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THE TEMPERATURE EFFECT ON IMPULSE DIELECTRIC STRENGTH OF POLYMER FILMS

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Abstract. In the paper, the experimental data on the temperature effect on the breakdown strength of polypropylene, polycarbonate and poly(ethylene terephthalate) films 2.0-2.5 μm thick have been obtained over a temperature range from 293 to 363 K. When the film samples were heated, the breakdown electric field strength was found to decrease slightly but to reveal a significant scatter of values. It was shown that the experimental results on the pulsed electrical breakdown of the polymer films could be described basing the notion of the ionization mechanism of the polymer breakdown, not related to the development of an impact ionization of the polymer molecules.

Keywords: impulse dielectric strength, temperature, polypropylene, polycarbonate, poly(ethylene terephthalate)

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ВЛИЯНИЕ ТЕМПЕРАТУРЫ НА ИМПУЛЬСНУЮ ЭЛЕКТРИЧЕСКУЮ ПРОЧНОСТЬ ПОЛИМЕРНЫХ ПЛЕНОК

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Аннотация. В работе исследовано влияние температуры на пробивную напряженность пленок полиэтилена, поликарбоната и полиэтилентерефталата толщиной 2.5-2.0 мкм в температурном диапазоне 363-293 К. Установлено, что при нагреве образцов пленок пробивная напряженность электрического поля уменьшается незначительно, но обнаруживает существенный разброс значений. Показано, что экспериментальные данные по импульсному пробою полимерных пленок можно описать на основе представления об ионизационном механизме пробоя полимеров, не связанном с развитием ударной ионизации их молекул.

Ключевые слова: полимерная пленка, импульсный пробой, пробивная напряженность, полиэтилен, поликарбонат, полиэтилентерефталат, температура

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Introduction

Steady interest towards the dielectric strength of polymer dielectrics has persisted for many decades, due to the great scientific and practical implications of this characteristic for studies of the breakdown phenomenon. The vast majority of publications on this issue consider electrical breakdown in polymers exposed to DC and AC voltages. Considerably scarcer data are available on the picture of impulse breakdown of polymers, mainly consisting of results obtained decades ago [1–3]. Technical advances made in recent years allow detecting breakdown characteristics under nanosecond voltage pulses, recording breakdown times and voltages with high accuracy [4–9].

One of the most important factors influencing the evolution of electrical degradation and breakdown in polymer dielectrics is temperature. Its role under the influence of voltage pulses was studied in large samples of poly (methyl methacrylate) (PMMA) [2], polyethylene (PE) and polytetrafluoroethylene (PTFE) [3]. The effect of temperature on the breakdown strength F_{br} turned out to be different for different polymers. F_{br} decreased by more than 25% in PMMA samples under heating from 293 to 433 K, while the value of F_{br} for PE and PTFE remained virtually constant in the same temperature range.

Polymer films are a special group in the diverse range of dielectric materials. They are widely used in electrical engineering, for example, for manufacturing high-voltage capacitors, or for synthesis of films ranging in thickness from several to tens of µm that serve as convenient samples for studies of electrical aging and breakdown in polymer material. However, the effect of temperature on the impulse strength of polymer films is yet to be fully explored.

The goal of this study is to understand the nature of the temperature effect on the characteristics of impulse strength of various polymer films at room and elevated temperatures.

Experimental procedure

Industrial films made of polypropylene (PP) with the thickness of 2.0 μ m, polycarbonate (PC) and polyethylene terephthalate (PET) with the thickness of 2.5 μ m were used. The polymers used to synthesize these films are characterized by varying degrees of polarity, glass transition and softening temperatures as well as different morphology of the material structure.

The films were fixed in a special circular holder and placed between steel electrodes, one disc-shaped (40 mm in diameter) and the other sphere-shaped (6 mm in diameter). The surface of the electrodes was polished to a specular gloss. The space between the spherical electrode and the film was filled with capacitor oil to prevent edge and surface discharges. A high-voltage pulse of negative polarity with an amplitude of 2.5 kV and rise time of about 130 ns was applied once to the given film samples.

The electrode system used provided an electric field close to homogeneous in the region of film breakdown. Since breakdown of films occurred at the leading edge of the pulse, it is preferable to use the value of the breakdown strength F_{br} to evaluate their impulse strength, defining it as

$$F_{br} = U_{br}/d,$$

where U_{br} is the voltage in the sample at the time of breakdown, d is the thickness of the film.

 U_{br} was measured with an ADS-2332 broadband storage oscilloscope and a high-voltage broadband divider with a cutoff frequency of 300 MHz, allowing to directly record the voltage variations in the sample. The time of film breakdown was detected in the waveforms by a sharp voltage drop and occurrence of oscillations. A typical waveform recorded at the time of breakdown at the leading edge of the pulse is shown in Fig. 1.

Electrical tests were carried out in the temperature range of 293–363 K. We should note that the chosen upper temperature limit was lower than the softening temperature of any of the polymers considered to prevent the spherical electrode from penetrating through the film with a consequent decrease in its breakdown voltage.

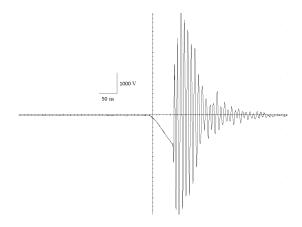


Fig. 1. Typical signal waveform at time instant of breakdown for 2.0 or 2.5 µm thick polymer film at pulse leading edge

Experimental results and discussion

The voltages U_{br} of polymer dielectric materials, including polymer films, are characterized by a significant scatter, therefore, breakdown tests should be repeated multiple times, and their results should be statistically processed. At least 50 samples of each polymer film were tested for breakdown at each temperature; the values of F_{br} found were used to calculate the empirical distribution function f(F) determining the breakdown probability in polymer film when the voltage U (electrostatic field strength F) is reached. The values of the function f(F) were calculated by the formula

$$f(F) = n/N, (1)$$

where N is the number of tests performed,

 $N \ge 50$; n is the number of samples experiencing breakdown when the field strength F is reached. It was established in [10-12] that the Weibull model of failure probability is applicable for statistical analysis of the results obtained for electrical breakdown under both DC and pulse voltages applied to polymer dielectrics. For this reason, a two-parameter Weibull distribution was used to approximate the empirical distribution function determined by relation (1), taking the form

$$f(F) = 1 - \exp[-(F/F_0)^m], \tag{2}$$

where m is the shape parameter, F_0 is the scale parameter. The parameters m and F_0 are easily evaluated using the least squares method with expression (2) linearized.

Fig. 2 shows a typical form of the linearized functions f(F) for polymer films studied for breakdown at different temperatures. The correlation coefficient of the approximating lines turned out to be higher than 0.97 in all cases, confirming the validity of the linear approximation used. The values of the parameters m and F_0 were calculated by the least squares method, and then used to calculate the first four moments μ_k of the distribution function [13]:

$$\mu_k = F_0^k \int_0^\infty x^{k/m} \exp(-x) dx. \tag{3}$$

This is the expected value of the Weibull distribution F_{br} (at k=1), its variance σ (at k=2), skewness μ_3 (at k = 3) and kurtosis μ_4 (at k = 4).

Fig. 3 shows the temperature dependences for the skewness and kurtosis of the function f(F) of the studied polymer films. Evidently, $\mu_x \neq 0$ for all temperatures, and the skewness values calculated at different temperatures are close; therefore, the functions f(F) cannot be considered symmetric. However, the values of skewness μ_3 are small and predominantly positive, which indicates a slight positive skewness of the function f(F).

The kurtosis μ_4 of this function varies from 2.5 to 3.5 in the studied temperature range, and does not depend on temperature. Note that the normal distribution should have zero asymmetry and a kurtosis equal to 3 [13]. The values of μ_3 and μ_4 that we calculated are close to these values, however, the breakdown voltage distributions that we obtained cannot be considered normal.

The temperature dependences of the most probable breakdown strength and the corresponding variances at each temperature are shown in Fig. 4 for all three types of polymer films. The values of F_{br} show a significant scatter for all polymers in the temperature range considered, showing a trend towards decrease with increasing temperature (at least for PP and PC films).

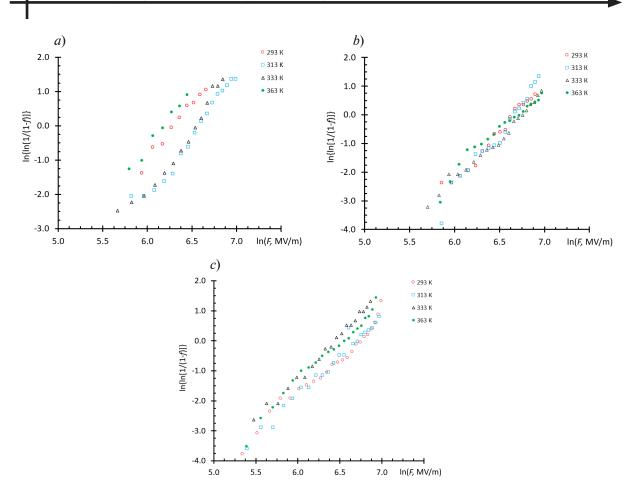


Fig. 2. Linearized Weibull distribution functions for PP (a), PET (b) and PC (c) films at different temperatures in the range of 293–363 K

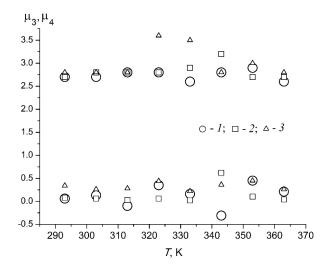


Fig. 3. Temperature dependences for skewness (μ_3 , lower symbols) and kurtosis (μ_4 , upper symbols) of breakdown strength distribution in PET films (symbols *I*), PP (*2*) and PC (*3*)

Student's *t*-test was applied to statistically validate the hypothesis of the decrease in F_{br} in polymer films under heating taking into account a small number of sample elements (determined by the number of temperature points for which the tests were carried out). The sample of F_{br} values for each polymer was divided into two groups with equal number of temperature points, low-temperature and high-temperature ones; next, Student's t-test (with a confidence interval of 90%) was used to compare the corresponding average values calculated for each of these groups [14]. The results obtained by this technique indicate that the hypothesis formulated about the effect of temperature on F_{br} can be accepted only for the PC film within the given confidence interval. The effect of temperature on F_{br} cannot be estimated as significant for PP and PET films.

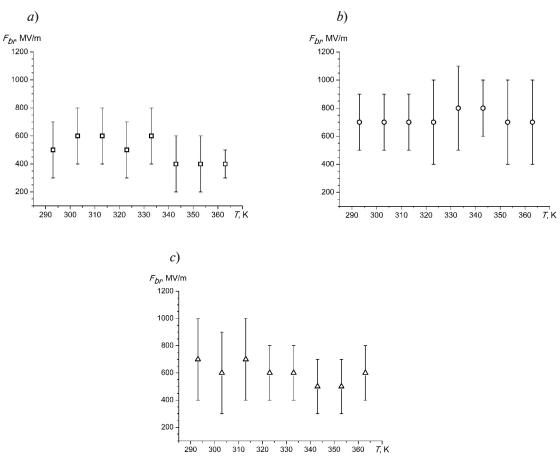


Fig. 4. Temperature dependences of most probable breakdown strength for PP (a), PET (b) and PC (c) films

Notably, neither the PP film nor the PC film changed their phase state during the heating process, since in the first case, the glass transition region is 253–263 K [15] and the PP film was in elastic state at a temperature above room temperature; on the contrary, in the second case, the glass transition temperature $T_g \approx 415$ K and the PC film was in a glassy state, even at the highest temperature of 363 K (used in our study). As for the PET film, its glass transition temperature is $T_g \approx 343$ K, i.e., T_g lies in the temperature range we considered but no noticeable change in F_{br} was observed at this point.

Additional data obtained during electrical tests of polymers in a DC field confirm that a sharp decrease in their breakdown strength occurs in the glass transition temperature region [16, 17]. Thus, it can be assumed that unfreezing of molecular mobility in polymers does not have a noticeable effect on impulse strength of polymer dielectric films, which is likely due to the short-term effect of the electric field on the polymer. Indeed, this time interval does not exceed 100 ns for film breakdown at the leading edge of the pulse.



It was found that the time interval for the final stage of electrical breakdown in polymer films, when fracture of the dielectric material occurs accompanied by the formation of a breakdown channel, is approximately $10^{-9}-10^{-8}$ s; the amplitude of breakdown current density reaches values of about 10^7 A/cm² [17, 18].

Note that the voltage oscillations in the sample, always detected in the waveforms after breakdown, can last for several hundred nanoseconds (see Fig. 1), but their occurrence is due to response of the measuring circuit to a short-term pulse of the breakdown current [18]. Evidently, finding out the reason behind such a rapid increase in current during breakdown remains one of the most important goals, which should allow to gain a better understanding of the picture of impulse breakdown in polymer dielectric films, as no consensus has been reached on this phenomenon.

Two possible physical mechanisms governing electrical breakdown in polymers are collisional ionization generating an electron avalanche [4, 9, 19, 20] and field-induced (tunneling) ionization of macromolecules [6–8]. We should note that the hypothesis of collisional ionization in polymers has faced much criticism in the recent years (see, for example, [6-8]). The cause for criticism is that the mean free path of electrons in polymer dielectrics does not exceed 1–3 nm, while electrons cannot gain the energy of 6–7 eV necessary for ionization of polymer macromolecules in a realistically achievable electric field.

On the other hand, the probability of large nanopores appearing in polymers increases with increasing temperature [21]. It is hypothesized in [22] that collisional ionization occurs precisely in such pores. The theory based on these hypotheticals [22] suggests a sharp dependence of breakdown strength on temperature, especially pronounced at temperatures above the glass transition temperature of the polymer. However, as mentioned above, the value of F_{br} is virtually independent of temperature in the case of impulse breakdown of polymer films.

As discussed in [6-8], the theory behind field-induced (tunneling) ionization of macromolecules can be used to explain the picture of impulse breakdown in polymers. According to this theory, electrons and positively charged molecular ions (holes) appear as a result of field-induced ionization of macromolecules. The explosive increase in their concentration is due to the Debye screening effect, manifesting when a certain critical charge density is reached. Debye screening leads to a decrease in the ionization energy of macromolecules and consequently to self-acceleration of the field-induced ionization process. The theory was expanded in [8], suggesting exposure to voltage pulses accelerates the field-induced ionization of macromolecules at the leading edge of the pulse. The reason for this effect is that electrons injected from the cathode can gain energy sufficient to excite molecules but not to ionize them in an electric field. Excited polymer molecules are ionized at lower field strengths than molecules in the ground state. Therefore, the critical concentration of electrons and holes is achieved in less time, which is the actual reason for breakdown at the leading edge of the pulse. According to the explanation given in [8], a slight decrease in the breakdown strength of polymer films under heating is associated with an increase in the free electron concentration both due to an increase in injected current density and due to an increased probability of thermally stimulated electron emission from traps.

Conclusion

This paper reports on the experimental study of the temperature effect on the breakdown strength of thin polymer dielectric films of polypropylene, polycarbonate and polyethylene terephthalate in the temperature range from 293 to 363 K. We found that the two-parameter Weibull distribution can be used for statistical processing of experimental results. The parameters of this distribution, determined at each temperature for each film, allow to calculate the most probable breakdown strength F_{br} and its variance σ . It was found that the value of F_{br} decreases slightly in polymers heated in the given range but is characterized by significant scatter that does not depend on temperature.

We established that the experimental data on the temperature effect on the impulse strength of polymers can be explained via the ionization mechanism of polymer breakdown, which is not associated with collisional ionization developing in the polymers.

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