Importance of Chemomechanical Processes for the Applications of Biaxially Oriented Polypropylene Films in Power Capacitors I. Phenomenological Model of Interaction

J. POLAKOVIČ

Department of Physical Chemistry, Faculty of Chemical Technology, Slovak University of Technology, SK-812 37 Bratislava
e-mail: polakovi@chtf.stuba.sk

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The chemomechanical processes between the liquid dielectric and the biaxially oriented polypropylene (BOPP) film have been studied by means of measurement of dimension changes of the film by this interaction. The dimension changes are caused by the change of state parameters (physical and chemical conditions) during the interaction. If specific potential Φ_L^s for molecules of liquid dielectric L in BOPP film, *i.e.* in solid phase, is greater than the chemical potential of molecules of liquid dielectric μ_L, *i.e.* in liquid phase, the molecules of liquid dielectric do not penetrate into the solid phase (BOPP film) and no dimension change is observed. Thus a high-quality power capacitor is produced. If this condition is not obeyed, the dimension changes may cause formation of vacancies, which consequently deteriorate the quality of power capacitor. On this basis a phenomenological model describing the interaction has been devised as a result of 400 measurements of dimension changes. The application of this model is demonstrated.

Because of their excellent mechanical strength, moisture-barrier property, and electric properties, biaxially oriented polypropylene (BOPP) films are widely used as a solid dielectric in power capacitors, particularly in "all film power capacitors". In order to reach high quality of the power capacitors the space between polypropylene films must be completely filled with the liquid dielectric where the dimensions of the films must not change. In an opposite case, vacancies (the small spaces between the polypropylene films, which are not filled with the liquid dielectric) arise. The vacancies deteriorate the quality of power capacitors because of their low dielectric strength (in comparison with the polypropylene film) and their influence on the electric field in the capacitor (they cause strong nonhomogeneity in electric field). It is generally supposed that to fill completely the space between polypropylene films is a satisfactory condition for reaching the high quality of power capacitors. The changes of the film dimensions are not taken into account [1]. The attention has been paid mainly to rough-surface biaxially oriented polypropylene films [2—8].

The BOPP film dimension changes deteriorate the quality of power capacitors because of their influence on the electric field in the capacitor. Also, in the case when the space between BOPP films is filled, the electric field is not homogeneous. *Blok* and *LeGrand* [9] have defined the compressive force, $\boldsymbol{f}$, which causes electromechanical thinning of the film

$$\boldsymbol{f} = (1/2)\varepsilon_r\varepsilon_0(V/d)^2 A \qquad (1)$$

where A is stressed sample area, ε_r the relative permittivity of the dielectric, ε_0 the permittivity of free space, V the applied voltage, and d the sample thickness.

In various parts of the capacitor design various forces $\boldsymbol{f}$ occur. Local regions subjected to higher electric fields thus experience a mechanical shear stress tending to form an indentation. As the electric field is raised, a weak spot in the film deforms slightly. The sharp depression thus formed concentrates the field at this point so leading to further increase in the stress. The stress is no longer normal to dielectric, so the material at the centre of the depression will flow outward and forms a "ridge" around the depression. When the field is raised higher, the dielectric strength (either intrinsic or electromechanical) can reach (at the centre of the depression) the value where the breakdown occurs [1]. This mechanism takes place also in the case when the liquid dielectric is not present in the space between BOPP films.

In the case of interaction between BOPP film and liquid dielectric, the dimension changes may be caused by: penetration (diffusion) of molecules of liquid L into the biaxially oriented polypropylene film F; dissolution of a part of BOPP film F in liquid L; various pro-

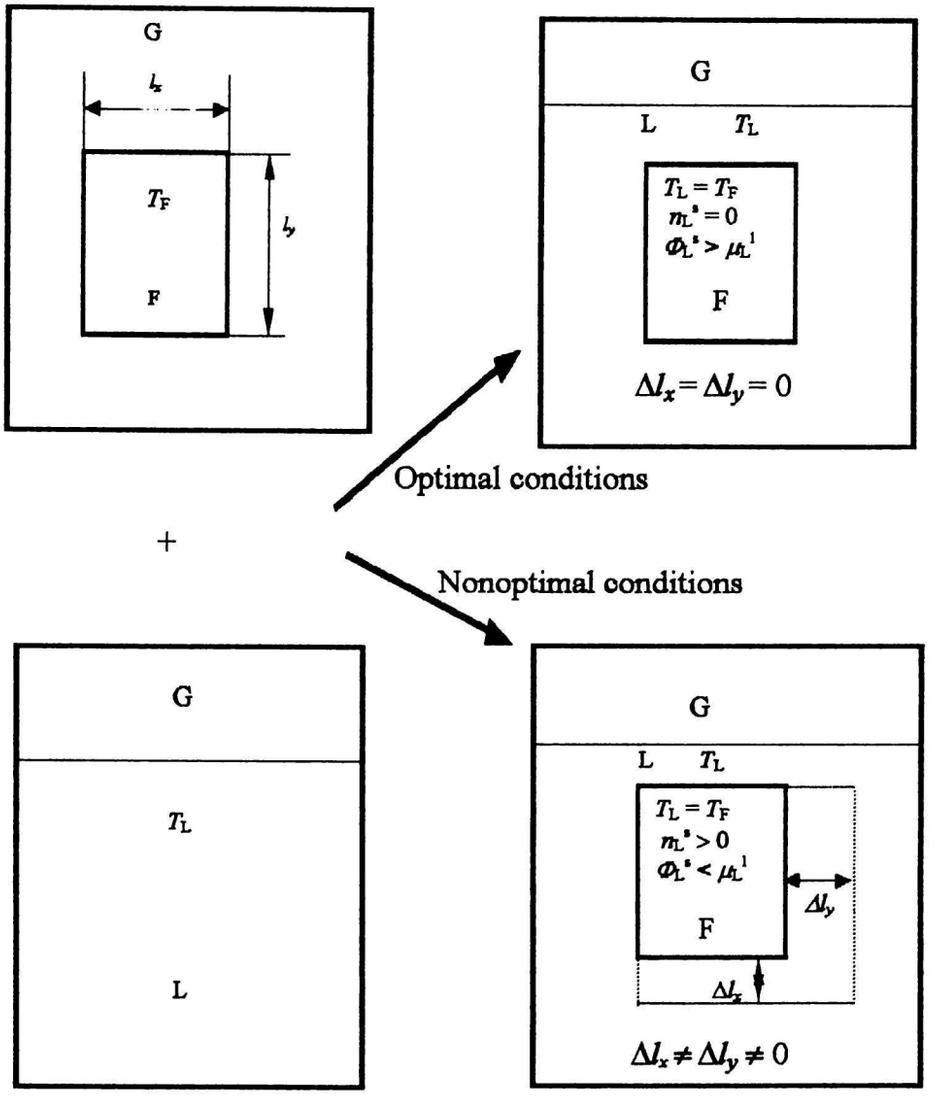

Fig. 1. The optimal and nonoptimal conditions for the interaction process.

cesses taking part on the surface of BOPP film (they are very important because of the thickness (10^{-5} m) of the film); relaxation processes.

All these processes are designated as chemomechanical processes. Thus, for the power capacitor production, the optimum process is the one where the dimensions of the BOPP film do not change and the space between BOPP films is filled with liquid. Information about the chemomechanical processes enables us to find the optimum conditions for the power capacitors production.

THEORETICAL

The chemomechanical processes are caused by the change of the state parameters (physical and chemical conditions) of macroscopic system (BOPP film) [10]. In the studied case the change begins when film F is contacted with the liquid dielectric L (l – liquid) (Fig. 1). *Erdos* [11, 12], and *Šiškova* and *Erdos* [13, 14] defined the specific potential Φ_L^s for liquid molecules L in BOPP film F (s – solid). When the relationship

$$\Phi_L^s > \mu_L^l \tag{2}$$

is valid, where μ_L^l is chemical potential of molecules L in liquid phase (l), the molecules of liquid L do not penetrate into the film F (Fig. 1, optimal conditions) and no dimension changes are observed. When the relationship

$$\Phi_L^s < \mu_L^l \tag{3}$$

is valid, the molecules of liquid L penetrate into film F (Fig. 1, nonoptimal conditions), which causes the dimension changes of the BOPP film and the elastic

properties of the film are changed, too.

The flux $\dot{n}_L^s$ is defined by the equation

$$\dot{n}_L^s = (dn_L^s)/(dt) \tag{4}$$

where n_L^s is the amount of liquid molecules L in the film F at time t.

Up to now, little attention has been paid to the study of the interaction process between the liquid dielectric and the BOPP film, because the works have been concerned mainly with thermoelastic properties of the film in the equilibrium with liquid [15—17].

The flux $\dot{n}_L^s > 0$ causes the dimension changes of the BOPP film. These dimensions are denoted as l_x, l_y, and l_z (x is the direction in which the BOPP film is wound on to a reel, z is thickness of the film, and y is perpendicular to x and z). When the relaxation processes do not take place (no change of the film orientation (texture) occurs by interaction) these equations are valid

$$\begin{aligned} l_x &= f_x(a_1, a_2, \ldots a_n, \Phi_L^s, n_L^s) \\ l_y &= f_y(a_1, a_2, \ldots a_n, \Phi_L^s, n_L^s) \\ l_z &= f_z(a_1, a_2, \ldots a_n, \Phi_L^s, n_L^s) \end{aligned} \tag{5}$$

where a_1, $a_2, \ldots a_n$ are symbols for physical and Φ_L^s for chemical conditions (state parameters) of the interaction process.

Because the equation

$$n_L^s = f(t) \tag{6}$$

is valid (t is time), eqns (5) may be written in the form

$$\begin{aligned} l_x &= f_x(a_1, a_2, \ldots a_n, \Phi_L^s, t) \\ l_y &= f_y(a_1, a_2, \ldots a_n, \Phi_L^s, t) \\ l_z &= f_z(a_1, a_2, \ldots a_n, \Phi_L^s, t) \end{aligned} \tag{7}$$

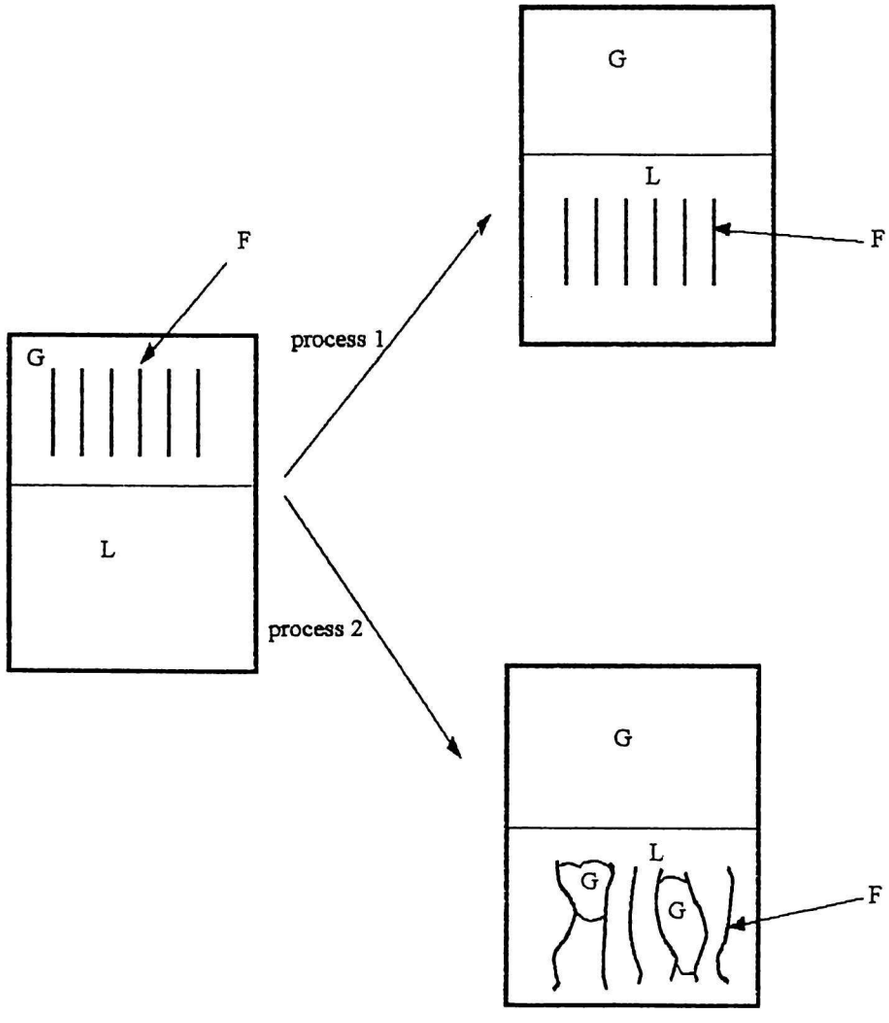

Fig. 2. The scheme for interaction between BOPP films F, liquid dielectric L, and gas G. Process 1 is optimal, and process 2 is nonoptimal.

Eqns (7) show that the dimension of the BOPP film may not be uniform (the same). This nonuniformity is responsible for the changes of the dimensions of BOPP films as they are shown in Fig. 1.

The purpose of the study of interaction between the liquid dielectric L and the BOPP film F is to choose such physical and chemical conditions (state parameters) which prevent the dimension changes and the vacancies formation. In such a case the properties (quality) of the power capacitors are determined only by the dielectric strength of the polypropylene film which is very high [18]. Such process is shown in Fig. 2 as the process 1. The situation, when the dimensions of the BOPP films are changed due to the interaction between BOPP films and liquid dielectric is shown in Fig. 2 as the process 2.

This phenomenological model (eqns (7)) has been devised as a result of 400 measurements.

EXPERIMENTAL

To demonstrate the phenomenological model these conditions were selected: biaxially oriented polypropylene film "Tatraphan 15", product of Chemosvit, Svit, Slovak Republic, and liquid dielectrics: No. 1 – dibutyl phthalate, product of Lachema, Brno, Czech Republic, No. 2 – silicone oil "Lukoil oil MF", product of Syntesia, Kolín, Czech Republic, were used. The changes of the film dimensions (by interaction) were measured by means of a special equipment [19] based on transformation of Δl_x or Δl_y to electric signal (potential difference) by means of differential transformer. Due to special construction, this equipment enables us to measure the length changes (Δl_x or Δl_y) with high precision (0.02 mm).

RESULTS AND DISCUSSION

The influence of these physical conditions was studied:

$a_1 = T_F$, temperature of BOPP film before interaction

$a_2 = T_L$, temperature of liquid dielectric before interaction

$a_3 = \boldsymbol{F}$, external mechanical force which acts on the film in course of the interaction

Fig. 3 shows the Δl_x time function ($\Delta l_x = f(t)$) for the same films in the different liquids: No. 1 and No. 2 at the same physical conditions T_F, T_L, $\boldsymbol{F}$ but at different potentials $(\Phi_L^s)_1$ and $(\Phi_L^s)_2$.

The Δl_xtime function is defined (as the difference between l_x at time t and l_x, when $t = 0$) by the equation

$$\Delta l_x(t) = l_x(T_L, T_F, F, \Phi_L^s, t) - \\ -l_x(T_L, T_F, F, \Phi_L^s, t = 0) \qquad (8)$$

The Δl_y time function is defined equally. As Fig. 3 shows, the liquid dielectric No. 1 causes greater Δl_x than the dielectric No. 2 does. This may be expressed

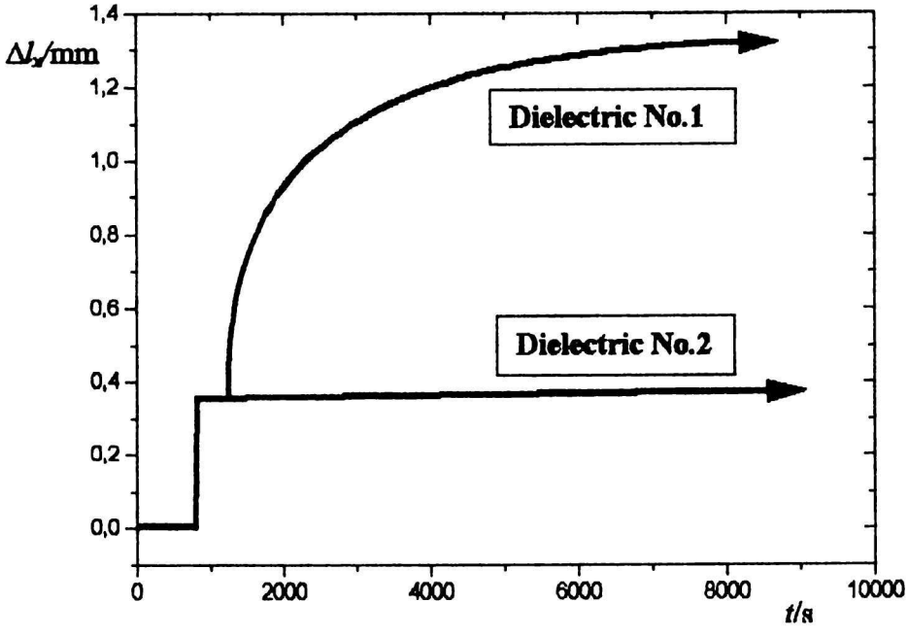

Fig. 3. The Δl_x time function for the same films in the different liquids, dibutyl phthalate (No. 1) and silicone oil (No. 2) at the same physical conditions: T_L – temperature of the liquid dielectric, T_F – temperature of the BOPP film, $\boldsymbol{F}$ – external mechanical force which acts on the film in course of interaction and different potentials $(\Phi_L^s)_1$ (liquid dielectric No. 1) and $(\Phi_L^s)_2$ (liquid dielectric No. 2).

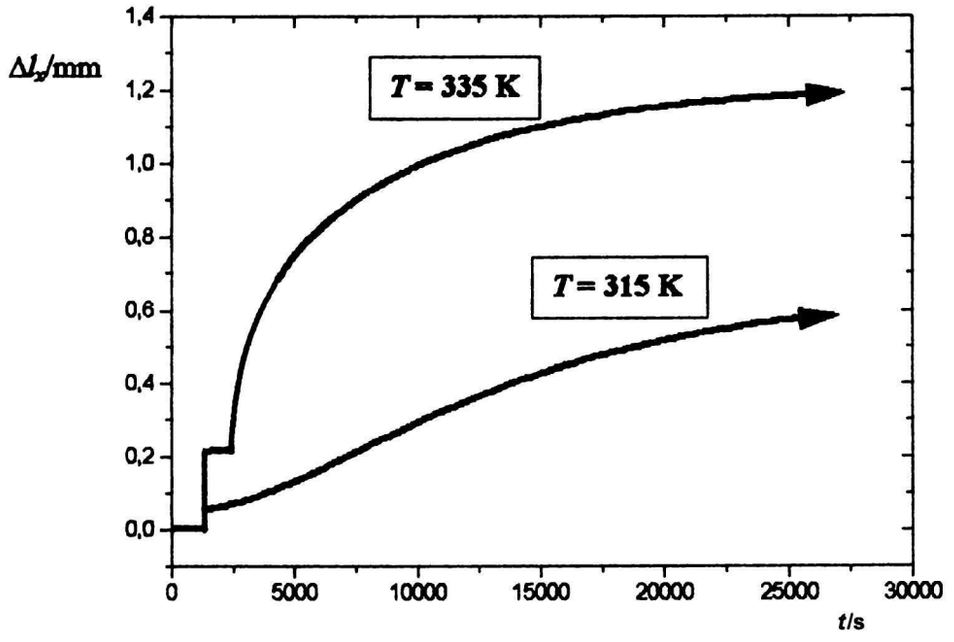

Fig. 4. The Δl_x time functions at the different temperatures of liquid dielectric T_L for the same value of: T_L – temperature of the film, $\boldsymbol{F}$ – external mechanical force which acts on the film in course of interaction and the same liquid dielectric (No. 1).

in the form

$$(\Delta l_x)_1(t \to \infty) > (\Delta l_x)_2(t \to \infty) \qquad (9)$$

where the symbol "∞" indicates that time approaches infinity. The relationship (*9*) may be interpreted in this way: the molecules of the liquid dielectric No. 2 penetrate into the same film in a smaller extent than the molecules of the liquid dielectric No. 1 do, because of the differences in molecular structure of liquid dielectrics No. 1 and No. 2. The differences in structure bring about the differences in Φ_L^s and, consequently, in Δl_x. For the production of high-quality power capacitors, the liquid dielectric No. 2 is more appropriate for the given film than the liquid dielectric No. 1.

Fig. 4 shows Δl_x time functions at different temperatures T_L for the same values of T_F, $\boldsymbol{F}$, and Φ_L^s. The results show that the liquid temperature T_L is the dominant physical parameter which influences both the kinetics and extent of interaction. The temperatures 335 K and 315 K correlate with production of power capacitors.

Fig. 5 shows both Δl_x and Δl_y time functions for different directions "x" and "y" in the film and for the same liquid, film, T_L, T_F, $\boldsymbol{F}$, and Φ_L^s. The results may be written in the form

$$\Delta l_x(t \to \infty) > \Delta l_y(t \to \infty) \qquad (10)$$

and may be interpreted in this way: the interaction between the liquid dielectric and BOPP film changes the film dimensions l_x and l_y to different extents $\Delta l_x(t \to \infty)$ and $\Delta l_y(t \to \infty)$ because the structure of BOPP film depends on the direction in the film. For the power capacitor properties, this fact is important because the probability of dimension changes and vacancies formation increases when the ratio R

$$R = \{\Delta l_x(t \to \infty)\}/\{\Delta l_y(t \to \infty)\} \qquad (11)$$

increases.

CONCLUSION

The phenomenological model of interaction, given by eqns (*7*), can be used for the choice of such liquid dielectric and BOPP film and such physical conditions (state parameters) that prevent the dimension changes and vacancies formation. According to *Tomago et al.* [20] these conditions should be formulated in agreement with the relation (*2*). In this case, both the changes of BOPP film dimensions and the probability of the vacancies formation (Fig. 1, optimal conditions, and Fig. 3, dielectric No. 2) are minimal.

If the interaction between the film and given liquid dielectric does not obey the relationship (*2*), the relationship (*3*) is valid (Fig. 1, nonoptimal process, and Fig. 3, liquid No. 1). In this case such physical conditions may be chosen (T_L, T_F, and $\boldsymbol{F}$) which enable the liquid dielectric to fill the space between BOPP films without vacancies.

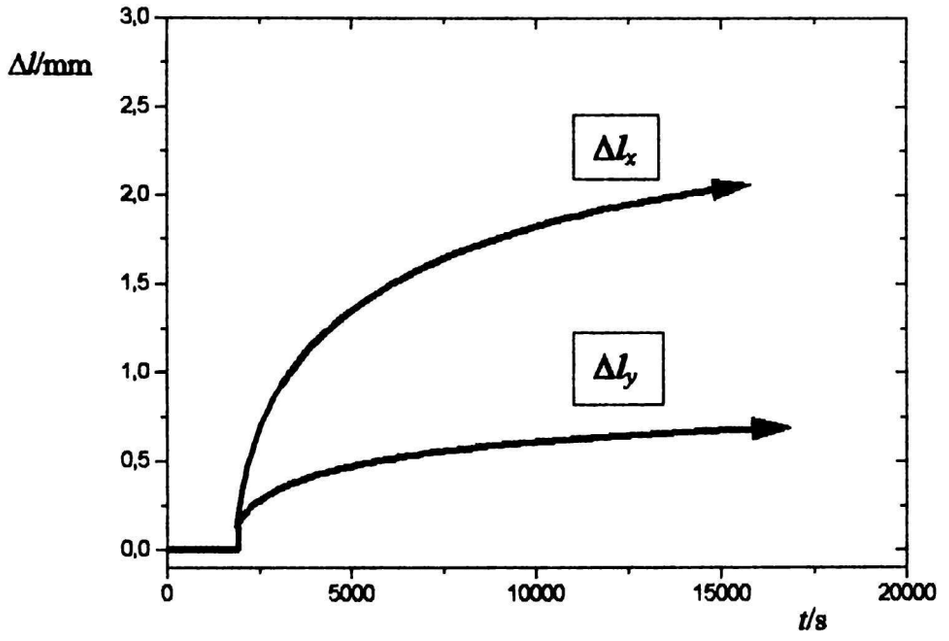

Fig. 5. The Δl time functions for different directions "x" and "y" and for the same: liquid dielectric (No. 1), T_L – temperature of the liquid, $\boldsymbol{F}$ – external mechanical force which acts on the film in course of interaction and potential Φ_L^s.

REFERENCES

1. Nash, J. L., *Polym. Eng. Sci. 28,* 862 (1988).
2. Fujiyama, M., Kawamura, T., Wakino, T., and Okamoto, T., *J. Appl. Polym. Sci. 28,* 1061 (1988).
3. Fujiyama, M., Kawamura, T., Wakino, T., and Okamoto, T., *J. Appl. Polym. Sci. 28,* 985 (1988).
4. Fujiyama, M., Kawamura, T., Wakino, T., and Okamoto, T., *J. Appl. Polym. Sci. 28,* 995 (1988).
5. Fujiyama, M., Kawamura, T., Wakino, T., and Okamoto, T., *J. Appl. Polym. Sci. 28,* 1011 (1988).
6. Fujiyama, M., Kawamura, T., Wakino, T., and Okamoto, T., *J. Appl. Polym. Sci. 28,* 1025 (1988).
7. Fujiyama, M., Kawamura, T., Wakino, T., and Okamoto, T., *J. Appl. Polym. Sci. 28,* 1035 (1988).
8. Fujiyama, M., Kawamura, T., Wakino, T., and Okamoto, T., *J. Appl. Polym. Sci. 28,* 1049 (1988).
9. Blok, J. and LeGrand, D. G., "Dielectric Breakdown of Polymer Films", General Electric Research and Development Center, Report No. 68-C-251 (1968).
10. Kvasnica, J., *Thermodynamics,* p. 22. Nakladatelství technické literatury (Publishers of Technical Literature), Prague, 1965.
11. Erdos, E., *Chem. Listy 48,* 1287 (1954).
12. Erdos, E., *Collect. Czech. Chem. Commun. 29,* 2406 (1964).
13. Šiškova, M. and Erdos, E., *Collect. Czech. Chem. Commun. 25,* 1729 (1960).
14. Šiškova, M. and Erdos, E.,*Collect. Czech. Chem. Commun. 25,* 2599 (1960).
15. Vaško, M., Bleha, T., and Romanov, A., *J. Macromol. Sci., Rev. Macromol. Chem. C 15*(1), 1 (1976).
16. Valko, L., *Chem. Zvesti 18,* 641 (1964).
17. Valko, L., *Chem. Zvesti 18,* 801 (1964).
18. Valko, L., *Polym. Bull. 9,* 144 (1983).
19. Polakovič, J. and Polakovič, J., Jr., Equipment for estimation of small changes of film length. *Czech.* 217 698 (1982).
20. Tomago, A., Shimidzu, T., Iijima, Y., and Yamaushi, I., *IEEE Trans. Electr. Insul.* Vol. *EI-12,* No. 4, 293 (1977).