Charge-carrier injection and extraction at metal-dielectric contact under an applied electric field

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There is charge injection/extraction at a metal-dielectric contact. In general, the problem is treated by analogy with that for metal-semiconductor contact. However, the electrical conduction through dielectrics is bulk and electrode limited. Consequently, it is difficult to discriminate between the two effects and to have specific information only about one of the mechanisms. We propose a method that allows to determine the sign and the value of the electric charge injected/extracted at the metal-dielectric contact. The method is based on the modification of the external electric field of a dielectric when electric charge is injected/extracted onto its surface. The lowest surface charge density measured in the presented experiments was around $1.8 \times 10^{-6}$ cm$^{-2}$ which corresponds to a medium distance of about 300 nm between two trapped charges. The injection/extraction process takes place no matter if there is a conduction current through the sample.

Keywords: Dielectrics, Metal-dielectric contact, Charge injection/extraction, Traps

1 Introduction

Charge-carrier injection/extraction at a metal-dielectric contact is an important factor in many electronic devices, especially in applications with insulators, where virtually all charge-carriers have to be injected from the electrodes. The electronic properties of metal-dielectric contact have received intensive attention for many years. Studies have been hampered by the inapplicability of techniques normally used to study the metal-semiconductor Schottky barriers like the junction capacitance$^{1,3}$.

According to the Schottky model$^4$ when a metal and a semiconductor or a dielectric form an interface, there is no charge transfer across the interface and the barrier height for the electrons is given by the difference between the work function of the metal in vacuum and the electron affinity of the semiconductor. However, it was observed experimentally that the Schottky model is not, generally, obeyed. Bardeen then proposed the well-known surface state model to explain this observation.$^5$ Bardeen’s model postulates a high density of surface states of the order of 1 per surface atom.

Under the applied electric field, electrons are injected at the cathode and a significant number are trapped. In the case of very low mobility materials (the mobility lower than $10^{10}$ m$^2$V$^{-1}$s$^{-1}$) the charge injected into material moves very slowly and the current density is time dependent. When a particle current is injected into an insulator, and the charge is partially trapped in the insulator, the field is often unequal to the average Laplace field $E = V/b$, where $V$ is the applied field and $b$ is the thickness of the sample. The local field is space and time dependent. The Coulombic interaction between the trapped electrons and the injected charge-carriers from metal into the dielectric material modifies the barrier height at the contact and into the bulk. Under these conditions, the current density varies with time, and this variation can be analyzed using isothermal and non-isothermal current-voltage measurements$^6-8$. The trapped electrons are highly concentrated near the injecting contact. The decay of the charging current can be explained equally well in terms of polarization effects.$^9$ It is difficult to discriminate between the two effects. The nature of the current is important to obtain the best results by measuring the isothermal charging current and the isothermal discharging current.

The aim of this paper is to demonstrate that there is always a charge-carrier injection/extraction at the
metal-dielectric contact. By the present method, the polarity and the value of the space charge injected/extracted have been determined at the metal-dielectric contact.

2 Experimental Details

The isothermal charging current, the isothermal discharging current and of the actuation voltage, at atmospheric pressure in dry N$_2$, were measured with polyethylene terephthalate (PET) films of 8 µm thickness over a range of electric fields from 0.5 MV m$^{-1}$ to 30 MV m$^{-1}$ and in the temperature range 20-90°C. PET is often used as a dielectric in high performance foil capacitors (class-E insulators). The dielectric was provided with a vacuum deposited Al electrode onto its surface and it was glued, using an electric conductive paste, on a 35 mm diameter metallic ring. So the sample behaves like an elastic membrane and the elastic constant for small displacements is equal with $k$ (the spring constant). The samples were theromally conditioned after each measurement (i.e. heated for 4 h under short-circuited condition, at a temperature of 90°C to completely remove the parasitic charge stored in deep traps). Particular care has been taken to control the experimental conditions especially, for keeping the current noise level within the limits of the electrometer noise (Keithley 616). The electrometer is connected to a data acquisition system. The sample is introduced into an oven and the temperature is controlled to better than 0.2°C. A low noise regulated power supply was used as voltage source. A 12.5 µm thickness Teflon ring was used as a spacer between the lower electrode and the non-metallized side of the PET film. The position of the higher electrode can be observed through an optical system.

3 Theory

Figure 1 shows the experimental arrangement. In order to deduce the value of the electric field $E_1$ in the air gap between the dielectric and the lower electrode, we will use Gauss’s law at the interface $x = 0$ between the dielectric and the air gap:

$$ -\varepsilon_0 E + E_1 = \sigma_2 / \varepsilon_0 $$

and Kirchhoff’s second law for the metal-dielectric-air gap-metal system:

$$ E b + E_1 a = V $$

where $\sigma_2$ is the injected/extracted charge density (cm$^{-2}$) (charge density per unit area), $\varepsilon$ the relative dielectric permittivity of the dielectric, $\varepsilon_0$ the dielectric permittivity of the vacuum, $E$ the electric field in the dielectric, $a$ the thickness of the air gap, $b$ the thickness of the dielectric and $E_1$ is the electric field in the air gap.

When a voltage $V$ is applied, the electric field $E_1$ will be:

$$ E_1 = \frac{V + \sigma_2 b}{b / \varepsilon_0 + a - x} $$

where $x$ is the displacement of the dielectric foil under the electrostatic force and $a - x$ is the thickness of air gap.

The electric force $F_\varepsilon$ acting on the dielectric will be:

$$ F_\varepsilon = \frac{E_1^2 S \varepsilon_0}{2} $$

where $S$ is the cross-sectional area of dielectric film. In first approximation, we can assume that the dielectric film is equivalent to an ideal spring. The film will move down with a distance $x$ so that:

$$ F_m = kx $$

where $k$ is the spring constant of the film.

From Eqs (3 - 5), we get:

$$ V + \frac{b \sigma_2}{\varepsilon_0} = \frac{2kx}{b / \varepsilon_0 + a - x} \sqrt{\varepsilon_0 S} $$
Eq. (6) gives the basic relationship to analyze the displacement $x$ of the dielectric as a function of the applied voltage $V$, the injected/extracted real charge $\sigma_2$, the relative dielectric permittivity of the dielectric layer, the spring constant of the system and the geometric factors of the device.

If the applied voltage increases the film will move down under the electrostatic force and for $x = a$ the dielectric will lend on the lower electrode. From Eq. (6), it results that

$$V_{pi} = -\frac{b\sigma_2}{\varepsilon\varepsilon_0} + \frac{2ka}{\varepsilon\varepsilon_0} \sqrt{\frac{d}{S\varepsilon_0 \varepsilon}}$$

... (7)

The actuation voltage $V_{pi}$ is almost equal with the so-called pull-in voltage in terms of RF micro-electromechanical systems capacitive switches. If there is no real charge in the dielectric film, from Eq. (7) we get:

$$V_{10} = \frac{2ka}{\varepsilon\varepsilon_0} \frac{d}{S\varepsilon_0 \varepsilon}$$

... (8)

Thus, we can check if there is no charge $\sigma_2$ by changing the polarity of the applied voltage $V$. If the actuation voltage $V_{pi}$ has the same value for both polarities of the applied voltage, the conclusion is that $\sigma_2 = 0$. If the two values are different, it is necessary to heat the sample in short circuit to remove the charge existing in the sample. This way, we can determine the value for $V_{10}$ and actually, it is not necessary to know the value of the spring constant $k$.

From Eqs (7) and (8), we get:

$$\sigma_2 (t) = \left( V_{10} - V_{pi} (t) \right) \frac{\varepsilon_0 \varepsilon}{b}$$

... (9)

Eq. (9) allows to determinate the injected/extracted charge by measuring the change in the actuation voltage. An increase of the actuation voltage will indicate that $\sigma_2 (t)$ is negative which means that electrons are injected from the lower electrode into the dielectric when the lower electrode is in contact with the dielectric. A decrease of the actuation voltage will indicate that $\sigma_2 (t)$ is positive which means that electrons are ejected from the dielectric into the lower electrode when the lower electrode is in contact with the dielectric and the lower electrode is biased positively in respect to the upper electrode which is at ground. Eq. (9) also allows us to study how $\sigma_2 (t)$ varies with time by monitoring the variation of $V_{pi} (t)$ with time.

4 Experimental Results

Figure 2 shows a typical isothermal charging current (ICC) for the situation when the applied voltage is lower than $V_{10}$. In this case, there is an air gap between the dielectric and the lower electrode. The upper electrode was positively biased and electrons are extracted at the contact between the dielectric and the upper electrode. The experimental data (data not presented here) shows that the injection current is identical with the extraction current for the given experimental conditions.

Figure 3 shows a typical ICC and a isothermal discharging current (IDC) for the situation when the applied voltage is higher than $V_{10}$ and the dielectric is in contact with the lower electrode. The ICC and IDC are very similar with those reported for similar conditions for a sample metallized on both sides. For
easy comparison, Fig. 3 shows the ICC and IDC for $V = 47$ V, i.e., for the situation when the dielectric is not in contact with the lower electrode. It can be observed that, for the situation when the applied voltage is lower than the actuation voltage, the isothermal charging current is almost identical with the isothermal discharging current.

To further focus on the injected or extracted charge, we conditioned the sample to remove the parasitic charge. After this, the value measured for $V_{10}$ was $V_{10} = 158$ V. During the next two weeks, we did not observe any variation of the actuation voltage in the limit of an experimental error of ± 0.4 V. After this, we kept the sample under an applied voltage of 210 V for 3 h. We measured the actuation voltage for the next two months. During the measurements, we stopped to increase the applied voltage immediately as the dielectric moves down and make contact with the lower electrode. The applied voltage was switched off immediately just to prevent further injection of electrons from lower electrode into dielectric. Using Eq. (9), the charge density per unit area $\sigma_2(t)$ was calculated. The results are shown in Fig. 4. It can be observed that $\sigma_2(t)$ decays very slowly. The full curve in Fig. 4 is the best fit line of the experimental data to an exponential decay function to determine the relaxation time of the injected charge. The value obtained was $\tau = 98.9$ days. This result is in very good agreement with data reported in literature for PET samples.\textsuperscript{11}

The actuation voltage measured with an opposite polarity was 130.5 V, indicating that when the higher electrode is negatively biased in respect with the lower electrode, the electric fields produced by the applied voltage and negative charge $-\sigma_2$ are in the same direction and consequently, the actuation voltage is lower. To check that a positive voltage applied to the lower electrode will determine electrons’ extraction from dielectric and consequently $-\sigma_2(t)$ will increase, i.e., $\sigma_2(t)$ will decrease, we applied a voltage of 180 V and the actuation voltage was measured every 10 min during the first hour and every hour during the next 10 h. The actuation voltage was increasing steadily from experiment to experiment from 130.5 V up to 171.2 V. This indicates that the initial negative charge was completely removed and more negative charge was extracted, eventually by ionizing the donor impurities, so that finally the dielectric was positively charged with a charge $\sigma_2(t)$.

A good confirmation of the positive side of the curve shown in Fig. 5 was obtained by integrating the ICC measured for a charging voltage of 180 V. The result is shown in Fig. 6. The charge density increases with time very similarly with the positive side of the real charge density as shown in Fig. 5.

5 Discussion

The long term transient charging and discharging currents in insulators can be ascribed either to dipole relaxation or to charge trapping and release of trapped charge. There is a very general result that the
measured current is the sum of particle and displacement currents. It is difficult to discriminate between the particle and displacement currents. The experimental data are analyzed in terms of polarization effects or in terms of space charge effects. The currents in Fig. 2 are measured when there is an air gap between the dielectric and the electrode, this means that we are measuring the open-circuit isothermal charging current and the open-circuit isothermal discharging current. In this case, we cannot describe about a conduction particle current. The problem is if the current in Fig. 2 represents only a polarization current.

A comparison of data in Figs 2 and 3, for the case when the dielectric is not in contact with the lower electrode and data in Fig. 3 for the case when the dielectric is in contact with the lower electrode, shows that the ICC and IDC are higher in the last case. To explain, the ICC in Fig. 3 has at least four components: (i) polarization current, (ii) injection current, (iii) extraction current and (iv) conduction current. A comparison of data in Figs 2 and 3 shows that the currents are in many respects similar. The fast decay observed during the first seconds is related to dipolar current. The next portion, for which the current follows the empirical Curie-Von Schweidler law $t^{-n}$ where $t$ is a time after the application of the field and $n$ is a constant often observed to be lower than 2, is related, to the injected/extracted current and the conduction current.

The experimental results obtained by us demonstrate that the actuation voltage is independent of the charge injected or extracted at the interface between the upper electrode and the dielectric. The actuation voltage does not change if we keep the sample under field for days, in the situation when the voltage is lower than the actuation voltage. This means that the actuation voltage is well independent of the polarization of the sample which is in good agreement with the presented model. To be more specific, this means that for the given experimental polarization conditions, there is not a remanent polarization in the sample. The actuation voltage changes only when the dielectric is in contact with the lower electrode and there is charge injection or extraction at the metal-dielectric contact, depending on the polarity of the applied voltage. A comprehensive study of the currents in Fig. 2 can provide good indications about the nature of the mechanisms of the isothermal charging current and the isothermal discharging current.

It can be observed that when the applied voltage is lower than the actuation voltage, the isothermal charging current is almost identical with the isothermal discharging current. The fact that the two currents are equal, is used as an argument to sustain that the ICC is of dipolar origin. In our opinion, this argument is valid for time shorter than about 10 s. The equality of the two currents for longer time indicates that the injected charge is trapped in the superficial traps very close to the surface. This observation is in
good agreement with our previous observations obtained using a combined protocol to study charge injection, trapping and transport in low mobility materials.\textsuperscript{13-15}

By measuring the actuation voltage with an error of 0.5 V, from Eq. (9) it results that the minimum charge density that can be measured is around $1.8 \times 10^{-6}$ cm$^{-2}$. This means that the medium distance between two trapped charges is around 300 nm. The error can be diminished by measuring the actuation voltage more precisely using a more sophisticated equipment. For example, the actuation voltage can be more precisely measured by measuring the change of the capacitance of the system\textsuperscript{10} under the applied voltage or by using a triangular voltage and measuring the current through the circuit\textsuperscript{11}.

The method can be used as well as for solid dielectrics. In this case, a thin elastic lower electrode of appropriate elastic constant $k$ has to be used.

### 6 Conclusions

The experimental results presented in this paper demonstrate that for a metal A-dielectric-air gap-metal B structure, at the contact between the metal A and the dielectric, there is electron injection if the metal A is negatively biased with respect to the metal B and electron extraction if the metal A is positively biased with respect to the metal B. From the measurements at room temperature and in a field range 0.5-30 MV m$^{-1}$, there is no indication that there is a threshold field for charge injection/extraction at the metal-dielectric contact. By measuring the change in the actuation voltage, it is possible to determine the polarity and the value of the space charge injected/extracted at the metal-dielectric contact. The results presented here are important both from theoretical point of view and for applications. A systematical study will allow us to understand, for example, the nature of the origin of the isothermal charging and discharging currents with direct implications in time domain spectroscopy and dielectric relaxation or to find solutions to decrease the injected/ejected charge and improve the reliability of RF micro electro mechanical systems capacitive switches, to give only two examples.

### References