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A review of hydrogen production optimization from the reforming of C1 and C2 alcohols *via* artificial neural networks

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ABSTRACT

Hydrogen production from different fuels has received extensive study interest owing to its environmental sustainability, renewability, and lack of carbon emission. This research aims to investigate how artificial neural networks (ANNs) are employed to optimize operating parameters for the catalytic thermochemical conversion of methanol and ethanol and their impact on hydrogen production. According to the ANN model, peak methanol conversion (99%) occurs at lower temperatures of 300 °C with a maximum hydrogen yield of 2.905 mol, whereas peak ethanol conversion (85%) occurs at 500 °C owing to dehydrogenation and the C-C bond-breaking process. A steam-to-carbon (S/C) ratio of (3.5) was advantageous for methanol steam reforming (MSR), and a high ethanol concentration of 10–15 vol% was favorable for ethanol steam reforming (ESR). Ni (10 wt%), and Co (10 wt%) were the optimum metal combinations in the catalyst for ethanol reformation at a reforming temperature of 450 °C. The optimum metal catalysts for producing hydrogen and converting ethanol were those synthesized through co-precipitation. The peak hydrogen yield was attained at the sintering temperature of 560–570 °C. ANN technique is cost-effective, quick, and precise, with vast potential to produce hydrogen energy, and may give significant benefits for industrial applications.

1. Introduction

The modern world's primary energy source is the combustion of conventional and nonrenewable fossil fuels, which are quickly depleting and emitting a huge amount of greenhouse gases. As a result, the atmosphere's temperature is rising, causing global warming [1]. One of our era's most imperative scientific challenges is the reduction of greenhouse gas emissions and rectification of global warming before it wipes out civilization [2]. Scientists in a range of disciplines are tackling this problem with diverse approaches. One of the key strategies

Abbreviations: ANN, Artificial neural network; ATR, Autothermal reforming of ethanol; CI, Continuous integration; ESR, Ethanol Steam reforming; EtOH, Ethanol (Chemical formula: CH_3CH_2OH, colorless, volatile, flammable, and polar solvent); GHSV, Gas hourly space velocity; LM, Levenberg-Marquardt; MD, Methanol decomposition; MEOH, Methanol (Chemical formula: CH_3OH, colorless, volatile, flammable, and polar solvent); MSE, Mean square error; MSR, Methanol steam reforming; POM, Partial oxidation of methanol; POX, Partial oxidation of ethanol; S/C, Steam-to-carbon ratio; S/E, Steam-to-ethanol ratio; S/M, Steam-to-methanol ratio; WGSR, Water gas shift reaction.

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undertaken by scientists to combat global warming is producing clean, renewable, and sustainable energy that produces zero emissions. Hydrogen is a potentially clean and green energy source since it combusts to produce only water or steam but no carbon dioxide. Thus, one potential component for environmental sustainability and renewable energy is utilizing hydrogen in different fields [3,4]. In addition to being depleted over time, coal, oil, and natural gas are unevenly distributed among the continents [5,6]. Renewable energies are derived from environmental sources, which fluctuate based on geographic location, climate, and environmental conditions [7]. For these reasons, establishing a widely utilizable, and sustainable energy source is indispensable. Hydrogen is an energy source that can be manufactured anywhere on the globe. Methanol and ethanol are two potential raw materials frequently used for producing hydrogen. They may dissociate in different thermochemical processes to produce hydrogen. Methanol is an advantageous raw material since its energy cost for hydrogen production is about half that of ethanol. Ethanol, which is primarily produced by biomass fermentation, is also a suitable raw material for hydrogen production as it is easy to manufacture, and has more constituent hydrogen [8,9]. Moreover, methanol and ethanol are liquid fuels that are easily transported. Therefore, they have high flexibility for hydrogen production without location restrictions.

Methanol and ethanol are both transparent, volatile, and polar due to their hydroxyl group. The catalytic reaction of syngas, including H_2 and CO, is usually utilized for commercial methanol production [10–12]. The current focus of scientists regarding hydrogen production from methanol and ethanol is the minimization of the energy and cost requirements, and carbon dioxide and carbon monoxide emissions reduction [10–12]. Moreover, bioethanol is a promising and renewable biofuel due to its low greenhouse gas emissions during production and low environmental impact [13]. Van Rens et al. [14] investigated the efficiency and exergy analyses of fuels made from biomass, including methanol and hydrogen. Modern technology is most effective in producing methanol, with an exergetic efficiency of 55%. Chen et al. [10] demonstrated the exergetic efficiency of ethanol in an ethanol steam reforming technique is 80%. The efficiency of a system is calculated using an exergy analysis based on how well it functions in a reversible model. Colombaroil et al. [15] evaluated the exergy for hydrogen generation in the ethanol reforming reaction. The fermentation of sugarcane produced ethanol. This study demonstrates that the quantity of carbon dioxide released during the MSR reaction was lower than that of the amount taken during sugarcane growth, indicating a net negative emission. The exergy analysis is developed by considering thermodynamic efficiency (η_{system}), pollution indicator (Π_g), and ecological efficiency (ε). The ecological efficiency for ESR reaction is nearly 90% when the pollution indicator (carbon cycle) is also considered in the system. The ecological efficiency is written as follows (Eq. (1)).

$$\varepsilon = \left(\frac{0.204 \times \eta_{system} \times \ln(135 - \Pi_g)}{\eta_{system} + \Pi_g} \right)^{0.5} \quad (1)$$

Steam reforming, partial oxidation, and autothermal reforming are the most appealing processes for producing hydrogen from methanol and ethanol [11,12,16]. Nevertheless, MSR and ESR is the most widely accepted technique since it produces a lot of hydrogen and is currently more economically viable than other processes. MSR and ESR processes both consume water, but the reaction temperatures vary between the two techniques [11,12,17,18]. The MSR procedure is often carried out at lower temperatures, between 150 °C and 400 °C, owing to the lack of a C-C bond in methanol [12]. In contrast, ESR occurs most often at relatively high reforming temperatures between 300 °C and 700 °C. In ESR, the C-C bond breaking starts after 400 °C, and the elimination of hydrogen occurs primarily at temperatures between 200 °C and 400 °C (Fig. 1) [19,20]. POM and POX involve only oxygen uptake, whereas ATR of methanol and ethanol involves oxygen and steam intake [11,12,21,22]. Artificial neural networks (ANN) are computer algorithms developed by emulating the function of the brain's neuron networks. Input, hidden, and output layers are the three levels that make up a standard artificial neural network. Each layer has a different number of nodes known as neurons [23–26]. These nodes are mathematically interconnected and can similarly transmit functions or output as brain cells operate. The weights in the transfer function regulate how smoothly the signal flows into the nodes. The computer-programmed algorithm selects the weights once the input is provided, allowing the output results to be predicted with acceptable error limits, and the predicted output is aligned with the experimental output [23–27]. An ANN model may predict the response corresponding to the new range of inputs not included in the training database when the model is

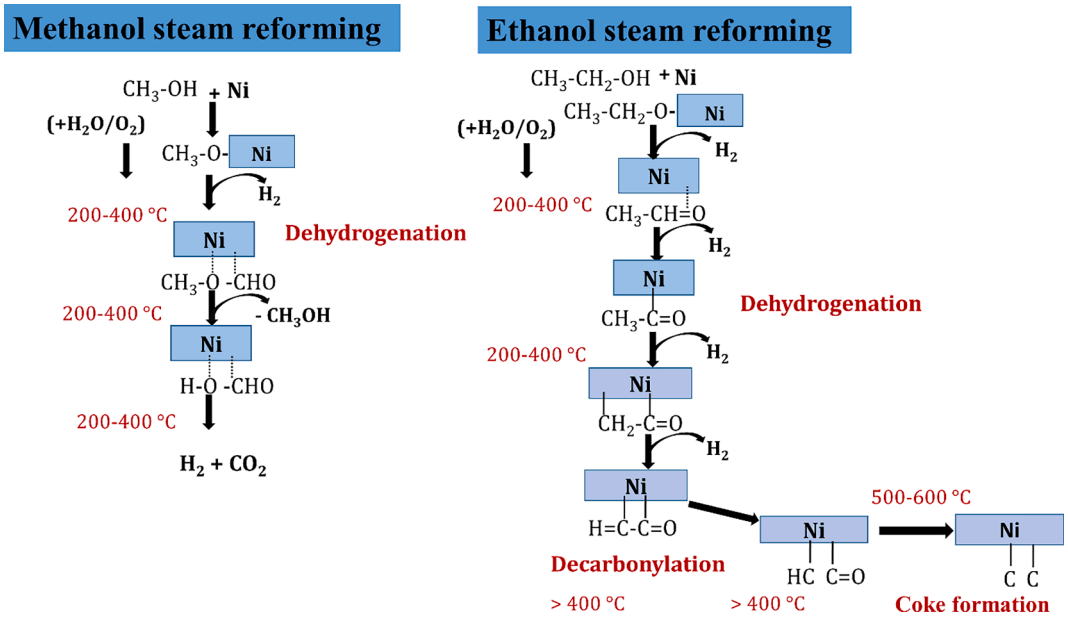

Fig. 1. The mechanism of methanol and ethanol-reforming. The MSR and ESR processes both involve water intake. Nevertheless, the reaction temperatures vary between the two techniques. The MSR procedure is often carried out at temperatures ranging from 150 °C to 400 °C due to the lack of a C-C bond. The dissociation of both methanol and ethanol is related to the absorption of alcohol onto the metal catalysts, and the formation of intermediates [99,100].

adequately trained. ANNs are an advanced optimization method used in chemical and process engineering and catalyst development. One of the greatest advantages of ANN optimization technology is that it doesn't have to be developed by predefined guidelines. Any adaptive learning algorithm can be employed to develop any flexible model of an artificial neural network from complexly formulated experimental conditions if a complete collection of training data or numerical values of parameters is provided [23–26,28]. Thus, compared to other optimization techniques, the ANN model has versatility in predicting the optimum operating parameters and the higher yield.

Most of the research on hydrogen energy is focused on improving hydrogen yield, increasing fuel conversion, decreasing energy requirements, lowering carbon emissions, and developing cost-effective processes. Metal-based catalysts' physical and chemical characteristics strongly determine hydrogen production and fuel conversion. They also have a great influence on reaction rate. The reaction temperatures, fuel flow rate, steam-to-fuel ratio (S/M or S/E), and oxygen-to-fuel ratio are vital reaction factors that govern overall methanol/ethanol conversion and hydrogen production [29–32]. Thus, setting these parameters is crucial for controlling product distribution and developing high hydrogen yields in various methanol and ethanol reforming technologies.

Hydrogen production from methanol and ethanol has received much scientific attention over the past few decades, leading to an increase in the number of literature reviews published. Most reviews focus on understanding the thermochemical conversion of methanol/ethanol reforming, catalyst preparation, reactor design, and operating conditions. Very few reviews concentrate on machine learning or computational intelligence for optimizing hydrogen production from various biomass feedstocks and fuels (e.g., hydrocarbons and biological compounds) via multiple chemical routes. A few of these studies are briefly described below.

To determine which computational intelligence method(s) performs best in the prediction, evaluation, and optimization of hydrogen production, Faizollahzadeh Ardabili et al. [33] reviewed the state-of-the-art continuous integration (CI) approaches applied in hydrogen production from methane reforming, biomass fermentation, and gasification, as well as from the fuel cell power plants. Le et al. [34] reviewed the *meta*-analysis of hydrogen production using different catalysts in the dry reforming of alcohols, hydrocarbons, and biological compounds via artificial neural networks (ANNs). Besides these, several research papers and review articles on fuels production, different reformation approaches (MSR, POM, ATR, ESR, POX, and EATR), and the application of different optimization techniques for determining the optimal process parameters and maximum yield related to those processes have been published in recent years. The original research is mostly focused on (1) biochar and hydrogen production from non-catalytic pyrolysis of biomass [35], (2) the use of analysis of variance (ANOVA) and Taguchi optimization methodologies to optimize the process parameters of bio-methanol reformation over Ni-Cu catalyst [29], and (3) application of ANN technology to optimize the process parameters of sewage sludge gasification for hydrogen production [36]. Whereas the review articles are mostly concerned with (1) optimizing the process parameters of methanol and ethanol reformation through the design of experimental methods (statistical optimization) [37], (2) production of hydrogen via anaerobic digestion of sewage sludge [38], (3) hydrogen production via methane reformation [39], and (4) application of ANN technology to optimize the biodiesel production [40]. According to original research and review papers published on hydrogen production, a review article regarding the application of ANN optimizing technology for the methanol and ethanol reformation techniques is still essential for comprehending the comprehensive study of the prediction of hydrogen yield and sensitivity test of reforming parameters.

According to our assessment of the literature, no review article has yet been published on the application of artificial neural networks (ANNs) to optimize operating parameters for catalytic thermochemical conversion of methanol and ethanol, along with the physical and chemical characteristics of various catalysts and their implications on hydrogen production. In this regard, the present study intends to encapsulate the application of artificial neural network training technology on the operating conditions of fuel reforming, including reforming temperatures, feedstock content, and flow rate, additionally the catalyst synthesis parameters, including catalyst formation techniques, sintering temperature, and stirring time, will be analyzed for their forthcoming benefits, and drawbacks. This study will also emphasize how quickly and accurately ANN technology can predict the maximum yield of hydrogen production from fuel reforming. Quantifying the optimal value for each process parameter may simplify future research; for example, an ANN prediction model might indicate whether a parameter should be quantitatively increased or decreased during each experiment.

In summary, the emission of greenhouse gases degrades the ozone layer, leading the net atmospheric temperature to rise drastically. Global warming might be one of the causes of human extinction [2,41]. Experts from several research sectors worldwide are attempting to address this impending environmental catastrophe. One of the policies scientists pursue is achieving global net zero emissions [10,11]. Water is the only byproduct left over when hydrogen is burned, making it a possible source of clean energy. Hydrogen production from methanol and ethanol reforming has been extensively investigated in recent years to determine this approach's feasibility and economic viability [16]. The reaction conditions for MSR and ESR are different, and the conversion of C1 and C2 alcohols and the production of hydrogen are highly dependent on the reaction parameters. According to the literature review, hydrogen production varies between studies since they used diverse reaction conditions and catalysts [16]. Therefore, optimizing the process parameters of various reforming processes to maximize hydrogen production and make the process both energetically and economically viable is vital. The ANN is a simple, flexible, and low-cost optimization approach that can run various parameters and data sets using various algorithms, which might open up a large opportunity for industrial-scale hydrogen production [23–26]. Hence, a critical review of the optimization of process parameters of methanol and ethanol reforming using ANN technology is highly demanded to summarize the original articles which have been published to date. A detailed assessment of this literature review is certainly essential in order to determine the numerical values of various parameters and their impact on the thermochemical conversion of C1 and C2 alcohols to obtain optimum hydrogen production. ANN optimization technology can simulate the data for large-scale industrial applications and evaluate which parameters need to be adjusted and which need to be eliminated or lowered.

2. Application of artificial neural networks for experiment design in fuel reforming and water gas shift reaction

2.1. Artificial neural networks learning process

ANN technology, combining both supervised and unsupervised learning, has a very high capacity to understand the interrelationships and connections between input and output databases [42–44]. The input factors for thermochemical catalytic methanol and ethanol conversion include reforming temperature, S/C ratio, carbon-to-oxygen ratio, feedstock flow rate, and catalyst types or their corresponding physicochemical characteristics. The output parameters are primarily the hydrogen yield and alcohol conversion (Eqs. (2)–(5)) [42–44].

$$\text{MeOH conversion } (\%) = \left(\frac{moles\ of\ fed\ MeOH - moles\ of\ excited\ MeOH}{moles\ of\ fed\ MeOH}\right) \times 100 \quad (2)$$

$$\text{EtOH conversion } (\%) = \left(\frac{moles\ of\ fed\ EtOH - moles\ of\ excited\ EtOH}{moles\ of\ fed\ EtOH}\right) \times 100 \tag{3}$$

$$H_2 \text{ selectivity } (\%) = \left(\frac{moles\ of\ produced\ H_2}{3\ moles\ of\ fed\ EtOH}\right) \times 100 \tag{4}$$

$$\text{Carbon product selectivity } X\ (\%) = \left(\frac{moles\ of\ C\ in\ product\ X}{Total\ moles\ of\ C\ in\ products}\right) \times 100 \tag{5}$$

The most widespread supervised learning approach simultaneously feeds neural networks with input and intended output data [44]. After that, the ANN model will generate output data with the error function using a random matrix, and these data will be compared to the expected output data [42–44]. Song et al. [42] implemented supervised learning for the ANN, where the catalytic synthesis preparation techniques used Ni and Al loadings as input parameters and EtOH conversion, H_2 yield percentage, and selectivity as output parameters. The input and outcome data pairs are expressed in supervised learning as follows (Eq. (6)).

$$(p_1, t_1), (p_2, t_2), \cdots\cdots (p_n, t_n), \tag{6}$$

In this equation, the variables p and t refer to the network's input and "corresponding output" parameters, respectively.

Prior to running the test, the data for the input and targets were generated by experiments and stored as training data. Contrarily, unsupervised learning trains neural networks to operate autonomously without external supervision. Weights for training neural networks cannot be modified in unsupervised learning due to the lack of expected output data. Utilizing supervised or unsupervised ANN is a challenging decision to make. Unsupervised learning is acceptable when the required output data are unknown and vice versa [42–44]. The key difference between the supervised and unsupervised learning processes is that the supervised algorithm uses the labeled data (input and output data) to anticipate the expected output. In contrast, unsupervised learning methods employ unlabeled data [45]. The supervised learning method is based on two principles: classification and regression [46]. Classification is a method for categorizing data into various groups by using an algorithm (random forest or decision tree) [47]. The random forest algorithm combines several sets of output (trees) to get at a single value. At the same time, regression is a method that uses an algorithm to determine the connection between independent and dependent variables. Regression models are widely used to predict numerical values from input and output data ranges [46]. The decision tree applies several algorithms to divide a node into multiple nodes. The unsupervised learning method is built based on three principles: association, clustering, and dimension reduction [48]. In the case of an association, the learning methods attempt to identify the interconnection between variables in a data set. In the case of clustering, unlabeled data is categorized by tracking the differences and similarities between unlabeled data in each data set using learning methods. In the case of dimensionality reduction, the dimensions or data for a given data are minimized while maintaining their consistency [48].

2.2. Multilayer feed-forward neural networks learning process

A multilayer feed-forward neural network is widely employed as part of ANN technology. An artificial neural network usually has three layers, each of which has a different number of nodes known as neurons. The artificial neural layer has an input layer, a hidden layer, and an output

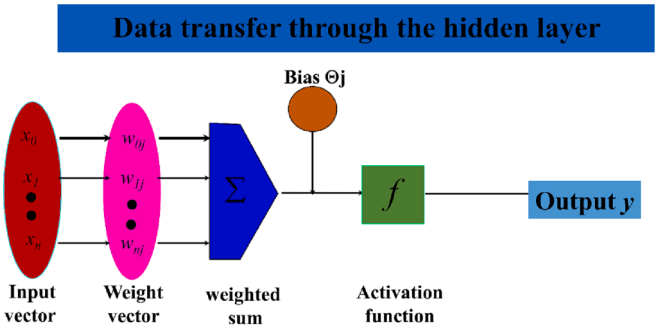

Fig. 3. The representation of the data transfer through a hidden layer of artificial neural network.

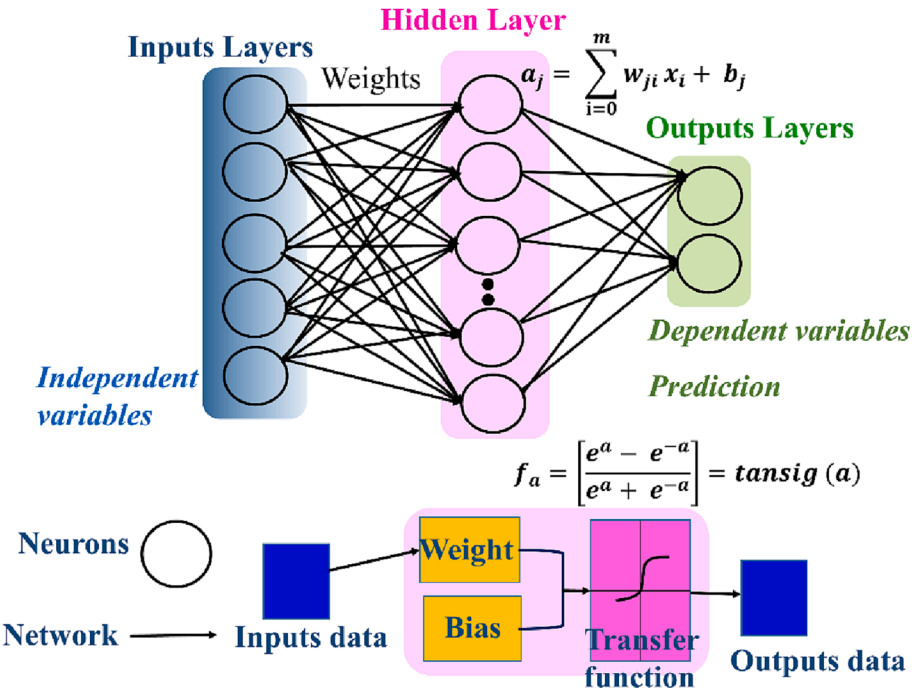

Fig. 2. The representation of the data transfer through a hidden layer of the artificial neural network. One of the most common models for artificial neural networks is one with a single hidden layer, in which every node is linked to every other node from any preceding layer. Thus, this node adds the weighted inputs and also the bias before transferring the results through a linear function. The sigmoidal tangent function is typically employed as an activation function for the hidden layers. Whereas the final output function could be both linear or sigmoidal function [42,51].

layer, all containing many neurons [23–26,42] (Figs. 2-3). The details of the three layers are described below.

(1) Input layer: The input layer's primary function is to obtain data from an external source, such as experimental data from various publications, past research, or experiments. The input layer has one neuron for each input parameter, such as reforming temperature, catalyst types, and S/M ratio [42].
(2) Hidden layer: Users decide how many nodes to include in the hidden layer based on their experience and trial and error methods. However, the number of neurons fluctuates according to the experiment's complexity. The large number of nodes produced by the complicated problems may occasionally contribute to overfitting issues [42].
(3) Output layer: The output layers process the results of the hidden layers. The number of output objectives determines how many neurons will be present in the output layers. The output layers contain the data calculated by ANN, such as the estimated hydrogen production at different reforming temperatures [42].

The most fundamental properties of the ANN are nonlinear functional interactions [23–26]. Due to this characteristic, ANNs are pretty effective techniques for analyzing data containing ambiguous relationships. The input layers receive the input data, which are then processed by the input layer to provide the input for the hidden layer. The output data or results from the input layer are then combined with the weights and biases of the hidden layer to produce the output data from the hidden layers. The results or outputs from the hidden layer are then directed into the last layer, known as the output layer [42].

One of the most common models for ANNs is one with a single hidden layer, in which every node is linked to every other node from any preceding layer [49,50]. Thus, this node adds the weighted inputs and also the bias before transferring the results through a linear function (Eq. (7)) [42] (Figs. 2-3).

$$a_j = \sum_{i=0}^{m} w_{ji} x_i + b_j \tag{7}$$

w_{ji} refers to the weight value that transfers from the input (i) to the hidden layer (j), b_j refers to the bias of the j nodes, and x_i refers to the input unit of the corresponding neurons. The output neuron may be represented as the following by applying an activation function (Figs. 2-3) (Eq. (8)) [42,51]. In this equation, z represents the neuron output, a represents the linear function, and f refers to the activation function.

$$z_j = f(a_j) \tag{8}$$

The sigmoidal tangent function (Eq. (9)) is typically employed as an activation function for the hidden layers. In contrast, the final output function could be both linear (Eq. (10)) or sigmoidal [42,51] (Figs. 2-3).

$$f_a = \left[\frac{e^a - e^{-a}}{e^a + e^{-a}}\right] = tansig(a) \tag{9}$$

The linear transfer function is as follows,

$$\text{purelin}\,(\mathrm{x}) = x \tag{10}$$

The activation function determines a neuron's activation, which connects a neuron's input and output variables. Three types of activation functions are commonly used for data transfer: sigmoid, linear, and Elliot [52]. The linear transfer function is written as follows (Eq. (11)):

$$\Phi\,(u) = u \tag{11}$$

where u is the total input, which is mostly employed at the output layer of the neural network. The non-linear functions, such as sigmoid and Elliott, range between −1 and + 1. A sigmoid function is an S-shaped function that is linear at the center but non-linear towards the end. The sigmoid function ranges between 0 and 1 [52]. This transfer function is the most employed in the reforming reaction of methanol and ethanol since it could be easily differentiable as compared to the other transfer function [52]. The sigmoid transfer function is written as follows (Eq. (12)):

$$\Phi\,(u) = \left[\frac{1}{1 + e^{-u}}\right] \tag{12}$$

Elliott is another easily differentiable function that ranges from −1 to 1 and is mostly employed in implementing the backpropagation algorithm [52]. The Elliott transfer function is written as follows (Eq. (13)):

$$\Phi\,(u) = \left[\frac{u}{1 + |u|}\right] \tag{13}$$

In ANN technology, the algorithm is used in the training process. Using a combination process, an artificial neural network can execute and analyze nonlinear relations in a complex data set and describe each explanatory factor's influence on the results, which a linear regression model may not be able to assess.

2.3. The details organization and topology of artificial neural network in alcohol reforming

To employ the ANN technology in various engineering applications and achieve their desired outcome, scientists have used various methods to customize network configurations. According to the literature survey, ANNs can be configured in several ways, such as ANNs with one hidden layer, ANNs with two hidden layers, and ANNs with three hidden layers [49,50,53]. Tompos et al. [54] demonstrated the ANN, which has nine input units corresponding to the proportion of nine elements (N = 9; Zr, Cr, Ce, Co, Cu, La, Au, Pd, and Pt) utilized in the manufacture of catalysts. There is one output node in the network's output layer. Since ANNs are a trial-and-error process, the number of neurons in the hidden layer might be varied with the different operations. However, for this study, the authors organized the hidden neurons from 5 to 50 in increments of 5, resulting in 10 different ANN structures. ANNs with two (N_{input}–$N_{hidden1}$–$N_{hidden2}$–N_{output}) and three (N_{input}–$N_{hidden1}$–$N_{hidden2}$–$N_{hidden3}$–N_{output}) hidden layers were organized as (9-5-5-1, 9-10-15-1, 9-14-10-1, 9-15-10-1, 9-20-10-1, 9-15-15-1, 9-20-20-1) and (9-5-3-2-1, 9-20-10-10-1) respectively [54]. Kim et al. [55] adopted a topology with five hidden layers (14-56-28-14-7) to optimize the water gas shift reaction (WGSR) using a multilayer perceptron ANN, and the data was determined by trial and error. The literature review indicated that the topology of the ANN model was usually determined depending on the experimental conditions, purpose, and data availability and that there are no strict guidelines for picking any specific topology for a certain ANN model.

2.4. The algorithm used for the artificial neural network in alcohol reforming

The ANN must be trained before learning can occur. The learning algorithm is a series of formulas or rules that the ANN model follows. According to the literature review, various algorithms such as backpropagation, Levenberg-Marquardt, and Nelder-Mead are used to govern the ANN model to simulate optimum hydrogen production and determine the optimal operating conditions and their impact on hydrogen production [42,56]. As a result, the aim was to employ different algorithms to train the process in accordance with different sets of defined criteria.

2.4.1. Backpropagation algorithm

The simplest case for backpropagation learning is when it modifies the network's weights and biases, quickly decreasing the performance function, which is the gradient's negative. This algorithm's first iteration can be expressed as follows (Eq. (14)). This gradient descent

algorithm can be achieved by batch or incremental mode. After every input is employed to the network, the weights are modified, and the gradient is calculated in the incremental mode [55,57]. Before the weights are adjusted, all inputs are employed in batch mode to feed the network.

$$x_{k+1} = x_k - a_k g_k \tag{14}$$

where g_k refers to the current gradient, a_k denotes the learning rate, and x_k denotes the vector containing the current weights and biases. Kim et al. [55] examined the ANN optimization strategy utilizing WGSR data followed by a backpropagation algorithm.

The backpropagation algorithm usually develops the partial derivates of a cost function (C) based on the weight (w) and bias (b) of the network (Eqs. (15)–(16)). The cost function analyses the difference between the experimental output and the predicted output, which is referred to as the performance of the backpropagation algorithm. The cost function is derived as follows (Eq. (17)),

$$a_k^l - \delta_j^l = u\frac{\partial C}{\partial w_{jk}^l} \tag{15}$$

$$\delta_j^l = u\frac{\partial C}{\partial b_j^l} \tag{16}$$

$$C = \frac{1}{2n}\sum_x ||y(x) - a^L(x)||^2 \tag{17}$$

where n denotes the number of training, y(x) denotes the expected output and $a^L(x)$ denotes the vector of activations, L is the number of layers and x denotes the input [58]. The error of the output layer is written as follows (Eq. (18)),

$$\delta_j^L = u\frac{\partial C}{\partial a_j^L}\sigma'(z_J^L) \tag{18}$$

$\partial C/\partial a^L$ calculates the cost change based on the function of the activation of the j^{th} output neuron. $\sigma'(z_j^L)$ measures the change of activation function at the j^{th} neuron of the output layer, whereas Δz^L is the little change in the neuron's weight [58]. The backpropagation algorithm can be trained without previous knowledge, accounting for its simplicity and flexibility. The implementation of this algorithm is quick. Backpropagation is a frequently used algorithm in machine learning optimization due to its extraordinarily high performance. Backpropagation algorithms have certain drawbacks, including the fact that their performance is largely dependent on the quality of the training data, training takes a relatively long time, and they could potentially result in noisy data [58]. Using a backpropagation algorithm, Chen et al. [59] investigated methanol-reforming for hydrogen production. Several data sets were employed in this study, and the model's accuracy increased with the quantity of data points in the dataset. The R^2 value for the MSR was 0.7 when 36 data sets for different parameters were utilized, but it increased significantly to 0.92 and 0.98 when 48 and 60 data sets were used, respectively. The ratio of training to test data sets was 5:1.

2.4.2. Levenberg–Marquardt algorithm

The Levenberg-Marquardt (LM) algorithm was developed to obtain second-order training speed in the ANN model without computing the Hessian matrix. The Hessian matrix can be represented as follows (Eq. (19)) if the artificial neural network learning performance function is a form of a sum of squares. The gradient is computed as follows (Eq. (20)) [42].

$$H = J^T J \tag{19}$$

$$g = J^T e \tag{20}$$

$$x_{k+1} = x_k - \left[J^T J + \mu I\right] J^T e \tag{21}$$

J represents the Jacobian matrix, which can be generated using the backpropagation method and is much easier to calculate than the Hessian matrix [56]. This matrix contains the network error corresponding to the bias and weight, whereas e denotes a vector of network errors. A Newton-like update is being used by the Levenberg-Marquardt method as an alternative to the Hessian matrix (Eq. (21)), where μ is a scalar quantity and the algorithm changes based on its value. When μ=0, the model adapts the newton design. As the goal is to achieve the 0 value for μ, the value of μ will be decreased after a successful step [56]. The gradient's descent adjusts with each iteration, and the sum of square values progressively decreases to the direction of descent, updating the solution. The mean square and solution have two directions of progression with each iteration, even though it tends to find the steepest descent; this is one of the major benefits of the LM algorithm [60]. Compared to Gaussian-Newtonian approaches, the LM algorithm converges substantially quicker to the optimal value. This approach applies to various parameters that are not even defined or specified. When the initial prediction is away from the actual value, this method attempts to converge toward the optimal solution. Therefore, the LM model is the most widely employed algorithm for machine learning optimization technology [60]. Nevertheless, if the parameters are greater than ten, this algorithm will function slowly or converge slowly to the optimal value, causing complications in the implementation of those models.

Song et al. [42] used the Levenberg-Marquardt and backpropagation algorithms to optimize the operating parameters for synthesizing catalysts (Ni/Al_2O_3) in EtOH reforming and identify the maximum conversion of EtOH and H_2 production. The highest EtOH conversion and H_2 selectivity were 79.6% and 91.4 %, respectively, while the optimum hydrogen production was 4.4 mol%, with a mean square error of 0.006, indicating that the learning algorithm is suitable for this ANN model. Doicin et al. [61] employed the Levenberg-Marquardt algorithm in an ANN model with successive iterations (3, 10, and 20) to determine which iteration resulted in the maximum ethanol conversion and hydrogen production in a predicted model. The relative errors were between −3 and 3% when predicting a high hydrogen yield after 10 iterations, indicating the learning algorithm is appropriate for simulating predicted data. Matei et al. [25] employed the Levenberg-Marquardt algorithm in an ANN model to optimize the operational factors for ESR over a Pd/SBA-15 catalyst. The data regression analysis of target versus predicted results (ethanol conversion and hydrogen production) exhibited that the R-value is near 1, indicating that the training method was efficient.

2.4.3. Nelder-Mead algorithm

The Nelder-Mead simplex algorithm is one of the most extensively employed direct search techniques for resolving optimization problems [56], where f: Rn → R, R refers to the objective function with n dimension. A simplex refers to a geometric figure with n dimensions with the convex hull of x_{n+1} vertices. A geometric object with n dimensions and a convex hull of xn + 1 vertices is called a simplex. The Nelder-Mead technique repeatedly creates a series of simplices to approach an ideal point of (min f (x)) [62]. The simplex's vertices are arranged based on the values of the objective function at each iteration. This algorithm has multiple steps, including shortening the vertices, computing the expansion and reflection points, and the inside and outside computation of the shrinkage points [62]. Chen et al. [56] employed the Nelder-Mead algorithm in an ANN model to optimize MSR processes' operating conditions over a Cu-Zn catalyst. The predicted vs. actual methanol conversion and hydrogen production graphs had R^2 values of 0.98 and 0.98, respectively, signifying the suitability of the learning algorithm for this ANN model.

3. Optimization through artificial neural networks application

The efficiency of H_2 production via steam reforming of different biofuels, such as methanol or ethanol, is determined by the kind of

catalyst used, its physical and chemical characteristics, and, most crucially, the operating conditions of the reforming processes. Methanol or ethanol thermochemical conversion is one of the most extensively used, efficient, cost-effective, and rapid techniques for producing hydrogen. According to a literature search, the ANN optimization technique is widely employed for steam reforming of methanol (MSR) and ethanol (ESR) because of its outstanding performance for methanol and ethanol conversion and hydrogen production. MSR and ESR approaches are both involved with the reaction of fuels with water and water–gas shift reaction [10,21,37]. Steam reforming reactions are endothermic reactions in which the reactants absorb heat from their surroundings, and the temperature drops in the vicinity. As a result, the catalytic deactivation of the steam reforming reaction is lower when compared to partial oxidation, which is an exothermic reaction associated with oxygen intake (Eqs. (22)–(26)) [10,37]. Methanol conversion reached nearly 100% at temperatures ranging from 350 to 400 °C, whereas ethanol conversion peaked at 500 to 600 °C for a non-noble metal catalyst. With the noble metal catalyst, the reforming temperature for maximum ethanol conversion is even higher (greater than600 °C). Copper is a widely employed metal catalyst for the MSR reaction, although Ni, Pd, Rh, and Pt are also tested for MSR. Co and Ni are the most often utilized catalysts for the ESR, yet other metals such as Rh, Pd, Pt, Ag, and Au are also investigated to see their performance for ethanol conversion [10,37]. Al_2O_3, ZnO, and MgO are the most tested support material for the MSR and ESR reaction due to their great performance and metal-support complex stability. Non-noble metals are comparatively superior catalysts due to their vacant d-shell electrons and electron exchange capacity, which improves reaction performance [10,37].

$$CH_3OH + H_2O \rightleftharpoons 2H_2 + CO_2,\ \Delta H^0_{298} = 49.5\ kJ{\cdot}mol^{-1} \quad (22)$$

$$CH_3OH + 0.5O_2 \rightleftharpoons 3H_2 + CO_2,\ \Delta H^0_{298} = -192\ kJ{\cdot}mol^{-1} \quad (23)$$

$$CH_3CH_2OH + 3H_2O \rightleftharpoons 6H_2 + 2CO_2,\ \Delta H^0_{298} = 174\ kJ{\cdot}mol^{-1} \quad (24)$$

$$CH_3CH_2OH + 1.5O_2 \rightleftharpoons 3H_2 + 2CO_2,\ \Delta H^0_{298} = -545\ kJ{\cdot}mol^{-1} \quad (25)$$

$$H_2O + CO \rightleftharpoons H_2 + CO_2,\ \Delta H^0_{298} = -41.2\ kJ{\cdot}mol^{-1} \quad (26)$$

Thus, optimizing the catalytic conditions and process parameters of methanol or ethanol reforming is critical for understanding their practical and commercial application and increasing hydrogen production. This literature review shows that most prior analyses emphasized the optimization of operating parameters and catalyst type for ethanol reforming through ANN technology, with hardly any consideration of the designs of reactors [11,25,42,61,63,64]. In recent years, heterogeneous catalysts have been designed using ANNs [42,53,65], and several studies have been published. The following part sought to offer a comprehensive review of the design and training of an ANN in the steam reforming of methanol/ ethanol.

3.1. Catalyst development and optimal parameters for methanol reforming

ANN technology is a widely used mathematical model of machine learning, and it has been employed to enhance the process parameters of methanol reformation and catalyst development. The literature study reveals that most studies attempted to improve hydrogen production by modulating the methanol-reforming temperature, the steam-to-feed (carbon) ratio, and the volume percentage of feedstock [56,64]. Methanol is regarded as one of the most efficient sources of hydrogen for application in fuel cells. The amount of hydrogen produced during methanol reformation can fluctuate significantly depending on the process parameters, impacting how efficiently fuel cells operate. Consequently, this study intends to identify the most efficient parameters of MSR. The two major processes associated with the MSR technique are steam uptake and WGSR [12]. Most tests for the MSR reaction are conducted at temperatures between 150 °C and 300 °C (Eqs. (22), (23), and (27)) [12].

$$CH_3OH \rightleftharpoons 2H_2 + CO,\ \Delta H^0_{298} = 90.1\ kJ{\cdot}mol^{-1} \quad (27)$$

Chen et al. [56] applied an ANN to optimize the operation conditions of MSR reaction over a Cu-Zn catalyst (Fig. 4 and Table 1). The reforming temperature (200 °C-300 °C), steam-to-carbon ratio (0.5–3.5), and Reynolds numbers (50–500) were the input parameters for this experiment. At the same time, the percentage of methanol conversion and hydrogen yield were designated as the output variables. The results of this investigation revealed that higher reaction temperatures (300 °C) and a higher steam-to-feed (carbon) ratio (3.5) were both favorable for the MSR reaction. Maximum hydrogen yield was attained with the optimal parameter combinations (2.905 mol (mol $CH_3OH)^{-1}$). The data regression analysis of target versus predicted results (methanol conversion and hydrogen production) exhibited that the R^2-value is near 1, indicating that the training method was efficient. Methanol conversion proceeds at substantially lower temperatures than ethanol conversion because the C-C bond is absent in methanol, and the elimination of hydrogen often occurs at lower temperatures. Mobarake et al. [64] optimized the operation parameters of MSR reaction over a Cu/ZnO/Al_2O_3 catalyst using an artificial neural network (Table. 1). The input parameters for this experiment were the reforming temperature, steam-to-feed ratio, CO_2, and H_2 in the feed. The output variables were the percentage of methanol conversion, hydrogen selectivity, and hydrogen yield. This research investigation demonstrated that higher reaction temperatures and a steam-to-carbon ratio were both favorable for methanol steam reforming. The MSR reaction was frequently tested at lower temperatures than the ESR reaction, with temperatures between 150 °C and 400 °C. Methanol may be transformed entirely at temperatures close to 350 °C [66], so a temperature of more than 350 °C is unnecessary for methanol-reforming. However, only 75% of ethanol is converted at 400 °C and 100% at nearly 600 °C [10]. The major reaction of both ethanol and methanol-reforming is the elimination of hydrogen, which usually occurs between 200 °C and 400 °C, whereas the C-C bond of ethanol breaks above 400 °C [10]. The literature review revealed that the steam-to-feed ratio and the reforming temperatures benefit the MSR process [56,64]. The S/M ratio range (0.5–3.5) for methanol steam reforming is lower than the S/E ratio range (3.5–5) for ethanol steam reforming [56,67]. Both methanol and ethanol conversion increase as the reforming temperature increases. MSR more efficiently occurs at lower temperatures than ESR due to methanol's lack of a C-C bond. The key mechanism, such as eliminating hydrogen reaction, is similar at lower temperatures (<400 °C) in both methanol and ethanol reformation. C-C bond breaking only occurs at higher temperatures in the ESR process.

3.2. Catalyst development and optimal parameters for ethanol reforming

The principal mathematical approach of machine learning for optimizing the process parameters of ethanol reformation is computer programming-assisted ANN technology. The literature study reveals that the vast majority of studies attempted to maximize hydrogen production by modifying reaction temperatures, the S/E ratio, catalyst type, and preparation methods of catalysts [25,42,61,63]. In the context of fuel cells, ethanol is thought to be one of the most effective sources of hydrogen. The conditions under which ethanol reformation is conducted may significantly alter the amount of hydrogen produced, affecting how well fuel cells function. Thus, this study aims to determine the most influential parameters and their quantification to identify whether a variable should be raised or dropped.

Cursaru et al. [63] employed the ANN technology in the ESR reaction over a Co/MCM-48 catalyst and targeted it to optimize the synthesis

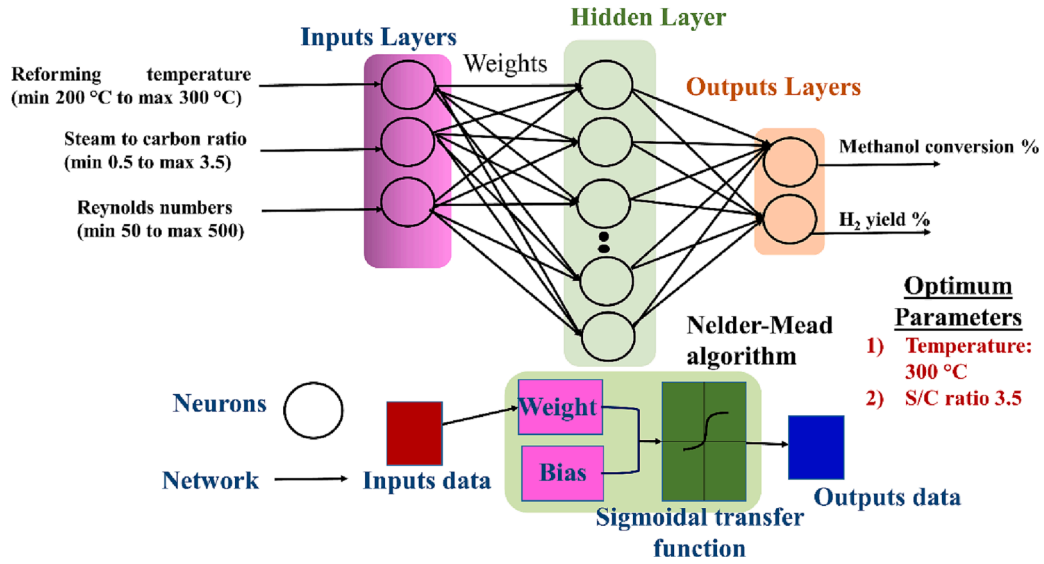

Fig. 4. The illustration of artificial neural network model application in the MSR reaction, where data are collected from Chen et al. [56]. The results of this investigation revealed that higher reaction temperatures (300 °C) and a higher S/C ratio (3.5) were both favorable for the MSR reaction. Maximum H_2 yield was obtained with the optimal parameter combinations (2.905 mol (mol $CH_3OH)^{-1}$).

Table 1
The artificial neural network model for MSR for H_2 production.

Reference	ANN type	Aim	Topology	Input and output parameters	Validation	Optimizing parameters and application
Chen et al. [56]	Supervised	The ANN to optimize the operation conditions of MSR over a Cu-Zn catalyst	This model has three ANN types. Layer: Input, hidden, and output. Neuron per layer: ANN1, 5–5-4; ANN2, 5–5-3; and ANN1, 5–5-3. Transfer function: sigmoidal. Learning algorithm: Levenberg–Marquardt and backpropagation	Input parameters: temperature (min 200 °C to max 300 °C), S/C ratio (min 0.5 to max 3.5), and Reynolds numbers (min 50 to max 500). Output parameters: MeOH conversion and H_2 yield	Mean square error: ANN1, 0.008; ANN2, 0.006, and ANN3, 0.009	Higher reaction temperatures (300° C) and a higher S/C ratio (3.5) were both favorable for the MSR reaction. Maximum H_2 yield was attained with the optimal parameter combinations (2.905 mol (mol $CH_3OH)^{-1}$).
Mobarake et al. [64]	Supervised	ANN model to predict the high hydrogen yield through MSR over a $Cu/ZnO/Al_2O_3$	Neuron per layer: ANN1, 5–8-3 Transfer function: sigmoidal. Learning algorithm: Levenberg–Marquardt	Input parameters: temperature, water-to-feed ratio, carbon dioxide, and hydrogen in the feed. Output parameters: percentage of H_2 yield and EtOH conversion	The R^2 (0.983) and MSE (0.00172) values show a respectable agreement between the predicted and actual values	Higher reaction temperatures (500.1 °C) and S/M feed ratio are both favorable for ESR

parameters of the catalysts (Fig. 5 and Table 2). Another goal of this study was to establish a relationship between the chemical characteristics of the catalyst and their performance in ethanol reformation and hydrogen production. In the first aim of this study, calcination temperature (520 °C, 560 °C, and 600 °C) and stirring times of synthesis (2 h, 9 h, and 15 h) were input factors, and the BET surface area, pore volume (PV), and pore size were the outputs variables of ANN-1. In the second aim of this study, calcination temperature and stirring periods of synthesis were input parameters, while the ethanol conversion and hydrogen yield were the outputs variables of ANN-2. This investigation revealed that the maximum values for the intended outcome, such as the conversion of EtOH and H_2 yield, were attained when the stirring duration for the synthesis of the catalysts was 15 h and the sintering temperature was roughly 560 °C −570 °C, which was equivalent to the optimum results of stirring time (15 h) and sintering temperature (560 °C −600 °C) stored for the training data. The data regression analysis of target versus predicted results (EtOH conversion and H_2 production) exhibited that the R-value is near 1, indicating that the training method was efficient [63]. The increase in the surface area caused by the higher sintering temperature was likely to enhance the metal dispersion and catalytic activities, resulting in increased production of hydrogen [68,69]. In contrast, high sintering temperatures might promote catalyst deactivation. According to a previous study, the catalyst in steam reforming sometimes is deactivated due to a reduction in nickel dispersion mediated by nickel sintering [70,71]. The production of large particles by Ostwald ripening at sintering temperatures above 650 °C reduced the catalytic surface area. As the surface area decreased, the fraction of ethanol sorption dropped, potentially limiting ethanol conversion [72].

Song et al. [42] employed ANN in the ESR reaction over Ni/Al_2O_3 catalysts, attempted to comprehend the optimum parameters for the catalyst preparation methods and reaction parameters, and determined the highest ethanol conversion and hydrogen production (Fig. 5 and Table 2). In the first objective of this study, the preparation methods (co-precipitation, precipitation, and impregnation) and weight percentage of Ni (10, 15, 20, and 25 wt%) and Al (47, 44, 42, and 39 wt%) in the

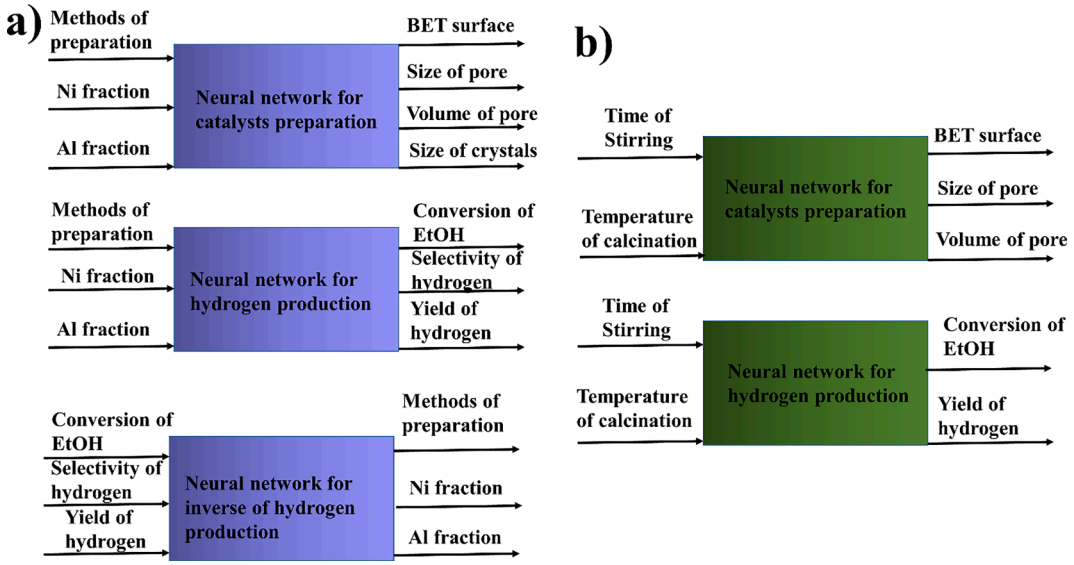

Fig. 5. The illustration of artificial neural network model application in the ESR reaction. (a) In order to understand the optimal parameters for the catalyst preparation techniques and reaction parameters and identify the maximum levels of EtOH conversion and H_2 production, Song et al. [42] applied the ANN application in the ESR reaction over Ni/Al_2O_3 catalysts. The first goal was to use artificial neural networks (ANNs) to prepare catalysts; the second was to use them to produce H2; and the third was to produce inverse H_2 production. (b) In the ESR reaction over a Co/MCM-48 catalyst, Cursaru et al. [63] used the ANN technique to optimize the catalysts' synthesis parameters. Another objective of this investigation was to establish a link between the catalysts' chemical and physical characteristics and their efficiency in EtOH reformation and H2 production.

Table 2
The ANNs model for the ESR reaction for H_2 production.

Reference	ANN type	Aim	Topology	Input and output parameters	Validation	Optimizing parameters and application
Song et al. [42]	Supervised	ANN application for the characteristics of the Ni/Al_2O_3 catalysts in ESR reaction and attempted to comprehend the optimum parameters for the catalyst preparation methods and reaction parameters, as well as determined the highest EtOH conversion and H_2 production.	This model has three ANN types. Layer: Input, hidden, and output. Neuron per layer: ANN1, 5–5-4; ANN2, 5–5-3; and ANN1, 5–5-3. Transfer function: sigmoidal. Learning algorithm: Levenberg–Marquardt	Input parameters ANN1 and ANN2: (1) preparation methods and (2) Ni and (3) Al loading.; ANN3: (1) EtOH conversion, (2) H_2 selectivity, and (3) yield; Output parameters: ANN1, (1) BET surface, (2) size of pore, (3) volume of pore, and (4) crystallite size; ANN2, (1) EtOH conversion, (2) H_2 selectivity and (3) yield; ANN3, (1) preparation methods and (2) Ni and (3) Al loading.	Mean square error: ANN1, 0.008; ANN2, 0.006, and ANN3, 0.009	In the co-precipitation method, aluminum, and nickel were 42.49 % and 12.35 % are optimum. The conversion of EtOH and H_2 selectivity for the optimal H_2 production was 79.6 and 91.4 mol %
Doicin et al. [61]	Supervised	ANN model to predict the high H_2 yield through ESR over a Co/Al_2O_3	Neuron per layer: ANN1, 3–10-2 Transfer function: sigmoidal. Learning algorithm: Levenberg–Marquardt	Input parameters: temperature (350 °C, 400 °C, and 450 °C), EtOH concentration (8 vol% and 10 vol%), and flow rate (1 mL/min and 3 $mL.min^{-1}$), Output parameters: H_2 yield and EtOH conversion	The error histogram results of this study varied from −1.595 to 1.338	The optimal number of iterations was 10, and relative errors ranging between −3 and 3% with a very high H_2 yield (77%-86%) estimated by the predicted model
Matei et al. [25]	Supervised	ANN for optimizing the operation condition of ESR over a Pd/SBA-15 catalyst	Neuron per layer: ANN1, 3–10-2 Transfer function: sigmoidal. Learning algorithm: Levenberg–Marquardt	Input parameters: temperature (300 ℃, 350 ℃, 400 ℃, 450 ℃, and 500 ℃), EtOH concentration (5 vol%, 10 vol % and 15 vol%), and WHSV (h^{-1}); output parameters: EtOH conversion and H_2 yield	Testing and training R values of 0.905 and 0.972	Higher reaction temperatures and EtOH concentrations are both favorable for ESR
Cursaru et al. [63]	Supervised	ANN technology in the ESR reaction over a Co/MCM-48 catalyst and aimed to optimize the catalyst synthesis parameters	Neuron per layer: ANN1, 3–10-2 Transfer function: the sigmoidal tangent. Levenberg–Marquardt	Input parameters: calcination temperature and stirring times of synthesis; Output parameters: the BET surface area, volume of pore, and size. of pore	The R-value is near 1	Stirring Duration: 15 h and the sintering temperature was roughly 560–570 °C

catalyst were designated as the input parameters, where the calcination temperature (600 °C) of the catalyst and reaction temperature (400 °C) were set up to be constant. The BET surface area, pore volume (PV), pore size, and crystallite size (CS) were the final output variables of ANN-1 ($5N_{input}$–$5N_{hidden1}$–$4N_{output}$, N refers to Neurons). In the second objective of this study, the preparation methods (co-precipitation, precipitation, and impregnation) and weight percentage of Ni and Al were the input function, when the calcination temperature of the catalyst was constant. The EtOH conversion, H_2 yield, and selectivity are the final output parameters of ANN-2 for the function X = f (Y) ($5N_{input}$–$5N_{hidden1}$–$3N_{output}$) [22]. In the third objective, since the H_2 yield, H_2 selectivity, and EtOH conversion were known, it might be useful to know both the characteristics of the catalysts and the operating conditions of the reactions following by the inverse scenario, which was designated as ANN-3 for the function of $X = f^{-1}$ (Y) ($5N_{input}$–$5N_{hidden1}$–$5N_{output}$). Table 3 and Fig. 6 demonstrate the actual and predicted data of EtOH conversion and H_2 selectivity, which were obtained by experiments and the ANN model, respectively. The EtOH conversion and H_2 selectivity for the optimum H_2 production were 79.6 and 91.4%, respectively, while the optimum hydrogen production was 4.4 mol%. The co-precipitation method was the most effective way to produce a catalyst for hydrogen production; the optimum weight percentage of aluminum and nickel in the catalyst was 42.49 and 12.35, respectively. A previous study investigated the efficiency of a catalyst produced by co-precipitation and impregnation during an ESR reaction [73]. The weak interface between cobalt and CeO_2 resulted in greater cobalt dispersion in the CeO_2 lattice when additional cobalt ions were integrated into the co-precipitated metal-support complex (Co_3O_4/CeO_2). Furthermore, this process made it easier to convert cobalt oxides into cobalt metal, actively regulating ethanol's steam reforming [73].

Szijjárto et al. [53] investigated the optimization of multicomponent catalyst formation for ethanol reforming using ANNs with holographic maps. They identified the optimal metal combination in the catalyst and the optimum temperature for the reaction (Table 2). Apart from the optimization feature, the holographic research technique can depict multidimensional experimental space and multidimensional space in a two-dimensional structure. As a result, the holographic map with ANNs both virtual optimization and data visualization. Holographic maps are used to represent multidimensional functions in two dimensions. The variables (the proportion of different metals in the catalyst) are organized along the X and Y axes in the holographic maps. Different wave-like lines indicate different variables, and the length of the line is proportional to the data point along that line [74]. This allows for the visual analysis of complex hypersurfaces and the revelation of interconnections in multidimensional spaces [75,76]. This research studied several catalysts such as Pt (0–3 wt% in compared to the $MgAl_2O_4$ support), Ni (0–10 wt%), Co (0–10 wt%), Cu (0–10 wt%), Zn (0–5 wt%), La (0–6 wt%), Ce (0–7 wt%), and Zr (0–6 wt%) for ethanol reformation at temperatures ranging from 350 °C to 450 °C. This investigation revealed that Pt catalyst would moderately impact ethanol reformation and hydrogen production. However, hydrogen production was increased with increasing Ni concentration. Co had no noticeable effect when Ni was absent, but hydrogen production increased noticeably when Ni and Co were together in the catalyst. Cu improved catalytic performance when the Ni level was zero or low, but it reduced hydrogen production activity when the Ni level was high. In contrast to earlier studies [77], adding Ce metal to the catalyst decreased hydrogen production. Ce had a positive effect when Zn was not present in the catalyst. As Zn had a negative impact, its absence increased hydrogen production. Pt (3 wt%), Ni (10 wt%), Co (10 wt%), Cu (0 wt%), Zn (0 wt%), La (0 wt %), Ce (7 wt%), and Zr (0 wt%) were the optimum combinations of different metal percentages in the catalyst for ethanol reforming at 450 °C. The hydrogen production was shown to be positively influenced by Ni, Co, and the reaction temperature. However, the interplay between Ni and Co and Ni, Co, and Ce could only be determined using ANNs [53]. Ni and Co are non-noble metal catalysts that can function well even at temperatures below 400 °C [78–80]. In contrast, Pd, Pt, Rh, and Ru are noble metals that frequently exhibit low catalytic activity at temperatures below 400 °C and function effectively at temperatures above 400 °C [81,82].

Matei et al. [25] employed an ANN to optimize ESR's operation conditions over a Pd/SBA-15 catalyst. The input parameters for this experiment were the reforming temperature (300 ℃, 350 ℃, 400 ℃, 450 ℃, and 500 ℃), ethanol concentration (5 vol%, 10 vol%, and 15 vol %), and gas hour space velocity (3 h^{-1}, 9 h^{-1}, and 15 h^{-1}), while the output data was the percentage of EtOH conversion and H_2 yield. The data regression analysis of target versus predicted results (ethanol conversion and hydrogen production) exhibited that the R-value is near 1, indicating that the training method was efficient. This study revealed that higher reaction temperatures and ethanol concentrations are both favorable for ethanol steam reforming, which is in line with previous studies [10,81,83]. At temperatures between 200 °C and 400 °C, the

Table 3
Actual and predicted data of EtOH conversion and H_2 selectivity were obtained by experimental and ANNs models, respectively.

Process related	Catalyst Composition		Experimental	Estimated	Experimental	Estimated	Reference
	Ni (wt%)	Al (wt%)	EtOH conversion %		H_2 selectivity (%)		
Co-precipitation	11.5	45.8	40.0	45.8	89.0	90.7	[42]
	13.5	45.7	65.0	64.6	90.0	90.3	
	16.5	44.2	62.0	71.5	88.0	89.8	
	18.5	43.1	57.0	61.6	91.0	89.7	
Precipitation	11.5	45.8	45.0	56.4	87.0	85.4	
	13.5	45.7	60.0	72.9	86.0	83.4	
	16.5	44.2	81.0	84.4	84.0	81.9	
	18.5	43.1	83.0	83.6	83.0	81.9	
Impregnation	11.5	45.8	43.0	44.9	86.0	85.8	
	13.5	45.7	45.0	46.1	86.0	87.0	
	16.5	44.2	47.0	47.0	88.0	87.3	
	18.5	43.1	47.0	46.9	86.0	86.5	
3 iteration	Co/Al_2O_3		60.3	60.9	77.1	73.3	[61]
			60.0	60.2	74.2	74.1	
			59.8	60.7	81.5	81.2	
			59.5	59.7	79.5	81.6	
			59.7	59.6	87.9	85.6	
10 iteration			60.3	60.3	77.1	77.1	
			60.0	60.2	74.2	73.6	
			59.8	59.9	81.5	83.4	
			59.5	60.1	79.5	81.2	
			59.7	59.7	87.9	87.3	

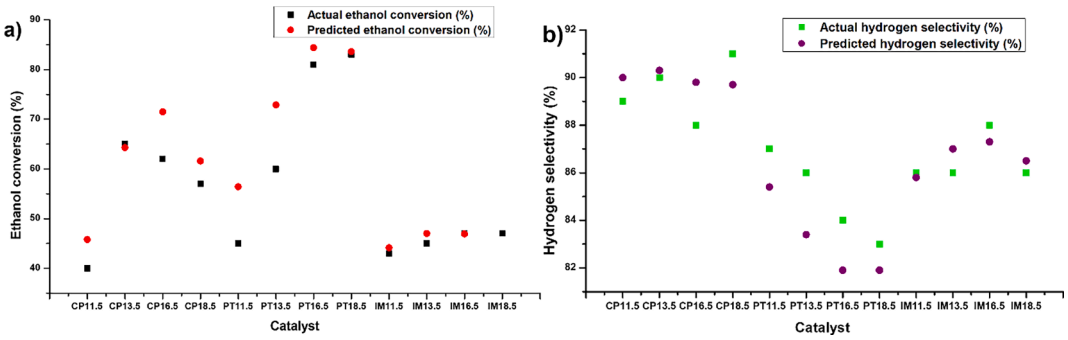

Fig. 6. The illustration of artificial neural network model application in the ESR reaction. To understand the optimal parameters for the catalyst preparation techniques and reaction parameters and identify the maximum levels of EtOH conversion and H_2 production, Song et al. [42] applied the ANN application in the ESR reaction over Ni/Al_2O_3 catalysts.). This image demonstrated the actual and predicted data of EtOH conversion and H_2 selectivity, which experiments and artificial neural networks model, respectively, obtained. EtOH conversion and H_2 selectivity for the optimum H_2 production was 79.6 and 91.4 %, respectively. The co-precipitation method was the most effective way to produce a catalyst for hydrogen production; the optimum weight percentage of Al and Ni in the catalyst was 42.49 and 12.35, respectively.

hydroxyl group of ethanol binds to the active site of the metal catalyst and initiates a dehydrogenation process, which releases hydrogen. The C-C bond is often broken at temperatures over 400° C, and hydrogen selectivity can approach 90% [84–87]. The steam reforming of ethanol increased with increasing reaction temperature from 200 °C to 400 °C. According to the literature review, hydrogen selectivity reached 80 to 90% at temperatures ranging from 600 °C to 700 °Cover Ni or Co-based catalyst. Over noble metal-based catalysts, the ESR reaction occurs at a higher temperature (600 °C −800 °C). As the reforming temperature of ethanol varies for various catalysts and their physical and chemical characteristics, the other experimental parameters may be adjusted based on the catalyst types.

In accordance with the literature, one of the most strongly influenced parameters for ethanol reformation is the reforming temperature determined by ANN optimization technology. The ethanol conversion gradually increased from 300 °C to 400 ℃, whereas maximum ethanol conversion occurred at 500 °C [25]. Although ANN technology did not take into account reforming temperatures over 500 ℃, the RSM optimization methodology revealed that temperature increased ethanol conversion (above 85%) at a temperature above 500 °C for ESR [67]. Therefore, 500 °C might be an adequate reforming temperature for ESR techniques. According to the literature review, when the volume % of ethanol increased from 5 to 10% at a temperature of 500 °C, ethanol conversion increased from 80 to 88% [25]. The ethanol conversion is regulated not only by the feedstock content (Vol%) but also by the steam content and the catalyst concentration, and ethanol may reform efficiently if all three parameters increase simultaneously. The weak interaction between metal and supports led to higher metal dispersion in the support lattice when more metal ions were added through the co-precipitation process, rendering this the optimum technique for synthesizing catalysts [42]. The non-noble metals, such as Ni and Co, are optimum metal catalysts due to their substantial hydrogen elimination capability. They also have high catalytic activity at even lower temperatures, allowing for various intermediate reactions and regulating hydrogen and ethanol conversion production. The ANN investigation also revealed that the optimum sintering temperature for catalyst synthesis was roughly 560 °C −570 °C [63].

3.3. Catalyst development and optimal parameters for water gas shift reaction

This study also intends to understand the optimization of the operating parameters of the WGS reaction using ANNs and compare the processes with ESR and MSR to identify similarities and differences in various hydrogen production processes. The reaction temperatures, various metal catalyst types, the steam-to-feed (carbon) ratio, and GSHV are the independent variables or input parameters for the WGSR [55,88]. Kim et al. [55] investigated ANN optimization strategy utilizing WGSR data. The WGS reaction was carried out using $Pt/Ce_xZr_{1-x}O_2$ catalysts to determine the optimal reaction parameters, including temperatures ranging from 200 °C to 450 °C, Pt contents ranging from 0.3 to 3.0 wt%, and steam-to-feed (carbon) ratio ranging from 5 to 9 (Fig. 7 and Table 4). Kim et al. [55] demonstrated that a low fraction of Pt content (0.3 wt%) in a catalyst positively affects the performance of $Pt/Ce_xZr_{1-x}O_2$ catalyst owing to the high reactivity of Ce fraction. When the Ce concentration was high, CO was usually converted at a lower temperature (250 °C). However, Ce did not significantly affect CO conversion when the temperature exceeded 400 °C [55]. Zr had a lesser activity than Ce-based catalysts. The high steam-to-feed ratios, roughly 9 to 10, resulted in significant CO conversion. Chen et al. [89] optimized ESR using the Taguchi optimization methods and demonstrated that increasing the temperature of the WGSR (400 °C) was beneficial for hydrogen production, and temperatures above 400 ℃ might not have a significant impact on the conversion of steam and CO *via* the WGSR. Chen et al. [90] revealed that water decomposition via the WGSR was higher, resulting in a higher hydrogen yield than carbon dioxide. If the S/C ratio increased, the WGSR accelerated [90].

Cavalcanti et al. [88] (Table 4) applied the model of an ANN for optimizing the operational parameters of the WGSR. The input layer comprises the data of catalyst and reaction conditions, such as the WGSR temperature (200–450 °C) and pressure (0.8–27.6 bar), hourly gas space velocity (795–1,200,000 h^{-1}), carbon monoxide feed content (1.30–37.2 vol%), carbon dioxide feed content (0–96 vol%), water feed content (1.5–69.2 vol%), nitrogen (inert gas) feed content (0–96.5 vol %), methane feed content (0–0.70 vol%), metal in the catalyst (Co, Cu, Ni, Ru, Ir, Pt, Pd, Ag, Cr, Au, and Zn), support type (CNT, Mo_2C, Fe_2O_3, AC, CeO_2, MgO, Al_2O_3, La_2O_3, ZrO_2, and TiO_2), promotor (Na, Ba, B, K, Mg, Al, Si, Hg, Y, Pb, S, Ti, Ce, Zr, La, and Fe), surface area (1.1–1487 m^2g^{-1}), and calcination temperature (25–800 °C) and time (0–10 h). This investigation revealed that ceria-support and Ni, Cu, and Ru metal catalysts, temperatures (300 °C), and gas hour space velocities (2000–6000 h^{-1}) were the optimum parameters for the WGSR. The R^2 (0.983) and mean square error (MSE) (0.00172) values show a good agreement between the predicted and observed values. Due to the elimination of hydrogen and C-C bond breakage, 90% of ethanol could be converted at temperatures between 500 °C and 600 °C [84–87], whereas 95% of methanol could be converted at temperatures of 350 °C [66]. The WGS reaction occurs at a temperature lower than ESR but higher than MSR.

The S/C ratio is one of the most influential factors in the WGS reaction [55,88]. Another optimum parameter obtained by the ANN model is the reaction temperature, which is crucial for the production of

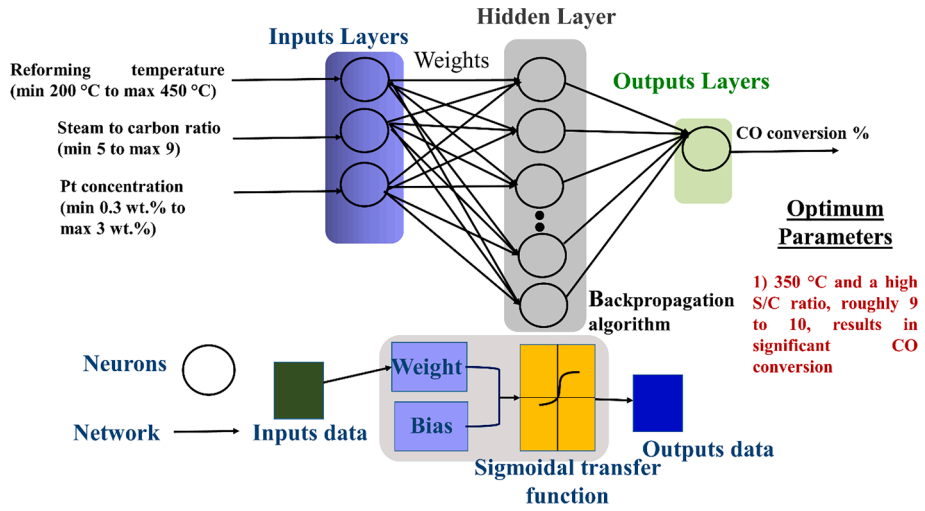

Fig. 7. The illustration of ANNs model application in the WGS reaction, where data are taken from Kim et al. [55]. Utilizing WGSR data, this study investigated the ANN's optimization approach. Pt/CexZr$_1$-xO$_2$ catalysts were employed in the WGS reaction to determine the optimum process parameters. At temperatures more than 350 °C, all cases achieved maximum CO conversion. The high amount of S/C ratio, roughly 9 to 10, results in significant CO conversion.

Table 4
Optimization of WGSR via ANN method.

Reference	ANN type	Aim	Topology	Input and output parameters	Validation	Optimizing parameters and application
Kim et al. [55]	Supervised	ANNs optimization strategy utilizing WGSR data. The WGSR was carried out using Pt/$Ce_xZr_{1-x}O_2$ catalysts.	Neuron per layer: 5_h14_h56_h28_h14_h7_1 Transfer function: sigmoidal. Learning algorithm: backpropagation	Input parameters: (Pt loads, support ratio (x: Ce/[Ce + Zr]), calcination temperature, calcination time, operating temperature, W/F, CO ratio, CO_2 ratio, H_2O ratio, and H_2 ratio Output parameters: CO conversion	obtained 0.988 R^2, 12.639 MSE, and 3.555 RMSE; thus, the developed model is reliable.	Ce concentration was high; the CO was usually converted at a lower temperature (250 °C). However, Ce had no significant effect on CO conversion when the temperature exceeds 400 °C. Zr had a lesser activity than Ce-based catalysts.
Cavalcanti et al. [88]	Supervised	Thermochemical catalytic conversion of the WGSR process for the H_2 production	Neuron per layer: 51 input, 1 output. Transfer function: sigmoidal. Learning algorithm: Backpropagation	Input parameters: temperature, pressure, hourly gas space velocity, CO feed content, CO_2 feed content, H_2O feed content, N_2 (inert gas) feed content, CH_4 feed content, metal in the catalyst Output parameters: percentage of H_2 yield and EtOH conversion	After training the network, the model found that the MSE value is 0.297	The ceria-supported catalysts with higher surface areas and Ni, Cu, and Ru metal catalysts, temperatures (300 °C), and space velocities (2000–6000 h^{-1}) were the optimum parameters for the WGSR found by this study.

H_2 and the conversion of CO. The temperature is somewhat less than ESR. Although the S/C ratio and temperature ranges for the MSR, ESR, and WGSR techniques differ, the basic concepts of the optimal variables were comparable.

4. Prediction of hydrogen yield *via* artificial neural network

Using ANN-driven modeling can identify a range of operational conditions for methanol and ethanol reforming, optimum hydrogen yield, and ethanol conversion, which are both theoretically and practically feasible regarding hydrogen production. One benefit of using ANN-driven simulation software in engineering applications is its ability to adjust to factors based on prior knowledge about an expected outcome. It also assists in obtaining high product yield through simulation (Tables 2 and 5).

Doicin et al. [61] (Fig. 8) employed an ANN model to investigate its efficiency and predict the high hydrogen yield through ESR reaction over a Co/Al_2O_3 catalyst and operational conditions. The input parameters of this model were temperature (350 °C, 400 °C, and 450 °C), ethanol concentration (8 vol% and 10 vol%), and flow rate (1 mL·min^{-1} and 3 mL·min^{-1}), where the percentage of hydrogen yield and ethanol conversion was selected as output variables. The ANN technology was then utilized for training the hidden layer (10 neurons) data using the Levenberg-Marquardt algorithm *via* artificial neural fitting. The error histogram results of this study varied from −1.595 to 1.338, which was close to zero, implying that model training was accomplished efficiently. Similar data was trained after 3, 10, and 20 iterations to increase the precision of data training. According to this study, the relative error decreases as the number of iterations increases. Thus, this proposed training was trustworthy, and the optimal number of iterations was 10, and relative errors ranging between −3 and 3% with a very high hydrogen yield (77%-86%) estimated by the predicted model. This study concluded that increasing reformation temperature improved hydrogen selectivity, which was consistent with earlier research.

Ebiad et al. [79] demonstrated that when the ESR temperature increased from 300 °C to 600 °C, H_2 selectivity over a Ni/CeO_2-ZrO_2 catalyst enhanced from 50.72% to 73.45%. Liu et al. [91] discovered that increasing the ESR temperature from 300 °C to 600 °C over a Ni (30%)Cu(5%)/Al_2O_3-MgO catalyst increased H_2 selectivity from 52.6% to 94%. Cursaru et al. [63] employed the ANN technology in the ESR

Table 5
Important results of MSR and ESR obtained by ANN method.

Parameters	Important finding	Reference
Catalyst type and content	The optimum weight percentage of Al and Ni in the catalyst was 42.49 and 12.35, respectively for the ESR reaction. The conversion of EtOH and H_2 selectivity for the optimal H_2 production was 79.6 and 91.4 mol %, respectively	[42]
	The co-precipitation method was the most effective way to produce a catalyst for the H_2 production	[42]
	Low Pt concentration (1.8 wt%) is not beneficial for WGSR.	[55]
	Pt (3 w/w%), Ni (10 w%), Co (10 w%), Cu (0 w %), Zn (0 w%), La (0 w%), Ce (7 w%), and Zr (0 w %) were the optimum combinations of different metal percentages in the catalyst for the ESR at 450 °C	[53]
	The ceria-supported catalysts with higher surface areas and Ni, Cu, and Ru metal catalysts were optimum parameters for the WGS reaction.	[88]
Reaction temperature	The maximum conversion of EtOH (nearly 85–90%) occurs at 500 °C, while relatively lower temperatures ranging from 300 °C to 400 ℃ may have a significant impact on the gradual increase of ESR.	[25]
	The higher reaction temperatures (300 °C) were favorable for the steam reforming of methanol.	[56]
	The temperatures (300 °C) were the optimum parameters for the WGS reaction	[98]
S/C ratio	A High EtOH concentration of 10–15 vol% was beneficial for the ESR reaction.	[25]
	The S/C ratio (3.5) was favorable for the MSR.	[56]
GHSV	The space velocities (2000–6000 h^{-1}) were the optimum parameters for the WGSR	[88]

reaction over a Co/MCM-48 catalyst to predict the highest ethanol conversion and hydrogen production. This investigation revealed that the maximum values for the intended outcome, such as ethanol conversion and hydrogen yield, were 85.31% and 61.77%, respectively. Szijjárto et al. [53] utilized the ANNs with holographic maps in the ESR reaction over various catalysts such as Pt, Ni, Co, Cu, Zn, La, Ce, Zr/$MgAl_2O_4$ to predict the highest EtOH conversion and H_2 production. The experimental data of hydrogen production (60.7%) was slightly greater than the predicted data from ANN (53.1%) at 450 °C. According to the experimental data, 75% ethanol was converted at 400 °C and 100% at nearly 600 °C over different non-noble metal-based catalysts [10]. In the ESR reaction, the noble metal-based catalyst converted ethanol more slowly.

ANN technology is beneficial for optimizing operating parameters and catalytic characteristics and estimating high H_2 yield and EtOH conversion. This methodology is particularly beneficial for the rapidity and adaptability of data training. Considering the preceding results, the ANN model can efficiently predict a reliable model. Thus, these simulation approaches are the simplest way to determine the operating conditions of high H_2 yield from methanol and ethanol reforming without generating excessive data from various experiments. ANN technology is the most cost-effective, rapid, systematic, and organized experimental design, with enormous potential to develop hydrogen energy from various fuel cells and may provide huge benefits not only for research purposes but also for industrial-level applications to produce green energy.

5. Future prospectus and challenges

The application of ANN technology to optimize diverse biofuels, such as methanol and ethanol, is limited [11,25,42,61,63,64]. More operational parameters, catalyst types, and research methods (models) must be optimized before concluding which factors benefit reform operations (Figs. 9-10). Thus, there could be adequate room for this computational study, which can be performed quickly and cost-effectively. Previous data from numerous publications might be utilized as input parameters. However, the ANN model may overfit if the input data is insufficient and is not obtained systematically from the experiments. As a result, it is necessary to collect sufficient systematic data before implementing an ANN model. One of the key advantages of the ANN model in methanol and ethanol reforming is its capacity to recognize the nonlinear relationship between dependent and independent components (Fig. 9). Although there is a significant amount of literature available, further study into ANN optimization technology is required for high hydrogen production and cost-effectiveness when compared to conventional reforming operations. Nowadays, the ANN model is used to optimize the process parameters of different fuel cells. For example, Jienkulsawad et al. [92] employed an ANN model to optimize the energy of a direct methanol fuel cell in a renewable power system. Rafe Biswas et al. [93] implemented the ANN model to predict the performance of a methanol fuel cell. Both advantages and disadvantages can be observed in implementing ANN technology to predict the data of methanol and ethanol conversion in their reforming reactions. The ability to recognize the nonlinear relationship between dependent and independent components is one of the key advantages of the ANN model for reforming the model. Compared to the other optimization methods, ANN technology is a fast and affordable process that may be employed to analyze a variety

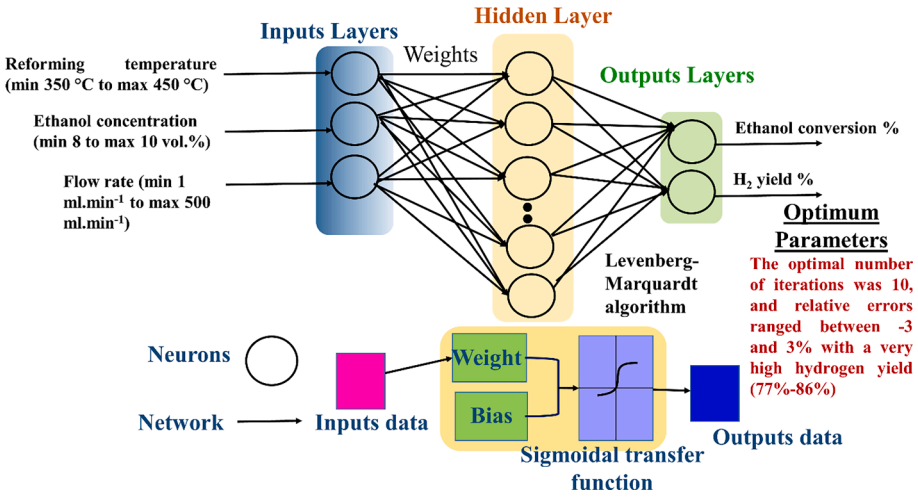

Fig. 8. The illustration of ANNs model application in the ESR reaction, where data are taken from Doicin et al. [61]. The optimal number of iterations was 10, and relative errors ranged between −3 and 3% with a very high yield of H_2 (77%-86%) estimated by the predicted model.

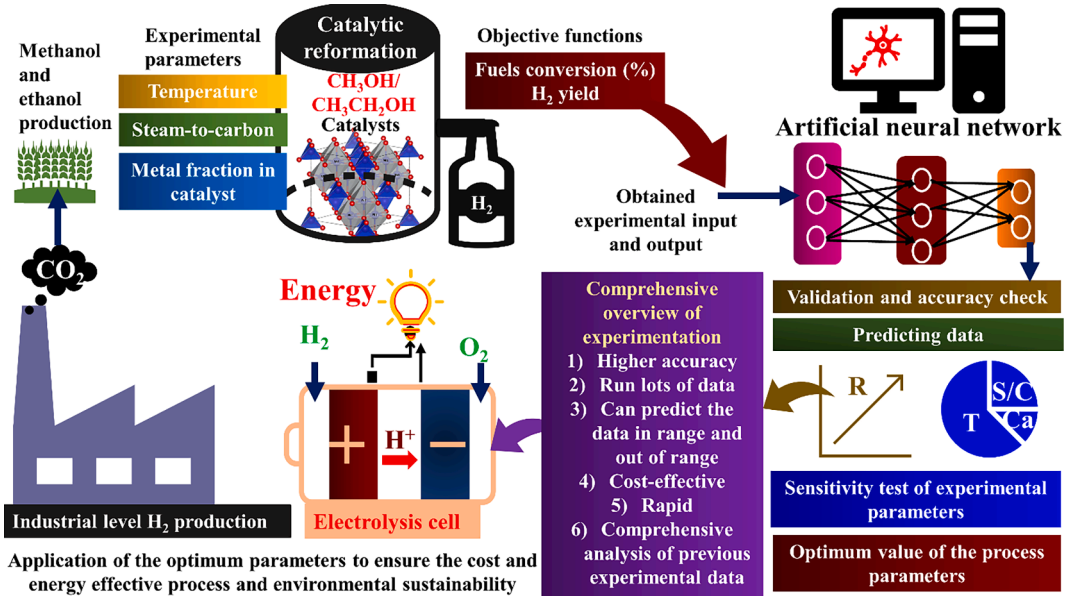

Fig. 9. The schematic diagram of the feasibility and implementation of ANNs technology in reforming various fuels, including methanol and ethanol. Identifying the nonlinear link between dependent and independent components is one of the ANN model's primary advantages in reforming the model. ANN technology is a simple and adaptable optimization model; for instance, depending on the experiments' goals, ANN technology requires less training and may employ various techniques. Massive amounts of information from diverse sources may be compared using ANN technology, which can comprehend the entire model and may benefit industrial-level applications. ANN technology is a quick and affordable technique. The sensitivity test of the ANN technology is used to evaluate the relative importance of different factors, which is useful for determining the values of different parameters for later implication in fuel cells, automobiles or in industries.

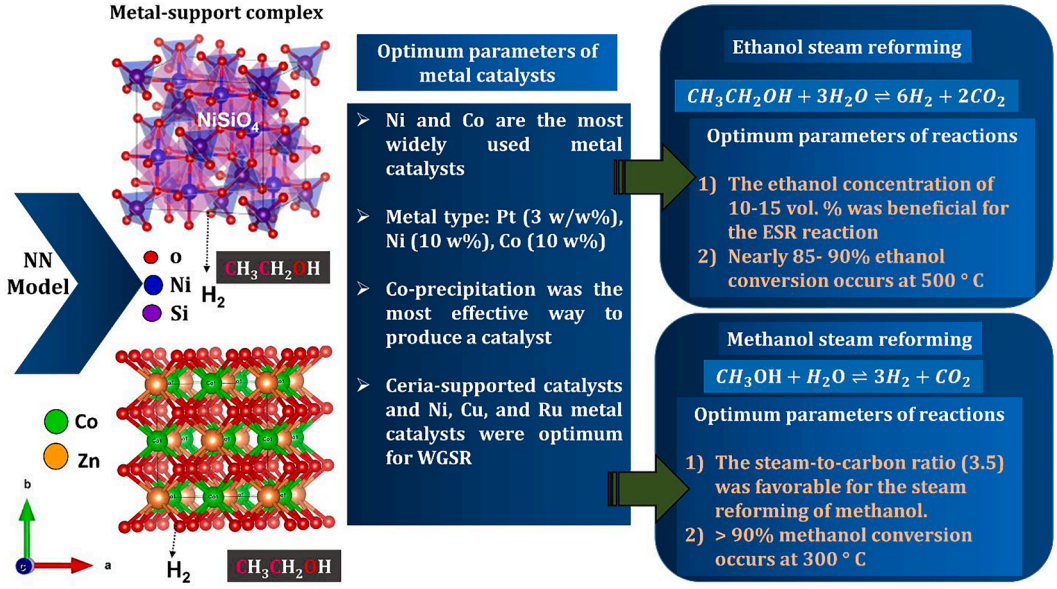

Fig. 10. Artificial neural networks (ANNs) model application to optimize operating parameters for catalytic thermochemical conversion of fuels (methanol and ethanol), in addition to the physical and chemical characteristics of various catalysts and their implications on operational processes and production of H_2.

of data obtained through experimentation or previous research [94]. The relative impact of various variables is assessed using the sensitivity test of the ANN technology, which is beneficial for figuring out the values of various parameters for subsequent studies [95]. Compared to other machine learning technologies, ANN technology is a simple and flexible optimization model; for example, less training is required for ANN technology, and different algorithms can be used for ANN technology depending on the purpose of the experiments [96]. ANN technology is useful for comparing large amounts of data from various literature, which can comprehend the entire model and may be useful for industrial-level applications. The primary disadvantage of an artificial neural network is its natural inclination to overfit [97]. There is a possibility that the predicted values won't match the experimental value if the data quality is insufficient and not good enough for both the input and output parameters. Several methodological issues need to be resolved in ANN technology since the neural network model is a numerical analysis constructed using empirical values. The ANN model needed computational resources and programming ability to train the problems and calculate the sensitivity tests.

6. Conclusions

An ANN optimization technology can efficiently and precisely predict the optimal parameters of MSR and ESR and the maximum hydrogen yield. Thus, without producing enormous data from several tests, these modeling methodologies are the most straightforward way to identify the operating parameters of high hydrogen output from MSR and ESR. According to the ANN model, the elimination of hydrogen reaction was mainly responsible for the 100 % methanol conversion observed at 300 °C. In contrast, the elimination of hydrogen and C-C bond-breaking reactions caused the ethanol conversion to increase with temperature and reach its maximum at 500 °C. The optimum temperature for the WGS reaction was 300 ℃. High H_2 yield could be predicted using the ANN model with minimal error. The breaking of the C-C bonds was assisted by steam or water. Therefore, a steam-to-carbon ratio of 3.5 benefited MSR, whereas a high ethanol concentration of 10–15 vol% was favorable for the ESR reaction. Ni and Co are the most often used metal catalysts for the ESR reaction due to their high efficiency for the steam reforming reaction. The optimal metal percentage combinations in the catalyst for ethanol reformation at 450 °C were Ni (10 w%) and Co (10 w%). Metal catalysts created by co-precipitation were the most effective at producing hydrogen because of their great dispersion, resulting in high catalyst activity. The maximum values for EtOH conversion and H_2 production for the targeted outcome were achieved when the stirring time for the catalyst production was 15 h, and the sintering temperature was 560 °C −570 °C due to the smallest crystallite size and significant dispersion of these metal catalysts. Future research might be streamlined by quantifying the optimal value for each process parameter. For instance, an ANN prediction model could suggest whether a parameter should be quantitatively increased or decreased throughout each experiment.

CRediT authorship contribution statement

Wei-Hsin Chen: Conceptualization, Funding acquisition, Project administration, Supervision, Writing – review & editing. **Partha Pratim Biswas:** Methodology, Validation, Visualization, Writing – original draft. **Aristotle T. Ubando:** Formal analysis, Writing – review & editing. **Eilhann E. Kwon:** Formal analysis, Writing – review & editing. **Kun-Yi Andrew Lin:** Formal analysis, Writing – review & editing. **Hwai Chyuan Ong:** Formal analysis, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The data that has been used is confidential.

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