Non-ohmic electric conduction in polycrystalline thulium oxide

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Abstract. The current voltage behaviour of polycrystalline Tm_2O_3 has been investigated at 77 K and 300 K to 350 K. At low voltages, the conduction is ohmic while at higher voltages the current has a quadratic dependence on voltage. The voltage V_T at which the transition from ohmic to non-ohmic conduction takes place changes from 180 V to 100 V as temperature increases from 77 K to 350 K. The results are interpreted in terms of the Rose theory of space charge limited currents (SCLC) in defect insulators containing shallow traps.

Keywords. Non-ohmic conduction; space charge limited currents; thulium oxide; electrical conductivity.

1. Introduction

Injection current is a powerful probe for the study of transport properties and defect states in insulators. Recently theoretical studies in space charge limited currents (SCLC) has been reported by Gould (1982) and Sharma (1982). Non-ohmic conduction and SCLC have been experimentally studied in GeSeGa (Mycielski *et al* 1982), SnO (Agarwal and Saxena 1982), SiO_x (Herdt and Adachi 1981), Ge (Koren and Borisenko 1981), In_2O_3 (Jain and Garg 1980), TeO_2 (Jain and Garg 1979), Bi_2O_3 (Arya and Singh 1979; Alvin and Sathianand 1974), Tm_2O_3 (Zdanowicz and Zdanowicz 1979; Zdanowicz *et al* 1980), Eu_2O_3 (Nakane *et al* 1979), Dy_2O_3 (Goswami and Verma 1974) and TiO_2 (Katsuta *et al* 1971). Electric and dielectric properties of rare earth oxide, used in the preparation of garnets, orthoferrites and molybdates have been studied extensively (Brueil *et al* 1973; Samsonov *et al* 1974; Lal and Dar 1976; Koleshko and Bapushkina 1979). Electric and dielectric properties of Tm_2O_3 (bandgap 5.1 eV) in thin film and pressed pellet form have also been studied (Zdanowicz *et al* 1980; Lal and Ashok Kumar 1975). Since little information is available in literature on the SCLC in Tm_2O_3 we have studied the field dependent electrical conductivity in the Ag- Tm_2O_3 -Ag system as a function of thickness and temperature and the possible nature of defects has been discussed.

2. Experimental

Polycrystalline thulium oxide specimens in pellet form have been prepared from spec-pure (JM grade) powder by pressing the powder at a pressure of $(1-4.6) \times 10^7$ kgm/m²

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and baking them in an N_2 atmosphere at ~ 1500 K for 12 hrs. After lapping off the surface layers and polishing of the pellets (cross-section area $2.3 \times 10^{-4} \text{ m}^2$) silver electrodes were deposited by vacuum evaporation on all the samples using molybdenum as boat material at a residual pressure of 5×10^{-5} Torr to form Ag-Tm₂O₃-Ag sandwich systems. The impurity contents in the starting material are listed below:

Element	Er	Al	Ca	Cu	Fe	Ni	Ta
(ppm)	100	100	1000	100	200	500	6000

All the measurements have been carried out in air. The current was measured by an electrometer amplifier (BARC make EA810A). Voltages upto 250 V were supplied from an electronically regulated power supply of range 0–250 V and higher voltages (> 250 V) were applied by a regulated power supply of range 250–3000 V (Aplab 7341 P). The temperature of the specimen was measured by using a precalibrated chromel-alumel thermocouple placed in contact with the specimen. The thermocouple was connected in series with a high impedance panel meter of range $(200 \pm 0.01) \text{ mV}$. Sufficient time was allowed for the constancy of the temperature of the specimen while taking observations at different voltages. The observations were repeated and found to be reproducible. The electrical conductivity of the sample at liquid N_2 temperature (77 K) was measured by dipping the sample holder and the connecting leads (properly shielded) directly into a liquid N_2 bath. Figures 1a, b show the experimental arrangements for J - V charac-

Figure 1. Schematic experimental set-up used for J - V characteristics, (a) at room and high temperatures; (b) at 77 K.

teristics at room temperature and above and at 77 K and are self explanatory. Figure 2 shows current density-voltage (J - V) characteristics, on log-log scale for sintered samples prepared at a pressure $1 \times 10^7 \text{ kgm/m}^2$ and having different thicknesses at 300 K. J varies linearly with the applied voltages upto 100–180 V at different temperatures (77–350 K) and no change in the magnitude of J is observed while reversing the polarity of the applied voltage. Hence silver contacts formed on the pellets are ohmic. Electrical conductivity is found to vary linearly with the pressure $[(1\text{--}4.6) \times 10^7 \text{ kgm/m}^2]$ applied while preparing the samples. Figure 3, on the other hand, illustrates the J - V characteristics of one of the samples (thickness $0.8872 \times 10^{-3} \text{ m}$) at different temperatures. J - V characteristics follow the same trend in samples prepared at different pressures from $(1\text{--}4.6) \times 10^7 \text{ kgm/m}^2$.

Figure 2. J - V characteristics at 300 K for pellets of different thicknesses — $\bullet$ $2.6154 \times 10^{-3} \text{ m}$, $\blacktriangle$ $1.9624 \times 10^{-3} \text{ m}$, $\circ$ $1.258 \times 10^{-3} \text{ m}$, Δ $1.008 \times 10^{-3} \text{ m}$, $\square$ $0.8872 \times 10^{-3} \text{ m}$. (Inset) Variation of J/V^2 with s^{-3} at 300 K.

Figure 3. J - V characteristics for a pellet ($s = 0.8872 \times 10^{-3} \text{ m}$) at different temperatures — Δ 77 K, $\bullet$ 300 K, $\circ$ 313 K, $\square$ 350 K. (Inset) Variation of $\ln \theta$ with $10^3/T$.

3. Discussions

J - V characteristics (figures 2 and 3) show that in all samples at low voltages, the conduction is ohmic while at higher voltages, the current has a quadratic dependence on voltages. It is also seen that transition voltage (V_T) varies from 180 V to 100 V as temperature increases from 77 K to 350 K (table 1). The observed ohmic dependence of electrical conductivity at low fields can be explained by the fact that in the studied samples bulk limited current exceeds the SCLC. The plots of $\log J/V^2$ versus V^{-1} are found to be nonlinear (not shown) which means that non-ohmic regions in J - V curves can not be explained by the tunnel effect of Fowler-Nordheim type. The plots of $\log J$ versus $V^{1/2}$ and $\log (J-J_0)$ which is the algebraic subtraction of ohmic current density J_0 from the total current density J versus $V^{1/2}$ have also been found to be nonlinear

(not shown), thereby rejecting the possibility of an explanation of J - V curves on the basis of the Rose theory of exponential trap distribution and the Poole-Frenkel effect respectively. Hence the non-ohmic portions of the J - V curves might be explained on the basis of the shallow trap model (Rose 1955), according to which current density J is given by

$$J = (9 \mu \epsilon_r \epsilon_0 \theta V^2) / (8 s^3) \quad (1)$$

where μ is the mobility, ϵ_r the relative permittivity of the medium, ϵ_0 the permittivity of

Table 1. The values of V_T , θ and n_0 at different temperatures

T(K)	V_T (Volts)	$\theta \times 10^5$	$n_0 \times 10^{-12} (\text{m}^{-3})$
350	100	3.268 ± 0.049	2.5228
323	120	2.937 ± 0.059	2.7207
313	135	1.753 ± 0.049	1.8267
300	165	1.635 ± 0.009	2.0825
77	180	1.145 ± 0.053	1.5910

Figure 4. Variation of $J-J_0$ with V^2 at 300 K for pellets of different thicknesses —●— $2.6154 \times 10^{-3} \text{ m}$, ▲ $1.9624 \times 10^{-3} \text{ m}$, ○ $1.258 \times 10^{-3} \text{ m}$, △ $1.008 \times 10^{-3} \text{ m}$, □ $0.8872 \times 10^{-3} \text{ m}$.

free space, s the thickness of the specimen, V the applied voltage and

$$\theta = (N_c/N_t) \exp(-E_t/k_B T) \quad (2)$$

where N_c is the effective density of states in the conduction band, N_t is the trap density of a shallow trap at energy E_t below the conduction band and k_B the Boltzmann constant; it was assumed that free space charge density is negligible in comparison to the total trapped space charge density ($\theta \ll 1$). Voltage at which conduction changes from ohmic to non-ohmic is given by (Lampert *et al* 1959):

$$V_T = (en_0 s^2)/(\theta \epsilon_r \epsilon_0) \quad (3)$$

where e is electronic charge and n_0 the volume generated free carriers.

The straight lines obtained in $J-J_0$ versus V^2 curves on the log-log scale in the square law region for pellets of different thickness at 300 K (figure 4) and for a pellet of thickness 0.8872×10^{-3} m at different temperatures (figure 5) show the validity of (1). The straight line (inset of figure 2) in J/V^2 versus s^{-3} graph further supports the

Figure 5. Variation of $J-J_0$ with V^2 for a pellet of thickness 0.8872×10^{-3} m at different temperatures — △ 77 K ● 300 K, ○ 313 K, □ 350 K.

validity of (1). The horizontal bars in the graph represent the errors in the measurement of s .

The calculated values of θ from (1) by the least square fit method taking $\epsilon_r = 11$ (Koleshko and Bapushikna 1979), $\mu = 10^{-2} \text{ m}^2/\text{V sec}$ (assumed) are listed in table 1. It also includes the values of transition voltages (V_T) obtained from figure 3 and n_0 calculated from (3).

The plot of $\ln \theta$ versus $10^3/T$ has been represented in the inset of figure 3 where horizontal bars represent the errors in the measurement of temperature. Assuming that N_c and N_t are not strong functions of temperatures, the intercept at $10^3/T$ being equal to zero yields as ratio $N_c/N_t = 1.8 \times 10^{-2}$ which on substitution in (2) gives a trap level at a depth of $\sim 0.12 \text{ eV}$ below the conduction band.

4. Conclusions

The quadratic dependence of current density on voltage ($J \propto V^2$) in the non-ohmic region of J - V characteristics in polycrystalline Tm_2O_3 at different temperatures has been interpreted in terms of SCLC in defect insulators containing shallow traps which exist at energy level $\sim 0.12 \text{ eV}$ below the conduction band.

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