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.

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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 H₂, NaOH, HCl, heavy water (D₂O) and others could be obtained as well depending on the manufacturing conditions. They can influence significantly the vital processes in living organisms either activating...
1. INTRODUCTION

The electromagnetic hydrogen bonds (O–H…O) are among H2O molecules. The water molecules could be bound into complex intermolecular clusters described by a general formula (H2O)n. Hydroxyl groups (–OH) in H2O 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 (H2O)n are with the following methods—X-Ray, EXAFS-spectroscopy, 1H-NMR, neurons diffraction, IR spectroscopy, NES and DNES spectral methods. There were charged ionic clusters [(H2O)n]n and [(H2O)n]−. With computer calculations were made water clusters (H2O)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++ proton is released from H2O molecule and then transferred and accepted by the neighboring H2O molecule resulting in formation of hydronium ions as H3O+, H4O2−, H5O33−, H6O44−, etc. Thus, water should be considered as an associated liquid composed from a set of individual H2O 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 Spectrum) and DNES (Diferential Non-equilibrian 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 explaining the different effects of the electrolyzed water on living things. Influence on the nervous system and antiinflammatory 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 device 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.
The parameters are:
- Temperature (+22–24°C);
- Monochromatic filter with wavelength $\lambda = 580\pm7$ 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 $(E_{O...H})$ of hydrogen O...H-bonds among $H_2O$ molecules in water sample is measured in eV. The function $f(E)$ is called spectrum of distribution according energies. 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$^{-1}$. DNES is defined as the difference

$$\Delta f(E) = f(\text{samples of water}) - f(\text{control sample of water})$$

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

The acidic anolyte (A) is with pH < 5.0 and ORP = -700...-820 mV. The active components of A are $O_2$, $HO_2^-$, $HO_2^*$, $OH^-$, $HO_2^-$, $O_2$;

The neutral catholyte (CN) is with pH ≤ 9.0 and ORP = -300...-500 mV. The active components of CN are $O_2$, $HO_2^-$, $HO_2^*$, $H_2O_2$, $H^+$, $OH^-$;

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 $HO_2^*$;
The neutral anolyte (AN) is with pH = 6.0 and ORP = +600...+900 mV). The active components of AN are O3, HO3, HO2+.

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 discretely 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 a model of the state of water for the average energy of hydrogen bonds in a given sample. Luck's model is used [19].

The relation between the wetting angle and the energy among the hydrogen bonds of water $E$ molecules is: $\theta = \arccos (-1.14).$

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

The average energy ($\Delta E_{O\cdots H}$) of hydrogen $H\cdots O$-bonds among individual molecules $H_2O$ was calculated for the catholyte and anolyte by NES- and 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 $\Delta E_{O\cdots H}$ 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 $\Delta E_{O\cdots H}$ values among individual $H_2O$ molecules with a statistically significant increase of local extemums in DNES-spectra of the catholyte and the anolyte (Table 1) where local extemums (eV) 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; $\lambda=8.95 \mu m$; $\tilde{\nu}=1117$ cm$^{-1}$. 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; $\lambda=8.95 \mu m$; $\tilde{\nu}=1117$ cm$^{-1}$. In DNES the local extremum at 8.95 $\mu m$ 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$^{-1}$.

For the catholyte the biggest local extremum is at -0.1312 eV, or 9.45 $\mu m$. Recent research on Graffi tumor in hamsters has demonstrated the antitumor effects of catholyte water [33].
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$^{-1}$

<table>
<thead>
<tr>
<th>$-E$(eV)</th>
<th>Catholyte f (E)</th>
<th>Anolyte f (E)</th>
<th>Control sample f (E)</th>
<th>DNESS Catholyte $\Delta f$ (E)</th>
<th>DNESS Anolyte $\Delta f$ (E)</th>
<th>-E(eV)</th>
<th>Catholyte f (E)</th>
<th>Anolyte f (E)</th>
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<th>DNESS Catholyte $\Delta f$ (E)</th>
<th>DNESS Anolyte $\Delta f$ (E)</th>
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<tr>
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<td>59.3</td>
<td>0</td>
<td>59.3</td>
<td>0</td>
<td>0.1187</td>
<td>0</td>
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<td>54.2</td>
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<tr>
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</table>

Notes: $E=-0.1112$ eV ($\lambda=11.15$ μm)($\nu=897$ cm$^{-1}$) is the local extremum responding to the stimulation effect on the nervous system and improvement of nerve conductivity; $E=-0.1212$ eV ($\lambda=10.23$ μm)($\nu=978$ cm$^{-1}$) is the local extremum for anti-inflammatory effect; $E=-0.1387$ eV ($\lambda=8.95$ μm)($\nu=1117$ cm$^{-1}$) is the local extremum related to the inhibition of development of tumor cells at the molecular level.
Table 2. Distribution of the number of water (H₂O) molecules according to the energy of hydrogen bonds in catholyte and anolyte

<table>
<thead>
<tr>
<th>-E(eV) x-axis</th>
<th>Catholyte y-axis</th>
<th>Anolyte y-axis</th>
<th>-E(eV) x-axis</th>
<th>Catholyte y-axis</th>
<th>Anolyte y-axis</th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Number of H₂O</td>
<td>Number of H₂O</td>
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<td>Number of H₂O</td>
<td>Number of H₂O</td>
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<td>molecules</td>
<td>molecules</td>
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<td>7</td>
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<tr>
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<td>0</td>
<td>0</td>
<td>0.1212</td>
<td>9</td>
<td>22</td>
</tr>
<tr>
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<td>11</td>
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</tr>
<tr>
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<td>4</td>
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<tr>
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<td>4</td>
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<td>9</td>
<td>7</td>
<td>--</td>
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</tr>
</tbody>
</table>

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 Al(OH)₃. The local extremum 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 H₂O molecules with different values of distribution of energies is presented above. These distributions are basically connected with the restructuring of H₂O molecules with the same energies. This serves as a mathematical model explaining the behavior of anolyte and catholyte regarding the distribution of H₂O 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 μm; 756 cm$^{-1}$) to (-0.1387 eV; 8.95 μm; 1117 cm$^{-1}$).

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 $\Delta E_{\text{H...O}}$ 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$^{-1}$ at (-0.1387 eV; 8.95 μm; 1117 cm$^{-1}$). This value is responsible for its antitumor effect. For the anolyte the highest local extremum is 54.2 eV$^{-1}$ at (-0.1212 eV; 10.23 μm; 978 cm$^{-1}$), which is responsible for its antiinflamatory effect.

The results from NES for $E_{\text{H...O}}$ and DNES for $\Delta E_{\text{H...O}}$ 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.

REFERENCES

17. Shu Li, Jegatheesan L, Jegatheesan V, Chun QL. The structure of water, fluid phase equilibria. 2020;511.
31. Shimada K, Ito K, Murai SA. Comparison of the bactericidal effects and cytotoxic