Solid State Tin Oxide based Gas Sensor for Liquefied Petroleum Gas Detection at Room Temperature

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Abstract

This paper presents the Liquefied Petroleum Gas sensing of tin oxide. Variations in resistance with exposure of LPG to the sensing element were observed. The average sensitivity for different volume percentage of gas was estimated. The maximum value of average sensitivity was found 2.0 for 2 vol.% of LPG. Percentage sensor response as a function of time was calculated and its maximum value was 79%. Response time of the sensor was 70 sec. Scanning electron micrographs and X-ray diffraction studies of samples have been done. SEM shows that grains are spherical in shape. XRD pattern reveals the tetragonal crystalline nature of the material. The crystallites size of the SnO_2 was found 22 nm. The sensor was quite sensitive to LPG and results were found reproducible.

Key words-LPG Sensor, TiO₂, SEM, XRD, Sensitivity

I. INTRODUCTION

SnO₂ is an important metal-oxide semiconductor, widely studied in gas sensing applications due to special properties such as chemical and thermal stability, natural non stoichiometry and stable phase SnO2 has been investigated for its gas sensing properties towards various reducing gases such as LPG, acetone and ethanol. Recently Liquefied Petroleum Gas (LPG) widely used as a fuel for industrial and domestic purposes, has often proved to be hazardous because of explosions caused by leaks [1-3]. It is therefore important to develop good sensors for the detection of LPG. A good chemical and thermal stability under operating conditions along with high mobility of conduction electrons are the important features of the SnO2 based gas sensors [4-7]. Most of the sensors based on commercially available bulk tin oxide is severely affected by the small surface to volume ratio and also operate at relatively high temperatures of over 573 K [8-11].

In the present study, we report LPG sensing behaviour of the bulk tin oxide. The structural and surface morphological properties of bulktin oxide were investigated.

II. EXPERIMENTAL DETAILS

The starting material was SnO₂. These were made fine on grinding in mortar with pestle for 6-8 h. The pelletization of this material having dimensions 9 mm in diameter and 5 mm in thickness was done by using hydraulic press under a uniaxial pressure of 462 MPa at room temperature. It was exposed to Liquefied Petroleum Gas in specially designed gas chamber at controlled conditions. Corresponding variations in resistance with the time were recorded by using a Keithley Electrometer.

III. CHARACTERIZATION

A. Surface Morphology

The surface morphologies of the bulk SnO2 was analyzed using a Scanning Electron Microscope unit and are visualized in Fig. 1. This reveals that the molecules of SnO₂ consist of spherical grains having diameter lie between 30-210 nm. Each grain is evenly dispersed over the surface.



Fig. 1 : SEMof SnO₂ pellet at nanoscale

B. X-ray Diffraction

The crystal structure and phase of the powdered sample was analysed using X-ray Diffract meter with Cu-K_aradiation as source having wavelength 1.546 Å. Crystallite size was estimated using the broadening of XRD peaks by the Debye-Scherer formula which is as follows:

$$D = \frac{K\lambda}{\beta \cos \theta}$$

Where β is the full width at half maximum (FWHM) of the peak, λ is X-ray wavelength, θ is the Bragg angle and K= 0.94, a dimensionless constant.

Fig. 2 shows the XRD patterns of bulk SnO_2 . These patterns shows tetragonal crystalline SnO_2 phase. Theaverage crystallite size was found 110 nm and minimum crystallite size was 22 nm corresponding to the plane (311).



Fig.2 XRD of SnO₂ in the form of powder I. Gas Sensing Characteristic

Sensitivity is the measure of physical and chemical properties of the sensing material when it is exposed to a desired gas. This term is also used to indicate either to the lowest level of chemical concentration or to the smallest increment of concentration that can be detected in the sensing environment. Greater is the change in physical properties of the sensing element under consideration, greater will be the sensitivity of the sensor. Here we have estimated resistance of tin oxide as monitoring quantity and therefore the sensitivity of LPG sensor may be defined as the ratio of the resistance in presence of the target gas (R_g) to the resistance in presence of the air (R_a), i.e.

$$\mathbf{S} = \frac{Rg}{Ra}$$

Where $R_a \& R_g$ stand for the resistance of the sensor in air and in the sample gas respectively.

The sensor response (SR) which is to be evaluated for each set of observations, given as follows:

$$\mathbf{SR \%} = \left| \frac{Ra - Rg}{Ra} \right| \mathbf{x100}$$

IV. RESULTS AND DISCUSSION

During experiment each time of exposition of LPG to the SnO₂ pellet, it was allowed to equilibrate inside the gas chamber for 20-25 minutes and the stabilized resistance was taken as Ra. The variations in resistance with the exposure time for different concentrations of LPG were observed as shown in Fig.3. Fig. 3 illustrates variations in resistance of pellet with time after exposure for different vol.% of LPG at room temperature. Finally the outlet of the chamber was opened, the resistance approaches to their initial value of stabilized resistance in air (Ra) for further range of time. Fig. 4 illustrate variations in average sensitivity of tin oxide pellet with time after exposure for different vol.% of LPG and it is found that as the concentration of LPG inside the chamber increases, average sensitivity of the sensor increases linearly. The maximum average sensitivities were found 1.5 and 2.0 for 1.5 and 2 vol.% of LPG.Sensor response curves are shown in Fig. 5. Curves SR1, SR2, SR3 and SR4 show variation in percentage sensor response with time after exposure for different vol.% of LPG upto 1100 sec and the maximum values of sensor responses were 64 and 79 for 1.5 and 2 vol.% of LPG.



Fig.3. Variations of resistance of SnO₂ pellet with time after exposure for different vol.% of LPG



Fig.4. Variations of average sensitivity of sensing material for different vol.% of LPG.



Fig.5. Sensor Response curves of SnO₂ sensing materials with time after exposure for different vol.% of LPG

Prima-facie the interaction of gas molecules with the surface of pellet causes the transfer of electrons between semiconducting surface and reducing gases. The atmospheric oxygen molecules (O2) are adsorbed on the surface of the pellet of tin titanium oxide. They capture the electrons from conduction band of the sensing material as below:

$$O_2(gas) \leftrightarrow O_2(ads)$$

$$O_2(ads) + e^- \rightarrow O_2^-$$

Due to this, electronic conductivity decreases, which results an increase in the resistance of the sensing material. Thus the equilibration of the chemisorption process results in stabilization of the surface resistance. Any process that disturbs this equilibrium gives rise to change in conductance of semiconductors.

Also when we expose the liquid petroleum gas to the surface area of semiconducting oxide, then reaction takes place between LPG and oxygen molecules, which are absorbed by the tin oxide pellet, i.e., a surface charge layer is formed, so that the bulk charge is generated. LPG is able to react with the adsorbed oxygen then the combustion products such as water depart, and a potential barrier to charge transport is developed i.e., this mechanism involves the displacement of adsorbed oxygen species by formation of water. This can be understood by following reaction. The formation of barrier is due to reduction in the concentration of conduction carriers and thereby, results in an increase in resistance of the sensing element with time. As the pressure of the gas inside the chamber increases, the rate of the formation of such product increases and potential barrier to charge transport becomes strong which stops the further formation and resistance decreases and becomes constant.

V. CONCLUSION

The research in this paper has demonstrated the possibility and potential of a nanosized SnO₂ based sensor operable at room temperature for sensing of LPG. SEM micrographs reveals that the molecules of SnO₂ consist of spherical grains. From XRD, minimum crystallite size was found 22 nm. The average sensitivity of sensor was 2.0. Rapid sensor response, good stability and satisfactory sensitivity demonstrate the promise of this sensor for LPG determination in the industrial and environmental monitoring.

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