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Distant transmission of medicinal substance properties to water is the result of the phonon mechanism of dispersed bodies' surface forces

S.N. Novikov[1], A.I. Ermolaeva[1], S.P. Timoshenkov[1], E.P. Germanov[2]

***Abstract*—A comprehensive study in the transmission of medicinal substance properties to liquid water (project "Emission" by DST fund) has been carried out using kinetic methods of gravimetry, work function of electron, scanning force and capacitive microscopy. Assumption based on model experiments, theory of water's coherent dynamics and conceptions about phonon mechanism of disjoining pressure structural component at phase boundary has been proposed: the observed transmission effect of medicinal substance properties to the liquid water (www.dst-fond.ru and www.newpharm.ru) was caused by interaction of dispersed electric surface micro-relief of carriers (oxide surface, CDR), modified by medicine, with the dipole-dipole emission from the decay of the coherent domain by water evaporation.**

I. INTRODUCTION

Properties of the phase boundary surface are always in the spotlight of physicists and chemists, and inspire new theoretical and experimental studies. An important event was publication of the book by leading experts in this field ("Surface Forces" by B.V. Deryagin, N.V. Churaev, V.M. Muller, 1987), devoted to various aspects of surface forces [1]. Analysing vast experimental results showing the processes occurring at the phase boundary surfaces of "gas-solid", "liquid-solid", the authors concluded that there is not enough to use the conception of the traditional dispersion and electrostatic forces for explaining the observed phenomena.

The first possibility of a "third type" of force existence associated with structure changes of solvent boundary layers, has been pointed out in works by (Deryagin B.V. and Kusakov M.M., 1936-1937). Later, it was called structural forces or structural component of disjoining pressure (Deryagin B.V. and Churaev N.V., 1972-1974). "The hydrogen bond grid presence in the water implies that change in mutual arrangement of water molecules in the boundary layer imposed by active centers arranged in a definite way on the surface, attenuates slowly with distance spreading over considerable distances (up to 100 Å, sometimes more). A surface atoms and groups act as active centers... It should be noted that the structure of the boundary layers, and consequently, their properties strongly depend upon place of active centers positioned on the surface and their number per area unit... Structural long-range action of surface isn't restricted by one or two layers of water molecules. The higher the surface hydrophilicity and the lower temperature and electrolyte concentration, the more appreciably it occurs." In conclusion the authors note [1]: "In fact, the theory of structural component of disjoining pressure in polar liquids with hydrogen bonding (macro-structural forces) is just starting to develop."

We put such an extensive quotation from [1] here as it contains the basic ideas used in discussion of our experimental results. Ideas in [1] were enhanced in articles of B.V. Deryagin school [2], [3]. The authors proposed a phonon mechanism of surface forces, binding surface characteristics with a change of liquid frequency spectrum, as a result of a thorough theoretical study of structural forces problems and analysis of experimental studies.

Literature dedicated by water structure has about 8 million references. Attention to this matter especially increased after theoretical works of the Milan physics school [4] used the quantum electrodynamic approach to the liquid water thermodynamics (a coherent water theory - CWT).

Studies of the water structure carried out in our laboratory have shown that our method - kinetics of electron work function (EWF) and derivatography (DTA + DTG) - allows getting objective information about permolecular water structure. In one study we have observed an interesting phenomenon – distant transmission of information about water solutions structure of Zn, Ba, Na and K chlorides to distilled water [5]. Similar observations were also noted in other studies [6].

Distant transmission of medicinal substances properties to liquid water has been introduced into practice recently. An example of such work is the project "Emission" performed by "DST Fund". The main operations by transferring properties of medicinal substances to water are the following:

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[1] National Research University MIET, *aviary@mail.ru* (S.N. Novikov)

[2] DST Foundation, *dstfund@gmail.com*

- transmission of information about medicine properties to the compact-disk (CDR) by laser scanning;
- transmission of this information through the Internet to the consumer's CDR;
- distant transmission of information about medicinal substance properties from the last CDR to the liquid water by covering the vessel with water by "CDR-cap" or installing water vessel on the CDR;
- using of "irradiated" water for treatment of a particular disease.

Omitting medical aspect of project "Emission"[1], from the standpoint of modern scientific knowledge in the present work we are trying to explain the possible physical mechanism above mentioned operations. Essentially, it's necessary to understand the mechanism of two main processes:

a) transmission of information about medicine to CDR;

b) transmission of information from CDR to liquid water.

To clarify these questions, this study was carried out in two steps.

The first step, an attempt of the simulation process of transferring information about the solid surface structure to distilled water was done. In the second step, we studied the water structure changes after it was affected by the surface of CDR with "emission" of different medicinal substances.

Discussions of the results were based on the above-mentioned ideas about the structural long-range action [1] and ideas obtained in the CWT [4].

We used EWF kinetic measurement methods, derivatography and the force capacitance scanning microscopy.

II. Experimental part

In research related to the water structure, special attention is given to purity of water, i.e. absence of uncontrolled impurities. It should be noted that getting pure water is a particular problem. Essentially, there is no absolutely pure water in nature. Even when water is placed in a particular vessel at atmospheric conditions, noticeable impurities appear. Therefore, in this work we used distilled water samples, obtained by a standard distillation apparatus. The impurities present in the water were not subjected to special control. However, in each experiment of the affecting the water structure by dispersion solid state body we used the same "fresh" distilled water sample for the relative comparison of results. In cases where we used solutions of KCl and $BaCl_2$, they are also prepared by the same distilled water.

The study was performed as follows. The distilled water sample (or solution) was placed in a standard chemical glass crystallizer (diameter of 10.5 cm and a depth of 1.5 cm), so the face of liquid was approximately at half of the crystallizer height (~ 5-7 mm) (Fig. 1). Solid body (an aluminum wafer or CDR) was the cap (the "Al-cap", the "CDR-cap") of this crystallizer. We paid special attention to the fact that the liquid had a free connection with atmosphere air (the system should be open). This procedure was always performed thoroughly in this study.

During the first stage a model experiment was performed in which the "Al-cap" was used as the temporary carrier; at the second stage a CDR was used as the temporary carrier. The experiment consisted of water (or solution) exposition in a vessel under Al-cap (or CDR), and then the isothermal evaporation rate of the exposed water samples was determined. Surface structure of the Al-caps had been studied previously in [8], [9], the results are given in the discussion.

Figure 1. Crystallizer with the water and Al-cap, a crucible of derivatograph for placing water sample.

Isothermal evaporation kinetic curves of water samples were obtained by derivatograph, the methodology is described in [10]. The sample of liquid was collected with a glass pipette (~ 0.5 g) and transferred to a crucible of derivatograph for evaporation study at atmosphere conditions.

In the first stage the study was performed to clarify the following questions:

- What is the structure of distilled water at base (initial) state, before exposure under the Al-caps;
- The same with the Al-cap oriented to water by metal or oxidized surface;
- What is the structure of one molar solution as KCl and $BaCl_2$ (without Al-caps);
- What is the structure of water under Al-cap, with surface modified by a 1M solutions of KCl and $BaCl_2$.

Solutions of KCl and $BaCl_2$ were chosen as model surface modifiers for Al-caps because their effect to the kinetic evaporation of water is very different, as it was previously shown in [11].

To modify the surface of the Al-caps we were dipped them in a one molar solutions (~ 1-2 days) with subsequent washing and drying (1 day). Monitoring the surface properties change in the Al-caps after modification was carried

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[1] Currently, significant statistic of biological and medical data showing about advisability such method of treatment disease has been accumulated [7].

out by measuring changes of the electron work function (EWF) (Table I).

Table I
Electron work function ϕ and surface potential ψ of initial and modified Al-caps.

NN	Surface without AOF		Surface with AOF		Remarks
	ψ, mV	ϕ, eV	ψ, mV	ϕ, eV	
1	668	5,17	-140	4,36	Base state
2	590	5,09	-119	4,35	of Al foil
1a	482	4,98	-184	4,32	1 day over
2a	618	5,12	-570	4,06	distilled water
1b	830	5,33	10	4,51	1 day of treatment in 1M of KCl solution
2b	1400	5,64	608	5,11	1 day of treatment in 1M of $BaCl_2$ solution

The results of the above listed experiments obtained in the first stage are shown at Fig. 2 and 3 as sample weight P dependency of evaporation time τ with constant temperature (19-22 °C) under atmosphere conditions, and also in Table II.

Dependence P=f(τ) in each experiment was recorded automatically; simultaneously the temperature of crucible with sample and the temperature difference between the test sample and etalon was recorded (DTA). This difference is characterized change of enthalpy in the sample.

Typical dependence P=f(τ) for distilled water is shown in Fig. 2. Maximum weight loss of water samples for selected observation time ($\tau_2 = 4000$ s) is $\sim 18 \cdot 10^{-4}$ g, the accuracy of measurement of weight $\pm 0,5 \cdot 10^{-5}$ g. The behaviour of dynamic curves P=f(τ) of the water and solutions evaporation was discussed in detail previously [11] where it was shown that a coherent phase having lower density evaporates primarily [4]. Figure 2 shows that dependence P=f(τ) for sample of distilled water has a typical “piecewise-linear” nature. Point τ_1, in which the evaporation rate changes, indicates the rates superposition of evaporation coherent domain (CD) and non-coherent molecular water, situated between CD according to [4].

Thus,

$$V_1 = V_{CD} = \frac{P_{CD}}{\tau_1}, \quad and \quad V_2 = V_{H_2O} = \frac{P - P_{CD}}{\tau_2} \qquad (1)$$

Data results of the evaporation rates are shown in Table II.

Methodology of the study at the second stage was the same but with one difference: we used CDR with medicinal information as “caps”.

III. Discussion

A. Stage I - A model experiment using oxidized Al-foil as a “temporary carrier” [2]

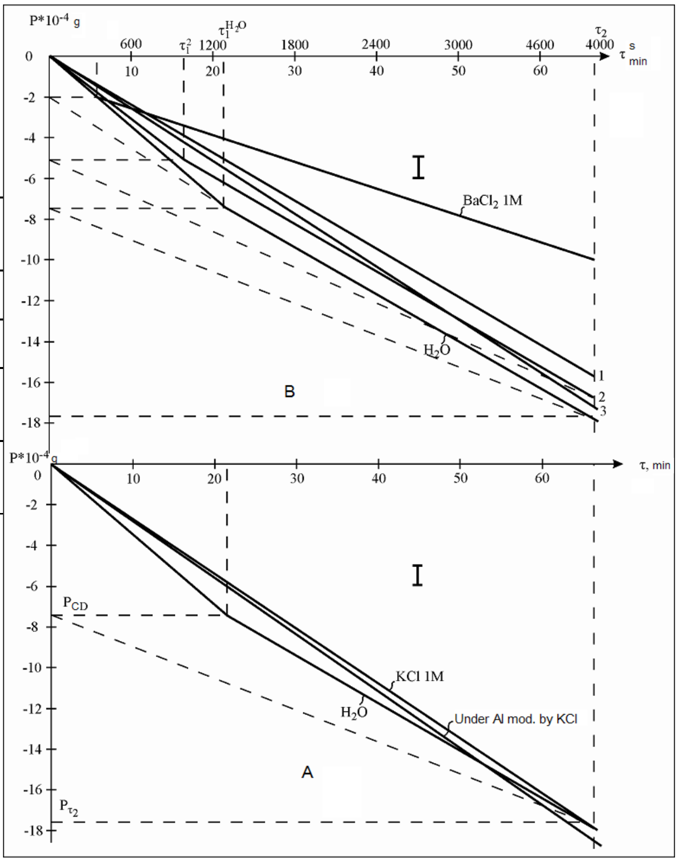

Figure 2. Isothermal evaporation kinetic curves of water samples with observed time τ_2=4000 s. A) The dependences of P=f(τ) in Al-cap modified by KCl. B) The dependences of P=f(τ) in Al-cap modified by $BaCl_2$.
1. Exposure for 1 day.
2. Exposure for 2 days.
3. Exposure for 6 days.

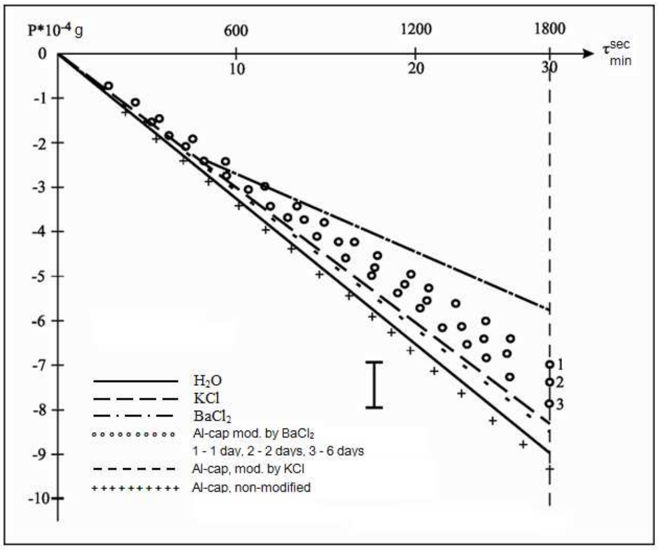

Figure 3. Kinetic curves isothermal evaporation of distilled water and solutions observed by τ_2=1800 s.

1) The surface structure of anodic-oxidized aluminium (“Al-caps”): Structure and properties of anode-oxidized film (AOF) of aluminum foil used for Al-caps were studied in [8], [9]. Using EWF measurements, scanning force

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[2] The results obtained in the first stage were presented and discussed at the VI International Congress of “Low and super-low fields and radiation in biology and medicine”, 2012, St. Petersburg, p.25 [12].

Table II
Average characteristics of the isothermal evaporation kinetic dependences for different water samples.

NN	Sample, exposure time	$P \cdot 10^{-4}$, g per 4000 s	$P_{CD} \cdot 10^{-4}$, g	$C = \frac{P_{CD}}{P}$	$V_{\text{evap.}}$, g/s		Δt, $^{\circ}C$	Conditions remarks
					$V_1 \cdot 10^{-6}$	$V_2 \cdot 10^{-6}$		
1	Distilled water	18,0	7,5	0,42	0,60	0,46	4	Initial from the flask
2	Distilled water	18,5	7,5	0,42	0,60	0,46	4	In the crystallizer under the Al-cap non-modified
3	Solution of KCl 1M	18,5	-	-	-	0,46	5	From flask
4	Solution of $BaCl_2$ 1M	11,2	2,6	0,25	0,47	0,21	8	From flask
5	Distilled water, 1 day	18,5	-	-	-	0,46	5	Under the Al-cap modified by KCl
6	Distilled water, 1 day	15,8	-	-	-	0,39	6	Under the Al-cap modified by $BaCl_2$
7	Distilled water, 2 days	17,4	2,7	0,15	0,60	0,37	6	The same
8	Distilled water, 6 days	16,5	5,0	0,30	0,50	0,29	4	The same

microscopy (SFM) and getting water absorption isoterms by weight method has been shown that AOF on Al is in a stationary-polarized state caused by production of molecules associates, i.e. coherent domains at surface of absorbed water [8]. Further research [9] using scanning capacitance microscopy (SCM) besides of SFM showed that:

a) AOF on Al foil consists of disk-shaped structures with an average size 200x200x30 nm;

b) Space between these "disks" has a higher electric potential;

c) Fluctuations of surface potential in AOF pores leads to development of long-range (up to ~ 0.8 microns) surface forces.

Thus, the system of permolecolar associates (coherent domains) of sorbed water causes structural-phase micro-relief of AOF surface and reduces aluminum EWF. The characteristic geometric and electrical AOF microstructures of Al-caps surface are shown in Fig. 4–7.

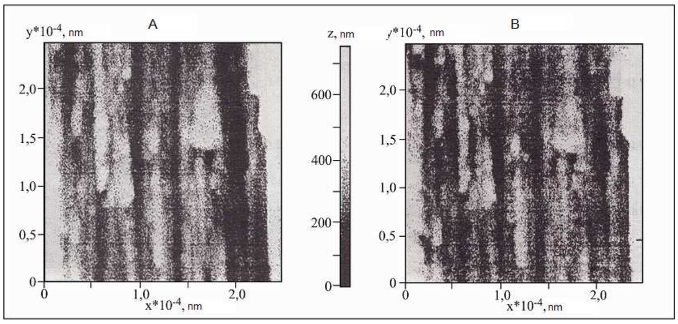

Figure 4. Surface of Al-cap without AOF: a) SCM; b) SFM.

Photomicrographs of the same Al-caps surface part made in semi-contact mode SFM (geometrical structure) and non-contact mode SCM (electric micro-relief) are presented in these figures. Fig. 4 and 5 shows the structure of Al foil surface (without AOF); Fig. 6 and 7 shows the research results of oxidized side wafer (AOF). Comparing these figures we may conclude that as AOF makes more advanced geometric surface structure, it qualitatively changes the electric micro-relief features.

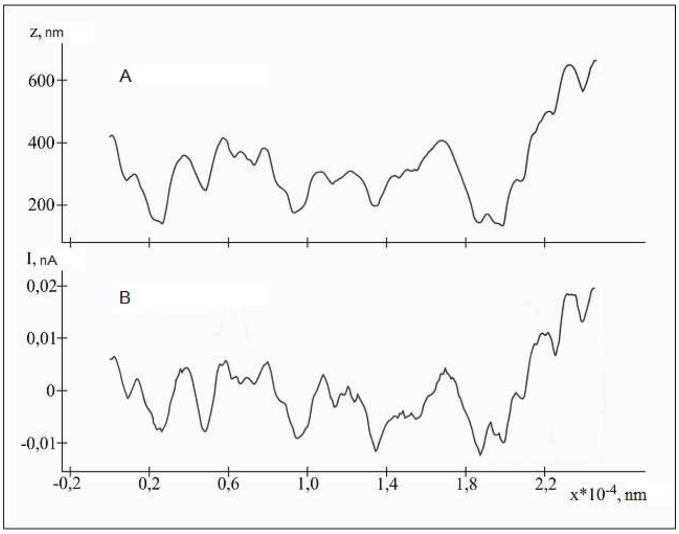

Figure 5. Profilograms of Al-cap surface without AOF: a) SCM; b) SFM.

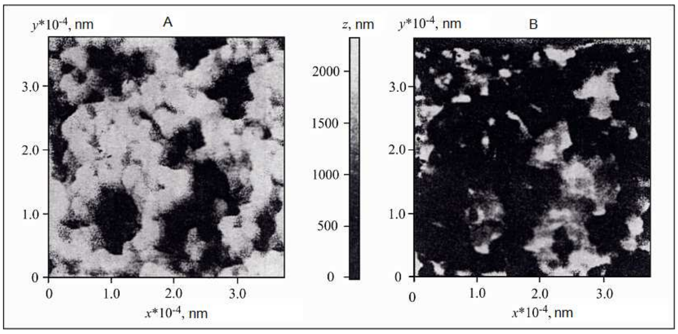

Figure 6. Surface of Al-cap with AOF: a) SCM; b) SFM.

Fig. 8 shows how electric relief is changed as distance H between cantilever and sample surface grows. As follows from Fig. 8b cantilever stops to "feel" surface only when H=0,8 microns. However, it does not mean that the structural component of surface pressure [1] at much greater distances between phase borders completely disappears. This is evidenced by data obtained in the present research (see below).

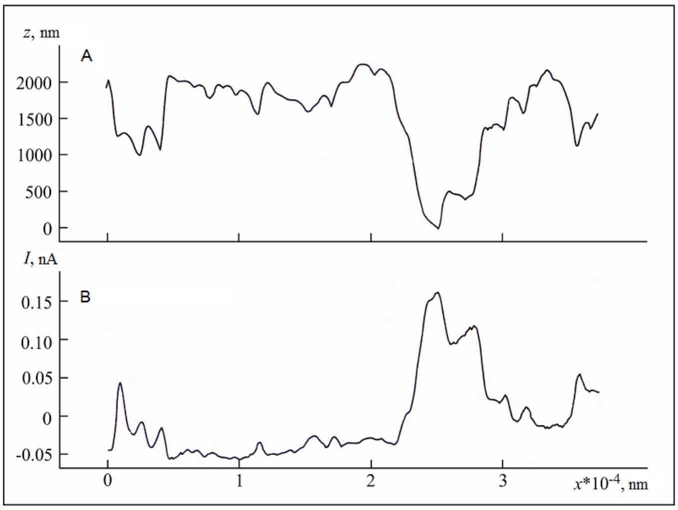

Figure 7. Profilograms of Al-cap surface with AOF: a) SCM; b) SFM.

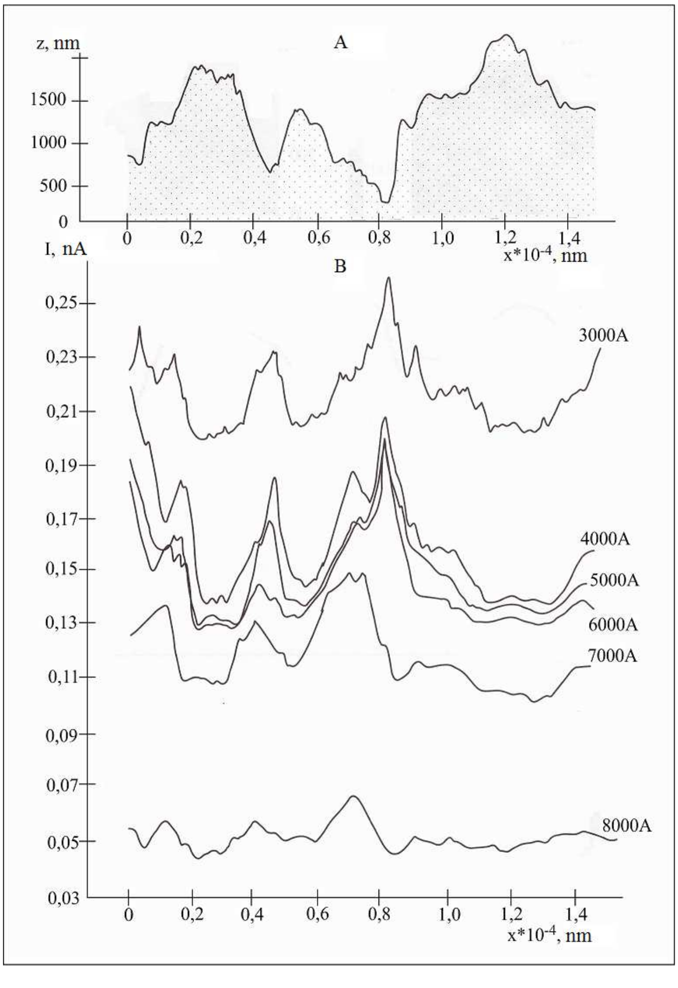

Figure 8. Change of the electrical micro-relief from different distances (H) of cantilever to AOF surface (profilograms): a) geometric micro-relief; b) electric relief.

Of course, modification of AOF surface should affect their electric relief and change the surface potential ψ. This is shown by EWF measurements at two Al-caps in base and modified state (Table I).

Table I shows that the surface of Al-caps without AOF weakly changes their structure because ϕ values are consistent with the reference data ϕ_{Si}. Also it weakly changes electronic properties as a result of its modification by water and KCl and $BaCl_2$ solutions, keeping EWF value close to the tabular one [13].

It looks differently for Al-cap surface having AOF. In this case, the surface significantly changes their electrical microstructure due to water presence or solutions exposure. Water absorption in coherent state reduces measured EWF value from $\phi_{Si} = 4,9 \div 5,0$ to $\sim$ 4,0÷4,4 eV [14] in both initial AOF and AOF modified by KCl. AOF modified by $BaCl_2$ has another effect: the value of EWF becomes equal to their reference value, i.e. increases by $\sim$ 0.6 eV. We explain this by collapse of water CD, absorbed on the surface modified by doubly-charged ion Ba^{2+}. This surface has an increased electrical activity and reduces dipole moment of sorbed water after CD collapse down to a normal value 1.83D [15].

2) Distant influence of Al-caps modified by $BaCl_2$ on permolecular structure of distilled water: The results shown on Fig. 2, 3 suggest that surface of Al-caps modified by $BaCl_2$, not contacting with distilled water (distance 5-7 mm), affects the evaporation rate, i.e. affects the permolecular structure of water

Indeed, Fig.2, 3 shows that dependences P=f(τ) obtained after distant influence on water of Al-caps modified by $BaCl_2$, are much closer to similar dependence for a 1M of $BaCl_2$ solution (the evaporation rate decreased significantly).

When we used Al-cap modified by KCl, as expected, the evaporation rate did not change. For KCl solution it was similar to the evaporation rate of distilled water. It must be emphasized that special reference chemical analyses (an ion Ba^{2+}) of distilled water in the crystallizer under the Al-cap modified by $BaCl_2$ did not detect traces of Ba. Consequently, $BaCl_2$ was absent in the water of crystallizer.

Thus, the above-discussed results make it possible to conclude that the presence of the Al-cap with oxidized surface modified by $BaCl_2$ at a distance of 5-7 mm from the water surface induces permolecular structure in water, similar to $BaCl_2$ solution. Apparently, this structure is an "informational copy" of $BaCl_2$ solution structure. Fig. 3 shows that the "copy" with increasing exposure time is relaxing and permolecular structure is returned to the characteristic of initial distilled water again. Reasons for decreasing the evaporation rate of $ZnCl_2$, $BaCl_2$ solutions were studied earlier [11].

The authors showed in [15] that during isothermal evaporation of water coherent domains which are being a metastable phase [4] and present in the water emit energy excess in the form of dipole-dipole radiation (Dicke super radiation) passing into the incoherent state. The radiation wavelength and power depend upon the CD size and concentration. Trying experimental determination of water radiation parameters has not given results yet. However, the indirect evidence of its presence noted in a number of works, in particular [16].

Thus, as a result of discussing experimental data obtained in step 1 (model experiment) we can conclude:

- there are structural long-range forces between the advanced AOF surface of the solid body (the “Al-cap” modified by $BaCl_2$) and the surface of distilled water, leading to the formation of “informational copy” of solid surface in water;
- apparently, the “informational copy” appears as a result of interaction between electric and phonon microstructure of solid surface with a dipole-dipole coherent radiation occurring on water evaporation;
- “informational copy” of surface modified by $BaCl_2$ in permolecular water structure has a limited “lifetime” ($\sim$ 5 days) as it returns to the permolecular structure, intrinsic for distilled water, due to the relaxation process.

B. Stage II. Changing the permolecular structure of water as a result of medicinal substances, grafted on CDR surface

1) Structure of the CDR surface: We describe here data from Wikipedia, due to the lack of CDR samples at different stages of technology for research.

It is known that CDR production is a complex multistep technological process. The result of it is a solid body having a multilayer structure and containing different materials. The fact that CDR is a solid body having an advanced surface (dispersion system) is important for the problem discussed in this work. This surface appears at the sixth stage of the technology by molding polystyrene under the pressure using prepared nickel die having a certain geometric relief. Solid body with an advanced surface arises as a result of this operation. Following three stages of technology, in fact, introduced nothing fundamentally new in CDR dispersed structure (metallizing, coating of protective layer and coating of label).

Thus, the work (informational) layer of disc has a certain geometric micro-relief in the form of a spiral track covering the entire CDR surface. It consists of pits extruded in a polycarbonate base. Each pit has depth of $\sim$ 100 nm and width of $\sim$ 500 nm. Pit length ranges from 850 nm to 3.5 microns. The pitch of spiral is $\sim$ 1.6 microns (Fig. 9).

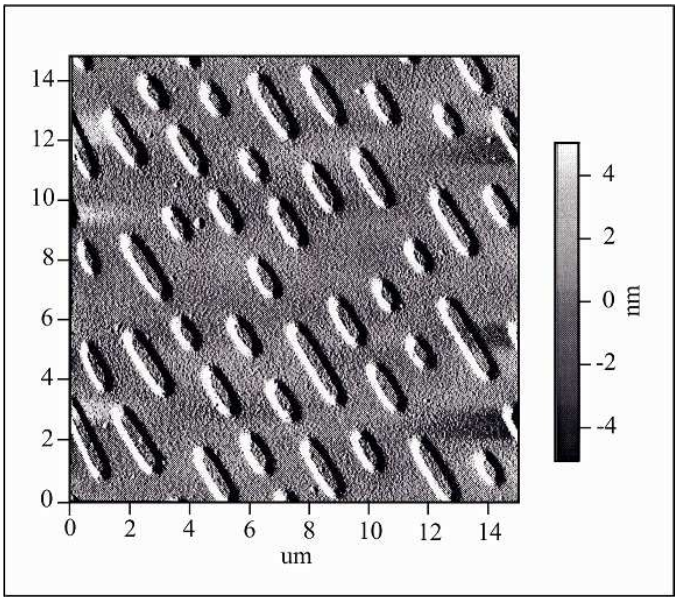

Figure 9. Microstructure of informational CDR layer.

If we make a rough estimate for the average characteristic size of the dispersion medium from this data (which is common practice [17]) it will be about 500 nm (0.5 microns). As shown above, we shall note that average characteristic size of AOF on Al estimated at $\sim$ 200 nm. Thus, values of the specific surface of Al-caps and CDR are comparable. Consequently, above-discussed physical mechanism of transmission to water of information of the structure of chemical compound grafted on surface ($BaCl_2$) works in case of CDR with the “grafted” medicinal substance too. Method of “grafting” can be different: the Al-cap in the model experiment was placed in the 1M solution of $BaCl_2$. In case of CDR the “grafting” is going on by laser scanning of medicine distributed over the CDR surface. Modification of electrical and phonon micro-relief dispersed surface occurs in both cases (Table I).

Fig. 10 shows data about kinetic evaporation of distilled water after its exposure under the surface of three CDR samples in base (clean) state.

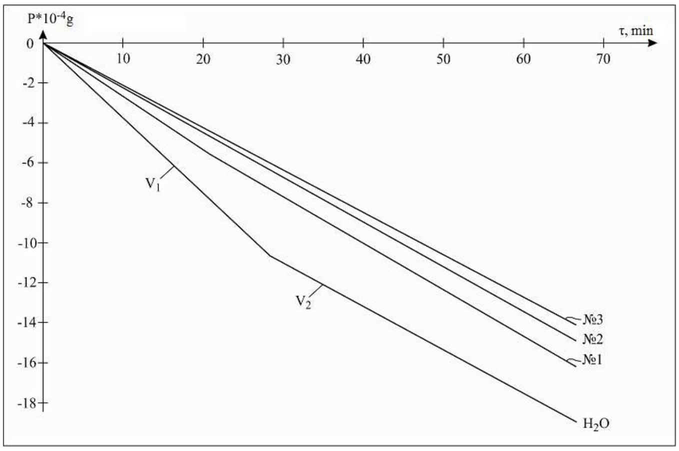

Figure 10. Kinetic curves of evaporation for distilled water affected by CDR in base (clean) state. H_2O is the distilled water without CDR-cap.

Curves P=f(τ) of distilled water without cap and a similar curve for distilled water under an unmodified Al-cap are shown for comparison. It follows from the data that CDR caps affect evaporation kinetics of water at a distance of $\sim$ 5-7 mm. Curves of three randomly selected disks are somewhat different and differ greatly from P=f(τ) for the Al-caps. Apparently, it indicates that specific surface of CDR dispersed phase exceeds the AOF surface on the Al-caps.

Thus, the results given in Fig. 10 clearly indicate the existence of structural long-range action effect for standard CDR. Based on our studies [15] we may conclude that structural forces, arising from CDR, contribute to the destruction of water domain structure (CD) and make it similar to the homogeneous structure of "molecular" water. Evaporation rates $\frac{dP}{d\tau}$ for base CDR-caps are close to rate V_2 of incoherent distilled water (see Table III). It should be noted that a thick polycarbonate layer ($\sim$ 1 mm), which protects an information dispersed CDR layer, does not shield action of structural forces.

Table III
Effect of the CDR-caps at the water evaporation rate.

NN	$V \cdot 10^{-6}$ g/s	Type of cap
1	0,40	CD-R, clear
2	0,36	- // -
3	0,35	- // -
Avg.	0,37	- // -
4	0,45	Al with AOF
5	0,47	- // -

2) Distant effect of CDR-caps modified by medicine substances to the water evaporation kinetics: As mentioned above, the methodology of research in this part does not differ from that described for stage I. In this case, the role of "cap" was performed by a CDR with "grafted" (by laser scanning) electromagnetic and phonon micro-relief of medicinal substances.

The results of these studies are shown in Fig. 11 as dependencies P=f(τ), where evaporation kinetics was compared in each case with base CDR-cap and water without cap.

Fig. 11 and Table IV shows the results, obtained for three medicinal substances: arbidol, fasamax and tyrosine.

Table IV
Evaporation characteristics of "medicinal" water.

NN	Medicine name grafted on CDR	$V_{1500} \cdot 10^{-6}$ g/s	$V_{\text{ph}} \cdot 10^{-6}$ g/s	$P_{\text{cdph}} \cdot 10^{-4}$ g
1	Arbidol	0,35	-0,003	3,9
2	Fasamax	0,45	0,10	15,0
3	Tyrosine	0,55	0,20	12,0

Assessing this information, we can point out that the main difference between the water evaporation kinetics is observed for any grafted medicine at the first period of exposure time τ_1 (up to $\sim$ 1500 s). Evaporation rates V with τ from 1500 to 4000 seconds in all cases are similar and equal $\sim (0,3 - 0,4) \cdot 10^{-6}$ g/s which corresponds to incoherent water. It follows that for comparison of different medicine we can introduce the common characteristic. It is the evaporation rate during the observation τ=1500 s. If the CDR (clean) cap leads to CD destruction in distilled water, the evaporation rate of medicinal (pharmaceutical) water is $V_{\text{ph}} = V_{1500} - (0,35 \div 0,40)$ g/s and the amount of coherent phases is $P_{\text{cdph}} = 1500 \cdot V_{\text{ph}}$.

Table IV shows that arbidol are not affect to the water structure, while the other two information copies are significantly increased proportion of CD in water.

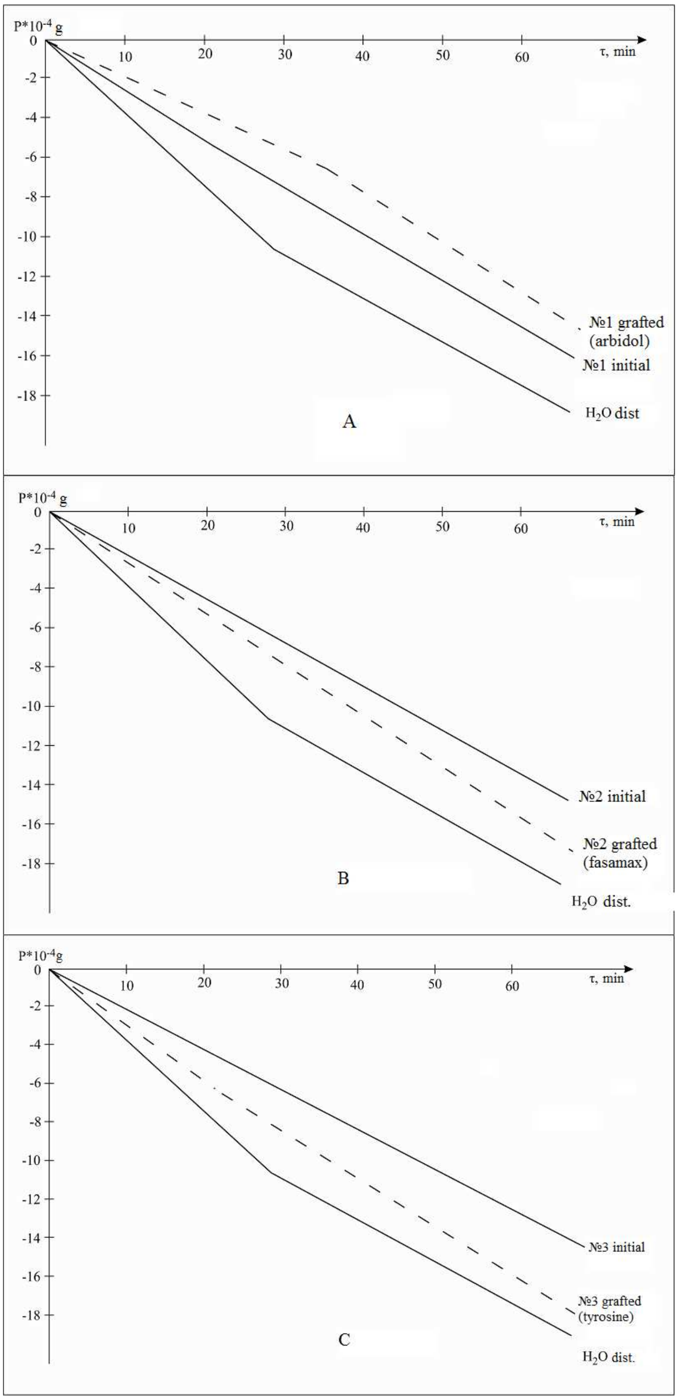

Figure 11. Kinetic curves of evaporation for distilled water impacted by CDR-caps modified by medicinal substances: A) Arbidol; B) Fasamax; C) Tyrosine.

IV. Conclusions

The results obtained through comprehensive study of the transmission of medicinal substances properties from CDR to liquid water allows us to give some conclusions about the physical mechanism of this process.

1) A necessary condition for providing the opportunity of transfer medicine properties to intermediate carrier (oxidized Al-wafer or CDR) is the presence of dispersed phase in last system (AOF at Al or relief

at information layer of CDR). The nature of carrier material, apparently, does not matter.

2) It was shown that the dispersed phase has an electric relief, i.e. the electromagnetic field acting at a distance (this effect has been observed in experiments with AOF (Fig. 8) at a distance of ∼ 1 micron). These results show that the surface field effect of solid body is detected at a distance of 0.5-0.7 cm.
3) Transmission of information about substances properties grafted on dispersed surface of intermediate carrier (by treatment in solution of $BaCl_2$ or laser scanning of medicine to CDR) occurs by “grafting” the surface compounds [18] due to changing electromagnetic relief of the carrier surface.
4) Transmission of information about grafted substance properties from a carrier to water, apparently, occurs due to interaction of the phonon spectrum (electromagnetic micro-relief) of the surface and dipole-dipole radiation (Dicke super radiation [19]), arising from the decay of CD evaporating water. The result of this interaction is the “informational copy” of grafted substance fixed by permolecular water structure (see the phase boundary scheme, Fig. 12). It should be pointed out that the long-range action according to the proposed scheme (distance between interfaces) is determined by water radiation parameters and can be very significant.

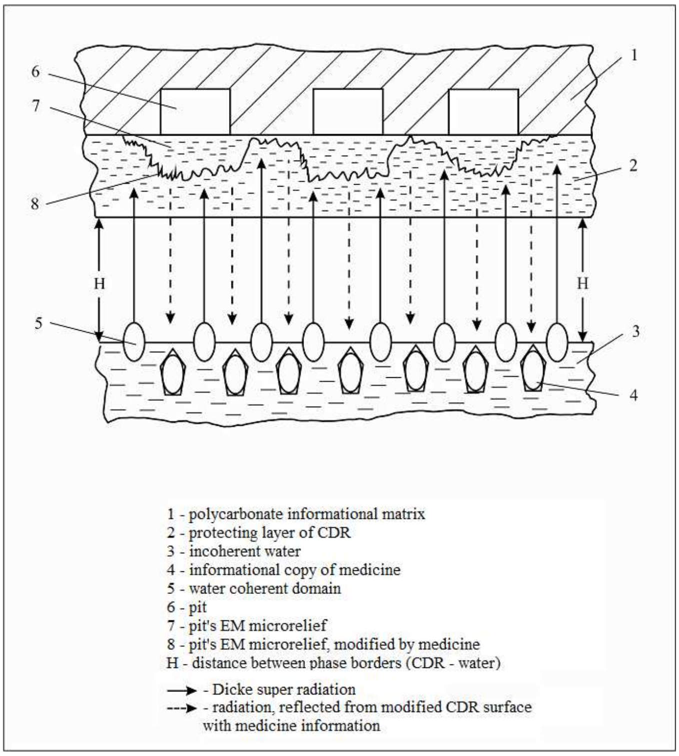

Figure 12. Proposed scheme of the process for formation “ informational copy” of medicine in water.

5) “Informational copy” which emerged in water (has a “medicinal properties”), has a limited “lifetime” (from 1 to 10 days); during long observation times water returns to the base permolecular structure. However, for example, in the case of mineral water (Essentuki №17) it was discovered that in solution of complex composition there may occur chemical processes that significantly affect permolecular water structure, that has a fundamental importance and requires further study.

Given the experimental results of this work, by the authors opinion, allows to discuss the proposed mechanism for transmission of information through the water and identify ways of further experimental investigations of such phenomenon important for practical usage.

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