



Integrated Approach to the Optimization, Synthesis, Fabrication, and Application of ZnO-Based Sensors for Portable LPG Leakage Detection Systems

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Received November 20, 2023

Revised March 22, 2024

Accepted April 15, 2024

Published online: 28 June 2024

Keywords

ZnO

Sensor

Energy

Metal Oxide

LPG Gas

Material Science

Abstract: Liquefied petroleum gas (LPG) is used for cooking as well as in vehicles in the form of compressed natural gas (CNG) and in industries. As gas is volatile, there is the possibility of leakage which will be turned into take explosion or fire. It is a keen need to use the detection system for gas leakage for security purposes. ZnO thick film sensor was synthesized and fabricated by using the standard screen-printing method. The XRD pattern revealed that the ZnO thick film was polycrystalline, with an average crystallite size of around 27.1174 nm. The SEM scan revealed a ZnO particle size of around 1.88µm. Additionally, electrical and thermal properties were examined. This paper discussed the synthesized ZnO thick film LPG gas leakage detector prototype using Arduino ATMEGA8.

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1. Introduction

The main source of fuel in this era is liquefied petroleum gas (LPG) not only in urban areas but also in rural areas. Compared to traditional fuel material LPG is clean and easy to handle. Because of the different government schemes most of the families in the rural areas benefited from using LPG. Today gas sensors are marked and demanded chemical sensors (Shankar and Rayappan 2015). Gas detection and indicators are widely needed in industries, industrial safety, monitoring and controlling environmental conditions, and processing control. The change in any one or more properties for instance resistance, temperature, humidity, etc. in the presence of a gas environment identifies the detection of gas leakage (Sowmya et al.

2021). The working of a metal oxide-based thick film gas sensor is based on the reaction of gas molecules with the surface of a thick film (Gusdevi et al. 2021). In this Oxygen, ions absorb on the surface by removing the electrons from bulk material and creating the barrier which limits the electron movement and results in the conductivity of the sensor (Hussain et al. 2017). For the detection of gas by metal oxide semiconducting material smaller grain size, stability, and good structural and electrical properties are the most important parameters (Bhavithra et al. 2021). The structural and electrical properties can be changed and improved by adding the additive and or dopant material (Ménil et al. 2005). Metal oxide materials are widely used for different applications such as solar cells, sensors, supercapacitors, optoelectronics, cosmetics,

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ENERGY AND THERMOFLUIDS ENGINEERING | ETE | ISSN 2716-8026 (PRINT)

Available online at <https://asps-journals.com/index.php/ete>

<https://doi.org/10.38208/ete.v4.775>

piezoelectric, etc. (Naeem et al. 2023). For commercial purposes, LPG is considered fuel in all countries (Devi et al. 2006). Today LPG is vastly and primarily used in different applications, mostly for cooking and recently in vehicles since it is clean and convenient (Shinde et al. 2007). The LPG gas is stored as a liquid at high pressure and low temperatures thus the gas converts into liquid in the cylinder which is use for cooking purposes (Arshak and Gaidan 2005). LPG means Liquefied Petroleum Gas having a mixture of different hydrocarbon elements and originating from natural gas (Singh et al. 2023). For commercial use, LPG is stored in a cylinder whereas the LPG storage area uses strong and not easily leak gas tubes (Patil et al. 2023). But we know that LPG is a flammable gas so the main issue of the leakage problem of LPG is a serious problem for home security and residential area. Many commercial gas sensors are available in the market. Out of that MQ series metal oxide-based gas sensors are used widely. Use of MQ gas sensor for detecting different reducing gases like LPG, smoke methane, and butane directly or with Arduino from 200 to 10000 ppm gas (Singh and Yadav 2023). This gas sensor cannot identify which gas was leaked, if a mixture of gas is present in the air, it detects the gas having the highest ppm. Similarly, most of the research was done on metal oxide semiconducting-based gas sensors (Reshma et al. 2023). The adsorption properties of the gas sensor change with respective changes in temperature and simultaneously noted the change in resistance value (Ingale et al. 2023). The sensitivity of the gas sensor depends on the gas molecules distributed on the surface of the sensor material, the morphology of the surface of the sensor, the grain size of the material, the surface area, etc (Panwar et al. 2023). The oxide materials like tin oxide, iron oxide, zinc oxide, cadmium oxide, aluminum oxide, molybdenum oxide, nickel oxide, and tungsten oxide, were studied for a long time as gas sensors. However, these oxide materials are sensitive to gas at very high temperatures (Shkir, 2023).

Zinc oxide (ZnO)-based gas sensors have become increasingly popular due to their rapid response, low detection limits, high selectivity, reliable performance, and cost-effectiveness. ZnO offers additional advantages such as biocompatibility, chemical stability, environmental friendliness, and low synthesis costs, making it a focal point in research for detecting toxic and harmful gases. A comparative analysis of a developed ZnO sensor prototype with available ones would involve evaluating sensitivity,

selectivity, response time, detection limit, stability, reliability, manufacturing cost, environmental impact, and long-term performance. This analysis helps researchers understand the strengths and weaknesses of the developed sensor and identify areas for improvement, ultimately advancing the field of gas sensing technology. The comparison aims to highlight the prototype's performance against existing sensors, showcasing its potential for practical applications. Researchers analyze various parameters, including sensitivity, selectivity, and response time, to gauge the prototype's effectiveness. By addressing these aspects, the study contributes valuable insights to enhance ZnO-based gas sensor technology.

Nowadays, interfacing of commercially available gas sensors with Arduino is widely used for sensing various gases and indicating the leakage of gas. Many researchers continuously work on thin/thick film semiconductor material gas sensors and try to improve the sensitivity, response, and recovery time but very few used the material for a working model. In this paper ZnO is used as a gas sensor prototype for LPG gas sensing using Arduino has been discussed. The advantage of this prototype system is that we can change the thick film sensor as well as the operating temperature (less than or equal to 300 °C) for the different gases as studied in characterization.

2. Experimental details

The methodology includes different stages, an experimental technique for the synthesis of ZnO powder, fabrication of thick film sensor, structural characterization of thick film sensor, electrical and gas sensing characteristics of thick film, and prototyping of ZnO thick film using Arduino.

2.1. Synthesis of ZnO

The ZnO powder was prepared by using the high-temperature self-combustion sol-gel method (Nehru et al. 2016). In this method, the precursors are Zinc nitrate hexahydrate as a source material and Dextrose as a fuel was used. The mixture of source material and fuel was dissolved in water and kept on a hot plate. As the solution becomes gel-type it was kept in a pre-heated muffle furnace at 550 °C for 5 to 10 minutes. The solution boils, ignites, and after completion of the reaction yields an amorphous-like powder. The powder was calcinated at 650 °C to get pure ZnO. The XRD of the powder proves the formation of ZnO.

2.2. Fabrication of ZnO thick films

The ZnO thick films were fabricated using a well-known screen-printing technique. For making the paste and for fabrication the ratio of organic to inorganic material was 70:30 throughout the work. For the formation of proper paste, in inorganic material ZnO powder and glass powder as a permanent binder were used. Whereas the inorganic part contains Ethyl cellulose as a temporary binder and Butyl Carbitol Acetate as a vehicle. The prepared paste was printed on the alumina substrate using the screen-printing technique. After drying the films in air and under an IR lamp the films were fired at 700 °C for 1 hour to remove the organic vehicle and stick the film material evenly on the surface of the alumina substrate. To decrease the wake-up time of the sensor of thick films one can mix the smaller amount (1 to 10%) of the different oxide material additives like Al_2O_3 and TiO_2 are added to the ZnO powder (Lamdhade 2015). And then analyze the electrical and gas sensing characteristics of the sensor films. The flow chart of the fabrication process shows the detailed method. Fig. 1 shows the block diagram of the process of fabrication of ZnO thick films.

3. Results and Discussion

3.1 Structural and morphological analysis

The structural characteristics of ZnO films were studied using X-ray diffraction (XRD) and morphology characteristics were studied using a scanning electron microscope (SEM). ZnO thick film was prepared at 550 °C

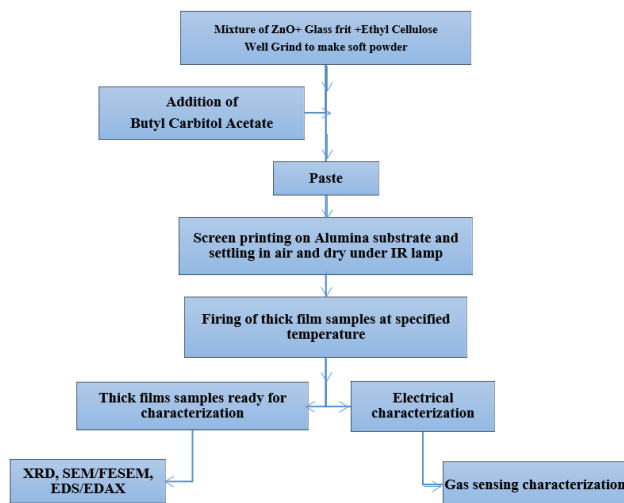


Fig. 1. Block Diagram of preparation and fabrication of thick film samples.

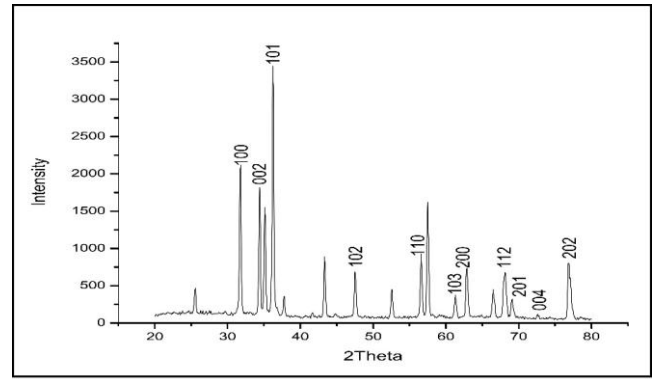


Fig. 2. XRD pattern of ZnO thick film.

temperature. Fig. 2 shows the XRD pattern of ZnO thick film.

The nature of the ZnO thick film was polycrystalline from the XRD pattern. To find the crystallite size Scherrer's formula was used below Eq. (1).

$$\text{Crystallite size (D)} = \frac{k\lambda}{\beta \cos\theta} \quad (1)$$

Where λ is the wavelength of X-ray radiation and β is the FWHM.

Fig. 3 shows the SEM image of the ZnO thick film. The grains were agglomerated with space in between. The ZnO particle size of about 1.88 μm was observed by SEM image. The specific surface area of the spherical particles of ZnO thick film material was derived by using the following Eq. (2).

$$S_w = \frac{6}{\rho d} \quad (2)$$

Where ρ the density and d is the diameter of the particles.

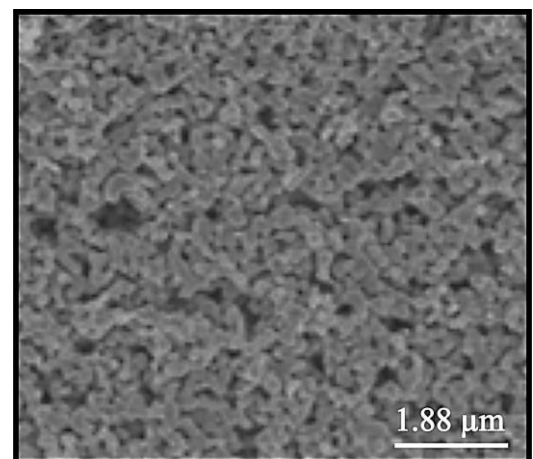


Fig. 3. SEM image of ZnO thick film.

Table 1 Crystallite size from XRD and Particle size, specific surface area from SEM.

Sr. No.	2θ	Plane (hkl)	Observed Values				
			2θ	d(A°)	Intensity (I/I ₀)	FWHM (β)	D (nm)
1	31.78	100	31.80	2.81	62.50	0.2553	34.1529
2	34.44	002	34.41	2.61	53.71	0.2551	34.4118
3	36.26	101	36.26	2.48	100.00	0.3130	28.1908
4	47.55	102	47.48	1.92	20.21	0.3023	30.3037
5	56.62	110	56.57	1.63	27.60	0.3600	26.4517
6	62.88	103	61.27	1.51	11.71	0.3375	28.8766
7	63.40	200	62.84	1.48	22.10	0.3867	25.4112
8	67.98	112	66.48	1.41	12.92	0.3946	25.4082
9	69.12	201	68.15	1.38	20.60	0.5101	19.8468
10	72.59	004	69.08	1.36	8.95	0.4183	24.3368
11	76.98	202	76.84	1.24	23.64	0.5121	20.9004

Table 1 contains the standard values of XRD data of ZnO from JCPDS 36-1451 card and calculated values of 2θ, d, FWHM, and peaks (hkl). The estimated crystallite, particle size, and specific surface area from XRD characterization. The average size of crystallite is about 27.1174 nm.

3.2 Electrical and Thermal Characterization

The electrical and gas sensing characteristics of prepared ZnO thick film were studied in the presence of air and LPG gas environments with respective changes in temperature. As the temperature rises, the decrease in sensor resistance takes place because of desorption of hydroxyl groups indicates the increase in sensitivity (Nehru et al. 2016). The gas sensing characteristics were observed in the presence of 1000 ppm LPG gas. Figure 4(a) shows the variation of resistance of prepared ZnO thick film concerning the change in temperature from room temperature to 400 °C. As the temperature increases the resistance of ZnO film decreases. Also, it was clear that the resistance of the film was lower in the presence of a gas environment. Fig. 4 (b) shows the graph for finding the operating temperature. The operating temperature is the temperature at which the thick film sensor senses the gas and changes its resistance. The operating temperature was calculated using the ratio of the resistance of the film in the air to the resistance of the film in a gas environment.

The ZnO gas sensors require a higher temperature (between 200 °C to 400 °C) to sense and respond to the reducing or oxidizing gases is the main disadvantage. Figure 5 clearly shows the two different peaks at temperatures 170 °C and 240 °C with the sensitivity of 3.5 and 4.29, respectively. The sensitivity increases with an increase in temperature.

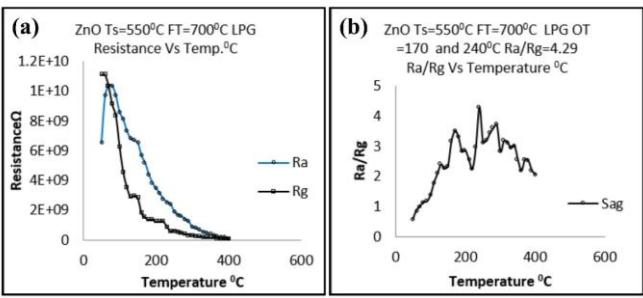


Fig. 4. (a) Electrical characteristics of ZnO thick film in the presence of air and LPG gas (b) Response curve of Film resistance V/s Operating temperature.

3.3 ZnO thick film-based LPG gas sensor prototype

As metal oxide-based gas sensors work at higher temperatures the heater and its temperature are the most important parameters while designing a prototype gas sensor. In this study, a Nichrome wire wound around a ceramic core having 1Ω resistance was used as a heater with a high power rating from 50 to 100 watts. The size of the heater is 10 X 10 X 30 mm. The Maximum working surface temperature of the heater is up to 400 °C. To power up the heater 12V power supply was provided. To control and maintain the operating temperature thermocouple is used. The output of the thermocouple is connected to the ADC through the AD595 instrumentation amplifier. The Atmel ATmega8A board is used to interface and to show the result on an LCD screen. Figure 5 shows the circuit diagram of ZnO thick film-based LPG gas sensor prototype.

This circuit can be used for different types of thin/thick metal oxide-based gas sensors. The sensor film can be removed and replaced for testing of different gases. Once the heater is ON, it increases the surface temperature so that ZnO thick film temperature. As soon as the heater

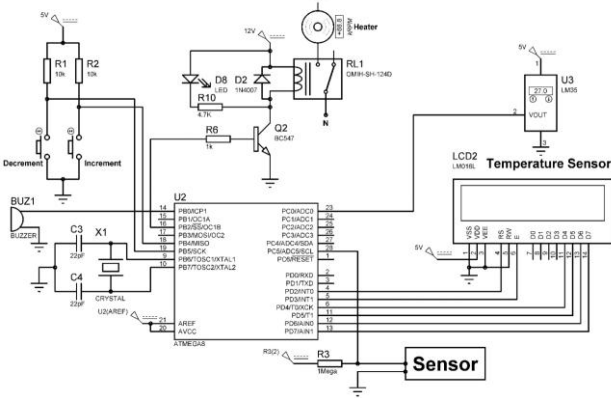


Fig. 5. Circuit diagram of ZnO thick film-based LPG gas sensor prototype.

temperature reaches the operating temperature heating stops and maintains the temperature. Buzzer used to indicate the heater temperature reached up to operating point temperature. Once this buzzer is ON, the circuit is ready for testing gas leakage. The buzzer starts only once initially when the circuit is powered up and the heater starts to heat up to the operating point temperature after that control circuit maintains the operating temperature and heater ON and OFF as per requirement. It takes different times for different operating temperatures. It takes around 4 minutes to reach a temperature of 150 °C. At this temperature around 50 mW power is required to the circuit. After exposure of LPG gas, the surface of the sensor reacts with LPG result to drop the resistance of sensor gradually.

The change in the sensor resistance is displayed on the LCD screen indicates the leakage of gas. This sensor circuit i.e prototype gas sensor works for different type of thick/thin films at variable operating temperatures by simply exchanging the first sensor film with the other sensor film. The maximum temperature of the heater is 300 °C so one can use it for an operating point up to 300 °C. Fig. 6 (a) shows the sensing circuit with display (portable prototype sensor kit) Fig. 6 (b) shows the actual circuit's internal connection with Arduino and Portable prototype kit.

Figure 7 (a) and (b) shows the complete prototype sensor under testing condition. The screen displays the heater's real-time temperature means sensor temperature by the word 'TMP', the operating temperature of the sensor by the word 'SET', and the resistance of the sensor by the word 'REST'. Two push buttons are used for setting the operating temperature as per requirement one for increasing and the other for decreasing the set temperature. After power on the circuit first set the operating temperature and the power on the heater, it increases to the set point, and the heater is automatically off. If the heater temperature decreases or increases,

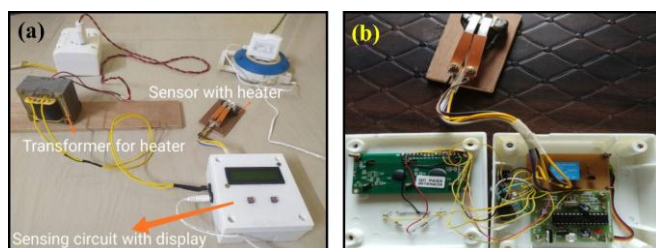


Fig. 6 (a) Portable Prototype gas sensor kit (b) Internal view interfacing with Arduino.

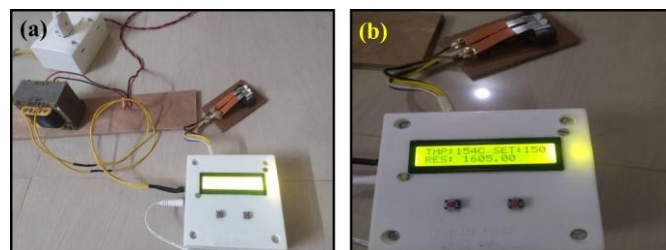


Fig. 7 (a) Portable Prototype ZnO gas sensor kit under testing condition (b) Output LCD display for readings.

control the circuit ON and OFF the heater and maintains the operating point. This complete process is completed in 5-7 minutes. After that, if LPG exposes on the surface of the thick film sensor it is indicated on display by decreasing the value of resistance.

4. Conclusion

The usual screen-printing approach was used to synthesize and produce a ZnO thick film sensor. According to the XRD pattern, the ZnO thick film is polycrystalline, with an average crystallite size of roughly 27.1174 nm. The SEM scan revealed that the ZnO particle size was approximately 1.88μm. Electrical and thermal properties have also been investigated. A prototype LPG gas leakage detector based on ZnO film sensor and Arduino ATMEGA8 has been constructed and tested successfully. Petroleum liquefied gas LPG is used not just for cooking but also in vehicles as compressed natural gas (CNG) and in industry. Because gas is flammable, there is a risk of leakage, which could result in an explosion or fire. It is critical to use a gas leak-detecting system for security reasons.

Disclosures

Free Access to this article is sponsored by SARL ALPHA CRISTO INDUSTRIAL.

Declaration

Contribution of the authors

All authors contributed equally to the preparation of this manuscript. All authors reviewed the manuscript.

Conflict of Interest

This research work has been done by all the listed authors with mutual interest. All the data used in this research work are cited in the manuscript. Therefore, no conflict of interest related to any person or agency for this manuscript.

Funding: Not Applicable

Acknowledgement:

We'd like to thank Dr. R.Y. Borse (Head, Department of Electronics, MSG College Malegaon) for overseeing and inspiring us to work on the experimental and characterization-based research article mentioned above.

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