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AC conductivity, dielectric relaxation and modulus behavior of Sb₂S₂O new kermesite alloy for optoelectronic applications



M. Haj Lakhdar*, T. Larbi, B. Ouni, M. Amlouk

Unité de physique des dispositifs a semi-conducteurs, Faculté des sciences deTunis, Tunis El Manar University, 2092 Tunis, Tunisia

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ABSTRACT

In this work, we present some physical properties of Sb_2S_2O thin films obtained through heat treatment of Sb_2S_3 thin films under an atmospheric pressure at 350 °C. The electrical conductivity, dielectric properties and relaxation model of these thin films were studied using impedance spectroscopy technique in the frequency range from 5 Hz to 13 MHz at various temperatures from 350 °C to 425 °C. Besides, the frequency and temperature dependence of the complex impedance, AC conductivity and complex electric modulus has been investigated.

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1. Introduction

Metal oxysulfides compounds have been recognized for their promising applications in various fields based on the electronic, magnetic, optical, and chemical characteristics oxysulfides are attractive materials due to their chemical and thermal stability which makes them very promising in various fields such as Field emission display [1], photocatalysts [2], up conversion luminescence efficiency [3]. Commonly, the oxysulfides could be synthesized using several methods as r.f. magnetron sputtering [4], hydrothermal method [5], solid state reaction [6], and melting technique [7] or with direct sulfidation of oxides by sulfur-based gases [8]. Antimony oxysulfide (Sb₂S₂O) has a triclinic structure which is derived from the Sb₂S₃ by replacing one of the sulfur atoms with a oxygen atom. On the other hand, when sulfur atoms partially substitutes oxygen atom in oxysulfides, the band gap generally expands. Indeed, the modulation in band gap from 1.8 eV (Sb_2S_3) [9] to 3.93 eV (Sb_2O_4) [10] was reported with an intermediate band gap of 3 eV (Sb₂S₂O) [11] which makes them as a potential material for optoelectronic and solar cell applications. Ahn et al. reported that Zinc oxysulfide, which is a solid solution of ZnO and ZnS may be suitable in the area of heterojunction thin film solar cells [12]. Besides, the temperature and frequency dependence on AC conductivity, dielectric property, relaxation mode of semiconductors materials have been extensively studied to

understand transport mechanism of charge carriers, which play a crucial role in optoelectronic field. Here, $\mathrm{Sb_2S_2O}$ was investigated as a new oxysulfide in order to achieve the electrical and dielectrical properties. Since no physical properties of this material have been reported yet, we investigated the frequency and temperature dependence of the complex impedance, AC conductivity and complex electric modulus in order to identify conduction mechanism in this paper.

2. Experimental details

2.1. Sb₂S₂O thin films

 Sb_2S_2O thin films are deposited on glass substrates on three stages. Firstly, antimony thin films have been prepared by thermal evaporation under vacuum $(10^{-4}-10^{-5}\,\text{Pa})$ The antimony was deposited on glass substrates raised to the temperature $100\,^{\circ}\text{C}$. Then, Sb thin films are heated at $300\,^{\circ}\text{C}$ during 6 h under sulfur atmosphere [9]. Finally, Sb_2S_3 thin films thus obtained are oxidized in air during 6 h at $350\,^{\circ}\text{C}$ to form thin layers of Sb_2S_2O [11]. We estimate that the value of the thickness of Sb_2S_2O thin film is in the same order of magnitude as that of Sb_2S_3 ($\approx 600\,\text{nm}$) [9].

2.2. Characterization technique

The measurements were carried out in the temperature range of 623–698 K by using a tube furnace (Vecstar FURNACES) and the

^{*} Corresponding author.

E-mail address: hajlakhdar_mourad@yahoo.fr (M. Haj Lakhdar).

electrical conductivity was measured by HP4192A impedance for high frequency and Autolab PGSTAT30 for low frequency. Both devices are controlled by programs that allow received and safeguard measures.

The electrical measurement was conducted using two shaped electrodes of band painted on either end of the sample by using the silver paste. AC conductivity was obtained from experimental impedance data using the relation: $\sigma_{AC}=d/Z'S$, where Z' is the real components of complex impedance, d is the distance between electrodes and S is the cross-sectional area.

3. Results and discussion

Shown in Fig. 1 are the complex impedance spectra of $\mathrm{Sb_2S_2O}$ thin films at different temperatures. It is clearly observed from this plot that the semicircles are depressed and their centers are shifted down to the real axis indicating non-Debye type relaxation processes in the material [13,14]. The experimental impedance data can be modeled by an equivalent circuit consisting by a parallel combination of a constant phase element (CPE) and a resistance (R). Indeed, the variation of the real part Z' with the imaginary part of impedance Z'' (Nyquist plots) can be described by the Cole–Cole model [15,16], which is given by:

$$Z = \frac{R}{[1 + (j\omega\tau)^{\alpha}]}$$

Where ω is angular frequency, $\tau = RC$ is the relaxation time and α is a parameter which characterizes the distribution of relaxation times. The values of the equivalent circuit element have been listed in Table 1.

Fig. 2 shows the experimental values of Z' versus frequency at different temperatures. As seen in Fig. 2, Z' magnitude decreases with increasing frequency as well as temperature, which indicates a semiconductor behavior.

The variation of imaginary part Z'' with frequency at different temperatures is plotted in Fig. 3. It is clearly observed from this plot that the imaginary part Z'' increase with frequency reaching a maximum peak $Z_{\text{max}}^{\text{N}}$ then decrease. Furthermore, the relaxation peak position shifts to higher frequencies when the temperature increases, we also note that $Z_{\text{max}}^{\text{N}}$ values decrease with temperature. The imaginary impedance spectra can be employed to evaluate the relaxation time τ of the electrical phenomena in the material. The relaxation time has been evaluated from the following equation: $\omega_{\text{m}}\tau=1$

The temperature variation of the relaxation frequency at maximum of Z'' is shown in Fig. 4, which satisfies the Arrhenius law [17] given by:

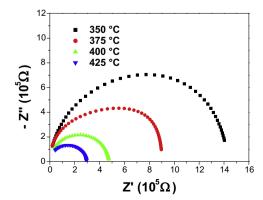


Fig. 1. Complex impedance spectra at different temperatures.

Table 1Cole-Cole values fitting parameters.

Temperature (K)	$R(\Omega)$	$C (10^{-13} \text{ F})$	α
623	1628460	7.84	0.908
648	1107325	7.96	0.844
673	505797	7.51	0.899
698	311710	7.46	0.901

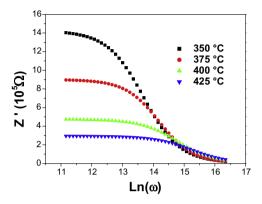


Fig. 2. Frequency dependence of Z' at different temperatures.

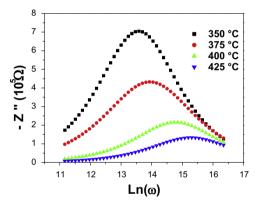


Fig. 3. Frequency dependence of Z'' at different temperatures.

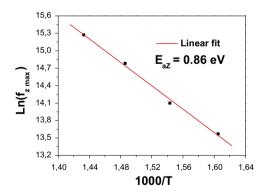


Fig. 4. Temperature dependence of relaxation frequency for Z''.

$$f_{\text{max}} = f_0 \exp\left(-\frac{E_a}{k_B T}\right)$$

Where f_0 is the pre-exponential factor, E_a is the activation energy, T is the measuring temperature and k_B is the Boltzmann constant. The activation energies are calculated from straight line fit (Fig. 4) and is found to be $E_a = 0$. 86 eV. This value supports the idea that the conduction mechanism for Sb₂S₂O is due to charge carriers hopping [18,19].

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