Հայաստանի Գիտությունների Ազգային Ակադեմիա
Национальная Академия Наук Армении
National Academy of Sciences of Armenia

Հայաստանի Կենսաբանական Հանդես
Биологический Журнал Армении
Biological Journal of Armenia

•Փորձարարական և տեսական հոդվածներ •Экспериментальные и теоретические статьи•
•Experimental and theoretical articles•

Biolog. Journal of Armenia, 3 (66), 2014

STUDY OF MOLECULAR DYNAMICS OF LAMELLAR PHASE OF AMPHIPHILE/WATER SYSTEMS

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The lamellar phase of amphiphile/water system was investigated by using atomic scale simulation. A 300 ns parallel molecular dynamics (MD) simulation was carried out for amphiphile/water system consisting of 128 sodium pentadecyl sulfonate (SPDS) and 2251 water molecules using GROMACS software package. At T=323K temperature we got both tilted fully interdigitated and liquid crystalline like disordered hydrocarbon chains, i.e. we observe a gel phase that coexists with a lamellar L_α phase.

Molecular dynamics (MD) simulations – alkyl sulfonates

Ուսումնասիրվել է երկսեր նյութ/ջուր լամելային փուլը օգտագործելով ատոմական մակարդակի համակարգչային մոդելավորում: Իրականացվել է 300 նվ զուգահեռ մոլեկուլային դինամիկ ուսումնասիրություն որպես համակարգ վերցնելով՝ երկսեր նյութ/ջուր համակարգը, բաղկացած 128 նատրիումի պենտադեցիլ սուլֆոնատի և 2251 ջրի մոլեկուլներից օգտագործելով GROMACS ծրագրային փաթեթը: T=323K ջերմաստիճանի դեպքում, ստացվել են պոչերի իրար մեջ լրիվ ներթափանցում, ինչպես նաև հեղուկ բյուրեղական չկարգավորված պոչեր, այսինքն դիտվել է «գել» փուլ, որը համագոյակցում է լամելային L_α փուլի հետ:»

Մոլեկուլային դինամիկ (ՄԴ) ուսումնասիրություն – ալկիլ սուլֆոնատ

При помощи компьютерного моделирования на атомарном уровне была исследована ламелярная фаза системы амфифильное вещество/вода. Была проведена параллельная молекулярно-динамическая (МД) симуляция длительностью 300 нс для системы, состоящей из 128 молекул пентадецил сульфоната натрия (ПДСН) и 2251 молекулы воды, с использованием программного пакета GROMACS. При температуре T=323K мы получили как наклоненные полностью взаимопроникающие, так и жидко-кристалличесские неупорядоченные углеводородные цепи, то есть наблюдалось сосуществование гель и ламелярной L_α фаз.

Молекулярно-динамическая (МД) симуляция – алкил сульфонат

Amphiphilic molecules, which consist of a polar headgroups and alkyl tails, in water solution, can generate self-organized lyotropic liquid crystalline structures composed of bimolecular layers of fluid amphiphile molecules separated by water layers. This kind of bilayers has been studied for many years as simplified models for phospholipid membranes [1-2], as well as they have been widely intensively used in drug delivery [3-5]. Experimental methods, such as X-ray diffraction and neutron scattering have been mainly used to reveal the structures and features of bilayers [1, 6].

Recently, with the increase of hardware power, the atomic-level computer simulations have been used as an additional method to study such systems [7].

The aim of this paper was the detailed study of the lamellar phase of ionic sodium pentadecyl sulfonate (SPDS)/water systems by means of molecular dynamics simulation. The determination of structural parameters (interlayer spacing and area per molecule) has been also done with comparison to experimental findings.

Materials and methods. A sodium pentadecyl sulfonate (C15H31SO3Na) molecule was used, which was extracted from our previous simulation [8]. We build a system with 128 SPDS and 2251 water molecules using our own code and GROMACS [9] modules. After final construction, the energy of the system was minimized using steepest descent method (5000 steps). The force field parameters for SPDS molecule were in detail discussed in [8]. The SPC models were used [10] for water molecules and the chemical bonds were maintained using with LINCS constrain algorithm [11]. The temperature was set to 323 K and was maintained using V-rescale algorithm. An isotropic normal pressure (1 atm) was maintained by the Berendsen coupling algorithm [12]. The PME [13] was used for long-range electrostatic interactions, and the van der Walls interactions were truncated at 1.2 nm. The equations of motion are integrated using leapfrog Verlet integrator with timestep of 2 fs.

The 128SPDS/2251 water system was subjected to a 300 ns simulation in NPT ensemble using GROMACS software package. The parallel MD simulation was performed on ArmGrid sites (http://www.grid.am).

Results and Discussion. The important characteristics of the system, such as the interlayer spacing and the area per molecule are determined and are shown in fig. 1 and 2.

As one can see from both curves, at the beginning of simulation run, we track the variation of interlayer spacing and area per molecules, which is probably due to the reorientation of disordered hydrocarbon chains. However, at the end of simulation, we track an equilibrium of system and as a result we receive ~ 6,3±0,2 nm and ~0.32 nm^2 for interlayer spacing and area per molecule, respectively.

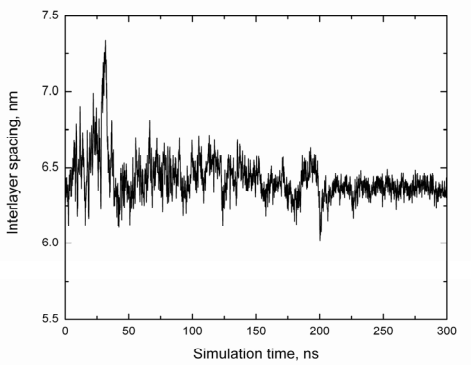

Figure 1. The interlayer spacing depending on simulation time.

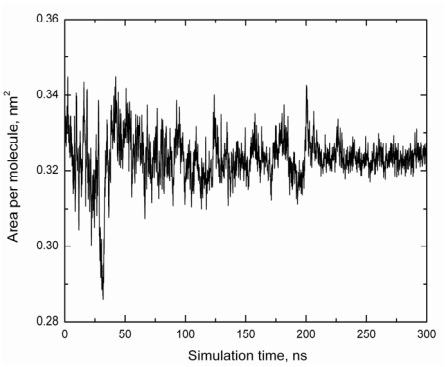

Figure 2. The area per molecule depending on simulation time.

Note that the obtained finding on interlayer spacing and area per molecule are in agreement with our X-ray diffraction data [14].

To visualize the structural changes in detail, we also provide the snapshot of SPDS/water systems at the end of simulation run. In Figure 3 the cross-sectional view perpendicular to the bilayer plane is shown at 300 ns point.

We see that the packing is an interdigitated gel phase, i.e. an ordered state with a tilt angle, as clearly seen by the snapshots. This phase is generally known as gel phase. Thus, the inspection of trajectories and snapshots reveals the fully interpenetration of the SPDS alkyl chains located on opposite sides of the bilayer (fully interdigitated gel state), however, together with ordering, the disordered domains called "clusterized regions"[15] are also found.

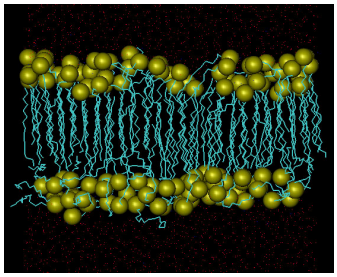

Figure 3. The cross-sectional view perpendicular to the bilayer plane.
The counterions and water hydrogens have been omitted for clarity.

Hence, we assume the formation of the gel phase, which coexists with a lamellar L_α phase. This kind of phases, on the other hand can be treated as a metastable gel phase with some fraction of disordered domains, which are also observed in phospholipid membranes [16]. Thus, the analysis of parameters and the snapshots implies either the formation of a coexistence between fully interdigitated and randomly phases or the presence of metastable gel phases.

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Received on 26.06.2014