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1997 J. Phys.: Condens. Matter 9 9289

(http://iopscience.iop.org/0953-8984/9/43/013)

J. Phys.: Condens. Matter **9** (1997) 9289–9298. Printed in the UK PII: S0953-8984(97)84954-5

Dynamics aspects of the charging behaviour of polymers under focused electron beam irradiation

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Received 10 June 1997

Abstract. A set of classical electrostatic equations is developed to study the charging behaviour of polymers under focused electron beam irradiation. The calculated results reveal the dependences of the space charge trapped in a polymer under focused electron beam irradiation on the relative permittivity, the radiation-induced conductivity of the sample and the irradiation conditions, which agree well with previous experimental results.

1. Introduction

Polymers, mostly as insulating materials, have been applied in a variety of fields such as packaging materials in microelectronics and HV transportation cables. When they are irradiated by a nonpenetrating electron beam, a space charge, resulting from electron trapping, is observed and has been studied by various authors [1–7]. Recently, many experimental techniques such as the thermal pulse method [8, 9], the pressure wave propagation method [10–12], the pulsed electro-acoustic method [13–15], the Kerr electro-optic method [16, 17], the mirror image method [18–22] and the time-resolved current method [23] have been proposed to measure the space charge and its distribution in a polymer. It is found experimentally that the space charge trapped in the polymer is strongly related to the relative permittivity [24, 25] and the radiation-induced conductivity [26] of the polymer, as well as the experimental conditions [23] such as the electron-beam energy and current under which the polymer is irradiated.

On the theoretical side, much effort has been concentrated on the simulation of the space charge spatial distribution by Monte Carlo simulation (MCS) [27, 28], where the single-scattering technique is used and the influence of the accumulated charge in the polymer on the forthcoming electrons is considered. Another method is to solve the continuity and Poisson's simultaneous equations under appropriate boundary conditions [19–31]. However, none of these methods is able to describe the evolution of the space charge trapped in the polymers under electron beam irradiation with irradiation time satisfactorily.

In the present paper, we attempt to derive a set of formulae by classical electrostatics to describe the evolution of the space charge trapped in a polymer under focused electron beam irradiation, where the influence of the trapped charge on the incoming electrons is taken into account.

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0953-8984/97/439289+10$19.50

2. Calculation

When a polymer is irradiated by an electron beam, both secondary and backscattered electrons are generated. However, some of electrons are trapped in the polymer, forming a space charge distribution and resulting in a displacement current [32]. As the electrons build up, a potential created by the space charge will decelerate the incoming beam electrons and so lower the effective beam energy, leading to the emission of more secondary electrons. On the other hand, some of the trapped electrons can also be detrapped or diffused due to the electric field arising from the space charge, leading to a leakage current. Therefore, according to the principle of current conservation, one has

$$I_0 = I_{bs} + I_d + I_l \tag{1}$$

where I_0 is the incident electron beam current; I_{bs} is the sum of the secondary and backscattered electron currents; I_d is the displacement current and I_l is the leakage current. We assume that the amount of space charge trapped in the polymer at irradiation time t is $Q(t)$ and its spatial distribution to be a Gaussian distribution expressed as follows,

$$\rho(r', t) = \frac{2Q(t)}{(2\pi)^{3/2}R^3} \mathrm{e}^{-r'^2/2R^2} \tag{2}$$

where r' is the radial distance from the electron beam incidence point and R is the standard deviation of the Gaussian distribution, referred to as the space charge distribution range, which is found to increase with time at the beginning and eventually reach a saturated value as described in our previous paper [33]. However, the dependence of R on t is difficult to express analytically. Here, we choose to take the Kanaya–Okayama range [34] as an approximation of R which does not vary with time and can be expressed as

$$R = 0.0276 A E_0^{1.67}/Z^{0.89} d \tag{3}$$

where E_0 is the incident electron beam energy in keV, A is the atomic weight of the sample in g mol^{-1}, Z is the atomic number and d is the density in g cm^{-3}.

Furthermore, from equation (2), the time variation of surface potential ($\varphi(t)$) at the electron beam incidence point can be written as

$$\varphi(t) = 4\frac{K}{4\pi\varepsilon_0}\int_0^{\pi/2}\mathrm{d}\theta\int_0^{\pi/2}\mathrm{d}\varphi\int_0^{\infty} r'^2 \frac{2Q(t)}{(2\pi)^{3/2}R^3}\mathrm{e}^{-r'^2/2R^2}\frac{\mathrm{d}r'}{r'} = \frac{KQ(t)}{(2\pi)^{3/2}\varepsilon_0 R} \tag{4}$$

where $K = 2.0/(1+\varepsilon_r)$, ε_r is the relative permittivity of the sample and ε_0 is the vacuum permittivity. Since the electrons impinging at the surface of sample will be decelerated by the surface potential, the effective incident electron beam energy ($E_{eff}(t)$) can be determined by E_0 and $\varphi(t)$ as follows

$$E_{eff}(t) = E_0 - e\varphi(t) \tag{5}$$

where e is the absolute value of the electronic charge.

The sum of the secondary and backscattered electron yields can be expressed as follows according to the Burke universal law [35],

$$\sigma(t) = K_1 E_{eff}(t)^{-n_1} + K_2 E_{eff}(t)^{-n_2} \tag{6}$$

where the values of K_1, n_1, K_2, n_2, are only materials dependent [35].

At any given irradiation time t, if the irradiation time increases by a short period of time Δt, the increase in $Q(t)$ during Δt is approximately calculated by

$$\Delta Q(t) = I_0(1-\sigma(t))\Delta t - I_l(t)\Delta t. \tag{7}$$

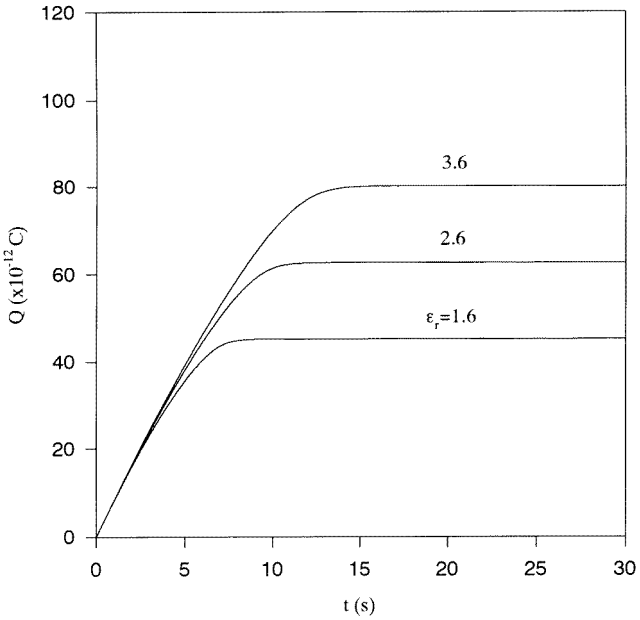

Figure 1. The evolution of the trapped charge with irradiation time for different relative permittivity.

Table 1. The parameters in our calculation.

Parameter	Value
A	6.7
Z	3.6
d	1.2 (g cm^{-3})
ε_0	8.85×10^{-12} (C^2 N^{-1} m^{-2})
k_1	1.032
k_2	0.115
n_1	0.725
n_2	0.223
ε_r	1.6, 2.6, 3.6
g_i	1.0×10^{-15}, 5.0×10^{-15}, 5.0×10^{-14} (S cm^{-1})
E_0	20, 25, 30 (keV)
I_0	5.0×10^{-12}, 1.0×10^{-11}, 5.0×10^{-11} (A)

We define $\tau = \varepsilon_0 \varepsilon_r / g_i$ [36] as the relaxation time of the charge trapped in the polymer, which is equal to the lifetime of a trapped charge. The leakage current, contributing from detrapping, therefore can be expressed as

$$I_l(t) = \frac{Q(t)}{\tau} = \frac{g_i Q(t)}{\varepsilon_0 \varepsilon_r} \tag{8}$$

where g_i is the radiation-induced conductivity. Strictly speaking, g_i is determined by the radiation dose rate [37]. However, experimental results demonstrate that with the onset of irradiation, $g_i(t)$ increases rapidly with time and reaches a steady-state or equilibrium value [38]. So, in our calculation, g_i is referred to as the steady-state value.

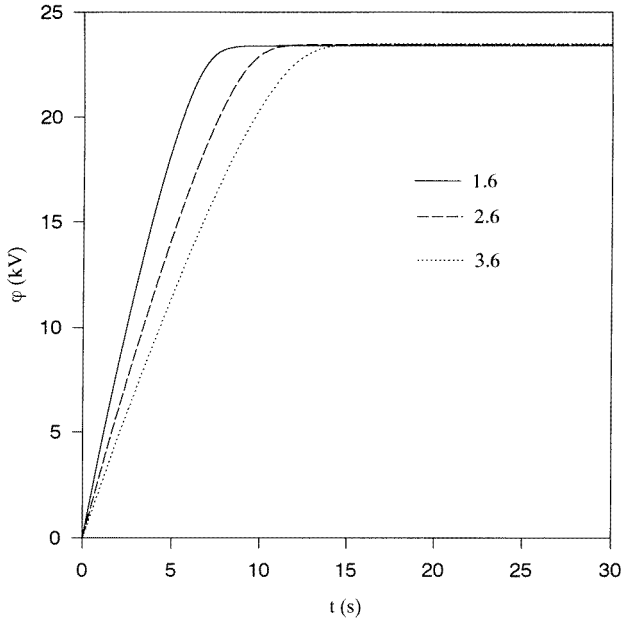

Figure 2. The evolution of the surface potential with irradiation time for different relative permittivity.

The procedures of calculation are summarized as follows. When $t = 0$, assuming $Q(0) = 0$, we have $\varphi(0) = 0$ and $I_l(0) = 0$ according to (3), (4) and (8). $\sigma(0)$ can be calculated by (5) and (6) as E_0 is known. If we set the time step to be 0.1 s, $\Delta Q(0.1)$ can be calculated by (7) as I_0 is known. Therefore at $t = 0.1$, we have $Q(0.1) = Q(0) + \Delta Q(0.1)$. Reiteration of the above procedure allows determination of the evolution of the space charge trapped in the polymers under a focused electron beam irradiation with irradiation time until saturation of the trapped charge.

3. Calculation results and discussion

Our calculation is carried out on a sample of PMMA. Its basic parameters used in the calculation are listed in table 1. Its actual relative permittivity and radiation-induced conductivity are reported to be 2.6 and about 5.0×10^{-15} S cm^{-1} [14] respectively. However, in order to study the dependence of the charging behaviour of the polymer on the relative permittivity and the radiation-induced conductivity, several values are used (see table 1). The relationship between the charging behaviour of the polymer and the irradiation conditions is also studied.

3.1. The relative permittivity dependence

The evolution of the trapped charge and the surface potential of the sample with irradiation time has been calculated and shown in figure 1 and figure 2 for various values of relative permittivity. The radiation-induced conductivity, electron beam energy and current are kept constant at 5.0×10^{-15} S cm^{-1}, 25 keV and 1.0×10^{-11} A respectively. One can find that the trapped charge and the surface potential of the polymer initially increase with the irradiation

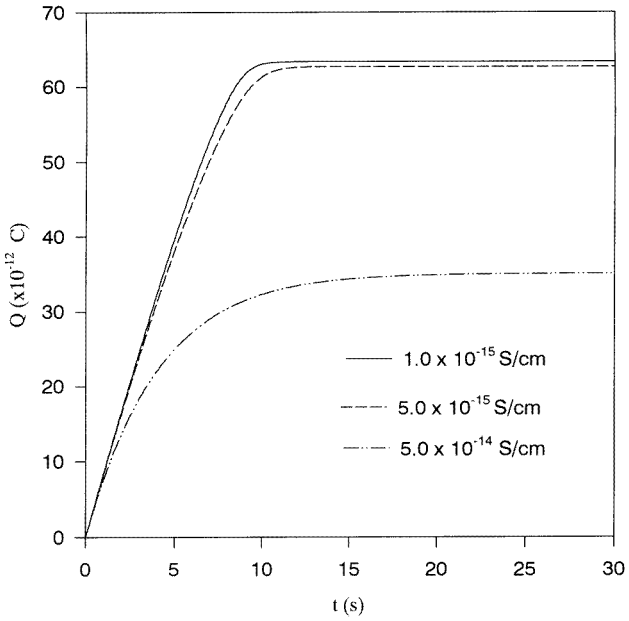

Figure 3. The evolution of the trapped charge with irradiation time for different radiation-induced conductivity.

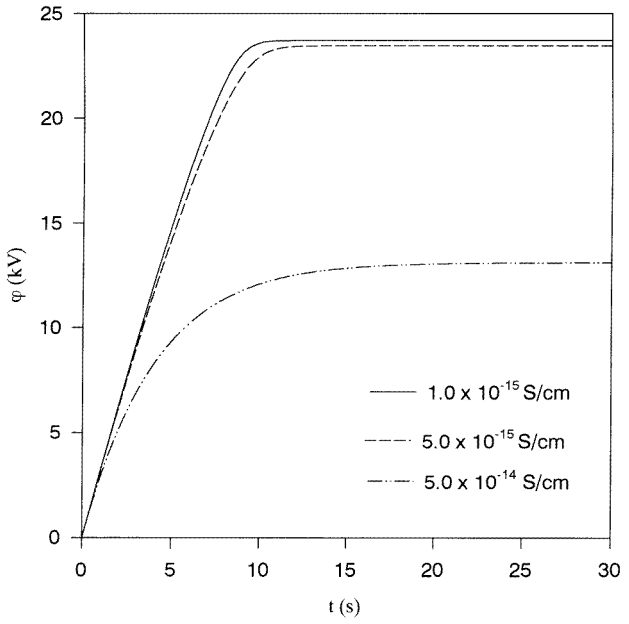

Figure 4. The evolution of the surface potential with irradiation time for different radiation-induced conductivity.

time and eventually saturate. However, the saturated values for the surface potential are almost the same, while the saturated values for the trapped charge increase with increasing

relative permittivity of the material. This agrees well with the experimental results reported before [24]. This can be explained as follows. In our calculation, the breakdown is not taken into account, so, the surface potential of the sample can increase nearly to the potential of the electron gun for different values of relative permittivity. As a result, the trapped charge should be proportional to the relative permittivity of the material.

Figures 1 and 2 also show that the irradiation time for the trapped charge to saturate is different for different values of relative permittivity. This is due to the same incident electron beam current, but different saturated trapped charge.

3.2. The radiation-induced conductivity dependence

To study the radiation-induced conductivity dependence, we vary g_i and keep the relative permittivity, incident electron beam energy and current constant at 2.6, 25 keV and 1.0×10^{-11} A, respectively. The calculated results are shown in figures 3 and 4. Figures 3 and 4 demonstrate that both the saturated values of the trapped charge and the surface potential of the sample decrease as the radiation-induced conductivity increases. This is because the higher radiation-induced conductivity causes more leakage of the trapped charge, resulting in a reduction of the saturated value of the trapped charge.

3.3. The incident electron beam energy dependence

Figures 5 and 6 show the evolution of the trapped charge and the surface potential of a sample with irradiation time for a radiation-induced conductivity of 5.0×10^{-15} S cm^{-1} and relative permittivity of 2.6, where the sample is irradiated by different-energy electron beams with the same current of 1.0×10^{-11} A. As expected, both the saturated values of the trapped charge and the surface potential of the sample increase with increasing the incident electron beam energy. This is because when no breakdown occurs, saturation takes place when the surface potential approaches the potential of the electron gun, leading to the net current being injected into the sample becoming zero. Therefore higher electron beam energy leads to higher surface potential and hence higher trapped charge in the sample. Figures 5 and 6 also indicate that the higher the incident electron beam energy, the longer the time needed for saturation of the trapped charge. This is due to the same factor as discussed in section 3.1.

3.4. The incident electron beam current dependence

Figures 7 and 8 show the charging behaviour of the sample with radiation-induced conductivity of 5.0×10^{-15} S cm^{-1} and relative permittivity of 2.6, irradiated by an electron beam with energy of 25 keV, for different currents. Figures 7 and 8 indicate that the saturated values of the trapped charge and the surface potential of the sample are slightly different for different incident electron beam currents. This is consistent with our experimental results. However, because of the difference of the spot area for different beam currents [39], the difference of the experimental data for different beam currents is bigger than the calculated data. Again, a sharper slope of the curve and shorter time for the trapped charge to saturate for a larger beam current is due to the larger charging rate resulting from a larger beam current.

It should be mentioned that in actual experiments, when a sample is irradiated by high-energy and large-current electron beams, breakdown is often initiated [40, 41] which will destroy the established space charge distribution and minimize the surface potential of the

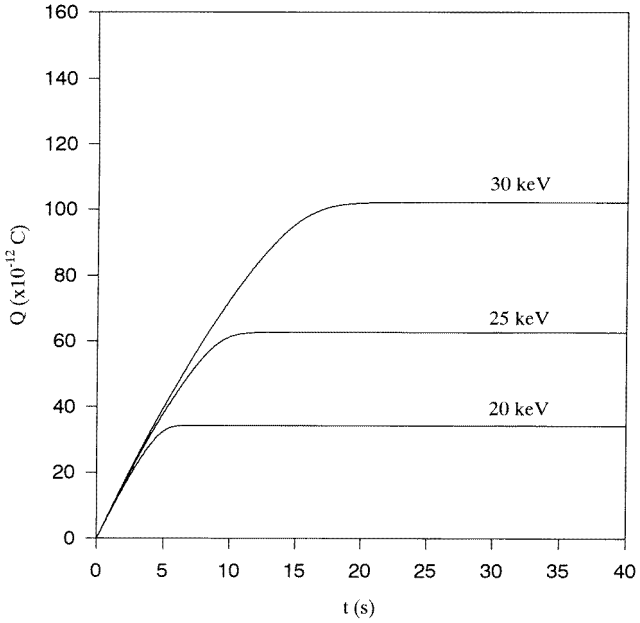

Figure 5. The evolution of the trapped charge with irradiation time for different beam energy.

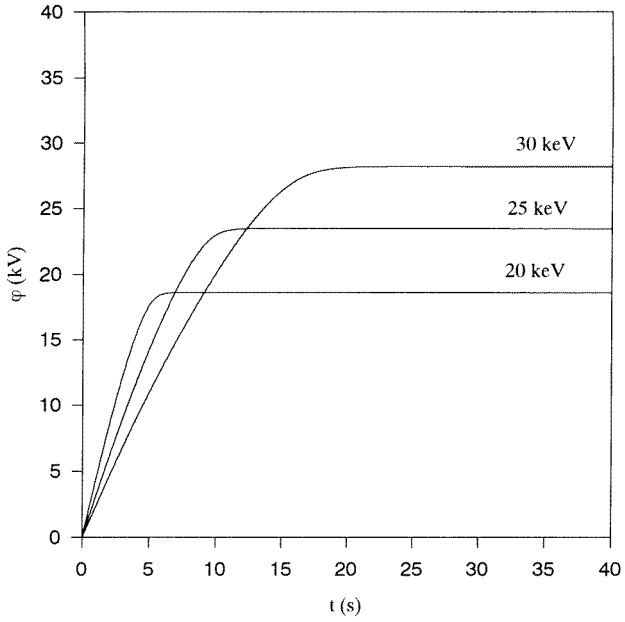

Figure 6. The evolution of the surface potential with irradiation time for different beam energy.

sample before attaining the incident electron beam accelerating voltage. After that, the charge will be trapped again and the above process will repeat itself. In our calculation, no breakdown is taken into account, but the simulation results such as the dependence of

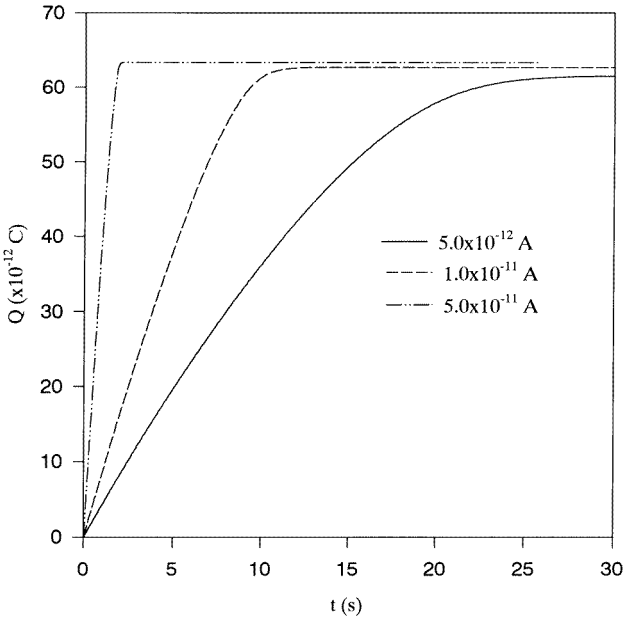

Figure 7. The evolution of the trapped charge with irradiation time for different beam current.

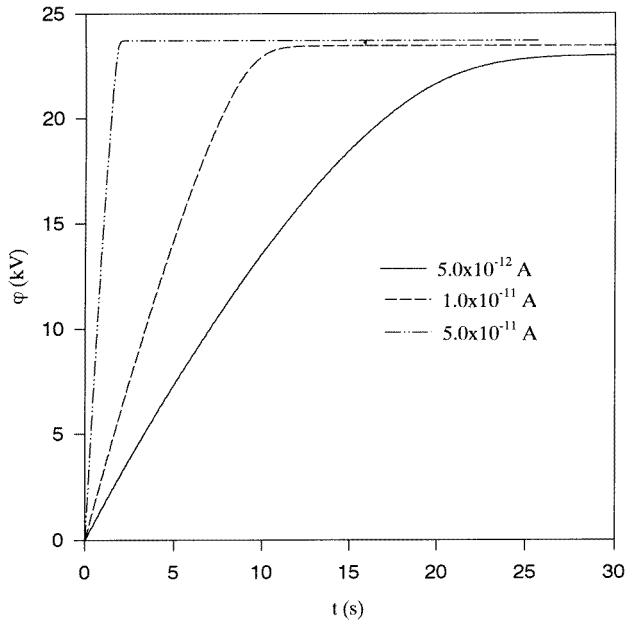

Figure 8. The evolution of the surface potential with irradiation time for different beam current.

the trapped charge on the relative permittivity, the radiation-induced conductivity, incident electron beam energy and current agree well with the experimental results. Hence if breakdown occurs, the calculation still gives a good description of the charging behaviour of the sample prior to occurrence of breakdown.

We also note that the calculated values of the saturated trapped charge are less than the corresponding experimental data. This is because (i) the surface potential of the sample may deviate the electron beam from the normal direction, leading to a larger electron beam spot and (ii) the radiation-induced conductivity [42] and the non-zero mobility [43, 44] of charge in the material result in a progression outwards of the trapped charge under an electric field arising from the trapped charge. Both lead to an increase of the space charge distribution range [45] which makes the material trap more charge. However, this does not affect the dependence of the trapped charge on the relative permittivity, radiation-induced conductivity and incident electron beam energy and current.

4. Conclusion

We have calculated the charging behaviour of polymers under focused electron beam irradiation. The calculated results reveal that the trapped charge is related to the properties of the material and increases as the relative permittivity of the material increases and as the radiation-induced conductivity decreases. The trapped charge is also dependent on the irradiation conditions and increases with increasing incident electron beam energy and current. These predictions agree well with the previous experimental results.

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