Hybrid Artificial Neural Network-Genetic Algorithm Technique for Modeling and Optimization of Plasma Reactor

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Hybrid Artificial Neural Network—Genetic Algorithm Technique for Modeling and Optimization of Plasma Reactor

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A hybrid artificial neural network—genetic algorithm (ANN—GA) numerical technique was successfully developed to model, to simulate, and to optimize a dielectric barrier discharge (DBD) plasma reactor without catalyst and heating. Effects of CH₄/CO₂ feed ratio, total feed flow rate, and discharge voltage on the performance of noncatalytic DBD plasma reactor were studied by an ANN-based simulation with a good fitting. From the multiojective optimization, the Pareto optimal solutions and corresponding optimal process parameter ranges resulted for the noncatalytic DBD plasma reactor owing to the optimization of three cases, i.e., CH₄ conversion and C₂⁺ selectivity, CH₄ conversion and C₂⁺ yield, and CH₄ conversion and H₂ selectivity.

1. Introduction

It is very promising to utilize methane and carbon dioxide in natural gas as raw materials for the production of highly valuable chemicals and clean fuels. A major difficulty for such direct methane conversion is to activate the stable C—H bonds in methane molecules using conventional catalysis. Methane conversion in the presence of oxidants, such as oxygen, is thermodynamically favored for the production of CO₂. It is very important to improve the conventional catalysts and, at the same time, to exploit other potential techniques for methane conversion. Because of high-energy electrons of the plasma reactor, it is expected that methane and carbon dioxide gases could be activated easily in the plasma environment and converted into synthesis gases and higher hydrocarbons. It is expected that a nonconventional dielectric barrier discharge (DBD) plasma reactor is an efficient tool for converting CH₄ and CO₂, greenhouse gas contributors, to synthesis gas and higher hydrocarbons at low temperature and ambient pressure.¹—⁵

Numerical modeling and simulation on plasma reactors are essential for resolving the difficulties met in the measurements in order to understand the influences of modifications of the reactor configuration and material on the microdischarges.³ Until recently, only a few researchers focused on numerical studies dealing with DBD. Eliasson and Kogelschatz⁶ showed information on a single microdischarge combined with reaction chemistry in a single barrier reactor. Another study, by Kang et al.,⁷ presented a numerical study for understanding the influences of barrier arrangements on the evolution and characteristics of discharges in a DBD. The motions of electrons, positive ions, and negative ions in DBD were described by a set of continuity equations. The model includes the direct interactions of electrons and photons with neutral gas particles, such as electron impact ionization, electron attachment, and photon impact ionization. However, it is rare to find comprehensive plasma modeling in a DBD reactor in relation to optimization of process parameters, which is vital in industrial applications. Comprehensive plasma reactor modeling should take into account various points of view, such as chemistry, chemical reaction and kinetics, catalysis, and physics. Accordingly, the plasma model becomes very complex.

Artificial neural networks (ANNs) have been widely used in chemical engineering applications for complex process modeling, process control, and fault detection and diagnosis.⁸—¹¹ Meanwhile, the combination of ANN and genetic algorithm (GA) has been used for integrated process modeling and optimization.¹²—¹⁴ The hybrid ANN—GA technique is a powerful method for process modeling and optimization that is better than other techniques such as response surface methodology, particularly for complex process models. The phenomenologically comprehensive plasma reactor model needs a robust numerical solver and is time-consuming to solve, which is not suitable for rapid prediction in optimization and process control. Artificial neural network (ANN) modeling is chosen to model the complex behavior between input and output in the DBD plasma process, either with or without a catalyst. Therefore, the development of a robust hybrid ANN—GA algorithm is necessary to simulate the plasma reactor.

The present contribution is intended to develop an integrated algorithm of artificial neural network—genetic algorithm (ANN—GA) to facilitate modeling and optimization of a DBD plasma reactor. The integrated approach is intended to simplify the complex process model particularly in the DBD plasma reactor. Multiobjective optimization is implemented to obtain optimal operating parameters and maximum reactor performances.

2. Experimental and Numerical Methods

2.1. Apparatus of DBD Plasma Reactor. The experimental apparatus of a DBD plasma reactor is schematically depicted in Figure 1, while the configuration or design of the DBD plasma reactor is displayed in Figure 2. A high-voltage ac generator supplying a voltage from 0 to 17.5 kV with a pulsed waveform at a frequency of up to 10 kHz was used. The voltage measurement was conducted using an oscilloscope (ISO-TECH ISR 622) equipped with a high-voltage probe (manufactured by Atama Tech Sdn. Bhd.). Atama’s high-voltage probe was calibrated using a Tektronix P6015 high-voltage probe. The circuit for a high-voltage pulse ac generator as depicted in Figure 3 can be divided into two main sections: the oscillator and the power drive. The oscillator was built around a CMOS 4093 (4-nand gates) and was configured as a pulse generator (duty
cycle controlled). Either on or off time may be set by using the 150 kΩ potentiometer (high and low potentiometers). The main working frequency band can be set by proper selection of the 10 nF capacitor. The on/off time control gives very good regulation of the maximum output voltage. In this case, when the frequency was increased the voltage was decreased, and vice versa. The power drive was built around a power switching transistor (2N3055) which was driven by a 2N2222 transistor. The power transistor receives the command signal from the oscillator that opens and closes many times per second the primary coil of the transformer, thus inducing a high voltage in the transformer secondary coil. The power transistor needs a good heat sink because it performs heavy currents. To limit this high current, a current-limiting resistor (ballast resistor), one or two 10 Ω/10 W resistors, is applied.

The variables that may be involved in the operation of a DBD plasma reactor are discharge gap width, discharge power (voltage and frequency), reactant (CH₄/CO₂) ratio, and total feed flow rate. The DBD plasma reactor was operated without heating and catalyst. The reactor temperature may increase to about 60 °C due to electron heating on the surface of the electrode. During the process, the gas temperature can be within the range of room temperature, while the electrons can reach temperatures of 10⁴—10⁵ K in a dielectric barrier discharge. The nonthermal plasma
can be generated by applying a high voltage to a gas space and incurring gas breakdowns. The gas breakdowns generate electrons that are accelerated by an electric field forming a nonthermal plasma. The electrical discharges can be realized in several ways depending on the types of voltage applied and reactor specification. In the plasma reactor, the energetic electrons collide with molecules in the gas, resulting in excitation, ionization, electron multiplication, and the formation of atoms and metastable compounds. When the electric field in the discharge gap is high enough to cause breakdowns in most gases a large number of microdischarges are observed. The active atoms and metastable compounds subsequently collide with molecules, and reactions may occur.

2.2. Hybrid Artificial Neural Network–Genetic Algorithm (ANN–GA) Approach for Modeling and Optimization. In the past decade, artificial neural networks have emerged as an attractive tool for nonlinear modeling especially in situations where the development of phenomenological or conventional regression models becomes impractical. Genetic algorithm (GA) was originally developed as the genetic algorithm models duplicating population evolution in natural systems. Several researchers employed ANN–GA in their applications to get optimal operating conditions or parameters set up for the input variables in order to gain maximum performances or output variables such as in fluidized controlled catalytic cracking, in a chiller system, and in coal combustion. The objective of the process optimization is to maximize the process performances simultaneously such that the optimal process inputs are obtained. The integrated ANN–GA strategy meets the objective based on two steps: (a) development of an ANN-based process model where the model has inputs of process operating parameters and output(s) of process output/response variables, and (b) optimization of the input space of the ANN model by using the GA technique such that the optimal responses are obtained.

2.2.1. Artificial Neural Network (ANN) Based Modeling. In this research, the ANN algorithm was developed in a MATLAB environment by utilizing the Neural Network Toolbox to model and simulate the DBD plasma reactor. The feedforward ANN model was used in this model development and was trained using a back-propagation training function. The network training was performed using the Levenberg–Marquardt algorithm due to its fast convergence and reliability in locating the global minimum of the mean square error (MSE) (Levenberg–Marquardt). The back-propagation training procedure aims at obtaining an optimal set (W) of the network weight matrices W and Wb, which minimize an error function. The commonly employed error function is the mean square error (MSE). For the MSE minimization, several training iterations are usually necessary. The MSE minimization procedure by itself does not ensure that the trained network would possess the much desired generalization capability. To ensure that the feedforward neural network model possesses satisfactory generalization capability, special treatments must be investigated to avoid the network overfitting, as follows:

(a) Partition the available example input–output data into two sets, namely training and test sets.

(b) Fix the number of nodes in multilayer perceptron (MLP)'s hidden layer to a small number (1 or 2).

(c) Initialize the network weight set, W, randomly.

(d) Train the network over several iterations using input–output vectors in the training set such that the MSE with respect to the test (e_test) set is minimized. Store the weight matrix that has resulted in the lowest e_test magnitude.

(e) Repeat steps c and d many times by changing the seed value of the random number. This ensures that the weights are initialized differently in each repeated training run.

(f) Go to step b and repeat steps c and d by systematically incrementing the number of hidden nodes (S). The network architecture and the weight set, W, that result in the overall least e_test magnitude are taken as optimal.

2.2.2. Genetic Algorithm (GA) Based Optimization. In general, the genetic algorithm (GA) is a method for solving optimization problems based on the principle of survival of the fittest during evolution. The GA is one of the strategic randomized search techniques, and is well-known for its robustness in finding the optimal or near-optimal solution since it does not depend on gradient information in its walk of life to find the best solution. Various algorithms were reported by previous researchers.

In this research, the MATLAB environment was used for developing computer codes of genetic algorithm optimization by utilizing the Genetic Algorithm Toolbox. The important thing is on how to combine the ANN and the GA algorithms in a hybrid system that supports modeling and optimization simultaneously. The principal features of the GA are as follows: (a) it requires only scalar values and not the second- and/or first-order derivatives of the objective functions, (b) it is capable of handling nonlinear and noisy objective functions, (c) it performs global searches and thus is more likely to arrive at or near the global optimum, and (d) it does not impose preconditions, such as smoothness, differentiability, and continuity, on the form of the objective function. Because of these attractive features, the GA is useful in solving the diverse optimization problems in chemical engineering.

2.2.3. Hybrid ANN–GA Algorithm. Having developed an ANN-based DBD plasma reactor model, a GA is used to optimize the R-dimensional input space (x) of the ANN model. The corresponding optimization objective can be defined as finding the R-dimensional optimal decision factor, x* = [x1*, x2*, ..., xn*]T, such that it maximizes the K-dimensional objective function vector f, which is defined as

\[ f(x*, W, b) = \sum_{k=1}^{K} f_k(x*, W, b) \]  (1)

where W = {W, W} and b = {b, b} are weight and bias parameters, respectively, for hidden and output layers of the network. It can be remarked in the above formulation that the problem belongs to multiobjectives optimization, which is simultaneously maximization of \( f(x*, W, b) \), where \( k \) represents the number of objective functions. A simple approach to solve the multiobjectives optimization problem is by aggregating the objectives into a single scalar objective function, namely the weighted sum of squared objective function (WSSOF) algorithm. In the WSSOF method, each objective function is multiplied by a weighting coefficient (\( w_k \)) and combined to form a single scalar objective function to be maximized. The maximization of the single objective function can be expressed as

\[ \max f(x, W) = \sum_{k=1}^{K} w_k f_k(x, W) \]  (2)

where \( 0 \leq w_k \leq 1 \) and \( \sum_{k=1}^{K} w_k = 1 \).

The detailed stepwise procedure for the hybrid ANN–GA algorithm for modeling and optimization is described below and is depicted schematically in Figure 4.
Step 1 (Development of an ANN-Based Model). Specify input and output experimental data of the system used for training and testing the ANN-based model. Create the network architecture involving input, hidden, and output layers. Investigate the optimal network architecture (optimal number of hidden layers) and make sure that the network is not overfitted.

Step 2 (Training of the ANN-Based Model). Normalize the experimental input and output data to be within the range \([-1 1]\). Normalize the data to be within the range \([-1 1]\). Simulate normalized \(x_i\) to the ANN-based model to compute \(K\)-dimensional output vector \(y_j\), where \(y_j = f(x_i, W, b)\). Re-transform the output vector \(y_j\) to the original values. Score/evaluate each member of the current population by computing its fitness of the \(j\)th individual using \(y\).

Step 3 (Initialization of Solution Population). Set the initial generation index \(Gen = 0\) and number of population \(N_{pop}\) and number of independent variables, \(nvars\). Create a random initial population of \(N_{pop}\) individuals.

Step 5 (Scaling the Fitness Scores). Scale/rank the raw fitness scores to values in a range that is suitable for the selection function. In the GA, the selection function uses the scaled fitness values to choose the parents for the next generation. The range of the scaled values influences the performance of the genetic algorithm. If the scaled values vary too widely, the individuals with the highest scaled values reproduce too rapidly, taking over the population gene pool too quickly and preventing the genetic algorithm from searching other areas of the solution space. On the other hand, if the scaled values vary only a little, all individuals have approximately the same chance of reproduction and the search will progress slowly. The scaling function used in this algorithm scales the raw scores based on the rank of each individual instead of its score. Because the algorithm minimizes the fitness function, lower raw scores have higher scaled values.

Step 6 (Parents Selection). Choose the parents based on their scaled values by utilizing the selection function. The selection function assigns a higher probability of selection to individuals with higher scaled values. An individual can be selected more than once as a parent.

Step 7 (Reproduction of Children). Reproduction options determine how the genetic algorithm creates children for the next generation from the parents. Elite count \(E_{child}\) specifies the number of individuals with the best fitness values that are guaranteed to survive to the next generation. Set elite count to be a positive integer within the range \(1 \leq E_{child} \leq N_{pop}\). These individuals are called elite children. Crossover fraction \(P_{cross}\) specifies the fraction of each population, other than elite children, that are produced by crossover. The remaining individuals in the next generation are produced by mutation. Set crossover fraction to be a fraction between 0 and 1.

Crossover. Crossover enables the algorithm to extract the best genes from different individuals by selecting genes from a pair of individuals in the current generation and recombining them into potentially superior children for the next generation with the probability equal to crossover fraction \(P_{cross}\) from step 7.

Mutation. Mutation function makes small random changes in the individuals, which provide genetic diversity and thereby increases the likelihood that the algorithm will generate individuals with better fitness values.

Step 8 (Replaces the Current Population with the Children). After the reproduction is performed and the new children are obtained, the current population is replaced with the children to form the next generation.

Step 9 (Update/Increment the Generation Index). Increment the generation index by 1: \(Gen = Gen + 1\).

Step 10 (Repeat Steps 4–9 until Convergence Is Achieved). Repeat steps 4–9 in the new generation until convergence is achieved. The algorithm stops if any one of the following five conditions is met. The genetic algorithm uses the following five criteria to determine when the algorithm stops:

(a) Generations: The algorithm stops when the number of generations reaches the maximum value \(Gen_{max}\).

(b) Fitness limit: The algorithm stops when the value of the fitness function for the best point in the current population is less than or equal to Fitness limit.

(c) Time limit: The algorithm stops after running for an amount of time in seconds equal to Time limit.

(d) Stall generations: The algorithm stops if there is no improvement in the objective function for a sequence of consecutive generations of length Stall generations.

(e) Stall time limit: The algorithm stops if there is no improvement in the objective function during an interval of time in seconds equal to Stall time limit.
Step 11 (Assign the Top Ranking of Children to the Optimal Solution Vector). After the GA convergence criterion is achieved, the children possessing the top ranking of the fitness value are assigned to the optimized population or decision variable vector, \( \mathbf{x}^* \).

2.3. Design of Experiment for Collecting the Training Data. The ANN-based model requires more example data which are noise-free and statistically well-distributed. However, the ANN-based model is not good at extrapolation. This weakness can be solved by collecting more example data in the regions where extrapolation is required. The design of the experiment was performed using central composite design (CCD) with full factorial design for designing the training and test data sets.

The CCD method is chosen since the method provides a wider covering region of parameter space and good consideration of variable interactions in the model. The CCD considers low, center, and high levels of each independent variable. Another advantage of central composite design is to reduce the number of experimental works, but the experimental design includes a wider region of parameter space. The variable range and levels of the CCD are presented in Table 1. It is well-known as a limitation of the ANN model that the model is not good at extrapolation. The limitation can be solved by collecting more example data in the regions where the extrapolation is required. This is the reason that the central composite design with full factorial design was used; it can cover a wider range and interactions of variables.

3. Results and Discussion

3.1. Artificial Neural Network Model Development for MIMO System. The multi-input and multi-output (MIMO) system with three inputs and four outputs of an artificial neural network model was developed for CH\(_4\) and CO\(_2\) conversion in a noncatalytic DBD plasma reactor without heating. The development is intended to obtain the optimum topology for the MIMO system. In this network model, three independent variables, i.e., CH\(_4\)/CO\(_2\) feed ratio \((X_1)\), discharge voltage \((X_2)\), and total feed flow rate \((X_3)\), are used as inputs of the MIMO network, while four dependent variables or objectives, i.e., CH\(_4\) conversion \((Y_1)\), C\(_2\)\(_\text{gas}\) selectivity \((Y_2)\), H\(_2\) selectivity \((Y_3)\), and C\(_2\)\(_\text{gas}\) yield \((Y_4)\), are selected as outputs of the MIMO network. The product gases include H\(_2\), CO, and C\(_2\)\(_\text{gas}\); hydrocarbons (C\(_2\)H\(_6\), C\(_2\)H\(_4\), C\(_3\)H\(_2\), C\(_4\)H\(_4\)). Prior to the network training, the 19 numbers of experimental data as presented in Table 3 are partitioned into training (15 patterns) and test (4 patterns) sets, where the test set patterns are marked separately in the table. The data are obtained based on the experimental design as revealed in Table 2. In each network training iteration, the training set was utilized to adjust the weight matrix set, \( \mathbf{W} \), and the test set was used for gauging the network’s generalization performance. The additional data for the noncatalytic DBD plasma reactor without heating are presented in Table 4 at extended conditions and with their comparison with a feed of pure CH\(_4\).

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Table 1. Central Composite Design with Full Factorial Design for the Noncatalytic DBD Plasma Reactor

<table>
<thead>
<tr>
<th>factors</th>
<th>range and levels</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH(_4)/CO(_2) ratio, (X_1)</td>
<td>(-\alpha) 0.3  1 2 3  3.7  (+\alpha)</td>
</tr>
<tr>
<td>discharge voltage, (X_2) (kV)</td>
<td>12.6 13.4 14.6 15.8 16.6</td>
</tr>
<tr>
<td>total feed flow rate, (X_3) (cm(^3)/min)</td>
<td>26.4 40 60 80 93.6</td>
</tr>
</tbody>
</table>

\(^a\)

Table 2. Experimental Design Matrix of the Noncatalytic DBD Plasma Reactor for Training Data

<table>
<thead>
<tr>
<th>(X_1)</th>
<th>(X_2)</th>
<th>(X_3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.0</td>
<td>13.4</td>
<td>40.0</td>
</tr>
<tr>
<td>3.0</td>
<td>13.4</td>
<td>80.0</td>
</tr>
<tr>
<td>3.0</td>
<td>15.8</td>
<td>40.0</td>
</tr>
<tr>
<td>3.0</td>
<td>15.8</td>
<td>80.0</td>
</tr>
<tr>
<td>1.0</td>
<td>13.4</td>
<td>40.0</td>
</tr>
<tr>
<td>1.0</td>
<td>13.4</td>
<td>80.0</td>
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<tr>
<td>1.0</td>
<td>15.8</td>
<td>40.0</td>
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<tr>
<td>1.0</td>
<td>15.8</td>
<td>80.0</td>
</tr>
<tr>
<td>0.3</td>
<td>14.6</td>
<td>60.0</td>
</tr>
<tr>
<td>0.3</td>
<td>14.6</td>
<td>60.0</td>
</tr>
<tr>
<td>2.0</td>
<td>12.6</td>
<td>60.0</td>
</tr>
<tr>
<td>2.0</td>
<td>16.6</td>
<td>60.0</td>
</tr>
<tr>
<td>2.0</td>
<td>14.6</td>
<td>26.4</td>
</tr>
<tr>
<td>2.0</td>
<td>14.6</td>
<td>93.6</td>
</tr>
<tr>
<td>2.0</td>
<td>14.6</td>
<td>60.0</td>
</tr>
<tr>
<td>2.0</td>
<td>14.6</td>
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<td>2.0</td>
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<tr>
<td>2.0</td>
<td>14.6</td>
<td>60.0</td>
</tr>
</tbody>
</table>

\(^b\)

\(^b\)

\(^c\)

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The performance of the ANN model is considered as fitness tests of the model, i.e., mean square error (MSE), correlation coefficient \((R)\), and epoch number (epochs). The comparison of the ANN model performance for various topologies is presented in Table 5 for one hidden layer network, while the comparison of a two hidden layers structure is demonstrated in Table 6. In Table 5, the network structure of 3–11–4 (for example) consists of 3 inputs, 11 hidden layers, and 4 outputs, while the network of 3–9–11–4 contains 3 inputs, 9 first hidden layers, 11 second hidden layers, and 4 outputs.

From Table 5, it can be shown that increasing the number of nodes in the hidden layer leads to decreasing the mean square error (MSE) and increasing the correlation coefficient \((R)\). The epoch number also decreases with increasing the number of nodes in the hidden layer. From Table 5, the 3–11–4 ANN structure shows a potential topology of the MIMO network for the DBD plasma reactor without catalyst. Table 6 demonstrates the comparison of a two hidden layers system, and the results shows that the MSE and the correlation coefficient of the two hidden layers system are better than those of the one hidden
layer system with a smaller epoch number. From the comparison, the 3–9–11–4 structure ANN model is chosen for the next simulation. Pertaining to the best ANN model (3–9–11–4 structure), the comparison of the network-predicted and observed values of the four responses (CH4 conversion (Y1), C2+ selectivity (Y2), H2 selectivity (Y3), and C2+ yield (Y4)) are depicted in Figure 5.

3.2. Multiresponses Optimization of DBD Plasma Reactor without Catalyst Using Hybrid ANN–GA Strategy. In this system, there are four responses/objectives to be maximized, i.e., CH4 conversion, C2+ selectivity, C2+ yield, and H2 selectivity. The multiojectives optimization is intended to obtain the recommended optimal operating parameters with simultaneous maximum performances. The simultaneous maximization is intended to come close to the real conditions of the process, but the higher number of objectives to be optimized simultaneously, the more difficult the optimization process is. To simplify the optimization problem, the multiobjectives optimization is intended to come close to the real conditions of the simultaneous maximum performances. The simultaneous maximization of the real process performances is intended for simplicity. For the multiresponses optimization, the decision variables/operating parameters bounds are chosen from the corresponding bounds in the training data as listed in Table 7. The Pareto optimal solutions are obtained from multiobjectives optimization using the genetic algorithm (GA) tool. Table 8 lists the numerical parameter values used in the GA for all optimization runs. In this optimization, the rank method is used for fitness scaling, while stochastic uniform is used for the selection method to specify how the genetic algorithm chooses parents for the next generation. From the 50 numbers of population size, two of them are elite and used in the next generation, while 80% of the rest of the population is used for crossover reproduction and 20% is used for mutation reproduction.

Because of the minimization feature in the default function of the genetic algorithm, the objective functions are converted according to eq 3 in the case of maximizing the two objective functions (F1 and F2) simultaneously. The equation is based on the popular approaches for inversion.27,28 The equation was implemented for manipulating the maximization problem by converting to a minimization problem for solving cases 1, 2, and 3:

$$F_i = \frac{1}{1 + F_{i,o}}$$  \hspace{1cm} (3)

where $F_{i,o}$ denotes the real objective functions, while $F_i$ are the inverted objective functions for the minimization problem.

Pertaining to case 1, the optimal operating parameters of the DBD plasma reactor for the CH4–CO2 conversion process are determined corresponding to simultaneous maximization of CH4 conversion and C2+ hydrocarbons selectivity. The Pareto figure owing to the solution of case 1 optimization and the corresponding variables are shown in Figures 6 and 7, respectively. The Pareto optimal solution points are obtained by varying the weighting factor ($w_j$) in eq 2 and performing the GA optimization corresponding to each $w_j$ variation. From Figure 6, it is found that if the CH4 conversion improves, the C2+ selectivity deteriorates, and vice versa. Theoretically, all sets of noninferior/Pareto optimal solutions are acceptable. From Figure 7, the three operating parameters show the key role in determining the optimal solutions. CH4 conversion as high as 20.3% is achieved.
with $C_2$ selectivity equal to 27.5%, while $C_2$ selectivity as high as 17.7% can be obtained with CH$_4$ conversion equal to 33.3%. This could be achieved by altering the CH$_4$/CO$_2$ feed ratio ($X_1$), discharge voltage ($X_2$), and total feed flow rate ($X_3$) from 3.4, 16.4 kV, and 32.6 cm$^3$/min to 1.9, 16.5 kV, and 32.2 cm$^3$/min, respectively. It is obvious in the Pareto optimal solutions (Figures 6 and 7) that the CH$_4$ conversion improves at lower CH$_4$/CO$_2$ ratio, since high CO$_2$ concentration in the feed enhances the decomposition of methane. Increasing the CO$_2$ concentration in the feed enhances the CH$_4$ conversion due to promoting the CH$_4$ conversion by oxygen from CO$_2$ decomposition. From Figure 7, a larger CH$_4$ amount in the feed and higher feed flow rate enhance the $C_2$ hydrocarbons selectivity, which is corroborated by the results of Eliasson et al.$^{29}$ and Liu et al.$^{30}$ However, increasing the total feed flow rate reduces the CH$_4$ conversion due to decrement of contact time between highly energetic electrons and gas molecules.

From the Pareto optimal figure, suitable optimal process parameter ranges for the noncatalytic DBD plasma reactor owing to simultaneous maximization of CH$_4$ conversion and $C_2$ yield can be recommended. Here, the ranges of CH$_4$/CO$_2$ feed ratio ($X_1$), discharge voltage ($X_2$), and total feed flow rate ($X_3$) of 1.9–3.4, 16.4–16.5 kV, and 32.2–32.6 cm$^3$/min, respectively, are recommended to achieve CH$_4$ conversion of 20.3%–33.3% and $C_2$ hydrocarbons selectivity of 17.7%–27.5%.

In the solving of case 2 optimization, the Pareto optimal solutions of the simultaneous maximization of CH$_4$ conversion and $C_2$ yield are shown in Figure 8. Figure 9 demonstrates the corresponding operating parameters with respect to the Pareto optimal solutions. The different trend is obvious in Figure 8, where the noninferior solution points distribute entirely within the range of the objectives. It is exhibited that the optimal CH$_4$ yield is distributed narrowly about 6.80%–6.84%, while the corresponding optimal CH$_4$ conversion is within the range of 23.5%–24.5%. This fact is also confirmed by the operating parameter effects where the $C_2$ yield changes slightly at the corresponding operating parameters variation. In particular, if the CH$_4$ conversion rises the $C_2$ yield shows a decrement, and vice versa. From the Pareto optimal solutions, the suitable operating condition ranges for the noncatalytic DBD plasma reactor owing to simultaneous maximization of CH$_4$ conversion and $C_2$ yield can be recommended based on Figures 8 and 9. In this case, the optimal ranges of CH$_4$/CO$_2$ feed ratio ($X_1$), discharge voltage ($X_2$), and total feed flow rate ($X_3$) are 3.0–3.5, 16.3–16.5 kV, and 20–24 cm$^3$/min, respectively, while the simultaneous maximum CH$_4$ conversion and $C_2$ hydrocarbons yield are 23.5%–24.5% and 6.80%–6.84%, respectively.

With respect to case 3 for simultaneous maximization of CH$_4$ conversion and H$_2$ selectivity, the Pareto optimal solution and the corresponding optimal operating parameters for this case are presented in Figures 10 and 11, respectively. From Figure 10, it is found that the Pareto optimal solution shows a noninferior feature where CH$_4$ conversion increases with decreasing H$_2$ selectivity, and vice versa. From the figure, CH$_4$ conversion as high as 25.5% is achieved with H$_2$ selectivity equal to 7.5%, while the H$_2$ selectivity as high as 15.5% can be obtained with CH$_4$ conversion equal to 11.6%. It is obvious
from Figure 11 that altering three operating parameters has a small influence on CH$_4$ conversion and H$_2$ selectivity. From Figure 11, the optimal operating parameters for simultaneous optimization of CH$_4$ conversion and H$_2$ selectivity can be recommended in the ranges of CH$_4$/CO$_2$ feed ratio ($X_1$), discharge voltage ($X_2$), and total feed flow rate ($X_3$) of 2.3–2.8, 13.0–14.5 kV, and 20–22 cm$^3$/min, respectively. The recommended process parameters are intended to achieve CH$_4$ conversion of 11.6%–25.5% and H$_2$ selectivity of 7.5%–15.5%.

4. Conclusions

A hybrid artificial neural network–genetic algorithm (ANN–GA) was successfully developed to model, simulate, and to optimize the noncatalytic DBD plasma reactor. A study on the effects of CH$_4$/CO$_2$ feed ratio, total feed flow rate, and discharge voltage on the performance of noncatalytic DBD plasma reactor without heating was successfully addressed by ANN-based model simulation with a good fitting. It can be concluded that three factors, i.e., CH$_4$/CO$_2$ feed ratio, total feed flow rate, and discharge voltage, in the noncatalytic DBD plasma reactor systems showed significant effects on the reactor performances. From the multiobjectives optimization by the hybrid ANN–GA technique, the Pareto optimal solutions and corresponding optimal process parameters ranges can be suggested which are suitable for the noncatalytic DBD plasma reactor owing to simultaneous maximization of CH$_4$ conversion and C$_2$H$_4$ selectivity, CH$_3$ conversion and C$_2$H$_4$ yield, or CH$_4$ conversion and H$_2$ selectivity.

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