P h y s i c s

STUDY OF THE SWELLING OF THE PHOSPHOLIPID BILAYER, DEPENDING ON THE ANGLE BETWEEN THE INCLINATION OF THE DIPOLE FRAGMENTS AND THE PLANE OF THE LAMELLA

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Using computer simulations and simultaneous X-ray diffraction at large and small angles, the dependence of the swelling coefficient of the phospholipid bilayer on the angle between the inclination of dipole fragments and the plane of the lamella was investigated. It was shown that in this case the balance between electrostatic, repulsive and van der Waals attraction forces is a particular importance. As the dipole angle increases, the repulsive forces of the electrostatic dipole-dipole interaction increase, resulting the increase of the bilayer thickness as a function of the number of –CH-groups in the phospholipid. At a certain angle the bilayer is destroyed. It is also shown that at certain small angles the bilayer takes a minimum thickness. In this case a phase transformation takes place and the system enters the "gel"-phase.

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Introduction. Phospholipids are complex compounds which, forming a bilayer, make the main part of the cell membrane and at the same time are its fundamental structural unit. Like other biological films, the thickness of the phospholipid bilayer does not exceed 10 *nm* (100 *Å*) and contains both hydrophilic regions (phosphoric acid residues included in the heads) and hydrophobic regions (fatty acid radicals forming the tails: –CH-groups). These properties allow the phospholipids, being in an aqueous medium, to form a bilayer, causing new physical properties of the biosystem [1–3].

This follows from the experimental data, that the angle between the dipolar fragments and the plane of the bilayer depends both on the intermolecular distance of the phospholipids and the distance between the layers. At different values of these distances the angle of inclination is being changed, which causes violation of both the structure and the aggregate states in the bilayer. At present, software programs are

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being used for molecular modeling of complex biosystems, which makes it possible related to solve some problems with the phospholipid bilayer and lamellar density. Methods of molecular dynamics and quantum mechanics are widely applied in the computer modeling of atomic and electronic structures of molecular and crystalline complex systems. One of the more convenient and accurate methods is the Hyper Chem 8 simulation program.

It is rather difficult [4] to determine experimentally the swelling of phospholipid membranes. This swelling is an important parameter of the biophysical function of the vitreous membranes. Modern computer programs make it possible to obtain accurate data on this parameter for their further comparison with experimental data. Thus, the study of membrane swelling has not been sufficiently investigated yet and is an urgent problem.

The aim of this work is to study the swelling of a phospholipid bilayer, which depends on the angle between the dipole fragments and the plane of the lamellae, using the computer simulation.

Experimental Part. A computer model of the phospholipid bilayer was made up in order to solve this problem. For this purpose, the Model build subroutine is used (Fig. 1).
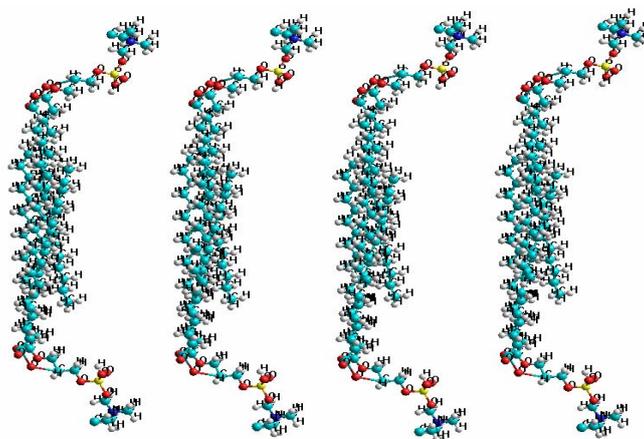

Fig. 1. Computer model of a phospholipid bilayer.

For the computer model of a phospholipid bilayer, the distance of localized dipole fragments in the upper and lower regions, i.e. the thickness of the bilayer is $L = 41$ Å, the distance between phospholipid molecules is $a = 15$ Å, the number of molecules is $2n = 8$, and the slope angle of the dipole fragment to the lamell plane (θ) is $60°$. The change of these parameters brings to a change in the angle θ and in some cases is the cause of the destruction of the bilayer.

The molecular mechanics method is used in the force field AMBER for geometric optimization.

The present paper also discusses the transitions phase [5] in lipid membranes with an emphasis on the lipid bilayer model. It reveals a microscopic image and

examines the key factors that leads to the phenomena described above. A general discussion of the model has been carried out and some specific examples are presented in more details. Experimental approaches for the study of the phase behavior of lipid membranes are also considered, and the biological results are evaluated.

An example of that are the amphiphilic molecules in mixture. As it was mentioned earlier [6–9], these molecules had a group of hydrophilic polar heads and two hydrophobic hydrocarbon tails. This means that in the amphiphilic–water system the heads of lipids are oriented in the polar aquatic environment. At that time the hydrophobic tails tend to minimize their contact with water (Fig. 2).
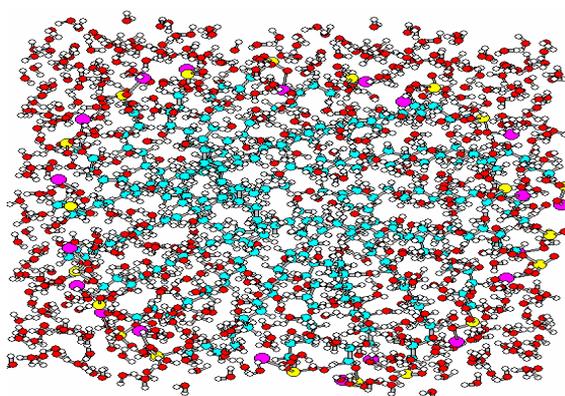

Fig. 2. Model of a mycel for the amphiphile–water system.

If there is a surface, they will migrate there and subsequently reduce the surface tension.

Results and Discussion. The dependence of the slope angle of the dipole fragment θ on distance L, for $N = 6,\ 8,\ 10,\ a = 15$ Å, has the following form (Fig. 3).
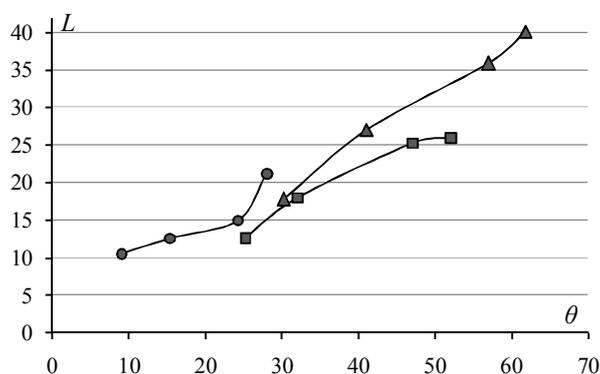

Fig. 3. Dependence of the thickness of the bilayer L on the angle of the dipole fragment at

$N = 6 - \bullet$;
$N = 8 - \blacksquare$;
$N = 10 - \blacktriangle$;

$a = 15$ Å $(K = 1.2;\ 1.4;\ 1.5)$.

Using the formula for swelling [10] and measuring the angle of inclination for the graphs in Figs. 3 and 4, we determine the coefficient of swilling K of the phospholipid bilayer for each graph.

With increase of L, the Van der Waals attraction force between the –CH-groups decreases [11–13], which breaks the balance between the electrostatic repulsive forces of the dipoles, and this leads to the destruction of the bilayer at distances $L > 44$ Å. That is to say, a phospholipid bilayer with such parameters and inclination angle $\theta > 82°$ cannot exist (Fig. 4).
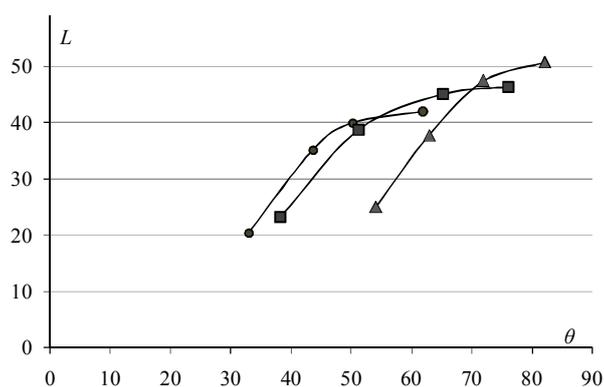
Fig. 4. Dependence of the thickness of the phospholipid bilayer L on the angle θ of the dipole fragment at
$N = 12 \ - \ \bullet$;
$N = 14 \ - \ \blacksquare$;
$N = 16 \ - \ \blacktriangle$;
$a = 15$ Å ($K = 1.7$; 1.85; 1.9).

Theoretical calculations show that this critical angle $\theta = 83°$ [10] increases with the number N of the –CH-groups (Fig. 5).
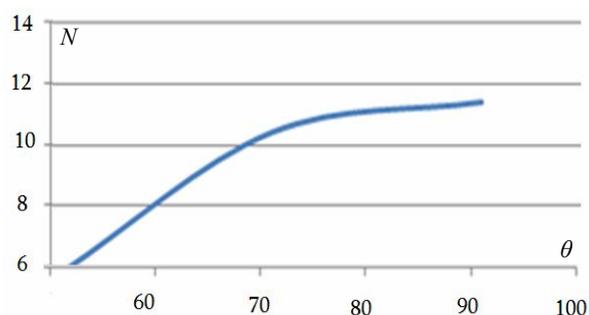
Fig. 5. Dependence of number N of the –CH-groups on the slope angle.

Conclusion. Comparing the results of the computer simulation with the experimental data, we obtain that with an increase of the angle θ of the dipole fragment, the thickness of the phospholipid bilayer L increases, so the bilayer swells. There are critical values of the inclination angles θ_{cr}^{u} and θ_{cr}^{l}. For the values of $\theta > \theta_{cr}^{u}$ the bilayer is being destroyed, and at $\theta < \theta_{cr}^{l}$, a phase transition occurs and the bilayer turns into the "gel" phase. The values of the angle grow with increasing numbers N of –CH-groups irregularly, and with a large number of N of the –CH-groups, the increase of the angle θ_{cr}^{u} is slowed down.

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