

# **Introduction to the Biophysics of Activated Water**

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# 1. INTRODUCTION TO THE BIOPHYSICS OF ACTIVATED WATER

## 1.1 STRUCTURE AND PROPERTIES OF A WATER MOLECULE

Water is the foundation of life on Earth. The first live organisms emerged there. The composition of the intracellular fluid of any live organism is identical to the composition of the primary ocean. Every human being spends the first nine months of his life before birth in water. Since a human body in middle age consists of water by 65% (muscles contain about 75% of water, while the blood – about 80%), it would be rightful to say that we – like our ancient evolutionary ancestors – continue to live in water past our birth. Children normally have a little bit greater quantity of water in organism, and old men have a little bit less. If the amount of water in a human body were reduced only by 10% in relation to the normal, a human being would perish.

Therefore, water accompanies us throughout the entire life, regardless of whether we habituate in the Arabian desert, the mountains of Tibet or New York's Manhattan.

At the first glance it appears that we know everything about water. In reality, water is one of the most mysterious substances.

Water's structure has been investigated for the last hundred years. The scientists, who have contributed significantly to this field include Bernal and Fowler (1933), Pople (1951), Frank and Wen (1957), Pauling (1959), Nemetry and Sheraga (1962), Davis and Litovitz (1966), Sceats and Rice (1980), Dahl and Anderson (1983).

Before analyzing specific properties of the water structure in macroscopic volumes, let's examine the structure of an  $H_2O$  molecule. Three nuclei form an isosceles triangle in an  $H_2O$  molecule with two protons ( $H^+$  nuclei) in its foundation and a nucleus of oxygen at the top. Interaction between hydrogen and oxygen is achieved thanks to the *S-P*-link by two *S*-electrons of oxygen. The distance  $OH$  (length of the triangle's sides) in a molecule of water steam is 0.957  $\text{Å}$ . It is a little bigger in ice, equal to 0.99  $\text{Å}$ . The distance  $H^+ - H^+$  in a molecule of water steam is equal 1.54  $\text{Å}$ . Ten electrons surround the three nuclei in a water molecule. Two electrons are mainly located close to an oxygen nucleus.

Several works (Lennarri-Jones, Pople, 1951, Pople, 1950) used the precise method of molecular orbits for studying the electronic structure of water molecules. In those works, it was demonstrated for the first time that conditions of the other eight electrons might be described as movements along the four oblong ellipsoid orbits. The axes of two of those orbits are oriented along the direction of the link  $O-H$ . The axes of the other two electronic orbits lie in the plane passing through an oxygen nucleus and perpendicular to the plane  $HOH$ . The axes of the four ellipsoid orbits are directed towards the tips of the tetrahedron with the center in the middle of the water molecule. The electrons move along the oblong orbits in couples (according to orientation

of spins). The protons  $H^+$ , located inside the two orbits, are linked to two poles of positive electric charges in the peripheral part of the water molecule. The electrons moving along the other two orbits form the so-called lone-pair electrons, which play a very significant role in interaction between water molecules in ice and water. These electrons explain a comparatively bigger value for electronic density in the peripheral part of the molecules of water, opposite to the other part, where hydrogen is located. Two poles of negative charges of water molecules are linked with them. The currently adopted model of a water molecule is practically completely identical to the model, which had been first proposed in the work (Bernal, Fowler, 1933).

According to calculations accounting for all the peculiarities of ionic links (Coulson, 1965), it was demonstrated that the hybrid-forming process involving  $2S-$ ,  $2P_y-$  and  $2P_z-$  orbitals of an atom of oxygen during formation of links with oxygen atoms is a more important factor than ionicity. Such hybrid-forming process results in spatial redistribution of charge and altering the shape of electronic orbits participating in establishing of a link.

Presently, intracellular distances and angles for a water molecule are quite well known. Actual values of electric charges are somewhat less definite. A positive charge of an oxygen nucleus is almost completely screened off, while proton charges are screened off by more than 70%.

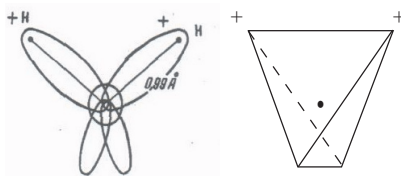
Certain parameters of a water molecule  $H_2O$ , as well as other similar molecules of intermediate (HDO) and heavy ( $D_2O$ ) water are presented in *Table 1.1* (Krasnov, 1968)

*Table 1.1*

Molecule	$H_2O$	HDO	$D_2O$
Length of link	0.9572 $\text{Å}$	0.9571 $\text{Å}$	0.9575 $\text{Å}$
Molecular angle	104.523°	104.529°	104.474°

Some authors provided different data for these parameters from those presented in *Table 1.1* (e.g., (Zatsepina, 1998)).

Each water molecule has four poles of electric charges (two positive and two negative), which form a tetrahedron. In order for such model to have a dipole moment of 1.87 Debye (such moment corresponds to experiments on water polarization), charges concentrated at the ends of the tetrahedron should be equal to  $+0.171 e$  ( $e$  – an electron's charge).



*Fig. 1.1 Model of a water molecule*



An adequate description of a water molecule can only be done by quantum mechanics. In the general case, a precise solution of the corresponding three-dimensional Schrodinger equation presents a very formidable task. There is one possibility, however, which makes that calculation not only simpler, but more graphic as well. That possibility is based on the assumption that the so-called adiabatic approximation could be used here, which would be possible if different degrees of freedom need a widely fluctuating specter of energies required for their inducement. In the case of a water molecule, an electronic subsystem and an ionic subsystem could be identified and, ultimately, movement of the whole molecule could be studied from the perspective of a separate object. These three subsystems determine the electronic, fluctuating and rotating degrees of freedom of a molecule. Characteristic energies of inducement for these degrees of freedom correspond to the ultraviolet (UV), close infrared (IR) and remote infrared (as well as microwave) frequencies of the electromagnetic specter.

Big difference of frequencies between each of these subsystems allows neglecting mutual interference in their interaction in calculations. Therefore, electronic transfers determining the main maximum of absorption of water molecules in the ultraviolet range of the specter have no significant effect on conditions of fluctuations of a water molecule, which parameters are represented by the sum of harmonic oscillations and characterized by frequencies, belonging to the close infrared area. Accordingly, vibrating conditions have a weak effect on the rotating specter, which frequencies lay in the far infrared range and the range of sub millimeter frequencies. Several important characteristics of a molecule such as the inertia moment, the dipole moment, mass, atomic link force can be extrapolated from the analysis of the rotating specter.

The centrifugal force, generated by rotation of a water molecule, causes its deformation. For example, when rotation is accelerated to the state corresponding to the quantum number  $j = 11$  (in this condition, the wave number of radiation of a rotating molecule  $k = 1/\lambda = 280 \text{ cm}^{-1}$ ) the molecular angle between directions of links between the nucleus of oxygen and the protons (HOH) decreases from the initial value of  $104^{\circ}27'$  to  $98^{\circ}52'$  while the distance of a link OH increases to  $0.964 \text{ \AA}$ .

Deformation of a molecule of water may also be attributed to agitation of its electronic levels, which alters distribution of the electrons and, subsequently, to different constants of links. For example, during irradiation of water steam molecules by various specter lines of vacuum ultraviolet light with wave length  $\lambda = 1219 \text{ \AA}$ , the length of the OH link increased to  $0.067 \text{ \AA}$  and the molecular angle increased by  $8.3^{\circ}$ . Accordingly, irradiation with a different wave length (at  $\lambda = 1240 \text{ \AA}$ ) caused the angle to increase by  $5.5^{\circ}$  (Bell, 1965).

Condition of a fluctuating specter also affects the size of a water molecule – it increases with more agitation. This is due to an obvious fact that with increasing amplitude of fluctuations, non-symmetric inharmonic oscillator properties begin to manifest themselves. The non-symmetric inharmonic oscillator is created by ions of equal mass, which always leads to shifting of ions from their middle position.

These factors show that even without considering the specific nature of water molecules' property to form a rigid quasi-crystalline macro system (consideration of that aspect is provided

below) external non-ionizing electromagnetic fields significantly affect the parameters of water molecules. Taking this into consideration explains some features of the effect that non-ionizing electromagnetic fields (for example fields of radiation emitted by mobile telephones or fields produced by various home appliances such as microwave ovens) have on live organisms.

## 1.2 THE AREA OF STABLE EXISTENCE OF WATER IN LIQUID STATE

It is commonly known, that water may exist in solid (ice), liquid and gaseous state. Some of the main properties of water are presented in *Table 1.2*.

In solid or liquid state, each molecule of water is surrounded by four neighboring molecules. In the solid state molecules are arranged in such a way that they are connected by opposite polarities. In this structure, each molecule is encircled by four nearest molecules (*Fig. 1.2*).

Interaction between molecules is determined by hydrogen links. Four hydrogen links of a water molecule are approximately aimed at the apexes of a right tetrahedron. Properties and orientation of these links is strongly dependent on temperature  $T$  and pressure  $P$ .

Depending on the ratio of  $P$  to  $T$ , the existence of both overcooled water and ice with a well-formed crystalloid structure is possible. The record temperature for overcooled liquid water is  $-92^{\circ}\text{C}$  may be achieved at the pressure of about  $P = 2000 \text{ Psa}$ . The reason for water existing in the liquid state in such a low temperature is significant deformation of hydrogen links caused by big pressure and its higher density, which impedes its crystallization.

Table 1.2 Main physical and chemical parameters of light and heavy water  
(Physical Constants, 1991).

	H <sub>2</sub> O	D <sub>2</sub> O
Density:		
Ice at 0°C	0.917 g/sm <sup>3</sup>	1.017 g/sm <sup>3</sup>
Water at 20°C	0.998 g/sm <sup>3</sup>	1.105 g/sm <sup>3</sup>
Viscosity at 20°C	1.005 cp	1.251 cp
Melting temperature	273.16 K	276.97 K
Boiling temperature	373.16 K	374.59 K
Specific heat ( $C_p$ ):		
Ice at 0°C	2.038 J/g °K	2.202 J/g °K
Water at 0°C	4.186 J/g °K	4.23 J/g °K
Steam at 0°C	1.905 J/g °K	1.68 J/g °K
Permittivity:		
Ice at -10°C	95	92
Water at 25°C	78.5	78.2
Thermal conductivity:		
Ice at 0°C	0.235 W/°K*m	
Water at 0°C	0.561 W/°K*m	0.560 W/°K*m
Water at 100°C	0.679 W/°K*m	0.644 W/°K*m
Steam at 100°C	0.025 W/°K*m	–
Time of dielectric relaxation:		
Ice at -10°C	6*10 <sup>-5</sup> s	9.1*10 <sup>-5</sup> s
Water at 25°C	9.2*10 <sup>-12</sup> s	11.9*10 <sup>-12</sup> s
Molecular magnetic sensitivity at 20°C	-1.297*10 <sup>-5</sup>	-1.295*10 <sup>-5</sup>
Refraction parameter at 20°C	1.333	1.328

There are 10 known modifications of ice crystalloid structure with hexagonal, tetragonal, cubic, trigonal, monoclinic and tetraclinic syngonies. Moreover, there is also amorphous ice.

Density of ice may be both below the density of water (with tetragonal and cubic lattice for the so-called ice 1 [ordinary ice] it is equal 0.94 g/sm<sup>3</sup>, staying mostly constant with temperature change) as well as significantly above it. For example, density of the so-called ice 8 with the cubic lattice is 1.5 g/sm<sup>3</sup> at the temperature of -50°C.

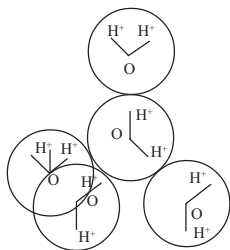


Fig. 1.2 Close vicinity of a water molecule

Higher water density during melting of ordinary ice is due to reduction of its volume caused by deformation of hydrogen links and shifts in location of molecules breaking the ideal tetrahedron configuration. Simultaneously, there is another tendency – increase of the length of hydrogen links with increasing temperature. Convergence of these two factors ultimately leads to a peculiar phenomenon of achieving the maximum density of water not at the temperature of melting of ice, but rather at 4°C. At a higher temperature, the spatial framework, characteristic of ice 1, begins to disintegrate, while “irregularly located” molecules of water begin to appear between the joints of the lattice. In the gaseous state hydrogen links disappear completely and steam behavior begins to be regulated according to the principles of gaseous state.

### 1.3 SPECIAL FEATURES OF INTERACTION OF IONIZING AND NON-IONIZING RADIATION WITH WATER. DIELECTRIC AND DISPERSING PROPERTIES OF WATER

It will be shown in greater detail in subsequent chapters that dielectric properties of water play a decisive role in the processes of interaction of ionizing and non-ionizing radiation with biological objects. In particular, even such a unique and paradoxical phenomenon as positive influence of small doses of ionizing radiation on live objects (called *hormesis*) can, to a great extent, be explained by dielectric properties of water and their variation under the influence of radiation. Let's consider the factors affecting the dielectric permittivity of water.

It is known, that each of the water molecules has its dipole moment  $p$ . It exists thanks to a spatial dispersion of an electric charge (i.e. the specific type of localization of the wave function of the electrons).

In the area of low frequencies, this type of dielectric permittivity of water is described by the formulae

$$\varepsilon_{wp}(\omega) = [1 + (8\pi/3)n_w \alpha(\omega)] / [1 - (4\pi/3)n_w \alpha(\omega)] \quad (1.1)$$

In this range of frequencies permittivity is determined exclusively by the mechanism of orientating polarizing ability of H<sub>2</sub>O molecules. The simple form of polarizing ability  $\alpha(\omega)$  is determined by the Debye relationship

$$\alpha(\omega) = \alpha_0 / (1 - i\omega\tau_r),$$

where  $\alpha_0 = p^2/3k_B T$  – static orientating polarizing ability,  $p$  – dipolar electric moment of a  $H_2O$  molecule,  $\tau_r \approx 4\pi\eta_0 \langle R \rangle^3/k_B T$  – time of relaxation of the orientating polarizing determined by the viscosity coefficient  $\eta_0$ , temperature  $T$  and the average radius of a molecule,  $n_w$  – volume concentration of water molecules.

In particular, relaxation  $\tau_r$  in clear water at  $\langle R \rangle \approx 1.5 \text{ \AA}$  and a normal viscosity coefficient for distilled water  $\eta_0 \approx 10^{-2}$  poise is equal to  $10^{-11} \text{ s}$ .

This mechanism of orientating polarizing polarization determines the form of  $\epsilon_w(\omega)$  for water from the lowest frequencies to the frequency  $\omega \approx 1/\tau_r \approx 10^{11} \text{ s}^{-1}$ . Therefore, interaction of low frequency electromagnetic waves (up to waves of the centimeter frequency range) with water is completely regulated by processes of reorientation of water molecules under the influence of the wave's electric field vector and also depends on its temperature and viscosity.

The next (by the order of increasing frequencies of the electromagnetic field) mechanism of forming a particle structure  $\epsilon_w(\omega)$  is ionic polarizing ability, which maximum matches the infrared frequency range. This polarizing mechanism is connected to various molecular fluctuations of water and can be described, with a high degree of precision, by an expression in the form of the sum of four of the dispersion components (Vysotskii, 1997)

$$\epsilon_{WIR}(\omega) = \sum_{K=1}^4 \Delta\epsilon_K \omega_K^2 / (\omega_K^2 - \omega^2 - i\omega\Gamma_K), \quad (1.3)$$

each one of which is determined by the main lines of the fluctuating specter with wave lengths  $\lambda_K$ , central frequencies  $\omega_K$  and amplitudes  $\Delta\epsilon_K$  (see *Table 1.3*).

*Table 1.3* Main resonance of water in the infrared range of the specter (Yukhnevich, 1973)

Average resonance wave length $\lambda_K$ (mkm)	Resonance frequency $\omega_K$ ( $10^{14} \text{ s}^{-1}$ )	$\Delta\epsilon_K$	Nature of resonance
$\lambda_1 \approx 52$	$\omega_1 \approx 0.36$	0.8	Inhibited translation of a molecule of $H_2O$
$\lambda_2 \approx 14.2$	$\omega_2 \approx 1.27$	0.9	Libration mode
$\lambda_3 \approx 6.07$	$\omega_3 \approx 2.97$	0.2	Deformation mode
$\lambda_4 \approx 2.86$	$\omega_4 \approx 6.3$	0.2	Valency vibrations of the OH groups of water molecules

At even higher frequencies the following and the principal maximum of interaction of the electromagnetic radiation with molecules of water  $H_2O$  is related to electronic polarizing ability and is characterized by the array of closely located spectral lines with average frequency

$\omega_{UV} \approx 2.2 \cdot 10^{16} \text{ s}^{-1}$ , average wave length  $\lambda_{UV} \approx 0.09 \text{ mkm}$  and the sum of amplitudes  $\Delta\epsilon_{UV} \approx 0.75$ . The expression for the dielectric permittivity in this frequency range has the form

$$\epsilon_{wUV}(\omega) \approx \Delta\epsilon_{UV} \omega_{UV}^2 / (\omega_{UV}^2 - \omega^2 - i\omega\Gamma_{UV}) \quad (1.4)$$

The main features of interaction of radiation with water within the boundaries of this frequency range refer to processes, related to ionization of atoms of hydrogen H and oxygen O.

The spectral width of each resonance  $\Gamma_i$  is small in comparison with corresponding resonance frequencies  $\omega_i$ . It has an intricate dependency on temperature and impurities in water. It is worth noting, that the expression for dielectric permittivity as a function of a pseudo frequency  $\omega = i\xi$  is important in the analysis of the question of the influence of water characteristics on the nature of interaction between biological macromolecules and their fragments (this fundamental problem will be addressed later). This circumstance is the result of the causation principle and it is determined by analytical properties of dielectric permittivity as a complex function of frequency. This result will be examined in greater detail in the following chapters. With such a substitution, any expression with the same form as in (1.4) can be worked into form

$$\epsilon_{wUV}(i\xi) \approx \Delta\epsilon_{UV} \omega_{UV}^2 / (\omega_{UV}^2 + \xi^2 + \xi\Gamma_{UV})$$

Proceeding from the obvious condition that in the vicinity of any electromagnetic resonance its spectral width is always much less than resonance frequency, which means that  $\Gamma_i \ll \omega_i$ ,  $\Gamma_i \ll |\xi|$ , we finally come to

$$\epsilon_{wUV}(i\xi) \approx \Delta\epsilon_{UV} \omega_{UV}^2 / (\omega_{UV}^2 + \xi^2)$$

We can see from the obtained formulae that specific expression for the width of each resonance  $\Gamma_i$  is insignificant and it doesn't affect the final solution.

At an even higher frequency (in the range of soft and hard Roentgen radiation with energy exceeding ionization potentials of the internal electronic cores of hydrogen and oxygen) all electrons in water can be treated as free.

In this case, the dielectric permittivity of water has the form

$$\epsilon_{wK}(\omega) \approx 1 - \omega_p^2 / \omega^2,$$

which is typical for plasma-like environments (the “plasmatic” approximation in the theory of dispersion of dielectric permittivity).

Here,  $\omega_p = (4\pi n_e e^2 / m)^{1/2}$  – plasma frequency,  $n_e$  – total concentration of all electrons in water,  $m$  – mass of an electron.

Beside the mentioned three main mechanisms of forming the structure of dielectric permittivity of water  $\epsilon_w(\omega)$ , there are two more mechanisms, one of which is related to the presence of charged free radicals (electrons  $e^-$ , protons  $H^+$ , ions  $H_2O^+$ ,  $H_2O^-$ ) in water and the other

one is determined by the effect of atoms  $\text{H}_2\text{O}^*$ , disturbed by thermal movement and external radiation.

There are different direct causes of appearance of the ions.

Specifically, there is always a probability of the balanced fluctuation dissociation



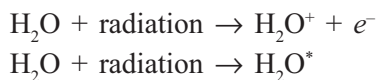
with thermodynamic probability determined from the Boltzmann expression

$$W \approx \exp(-E_\alpha/kT) \quad (1.6)$$

being dependent on temperature  $T$  and the energy of dissociation  $E_\alpha$ . In particular, at  $T = 300 \text{ K}$  the value  $W \approx 10^{-10}$ , indicating the presence of  $n^+ \approx Wn_w \approx 10^{13}$  pairs of ions  $\text{H}^+$  and  $\text{OH}^-$  in a unit of volume of water. This mechanism, involving the process of thermally stimulated water hydrolysis, will be investigated more closely in section 1.4.

Another mechanism is still more important, distinguished by generation of additional ions in the course of radiolysis during water irradiation with ultraviolet or Roentgen quanta. All products generated by water radiolysis (including charged free radicals, disturbed molecules  $\text{H}_2\text{O}^*$ , neutral free radicals  $\text{H}$ ,  $\text{OH}$ ,  $\text{HO}_2$  and molecules  $\text{H}_2$  and  $\text{H}_2\text{O}_2$ ) play a certain role in radiation and chemical transformations occurring in water solutions. Let's investigate this process more attentively (Amiragova, 1964)

With absorption of each  $100 \text{ eV}$  in the course of electrolysis there is an average of about four splits of water molecules. Under the influence of radiation on water, there are, initially, ionized and agitated water molecules  $\text{H}_2\text{O}^+$  and  $\text{H}_2\text{O}^*$ . Initial radiolysis processes, leading to generation of free radicals, are executed according to the patterns



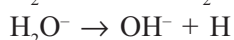
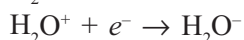
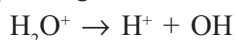
The average energy of generation of one pair of ions from a single molecule is equal  $\langle E \rangle = 34 \text{ eV}$ . Model estimates and the results of experiments testify that with absorption of  $100 \text{ eV}$  of energy from ionizing radiation an average of 2.6 ions  $\text{OH}^-$  and 2.6 ions  $e^-$  are generated. Moreover, in the process of interaction, with absorption of the same  $100 \text{ eV}$  of energy about 12 agitated molecules of water are also generated. Out of those 12 molecules about three are split by ionization and about nine – by agitation. In this case, 62% of the absorbed energy will be spent on dissociation.

Radicals generated in the course of radioactive radiation (ions  $\text{OH}^-$ , electrons and agitated molecules  $\text{H}_2\text{O}^*$ ) move in the water environment by means of diffusion. Further evolution of these radicals corresponds to a chain of chemical transformations.

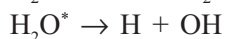
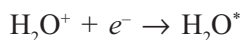
There are two hypothesis regarding subsequent transformations of ion  $\text{H}_2\text{O}^+$ . According to one of them, ionized water molecules dissociate with generation of ion  $\text{H}^+$  and a radical  $\text{OH}$ , while the electron, having been slowed down as a result of interaction with water, is attached to a water molecule with generation of a negatively charged ion  $\text{H}_2\text{O}^-$ . During dissociation, that ion disintegrates into an ion  $\text{OH}^-$  and a neutral radical  $\text{H}$ .

Theoretically, another variant is also possible, when the ion  $\text{H}_2\text{O}^+$ , generated during dissociation of a water molecule, quickly captures the electron in the liquid environment, which leads to generation of an agitated molecule of water  $\text{H}_2\text{O}^*$  with high energy of agitation. Later, that molecule disintegrates into two neutral radicals  $\text{H}$  and  $\text{OH}$ .

Corresponding reactions have the form



Then, in the chain of consecutive transformations, atomic hydrogen is generated



Currently, it is impossible to say, which of the variants has bigger probability. The two demonstrated methods of radicals' generation from ionized water molecules are different by mutual position of the radicals  $\text{H}$  and  $\text{OH}$  in space.

In the first case, the radicals  $\text{OH}$  are generated in spots where ions appear, i.e. directly along the track of an ionizing particle while radicals  $\text{H}$  are generated at some distance, equal to a free run of an electron until its collision with a water molecule. That distance is approximately equal to  $150 \text{ \AA}$ , which is about the length of 70 water molecules placed side by side.

In the second case, the radicals  $\text{H}$  and  $\text{OH}$ , generated during dissociation of an agitated molecule of water  $\text{H}_2\text{O}^*$ , appear close to each other.

Behavior of agitated water molecules  $\text{H}_2\text{O}^*$  is quite interesting. Absolute majority of them is agitated to levels of energy, representing the lower states with energy 5.6 and 7.5 eV. Such molecule can dissociate. In order to achieve that effect, dissociation energy of 5.2 eV is required. Radicals generated during dissociation have a small surplus of kinetic energy (from 0.4 to 2.3 eV). In the process of collision with neutral molecules of water the radicals are quickly slowed down along the length of the track

$$l \approx 1/\sigma_c n_w \approx 20\text{-}30 \text{ \AA}$$

and cannot diverge away from each other by a large distance. Here,  $\sigma_c \approx 10^{-16} \text{ sm}^2$  – cross section of elastic diffusion of the radicals on water molecules.

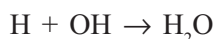
Due to such a rapid braking, these radicals are capable of repeated interaction (forming combinations) after stopping. That effect, localized in space, limits the mutual location of the radicals in water and is called “Frank-Rabinivitch cells”. It plays an important role in the radiolysis process. Due to that effect the free radicals, generated during dissociation of the initially agitated molecules, do not participate in later water radiolysis.



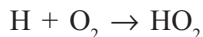
Further transformation of free radicals H, OH and HO<sub>2</sub> takes place in the course of several alternative reactions. In particular, when water is depleted of free oxygen, a reaction of synthesis of a very powerful oxidizer (hydrogen peroxide) takes place



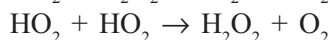
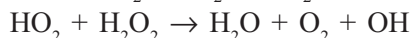
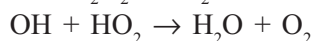
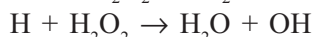
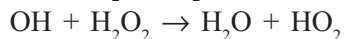
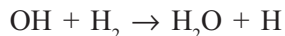
as well as a reaction of water reduction



When there is a lot of dissolved oxygen in water, another reaction occurs



Beside that, there are also secondary reactions in the water environment



All free radicals and molecular products, generated during water radiolysis (ions H<sub>2</sub>O<sup>+</sup>, H<sub>2</sub>O<sup>-</sup>, agitated molecules H<sub>2</sub>O\*, free radicals H, OH, HO<sub>2</sub> and molecular products H<sub>2</sub> and H<sub>2</sub>O<sub>2</sub>) play their parts in radioactive and chemical transformations in water.

Specifically, radioactive and chemical transformation processes in dissolved water solutions are explained by the presence of free radicals H, OH, HO<sub>2</sub> as well as hydrogen peroxide H<sub>2</sub>O<sub>2</sub>. Probability of reactions between radicals decreases rapidly as distances between them increase in the result of diffusion. Based on that, a clear difference is established between radicals contributing to generation of molecular products H<sub>2</sub> and H<sub>2</sub>O<sub>2</sub> and radicals, which have avoided transformation into molecular products in the result of diffusion or a reaction with a dissolved substance. According to this model, the final yield of products of radiolysis should not change with adding more dissolved substance.

Counter to that, the yield of molecular products in the same process may alter considerably. Secondary reactions of radicals with molecular products occur primarily in clear water and they don't have a significant influence on radiolysis of water solutions.

The radical OH reacts mostly as an oxidizer and because of that many radioactive and chemical transformations of oxidization in water solutions are explained by its participation. In oxidization reactions it can act as an acceptor of electrons or acceptor of atoms of hydrogen. Oxidizing capacity of OH is determined by the oxidization-reduction potential of a system and

depends on the environment's acidity. In particular, oxidizing ability of the radicals OH decreases with higher  $pH$ .

In most cases, a neutral radical H enters into chemical reactions as a reducer. The reducing capacity of atoms of H usually increases with higher  $pH$ . In acid liquors, the radical H can sometimes act as an oxidizer.

Depending on pH and the oxidization-reduction potential of the environment, radical  $HO_2$  can act both as an oxidizer and reducer. Its oxidization capacity decreases with higher  $pH$ , while its reduction capacity increases.

The provided brief analysis of the mechanism of radiolysis of water shows a wide variety of chemical reactions and final products, among which free radicals are dominant. Another very important conclusion is that the complicated chain of consecutive chemical transformations shown earlier may be initiated not only by an act of initial radiological impact on a water molecule but also at the presence of certain chemical and thermodynamic factors characteristic of initial dissociation.

We shall demonstrate in the coming chapters that the influence of these factors and the possibility of reducing the effect of free radicals generated by a non-radioactive way is inseparably connected with one of the most probable scenarios of the phenomenon of positive effect of small doses (called *hormesis*).

Let's now turn to considering the effect of electrons, generation of which represents one of the stages of the process of water radiolysis. It appears at first, that the role of free electrons is incommensurably weaker than the direct effect of very aggressive heavy radicals. In reality, it is quite different.

It follows from the expression (1.5) that the contribution of "plasmatic" approximation to the overall dielectric permittivity is inversely proportional to the mass of a charged object  $m$ . Due to a smallness of mass of an electron  $m_e$  (compared with masses of much heavier ions of hydrogen and oxygen) it is most interesting from the point of view of its influence on the structure  $\epsilon_w(\omega)$ . Obviously, the "plasmatic" approximation takes place in the area of extremely high frequencies (i.e. in the Roentgen frequency range), where electrons tied in atoms and molecules can be regarded as free, as well as in the case when electrons are actually free (in the result of spontaneous ionization or radiolysis). Electrons, generated in the process of radiolysis, after their rapid slowing down in the water, are transferred into a quasi stationary condition of a "hydrated electron" with a longer life span  $\tau_e \approx 7.2 \cdot 10^{-4} s$ . The life span  $\tau_e$  corresponds to the process of repeated combination (deionization) of an "almost free" hydrated electron. Meanwhile, an additional contribution to the general dielectric permittivity of water made by a system of such elements may be calculated by analogy with the case of plasma-like environments.

The equation for the movement of such electrons in the presence of an external electric field  $\vec{E} = \vec{E}_0 \exp(-i\omega t)$ , which may bear the mark of an external influence as well as have purely fluctuational features and be regulated by the features of quantum electrodynamics, has the form

$$m \frac{d^2 \vec{r}}{dt^2} + (m/\tau_e) \frac{d\vec{r}}{dt} = e \vec{E}_0 \exp(-i\omega t) \quad (1.7)$$

After completion of transformational processes, forced movement of each of these electrons is described by the solution

$$\vec{r} = - [e \vec{E}_0 / m(\omega^2 + i \omega / \tau_e)] \exp(-i\omega t) \quad (1.8)$$

The dipole moment created by such an electron is  $\vec{p}_g = e\vec{r}$  equal and the induction vector  $\vec{D} = n_g \vec{p}_g$ . Here,  $n_g$  – concentration of hydrated electrons.

Using the connection  $\vec{D} = \vec{E} + 4\pi\vec{P} \equiv \Delta\epsilon_{wg}\vec{E}$  we finally find the expression for the contribution of hydrated electrons

$$\Delta\epsilon_{wg}(\omega) \approx -4\pi n_g e^2 / m(\omega^2 + i\omega\Gamma); \Gamma = 1/\tau_e \quad (1.9)$$

to the general dielectric permittivity of water.

The value of  $\Delta\epsilon_{wg}(\omega)$  determining the influence of hydrated electrons has a negative actual part

$$\text{Re}\{\Delta\epsilon_{wg}(\omega)\} \approx -4\pi n_g e^2 / m(\omega^2 + \Gamma^2) \approx -4\pi n_g e^2 / m\omega^2 \quad \text{with } \omega \geq \Gamma \quad (1.10)$$

which is completely analogous to plasma-like environments.

In this case there is a minimal frequency  $\omega_{min}$ , determined from the condition  $\epsilon_w(\omega_{min}) = 0$ . Below this frequency (with  $\omega < \omega_{min}$ ) we have  $\epsilon_w(\omega < \omega_{min}) < 0$ .

In the range of frequencies  $\omega < \omega_{min}$  existence of electromagnetic waves is impossible, since for them, the wave number

$$k = -(\omega/c)\{\epsilon_w(\omega)\}^{1/2}$$

becomes purely imaginary. Such waves cannot spread in the environment. This “cut-off frequency”  $\omega_{min}$  plays an important role in the analysis of interaction of biological macromolecules in water-containing environments (including volume of a cell). Another mechanism exerting additional influence on dielectric permittivity of water is related to the presence of agitated atoms in the structure  $\epsilon_w(\omega)$ . This mechanism provides for the existence of additional resonance of electronic absorption into a softer range of frequencies than that of non-agitated atoms as well as the need to account for dispersion characteristics of a system of agitated atomic electrons in a frequency range exceeding the ionizing potential of water molecules. The number of agitated molecules of water during irradiation and radiolysis can be estimated with reference to a fact, that with absorption of each 100 eV of energy from external radiation, an average of 9 agitated molecules H<sub>2</sub>O are generated. Durations of those agitated states lies within the range from  $\tau_1 \approx 0.1$  s (for metastable levels with quadruple transfers of the E2 type) to  $\tau_2 \approx 10^{-7}$  s (for permissible dipole transfers of the E1 type), given that because of the effect of “radiation capture” related to resonance

overabsorption of quants in the volume of the same environment, the actual duration may be significantly increased.

The presence of hydrogen links in water causes one more specific phenomenon determining dielectric parameters of water and special features of behavior of biological objects in water. In some cases, water displays the effect of non-local polarizing. Then, instead of an ordinary linear and localized equation describing the relation of induction  $\vec{D}(\vec{r})$  and tension  $\vec{E}(\vec{r})$  in an electric field

$$\vec{D}(\vec{r}) = \varepsilon \vec{E}(\vec{r})$$

there exists an equation for non-local relation

$$\vec{D}(\vec{r}) = \int_v \varepsilon(\vec{r}, \vec{r}') \vec{E}(\vec{r}') dV'$$

This kind of relation is natural only for crystals and corroborates the fact that a component of an electric field in one part of a crystal determines induction in another part. In crystals, such non-local character is a natural consequence of the periodicity of lattice and full correlation in positioning of its elements. In water, due to a large number of hydrogen links, there is also a strong correlation of polarizing in points that are spread out in space (in a much lower volume of space, of course).

#### 1.4 SPATIAL STRUCTURE WATER MODELS

Before we conduct an analysis of main models characterizing the structure of mutual situation of water molecules in space, let's turn to experimental results related to the study of such structure. Anomalies in water characteristics were the reason for making a suggestion that water is a mix of different types of associated molecules:

“hydrol”  $\text{H}_2\text{O}$ , “di-hydrol”  $(\text{H}_2\text{O})_2$ , “tri-hydrol”  $(\text{H}_2\text{O})_3$  and so forth. Sometimes, these hypothetical associates are called polymers. The question about the maximum size for a polymer can be solved based on comparison of experimental data and theoretical results.

First of all, it is evident, that since liquid water is a fluid, capable of easy penetration of pores with diameter over 3 Å, it is fundamentally different from ice and cannot have rigid crystalline structure, which is characterized by the presence of a “distant order”. By the term “distant order” we mean such a way of distribution of an ordered system in space, when features of periodicity and parameters of translation symmetry don't depend on absolute shift, if such shift is a multiple of one of the spatial periods. Meanwhile, many facts suggest that water is actually characterized by a specific spatial order. Based on this, we deem it evident, that liquid water is most ordered near each of the molecules in a small volume of space. It corresponds to the fact that there must be “close order” in water, while a definite “distant order” is missing. Because of

that, it would be useful to consider the function of radial distribution of the main structural elements of water (for example, nuclei of oxygen or water molecules, taken as a whole).

The simplest variant of such analysis is related to determination of the function

$$g(r) = \langle n_w \rangle^{-1} dN_w(r)/dV \equiv n_w(r)/\langle n_w \rangle \quad (1.11)$$

of radial distribution of molecules of water in relation to any chosen molecule or, on the other hand, relation of local density of molecules  $n_w(r)$  of water to average density  $\langle n_w \rangle$ .

This function also determines the probability of finding a concrete quantity of molecules  $N_w(r)$  in a single volume located at a distance  $r$  from a chosen molecule. Considering, that this function does, in fact, determines the correlation (mutual relation) between positions of centers of different molecules, it (in accordance with the terminology of the theory of random events) has another name – paired correlation function.

Proceeding from such definition, it is easy to find an expression for total quantity of water molecules in a spherical layer with the radius  $r$ , volume  $dV$  and width  $dr$

$$dN_w = g(r)\langle n_w \rangle dV = g(r)\langle n_w \rangle 4\pi r^2 dr \quad (1.12)$$

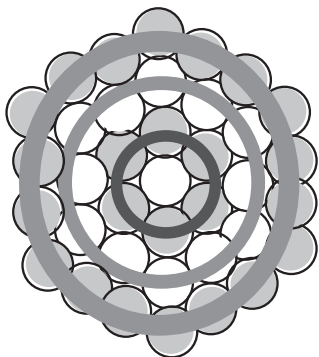
We can easily make some modeling estimates determining the behavior of the variable  $g(r)$  at small values of  $r$ . Let's assume, that the variable  $r$  characterizes the current position of the center of a molecule. If we suppose that all water molecules are balls with diameter  $R$ , it follows directly from (1.11) that at  $r < 2R$  we have  $g(r) = 0$  (meaning that one molecule cannot penetrate the volume of another).

Correspondingly, with increasing  $r$  the value of  $g(r)$  increases and with  $r = 2R$  we have  $g(r) > 1$ . This result becomes immediately clear, if we use a simplified version of the model of water, which states that water molecules are in tight contact with each other. Based on the same model, with further increasing of  $r$  the value of  $g(r)$  decreases and at  $r = 3R$  we have  $g(r) < 1$  (in an ideally periodical system  $g(r) = 0$  in this case). With further increase of the distance  $g(r)$  would also increase and so forth.

The quantity of water molecules corresponding to the first maximum of the probability density  $g(r)$  is usually called the coordination number (and hypothetical position of these molecules is called the first coordination sphere). The same terminology is used for describing the number of molecules, whose position in space corresponds to the second, third and subsequent maximums of the function  $g(r)$ , which corresponds to the second, third and subsequent coordination spheres (*Fig. 1.3*).

If water structure was characterized by the distant order, the function of radial distribution  $g(r)$  would be oscillating with a constant period and a constant amplitude of oscillations. However, because of unavoidable violations of periodicity with increasing distance, there is a rather fast decrease of the amplitude of oscillations  $g(r)$  and with  $r \gg R$  we have  $g(r) = 1$ . This result

follows directly from *Fig. 1.3* (with a higher coordination sphere number there is a higher uncertainty about the size of its radius).

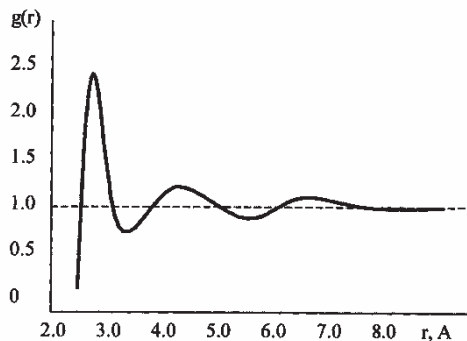


*Fig. 1.3 Spatial distribution of the first three coordination spheres with maximally dense water molecules*

The concept of coordination number can, strictly speaking, be applied only to crystals. Coordination numbers are just as useful for describing structures of liquids, but they have mainly statistical meaning in that case. In the case of liquids, they actually determine how close crystalline structure is to liquid structure.

In practice, the function  $g(r)$  is determined from the analysis of diffraction of Roentgen rays and thermal neutrons. On *Fig. 1.4* there is a paired correlation function of distribution of centers of water molecules (i.e. distribution of distances between nuclei of hydrogen atoms in different molecules of water).

It follows from this picture, that in the vicinity of any molecule of water there are three coordination spheres. Position of the first maximum and its area allow us to conclude that each molecule is surrounded (on average) by less than five other molecules located at a distance, close to the length of hydrogen links. Position of the second maximum (about  $4.5 \text{ \AA}$ ) matches the length of an edge of a tetrahedron hypothetically attached to a molecule of water. This serves as one of the evidence supporting the theory that water represents a three — dimensional tetrahedron lattice consisting of interlinked molecules.



*Fig. 1.4 Paired correlation function of distribution of centers of water molecules*

The main question in any cohesive theory dealing with water is how well the known properties of a molecule of water (i.e. properties of its microscopic structural element) can explain all the numerous effects, which characterize the macroscopic volume of water. Due to importance of this problem it is beneficial to investigate it based on historical approach.

Let's review the evolution of understanding of the structure of water.

One of the first cohesive theories, which explained the structure of water on the basis of mathematical calculations, presumed that there is a set of similarities between properties of solid objects and liquids (quasi-crystalline theory of the liquid state). There were good reasons for such an analogy. Indeed, the density of water and the concentration of molecules in it is quite comparable with similar parameters of solid objects (including in crystals). Many liquids have the ability to preserve their form (for example, glass and sap). Thermodynamic, acoustical and electrophysical properties of many liquids are little different from the corresponding properties in solid objects. Of course, there are important differences, too. For example, ionic conductivity of electric current, non-existent in crystals, plays an important role in liquids. The diffusion coefficient in liquids is high, although, in all fairness, we should note, that coefficient of diffusion of atomic hydrogen in crystalloid palladium, for example, is so high, that it can well be compared with diffusion in some liquids.

The quasi-crystalloid theory of the liquid state was created in 1939–1954 (Zatsepina, 1998). According to that theory, the solid and the gaseous states of the matter were treated as two extreme cases of the liquid state. Within the framework of that theory, a peculiar symbiosis of two different processes – constant movement of atoms or molecules, as well as the presence of an oscillating movement around the equilibrium position – each of which characteristic either for gas or for a solid object.

Qualitatively, this model of liquid could be described the following way. Potential relief (spatial structure of potential energy for each of the molecules) of a quasi-crystalline liquid is an almost periodic three-dimensional system of potential pits and barriers. These pits and barriers are the result of self-regulated movement of all atoms and molecules. By the way, this is the approach implemented in one of the principal variants of the theory of nuclear matter (the “drop” or “hydrodynamic” model of a nucleus), where a potential barrier is created by coordinated movement of all protons and neutrons in the volume of a nucleus.

Movement of molecules, according to this model, is a combination of two independent movements – the oscillating movement in the volume of each of the potential pits and, then, a random (fluctuational) jump into a neighboring pit. During this process, the average frequency of oscillations in a potential pit is approximately the same as regular Debay frequencies in a solid object (i.e. about  $\omega_D \approx 10^{13} \text{ s}^{-1}$ ). Accordingly, the average duration of a jump into a neighboring pit is equal to  $\tau_0 \approx 10^{-13} \text{ s}$ .

Average time of staying in the same pit  $\langle \tau \rangle$  is much longer than  $\tau_0$ . It can be easily determined with the help of simple thermodynamic estimates, provided that in order for a molecule to leave a potential pit it must receive additional kinetic energy, exceeding pit's depth  $\Delta W$ . A molecule can receive that energy only from the whole system's overall thermal energy. From the



Boltzman distribution follows, that the probability of fluctuational concentration of energy on one particle at a temperature  $T$  equals to  $\exp(-\Delta W/kT)$ , and the probability of a molecule leaving a pit within a unit of time equals  $\exp(-\Delta W/kT)/\tau_0$ . Hence, we find that the average time of a molecule's stay in one potential pit equals

$$\langle \tau \rangle = \tau_0 \exp(\Delta W/kT)$$

Since  $\Delta W \gg kT$ ,  $\langle \tau \rangle$  turns out to be much larger than  $\tau_0$ .

By the same logic, we can find an approximate expression for the average speed of movement of a molecule in a liquid. Assuming that the average distance between centers of neighboring potential pits equals  $2R$ , average speed is

$$\langle v \rangle \approx 2R/\langle \tau \rangle = (2R/\tau_0) \exp(-\Delta W/kT)$$

This expression agrees very well with the experimental data obtained from studying diffusion and ionic electric conductivity in water. In addition, it's obvious that such a simplified model of liquid is based on very rough assumptions and cannot explain many facts observed in experiments (in particular, its irregular compressibility and "water's memory").

Another approach to a model of liquid as an analogue of solid objects was made in the works of Bernal (1959, 1960). Based on specific rules of symmetry, he came to the conclusion that correlation features in distribution of water molecules neatly match the experimental data provided that spatial distribution of molecules of water is characterized by pentagonal symmetry. This model had allowed Bernal to explain accurately such anomalies in simple liquids as their capacity for excess chilling without a transition to the solid state, high entropy of water, its liquidity and several other effects.

The "dodecahedron model" of L. Pauling (1959) holds a special place among numerous models of water structure. In the basis of this model there is the idea that hydrates of gases can create spatial tetrahedral frames with vary large (in the scale of a water molecule) internal micro cavities with rigid atomic walls. Such systems have been dubbed clathratic hydrates. The main elements of this structure are right polyhedrons (dodecahedrons), each one of which is interlinked by hydrogen links. Each one of them has 12 pentagonal sides, 30 edges connecting those sides and 20 ends with 3 edges converging in each one of them. Water molecules are located in the joints of this spatial frame. If there are alien dissolved gases in water, they are located in internal micro cavities. In the Pauling model, frame molecules of water form the walls of internal micro cavities, each one of which can be characterized by an inscribed sphere with diameter  $5.2 \text{ \AA}$ . Molecules of methane  $\text{CH}_4$ , oxygen  $\text{O}_2$ , nitrogen  $\text{N}_2$ , chlorine  $\text{Cl}_2$  may be located in the volume of micro cavities. Micro cavities are connected with external environment by windows with diameter about  $2.5 \text{ \AA}$ , which is little less than diameter of a water molecule ( $\approx 2.76 \text{ \AA}$ ). These micro cavities are separated from the "outside world", i.e. from the main volume of water, by a certain potential barrier.



We can easily make sure, that the internal size of these micro cavities is much larger than the diameter of a water molecule. Therefore, molecules of water may be located in those cavities without forming any hydrogen links! Moreover, due to high symmetry of the electrostatic field inside micro cavities there is a certain limitation on formation of hydrogen links for such molecules of water, which makes them hydrophobic. Calculations have shown, that the density of such structure (without filling it with water molecules) equals  $0.80 \text{ g/sm}^3$ . If micro cavities are filled with water molecules, density is close to normal water density  $1 \text{ g/sm}^3$ .

Due to the named circumstances, existence of micro cavities not filled with water is possible, along with a stable and isolated from the outside environment condition of water in those micro cavities.

Minute calculations show, that within the temperature range from  $0^\circ$  to  $30^\circ\text{C}$  the Pauling model can sufficiently explain all features of water (including its unusual compressibility with a minimum at the temperature of  $4^\circ\text{C}$ ).

It will be shown in chapter 2 that many anomalous features of water (even the most important one of them – the –“water memory”) can be successfully explained on the basis of the Pauling model in its comprehensive application.

In his works, Frank studied the possibility of uniting the clathrate Pauling model with the cluster model. In that case, the dodecahedron frames can sometimes connect with each other by hydrogen links and, thus, form groups with ordered structure (i.e. clusters). Since there is a very strong correlation between close hydrogen links, appearance and removal of hydrogen links takes place in a coordinated fashion, being synchronized in time. Such nature of the link allows us to suggest that “flickering clusters” appear and disappear in water. The mechanism of forming such clusters is the following.

At the room temperature more than half of hydrogen links in water is severed. Nevertheless, an even distribution of severed links over the molecules is not advantageous due to collective (cooperative) character of their generation. Remaining links are redistributed in a way that allows achieving their maximum concentration forming associates with the maximum number of links per molecule. It corresponds to formation of a structure, close to the structure of ice, providing the maximum number of links. These associates are rather unstable. Local fluctuations of energy lead to their disintegration and appearance of new ordered associates – clusters. The life time of clusters – about  $10^{-10} \text{ s}$ , or about 1000 molecular oscillations.

Later, Frank and Quist (1961) as well as Frank and Wen (1957) both considered another variant of a double structured cluster model of water. They studied water as a balanced mix of monomeric molecules and molecules, belonging to ice remains. In this model, great significance is given to a collective mechanism of appearance and disappearance of hydrogen links. In liquid water the “flickering clusters” with ordered internal hydrogen connection (there are ice-like groups of water molecules in those clusters) alternate with vast areas, where hydrogen links don’t exist.

Various aspects of this problem are actively discussed at the present time. In one of the most comprehensive reviews of late (Buljonkov, 1991) verified data related to the tetrahedron location of hydrogen links is used to create a universal clathrate of water consisting of three