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Quantitative recognition of flammable and toxic gases with artificial neural network using metal oxide gas sensors in embedded platform

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ABSTRACT

Artificial Neural Network (ANN) based pattern recognition technique is used for ensuring the reliable evaluation of responses from an array of Zinc Oxide (ZnO) based sensors comprising of pure ZnO nanorods and composites of ZnO–SnO₂. All the sensors were fabricated in the lab. The paper first reports the development of an artificial neural network based model for successfully recognizing different concentration of hydrogen, methane and carbon mono-oxide. Feed forward back propagation neural network was used for the classification of the gases at critical concentrations. The optimized ANN algorithm is then embedded in the microcontroller based circuit and finally verified under lab conditions.

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1. Introduction

Qualitative and quantitative detection of flammable and toxic gases like hydrogen (H₂), methane (CH₄) and carbon mono-oxide (CO) in coal mines, petroleum industries and gas storage plants are of great importance for the safety of workers, health hazards and to avoid risk of accidental explosion. H₂ is a potential source of energy of future and is used in fuel cell [1], H₂ engine cars, chemical industry etc. H₂ is a colorless, odorless and highly explosive gas having flammability in the range (4–75%) in air [2]. In coal mines presence of methane is always expected, which has caused most loss of life than any other gas. Methane in the range (5–14%) in air is flammable [2]. CO is also a colorless and tasteless gas which is believed to be the most dangerous gas in mine. CO in the range of 50 ppm is fatal and is also flammable and explosive [2] in higher range of concentrations.

Oxide based semiconductor sensors present advantages like high sensitivity to low concentration of gases, less fabrication cost, long term stability, low power consumption etc. which makes them

attractive material for the detection of toxic, hazardous and combustible gases. A wide range of sensors based on semiconductor metal oxides including ZnO, TiO₂, SnO₂, WO₃, In₂O₃, etc. were used for the detection of gases like H₂ [3], CH₄ [4], LPG [5], CO [6] etc. Both ZnO and SnO₂ are n-type wide band semiconductors which have attracted remarkable attention as highly sensitive sensor material. These material were expected to be superior material for sensing gases like H₂ [7,8], CO [9,10], and CH₄ [10,11]. In spite of the high response, metal oxide semiconductor (MOx) based sensor presents a serious drawback of poor selectivity, which hinders their commercial application. Therefore, MOx sensors fall short when a precise detection of specific concentration of gaseous component is required or when they are used to discriminate a mixture of gas. Drift in the sensor output due to ageing, poor stability or high sensitivity to humidity of the ambient environment are also serious drawbacks of metal oxide based sensors. Both drift and poor selectivity make the sensor output unreliable [12].

Many authors have suggested various ways to overcome the problem of cross-sensitivity to achieve selective sensor which may include, a) addition of noble catalytic metal to promote the reaction to specific gas [13], b) operating the sensor in dynamic mode [14], c) surface modification [15] and d) use of multi-compositional sensing film [16,17]. However, extremely selective sensors are very

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expensive and operating the sensor in dynamic mode makes the measurement system complex. Although, the problem of cross-sensitivity can be addressed with any of the above mentioned ways, the troubles of reliability due to the drift in sensor output still persist. A potentially more cost effective way to perform such measurement is to use a sensor array coupled with a pattern classification system [18–20]. The use of multiple sensors with an appropriately modeled pattern classifier is therefore expected to improve the reliability in the recognition of specific concentration of gaseous components. This will also improve the selectivity for a particular gas when there is a mixture of gases.

Various pattern analysis methods like principle component analysis (PCA) [21], partial least square analysis (PLS) [22], and artificial neural network [23] etc. can be used with gas sensor arrays for the classification and identification problem. Among these methods artificial neural network is reported to provide superior performance when quantification or multicomponent mixture classification is required [24]. Multilayer perceptron (MLP) and radial basis function (RBF) are the two commonly used neural network algorithms used for pattern recognition. However, the neuron connectivity in RBF is comparatively lower than MLP. This is the reason why the mean square error of an RBF network is less compared to an MLP one having the same number of neurons. Therefore the parameters for tuning the network are higher in MLP. MLP with nonlinear transfer function, like sigmoid function, allow the network to learn any input–output relationship adjusting the weights and biases in the network by a gradient descent of technique known as back propagation of errors. Many authors have reported the successful utilization of ANN in many applications including pattern classification, identification, decision making etc. [25–28]. Multilayer perceptron neural network have been used by Joo et al. to identify breast nodule malignancy using sonographic images [26]. Lv et al. reported the detection of low concentration of formaldehyde in binary gas mixtures using micro gas sensor array and a neural network [27]. Lee et al. reported about a combustible gas recognition system including sensor array and neural network based pattern recognition [28].

Most of the commercially available gas recognition systems are PC based and hence are bulky. Embedded systems can be developed with a suitable training function which can automatically read the sensor input from the sensor array and produce the neural network output. This may overcome the drawback of PC based gas pattern recognizer, making them portable and convenient to the user. In this work, we report the development of a prototype toxic gas recognizer using sensor array and multilayer neural network. The sensors were based on pure ZnO nano-rods and ZnO–SnO₂ composites which were fabricated in the lab. The sensors were exposed to H₂, CH₄ and CO in different concentration ranges. The data generated from the sensors were used to train a multilayer neural network with back propagation learning algorithm, which can reasonably produce output to unseen input data. A process flow chart for realization of the ANN in microcontroller is also formulated and verified by downloading the network in the microcontroller. The use of an array of sensors instead of one or two along with a suitably trained neural network can increase the reliability of the measurement.

2. Sensor fabrication

The sensor used in the experiment consists of pure oxide of zinc as well as composite oxide of zinc and tin, as the active gas sensing element. For the fabrication of sensors separate sols were prepared for pure ZnO and composite ZnO–SnO₂ with polyvinyl (PVA) as capping agent. The detail material synthesis and methodical characterization of the sensor materials were reported in our previous

communications [29,30]. In a typical procedure, sol for pure ZnO is prepared by mixing 0.6 g of zinc acetate di-hydrate [Zn(CH₃COO)₂·2H₂O] with aqueous solution of PVA under constant stirring. The 1.0 M sodium hydroxide (NaOH) solutions was then mixed drop wise to the above solution until a pH value reached 7.0. The solution was then stirred at 80 °C for 1 h. Sol for ZnO–SnO₂ sensor is prepared by mixing appropriate amount of zinc acetate dehydrate (Zn(CH₃COO)₂·2H₂O) and stannic chloride (SnCl₄·5H₂O) with PVA. 1.0 M NaOH solution was used as reducing agent until pH of the solution becomes 7.0. The final solution was stirred at 110–120 °C for 1 h. Finally, the resulting precursors were spin-casted on the cleaned <100> oriented Si/SiO₂ substrates followed by annealing at 550 °C for 30 min in a tube furnace.

3. Measurement techniques

A total of six numbers of sensors were used for the collection of data generated from the sensors when exposed to three different gases namely hydrogen, methane and carbon monoxide. The sensors were exposed to each test gas in different concentrations (0.1, 0.3, 0.5 and 1.0%) separately. The sensor types and their operating conditions are given in Table 1. The detail arrangement of the sensor measurement set up and sensor structure were reported elsewhere [29,30]. Typically, the sensors were placed inside a closed test chamber with suitable arrangement for gas inlet and outlet. Measured amount of test gas was fed to the test chamber using Alicat[®] mass flow controllers (MFC). The test chamber was fitted with heating arrangement that would maintain a uniform temperature (with an accuracy of ±3 °C) inside the chamber. The sensors were designed to operate in a resistive mode and Fig. 1 shows a model of the sensor measuring circuit. Pd–Ag contact electrodes were made on the top of the sensing layer by e-beam evaporation technique in order to provide electrical connection. An electrometer (Agilent U1253B) was used to monitor the variation of sensor resistance. The resistance variation of the sensor is converted into voltage output by the measurement circuit.

4. Design of ANN in matlab[®]

A feed forward multilayered neural network (Fig. 2) trained by a gradient descent technique, also called back propagation of errors, was used to process the data arising from the sensors to investigate the feasibility of discrimination of gases. The network was designed in the Neural Network toolbox in MATLAB[®] with six input neuron equal to the number of sensors used, two hidden layers with ten and twelve neurons in the hidden layer 1 and 2, respectively, and thirteen output nodes corresponding to a specific concentration of a specific gas, as shown in Table 2. Sigmoid function was used as the activation function for the hidden and the output neurons. Training of the network was performed using supervised back propagation learning algorithm which was first initialized with random weights and biases. During the training process, weights and biases of the network were iteratively adjusted in order to minimize the

Table 1
Sensor type and operation temperature.

Sensor	Sensing material	Operating temperature (°C)
S1	ZnO	150
S2		200
S3		250
S4	ZnO–SnO ₂	150
S5		200
S6		250

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