A Surface Potential Investigation on Trap Creation in an Organic Dielectric

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SYNOPSIS

In view of the characteristics of space charge positions in an organic insulation dielectric under high electrical field, a surface potential model was put forward to investigate the dynamics of charge carrier capture. By the suggested comparative experimental method, trap creation under high field was expressed in the increment of surface potential. It is shown that the structural degradation is proportional to the time of the field application and to the exponential of the inverse of the field. Such a result was also verified by photoconductive current research. Taking a certain extent of trap creation as the critical condition of electrical breakdown, a widely used empirical formula about electrical aging was finally deduced. © 1996 John Wiley & Sons, Inc.

INTRODUCTION

It is well known that space charges have a great effect on the electrical properties of a dielectric and that they are the main causes of electrical breakdown.[1] Therefore, it is necessary to acquire a better understanding of the dynamics of space charge formation and on the relationship between trap the creation and high electrical field to find the breakdown mechanism. Until now, there have been many models about electrical breakdown in solid dielectric, such as avalanche breakdown,[2] electronic current runaway,[3] local defect extension,[4] and heat tunnel model,[5] etc., but they cannot illustrate the phenomena completely. This is due mainly to the fact that electrical breakdown is a combined result of electrical strength and the time of the field application. Today, two kinds of empirical formulas on electrical aging, i.e., the inverse power and exponential law, are widely used to describe the regularities of electrical aging in a dielectric.[6] Many scientists have tried to explain these empirical formula but failed at last, owing to lack of a suitable physical parameter which connected chemical structure and dielectric properties. In fact, impurities and defects are inevitably introduced into the dielectric. On the conception of the energy band, they can be expressed as local levels between conduction and valence band, which can trap carriers to form space charges. Electrical aging will eventually lead to structural degradation in the form of either new defect creation or an extension of the original ones. From the general characteristic of structure degradation in the dielectric, trap creation under high field is analyzed first by the increment of surface potential and of photoconductive current; then the electrical aging law is to be deduced.

DYNAMICS OF ELECTRON TRAPPING

Theory[7] and experiment[8,9] analyses on the place of space charge formation under high electrical field in a solid organic dielectric have stated that the carriers (electrons and holes) injected into the dielectric from electrodes are the same polars as the field; and the positions of the trapped carriers are usually in the range of 2 μm from the surface, which does not change even for the field application up to 18 days.[10] By the reported results, an approximate expression about space charge distribution in the dielectric after high field application is suggested as shown in Figure

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1, where the space charge densities are uniformly distributed near the electrodes, and both have the same magnitudes of ρ. When one side of the sample is connected with ground and the other is in the open circuit, the surface potential determined by space charges can be described by

$$V_f = \delta d \rho / \varepsilon_o \varepsilon \quad (1)$$

where ε_o is the permittivity of free space and ε of a dielectric; d, the thickness of sample; ρ, the charge density distributed in x-axis with the assumption of uniform distribution in y- and z-axises; and δ, the approximate thickness of the space charge distribution in x direction. With qn_t substituting ρ, eq. (1) can be expressed as

$$V_f = \delta d q n_t / \varepsilon_o \varepsilon = A n_t \quad (2)$$

where A is a constant; n_t, the number of the carriers captured by traps in unit volume of dielectric; and q, the electronic charge. Equation (2) shows that the surface potential is determined by the space charges on the nonground surface, and the magnitudes directly express the number of the carriers captured by traps in the sample.

The following only discusses the characteristic of electron injection, but it can be used for the hole.

The number n_t of the trapped electrons in a dielectric can be described by the first-order capture kinetic equation.[11] Assuming that the DC field in investigating the characteristic of electron capture is F, and that the original trap density is N_{to}, the rate of electron capture can be written as[11]

$$(\partial n_t / \partial t)_+ = n v \sigma_n (N_{to} - n_t) \quad (3)$$

where σ_n is capture cross section of the trap centers; n, the electron density in the conduction band of the dielectric, which is injected by the field F; and v, the thermal velocity of free electrons. However, eq. (3) is still difficult in explaining many capture phenomena,[10] and we must consider the detrapping process of the trapped electrons. When the field F is in the range of 1×10^5 to 1×10^6 V/cm, thermal emission of the trapped electrons is considered as the main detrapping mechanism and can be expressed as[12]

$$(\partial n_t / \partial t)_- = -e_n n_t \quad (4)$$

where thermal emission rate

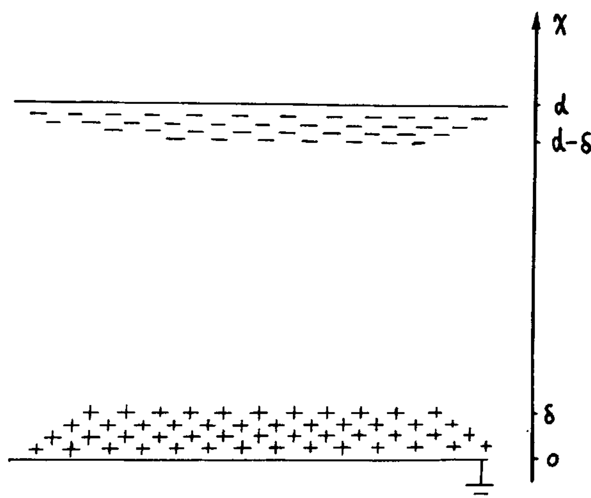

Figure 1 Space charge distribution in the organic dielectric after a high field was applied.

$$e_n = \omega \exp(-E_t / KT) \quad (5)$$

ω is an escaping frequency; E_t, the depth of the energy level of trap; T, absolute temperature; and K, the Boltzmann constant. The total capture rate is worked out by eqs. (3) and (4):

$$d n_t / dt = n v \sigma_n (N_{to} - n_t) - e_n n_t \quad (6)$$

At $t = 0$, field F has not yet injected electrons into the sample, i.e., $n_t|_{t=0} = 0$. The solution of eq. (6) can be written as

$$n_t(t) = [n v \sigma_n N_{to} / (n v \sigma_n + e_n)] \times \{1 - \exp[-(n v \sigma_n + e_n) t]\} \quad (7)$$

When expressed by surface potential, eq. (7) becomes

$$V_f(t) = V_f(\infty) \{1 - \exp[-(n v \sigma_n + e_n) t]\} \quad (8)$$

where

$$V_f(\infty) = A n v \sigma_n N_{to} / (n v \sigma_n + e_n) \quad (9)$$

is a steady state surface potential at dynamic equilibrium. Equation (9) shows that at a certain condition, the larger the N_{to}, the higher the $V_f(\infty)$. At a given field F, the drift current of electrons can be written as

$$j = n q \mu_e F \quad (10)$$

where μ_e is the drift mobility of electrons. In this range of fields, the electronic current injected into dielectric can be described by the Schottky emission current,[3] i.e.,

$$j = j_o \exp(\beta_s F^{1/2}/KT) \quad (11)$$

where j_o does not relate to F, $\beta_s = (q^3/4\pi\varepsilon_o\varepsilon)^{1/2}$. From eqs. (10) and (11), n can be deduced as (neglecting diffuse current)

$$n = J_o \exp(\beta_s F^{1/2}/KT)/q\mu_e F \quad (12)$$

It is seen from eq. (12) that with the increase of F, n increases exponentially with the field, resulting in $n_t(t)$ [or $V_f(t)$] tending to steady-state value quickly.

Once the trapped electrons were removed, the dielectric is taken to undergo the application of high field F_f, that is the process to research trap creation. Here, the F_f is the field used to study create traps, assuming that the newly created trap density is $\Delta N_t(F_f, t_f)$, which is the function of F_f and time t_f of F_f application, and that the capture cross section of the new traps is σ_{nf}. After getting rid of the trapped electrons captured during F_f application in the sample, the same field F as that in eq. (6) is applied. Consequently, the first-order capture kinetic equation containing newly created traps can be written as

$$dn_t/dt = nv\sigma_n(N_{to} - n_{t1}) - e_n n_{t1} + nv\sigma_{nf}(\Delta N_t - n_{t2}) - e_n n_{t2} \quad (13)$$

where $n_t = n_{t1} + n_{t2}$ is the total captured electron number; n_{t1}, the electron number captured by the original traps; and n_{t2}, the number by the newly created traps. The integration on eq. (13) gives

$$n_t(t) = [nv\sigma_n N_{to}/(nv\sigma_n + e_n)] \times \{1 - \exp[-(nv\sigma_n + e_n)t]\} + [nv\sigma_{nf}\Delta N_t/(nv\sigma_{nf} + e_n)] \times \{1 - \exp[-(nv\sigma_{nf} + e_n)t]\} \quad (14)$$

Correspondingly, the surface potential can be expressed as

$$V_f(t) = V_{f1}(\infty)\{1 - \exp[-(nv\sigma_n + e_n)t]\} + V_{f2}(\infty)\{1 - \exp[-(nv\sigma_{nf} + e_n)t]\} \quad (15)$$

where

$$V_{f1}(\infty) = Anv\sigma_n N_{to}/(nv\sigma_n + e_n)$$

and

$$V_{f2}(\infty) = Anv\sigma_{nf}\Delta N_t/nv\sigma_{nf} + e_n)$$

are the steady-state values of the potential resulted from electrons captured by the original and the newly created traps, respectively. The variation of surface potential increment caused by the captured electrons in newly created traps with the time of the F application is

$$\Delta V_f(t) = [Anv\sigma_{nf}\Delta N_t/(nv\sigma_{nf} + e_n)] \times \{1 - \exp[-(nv\sigma_{nf} + e_n)t]\} \quad (16)$$

If capture cross section of the newly created traps is the same as the original, i.e., $\sigma_n = \sigma_{nf}$, then eq. (13) can be simplified as

$$dn_t/dt = nv\sigma_n(N_{to} + \Delta N_t - n_t) - e_n n_t \quad (17)$$

Therefore, the kinetic characteristic of electron capture by the newly created traps is obtained

$$\Delta n_t(t) = [nv\sigma_n\Delta N_t/(nv\sigma_n + e_n)] \times \{1 - \exp[-(nv\sigma_n + e_n)t]\} \quad (18)$$

or expressed in surface potential

$$\Delta V_f(t) = [Anv\sigma_n\Delta N_t/(nv\sigma_n + e_n)] \times \{1 - \exp[-(nv\sigma_n + e_n)t]\} \quad (19)$$

Once dynamic equilibrium is reached, eq. (19) or (16) can be described as

$$\Delta V_f(\infty) = Anv\sigma_n(\sigma_{nf})\Delta N_t/[nv\sigma_n(\sigma_{nf}) + e_n] \quad (20)$$

Eq. (20) shows that except for $\Delta V_f(\infty)$ [or $\Delta V_f(t)$], any other parameters do not connect with ΔN_t; and whether σ_n is the same as σ_{nf} or not, the density of the newly created traps under high field can be expressed directly with the increment of surface potential.

STUDY METHOD FOR TRAP CREATION

Owing to the dispersity of the tested results in different samples, it is still very difficult to explain the mechanism of electrical aging. This paper views the electrical aging process from a relative variation of a physical parameter and puts forward a comparative experimental method, i.e., using a nondestructive physical parameter–surface potential to investigate

the process. For a general dielectric, there is still no other method that can be used to descrcibe the kinetic characteristic of electron trapping under a high electrical field, not mention to study the law of trap creation.[13] With the help of surface potential, the characteristic of electron capture under a high electrical field can be studied. It is shown that electron capture depends on the applied field and that near the breakdown field, impacting ionization detrapping between the free and the trapped electrons will happen.[13] Therefore, it is very hard to study trap creation directly from the kinetic characteristic of electron capture. Yet, eq. (16) tells us that the increment ΔV_f of surface potential can describe the kinetics of trap creation. In order to obtain the variation of ΔV_f with the high field F_f, a comparative experimental method is designed, as shown in Figure 2. At first, a reference DC field F is applied to the sample for a fixed time t_o, and the surface potential V_{fo1} at time t_o is written down. The relationship between the number of electrons captured by original traps and the time t of the F application is obtained, as shown in Figure 2 (Curve 1). The principle to choose the reference voltage lies in that under such a field application for time t_o; the new trap creation is negligible. On this principle, the DC field $F = 4 \times 10^5$ V/cm and $t_o = 30$ min are selected in our test. Then, after removing the trapped electrons which formed during the first step, the samples are used to perform the aging experiment at a high field F_f for time t_f.

Assuming the new traps created in the dielectric during the second step experiment equals $\Delta N_t(F_f, t_f)$, the third step test is performed, i.e., removing the trapped electrons obtained in the second step experiment from the sample to make the surface potential equal zero and then repeating the same procedure as that in the first step. Curve 3 in Figure 2 is obtained, and the surface potential V_{fo3} at $t = t_o$ is written down. If new traps have been created in the dielectric during the second step experiment, V_{fo3} will be larger than V_{fo1}. Such an increment, $\Delta V_f = V_{fo3} - V_{fo1}$, represents the magnitude of the new trap creation under F_f application for time of t_f. If no trap is created, ΔV_f will be zero, and Curve 3 in Figure 2 will superpose on Curve 1. By the process, the kinetics of trap creation under the field F_f in dielectric can be obtained.

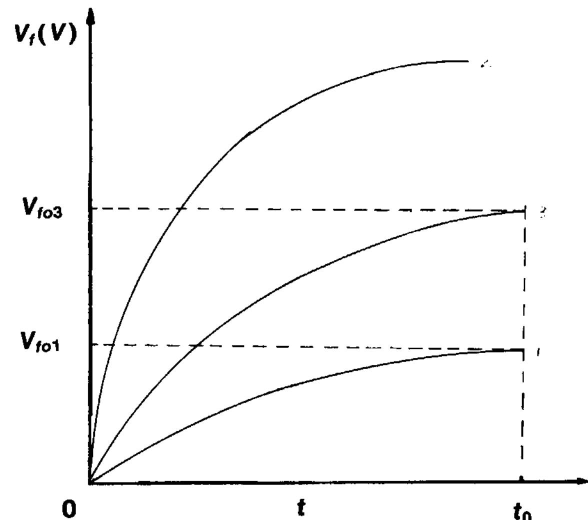

Figure 2 A demonstration of surface potential in the comparative experimental method for investigation into trap creation. Curves 1, 2, and 3 are the variation of surface potential with the time t in the first, second, and third step experiments, respectively. Curve 3 is obtained in the same condition as the first step. $\Delta V_f = V_{fo3} - V_{fo1}$ represents the created traps. If no trap creation, Curve 3 will superpose on 1.

EXPERIMENTAL RESULTS AND DISCUSSION

Polypropylene (PP) and polyethylene terephthalate (PET) organic films with the thicknesses of 12 and 20 μm, respectively, are selected as the test samples. Aluminium electrode with a diameter of 30 mm is coated by vacuum deposition on one side of the sample, which is grounded as a reference potential. A polished aluminium bulk pressed on the sample is taken as another electrode, through which the continuously adjusted dc F and F_f are applied, respectively. Once a given condition is reached, remove the field and the free electrons on the surface of sample with a large resistance.

The surface potential is measured by noncontacting method, a power-driven capacitance probe method, with an area of probe of 5×5 mm^2. All experiments are carried out in the temperature of 20°C and dry atmospheric conditions.

If a negative dc field F has not yet been applied, the surface potential of sample is zero; but it is not zero once F is applied. Such a result indicates that the surface potential is caused by electrons injected by the field and captured by traps in the sample. The experiments show that after the second step dc field F_f was applied, the capture curve in the third step has the same shape as that in the first, but they have different amplitudes. This means that the application of the dc F_f has almost no effect on the transient characteristic of the electron capture, and the nature of the newly created traps in the sample

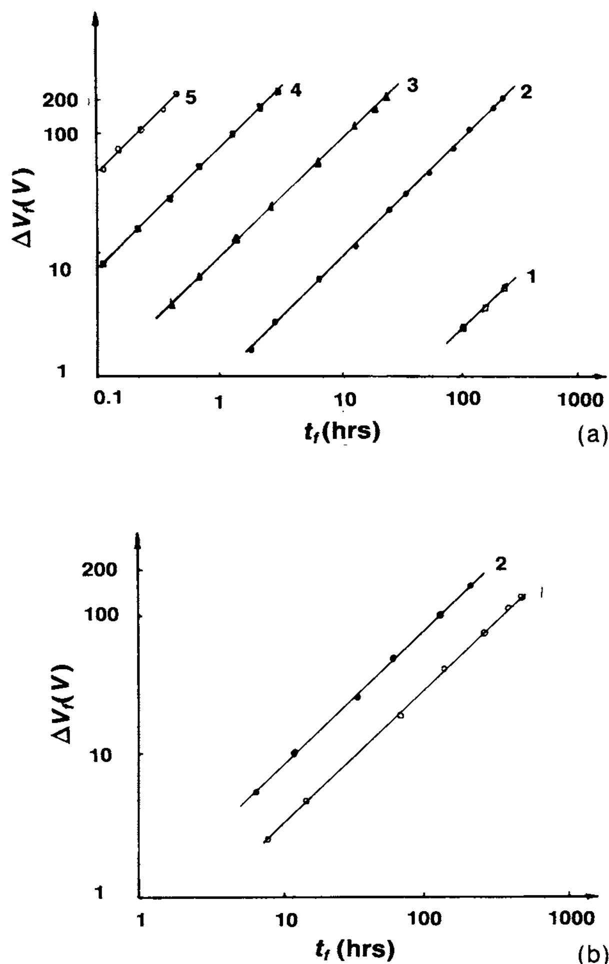

Figure 3 The increment ΔV_f of surface potential as a function of the time t_f of the field F_f application. (a) PET films; Curves 1, 2, 3, 4, and 5 correspond to the fields of 7.5×10^5, 1×10^6, 1.5×10^6, 2×10^6, and 2.5×10^6 V/cm, respectively. (b) PP films; Curves 1 and 2 correspond to the fields of 1×10^6 and 1.25×10^6 V/cm, respectively.

is the same as that of the original. Therefore, it is certain that the increment of the surface potential is caused by the electrons captured by new traps. Figure 3 shows the variation of the surface potential increment in PET and PP films with the dc F_f and the time t_f of the F_f application, which is obtained by the comparative method mentioned above. Each tested datum is an average value of three samples. The highest dc F_f is 2.5×10^6 V/cm. The field higher than that will easily result in electrical breakdown so that it is hard to obtain the experimental results. At the time of the experimental performance, new trap creation in the dielectric is not effective if the dc F_f is less than 1×10^6 V/cm. This is due mainly to the fact that when the field is higher than 1×10^6 V/cm, electron density injected into the dielectric by fields is very large, and many electrons obtain high energy from the field, which make the rate of molecular chain breakdown or the probability of the original defect extension increase. When the field less than that, electron density injected into it is small, and a fewer number of electrons get high enough energy to destroy molecular structure, resulting in less trap creation. Figure 3 shows us that trap creation is proportional to the time of the field F_f application.

Figure 4 is the variation of the increment ΔV_f of surface potential in PET with F_f, which indicates that the increment is proportional to the exponential of the inverse of the field. It is seen from Figures 3 and 4 that for different field F_f applications or different times t_f of the field application, the magnitude of created traps tends to be of the same values, which are higher values than that resulting in electrical breakdown. Such a result states that the lifetime of the sample will be ended once trap density reaches the critical value. For PET films, the lifetime t_f of the sample as a function of $1/F_f$ is shown in Figure 5, in which each datum is taken from the average of ten samples, and the vertical lines stand for standard deviation. The results demonstrate that the lifetime in PET films is proportional to the exponential of the inverse of the field.

In order to verify the results further, a photoconductive current spectrum is used to analyze the energy level depth and the density of traps. The peak positions at the wavelength axis display the level depth, and the peak amplitudes indicate the trap density. Figure 6 is a group of photoconductive current curves of the original and the aged PP samples at a field of 1×10^6 V/cm for different aging times.

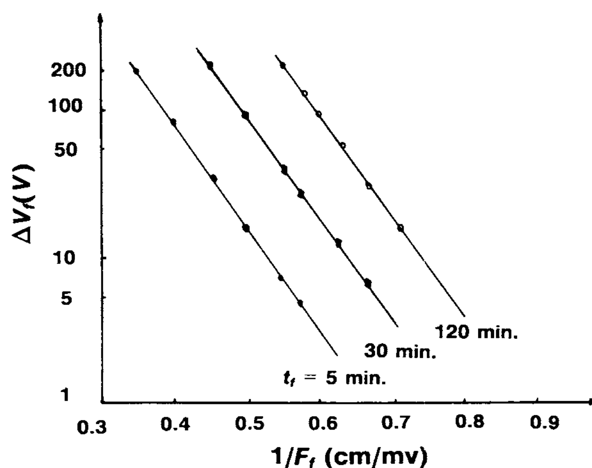

Figure 4 The increment ΔV_f of surface potential versus the inverse of F_f in PET films.

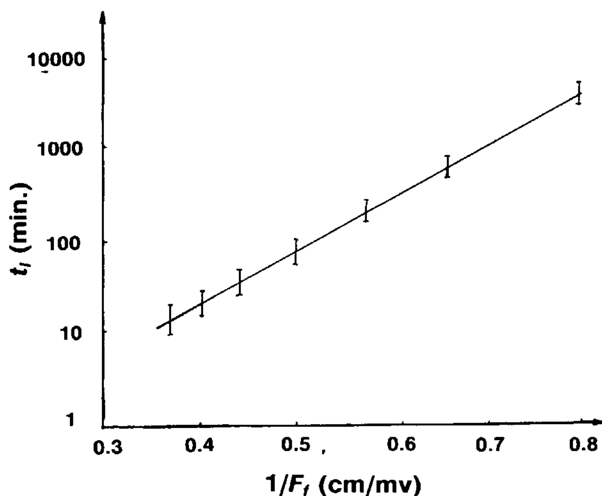

Figure 5 The lifetime t_l against F_f in PET films.

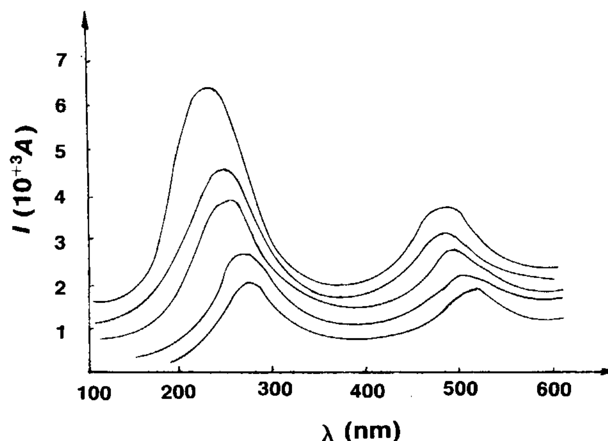

Figure 6 Photoconductive current spectrum of PP films at a field of 1×10^6 V/cm for different of times. Curves 1, 2, 3, 4, and 5 correspond to the aging time of 0, 60, 120, 240, and 500 h, respectively.

With an increase in the aging time, both current amplitudes at the wavelength of 265 and 507 nm increase, and their peak positions move toward the shorter wavelength. As a function of aging time, the increment of photoconductive current is shown in Figure 7. It is conspicuous that the increment of the current amplitude is proportional to the aging time.

There are many factors, such as radiation, oxidation, ion injection, high electrical field, etc., which can result in trap creation in the solid dielectric. This paper considers electrical degradation, and only the field effects on dielectric are considered. Under a high field F_f application, the field F_f injects electrons into the dielectric, and the electrons will get a lot of energy from the field. The experiments have shown that it is hot electrons under high field that create new traps in the solid dielectric.[14,15] Therefore, trap creation is dependent on the number of electrons injected by the field. Assuming that under high field F_f, the field injects electronic current J; and the number of electrons injected in the period of time dt is Jdt/q, where q is electronic charge. Introducing a trap creation coefficient γ, which is the function of field,[16] the rate of new trap creation in the dielectric under the field F_f can be expressed by

$$dN_t = \gamma J dt/q \qquad (21)$$

where

$$\gamma = \gamma_o \exp(-H/F_f) \qquad (22)$$

γ_o and H have nothing to do with field. When the field F_f is larger than 1×10^6 V/cm, the current J injected by the field can be described by Fowler-Nordheim tunnel current[3]

$$J = D_o F_f^2 \exp(-B/F_f) \qquad (23)$$

Combining eqs. (21), (22), and (23), we obtain that

$$dN_t = (\gamma_o D_o F_f^2/q)\exp[-(H+B)/F_f]dt \qquad (24)$$

Assuming that the primary trap density in dielectric is N_{to}, the trap density in the period t_f of the field F_f application will be

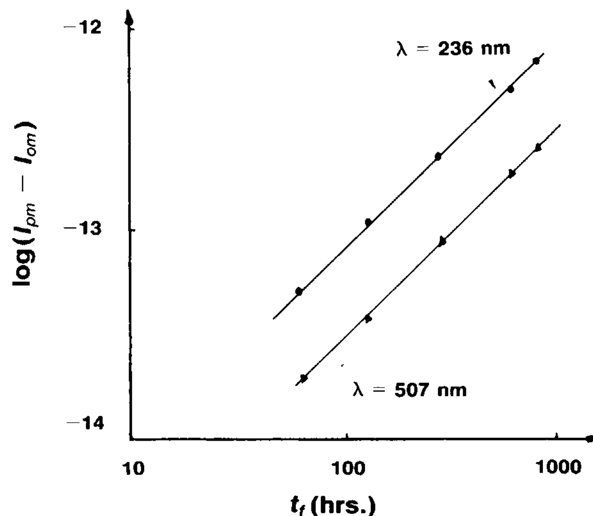

Figure 7 The increment of photoconductive current amplitude, i.e., $(I_{pm} - I_{om})$ as a function of the electrical aging time. I_{pm} is the amplitude of the photoconductive current at different aging time; I_{om} is that of the original PP sample.

$$N_t(F_f, t_f) = N_{to} + (\gamma_o D_o t_f F_f^2/q) \times \exp[-(H+B)/F_f] \quad (25)$$

If the variation of F_f lies in the range from 1×10^4 to 1×10^6 V/cm, J can be expressed by eq. (11). Inserting eqs. (11) and (22) into (21), N_t can be worked out as

$$N_t(F_f, t_f) = N_{to} + (\gamma_o J_o t_f/q) \times \exp(-H/F_f + \beta_s F_f^{1/2}/KT) \quad (26)$$

It can be seen from eq. (26) that ln N_t is the hyperbolic function of F_f in H/F_f, and the parabolic function of F_f in $(\beta_s F_f^{1/2}/KT)$, so the change of the first term with field F_f is faster than that of the second. Eqs. (25) and (26) show that under high field, new trap creation is proportional to the exponential of the inverse of a field and to the time of the field application. Substituting eqs. (25) and (26) into eq. (20), the kinetic process of trap creation can be expressed by

$$\Delta V_f(\infty) \approx G t_f \exp[-(M/F_f)] \quad (27)$$

where G and M do not relate to field F_f. ΔV_f essentially represents the degradating degree of the dielectric microstructure after the F_f was applied for the time of t_f. Once the dielectric degradates to the degree of $\Delta N_t = \Delta N_{t\,\max}$, breakdown occurs unavoidably. When expressed by the increment of surface potential, the lifetime t_{lc} of the dielectric under a high field F_f can be worked out from eq. (27), i.e.,

$$t_{lc} \exp(-M/F_f) = \text{const} \quad (28)$$

which is consistent with the empirical formula of electrical aging[6] and also with many experimental and numerical calculating results.[17,18]

By the results obtained in this paper and the impacting ionization between the free and the trapped electrons reported in,[13] an electrical aging and breakdown mechanism is suggested as follows.

Under the application of high electrical fields, carriers (electrons or holes) are injected into the dielectric. For simplicity, only electrons are considered. Electrons injected into the conduction band of a dielectric will obtain energy from the field and appear in the form of energy distribution.[19] These electrons behave mainly in three aspects: Part of electrons are to be trapped; part of them get a certain energy from the field resulting in the trapped electrons detrapping while they impact with the trapped electrons; and part of them obtain high energy from the field to cause new trap. Generally, trap density in solid dielectric exists most likely in the form of energy distribution and all those energy levels may capture electrons. The energy of electrons that makes the trapped electrons detrapping does not require too much, but 1.5 times the trapped energy level will be high enough to make the trapped electrons in the level detrap by impacting ionization.[20] Consequently, the impacting ionization between the free and trapped electrons in the dielectric under high field is obvious. At the same time, the third part of electrons make the dielectric create new traps, which also capture electrons and cause impacting ionization. Hence, impacting ionization enhances the ionization coefficient, and the detrapping rate will be reported elsewhere. With the prolongation of the time of the field application, a new trap generates continuously, resulting in further impacting ionization until the local energy levels the extending and connecting among them. The trapped electrons in this local level will become a nearly free state and obtain higher energy from the field, which makes the trapped electrons in the deeper level away from them about 1.5 times the energy level detrap. Such a degrading circulation of trap creation, and the trapped electrons detrapping due to the impacting ionization, finally form large current and electrical breakdown occurs.

CONCLUSION

From the characteristic of space charge distribution in the organic dielectric under high electrical field, a surface potential model is suggested in this paper, with which the kinetic properties of electron capture are described. To limit the dispersity of electrical aging data, a comparative experimental method is designed to investigate trap creation in the organic dielectric. It is shown that the structural degradation is proportional to the time of the field application and to the exponential of the inverse of a field. Introducing a trap creation coefficient, a kinetic theory on trap creation in the organic dielectric under high field is deduced. Taking a certain amount of trap creation as the critical condition of electrical breakdown, it is found that the electrical lifetime of organic dielectric is proportional to the exponential of the inverse of a field. Such a theory is consistent with the widely used empirical formula and agrees with experimental results. Finally, the electrical breakdown process is explained from the combined effects of the field and the time of a field application.

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