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# Dynamic Nano Clusters of Water on Waters Catholyte and Anolyte: Electrolysis with Nano Membranes

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#### Authors' contributions

This work was carried out in collaboration among all authors. Author II designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors GG and SK managed the analyses of the study. Author II managed the literature searches. All authors read and approved the final manuscript.

## Article Information

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# ABSTRACT

The electrolysis is a base of many industrial processes like galvanotechnics and galvanoplastics. It is also used for hydrogen, and oxygen production. Different products such as  $H_2$ , NaOH, HCl, heavy water ( $D_2O$ ) and others could be obtained as well depending on the manufacturing conditions. They can influence significantly the vital processes in living organisms either activating

or diminishing their rate [1,2]. The catholyte and anolyte waters are produced in the cathode and anode parts of the electrolysis cell. The catholyte is alkaline and has a negative oxidation reduction potential (ORP). The anolyte is acidic and of positive ORP. The value for ORP is in milivolts (mV). It was demonstrated that the catholyte stimulated the metabolitic processes and immune system, and had an anti-oxidant action. Recent investigation of catholyte water on *Graffi* tumor in hamsters has demonstrated its antitumor effect [3]. The anolyte however has demonstrated strong biocidal and antiinflamatory action [4,5] Cha, Chun-Nam et al. [6]. In spite of many investigations carried out all over the world no satisfactory explanation of these properties has been suggested until now. In the report a model of the number of water molecules related to the energy of hydrogen bonds is described.

Keywords: Electrochemical treatment of water; anolyte; catholyte; electrolysis; number of water molecules; energy of hydrogen bonds; NES; DNES.

#### **1. INTRODUCTION**

The electromagnetic hydrogen bonds (O–H...O) are among H<sub>2</sub>O molecules. The water molecules could be binded into complex intermolecular clusters described by a general formula  $(H_2O)_n$ . Hydroxyl groups (-OH) in H<sub>2</sub>O molecule are polar. Water acts as a reagent for a big number of chemical reactions with hydrophilic effects. There are reactions of chemical exchange in water and high ionizing ability. There was performed research with Raman spectroscopy that hydrogen bonds among water molecules are constantly tearing, changing and moving (Geissler, Saykally and Smith, 2005). The model of Keutsch, Saykally is between 3 to 50 water molecules [7,8]. These results correlate with our quantum-mechanical analysis of water spectrum [9]. Fowler, Quinn, Redmond [10,11] and Ignatov, Mosin show the models with n from 3 to 60 water molecules [9]. There are different models of water clusters in the research of Choi, Jordan, [12], Loboda O, Goncharuk V. [13] Sykes [14], Chaplin [15], Liu, Cruzan, Saykally [16], Shu et al. [17].

The research of water clusters  $(H_2O)_n$  are with the following methods–X-Ray, EXAFSspectroscopy, <sup>1</sup>H-NMR, neurons diffraction, IR spectroscopy, NES and DNES spectral methods. There were charged ionic clusters  $[(H_2O)_n]^*$  and  $[(H_2O)_n]^-$ . With computer calculations were made water clusters  $(H_2O)_n$ , where n = 3–20.

However, none of the suggested models can explain how the water clusters change during the electrolysis and how these changes are connected to the unusual properties of the obtained catholyte and anolyte.

In this research the authors make mathematical models of water molecules in anolyte and

catholyte. It is accepted that the aqueous solutions may undergo autoprotolysis, i.e. the H<sup>+</sup> proton is released from H<sub>2</sub>O molecule and then transferred and accepted by the neighboring H<sub>2</sub>O molecule resulting in formation of hydronium ions as  $H_3O^+$ ,  $H_5O_2^+$ ,  $H_7O_3^+$ ,  $H_9O_4^+$ , etc. Thus, water should be considered as an associated liquid composed from a set of individual H<sub>2</sub>O molecules, linked together by hydrogen bonds and weak intermolecular van der Waals forces [16]. The simplest example of such associate can be a dimer of water.

Methods NES (Non-equilibrian Energy DNES (Diferential Spectrum) and Nonequilibrian Energy Spectrum) are used for the investigation of catholyte and anolyte [18,19]. They allow the evaluation of hydrogen bonds energy. It is expected that these models could help explaning the different effects of the electrolyzed water on living things. Influence on the nervous system and antiinflamatory and antitumor effects have been reported [9]. In 2019 the presence of nascent hydrogen H\* was discovered in catholyte and it is this nascent hydrogen that activates the catholyte reactions and is responsible for catholyte's beneficial physiological effects [20].

#### 2. MATERIALS AND METHODS

#### 2.1 NES and DNES Spectral Analyses

The device for spectral analysis with methods NES and DNES is based on an optical principle. The author of the dvice is A. Antonov [18]. It uses a hermetic camera for evaporation of water drops on a water-proof transparent pad which consists of thin maylar folio and a glass plate.

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The parameters are:

- Temperature (+22-24°C);
- Monochromatic filter with wavelength  $\lambda$  = 580±7 nm (yellow color in visible spectrum).
- Angle of evaporation of water drops from 72.3°to 0°;
- Range of energy of hydrogen bonds among water molecules is E=-0.08– -0.1387 eV or  $\lambda = 8.9$ –13.8 µm;

The energy  $(E_{H...O})$  of hydrogen O...H-bonds among H<sub>2</sub>O molecules in water sample is measured in eV. The function f(E) is called spectrum of distribution according energues. The energy spectrum of water is characterized by a non-equilibrium process of water droplets evaporation and this is non-equilibrium energy spectrum (NES) and is measured in eV<sup>-1</sup>. DNES is defined as the difference

 $\Delta f(E) = f$  (samples of water) – f (control sample of water)

DNES is measured in  $eV^{-1}$ where f(\*) denotes the evaluated energy.



Fig. 1. Scheme of the electrolyzer

#### 2.2 Electrolyzer

To obtain catholyte and anolyte, water electrolysis is performed. For this a direct current is applied to the electrodes of the electrolyzer. There is semipermiable membrane, which separates the electrolyzer of two parts for anolyte and catholyte. In the electrolyzer the membrane separates the alkaline fraction (catholyte) obtained at the cathode. The acidic fraction (anolyte) is obtained at the anode. When the current passes through the water there are redox reactions on the surface of the electrodes. The functional scheme and the reactions are shown in Fig. 1.

#### **3. RESULTS AND DISCUSION**

# 3.1 Physiological Effects of Anolyte and Catholyte

The enhanced electron-donating property of the catholyte can also enhance the electron background of the physiological fluids in an organism. The catholyte water is improving metabolic processes and enzyme activity, increases the DNA and ATP synthesis, influences on bio molecules with anti oxidant effects and normalizing ORP. There is improvement of hydrogen ion exachange throght cell membran [21-23].

The main parameters pH and ORP (Oxidation-Reduction Potential) of the anolyte and catholyte depend on the strength of the electrical current, the initial water temperature, the processing time and the presence of dissolved matter.

The catholyte may be classified into a few categories as follows [23]:

The alkaline catholyte (C) is with pH > 9.0 and ORP = -700...-820 mV. The active components of C are  $O_2$ ,  $HO_2^-$ ,  $HO_2^+$ ,  $OH^-$ ,  $OH^+$ ,  $HO_2^-$ ,  $O_2$ ;

The neutral catholyte (CN) is with pH  $\leq$  9.0 and ORP = -300...-500 mV. The active components of CN are O<sub>2</sub>, HO<sub>2</sub><sup>-</sup>, HO<sub>2</sub><sup>+</sup>, H<sub>2</sub>O<sub>2</sub>, H<sup>+</sup>, OH<sup>-</sup>.

On the contrary, the anolyte obtains acidic reaction, the ORP and conductivity increases, the amount of the dissolved oxygen in water also increases, whereas the amount of hydrogen decreases [24]. The anolyte is a brownish acidic liquid with a characteristic odor and taste with pH = 3-5, and ORP = +250...+800 mV. The electrochemically activated solutions of the anolyte are divided into two main types:

The acidic anolyte (A) is with pH < 5.0 and ORP = +800...+1200 mV. The active component of A is HO<sub>2</sub>\*;

The neutral anolyte (AN) is with pH = 6.0 and ORP = +600...+900 mV). The active components of AN are  $O_3$ , HO<sup>-</sup>, HO<sub>2</sub>\*;

The anolyte has antiviral, anti-inflammatory, antibacterial and antifungal effects [4,5]. Anolyte has effects on bacterial infections such as staphylococcal Enterotoxin-A [25-31].

#### 3.2 Results with NES and DNES Methods for Catholyte and Anolyte

It has been experimentally proven that during the evaporation of a drop, the wetting angle  $\theta$  decreases discreetly to 0, and the diameter of the drop bottom slightly changes, and this constitutes a new effect in physics [18]. Through measurement of this angle at regular intervals of time one can determine the function of distribution according to values of the last f( $\theta$ ). The function is called spectrum of the state of water [18].

For practical purposes it is required that information is to be received from the spectrum of the state of water for the average energy of the hydrogen bonds in a given sample. Luck's model is used (1980). He considers water as composed of O-H...O groups [32]. A greater part of these groups is related to energy of the hydrogen bond (-E), and the remaining are free (E=0). The function f(E) can be modified under influence. It is measured in reciprocal electron volts (eV<sup>-1</sup>) [18]. A correlation is established between  $f(\theta)$  and the energy of the hydrogen bonds among water molecules. The energy E is measured in electron volts (eV) and f(E) is called spectrum of distribution according to energies. The water spectrum is received as a result of a non-equilibrium process of evaporation of water drops and because of this the term Non-equilibrium spectrum of water (NES) is used.

The difference:  $\Delta f$  (E)=f (sample) - f (control sample) is called Differential non-equilibrium energy spectrum (DNES).

DNES is a measure for the modification of water's structure as a result of influence. The total influence of all remaining factors besides the examined influence is the same for the control sample with water and the sample, which is influenced.

For calculation of the function f(E) experimental dependence between the water surface tension

 $(\theta)$  and the energy of hydrogen bonds (E) was established:

$$f(E) = b \times f(\theta)/1 - (1 + b \times E)^2)^{1/2}$$
,

where  $b = 14.33 \text{ eV}^{-1}$ .

The relation between the wetting angle and the energy among the hydrogen bonds of water E molecules is:  $\theta$ =arcos (-1-14.33E).

The hydrogen bonds energy E is measured in electron-volts (eV) and is also related to the spectrum of energy distribution.

The average energy ( $\Delta E_{H...O}$ ) of hydrogen H...Obonds among individual molecules H<sub>2</sub>O was calculated for the catholyte and anolyte by NESand DNES-methods. We studied the distribution of local extemums in the catholyte and anolyte solutions. The result for catholyte in the NES-spectrum is E=-0.1251 eV, for the anolyte is E=-0.1130 eV and for the control sample of water - E=-0.1191 eV. The values of  $\Delta E_{H_{u},0}$  for the catholyte with the DNES method are within the interval (-0.0121±0.0011 eV), and for the anolyte the interval is (+0.0061±0.0011 eV). These results suggest the restructuring of  $\Delta E_{H_{...O}}$  values among individual H<sub>2</sub>O molecules with а statistically significant increase of local extemums in DNESspectra of the catholyte and the anolyte (Table 1) where local extremums  $(eV^{-1})$  of the function of distribution of energies of hydrogen bonds for both catholyte and anolyte are presented.

For the catholyte the biggest extremum was detected at E=-0.1387 eV;  $\lambda$ =8.95 µm;  $\tilde{v}$  =1117 cm<sup>-1</sup>. In 1995 A. Antonov performed experiments with the impact of different types of water on mice tumor cells [18]. These experiments detected a decrease in the NES-spectrum compared with the control sample of cells from healthy mice. At the same time there was a decrease of the local extremum at E=-0.1387 eV;  $\lambda$ =8.95 µm;  $\tilde{v}$ =1117 cm<sup>-1</sup>. In DNES the local extremum at 8.95 µm was with a negative value. It should be noted that for the catholyte the local extremum in DNES was detected with the positive value at 87.0 eV<sup>-1</sup>.

For the catholyte the biggest local extremum is at -0.1312 eV, or  $9.45 \mu \text{m}$ . Recent research on *Graffi* tumor in hamsters has demonstrated the antitumor effects of catholyte water [33].

-E(eV)	Catholyte f (E)	Anolyte f (E)	Control sample	DNES Catholyte	DNES Anolyte	-E(eV)	Catholyte f (E)	Anolyte f (E)	Control Sample	DNES Catholyte	DNES Anolyte
	. ,	(eV <sup>-1</sup> )	f (E) (eV <sup>-1</sup> )	Δf (E)	Δf (Ĕ)		(eV⁻¹)	(eV <sup>-1</sup> )	f (E) (eV <sup>1</sup> )	Δf (E) (eV <sup>-1</sup> )	Δf (Ĕ) (eV⁻¹)
0.0937	0	59.3	0	0	59.3	0.1187	0	29.6	52.2	-52.2	-22.6
0.0962	0	0	0	0	0	0.1212	34.8	89.9	34.8	0	54.2
0.0987	0	44.4	17.4	-17.4	27.0	0.1237	0	29.6	34.8	-34.8	-5.2
0.1012	0	0	0	0	0	0.1262	34.8	0	0	34.8	0
0.1037	34.8	0	52.2	-17.4	0	0.1287	34.8	14.8	34.8	0	-20.0
0.1062	17.4	0	0	0	0	0.1312	52.2	14.8	0	52.2	14.8
0.1087	17.4	59.3	34.8	-17.4	24.5	0.1337	0	0	52.2	0	0
0.1112	17.4	0	0	17.4	0	0.1362	34.8	14.8	0	34.8	14.8
0.1137	0	0	0	0	0	0.1387	87.0	14.8	17.4	69.6	-2.6
0.1162	34.8	29.6	0	34.8	29.6	_	_	_	_	-	-

Table 1. Local extremums of catholyte and anolyte solutions in NES- and DNES-spectra with function of didtribution of energy f (E) for NES and  $\Delta f$  (E) for DNES in eV<sup>-1</sup>

Notes:  $E=-0.1112 \text{ eV} (\lambda=11.15 \mu\text{m})(\tilde{v}=897 \text{ cm}^{-1})$  is the local extremum responding to the stimulation effect on the nervous system and improvement of nerve conductivity;  $E = -0.1212 \text{ eV} (\lambda=10.23 \mu\text{m})(\tilde{v}=978 \text{ cm}^{-1})$  is the local extremum for anti-inflammatory effect;  $E=-0.1387 \text{ eV} (\lambda=8.95 \mu\text{m})(\tilde{v}=1117 \text{ cm}^{-1})$  is the local extremum related to the inhibition of development of tumor cells at the molecular level

-E(eV) x-axis	Catholyte y-axis Number of H <sub>2</sub> O molecules	Anolyte y-axis Number of H <sub>2</sub> O molecules	-E(eV) x-axis Number of H₂O molecules	Catholyte y-axis Number of H₂O molecules	Anolyte y-axis Number of H₂O molecules
0.0937	0	15	0.1187	0	7
0.0962	0	0	0.1212	9	22
0.0987	0	11	0.1237	0	7
0.1012	0	0	0.1262	9	0
0.1037	9	0	0.1287	9	4
0.1062	4	0	0.1312	13	4
0.1087	4	15	0.1337	0	0
0.1112	4	0	0.1362	9	4
0.1137	0	0	0.1387	21	4
0.1162	9	7	_	_	-

Table 2. Distribution of the number of water ( $H_2O$ ) molecules according to the energ	jy of
hydrogen bonds in catholyte and anolyte	



Fig. 2. Number of the water molecules as a function of the energy of hydrogen bonds

It should be noted that some medical drugs for the treatment of influenza contain aluminum hydroxide  $AI(OH)_3$ . The local extemum in this case was detected at -0.1326 eV, or at 9.35 µm.

A mathematical model of the number of water molecules according to the energy of hydrogen bonds in catholyte and anolyte has been developed (Ignatov, Gluhchev, 2020) (Table 2; Fig. 2). The evaluation of the possible number of hydrogen bonds as percent of  $H_2O$  molecules with different values of distribution of energies is presented above. These distributions are basically connected with the restructuring of  $H_2O$  molecules with the same energies. This serves as a mathematical model explaining the behavior of anolyte and catholyte regarding the distribution of  $H_2O$  molecules to the energies of hydrogen bonds (Ignatov, Mosin, 2013). The new

model shows the number of water molecules and their structuring in clusters.

## 4. CONCLUSION

The separation of catholyte and anolyte at the cathode and anode in electrolysis cell respectively, is by a semipermeable membrane. The membrane is transperant for water ions, molecules and clusters. However, this process is not quite clear and there is no theoretical satisfactory explanation of what actually happens.

Two of the authors (Ignatov and Gluhchev) suggest a mathematical model for the number of water molecules using the distribution of the hydrogen bonds energy E in the range (-0.0937 eV; 13.23  $\mu$ m; 756 cm<sup>-1</sup>) to (-0.1387 eV; 8.95  $\mu$ m; 1117 cm<sup>-1</sup>).

The measurements with spectral methods NES and DNES show significant difference between anolyte and catholyte. The result for catholyte in the NES-spectrum is -0.1251 eV, while for the anolyte it is -0.1130 eV. The values of  $\Delta E_{H...0}$  for catholyte measured by the DNES method are in the interval (-0.0121±0.0011 eV), while the corresponding interval for the anolyte is (+0.0061±0.0011 eV). The highest local extremum for the catholyte is 69.6 eV<sup>-1</sup> at (-0.1387 eV; 8.95 µm; 1117 cm<sup>-1</sup>). This value is responsible for its antitumor effect. For the anolyte the highest local extremum is 54.2 eV<sup>-1</sup> at (-0.1212 eV; 10.23 µm; 978 cm<sup>-1</sup>), which is responsible for its antiinflamatory effect.

The results from NES for  $E_{H... O}$  and DNES for  $\Delta E_{H... O}$  show that the angle of moisture at the catholyte is larger than the one at the anolyte.

The present investigation points at the relationship between the number of water molecules and the energy of hydrogen bonds, which may serve as a starting point for future research.

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# **COMPETING INTERESTS**

Authors have declared that no competing interests exist.

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