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Short Note

A Simple Graphical Method for Evaluating Dipole Relaxation Time in Dielectric

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A simple graphical method for evaluating the dipole relaxation times (τ) in a dielectric is proposed. From isothermal measurements of the transient discharge current due to dipole relaxation, τ can be obtained graphically by means of discharge current transient spectroscopy (DCTS). In other words, the time at which the DCTS signal is a maximum corresponds to τ. Since τ and the steady-state polarization (P_s) in dielectrics at electric field (F_p) and temperature (T) can be obtained using DCTS, the dependencies of τ and P_s on F_p or T can be investigated in detail.

KEYWORDS: dielectric, ferroelectric, dipole relaxation time, polarization, depolarization, discharge current transient spectroscopy (DCTS)

Dielectric thin films with high dielectric constants have been studied intensively in order to enlarge the capacitance of the capacitors in a dynamic random-access memory (DRAM). Moreover, in order to fabricate a metal-insulator-semiconductor field-effect transistor (MISFET) which has a memory itself, ferroelectric thin films have been investigated as possible gate insulators in MISFETs. In order to realize these devices, an understanding of the mechanism of dipole relaxation in these materials is essential.

In order to evaluate the dipole relaxation time (τ), the thermally stimulated depolarization current (TSDC) method and similar methods have been proposed and tested.[1–5] However, these methods require not only a rigid constant heating rate but also an assumed temperature dependence of τ. An isothermal measurement is more suitable for evaluating τ experimentally.

Discharge current transient spectroscopy (DCTS) has recently been developed for evaluating the densities and energy levels of traps in an insulator.[6,7] Since, using DCTS, the transient discharge current can be measured isothermally in a capacitor which has a dielectric between two electrodes, this method is suitable for evaluating τ in dielectrics.

The time and temperature dependencies of dipole polarization in a dielectric are determined by competition between the orienting action of an electric field and the randomizing action of thermal motion. A capacitor, which has a dielectric between two electrodes of unit area, is considered here. In the elementary theory of dielectrics,[1,3] the buildup of polarization $P_p(W)$ in the capacitor during time W after the application of an electric field F_p at a temperature T is given by an exponential function of time

$$P_p(W) = P_s\left[1 - \exp\left(-\frac{W}{\tau}\right)\right], \tag{1}$$

where P_s is the steady-state polarization.

Provided that the relaxation times for polarization and depolarization of the dielectric can be considered identical, the decay of polarization after removal of the electric field is given by

$$P(t) = P_p(W)\exp\left(-\frac{t}{\tau}\right) \tag{2}$$

and the corresponding depolarization current density (i.e., transient discharge current density) $J_{dis}(t)$ can be written

$$J_{dis}(t) = -\frac{dP(t)}{dt} = \frac{P(t)}{\tau}. \tag{3}$$

The values of τ and P_s are determined by considering data for $J_{dis}(t)$. If one can obtain an expression in the form

$$P_p(W)x\exp(-x)$$

where $x = t/\tau$, this has a maximum value of $P_p(W)\exp(-1)$ at $x = 1$. This indicates that τ and $P_p(W)$ can be determined independently using a graphical method. The results from this method are more accurate than those results obtained from fitting a curve to $J_{dis}(t)$ data.

If one defines a function

$$S(t) = tJ_{dis}(t)\exp(1), \tag{4}$$

a function with the desired form can be obtained:

$$S(t) = [P_p(W)\exp(1)]\,\frac{t}{\tau}\exp\left(-\frac{t}{\tau}\right). \tag{5}$$

Since this function has a maximum at $t_m/\tau = 1$, the values of τ and $P_p(W)$ can be determined independently as follows:

$$\tau = t_m, \tag{6}$$

and

$$P_p(W) = S(t_m). \tag{7}$$

Using eq. (1), P_s is derived from W and the graphically obtained values of τ and $P_p(W)$.

When the dielectric has many discrete dipole relaxation times, the function becomes

$$S(t) = \sum_i [P_{pi}(W)\exp(1)]\,\frac{t}{\tau_i}\exp\left(-\frac{t}{\tau_i}\right), \tag{8}$$

which suggests that $S(t)$ has many peaks corresponding to each condition $t_{mi}/\tau_i = 1$, where t_{mi} is the time corresponding to the i-th peak in $S(t)$, τ_i is the i-th dipole relaxation time, and $P_{pi}(W)$ is the polarization of dipoles with τ_i.

In order to demonstrate that DCTS is a powerful method for evaluating τ in dielectrics, the following

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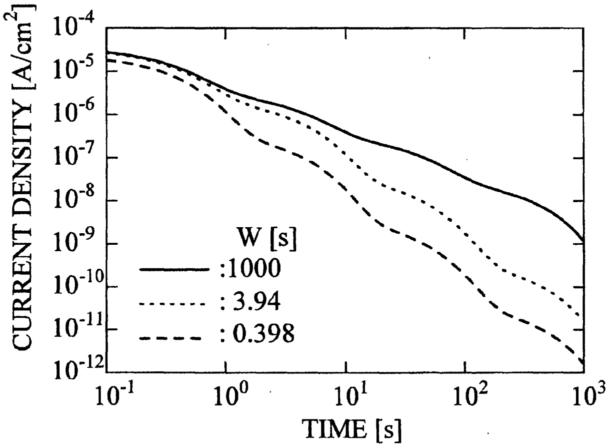

Fig. 1. Transient discharge current density $J_{\rm dis}(t)$ in a dielectric at three polarization times W of 0.398 s, 3.94 s and 1000 s. The curves were calculated using eqs. (1)–(3) with the parameters (τ [s], $P_{\rm s}$ [C/cm^2]) for four dipoles: (0.3, 1x10^{-5}), (3.0, 1×10^{-5}), (30, 1×10^{-5}) and (300, 1×10^{-5}).

$J_{\rm dis}(t)$ is considered. The solid line in Fig. 1 shows $J_{\rm dis}(t)$ calculated from eqs. (1)–(3) using the values $W = 1000$ s, $P_{\rm s1} = 1{\rm x}10^{-5}$ C/cm^2, $\tau_1 = 0.3$ s, $P_{\rm s2} = 1\times10^{-5}$ C/cm^2, $\tau_2 = 3.0$ s, $P_{\rm s3} = 1\times10^{-5}$ C/cm^2, $\tau_3 = 30$ s, $P_{\rm s4} = 1\times10^{-5}$ C/cm^2, and $\tau_4 = 300$ s, where $P_{{\rm s}i}$ is the steady-state polarization of dipoles with τ_i. The broken and dotted lines represent $J_{\rm dis}(t)$ for W values of 0.398 s and 3.94 s, respectively.

The $S(t)$ signal, which was calculated using eq. (4) and the data for $J_{\rm dis}(t)$ in Fig. 1, is shown in Fig. 2. In the solid line ($W = 1000$ s), there are four distinct peaks at $t_{\rm m1} = 0.398$ s, $t_{\rm m2} = 3.94$ s, $t_{\rm m3} = 37.8$ s, and $t_{\rm m4} = 296$ s. Thus, it is found that the dielectric has four dipole relaxation times in the range $0.1\,{\rm s} \le \tau \le 1000\,{\rm s}$. The values of τ_i and $P_{{\rm p}i}(1000)$ can easily be obtained from eqs. (6) and (7) using $t_{{\rm m}i}$ and $S(t_{{\rm m}i})$, respectively. The values of τ_i ($i = 1, 2, 3$) obtained by DCTS are greater than the actual values. For example, the value of τ_1 obtained graphically from the solid line in Fig. 2 is 0.398 s, but the actual value is 0.3 s. This difference arises because the first peak time is affected by dipoles corresponding to the second peak.

Consider the elimination of this effect. The broken line in Fig. 2 shows the $S(t)$ signal for $W = 0.398$ s (corresponding to the first peak in the solid line). From the broken line, the values of τ_1 and $P_{\rm p1}(0.398)$ are 0.312 s and 7.66×10^{-6} C/cm^2, respectively. The value of $P_{\rm s1}$ is 1.06×10^{-5} C/cm^2 from eq. (1) using $P_{\rm p1}(0.398) = 7.66\times10^{-6}$ C/cm^2, $\tau_1 = 0.312$ s and $W = 0.398$ s. These values are similar to the actual values of τ_1 and $P_{\rm s1}$. Therefore, it is found that reliable values of τ_1 and $P_{\rm s1}$ can be obtained using DCTS.

The $S(t)$ signal for $W = 3.94$ s (corresponding to the second peak in the solid line) is shown by the dotted line in Fig. 2. The values of τ_2 and $P_{\rm p2}(3.94)$ are estimated to be 3.08 s and 7.63×10^{-6} C/cm^2 from this data, which

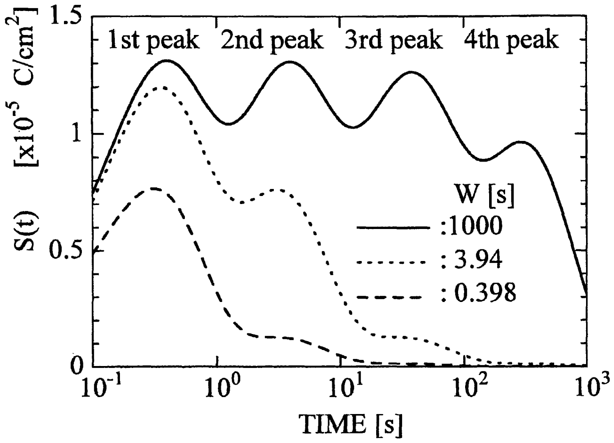

Fig. 2. DCTS signals calculated from the data in Fig. 1 using eq. (4).

gives $P_{\rm s2} = 1.06\times10^{-5}$ C/cm^2 using eq. (1). In the same manner, reliable values for τ_3, τ_4, $P_{\rm s3}$, and $P_{\rm s4}$ can be obtained using DCTS.

This method has the following advantages. By changing $F_{\rm p}$ or T in the DCTS measurement, the dependence of $P_{\rm s}$ on $F_{\rm p}$ or T can be investigated in detail. Moreover, the temperature dependence of τ, which is usually unknown, can be obtained exactly by changing T.

Consider the sensitivity of $P_{\rm s}$ in the DCTS measurement. When the measuring time is in the range 0.1 s to 1000 s, one can find values of τ between 0.1 s and 1000 s. If the minimum measurable current of an ammeter is of the order of 10^{-15} A, reliable values of $P_{\rm s}S$ are approximately 3×10^{-16} C at $\tau = 0.1$ s and 3×10^{-12} C at $\tau = 1000$ s according to eqs. (4), (6) and (7), where S is the electrode area of the capacitor.

DCTS was applied to the evaluation of dipole relaxation times (τ) in a dielectric. After the transient discharge current $J_{\rm dis}(t)$ is measured isothermally in a capacitor with a dielectric between two electrodes, the product of t and $J_{\rm dis}(t)$ is calculated. Since a graph of this product has many peaks corresponding to each condition $t/\tau = 1$, the values of τ can be evaluated graphically without assuming the temperature dependence of τ. Moreover, by changing T the temperature dependence of τ can be investigated in detail. Therefore, DCTS is suitable for evaluating τ in dielectrics.

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