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Regular paper
Investigation of SnO_2-based Thick Film Gas Sensors through Optimization of Artificial Neural Networks Parameters for Hazardous Gas Detection

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Abstract: - In the present study, a systematic approach for optimizing artificial neural network parameters for the detection of a greenhouse gas carbon di-oxide (CO_2) through SnO_2-based thick film gas sensors for a wide range of concentrations $\mathbf{0-6000}\ \boldsymbol{ppm}$ is presented. The un-doped and Pd-doped SnO_2 thick film is used as the sensor. The approach includes dataset preparation, input and output variable selection, artificial neural network architecture selection, training, validation, hyperparameter tuning, and testing. The input variables were selected based on their relevance to the problem at hand, such as the concentrations of the gas. The output variables were the changes in resistance of the sensor in response to the corresponding input variables. The artificial neural network architecture was carefully chosen, considering factors like the number of layers, neurons per layer, activation functions, and learning rate. Using the prepared dataset, the artificial neural network was trained by adjusting the connection weights between neurons to minimize the disparity between actual and predicted sensor responses. After training, the accuracy of the artificial neural network and generalization ability were assessed using a separate dataset for validation. The approach in the presented study can be used for different types of gases, and can be utilized in various applications

Keywords: Gas sensors, SnO_2-Thick film, Greenhouse gases, Artificial Neural Networks, Environmental Monitoring.

I. INTRODUCTION

Detecting toxic/greenhouse gases is crucial in industries like mining, chemical, petrochemical etc. SnO_2-based thick film gas sensors are reliable and cost-effective for this purpose. These sensors are favored for their sensitivity, affordability, and ease of production. However, the accuracy and sensitivity of these gas sensors can be significantly improved by optimizing the Artificial Neural Network (ANN) parameters used in gas sensing applications. The ANN is a machine learning technique that can learn and recognize patterns in large datasets, making it an attractive option for gas sensing applications.

The optimization of ANN parameters, such as the number of hidden layers, number of neurons, learning rate, and activation function, is essential for enhancing the accuracy and sensitivity of gas sensors. Various studies have focused on this optimization process, employing techniques such as genetic algorithms, backpropagation algorithms, and evolutionary computation to achieve high accuracy and sensitivity in gas sensing applications [[1]]. Previous research has highlighted the development of gas sensors for detecting toxic/greenhouse gases using SnO_2-based thick film gas sensors and the optimization of ANN parameters to improve their performance [[2], [3]].

For instance, Cheng *et al.* (2023) developed an ANN-based gas sensor for detecting hydrogen gas, utilizing doped SnO_2 thick film gas sensors. They optimized the ANN parameters using a genetic algorithm, achieving high accuracy metrics for hydrogen detection, including a regression coefficient of 0.9882, an average absolute deviation of 2.74, and a root mean square error of 8.05 [[4]]. Another study by Martinelli *et al.* (1995) reviewed the use of thick-film gas sensors for detecting hazardous gases. They reported that SnO_2-based sensors are widely used due to their high sensitivity, low cost, and ease of fabrication, and they emphasized the importance of optimizing ANN parameters for accurate gas detection [[5]].

In a study by Ismail *et al.* (2023), researchers optimized an ANN structure for detecting alcohol aroma using an MQ-3 gas sensor. The optimized ANN, with 2 hidden layers, 10 neurons, and 1000 iterations, achieved high accuracy in classifying alcoholic and non-alcoholic liquids [[6]]. Zhai *et al.* introduced a real-time gas classification system utilizing a multi-layer perceptron (MLP) ANN designed to swiftly detect and categorize gas sensor data, ensuring high accuracy with low latency [[7]].

Guerbas *et al.* focused on diagnosing power transformer insulating oil using MLP neural networks optimized by improved particle swarm optimization (PSO), demonstrating robustness and high accuracy in gas concentration

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prediction, outperforming classical diagnostic methods [[8], [9]]. These sensors operate on the principle of changes in electrical properties, such as resistance, in response to various gases. They are cost-effective, robust, and capable of detecting a wide range of gases.

The accurate detection of toxic/greenhouse gases through SnO_2-based gas sensors requires optimizing ANN parameters. ANNs are computational models that imitate the design and capability of human intelligence and are widely used in gas sensor data analysis. ANN-based gas detection systems offer advantages over traditional methods, including higher accuracy, robustness, and the ability to detect multiple gases simultaneously. However, the performance of these systems depends on optimizing various parameters, including dataset preparation, input and output variable selection, architecture selection, training, validation, hyperparameter tuning, and testing.

Dataset preparation is the first step in optimizing ANN parameters for gas detection [[10]]. A dataset should be prepared by collecting sensor responses to various gas concentrations, covering a wide range of toxic/greenhouse gases [[11], [12]]. The input variables should be selected based on their relevance to the problem at hand. The output variables should represent the modification in the electrical behavior of the sensor in response to the corresponding input variables.

The next step is choosing an ANN architecture, which includes deciding the number of hidden layers, the number of neurons in each layer, the activation functions, and the learning rate. The choice of ANN architecture depends on the complexity of the problem and the size of the dataset. A complex problem necessitates a greater number of hidden layers and neurons, whereas a smaller dataset requires fewer hidden layers and neurons.

After selecting the ANN architecture, the ANN is trained using the prepared dataset. The training involves adjusting the weights of the connections between neurons in the network to minimize the difference between actual sensor responses and predicted sensor responses. This process involves forward propagation of input signals through the network, followed by backpropagation of errors to adjust the weights [[13], [14]]. Once trained, the ANN needs to be validated to test its accuracy and generalization capability using a separate dataset not used in training.

If the ANN performs well on the validation dataset, it is ready for hyperparameter tuning. Hyperparameters, such as learning rate, regularization rate, and momentum rate, need to be optimized to improve the performance of ANN. This optimization can be done using a trial-and-error approach to find the best combination of parameters for the highest accuracy. The final step is testing the ANN on unseen data to evaluate its accuracy and reliability. Testing involves inputting new gas concentrations into the trained ANN to predict sensor responses, and accuracy is evaluated by comparing predicted sensor responses with actual responses.

The optimization of ANN parameters is crucial for the accurate detection of toxic/greenhouse gases through SnO_2-based thick film gas sensors. ANN-based gas detection systems have several advantages over traditional methods, including higher accuracy, robustness, and the ability to detect multiple gases simultaneously. The systematic approach presented in this study can be used to optimize ANN parameters for different types of gas sensors, and it can be utilized in various applications

II. METHODOLOGY

The methodology for the proposed work to be carried out can be divided into three main parts: sensor fabrication, experimental setup, and data analysis using ANNs. The crucial steps are however shown in Fig. 1.

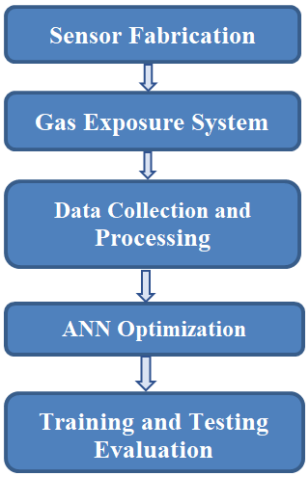

Fig. 1 Flow chart describing the chronology of the important steps.

A. Sensor Fabrication

Screen printing method is generally used to create SnO_2-based thick film gas sensors. The SnO_2 powder was combined with a binder and solvent to create a paste that was then screen-printed onto an alumina substrate. After drying, the sensors were burned at high temperatures to produce a thick coating.

B. Experimental Setup

The experimental setup consisted of a gas chamber, gas cylinders containing target gases (e.g. CO_2, NO_2, and H_2S), a power supply, and a data acquisition system. The sensors were mounted inside the gas chamber and connected to the data acquisition system. The target gases were injected into the chamber at different concentrations using mass flow controllers. The sensors were exposed to the gases for a certain period, and the response of the sensors was recorded by the data acquisition system.

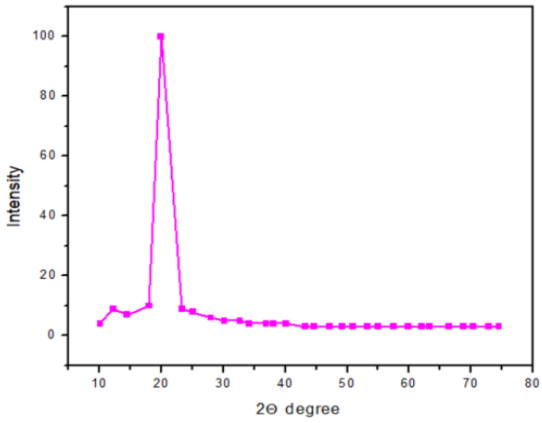

Fig. 2 X-ray beam interacting with the SnO_2 sample.

The non-destructive analytical technique of X-ray diffraction (XRD) is used to analyze the crystallographic structure of materials. The XRD graph of screen-printed SnO_2 thick films shows the diffraction pattern produced by an X-ray beam interacting with the SnO_2 sample as shown in FIG. 2. The graph depicts a succession of peaks that correlate to the various crystal planes seen in SnO_2. The locations and intensities of these peaks reveal information about the material's crystal structure and orientation. The XRD graph of screen-printed SnO_2 thick films will display several peaks corresponding to the (110), (101), (200), (211), and (220) crystal planes. The peak X-ray diffraction (XRD) value in the graph is prominently shown around the 2θ degree of approximately 26. This is where the peak intensity reaches its highest point, near 100 on the intensity scale. This peak is typically indicative of the crystalline structure in the material being analyzed, and its specific position (around 26 degrees in 2θ) could suggest the identity of the crystalline phase, depending on the material and the experimental conditions. For SnO_2 (tin dioxide), this particular peak likely corresponds to the (110) crystal plane, which is one of the common and strong diffraction peaks for cassiterite, the mineral form of SnO_2. The (110) plane in cassiterite SnO_2

is often the most intense and sharply defined peak, reflecting the ordered atomic arrangement within the crystal structure. This peak is crucial for confirming the presence of the crystalline phase of SnO_2 and can be instrumental in analyzing the quality and orientation of the crystal lattice within the sample.

C. Data Analysis using ANN

The acquired data was pre-processed into two parts: training data and testing data. The training data was used to train the ANN, while the testing data was utilized to assess the performance of the trained ANN. The pre-processed data was then fed into the ANN, which had three layers: input, hidden, and output. A grid search technique was used to maximize the number of neurons in the hidden layer, learning rate, and momentum.

A grid search technique was employed to discover the optimum combination of ANN parameters to optimize. The number of neurons in the buried layer was increased in increments of 5 from 5 to 20. The learning rate was altered from 0.01 to 0.1 in 0.01 increments, while the momentum was adjusted from 0.1 to 0.9 in 0.1 increments. The ANN's performance was assessed using two metrics: mean absolute error (MAE) and root mean square error (RMSE). The ANN was trained using the training data once the optimal combination of parameters was discovered. The trained ANN was then applied to the testing data to forecast the concentration of target gases. The MAE was used to assess the ANN's performance.

D. Data collection of sensor responses to different gas concentrations

Data collection is critical for assessing SnO_2-based thick film gas sensor performance. The gathered data helps evaluate and analyzed the sensitivity and selectivity of the sensor to various gases and concentrations. The sensor is exposed to a range of gas concentrations, and its reaction is recorded and enables to calculate sensitivity, selectivity, and reaction time. During exposure, the gas flows over the sensor surface, and conditions such as gas flow rate, duration, and temperature are controlled for reproducibility and accuracy. The sensor response is recorded in terms of resistance changes over time and stored for further analysis. Data visualization through graphs helps identify the detection limit and dynamic range. The data acquisition system ensures accurate and reliable data collection by controlling gas flow rate and exposure time, and recording data at specified sampling rates and resolutions.

E. ANN Technique

ANN models can be used as a substitute for traditional methods of examining technology and doing computations in Matlab. ANN models are made up of neurons and their connections, each with their weights. Neurons process information, whereas connections store it. Each processing node produces a weighted sum of input signals and applies a transfer function during training. "Feed propagation" refers to the training approach utilized during error backpropagation. The difference between the desired and actual output is calculated, and the weights in the hidden and output layers are adjusted using a back propagation technique with an adaptive learning rate. The result is a network that uses neurons to convert input values to output values. The feed-forward propagation approach is appropriate for testing and training data.

F. Techniques for Detecting Carbon Dioxide

Infrared (IR) Absorption: CO_2 molecules absorb specific wavelengths of infrared light, which can be detected by an IR sensor. IR sensors are commonly used in CO_2 sensors for indoor air quality monitoring [[14]].

Electrochemical Detection: Electrochemical sensors utilize a chemical reaction that occurs when CO_2 comes into contact with a specific electrode. This reaction produces an electrical current that can be measured and used to determine the CO_2 concentration [[15]].

Photoacoustic Detection: Photoacoustic sensors use a laser to heat a gas sample containing CO_2. As the gas heats up, it expands, producing sound waves that can be detected and used to calculate the CO_2 concentration [[16]].

Solid-state Detection: Solid-state CO_2 sensors use a solid material that absorbs CO_2 molecules, causing a change in electrical conductivity. This change in conductivity can be measured and used to determine the CO_2 concentration. Overall, these sensing mechanisms are used in various CO_2 monitoring devices, such as industrial gas detectors, indoor air quality monitors, and greenhouse gas monitoring systems [[16]].

Carbon dioxide, is a greenhouse gas, a by-product of human activities, such as burning fossil fuels and deforestation. It is also naturally occurring and a necessary component of the Earth's atmosphere, to regulate temperature. However, excessive levels of CO_2 in the atmosphere can contribute to climate change and have negative impacts on human health and the environment. To monitor and regulate CO_2 levels, sensors are used to detect the concentration of the gas in the air. SnO_2 (tin dioxide) sensor is one type of sensor that is commonly used for CO_2 detection. In this article, the sensitivity of SnO_2 sensor towards different concentration of CO_2 is explored.

SnO_2 sensors work by detecting changes in the electrical conductivity of the sensor material when it comes in contact with CO_2. As the concentration of CO_2 increases, the electrical conductivity of the SnO_2 material change. This change in electrical conductivity can be measured and used to determine the concentration of CO_2 in the air. The sensitivity of SnO_2 sensors for CO_2 detection can vary depending on the specific sensor and the conditions under which it is being used. In general, SnO_2 sensors have a sensitivity range of $0 - 6000\ ppm$ for CO_2, with an accuracy of $\pm 20\ ppm$ or $\pm 5\%$ of the reading (whichever is greater) [[17]].

III. RESULTS

The sensitivity in terms of change in resistance of the undoped SnO_2 thick film as a sensor for detecting CO_2 at different concentrations ranging from $0 - 6000\ ppm$ 350°C is shown in Table 1.

Table 1 Sensitivity using two different transfer functions at 350°C for undoped SnO_2 Sensor with Random Weight /Bias Rule.

Concentration of CO_2 gas (ppm)	**Sensitivity for logsin transfer function (%)**	**Sensitivity for purelin transfer function (%)**
0	5.00	4.00
1000	10.00	8.50
2000	15.00	13.00
3000	20.00	17.50
4000	24.00	19.00
5000	25.08	20.12
6000	25.00	20.00

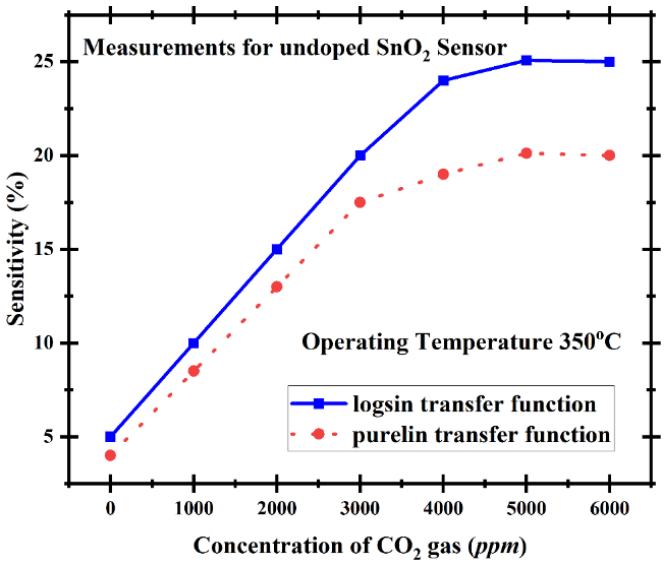

Fig. 3 Sensitivity measurements using two different transfer functions at $350°C$ for undoped SnO_2 Sensor.

From Fig. 3 obtained for the dataset given in Table 1 following points are noteworthy:

- Sensitivity for the undoped SnO_2 increases as the concentration of CO_2 rises.
- The logsin transfer function starts at a display higher sensitivity and maintains a lead over the purelin across all concentrations.
- Both logsin and purelin transfer functions reach their peak for the sensitivity around $5000\ ppm$, as suggested by the highest values provided (logsin at 25.08% and purelin at 20.12%) in Fig. 3.

Further testing was done using Matlab software and a neural network tool with various transfer functions. The highest sensitivity of 25.08% was found using the logsin transfer function. Another test using a random weight/bias rule [[18]] also resulted in a maximum sensitivity of 20.12% using the purelin transfer function. These findings suggest that the logsin transfer function may be the most effective for achieving high sensitivity in gas sensors.

The sensitivity of the 1.5% Pd-doped SnO_2 thick film as sensor for detecting CO_2 at different concentrations ranging from $0 - 6000\ ppm$ at 350°C for three different transfer functions is obtained in Table 2.

Table 2 Sensitivity using three different transfer functions at 350°C for 1.5% Pd-doped SnO_2 Sensor with Random Weight /Bias Rule.

Concentration of the CO_2 gas (*ppm*)	**Sensitivity for logsin transfer function (%)**	**Sensitivity for transin transfer function (%)**	**Sensitivity using another transin transfer function (%)**
0	45.00	42.00	40.00
1000	55.00	52.00	50.00
2000	65.00	62.00	60.00
3000	72.00	69.00	67.00
4000	75.00	72.00	70.00
5000	78.00	75.00	73.00
6000	79.54	79.28	76.00

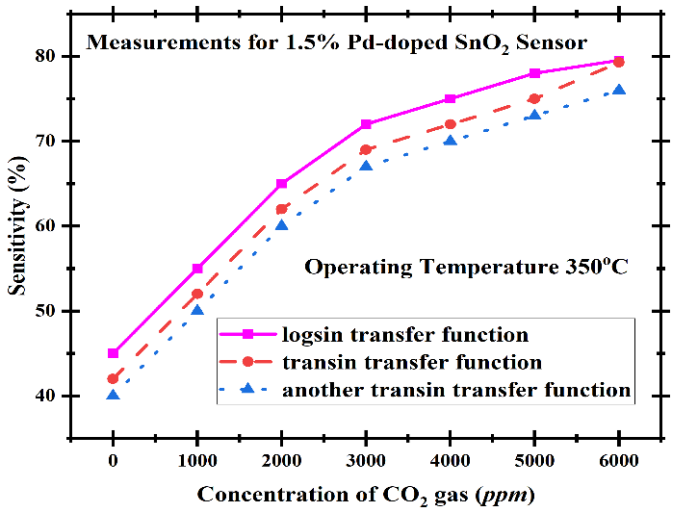

Fig. 4 Sensitivity measurements using three different transfer functions at 350°C for 1.5% Pd-doped SnO_2 Sensor with Random Weight /Bias Rule at 350°C.

From Fig. 4 obtained for the dataset given in Table 2 few important observations are outlined below:

- The logsin transfer function showcases the highest sensitivity reaching up to 79.54% at 6000 *ppm* similar to as observed in Fig. 3 for the undoped SnO_2 thick film.
- The transin transfer function also achieves high sensitivity, slightly less than logsin, peaking at 79.28% at 6000 *ppm*.
- The additional transfer function (referred to as "transin TF") was included for comparison purposes. It exhibits a more linear increase in sensitivity, peaking at 76% when the gas concentration reaches 6000 ppm. However, it demonstrates lower efficiency compared to the above two mentioned transfer functions.

The Levenberg-Marquardt feedforward propagation algorithm [[17]] revealed a maximum sensitivity of 79.54% in the logsin network transfer function at the same temperature (Fig. 4), outperforming the other transfer functions in the ANN. Moreover, when compared to the algorithm in the transin network transfer function, the random weight/bias rule achieved a maximum sensitivity of 79.28% in the network at an operating temperature of $350°C$. Amongst the three transfer function networks, the logsin function was deemed the most suitable, as it

achieved the maximum validation performance at zero epoch for the Levenberg-Marquardt feedforward propagation algorithm.

Table 3 Sensitivity using three different transfer functions at 350°C for 2% Pd-doped SnO_2 Sensor with Random Weight /Bias Rule.

Concentration of the CO_2 gas (*ppm*)	**Sensitivity for logsin transfer function (%)**	**Sensitivity for purelin transfer function (%)**	**Sensitivity using another transfer function (%)**
0	48.00	45.00	43.00
1000	58.00	55.50	53.00
2000	68.00	65.00	63.00
3000	75.00	72.50	70.00
4000	78.00	75.50	73.00
5000	81.00	78.50	76.00
6000	82.54	80.28	78.00

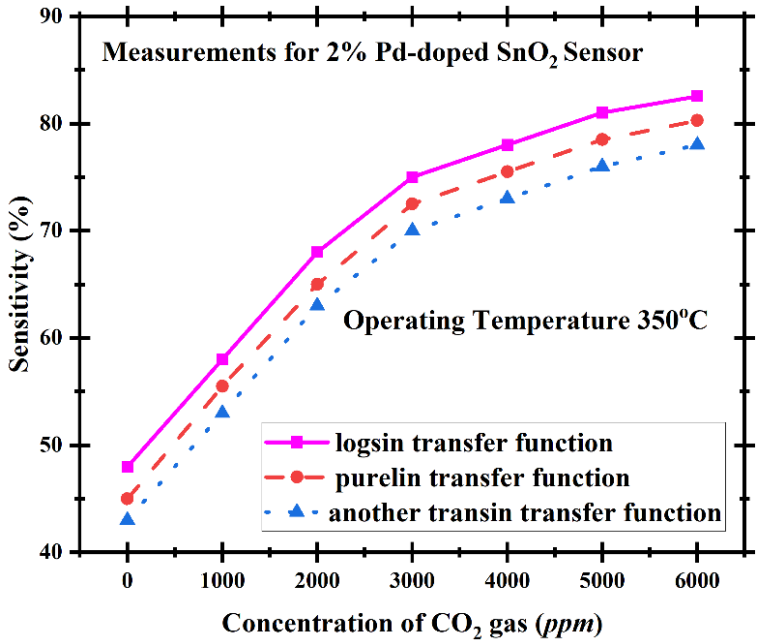

Fig. 5 Sensitivity measurements using three different transfer functions at 350°C for 2% Pd-doped SnO_2 Sensor with Random Weight /Bias Rule at 350°C.

In order to achieve optimum sensitivity of the Pd-doped SnO_2 thick film, measurements for 2% Pd-doped SnO_2 thick film as sensor were also done for detecting CO_2 at different concentrations ranging from $0 - 6000$ ppm at 350°C for three different transfer functions given in Table 3. The sensitivity data for 2% Pd-doped sensors, analyzed using the Random Weight/Bias Rule at $350°C$, exhibits a progressive increase in sensitivity as the concentration of the target gas increases, across all transfer functions (logsin, purelin, and another transin (TF)). Starting from a base sensitivity that ranges between 43% to 48%, the sensitivity values rise consistently, reaching up to approximately 78% to 82.54% at the highest tested concentration of 6000 ppm. This pattern suggests that the 2% Pd doping enhances the response of the sensor to gas concentration, which is crucial for effective detection in hazardous environments. The differences in sensitivity among the transfer functions highlight how the choice of activation function in the neural network model can affect the output of the thick film sensor, with each function offering varying degrees of linearity and response dynamics.

IV. CONCLUSION

The study undertaken focused on the optimization of ANN parameters to enhance the efficacy of SnO_2-based thick film gas sensor for detecting CO_2 gas. The research employed a comprehensive methodology that included dataset preparation, input and output variable selection, and ANN architecture optimization, followed by rigorous training, validation, hyperparameter tuning, and testing phases. Through experimentation, the study confirmed that optimizing ANN parameters significantly improves the accuracy and sensitivity of the gas sensors, which is crucial for applications in industries where detecting hazardous gases is imperative for safety and environmental monitoring. The results were particularly promising, demonstrating that the Levenberg-Marquardt feedforward propagation algorithm with an adaptive learning rate technique substantially outperformed other tested algorithms. This optimization not only provided a high degree of reliability in the response of the sensor but also showcased the potential of ANNs in refining sensor technology to achieve superior performance. Such advancements underscore the critical role of machine learning techniques in enhancing the functional capabilities of sensor-based technologies, thus offering robust tools for ensuring environmental safety and occupational health in various industrial settings.

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