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Thermoelectric power (α) and I-V characteristics study of SnO_2 thin films as a function of thickness

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ABSTRACT

Thin films of tin (Sn) of varying thicknesses have been deposited on glass substrates in a vacuum. After annealing thin films of tin were heated in an air, forms tin oxide (SnO_2) films. Thickness dependent thermoelectric power (α) of the films has been measured. The current for different voltages and different thicknesses of SnO_2 thin films were measured. The current and voltage characteristics of SnO_2 thin films for different thickness have been predicted.

Keywords: SnO_2, thin films, thickness, thermoelectric power, current, voltage.

INTRODUCTION

Cassiterite which is an important mineral of tin has stannic oxide, SnO_2 as its main constituent, crystallizes in the tetragonal rutile structure and is strongly resistant to chemical reagents and heat treatment in air or oxygen. It belongs to the structural space group P4/mm having two molecules per unit cell. Kutomi and Nobusawa [1] prepared bulk polycrystalline SnO_2 by heating metallic tin in air and thermo electrical properties of bulk SnO_2 investigated. The thermal conductivity, electrical resistivity and Seebeck coefficient were measured and a figure of merit was calculated. Some of the authors[2-3] have studied the thermoelectric power of SnO_2 films deposited by spray pyrolysis.

Thermoelectric power (TEP) of SnO_2 films prepared by pyrosol process has been measured by Islam and Hakim [4] in the concentration range 9 x 10^{18} to 1.6 x 10^{21} cm^{-3} between 27 to 160 ^{O}C. The dielectric properties, the I-V characteristics and electrical resistance of SnO_2 films found by oxidation of tin films have been reported by [5-7]. The sheet resistivity of tin oxide films deposited by electron beam evaporation has been studied during annealing both as function of time and temperature [8]. The electrical and optical properties of stannic oxide have been widely investigated [9-10]. Deshpande and Bhide [11] reported dielectric properties of SnO_2.The permittivity as a function of temperature was measured in ceramic and compact powder samples of SnO_2. There is a permittivity peak in the region at 25 ^{O}C and dielectric non linearity is observed below this temperature. They further suggested that SnO_2 is ferroelectric. According to Van Daal [12] the static dielectric constant of SnO_2 for directions parallel and perpendicular to c-axis (tetragonal) has values of 9.0 (+ -) 0.5 and 14(+ -) 0.5 respectively. Tin oxide in its pure form is an n-type semiconductor with a wide band gap. In recent years a great interest has been developed in the studies of stannic oxide films owing to the number of practical applications such as solar cell [13], liquid crystal displays [14] and gas sensors [15]. Therefore it can be said that tin oxide in thin film state is technologically important. Thin films of SnO_2 have been prepared by Agarwal and Saxena [16] from thermal oxidation of vacuum evaporated tin films at 140 ^{O}C. Tin oxide films were successfully prepared by the reactive evaporation method with a dc Glow of oxygen. The structure and composition of the films were characterized by Mossbauer spectroscopy, X-ray diffraction and scanning electron microscopy [17].

The highly conducting and polycrystalline SnO_2 films were prepared [18] by the oxidation of tin films at 450 to 500 ^{O}C. Ogawa et al [19] prepared tin oxide films from ultrafine particles . The particle size and film structure were studied under various deposition conditions, that is oxygen gas pressure, rf activation power, evaporation temperature and others.

From the survey of literature there are various methods for preparing SnO_2 films, Their structure is found to vary from one to another. Films of SnO_2 prepared by vacuum evaporation method are found to be amorphous and disordered [20]. Dissociation and formation of new species have been observed during the vacuum deposition of several oxides [21]. In order to avoid the formation of amorphous and disordered SnO_2 films and dissociation of SnO_2 films formed by vacuum evaporation method, we obtained SnO_2 films by oxidation of tin films for the measurement of thermoelectric power (α) and I-V characteristics**.**

MATERIALS AND METHODS

1. Preparation of tin thin Films

Tin metal films of varying thicknesses were obtained on backed and cleaned microscopic glass slides by vacuum evaporation of 99.999 % pure metal with a pressure of ~ 10^{-4} torr using a conventional vacuum system.

The films were thermally oxidized in air at a temperature of 410^{O}K with an atmospheric pressure for 36-48 hours, forms tin oxide (SnO_2) films.

The SnO_2 thin films were annealed at 383^{O}K for 6-8 hrs, after annealing the stoichiometric films of SnO_2 were used for measurement purpose. The Large area ohmic contacts of evaporated Al were made at the end of the films.

2. Measurement of Thickness

The film thickness (d) of SnO2 thin films was measured by gravimetric and optical interference methods, reported by [22-25] (±100Å) using the relation

$$d = \frac{M}{g \times A} cm \qquad (1)$$

Where
A - Surface area of the film
M - Mass of the film material
g - Density of the film material,

Both techniques of thickness measurements agreed within ±10% for very thin films and ± 5% for thick films.

3. Electrical properties

The thermoelectric power (α) is measured by integral method [26].

In Integral method, one end of the sample is heated while other end held at constant temperature. The temperature difference (ΔT) between two ends of the film causes the generation of thermo emf.

Thermoelectric power was calculated from the relation

$$\alpha = \Delta V/\Delta T, \mu V/^{O}K \qquad (2)$$

The measurement of I-V characteristics [27] were carried out at different temperatures ranging between room temperature to 473 ^{O}K. The varying electric field was applied to the sample by an electronically regulated DC power supply and the current was recorded with the help of microvolt meter.

To eliminate the effect of adsorbed moisture, the measurements were made at reduced pressure of ~ 10^{-2} torr keeping the film inside a glass vacuum tube connected to rotary pump. For all these measurements, leads were connected to Al electrode using pressure contacts.

RESULTS AND DISCUSSION

The variation of **α** with thickness of the film is shown in Fig.1, it is seen from this fig. that thermoelectric power decreases with increase of thickness irrespective of type of conductivity. This decrease of α with increase of film thickness is related to removal of defects with increase of film thickness and decrease in inter crystalline barrier heights.

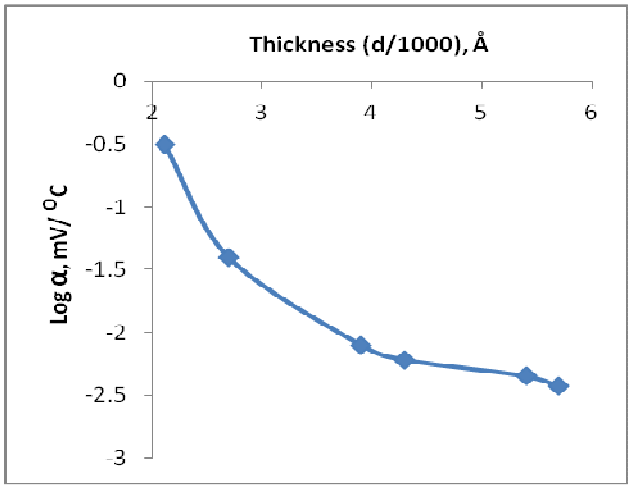

Fig. 1: Plot of log α versus thickness (d).

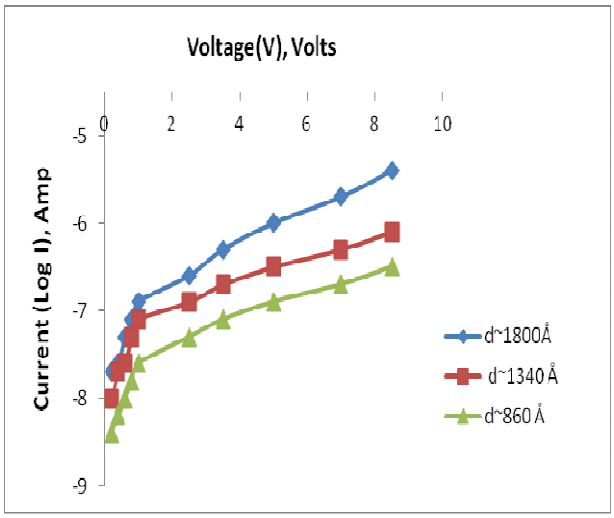

Fig.2 : Plots of current (Log I) versus voltage (V).

Fig.2 represents I-V characteristics of films having different thicknesses. It is seen that each curve starts with ohmic region (I α V) and then changes to nonohmic region. (I α V^n). The observed ohmic dependence of current on voltage at low fields can be explained on the basis that in the sample studied, bulk limited current exceeds the space charge limited current (SCLC). At higher fields, the observed value of n (I α V^n) which is less than two, for all curves, rejects the possibility of an explanation of nonohmic region on the basis of Rose [28] theory of SCLC in defect insulators containing exponential distribution of traps or shallow traps. However I-V characteristics of thermally grown $Sn0_2$ films as reported by Agarwal et al [29] Showed SCLC conduction with n = 2 or more. In our present investigation n is always found to be less than 2.

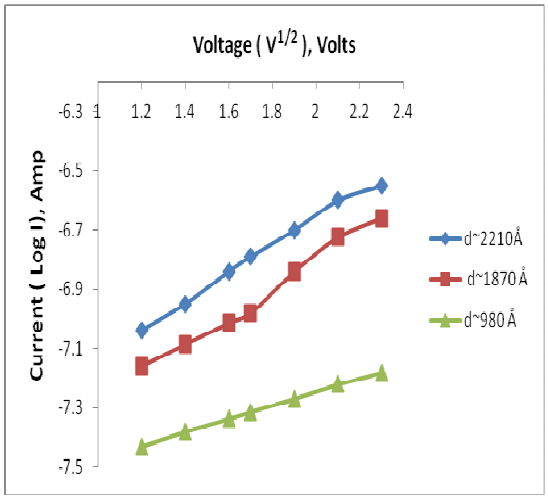

Fig.3 : Plots of current (log I) versus voltage ($V^{1/2}$).

Plots of log I versus $V^{1/2}$ shown in fig.3 are straight lines in the high field region, suggesting that Schottky emission or Poole-Frankel emission might be responsible for the non linearity in I-V curves and may be expressed by the relations

$$I = AT^2 \exp [(\beta_S V^{1/2} - \phi_0) / k_B T] \quad (3)$$

(Schottky emission)

$$I = B \exp [(\beta_{PF} V^{1/2} - \phi_1) / k_B T] \quad (4)$$

(Poole-Frankel emission)

Where

$\beta_{PF} = 2 \beta_S = 2(e^3 / \mathbf{4}\pi \varepsilon_0 \varepsilon_r d)^{1/2}$

and

e = electronic charge,

d = thickness of the film,

k_B the Boltzmann constant and ε_r and ε_0 are high frequency dielectric constants of the material and air respectively, and A and B are constants.

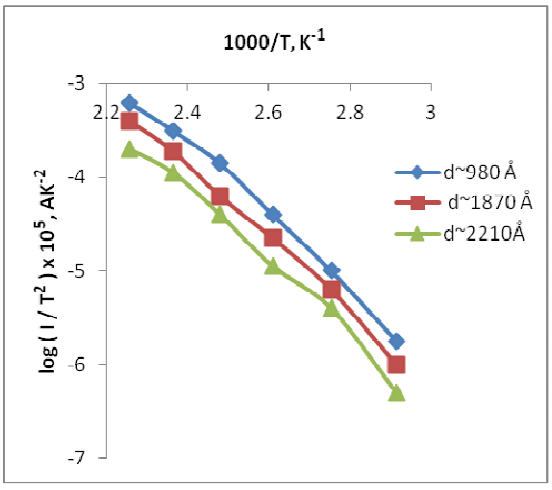

Fig.4 : Plots of log (I/T^2) Versus reciprocal temperature (1000/T)

Fig.4 depicts the graph between log (I/T^2) and $10^3/T$. The straight lines for films of different thicknesses show that Schottky emission is responsible for nonohmic conduction in $Sn0_2$ films. The barrier heights (ϕ) are calculated from the slopes of graphs of log $1/T^2$ versus $10^3/T$. It is seen that barrier height decreases as thickness increases.

The thermoelectric power of tin oxide samples in thin film state was measured by integral method. The samples containing excess of Sn^{+2} showed p-type conductivity while those containing excess of Sn^{+4} showed n-type conductivity.

In other words p-type conductivity is observed in the films corresponding to Sn0 stoichiometry and n-type conductivity in samples corresponding to $Sn0_2$ stoichiometry. Further it can be said that the type of conductivity of tin oxide film is governed by the ratio of Sn^{4+}/Sn^{2+}. As the value of this ratio increases the p type conduction disappears. Our observation for transformation of type of conductivity from p to n on the ratio of Sn^{4+} / Sn^{2+} is in excellent agreement with that of Uen et al [17].

CONCLUSION

Thin films of tin (Sn) have been well prepared in vacuum by evaporation method and oxidized in air form SnO_2 films. Measurement of thermoelectric power (α) and current (I) of such films are the functions of thickness and temperature**.**

Thermoelectric power decreases with increase of thickness irrespective of type of conductivity. The I-V characteristics of films for different thicknesses have been plotted, it is seen that each curve starts with ohmic region ($I \alpha V$) and then changes to nonohmic region as ($I \alpha V^n$). log I versus $V^{1/2}$ are straight lines in the high field region, suggesting that Schottky emission or Poole-Frankel emission might be responsible for the non linearity in I-V curves. The log (I/T^2) verses $10^3/T$ curves show straight lines for films of different thicknesses, showed that Schottky emission is responsible for nonohmic conduction in $Sn0_2$ films.

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