

---

<https://doi.org/10.15407/ujpe69.9.632>

I. IGNATOV,<sup>1</sup> Y. MARINOV,<sup>2</sup> F. HUETHER,<sup>3</sup> G. GLUHCHEV,<sup>4</sup> M.T. ILIEV<sup>5</sup>

<sup>1</sup> Scientific Research Center of Medical Biophysics  
(32, N. Kopernik Str., Sofia 1111, Bulgaria; e-mail: mbioph@abv.bg)

<sup>2</sup> Georgi Nadjakov Institute of Solid State Physics, Bulgarian Academy of Sciences  
(Sofia 1784, Bulgaria)

<sup>3</sup> EVODROP AG  
(Brüttisellen 8306, Zürich, Switzerland)

<sup>4</sup> Institute of Information and Communication Technologies, Bulgarian Academy of Sciences (BAS)  
(Sofia 1113, Bulgaria)

<sup>5</sup> Faculty of Physics, Sofia University  
(Kliment Ohridski Str., Sofia 1000, Bulgaria; e-mail: mbioph@abv.bg)

## MODELING WATER CLUSTERS: SPECTRAL ANALYSES, GAUSSIAN DISTRIBUTION, AND LINEAR FUNCTION DURING TIME

---

*Our experimental and theoretical studies have consistently revealed the presence of water clusters in various environments, particularly under hydrophobic conditions, where slower hydrogen ion interactions prevail. Crucial methods like Nuclear Magnetic Resonance (NMR) and Fourier Transform Infrared (FTIR) method have played a pivotal role in our understanding of these clusters, unveiling their potential medical applications. The stability and behavior of these clusters can be influenced by factors such as metal ions' presence, leading to stable clusters' formation. This potential for medical applications should inspire hope and further research. Moreover, our research has revealed that water clusters exhibit characteristics of dissipative structures, demonstrating the self-organization under physical, chemical, or thermal changes akin to Rayleigh–Benard convection cells. This dynamic and significant behavior supports the notion that water's role transcends simple chemistry, potentially influencing biological processes at a fundamental level. The interaction of water clusters with their environment and the ability to maintain non-equilibrium states through the energy exchanges further underscores their complexity and significance in both natural and technological contexts. Water filtration is a process for improving water quality. The effect is re-structuring hydrogen bonds and structuring water clusters, most of which are hexagonal. In our research, we applied filtered water using patented EVODrop Swiss technology.*

*Keywords:* water clusters, Gaussian distribution, linear function, spectral analyses

### 1. Introduction

Water, a substance of profound importance due to its unique properties and role as a universal solvent, continues to be a subject of intense scientific interest. Particularly, its structure and behavior at the molecular level have been a source of fascination. With their polar nature and electromagnetic hy-

Citation: Ignatov I., Marinov Y., Huether F., Gluhchev G., Iliev M.T. Modeling water clusters: spectral analyses, gaussian distribution, and linear function during time. *Ukr. J. Phys.* **69**, No. 9, 632 (2024). <https://doi.org/10.15407/ujpe69.9.632>.

© Publisher PH “Akademiya” of the NAS of Ukraine, 2024. This is an open access article under the CC BY-NC-ND license (<https://creativecommons.org/licenses/by-nc-nd/4.0/>)

drogen bonds, water molecules interact complexly, influenced by van der Waals forces and dipole-dipole interactions. This intricate behavior defies a simple explanation based on single parameters, underscoring the need for comprehensive studies.

The presence of water clusters is confirmed through various methods. The clusters consist of two, three, four, five, six, and more water molecules. The most stable clusters are accepted to consist of six water molecules. Configurations of up to 30 water molecules are also formed. They are combinations of hexagonal (hexamer) and smaller cluster formations.

Experimental studies demonstrate the existence of water clusters for a longer time.

The most indicative experiment is in a hydrophobic environment with slower hydrogen ions ( $H^+$ ) or proton interactions between water molecules in clusters. A signal from the water clusters is obtained within three days [1].

Density Functional Theory (DFT) was used to analyze the stability of water clusters depending on the number of hydrogen bonds and the intermolecular interactions of the water clusters [2]. Using Nuclear Magnetic Resonance (NMR) and Fourier Transform Infrared (FTIR) methods, dimer, tetramer, and hexamer water clusters have been studied with applications in medicine [3].

Water can be considered a convenient, but complex model for investigating cooperative interactions and structural organization of molecular clusters sized over the nano-scales. NMR-based parameters demonstrate the number of water molecules and hydrogen bonds in water clusters [4, 5].

In the presence of an impact on water, new hydrogen bonds are formed, and water molecules restructure into clusters. An equilibrium is established, where they maintain balance with hydrogen ions. These effects are measured using pH, NMR, and spectral methods. Let us consider chemically pure water with a pH of 7.0. If this water is in an open container, it undergoes a chemical reaction with carbon dioxide. Carbonic acid is formed, and the pH can reach a certain level after a certain time, depending on the water volume, temperature, humidity, *etc.* Interestingly, an equilibrium occurs between dissolved carbonic acid and protons ( $H^+$ ). The quantity of  $H^+$  is measured by pH. The result is sustained over time. After one or two weeks, or longer, it remains unchanged. This demonstrates that unstable hydro-

gen ions can reach an equilibrium state. Stable cluster formations are created with metal ions such as  $Ca^+$ ,  $Ca^{2+}$ ,  $Mg^{2+}$ ,  $Zn^{2+}$ , *etc.*, even in small quantities. In these systems, the ion balance leads to stable clusters during the time. In drinking water, we have dissolved positive and negative ions. Stable water clusters are formed around the metal ions. Water clusters of calcium ions  $Ca^+(H_2O)_n$  have been studied in [6–8]. There are results with magnesium ions structured in clusters with the formula  $Mg^{2+}(H_2O)_n$  can be found in [9]. The zinc ions are in clusters with the formula  $Zn^{2+}(H_2O)_n$  were studied in [10]. There are stable clusters water clusters  $[H_2O]_n$  around the negative ions such as  $CO_3^{2-}$  [11],  $SO_4^{2-}$  [12], and  $PO_4^{3-}$  [13].

Experiments were conducted using a permanent magnetic field with inductions ranging from 1 to 10 T on water [14]. The experiments were performed with a magnetic field with induction 0.3 T with research for the stable effects during 24 h [15]. As the magnetic field increases, the number of hydrogen bonds increases as well. Using the Molecular Dynamic Simulation method, the structuring of water clusters was demonstrated. Changes in water parameters are preserved during the time. The self-diffusion coefficient changes while preserving the parameters.

Analyses of hydrogen bonds are also carried out using the hierarchical cluster structure method [16].

The Nobel laureate Prigogine created the theory of dissipative structures [17]. The relationship between dissipative structures and water clusters can be seen as dependencies between systems' dynamic and non-equilibrium characteristics. For instance, water clusters may self-organize within a water environment under the physical fields and chemical compounds. This is similar to the dynamics of dissipative structures. Thus, we can observe interesting examples of a complex behavior in systems far from thermodynamic equilibrium. Water clusters interact with their environment through the energy exchange, such as the heat absorption or release during the formation or breakdown of hydrogen bonds. This process contributes to maintaining the clusters in a non-equilibrium state akin to dissipative structures.

The water clusters are dissipative structures similar to self-organized Rayleigh–Benard connective cells [18]. Self-organized hexagonally patterned microdroplet clusters over locally heated water surfaces have been recently observed. These clusters are stable in time and result in increases after 80 seconds. The

time-dependent Voronoi entropy of the clusters was calculated using software developed by the Department of Physics and Astronomy at the University of California.

The paper [19] presents theoretical and experimental proofs for dissipative structures. Three physical perturbations were performed to form dissipative structures in liquid water with low energy impact. The methods included extremely diluted solution (EDS), iteratively filtered water (IFQ), and iteratively carbonated water. Special attention is given to the recent achievement that such structures exhibit remarkable persistence even in the solid phase, presenting as large quantities of supramolecular water aggregates. Each aggregate acts as a nucleus, measuring hundreds of nanometers in size. Observations of dissipative structures occurred under ambient pressure and temperature conditions using reproducible experimental techniques. An analysis of the nature of these dissipative structures is provided, with an explanation grounded in the thermodynamics of far-from-equilibrium systems and irreversible processes. Furthermore, their spontaneous quantum origin is demonstrated.

Josephson, a theoretical physicist and Nobel laureate, has indeed delved into the field of water clusters, particularly focusing on their quantum properties and potential implications.

In his article "Possible Role of Josephson Tunneling and the Operation of Life," Josephson discusses the potential involvement of Josephson tunneling, a quantum phenomenon, in biological systems, including its role in water cluster behavior within living organisms [20].

Josephson's interest in the quantum aspects of water's behavior extends to its role in biological processes, suggesting that quantum coherence could be relevant to understanding phenomena such as the energy transfer and information processing in biological systems.

In 1988, Del Giudice, Preparata, and Vitiello described the coherent dynamics structures of water and its implications for biological systems. The investigation proposes that water molecules can exhibit coherence at the quantum level, forming coherent domains or clusters [21].

The Italian team explores the possibility that water molecules can organize into coherent structures, which may have profound implications for under-

standing various biological phenomena, including the cell signaling, energy transfer, and consciousness.

In 2015, Montagnier, Del Giudice, Vitiello *et al.*, performed very interesting experiments on the informational properties of water [22]. The article describes the experimental conditions under which diluted aqueous solutions of bacterial DNA can emit low-frequency electromagnetic signals. Analyses of the effects were shown in [23].

In 2017, McDermott *et al.* published a paper on structuring chiral water superstructures surrounding DNA under ambient conditions [24]. The publication confirms the results obtained by Montagnier *et al.* on the effects of information transfer via DNA with magnetic fields [25]. Under ambient conditions, chiral water structures surrounding DNA reflect a process, where water molecules can organize into specific, chiral structures around DNA molecules under ordinary conditions. This implies that the microenvironment around DNA in living cells includes water that is not randomly arranged, but has a specific organized form with stable water structures.

In 1996, Liu, Cruzan, and Saykally connected chiral water structures with trimer and pentamer water clusters [26].

Proton transfer and chiral conversion via hydrogen bonds are important in applications such as chiral recognition, enzymatic catalysis, and drug preparation.

Vibrational circular dichroism (VCD) spectra exhibit distinct chiral recognition peaks from 3000 to 3500  $\text{cm}^{-1}$ . The molecular orbitals involved in the interlayer interaction predominantly comprise O 2p atomic orbitals. The energy of these orbitals increases by up to 0.1 eV due to the CIPT processes, indicating corresponding recognition between monolayer water clusters [27].

The concept indicates that clustered water (CW) possesses certain informational properties. A series of investigations were performed with Quantum FAFA equipment to register the frequency signals in water. The resonant frequency profile of CW was notably distinct from that of distilled water (DW). In separate experiments, standard CW served as the control, while other samples of CW were exposed to an additional low-frequency signals. The results indicate the presence of several new resonant frequency peaks in the exposed CW compared to the non-exposed CW control. These results show a novel approach to un-

raveling the informational properties of the Meridian system and its association with bio-signal networks [28].

The Nobel laureate Agre demonstrates that, till 1990, there was no clear understanding of the water transport across a cell membrane. He discovered aquaporins and the informational effects of hydrogen bonds. Water molecules may thus tumble from one set of tetrahedral hydrogen bonds to the next and pass through the channel without noticing it [29].

In a tetrahedral cluster, four others form a pentamer and surround the central water molecule.

One realistic model for the fusion of two membranes assumes a proteinaceous initial fusion pore, since biological “fusion pores” can be as small as ion channels or gap junctions. However, another model proposes a lipid initial pore. Between the contacting monolayers, a semi-confluent zone is created, where the continuity between the two adherent membranes is lipid rather than aqueous [30, 31].

With its potential applications in various fields, the WAT FOUR water cluster model combines approximately 11 water molecules with a tetrahedral cluster of four beads. This model increases bead-bead interactions and degrees of freedom, opening up a new possibilities for research and development [32, 33].

Water clusters play an active and essential role in biochemical reactions within living organisms. Understanding their formation and behavior is crucial for comprehending water’s interactions. Further research into water clusters, particularly in plant cells, holds promise for uncovering new insights and applications in various fields [34].

Water deficits are a reality to which procedures are increasingly susceptible due to climate change and its consequent shift in rainfall regimes. As such, technology and management that seek to reduce the impacts caused by these changes are of fundamental importance and need to be elucidated.

The investigation aims to create water cluster models after the filtering with an EVOdrop device, using spectral analyses, a Gaussian distribution, and a linear function.

## 2. Methods

### 2.1. Water Samples

Water samples were prepared for investigation at  $t = 22$  °C. The control samples consist of tap water,

while the experimental samples are filtered from tap water using patented Swiss technology [35, 36].

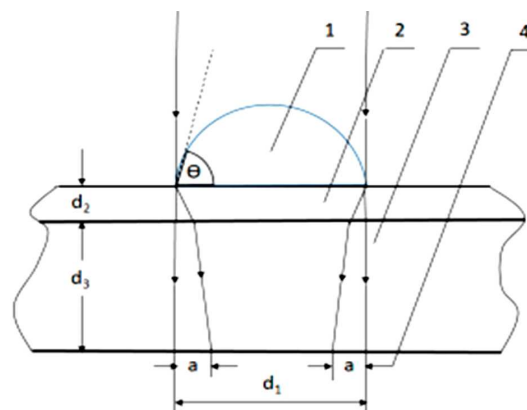
### 2.2. Non-equilibrium energy spectrum (NES) and differential non-equilibrium energy spectrum (DNES) spectral analyses

The wetting angle  $\theta$  was measured with a spectral device invented by A. Antonov [37–39]. The water drops were evaporated in a sealed chamber at a stable temperature of 22 °C (Fig. 1). The drops were placed on a 350  $\mu\text{m}$  thick BoPET (biaxially oriented polyethylene terephthalate) sheet.

The device has the following technical features:

- Monochromatic filter with wavelength  $\lambda = 580 \pm 7$  nm.
- Water evaporation angle ranging from 72.3 deg to 0 deg.
- The measured range of energy of hydrogen bonds among water molecules is  $\lambda = 8.9\text{--}13.8$   $\mu\text{m}$  or  $E = -0.08\text{--}-0.1387$  eV [35, 36].

Luck considers that water consists of hydrogen bonds between one water molecule’s hydrogen atom and another’s oxygen [40]. Most of them are bound by the energy of the connection ( $-E$ ), and the remaining are free ( $E = 0$ ). It is accepted that  $E$  has a negative value. This is known as the Luck two-state model [41]. The number of hydrogen bonds between the hydrogen atom of one water molecule and the oxygen of another one in a particular volume of water is twice as high as the number of molecules it contains.



**Fig. 1.** Operating principle of the method for measuring the wetting angle of liquid drops on a hard surface: drop (1), thin mylar sheet (2), glass plate (3), refraction ring width (4). The wetting angle  $\theta$  is a function of  $a$  and  $d_1$

A part of the hydrogen bonds is restructured in proximity to the spherical part of the drop surface, and, as a result, we obtain the dependence between the surface tension  $\delta$  and the hydrogen bond energy [42].

Based on the statistical mechanics, the following formula is valid for the surface tension water drop-air [43]

$$\delta = -kTI \ln[1 + \alpha/[\exp(-\beta E) + \alpha]]. \quad (1)$$

Here,  $k$  is the Boltzmann constant,  $\beta = 1/kT$ ,  $T$  – is the absolute temperature,  $E$  – is the hydrogen bond energy,  $\alpha$  is the ratio of two subvolumes of the phase space related to the structuring and restructuring of hydrogen bonds,  $\alpha = 8 \pm 8$  and  $I = 5.03 \times 10^{18} \text{ m}^{-2}$  is the density of water molecules in the hydrophobic surface layer.

The values of  $E$  and  $\alpha$  are determined by comparison with the experiment. Expression (1) explains the fraction  $C$  of the actual surface tension  $\gamma$ , i.e.,  $\delta = C\gamma$  [42, 44]. According to [44], the contribution of the non-hydrogen bond interaction amounts to 20% of the real value of  $\gamma$  and  $C = 4/5$ .

Consider the Helmholtz free surface energy  $F = \gamma\Sigma$ , where  $\Sigma$  is the spherical part of the drop surface [18].

At the instant of mechanical equilibrium,  $F$  should be minimal, i.e.,  $dF = 0 = d(\gamma\Sigma)$  [42]

$$0 = \gamma\Sigma - \gamma_0\Sigma_0. \quad (2)$$

The expressions for  $\Sigma_0$  and  $\Sigma$  are as follows [38]:

$$\Sigma = \pi D^2/2(1 + \cos \theta); \quad \Sigma_0 = \pi D^2/2(1 + \cos \theta_0), \quad (3)$$

$$-E/kT = C\gamma/IkT, \quad (4)$$

$$E = C\gamma_0(1 + \cos \theta)/I(1 + \cos \theta_0). \quad (5)$$

During the process, the wetting angle changes in discrete steps and characterizes the average energy of hydrogen bonds as follows:

$$\theta = \arccos(-1 + bE), \quad (6)$$

where  $\theta$  is the wetting angle,  $E$  is the average energy of hydrogen bonds, and  $b = I(1 + \cos \theta_0)/C\gamma_0$  is a temperature-dependent parameter [31, 32]. The development of the method is the Non-equilibrium energy spectrum (NES) and Differential non-equilibrium spectrum (DNES) [38, 39].

This non-equilibrium energy spectrum (NES) is measured in  $\text{eV}^{-1}$ . DNES is defined as the difference [38, 39]:

$$\Delta f(E) = f(\text{water sample}) - f(\text{control water sample}), \quad (7)$$

DNES is measured in  $\text{eV}^{-1}$ , where  $f(*)$  denotes the evaluated energy [38, 39, 45, 46].

### 3. Gaussian Distribution of Water Clusters

The distribution of clusters according to the average energy of the hydrogen bonds in them with the Gaussian function in the following formula [47]:

$$y = [1/s\sqrt{2\pi}] \exp[-(x - x_s)^2/2s^2]. \quad (8)$$

The value  $x$  represents the average energy of hydrogen bonds in a particular group of water clusters. The quantity  $y$  is the number of clusters with the same energy of hydrogen bonds (denoted by  $x$ ) and the same number of water molecules. Essentially,  $y$  gives the number of clusters with the same energy of hydrogen bonds (denoted by  $x$ ) and the same number of water molecules. Essentially,  $y$  gives the cluster's probability density at a specific energy level,  $x$ .  $X_s$  is the expected value, and  $x_s$  represents the average energy of hydrogen bonds across all groups of clusters. The quantity  $s$  is the standard deviation, which measures the dispersion of the energy values around the mean ( $x_s$ ). Low values of  $s$  indicate that the energies of the hydrogen bonds in the clusters are closely grouped around the mean, suggesting the uniformity in the bonding energy across clusters. Conversely, a larger  $s$  would indicate a wider variation in the energy of hydrogen bonds, implying a more diverse set of clusters in terms of the energy stability.

## 4. Results

### 4.1. Gaussian distribution of water clusters

The parameters of water clusters were described by Gaussian distribution [47].

The Non-equilibrium energy spectrum (NES) and Differential non-equilibrium energy spectrum (DNES) analyze parameters of hydrogen bonds between water molecules due to external influences or dissolving inorganic and organic substances.

In a previous model, the number of clusters of a particular type in water is related to their average energy of hydrogen bonds [47]. The distribution of the numbers of particular types of clusters is Gaussian.

**Table 1. Results of NES spectrum of EVOdrop filtered water and control samples of tap water over 7 days**

The average energy of hydrogen bonds ( $-E$ ) (eV)	Day							Average result
	1 <sup>st</sup>	2 <sup>nd</sup>	3 <sup>d</sup>	4 <sup>th</sup>	5 <sup>th</sup>	6 <sup>th</sup>	7 <sup>th</sup>	
EVOdrop filtered water	0.1257	0.1254	0.1251	0.1250	0.1248	0.1249	0.1247	0.1251
Control sample (tap water)	0.1132	0.1136	0.1137	0.1139	0.1130	0.1128	0.1137	0.1134

**Table 2. Gaussian distribution results from the results with the NES spectrum of EVOdrop filtered water**

Value	Day							Sum
	1 <sup>st</sup>	2 <sup>nd</sup>	3 <sup>d</sup> day	4 <sup>th</sup>	5 <sup>th</sup>	6 <sup>th</sup>	7 <sup>th</sup>	
$y_i$	266.5563	787.5874	1180.1	1085.7	787.5874	962.5653	594.723	5624.1
Normalized quantities	0.0474	0.14	0.201	0.193	0.14	0.171	0.1057	$\approx 1$

Mehandjiev *et al.* described the distribution of clusters according to the average energy of the hydrogen bonds in them with the Gaussian function in the following formula [47]:

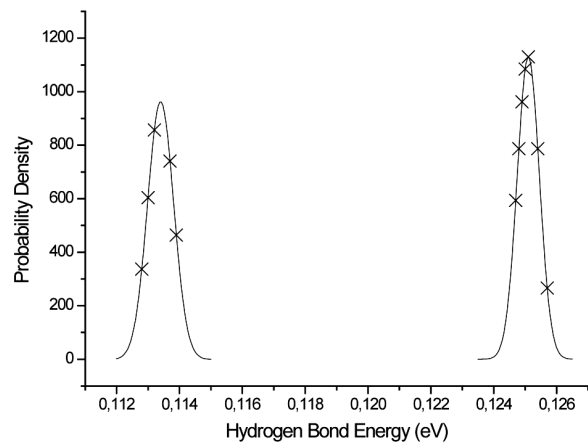
$$y = [1/s\sqrt{2\pi}] \exp[-(x - x_s)^2/2s^2]. \tag{9}$$

The Gaussian distribution has the following two components:  $[1/s\sqrt{2\pi}]$  is the normalizing factor ensuring that the total area under the Gaussian curve equals 1. It adjusts the height of the curve based on the deviation,  $s \exp[-(x - x_s)^2/2s^2]$  is an exponential component that shapes the curve, where the distribution of cluster energies around the mean ( $x_s$ ) follows a bell-shaped curve. The exponent shows that, as the difference between  $x$  and  $x_s$  increases, the value of  $y$  decreases exponentially, which is typical of a normal distribution.

Let us consider the results obtained from water filtration using a patented EVOdrop device [35, 36]. Measurements have been conducted over 7 days using NES and DNES methods.

Table 1 illustrates the results of NES spectrum of EVOdrop filtered water and control samples of tap water over 7 days. With the application of the *t*-test Student, the results are:  $p < 0.001$ ;  $r = -0.013$ .

Table 2 shows the Gaussian distribution results from the results with NES spectrum [47, 48] of EVOdrop filtered water. The value  $y_i$  is the probability density. The normalized quantities are for the probability.



**Fig. 2.** Comparative analysis of the Gaussian probability distribution according to the average energies ( $-E$ ) of hydrogen bonds of experimental and control groups

Table 3 presents the Gaussian distribution results from the NES spectrum of the control tap water sample. The quantity  $y_i$  is the probability density. The normalized quantities are for the probability.

Fig. 2 illustrates a comparative analysis of the Gaussian probability distribution according to the average energies of hydrogen bonds of experimental and control groups. Figure 2 depicts that the probability density for the experimental group is 1180.1, whereas, for the control group, it is 857.5. Comparing these two values reveals that the probability density in the experimental group is higher around the mean value compared to the control group. This indicates

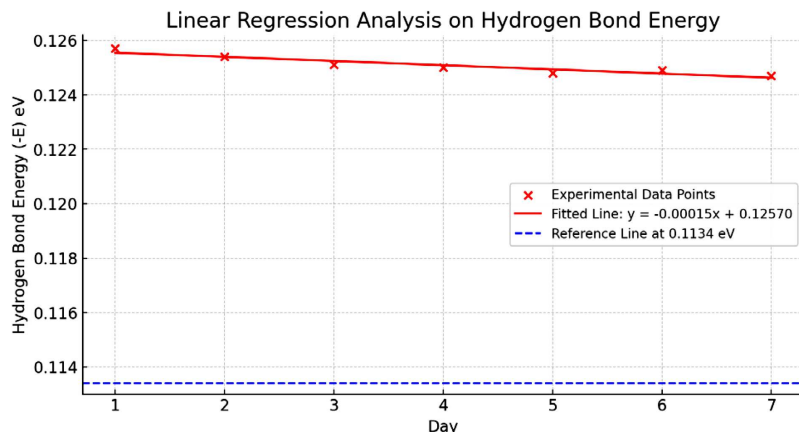


Fig. 3. Linear function  $E(t) = -0.000154t + 0.1257$  of the experimental sample with filtered water EVOdrop

Table 3. Gaussian distribution results from the results with the NES spectrum of the control tap water sample

Value	Day							Sum
	1 <sup>st</sup>	2 <sup>nd</sup>	3 <sup>d</sup> day	4 <sup>th</sup>	5 <sup>th</sup>	6 <sup>th</sup>	7 <sup>th</sup>	
$y_i$	857.5	857.5	741.115	764.704	604.228	337.143	741.115	4603.305
Normalized quantities	0.1863	0.1863	0.161	0.101	0.1313	0.073	0.161	$\approx 1$

Table 4. Results with EVOdrop filtered water and the NES spectrum of *Haberlea rhodopensis* Friv

Time	Day							Average result
	1 <sup>st</sup>	2 <sup>nd</sup>	3 <sup>d</sup>	4 <sup>th</sup>	5 <sup>th</sup>	6 <sup>th</sup>	7 <sup>th</sup>	
$(-E)$ (eV)*	0.1203	0.1201	0.1200	0.1199	0.1201	0.1201	0.1997	0.1996

\*The average energy of hydrogen bonds  $(-E)$  (eV) 1% solution of *Haberlea rhodopensis* Friv in EVOdrop water.

that the data in the experimental group are more concentrated around the mean value than in the control group.

The higher probability density around the mean value in the experimental group with EVOdrop filtered water indicates that the values for the energy of hydrogen bonds are more distributed around the mean value.

This demonstrates the greater stability and less variability in the strength of hydrogen bonds between water molecules in clusters. The investigation illustrates that most of them are hexagonal [45].

#### 4.2. Linear function

In the research, we applied a linear function of the type  $y = mx + b$ , where  $y$  is the dependent value of the

energy of the hydrogen bonds,  $x$  is the independence variable (time),  $m$  is the slope coefficient, and  $b$  is the intercept of the line with the  $y$ -axis.

The linear function  $E(t) = -0.000154t + 0.1257$  illustrates the stability in time of the energy of hydrogen bonds of water molecules and clusters (Fig. 3) of the experimental sample with filtered water EVOdrop.

The slope  $(-0.000154)$  indicates a slight decrease in the energy of hydrogen bonds with each elapsed unit of time. The correlation coefficient is  $r = -0.013$ .

The following conclusions from the linear function are valid. The slope indicates that the average energy of the hydrogen bonds decreases by approximately  $(-0.00015 \text{ eV})$  per day. The intercept of

( $-0.1253$  eV) is the initial value of the energy of the hydrogen bonds, which would be measured on the day zero. This is the theoretical start of the measurements. The correlation coefficient  $r = -0.013$  indicates no correlation between the samples. This indicates an inverse dependence between the time and energy of hydrogen bonds. The  $p$ -value  $0.000048$  is very low, indicating a statistically significant linear relationship between time and the energy of the hydrogen bonds in the data. This linear function can be used to predict the energy of the hydrogen bonds in the future days.

#### 4.3. Binary water clusters and bigger associates with the Bulgarian endemic plant *Haberlea rhodopensis* Friv

The water clusters were proved during osmosis and diffusion [49]. The osmotic process is essential for plants, and the authors showed indicators for hydration of plant cells and protection against osmotic stress [50].

The Bulgarian endemit *Haberlea rhodopensis* Friv is related to the interaction between the water molecules during the biosis–anabiosis–biosis cycle. Binary water clusters were proved [51].

The method was used to investigate the EVOdrop filtration technology of tap water and its enrichment with hydrogen in terms of hydration and water molecule clustering with *Haberlea rhodopensis* Friv (Table 4) [52]. The statistically significant results demonstrated that EVOdrop treatment shifts the hydrogen bonds' energy distribution toward larger values and the corresponding formation of local maxima.

The EVOdrop filtration technology enriches tap water with hydrogen [48], notably affecting water molecules' hydration dynamics and clustering.

According to the control sample with tap water, the treatment alters the energy distribution of hydrogen bonds, shifting it toward the highest values and forming bigger peaks. This suggests that EVOdrop technology could potentially enhance water quality by modifying its molecular structure, thereby offering benefits in terms of hydration efficiency and, possibly, other health-related aspects as the positive effects of Hydrogen-rich water after the hydration with myeloid tumor of hamsters [53].

## 5. Conclusions

Extensive research into water clusters highlights their pivotal role in physical and biological systems, elu-

cidating their complex molecular interactions and structural dynamics through techniques like NMR, FTIR spectroscopy, and DFT. These clusters exhibit notable stability and reactivity under various conditions, which are crucial for biological functions such as the proton transfer and DNA structuring.

Theoretical studies and practical experiments, including those by Nobel laureates Prigozhin, Montagnier, Josephson, and Agre, have opened new perspectives on the quantum properties of water clusters. They suggest their intricate involvement in cellular processes through quantum coherence and tunneling mechanisms.

Filtration can enhance water quality, which restores hydrogen bonds between water molecules.

During this process, water clusters are organized with a significant proportion adopting hexagonal structures. We employed the patented EVOdrop Swiss technology in our research and analysis. The effects of this technology were demonstrated through the analysis of water cluster models using spectral analyses, Gaussian distribution, and linear function. These methods allowed us to elucidate the structural changes and improvements in the molecular arrangement of water post-filtration.

The Gaussian distribution analysis indicates that probability density in the experimental group with EVOdrop filtered water is higher around the mean value than in the control group with tap water. This suggests a greater concentration of data points around the mean in the experimental group, implying more consistency in the measurement obtained. Moreover, the elevated probability density around the mean value in the experimental group suggests that the energy values of hydrogen bonds are more consistent around the mean. This demonstrates a reduced variability in the energy of hydrogen bonds within the water clusters.

The finding indicates enhanced stability and reduced variability in the strength of hydrogen bonds among water molecules within clusters. The linear function  $E(t) = -0.000154t + 0.1257$  exemplifies the temporal stability of the hydrogen bond energy in clusters.

Furthermore, the results from EVOdrop filtration technology on *Haberlea rhodopensis* Friv. demonstrate that this method enriches the water with molecular hydrogen, significantly influencing the hydration dynamics and clustering water molecules.



1. K. Oka, T. Shibue, N. Sugimura, *et al.* Long-lived water clusters in hydrophobic solvents investigated by standard NMR techniques. *Sci Rep.* **9**, 223 (2019).
2. A.D. Kulkarni. Unraveling hydrogen bonded clustering with water: Density Functional Theory Perspective. *Book: Density Functional Theory – Recent Advances, New Perspectives and Appl.* **10**, 173 (2021).
3. K. Kristinaityte, L. Dagys, J. Kausteklis *et al.* NMR and FTIR studies of clustering of water molecules: From low-temperature matrices to nano-structured materials used in innovative medicine. *J. Mol. Liq.* **235**, 1 (2017).
4. N. Sun, Z. Li, N. Qiu, X. Yu *et al.* Ab initio studies on the clathrate hydrates of some Nitrogen- and Sulfur-containing gases. *J. Phys. Chem. A* **121**, 2620 (2017).
5. N. Yang, C.H. Duong, P.J. Kelleher, M.A. Johnson. Capturing intrinsic site-dependent spectral signatures and lifetimes of isolated OH oscillators in extended water networks. *Nat. Chem.* **12**, 159 (2020).
6. E. Kochanski, E. Constantin. Theoretical and experimental studies of the system  $\text{Ca}^+(\text{H}_2\text{O})_n$  for  $n = 1$  to 10. *J. Chem. Phys.* **87**, 1661 (1987).
7. H. Watanabe, S. Iwata. Molecular orbital studies of the structures and reactions of a singly charged Calcium ion with water clusters,  $\text{Ca}^+(\text{H}_2\text{O})_n$ . *J. Phys. Chem. A* **101**, 487 (1997).
8. I.R. Ariyaratna, E. Miliordos. Geometric and electronic structure analysis of calcium water complexes with one and two solvation shells. *Phys. Chem. Chem. Phys.* **39** (2020).
9. A.A. Delgado, D. Sethio, E. Kraka. Assessing the intrinsic strengths of ion–solvent and solvent–solvent interactions for hydrated  $\text{Mg}^{2+}$  Clusters. *Inorganics.* **9**, 31 (2021).
10. P. Wang, Y. Su, R. Shi *et al.* Structures and spectroscopic properties of hydrated zinc(II) ion clusters  $[\text{Zn}^{2+}(\text{H}_2\text{O})_n$  ( $n = 1-8$ )] by Ab initio Study. *J. Clust. Sci.* **34**, 1625 (2023).
11. A.K. Pathak, T. Mukherjee, D.K. Maity. IR spectra of carbonate-water clusters,  $\text{CO}_3^{2-}(\text{H}_2\text{O})_n$ : A theoretical study. *Synthesis and Reactivity in Inorganic, Metal-Organic, and Nano-Metal Chem.* **38**, 76 (2008).
12. R. Custelcean, N.J. Williams, Ch.A. Seipp. Aqueous sulfate separation by crystallization of sulfate–water clusters. *Angewandte Chemie.* **54** (36), 10525 (2015).
13. W.W. Rudolph, G. Irmer. Raman and infrared spectroscopic investigations on aqueous alkali metal phosphate solutions and density functional theory calculations of phosphate–water clusters. *Appl. Spectr.* **61**, 1312 (2007).
14. K.-T. Chang, Ch.-T. Weng. (2006) The effects of an external magnetic field on the structure of liquid water using molecular dynamic simulation. *J. Appl. Phys.* **100**, 043917 (2006).
15. I. Ignatov, I.K. Stankov. Parameters and effects of magnetic field and potassium carbonate in water, applications. *Ukr. J. Phys.* **69**, 321 (2024).
16. Y. Gao, H. Fang, K. Ni. A hierarchical clustering method of hydrogen bond networks in liquid water undergoing shear flow. *Sci Rep.* **11**, 9542 (2021).
17. A.A. Chernyuk, V.I. Sugakov. Ordered dissipative structures in exciton systems in semiconductor quantum wells. *Phys. Rev. B* **74**, 085303 (2006).
18. X. Zhu, Q. Zhou. Flow structures of turbulent Rayleigh–Bénard convection in annular cells with aspect ratio one and larger. *Acta Mech. Sin.* **37**, 1291 (2021).
19. V. Elia, R. Germano, E. Napoli. Permanent dissipative structures in water: The matrix of life? Experimental evidences and their quantum origin. *Curr. Top. Med. Chem.* **15**, 559 (2015).
20. B.D. Josephson. Possible role of Josephson tunneling in the operation of life. *Physica Scripta* **40**, 786 (1991).
21. E. Del Giudice, G. Preparata, G. Vitiello. Water as a free electric dipole laser. *Phys. Rev. Lett.* **61**, 1085 (1988).
22. L. Montagnier, E. Del Giudice, E.G. Vitiello *et al.* Transduction of DNA information through water and electromagnetic waves. *Electromagn Biol Med.* **34**, 106 (2015).
23. M. Scalia, P. Avino, M. Sperini *et al.* Some observations on the role of water states for biological and therapeutical effects. *Inov. Biosyst. Bioeng.* **231**, 149 (2018).
24. H.L. McDermott, H. Vanselow, S.A. Corcelli, P.B. Petersen. DNA’s chiral spine of hydration. *ACS Cent. Sci.* **3**, 708 (2017).
25. L. Montagnier, J. Aissa, G. Vitiello *et al.* Water bridging dynamics of polymerase chain reaction of Gauge theory paradigm of Quantum field. *Water.* **9**, 339 (2017).
26. K. Liu, J.D. Cruzan, R.J. Saykally. Water clusters. *Science.* **271**, 929 (1996).
27. B. Wang, W. Jiang, Y. Gao, Y. *et al.* Chirality recognition in concerted proton transfer process for prismatic water clusters. *Nano Res.* **9**, 2782 (2016).
28. J. Pan, L. Lorenzen, F. Carrillo *et al.* Clustered water and bio-signal networks. *Conference on IEEE Cybernetics and Intelligent Systems* (2004).
29. A. Engel, Y. Fujiyoshi, P. Agre. The importance of aquaporin water channel protein structures. *EMBE J.* **19**, 800 (2000).
30. A. Chanturiya, L.V. Chernomordick, J. Zimmerberg. Flickering fusion pores comparable with initial exocytotic pores occur in protein-free phospholipid bilayers. *PNAS* **94** (26), 14423-28 (1997).
31. J. Milhaud. New insights into water–phospholipid model membrane interactions. *Biochim. Biophys. Acta* **1663** (1–2), 19 (2004).
32. L. Darre *et al.* Another coarse grain model for aqueous solvation: WAT FOUR? *J. Chem. Theory Comput.* **6** (12), 3793 (2010).
33. F. Villanelo *et al.* Accessing gap-junction channel structure-function relationships through molecular modeling and simulations. *BMC Cell Biol.* **18** (Suppl 1), 5 (2017).
34. A.F.E. Tomsic. Collision between water clusters and surfaces. *Ph. D. dissertation* (Goteborg University, 2003).
35. F. Huether. Filter System. Fabio and Markus Membrane ENG GMBH. CH Patent WO2020169852A1. 3 January 2019.

36. F. Huether. Water Purifier, Fabio and Markus Turbine ENG GMBH. CH Patent WO2020178200A1. 3 January 2019.
37. A. Antonov, L. Yeskesseliva, I. Teodossieva. Influence of ions on the structure of water conditions for away from equilibrium. *Physiology* **26** (4), 255 (1989).
38. P. Gramatikov, A. Antonov, M. Gramatikova. Study of the properties and structure variations of water systems under the stimulus of outside influences. *Fresenius J. Anal. Chem.* **343**, 134 (1992).
39. L. Todorova, A. Antonov. Note on the drop evaporation method for studying water hydrogen bond distribution: I. A filtration application. *C. R. Acad. Bulg. Sci.* **53**, 7 (2000).
40. W. Luck. A model of hydrogen-bonded liquids. *Angewandte Chemie*. **19**, 28 (1980).
41. G.M. Kontogeorgis, A. Hoster, A. Kottaki, *et al.* Water structure, properties and some applications – a review. *Chemical Thermodynamics and Thermal Analysis* **6**, 100053 (2022).
42. A. Luzar, S. Svetina, B. Žekš. The contribution of hydrogen bonds to the surface tension of water. *Chem. Phys. Lett.* **96**, 485 (1983).
43. L.F. Vega, F. Lovel. Review and new insights into the application of molecular-based equation of state to water and aqueous solutions. *Fluid Ph. Equilib.* **416**, 150 (2016).
44. B.J. Zhang, J. Kim, T.Ch. Lee. Behavior of an evaporating water droplet on a lubricant-impregnated nano-structured surface. *Exp. Therm. Fluid Sci.* **96**, 216 (2018).
45. I. Ignatov, M.T. Iliev, T.P. Popova, G. Gluhchev, P. Gramatikov, P. Vassileva. Meteorological data and spectral analyses of non-equilibrium processes in water during the total Solar eclipse of 11.08.1999. *Ukr. J. Phys.* **69**, 85 (2024).
46. I. Ignatov, M.T. Iliev, P.S. Gramatikov. Education program on physics and chemistry for non-equilibrium processes at the interfaces between solid-liquid-gaseous media. *Eur. J. Contemp. Educ.* **12** (3), 862 (2023).
47. D. Mehandjiev, I. Ignatov, N. Neshev, F. Huether, *et al.* Formation of clusters in water and their distribution according to the number of water molecules. *Bulg. Chem. Commun.* **54**, 211 (2022).
48. M.T. Iliev, F. Huether, I. Ignatov, P.S. Gramatikov. Education of students on Physics and Chemistry with effects of water filtration. Modeling of water clusters and hexagonal structures. *Eur. J. Contemp. Educ.* **12**, 1546 (2023).
49. D. Mehandjiev, I. Ignatov, N. Neshev *et al.* History-dependent hydrogen bonds energy distributions in NaCl aqueous solutions undergoing osmosis and diffusion through a ceramic barrier. *J. Chem. Technol. Metall.* **58**, 340 (2023).
50. I. Ignatov, F. Huether, T.P. Popova *et al.* Effects of electromagnetic waves on parameters, hydration and in vitro antimicrobial activity of the Brassica oleracea L. var. italica Plenck. and water. *Plant Sci. Today*. **11**, 553 (2024).
51. S. Kuroki, R. Tsenkova, D. Moyankova, J. Munkan, J.H. Morita, S. Atanassova, D. Djilianov. Water molecular structure underpins extreme desiccation tolerance of the resurrection plant *Haberlea rhodopensis*. *Sci. Rep.* **9**, 3049 (2019).
52. I. Ignatov, F. Huether, N. Neshev *et al.* Research of water molecules cluster structuring during. textitHaberlea rhodopensis Friv. hydration. *Plants* **11**, 2655 (2022).
53. R. Toshkova, N. Neshev, F. Huether *et al.* Effects of hydrogen-rich water on hamsters with experimental myeloid tumor. *Libri Oncoi.* **51** (2), 85 (2023).

Received 15.05.24

I. Ігнатов, Й. Марінов,  
Ф. Хютер, Г. Глухчев, М.Т. Ілієв

МОДЕЛЮВАННЯ ВОДНИХ  
КЛАСТЕРІВ: СПЕКТРАЛЬНИЙ АНАЛІЗ,  
РОЗПОДІЛ ГАУСА І ЛІНІЙНА ФУНКЦІЯ ЧАСУ

Наші експериментальні та теоретичні дослідження виявили наявність кластерів води в різних середовищах (особливо в гідрофобних умовах), де переважають повільніші взаємодії іонів водню. Ключові методи, такі як ядерний магнітний резонанс (ЯМР) і інфрачервона Фур'є-спектроскопія (ІЧФС), відіграли вирішальну роль у нашому розумінні цих кластерів, розкриваючи їх потенційне застосування в медицині. На стабільність і поведінку цих кластерів можуть впливати такі фактори, як присутність іонів металів, що приводить до утворення стабільних кластерів. Цей потенціал для медичного застосування повинен вселити надію та ініціювати подальші дослідження. Крім того, наші дослідження виявили, що водні кластери проявляють характеристики дисипативних структур, демонструючи самоорганізацію під час фізичних, хімічних або теплових змін, подібних до конвекційних комірок Релея–Бенара. Ця динамічна та показова поведінка підтверджує уявлення про те, що роль води виходить за межі простої хімії, потенційно впливаючи на біологічні процеси на фундаментальному рівні. Взаємодія водних кластерів з навколишнім середовищем і здатність підтримувати нерівноважний стан через обмін енергією ще більше підкреслює їхню складність і значення як у природному, так і технологічному контекстах. Фільтрування води – це процес, який використовується для покращення її якості. Ефект виникає внаслідок реструктуризації водневих зв'язків і структурування водних кластерів, більшість з яких є гексагональними. У наших дослідженнях ми застосували воду, фільтровану за патентованою швейцарською технологією EVOdrop.

*Ключові слова:* водні кластери, розподіл Гауса, лінійна функція, спектральний аналіз.