Fabrication And Characterizations Of Screen Printed Ceo_2 Loaded Nb_2O_5 Thick Films

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Abstract:

This research focuses on the fabrication and characterization of CeO_2 loaded Nb_2O_5 thick films using the screen-printing technique. In this study, Nb_2O_5 was utilized as the base material, while CeO_2 served as an additive. The electrical properties of the fabricated films were evaluated through resistivity, temperature coefficient of resistance (TCR), and activation energy analyses at high and low-temperature regions. The resistivity, TCR and activation energy was found to be 493999.44 Ω.m, -0.001844/°C and 0.11951eV for fabricated CeO_2 loaded Nb_2O_5 thick films respectively. The gas-sensing performance of the films was investigated, revealing maximum sensitivity to NH_3 gas at an optimal operating temperature of 120°C and a concentration of 200 ppm. The results demonstrate the potential of CeO_2 loaded Nb_2O_5 thick films for applications in high-performance ammonia gas sensors. Fabricated films also shows quick response and recovery time to NH_3 gas.

Keywords: *Additive, gas sensors, thick films, electrical properties, recovery time, NH_3 gas.*

1. Introduction

The need for gas sensors has become increasingly critical due to the growing concerns over air quality, environmental pollution, industrial safety, and public health. In urban environments, exposure to harmful gases lead to serious health issues, including respiratory diseases, cardiovascular problems, and neurological disorders [1, 2]. Gas sensors enable real-time monitoring of these gases, allowing for early detection of pollution levels and the prevention of health risks. In industrial settings, gas sensors are essential for detecting hazardous leaks of toxic or flammable gases, thus preventing accidents, explosions, and potential fatalities [3, 4]. Furthermore, gas sensors play a vital role in the automotive industry by monitoring exhaust emissions and ensuring compliance with environmental regulations. With the rise of smart cities and the increasing demand for environmental monitoring, portable and reliable gas sensors are also essential for outdoor air quality monitoring, especially in areas prone to industrial emissions or traffic congestion [4, 5]. As environmental sustainability becomes a top priority globally, the development of advanced, cost-effective, and sensitive gas sensors is crucial for protecting public health, ensuring workplace safety, and supporting sustainable practices across various sectors [4-6].

Binary metal oxide gas sensors are an important class of materials used for detecting a wide range of gases, such as CO, NO_2, NH_3, and volatile organic compounds (VOCs). These sensors typically consist of two metal oxides, each contributing distinct electronic and catalytic properties that enhance gas-sensing performance [6, 7]. The combination of metal oxides, such as SnO_2–ZnO, TiO_2–CuO, or Fe_2O_3–CuO, offers several advantages over single metal oxide systems, including improved sensitivity, selectivity, stability, and response time. One key feature of binary metal oxide sensors is the synergistic effect between the two oxides, where one oxide can act as a catalyst to facilitate gas adsorption, while the other can modulate the charge carrier dynamics or enhance the surface reactivity [8, 9]. For instance, one oxide may serve as an electron donor, while the other may accept electrons during the gas-sensing process, leading to more efficient gas detection [10, 11]. Additionally, binary metal oxide sensors tailored by adjusting the stoichiometry, doping with other elements, or optimizing their morphology to enhance their surface area and porosity, further improving sensor performance [12, 13]. These sensors are widely used in environmental monitoring, industrial safety, and automotive applications, thanks to their low cost, ease of fabrication, and ability to detect gases at low concentrations [13, 14]. However, challenges such as cross-sensitivity, long-term stability, and the need for operational temperature control remain areas of active research and development [14, 15].

The physicochemical and electrical properties of CeO_2 loaded Nb_2O_5 nanoparticles play a significant role in their performance in various applications, including gas sensing and catalysis. CeO_2 is a highly versatile material, known for its excellent redox properties due to the reversible conversion between Ce^{3+} and Ce^{4+} oxidation states [16, 17]. This gives CeO_2 nanoparticles high oxygen storage capacity and enhanced catalytic activity, particularly in reactions involving the adsorption and activation of oxygen or reactive gases [18]. Their high surface area, coupled with the presence of oxygen vacancies, enables efficient interaction with gas molecules, making CeO_2 nanoparticles ideal for gas sensing and environmental remediation. The material's wide bandgap (~3.1 eV) ensures stability under varying environmental conditions, while its ability to undergo reversible transformations at the surface enhances its electronic conductivity and charge transfer properties [19, 20].

Niobium pentoxide (Nb_2O_5) is a wide-bandgap semiconductor of around 3.4 eV with a layered structure that provides high stability and robustness, even at elevated temperatures. Nb_2O_5 exhibits interesting electrical properties, such as its ability to modulate charge carriers such as electrons and holes when exposed to gases, making it a suitable candidate for sensor applications [21, 22]. The material's surface can be easily modified through doping or functionalization, which significantly enhance its conductivity and sensitivity toward targeted gases [22]. Nb_2O_5 also exhibits high dielectric constant and low leakage current, further improving its performance in devices that require stable electrical behavior [22, 23]. When combined, CeO_2 and Nb_2O_5 nanoparticles exhibit synergistic effects CeO_2 enhances the redox activity and gas adsorption, while Nb_2O_5 contributes to charge transport and material stability. This combination optimizes their physicochemical and electrical properties for advanced gas-sensing applications, enabling the detection of gases with high sensitivity, selectivity, and rapid response times [24-26].

CeO_2 loaded Nb_2O_5 nanoparticles have gained significant attention in gas sensing applications due to their unique structural and electronic properties. The incorporation of cerium oxide onto niobium pentoxide nanoparticles enhances the sensitivity and selectivity of the sensor, particularly for detecting harmful gases such as NO_2, CO, and ethanol. CeO_2, with its redox-active nature and ability to undergo changes in oxidation states (Ce^{3+}/Ce^{4+}), contributes to the modulation of the sensor's surface reactivity, thereby improving the interaction with target gas molecules [23-25]. Nb_2O_5, known for its high chemical stability and wide bandgap, provides a solid framework for the CeO_2 loading, enhancing the overall mechanical and thermal stability of the sensor. The synergy between CeO_2 and Nb_2O_5 facilitates enhanced charge transfer, better electron mobility, and increased surface area, leading to improved gas detection performance. Additionally, the modification of the CeO_2-Nb_2O_5 composite nanoparticles by doping, tuning their morphology, or optimizing their surface properties can further refine the gas sensing response, making these materials promising candidates for real-time environmental monitoring and industrial applications [24-26].

The present work is deals with the fabrication of CeO_2-loaded Nb_2O_5 films using screen printing technique and the subsequent investigation of their electrical and gas-sensing properties of fabricated films. The screen printing method offers a cost-effective and scalable approach for the fabricated thick films making it suitable for large-area applications such as gas sensors [27, 28]. In this study, CeO_2 nanoparticles are incorporated into Nb_2O_5 matrices to capitalize on the synergistic effects of both materials.

2. Materials and methods

Commercially available AR grade (99.99 % purity) CeO_2 and Nb_2O_5 nanoparticles were used for the fabrication of the films by screen printing method. In this work, CeO_2 is used as additive and Nb_2O_5 is the base material. The 7 wt. % of CeO_2 additive is added in to Nb_2O_5 by weight percentage method as reported previous work. For the fabrication of films the 70 and 30 % inorganic and organic materials composition ratio was used respectively. The 70% inorganic material consist of CeO_2 (0.049 g) and Nb_2O_5 (0.651 g) nanoparticles. The 30% organic part consist of BCA and ethyl cellulose for the preparation of thixotropic paste of selected materials. By utilizing screen printing setup (Fig. 1) the films were fabricated. After fabrication of films, all films were dried under IR lamp for 20-30 minutes then annealed at temperature of 300°C for 2 hours in the muffle furnace. Then used for electrical and gas sensing characterizations [28-30].

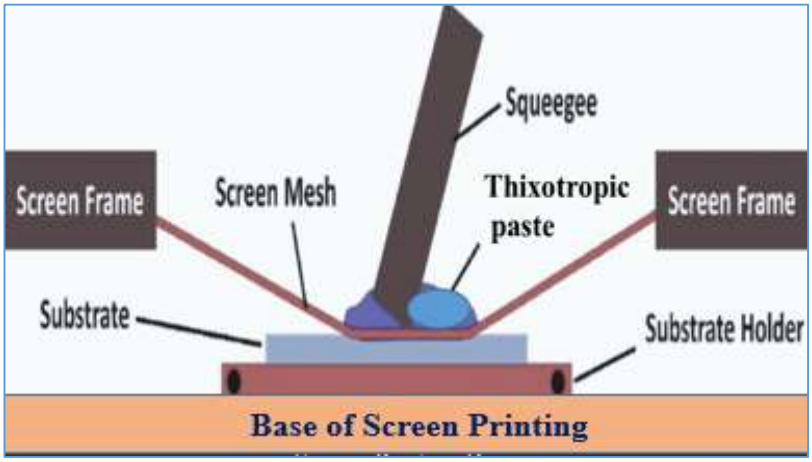

Fig. 1. Schematic diagram of screen printing setup

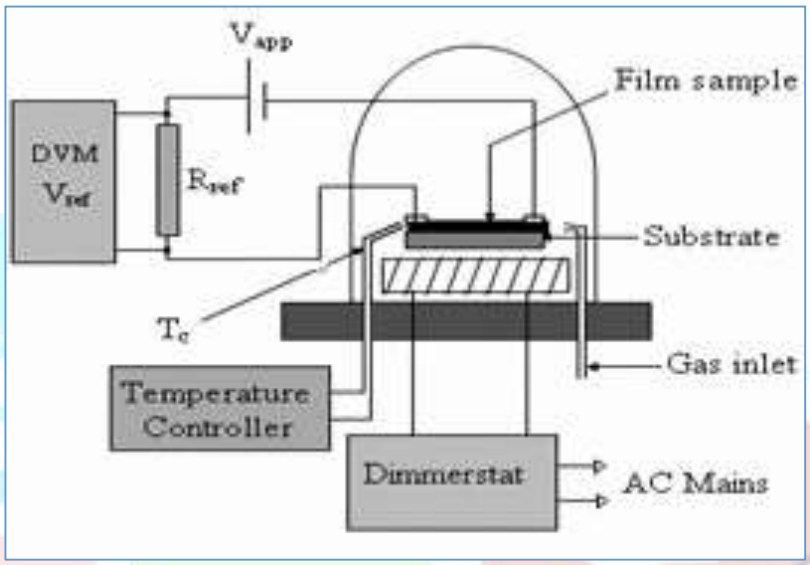

Fig. 2. Schematic diagram of static gas sensing device

The electrical characterizations were performed by investigating resistivity, TCR and activation energy at high and low temperature regions of CeO_2 loaded Nb_2O_5 thick films. Electrical characterizations of films were carried out using half bridge method and static gas sensing device as display in Fig. 2. In this device, a glass dome, thermocouple, electric coil, film substrate holders, temperature controller, digital temperature indicator, multimeter and references resistor and other components. The resistivity, temperature coefficient of resistance and activation energy of the films were determined by using Eqs. 1, 2 and 3 respectively [31].

$$\rho = \left(\frac{R \times b \times t}{l} \right) \Omega.m \qquad \text{(Eq. 1)}$$

Where, ρ = Resistivity of prepared film, R = resistance at normal temperature, b = breadth of film, t = thickness of the film, L = length of the film.

$$TCR = \frac{1}{R_o}\left(\frac{\Delta R}{\Delta T} \right) /^{O} C \qquad \text{(Eq. 2)}$$

Where, ΔR = change in resistance, ΔT = temperature difference, and R_o = Initial resistance of the film.

$$\Delta E = \frac{log\, R}{log\ Ro} \times KT \qquad \text{(Eq. 3)}$$

Where, ΔE = Activation energy, R = Resistance at raised temperature, R_0 = Resistance at room temperature.

3. Result and discussion

Fig. 3 (a) shows the plot of temperature versus resistance for the CeO_2 loaded Nb_2O_5 film, revealing a clear trend as the ambient temperature of the film increases, its resistance decreases. This behavior suggests the semiconducting nature of the CeO_2-loaded Nb_2O_5 films. In semiconductors, the conductivity typically increases with temperature, which is a result of the generation of additional charge carriers. At higher temperatures, thermal energy excites electrons from the valence band to the conduction band, thereby creating electron-hole pairs. This process effectively increases the number of charge carriers available for conduction, leading to a reduction in the overall resistance of the material [31, 32]. This temperature-dependent behavior is characteristic of semiconductors, where the increased thermal energy facilitates the movement of charge carriers, enhancing the electrical conductivity. The observed decrease in resistance with rising temperature further supports the CeO_2 loaded Nb_2O_5 films exhibit semiconducting properties, making them suitable for gas sensing applications where temperature variation plays a critical role in sensor response [32, 33].

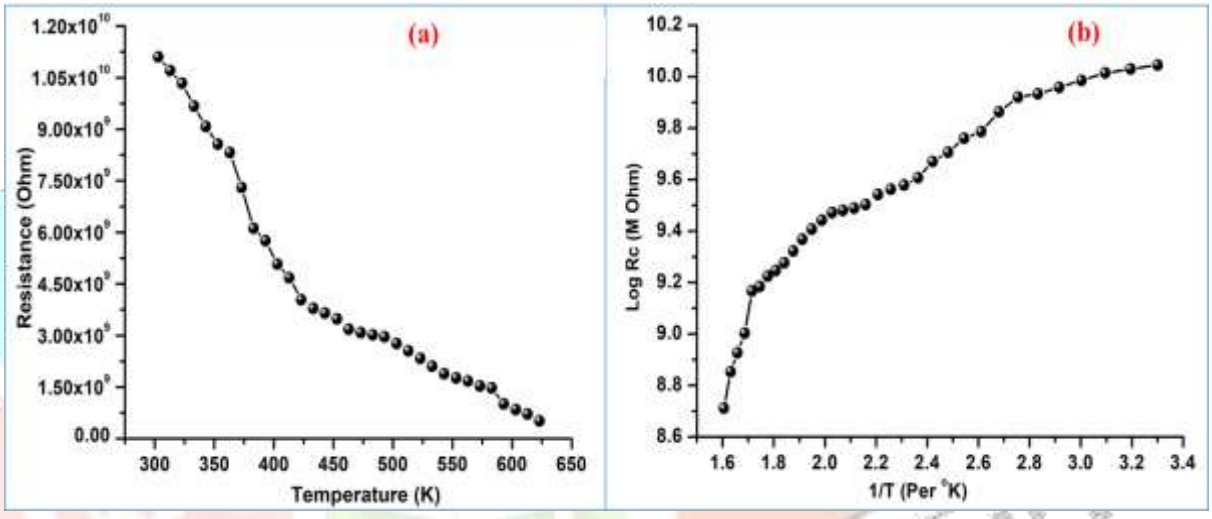

Fig. 3. Plot of (a) Resistance verses temperature and (b) Log Rc vs. 1/T CeO_2 loaded Nb_2O_5 films

The activation energy is a crucial parameter for understanding the temperature-dependent conductivity of semiconductors and other materials. A lower activation energy typically indicates that charge carriers can more easily transition between energy bands, which enhances conductivity, particularly at higher temperatures. To determine the activation energy experimentally, the temperature-dependent conductivity of the material is measured and analyzed using models like the Arrhenius equation. The Arrhenius plot is plotted Log Rc versus 1/T and it is shown in Fig. 3 (b). In the case of Nb_2O_5, doping with CeO_2 enhance electrical conductivity by introducing defects or impurities into the Nb_2O_5 lattice. These defects generate additional charge carriers, which facilitate charge transport and reduce the activation energy, leading to increased conductivity, particularly at elevated temperatures [30, 31].

In gas sensing study, the gas sensing parameter such as sensitivity, selectivity, limit of detection, response time and recovery time of CeO_2 loaded Nb_2O_5 films were investigated. Sensitivity refers to the ability of the sensor to detect changes in the concentration of a target gas. It is usually expressed as a ratio or percentage change in the sensor's output signal in the form of resistance or current per unit change in gas concentration. High sensitivity ensures the detection of even small gas concentrations. Fig. 4 (a-d) showcases several performance metrics of CeO_2-loaded Nb_2O_5 thick films for gas sensing applications [32, 33]. Fig. 4 a

highlights the selectivity of the CeO_2-loaded Nb_2O_5 thick films for different gases. The gas sensing mechanism of CeO_2-loaded Nb_2O_5 thick films to NH_3 gas operates on a chemiresistive principle involving surface interactions between the gas molecules and the sensing material. When NH_3 gas is introduced, the NH_3 molecules react with the adsorbed oxygen species. This reaction releases the trapped electrons back into the conduction band, reducing the electron-depletion layer and decreasing the film's resistance and recorded sensitivity of the films [30, 32].

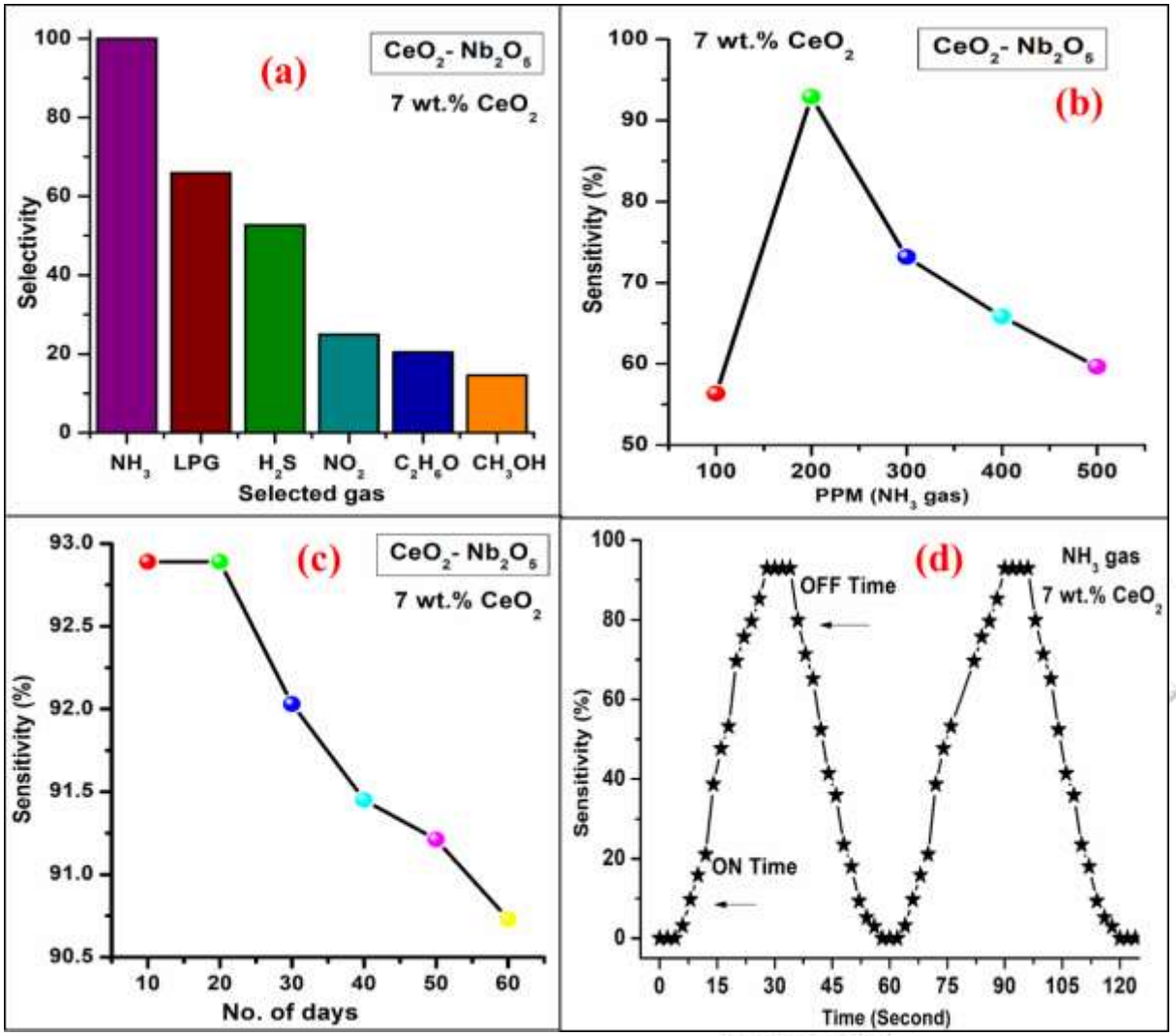

Fig. 4. Plots of (a) Selectivity histogram, (b) Limit of detection, (c) Reproducibility and (d) Response and recovery time of CeO_2 loaded Nb_2O_5 thick films

Selectivity is the sensor's ability to differentiate and respond exclusively to the target gas while ignoring other gases in the environment. High selectivity minimizes interference from other gases, ensuring accurate detection of the intended analyte. The highest selectivity is observed for NH_3 (ammonia), followed by LPG, H_2S, NO_2, C_2H_6O, and CH_3OH. Limit of Detection (LOD) is the smallest concentration of the target gas that the sensor can reliably detect. A lower LOD indicates a more sensitive sensor, capable of identifying trace amounts of gas. Fig. 4 b displays sensitivity varies with the concentration of NH_3. The sensitivity peaks around 200 ppm and decreases at higher or lower concentrations. Response time is the time required for the sensor to reach a significant percentage (commonly 90%) of its final signal after exposure to the target gas. Recovery time is the time taken for the sensor to return to its baseline signal after the gas is removed [33, 34]. Fig. 4 c shows the stability of the sensor over time, with a gradual decline in sensitivity over 60 days, indicating

some degree of degradation. Fig. 4 d depicts the dynamic behavior of the sensor when exposed to NH_3 gas. The sensitivity rapidly rises during the ON time (12 sec) and recovers quickly during the OFF time (35 sec), suggesting a fast response and recovery cycle [34, 35].

Conclusion

The research successfully demonstrated the fabrication and characterization of CeO_2-loaded Nb_2O_5 thick films using the screen-printing technique. Nb_2O_5 served as the base material, while CeO_2 acted as an effective additive, enhancing the overall performance of the films. Gas sensing studies highlighted the film's maximum sensitivity to NH_3 gas at a relatively low optimal operating temperature of 120°C and a concentration of 200 ppm. Obtained results shows the potential of CeO_2-loaded Nb_2O_5 thick films for high-performance NH_3 gas sensors in industrial and environmental monitoring.

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Conflicts of Interest

The author declare no conflict of interest.

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