JOURNAL OF MATERIALS SCIENCE 25 (1990) 2901–2905

Electrical conduction through co-evaporated SiO_x–SnO thin films

G. A. KHAN, C. A. HOGARTH
Department of Physics, Brunel University, Uxbridge, Middlesex UB8 3PH, UK

D.c. electrical properties of SiO_x–SnO (300 nm) thin films forming part of a metal–insulator–metal (MIM) structure are reported. The conduction process is related to the field-assisted thermal excitation of electrons from the trapping centres in the forbidden band gap to the conduction band (Poole–Frenkel effect). The energy gap between the trapping level and the bottom of the conduction band in 80% SiO_x–20% SnO in thin films (300 nm) is found to be equal to 0.50 eV. The level of conduction in the composite films is observed to increase with an increase in the content of SnO.

1. Introduction

The technological importance of dielectric films in the electronics industry is reflected to some extent in the fundamental study of their electrical properties. The amorphous or glassy structure of most dielectric films in common use does not represent a well-defined state of matter and may also encourage the migration of ionic species, giving rise to time-dependent phenomena [1]. In principle, any dielectric film should show an ohmic region of current–voltage characteristic at sufficiently low electric fields [2]. In practice, many films other than those which are heavily doped or contaminated do not show this region because of very low-voltage polarization phenomena at the contacts or in the bulk [3]. The ohmic region is normally characterized by high activation energies of the order of 1 eV associated with the ionic conduction. There have been numerous reports on the d.c. behaviour of evaporated dielectric films and most of the results show an exponential dependence of current on the square root of applied field (Poole–Frenkel or Schottky effect) in the region of high field of the order of $10^6\,V\,m^{-1}$ [4–9]. The difference between the equations for Schottky and Poole–Frenkel effects are the pre-exponential factor, the activation energy and the slope of the straight part of curve of the log I against $V^{1/2}$ plot [10].

Silicon monoxide is a material which is commonly used as a dielectric medium in thin-film capacitors and as an insulating layer in various vacuum-evaporated devices, such as thin-film transistors [2]. The problem of the structure of dielectric SiO_x remains unresolved except for the agreed fact that the material is amorphous. Electron spin resonance studies by Timson and Hogarth [11] have revealed the presence of unpaired spins in amorphous SiO_x thin films and have shown a correlation between the concentration of spins and the level of d.c. conductivity.

Hill [12, 13] suggested the conduction mechanism as field-enhanced thermally activated hopping from donor-like centres (the Poole–Frenkel mechanism) in the higher range of temperature and tunnelling in the lower-temperature range. Similarly other authors [14–16] suggested the Poole–Frenkel conduction mechanism as being dominant in SiO_x thin films. Johansen [9] and Hartman *et al.* [10] proposed Schottky emission as the high-field conduction mechanism through amorphous thin films of SiO_x.

Taheri *et al.* [17] and Bidadi and Hogarth [18] have shown that co-evaporated layers of SiO with B_2O_3 resulted in improved dielectric properties as compared with the pure SiO layers. Hogarth and Wright [19] investigated amorphous co-evaporated thin films of SiO–TiO and the electrical conductivity of composite oxide films was observed to increase with the increase in TiO content. Bidadi and Hogarth [20] found that SiO–TiO thin-film metal–insulator–metal (MIM) structures undergo an electroforming process when a sufficiently high electric field was applied, having some of the features of dielectric break-down but of non-destructive character, and resulting in an effective decrease in the sample resistance by several orders of magnitude.

SnO_2 has been widely investigated as a relatively high-conducting oxide and used for transparent conducting electrodes on solar cells, transparent heating elements and antistatic coatings on glass. Antimony or phosphorus added to tin oxide increases its conductivity. Ilyas *et al.* [21] studied the high-field electrical conduction in SiO–SnO_2 thin films and confirmed the Schottky high-field process to be the dominant one. It is therefore of some interest to study the behaviour of the SnO when mixed with SiO, and we have studied the electrical conduction processes before electroforming through co-evaporated SiO_x–SnO layers of different compositions.

2. Experimental procedure

The samples for electrical measurements were deposited on 3 in. × 1 in. (76 mm × 25 mm) Corning 7059 borosilicate glass slides. In order to prepare MIM sandwiches, a metal strip was evaporated as the base

0022-2461/90 $03.00 + .12 © *1990 Chapman and Hall Ltd.*

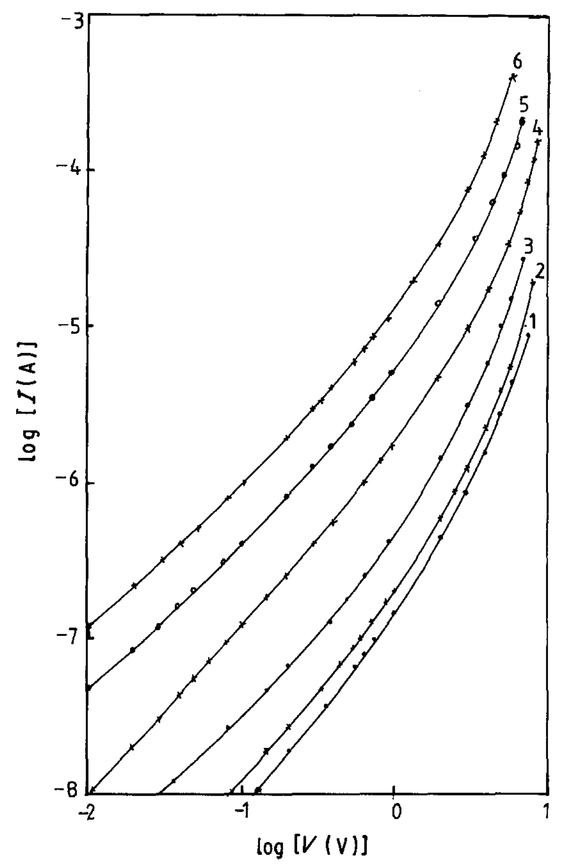

Figure 1 Log I against log V of 80% SiO_x–20% SnO (300 nm) thin films at different temperatures: (1) 182 K, (2) 228 K, (3) 263 K, (4) 297 K, (5) 318 K, (6) 343 K.

electrode followed by mixed oxides from separate sources as insulator using the co-evaporation technique of Hogarth and Wright [19], and finally evaporating metal for the top electrode. The rates of evaporation and film thickness could be controlled. Six devices, each having an effective area 0.1 cm^2, were made on each substrate.

All electrical measurements were taken in a subsidiary vacuum system at a pressure of about 10^{-5} torr (1.33×10^{-3} Pa) and equipped with the necessary facilities. The device was firmly attached to the copper base by pressure contacts, and electrical connections were made to the devices by means of copper strips joined with silver paste. A stainless steel tank incorporated in the top plate was used for low-temperature measurements by pouring liquid nitrogen into it. An insulated resistive wire made of molybdenum ribbon was inserted in the holes at the copper base and used as a heater element. The temperature measurements were made by attaching a Chromel–Alumel thermocouple to the substrate which was connected to a Comark electronic thermometer type 1601 Cr/A. The d.c. voltage–current characteristics were measured in a conventional manner. The bias voltage was supplied by a Coutant type LA100.2 power supply, and the circulating current was recorded by a Keithley 610C solid-state electrometer. The voltage across the sample was measured by a Farnell digital multimeter model DM131.

The capacitance of the sample was measured by a Hewlett-Packard impedance analyser (5 Hz to 13 mHz) type 4192 ALF using an a.c. signal of 500 mV.

3. Results and discussion

The possible conduction mechanisms through thin insulating films are (a) tunnelling, (b) ionic conduction, (c) space-charge-limited flow, (d) Schottky effect and (e) Poole–Frenkel effect. As the thickness of our films is well above 10 nm, the tunnelling process is ruled out for our samples. The values of the activation energy calculated (see Fig. 5 below) in our case are ≈ 0.40 eV and suggest the conduction to be due to electronic charge transfer and not to the motion of ions. Fig. 1 shows the plots of log(current) against log(voltage) of 80% SiO_x–20% SnO (300 nm) thin films in an MIM sandwich structure. It can be seen that the conduction is almost ohmic for low voltages and non-ohmic for higher voltages. The ohmic behaviour may be attributed to the presence of silicon and tin ions as previously observed [4]. If the non-ohmic conduction is due to space-charge-limited current flow then we should get a straight line in I against V^2 plots which is not found in our case, as can be seen in Fig. 2. So the only possible conduction mechanisms at higher fields in the present case could be the Schottky effect or the Poole–Frenkel effect.

The I–V characteristics of thin insulating films according to the Poole–Frenkel effect or Schottky effect take the form

$$I \propto \exp\left(\frac{\beta E^{1/2} - V_g}{kT}\right) \tag{1}$$

where E is the electric field applied across the device, k is Boltzmann's constant and β is the barrier-lowering coefficient; V_g is the energy interval between the trap level and the bottom of the conduction band in the case of the Poole-Frenkel effect, or the work function in the case of the Schottky effect. The constant β is defined as

$$\beta = (e^3/\alpha\pi\varepsilon_0\varepsilon_r)^{1/2} \tag{2}$$

where e is the electronic charge, ε_r is the relative dielectric constant of the material and α is a constant with its value equal to 1 or 4 for Poole–Frenkel or Schottky effect, respectively. Fig. 3 shows the plots of log I against $V^{1/2}$ of 80% SiO_x–20% SnO (300 nm) at different temperatures according to Equation 1. The existence of linear relations for high voltages can be clearly realised, which suggests the occurrence of either the Poole–Frenkel or the Schottky conduction mechanism. The experimental value of the constant β is determined from the slope (m = 0.76 at room temperature) of the straight parts of the curves in Fig. 3 according to Equation 1 using the relation

$$\beta = mkTd^{1/2}/0.4343$$

where d is the thickness of the film. The calculated value of β comes out to be 2.4×10^{-5} eV $V^{-1/2}$ $m^{1/2}$. The theoretical Poole–Frenkel value $\beta_{P\text{-}F}$ determined according to Equation 2 at high frequency (100 kHz) where the relative dielectric constant is 8.3 at room temperature is 2.39×10^{-5} eV $V^{-1/2}$ $m^{1/2}$, whereas the Schottky value β_S is 1.19×10^{-5} eV $V^{-1/2}$ $m^{1/2}$. It is obvious that the experimental value of β is almost equal to the theoretical value of $\beta_{P\text{-}F}$, while β_S is not comparable with the experimental value of β. It can'

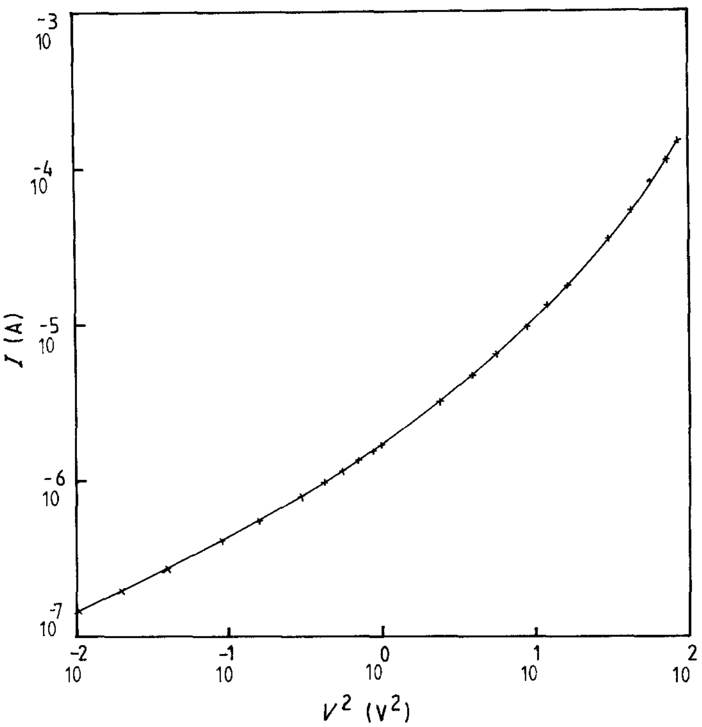

Figure 2 I against V^2 for 80% SiO_x–20% SnO (300 nm) thin films at 318 K.

also be seen in Fig. 3 that the slopes of the plots at different temperatures give almost a constant value of β_{exp}, indicating that β_{exp} is independent of temperature. This further suggests that β_{exp} in our case does not correspond to the Schottky effect since β_S is expected to be temperature-dependent.

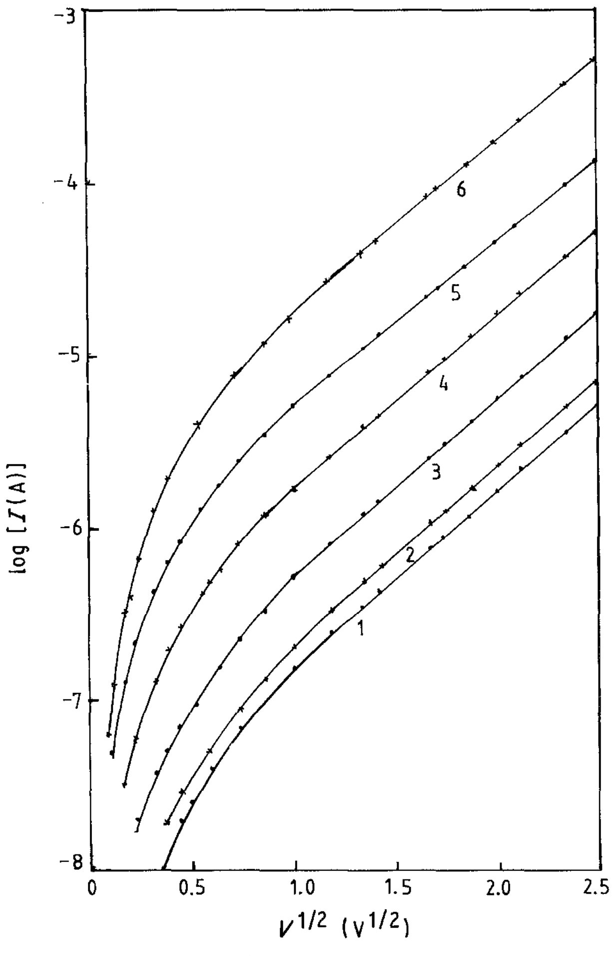

Figure 3 Log I against $V^{1/2}$ of 80% SiO_x–20% SnO (300 nm) thin films under the same conditions as in Fig. 1.

Fig. 4 shows the I–V characteristics of 80% SiO_x–20% SnO (300 nm) for both positive and negative polarity of top electrode. There is no change in the conductivity, indicating the conduction process to be bulk-limited and not electrode-limited. Fig. 5 represents the plots of log I against $1/T$ for the same device at different fields. The activation energies calculated from the slopes of the straight parts of the curves lie in the range 0.37 to 0.47 eV for the different

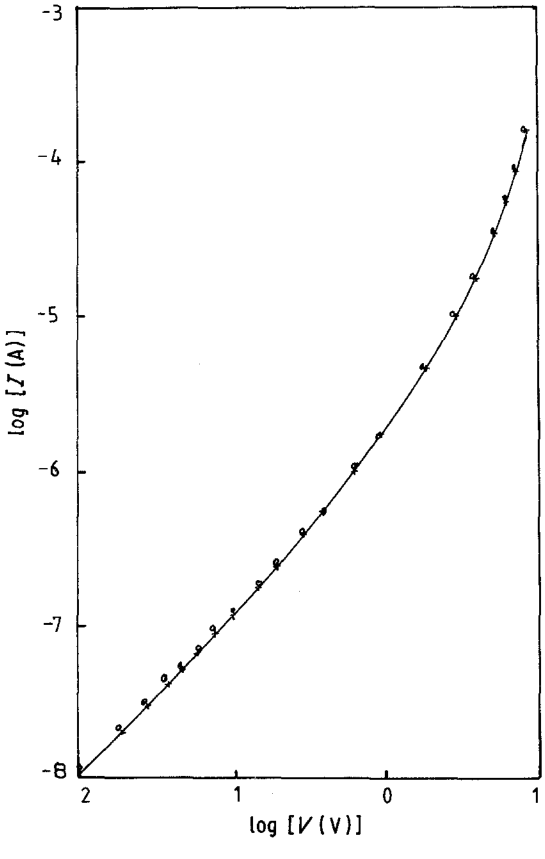

Figure 4 Log I against log V of 80% SiO_x–SnO (300 nm) thin films at different polarities of top electrode.

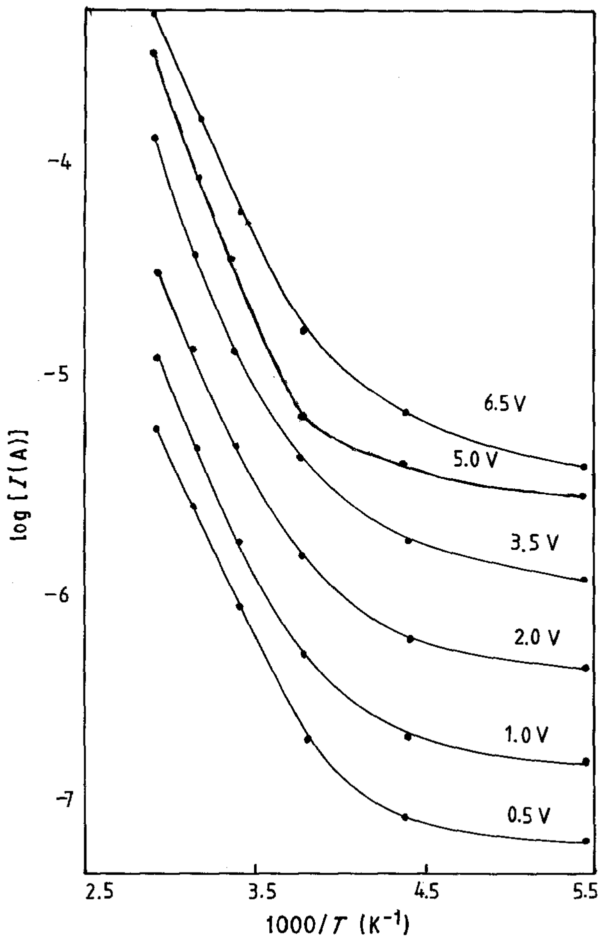

Figure 5 Log I against inverse of temperature for 80% SiO_x–20% SnO (300 nm) thin films at different voltages.

fields. The activation energy first increases and then decreases with the field, a result which is in good agreement with the behaviour observed earlier by Hirose and Wada [4] in SiO_x thin films.

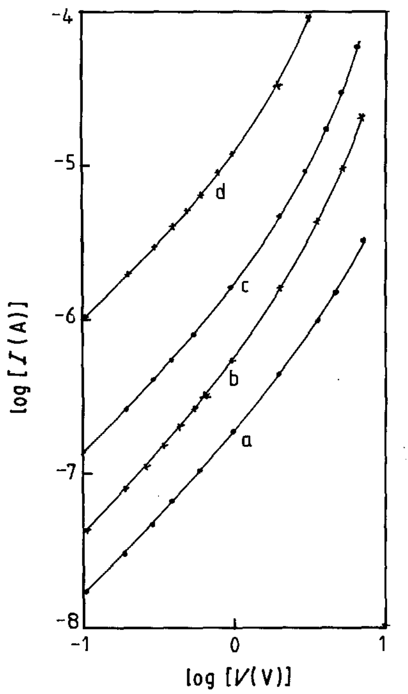

Figure 6 Log I against log V of SiO_x–SnO (300 nm) thin films with different compositions: (a) 10%, (b) 15%, (c) 20%, (d) 30% SnO.

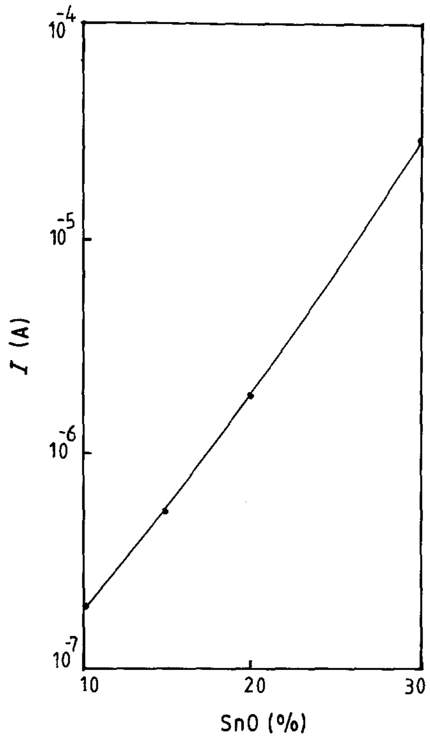

Figure 7 Plot showing variation of circulating current with SnO content in SiO_x–SnO (300 nm) thin films for a fixed applied voltage of 2 V.

The current flowing through the sample in terms of activation energy $\Delta\phi$ can be written as

$$I \propto \exp(-\Delta\phi/kT) \tag{3}$$

By comparison of Equations 1 and 3 we can write

$$V_g = \Delta\phi + \beta E^{1/2} \tag{4}$$

By using $\Delta\phi$ as 0.40 eV and the corresponding values of β and E in Equation 4 we obtain the value of the energy gap between the trap level and the bottom of the conduction band as 0.50 eV. The value of $\Delta\phi$ for pure SiO_x thin film as found by Hirose and Wada [4] for the Poole–Frenkel effect is 0.54 eV. So we propose that the energy interval between the trap level and the conduction band decreases with an increase in the content of SnO in SiO_x–SnO composite thin films.

In the light of the above observations we propose that the conduction process in our case is due to field-assisted excitation of trapped electrons in the forbidden gap into the conduction band (Poole–Frenkel effect) of thin-film samples. The level of conduction in SiO_x–SnO films is observed to increase with SnO content as can be seen in Figs 6 and 7. This can be attributed to the decrease in $\Delta\phi$, i.e. to the creation of more donor-like states within the forbidden gap with the increase in SnO concentration. This behaviour is in good agreement with the observation made by us elsewhere while considering the optical energy gap of the same material, where the optical energy gap is observed to decrease with an increase in the content of SnO in SiO_x–SnO composite films.

4. Conclusion

The conduction process at high fields but before electroforming in SiO_x–SnO thin films is governed by the Poole–Frenkel effect, the experimental value of β_{P-F} being in very good agreement with the theoretical value. The conductivity of our co-evaporated thin films increases with an increase in the content of SnO, which is related to the increase in impurity states in the band gap. The value of V_g for 80% SiO_x–20% SnO is 0.50 eV, which is somewhat less than the value for pure SiO_x (0.54 eV).

References

1. A. E. HILL, A. M. PHAHLE and J. H. CALDERWOOD, *Thin Solid Films* **5** (1970) 287.
2. F. ARGALL and A. K. JONSCHER, *ibid* **2** (1968) 185.
3. A. SERVINI and A. K. JONSCHER, *ibid* **3** (1969) 341.
4. H. HIROSE and Y. WADA, *Jap. J. Appl. Phys.* **4** (1965) 639.
5. C. A. MEAD, *Phys. Rev.*, **128** (1962) 2088.
6. P. R. EMTAGE and W. TANTRAPORN, *Phys. Rev. Lett.* **8** (1962) 267.
7. S. R. POLLOCK, *J. Appl. Phys.* **34** (1963) 877.
8. M. HACKSKAYLO, *ibid.* **35** (1964) 2943.
9. I. T. JOHANSEN, *ibid.* **37** (1966) 499.
10. T. E. HARTMAN, J. C. BLAIR and R. BAUER, *ibid* **37** (1966) 2468.
11. P. A. TIMSON and C. A. HOGARTH, *Thin Solid Films* **8** (1971) 237.
12. R. M. HILL, *Phil. Mag.* **23** (1971) 59.
13. *Idem, ibid.* **24** (1971) 1307.
14. M. MEADURE and R. MEADURE, *ibid.* **40** (1977) 401.
15. M. DEERY, J. G. PERKINS and K. G. STEPHENS, *Thin Solid Films* **8** (1971) R16.
16. A. K. JONSCHER and C. K. LOTT, *J. Phys. C.* **4** (1971) 1341.
17. E. H. Z. TAHERI, C. A. HOGARTH and R. D. GOULD, *Phys. Status Solidi* **12** (1972) 563.
18. H. BIDADI and C. A. HOGARTH, *Int. J. Electron.* **36** (1974) 287.
19. C. A. HOGARTH and L. A. WRIGHT, in Proceedings of International Conference on the Physics of Semiconductors, Moscow, 1968 (edited by S. M. Ryvkin, Nauka, Leningrad, 1968) p. 1274.
20. H. BIDADI and C. A. HOGARTH, *Thin Solid Films* **27** (1975) 319.
21. M. ILYAS, C. A. HOGARTH and R. HARANI, *J. Mater. Sci. Lett.* **3** (1984) 703.

*Received 11 August
and accepted 12 September 1989*