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DIELECTRIC DISPERSIVE BEHAVIOUR OF SILICON MONO OXIDE THIN FILM SANDWICHED STRUCTURE ANNEALED AT DIFFERENT TEMPERATURES

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Dielectric properties of evaporated silicon mono oxide film sandwiched between aluminium electrodes have been studied in the frequency range from 0.1 Hz to 1MHz. The structures were prepared on glass slide by in situ deposition of aluminium through successive evaporation in vacuum of the order of 10^{-5} torr. The structures were annealed at temperatures of 303 °K to 378 °K for five minutes and the effect of annealing is discussed. Dielectric paramters were measured by alternating current (A.C) impedence technique of Frequency Response Analyzer. The capacitance C′ (permitivity), conductance G and the loss factor g″ were found to depend on frequency and temperature, within high frequency range the capacitance varies very slowly, showing the dielectric dispersive nature and temperature independent at high frequencies. The experimental evidence on the frequency domain behaviour of this structure have been discussed. The results were best analyzed by comparing them with the power law frequency dependence curve which departs from Debye like dielecric behaviour. The a.c conductance G of such samples varies directity to (ω^n) , ω being the circular frequency, and n is a number, which is less than one and is a temperature dependent quantity. The slope of loss curve is not symme-trical with the loss peak. It is emphasised that these experimental results cannot be analysed in terms of quantum hopping conduction mechanism proposed by Johnschere. The aim of this investigation is to study the dielectric response in the specific temperature range of deposited SiO films and to prepare initially the low cost Schottky diodes and other basic components.

Key words: Capacitance, Conductance, Loss factor, Power law, Frequency dependence

Introduction

Dielectric spectroscopy is one of the most powerful and wide spread analytical technique used to study the role of charge carriers in transport and polarization mechanism. In a physical system, the materials such as, inorganic crystalline solid, amorphous semiconductor, glass and polymers, are the well known class of compounds which exhibit unusual rapid ion transport behaviour fairly at low temperature i.e well below their melting points. (Kumar and Ganapati 1991).

According to Jonschere (1977) and Hill (1978) the frequency dependence of the dielectric loss of all kinds of dielectric solids follows a universal law in which loss is proportional to ω^{n-1} with n<1 regardless of their physical and chemical nature. The common approach to the meaning of the frequency dependence of dielectric loss is based on the Debye polarization mechanism for which the complex dielectric suscesptibility$\chi^*(\omega)$ is given by,

$$\chi^*(\omega)=\chi'(\omega)-\chi''(\omega)=A(i\omega)^{n-1} \quad \text{.......}(1)$$

which is equal to, $[\varepsilon(\omega)-\varepsilon_\infty]/\varepsilon_0=1/1-\iota\omega\tau$ ....(2)

where ε. (ω) is the complex dielectric permitivity at frequency ω. and ε_∞ is the limiting high frequency value of permitivity and ε_∞... is the permitivity of free space and 'τ' is the dielectric relaxation time. 'A' is a constant and the exponent 'n' falls in the range 0<n<1. This produces the familiar Debye loss peak at ωτ=1 (Jonschere 1975). The loss in most of the dielectric materials shows a component that is flat in frequency over several decades and depends on temperature. In most of the materials the frequency dependence of dielectric loss can be expressed by the empirical relationl (Jonschere 1974).

$$\chi''(\omega)=AW^{n-1} \quad \text{......}(3)$$

extending over the several decade of frequency from the lowest audio and subaudio range to $\simeq 10^9$ Hz. This can be interpreted as the power low frequency dependance. We note here the absence of the pure Debye response. as equation (4)

$$1/\chi''(\omega)=(\omega/\omega_2)^{-m}+(\omega/\omega_1)^{1-n} \quad \text{......}(4)$$

where ω_1 and ω_2 are generally thermally activated parameters and the exponent m and 1-n are both smaller than unity and decreases with decreasing temperature. This law of reciprocal addition gives a loss peak at a frequency ω_p, which increases with temperature. In many dielectric materials the polarization is caused by hopping charges rather than by dipoles, the peak is not readily observed because of the onset of DC conduction. This is called 'Universal Law Dielectric Relaxation.

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The most widely used theories of a.c conductivity of hopping system has been based on a pair of approximations, in which the individual hop of a carrier is assumed to be independent of each other (Elvin and Moore 1974). The stochastic nature of the conduction is then taken into account by averaging the overall possible pairs of sites between which hops can occur. Although these theories have yielded considerable information about the various processes which can produce a single hop. Theories in which multiple hopping process are involved have been developed by Scher and Lax (1973).

The experimental results are usually interpreted in terms of a model initially proposed by Pollak and Geballe (1961), this model involves hopping conduction mechanism, thermally assisted, between localised states. However, the interpretation is not unique and it has been pointed out (Nadkarni and Simmon 1972) that the classical hopping over the barrier seperating the localised states can occasionally exist.

Jourdain (1979), have evaluated the a.c conductivity $\sigma_{a.c}$ of evaporated silicon monoxide film sandwiched between gold electrodes within high frequency limit and a wide range of temperatures from 173° K to 415°K. They established their results that for most of the annealed SiO sandwiched structures, measured at low frequency, the a.c conductivity $\sigma_{a.c}$ is proportional to ω^n, ω being the circular frequency and n is a temperature dependent quantity and the value of n decreases from 0.82 to 0.48. When the temperature is raised to 435°K, this quantity 'n' is found to depend on some power of frquency (Argall and Jonscher 1968; Frost and Johnscher 1975). The weak localization and electron-electron interaction (Giordano and Dilley 1993) in some samples which consists of two thin metal films seperated by insulator film of SiO. When the thickness of such insulating film is less than or equal to 200A, the electron-electron scattering rate was enhanced with respect to that found in isolated thin films.

Here, the silicon monoxide is being used as an evaporated dielectric to evaluate its capacitance and a.c conductance G against their frequency response, which is normally measured by alternating current impedence technique, The data has been compiled and are usually plotted in the form of complex impedence diagrams from which it is possible to extrapolate the true a.c. and d.c. conductivity. To proceed further the parameters were evaluated on basis of its dielectric properties and loss factor measured in thin film form, annealed under certain conditions (Thickness and vacuum) or in the form of sandwich strucure.

Experimental

Preparation of the sample. By selecting corning glass slide of size 25x 25x 0.8 mm^3 as a substrate and washed it with detergent (Mixture of $K_2Cr_2O_7 + HNO_3$) and were dried and cleaned through attached cathodic glow discharge process in vacuum. A vacuum of order 10^{-5} mm of Hg were established and maintained through ion pump bell jar, and without breaking the vacuum so that, it is not exposed to ambient gases between the deposition of each layer. The vacuum was measured through Pirani and Penning Ionization gauge, during deposition of pure Aluminium and SiO. The purity of the material chosen was of spectroscopic grade 99.99% (supplied by Koch Light Laboratories, France) and kept it in a molybedenium boat as an evaporant in the chamber. After passing a current of 45-50A through the source of evaporation for 5 min, we can deposit the aluminium film of thickness 2000°A on glass substrate within the vacuum chamber of Edwards high vacuum coating unit. The rate of deposition of aluminium was kept at 20°A sec^{-1}. The thickness of aluminium films were 0.2 micron and that of SiO film were measured to be 0.1 micron recorded by Film Thickness Monitor-3, The structures were annealed at temperatures of 303, 328, 353, and 378° K for 5 min. The rate of deposition and the thickness of the film were monitored by quartz crystal oscillator. Then silicon monoxide film was deposited again on the same substrate (Each substrate) which is already coated with aluminium. The thickness of the insulating film was 1000A. For annealing the sandwiched structure Edwards radiant heater with temperature recorder Model RPH-86 was used to anneal at different temperatures and the annealing time was kept constant (Siddall 1959). The thermocouple probes connected with this system were used for measuring the temperature (Inside) 328°K, or 353°K and 378°K and the time of annealing was recorded by a stop watch. The temperature were recorded by radiant heater. The contacts on these capacitors were made by a very thin layer of silver paint. The thickness of aluminium film were 0.2 micron and that of SiO films were 0.1 micron recorded by Film Thickness Monitor-3.

Measurement of thickness of film. The ability to measure the continuously depositing vapours in high vacuum is well suited to the measurement of thickness of mass deposited on a particular glass substrate. Quartz Crystal Oscillator were used as a monitor for measuring the thickness of thin film during vacuum deposition. The crystal oscillator utilizes the thickness shear mode of the piezoelectric quartz. The frequency shift due to film deposition is given by Equation (5).

$$\Delta f = C_f \, p \Delta t \qquad (5)$$

where Δt is the thickness of the film. C_f is the constant of material which is being deposited, p being the density of the film material. (Juh-Tzeng 1982).

Measurements by frequency response analyzer (chelsea). The block diagram of dielectric spectrometer was given earlier (Yaqub and Shakeel. 1998) with temperature and frequency as parameter and by using a fully automatic system based on Solartron Frequency Response Analyzer 1255A;a Chelsea Dielectric Interface CDI and a PC along with XY plotter. The FRA is used to measure the voltage across the sample and the current flowing through the sample. A complete computer control is available within line calculation of results enabling the frequency sweeps to be performed in the minimum time. This instrument can measure capacitance from 0.1 pF to 0.1 μ F and G values from O.1mhos to 10^{-12}mhos. This instrument is capable of providing print out of the measured data. This data can further be used for additional plots for detailed study of the parameters. FRA can produce sine wave which are applied to the sample. The correlator of this analyzer rejects noise harmonics of the fundamental signal and only the fundamental response is single XY recorder. The analyzer can measure both the in phase and quadrature component (imaginary part) to an accuracy of ± 1% and ± 5% and on full scale deflection. The instrument described above has been designed to measure the a.c properties. The system is capable of generating a.c signals of frequency ranging from 10^{-2} to 10^{6} Hz. A. D.C bias upto 41 volt can be applied to an a.c signal up to 3.0 volts. The measurements were made 1 volt rms a.c signal and 0 volt d.c bias. The readings were taken three points per on decade a log-log scale.

Results and Discussions

Figure 1 shows the frequency dependence curves capacitance C of complex capacitance is log-log representation, showing the dispersive behaviour at high frequencies. The four curves are drawn (Interpolated from the print out) at four different annealing temperatures 303, 328, 353, 378°K for frequencies 0.1Hz to 1 MHz.

The data taken within the temperature range down to 303°K. The response becomes virtually temperature independent at low temperature.

Figure 2 shows the measured values of a.c conductance G of sandwiched structure at four annealing temperatures as shown in log-log representation in the frequency range from 0.1Hz to 1 MHz.

Figure 3 represents the measured values of imaginary part (loss) of complex dielectric constant ε* of SiO sandwiched structure at four annealing temperatures for frequencies 0.1Hz to 1MHz. The behaviour emerged from this curve shows a departure from the Debye like dielectric dispersion properties and the slopes above and below the loss peaks are not symmetrical. According to Jonschere the power law frequency dependence is observed.

High frequency dispersive nature of the material in capacitance or permitivity is observed as a function of frequency at low temperature:

(1) The high frequency dispersion is due to the small values of exponent 'n' in equation (1) and it is a feature of the response of many material in which high densities hopping charge carrier dominate the dielectric behaviour (Deori and Jonschere 1979).

(2) The need for recourse to a.c measurement because of electrode polarization phenomena in such structure which is due to the incomplete replenishment of ions at the electrodes. This makes the measurement difficult. The complex impedence approach has the advantage of simplicity and conceptual neatness, true and a more detailed interpretation of results often reveals these features which complicate this picture while incidently throwing interesting fresh light on the dielectric response of compounds.

(3) A decrease in capacitance is observed with increase in frequency, in decades but the dispersion at low temperature never goes down to zero as well as other temperatures. The sample capacitance is a temperature dependent quantity. At higher frequencies, the material of the film is showing a more typical universal response with value of 'n' closer to unity. (Jonschere 1978).

Effect of temperature. 1. There are three basic trends at these temperatures. At lower temperature there is a well developed universal law, loss is proportional to frequency to the power n~1. At intermediate temperature a decrease in capacitance and increase in G appears, while at the highest temperatures and over a wide range of frequencies onset of a strong low frequency dispersion is found.

Capacitance is comparably lower at high frequencies and higher temperature.

The dispersion in capacitance is observed at low temperature. The higher capacitance at low frequency and higher annealed temperature may be attributed to the siginificant polarization of the charge carriers giving rise to ionic component of conduction. Here the dipoles cannot orient themselves at these frequencies and hence capacitance decreases. The variation of capacitance with frequency follows to (Juh-Tzeng 1982). The following relation holds,

$$C = C_g + [G\tau/\omega^2\tau^2+1] \text{ ....} (6)$$

Where C_g is the geometrical capacitance, G is the conductance corresponding to the absorption current, τ is the dipole relaxation time and ω is the angular frequency. The capacitance is maximum when frequency ω is zero and C is minimum when ω =α,

$$Xc = 1/2\pi f C \text{ ....} (7)$$

Thus the decrease in capacitance with increase in frequency (Fig 1) is in accordance of equation (4). The experimental data can be used to determine the activation energy ΔE for the relaxation process. For any constant capacitance, the relation between the frequency f and the temperature can be expressed as,

$$f = AR^{-1} \exp(-\Delta E/KT) \ldots\ldots\ldots\ldots (8)$$

where the value of the constant 'A' depends on the value of the capacitance under consideration, 'R' is a resistive parameter of the film which depends on the deposition condition like vacuum, substrate temperature, deposition rate etc. The experimental data covering given range of temperatures were fitted by choosing the best values of 'C'to substract from $C_\infty$of the capacitance.

Effect of frequency on capacitance measurement. The frequency range over which the experiments are accomplished is restricted between 0.1Hz to 1MHz. The lower limit is imposed by the impedence of the matching circuit i.e detector sensitivity. The increase in capacitance shows a maximum for the low frequency in all the sample. The capacitance exhibits increasing tendency with further increase in temperature. With n=0.37 at high temperature to n=0.88 at low temperature. For higher frequencies, the decrease of sample capacitance can be explained as;

a. The decrease in capacitance at higher frequencies and high temperatures may be attributed to the decrease in the orderliness of the orientations of polarization charges.

b. At low frequencies the a.c conductivity is viewed highly as an extension of traditional d.c electrical measurements which adds frequency to the more familiar parameter of such measurements. As the excitation frequency increases, contact of the film effect and losses are expected to directly reflect the bulk dissipative mechanism.

c. It is well known that the time required for electronic or ionic polarization to set in is very small as compared to time of voltage sign change i.e half period of applied a.c voltage. At higher frequency the half period of a.c voltage becomes shorter. When the space charge polarization fails to settle itself, the capacitance begins to drop.

d. At a higher frequency the system requires higher thermal energy to attain a maximum capacitance resulting in the shift of height of the curve towards the higher temperatures.

e. For each of the dispersion curve the value of n is different, particularly at lowest temperature the power law relation is clearly discernible at high frequencies, n=0. 88.

Variation of conductance G as a function of frequency at different temperatures. The total conductance G as obtained in the normalized plots of the conductance vs frequency in (Fig 3), it is almost frequency dependent. At low frequencies below 1Hz, G is almost constant, as the experimental range of frequencies increases, G increases in 10 decades. This vary slowly at low frequencies. This G, consists of a.c as well as d.c conductance. At higher annealing temperature, G shifts towards higher values. Since more electrically excited thermal current is generated by thermally excited electrons. At low annealing temperature, there is a characteristic depolarization current of frozen particles in polarization, it is called DRC-Dielectric Relaxation Current. (Nadkarni and Simmon, 1972).

Conclusion

In order to interpret our experimental results, it is proved that the dielectric response of such amorphous film in the frequency response departs from the Debye response and it falls into a universal pattern in which the frequency dependence of the dielectric loss follows the empirical relation in which loss is proportional to ω^{n-1} which is extending over several decades of frequency.

Under the above discussions, it is concluded that the a.c conductance G of such film is proportional to ω^n over many decades of frequency, where $n \leq 1$. In such amorphous compound it increases with frequency, which is in accordance with the work of Jonschere (1978).

The results show that the value of 'n' is a temperature dependent quantity, which is entirely controlled by mutiple hopping process.

It is concluded that both the real and the imaginary part of a.c (Conductance G) of the present model have the same frequency dependence specially at high frequencies

If the electrode contacts are poor in thin film structure, no d.c polarization will be observed and the effect of series resistance will be prominent.

The total conductance G of the samples which we have examined consists of a.c as well as d.c conductance over a given range of temperatures from 300 K to 378°K, frequencies up to 1MHz, hence the a.c conductivity is proportional to some power of circulr frquency ω^n (Jonschere 1977). At normal temperature electrodes effects produce an apparent dispersion in a.c conductance G.

As we increase the annealing temperature of the sandwiched structure the value of 'n' will decrease and the conductance will shift towards higher values.

The capacitance C decreases with incraease in frequency though very slowly i.e the permitivity decreases but lower

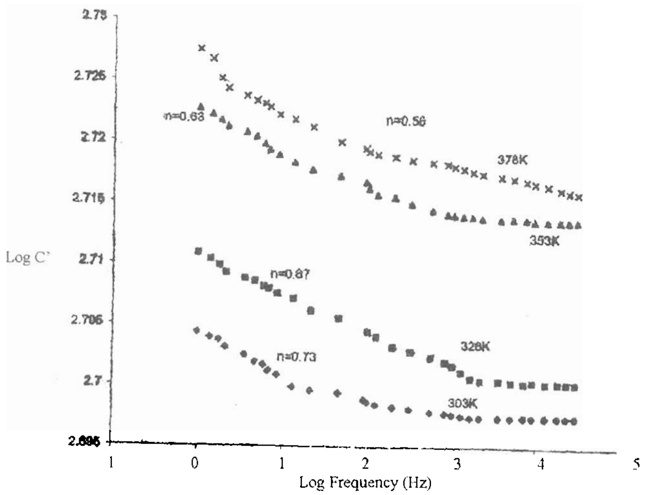

Fig 1 The frequency dependence curves showing C' of complex capacitance in log-log representation, showing the dispersive nature at high frequecies.

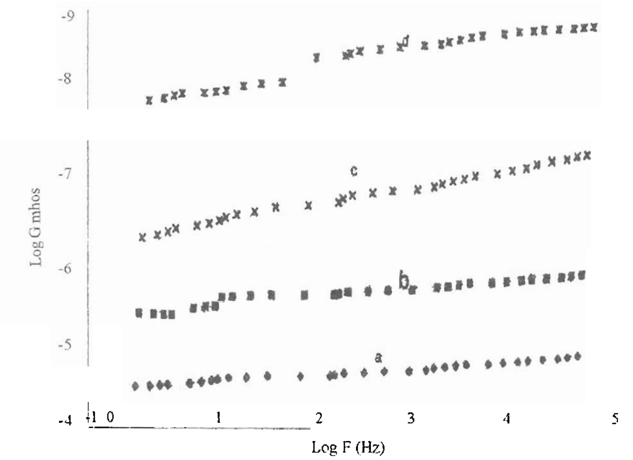

Fig 2 The Compilation of a.c Conductance data for a range of temperature arranged on a common log -f basis (Hz) but displaced vertically a=303K, b=328K,c=353K and d=378K for higher temperatures.

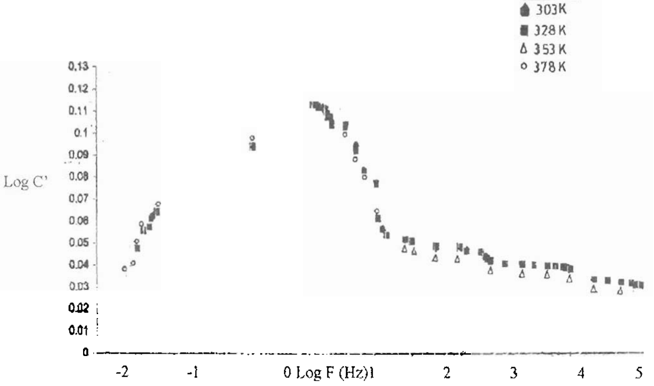

Fig 3 The frequency dependence of dielectric loss of Al-SiO-Al sandwiched structure annealed at three different termperatures. 303,328,353 and 378 K.

temperature, it does not reaches to zero and dispersion is observed at high frequency.

To achieved the best results of thin film, a vacuum requirement is of the order of 10^{-5} torr, otherwise the measured properties will not show a desired effect.

The dispersion in permitivity is observed on the low temperature annealed sample observed at frquency range 10Hz-1MHz.

This method can yield valid informations on the precise nature of interaction of electric field on amorphous materials as well as on polymeric materials. (Shickiwski *et al* 1998).

This experimental study of low frequency polarization effect in evaporated silicon monoxide films has shown that the frequency at which the loss is maxium coincide with the relaxation frequency (Fig 3) which is determined by the capacitance measurements. This is the distinct feature of Debye like dispersion properties.

Effect of frequency on the variation of imaginary part ε *of complex dielectric constant.* As shown in the plots of Fig 3, the imaginary part ε'' (ω) (dielectric loss) at various temperatures, (with different symbol corresponds to a different temperatures) shows a dependence on temperature. The dispersion is also observed in imaginary part at low frerquency range 0-100 Hz.

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