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Charge trapping and ac conductivity in Amorphous Silicon Oxide

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Abstract

In this paper we have studied the effect of accelerated thermal ageing on the electrical properties of amorphous silicon oxide films a-SiO_2. In order to study the charge trapping phenomenon in this material, we have performed the mirror method using a secondary Electron Microscope (SEM). This method consists to inject a negative space charge in the specimen with a high energy electron beam. Results show that trapped charges increase with thermal ageing time. Dielectric investigations performed in the frequency range between 20 Hz and 1 MHz, showed that the relative permittivity increases with thermal ageing time. The ac conductivity has been found to follow the Jonsher law $\sigma_{ac}\ \alpha\omega^{n}$. The decrease of ac conductivity has been interpreted.

PACS: Type pacs here, separated by semicolons ;

Key words: a-SiO2; capacitance; Cole-Cole; mirror method; space charge.

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1. Introduction

Since 1970, Silicon oxides SiO_x with $0 < x < 2$ have attracted a great interest in fiber technology and opto electronic research [1, 2]. This attention is made for the low cost of silicon and for specific physical properties [3-5]. Different allotropic forms of SiO_x were reported in order to investigate their structural and optical quality [6-9]. Refractive indexes and dielectric permittivity of thin amorphous SiO_x films have been extensively investigated [10-12]. This paper focuses on the effect of accelerated thermal ageing on electrical properties of amorphous silicon oxide thin films. The electrostatic mirror method is used to analyse the trapped charges evolution with thermal ageing. This characterisation is completed by Cole-Cole analysis to determine the relative permittivity and to follow the evolution of ac conductivity with thermal ageing.

2. Experimental

The samples studied in this work are commercial silicon oxide films delivered by Glassware Marienfeld laboratory. They have dimensions of (20x20x0.16) mm^3. They were submitted to thermal constraints in a Memmert

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ULE 500 steam room at different times. Thermal ageing consists in applying several heating-cooling cycles. The cycle is composed of four stages: 1: heating from 23°C to 85°C during 2hours, 2: keeping at 85°C during 1hour, 3: cooling to 23°C during 4hours, 4: keeping at 23°C during 1hour. Thus a whole cycle lasts 8 hours.

3. Results and discussion

The mirror method elaborated by C. Le Gressus [14] and developed by B.Vallayer [15] consists in injecting a charge Q_i in an insulating material using scanning electron microscope (SEM). Samples were introduced in the scanning electron microscope room (Philips XL30 brand). Each sample was irradiated by an electron beam with energy of E_i =30 keV during 30s. Then we applied a reading tension and we observe a mirror picture. In figure 1, we have presented a mirror picture got for a reading tension of 800 V for the virgin sample as an example.

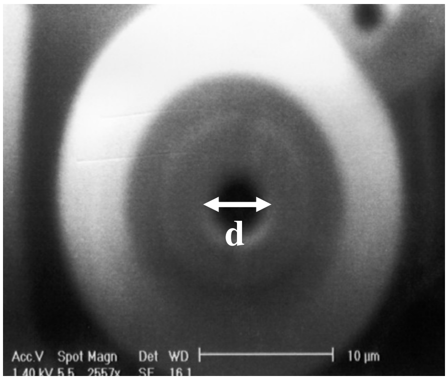

Figure 1: mirror picture got for a reading tension Vs = 800V

d is the apparent diameter of the electron canon.

The 1/d curves, as a function of the reading tension, were deduced from the mirror pictures of aged samples. Figure 2 shows the charge spreading characteristics.

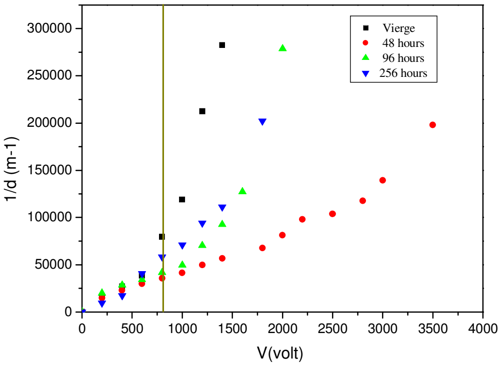

Figure 2: mirror plot of the aged Silicon oxide samples.

One can see that the 1/d curves exhibit straight line shape up to 800 V. Moreover when the charge has spread in the sample we observe a drift according to the linear zone from Vs =800V (Fig 2). The charges spread is due to a local conduction phenomenon. In deed, Sudarshan et al [16] have demonstrated that the trapped charge can cause the energy band to bend within the region where the charge is localized and therefore promotes the electron tunnelling from trap states into the conduction band. This explains the conduction within the charged area.

The linear part of the curve 1/d = f(V) is described by the punctual charge approximation satisfying the relation (1) [13-15]. Its slope is linked with the amount of trapped charges Q_t and the relative permittivity ε_r .

$$\frac{1}{d}=\frac{4L}{d'}\frac{2\pi\varepsilon_0(\varepsilon_r+1)}{Q_2K(h)}V \qquad (1)$$

where L is the work distance between the sample's surface and the last diaphragm of electrons canon, d' is the real diameter of the electrons canon and K(h) is a parameter depending on the sample's height.

In order to determine the quantity of trapped charge, we have measured the ε_r using Cole-Cole analysis. We have performed a capacitance measurement at room temperature by a HP 4284A impedance analyzer for the frequency range between 20 Hz and 1 MHz.

The experimental results (ε″ versus ε′) are represented on a Cole-Cole plot. Figure 3 shows the Cole-Cole representation for unaged sample as an example.

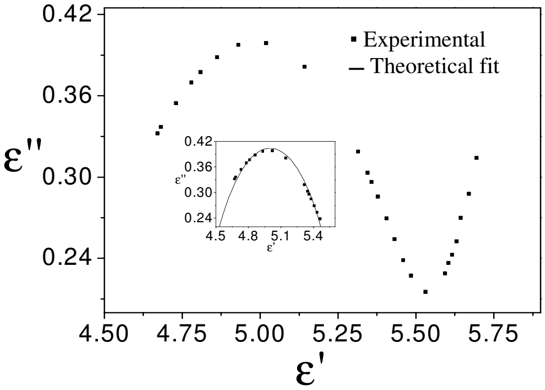

Figure 3: Arc plot for unaged sample fitted with a Cole-Cole approach

According to the Cole-Cole approach, the complex permittivity is expressed by [17]:

$$\varepsilon^*(\omega)=\varepsilon_\infty+\frac{\varepsilon_s-\varepsilon_\infty}{1+(j\omega\tau)^{1-\alpha}} \qquad (2)$$

where ω is the angular frequency, j is the imaginary unit, ε_∞ is the high frequency limit of the complex dielectric permittivity $\varepsilon^*(\omega)$, ε_s is a static dielectric permittivity, τ is a mean relaxation time and α is discussed as the parameter of the distribution of the relaxation time. ε_r values were deduced from fitting parameters for aged samples. The obtained values of ε_r as well as the deduced Q_t ones are gathered in table 1.

Table 1: ε_r values deduced from Cole–Cole plots and Qt deduced from the 1/d curves slopes.

Aging time (hours)	ε_r	Q_t
0	4.61	77.6 pC
48	5.10	142 pC
96	5.68	200.7 pC
256	6.03	256 pC

We notice that thermal aging leads to an increase of the relative permittivity and the trapped charges quantity. This demonstrates that the capacity of trapping charges increases in the thermally aged samples. One can conclude that thermal ageing creates charge traps. The excess of trapped charges is a sign of material ageing. To investigate the influence of thermal aging on the conductivity phenomenon, the ac conductivity evolution was studied using the Jonsher law [18]:

$$\sigma_{ac} \propto \omega^n \qquad (3)$$

where ω is the angular frequency and $n < 1$ is temperature-dependent parameter.
Moreover, σ_{ac} was related to ε'' by $\sigma_{ac} = \varepsilon_0 \varepsilon'' \omega$ [19].

The normalized conductivity curves for aged samples are reported in Figure 4. They show a decrease of the ac conductivity after thermal aging.

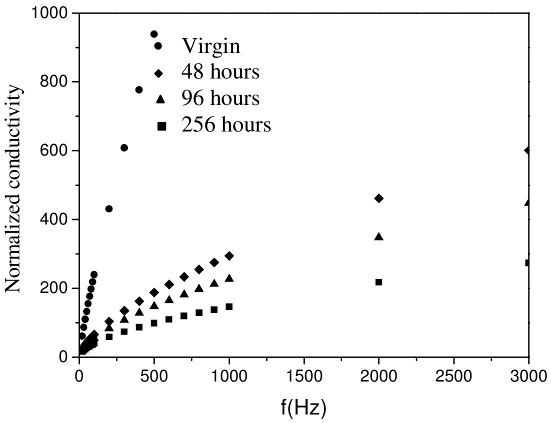

Figure 4: Conductivity normalized by $4\pi\varepsilon_0$, in aged samples.

The decrease of the conductivity is attributed to the stabilisation of the trapped charges in the deeper energetic levels, induced by thermal aging, as has been demonstrated by mirror results.

Thus, the thermal ageing clearly favours the trapping of charges. However, the presence of ac conductivity might be related to the impurities in SiO_2 such as Ti and Sn which were detected by fluorescence X [20]. In deed, we have demonstrated in previous work using the IR spectroscopy [20] that the molecular mobility is reduced after thermal aging, which leads effectively to a stabilisation of trapped charges in deeper energetic levels.

4. Conclusion:

In this paper, we have reported the results of electrical characterization of amorphous silicon oxide thin films by Mirror method and dielectric spectroscopy. The mirror effect due to a localized trapped charge near the surface of the studied samples scanned by low energy electron beam has been explained. This phenomenon has been used in order to determine the quantity Qt of trapped charges. Results show that the capacity of trapping charges increases in the thermally aged samples. This was related with the increase of ε_r . The decrease of the ac conductivity with the ageing time is attributed to the creation of deeper charge traps which leads to a stabilization of trapped charges.

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