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Electrical conductivity and dielectric relaxation in non-crystalline films of tungsten trioxide

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Abstract

Amorphous tungsten trioxide (a-WO_3) thin films were prepared by thermal evaporation technique. The electrical conductivity and dielectric properties of the prepared films have been investigated in the frequency range from 100 Hz to 100 kHz and in the temperature range 293–393 K. In spite of the absence of the dielectric loss peaks, application of the dielectric modulus formulism gives a simple method for evaluating the activation energy of the dielectric relaxation. The frequency dependence of $\sigma(\omega)$ follows the Jonscher's universal dynamic law with the relation $\sigma(\omega) = \sigma_{dc} + A\omega^s$, where s is the frequency exponent. The conductivity in the direct regime, σ_{dc}, is described by the small polaron model. The electrical conductivity and dielectric properties show that Hunt's model is well adapted to a-WO_3 films.

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Keywords: Conductivity; Dielectric properties; Relaxation; Electric modulus

1. Introduction

Thin film technology is well established and widely used in the fabrication of electronic devices. The technique has been successfully used to fabricate thin film resistors, capacitors, photoelectronic devices etc. The use of this technique in fabricating electronic devices makes it necessary to understand the electrical properties of the material in thin film form.

Tungsten trioxide (WO_3) is a wide band-gap n-type semiconductor. Films of WO_3 are considerable interest because of their potential applications in electrochromic devices [1–4] and gas sensors [5,6]. These films can be amorphous or polycrystalline depending on preparation method. These films have previously been deposited by various different techniques such as sputtering [7–10], pulsed laser [11,12], thermal evaporation [13–15], wet chemical method [16–21], sol–gel [22], and spray pyrolysis [23,24]. However, the properties of the films are significantly affected by the film crystallinity.

Development of devices based on WO_3 thin films is clearly dependent upon knowledge of general electrical behaviour of these materials. In a previous work, we observed that WO_3 films deposited by thermal evaporation technique are amorphous with optical gap of 3.28 eV [15]. According to the available literature, AC conductivity and dielectric properties of the thermally evaporated WO_3 films were not studied.

AC conductivity measurement of semiconductors, as is a powerful tool for obtaining information about the defect states in amorphous semiconductors [26,27], has been extensively used to understand the conduction process [25]. Various models have been proposed to explain the

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ac conduction mechanisms [25–30]. Dielectric relaxation studies are important to understand the nature and the origin of dielectric losses, which in turn, may be useful in the determination of structure and defects in solids. The dielectric behaviour of thin film devices depends not only on their material properties, but also on the substrate used for fabrication and the type of the metal electrodes. Fringing effects at the edges of thin film dielectrics is usually negligible because the thickness of the dielectric is usually very small compared to its lateral dimensions. The magnitude of geometric and measured capacitance may differ if the electric field at the metal insulator interface varies over this region.

In this work, amorphous tungsten oxide films have been prepared by thermal evaporation technique. The temperature and frequency dependence of the electrical conductivity, the dielectric constants for a-WO_3 films in the frequency range 0.1–10 kHz and in the temperature range 293–393 K have been investigated and analyzed to determine some related parameters and predict the electronic conduction mechanisms.

2. Experimental techniques

Tungsten trioxide (WO_3) powder used in this study was obtained from BDH chemical Ltd Company with purity of 99.986%. Thin films of different thicknesses were deposited by vacuum thermal evaporation method, using a high vacuum coating unit (Edwards, E306A). Thin films were deposited from a molybdenum evaporator charged by WO_3 in a vacuum of 10^{-5} Pa. The deposition rate was controlled at 1 nms^{-1} using a quartz crystal thickness monitor (FTM6, Edwards). The film thickness ranged between 100 and 500 nm. Accurate thickness measurements were derived after deposition using interferometric method. For AC measurements, films of thickness 500 nm were sandwiched between ITO and Au electrodes as lower and upper electrodes respectively. A programmable automatic RCL bridge (Fluke PM6306) was used to measure the impedance Z, the capacitance C, and the loss tangent ($\tan\delta$) directly. The real part of the total conductivity was calculated from the equation: $\sigma(\omega) = t\sin\phi/ZS$, where t is the thickness of the film, S is the cross-sectional area and ϕ is the phase angle between the input and out put signals. The dielectric constant was calculated from the equation: $\varepsilon_1 = tC/\varepsilon_0 S$, where C is the capacitance of the film, and ε_0 is the permittivity of free space. The dielectric loss ε_2 was calculated from the equation: $\varepsilon_2 = \varepsilon_1 \tan\delta$, where ($\delta = 90 - \phi$).

3. Results and discussion

3.1. Dielectric properties of a-WO_3

Dielectric dispersion implies the variation of real and imaginary parts at fixed temperatures. The dielectric constant is associated with the polarization of the material under the influence of sub-switching AC field [31]. The frequency dependence of the real $\varepsilon'(\omega)$ and imaginary $\varepsilon''(\omega)$ parts of the dielectric permittivity at different temperatures for a-WO_3 film are shown in Fig. 1(a,b). As the temperature increases, the dielectric constants at low frequency show a dispersive behaviour and rise rapidly and a strong dispersion is also observed at frequencies below ~400 Hz.

The study of dielectric properties is an important source for information in a thin film; since we can determine the electrical and dipolar relaxation time and its activation energy [32] but, it is very difficult to observe the dielectric relaxation peak in transition metal oxide (TMO) glasses because its dielectric loss current is masked by the dominant conduction current [33]. It has been suggested by Moynihan, Boese and Laberage [34,35] that in the absence of a well-defined $\varepsilon''(\omega)$ peak, information about the relaxation mechanism can be obtained from the dielectric modulus representation, which is defined as the reciprocal of the dielectric permittivity [36,37]:

$$M^*(\omega) = 1/\varepsilon^*(\omega) = M' + iM'', \tag{1}$$

M' and M'' are defined as:

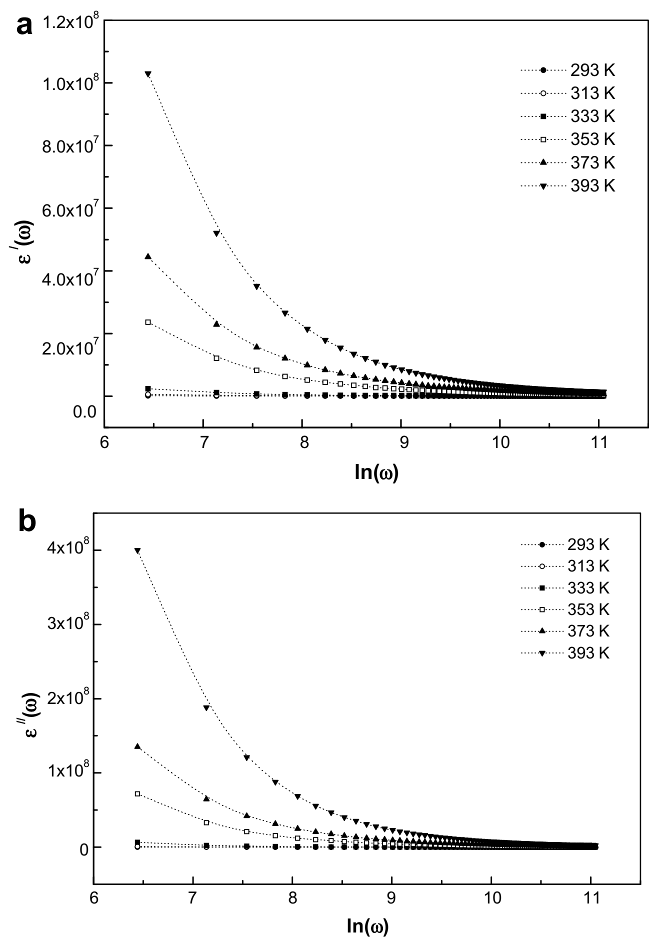

Fig. 1. Frequency dependence of the: (a) real part $\varepsilon'(\omega)$ and (b) imaginary part $\varepsilon''(\omega)$ of the dielectric constant for a-WO_3 thin film at different temperatures.