JOURNAL OF MATERIALS SCIENCE 27 (1992) 5040–5044

D.c. conduction studies on thermally evaporated neodymium oxide thin films

M. D. KANNAN, SA. K. NARAYANDASS, C. BALASUBRAMANIAN, D. MANGALARAJ
Department of Physics, Bharathiar University, Coimbatore 641 046, India

Aluminium–neodymium oxide–aluminium thin film capacitors have been prepared by thermal evaporation and the d.c. conduction properties of these films have been studied. The thicknesses of the films have been determined by a multiple beam interferometer. The current–voltage power-law dependence showed that the conduction in these films is space-charge limited. The linear dependence of the current density on the square root of the applied field confirmed the exponential trap distribution. The trap density has been found to be of the order of 10^{26} m^{-3}. It has also been observed that the Schottky type of conduction is predominant in the high-field region and the height of the Schottky barrier has been determined. It is seen that the conduction mechanism is an activated process with the activation energy decreasing with increasing field.

1. Introduction

Rare-earth oxides, because of their high thermal and chemical stability and good radiation resistance seem to be an important group of materials which can be used in thin-film technology. Fromhold and Foster reported that some of these oxides showed most promising properties as potential materials for use in thin-film capacitors [1]. In recent years, considerable interest has been stimulated in the study of direct current conduction [2–4] in thin dielectric layers because of their use in passivated devices, field-effect transistors and microcircuitry. Electrical conduction in rare-earth oxide films has been studied by various workers [5–14]. Neodymium oxide is representative of this group, on which some work has been done [15–22], although as far as we know, a d.c. conduction study on thermally evaporated neodymium oxide thin films has not yet been investigated. Dharmadhikari has reported [18] the electrical charge transport in amorphous Nd_2O_3 thin films prepared by electron beam gun evaporation and the measurements were made over smaller temperature and voltage ranges. With this in mind an attempt has been made to study the d.c. conduction properties of thermally evaporated neodymium oxide thin films.

2. Experimental procedure

Aluminium–neodymium oxide–aluminium film capacitors were prepared, using a 0.3 m conventional vacuum coating unit (Hind Hivac, India), on well-cleaned glass substrates. First, pure aluminium (99.999%) was evaporated through a suitable mask to form the base electrode. Then neodymium oxide powder (99.99%) was evaporated from a tungsten conical basket to form the dielectric layer. Prior to evaporation, the neodymium oxide powder was degassed for about 30 min. The rate of evaporation was properly controlled and was maintained at 0.05 nm s^{-1}. Finally, aluminium was again evaporated to complete the metal–insulator–metal structure. The entire evaporation process was carried out under a vacuum of 2.66×10^{-3} Pa.

The thickness of dielectric films was measured using a multiple beam interferometer (Fizeau fringes). The d.c. electrical measurements were made on well-stabilized capacitors in a rough vacuum ($< 10^{-2}$ torr; 1 torr = 133.322 Pa) at different temperatures, using an ECIL electrometer amplifier (Model EA815) and a d.c. dual power supply. The temperature of the sample was determined using a pre-calibrated copper–constantan thermocouple.

3. Results and discussion

Fig. 1 presents the current–voltage (I–V) characteristics of neodymium oxide film of thickness 97 nm for different temperatures on a double logarithmic scale. The current, I, exhibits a voltage, V, dependence of the form $I \propto V^n$, where n is found to vary from 1.4–1.5 at low fields and attains a value greater than 4 at high fields. However, in very low fields, $n \approx 1$ and as the temperature increases the two regions observed (below 393 K) tend to disappear.

When referring to the power-law dependence of current upon voltage in the context of dielectric films, one naturally tends to think of space-charge limited conduction (SCLC) in solids [23]. Also, in the SCLC theory [24, 25], the current requires a thickness dependence of the form $I \propto d^{-n}$, where n is a parameter that depends on the trap distribution ($n \geqslant 3$). The thickness dependence of the current has been studied and is shown in Fig. 2. The slope of the I versus d plot (-4.4) being greater than -3 confirms that the

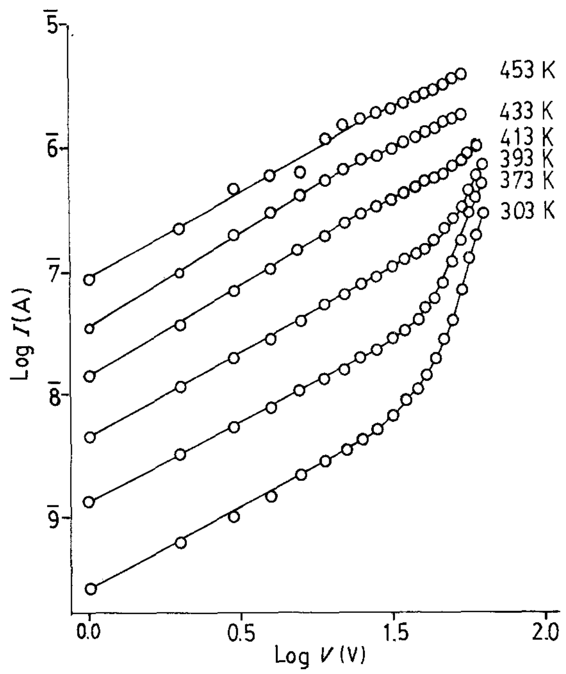

Figure 1 Current–voltage characteristics (d = 97 nm).

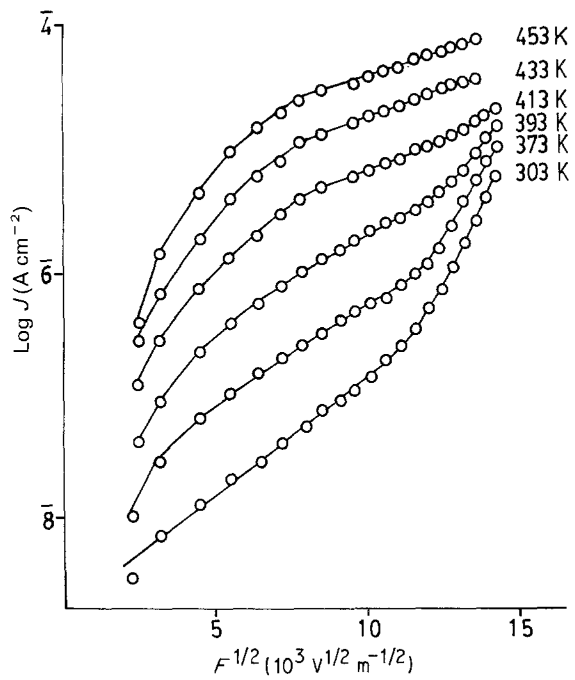

Figure 3 Variations of the current density with the square root of the field at different temperatures.

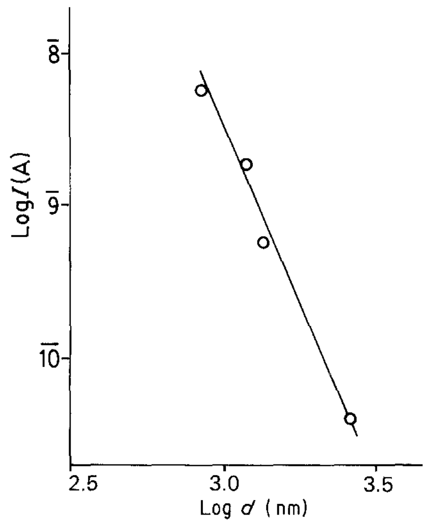

Figure 2 Thickness dependence of the current for $V = 3$ V (slope = − 4.4).

observed currents are actually space-charge limited. The field dependence of the current density is shown in Fig. 3 ($\log J$ versus $F^{1/2}$). Such a linear plot in the moderate high-field region indicates that the conduction mechanism may be of either Schottky or Poole–Frenkel type. The Schottky effect is the conduction of electrons by thermal excitation over the potential barrier between electrons at the Fermi level of the injecting electrode and electrons in the insulator conduction band. The high field, which exists in the film, causes a lowering of the potential barrier height and the conduction relation is

$$J = J_{Sc} T^2 \exp[(\beta_{Sc} F^{1/2} - \phi)/(kT)] \qquad (1)$$

where β_{Sc} is the Schottky field-lowering coefficient, F is the field and ϕ is the barrier height.

Poole–Frenkel conduction occurs as a field emission of electrons from localized donors located at an energy, ϕ, below the insulator conduction band. The current density equation in this case is

$$J = J_{PF} \exp[(\beta_{PF} F^{1/2} - \phi)/(kT)] \qquad (2)$$

The field-lowering coefficient, β, is given by

$$\beta = [e^3/(a\pi\varepsilon_0\varepsilon_r)]^{1/2} \qquad (3)$$

where e is the electronic charge, $a = 1$ for PF emission and 4 for Schottky emission, ε_0 is the permittivity of free space and ε_r is the high frequency dielectric constant. However, Soukup and Speliotis [26] have observed that there could be no *a priori* reason for taking the high-frequency dielectric constant, and Hill [27] has also pointed out that β_{Sc} is a function of both the low- and the high-frequency values of the dielectric constant. The values thus obtained for Schottky, β_{Sc}, and Poole–Frenkel, β_{PF}, are 1.11×10^{-24} and 2.22×10^{-24} eV V$^{-1/2}$ cm$^{1/2}$, respectively. The dielectric constant, ε_r, for the calculation has been taken as 5.65 [15, 16]. The experimental value of β (1.256×10^{-24} eV V$^{-1/2}$ cm$^{1/2}$) has been determined from the plots of $\log J$ versus $F^{1/2}$ (Fig. 3) and g versus the inverse absolute temperature (Fig. 4). The values of g are the slopes determined from Fig. 3 in the high-field region. Table I compares the experimental and theoretical values of β. From the table it is clear that the experimental value is closer to the calculated β_{Sc} than β_{PF}. Hence it can be proposed that the dominating conduction mechanism for thermally evaporated neodymium oxide thin films may be of the Schottky type.

TABLE I Theoretical and experimental values of β

β (10^{-24} eV V$^{-1/2}$ cm$^{1/2}$)	
Theoretical	Experimental
β_{Sc} 1.11	1.256
β_{PF} 2.22	

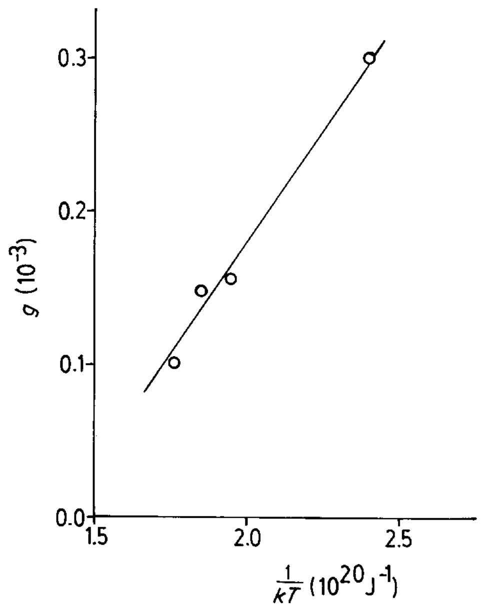

Figure 4 The inverse absolute temperature dependence of the slopes observed from the log J versus $F^{1/2}$ plots in the high-field region.

However, Hughes and Jones [28] and Gould and Hogarth [29] have reported that a mere coincidence of the experimental β_{Sc} with the theoretical value cannot be taken as a deciding factor for the conduction mechanism responsible. Therefore, in order to confirm the observed β_{Sc} dependence with the theoretical β [30], a graph between $\log(J/T^2)$ and $1/T$ (Schottky plot) (Fig. 5) was plotted for different voltages. The resulting straight lines observed in Fig. 5 confirm the Schottky type of conduction mechanism.

Thus from the above discussion it can be concluded that the conduction mechanism in Nd_2O_3 thin films is of the Schottky type. The next step is to determine the height of the Schottky barrier. J/T^2 is plotted on a logarithmic scale against inverse absolute temperature (Fig. 5) for various voltages and from the slopes $\phi = \beta_{Sc} F^{1/2} - \phi_0$ has been determined. The values of

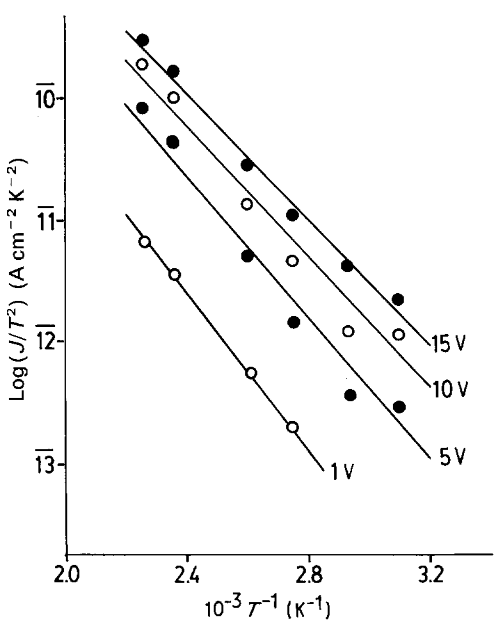

Figure 5 Plot of $\log(J/T^2)$ versus $1/T$ at various applied voltages (d = 97 nm).

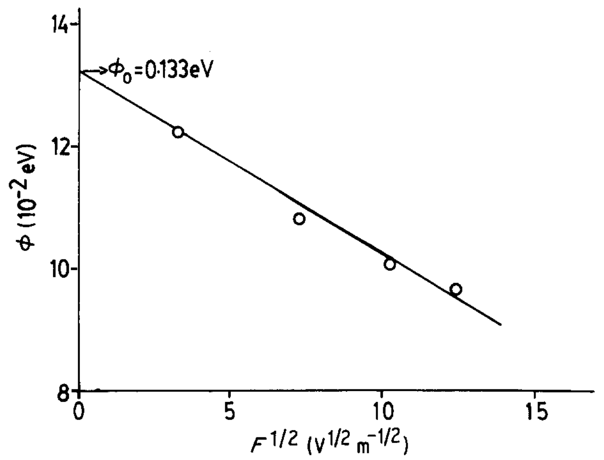

Figure 6 ϕ versus $F^{1/2}$ for the same film as in Fig. 5.

ϕ thus determined have been plotted against the square root of the applied field and an extrapolation of these linear plots to zero field ($F^{1/2} = 0$) yielded ϕ_0 (0.133 eV) which is shown in Fig. 6.

The zero-field current density, J_0, was determined from Fig. 3 by extrapolating the linear plots to zero field. When $\log(J_0/T^2)$ was plotted against inverse absolute temperature (Fig. 7) a straight line plot was obtained and from the slope of which ϕ_0 was calculated. The values of ϕ_0 determined by the above methods are shown in Table II.

The activation energy was determined at different constant applied voltages using the relation

$$I = I_0 \exp[-E/(kT)] \qquad (4)$$

by plotting the current density on a logarithmic scale against the inverse absolute temperature (Fig. 8). The

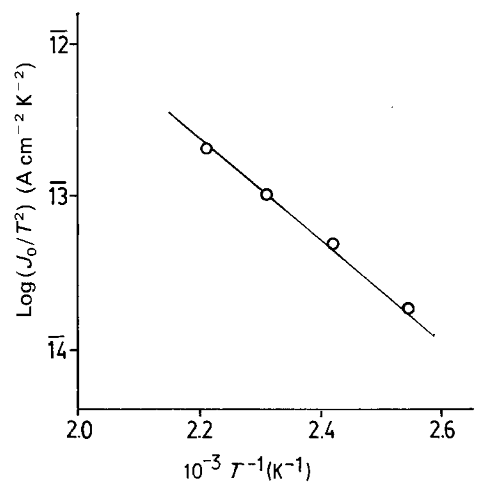

Figure 7 Log (J_0/T^2) versus $1/T$ for the same film as in Fig. 5.

TABLE II Schottky barrier height; ϕ_0 determined by different methods

	Method I[a]	Method II[b]	Method III[c]
ϕ_0 (eV)	0.133	0.136	0.130

[a] From the plot of log (J/T^2) against $1/T$.
[b] From the plot of log (J_0/T^2) against $1/T$.
[c] From the plot of ϕ versus $V^{1/2}$.

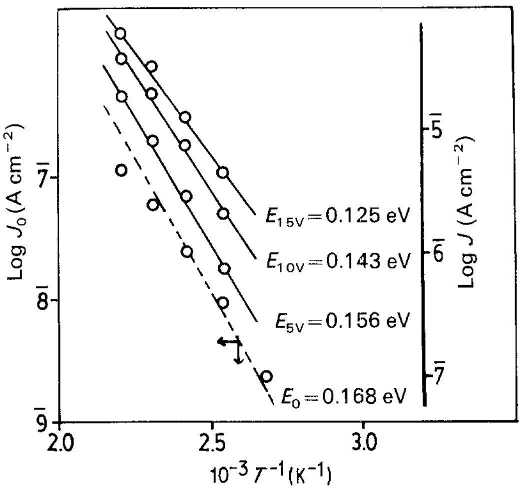

Figure 8 The inverse absolute temperature dependence of $\log J_0$ and $\log J$ at different applied voltages (slopes: at zero field = − 4500, at 5 V = − 4167, 10 V = − 3833 and 15 V = − 3333).

TABLE III Activation energy as a function of applied voltage

Voltage (V)	Activation energy (eV)
5	0.156
10	0.143
15	0.125

activation energy determined by this method is shown in Table III. It can be seen that the activation energy decreases with increasing applied voltage. The zero-field activation energy has been found to be 0.168 eV. This value of activation energy may be associated with traps in the energy gap. Also the linear dependence of these curves along with the significant variations in the slope between the curves obtained for different voltage levels (0 V = − 4500, 5 V = − 4167, 10 V = − 3833 and 15 V = − 3333) implied the existence of an exponential distribution of trapping levels rather than of a single discrete trapping level below the conduction band edge of the oxide.

The exponential distribution of traps below the conduction band edge is defined by Lampert [31] as

$$N(E) = N_0 \exp[-E/kT_t)] \quad (5)$$

where $N(E)$ is the trap density per unit energy range at an energy E below the conduction band edge, N_0 is the value of $N(E)$ at the conduction band edge, T_t is a temperature parameter characterizing the exponential trap distribution and k is the Boltzmann's constant. The current density–voltage power law characteristic is also given by

$$J \approx e\,\mu\,N_c\,[\varepsilon/(eN_{t(e)})]^l\,V^{l+1}/d^{2l+1} \quad (6)$$

where e is the electronic charge, μ the mobility, N_c the effective density of states in the conduction band, ε the permittivity, $N_{t(e)} = N_0\,kT_t$ [32] the total density of trapping levels in the exponential distribution, and $l = T_t/T$, where T is the lattice temperature.

Gould [33] has shown that the slopes obtained from Fig. 8 for different constant voltages is given by

$$\text{slope} = T_t \log[\varepsilon V/(ed^2 N_t)] \quad (7)$$

From this the trap density, N_t, and the corresponding value of the trapping density per unit energy range at the conduction band edge, N_0, thus determined for $V = 5$ V are $1.091 \times 10^{26}\ \text{m}^{-3}$ and $1.56 \times 10^{46}\ \text{J}^{-1}\,\text{m}^{-3}$, respectively.

4. Conclusion

The current–voltage characteristics for thermally evaporated Nd_2O_3 thin films have been studied. In the wider field and temperature ranges studied it has been shown that both the space-charge limited and Schottky type of conduction contribute to the conduction in these films. The activation energy for the conduction process has been found to be associated with the traps in the energy gap and is field dependent. It has also been confirmed from the plot of current density versus inverse absolute temperature that the distribution of trap density in the band gap at energies below the conduction band edge is exponential and the trap density has been found to be of the order of $10^{26}\ \text{m}^{-3}$.

Acknowledgement

One of the authors (M. D. K.) thanks the Bharathiar University for the award of a research fellowship.

References

1. A. T. FROMHOLD Jr and W. D. FOSTER, *Electrocomponent Sci. Technol.* **3** (1976) 51.
2. D. L. PULFREY, A. H. M. SHOUSHA and L. YOUNG, *J. Appl. Phys.* **41** (1970) 2838.
3. L. SULLIVAN and H. C. CARD, *J. Phys. D* **7** (1974) 1531.
4. A. M. PHAHLE, *Thin Solid Films* **46** (1977) 315.
5. A. T. FROMHOLD Jr, *Phys. Status Solidi* **36** (1969) K129.
6. M. SAYER, M. SUZANNE MARTIN and N. J. HELLICAR, *Thin Solid Films* **6** (1970) R61.
7. E. RIEMANN and L. YOUNG, *J. Appl. Phys.* **44** (1973) 1044.
8. T. ZDANOWIEZ, B. JANKOWSKI and L. ZDANOWICZ, *Acta Phys. Polonica* **A57** (1980) 151.
9. A. GOSWAMI and R. RAMESH VARMA, *Thin Solid Films* **22** (1974) S2.
10. C. R. DUTTA and K. BARUA, *ibid.* **103** (1983) 295.
11. S. R. JAWALEKAR, *Ind. J. Pure Appl. Phys.* **7** (1969) 801.
12. C. K. CAMPBELL, *Thin Solid Films* **6** (1970) 197.
13. A. GOSWAMI and R. GOSWAMI, *ibid.* **20** (1974) 3.
14. A. GOSWAMI and R. RAMESH VARMA, *ibid.* **28** (1975) 157.
15. M. D. KANNAN, SA. K. NARAYANDASS, C. BALASUBRAMANIAN and D. MANGALARAJ, *Phys. Status Solidi (a)* **121** (1990) 515.
16. *Idem*, in "Proceedings of the 78th Indian Science Congress', S. N. Thakur (Sec. Pdt.), ISCA, Calcutta, January 1991, p. 6.
17. V. S. DHARMADHIKARI and A. GOSWAMI, *Thin Solid Films* **87** (1982) 119.
18. VINEET S. DHARMADHIKARI, *ibid.* **103** (1983) 311.
19. D. I. CHERNOBROVKIN and V. V. BAKHTINOV, *Izv. Vyssh. Uchebn. Zaved. Fiz.* **14** (1971) 148.
20. V. S. KRIKOROV, B. J. KRASOV and A. E. MARKARYANTS, *Electron Technol.* **12** (1971) 79.
21. A. T. FROMHOLD Jr and W. D. FOSTER, *Electrocomp. Sci. Technol.* **3** (1976) 51.
22. R. D. GOULD, *Thin Solid Films* **110** (1983) L121.
23. A. K. JONSCHER, *ibid.* **1** (1967) 213.
24. A. C. RASTOGI and K. L. CHOPRA, *ibid.* **26** (1975) 61.
25. L. V. GREGOR and L. H. KAPLAN, *ibid.* **2** (1968) 95.
26. R. J. SOUKUP and D. E. SPELIOTIS, *J. Appl. Phys.* **41** (1970) 3229.

27. R. M. HILL, *Philos. Mag.* **23** (1971) 59.
28. D. M. HUGHES and M. W. JONES, *J. Phys. D* **7** (1974) 2081.
29. R. D. GOULD and C. A. HOGARTH, *Int. J. Electron.* **38** (1975) 577.
30. P. J. REUCROFT and S. K. GHOSH, *Thin Solid Films* **20** (1974) 363.
31. M. A. LAMPERT, *Rep. Prog. Phys.* **27** (1964) 329.
32. R. D. GOULD and M. S. RAHMAN, *J. Phys. D* **14** (1981) 79.
33. R. D. GOULD, *J. Appl. Phys.* **53** (1982) 3353.

Received 22 May
and accepted 27 November 1991