Experimental analysis and parameter optimization on the reduction of NOx from diesel engine using RSM and ANN Model

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Abstract
The major emission sources of NOx are from automobiles, trucks, and various non-road vehicles, power plants, coal fired boilers, cement kilns, turbines, etc. Plasma reactor technology is widely used in gas conversion applications, such as NOx conversion into useful chemical by-product. Among the plasma treatment techniques, nonthermal plasma (NTP) is widely used because it does not cause any damage to the surfaces of the reacting chamber. In this proposed work, the feasibility of Dielectric Barrier Discharge (DBD) reactor–based nonthermal plasma (NTP) process is examined based on four operating parameters including NOx concentration (300–400 ppm), gas flow rate (2–6 lpm), applied plasma voltage (20–30 kVpp), and electrode gap (3–5 mm) for removing NOx gas from diesel engine exhaust. Optimization of NTP process parameters has been carried out using response surface-based Box-Behnken design (BBD) method and artificial neural network (ANN) method and compared with the performance measures such as $R^2$, $MSE$ (mean square error), $RMSE$ (root mean square error), and $MAPE$ (mean absolute percentage error). Two kinds of analysis were carried out based on (1) NOx removal efficiency and (2) energy efficiency. Based on the simulation studies carried out for Nox removal efficiency, the RSM methodology produces the performance measures, 0.98 for $R^2$, 1.274 for $MSE$, 1.128 for $RMSE$, and 2.053 for $MAPE$, and for ANN analysis method, 0.99 for $R^2$, 2.167 for $MSE$, 1.472 for $RMSE$, and 1.276 for $MAPE$. These results shows that ANN method is having enhanced performance measures. For the second case, based on the energy efficiency study, the $R^2$, $MSE$, $RMSE$, and $MAPE$ values from the RSM model are 0.97, 2.230, 1.493, and 2.903 respectively. Similarly based on ANN model, the $R^2$, $MSE$, $RMSE$, and $MAPE$ values are 0.99, 0.246, 0.46, and 0.615, respectively. From the performance measures, it is found that the ANN model is accurate than the RSM model in predicting the NOx removal/reduction and efficiency. These models demonstrate that they have strong agreement with the experimental results. The experimental results are indicated that optimum conditions arrived based on the RSM model resulted in a maximum NOx reduction of 60.5% and an energy efficiency of 66.24 g/J. The comparison between the two models confirmed the findings, whereas this ANN model displayed a stronger correlation to the experimental evidence.

Keywords Air pollution control · Artificial neural network (ANN) · Dielectric barrier discharge (DBD) · NOx reduction · Nonthermal plasma (NTP) · Response surface method (RSM)

Introduction
Since the second half of the twentieth century, global warming has gained the status of a major concern due to a rise in the earth’s temperature. In one third of the USA, pollution has greatly increased, allowing gases to enter the atmosphere, and due to this, global warming has accelerated at a much greater rate. Around 40% of NOx emissions are due to vehicle use of the road. Emissions of gases such as sulfur oxides, nitrogen oxides, and carbon monoxide contribute to toxicity, which has both human
and environmental consequences (Chen & Liu 2010, E et al. 2020, Kampa & Castanas 2008). The release of NOx among these gases has a significant impact on environmental degradation. The cause of NOx pollution is automotive exhausts, exhaust gases generated in industries like turbines, power plants, and cement kilns (Amoatey et al. 2019; Sohn et al. 2021). When combustion occurs at high temperature, NOx is likely to be formed. Oxidation of nitric oxide (NO) results in the production of a tropospheric greenhouse gas with a strong odor (E et al. 2020, Zhao et al. 2021). These exhaust sources, while feasible in theory, are still challenging to get rid of the NOx entirely (Maheswari 2014, Li et al. 2011). There is a need for an efficient method for reducing the amount of NOx emitted by these sources.

The Indian government strongly demands that diesel generator manufacturers adhere to the permissible NOx and hydrocarbon emission limits in the atmosphere. With respect to commercial vehicles, India’s National Emission Standards-EU has proposed a drop from 0.39 g of hydrocarbon emission limits per kilometer in 2017 to 0.20 g of NOx limits in 2020 as illustrated in Fig. 1a. There are over a million vehicles sold in the commercial vehicle sector for the first time in the fiscal year, an all-time high level of production for Indian trucks and buses in 2019. The most rapid growth in the year is for heavy trucks. As shown in Fig. 1b, according to the Central Pollution Control Board (CPCB), 3.5 g/kmHr of NOx is emitted into the atmosphere in 2020 as a result of heavy diesel vehicles (Source CPCB, 2020).

The use of emerging technology and improvements to existing techniques have become needed to confront the constraints on NOx pollution. Selective of non-catalytic reduction (SCR) is commonly used nowadays, especially for reducing NOx emission during coal combustion. Several other methods, such as mechanical scrubbing, adsorption, absorption, electron beam, electrochemical cell, or vapor diffusion, are often used (Arun kumar 2019, C Maheswari 2015, Maheswari 2013, Skalska et al. 2010). For example, Kinoshita et al. (2022) reported, when using glass fiber plastics as an adsorbent, a maximum of 52% of NOx removal was achieved. Feng et al. (2022) investigated the viability of hydrocarbon adsorber and converter to control the hydro carbon emission; the result shows that it reaches the removal efficiency of 50% within 102 s. Nonetheless, each of these techniques has its limits and drawbacks. Additionally, certain developing nations have placed highly stringent limits on NOx emissions (Maheswari et al. 2017). As a result, research is being conducted worldwide to develop more reliable approaches. Plasma-assisted deletion is an important and superior suggested approach for cleaner air among NOx pollution control techniques (Tang et al. 2021). The removal of nitrogen oxides using nonthermal plasma (NTP) has been put forward as a viable option (Takaki et al. 2004). In the NTP systems, toxins are easy to remove, there are no organic contaminants and maintenance costs, and primary processing is at ambient pressure (Suresh et al. 2021a, 2021b). Jolibois et al. (2012) explored the maximum of 55% of removal efficiency was achieved when applying the surface-based plasma discharge in wet conditions.

In view of the benefits of NTP, even though the treatment efficiency of NOx reduction is substantial, the key difficulty of using this technology is energy consumption. Many parameters, such as initial concentration of NOx, gas flow rates, treatment time, duty cycle, electrode gap, electrode geometry, and applied voltage, will affect purification performance and energy rate of plasma system (Kuwahara et al. 2016; Mizuno 2007, 2013; Takaki et al. 2004). In the present study, the dielectric barrier discharge (DBD)–based NTP reactor is developed to reduce the NOx which will clearly provide a homogeneous, low-energy top discharge with higher efficiency when placing dielectric material between the high-voltage and ground electrodes (Ansari et al. 2020, Khan & Kim 2020). NTP tends to be more ideal for the removal of NOx due to its ability to efficiently convert nitrogen to OH radicals and the ease of installation, increasing the risk of the formation of free radicals, such as hydroxyl radical (OH) (Krosuri et al. 2021). Electrons with energy ranges from 1 to 10 eV are emitted during plasma process and exposed to nitrogen in gases to process nitric oxide to nitrogen dioxide (Tang et al. 2021). Zhu et al. (2020) found an alternative approach that used the DBD. Only ammonia
is used to make DBD radicals, which are combined with the flue gases to start a reaction. The removal efficiencies ranged between 93.89% and energy density of 500 J/lit.

Exhaustive simulation and process optimization efforts have been devoted to understanding the system behavior and finding the global optimal for chemical processes so far, and some have used the statistical simulation approach like mathematical modeling, response surface methodology (RSM), and artificial neural network (ANN) (Bhatti et al. 2011; Zhao et al. 2020). Each approach has its own set of benefits and drawbacks. Mathematical modeling, for example, involves a system comprehension that includes functions that apply to the system. It can contain calculations, equations, constitutive equations, and restrictions. The consistency of the model works relatively well when the dependent and independent variables maintain a constant relationship but decreases dramatically when the equation exhibits nonlinear behavior (Chen & Tan 2012). Other tools, such as RSMs or ANNs, have been used with nonlinear systems to see how input variables impact the behavior of the system (Zhao et al. 2020). The DOE research will investigate all the independent variables within the bounds of each variable and integrate the data. Out of all techniques, in specific, RSM has actually been used in numerous technical fields.

Complex activities like learning and pattern recognition have given rise to a vast quantity of computational investigation into small networks of basic components termed ANNs. In the last few years, scientists in a number of scientific fields have taken an interest in the possible mathematical usefulness of artificial intelligence techniques to tackle a number of issues. ANNs are designed to help understand the roles of the brain in mammalian. Additionally, ANNs are used to render an approximation of hidden feature. It’s quite powerful for managing very subjective data, particularly in those areas where analysis is essential. Other methods of neural network creation are in progress, in addition to ANN. To name a few, feed-forward networks and real-time control systems are adopted, and an adaptive neural network is applied to systems that needed it (Agatonovic-Kustrin & Beresford 2000, Picos-Benítez et al. 2017; Zhao et al. 2020).

Research on DBD reactors based on NTP in the treatment of NOx is rarely found. Similarly, there are just a few publications on DBD reactor optimization utilizing response surface technique and ANN. Therefore, a test to explore the feasibility of using DBD plasma technology with minimizing the emissions of NOx and optimizing the process parameters utilizing response surface approach is being carried out. In this way, new treatment options for NOx pollution may be opened up, and continued work in this area may help overcome the numerous obstacles that stand in the way of identifying and applying the appropriate treatment plan. The purpose of this work is to develop a DBD reactor with a larger efficiency for eliminating NOx. To enhance the reactor efficiency of removing NOx, four parameters are analyzed via BBD design: the initial NOx concentration, gas flow rate, electrode gap, and voltage. Additionally, the energy efficiency of the DBD reactor is evaluated. The artificial neural network (ANN) uses both inter-variable interactions as well as both interactive factors in predicting and simulating interrelationships. This is used to determine the predictor importance of the resulting model.

**Experimental methodology**

**Experimental setup**

The schematic experimental setup for NOx reduction is illustrated in Fig. 2, and its specifications are mentioned in the previous study (Suresh et al. 2021b). This design is comprised of four main elements: gas supply, DBD reactor, power supply module, and gas-sensing apparatus. A graphical representation of the DBD reactor is shown in Fig. 2. Inner electrode has the dielectric shielding placed inside the reactor. The outer electrode is constructed from copper wires that is fixed to the dielectric layer. Stainless steel is used as the grounding electrode. Due to the existence of large electrical foils (centroids) in the middle of the dielectric tubes, flashover is avoided by using foil sides that is lower in voltage. In the reactor, glass is used as the dielectric barrier. Surface thickness of all components is 2 mm, as is shown as a strong dielectric tube in Fig. 2. The reactor is attached to a high-voltage supply to produce plasma that should be with peak intensity. Fluidised gases are introduced by a nozzle into the reactor. The specific quantity of NOx and oxygen are mixed thoroughly and entered into the DBD reactor. To measure the inlet and outlet NOx concentrations, the NOx sensor with transmitters is fixed. The flue gas from the diesel engine containing nitrogen oxide (NOx) is mixed with 10% oxygen (O2), and then it is injected to the reactor. By using air flow regulator, the mixture gas flow rate is regulated. Based on the NOx level in the untreated air, the NOx sensor is used to determine the NOx level. High-intensity arc pulses with a peak-to-peak voltage range of +15 kV to −15 kV (=30 kV) are used to create plasma in DBD reactor. The relay must be turned on and off repeatedly in order to achieve peak-to-peak voltage. DAQ is the data acquisition card that is used to retrieve sensor (NOx) data and pass control signals to the ultimate feedback controller. NOx emissions are detected and regulated using a customized ADu841 VVME processor-based VMAT DAQ card (Maheswari 2014, 2015).
RSM modeling

Experimental conditions are determined using the RSM and planning technique that decreases the workload significantly while still obtaining an optimal model result. One of the main operations in process optimization design studies, the Box-Behnken design (or BBD), describes how much more improvements in production output, or efficiency can be attained by the process as well as the conditions in which it can be carried out (Srikanth et al. 2021). For the experimental layout, Minitab 18 software is being used. Four parameters and three stages are involved in the BBD configuration. As seen in Table 1, the independent variables are NOx initial concentration, gas flow rate, high voltage, and electrode gap, and respective values have been labeled as $+1$, $0$, $-1$, and thus the response variable being NOx reduction and reactor energy efficiency. Experimental runs included 27 runs which typically consisted of three attempts each to correct and prove an error before they are incorporated into the calculations to determine experimental error (Table 2).

The NOx elimination BBD model displayed an association between independent variable as a secondary response surface model. The following equation (Eq. 1) demonstrates the relationship between independent and dependent variables:

$$Y_0 = \delta_0 + \sum_{i=1}^{k} \delta_i X_i + \sum_{i=1}^{k} \delta_{ii} X_i^2 + \sum_{i<j=2}^k \delta_{ij} X_i X_j + \vartheta_i$$

where $Y_0$ denotes the predicted variable response; $X_i$ and $X_j$ denotes independent variables; $\delta_i$, $\delta_{ii}$, and $\delta_{ij}$ denotes the $i^{th}$ linear, quadratic, and interaction coefficients; and $\vartheta_i$ denotes the error. The original concentration of NOx pumped into the reactor is referred to as $NO_{in}$, while the concentration of NOx at the reactor’s vent or exit is referred to as $NO_{out}$. As seen in Eq. 2, NOx removal is measured in ppm (parts per million).

$$NOx\text{ removal}\% = \frac{NO_{in} - NO_{out}}{NO_{in}} \times 100$$

The most commonly used metrics for measuring non-thermal plasma energy consumption are power density and energy efficiency, which are determined using the following equations (Eq. 3–4) (Mansouri et al. 2020; Suresh et al. 2021b).

$$\text{Power density (J/lit)} = \frac{\text{Power (W)} \times 60}{\text{Gas flow rate (lpm)}}$$

$$\text{Energy Efficiency (g/J)} = \frac{NO_{in} - NO_{out}}{\text{Power density}}$$

ANOVA is a form of analysis of variance that is employed in statistics, which looks for the combination of internal variability (randomness) and underlying trend (systematic) variables that accounts for the total variability discovered in the study. Statistical variables may or may not affect the
| Run order | Factors | Actual | BBD predicted | BBD error (%) | ANN predicted | ANN error (%) |
|-----------|---------|--------|---------------|---------------|---------------|---------------|
|           | A       | B      | C             | D             | NOx reduction (%) | Energy efficiency (g/J) | Energy efficiency (g/J) | NOx reduction (%) | Energy efficiency (g/J) | NOx reduction (%) | Energy efficiency (g/J) |
| 1         | 300     | 2      | 25            | 4             | 54.6           | 26.21          | 53.16          | 23.54          | 2.64          | 10.18          | 54.901          | 26.65          | -0.55          | -1.68          |
| 2         | 400     | 2      | 25            | 4             | 47.5           | 30.4           | 46.99          | 29.94          | 1.08          | 1.51           | 47.564          | 30.316         | -0.13          | 0.28           |
| 3         | 300     | 6      | 25            | 4             | 37.2           | 53.65          | 37.48          | 54.66          | -0.74         | -1.87          | 37.163          | 53.735         | 0.10           | -0.16          |
| 4         | 400     | 6      | 25            | 4             | 26.4           | 50.76          | 27.61          | 53.97          | -4.56         | -6.33          | 26.489          | 50.767         | -0.34          | -0.01          |
| 5         | 350     | 4      | 20            | 3             | 38.9           | 54.48          | 39.39          | 53.78          | -1.26         | 1.29           | 36.45          | 54.161         | 6.30           | 0.58           |
| 6         | 350     | 4      | 30            | 3             | 48.3           | 45.09          | 49.04          | 46.49          | -1.54         | -3.10          | 48.188          | 45.023         | 0.23           | 0.15           |
| 7         | 350     | 4      | 20            | 5             | 33.8           | 47.32          | 33.56          | 47.38          | 0.72          | -0.13          | 33.848          | 47.286         | -0.14          | 0.07           |
| 8         | 350     | 4      | 30            | 5             | 43.7           | 40.78          | 43.21          | 40.09          | 1.13          | 1.69           | 43.706          | 40.731         | -0.01          | 0.12           |
| 9         | 300     | 4      | 25            | 3             | 47.3           | 45.4           | 46.56          | 45.74          | 1.56          | -0.74          | 47.293          | 45.413         | 0.01           | -0.03          |
| 10        | 400     | 4      | 25            | 3             | 38.1           | 48.76          | 38.54          | 48.59          | -1.16         | 0.34           | 38.009          | 48.724         | 0.24           | 0.07           |
| 11        | 300     | 4      | 25            | 5             | 40.1           | 38.49          | 40.73          | 39.34          | -1.57         | -2.21          | 39.334          | 39.215         | 1.91           | -1.88          |
| 12        | 400     | 4      | 25            | 5             | 31.6           | 40.44          | 32.71          | 42.20          | -3.51         | -4.35          | 31.587          | 40.476         | 0.04           | -0.09          |
| 13        | 350     | 2      | 20            | 4             | 44.8           | 31.36          | 40.88          | 33.35          | 8.75          | -6.35          | 44.751          | 31.416         | 0.11           | -0.18          |
| 14        | 350     | 6      | 20            | 4             | 29.6           | 62.03          | 29.38          | 60.93          | 0.74          | 1.78           | 27.731          | 62.028         | 6.31           | 0.00           |
| 15        | 350     | 2      | 30            | 4             | 56.4           | 26.32          | 56.56          | 26.06          | -0.29         | 0.98           | 56.376          | 26.364         | 0.04           | -0.17          |
| 16        | 350     | 6      | 30            | 4             | 40.4           | 56.56          | 39.03          | 53.64          | 3.39          | 5.17           | 40.602          | 56.756         | -0.50          | -0.35          |
| 17        | 300     | 4      | 20            | 4             | 39.7           | 47.64          | 38.82          | 46.18          | 2.22          | 3.06           | 39.663          | 47.501         | 0.09           | 0.29           |
| 18        | 400     | 4      | 20            | 4             | 31.8           | 50.88          | 30.80          | 49.04          | 3.14          | 3.61           | 31.941          | 50.908         | -0.44          | -0.06          |
| 19        | 300     | 4      | 30            | 4             | 46.2           | 36.96          | 48.47          | 38.89          | -4.91         | -5.23          | 53.131          | 36.939         | -15.00         | 0.06           |
| 20        | 400     | 4      | 30            | 4             | 41.5           | 44.26          | 40.45          | 41.75          | 2.53          | 5.67           | 41.091          | 42.631         | 0.99           | 3.68           |
| 21        | 350     | 2      | 25            | 3             | 55.3           | 30.97          | 54.66          | 32.90          | 1.16          | -6.25          | 55.468          | 32.719         | -0.30          | -5.65          |
| 22        | 350     | 6      | 25            | 3             | 36.4           | 61.25          | 37.12          | 60.48          | -1.98         | 1.26           | 36.371          | 61.19          | 0.08           | 0.10           |
| 23        | 350     | 2      | 25            | 5             | 48.3           | 27.05          | 48.82          | 26.51          | -1.08         | 2.00           | 48.289          | 27.177         | 0.02           | -0.47          |
| 24        | 350     | 6      | 25            | 5             | 31.8           | 53.5           | 31.29          | 54.08          | 1.61          | -1.09          | 31.777          | 53.455         | 0.07           | 0.08           |
| 25        | 350     | 4      | 25            | 4             | 41.6           | 46.59          | 41.30          | 46.93          | 0.72          | -0.74          | 41.531          | 46.653         | 0.17           | -0.13          |
| 26        | 350     | 4      | 25            | 4             | 41.6           | 46.59          | 41.30          | 46.93          | 0.72          | -0.74          | 41.531          | 46.653         | 0.17           | -0.13          |
| 27        | 350     | 4      | 25            | 4             | 41.6           | 46.59          | 41.30          | 46.93          | 0.72          | -0.74          | 41.531          | 46.653         | 0.17           | -0.13          |
given results, depending on whether they are systematically distributed. This statistical analysis helps researchers estimate the effect that the study variables have about the results of a study’s dependent variable (McHugh 2011). When forecasting the result of the given occurrence, the coefficient of determination would be a mathematical calculation that evaluates the extent to which variations throughout one parameter could be justified by differences in another variable. In other words, this coefficient, more often referred to as $R^2$, quantifies the strength of a linear interaction between two variables and has been heavily used by researchers when doing pattern analysis (Armstrong et al. 2002; Keselman et al. 1998). To understand the significance of the effect of the responses to be assessed, ANOVA has been used. In accordance with the quadratic model calculation, 2-based contour plots are created, and they have been used to arrive at conclusions regarding the influence of each parameter as well as to examine the interplay between them (Zwanenburg et al. 2011).

**ANN modelling for future forecasting the NOx removal efficiency**

ANN has a biological analogy for the fundamental concept of a highly flexible and effective computer device. The network consists of several interconnected units to allow for data communications between them. These units are basic generators which run in parallel. They also are called nodes or neurons (Agatonovic-Kustrin & Beresford 2000). Each neuron is linked by a communication connection to another neuron. Each link has been linked to a weight that provides information on the input signal. This has been the most helpful input for the resolution of a specific problem by neurons since the weight normally stimulates or prevents the signal. The internal condition of each neuron has been known as a signal stimulation. Some signals are produced after being combined with an input rule; and signals resulting from combining the two would always be transmitted to other units (Agatonovic-Kustrin & Beresford 2000, Elmolla et al. 2010).

A back-propagation trained model is used for the neural network. Neural background nets with a single hidden layer are shown to provide reliable approximations to any continuous function if adequate hidden neurons are present. One essential aspect of back propagation neural networks has been that the relations among different variables are not defined. Instead, they benefit from the explanations that are shown to them. Additionally, individuals are capable of generalizing accurate answers, one that superficially mimics the information used during the learning process (Civelekoglu et al. 2009; Sakiewicz et al. 2020).

Modeling the ANN framework is done using the same dataset that is used for the modelling of RSM and is one of the exercises in a major upgrade in the RSM model. Based on experimental results (Table 2), numerical and numerical prediction, a full model is created to forecast the NOx reduction (using the MathWorks’ MATLAB programme). The application used a 3-layered neural network. There have been four input nodes, ten hidden nodes, and two output nodes as illustrated in Fig. 3. A back-propagation algorithm based on the Levenberg–Marquardt principle with a sigmoidal function has been introduced. The experimental neurons contained an NOx concentration, flow rate, voltage, and electrode gap, which are each fed into the evaluate to calculate the output NOx reduction and energy efficiency.

**Model validation using RSM and ANN**

The coefficient of determination ($R^2$), the adjusted $R^2$ value, and the mean squared error are used to measure and equate the RSM and ANN models’ accuracy. Equations 5–8 characterise MSE, RMSE, MAPE, and $R^2$, respectively (Kim et al. 2019; Soleimanzadeh et al. 2019).

\[
MSE = \frac{\sum_{i=1}^{n} (E_i - Y_i)^2}{n} \tag{5}
\]

\[
RMSE = \sqrt{\frac{\sum_{i=1}^{n} (E_i - Y_i)^2}{n}} \tag{6}
\]

\[
MAPE = \frac{\sum_{i=1}^{n} |E_i - Y_i/E_i|}{n} \times 100 \tag{7}
\]

\[
R^2 = 1 - \frac{\sum_{i=1}^{n} (E_i - Y_i)^2}{\sum_{i=1}^{n} (E_i - E_m)^2} \tag{8}
\]

The residuals, $E_i$, is the variance between the observed response. If the estimates from the model matched the experimental values exactly, $R^2$ is equal to 1.0. The $R^2$ is a slightly
changed abbreviation of the original that incorporates the number of predictors. From Eqs. 5–8, \( n \) denotes number of trials, \( Y_i \) denotes predicted response, and \( E_{\text{m}} \) denotes average observed response.

**Results and discussion**

**RSM modeling**

**Model validation**

Table 3 shows the experimental outcomes achieved by BBD model and reasonably similar the calculated data of the 3 replicated groupings showing the data are reproducible properly. Predicted accuracy and error rates and the experiment show agreement with the results, thereby proving that the prediction works. The discrepancy between the expected values and the actual value is lower than 2, which means that data fits as predicted. To determine the factors to be tested, the design is built using the second-order polynomial model. NOx concentration (A), flow rate (B), voltage (C), and electrode gap (D) are used as independent inputs to determine the dependent variable includes NOx reduction and energy efficiency. Equations 9–10 are effective for characterizing the effect of each variable’s coefficient.

\[
\text{NOx reduction(\%)} = -13.4 + 0.423 A - 4.49 B + 0.9650 C - 2.917 D - 0.000666 AXA + 0.418 BXB - 0.00925 AXB
\]  
(9)

\[
\text{Energy efficiency (g/J)} = -143.5 + 0.930 A + 19.97 B - 0.729 C - 3.197 D - 0.001186 AXA - 0.860 BXB - 0.01770 AXB
\]  
(10)

The above results show the model’s quadratic coefficient of flow rate to have the most beneficial effect on NOx elimination with its maximum negative coefficient and energy efficiency with its maximum positive coefficient. This is due to the decrease in NOx from increasing the gas flow rate, while the energy efficiency is enhanced by the rise in the flow rate. The negative symbol for the electrode gap is the second most important variable to reduce the reduction of NOx and energy efficiency by increasing the flow rate. Similar results are obtained by Mansouri et al. (2020) in the experiment of the NOx reduction by packed bed DBD reactor.

The normal test plot has been used to see whether results follow the bell curve or the normal distribution. According to Fig. 4a and b, the residuals are closely compared to the linear line and ranged between – 2 and 2 in terms of NOx reduction and – 4 to 4 in terms of energy efficiency. The pareto chart shows the prevalence of defects and also its overall effect, which would also be usually referred to as a bar chart. It would be very helpful to use pareto charts to identify flaws in order to locate the most prevalent elements of the model. From Fig. 5, the bars are listed from largest to smallest in ascending order (from tallest to shortest). The size of the largest bar represents the most critical aspect of the response. Figure 5a illustrates the pareto curve for NOx reduction, indicating that the most critical element is gas flow rate, followed by voltage, NOx concentration, and electrode gap. Similar to that is shown in Fig. 5b; the gas flow rate has the highest influencing factor compared to voltage, electrode gap, and NOx concentration being taken into consideration when measuring performance.

| Source       | DF | Adj SS | Adj MS | F value | p value | Remarks  |
|--------------|----|--------|--------|---------|---------|----------|
| Model        | 14 | 1548.49| 110.607| 71.62   | <0.0001 | Highly significant |
| A            | 1  | 193.60 | 193.603| 125.36  | <0.0001 | Highly significant |
| B            | 1  | 920.50 | 920.501| 596.03  | <0.0001 | Highly significant |
| C            | 1  | 279.37 | 279.367| 180.89  | <0.0001 | Highly significant |
| D            | 1  | 102.08 | 102.083| 66.10   | <0.0001 | Highly significant |
| A X A        | 1  | 17.04  | 17.041 | 11.03   | 0.006   | Significant |
| B X B        | 1  | 12.81  | 12.813 | 8.30    | 0.014   | Significant |
| C X C        | 1  | 0.12   | 0.120  | 0.08    | 0.785   | Significant |
| D X D        | 1  | 0.61   | 0.608  | 0.39    | 0.542   | Significant |
| Error        | 12 | 18.53  | 1.544  |         |         | Not Significant |
| Lack-of-Fit  | 10 | 18.53  | 1.853  | 5.24    | 0.345   | Not Significant |
| Total        | 26 | 1567.03|        |         |         |           |
| S            |    | 1.2427 |        |         |         |           |
| \( R^2 \)    |    | 0.9882 |        |         |         |           |
| \( R^2 \) (adj) |    | 0.9744|        |         |         |           |
| \( R^2 \) (pred) |    | 0.9319|        |         |         |           |
ANOVA analysis

Tables 3 and 4 demonstrate the ANOVA findings for the BBD model for NOx reduction and energy efficiency, respectively. The data illustrate the SS (sum of squares), DF (degree of freedom), MS (mean square), F value, and probabilities (p value) for all the linear and quadratic parameters. The studies have been performed to see the impact of both of individual factors as well as the synergistic ones as in BBD model is of several factors.

Table 4 displays the NOx reduction ANOVA effects of the BBD model. Many of the linear terms including NOx concentration, flow rate, voltage, and electrode gap in the RSM model had p values below 0.0001, which implies that they are important to the model. The most important of the four factors is in particular the gas flow rate which is contributed 58.74% in NOx reduction. By examining the ANOVA in Table 4, we learn how often independent variables impact on the reduction of NOx. By analyzing the significance level of the variables’ quadratical terms, we observed that the model is applicable to $A^2$, $B^2$, $C^2$, and $D^2$. The model should be subjected to the following conditions: $F > 0.1$, $R^2 > 0.95$, $R^2$ (pred) > 0.7, and $R^2$ (adj) $- R^2$ (pred) < 0.2. p value and F value are employed to validate the model. From Table 3, the fit of this model is good in the regression field, the F value of the model is 71.62 with p < 0.0001. The $R^2$ value of 0.9882, $R^2$ (pred) of 0.9319, and $R^2$ (adj)- $R^2$ (pred) of 0.0425 demonstrate a high level of consistency and credibility.

Similarly, Table 4 presented the ANOVA effects of energy efficiency in BBD model. The contribution of the overall model is greater than that is stated here, with a p value of < 0.0001, which implies that the variables do significantly impact the response. According to the model, the NOx concentration, flow rate, voltage, and electrode gap (linear terms) had a significant effect on energy efficiency of DBD reactor. Out of these factors, flow rate has a major significant one which contributes 82.33% for energy efficiency. The initial NOx concentration and flow rate affected the energy
efficiency of DBD reactor, which is quadratic rather than linear. From Table 4, the fit of this model is good in the regression field; the $F$ value of the model is 122.18 with $p < 0.0001$. $R^2$ value of 0.9783, $R^2_{(pred)}$ of 0.9357, and $R^2_{(adj)}$ of 0.0346 demonstrate a high level of consistency and credibility. Figure 6 shows a close association between the predicted and real values with an $R^2$ high coefficient value of 0.9788 for the reductions in NOx and energy efficiency. So as a result, measurement established a reasonable expectation of the expected response within the scope of the study.

**Effect of independent variables on the responses**

The two-dimensional contours shown in Figs. 7 and 8 examine the influence of the input factors in the DBD reactor, as seen in the BBD model, on NOx reductions and on the energy efficiency. Figure 7a, b, and c examines the impact of two variables, with a different flow rate, voltage, and electrode gap, respectively, that the NOx concentration is set. The darker portion of Fig. 7a, b, and c shows the greatest amount of nitrogen oxides removed, while the lightest part of Fig. 7b shows the lowest exclusion. In Eqs. 3–4, the flow improvement decreases energy density and, in essence, improves energy efficiency by enhancing gas flow (Shin et al. 2019). The greater the gas flow rate, the lower the gas holding duration in the reactor, leading to reductions in energy demand per unit mass and increased energy efficiency, leading to the

| Table 4 ANOVA results for the energy efficiency |
|-----------------------------------------------|
| **Source** | **DF** | **Seq SS** | **Adj MS** | **F value** | **p value** | **Remarks** |
| Model       | 7      | 2710.33    | 387.19     | 122.18      | <0.0001     | Highly significant |
| A           | 1      | 24.51      | 24.51      | 7.73        | 0.012       | Significant |
| B           | 1      | 2280.87    | 2280.87    | 719.72      | <0.0001     | Highly significant |
| C           | 1      | 159.43     | 159.43     | 50.31       | <0.0001     | Highly significant |
| D           | 1      | 122.69     | 122.69     | 38.71       | <0.0001     | Highly significant |
| A X A       | 1      | 34.59      | 56.29      | 17.76       | <0.0001     | Highly significant |
| B X B       | 1      | 75.71      | 75.71      | 23.89       | <0.0001     | Highly significant |
| Error       | 19     | 60.21      | 3.17       |             |             |              |
| Lack-of-fit | 17     | 60.21      | 3.54       | 6.24        | 0.547       | Not significant |
| Total       | 26     | 2770.54    |            |             |             |              |

$S$ = 1.78020
$R^2$ = 97.83%
$R^2_{(adj)}$ = 97.03%
$R^2_{(pred)}$ = 93.57%

**Fig. 6** Actual vs predicted plot. **a** NOx reduction and **b** energy efficiency
decrease in NOx level. Similar findings are occurred in NOx deduction by plasma method as stated the researcher Shin et al. (2019).

The graphs 7b and 8b illustrate the effect of two variables, NOx concentration and voltage, on NOx reduction and energy efficiency. Because the voltage’s value is so closely correlated with NOx concentration, it is apparent that this with respect to DBD revealed the DBD/NOx product voltage is the most dramatically influential. Additionally, the energy efficiency reduced as the voltage is increased in relation to the NOx concentration. Increased voltage greatly raises the energy density of micro discharge, which accelerates NOx reduction but reduces the DBD reactor’s energy efficiency. The similar findings are stated several researchers (Aerts et al. 2015; Chen et al. 2017; Talebizadeh et al. 2014). It is also found that in the process of raising the voltage, the electron density in the gas triggered a further reaction (Eqs. 11–14), which took place in the plasma phase (Talebizadeh et al. 2014).

\[
\begin{align*}
O_2 + e & \rightarrow O + O + e \\
O_2 + O & \rightarrow O_3 \\
NO + O & \rightarrow NO_2 \\
NO + O_3 & \rightarrow NO_2 + O_2
\end{align*}
\]  

Figure 7d shows the interrelationship of voltage to flow rate on NOx reduction with a given NOx concentration and distance of 350 ppm and 4 mm. Increasing the voltage as the flow rate increased had a beneficial effect on NOx reduction and energy consumption (Fig. 8d). Similarly, raising the flow rate with a distance has a negligible effect on energy efficiency (Fig. 8e), but the minimum flow rate with a small gap has the greatest effect on NOx reduction (Fig. 7e). Additionally, Fig. 7f illustrates the effect of increasing the voltage with an increase in the electrode gap on NOx reduction at a constant NOx concentration and flow rate, despite the fact that increasing the voltage with an increase in the electrode gap had a negative effect on NOx reduction. A similar finding is obtained for the DBD reactor’s energy efficiency (Fig. 8f). Zhu et al. (2020) found an alternative approach that used the DBD. Only ammonia is used to make DBD radicals, which are combined with the flue gases to start a reaction. The removal efficiencies ranged between 93.89% and energy density of 500 J/lit. Similarly, when catalytic based red mud packed plasma was used, more than 90% of NOx removal efficiency was achieved (Nishanth et al. 2021). In contrast when applying chemical-assisted hybrid plasma technique, 40% of NOx removal efficiency was achieved (Yamasaki et al. 2022).

Optimum conditions

The maximum response is used as a benchmark for optimization concerning NOx reduction and energy efficiency, as well as other parameters including NOx concentration, flow rate, voltage, and electrode gap in the experimental study region. Using experimental trials, the optimal values obtained for NOx concentration, flow rate, voltage, and Input voltage for reactor are presented in Table 5. As a result of this optimum condition, the overall NOx reduction is 60.5%, and the energy efficiency is 66.24 J/lit.

ANN modeling

ANN is used for the modelling and testing of the neuronal network model in NOx reduction experiments using the experimental results under the BBD operating conditions. To evaluate the optimal ANN model, a hidden layer of 1–14 neurons is used. Figure 9 showed the preparation, evaluation, and test network performance curves along with their
R-squared values for the R-terms (predictor-predicted), for comparison between all possible configurations of the network architectures. The accurate outcome presented here (network inputs equal output requirements) comes up with $R = 0.99$. The findings show a strong association between the output values and goals during training ($R = 0.99099$) and testing ($R = 0.9979$). The results showed good correlations between output values and objectives. From Table 2, predicted findings are checked by the experimental data, confirming that the model had a good correlation with the actual result. Additional tests also demonstrate that the non-linearity of the structure correctly as measured by the ANN model.

The “cause and effect” of the input variables is modeled by means of sensitivity analysis. Therefore, the system efficiency is determined to be improved by the results of the inputs of the predictor factors. In order to establish the value of the network variables for NOx reduction, the Garson equation and connection weight algorithm have been employed. ANN given the coefficients, which stand for the interactions between inputs and outputs in an NTP reactor, each of which is calculated based on how much of the signal they received. The Garson equation and connection weights method are used to determine the relative influence of four input variables, as seen in Eq. 15–16 (Goh 1995; Shin et al. 2019).

$$R_{IC} = \frac{\sum_{m=1}^{k} |w_{mn}w_{nh}| / \sum_{t=1}^{v} |w_{tn}|}{\sum_{m=1}^{k} \sum_{n=1}^{k} (|w_{mn}w_{nh}| / \sum_{t=1}^{v} |w_{tn}|)}$$ (15)

| Table 5 | Optimum conditions from BBD design |
|---------|-----------------------------------|
| NOx concentration (ppm) | Flow rate (lpm) | Voltage (kVpp) | Electrode gap (mm) | Result |
| NOx reduction  | 300 | 2 | 30 | 3 | 60.5% |
| Energy efficiency | 350 | 6 | 20 | 3 | 66.24 g/J |

Fig. 8 Contour plot for energy efficiency. a Flowrate vs NOx concentration, b voltage vs NOx concentration, c gap vs NOx concentration, d voltage vs flowrate, e gap vs flowrate, and f gap vs voltage
where \( RIG \) and \( RIC \) denotes relative importance by garsons and connection weights algorithm, \( v \) and \( k \) denote number of hidden and input neurons, \( w_{mn} \) and \( w_{nh} \) denote connection weight among input and hidden neuron and connection weight among hidden and output neuron.

\[
RIC = \sum_{n=1}^{k} |w_{mn}w_{nh}|
\]  

(16)

Table 6 Relative importance of input factors for NOx reduction

| Input variables | Connection weight method | Garson method |
|----------------|--------------------------|--------------|
|                | Relative importance | Rank | Relative importance | Rank |
| NOx concentration | 16.98 | 4 | 2.33 | 3 |
| Flow rate (lpm) | 25.25 | 3 | 2.67 | 2 |
| Voltage (kVpp) | 32.28 | 1 | 2.68 | 1 |
| Electrode gap (mm) | 28.42 | 2 | 2.32 | 4 |

Table 7 Comparison results for BBD and ANN

| Response         | Error | RSM | ANN |
|------------------|-------|-----|-----|
| NOx reduction    | MSE   | 1.274 | 2.167 |
|                   | RMSE  | 1.128 | 1.472 |
|                   | MAPE  | 2.053 | 1.726 |
|                   | R²    | 0.98  | 0.99 |
| Energy efficiency| MSE   | 2.230 | 0.246 |
|                   | RMSE  | 1.493 | 0.496 |
|                   | MAPE  | 2.903 | 0.615 |
|                   | R²    | 0.97  | 0.99 |

In all cases, the findings show that all relation weights displayed positive or negative values and in addition to being influenced. To prevent this, all relation weights have been determined in the adjusted Garson equation with their absolute value. Thus, the two approaches are compared on the end results and tested based on the weights that are derived from preparation (Zhou et al. 2015). The changes in initial weights influenced the final weight. As seen in Table 5, the parameters are ranked according to their relative significance. This is based on the findings of the link weight process, which showed voltage is the most influential in the rate of reduction of NOx emission, followed by electrode distance and NOx concentration.

In the data obtained by the Garson method (Table 6), it can be concluded that voltage and flow rate have the greatest effect on the removal rate, then NOx concentration and electrode gap, followed by NOx concentration. In terms of voltage and flow rate, this finding is not commensurate with the RSM result. The most important element (58.74% contribution) is that of voltage, according to the RSM model than flow rate (17.83% contribution). From this perspective, both flow rate and voltage have been the most critical variables, while electrode distance is the least critical. Garson method is highly consistent with the BBD paradigm, according to this comparison of the relative value of ANN to BBD. Sensitivity analysis can quantify the proportion of effect, but the quadratic function describes the positive/negative correlation.

RSM and ANN model validation

The predictions, which are gathered from the experimentally observed responses, are compared to find out whether an ANN or an RSM approach is more efficient. The good use of \( R^2 \)
values near 1.0, with the comparison of the statistic parameters acquired by ANN and RSM, is validated with the good usefulness of model’s prediction. The findings of RSM and ANN experiments in NOx reduction and energy efficiency have been seen in Table 7, which compared their values to those of $R^2$, MSE, RMSE, and MAPE. RSM models NOx drop, $R^2$, MSE, RMSE, and MAPE are, respectively, 0.98, 1.274, 1.128, and 2.053. In ANN, 0.99, 2.167, 1.472, and 1.276, respectively, for the $R^2$, MSE, RMSE, and MAPE model. Similarly, the $R^2$, MSE, RMSE, and MAPE values from the RSM models are, respectively, 0.97, 2.230, 1.493, and 2.903 for energy efficiency. A for ANN model, the $R^2$, MSE, RMSE, and MAPE models included 0.99, 0.246, 0.46, and 0.615. It is found that the ANN models are more accurate than the RSM model at predicting the NOx removal/reduction and efficiency data. Figure 10 depicts the distribution of experimental and predicted values of BBD and ANN model for NOx reduction of DBD system.

Conclusion

The experimentation was carried out in this study to limit NOx emissions from diesel engines using an NTP-based DBD reactor. The NOx elimination and energy efficiency of the DBD reactor have been modeled using Box-Behnken design (BBD) and artificial neural network (ANN) techniques. The current study’s key conclusions can be outlined as follows: the final output model for the NTP process outlined 98.82% of variation in NOx reduction and 97.83% of the variation in energy efficiency. The plots show that the rise in the flow rate has been a strong factor in the reduction of NOx, which has a direct effect on energy efficiency as the flow rate increases. NOx removal was calculated to be 60.5% under the optimal conditions with an energy efficiency of 66.24 g/J. With a cooperative correlation coefficient of 0.99 for NOx reduction and energy efficiency, this model is capable of capturing the uncertainties of the experimental data stronger than the RSM model. As a result, the ANN model’s accuracy in estimating the NOx removal process is higher than the RSM model. These studies have led to potential outcomes in wide-ranging application of plasma technologies for the NOx removal. Further in future, the same study may be carried out for simultaneous reduction of NOx and SO2 removal efficiency from the industrial flue gas emissions. Also, the NTP reactor can be studied with respect to catalytic medium to further enhance the energy yield. A detailed study can be carried out in future to estimate the cost involved while scaling up the reactor for Industrial applications.

Author contribution CM supervised and validated the whole project. ASR analyzed, interpreted, and wrote the manuscript. RB helped in the ANN analyze. NS, RR, and MV conducted the experiments.

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Declarations

Ethics approval and consent to participate Not applicable.

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