A.C. CONDUCTION BEHAVIOUR IN AMORPHOUS WO_3/CEO_2 THIN FILM.

B. YAGOUBI*, C.A. HOGARTH. A. BOUKORTT*.**

Electronic Department Signal and System Laboratory
Mostaganem University, 27000 Algeria*.
Department of Physics, Brunel University
Uxbridge, Middlesex UB8 3PH, Great Britain**.

Abstract

The a.c. conduction of amorphous thin film of Al-WO_3/CeO_2-Al in which aluminium is used as electrode, is studied in the frequency range 0.1kHz to 10^4kHz. The dielectric response of amorphous 58%WO_3/42%CeO_2 to the applied alternating field is believed to be Debye-like, where the polarisation could be caused by the transition of charge between W^{+6} and Ce^{+4} centres. It is assumed that these centres are sufficiently close in this composition that appreciable interaction occurs, thereby causing the polarisation responsible for this type of conduction. Whereas the a.c. conduction in 81%WO_3/19%CeO_2, behaving as ω^s where s=1 around 10kHz, could be interpreted in terms of Elliott model, that is two electrons are assumed to hop to gather from a D^- to D^+ defect centres over a potential barrier separating them. At frequencies higher than 10kHz, the a.c. conductivity behaves as ω^2 in 81%WO_3/19%CeO_2. This behaviour in this composition is very similar to that of chalcogenide glasses.

Introduction

The a.c. conductivity in a number of thin oxide films and amorphous materials has been investigated [1, 2, 3]. Many experimental results have been gathered, but the conduction mechanism itself in these materials, is not yet well understood. Thin film behaviour may differ from that of the corresponding bulk material. This behaviour depends mostly on the preparation technique used.

In the present work, thin films of mixed WO_3/CeO_2 were prepared by co-evaporation. Most workers agreed that films prepared by this technique present a high state of disorder, which can be observed by electron diffraction. In this paper, we discuss the mechanism of a.c. electrical conduction in two particular compositions of amorphous WO_3/ CeO_2 thin film.

1. Experimental work

The mixed oxide films of WO_3 and CeO_2 of thickness of order 200nm were prepared by co-evaporation in a vacuum better than 10^{-6} torr. The experimental aspect of this technique is presented elsewhere[3]. The a.c. measurements were carried out inside a vacuum system at about 10^{-5} torr to avoid any gaseous contamination and were determined by applying alternating voltage, which was supplied by a Hewlett Packard impedance analyser working in the frequency range 5Hz to 13MHz.

The layers for the electron diffraction investigation were deposited on a carbon-coated mica substrate held at 90°C. The thickness measurement procedure may be found in the references [3, 4].

2. Results and discussion

The mechanism of a.c conduction in 81%WO_3/19%CeO_2 above 10^2kHz shown in figure (1a), is due to hopping of carriers and it is supposed to be confined to immediate neighbouring sites, giving rise to a square law frequency dependence and conductance almost independent of temperature. It is observed that at low frequencies and low temperatures, the a.c. conductance varies as ω^s, where ω is the angular frequency and s is a quantity whose value is close to, but less than unity. Our results in 81%WO_3/19%CeO_2 are very similar to those obtained by Argall and Jonsher[5] on silicon oxide. This behaviour of a.c. conductivity has already been observed experimentally in many amorphous semiconductors and it may be interpreted in terms of Elliott[6] model. The latter is an extension of Pike[7] model which considers a case in which hopping between localised sites is over potential barriers, instead of tunnelling through them, with a height distribution caused by the random spatial distribution of these sites leading to ω^s behaviour with s decreasing linearly with temperature.

Pike [7] suggested that the exponent s should be temperature dependent, in contrast to Austin and Mott [9] formula where s is temperature independent, and it can be approximated by the following expression

$$1-s=\frac{6KT}{W} \tag{1}$$

where T is the absolute temperature, K the Boltzman constant and W is the energy required to remove two electrons from D^- to D^+ and it is given by

$$W=B+W_2-W_1 \tag{2}$$

where B is the band gap of the material, and W_2 and W_1 are approximately the distortion energies associated with the charged D^+ and neutral D dangling bonds [8].

Although equation (1) is, unfortunately, not easy to verify because the difficulty rises in the lack of accuracy in determining the exponent s and the band gap W from different experimental measurements and also to structural heterogeneities, we believe that the a.c. conduction in thin film of 81%WO_3/19%CeO_2, which is similar to that of chalcogenide glasses, can be described by Elliott[6] model. The latter considers that the loss in amorphous solids is caused by the hopping between localised defect states, which are randomly situated in space and energy, hence a broad distribution of relaxation times should ensure, instead of the fixed value that is necessary to cause a Debye-loss peak. In this case and in addition if the relaxation time is an exponential function of some random variable then the a.c. conductivity behaves as ω^s as shown in figure (1a).

Now, let us consider the application of alternating electric field to a sample, producing a polarisation P(t). In the frequency space we have the following relation

$$P(\omega)=\varepsilon_0\chi(\omega)E(\omega) \tag{3}$$

Where P (ω) and E(ω) are the Fourier transform of the polarisation and the electric field respectively, ε_0 is the free space permittivity and χ(ω) is the dielectric susceptibility which is in general a complex function of frequency:

$$\chi(\omega)=\chi'(\omega)-j\chi''(\omega) \tag{4}$$

The dielectric loss ε" is identical to χ"(ω) and further related to the a.c. conductivity by

$$\sigma(\omega)=\varepsilon_0\,\varepsilon''(\omega).\,\omega \tag{5}$$

Let us suppose that the dielectric response of the amorphous solid to the applied alternating field is Debye-like, that is the polarisation which is caused by the transition of a charge between two preferred sites, decays with first-order kinetics on removal of the exciting field, with a characteristic relaxation time, τ such

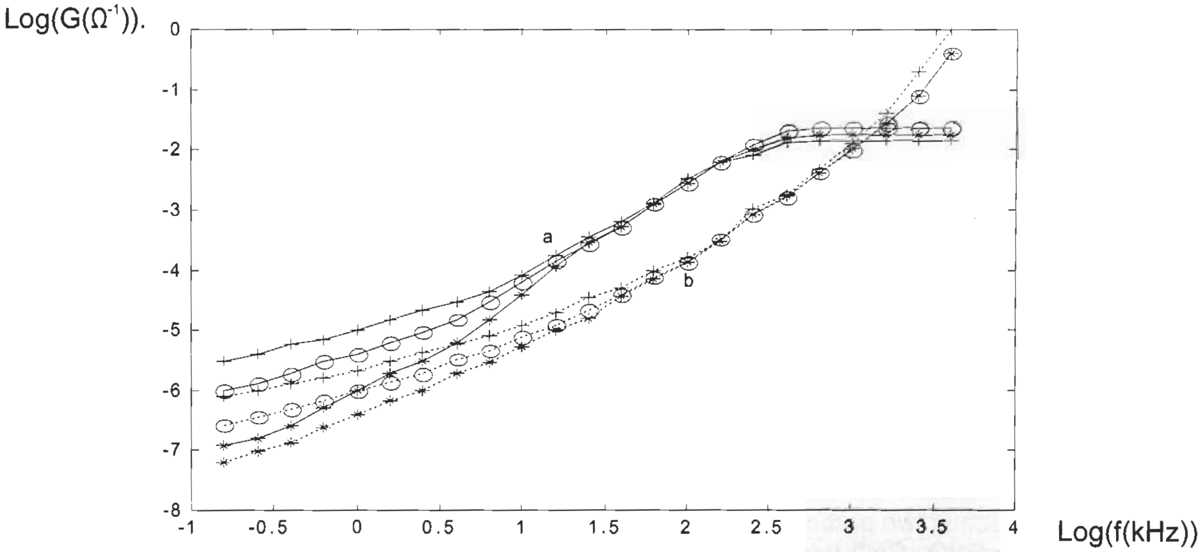
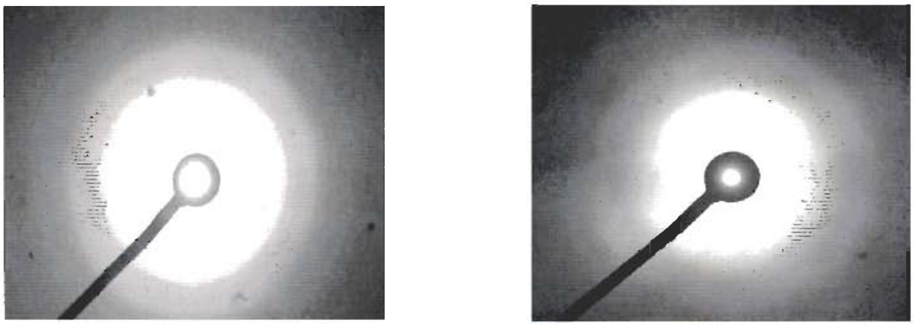


Figure 1. A. C. conductance versus frequency for a) 81%WO_3/19%CeO_2 and b) 58%WO_3/42%CeO_2 with thickness ≈ 200nm and at different temperatures: (*)140K, (0)200K, (+) 260K.



(a) (b)

Figure 2. Electron diffraction for a) 81%WO_3/19%CeO_2 and b) 58%WO_3/42%CeO_2 thin films.

$$\frac{dP}{dt} = -\frac{P}{\tau} \quad (6)$$

The solution is

$$P(t)=P_0e^{-t/\tau} \quad (7)$$

We get in the frequency space the dielectric loss

$$\varepsilon''(\omega)=\frac{\omega\tau\varepsilon''(0)}{1+\omega^2\tau^2} \quad (8)$$

Inserting this relation in equation (5) we obtain the expression of the a.c. conductivity

$$\sigma(\omega)=\varepsilon_0\frac{\omega^2\tau\varepsilon''(0)}{1+\omega^2\tau^2} \quad (9)$$

If τ is constant, theoretically, then the a.c. conductivity varies as ω^2 for $\omega<<1/\tau$. This is in good agreement with our experimental results on 58%WO_3/42%CeO_2 shown in figure (1b) and it corresponds to the region of the a.c. conductivity with frequencies less than 10^2kHz.

The a.c. conductivity tends to become constant for $\omega>>1/\tau$, hence a relaxation time value is estimated around 3.18 10^{-6}s. We believe, therefore, that the dielectric response of amorphous 58%WO_3/42%CeO_2 to the applied alternating field is Deby-like, where the polarisation could be caused by the transition of a charge between W^{+6} and Ce^{+4} centres. We think that these centres are sufficiently close in this composition that appreciable interaction occurs, thereby causing the polarisation responsible for this type of conduction.

Although both compositions 58%WO_3/42%CeO_2 and 81%WO_3/19%CeO_2 are amorphous as it is clearly shown by the electron diffraction in figure (2), the type of the conduction is not the same.

Conclusion

Both proportions are amorphous as it is shown by the electron diffraction. The sample with 81%WO_3 has shown in its a.c. conduction characteristic, a behaviour that is similar to that found in chalcogenide glasses. We believe that in this proportion the a.c. conduction can be interpreted in terms of Elliott model, that is two electrons are assumed to hop to gather from a D^- to D^+ defect centres over a potential barrier separating them. At frequencies higher than 10kHz, the a.c. conductivity behaves as ω^2. Whereas the a.c. conduction in the sample with 58%WO_3 is Deby-like, i.e the a. c conductivity varies as ω^2 for $\omega<<1/\tau$ and becomes constant for $\omega>>1/\tau$.

REFERENCES:

[1] J. KOCKA, O. KLIMA ,E. SIPEK, C. E. NEBEL, J. H. BATTER and M. HOHEISEL, Phys. Rev., B, 45, 6593, 1992

[2] C. NEBEL, R. A. STREET, N. M. JOHNSON and J. KOCKA, Phys. Rev., B, 46, 6789, 1992.

[3] B. YAGOUBI and C. A. HOGARTH ,Revue Internationale des Technologies Avancées, 12, 31, 2000.

[4] E. MARQUEZ, J. M. GONZALEZ-LEAL, R. JIMENEZ-GARAY, S. R LUKIE and D. M. PETROVIC., Phys., D: Appl. Phys., 30, 690 ,1997.

[5] F. ARGALL and A. K. JONSCHER., "Thin Solid Films", 2, 185, 1968.

[6] S. R. ELLIOTT., Phil. Mag., 36, 1291, 1977.

[7] G. E. PIKE., Phys. Rev., B6, 1572, 1972.

[8] D. AUSTIN and N. F. MOTT., Adv. Phys., 18, 41, 1969.

[9] N. F. MOTT, E. A. DAVIS and R. A. STREET., Phil. Mag., 32, 961, 1975.