International Journal of
INTELLIGENT SYSTEMS AND APPLICATIONS IN ENGINEERING
ISSN:2147-6799 www.ijisae.org Original Research Paper

Development of Discrimination Techniques for the Detection of Single and Multicomponent Gas Mixture using Tin Oxide (SnO2) based Sensor Array

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Submitted:12/03/2024 **Revised**: 27/04/2024 **Accepted**: 04/05/2024

Abstract: Tin oxide-based gas sensors have been widely used to detect single-target and multi-component gas mixtures in ambient atmospheres, utilizing sophisticated classification techniques such as Artificial Neural Networks (ANN). These techniques optimize pattern recognition, using sensor arrays formed by multiple commercially available sensors, like the TGS class from FIGARO, and Inc. The integration of ANN with these sensor arrays allows for the effective use of odor sensors, capable of processing data and extracting hidden information regarding the nature and concentrations of various gases, including toxic and residue components. The development and refinement of these discrimination techniques hold significant promise for advanced detection and identification of complex gas mixtures, potentially establishing SnO2-based sensor technology as a reliable and sophisticated method for gas analysis. Comparative analysis of discrimination techniques, including PCA, LDA, PLSR, SVM, and ANN, reveals that KNN regression outperforms gas concentration estimation.

Keywords: Tin Oxide, Pattern Recognition, Artificial Neural Network, Sensor Array

1. INTRODUCTION

As automation and worldwide connection grow, sensors will become more common, with gas sensors playing an important role in a variety of applications. [1]. Semiconducting metal oxide-based sensors are tiny, sensitive, long-lasting, and inexpensive [2], making them a popular choice for a variety of applications. Figaro Engineering Inc. developed the first commercially viable gas sensor in the 1960s, which used SnO2 [3]. SnO2 is frequently utilized in gas sensors, solar cells, photovoltaic devices, biological applications, and electrochemical processes [4]. Tin oxide is often employed in gas sensor applications because of its great sensitivity to reducing and oxidizing gases [5]. Nanostructured SnO2, with a particle size of less than 20 nm and a specific surface area of 100-200 m2/g, is especially promising for gas sensing and catalysis. The high surface-to-volume ratio of nanoparticles affects chemical behavior and increases their efficacy in heterogeneous processes [6]. The high adsorption and reactivity of nanostructured tin dioxide are related to the structure and concentration of surface active sites, notably oxygen vacancies and coordinately unsaturated tin cations [7].

Tin dioxide (SnO2), because of its high sensitivity, ease of use, and chemical and thermal stability, is a commonly used n-type metal oxide semiconductor (MOS) in gas sensing [8]. The gadget suffers from gas detection at ambient temperature, necessitating additional power and integration at higher working temperatures. Its usefulness in real-world applications is restricted by its inability to detect explosive gases at high temperatures, and its main flaw is a lack of selectivity [9]. Several tactics have been explored to overcome this, including functionalization, additives, particle size management, and chemical or physical filtration. It has been established that noble metal additions such as palladium, platinum, gold, and silver increase the sensitivity of SnO2-based gas sensors [10, 11]. MOS's capacity to detect single gases such as CO or NO2 has greatly increased, but its ability to discriminate between multiple gas species and recognize mixtures has not kept pace [12]. The MOS sensor's cross-sensitivity to different gases severely hampered its development. Nevertheless, distinct gases in a mixture cannot be identified separately by a single gas sensor device [13].

Research has indicated the possibility for qualitative analysis of gases; however, recommendations for analytical chemistry are frequently disregarded in these methods [14]. Consistencies in the variance of the training sample and an absence of a common boundary with clean air arise from the authors' frequent inability to combine points corresponding to different concentrations

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of analytics in a single area corresponding to the analyzed gas [15]. With successful adaptation, this procedure can be used to analyze data from multi-sensor electronic nose systems. Its original application was the problem of qualitative analysis of a gas mixture, and it aims to formalize that process [16]. Because typical scientific research is not focused on practical problems, despite the many publications on selective gas analysis using metal oxide sensors, these approaches remain far from commercial implementation [17]. This approach makes calibration devices for qualitative analysis of one-component systems challenging because it restricts the study to a discrete set of gas concentrations [18].

E-nose has been created using feature extraction and pattern recognition to address cross-sensitivity and discrimination across gas mixes [19]. In the past, sensor arrays were primarily concerned with gathering response values, which led to high costs and power consumption [20]. E-nose systems extract more characteristics from dynamic curves, which results in smaller sensor arrays and lower power usage [21]. This can help with gas identification by combining response and recovery time features [22]. Researchers employ dimensional reduction techniques such as principal component analysis, discrete wavelet transform, and Fourier transform to convert n-dimensional space into two- or three-dimensional projections [23, 24]. Data dimension reduction and gas classification are also achieved using artificial neural networks and unsupervised approaches such as KNN and linear discriminant analysis [25, 26]. Supervised algorithms, such as SVM and BPNNs, have been developed to improve gas identification accuracy [27]. Traditional data preparation for tin oxide gas sensors is complicated, necessitating initial feature extraction before pattern detection. Furthermore, SnO2 sensors frequently exhibit cross-sensitivity to multiple gases, reducing accuracy. Variations in humidity and temperature have an impact on performance as well.

The main contribution of the work is enumerated as

- High sensitivity and selectivity SnO2-based sensors have greatly enhanced the detection capabilities for both single gases and multi-component mixtures. For accurate and trustworthy gas analysis in a variety of industrial and environmental applications, this development is essential.
- The use of complicated algorithms such as ANN, PCA, LDA, SVM, and PLSR in sensor arrays improves gas detection and classification accuracy and usability.
- SnO2-based sensors are essential for gas analysis and detection in a variety of applications, including industrial safety, healthcare diagnostics, and environmental monitoring, due to their versatility and dependability.

The remainder of the paper is organized as follows: Section 1 provides an overview of tin oxide, Section 2 examines previous work, Section 3 details the technique analysis, Section 4 presents the work discussion, and Section 5 closes the study.

2. RELATED WORKS

Lee et.al [28] the work demonstrated Au-coated SnO2 NR gas sensors, which are very sensitive to VOC gases, notably BTXF. Heat treatment of SnO2 NRs increased their crystallinity, resulting in Au clusters on the sidewalls that serve as catalytically active sites for VOC oxidation processes. These highly organized arrays have a large surface area, are easily reducible, and have good mechanical strength, making them excellent for heterogeneous catalysts. The sensors demonstrated excellent sensing capability, with a high reaction time and a short response time of less than 2.5 seconds for detecting 10 ppm of each gas. The sensor was integrated into a mini-GC system to investigate the selective detection of each gaseous constituent in a BTXF mixture. However, due to the intricacy of the heat treatment technique and the usage of gold, au-coated SnO2 NR fuel sensors can be costly.

Kim et.al [29] the researchers used gas-flow thermal evaporation and atomic layer deposition to build a controlled porous structure for photoactive gas sensors. AR was introduced into the chamber during thermal evaporation to adjust the porosity of the SnO2 matrix. Atomic layer deposition was employed to manufacture nano-scale TiO2 layers on the surface of porous SnO2, which resulted in great sensitivity and rapid reaction. When exposed to CO at a concentration of 50 parts per million, the 0.2 Torr sensor demonstrated great sensitivity and reaction rate, as well as a low detection limit of 1 part per million. The inclusion of a TiO2 layer allowed for a 20% selectivity in response to HCHO. When exposed to CO, the SnO2 and SnO2@TiO2 heterostructure gas sensors had a moderate selective response to HCHO at 20%, showing decreased selectivity for particular gases. Their practical utility may be restricted due to their dependency on exact deposition pressures and UV light.

Li et.al [30] a cooperative strategy based on machine learning techniques and sensor integration provided for accurate NO2 and NH3 gas detection in mining environments. A wearable sensor array made of graphene and polyaniline composite has been created to increase sensitivity and selectivity in mixed gas settings. The partial least squares and backpropagation neural network methods show promise for real-world mining detection, with over 99% theoretical prediction accuracy on NH3 and NO2 values throughout a large relative humidity range. A wireless wearable wristband is designed to provide real-time warning of dangerous gases in mines under varying humidity circumstances. In practice, however, voltage acquisition resolution and Bluetooth transmission bit count may have an impact on calculation precision.

Verma et.al [31] the sensing behavior of Pd-doped SnO2 thick film sensors for LPG detection on alumina substrates was explored utilizing nonlinear ANN approaches. The sensors were made from a 1" x 1" alumina substrate and subjected to LPG gas. When exposed to 0.5% LPG, the sensors responded at their maximum rate of 72.08%. The sensors' sensitivity was evaluated using an ANN algorithm and two training techniques. However, because they are primarily optimized for single-gas detection, they may be unable to distinguish or detect additional gases in mixed-gas settings. Furthermore, their use in dynamic real-world contexts may be hampered by their dependency on precise operating temperatures and doping levels.

Tombel et.al [32] the study sought to develop effective approaches for extracting characteristics for machine learning algorithms that predict VOC categorization. Five supervised machine learning methods were used for a preliminary dataset, and 10 feature extraction approaches were utilized to classify three types of gases based on sensor response variations. The findings demonstrated that it is possible to categorize VOC chemicals based on sensor responses, despite gas sensor constraints such as limited sensitivity, selectivity, and signal noise.

Zang et.al [33] discussed SnO2, CuO, In2O3, and ZnO, distinct MOS Nano fiber, which were created using an easy-to-use single-needle electrospinning technique. The array exhibits high sensitivity to a variety of target analytic gases due to its smooth surface and constant nanofiber diameter (average of 150 nm). Four MOS nanofiber sensors yielded distinct response patterns, and a fabricated E-nose was used to measure five VOC gases related to human health. To decrease the dimension of the feature matrix, a PCA algorithm was used in conjunction with feature extraction from the response curves. As a result, the constructed E-nose system was able to distinguish between five distinct VOC gases. The main disadvantages of utilizing SnO2 in conjunction with other MOS nanofiber sensor arrays are potential problems with selectivity and cross-sensitivity in mixed gas environments, despite the high sensitivity and capacity to distinguish between different VOC gases. Furthermore, the system's computational requirements and complexity may increase if feature extraction and dimensionality reduction depend on sophisticated data processing methods like PCA.

As a result, Because of their intricacy and use of gold, au-coated SnO2 NR petrol sensors may be costly. Although highly sensitive, they require refinement in practical scenarios involving various gases. In mixed gas environments, SnO2 and SnO2@TiO2 gas sensors may not be able to distinguish between other gases despite having a moderately selective response to HCHO. Precise doping levels and operating temperatures may complicate their application. Selectivity and cross-sensitivity problems in mixed gas environments may arise when SnO2 is used in conjunction with other MOS nanofiber sensor arrays.

3. Advanced Gas Detection Using SnO2 Sensor Arrays: Discrimination Techniques Analysis

3.1 E-Nose Device

Hartman created the first scent analysis equipment in 1954, which consisted of a microelectrode and a mechanical sensor. Persaud and Dodd introduced the first intelligent artificial nose model in 1982, capable of recognizing up to 20 unique scents. Ikegami and Kaneyasu later created an integrated sensor with six metal oxide semiconductors.

Gardner and Bartlett invented the term "electronic nose" in 1988 at a pivotal moment, linking it to the biological sense of smell. Since then, this phrase has spread worldwide, emphasising the principles of technology. Notably, the electronic nose (e-nose) system is made up of several critical components, including a sensor array to record signals resulting from the interaction of sensing materials with volatile compounds, a sampling system to handle and store samples during analysis, and a computer to store, pre-process, and process data, as illustrated in Figure 1.

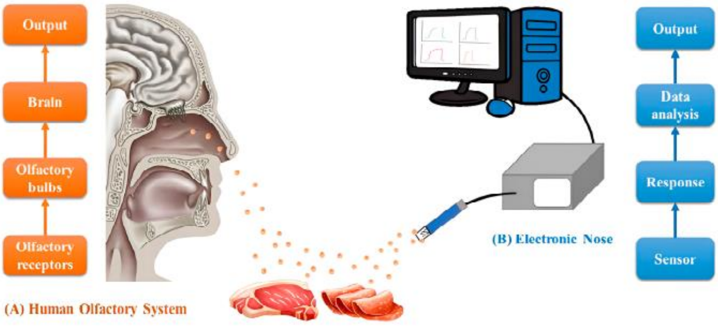

Fig 1: biological olfactory system (A) and e-nose technology (B)

Electronic noses were first presented in the 1990s by businesses like AlphaMOS, Neotronics, and Aromascan, and have since been commercially accessible. Because of its potential uses, the market for these systems has risen fast, resulting in the development of a wide range of devices utilizing diverse sensor technologies. Some e-nose systems that were formerly commonly used in books are no longer manufactured. Research and development have continued to create equipment that mimics human taste buds, such as Otto and Thomas' 1985 liquid sample analysis system. This novel device represents a shift in taste-sensing technology. E-nose technology has also been investigated for uses other than odor analysis, including respiration analysis for illness diagnosis, food quality assessment, environmental tracking, and security and defense. This diversification highlights e-nose technology's versatility and effectiveness in numerous situations, paving the way for its widespread usage in a variety of sectors.

The gas sensor chamber's adjustable shutter enabled simple exposure to volatiles. A pneumatic system was employed to keep the sensors clean in between readings. The nose was made up of 2 primary parts: the sensor probe and the main electrical device that linked to the computer. Figaro Co. of Japan produces a variety of metal oxide sensors. The primary device, an AT mega 328P-PU microcontroller, controlled the communication between the sensors and the computer.

MOX sensors demand temperatures in the hundreds of degrees Celsius, which necessitates inbuilt heaters that require 5 volts of electrical power. These sensors rely largely on heater voltage settings, which may be regulated by an electrical circuit to modify the sensor's heating voltage and maintain it at the required level. These changes can be performed during each sensor reading cycle.

Table 1: List of sensor models used in our electronic nose device and target odors and gases.

Sensors	**Target Gas Detection**
TGS 2600	The sensor is extremely sensitive to tiny amounts of air pollutants such as hydrogen and CO2 in cigarette smoke, detecting hydrogen at levels of just a few ppm.
2600	The sensor is extremely sensitive to minute amounts of gaseous air pollutants, such as and can detect hydrogen at levels of a few ppm.
2602	The sensor is very sensitive to low quantities of odorous gases and VOCs, such as ammonia and H2S, from waste items at the workplace and at home.
2603	The sensor is extremely sensitive to trace amounts of odorous gases and VOCs, such as ammonia and H2S.
2610	Uses filter material in its housing to eliminate the impact of interference gases like alcohol, resulting in a highly selective reaction to LP gas.
2611	Uses filter material in its housing to reduce the impact of interference gases like alcohol, resulting in a highly selective reaction to methane gas.
2612	The sensor is appropriate for monitoring methane, propane, and butane in LNG and LPG.
2620	It is very sensitive to organic solvents and other volatile vapors, making it excellent for use in organic vapor detectors and alarm systems.
2630	The device is extremely sensitive to R-404a, and R-410a, and it employs filter material to remove interference gases.

The electronic nose detects sensor reactions using MOX sensors, which measure the change in conductance during a short response. The sensor conductance is represented by the symbols $U/U0$ or $G/G0$. The reading cycle is repeated every 0.75 seconds, and the sensors experience transitory situations such as gas composition changes and heater voltage variations. 500 measurements were performed using a rectangular voltage drop/rise of 0.3 V.

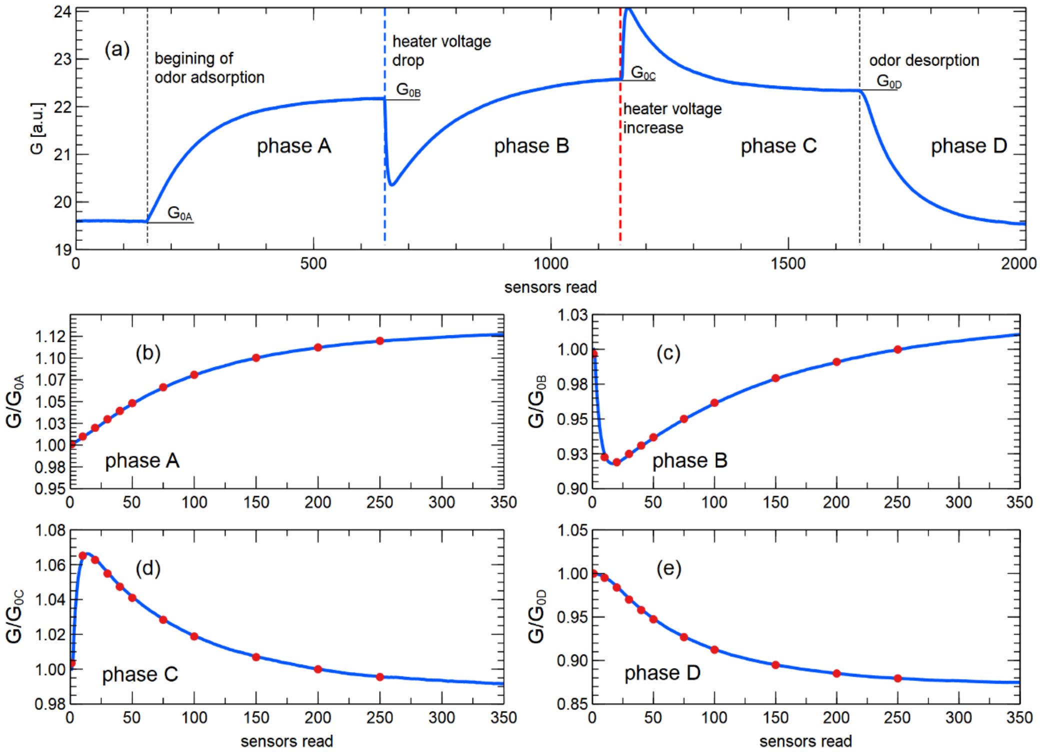

Fig 2: Curve during a measurement cycle

3.2 Discrimination Techniques

Electronic noses, often known as the "brain," are extremely important in the field of detection. They use visualization techniques and algorithms to achieve discriminating and selectivity in sensor array responses. The success of these systems is dependent on processing and synthesizing responses from numerous sensors, such as signal filtering, switching, and feature extraction. Data matrices are optimized, and the signal passes through a second round of pattern recognition analysis. The system learns the properties of the examined scent through training and predicts samples of unknown concentrations using a predetermined model.

3.2.1. Principal Component Analysis

PCA is a popular approach in sensing for data reduction in multivariate statistics. It turns the first N variables in a dataset into a new N-dimensional coordinate system, projecting the variable with the largest variance onto the principal axis. Created for huge datasets, it is now widely used in e-noses for qualitative categorization. The visualization of tiny variables in two or three dimensions facilitates qualitative categorization. However, because it relies on human interpretation, PCA is arbitrary and susceptible to bias. To promote impartiality, alternative categorization approaches have been created, which apply labels to fresh data points through algorithmic autonomy.

3.2.2. Linear Discriminant Analysis

That maximizes variance ratios across and within classes using linear combinations of original variables. It intends to project datasets into lower-dimensional spaces to decrease computational complexity and overfitting problems while enhancing class distinguishability. LDA, like PCA, is a linear transformation approach for dimensionality reduction, and it is equivalent in terms of dimensionality reduction, visualizations, and classification reliability. It improves differentiation by increasing inter-class separation and variance, making it simpler to split space into sections associated with certain classes. LDA is a supervised approach that creates models from a training dataset and compares them to fresh data, assuring accurate classification while eliminating subjective interpretation.

3.2.3. Partial Least Squares Regression

PLSR is a method that combines the best aspects of MLR and PCA to analyze or predict dependent variables using independent variables. It eliminates orthogonal elements from latent variables, resulting in the most powerful prediction insights. PLSR is especially beneficial for predicting dependent variables from a variety of independent variables, controlling multicollinearity, and understanding underlying patterns and connections.

3.2.4. Support Vector Machines

Its goal is to maximize the separation of distinct categories by projecting labeled data points onto an N-dimensional space and assigning each new data point a category based on its position.

SVM handles two classes at once and determines the optimum hyper plane to partition them. If a basic hyper plane is insufficient, SVM computes the greatest distance between data points. If a separate hyper plane is not possible, SVM employs nonlinear transformations via kernels to enhance data dimensionality, allowing effective classification without data modification.

SVM, a machine learning algorithm, tries to minimize mistakes by generating a hyper plane with $N - 1$ dimensions, maximizing the margin, and considering error tolerance. It can generate remarkable results in both classification and quantification problems by combining its classifier and regressor capabilities.

3.2.5. Artificial Neural Networks

ANNs mimic data processing and problem-solving capabilities of the brain. ANNs, which are made up of interconnected processing elements, are very good at handling nonlinear, complex data. They have three layers: input, hidden, and output. Nodes in them send signals that help in decision-making. The 1943 concept by McCulloch and Pitts served as the inspiration for ANNs, which were later used in electronic noses.

ANNs process input in backward-pass and forward-pass stages, modifying synaptic weights to minimize mistakes. They excel at supervised learning when trained on labeled data using methods such as the vanishing gradient approach.

ANNs in electronic noses use labeled data to identify patterns and generate precise predictions. Their ability to effectively capture and analyze intricate, multi-dimensional data is what makes them so effective in the varied and variable environments in which electronic noses are used.

KNN-based regression beat other current models, including ANN, RF, Decision Tree, and Linear Regression. The study sought to enhance the KNN's effectiveness in predicting gas concentrations in mixtures by selecting ideal parameters. The most successful alternatives for each dataset were distance weighting, the Euclidean distance measure, and the five nearest neighbors. Analyzing the algorithm's performance using these optimal parameter settings yielded promising findings. The model had one neuron on the output layer, six hidden layers, and a linear activation function in that layer. The analysis discovered that the best parameter combinations each gas mixture lowered the mean square error while increasing the coefficient of determination.

Linear regression describes the connection between dependent and independent variables, whereas decision tree regression employs a tree-like model to predict outcomes. However, these models may require greater accuracy and are susceptible to overfitting. Random forests combine decision trees to improve performance while reducing overfitting. Each tree is resistant to noise and outliers, can manage missing data, and offers a feature significance rating. However, they are computationally expensive for huge datasets and perform badly on imbalanced ones.

4. Discussion

Using machine-learning methods, the researchers attempted to recreate greenhouse gas concentrations and variability from low-cost sensors. Each substance was tested over three days, with setup periods lasting more than two hours. The electronic nose gathered sensor data and sent it to a computer for real-time visualization. The researchers studied field calibration strategies for low-cost sensors such as metal oxide, electrochemical, and tiny infrared sensors. They determined that artificial neural networks (ANN) were the most effective calibration approach and that employing a range of sensor types might aid in the resolution of cross-sensitivity concerns. However, due to the high cross-sensitivity to water vapor, the signal could not be correctly reconstructed using simply Figaro® TGS sensors.

In research comparing FFNN and DNN for calibrating trace gases, DNN was shown to be more accurate at recreating significant concentration fluctuations. The study also discovered that changing the temperature of a MOS sensor improves electronic nose evaluation characteristics, such as sensitivity, selectivity, noise reduction, and range. The sensor's resistance to temperature is inversely proportional and linear, and temperature varies exponentially with heating voltage. The study investigated four unique TM patterns and used them to a personalized electronic nose for lung cancer patients.

The study examined three analyses at varying concentrations: methane, CO_2, and butanone. The results indicated that unique TM patterns increased selectivity and concentration distinction. Combining square and triangular patterns with k-NN data clustering produced the best results. The TGS2600 sensor class demonstrated superior discrimination in both analyses and concentrations, demonstrating the advantages of square-granular architecture.

The pre-heating phase time was consistent with the datasheet recommendation, although drifts were seen in the AS-MLV-P2, whilst the four TGS2600 and two TGS822 achieved a steady baseline. This might explain the inconsistent results from the S-7 and S-8 sensors. The BME260 sensor measured temperature, humidity, and pressure. Following the wash-out phase, the humidity levelled off at 10% to 13%. Manufacturers give sensor resistance changes dependent on temperature and humidity, however they are beyond the practical range. Future studies will look at humidity's influence and build compensatory methods. The current study's findings are consistent with earlier studies demonstrating the benefits of temperature modulation in MOS sensors.

A sensor (SP3-AQ2) was used to detect 12 airborne analyses that had different TM patterns. The amounts tested were higher than the usual VOC concentrations in exhaled breath. Accurate temperature modulation, wavelet transforms, and machine learning-based categorization improved gas identification accuracy. One study reported 92% compound discrimination accuracy for analyses such as methanol, ethanol, and butanone. Recent tests with commercial MOS gas sensors demonstrated the feasibility of complex data processing based on support vector machines, as well as simultaneous gas categorization and concentration prediction using TGS2620 and TGS2602 sensor types.

5. Conclusion

The development of tin oxide represents a significant advancement in the detection and analysis of both single-target gases and complex multi-component gas mixtures in ambient atmospheres. By integrating advanced discrimination techniques such as Artificial Neural Networks (ANN) with sensor arrays, these sensors can effectively process and interpret complex data to identify and quantify various gases, including toxic and residue components. The use of commercially available sensors, like the TGS class from FIGARO, USA, Inc., has further enhanced the reliability and accuracy of these systems. Comparative analysis of various discrimination techniques, including PCA, LDA, PLSR, SVM, and ANN, has shown that KNN regression provides superior performance in estimating gas concentrations. These findings suggest that with continued refinement, SnO2-based sensor technology, coupled with sophisticated data analysis methods, holds great promise for advanced gas detection and identification, establishing it as a robust and sophisticated tool in the field of gas analysis. Future research should focus on addressing environmental factors such as humidity to further improve the accuracy and reliability of these sensors.

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