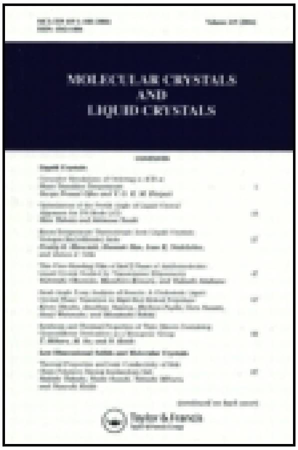

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Instability to Rupture of Anisotropic Viscoelastic Membranes: Dispersion Equation and Effects of Surface Elasticity and Viscosity

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INSTABILITY TO RUPTURE OF ANISOTROPIC VISCOELASTIC MEMBRANES: DISPERSION EQUATION AND EFFECTS OF SURFACE ELASTICITY AND VISCOSITY

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Abstract Instabilities of surface fluctuation waves can lead to rupture of liquid crystaline bilayer membranes. On the basis of the assumptions used in our previous works we derived the equations describing the instability of rupture of anisotropic, viscoelastic membranes under tension, in electric fields, and for arbitrary surface elasticities and viscosities. The surface elasticities increase the stability of the membrane, while the membrane tension destabilize it. External electric and mechanical constraints increase the rate of growth of the fluctuation waves. The fluctuation wave mechanism can describe at least qualitatively the experimental data for the time and tension dependence of the critical potential of rupture.

INTRODUCTION

Instability to rupture of biological and artificial membranes and thin films is involved in the mechanisms of wide variety of biological and engineering processes and can be important for applications to practice. Typical examples are: membrane fusion, membrane rupture and mechanical destruction, cell division and fusion, nerve exitation, colloid stability, etc.

Membrane rupture can be induced by mechanical, electrical and other constraints and can be due to several mechanisms[1-8]. One of them is based on the existence of surface perturbations, due to thermal motion or external vibrations. These perturbations in the shape of the membrane surfaces can be represented as a sum of surface waves of different wavenumbers. The growth of unstable modes can lead to formation of pores and membrane rupture. This is the fluctuation wave mechanism of membrane rupture, which is borrowed from the colloid chemistry where it describes well the rupture of thin liquid films.[10,11,12] Thermal fluctuations and external constraints can also lead to longitudinal waves. In this case instable modes can also give rise to pores. The pore expansion leads to membrane rupture. This is the stochastic pore formation and expansion mechanism of membrane rupture.[13,14] Originally the calculations based on this mechanism were based on the thermodynamic probability of pore formation. We presently are trying to derive these results on the basis of existence of longitudinal fluctuation waves (Zhelev and Dimitrov, in preparation). This can allow to find criteria for the limits of validity of both mechanisms: surface fluctuation waves mechanism and the stochastic pore formation mechanism.

Independently of what kind of mechanism is operating there is a very important question. Biological membranes are anisotropic and they can adsorb a lot of substances as proteins, ions, lipid molecules, nucleic acids. The adsorption will increase the transversal anisotropy of the membrane. How to account for those effects? Introducing different phenomenological constants for the directions normally and laterally along the membrane takes into account the fact that lipid molecules are anisodiametric. This can also be described more precisely by considering the membranes as liquid crystals. For the lake of simplicity here we do not consider explicitly the orientation changes. They are partly accounted for by the different constants in both directions. The adsorbed substances can be considered as a separate phase. It is, however, extremely thin. In addition, the adsorbed ions, for example, can be placed between the polar headgroups of the lipids. Therefore, there is a surface region which has specific properties and can not be described as a separate bulk phase. We will account for the elastic and viscous properties of this region by using the concept for surface elasticity and surface viscosity of membranes.[15]

Unlike previous works, where anisotropic viscoelastic membranes with zero or infinite surface elasticity[16] or isotropic membranes with arbitrary surface elasticity[15] this communication presents results for the more general case of an anisotropic viscoelastic membranes having arbitrary surface elasticities and viscosities. It also provides a detailed analysis of the role of the surface elasticity and viscosity.

THE MODEL AND BASIC EQUATIONS

The membrane is represented as a thin planar transversely isotropic viscoelastic lamella. The viscoelasticity is described by the Kelvin model. For this system the components of the stress tensor are:

$$p_{11} = -p\delta_{11} + 2G_1\varepsilon_{11} + 2\mu_1 e_{11} + (E_1 - G_1)(\varepsilon_{11} + \varepsilon_{22}) + (k_1 - \mu_1)(e_{11} + e_{22}) + K_v(\varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33}) + k_v(e_{11} + e_{22} + e_{33})$$

$$p_{22} = -p\delta_{22} + 2G_1\varepsilon_{22} + 2\mu_1 e_{22} + (E_1 - G_1)(\varepsilon_{11} + \varepsilon_{22}) + (k_1 - \mu_1)(e_{11} + e_{22}) + K_v(\varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33}) + k_v(e_{11} + e_{22} + e_{33})$$

$$p_{33} = -p\delta_{33} + E_2\varepsilon_{33} + k_2 e_{33} + K_v(\varepsilon_{11} + \varepsilon_{22} + \varepsilon_{33}) + k_v(e_{11} + e_{22} + e_{33}) \tag{1}$$

$$p_{12} = 2G_1\varepsilon_{12} + 2\mu_1 e_{12}$$

$$p_{13} = 2G_2\varepsilon_{13} + 2\mu_2 e_{13}$$

$$p_{23} = 2G_2\varepsilon_{23} + 2\mu_2 e_{23}$$

where: $\varepsilon_{ij} = (\partial u_i/\partial x_j + \partial u_j/\partial x_i)/2$, $(i,j = 1,2,3)$ are the components of the strain tensor; u_i are the components of the displacement vector; $e_{ij}=d\varepsilon_{ij}/dt$; G_1, E_1, G_2, E_2 and K_v are elastic coefficients; μ_1, k_1, μ_2, k_2 and k_v are viscosities; p – dynamic pressure; δ_{ij} are the components of the unit tensor.

Micropipette aspiration experiments show that the membranes behaves as incompressible[9]. The continuity equation for incompressible membrane is:

$$e_{11} + e_{22} + e_{33} = 0 \tag{2}$$

We assume that the membrane is transversely isotropic, but has two viscoelastic surface layers of different mechanical properties. For mathematical simplicity, only the variations in x_1 and x_3 directions are considered. Then the equations of motion for the membrane material, when there are perturbations expendable in two dimensional Fourier series are:

$$\begin{aligned}
\rho dv_1/dt = {} & -\partial p/\partial x_1 + (E_1 + G_1)\partial^2 u_1/\partial x_1^2 + \\
& (k_1 + \mu_1)\partial^2 v_1/\partial x_1^2 + G_2\partial^2 u_3/\partial x_1\partial x_3 + \\
& G_2\partial^2 u_1/\partial x_3^2 + \mu_2\partial^2 v_3/\partial x_1\partial x_3 + \mu_2\partial^2 v_1/\partial x_3^2 \\[1ex]
\rho dv_3/dt = {} & -\partial p/\partial x_3 + E_2\partial^2 u_3/\partial x_3^2 + \\
& k_2\partial^2 v_3/\partial x_3^2 + G_2\partial^2 u_1/\partial x_1\partial x_3 + G_2\partial^2 u_3/\partial x_1^2 + \\
& \mu_2\partial^2 v_1/\partial x_1\partial x_3 + \mu_2\partial^2 v_3/\partial x_1^2
\end{aligned} \tag{3}$$

where: ρ is mass density; u_i are the components of the displacement vector; v_j are the components of the velocity vector; x_1 and x_3 are the respective axes.

There are three types of derivatives in the right hand side of Eqs.(3) The biggest ones are derivatives of the dynamic pressure which are of the same order as the derivatives $\partial^2 u_1/\partial x_3{}^2$ and $\partial^2 v_1/\partial x_3{}^2$. The smallest derivatives are $\partial^2 u_3/\partial x_1{}^2$ and $\partial^2 v_3/\partial x_1{}^2$.

The terms in the left hand side of Eqs.(3) represent inertial forces and are of the same order as the smallest derivatives. In this work the smallest derivatives will be neglected. This means that the obtained results are valid for perturbation wavelengths larger than the membrane thickness. When the wavelengths are of the order of the membrane thickness they can be used as a rough approximation.

It is supposed that the solution of the differential equations (3) can be represented by the vector:

$$\begin{Bmatrix} p \\ u_1 \\ u_3 \\ v_1 \\ v_3 \end{Bmatrix} = \begin{Bmatrix} p_0(x_3)\cos kx_1 \\ u_{10}(x_3)\sin kx_1 \\ u_{30}(x_3)\cos kx_1 \\ v_{10}(x_3)\sin kx_1 \\ v_{30}(x_3)\cos kx_1 \end{Bmatrix} \exp(\omega t) \tag{4}$$

where: k is wavenumber and ω – angular velocity.

We use the following boundary conditions:

The normal components of the strain tensor at the interfaces are equal to the perturbation amplitudes,

$$u'_3 = \zeta_i\,, \quad \text{at} \quad x_3 = x'_3 + \zeta_i \tag{5}$$

$$(i = 1,2)$$

where: ζ_i is the perturbation amplitude at the i–th interface, and

x^1_3 is the x_3 coordinate of nonperturbed interface.

The tangential components of the stress tensor are balanced by the forces due to surface elasticity E^s and viscosity k^s:

$$p_{13} = E^s \partial^2 u_1/\partial x_1^2 + k^s \partial^2 v_1/\partial x_1^2 \tag{6}$$

The normal stress tensor components are balanced by the disjoining pressure Π and capillary pressure:

$$p_{33} = \Pi'\zeta + \sigma_i \partial^2 \zeta_i/\partial x_1^2 \tag{7}$$

$(i = 1,2)$

where: $\zeta = (\zeta_1 - \zeta_2)$ is membrane thickness fluctuation; $\Pi' = d\Pi/dh$ (h – is membrane thickness); and σ_i is surface tension at the i–th interface.

RESULTS

The dispersion equations found from the set of differential equations (3) and respective boundary conditions (5), (6) and (7) for symmetrical system $(\zeta_1 = -\zeta_2)$ is:

$$\begin{aligned} &\mathrm{th}(\lambda kH/2) - \lambda kH/2 - (\lambda^3/2P_2)(2\Pi' - \sigma k^2 - \\ &2P_2P_3(2P_1P_2 + \lambda^2 + 2)/((2P_1P_2 - \lambda^2)(2\Pi' - \sigma k^2) + \\ &2P_2P_3(1 + \lambda^2)^2) = 0 \end{aligned} \tag{8}$$

where: $H=(h+\zeta)$; $\lambda=(A_2+A_3+A_4-2A_1)/A_1$; $P_1=kA^s/A_1$; $P_2=1/kH$; $P_3=kA_1$; $A_1=(G_2+\omega\mu_2)$; $A_2=(E_2+\omega k_2)$; $A_3=(G_1+\omega\mu_1)$;

$A_4=(E_1+\omega k_1)$ and $A^s=(E^s+\omega k^s)$.

The dispersion equation for isotropic membrane can easily be found from (8) substituting $G_1 = G_2 = E_1 = E_2/2 = G$ and $\mu_1 = \mu_2 = k_1 = k_2 = \mu$.

In thin film approximation, when membrane thickness is much less than the perturbation wavelength the argument of the hyperbolic function is $(\mu kH/2) << 1$. Then the dispersion equation in thin film approximation for isotropic membrane reads:

$$(1/H)(8 - 3k^2H^2/2)A_1{}^2 + ((2\Pi' - \sigma k^2)x \tag{9}$$

$$x(k^2H^2/6 - 1) + 4A^s/H^2)A_1 - (2\Pi' - \sigma k^2)k^2HA^s/6 = 0$$

A_1 written here, is equal to $(G + \omega\mu)$, we keep the lowercase index 1 to remember that isotropic elasticity G and viscosity μ in this case are equivalent to transverse shear elasticity and viscosity. It is seen from this dispersion equation that in the long wave limit (k tends to zero) there are two asymptotic solutions. In the case of zero surface viscosity they are:

$$\omega_1 = - G/\mu \tag{10}$$

$$\omega_2 = (H\Pi'/4 - E^s/2H - G)/\mu \tag{11}$$

The first one is equivalent to that found from the dispersion equation for membrane with tangentially immobile surfaces, and the second one corresponds to the dispersion equation for a membrane with tangentially free surfaces. It is seen from (10) that for immobile surfaces (E^s tends to infinity) the asymptotic solution tends to infinity and there is one dispersion curve. When E^s is very small (membrane with tangentially free surfaces) in the thin film approximation there is one dispersion equation, which for small k corresponds to the second asymptotic solution.

Figure 1 and Fig.2 show the dispersion curves obtained by numerical solution of Eq. (8) for symmetric isotropic membranes with different surface elasticity for typical values of the membrane and the external field parameters: membrane elasticity $G = 10^4$ Pa; membrane viscosity $\mu = 10^2$ Pa.s; membrane surface tension $\sigma = 10^{-4}$ N/m; membrane thickness $h = 5.7\times10^{-9}$ m; transmembrane potential $U = 1$ V; relative membrane permittivity $\varepsilon_m = 5$. r is maximum perturbation characteristic size (in this case equal to 10^{-6} m).

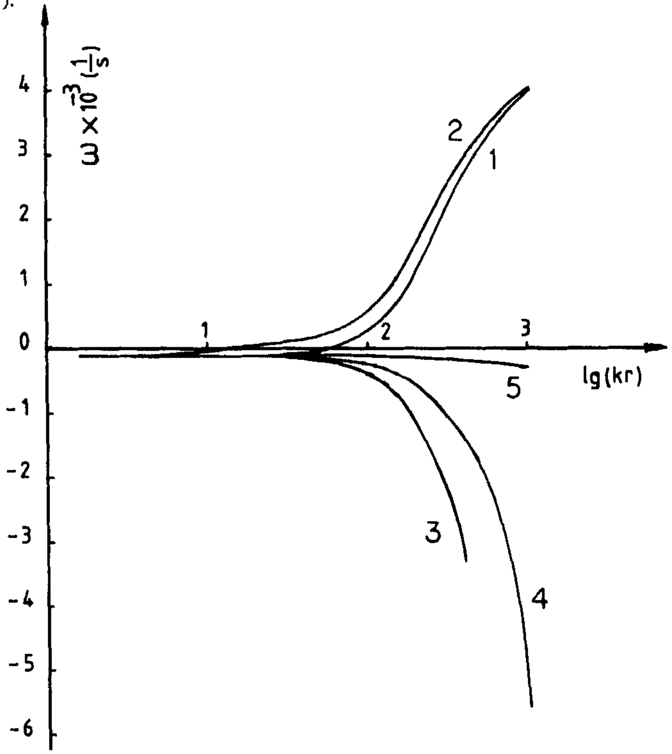

FIGURE 1. Dispersion curves from Eq.(8), for different surface elasticities (curve 1 – $E^s=10^{-1}$ N/m (or more); 2 – $E^s=4\times10^{-3}$ N/m; 3 – $E^s=2\times10^{-3}$ N/m; 4 – $E^s=10^{-3}$ N/m; 5 – $E^s=10^{-4}$ N/m), when $k^s = 0$. The corresponding asymptotic for small k is given by Eq.(10).

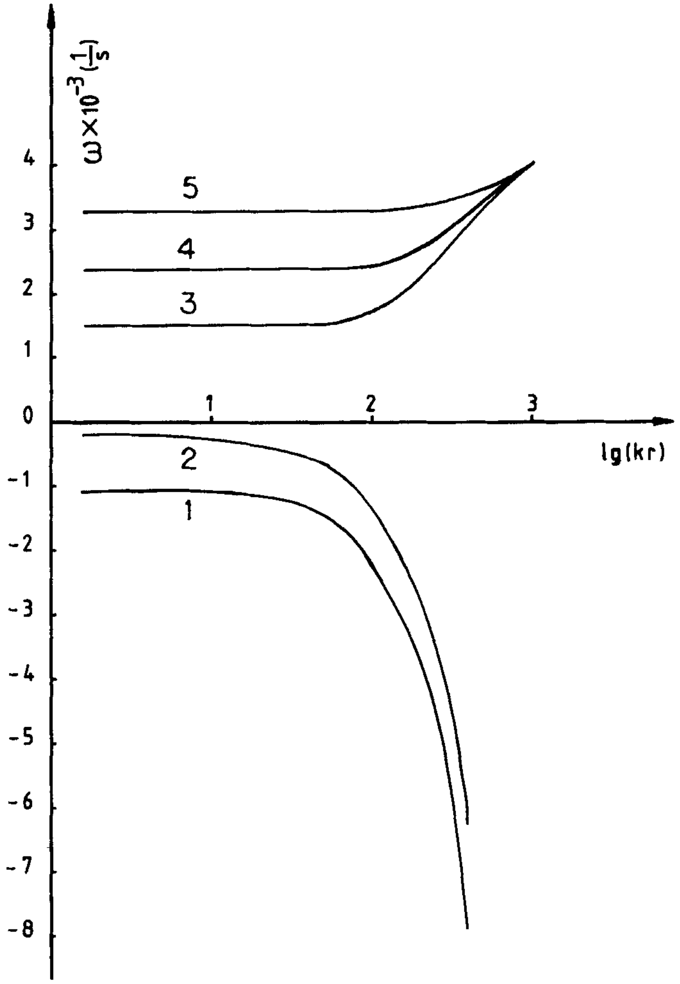

FIGURE 2. Dispersion curves from Eq.(8) for $k^s=0$ and surface elasticities – E^s (for curve 1 – $5x10^{-3}$ N/m; 2 – $4x10^{-3}$ N/m 3 – $2x10^{-3}$ N/m; 4 – 10^{-3} N/m; and 5 – 10^{-4} N/m (or less). The asymptotic of the dispersion equation for small k is given by Eq.(11).

The disjoining pressure presents the external forces. For transmembrane potentials from 0.1 V to 1 V (or more), where electric field membrane rupture occurs, the electrostatic component of the disjoining pressure is many orders of magnitude bigger than other components (e.g. van der Waals attraction of semiinfinite surrounding phases etc.). In this case the disjoining pressure is represent by the electrostatic component:

$$\Pi = - \varepsilon_0 \varepsilon_m U^2/2h^2 \tag{12}$$

where $\varepsilon_0 = 8.85 \times 10^{-12}$ F/m is vacuum permittivity.

For small k the dispersion curves approach the first and the second asymptotic solutions , described by Eqs. (10, 11). The increase of the surface elasticity leads to decreasing of the angular velocity of the unstable mode. It is interesting to note that rather large values of the surface elasticity (three orders of magnitude larger than GH for that particular case) are required to ensure tangential immobility of the membrane surfaces. On the other hand, for the same case, the membrane surfaces behave as tangentially free mobile when the surface elasticity is of the same order of magnitude as GH (or less). the Figures 3 and 4 presents the effects of the surface viscosity for finite values of the surface elasticity. The increase of the surface viscosity leads to decrease of the angular velocity for the unstable mode (Fig. 3).

CONCLUSIONS

The linear stability analysis of anisotropic viscoelastic membranes considered as consisting of a bulk three-dimensional medium and two surface layers leads to a relation between the wavenumber and the angular frequency (dispersion equation) which can be evaluated numerically and has several roots. Two limiting cases, isotropic membranes and the long-wave limit (thin film approximation), allow to investigate in more detail the effects of surface elasticity and surface viscosity. These two surface physical properties lead always to static or dynamic stabilization

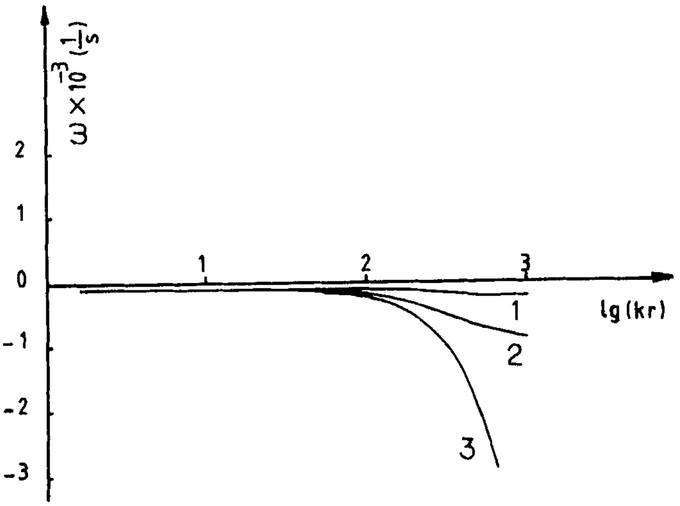

FIGURE 3. Dispersion curves corresponding to the curves shown on Fig. 1., for $E^s=10^{-3}$ N/m and different surface viscosities k^s (1 - $5x10^{-6}$ N.s/m; 2 - 10^{-6} N.s/m; and 3 - k^s = 0).

of the membrane, respectively. The surface elasticity can be important if the bulk elasticity is not too high. For long waves both types of elasticities appear as a sum of two constants to form one effective elasticity. By increasing the surface elasticity one can increase the membrane stability. For example, action of divalent cations on negatively charged lipid membranes, can lead to stabilization if there are not changes of the transmembrane potential. In this case the bulk membrane elasticity does not change so much. These effects can have practical application in membrane fusion. By adding different substances to one of the membrane one can increase only its stability.

The surface viscosity does not change the equilibrium condition for stability, but significantly affects the value of the angular velocity. It generally decreases the rate of growth of the fluctuation waves. For the particular values of the bulk elasticities chosen for the examples the dominant wavenumber does

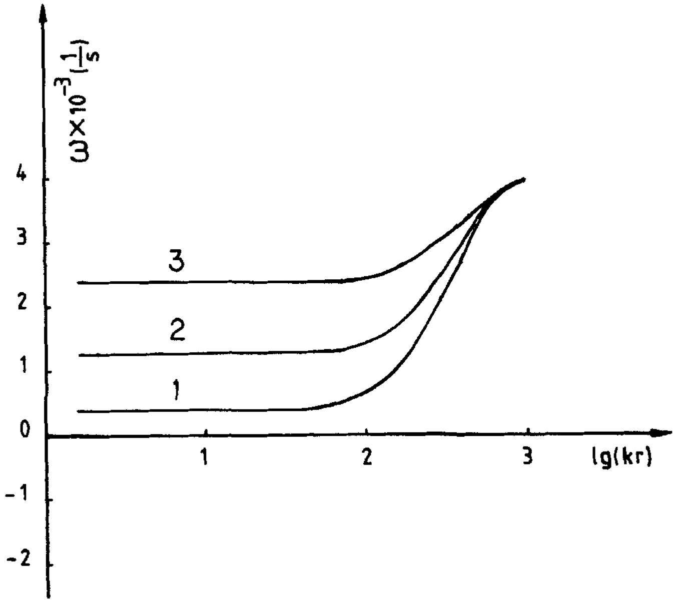

FIGURE 4. Dispersion curves corresponding to that shown on Fig. 2., for $E^s = 10^{-3}$ N/m and surface viscosities k^s (1 – 5×10^{-6} N.s/m; 2 – 10^{-6} N.s/m; and $k^s = 0$).

not depend on the surface viscosity. By decreasing the surface viscosity one can increase the rate of membrane rupture. For example, adsorption of proteins, ions, etc., can lead to increase of the characteristic time of rupture.

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REFERENCES

1. D. S. Dimitrov, J. Membrane Biol., 78, 53 (1984).
2. D. S. Dimitrov and D. V. Zhelev, J. Colloid Interface Sci., 99, 324 (1984).
3. D. S. Dimitrov and R. K. Jain, J. Colloid Interface Sci., 101, 489 (1984).
4. D. S. Dimitrov and R. K. Jain, Biochim. Biophys. Acta, 779 437–468 (1984).
5. D. V. Zhelev and D. S. Dimitrov, International Agrophysics, 1(2) 205 (1985).
6. D. S. Dimitrov, D. V. Zhelev, and R. K. Jain, J. Theor. Biol., 113, 353 (1985).
7. D. S. Dimitrov and D. V. Zhelev, Studia Biophysica, 110 105 (1985).
8. S. S. Dukhin, N. N. Rulyov, and D. S. Dimitrov, Coagulation and Dynamics of Thin Layers. Naukova Dumka, Kiev (1986).
9. E. A. Evans, Biophys. J., 13 926 (1973).
10. A. Sheludko, Proc. Koninkl. Nederl. Akad. Wet., B65 87 (1962).
11. I. B. Ivanov, Pure Appl. Chem., 52 1241 (1980).
12. D. S. Dimitrov, Progress in Surface Sci., 14 295 (1983).
13. B. V. Derjaguin and Yu. A. Gutop, Kolloid. Zh., 24 431 (1962).
14. Yu. A. Chizmadzhev, V. B. Arakelyan, and V. F. Pastushenko, Bioelectrochem. Bioenerg., 6 53 (1979).
15. A. Steinchen, D. Gallez, and A. Sanfeld, J. Colloid Interface Sci., 85 5 (1982).
16. C. Maldarelli and R. K. Jain, J. Colloid Interface Sci., 90 233 (1982).