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Article

Use of a New Method for the Estimation of the Structure of Drinking and Mineral Waters

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Abstract

Water quality is traditionally assessed using epidemiological, organoleptic, sanitary-toxicological, and radiation criteria. However, recently, increasing evidence points to the need to consider the structure of aqueous solutions, especially when used for therapeutic purposes. In this study, electrophysical methods were used to study aqueous solutions of sodium chloride of varying concentrations, drinking water, mineral water, and seawater. Patterns in the frequency and amplitude changes of a sinusoidal oscillator compared to distilled water were identified. Based on experimental data, the Ks coefficient is proposed for assessing the structure of drinking and mineral waters. It is defined as the ratio of the oscillation amplitude of the generator at its minimum frequency to the oscillation amplitude at its maximum frequency. A theoretical justification for the formation of the structural organisation of aqueous solutions depending on the concentration of dissolved salts is presented, taking into account the presence of associated and “free” water dipoles, as well as the formation of hydrate formations in a liquid medium. The need to account for intermolecular interaction forces involving water dipoles is emphasised.

Keywords: water structure; the structure of aqueous solutions; drinking and mineral waters; hydration of ions; forces of intermolecular interaction

1. Introduction

According to established global practice, the suitability of water for drinking and other purposes is assessed according to the following main criteria: epidemiological, organoleptic, sanitary-toxicological, and radiation [1]. However, in recent years, increasing evidence has emerged indicating the need to utilise another criterion—the structure of drinking and mineral water used in various technologies [1–4].

It has been established that the structure of water and aqueous solutions is determined by interactions occurring in the solution between atoms, molecules, and ions [5,6]. The structure of water, formed by molecules linked by hydrogen bonds, includes various water dipoles—clusters—and “free” (more mobile) dipoles [5,7,8]. According to [9], van der Waals bonds also appear in liquid water, which contribute to the formation of water structure. At the same time, the continuous formation and destruction of water molecule associations—clusters—occurs in water and its solutions.

Dielectrometry has previously been used to evaluate the structure of water, its aqueous solutions, and drinking and mineral waters [10]. During experiments, the liquids being studied were placed in a measuring cell (Figure 1), which contains a standard glass test tube with a diameter of 20 mm and a length of 200 mm, containing two capacitor plates. Based on the obtained results, the Ks

criterion was proposed, which allows for the evaluation of the ratio of water dipole mobility at different frequencies.

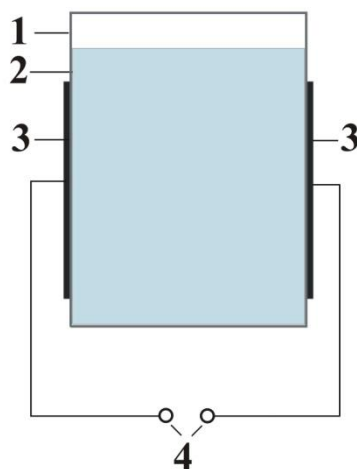


Figure 1. Schematic diagram of a measuring cell for assessing the structure of water and aqueous solutions: 1 – glass container for the liquids being tested, 2 – liquid being tested, 3 – capacitor plates made of non-magnetic material with an area of 60 cm² each, 4 – terminals for connecting the measuring cell in parallel with the inductance of the oscillatory circuit.

However, the surface material of the above-mentioned measuring cell significantly influences the properties of water and water-containing systems (the effect of the near-wall layer) [11]. Given the above data, for studies with distilled water and water-containing systems, it is advisable to use measuring cells in which the capacitor plates are located 5 to 10 cm apart and more than 1 cm from the tank walls [11,12].

In recent years, studies have appeared that utilise a method for assessing structural changes in water and aquatic systems, in which a measuring cell is connected in parallel with the inductance of a sine-wave generator's oscillatory circuit [13,14]. This method determines the change in the amplitude and frequency of the sine-wave generator's oscillations under various influences, and also evaluates the dynamics of the electrical capacitance and active resistance of the equivalent electrical circuit of the measuring cell.

This **review aims** to show the possibilities of new methods for the estimation of the structure of drinking and mineral waters.

2. Materials and Methods

The studies were conducted using measuring cells, a non-destructive and reproducible method for assessing the structure of water and water-containing systems, and data from [11–16]. These studies used distilled water with a specific conductivity of 1.2–1.4 μS/cm, as well as aqueous solutions of sodium chloride in concentrations from 1·10⁻⁶ M to 1·10⁻¹ M. The liquids under study were placed in a rectangular measuring cell with dimensions of 7x7x13 cm, on opposite walls of which two capacitor plates made of non-magnetic material, isolated from the liquid, were placed, each of which had an area of 60 cm² (Figure 1).

The measuring cell was connected in parallel with the inductance of the oscillatory circuit of the sinusoidal oscillator according to the method [15,16]. The change in the amplitude and frequency of the oscillations of the sinusoidal oscillator, as well as the electrical capacitance and active resistance of the equivalent electrical circuit of the measuring cell, were determined (Figure 2)

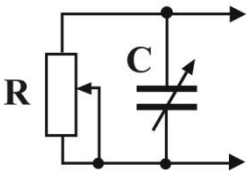


Figure 2. Equivalent electrical circuit of a measuring cell with liquid (variable capacitor and variable active resistance connected in parallel).

In the first series of experiments, at a liquid temperature of 20 °C, the effect of increasing the concentration of sodium chloride solution from $1 \cdot 10^{-6}$ to $1 \cdot 10^{-1}$ M on the amplitude and frequency of generator oscillations was assessed. Using distilled water, the generator frequencies (initial frequencies) were $f_1 - 3.62 \pm 0.02$ kHz; $f_2 - 10.03 \pm 0.01$ kHz; $f_3 - 32.38 \pm 0.01$ kHz; $f_4 - 117 \pm 0.05$ kHz; $f_5 - 327.87 \pm 0.10$ kHz and $f_6 - 1142.70 \pm 0.33$ kHz.

In the second series of experiments, the influence of drinking water (Fiji, Rogla, Yana), mineral water (DonatMg) and sea water from the Adriatic Sea with a mineralisation of 0.317, 0.348, 0.478, 14 and 37 g/liter, respectively, on the amplitude and frequency of oscillations of the generator was assessed.

Considering that the differences between the studied liquids increase with increasing reactive current frequency [10], generator frequencies of up to 1 MHz were used in the work.

To identify the differences between the studied liquids, a coefficient for assessing their structure (K_s) was proposed, which was calculated as the ratio of the amplitude of the generator oscillations at the minimum frequency – f_1 to the amplitude of the oscillations at the maximum frequency of the generator – f_6 .

3. Results of Research

3.1. Changes in the Frequency and Amplitude of Generator Oscillations with Increasing Concentration of Aqueous Solutions of Sodium Chloride

The effect of increasing the concentration of sodium chloride solutions on the change in frequency (f_1 , f_2 , f_3 , f_4 , f_5 and f_6) and the amplitude of generator oscillations at a liquid temperature of 20 °C is shown in [16], Figures 3 and 4.

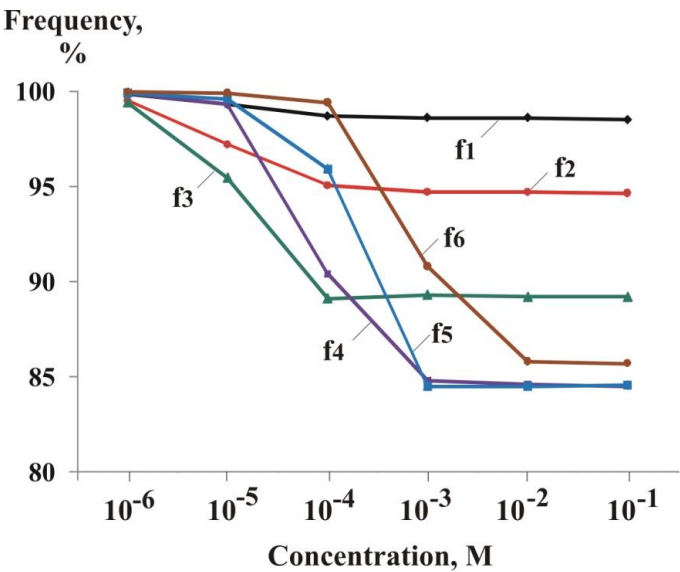


Figure 3. The effect of increasing the concentration of sodium chloride solution in the measuring cell on the change in frequencies (f_1 , f_2 , f_3 , f_4 , f_5 and f_6) of the sinusoidal oscillation generator (in % of distilled water).

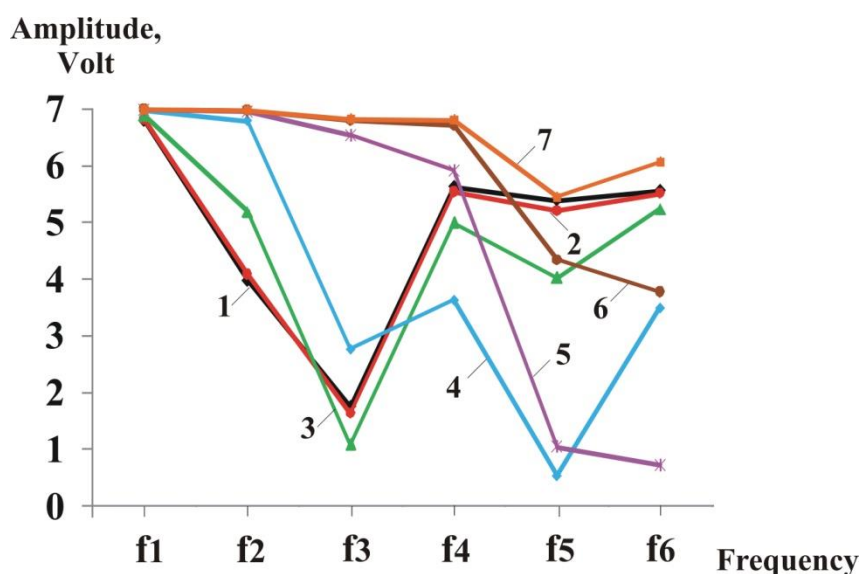


Figure 4. The effect of increasing the concentration of sodium chloride solutions in the measuring cell on the amplitudes (volts) of oscillations of the sinusoidal oscillation generator at different initial frequencies (f1, f2, f3, f4, f5 and f6). 1 – distilled water; 2, 3, 4, 5, 6, 7 – sodium chloride solutions in concentrations of $1 \cdot 10^{-6}$ M, $1 \cdot 10^{-5}$ M, $1 \cdot 10^{-4}$ M, $1 \cdot 10^{-3}$ M, $1 \cdot 10^{-2}$ M, $1 \cdot 10^{-1}$ M, respectively.

With an increase in the concentration of sodium chloride solutions from $1 \cdot 10^{-6}$ M to $1 \cdot 10^{-1}$ M, all initial frequencies of sinusoidal oscillations of the generator (f1, f2, f3, f4, f5 and f6) compared to distilled water monotonically decreased (Figure 3) by 1.5%, 5.4%, 10.8%, 15.5%, 15.4% and 14.3%, respectively ($P < 0.001$ in all cases). A more pronounced decrease in the frequency of oscillations of the generator was at high frequencies (f4, f5 and f6) and, obviously, indicated that with an increase in the concentration of solutions, the destruction of the cluster structure of distilled water occurs [6]. As a result, the number of “free” dipoles of water, which rotate in an alternating electric field at higher frequencies, increases, the electrical capacitance of liquids increases and, accordingly, the frequency of oscillations of the generator decreases.

With an increase in the concentration of sodium chloride solutions from $1 \cdot 10^{-6}$ M to $1 \cdot 10^{-1}$ M, the amplitude of the generator oscillations (Figure 4) at frequencies f1, f2, f3, f4, f5 and f6, compared to distilled water, increased by 2.9%; 75.4%; 286%; 21%; 1.2% and 9%, respectively ($P < 0.001$ in all cases). At the same time, at frequencies f1 and f2, with an increase in the concentration of sodium chloride solution to $1 \cdot 10^{-1}$ M, the amplitude of oscillations increased monotonically, and at frequencies f3, f4, f5 and f6, with an increase in the concentration of sodium chloride solution, the amplitude of the generator oscillations, compared to distilled water, initially decreased and then increased.

It should be noted that at frequencies f3 and f5 in the experiments with distilled water and at a sodium chloride solution concentration of $1 \cdot 10^{-6}$ M, $1 \cdot 10^{-5}$ M and $1 \cdot 10^{-4}$ M, there were pronounced local minima. At frequency f3 and a solution concentration of $1 \cdot 10^{-5}$ M, the oscillation amplitude decreased to 15.6%, and at frequency f5 and a concentration of $1 \cdot 10^{-4}$ M, the oscillation amplitude decreased to 7.6%. With an increase in the sodium chloride solution concentration to $1 \cdot 10^{-3}$ M and $1 \cdot 10^{-2}$ M, local minima at frequencies f3 and f5 were not observed ($P < 0.001$ in these cases). Only at a solution concentration of $1 \cdot 10^{-6}$ M was a slight local decrease in the amplitude to 78%.

According to [16], the presence of the above-mentioned local minima at frequencies f3 and f5 in experiments with distilled water and at sodium chloride concentrations of $1 \cdot 10^{-6}$ M, $1 \cdot 10^{-5}$ M and $1 \cdot 10^{-4}$ M obviously indicates a decrease in the quality factor of the oscillatory circuit at these frequencies, caused by a decrease in the mobility of water dipoles. These minima in distilled water are probably due to a significant number of water dipoles in it being in clusters [6,7]. In sodium chloride solutions, there is an increase in the number of “free” water dipoles and a decrease in the size of water clusters

[6], in which, obviously, the expression of the bonds between water dipoles increases and, accordingly, their mobility decreases

The absence of the aforementioned minima at frequencies f_3 and f_5 as the sodium chloride solution concentration in the measuring cell increases from 1×10^{-3} M to 1×10^{-2} M likely occurs due to an increase in the number of “free” water dipoles (which have greater mobility) and water dipoles located in hydrate formations in the region of distant hydration. Moreover, the total mobility of water dipoles increases with increasing sodium chloride solution concentration at all frequencies studied, resulting in a further decrease in frequency and an increase in the amplitude of the generator’s sinusoidal oscillations.

In evaluating the literature data on structural changes in water under various influences, it should be noted that, according to [17], water can exist in several structural states depending on the concentration of the sodium chloride solution and the temperature of the solution. The transition from one state to another occurs with an increase in the concentration of the solution or its temperature. Taking into account the obtained results and these literature data, it can be assumed that with a stepwise increase in the concentration of the sodium chloride solution to 1.10^{-4} M, the presence of local minima on the oscillation amplitude curves at frequencies f_3 and f_5 may be associated with the presence of structural formations in water (“water I” and “water II” according to [18]), which change relatively discretely with an increase in the concentration of the sodium chloride solution.

3.2. Changes in the Frequency and Amplitude of Generator Oscillations When Exposed to Drinking and Mineral Waters

When exposed to drinking and mineral waters (Fiji, Rogla, Jana, DonatMg, sea water), the differences between them at each frequency (f_1 , f_2 , f_3 , f_4 , f_5 and f_6) did not exceed 1.5%. The influence of drinking and mineral waters on the amplitude of generator oscillations was more pronounced and is shown in Figure 5.

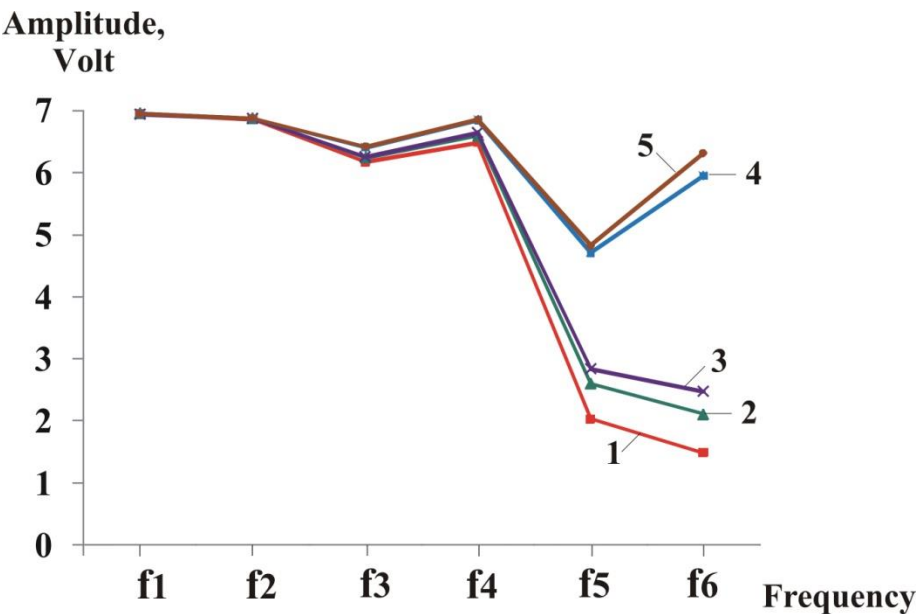


Figure 5. The influence of drinking and mineral waters on the amplitude (volts) of the sinusoidal oscillation generator at different frequencies (f_1 , f_2 , f_3 , f_4 , f_5 and f_6). 1, 2, 3 – drinking waters Fiji, Rogla and Jana, respectively; 4 – mineral water DonatMg; 5 – sea water.

It turned out that the differences between the minimum and maximum values of the generator oscillation amplitude at each frequency were f_1 , f_2 , f_3 , f_4 , f_5 and f_6 were 0.22%; 0.32%; 3.99%; 5.7%;

138% and 325.4%. More pronounced differences in the decrease in amplitude between the waters were at a high frequency (f6).

3.3. Changes in the Electrical Capacitance and Active Resistance of the Equivalent Electrical Circuit of a Measuring Cell with Increasing Concentration of Sodium Chloride Solutions and When Exposed to Drinking and Mineral Waters

With an increase in the concentration of sodium chloride solutions from 1.10-6 to 1.10-1 M, the electrical capacitance of the equivalent electrical circuit of the measuring cell at frequencies f1, f2, f3, f4, f5 and f6 increased by 6.3%; 44.7%; 112%; 153%; 164% and 153%, respectively (Figure 6) [16]. These changes were several times greater than the above-mentioned frequency changes.

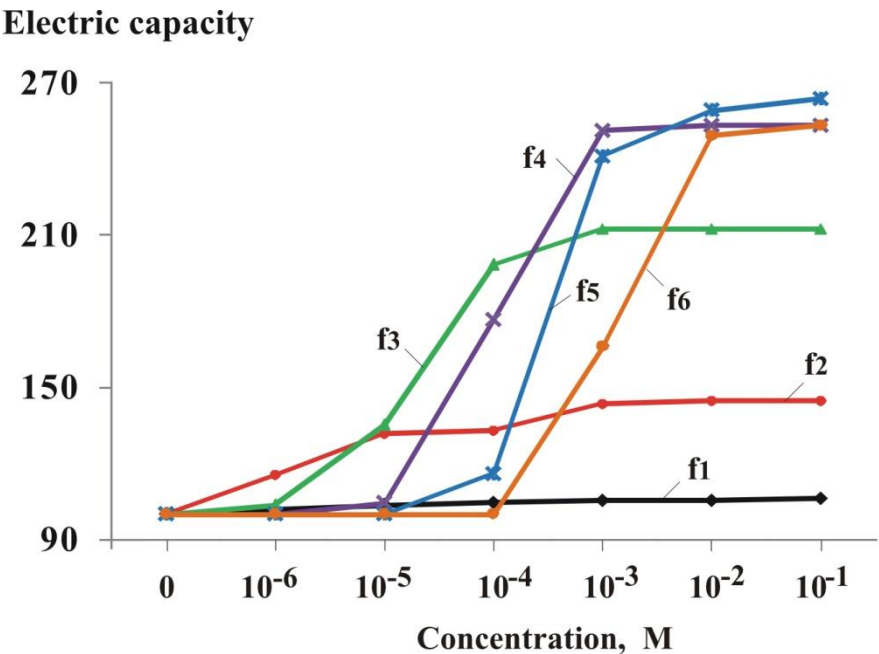


Figure 6. The effect of increasing the concentration of sodium chloride solution in the measuring cell on the change in the electrical capacitance of the equivalent electrical circuit of the measuring cell at frequencies f1, f2, f3, f4, f5 and f6 of the sinusoidal oscillation generator (in % of distilled water).

With an increase in the concentration of sodium chloride solutions from 1.10-6 to 1.10-1 M, the active resistance of the equivalent electrical circuit of the measuring cell at frequencies f1, f2, f3, f4 and f5 increased by 558%; 1080%; 1063%; 453%; 98.8%, and at frequency f6 the active resistance decreased by 61.5% [16]. These changes were also several times greater than the above-mentioned amplitudes of the generator oscillations.

It should be noted that the above-mentioned changes in the generator’s oscillation frequency with increasing sodium chloride solution concentration correlate with changes in the equivalent circuit’s capacitance at all frequencies used (f1, f2, f3, f4, f5, and f6). The correlation coefficient ranges from 0.95 to 0.99 (P < 0.001). This allows us to conclude that the equivalent circuit’s capacitance adequately reflects changes in the generator’s oscillation frequency and, consequently, the electrical capacitance of sodium chloride solutions.

The above-noted changes in the generator oscillation amplitude with increasing sodium chloride solution concentration correlate with changes in the active resistance of the equivalent circuit at frequencies (f1–f6). The correlation coefficient ranges from 0.82 to 0.97 (P<0.001 and P<0.05). This allows us to conclude that the equivalent circuit resistance adequately reflects the change in the generator oscillation amplitude when a measuring cell with water-containing systems is included in the oscillatory circuit and indicates a change in the mobility of water dipoles. The correlation data are consistent with the results of [16].

3.4. Evaluation of Changes in the Coefficient K_s for Assessing the Structure of Aqueous Solutions of Sodium Chloride, Drinking and Mineral Waters

Taking into account the above-obtained changes in the frequency and amplitude of the sinusoidal oscillation generator, as well as the data from [10], the coefficient K_s was proposed for assessing the structure of drinking and mineral waters, which was calculated as the ratio of the amplitude of the generator oscillations at the minimum frequency – f_1 to the amplitude of the oscillations at the maximum frequency of the generator – f_6 . The change in the value of the coefficient K_s with an increase in the concentration of the sodium chloride solution in the measuring cell is shown in Figure 7.

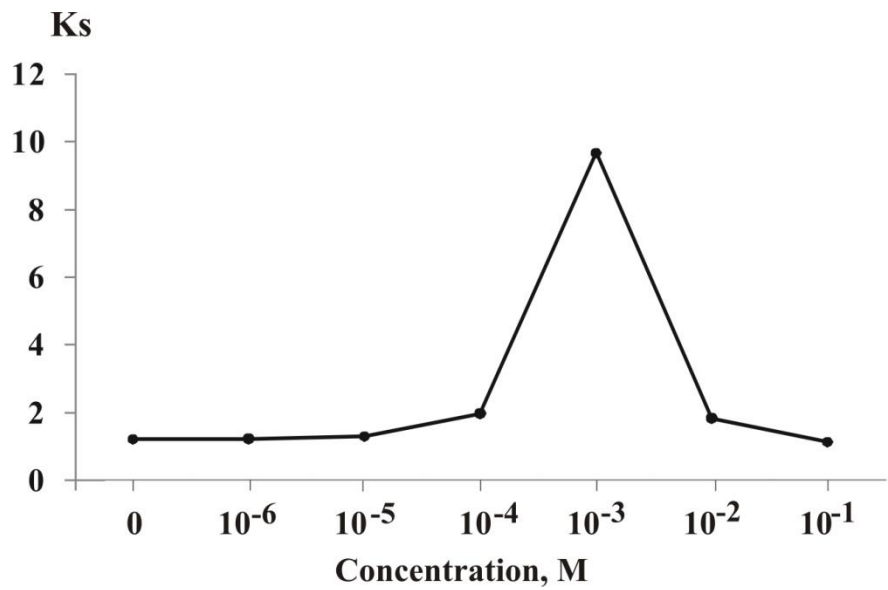


Figure 7. Change in the coefficient K_s with increasing concentration of sodium chloride solution in the measuring cell.

It turned out that in distilled water and at NaCl concentrations of $1 \cdot 10^{-6}$ M, $1 \cdot 10^{-5}$ M and $1 \cdot 10^{-4}$ M, the values of the K_s coefficient were 1.22, 1.24, 1.31 and 1.99, respectively (Figure 6). At a concentration of $1 \cdot 10^{-3}$ M, the K_s coefficient had a maximum value of 9.68. This means that the mobility of water dipoles at frequencies f_1 and f_6 at low NaCl concentrations does not differ significantly, but increases many times over with an increase in the solution concentration to $1 \cdot 10^{-3}$ M. With an increase in the concentration of calcium chloride to 10^{-2} and 10^{-1} M, the K_s values were 1.85 and 1.15, respectively. The indicated changes in K_s are consistent with the results of [10].

Next, the K_s coefficient values of drinking and mineral waters were determined in comparison with the value of this parameter for sodium chloride solutions (Figure 8). It turned out that the K_s coefficient of drinking water lies between the K_s of $1 \cdot 10^{-3}$ M of an aqueous sodium chloride solution and the K_s of $1 \cdot 10^{-2}$ M of a sodium chloride solution. Donat mineral water and sea water have K_s close to the K_s of a NaCl solution at a concentration of $1 \cdot 10^{-1}$ M. The indicated changes in K_s are consistent with the results of work [10].

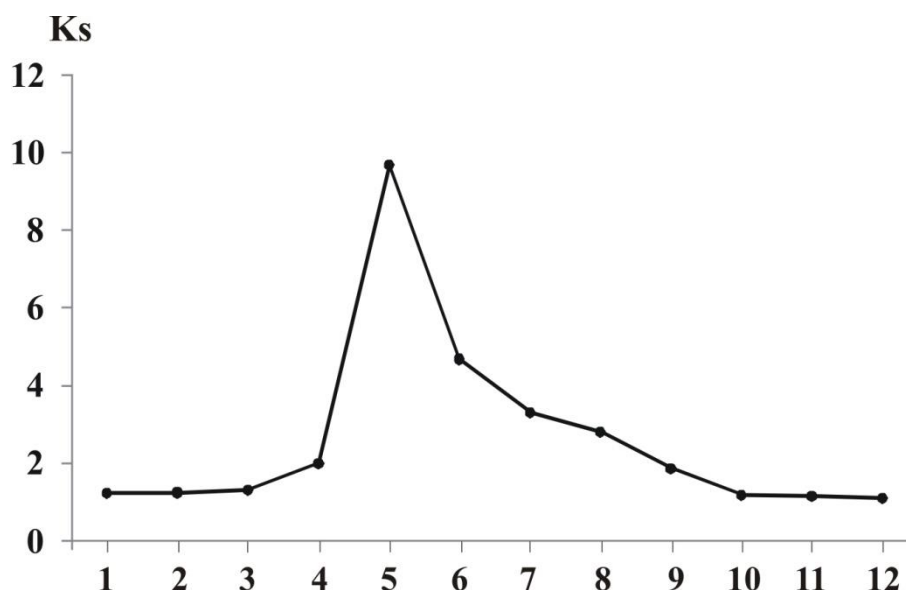


Figure 8. Change in the Ks coefficient with an increase in the concentration of sodium chloride solution in the measuring cell and under the influence of drinking and mineral waters. 1 - distilled water; 2, 3, 4, 5, 9 and 11 - sodium chloride solutions in concentrations of $1 \cdot 10^{-6}$ M, $1 \cdot 10^{-5}$ M, $1 \cdot 10^{-4}$ M, $1 \cdot 10^{-3}$ M, $1 \cdot 10^{-2}$ M and $1 \cdot 10^{-1}$ M; 6, 7 and 8 - Fiji, Rogla and Jana drinking waters, respectively; 10 and 12 - Donat mineral water and sea water, respectively.

From Figure 7,8 it follows that Ks is a quantitative characteristic of the qualitative transition of an aqueous salt solution from one structural state to another, which is probably due to an increase in ion hydration. The presence of ions in water leads to a decrease in the size of clusters [6] and their partial destruction already at a concentration of $1 \cdot 10^{-5}$ M [18]. In addition, when water molecules are oriented within the boundaries of the hydrate formation, van der Waals forces [9] begin to manifest themselves during their electrostatic interaction with the relatively free dipoles of water.

According to [19], if the number of water molecules in an associate is 10^3 units or more, then against the background of thermal motion of the molecules, they can stably exist as a single structurally organised kinetic whole (cluster) under given thermodynamic conditions. Such clusters are formed and are stable in distilled water and in aqueous solutions of sodium chloride at concentrations of $1 \cdot 10^{-6}$ M and $1 \cdot 10^{-5}$ M (points 1, 2, 3; Figure 7). It is their structures that influence the Ks values, which have similar values. With an increase in the solution concentration to $1 \cdot 10^{-4}$ M (point 4, Figure 7), the size and number of associates decrease, which leads to a slight increase in the Ks coefficient.

The Ks coefficient has a maximum value at an aqueous solution concentration of $1 \cdot 10^{-3}$ M (Figure 7). This concentration represents the boundary between competing structures of an aqueous sodium chloride solution. As the solution concentration increases further to $1 \cdot 10^{-2}$ M and $1 \cdot 10^{-1}$ M (points 9, 11; Figure 7), strong ion-dipole and ion-ion interactions begin to manifest themselves, resulting in the main contribution to the Ks value coming from the hydrated structures of the ions.

For drinking and mineral waters, the Ks value allows us to determine the structural state of the solutions. As Figure 7 shows, drinking waters contain associates of water dipoles and hydrated complexes, while mineral waters predominantly consist of hydrated ionic formations or more complex structures.

4. Conclusions

The data obtained in this study are consistent with current understanding of changes in the structure and other properties of water and water-containing systems under various influences and likely indicate that increasing the concentration of sodium chloride solution results in a relatively discrete transition from one structural state of water and water-containing systems to another.

The Ks criterion, along with the electrical capacitance of liquids, can be used to assess the structure of drinking and mineral waters and allows for the evaluation of the mobility of water dipoles at different frequencies. Drinking waters contain associates of water dipoles and hydrated complexes, while mineral waters predominantly consist of hydrated ion formations or more complex structures.

Incorporating a measuring cell or its equivalent electrical circuit into the oscillatory circuit of a sinusoidal oscillator under various influences can be used to indirectly assess changes in the number of water dipoles oscillating in an electric field, their mobility, and, accordingly, to evaluate the structure and other properties of water and water-containing systems. Moreover, the electrical capacitance and active resistance of the equivalent electrical circuit have a greater dynamic range of change and, accordingly, greater sensitivity when assessing changes in the structure and other properties of water.

Author Contributions: G.S.—methodology, investigation; B.L.—methodology, investigation; M.B.—conceptualisation, resources, writing, original draft preparation; N.G.—methodology, conceptualisation; L.A.—investigation, data curation; A.V.—conceptualisation, writing—original draft preparation; A.K.—conceptualisation, writing—original draft preparation. All authors have read and agreed to the published version of the manuscript.

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