Influence of Non-Thermal Intensity EHF Radiation on Properties of Water and NaCl Aqueous Solutions

D.Y. Mynziak, P.P. Loshitskiy Department of Physical and Biomedical Electronics National Technical University of Ukraine "KPI" Kiev, Ukraine minzyak@mail.ru, pepel@phbme.ntu-kpi.kiev.ua

Abstract — This paper reports on the investigation of concentration and time-dependent properties of water and NaCl solution under the effect of the electromagnetic fluctuations of extremely high frequencies (EHF). The maximum sensitivity of physiological solution to external physical effects is shown.

Keywords—water; NaCl aqueous solution; EHF radiation; dielectric constant; biological systems

I. INTRODUCTION

Water and aqueous solutions of mineral salts, proteins and other organic compounds are the main building blocks of the biological objects. Their prevalence in nature and pivotal role in functioning of the living organisms make aqueous solutions the object of research in many scientific areas [1]. One of the important aspects is the investigation of the influence of the external physical phenomena on the properties of aqueous solutions.

NaCl solutions are one of the most studied systems from their thermodynamic and dielectric properties point of view [1-3]. Therefore, these solutions can be used as a model system for investigating influence of external physical phenomena, in particular, of the radiation with extremely high frequencies (EHF) of non-thermal intensity. Aqueous solutions of NaCl belong to the group of strong electrolytes are their behaviour is described by Debye-Hückel theory, that is based on the assumption of total dissociation of strong electrolytes into ions and accounts for overall interaction of each ion with all other ions sounding it [4, 5]. To account for the effect of the solvent, the theory contains the concept of macroscopic dielectric constant. Around an ion a cloud with the opposite charge is formed. Therefore each point of the solutions experiences total effect of all these ions. The radius of ionic cloud is a characteristic length, which determines some statistical sphere that surrounds central ion. The existence of ionic atmosphere (cloud) distinguished real solutions from the ideal ones. Characteristic length depends on dielectric constant, concentration and ion charge [6].

This theory is based on the assumption that ion has no dimensions and considers a solvent as non-structured media with average value of dielectric constant. It cannot describe the microstructure of electrolytes aqueous solutions as well as explain a number of properties of aqueous solutions, in particular abrupt decrease of NaCl diffusion coefficient in O. Korostynska, A. Mason and A. I. Al-Shamma'a Built Environment and Sustainable Technologies (BEST) Research Institute, School of the Built Environment, Liverpool John Moores University Byrom Street, Liverpool, L3 3AF, UK o.korostynska@ljmu.ac.uk

aqueous solutions of small concentration [7], which probably is due to structural changes in the solution.

This work aims to investigate the changes in properties of water and NaCl aqueous solutions in various concentrations under the influence of EHF radiation of non-thermal intensity by using a novel approach that allows determination of fluctuations of fluid differential temperature [8].

II. EXPERIMENTAL PROCEDURES AND METHODS

As a test media, reagents with at least 99% purity and deionised water with more than 99.9% chemical purity were used for the experimental studies. The solutions with required concentrations were prepared by dissolving certain amount of NaCl in a fixed volume of deionised water. Weight concentration (C) of NaCl in solution was calculated using (1):

$$C = \frac{M_s}{M_p} *100\%$$
 (1)

where $M_p = M_w + M_s$ is the mass of the solutions, with masses of water M_w and salt M_s measured by weighing on analytical scales with 0.001 g accuracy.

Experimental investigations on dependence of noise dispersion of differential temperature between the control and measured samples on NaCl aqueous solution concentration and assessment of influence of UHF radiation on these solutions were conducted using a specifically developed laboratory apparatus, which consists of a micro-thermocouple with digital processing of measured values. The apparatus has the absolute uncertainty of measurements is 0.03 °C and provides for recording data with 2-3 s period. Detailed description of measurements methods, metrological characteristic and calibration parameters of the apparatus are given in [9]. Measurements of aqueous solutions parameters were taken at a room temperature of 23 ±1 °C. Apparatus "Oratoria - IVM" was used as a source of UHF radiation and it provided generation of broadband noise in 53-68 GHz working range with noise intensity spectral density S=10⁻¹⁹ W/Hz*cm² and integral power $P=10^{-9}$ W/cm², and has amplitude modulation with 6-18 Hz random frequency. The presence of random modulation allows reduction of irradiation time and, moreover, considerably increases repeatability and reproducibility of the results. The emitter (except for an antenna) and power supply and control unit were shielded and grounded.

III. RESULTS AND DISCUSSION

To determine which concentration of NaCl in aqueous solution is the most sensitive to the influence of UHF irradiation, the experiments were conducted for the solutions with the following concentrations: 0% (deionised water), 0.5%, 0.9% (physiological solutions), 1%, 2%, 3%, 4% and 5%. Each experiments in this series consists of three stages: (1) 10 min without irradiation (control sample), (2) 10 min under the UHF irradiation and (3) 10 min after the influence to assess the consequences of UHF irradiation influence. The results of recording of the differential temperature in a form of noise temperature dispersion are given in Fig. 1.



Fig. 1. Noise dispersion in NaCl solutions.

Analysis of the noise dispersion graphs showed that the biggest change in noise dispersion was demonstrated by 0.9 % NaCl (physiological solution).

To investigate the influence of UHF irradiation of various duration on 0.9% NaCl physiological solution, the measurements were taken as follows: 10 min before the irradiation, then for 2, 5, 10, 15, 20, 25 and 30 min UHF irradiation and finally after 10 min after the irradiation procedure. The average algebraic value of differential temperature dispersion in 5 min intervals was chosen as an assessment criterion of the UHF irradiation effect. Accounting for the fact that dispersion depends on the sampling range size, to exclude the results variations due to different size of sampling range, the following approach was used: each stage – before, during and after the irradiation – was separated into 5 min intervals in which the dispersion was calculated, and afterwards these values were processes as algebraic average. The results of noise dispersion processing are illustrated in Fig. 2 and Fig. 3.

As one can see from the above graphs, the noise dispersion curve reaches the maximum value at 15 min after beginning of irradiation (Fig. 2) and in another case at 20 min (Fig. 3).

To investigate the differential temperature noise dispersion under the influence of UHF irradiation on deionised water and 0.9% NaCl physiological solution, a number of experiments were conducted. The example of differential temperature recording in deionised water is given in Fig. 4, while Fig. 5 presents the graph of noise dispersion in NaCl solution.

From the analysis of average dispersion it can be concluded that the noise dispersion reaches its maximum value during UHF irradiation. To eliminate the influence of the external factors on the effect of noise dispersion, nine experiments were conducted were deionised water was effected with UHF. The recordings were made 40 min before UHF irradiation, 40 min during irradiation and 180 after the influence. The 40 min duration interval was chosen based on the previous experiments [8, 9] which revealed that the noise dispersion changed after 15 or 20 min from the start of UHF irradiation, while 180 min time period was chosen based on the assessment on behaviour of the water noise dispersion. All the results of temperature measurements were summed up and the average value of 9 recordings was plotted.



Fig. 2. Noise dispersion in NaCl solution with the maximum value of dispersion at 15 min of irradiation.



Fig. 3. Noise dispersion in NaCl solution with the maximum value of dispersion at 20 min of irradiation.



Fig. 4. Example of differential temperature recording in water for 260 minutes: for 40 min before irradiation, for 40 min during irradiation and for 180 min after the UHF was switched off.



Fig. 5. Noise dispersion in 0.9% NaCl solution recorded for 260 minutes: for 40 min before irradiation, for 40 min during irradiation and for 180 min after the UHF was switched off.

Experiments with 0.9% NaCl solutions were conducted with various time intervals after the influence of UHF irradiation. In the first case, the recordings were made in the same manner as for the distilled water, namely 40-40-180 min. Fig. 6 presents the noise dispersion for 20 recordings. In the second case, for the assessment of noise dispersion dynamics, the time interval after UHF irradiation influence was set to 1000 min, in order to determine the time interval during which the information memory transferred to the solutions is preserved after the low intensity UHF irradiation. Fig. 7 depicts the curve of differential temperature for 1080 min duration, while Fig. 8 illustrates the differential temperature noise dispersion for the same time duration.



Fig. 6. Noise dispersion in 0.9% NaCl solution for 20 recordings.



Fig. 7. Recording of differential temperature in 0.9% NaCl solution for 1080 min: for 40 min before irradiation, for 40 min during irradiation and for 1080 min after the UHF was switched off.

As one can see from the dynamics of noise dispersion, after over 13 hours after UHF irradiation the noise dispersion does not change. It can be assumed that "memory" of 0.9% NaCl solution, as well as of deionised water, exceeds this time period.



Fig. 8. Differential temperature dispersion in 0.9 % NaCl solution for 1080 min: for 40 min before irradiation, for 40 min during irradiation and for 1080 min after the UHF was switched off.

Therefore, long-term recordings are necessary to investigate the dynamics of water "memory", as supported by some authors in [10, 11], who suggest that water has a memory from few days to few months after the UHF irradiation. From the analysis of noise dispersion graphs it follows that noise dispersion during the influence of UHF irradiation reaches its

maximum value, but after this influence the dispersion is less than before it.

The structure of electrolytes aqueous solutions is described by various models, the most realistic of which seems Franck-Evans's model, which accounts for existence of three concentric regions around the solution particles. In the first region, as a result of ion influence the water dipoles are oriented in such a manner that they form fair rigid sheath around an ion. External region is the one where the structure of standard water is completely preserved, and the influence of ion has the effect of dielectric polarisation. Between these regions intermediate region exists, where the structure of water is disturbed by a competitive tension of the two extreme regions. Their influence is mutually opposite. The ions, depending on their charge sign, try to reorient the water dipoles accordingly. The region of standard structure acts on the intermediate region and attempts to preserve the tetrahedral structure [12].

The structure of NaCl aqueous solutions was studied by investigating its dielectric characteristics and irradiation spectra in broad wavelength range from 10 μ m to 10 cm [13-16]. These investigations didn't reveal any specific behaviour at small concentrations, including for physiological solutions, namely 0.9 % NaCl in deionised water, and is isotonic in relation to human cells. For the mass concentrations of 4.7 % and 21.7 %, specific behavioural features were observed, which were interpreted and accumulation of separate phases of water-salt solution [13].

Optimised model of hydro-separated ion pair of sodium chloride was given in [17], where the following bonds lengths have the values as given in Table 1.

Table 1. Bonds lengths values for ion pairs.

Ion pair	Bond length
$Na^+ - O(H_2O)$	0.211
Cl ⁻ - H (H ₂ O)	0.222
О - Н	0.099
О Н	0.155
$O(H_2O)O(H_2O)$	0.254
Na ⁺ - Cl ⁻	0.585

This means that in the nearest regions of ions there exists five water molecules, which are oriented according to the ion charge signs. Internal and intermediate sheath are within the radius of $R = \frac{1}{2} [0.585 + 0.211 + 0.222] = 0.509$ nm. Therefore, standard water surrounds hydro-separate ion pair of NaCl solution with radius R. Hexagonal plat mesh, which presents itself the structure of ice-type water frame, can be transformed into sphere surface or, more precisely, into "icosahedron" of any dimension, by substituting in it in a particular manner, the twelve hexagonal nods into the nods with five coordinations (pentamers) [18].

The surface of the system with six water molecules is $S_6 = A^2 \times 2.598$, while of the system with five molecules it equals $S_5 = A^2 \times 1.785$ (with A=0.254 nm), therefore, depending on the sphere radius R, it is easy to determine the number of molecules necessary to form such a sphere. The assessment of water molecules number, which are present in the second intermediate region, is made by accounting for a difference in volumes of hydro-separated ion pair and its surrounding

sphere. Using this method, each Na^+ - Cl⁻ ion pair needs 352-358 water molecules. The existence of the top and bottom values is due to the fact that correctness of the water molecules assembly and the presence of Bireme's defects were not accounted for. It was shown [18] that water molecules form fractal-cluster structure. Moreover, the order level of ideal diamond-type fractals is determined by pseudo-periods of triplet cores. The first-level cluster of the first order contains 356 water molecules. Therefore, for the "ideal" solution structure, each NaCl molecule (one ion pair) in solution needs 356 molecules or, in terms of the mass concentration, it corresponds to 0.9 % NaCl aqueous solutions, e.g. to physiological solution.

During the influence on physiological (ordered) solution with external physical factor, in particular with non-thermal intensity UHF irradiation, the processes of acceleration of the water dipoles combining into clusters – accumulation of the cluster energy – cluster breaking take place [19]. The ordered structure of the solutions is sensitive to disturbances, which considerably increase the fluctuations.

IV. CONCLUSIONS

Investigation of NaCl aqueous solutions in various concentrations has shown that the most sensitive to the external physical factors if the solutions with 0.9 % NaCl concentration, which corresponds to the physiological solution and concentration of NaCl in human blood. As the results of nonthermal intensity UHF irradiation influence on the water and physiological solution, the average values of differential temperature noise dispersion increases during the irradiation, and decreases after the irradiation, with the dispersion value being less as compared to non-irradiated solution. Water is very different from simple liquids formed by hydrate compounds of the sixth group of the periodic system, and most other liquids. One of the interesting properties of water is the "memory" effect, its means that water keeps properties for a long time after interaction with the external influences. Disordered motion of the liquid contributes to the temperature fluctuations, which depend on the impacts of high frequency. High frequency is able to increase the stability of the parameters of these oscillations.

ACKNOWLEDGMENT

This work is in part financially supported by the European Community's Seventh Framework Programme through the FP7-PEOPLE-2010-IEF Marie-Curie Action project 275201, Water-Spotcheck.

References

 K. M. Callahan, N. N. Casillas-Ituarte, M. Xu et al., "Effect of magnesium cation on the interfacial properties of aqueous salt solutions," Journal of Physical Chemistry A, vol. 114, no. 32, pp. 8359-8368, 2010.

- [2] L. A. Ferreira, P. Parpot, J. A. Teixeira et al., "Effect of NaCl additive on properties of aqueous PEG-sodium sulfate two-phase system," Journal of Chromatography A, vol. 1220, pp. 14-20, 2012.
- [3] E. Jager, A. Jager, T. Etrych et al., "Self-assembly of biodegradable copolyester and reactive HPMA-based polymers into nanoparticles as an alternative stealth drug delivery system," Soft Matter, vol. 8, no. 37, pp. 9563-9575, 2012.
- [4] B. Maribo-Mogensen, G. M. Kontogeorgis, and K. Thomsen, "Comparison of the Debye-Huckel and the mean spherical approximation theories for electrolyte solutions," Industrial and Engineering Chemistry Research, vol. 51, no. 14, pp. 5353-5363, 2012.
- [5] D. M. Zuckerman, M. E. Fisher, and S. Bekiranov, "Asymmetric primitive-model electrolytes: Debye-Huckel theory, critically, and energy bounds," Physical Review E - Statistical, Nonlinear, and Soft Matter Physics, vol. 64, no. 1 I, pp. 011206/1-011206/13, 2001.
- [6] A. I. Levin, Theoretical Fundamentals of Electrochemistry, Moskow: Mettalurgy, 1972.
- [7] A. A. Furman, M. P. Beldu, and I. N. Sokolov, Salt, Moskow: Chemistry, 1989.
- [8] P. Loshitsky, and V. Mamaev, "Investigation of changes properties of water and aqueous solutions of sodium chloride by the action EHF emission, nonthermal intensity," Medical Informatics and Engineering, vol. 1, pp. 53 - 60, 2008.
- [9] P. P. Loshitsky, and V. N. Mamaev, "Investigation of low intensity thermal influence of stochastic electromagnetic fluctuations of extremely high frequencies on physiological solution," Electronics and Communications, vol. 2, no. Special Issue "Problems in electronics", pp. 46-50, 2007.
- [10] V. D. Iskin, Y. V. Zavgorodnuy, N. M. Yatsenko et al., Biological effect of mirowaves, Moskow: Biophysics, 1987.
- [11] L. B. Oriabinskaya, P. P. Loshitsky, V. C. Mosienko et al., "Structured water and its role in biological systems functioning," Electronics and Communications, vol. 1, no. 6, pp. 183-189, 1999.
- [12] V. V. Silykov, Structure of single-atom fluids, water and aqueous solutions, Moskow: Science, 1976.
- [13] A. N. Romanov, "Influence of mass mineral salts concentration on dielectric characteristics of their aqueous solutions in microwave region," Radiotechnology and Electronics, vol. 49, no. 10, pp. 1235 -1242, 2004.
- [14] A. Y. Zacetsky, A. C. Lileev, and A. K. Liaschenko, "Dielectric properties of NaCl aqueous solutions on EHF region," Journal of Inogranic Chemistry, vol. 39, no. 6, pp. 1035-1040, 1994.
- [15] A. K. Liaschenko, T. A. Novskova, A. C. Lileev et al., "Rotational motion of water molecules in hydrant ions shealths and broadband dielectric spectra of the electrolytes solutions," Journal of Physical Chemistry, vol. 67, no. 8, pp. 1615 - 1622, 1993.
- [16] B. M. Liberman, and V. I. Gayduk, "Calculations of the dielectric and illumination spectra of electrolytes aqueous solutions in broad wavelength range. Hybrid model.," Radiotechnology and Electronics, vol. 44, no. 1, pp. 97-103, 1999.
- [17] V. A. Shaposhnik, "Kinetic theory of electrolytes aqueous solutions," Vestnik VGU: Chemistry, Biology, Farmocology, vol. 2, pp. 81 - 85, 2003.
- [18] N. A. Bulenkov, "On the possible role of hydrotation as the leading integral factor in biosystem organisation at various hierarchy levels," Biophysics, vol. 36, no. 2, pp. 181 -243, 1991.
- [19] P. P. Loshitsky, M. V. Kurik, and N. A. Nikolov, "Reaction of aqueous solutions to the influece of low-intensity physical factors," Electronics and Communications, vol. 16, pp. 80-84, 2002.