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Electrical Properties and Scaling Behavior of AgO Thin Films

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Abstract

The AgO sample, prepared by chemical reaction technique. The compound shows significant frequency dispersion in itsdielectric properties. The scaling behavior of dielectric loss and imaginary electric modulus suggest that the relaxationdescribe same mechanism at various temperatures. Impedance data presented in the Nyquist plot (Z_{00} versusZ_0) are used to identify an equivalent circuit and to know the bulk and interface contributions. The complex impedance analysis of AgO exhibits the appearance of both the grain and the grain-boundary contribution.The frequency dependent conductivity spectra follow the universal power law. The magnitude of theactivation energy indicates that the carrier transport is due to the hopping conduction.

Keywords: Spray pyrolysis, dielectric, imaginary electric, complex impedance, thin films, AgO

Introduction

Most of the I-III-VI2 compounds are direct gap semiconductors and they crystallize with the chalcopyrite structure [1, 2].They have attracted a lot of attention due to their potential applications in opto-electronic and photovoltaic devices. Although the information on Ag chalcopyrite compounds are scarce compared with Cu compounds, there is many studies about Ag compounds are found [3, 4].Among the various thin film deposition techniques, spray pyrolysis is one of the principle methods used to produce a large area and uniform coating at

simple and low cost [5].It is well known that the optical properties of thin films are highly sensitive to the preparation conditions and treatment conditions [6].
The interaction of laser beams with solid surfaces produce a variety of surface morphology changes, many of which show ripple structure with periods comparable to optical wavelength [7, 8]. Energy-beam shaping can be used to prevent heterogeneous nucleation and promote growth of long individual grains. Argon laser offers the potential for good control over the recrystallization process because of the ability to produce a narrow molten zone and to shape the beam with a variety of optical techniques. In this paper, deposition of AgO thin films by chemical spray pyrolysis and the effect of thermal annealing and laser radiation optical properties of thin films are studied [9].

Material & Method

0.5 g of silver nitrate ($AgNO_3$) solid was dissolved in 10mL of distilled water in a 100mL beaker. Then, triethanolamine TEA solution was added drop wise with constant stirring until the initially formed precipitate was dissolved (brownish solution becomes colorless). More distilled water was added to make a total volume of 80 mL. The pH of the bath was 8.0. Glass slides that have been preleased by degreasing in concentrated H_2SO_4, washed with water and detergent and rinsed with distilled water were vertically placed into the beaker and the bath was brought to and kept at 50∘C on a hot plate. After various periods of time 90 min, the coated four slides were removed from the bath, thoroughly rinsed with distilled water, and air-dried using electrical hand drier. The films were annealed at (100, 150 and 200^{O} C) for better adhesion and homogeneity on the substrates.
For the electrical measurement the sintered disc was polishedto make both their faces flat and parallel and electroded by highpurity ultrafine silver paste. To overcome the effect of moisture,on electric properties.Capacitance (C), impedance (Z), phase angle (w) and conductance(G), of the sample was measured in the frequency range100 Hz–1 MHz at various temperatures using a computercontrolled LCR-meter . The complex electric modulus
$M^* = (1/\varepsilon^*)$, the complex impedanceZ^* $(=1/\omega C_0 \varepsilon^*)$ and the ac electrical conductivity s $(= \omega\varepsilon_0\varepsilon_{00})$were obtained from the temperature dependence of the real $(\varepsilon_0)$and imaginary (ε_{00}) components of the complex dielectricconstant
$\varepsilon^* = (\varepsilon_0 - \varepsilon_{00})$, $\omega = (2\pi\nu)$ [10,11,12].

Results and Discussion

Dielectric relaxation studythe angular frequency ω= (2πν), n is the frequency of ac field) dependence of real (ε_0) and imaginary parts (ε_{00}) of the complexdielectric permittivity (ε*) of Ag O as a function of temperature isshown in Fig (1). A relaxation is observed in the entire temperaturerange as a gradual decrease in $\varepsilon_0(\omega)$ and a broad peak in ε_0 (ω). Thevariation of $\varepsilon_0$with frequency explains frequency relaxationphenomena of the material which are associated with a frequencydependent orientation polarization. If the frequency of the appliedac is much lower than the inverse of relaxation time (τ) of thedipoles (i.e. $\omega << 1/\tau$), the situation is essentially as for dc i.e.electric dipoles follow the field and we have $\varepsilon_0 \sim \varepsilon_s$ (ε_s = lowfrequency value of ε_0 or value of dielectric constant at quasi-staticfields). At very high frequency ($\omega << 1/\tau$), dipoles responsible forthe polarization can no longer able to follow the oscillations of theelectric field and $\varepsilon_0 \sim \varepsilon_1$($\varepsilon_1$= high frequency value of ε_0). The fieldreversal and the dipole reorientation becomes out-of-phase, givingrise to an energy dissipation within the sample at a particularfrequency. Such an effect is called dielectric relaxation and may beevidenced by a drop in ε_0 and a maximum in the imaginary part ofthe permittivity (ε_{00}) at the relaxation frequency Fig (2).The peak position of ε_{00} ,centered at the dispersion region of ε_0,shifts to higher frequency with increasing temperature Fig (2).The increase in angular frequency corresponding to ε_{00} maximawith temperature indicates an increase in charge carriers in thesample by thermal activation. It should be noted that the sharpincrease in ε_{00} at low frequencies is due to presence of dcconductivity. At low frequencies the ac conductivity is similar to dcconductivity (δ_{dc}). So δ_{dc} has an observable influence to the losscomponent of the dielectric response when it is relatively large.With increasing frequency, the influence of δ_{dc} would be ignorable.This kind of dependence of loss components on frequency istypically associated with losses by conduction. It seems clear thatthe width of the loss peaksFig (2) cannot be accounted for in terms of a mono dispersive relaxation process but point's towardsthe possibility of distribution of relaxation times. One of the mostconvenient ways to determine the poly dispersive nature of the dielectric relaxation is through Cole–Cole model where thecomplex dielectric constant (ε*) is described by the empirical relation [13]:

$$\varepsilon^* = \varepsilon_0 - i\varepsilon_0 = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1+(i\omega\tau)^{1-\alpha}} \qquad (1)$$

Whereas is the low frequency value and e1 is the high frequency value of e0, a is a measure of distribution of relaxation times, $\omega = (2\pi\nu)$ is the angular frequency of applied ac field and $\tau = \omega^{-1}$ is the most probable relaxation time, $= \sqrt{-1}$. We have fitted the experimental data using equation (1). The fitted data are shown in Fig (2), giving the values of (α) to be between 0.10 and 0.17 for the temperature range from 100 to 200^{O}C. A good agreement between the directly measured values of (ε_{00}) and those calculated using equation (1) suggests that the relaxation process differs from the monodispersive Debye process (for which $\alpha = 0$). As an alternate approach, the polydispersive nature of dielectric relaxation of AgO can also be confirmed by a representative complex Argand plane plots between (ε_{00}) and (ε_0) for (T = 100, 150 and 200)^{O}C as shown in Fig(3) . For a pure monodispersive Debye process, one expects semicircular plots with a centre located on the (ε_0) axis whereas, for polydispersive relaxation process ($\alpha \neq 0$), these Argand plane plots are close to circular arcs with end points on the axis of reals and a centre below this axis. The parameter α, can be determined from the angle subtended by the radius of Cole–Cole circle with the (ε_0)axis and is found to be 0.28 radian at 30 8C. The scaling behaviour of e00is shown in Fig (3) for AgO. We have scaled each (ε_{00}) by $(\varepsilon_{00})_m$ ($(\varepsilon_{00})_m$ is the peak value of loss ε_{00}) and each frequency by vm (vm is the frequency corresponding to $(\varepsilon_{00})_m$ i.e. the frequency at which (ε_{00})was found to be maximum). The overlapping of curves at various temperatures into a single master curve indicates that the relaxation describes the same mechanism at various temperatures in AgO. For the measurement of the characteristic relaxation time (τ_m), we can choose the inverse of frequency of the peak position of (ε_{00})versus log v plots in Fig (3) i.e. $\tau_m = \omega_m^{-1}$ .The temperature dependence of the characteristic relaxation time is shown in the inset of Fig (3), which satisfy the Arrhenius law: $\omega_m \tau_m \exp(E_a / k_{Bt}) = 1$. From the numerical fitting analysis, we have obtained the value of the activation energy $E\varepsilon_{00} = 0.35$ eV for the material ($E\varepsilon_{00}$ represents activation energy of ε_{00} obtained from the peak position of ε_{00}versus log Vs. plots). Such a value of activation energy indicates that the conduction mechanism in AgO may be due to the polaron hopping based on the electron carriers. In the hopping process, the electron distorts its surrounding by moving its neighboring atoms in the B-sites of the perovskite system.

The electrical properties of the material were determined byimpedance spectroscopy. Experimental complex impedancedata may well be approximated by the impedance of anequivalent circuit consists of resistors, capacitors and possiblyvarious distributed circuit elements. These equivalent circuitcan be physically interpreted and assign them to appropriateprocess. The display of impedance data in complex plane plotappears in the form of a succession of semicircles attributed torelaxation phenomena with different time constants due to

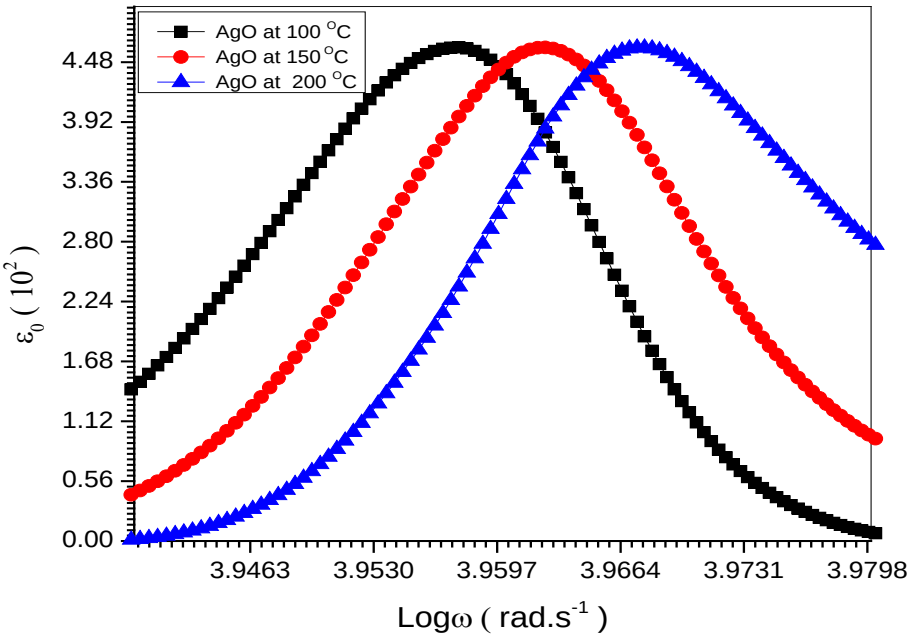
thecontribution of grain, grain-boundary and interface/electrodepolarization in the polycrystalline materials. Hence the contributionto the overall electrical properties by various componentsin the material are separated out easily. Cole–Cole representationis more convenient to distinguish between the bulk and grainboundary effects [14]. The complex impedance plane plot (Z^{**} Vs. Z^{*}) where real Z^{*} represents the resistive part and imaginary Z^{**}represents the capacitive part is generally used. Fig. (4) presents a typical complex impedance plane plot of AgO ceramic and the corresponding equivalent circuit (inset of Fig.(4) at 100, 150 And 200 ^{O}C. The two well resolved semicircular arcs on this complexplane plot are attributed to the grain and grain-boundary effect.The values of grain and grain-boundary resistances andcapacitances can be obtained by an equivalent circuit of twoparallel resistance-capacitance (RC) elements connected inseries. The equivalent electrical equations for grain and grain boundary are

$$Z^{*} = R_s + \frac{R_g}{1+(\omega R_g C_g)^2} + \frac{R_{gb}}{1+(\omega R_{gb} C_{gb})^2} \quad (2)$$

And

$$Z^{**} = R_g \left[\frac{\omega R_g C_g}{1+(\omega R_g C_g)^2}\right] + R_{gb} \left[\frac{\omega R_{gb} C_{gb}}{1+(\omega R_{gb} C_{gb})^2}\right] \quad (3)$$

Where, R_s is the series resistance, Z^{*} and Z^{**} are the real andimaginary parts of complex impedance (Z*), C_g and C_{gb} are the grainand grain-boundary capacitances and R_g and R_{gb} are the grain andgrain-boundary resistances. We have fitted our experimental data with equations (2) and (3) as shown by solid lines in Fig.(4). The fittedparameters for R_g are 6.83, 6.75 and 4.52 MV at temperatures 100,150 and 200 ^{O}C respectively and the corresponding graincapacitance (C_g) values are 7.81 X 10^{-11}, 8.50 X 10^{-11},8.62 X 10^{-11} F. Similarly, the grain-boundary resistance (R_{gb}) values at 100, 150 and 200 ^{O}C are 12.68, 12.50 and 11.64 MVrespectively. And the corresponding grain-boundary capacitance (C_{gb}) values are 9.45 X 10^{-11}, 9.58 X 10^{-11}, and 10^{-11}F.



Fig

Fig.(1):Angular frequency dependence of (ε_0)of AgO at various Temperatures at (100, 150, 200 ^{O}C).

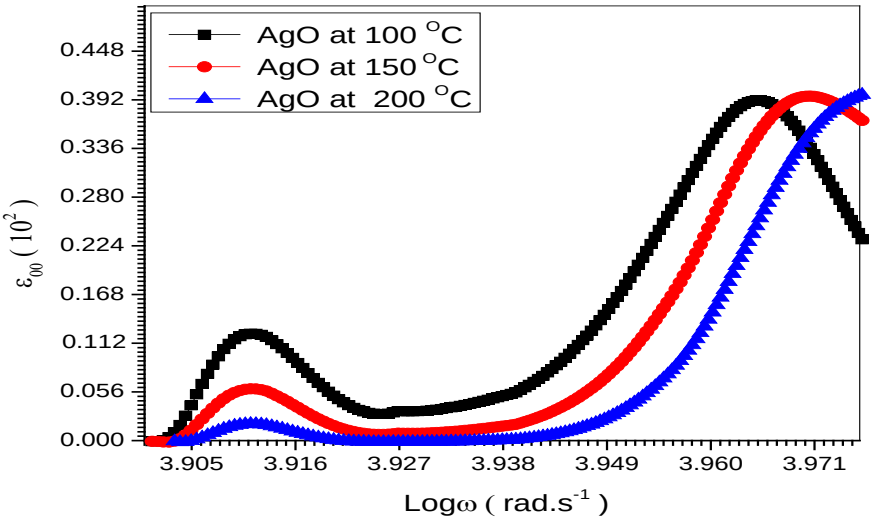


Fig. (2): Angular frequency dependence of (ε_{00}) of AgO at various Temperatures at(100, 150, 200 ^{O}C)

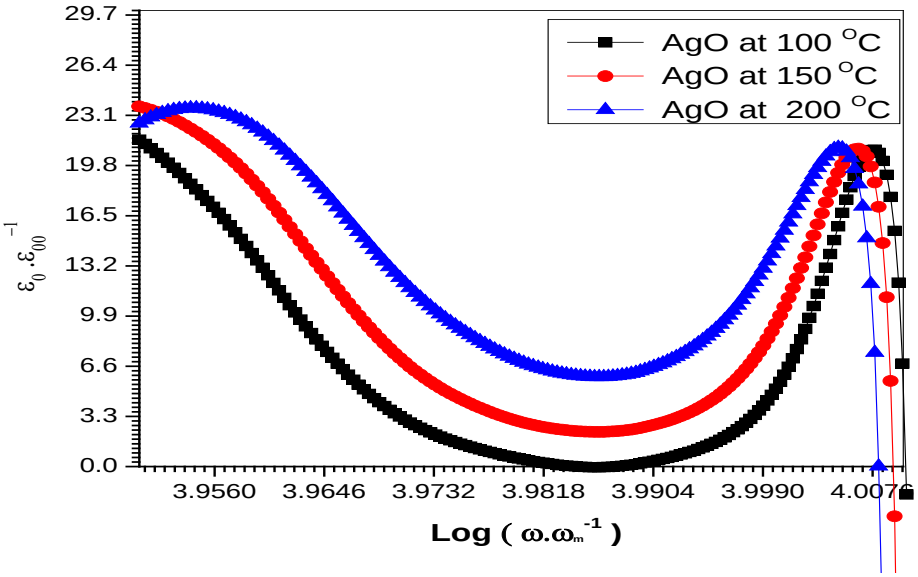


Fig.(3):Scaling behavior of (ε_{00})at various temperatures for Ag Oat 100, 150, 200 ^{O}C.

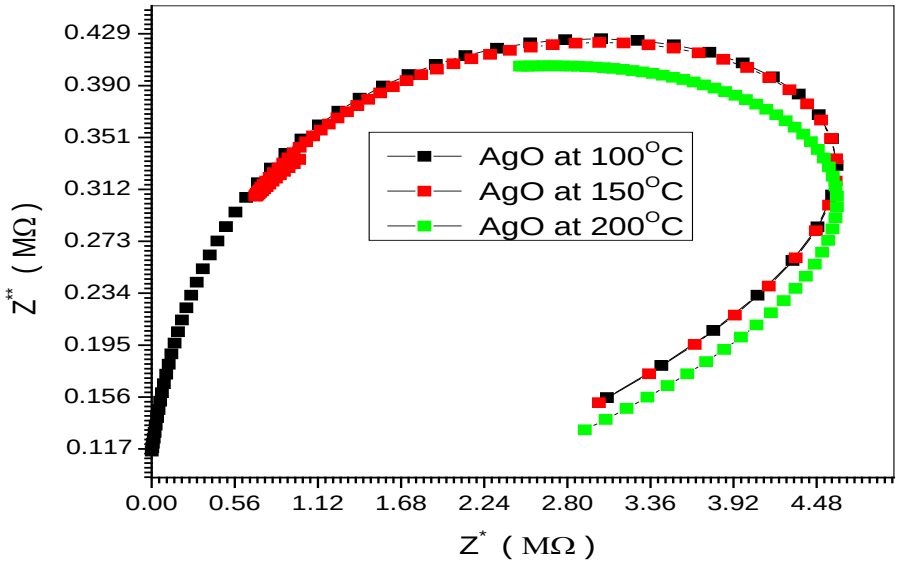


Fig.(4):Complex plane plot of impedance for Ag O at 100, 150, 200 ^{O}C

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