the feed, temperature, and CH₄/CO₂ ratio of 200000 h⁻¹, 12 mol%, 735°C, and 1, respectively[18,27,28]. This could help as a guide in determining what range of process parameters that could negatively or positively influence the performance of the catalysts in the methane dry reforming reaction.

Table 1. Parameter estimates for the level of importance analysis

Predictor		Predicted Hidden Layer 1								
		1	2	3	4	5	6	7	8	9
Input Layer	(Bias)	-0.464	-0.600	0.878	-0.452	0.473	-0.479	-1.285	-0.010	-0.290
	84000	0.367	-0.409	1.145	-0.059	-1.153	0.025	-1.309	1.091	-0.460
	113000	0.156	0.073	-0.343	0.051	-0.290	-0.144	-1.700	-0.019	-0.225
	142000	-0.836	0.123	-0.997	-0.247	1.167	0.099	1.100	-1.090	0.401
GHSV	171000	-0.624	-0.165	0.287	0.147	0.165	-0.149	0.843	0.543	0.011
	200000	0.842	-0.673	0.143	0.279	-0.202	0.087	-0.218	-0.416	-0.414
	5	-1.738	-0.541	1.322	-0.966	-0.347	-0.282	0.875	0.155	0.667
O ₂ Conc. in feed	8	-0.246	0.787	-0.340	-0.634	0.479	0.608	1.181	-0.199	0.768
	10	0.974	0.303	-0.915	0.238	-0.054	0.075	-1.823	-0.150	0.289
	12	1.242	-0.698	1.362	1.061	0.363	-0.149	-1.295	-0.006	-1.047
Temp.	700	1.598	-0.568	1.296	1.152	0.110	-0.166	-1.899	0.526	-0.976
	735	1.455	-0.359	1.335	0.156	0.261	0.221	0.996	0.031	-0.662
	750	-0.924	0.323	-1.283	-0.773	-0.266	-0.132	0.900	-0.509	1.085
	775	-2.477	0.026	-0.361	-1.028	0.203	0.768	-1.409	0.338	1.220
CH ₄ / CO ₂ ratio	1	-0.650	0.186	0.237	-0.516	-0.185	0.509	0.527	-0.239	0.745
	2	-0.179	-0.212	-0.310	-0.856	0.228	-0.235	0.054	-0.068	0.648
	3	1.256	-0.065	0.772	0.591	0.282	-0.029	-0.963	0.257	-0.702
	4	-0.875	0.153	0.225	-0.211	-0.716	-0.178	-1.140	-0.001	-0.427
Output Layer	CH ₄ conversi on	0.255	-2.706	0.516	-2.371	-0.953	1.399	-0.047	3.136	-1.212

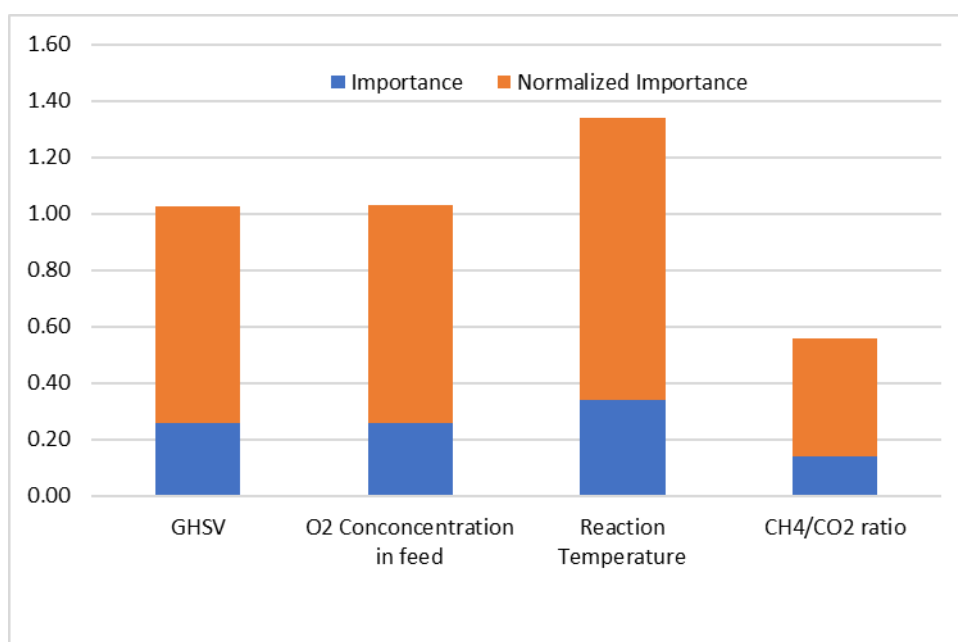


Figure 4. Level of importance analysis of each of the input parameters on the CH₄ conversions and hydrogen yield.

4. Conclusions

This study has demonstrated the robustness of using artificial intelligence techniques to investigate the effect of process parameters on hydrogen production and CH₄ conversion in methane dry reforming reaction. Ten ANN model architecture was configured and tested using datasets obtained from experimental studies. The best ANN configuration of 4-9-2 was subsequently employed for predictive modeling and parameter analysis. The model analysis revealed that the optimized ANN model accurately predicted the hydrogen yield and CH₄ conversion by the methane dry reforming as indicated by the low sum of square error and high R². Based on the level of importance analysis, the GHSV, O₂ concentration in the feed, the reaction temperature, and the CH₄/CO₂ ratio have varying levels of influences on the methane dry reforming reaction. Reaction temperature with the level of importance of 0.338 has the most influence on the methane dry reforming reaction. The outcome of this study could help in guiding the optimum design of experiment for hydrogen production by methane dry reforming.

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