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Study of Conduction Mechanism of Bi_2O_3 Doped Sr_2CoO_4 Nanocomposite Material and its Gas Sensing Behavior

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ABSTRACT

Nanocrystalline 10 Wt% Bi_2O_3 doped Sr_2CoO_4 was prepared by sol-gel citrate method, and calcined at different temperature. The Sr_2CoO_4 nanoparticles were characterized by Impedance analysis, and Gas sensing. The ac conductivity was studied for the sample 10 Wt% Bi_2O_3 doped Sr_2CoO_4 calcined at 650^OC, at temperatures 100 to 700^OC and over a wide range of frequencies (50 Hz to 200 kHz). Experimental results indicate that the ac conductivity depend on temperature, frequency and concentration of dopent. Nanocrystalline 10 Wt% Bi_2O_3 doped Sr_2CoO_4 was found to be good ammonia sensor with high sensitivity and selectivity.

Keywords: Sol- Gel Citrate, Gas, Sensing

I. INTRODUCTION

Ammonia is a natural gas that is present throughout the atmosphere in concentrations of low-ppb to sub-ppb levels as the result of emission from anthropogenic and natural sources [1]. It is produced in large quantities by chemical industry for the production of fertilizers and other nitrogen-containing compounds and for the use in refrigeration systems as cooling agent. Natural source include production by bacteria.

In the environment, high concentrations of ammonia lead to eutrophication and acidification of both ground and water, whereas in indoor environments it is a health hazard to humans [2-3]. Therefore ammonia is an important target gas in applications like leakage control in refrigeration systems and air conditioners on one hand or in the emission control and quality monitoring of waste and drinking water on the other hand [4-5]. Other application areas for ammonia sensors include high temperature sensing in the exhaust of cars and, as it is a product of biochemical processes, ammonia is also a useful reporter molecule in a variety of medial applications. This need have supported the development of devices capable of the detection and quantification of gaseous and dissolved ammonia. Many aspects of ammonia sensing are summarized in a recent review by Timmer et al. and information on ammonia sensing in solution can be found.

For the use in work place safety and indoor monitoring applications devices capable of detecting ammonia in concentrations of 1–50 ppm in ambient air are required. These devices must not be prone to strong humidity interferences. Several regulations on the allowed ammonia concentration and exposure times exist, as there are the acceptable exposure limits (AEL) in Germany/Switzerland, the long time (LTEL) or short time (STEL) exposure limits in the UK, or the permissible exposure limit (PEL), usually a time-weighted average (TWA), in the USA. The values given therein are in the range from 25 to 50 ppm. Many commercial ammonia gas sensors as well as analytical devices are offered for this application area and concentration range [6-10].

The scientific literature describes the development of many sensor types. Optical ammonia sensors detect ammonia by the change in absorbance of an acid-base indicator, fluorescence of an ammonia complex, or a change in refractive properties of a coating. Infrared (IR) based gas sensors measure the absorbance change induced by the ammonia molecule. Electrochemical sensors rely on a change in resistance, capacitance or

potential of the sensitive layer. Finally, resonator devices measure the mass of absorbed ammonia. Reported as sensitive layers in chemical gas sensors are different metal oxides, CuBr, TiN, and many different polymers and polymer blends [11]. In this paper we are presenting the nanocrystalline Sr_2CoO_4 as a ammonia gas sesnsor.

II. EXPERIMENTAL DETAILS

The nanocrystalline Sr_2CoO_4 specimens were prepared by using sol-gel citrate method. A stoichiometry mixture of strontium nitrate and cobalt nitrate were magnetically stirred with citric acid and ethylene glycol at 80°C for 2 h to get homogeneous and transparent solution. The solution was further heated at about 130°C for 12 h in a pressure vessel to form the gel precursor. The prepared product was subjected to 3 h heat treatment at 350°C in a muffle furnace and then milled to a fine powder. The dried powder then calcined in the range of 450-750°C in order to improve the crystallinity of materials. The solution of Bismuth nitrate was used as dopant in the precursors of Sr_2CoO_4.

III. RESULT AND DISCUSSION

Figure 1 shows Complex impedance formalism helps in determining inter-particle interaction like grain, grain boundary effects, etc. To study the contribution due to different effects, Cole–Cole analyses have been done at different temperatures. It also provides information about the nature of dielectric relaxation. For pure monodispersive Debye process, one expects semicircular plots with the centre located on the *Z'*axis whereas, for polydispersive relaxation, these argand plane plots are close to circular arcs with endpoints on the axis of reals and the centre below this axis. From the graph it is observed that the semicircular diameter decreases with increasing frequency, thus the resistivity of the sample decreases i.e. conductivity of the sample increases.

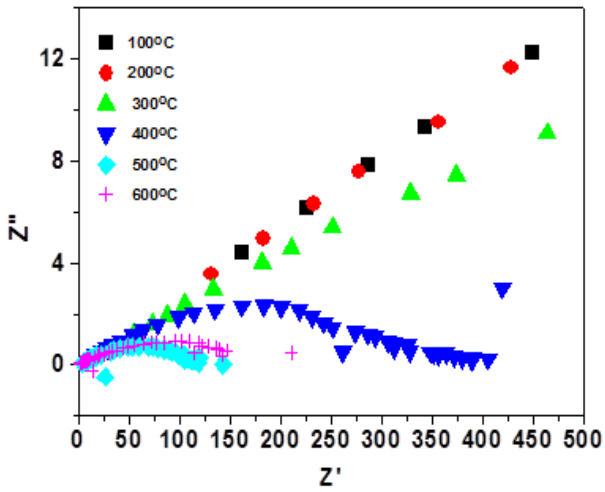

Figure 1. Nyquist plot for 10 Wt% Bi_2O_3 doped Sr_2CoO_3 nanocomposite.

The ac conductivity increases with increase of the frequency and temperature applied in the experiment. The rise of conductivity upon increasing the frequency and temperature is a common respond for semiconductor samples. It is due to the tremendous increase of the mobility of charge carriers in the composite film. As shown in Figure 2, there are two trends appeared, the first one is frequency independent conductivity and another is frequency dependent conductivity. The first trend is contributed by free charges available in the composite system whereas the second, which is frequency dependent conductivity, is due to trapped charges which are only active at higher frequency region.

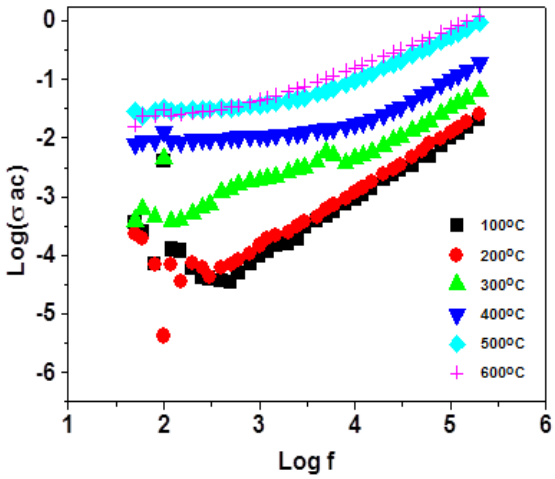

Figure 2. Variation of conductivity with frequency for the 10 Wt% Bi_2O_3 doped Sr_2CoO_3 Nanocomposite.

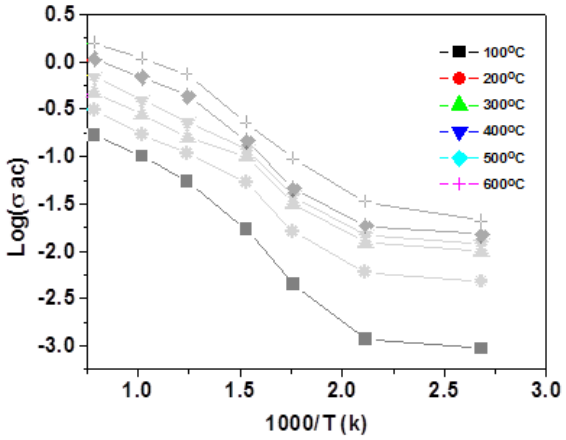

Figure 3. Arrhenius plot for conductivity for 10 Wt% Bi_2O_3 doped Sr_2CoO_3 nanocomposite.

Figure 3 presents the experimental results of the electrical conductivity for all orthophosphate samples in the standard Arrhenius plot

$$\sigma = \sigma_0 \exp(-Ea/kBT),$$

where Ea is the activation energy. These results show an increase of the conductivity with increasing temperature for all compounds indicating a characteristic activated behavior over the complete temperature range studied. Furthermore, plots of log(s) vs. 1/T were found to be linear in the temperature range considered. From impedance measurements, the total conductivity activation energies, Ea, were derived, yielding activation energies 0.000116, 9.59E-05, 9.57E-05, 9.11E-05, 8.87E-05 and 8.4E-05 eV for the frequencies 10, 50, 100, 120, 150 and 200 kHz respectively. From activation energy values it is clear that the activation energy decreases with increasing frequency thus the conductivity of the sample increases with the increasing frequency.

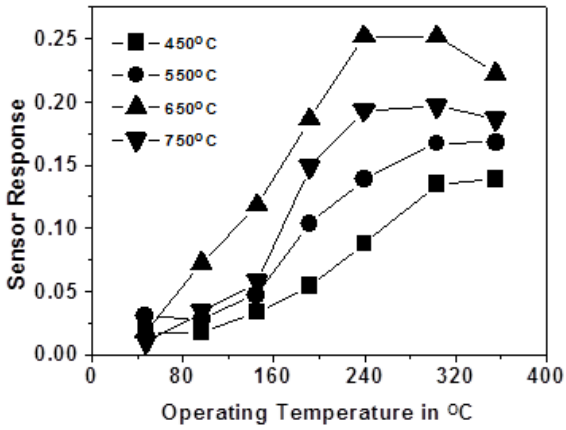

Figure 4. Sensor response to 1000 ppm NH_3 gas as a function of the operating temperature for undoped Sr_2CoO_4 films calcined at 450, 550, 650 and 750°C.

Figure 4 shows the effect of calcination temperatures on the response of Sr_2CoO_4 sensors to 1000 ppm ammonia gas at a various operating temperatures ranging from 50-350°C. Although the maximum responses of Sr_2CoO_4 obtained at different heating temperatures appear at different operating temperatures, it is clearly seen that the response to ammonia gas are greatly affected by the heating temperature. The response of Sr_2CoO_4 obtained at 650°C for 6 h reached a maximum at an operating temperature of 250°C.

Selectivity is the ability that a gas sensor to distinguishes between different kinds of gases. Figure 5 shows the cross sensitivity of Sr_2CoO_4 for NH_3, CO, LPG and H_2 gases as a function of operating temperature. It is evident from the figure that the Sr_2CoO_4 sensor was highly selectivity to NH_3 gas against CO, LPG and H_2 gases. The sensor shows high degree of selectivity towards NH_3 gas than other reducing gases at an operating temperature of 250°C.

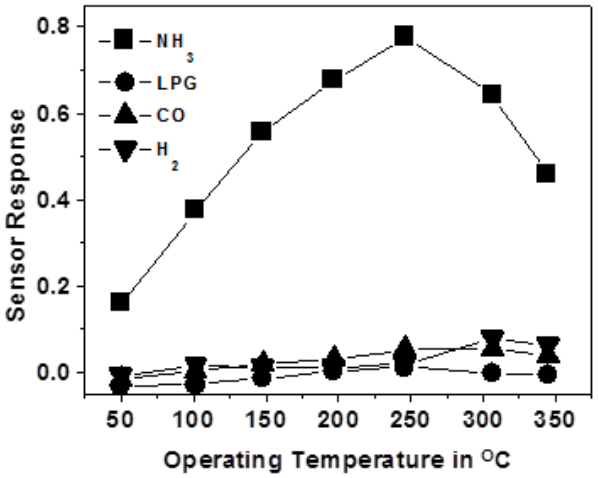

Figure 5. Sensor response of Sr_2CoO_4 thick films calcined at 650°C to 1000 ppm of NH_3, LPG, CO and H_2 as a function of operating temperature.

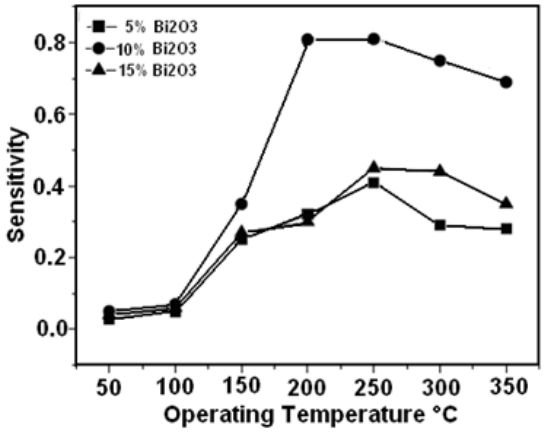

Figure 6. Sensor response of 5%, 10% & 15% Bi_2O_3 doped Sr_2CoO_4 films calcined at 650°C for 1000 ppm NH_3 gas at an operating temperature 200°C.

In order to promote gas sensitivity, dopants were shown to effectively influence the semiconductive properties of sensor materials. Fig. 6 shows the different wt% (5, 10 and 15) of Bi_2O_3 doped Sr_2CoO_4 calcined at 650^OC for 6h for 1000 ppm NH_3 gas at an various operating temperatures ranging from 50-350^{o}C. The gas response of the Sr_2CoO_4 was markedly promoted for 10wt% Bi_2O_3 doped Sr_2CoO_4 calcined at 650^{o}C at an operating temperature of 200°C.

IV. CONCLUSION

In this work, Sr_2CoO_4 have been presented as suitable semiconductor materials for selective NH_3 detection. The electrical conductivity of Sr_2CoO_4 is strongly dependent on the amount of Bi_2O_3 doped Sr_2CoO_4. The electrical conductivity increased with increasing Bi_2O_3 content and attained the maximum at 10 wt% Bi_2O_3 doped Sr_2CoO_4 calcined at 650^OC. The impedance spectra for 10 wt% Bi_2O_3 doped Sr_2CoO_4 shows the sample resistance decreases with increase in temperature. The sensor shows high degree of selectivity towards NH_3 gas than other reducing gases. The best sensor response was found at an operating temperature of 200°C for NH_3 gas.

V. ACKNOWLEDGEMENTS

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