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Artificial neural network modeling of hydrogen-rich syngas production from methane dry reforming over novel Ni/$CaFe_2O_4$ catalysts

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ABSTRACT

In this study, the application of artificial neural networks (ANN) for the modeling of hydrogen-rich syngas produced from methane dry reforming over Ni/$CaFe_2O_4$ catalysts was investigated. Multi-layer perceptron (MLP) and radial basis function (RBF) neural network architectures were employed for the modeling of the experimental data obtained from methane dry reforming over novel Ni/$CaFe_2O_3$ catalysts. The Ni/$CaFe_2O_3$ catalysts were synthesized and characterized by XRD, SEM, EDX and FTIR. The as-synthesized Ni/$CaFe_2O_3$ catalysts were tested in a continuous flow fixed bed stainless steel reactor for the production of hydrogen-rich syngas via methane dry reforming. The inputs to the ANN–MLP and ANN–RBF-based models were the catalyst metal loadings (5–15wt %), feed ratio (0.4–1.0) and the reaction temperature (700–800 °C). The two models were statistically discriminated in order to measure their predictive capability for the hydrogen-rich syngas production. Coefficient of determination (R^2) values of 0.9726, 0.8597, 0.9638 and 0.9394 obtained from the prediction of H_2 yield, CO yield, CH_4 conversion and CO_2 conversion respectively using ANN–MLP-based model were higher compared to R^2 values of 0.9218, 0.7759, 0.8307 and 0.7425 obtained for the prediction of H_2 yield, CO yield, CH_4 conversion and CO conversion respectively using ANN–RBF-based model. The statistical results showed that the ANN–MLP-based model performed better than ANN–RBF model for the prediction of hydrogen-rich syngas from methane dry reforming over the Ni/$CaFe_2O_4$ catalysts. Further t-test performed based on the target outputs from the ANN–MLP and ANN–RBF network shows that the models were statistically significant.

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Introduction

Energy derived from fossil sources such as crude oil, coal and natural gas has been the main contributor to the ever increasing global energy consumptions [1,2]. Unfortunately, the utilization of these fossil fuels for different human and industrial activities has been associated with the emissions of greenhouse gases which are the major contributors to global warming [3,4]. Besides, forecast by reservoir experts revealed that the rate at which the crude oil reserve is fast depleting might leads to energy crisis in the nearest future if there is continuous dependence on crude oil without sustainable alternatives [1,5].

Methane dry reforming is a potential technological option for mitigating the emission of greenhouse gasses as well as for increasing the global energy supply via production of hydrogen and syngas [6,7]. Methane dry reforming is a thermo-catalytic process that utilizes CH_4 and CO_2, the two main contributors to greenhouse gases for the production of hydrogen and syngas [8]. Theoretically, syngas ratio of unity is produced from methane dry reforming, making it suitable as chemical building blocks for the production of oxygenated fuels and other value added chemicals [9,10]. In addition, the hydrogen produced from the methane dry reforming possesses high energy content with 0% emission when utilized as fuel [11]. Liquid hydrogen has been used by the US national aeronautics and space management (NASA) as fuel to propel space shuttle and other rockets into orbit [12].

Despite the potential benefits of hydrogen and syngas production from methane dry reforming, this process is being constrained with catalyst deactivation from cooking and sintering [13]. As a result of this, researchers have designed, developed and tested the catalytic activities of supported metals based catalysts such as Ni/CeO_2, Ni/La_2O_3, Ni/Al_2O_3, Ni/SiO_2, Co/CeO_2, Co/La_2O_3 and so on in methane dry reforming [6,14–19]. However, based on the extensive search of literature to the best of the authors' knowledge, Ni/$CaFe_2O_3$ catalyst has not been investigated for the production of hydrogen and syngas via methane dry reforming. Methane dry reforming is a complex gaseous reaction due to the influence of side reactions such as reverse water gas shift reaction, methane cracking and reverse Boudouard reaction [20]. Hence, conventional mathematical modeling of the methane dry reforming process will be cumbersome and time consuming. Alternatively, ANN-based modeling has been successfully proven for development of empirical models for complex systems [21–24]. Nevertheless, there is presently no literature on the application of ANN for modeling hydrogen and syngas production from methane dry reforming over Ni/$CaFe_2O_3$ catalyst. Hence, the main objective of this study is to investigate the application of artificial neural network (ANN)-based model for the prediction of hydrogen and syngas production from methane dry reforming over Ni/$CaFe_2O_3$ catalysts.

Methods

Synthesis of catalysts

Prior to the synthesis of the Ni/$CaFe_2O_4$ catalysts, the $CaFe_2O_4$ support was prepared by sol–gel technique reported elsewhere [25]. In a typical synthesis, a stoichiometric ratio of $Ca(NO_3)_2.4H_2O$ and $Fe(NO_3)_3.9H_2O$ were mixed in 30% aqueous NH_3 solution, and the mixture was stirred at room temperature for 24 h. The solution was then slowly heated to 80 °C and maintained at that temperature level until the water evaporated. The resulting brown dry gel-like slurry was calcined at 450 °C for 2 h followed by heat treatment at 900 °C for 10 h using muffle furnace to obtained $CaFe_2O_4$ powder. Finally, $CaFe_2O_4$ powders were crushed in the mortar to obtain fine particle size. Thereafter, Ni/CaF_2O_4 was synthesized with the metal loading of 5–15 wt% using wet-impregnation method. Required amount of $Ni(NO_3)_2.6H_2O$ precursor was dissolved in aqueous solution and 1 g of $CaFe_2O_4$ was added to the solution under stirring for 3 h. Subsequently, the slurry was oven dried for 24 h at 120 °C, crushed and finally calcined at 800 °C for 5 h.

Catalyst characterization

The characterization of the catalyst in this study was based on the as-prepared 15%wt Ni/$CaFe_2O_4$. The phase identification and crystallinity of the as-prepared 15wt%Ni/$CaFe_2O_4$ catalyst was characterized by X-ray powder diffraction analysis using RIGAKU miniflex II X-ray diffractometer capable of measuring powdered diffraction pattern from 3 to 145° in 2θ scanning range. The X-ray source is Cu Kα with wavelength (λ) of 0.154 nm radiation. The XRD is equipped with the latest version of PDXL, RIGAKU full function powder-diffraction analysis software. The morphology and the elemental composition of the as-prepared catalyst were determined using JEOL field emission scanning electron microscopy (FESEM) equipped with energy-dispersive X-ray spectroscopy (EDX). The bond formations of the as-prepared 15wt%Ni/$CaFe_2O_4$ catalyst was confirmed by FTIR spectrum using Perkin Elmer Spectrum BX(II) spectrophotometer.

Catalytic activity test

The schematic diagram of the experimental set-up for the hydrogen-rich syngas production from methane dry reforming is represented in Fig. 1. The set-up comprised CO_2, CH_4, N_2 and H_2 gases. The main reactants for the methane dry reforming are CO_2 and CH_4, while N_2 and H_2 serve as a carrier gas and for reduction, respectively. The stainless steel fixed bed continuous reactor was packed with 200 mg of catalysts supported with quartz wool and heated inside a split-tube furnace that was equipped with a K-type thermocouple for measurement of the catalytic bed temperature. The catalyst was reduced in 60 ml/min of 20% H_2 and 80% N_2 prior to the commencement of the catalytic activity test. The flow rate of the inlet gas was maintained at 100 ml/min and individually