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ФИЗИКО-МАТЕМАТИЧЕСКИЕ НАУКИ

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POLARIZATION PROFILES IN CORONA POLED P(VDF-TFE) COPOLYMER

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Introduction. Ferroelectric polymers have attracted attention recently as possible candidates for partial replacement of ceramics in manufacturing piezoelectric and pyroelectric sensors and actuators due to good combination of mechanical and electrical properties. While polyvinylidene fluoride (PVDF) and its copolymer with trifluoroethylene P(VDF-TrFE) was extensively studied, much less is known about copolymer with tetrafluoroethylene P(VDF-TFE), although it also belongs to a class of ferroelectric polymers and possesses pyroelectric and piezoelectric properties. Very important properties of the materials for sensors are value of the residual polarization, its thermal and temporal stability, and uniform spatial distribution of polarization. In spite of their obvious importance, these properties have not been given enough consideration so far in P(VDF-TFE). Polarization profiles have not been generally studied, and data reported on other corona poled ferroelectric polymers were quite fragmentary.

Aim. The aim of this work was to study uniformity of polarization in the thickness direction of corona poled P(VDF-TFE) samples. We also tried to investigate the role played by the trapped charges in uniformity of the residual polarization. To accomplish this, polarization profiles in the corona poled films were measured under different conditions by the piezoelectrically induced pressure step (PPS) method [1]. Some experiments were also performed by poling two-layer films, in which only that contacting the positive electrode was metallized.

Materials and metods. P(VDF-TFE) copolymer samples of 20 μm thick film were composed of 95% of VDF and 5% of TFE. The film had been extruded from melt and uniaxially stretched by the supplier (Plastpolymer, Russia) and contained about 90% of ferroelectric *β*-phase crystallites according to IR spectroscopy data. Al electrodes were deposited on one surface of the samples by thermal evaporation in vacuum.

Poling was performed in a corona triode with the bare surface of the sample exposed to the negative corona initiated by a pointed tungsten electrode [2]. Ions and electrons passed through a control grid, kept at a constant negative potential with respect to the grounded rear electrode. The grid was made vibrating to allow simultaneous measurement of the apparent surface potential by Kelvin's method and the poling current. The samples were virtually short circuited for 15 min immediately after completion of poling by changing the corona polarity and grounding the control grid. Duration of the short circuiting was long enough to ensure zero field everywhere in the bulk of the sample.

Results and discussion. It has been found that poling of P(VDF-TFE) film at 25 ^{o}C under constant current density of 80 μA/m^2 for 15 min resulted in non-uniform distribution of polarization with a prominent peak near the positive electrode, while a broad zone near the surface bombarded by corona ions remained non-polarized. Field strength increased with time and was equal to 100 MV/m at the end of poling. One should consider that the value E=50 MV/m is close to the coercive field of VDF-containing ferroelectric polymers.

Thermal stimulation affected the magnitude of the polarization peak. In the samples poled at 85°C at a constant current density of 80 μA/m^2 and cooled to 25^{o}C under applied field of 100 MV/m, the peak was higher and closer to the positive surface than in the room-temperature poled samples. The two-layer samples having the total thickness of 40 μm were poled at 25^{o}C at the constant average field of 100 MV/m. Only the film attached to the positive electrode had high and rather uniform ferroelectric

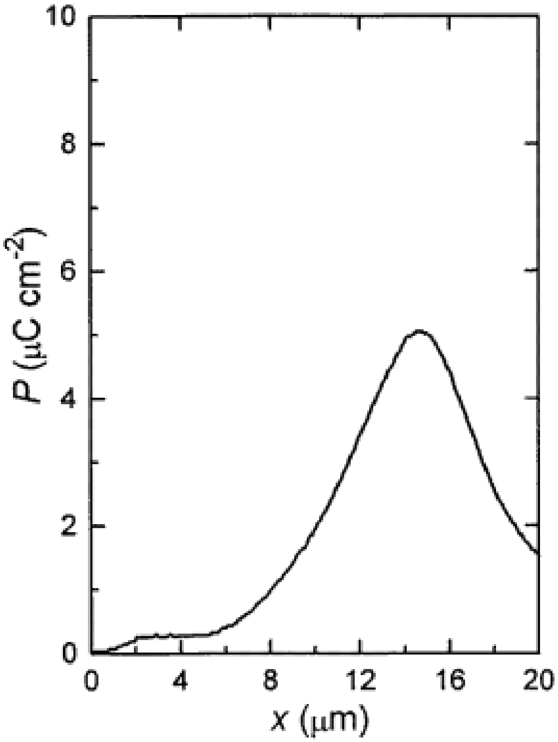

Fig. 1. Distribution of polarization in P(VDF-TFE) film poled at 25 ^{o}C at a constant current density of 80 μA/m^2 for 15 min

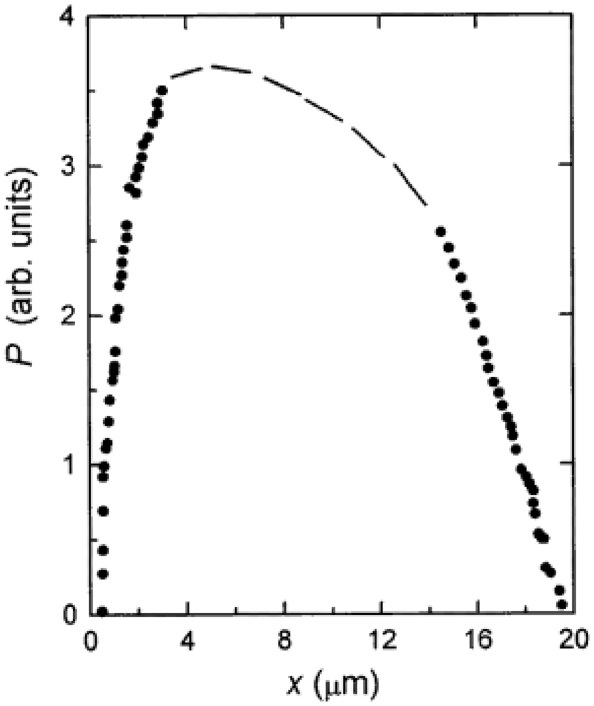

Fig. 2 Distribution of polarization in P(VDF-TFE) film poled at the constant field of 100 MV/m. Measurement by the LIMM method

polarization, while the upper film remained non-polarized indicating that the poling voltage was applied mainly to the lower film. Therefore, to obtain uniformly poled P(VDF-TFE) at a moderate poling field, it would be expedient to cover the sample by another auxiliary film.

Residual polarization in samples poled at high fields of 100 MV/m at room temperature has been uniform. However, even in this case, the polarized part of the volume was separated from the surfaces of the sample by a thin transition zone of about 0.5 μm thickness. This feature was revealed by using the laser intensity modulated method (LIMM) [3].

To explain the obtained results one should consider that the polarization non-uniformity in the thickness direction can be apparently caused by non-uniform distribution of the poling field. According to Poisson's equation, the uniformity of the field $E(x,t)$ is distorted by presence of either real charges $\rho(x,t)$, or the polarization charge $dP(x,t)/dx$. By analyzing experimental results it has been concluded that injection of negative charge carriers from the virtual electrode, formed on the surface bombarded by corona ions, dominated in most cases over separation of the intrinsic carriers. After short-circuiting, i.e. at equilibrium conditions ($E(x)=0$), the space charge $\rho(x)$ can be localized only at the boundaries of polarized zones where the derivative dP/dx is not equal to zero.

It is known that polarization charge appearing during poling creates a depolarizing field which tends to switch the ferroelectric polarization back to its original state after completion of poling. Thus, the residual polarization can be high and stable only if the depolarizing field is compensated or neutralized. We believe that in the case of ferroelectric polymers this compensation is performed by the space charge $\rho(x)$ trapped in transition zones, by which the polarized volume is separated from the non-polarized one. If the values of polarization charge $dP(x)/dx$ and real charge $\rho(x)$ are equal to each other, the depolarizing field is completely compensated, so that $E(x)=0$ everywhere in the sample. We consider the existence of transition zones coupled with the compensating space charge as a general feature of poled P(VDF-TFE) and, most probably, of all other ferroelectric polymers. The presence of space charge in transition zones is a guarantee of the remnant polarization high stability.

The thickness of the non-polarized zone depends on the penetration depth of injected carriers. The zones are particularly protracted in the case of moderate poling fields. On the other hand, if the poling field is high, the near-surface zones are very narrow, as if the negative charge is not injected deeply into the bulk. It takes a definite time for injected charge to penetrate deeply into the bulk. This can be accomplished only if regions it front of the charge are non-polarized as, for example, in case of low or moderate poling fields. If, however, ferroelectric polarization has been already formed near the surface, as in case of high poling fields, the injected charge cannot move easily through the polarized section. It seems that the apparent conductivity of the polarized areas is much lower than that of the non-polarized ones. This is the reason why polarization is uniform in case of high poling fields.

Conclusions. For obtaining high and stable residual polarization, required for application in the sensors, P(VDF-TFE) films should be poled at the field strength at least two times higher than the coercive field.

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