A Contribution to the Problem of the Sensitivity of the Space Charge on the Electrical Ageing of Irradiated Polymer Dielectrics

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Abstract – Processes of the electrical ageing by the dielectrics are usually connected with the accumulation of space charge during electrical loading and with the structural breaking in substance at molecular and over molecular level. The subject of this article is a research into the influence of the space charge to the electrical ageing and the service life of two polymer dielectrics which are widely used in construction of electro physical installations as isolation and construction materials: low-density polyethylene (LDPE) and polymethylmethacrylate (PMMA).

Practical interest is in a research of conducting polymer isolation under action of ionizing radiations because they make destructive influence to the polymer and change the terms for space charge accumulation in it as well. The analyzed materials were studied in a frequent influence regime of high voltage impulses of microseconds range by the following frequency till 1 kHz.

The space charge accumulation was studied by the methods of acoustic and proton sounding. As a result it could be established that in the researched diapason of duration and following frequency of high voltage pulses there is the space charge accumulation in both researched polymers neither before nor after radiation. Consequently electrical ageing of these dielectrics in electrical loading used regime is conditioned by the structural breaking processes.

1. Introduction

Interrelation and interconnection of ageing factors influence make difficult isolation ageing research. The task of this work was to research regularity in ageing by two polymers of different structure and quite different in ionic radiation receptivity – low-density polyethylene (LDPE) and polymethylmethacrylate (PMMA). by the influence of high voltage pulses of microsecond range after destructive influence of ionizing radiation.

2. Methods of Experiment

Proton irradiation of test specimens was made on the cyclotron U-120 in the Scientific Research Institute of Nuclear Physics of Tomsk Polytechnic University. Proton energy was $T_p = 10$ MeV, thickness of current beam was $j = 2.5 \cdot 10^{-8}$ A/cm^2.

Electron irradiation of test specimens was made on the installation ELU-4 in the Scientific Research Institute of Introscopy of Tomsk Polytechnic University. Electron energy was $T_e = 4$ MeV, current density was $j = 1$ μA/cm^2.

Space charge accumulation in irradiated dielectrics, which were exposed to aging, was studied by the methods of acoustic and proton sounding.

The method of acoustic sounding is based on stimulation of electrical current in electrified dielectric by the passing of acoustic pressure pulse through it. The physical grounds of the method are described in the works [1, 2]. The acoustic impulse is a plane wave with a free form of signal P, this impulse is directed into the plane-parallel test specimen through one of its surfaces and is moving to the opposite surface along the axis $\boldsymbol{x}$ with the longitudinal sound speed v. On the measure electrode with the area S is registered the potential $U(t)$, which is proportional to the integral of the whole current in the test specimen $j(t)$:

$$U(t) = -\int_0^t j(t')dt' \qquad (1)$$

by $h/v \ll RC$, where R and C are incoming resistance and capacity of registering apparatus. In the general case dielectric strain in dielectric is

$$D(x) = \varepsilon_0 \varepsilon\, E(x) + P_S(x), \qquad (2)$$

where $P_S(x)$ is a slow part of relaxation polarization which is not found by measuring of relative permittivity ε by alternating voltage. Then is the registered potential introduced as

$$U(t) = \frac{\chi v S \alpha}{Ch} \int_{t'}^{t} P(t-\tau)D(\tau)d\tau, \quad \frac{h}{v} << RC, \qquad (3)$$

where χ is a compressibility;

$$\tau = x/v;$$

$$t' = -\chi \int P(t-\tau)d\tau;$$

$$\alpha = \alpha_E + \alpha_S;$$

$\alpha_E = \alpha_{qn} + \alpha_\varepsilon$ is a coefficient showing the change of free charge (not concerned with polarization) space density and concentration of doublets (α_{qn}) and change of dipole moments of doublets (α_ε); α_S is a coefficient showing piezoeffect due to P_S .

In a particular case, if $\alpha_E >> \alpha_S$ (as by sounding of LDPE), the method could be used for determination of electric field intensity in the dielectric:

$$U(t) = \frac{\varepsilon_0 \varepsilon \chi v S \alpha_E}{h \cdot C} \int_{t'}^{t} P(t-\tau) \cdot E(\tau) d\tau. \qquad (4)$$

Numerical parameter of integral equation and pressure profile in the test specimen could be found by its sounding in electric field from outside source. In this case for the dielectric with the voltage V from (4) we have:

$$U(t) = \frac{\varepsilon_0 \varepsilon \chi v S \alpha_E V}{h^2 \cdot C} \int_{t'}^{t} P(t-\tau) d\tau =$$

$$= K_E \int_{t'}^{t} P(t-\tau) d\tau. \qquad (5)$$

Differentiation (5) with t gives

$$\frac{dU}{dt} = K_E \cdot P(t-t') \approx K_E \cdot P(t). \qquad (6)$$

It means that the measure signal derivative is proportional to the pressure onto the surface of the test specimen, through which the acoustic impulse is sending.

By combined solution of both equations (4) and (5) it is possible to find E. If the function $P(t-\tau)$ is known, so it is possible to find K, and, as following, α_E.

For research of charge accumulation in polymers while electrical ageing there was projected and build an especial plant in the Scientific Research Institute of High Voltages. This plant helps to sound the electrical field in test specimens in 0.1 s after taking off high voltage [2].

The plant ensures measure and registration of electrical fields with the voltage by 10^2 V/cm with the solving possibility not worth than 30 μm.

The method of sounding of electrical field in solid dielectrics by the accelerated ions was developed in the Scientific Research Institute of High Voltages. This method is based on the influence of the dielectric electrical field to the currents flowing in its space by the irradiation of charged parts stream with the controllable track [3].

A plane-parallel h thick test specimen with the electrodes on its surface is irradiated by the ions with the current density j and with an estimated projected track r in material, $r < h$. The electrode is earthed on the surface turned to the source of irradiation; from the opposite electrode the current of the density j is registered.

The relative tracks dispersion is decreasing with increasing of r and by the value of r in some microns is less than 3%. The capacity of ionization energy loss of ions is falling down very rapidly in the near of r. The dielectric could be examined as a two-parts one consisting of irradiating part with the thickness r and of not irradiated part with the thickness $h - r$.

The current density in irradiated dielectric is found from the equation

$$j(x,t) = j_n(x) + \gamma(x,t) \cdot E(x,t) + \varepsilon_0 \varepsilon \frac{dE(x,t)}{dt}, \qquad (7)$$

where x is a coordinate on the perpendicular axis to the surface of the test specimen; γ – a specific space electrical conducting of material; E – an electrical field intensity. The value γ in the not irradiated part of test specimen ($r < x < h$) is reasonable less than in irradiated and as following in the area $r < x < h$ where is the main absorption current. After integrating of the equation (7) from r till h we get

$$j(t) = \frac{\varepsilon_0 \varepsilon}{h-r} \cdot \frac{d\varphi(x,t)}{dt}, \qquad (8)$$

where φ – an electrical field potential on the ion track depth.

During the irradiation process the value j is decreasing in a close to exponential dependence and in a quasi-stationary state by $t = t_k$, if potential change in the test specimen practically goes to the end, $j << j_p$.

If the test specimen was preliminary charged and at the moment of irradiation beginning ($t = 0$) the potential on the ion track depth is $\varphi(r, 0)$, so at the irradiation moment t_k it is

$$\varphi(r,t_k) - \varphi(r,0) = \frac{h-r}{\varepsilon_0 \varepsilon} \cdot \int_0^{t_k} j'(t) dt =$$

$$= -\frac{h-r}{\varepsilon_0 \varepsilon} \cdot \sigma'(t_k), \qquad (9)$$

where σ' – a surface charge density on the measure electrode; j' – a current density from the electrode of the charged test specimen. From (9) follows

$$\varphi(r,0) = \frac{h-r}{\varepsilon_0 \varepsilon} \cdot (\sigma' - \sigma), \qquad (10)$$

where σ is in accordance with the case when $\varphi(r,0) = 0$ from the beginning. Accordingly by the irradiation of the test specimens with the discrete increasing track $r_1, r_2, ..., r_i$ we have

$$\varphi(r_i,0) = \frac{h-r_i}{\varepsilon_0 \varepsilon} \cdot (\sigma_i' - \sigma_i), \qquad (11)$$

where σ_i and σ_i' – meanings of the charge surface thickness inducted on the electrodes of not charged and preliminary electrified test specimens by some methods. These test specimens are irradiated by the ions with the track r_i. If the thickness of the charged and not charged test specimens are different and come to h and h_1, so

$$\sigma_i = \sigma_i^1 \frac{h_1 - r}{h - r},$$

where σ_i^1 – a meaning of the charge surface thickness inducted on the electrodes of charged test specimen with the thickness h_1. So, to increase discrete the ion track in the test specimen r and to measure charges σ' и σ inducted on electrodes of electrified and not electrified test specimens for every track meaning we could find the distribution of potential through the thickness of test specimen from (11). Further it is easy to count the field intensity distribution

$$E(r_i) = -\frac{d\varphi(r_i)}{dr_i} \tag{12}$$

and the space charge distribution

$$q(r_i) = -\varepsilon_0 \varepsilon \frac{dE(r_i)}{dr_i}. \tag{13}$$

For measuring of potential space distribution with participation of the author there was developed and produced an experimental plant which helps to discrete increase the energy of ions falling to the test specimens (it means to increase their track) and to register automatically an inducted charge for every track on the measuring electrodes of charged and not charged test specimens (σ, σ′, и σ′ – σ).

The researches of electrical field intensity distribution and the space density of the charge were made with the help of ion cycle accelerator U-120 of the Scientific Research Institute of Nuclear Physics at Tomsk Polytechnic University. By measures the test specimens were influenced by protons with the maximal energy till 10 MeV and the beam current density $j_n = 5 \cdot 10^{-9}$ A/cm^2.

The change of proton energy falling to the test specimen was made with the help of radiation absorbers as some layers of aluminum foil with the thickness 15 μm. The thickness of maximum absorber was 510 μm and was reduced for the next absorbers with the step 15 μm and 7 μm. The resolving capacity was about 30 and 15 μm accordingly.

For the charge distribution measure there were taken pairs of test specimens in identical thickness. One of test specimens from every pair was kept in the electric field; another one was left not electrified. Than the electrodes from aluminum foil with the thickness 7 μm were kept to both test specimens, the test specimens were kept to the collimator and put to the measure cell. The irradiation of test specimens was first made through the absorber with the maximum thickness. The test specimens were irradiated till coming of stationary station in measure of charges inducted on the measure electrodes. Than in front of the test specimens an absorber of less thickness was installed and the charges inducted on the electrodes were measured again.

So for every pair of test specimens we have got the value of charges inducted on electrodes by different meaning of proton track in the test specimens.

The high voltage tests were made after the method described in [4].

3. Results and Discussing

According to the put research tasks the thickness of LDPE test specimens was chosen to exclude their charging by the irradiation. As far as the main part of the charge injected by the irradiation is close to the thermalized protons, whose track in LDPE by the parts energy 10 MeV is 1.1 mm, so the test specimens were made with the thickness between electrodes not more than 0.5 mm. As following the protons could not bring the charge into the test specimens.

By γ-irradiation of LDPE the capacity of exposition dose was 1.5 MR/hour. Such capacity is not enough to make a charge as Compton or photo effect because the space charge was not found even in some minutes after the irradiation.

Irradiated test specimens were influenced by the high voltage unipolar impulses.

After influence of high voltage impulse the test specimens were influenced by acoustic and proton sounding.

After influence of high voltage impulses to the test specimens of LDPE there was not found charge accumulation.

The space charge accumulation absence in the test specimens by the influence of the electric field impulses in microsecond duration we could explain with the temporal dependence in moving of electrons and holes.

The electron drift mobility is comparative slowly decreasing with the time and during 10^{-4} s has been keeping up 10^{-6} cm^2/V·s. After finishing the influence of voltage impulse in microsecond duration the electron has been keeping high mobility for a comparatively long time and is moving to the electrode in the space charge electrical field E_q.

Our notes show that by the duration of charge impulse in ones and tens of microseconds and duration of the pause between the impulses in milliseconds and upper the electron injected into the test specimen by the charge impulse influence will be extracted during the pause between the impulses as result of big drift mobility and not very big primary charge penetration depth.

At the same way the holes work. In [5] it is shown that the mobility of electrons and holes is practically identical.

The control experiments show that by influence of millisecond duration impulses to LDPE, if the impulse duration is comparable with the pause duration and the penetration deep of electrons and holes to the dielectric is considerable, so the space charge is accumulated very effective as by influence of direct voltage.

The PMMA irradiation was made by protons, electrons and γ-quanta. The irradiation method and ion radiation sources are identical to those for LDPE. The special steps were made to prevent charge bringing to the polymer by the charged parts. So for proton irradiation there were used test specimens with the electrode system plane-to-plane and with the between electrode distance not more than 0.6 mm. This value is not more than 66% of proton track with the energy 10 MeV in PMMA. Under such conditions the irradiation is made "for shouting", the proton thermalization in the test specimen thickness does not happen. As far as the space charge is situated close to the thermalized protons so the charge is not brought into the test specimens by the protons under these conditions.

The electrons with the energy 4 МэВ by irradiation of PMMA test specimens with electrons there was a bigger track reserve. For irradiating we used the test specimens with the thickness 3 mm and the distance between electrodes was till 0.7 mm. The electron track with the energy 4 MeV in PMMA is upper than 40 mm. That is why the electrons do not bring the charge into test specimens too.

The space charge forming in PMMA as Compton or photo effect by γ-quant irradiation of polymers could not happen because of not sufficient capacity of γ-irradiation exposure dose.

The test specimen sounding after irradiation by acoustical and proton methods confirmed the space charge accumulation absence.

The influence of high voltage impulses in microsecond duration to PMMA does not bring to the dielectric charging what could be confirmed by the results of acoustical and proton sounding. The charge accumulation absence in the impulse electrical field with the impulse duration diapason 10^{-6}–10^{-5} s in PMMA could be explained by the low effective mobility of charge carrier in this polymer. The control experiments show that the space charge does not accumulate in this material even by the influence of direct electrical field.

4. Conclusions

The space charge in the studied polymers does not influence on the electrical ageing in the impulse electrical field. The preliminary influence of different ion irradiation to these polymers does not change the whole situation and conditions for space charge accumulation.

References

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