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THE FORMING MECHANISM OF CLUSTER STRUCTURE OF WATER

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Abstract. The influence of phase changes on structural parameters of water was researched by method of laser emission scattering depending on conditions of their implementation in order to determine the way it forms.

The formation of stable water clusters under its boiling and phase change water-vapour-water was defined. It is assumed that the required for their formation change of bivalent oxygen atoms of free water molecules into tetravalent state is provided by released repulsion energy while oxygen atoms of neighbouring molecules approach each other.

The obtained findings made it possible to draw a conclusion that forming of stable clusters was stipulated by mutual action of torsional and electrostatic factors. Torsional attraction provided spatial grouping of water molecules with unidirectional spins of cluster oxygen atoms causing hydrogen bonds to link them into hexagonal rings (H₂O)₆ and further forming clusters from them. Torsional attraction of cluster oxygen atoms ensures temperature and time stability of hydrogen bonds as well as internal stability of construction in clusters.

Under boiling and phase change water-vapour-water its structure alters due to the ratios of speeds in binding clusters among themselves as a result of inter-cluster torsional interaction and generation of fine clusters because of influence of cluster torsional fields on oxygen atoms spins of free molecules. In room temperature the change of water structure was determined mostly by inter-cluster torsional attraction.

The process of natural water structuring is determined by the similar mechanism.

The influence of water defrosting speed on the process of cluster forming in it after phase change water-ice-water is discovered. Strength integrity of large-scale clusters after defrosting is provided by

torsional attraction of cluster oxygen atoms. The conclusion is drawn that long-term water memory which is carried by a spin configuration of oxygen atoms in large-scale clusters is not destroyed by phase change water-ice-water.

Keywords: water structure, a cluster, torsional interaction, hydrogen bond, phase change.

Introduction. The research of water structure has a long history. [1-3] However, significant results in determining the true state of the aspect of water properties have not been achieved upon the recent time. Up to the late 20 century methods of X-ray structural analysis were mostly used as main sources of information about water structure that are optimal to study the structure of crystalline solids with rigid fixation of atomic site in a lattice (without considering the thermal oscillations of atoms) the space spreading of which is characterized by both a short-range and long-range order.

The absent long-range order in liquid phase of water sets limits on X-ray diffraction methods by fixing only the shortest order in atomic spreading of oxygen and hydrogen in the liquid water phase. This fact let experimentally determine the existing inhomogeneous density of the liquid phase 1-2 μm in size [2], which is equivalent to clusters consisting of several water molecules interconnected by hydrogen bonds with lifetimes of 10-12 sec order. The latter ones continuously collapse and recover which corresponds to the pattern of instantaneously varying configurations of these clusters spreading.

Numerous theoretical models of water structure based on experimental findings from X-ray methods didn't bring actually anything essentially new in understanding of water structuring.[4]

The absent advance of water structure parameters beyond the dimensional nanometric and time picosecond frames for a long time persuaded many researchers that those frames in particular reflected the true pattern of the structure and made skeptics deny the water structure in general [5], not to say anything about its memory.

The progress was noted in the case when other mostly optic methods of water structure research were used. About last two decades brought into experimental works giving evidence about possible forming and existence of stable large-scale clusters in the liquid water phase.

In the works [7,8] the existence of stable water clusters of 0,5-1 μm was determined by methods of fluidal chromatography and protonic magnetic resonance with phase-contrast microscope.[9] According to [8-10] their forming was provided by hydrogen bonds and remote Coulomb forces (a charge complimentary bond). It is assumed [10] that the discovered structure can perceive and store information.

Measuring of small-angle scattering of laser emission by water located the existence of giant heterogeneous clusters (GHCs) with average sizes of 1-100 μm and lifetimes more than 10 sec. [11, 12] These clusters exist because various charged particles OD^- , H^+ , OH^- , H_3O^+ , isotons of oxygen and hydrogen, molecules HOD that are present in the test sample take part in their forming. It is assumed that

some from the pointed components of the initial water sample can be “nucleating seeds” in growth centers of GHCs around which nanoclusters group forming fine and medium clusters of $\sim(1-4)$ μm sizes in various “foreign” content due to hydrogen bonds and ensemble of H_3O^+ and OH^- ions. The following binding of the clusters to GHCs can be explained by disperse interaction.[12]

The method of frosting water and photographic recording of ice microcrystals in the field of a microscope after mental and emotional influence on the initial liquid water made it possible to discover the dependency of their geometry on the character and degree of influence.[13] The form of ice microcrystals indicates the forming of stable clusters in the initial water sample. According to the author the forming of the observed clusters is determined by influence of wave field of unknown nature. [9,13]

The method of laser mission scattering (laser nephelometry) adapted to measure light scattering of water allowed to discover the constant existing in water clusters of the sizes ranging from tenths up to one and a half and two dozens of microns, life times of which can be limited only by external factors causing structural changes. It is experimentally determined [14-16] that the property of water to respond to external influence by changing their parameters (ensemble, sizes, form and concentrations) is connected with these clusters in particular.

Discrepancies in structural parameters given in various sources can be connected to various informative opportunities of the used research methods as well as to incapacity to interpret experimental findings within generally accepted perception about the mechanism to form the cluster structure by means of hydrogen bonds and Van der Waals forces.

The latter fact generates the number of questions as to the part of hydrogen bonds and other types of intermolecular interactions in forming the cluster water structure as well as to the mechanism of the process in a whole.

In the first place it is not stated in which way picosecond hydrogen bonds in nanoclusters transform into long-lasting ones under forming of large-scale stable clusters observed in [9-12].

Besides close-range (length) of a hydrogen bond α ($\alpha \approx 1,8\text{\AA}$ [17]) and especially of that of a disperse one (attraction energy $U \sim \alpha^{-6}$ [18]) is a factor that practically excludes forming of large-scale stable clusters due to this type of interactions in water. The speculations on the topic of a charge complementary [10] and disperse [12] bond are suppositions that have none experimental proof.

Findings obtained by the light scattering method suggest presence of an additional factor of non-electrostatic nature that provides forming of cluster water structure.

The given work studies the influence of phase changes on structural water parameters depending on conditions of their implementation in order to determine the mechanism of forming of its structure by method of light scattering.

Materials and methods. The study of phase changes of water-vapour-water and water-ice-water on water structure were conducted by measuring dependencies in scattering intensity of laser emission I from scattering angle Q (scattering indicatrices $I(Q)$) of initial water samples $I(Q)_{int}$ immediately after the phase change took place and in approximately 90 minutes upon its cease $I(Q)_{inf}$. Upon indicatrices being measured and their computational and graphic processing being done the ensemble and sizes of clusters, their relative concentrations as well as the degree of change in those parameters upon the influence and following exposition were determined. The methods of determining an ensemble and sizes of clusters are given in [19].

The degree of change in total concentration of clusters $N_{\Sigma} = \sum N_i$ (N_i – concentration of clusters of i type (size) was determined in relevance to the integral intensity of scattered light upon the influence I_{Σ}^{inf} to the initial one I_{Σ}^{int} .

$$N_{\Sigma}^{rel} \approx I_{\Sigma}^{rel} = I_{\Sigma}^{inf} (I_{\Sigma}^{int})^{-1} \cdot 100\%.$$

According to [19] in approximation of single scattering value I_{Σ} is numerically equal to the square under the curve $I(Q)$. The veracity of relation $N_{\Sigma}^{rel} \approx I_{\Sigma}^{rel}$ under this condition is proved in [19].

The degree of change in cluster concentration of certain sizes was determined on computational values of relative scattering indicatrices $R(Q)$ in every case of influence which are the relation of the measured scattering indicatrices of one and the same water sample after and before the influence:

$$R(Q) = I(Q)_{inf} \cdot (I(Q)_{int})^{-1}.$$

The use of $R(Q)$ allows to simultaneously determine direction and degree of change in cluster concentration of various sizes in the result of influence based on its numerical value. Thus, the value $R(Q) > 1$ identifies increase and $R(Q) < 1$ implies decrease in concentration of clusters forming scattering intensity in the corresponding angle sector. Additional data on informative properties $R(Q)$ are given in [19].

Drinking water from an artesian well was used for the study. The influence of boiling and phase change of water-vapour-water on its structure was researched on samples with various initial concentrations of fine clusters.

Duration of condensed vapour accumulation with the volume of $V=5 \text{ cm}^3$ sufficient to be measured was 4-5 minutes. It took about 5 minutes to cool it down up to room temperature $\sim 20^{\circ}\text{C}$. In the whole the

measurement of dependencies $I(Q)^{inf}$ was made in 9-10 minutes upon the commencement of the phase change.

The influence from phase change of water-ice-water was studied under various speeds of defrosting of the tested water samples which were previously refrigerated under $T=-18^{\circ}\text{C}$ in a freezing compartment where cuvettes with water were kept for ~ 90 minutes. Additional information on the conditions of the experiment is given in the part *Experimental Results*

The results of researches.

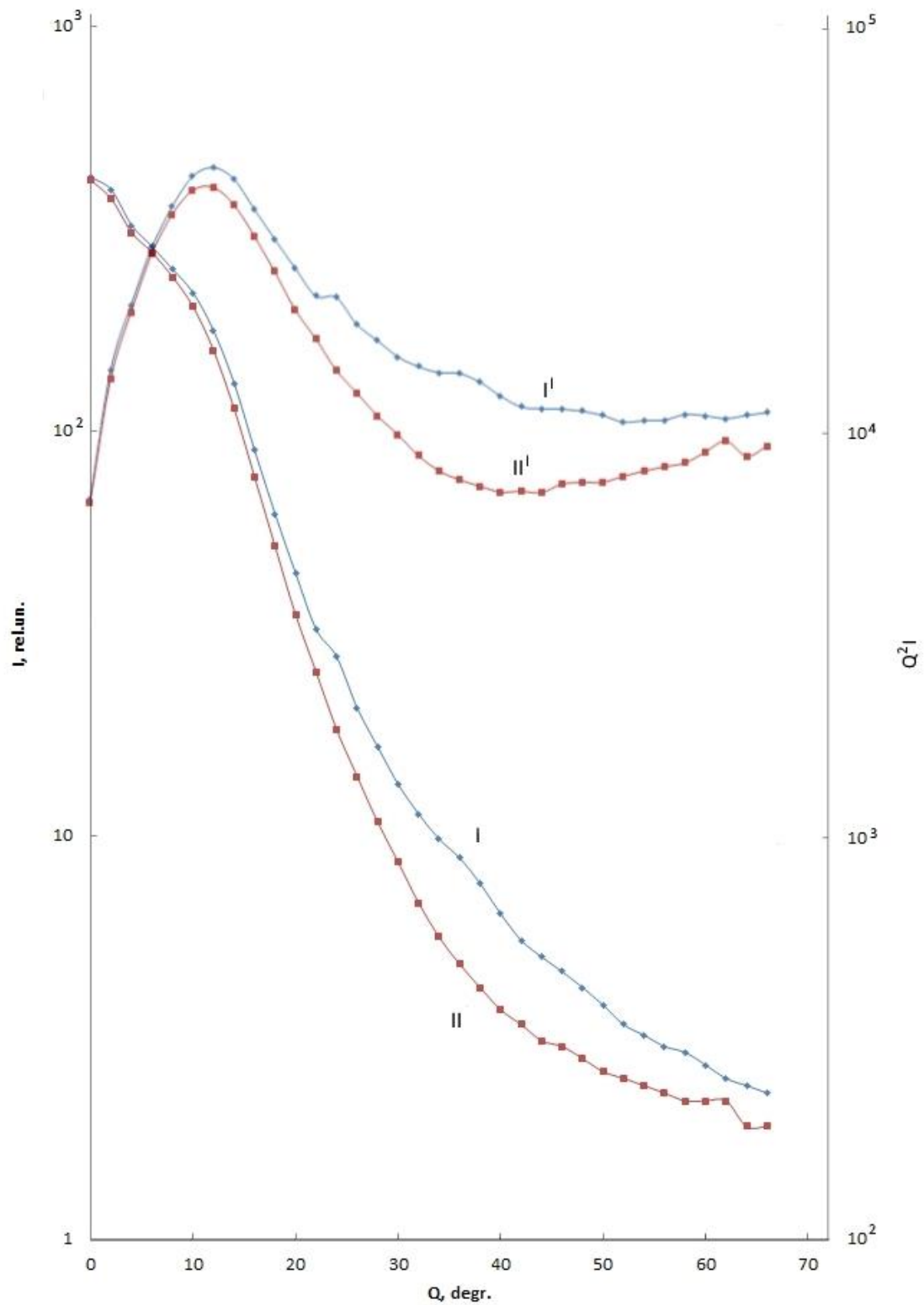
1. Scattering Indicatrices of Initial Water.

Scattering indicatrices of initial water are decreasing functions with increase in the scattering angle (pic.1). According to [14], scattering centers are clusters-ice microcrystals present in the liquid phase all temperature interval long throughout its existence. The observed character of curves $I(Q)$ in a small-angle plane ($Q \leq 10^{\circ}$) is determined by diffraction of the falling light on (conditionally) large-scale (further- large) clusters with radii in the interval $0,9 \mu\text{m} < r \leq 2,0 \mu\text{m}$; in the angle sector $10^{\circ} < Q \leq 30^{\circ}$ it is determined by the diffraction on medium-scale (further medium) clusters ($0,4 \mu\text{m} \leq r \leq 0,9 \mu\text{m}$); in the interval of angles $Q > 30^{\circ}$ the one is defined by scattering on fine-scale (further fine) clusters with $r < 0,4 \mu\text{m}$ partially due to diffraction as well as to reflection the significance of which grows with increase in Q . [20]

Insignificant fluctuations of intensity present on curves I and II as well as the corresponding ones on computational dependencies $Q^2 I(Q) = f(Q)$ (pic.1, I' and II') in the angle interval $Q > 25^{\circ}$ are connected to interference of reflected from the oversize clusters with $r > 2 \mu\text{m}$ which are present in small concentration in initial water and refracted by them rays. [16]

During the experiment the decrease in concentration of fine clusters took place in tested samples which was followed by increase in size of oversize clusters. This kind of change of structural parameters in initial water is connected with its natural structuring in the process of storage which implies binding of fine clusters with large-scale ones.

Conditional classification of tested water into two degrees of structuring was made in order to determine the influence of initial structure on character of influence from water-vapour-water phase change. Initial structure of I degree was characterized by higher concentration of fine clusters and smaller sizes of oversize clusters compared to the ones of structure of II degree. Shown on pic.1 scattering indicatrices scattering indicatrices of initial samples were numbered in decreasing order of fine clusters concentration. Structural parameters of these water samples are given in tables 1 and 2.



Pic.1 Scattering indicatrices $I(Q)$ of initial Water of the first (I) and second (II) structural degrees and corresponding to them calculation functions $Q^2 I(Q) = f(Q)(I', II')$.

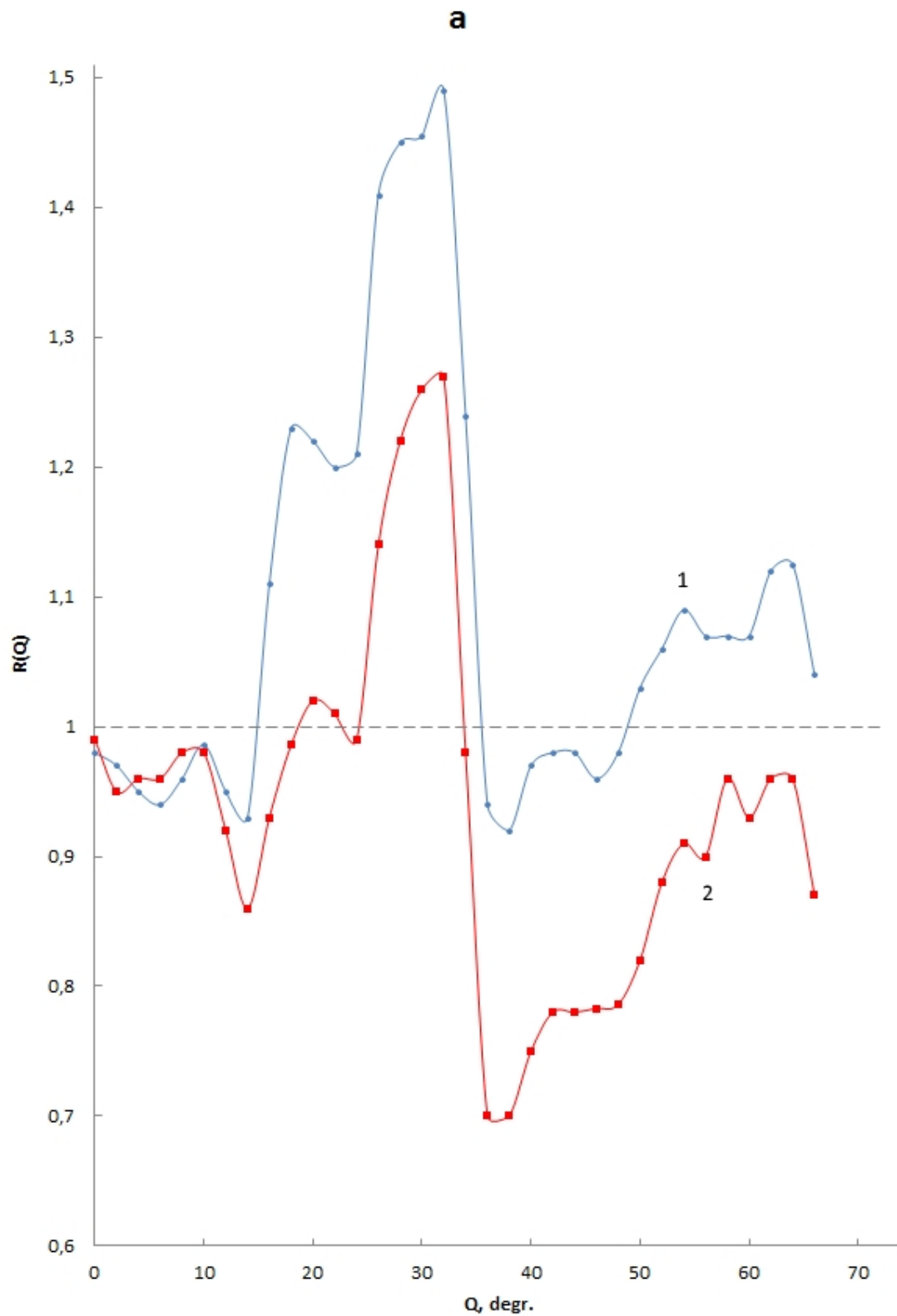
2. Influence of Boiling on Water Structure

Pictures 2.a and 3.a (curves 1) show relative scattering indicatrices $R(Q)$ of water samples of I and II degrees measured immediately after the cease of boiling within 10 minutes and cooling down up to room temperature. Pictures imply that boiling without phase change lead to the similar changes of structural parameters in samples of both degrees the main of which lie in the following:

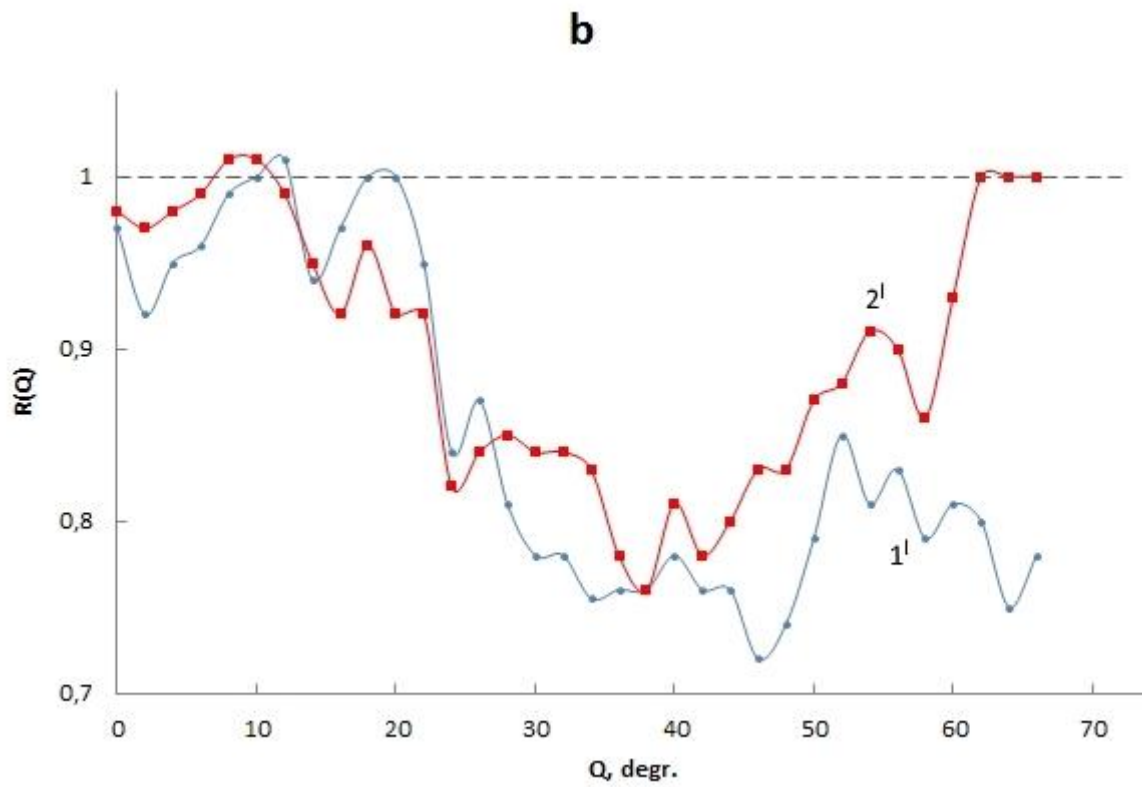
1. Values $R > 1$ in angle intervals of scattering $Q > 20^\circ$ and $Q > 30^\circ$ for samples of I and II degree correspondingly indicate significant increase in concentration of fine and medium clusters with sizes in interval $0,24\mu\text{m} < r < 0,47\mu\text{m}$ in the sample of I degree (table 1) and the one in concentration of fine clusters with sizes in interval $0,19\mu\text{m} < r < 0,26\mu\text{m}$ in the sample of II degree (table2).
2. Values $R < 1$ in angle intervals $Q < 20^\circ$ and $Q < 30^\circ$ for the same samples attest to insignificant decrease in concentration of medium and large clusters with sizes $0,48\mu\text{m} < r \leq 1,58\mu\text{m}$ and $0,34\mu\text{m} < r \leq 1,58\mu\text{m}$ correspondingly.
3. Presence and character of interferential component of curves 1 in pictures 2.a and 3.a indicate increase in sizes and concentrations of oversize clusters¹ (tables 1 and 2).
4. The changes in concentration of clusters of various sizes occurred while boiling determined growth of total concentration of N_{Σ}^{inf} clusters in water samples with various structural degrees (tables 1 and 2).

The following exposition led to various structural changes in samples of I and II degrees. The sample of I degree showed decrease in total concentration of N_{Σ}^{inf} clusters caused by decrease in concentration of large, medium and to the most degree fine clusters. Shifting of curve 2 on pic.2.a downward on vertical axis attests to it. The character of interferential component of the curve indicates increase in sizes and concentrations of oversize clusters within the exposition (table 1).

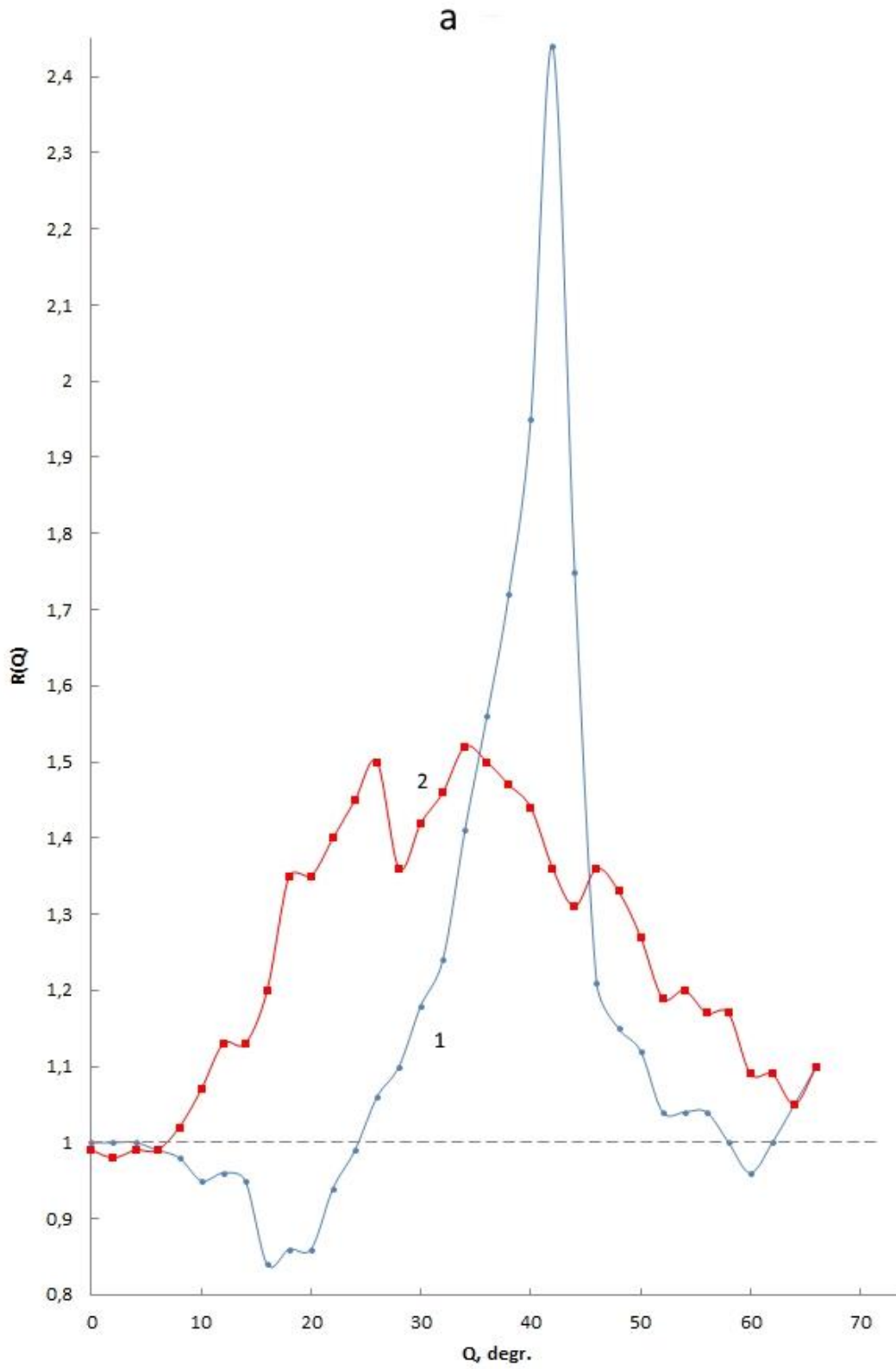
1) Present on curve 1 pic.3.a extreme interferential maximum of zero order attests to intensive forming of oversize clusters with $r \approx 4,1\mu\text{m}$. Periodic implicit maxima on its decay in angle sector $20^\circ \leq Q \leq 40^\circ$ are connected to overlapping of another interferential component determined by scattering on larger clusters with $r \approx 6,7\mu\text{m}$.



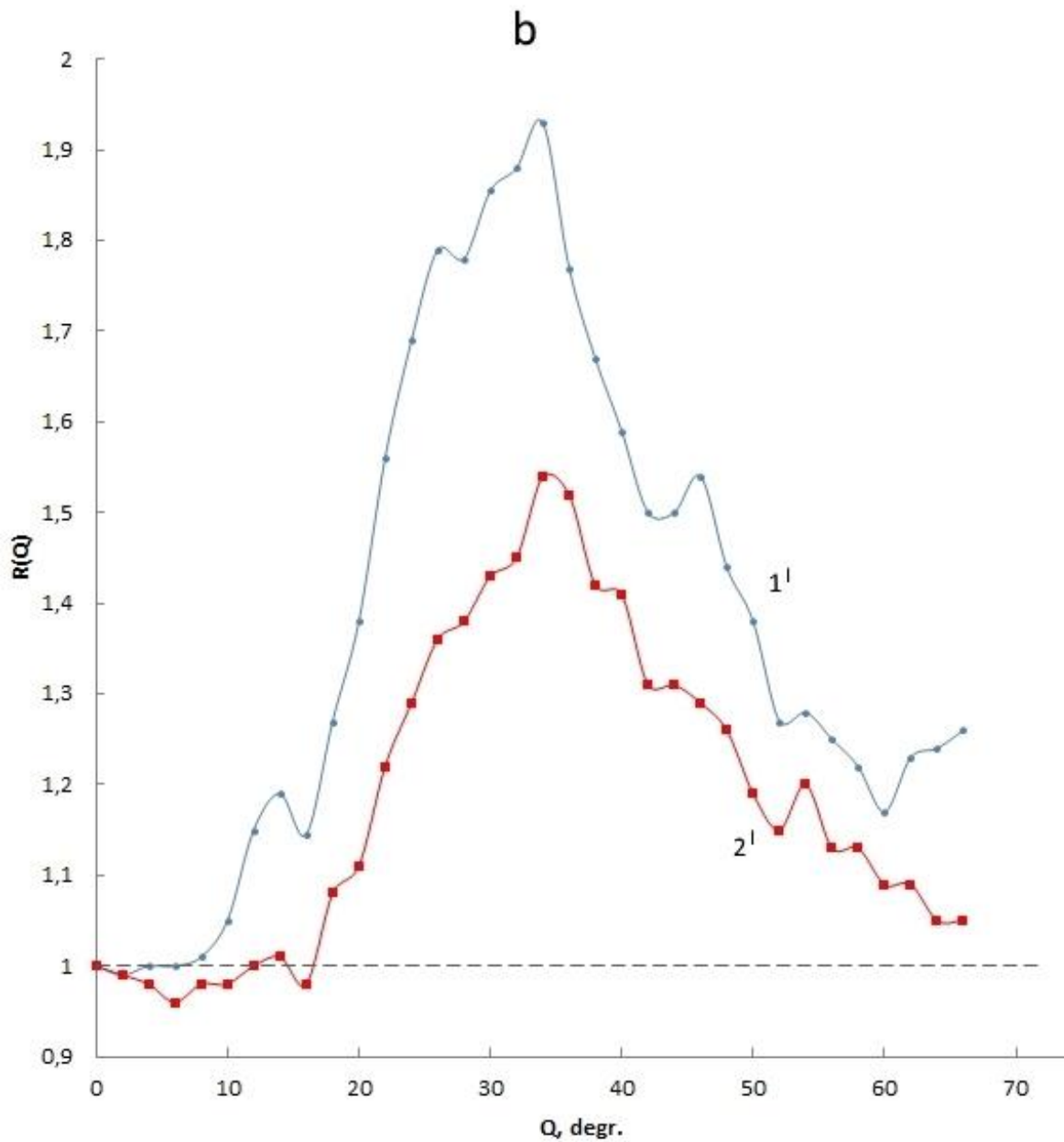
Pic.2,a. Relative scattering indicatrices of water sample of I structural degree after boiling (1) and the exposition (2)



Pic.2,b. Relative scattering indicatrices of condensed water sample of I degree after the phase change (1') and its exposition (2') .



Pic.3,a. Relative scattering indicatrices of water sample of II structural degree after boiling (1) and the exposition (2).



Pic.3,b. Relative scattering indicatrices of condensed water sample of II degree after the phase change ($1'$) and its exposition ($2'$)

Further increase in total concentration of N_{Σ}^{inf} clusters was typical for the sample of II degree due to significant generation of fine and medium clusters which is proved by values $R > 1$ in the angle interval $Q > 10^\circ$ as well as increase in sizes of oversize clusters (pic. 3.a. curve 2; table2).

3. Influence of Water-Vapour-Water Phase Change on Water Structure

Given on pic.2.b. curve 1' of the relative scattering indicatrix of condensed water of I degree sample immediately after cooling it down up to room indoor temperature implies that the most significant structural changes involved increase in sizes and concentration of oversize clusters compared to the observed ones after boiling, decrease in concentration of large, medium and to the most degree fine clusters. The corresponding interferential component of curve 1' and values $R < 1$ in the angle interval $Q > 20^\circ$ are indicative of these changes. The total concentration of N_{Σ}^{inf} clusters decreased according to the observed ones both in the sample being boiled and in the initial one (table 1).

Following exposition caused insignificant increase in total concentration of clusters due to generation of fine clusters which is proved by growth in R values with increase of scattering angle in interval $Q > 45^\circ$ (pic.2.b, curve 2'; table 1).

Immediately after the phase change in the condensed water of II degree sample significant increase in total concentration of clusters took place due to generation of medium and to the most degree fine clusters with $r \approx 0,26 \mu\text{m}$ which is proved by values of the relative scattering indicatrix $R \gg 1$ in angle interval $Q > 15^\circ$ (pic.3.b, curve 1').

As shown in pic 3.b, curve 2' the further exposition of condensed water of II degree sample determined decrease in concentration of medium and fine clusters due to their binding with oversize clusters. The character of interferential component of curve 2' and table 2 testify to the increase in sizes of the latter ones.

4. Influence of Water-Ice-Water Phase Change on Water Structure

Water corresponding to II structural degree was used in the experiment. The influence of this phase change on water structure depended on defrosting speed of the tested water. Table 3 shows structural parameters of water structure in various time periods after its defrosting in the streamlined way (the cuvette with frozen water was placed in the hand-locked palm).Picture 4 shows relative scattering indicatrices of sample A immediately after defrosting and increase of its temperature to the room one (curve 1) and after the exposition (curve 2).

As shown in table 3 sets and sizes of oversize and large clusters to a greater degree and the ones of medium clusters to a lesser degree coincided or were very close to the sizes of the tested sample in all its states. The peculiarity of *ice-water* phase change was increase in total concentration of clusters after defrosting and the further exposition compared to the initial one which is $N_{\Sigma}^{\text{int}} = 100\%$ (table 3). Values $R >$

1 of curves 1 and 2 on pic.4 in angle sector $25^{\circ} \leq Q \leq 65^{\circ}$ indicate increase in concentration of fine clusters with sizes $0,14 \mu \leq r \leq 0,38 \mu \text{m}$.

Increase in parameters of the interferential component of curve 2 (amplitudes, regularity property of fluctuations R and their angular resolution) observed after the exposition testifies to the increase in concentration of oversize clusters as the result of binding among fine clusters generated while the exposition.

Picture 4 shows relative scattering indicatrices of water sample B which was slowly defrosted by natural heating to room temperature (curve 3) and subjected to exposition in various periods of time (curves 4 and 5). Structural parameters of the sample in its diverse states are given in table 4.

As it follows from the obtained findings after defrosting sets and sizes of oversize and large clusters had not only significant similarity between themselves but also the one with the initial structure. Some changes of the parameters were observed in medium and fine clusters; however in most cases changes of their parameters did not exceed the error of measurements. [15]

The peculiarity of natural defrosting influence on water structure was no generation of fine and medium clusters. Values $R < 1$ of curves 3-5 in angle interval $Q > 12^{\circ}$ attest to decrease in their concentration compared to the initial ones. It may be caused by partial destruction of crystalline structure of ice to fragments with sizes less than the ones of medium and fine clusters of the initial structure.

Increase in time exposition determined gradual recovery in concentration of a finely divided structural fraction which is indicated by the upward shift of curve 5 on R axis and increase of N_{Σ}^{inf} (pic. 4, curves 5; table 4).

Parameters of water structure of I degree

#	State of water sample	t_{inf} , min	r , mm	I_{Σ} , rel.units	N_{Σ}^{rel} , %
1	Initial	0	2.2-2.8; 1.58; 0.95; 0.58; <0.4	17100	100
2	After boiling	10	(2.8-3.2)↑; 1.58↓; 0.95↓; 0.56↓; 0.43↑; (0.34-0.24)↑↑; ≤0,18↑	18710	109.4
3	Exposition	90	(5,5-6,5)↑; 1.57↓; 0.95↓; 0.53↓; 0.39↓; 0.28; <0.24↓;	16070	94
4	After evaporation and condensation	10	(4.5-6.5)↑; 1.58↓; 0.79; 0.53↓; 0.41; <0.36↓	14586	85
5	Exposition after condensation	90	4.5-6.0; 1.58↓; 0.73; 0.63; 0.43↓; <0.40↑	15390	90

Table 2.

Parameters of water structure of II degree

#	State of water sample	t_{inf} , min	r , mm	I_{Σ} , rel.units	N_{Σ}^{rel} , %
1	Initial	0	3.2-4.7; 1.58; 1.06; 0.59; 0.43; <0.40	15200	100
2	After boiling	10	(4.1-6.7) ↑; 1.58↓; 1.06; 0.58↓; 0.43↓; (0.26-0.19) ↑↑	17630	116
3	Exposition	90	5.3-7.2; 1.58; 0.95; 0.56↑; <0.47↑↑	18950	125
4	After evaporation and condensation	10	4.7-7.2; 1.58; 1.06; 0.53; <0.47↑↑	20976	138
5	Exposition after condensation	90	(5.3-7.4)↑; 1.58; 0.95↓; 0.56↓; <0.45↓	18040	119

Parameters of water structure after the phase change water-ice-water under accelerated defrosting

#	State of water sample	r, mm	N_{Σ}^{rel} , %
1	Initial	5.3-7.2; 1.58; 0.95-1.19; 0.59-0.70; 0.43; <0.40	100
2	After defrosting	5.3-6.7↓; 1.58; 0.95-1.19; (0.59-0.70)↑; 0.48↑; 0.32↑; <0.30↑	129
3	Exposition 90 minutes	4.7-6.7↑; 1.58; 0.95; 0.58-0.73; 0.43; 0.32; <0.30	132

Table 4.

Parameters of water structure after the phase change water-ice-water under natural defrosting

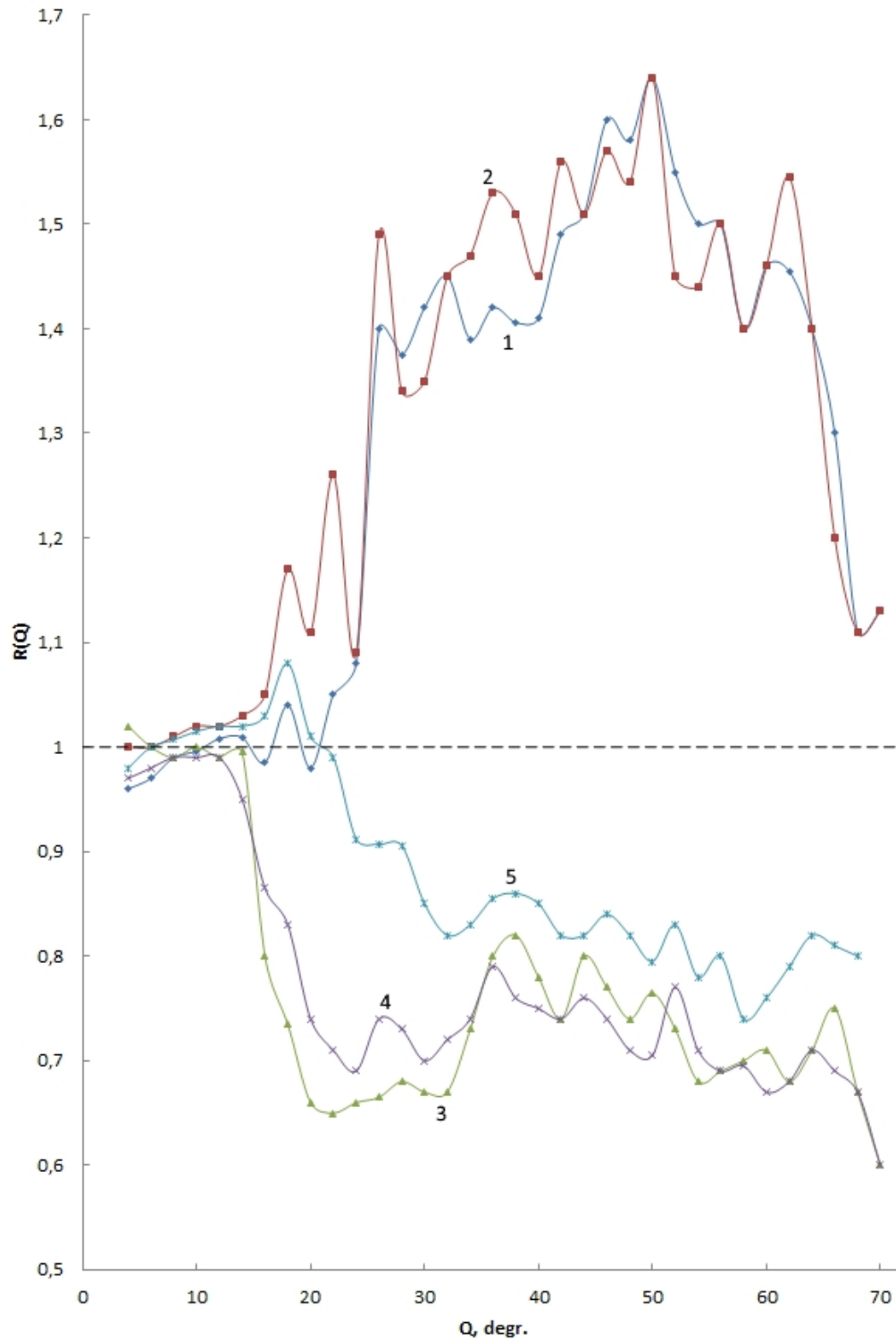
#	State of water sample	r, mm	N_{Σ}^{rel} , %
1	Initial	4.7-7.2; 3.5; 1.58; 0.95; 0.61; 0.32; <0.30	100
2	After defrosting	4.7-7.2; 1.60; 0.95; 0.68; 0.26; <0.26	76.4
3	Exposition 90 minutes	4.7-6.7; 3.5; 1.58; 0.79; 0.63; 0.32; <0.30	77
4	Exposition 14,5 hours	4.5; 7.2↓; 3.5↓; 1.58; 0.80↑; 0.61-0.53↑; 0.32↑; <0.30↑	87.5

Notes to tables 1-4:

1. Arrows at values of cluster radii indicate decrease (↓) or increase (moderate (↑), significant (↑↑)) in their concentration relative to the value of the previous state of the sample.

2. Table column N_{Σ}^{rel} contains values of total concentration of clusters in reference to the parameter of the initial sample the value of which was considered as 100% in each experiment to make it more comfortable to compare the character and degree of influence in phase change in relation to the initial structure and experimental conditions.

3. Symbol (<) at radii of fine clusters indicates fine clusters present in the set that have radii less than the ones given in the tables. Significant overlapping in “directional diagrams” of light scattered by fine clusters of various sizes doesn’t allow determining of their specific values.



Pic.4 Relative scattering indicatrices of water sample of II degree after the phase change water-ice-water under accelerated (1;2) and natural (3-5) defrosting, immediately after it (1;3), after the exposition for 90 minute time(2;4), after the exposition for 14,5 hour time (5)

Discussion of the Results. The following perceptions about structural aspect of water are generally accepted:

1. Clusters form themselves due to hydrogen bonds only. [1,2, 17]
2. Small lifetimes of hydrogen bonds don't let stable clusters form.
3. Hydrogen bonds collapse when temperature rises, and in a vapour phase only free water molecules H₂O can exist. [21-24]²

Given in the present work experimental findings have the number of peculiarities which do not correspond to generally accepted understanding about the process of water structuring. They are as follows:

1. forming of clusters in boiling process contrary to the anticipated collapse of theirs;
2. generation effect under the phase change water-vapour-water which comprises increase in concentration and sizes of clusters compared to their initial values;
3. all registered in the experiment clusters of various sizes are stable;
4. indestructability of large-scale clusters in the initial structure under phase changes of water.

The peculiarities and other findings indicate that other factor than hydrogen bonds plays primary role in forming stable cluster water structure determining the observed changes. Below there is the background of veracity for the given statement, and the mechanism of forming water structure is considered.

According to [17] molecules of water oxygen atoms of which are tetravalent take part in forming cluster structure.

Transition of bivalent oxygen atoms of free water molecules into tetravalent state under phase change water-ice in free enthalpy equilibrium occurs due to evolved by water warmth of crystallization which provides excitation in one of 2p²-electrons of each oxygen atom into 3s-state and practically instantaneous forming of monolithic ice structure.

The mechanism of transition for bivalent oxygen atoms of water molecules not bound in clusters into tetravalent state while boiling is as follows below. Thermal energy brought to water caused intensive movement in its molecules. The movement which is not directed and comprises not only progressive but also rovibrational components provides multiple short-term approaches of oxygen atoms of neighboring molecules. Under significant approach, when distance between their nuclei $l \rightarrow 0$ because of weak electronic screening of nuclear fields, repulsive force occur between them. [18] The evolved hereby energy $U \sim \exp(-l/r_1)$ where $r_1 = 0,53 \text{ \AA}$ is radius of the first Bohr orbit is sufficient for the transition of one of 2p²-electrons in some part of oxygen atoms into 3s-state.

² According to [25] pairwise (dimer) groups (H₂O)₂=H₂O...HOH form in water vapours under not very high temperatures.

As a result the oxygen atoms transit into sp^3 hybrid state forming 4 equal in form and energy sp^3 hybrid orbitals with unpaired electrons. Two of them are saturated, covalent, providing bonds of oxygen atom with two hydrogen atoms and the other two are unsaturated, coulomb - hydrogen ones providing intermolecular bonds. [17]

According to [17] water molecules with tetravalent oxygen atoms in liquid phase can exist only linked by hydrogen bonds as a component of hexagonal fragments of ice $(H_2O)_6$ which are *elementary cells* of clusters with the following binding of them into clusters by similar bonds.

However hydrogen bonds alone with their temperature degradation as the only factor of forming not only *elementary cells* of clusters but also clusters themselves in extreme conditions of boiling seems practically unlikely due to this reason. There is rationale for considering other factor to be responsible for temperature and time stability as well as for potential generation of new clusters. More specifically it is torsional attraction of tetravalent oxygen atoms both as individual ones (immediately after their forming) and the ones comprised in hexagonal rings $(H_2O)_6$ and clusters. The nature of the torsional interaction is as follows.

Every of tetravalent oxygen atoms comprises 4 unpaired valence electrons with not compensated spins providing significant value of atomic spin³. Due to essential excess of torsional long-range action over coulomb ones [27] the property of similar torsional charges (objects having unidirectional spins) to attract each other [27] provides fast space grouping of water molecules with parallel spins of tetravalent oxygen atoms with their following binding into hexagonal rings due to hydrogen bonds. Strong mutual torsional attraction of oxygen atoms in every ring determines its inner sustainability and temperature stability.

Succeedent forming of clusters from hexagonal rings while boiling is established by mutual actions of torsional and coulomb factors.

Turbulent character in movement of hexagonal rings provides manifold inception of short term multiple space configurations of their collective “spins” considering the latter ones as a vector sum of spins of oxygen atoms in hexagonal rings with approximately one-way orientations. The fact causes mutual torsional attraction of rings with the similar spin orientation forming “protoclusters” from them (assemblage of rings linked torsionally but not yet by hydrogen bonds). When neighbouring rings approach enough for coulomb interaction to arise among some atoms of oxygen and hydrogen in water molecules of neighbour rings multiple (up to 12 bonds per a ring [17]) hydrogen bonds form among rings linking them into fine clusters.

³ A spin of an atom is a vector sum of spins and orbital moments of impulses from electrons and intrinsic moment of momentum from a nucleus. [26]

The following evolution of structure lies in enlargement of fine clusters for both initial water and one formed while boiling by their binding among themselves and with separate hexagonal rings according to the considered above algorithm. It should be noted that higher concentration of fine clusters in initial water of I degree provided their binding into larger clusters practically simultaneously with forming of new fine clusters.

It streamlined the process of restructuring in the whole compared to the water sample of II degree in which forming of fine clusters occurred only when hexagonal rings accumulated in certain concentration. It is proved by forming of main amount of fine clusters in sample of I degree with $r \approx 0,24 \mu\text{m} - 0,34 \mu\text{m}$ and in sample of II degree with $r \approx 0,19 - 0,26 \mu\text{m}$ when boiling (tables 1 and 2).

Peculiarities of changes in water structure of water samples of I and II degrees under exposition after boiling were determined by speeds of clusters binding among themselves and generation of new fine clusters. The dissimilarity is caused by influence of two factors. On the one hand unidirectionality of collective “spins” in hexagonal rings under their binding into a cluster implies proportionality in intensity of torsional field of the latter to number of rings included into its composition i.e. to the size of a cluster⁴. It follows that increase in sizes of clusters amplifies increase in torsional interaction among them which should speed up their binding.

On the other hand abatement of temperature factor decelerates the processes in forming of tetravalent oxygen atoms and hexagonal rings which should slow down the speed of forming of new fine clusters.

With this background experimental findings about sizes of generated clusters while boiling and structural parameters after boiling stops indicate that in sample of I degree binding speed of clusters into oversize ones exceeded the speed in forming of fine clusters while the exposition period.

Such ratio of speeds of two opposite directed processes in change of total concentration of clusters caused its decrease⁵.

⁴ The given supposition is proved by higher mechanical durability of large clusters compared to the one of fine clusters under influence on water changing its structure. [19]

⁵ By estimate carried on the assumption of a spherical form of clusters and their strong packing about $10^3 - 10^4$ of fine clusters in various sizes in interval $r \approx 0,2 - 0,4 \mu\text{m}$ are “spent” to form an oversize cluster with $r \approx 6 - 7 \mu\text{m}$. Since fine and medium clusters make main part of total concentration significant decrease in N_{Σ}^{exp} is the result of their binding (table 1).

In sample of II degree the ratio of speeds was opposite to the one in sample of I degree which determined increase in N_{Σ}^{exp} due to generation of fine clusters (table 2).

As it follows from the above said generation of fine clusters can occur only if concentration of molecules with tetravalent oxygen atoms increases. Providing abatement of temperature factor the change of valence of bivalent oxygen atoms in free water molecules is possible only due to their spin reorientation of fine clusters existing in the liquid phase by total torsional field⁶. It lies in turning of oxygen atoms of those molecules the spin direction of which doesn't coincide with the operative direction of torsional field to a certain angle. With this whenever each oxygen atom turns the entire molecule will turn because of strength of its covalent bonds with its hydrogen atoms. Such a turn can cause the approach of oxygen atoms in neighbouring molecules that determines repulsive forces among them and further change of a certain part in oxygen molecules into tetravalent state similar to the one while boiling.

It is assumed that spin reorientation can create distance between a proton of hydrogen atom of one molecule and an atom of oxygen in a neighbouring molecule which is shorter than a length of a hydrogen bond in equilibrium which provides its essentially bigger energy. The released energy in such a intermolecular bond can be enough to excite one of $2p^2$ electrons of oxygen atoms in some molecule into $3s$ state with their further binding into hexagonal rings and new clusters.

The peculiarity in spin reorientation of molecules is time extension because the angles of their turns differ. The peculiarity along with single-shot act of turning of free molecules essentially decelerates the speed in change of oxygen atoms into tetravalent state compared to the one while boiling.

Veracity of the mechanism in change of valence of oxygen atoms of free molecules in a liquid phase is confirmed by induction of generating effect under room temperature by after-action of informational influence on water observed in [28,29].

Present developed structure in samples of I and II degree under phase change water-vapour-water indicates that in a vapour phase clusters were not destroyed being thermally-stable due to inter-cluster torsional attraction. In a fact if there were complete or partial destruction present in the vapour phase, the decrease in ensemble or sizes and concentration of clusters would be observed immediately after the phase change accordingly.

Retaining of clusters ensembles in condensed water vapour after the phase change, increase in sizes and concentration of oversize clusters in sample of I degree, increase in concentration of fine and medium

⁶ Taking into consideration high mobility of fine clusters which is determined by their small sizes and mass, one can assume certain space correlation of their individual torsional fields.

clusters in sample of II degree which occurred while in the vapour phase indicate that processes of restructuring taking place in them while boiling continue at higher speeds though.

Their growth while the phase change is connected to the increase in effective torsional inter-cluster inter-ring and mixed (cluster-ring) interaction which amplifies mutual attraction of corresponding structural elements. It is assumed to be determined by two factors:

1. absent friction force of clusters and hexagonal rings with liquid phase because of its absence in the vapour phase and increase of their mobility;
2. present temperature factor which accelerates not only formation of short term unidirectionalities of torsional fields and rings along with factor 1 but also attraction of bivalent oxygen atoms in unbound water molecules which provides their change into tetravalent state.

Invariance or similarity in a set and sizes of clusters of large-scale disperse structural fraction before and after the change water-ice-water indicate that they did not collapse in the process of the entire change. While frosting into solid phase only liquid phase changed which consisted of unbound into clusters water molecules and clusters of the initial structure happened to be built into this monolithic crystalline phase while pertaining its individual form and inner construction.

Strength integrity of oversize and large clusters was preserved due to inner cluster torsional attraction.

Differences in processes of water restructuring after the phase change in sample A and B can be presumptively determined by various modes of defrosting and values of thermal energy employed for the process. In the former case the amount of energy applied to sample A evidently exceeded the value of melting heat $E_{\text{melt}} = 6 \text{ kJ/mol}$ required for the phase change ice-water [17]. Excessive energy provided quick collapse of monolithic crystalline structure of ice into fine disperse phase which is mixture of molecular ice structure and free water molecules. [17]

In the latter case the required amount of energy in sample B gradually accumulated and the process of solid ice destruction occurred due to its crushing into large scale blocks with their slow collapse into finer disperse phase.

Gradual recovery of fine disperse fraction after a long exposition (pic.4, curve 5, table 4) is connected to the influence of torsional field of oversize clusters retaining memory about former structure in their spin configuration on liquid phase which induced generation of fine and medium clusters

Conclusions

1. It is experimentally determined forming of stable water clusters while its boiling and the phase change water-vapour-water. Required for their forming change of bivalent oxygen atoms of free water molecules into tetravalent state in such conditions is provided by evolved energy of repulsion while atoms of oxygen in neighbouring molecules approach each other.

2. Forming of stable clusters is conditioned by mutual actions of torsional and electrostatic factors. Because of essentially bigger long-range action than the Coulomb one torsional attraction provides fast space grouping of water molecules with undirected spins of tetravalent oxygen atoms which causes their binding with hydrogen bonds into hexagonal rings $(\text{H}_2\text{O})_6$ and the following forming clusters out of them.

Torsional attraction of cluster oxygen atoms provides temperature and time stability of hydrogen bonds and inner sustainability in structure of clusters.

3. While boiling and phase change water-vapour-water the change of its structure is defined by ratio of speeds in two competing processes i.e. binding of clusters among themselves with forming of large-scale disperse structure and generation of fine clusters with formation of fine disperse structure.

The former process is determined by inter cluster torsional interaction and is dominant under increased concentration of fine clusters in the initial water. The latter process is connected to interaction influence of torsional fields of clusters on spins of oxygen atoms in free water molecules and prevails under low concentration of fine clusters in the initial water.

4. In room temperature abatement of the temperature factor essentially decelerates the speed in forming of fine clusters that's why structural change under the exposition is mostly determined by binding of clusters due to inter-cluster torsional interaction and decrease of their total concentration and forming of large scale disperse structure.

The similar mechanism determines the process of natural water structuring.

5. The influence of water defrosting speed on process of cluster forming in it after the phase change water-ice-water is discovered.

Strength integrity of large-scale clusters after defrosting is provided by torsional mutual attraction of cluster atoms of oxygen.

Long term water memory the carrier of which is spin configuration of oxygen atoms in large-scale clusters is not destroyed by the phase change water-ice-water.

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