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NO₂ Gas Sensing Properties of Ni-Doped ZnO Thin Film with Sponge Like Morphology

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ABSTRACT: In this study, NO₂ sensing properties of Ni-doped ZnO thin film has been investigated. Gas sensing measurements revealed that Ni doping in ZnO significantly improved the sensor's response, towards NO₂ at optimized operating temperatures as compared to our earlier reported Ag-ZnO and Cu-ZnO thin film sensors. Ni doped ZnO film with sponge like morphology showed sensing response in the range between 2.1 – 6.31 for 10-100 ppm of NO₂. The response time of the sensor ranged between 109 - 138 s, while the recovery time varied from 106 to 428 seconds towards 10-100 ppm of NO₂. This study highlights the potential of Ni-doped ZnO as an efficient gas sensor towards NO₂ gas.

KEYWORDS: Ni-doped ZnO, Gas sensor, Chemical bath deposition Method, NO₂ gas.

I. INTRODUCTION

Nitrogen dioxide (NO₂) is a major atmospheric pollutant with significant effect on human health and environmental quality. It is a reddish-brown, highly reactive gas that functions as a strong oxidizing agent. NO₂ is primarily produced through the combustion of fossil fuels such as coal, oil and natural gas, with key emission sources including vehicle exhaust, power plants and pulp mills [1]. The threshold limit values for short-term and long-term exposure to NO₂ are 5 ppm for 10 minutes and 3 ppm for 8 hours, respectively [2]. Therefore, it is essential to enforce strict emission regulations and develop continuous monitoring procedures for NO₂ in both ambient and industrial settings to reduce the immediate health and environmental risks associated with excessive nitrogen oxide levels. Among the various metal oxide sensors, Zinc oxide (ZnO) is a highly promising metal oxide for gas sensing applications due to its exceptional chemical sensitivity to adsorbed gases, excellent stability, doping versatility, non-toxicity, and cost-effectiveness [3]. Nickel (Ni) is significant transition metal dopant in ZnO, owing to its effective ionic radius (Ni²⁺: 0.69 Å) being close to that of Zn²⁺ (0.74 Å), enabling the substitution of Ni²⁺ into the ZnO lattice. This doping induces high donor defect states, enhancing charge carrier separation and transport within ZnO. Therefore, Ni doping significantly improves the gas sensing properties of ZnO [4, 5]. Among various deposition methods, the chemical bath deposition (CBD) method was used for synthesizing Ni-doped ZnO films due to its cost-effectiveness, doping flexibility, tunable deposition parameters, and suitability for large-scale production [6].

II. MATERIALS AND METHODS

Ni-doped ZnO thin film (8wt%) was deposited on glass substrates using the chemical bath deposition (CBD) method at room temperature. The precursor solution was prepared by dissolving analytical-grade zinc nitrate [Zn(NO₃)₂·H₂O] and nickel nitrate [Ni(NO₃)₂] (SDFCL) in double-distilled water. An ammonia solution (NH₄OH, sp. gr. 0.91) (SDFCL) was added dropwise to control the pH of the bath solution, as reported in our previous study [7]. The Ni-doped ZnO (8wt%) film was exposed to NO₂ gas to evaluate its gas sensing properties using an indigenously designed a static gas sensing measurement setup at the host Institution of one of the authors [8]. The film was mounted on a heating probe and placed inside a sealed gas chamber. A known volume of NO₂ gas was introduced into the chamber using a calibrated airtight syringe and needle to ensure precise gas concentration control. The operating temperature was varied from room temperature to 350°C, while the gas concentrations ranged from 5 to 100 ppm.

III. RESULTS AND DISCUSSION

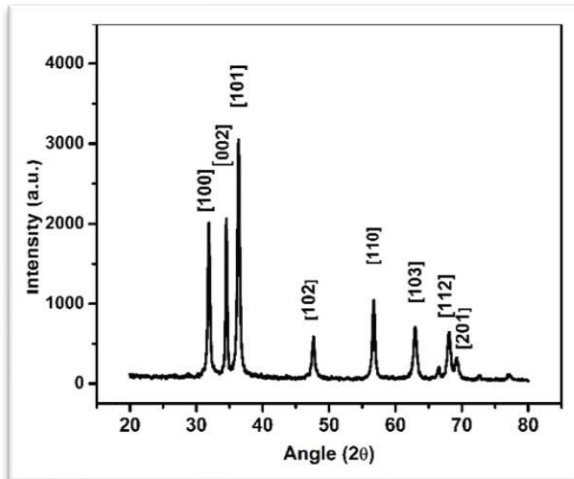


Figure 1: XRD pattern of Ni-doped ZnO [7]

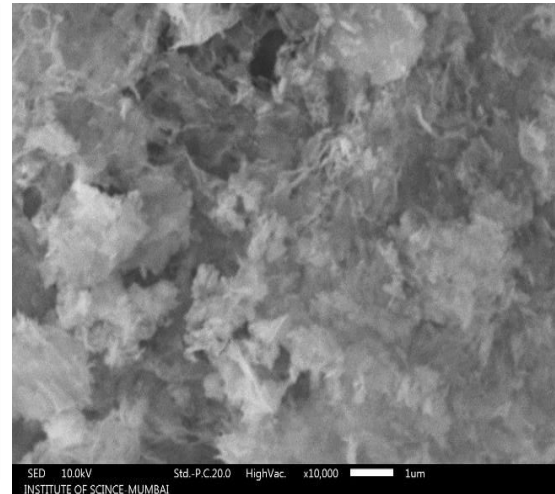
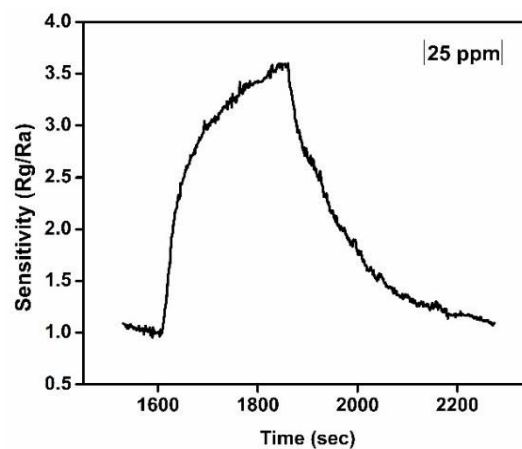
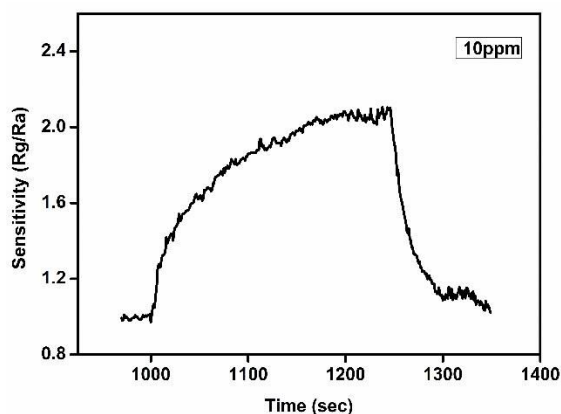


Figure 2: SEM image of Ni-doped ZnO [7]

Figure 1 shows XRD pattern of Ni doped ZnO film. The peak positions of XRD pattern reveal a hexagonal wurtzite phase and in good agreement with the data of Joint Committee on Powder Diffraction Standards (JCPDS) card no. 36-14519. Figure 2 presents the scanning electron microscope (SEM) image illustrating the morphology of Ni-doped zinc oxide film at 8 wt% Ni. The crystallites exhibit a distinctive sponge-like structure. The average crystallite size of Ni-doped ZnO is approximately 100–200 nm, with a uniform distribution across the surface.

IV. GAS SENSING STUDIES OF NI-DOPED ZNO TOWARDS NO₂ GAS

The gas sensing response curves of 8 wt% Ni-ZnO for 10 ppm, 25 ppm, 50 ppm and 100 ppm variation in NO₂ gas concentration are shown in figure 3. The response curves consistently exhibited typical gas response characteristics, with complete but extended recovery periods.



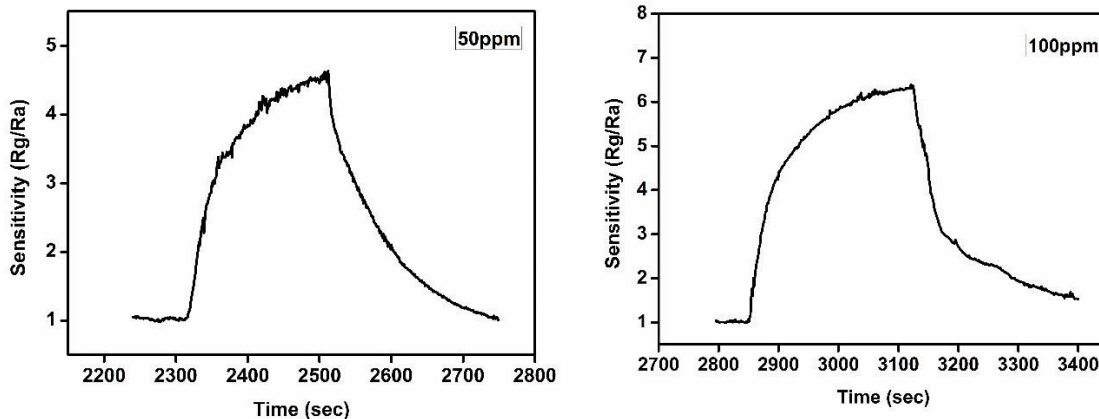


Figure 3: Response curves at 10 ppm, 25 ppm, 50 ppm and 100 ppm towards NO₂ at 200°C.

Figure 4 shows the maximum sensitivity of Ni-doped ZnO versus NO₂ gas concentration. It is observed that the sensitivity seems to be less for lower ranges but good for higher ranges. The gas sensitivity demonstrated a consistent linear increase, moving from 2.1 to 3.59 for a concentration range of 10 to 25 ppm of NO₂ gas concentration. It is again increased from 3.59 to 4.61 and then to 6.31 as the concentration increased from 25 to 50 ppm and 50 to 100 ppm, respectively.

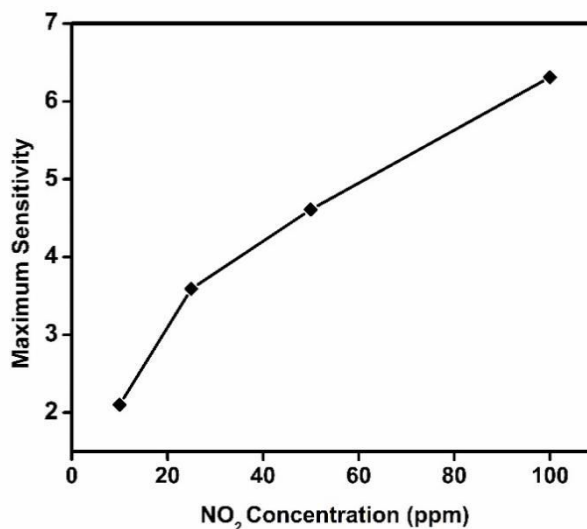


Figure 4: Maximum Sensitivity of Ni-doped ZnO versus NO₂ concentration

Figure 5 shows that the response time of Ni-doped ZnO towards NO₂. It is seen that the response time is in the range between 100-140 seconds. Figure 6 indicates that recovery time varies from 100 to 450 seconds, with lower concentrations (10 ppm and 50 ppm) showing shorter recovery times

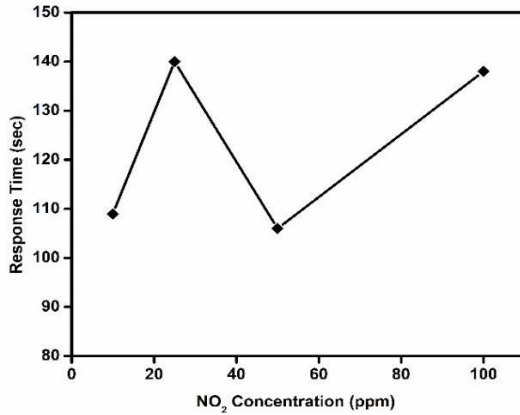


Fig 5: Response Time of versus NO₂ conc.

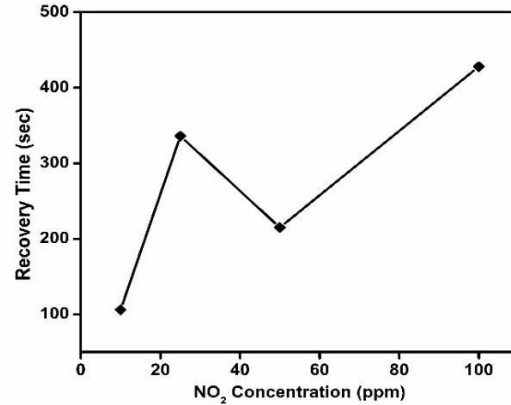


Fig 6: Recovery Time versus NO₂ conc

V. SENSING MECHANISM

On exposure of ZnO sensor to NO₂, the oxygen species already adsorbed on the ZnO surface interact with the molecules of the gas causing the electron depletion layer and junction potential barriers to widen, resulting in an increase in the sensor resistance and the generation of a sensing response [9-11]. The interaction is shown by the following reaction.

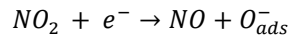


Figure 7 illustrates the comparison of sensitivity of Ni-ZnO with our earlier reported work Ag-ZnO and Cu-ZnO sensor [12,13]. It is seen that Ni-doped ZnO exhibits the enhancement in sensitivity than Ag-ZnO and Cu-ZnO. The ionic radius of Ni²⁺ is similar to that of Zn²⁺ (0.69 Å for Ni²⁺ and 0.74 Å for Zn²⁺), enabling Ni²⁺ to easily substitute Zn²⁺ in the ZnO lattice. This substitution results in the formation of donor defects, which are important for enhancing charge separation and transport within the ZnO material [14,15]. Furthermore, the porous structure of Ni-doped ZnO leads to a high surface-to-volume ratio. This characteristic increases the surface activity of the nanoparticles, as the adsorbed oxygen species and surface defects play a significant role in the material's response [16].

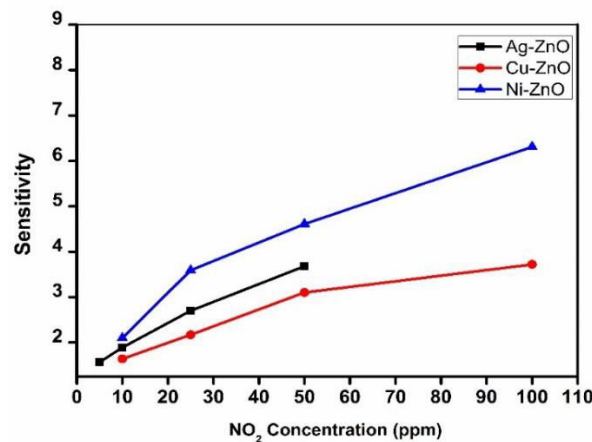


Figure 7: Sensitivity of doped ZnO versus NO₂ concentratio

VI. CONCLUSION

This study demonstrates that Ni-doped ZnO thin film with sponge like morphology exhibit enhanced NO₂ sensing properties compared to our previously reported work Ag-ZnO and Cu-ZnO sensors. The doping of Ni into ZnO significantly improved the sensor's response, with a response range of 2.1 – 6.31 for NO₂ concentrations between 10-100 ppm. The sensor displayed a response time of 109 -138 seconds and a recovery time ranging from 106-428 seconds. These results underscore the potential of Ni-doped ZnO as an efficient and effective gas sensor for detecting NO₂.

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