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Space-charge dynamics of polymethylmethacrylate under electron beam irradiation

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Abstract. Space-charge dynamics of polymethylmethacrylate (PMMA) under electron beam irradiation has been investigated employing a scanning electron microscope. Assuming a Gaussian space-charge distribution, the distribution range (σ) has been determined using a time-resolved current method in conjunction with a mirror image method. σ is found to increase with irradiation time and eventually attain a stationary value. These observations have been discussed by taking into account radiation-induced conductivity and charge mobility.

1. Introduction

It has been recognized for a long time that irradiation of polymers with a non-penetrating electron beam leads to the build-up of a space charge due to electron trapping [1]. Indeed, space charge is useful in electrets [2] or piezoelectric materials, but it can also produce disastrous effects, for instance in a DC or even AC high-voltage insulator. The accumulated space charge can distort the original electric-field distribution greatly, and sometimes even cause breakdown of the insulator [3]. Therefore, it is interesting and important to measure and study the space charge and its distribution in the insulator.

In recent years, many experimental techniques have been proposed to measure space charge and its distribution in insulators [4]. The most popular and well developed methods include the pressure wave propagation (PWP) method [5–7], the pulsed electro-acoustic (PEA) method [8–10] and the mirror image method (MIM) [11–15]. These methods have been successfully applied to the samples of polymethylmethacrylate (PMMA) [16], polyethylene (PE) [17], polyfluoroethylenepropylene (Teflon^{TM} FEP) [18], polyimide (Kapton^{TM} PI) [19] and polypropylene (PP) [20] etc. Some experimental results are confirmed by computer simulation, in which the one-dimensional continuity and Poisson's simultaneous equations are solved under appropriate boundary conditions, taking into account the electron-irradiation parameters such as radiation-induced conductivity (RIC) and the electron deposition profile [21–23]. Recently, with the help of the MIM employing a scanning electron microscope (SEM), the charging behaviour of an insulator by a focused electron beam has been studied extensively [11, 12, 24]. Chen, Gong and Ong [24] have determined the steady-state space-charge distribution volume in electron-beam-irradiated insulators by the MIM [11, 12, 14] employing an SEM. However, to deeply understand the

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RIC and charge transport mechanisms in an insulator, it is necessary to study the evolution of the space-charge distribution with irradiation time, i.e. the dynamics of space charge.

The present paper aims at studying the space-charge dynamics of PMMA under electron beam irradiation with a time-resolved current method [25] in conjunction with the MIM [11, 12, 24] employing an SEM. The details of the measuring method are described in section 2.

2. Measuring method

2.1. Experimental procedures

PMMA samples were cleaned carefully with hexane and stored in vacuum for at least 24 h prior to experiments. In our experiments, a JEOL-JSM-35CF SEM equipped with a JEOL 50 A-LNB liquid nitrogen baffle for obtaining a clean vacuum was employed. The sample was mounted on the sample holder which was connected to the ground through a Keithley 485 low-impedance picoammeter, and its upper free surface was perpendicular to the electron beam. The electron injection was carried out by a high-energy electron beam in the range 5–25 keV with current varying from 5×10^{-12} to 50×10^{-12} A at the spot mode. The trapped charge (Q) was obtained from the current recorded by the picoammeter according to the time-resolved current method which we have described in detail previously [25].

After electron injection in the sample, a potential distribution $V(r)$, where r is the distance from the field point to the charge centre, was set up by the trapped charge. The MIM experiment was performed by an 1 keV electron beam scanning across the charged area of the sample. The radius (r) of the equipotential surface that reflects the 1 keV electron beam can be determined by the mirror image method. For more details about the MIM, reference is made to previous publications [11, 12, 14].

2.2. The calculation of the space-charge distribution range (σ)

The density of trapped charge gradually decreases beyond the point of incidence of the beam on the insulator surface (non-penetrating beam), and a Gaussian distribution is assumed to be a good approximation [26, 27]:

$$\rho(r') = \frac{2Q}{(2\pi)^{3/2}\sigma^3} \mathrm{e}^{-r'^2/2\sigma^2} \tag{1}$$

where Q is the total trapped charge, r' is the radial distance from the electron-beam incidence point and σ is the standard deviation of the Gaussian distribution, referred to as the space-charge distribution range.

In the MIM experiment, the mirror image is produced by scanning across the charged area of the sample with a low-energy electron beam. It is generally accepted that r is much larger than σ. Furthermore, for mathematical simplicity, only the electrostatic potential at the symmetry axis normal to the sample surface is deduced and expressed as follows:

$$V(r) = \frac{KQ}{4\pi\epsilon_0}\frac{1}{r} - \frac{KQ\sigma}{(2\pi)^{3/2}\epsilon_0}\frac{1}{r^2} \tag{2}$$

where $K = 2/(\epsilon_r + 1)$, ϵ_r is the relative permittivity of the sample ($\epsilon_r = 2.6$ for PMMA) and ϵ_0 is the vacuum permittivity.

In equation (2), the first term (proportional to $1/r$) corresponds to the potential of a point charge of magnitude Q (the net total charge of the distribution) at the origin, which

is the well known monopole potential. The second term (proportional to $1/r^2$) is the dipole potential and the higher-order terms are neglected.

$[V(r), r]$ is obtained by the MIM and Q is determined by the time-resolved current method, so σ can be calculated by rearranging equation (2):

$$\sigma = \left(\frac{\pi}{2}\right)^{1/2} \left(1 - \frac{4\pi\epsilon_0 V(r) r}{KQ}\right) r. \tag{3}$$

3. Experimental results and discussion

The statics of space charge has been studied in detail before [25, 28]. Here, we focus our attention on space-charge dynamics, i.e. the evolution of the space-charge distribution range with the irradiation time.

The space-charge distribution range (σ) for various beam energies and a fixed beam current 1.0×10^{-11} Å as a function of time is depicted in figure 1. It is seen that σ increases with increasing beam energy, in good agreement with other authors' reports [20, 24]. More interestingly, one can find that, for each beam energy, σ increases with time within the first $\boldsymbol{u}$ 20 s and eventually attains a saturated value. This means that, with the onset of irradiation, the space charge moves rapidly outwards. Thereafter, the motion of the space charge slows down progressively and eventually the space-charge distribution remains approximately stationary. The origin of the observed space-charge dynamics can be explained as follows.

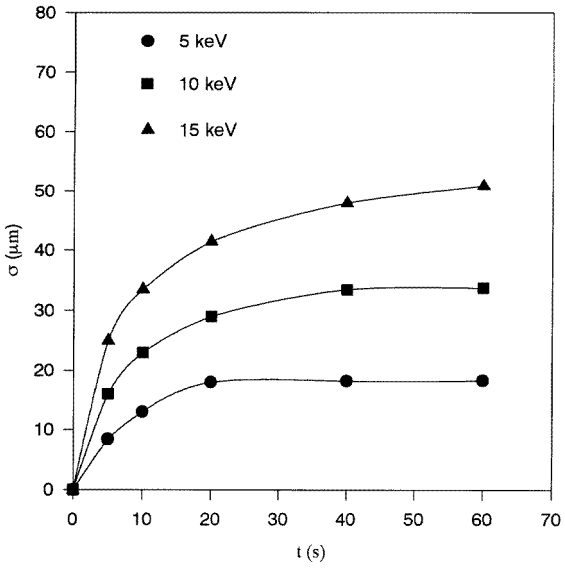

Figure 1. The space-charge distribution range σ as a function of irradiation time t for various beam energies with beam current 1.0×10^{-11} A.

When the PMMA sample is irradiated with an electron beam, some of the electrons are backscattered or lead to secondary electron emission, while the rest lose their excess energy and condense into electronic states in the material to form space charge [29]. Meanwhile, a radiation-induced conductivity is caused, whose value can be several orders of magnitude greater than that of the original conductivity [30, 31]. As irradiation proceeds, the energy

coming from the incident electrons leads to a temperature increase in the irradiated region, resulting in rapid detrapping of electrons in the shallow states, and as the electric field builds up in the polymer by the space charge, these electrons are driven more deeply into the material before becoming retrapped. This space-charge transport involves mainly two mechanisms: (1) the radiation-induced conductive process, due to the additional conductivity g_i induced by irradiation, which leads to a conductance current $g_i E$, where E is the electric field arising from the space charge, and (2) the original conductive process, due to the rest conductivity which the sample has even without irradiation [32, 33], which leads to a current $\mu\rho_f E$, where ρ_f and μ are the density and the mobility of free electrons, respectively. Other processes involved include hopping conduction, i.e. transfer of electrons between densely spaced localized states, field-assisted thermionic emission (Poole–Frenkel effect) and tunnelling of electrons into the conduction band under the influence of high electric stress [34, 35]. The transportation of space charge is controlled by a field-independent trapping time, followed by the capture of electrons in deep trap states [36], in which release of charge is difficult. Therefore, the space charge initially moves rapidly outward. As electrons are trapped in deep states, the speed of space charge movement slows down and σ reaches a fixed value asymptotically (see figure 1).

4. Conclusion

We have established an experimental procedure to study the space-charge dynamics in irradiated polymers using the time-resolved current method in conjunction with the mirror image method. A Gaussian space-charge distribution is assumed and its distribution range σ (the standard deviation) has been obtained. Experimental results reveal that σ initially increases with irradiation time and eventually reaches a symptotic value. The space-charge dynamics is caused by the electric field arising from the space charge, the radiation-induced conductivity and the non-zero electron mobility in the polymer.

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