Chapter 4

DIELECTRIC PROPERTIES OF a-Si AND a-Si:H

Abstract

Studies are reported on the dependence of the dielectric constant, loss factor, impedance and equivalent series resistance of a-Si and a-Si:H capacitors on the film thickness, frequency of measurement (from 10 kHz to 1 MHz) and ambient temperature. It is found that a-Si:H film has a higher dielectric constant than a-Si film of the same thickness, which is interpreted as due to the decrease in the density of states in the energy gap of the material as a result of hydrogenation. It is also found that the interfacial polarization is negligible in thinner films while in thicker films it is evident to some extent. The influence of the electrodes and the lead resistance on the dielectric properties is clear from the increase in the loss factor with frequency.
4.00 Introduction

In recent years a great deal of interest has developed in the use of high quality dielectric films. The preparation of thin dielectric films has been motivated by their use in small volume capacitors. This improved the flexibility of thin film circuits and substantially increased their ability to meet the needs of circuit designers. The fabrication of capacitors using techniques compatible with those employed to form resistors and conductors has been one factor responsible for the rapid development of all thin film circuits [1]. It has also been stimulated by the increasing quality and complexity of electronic circuits and paved the way for the birth of thin film microelectronics. From an analysis of the properties of several thin film insulators, it becomes evident that a large number of dielectric materials are available for the fabrication of thin film capacitors. But only those films which are thermally and chemically stable are actually used in the fabrication of capacitors. The materials which are usually employed in the fabrication of thin film capacitors are oxides of metals and semiconductors such as $\text{Ta}_2\text{O}_5$, $\text{Al}_2\text{O}_3$, $\text{SiO}$ and $\text{SiO}_2$, of which the last two are the most common and thoroughly investigated [2].

But there is another important aspect for the study of the dielectric properties of materials. These studies help
to understand certain fundamental physical properties of the system, like the presence of structural defects, microvoids and impurities present in the film. The various polarization and relaxation mechanisms and the behaviour of electrode insulator interfaces are also studied in detail by many schools of research.

A thin insulator film departs considerably from the idealized theoretical model. Kohn in 1958 has shown theoretically, that the dielectric constant of the insulator should be independent of film thickness to a few atomic layers [3]. But it is a commonly observed fact that in films obtained by vapour deposition, the dielectric constant falls rapidly with decreasing thickness. This is because the films obtained by this method are porous and as the film thickness decreases, porosity increases and in turn causes a decrease in the dielectric constant. The theoretical predictions of Kohn is true only in the case of structurally perfect organic films [4]. This convincingly proves the need for extensive experimental studies on the dielectric properties of a material prepared by different methods.

The bulk of the dielectric studies carried out are on insulating materials like the ones sighted earlier. Rare earth oxides are also subjected to thorough dielectric investigations. But a close survey of the literature reveals that
eventhough the electrical properties of a Si:H are investigated thoroughly their dielectric properties are seldom subjected to a systematic study. Even the few investigations carried out are on the optical frequency dielectric constant and that too with a view to explain certain optical properties of the material [5-7]. More exactly these studies are centred around the fact that the imaginary part of the dielectric function and density of states of a-Si obtained from experiments exhibit properties which could not be obtained from a simple averaging of the crystalline spectra.

It becomes obvious that the few experimental and theoretical studies carried out on the optical frequency dielectric properties of amorphous silicon are in fact to supplement and explain the optical data of these materials. Again the fact that even the intrinsic a-Si:H films are well inferior to the conventional insulators as a dielectric material further discouraged any attempt at a systematic study of their dielectric properties. Hence we felt it necessary to carry out an investigation on the dielectric properties of the a-Si and a-Si:H thin films prepared by the vacuum evaporation method.

The investigation is in no way an exhaustive one but only intended to understand the basic dielectric behaviour of the material. A brief but relevant theory is given in the
first part followed by experimental details and concludes with a discussion on the results.

4.10 Theory

A theory for the dielectric behaviour of a metal-insulator-metal system, in which the insulator is highly doped and in which Schottky barriers exist at the metal-insulator-metal interfaces, was formulated by Simmons et al. in 1970 [8]. This theory could explain the observed minimum in the quality factor vs temperature curve and dependence of capacitance on frequency and temperature of evaporated molybdenum oxide films [9]. Later, in 1973, Goswami et al. [10] developed another theory for the dielectric properties of insulators. It was found that the second theory was more suitable for the present case and hence it is given below.

In Goswami's theory each capacitor system was assumed to comprise of the following:

(i) An inherent capacity element (C) unaffected by frequency (f) and temperature,

(ii) a discrete resistance element (R) due to the dielectric film in parallel with C, and

(iii) a series resistance (r) due to the leads.
Fig. 4.10
(a) Capacitor elements (C, R, r)
(b) Equivalent circuit
It is assumed that $R$ is temperature dependent since $R = R_0 \exp(\Delta E/kT)$ where $R_0$ is the pre-exponential factor, $\Delta E$ activation energy and $T$ the absolute temperature. But the lead resistance $r \ll R$ is temperature independent. Figures 4.10(a) and (b) show the different elements and the equivalent series circuit.

Impedance ($z$) of the circuit in Figure 4.10(a) in terms of $C$, $R$, $r$ and $\omega$ ($=2\pi f$) is given by the equation

$$z = \frac{R}{1+j\omega CR} + r$$

$$= \frac{R+r(1+\omega^2 R^2 C^2)}{1+\omega^2 R^2 C^2} - \frac{j\omega C R^2}{1+\omega^2 R^2 C^2}$$

(4.10)

In the equivalent series circuit this can be represented by

$$R_s + \frac{1}{j\omega C_s} \quad \text{or} \quad R_s - j\frac{1}{\omega C_s}$$

(4.11)

where $R_s$ and $C_s$ are equivalent series resistance and equivalent series capacitance. From the real and imaginary parts of the above equations, we have,

$$C_s = \frac{1+\omega^2 R^2 C^2}{\omega^2 R^2 C^2} = (1+D^2)C$$

(4.12)
and \[ \frac{R_s}{R} = \frac{R+r(1+\omega^2 R^2 C^2)}{(1+\omega^2 R^2 C^2)} = r + \frac{D^2}{E^2} R \] (4.13)

where \( D = \frac{1}{\omega RC} \). Since \( \tan \delta = \frac{\omega C_s R_s}{R} \) we have

\[ \tan \delta = \frac{\omega (1+\omega^2 R^2 C^2) [R+r(1+\omega^2 R^2 C^2)]}{(\omega^2 R^2 C)(1+\omega^2 R^2 C^2)} \]

\[ = \frac{1}{\omega RC} + \frac{r}{\omega^2 R^2 C} + \omega RC \]

\[ = D(1 + \frac{r}{R}) + \omega RC \] (4.14)

When \( \omega^2 R^2 C \gg r \) or \( \frac{r}{R} \ll 1 \) which is true for all cases, equation (4.14) reduces to

\[ \tan \delta = \frac{1}{\omega RC} + \omega RC \] (4.15)

When \( \omega \) is small, \( \frac{1}{\omega RC} \gg \omega RC \). Then

\[ \tan \delta = \frac{1}{\omega RC} \] (4.16)

When \( \omega \) is large \( \frac{1}{\omega RC} \ll \omega RC \) and then

\[ \tan \delta = \omega RC \] (4.17)
Equations (4.16) and (4.17) predict conditions when the loss factor will be inversely or directly proportional to $\omega$.

The effect of temperature on capacitance is obtained from equation (4.12) which can be written as

$$C_s = \frac{1}{\omega^2 R^2 C} + C \quad (4.18)$$

Since $R$ depends on temperature $C_s$ will also vary with temperature.

4.20 Experimental

Amorphous silicon films of thicknesses ranging from 460 Å to 1300 Å were prepared on glass substrates pre-coated with aluminium electrodes of breadth 1 cm. The area of the a-Si films was 2 cm x 2 cm. Over this, aluminium counter electrodes were deposited so that the two metal electrodes had an overlapping area of 0.8 cm$^2$, with the a-Si film sandwiched between them. In the fabrication of a-Si:H capacitors the Si films were subjected to hydrogenation as described in Chapter 2,
before depositing the counter electrode. The hydrogenation temperature, partial pressure and time of annealing were 573K, 0.05 torr and 60 minutes respectively. In the case of unhydrogenated samples the films were subjected to annealing in a vacuum (~ 10\(^{-5}\) torr) at 573K for 60 minutes prior to the deposition of the counter electrode. The completed devices were subjected to dielectric measurements using the HP 4277 A LCZ bridge, in the frequency range 10 kHz to 1 MHz and temperature range 300K to 373K.

The parameters measured were capacitance (C) loss factor (\(\tan \delta\)) and equivalent series resistance (ESR).

4.30 Results and discussion

4.31 Unhydrogenated amorphous silicon (a-Si)

In this case the thickness of the a-Si film was 603 Å. In Figure 4.30 the frequency dependence of the dielectric constant (\(\varepsilon\)) and loss factor (\(\tan \delta\)) are given. The value of \(\varepsilon\) at 10 kHz is 4.4 and it increased to 16.4 as the frequency was increased to 275 kHz. Above 275 kHz it was not possible to find the value of \(\varepsilon\) as the capacitance value was above the range of the instrument. In the same figure the loss factor (\(\tan \delta\)) is plotted against frequency.
Fig. 4.30

Dielectric constant & tan δ vs Frequency

![Graph showing Dielectric constant (\(\varepsilon\)) and tan δ vs Frequency (kHz).]
Loss factor also increased with increase of frequency. The temperature dependence of $\varepsilon$ is presented in Figure 4.31. It is clear from this figure that $\varepsilon$ is independent of temperature in the range 300 to 373K. In Figure 4.32 the logarithm of the impedance ($Z$) of the capacitor is plotted against the logarithm of frequency. $Z$ decreased up to 300 kHz above which it increased. In the same figure the equivalent series resistance (ESR) of the capacitor is plotted against frequency. A sharp decrease in ESR is observed between 10 kHz and 60 kHz. From 100 kHz to 1 MHz the curve exhibits a negligible frequency dependence. It is interesting to note that at 274 kHz the capacitance increased sharply and went out of the range of the LCZ bridge. Again the capacitance appeared at 500 kHz with a negative sign (not plotted in Figure 4.30) and this negative capacitance began to decrease and approach zero as the frequency was increased. It was around this frequency (at 300 kHz) the impedance showed the minimum value and began to increase with the further increase of frequency. But the ESR did not exhibit any change at 300 kHz.

4.32 Hydrogenated amorphous silicon (a-Si:H)

Four films of a-Si:H with thicknesses 461 Å, 500 Å, 837 Å and 1312 Å were subjected to measurements. The frequency dependence of their dielectric constants ($\varepsilon'$) is given in
Fig. 4.31
Temperature dependence of the Dielectric constant ($\varepsilon$) of a-Si

Fig. 4.32
Frequency dependence of the Impedance (Z) of a-Si
Figure 4.33. It can be seen from this figure that for films 1 and 2 (i.e. 461 Å and 500 Å), there is negligible increase of $\varepsilon$ as the frequency is varied from 10 kHz to 100 kHz. But $\varepsilon$ becomes 12.4 and 12.5 for films 1 and 2 at 240 kHz and 260 kHz respectively. In the case of films 3 and 4 (837 Å and 1312 Å), $\varepsilon$ decreases at first reaches a minimum value and then increases. For film 3 this minimum occurs at 50 kHz and for film 4 at 90 kHz. For all the films the loss factor ($\tan \delta$) increased as the frequency was increased and at lower frequencies $\tan \delta$ increased as the thickness of the film was increased (Figure 4.34). Temperature dependence of $\varepsilon$ and $\tan \delta$ are given in Figures 4.60 and 4.70. While for films 1 and 2 $\varepsilon$ was independent of temperature, for film 3 $\varepsilon$ slightly increased up to 350K and became steady at that value in the temperature range studied. Loss factors of these films also were weakly dependent upon the temperature. But in the case of film 4, there was a strong dependence of both $\varepsilon$ and $\tan \delta$ on temperature. $\varepsilon$ increased by 123.5% and $\tan \delta$ by 114% as the temperature was varied in the range 300K to 373K. The thickness dependence of the dielectric constant is given in Figure 4.35. Between 461 Å and 500 Å there was a sharp increase in $\varepsilon$ and as the thickness was increased further there was a more gradual increase of $\varepsilon$. In Figure 4.36 a typical
Fig. 4.33

Dielectric constant vs Frequency

Dielectric constant (ε) vs Frequency (kHz)

- 837 Å
- 500 Å
- 1312 Å
- 461 Å
Fig. 4.34

Loss factor vs Frequency

$\tan \delta$

Frequency (kHz)
Fig. 4.35

**Thickness vs Dielectric constant**
(measured at frequency 10 kHz)

![Graph showing the relationship between thickness and dielectric constant. The x-axis represents thickness in Å (angstroms), ranging from 0 to 1500, and the y-axis represents dielectric constant (E), ranging from 0 to 10. The graph shows a positive trend, indicating an increase in dielectric constant with thickness.]
Fig. 4.36

Frequency dependence of the Impedance (Z) and ESR of a-Si:H thin films

In Z vs. ln ω

In Z

ESR

Slope = 0.83

200 400 600 800
Frequency

ESR

20 40 60 80 100 120

ln Z

ln ω
Fig. 4.60
Temperature dependence of the Dielectric constant ($\varepsilon$) of a-Si:H thin films.

Fig. 4.70
Temperature dependence of the Loss factor ($\tan \delta$) of a-Si:H thin films.
plot of $\ln Z$ vs $\ln \omega$ and ESR vs frequency for a-Si:H films are given. In this particular case the film thickness is 1312 Å. The slope of the straight line part of $\ln Z$ vs $\ln \omega$ plot is 0.83 unlike in the case of a-Si, where it was 1. $Z$ shows a minimum value at 500 kHz, beyond which it increases. ESR exhibits a sharp decrease when the frequency changes from 10 kHz to 25 kHz. From 200 kHz to 1 MHz ESR remains almost independent of frequency.

An analysis of the above results is presented below. It is well known that all the main polarizations are possible in the case of an amorphous thin film. In an MIS structure interfacial polarization can arise due to the accumulation of space charge at the metal-dielectric interfaces. Both bulk material properties and interfacial effects can contribute to the polarization mechanisms in these films. Interfacial polarization is not a characteristic of the material while polarizations resulting from imperfections, defects and ion-vacancies in the film structure are characteristic of the material [11].

It is generally observed that $\varepsilon$ increases towards the low frequency side in the case of interfacial polarization. In this case $\varepsilon$ increases with increase in temperature.
In the present case we have observed that in unhydrogenated samples $\varepsilon$ increases with increasing frequency. In this case $\varepsilon$ is independent of temperature in the range measured (Figure 4.20). So here interfacial polarization can be ruled out. Since electronic polarization becomes dominant at very high frequencies, the most probable mechanism will be polarization resulting from imperfections, defects and microvoids in the film.

In hydrogenated films, for certain film thicknesses (837 Å and 1312 Å) $\varepsilon$ decreases with increasing frequency up to a certain value of frequency and then increases with further increase in frequency. It is also observed that for these films $\varepsilon$ increases with increase in the measurement temperature (Figure 4.60). Hence it can be concluded that interfacial polarization is present in addition to polarization due to defects and microvoids in these films and also that the contribution due to the latter is comparatively less than in the case of unhydrogenated samples. This is more due to the passivation of Si dangling bonds by hydrogen than due to the annealing out of microvoids since both hydrogenated and unhydrogenated films were subjected to the annealing step up to 573K. But thinner a-Si:H films (461 Å and 500 Å) behaved like unhydrogenated samples.
It is usually observed that as the frequency increases, \( \tan \delta \) initially decreases, passes through a minimum and then increases. Here in hydrogenated and unhydrogenated cases and for different film thicknesses we have observed a steady increase in \( \tan \delta \) with frequency, in the frequency range studied. This is due to the effect of electrodes and lead resistances [24].

The dielectric constant of a solid is a property which depends on the atoms and molecules present and their arrangements. Hence it is a bulk property. However, practically in the case of thin films it depends on the thickness of the films [12,13]. The thickness dependence of the dielectric constant is thought to arise owing to the presence of voids in the thinner films. As the films become thicker the density of voids decreases resulting in a higher value of dielectric constant and when the films become sufficiently thick for the voids to disappear, the dielectric constant becomes thickness independent.

From the value of the dielectric constant, loss factor, impedance, equivalent series resistance and parallel conductance of the a-Si:H films with different thicknesses at 10 kHz frequency the values of these parameters at 603 Å...
are deduced. The following values are obtained. Dielectric constant ($\varepsilon$) = 7, loss factor ($\tan \delta$) = 0.11, impedance ($Z$) = 170 k$\Omega$, equivalent series resistance (ESR) = 0.021 k$\Omega$ and parallel conductance ($G$) = 0.6 ms. The corresponding values of a-Si film of the same thickness are $\varepsilon = 4.4$, $\tan \delta = 0.05$, $Z = 292$ k$\Omega$, ESR = 0.02 k$\Omega$ and $G = 0.19$ ms. So it becomes clear that after hydrogenation the dielectric constant increased considerably. This was accompanied by an increase of the loss factor. The increase in $\varepsilon$ is due to the decrease of the localized states in the energy gap. The increase in the loss factor is due to the increase in the parallel conductance which indicates that the main loss mechanism is conductive.

4.40 Conclusion

From the dielectric studies of hydrogenated and unhydrogenated amorphous Si thin films it was found that the unpaired silicon bonds known as dangling bonds contribute to the polarization mechanisms. In thinner films the interfacial polarization is negligible in the frequency range studied. But in thicker hydrogenated films interfacial polarization is evident to some extent. The increase of the loss factor with frequency indicated the role of electrodes and lead resistance in the measurements. From these studies it can be concluded that eventhough the resistivity of the a-Si and
a-Si:H films is very high their dielectric parameters are very much dependent upon the frequency and temperature of measurement.

From the study of the dielectric parameters of a-Si:H thin films with different thicknesses the values of those parameters at a thickness of 603 Å were found out and were compared with those of a-Si thin film of the same thickness. The appreciable increase in $\varepsilon$ noticed in the case of a-Si:H film is interpreted as due to the decrease in the localized gap states.
REFERENCES


