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TEMPERATURE AND CONCENTRATION DEPENDENCES OF pH IN AQUEOUS NaCl SOLUTIONS WITH DISSOLVED ATMOSPHERIC CO_2

Temporal variations in the temperature and concentration dependences of the acid-base balance (pH) in dilute aqueous sodium chloride (NaCl) solutions contacting with atmospheric carbon dioxide (CO_2) have been studied. The measurements are carried out for the inverse ion concentrations corresponding to 180, 215, 270, and 360 water molecules per sodium or chlorine ion and in a temperature interval of 294–323 K. The pH relaxation times in aqueous NaCl solutions with dissolved atmospheric CO_2 and the corresponding temperature and salt-concentration dependences are calculated. For aqueous salt solutions characterized by a temperature and an irreducible pH component, a principle for selecting the optimal states is formulated: optimal are those values that provide the minimum pH relaxation time. On this basis, the temperature interval of human activity is determined to extend from (30 ± 2) °C to 42 °C.

Keywords: aqueous solution, sodium chloride, acid-base balance, carbon dioxide, relaxation time.

1. Introduction

This work is devoted to the study of temporal variations in the temperature and concentration dependences of the acid-base balance (pH) in dilute aqueous solutions of sodium chloride (NaCl), when atmospheric carbon dioxide (CO_2) is dissolved in them.

The mechanism of pH formation in aqueous solutions is governed by the dissociation of water molecules occurring both as a result of their collision and under the influence of local electric fields created by the ions of dissolved salts [1]. The local electric fields, which arise as a result of polarization effects under the influence of salt ions, decrease the dissociation energy of water molecules. Therefore, the available energy of their thermal motion is enough for a substantial number of water molecules to dissociate. As a result, the concentration of hydrogen ions increases, which reduces the pH of the aqueous salt solution.

How does the presence of sodium chloride affect the acid-base balance in water contacting with atmospheric air? Theoretical calculations that were carried out in works [2, 3] showed an insignificant (by approximately 0.01) pH decrease in the aqueous solutions of sodium chloride saturated with carbon dioxide in comparison with pure water. The cited authors considered factors that, in the authors' opinion, should together partially compensate the salt effects on the pH in the aqueous solution of sodium chloride; namely, the presence of the salt reduces the solubility of carbon dioxide and the effective concentration of hydrogen ions, but enhances the dissociation of carbonic acid in the solution. As a result, a conclusion was drawn that the addition of salt has almost no effect on the pH value. However, it was proved in experimental works [4,5] that the acid-base balance value in aqueous sodium chloride solutions saturated with carbon dioxide *considerably* decreases with the growth of the NaCl concentration, if the temperature and the partial pressure of CO_2 remain constant. Such an inconsistency between the results of theoretical calculations and experimental data stimulates more thorough systematic experimental studies and exhaustive theoretical justifications.

This work was aimed at studying the peculiarities in the temporal evolution of non-equilibrium solution states corresponding to the establishment of equilibrium concentration values of atmospheric carbon dioxide in aqueous salt solutions contacting with the atmosphere, and finding a principle of selecting those equilibrium states that are consistent with the normal mode of human life.

2. Experimental Part

In the experiments, as-prepared distilled water of purity class II according to DSTU ISO 3696:2003 was used. It was produced by means of the Adrona Crystal EX Double Flow water purification system (Adrona SIA, Latvia). The 0.9 wt.% aqueous NaCl solution for infusions (saline) of pharmaceutical quality (Darnytsia, Ukraine) was used as the initial solution. The aqueous solutions were produced gravimetrically with the help of a Radwag AS 220.R2 balance (Radwag, Poland); the weighing error did not exceed 0.1 mg. The total relative measurement error of solution components did not exceed 0.05%. Thermostating was provided making use of a UTU-10 ultra-thermostat (Krakow, Poland) with an error of ±0.1 K.

In what follows, we will use the inverse concentration of ions in water, which is the ratio between the number of water molecules N_w and the number of salt anions or cations N_s in the solution, i.e.,

$$\zeta = \frac{N_w}{N_s}. \tag{1}$$

The experimental measurements of the acid-base balance were carried out in a temperature interval of 294–323 K and for the inverse NaCl concentrations $\zeta = 180$, 215, 270, and 360.

The solution pH was measured according to IUPAC recommendations [6, 7] using an AZ Bench Top Water Quality Meter 86505 device (AZ Instrument Corp., Taiwan) equipped with a temperature-sensitive probe. The total relative error of pH measurements was 0.5% [8, 9]. Before the measurements, the pH meter was calibrated using reference buffer solutions with pH = 4.00 and 7.00 at a temperature of 25.0 °C.

The solutions were prepared at room temperature via their stirring with the help of a magnetic stirrer. Afterward, the specimens were simultaneously placed into a thermostat for 30 min. Then the first measurement was started (the elapsed time was reckoned from this moment).

In the experiment, the sodium chloride solution was in contact with atmospheric carbon dioxide via the free surface of the solution, which was constant and equal to 8.14 cm^2 in all measurements. The solutions had the same volume of 30 cm^3 and were placed in identical containers, where the height of the solution column was 3.7 cm.

3. Results of Experimental Measurements of the Time Dependences of the Acid-Base Balance in Aqueous NaCl Solutions

Some typical time dependences of the acid-base balance in aqueous NaCl solutions are shown in Fig. 1. To approximate the obtained data taking the χ^2-criterion into account, the following function was chosen:

$$\mathrm{pH}(t) = \mathrm{pH(eq)} + A \exp\left(-\frac{t}{\tau_s}\right), \tag{2}$$

where the quantity $\mathrm{pH}(0) = \mathrm{pH(eq)} + A$ is equal to the initial pH value of the solution, pH(eq) is the final pH value of the solution (this is the pH value for the solution that is in equilibrium with atmospheric carbon dioxide), t is the time interval passed since the solution was prepared, and τ_s is the relaxation time of the solution pH.

The pH values of the solutions that were in equilibrium with atmospheric carbon dioxide, pH(eq), were determined under the following conditions: for water, the acid-base balance was equal to 7.00 at the temperature $T = 25$ °C and to 6.63 at $T = 50$ °C [1, 10, 11]; for water in equilibrium with atmospheric carbon dioxide, pH = 5.63 at $T = 20$ °C [2, 12, 13]; and, for the NaCl solution in equilibrium with atmospheric carbon dioxide, pH = 5.61 at $T = 20$ °C [2, 3]. In addition, the following assumptions were made: the difference between the pH of dilute aqueous NaCl solutions and the pH of water in equilibrium with atmospheric CO_2 is insignificant, and the temperature dependences of the pH of dilute NaCl solutions in equilibrium with atmospheric CO_2 are similar to that of the pH in water.

From the experimental time dependence of the pH of sodium chloride solutions, the pH relaxation times of the solutions were calculated according to expression (2) and applying the following algorithm. The values $\Delta\,\mathrm{pH} = \mathrm{pH}(t) - \mathrm{pH(eq)}$ and the natural logarithms of the inverse values $(1/\Delta\,\mathrm{pH})$ were calculated. Then, the dependence $\ln(1/\Delta\,\mathrm{pH})$ versus t was approximated by a straight line $C_1 + C_2 t$ using the least squares method. The obtained coefficient C_2 is the quantity reciprocal to the relaxation time τ_s of the solution pH.

Then, the pH of aqueous NaCl solutions in equilibrium with atmospheric CO_2 can be calculated using the formula

$$\mathrm{pH(eq)} = a - bT, \tag{3}$$

where T is the solution temperature in Celsius degrees, $a = 5.97$, and $b = 0.015$ [1].

The temperature dependences of the pH relaxation times in aqueous NaCl solutions within the examined interval of inverse concentrations of dissolved ions are presented in Table.

4. Universal Behavior of the Temperature Dependence of the pH Relaxation Time in Aqueous Solutions

Let us analyze the data quoted in Table and consider the details of the temperature and concentration dependences of the pH relaxation time, $\tau_s(T, \zeta)$, for various salt concentrations. Here, we proceed from the fact that primitive life appeared in the primordial ocean, which was just an aqueous salt solution, and the primitive living beings were emerging provided the maximum consistency between their properties and the physical properties of such solutions.

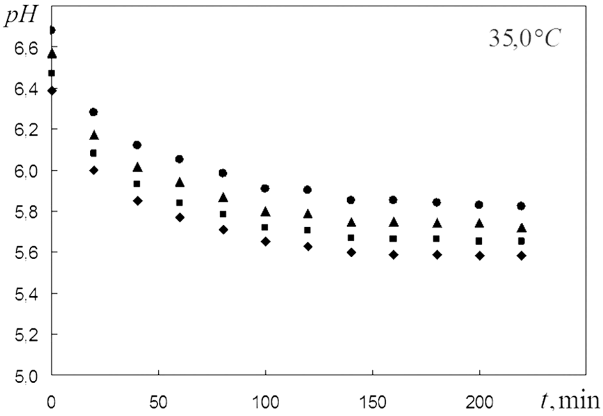

Fig. 1. Results of experimental measurements of the temporal dependences of the acid-base balance in aqueous sodium chloride solutions with various ion concentrations $\zeta = 180$ (♦), 215 (■), 270 (▲), and 360 water molecules per salt ion (•) at a temperature of 35 °C

The most general characteristics of living organisms remained almost the same, as they were formed in the primordial ocean. In our opinion, the most important of them are the following ones:

- the temperature interval in which such organisms exist;
- the optimal temperature for their vital functions;
- the interval of pH values or the concentrations of hydrogen ions arising at the dissociation of water molecules.

That is why our attention in this research was paid, first of all, to the values and variation intervals of the time τ_s required for the establishment of equilibrium pH values, their dependences on the salt concentration, and the pH dependence on ζ along every isotherm.

We emphasize that, in the framework of this approach, the temperature is taken into account as a general physical factor, and the pH reflects the influence of the electric fields of cations and anions of dissolved salts on the behavior of hydrogen ions, the most mobile cations in the solution. It should also be noted that monitoring the pH behavior in the carbon dioxide solution with available salts is an indicator of the effect exerted by the latter on the rates of physiological processes, in particular, the transfer of carbon dioxide and oxygen.

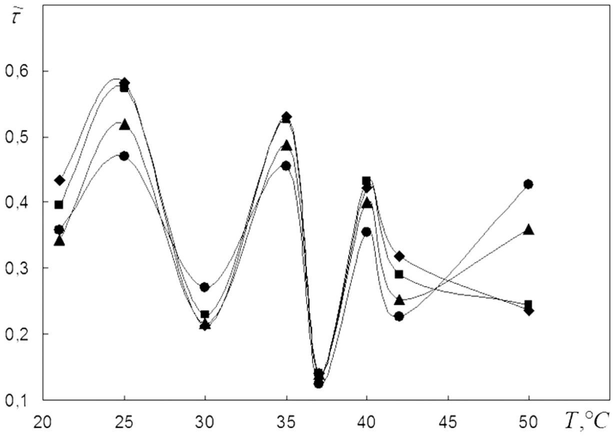

Fig. 2. Temperature dependences of the normalized pH relaxation time in aqueous NaCl solutions with various ion concentrations $\zeta = 180$ (◆), 215 (■), 270 (▲), and 360 water molecules per salt ion (●)

To analyze the temperature dependence of the pH relaxation time in NaCl solutions, let us change to the dimensionless quantities

$$\tilde{\tau}(T,\zeta) = \lambda(\zeta)\frac{\tau_s(T,\zeta)}{\tau_w(T)}, \tag{4}$$

where $\tau_s(T,\zeta)$ is the pH relaxation time in the sodium chloride solution with the inverse salt concentration ζ at the temperature T, $\tau_w(T) = \lim_{\zeta\to\infty}\tau_s(T,\zeta)$ is the pH relaxation time in water at the temperature T, and the constant $\lambda(\zeta)$ depends on the NaCl concentration in the solution and takes the values $\lambda(\zeta) = 5.17$, 4.30, 3.26, and 2.57 for the inverse concentrations $\zeta = 180$, 215, 270, and 360 water molecules per salt ion, respectively.

Figure 2 illustrates the temperature dependences $\tilde{\tau}(T,\zeta)$ of the normalized pH relaxation time in NaCl solutions with the examined concentration values. The analysis of this figure testifies that the curves $\tilde{\tau}(T,\zeta)$ are similar to one another and can be overlapped within the calculation error limits for the pH relaxation time. In our opinion, this fact has to be of crucial importance for the behavior of biochemical processes in an organism, because the essence of physiological processes is determined just by the mobility of hydrogen ions.

By its origin, $\tilde{\tau}(T,\zeta)$ [or $\tau_s(T,\zeta)$] reflects the dependence of the solution pH on the concentration of carbon dioxide in the atmosphere. It should be noted that the specific properties of atmospheric CO_2 are not used here. Therefore, we may assume that the quantity $\tilde{\tau}(T,\zeta)$ is the relaxation time of an arbitrary non-equilibrium value of the acid-base balance. This means that the quantity $\tilde{\tau}(T,\zeta)$ can serve as a basis for the description of relaxation processes in the blood plasma of mammals and humans. As one can see from Fig. 2, the relaxation process has several characteristic temperatures:

- $T_1 = 303$ K (30 °C), which plays the role of the lower limit for the existence of warm-blooded organisms;
- $T_2 = 310$ K (36.6 °C), which is the optimal temperature for the vital functions of warm-blooded organisms (the normal human body temperature);
- $T_3 = 315$ K (42 °C), which is the upper limit for the existence of human and most warm-blooded organisms.

Note that the dependence $\tau_w(T)$ determines the pH in water. This is the most important primary function, which affects the properties of all aqueous salt solutions, and its behavior is governed by the processes of thermal motion in water.

The main biochemical reactions that take place in human and mammalian organisms and provide their vital functions are coupled with the oxygen and carbon dioxide transport [14]. Such a transport would be impossible without the formation of non-equilibrium states that generate the corresponding flows of the oxidant and the final products of the reactions. It is clear that the intensity of vital activity would be higher, if the relaxation times of those non-equilibrium states were shorter. In this regard, we will consider, as optimal for life, those temperature and pH values at which the time required for the dissolution of atmospheric CO_2 is minimum.

Note that the minimum pH relaxation times in NaCl solutions are observed near a temperature of 36.6 °C. Since those minima are observed at all concentrations, and the concentration variation does not result in the shifts of the peaks, we may assert that the shortest lifetime reflects the specific features of thermal motion in the water itself. The statement about the establishment of pH equilibrium within the shortest time interval, if the temperature is in the vicinity of 36.6 °C, is in qualitative agreement with the conclusion made in works [10, 14, 15] that this temperature is optimal for the functioning of human and mammalian organisms. The authors of work [14] explain this feature as follows. On the one hand, at 36.6 °C and under the normal pressure, the isobaric heat capacity of water has a minimum, which is the most favorable for heat exchange in the bodies of mammals. On the other hand, the rate of oxygen transport by human vessels is maximum just at the temperature $T = 36.6$ °C [14]; so, the conditions are created for the intensity of biochemical processes at the cellular level to be maximum. The presence of minima for the pH relaxation time in sodium chloride solutions at the temperature $T = 36.6$ °C, which were revealed experimentally, is an extremely important fact for biomedicine and biophysics, because the rate of physiological processes is determined exactly by the mobility of hydrogen ions.

It was found that the optimal temperature for human vital functions is also determined by a characteristic function for water connected with the dissociation of water molecules. Near the temperature $T = 42$ °C, a peculiarity in water properties is observed, which is related to a substantial modification in the character of the thermal motion of molecules, namely, the vibrational motion of molecules (similar to that in crystals) transforms into a continuous thermal drift (similar to that in argon) [15–18]. This conclusion was drawn from the analysis of the temperature dependences of the following parameters: the kinematic viscosity of water [19], the time of the settled life of water molecules [18], the time of dipole relaxation of water molecules [20], and the entropy diameter on the liquid-vapor coexistence curve for water [17].

In works [15–18], it was shown that the character of the thermal motion of water molecules changes in a relatively narrow temperature interval. This circumstance allows this transformation to be interpreted as a dynamic phase transition in the system associated with substantial changes in both the translational and rotational modes of molecular motions, which are closely connected with modifications in the properties of the hydrogen bond network. Near the temperature $T = 42$ °C, this network decays into a set of small and short-living molecular associates.

We emphasize that the relaxation processes in NaCl solutions are combinations of the relaxation processes associated with hydrogen ions, as well as the relaxation processes occurring as a result of water-salt interaction.

Let us discuss the concentration dependence of the pH relaxation time in NaCl solutions. From the data in Table, it follows that the pH relaxation time in water is approximately 6 times longer than that in the saline with 180 water molecules per ion. The higher the salt concentration, the more distinct the manifestation of a certain oscillatory behavior of the pH relaxation time with the temperature variation. In particular, for water in this temperature interval, there is a maximum at $T = 35$ °C and a minimum near $T = 40$ °C. At the same time, for aqueous solutions

Temperature dependences of the pH relaxation time in aqueous NaCl solutions at various examined inverse solution concentrations

T, °C	pH(eq)	ζ	τ_s, min
21.0	5.67	180	68
		215	77
		270	86
		360	114
25.0	5.60	180	92
		215	113
		270	133
		360	152
30.0	5.52	180	37
		215	50
		270	60
		360	95
35.0	5.44	180	124
		215	153
		270	182
		360	216
37.0	5.41	180	28
		215	36
		270	45
		360	51
40.0	5.37	180	33
		215	42
		270	51
		360	57
42.0	5.34	180	34
		215	39
		270	43
		360	50
50.0	5.23	180	45
		215	58
		270	110
		360	165

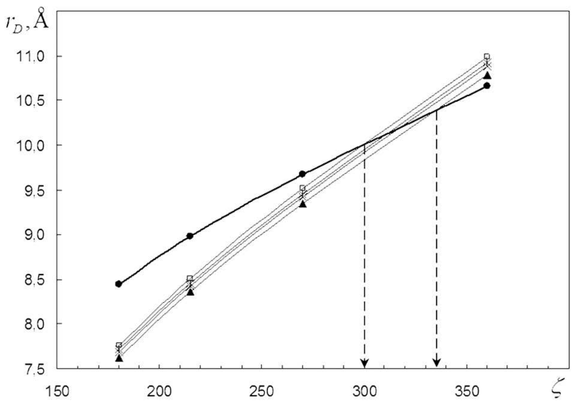

Fig. 3. Dependences of the Debye radius and the r_ζ-value (●) on the ion concentration ζ in aqueous NaCl solutions for various temperatures T = 25 °C (□), 35 °C (∗), 40 °C (×), and 50 °C (▲)

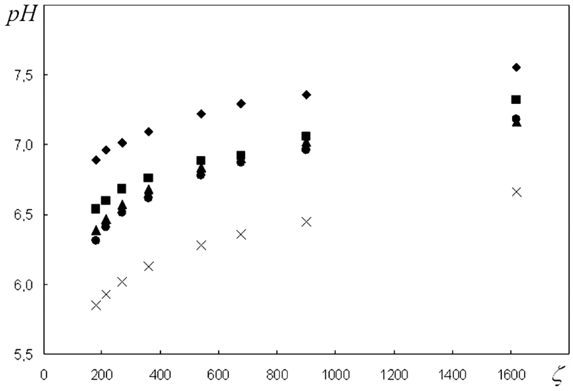

Fig. 4. Concentration dependences of pH in aqueous NaCl solutions at various temperatures T = 25 °C (◆), 30 °C (■), 35 °C (▲), 40 °C (●), and 50 °C (×)

of sodium chloride, there appears a local maximum at 25 °C; then, a local minimum at 30 °C; then, a local maximum at 35 °C; and so on.

On the other hand, despite that the pH relaxation time in NaCl solutions increases monotonically with the increasing salt concentration (within the calculation error of the pH relaxation time), this parameter demonstrates certain features. At the two-fold dilution of NaCl solutions (from 180 to 360 water molecules per sodium or chlorine ion), the rate of the pH relaxation time growth changes as follows:

- the minimum growth by a factor of 1.45 takes place at the temperature $T = 42.0$ °C,
- the maximum growth is observed at the temperatures $T = 30.0$ °C (by 2.6 times) and 50.0 °C (by 3.7 times),

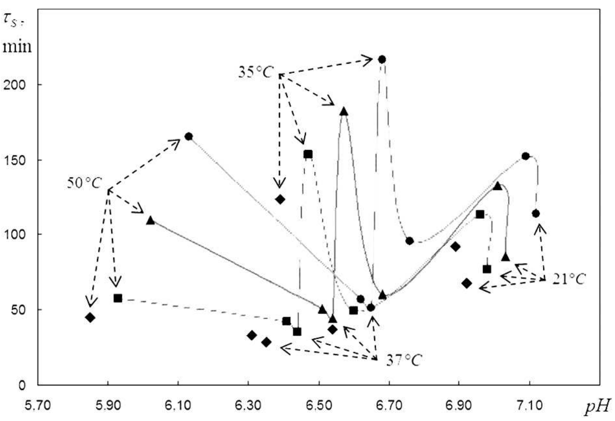

Fig. 5. Dependences of the pH relaxation time on the pH value in aqueous NaCl solutions for various inverse ion concentrations ζ = 180 (◆), 215 (■), 270 (▲), and 360 water molecules per salt ion (●)

- an average growth of 1.7 times is observed for other examined temperatures.

Hence, the following conclusions can be made. It is water that determines the main properties of the temperature dependence of the pH relaxation time in aqueous solutions of sodium chloride. At the same time, the presence of NaCl salt is responsible for the appearance of local peculiarities and characteristic features in the temperature dependence of the pH relaxation time in aqueous NaCl solutions.

It is worth noting that water molecules are less stable in electrolytes than in water, because the electric fields created by the salt ions contribute to the dissociation of water molecules. Therefore, while discussing the influence of the NaCl concentration on the solution pH, it is necessary to bear in mind that the electric fields of ions can change the equilibrium characteristics of electrolyte solutions. Let us consider, in more details, the influence of the electrostatic interaction between the ions on the behavior of molecular processes in aqueous NaCl solutions.

5. Qualitative Analysis of the Influence of Electrostatic Interaction Between Ions on Molecular Processes in NaCl Solutions

It is reasonable to begin a qualitative analysis of the influence of electrostatic fields created by salt ions on the establishment of the equilibrium pH values in aqueous sodium chloride solutions by comparing the average distances between the salt ions in the solution (they can be evaluated from geometric considerations) with the average distances at which the electrostatic field of charged salt ions is effective. The former parameter can be calculated as follows: $r_\zeta = 0.5\zeta^{1/3}r_w$, where r_w is the diameter of the water molecule. To evaluate the latter one, let us calculate the Debye radius for ions in aqueous NaCl solutions. Relevant theoretical considerations and calculation relations can be found in work [1]. Figure 3 illustrates the calculated Debye radii (in angströms) for various inverse concentrations ζ and temperatures T.

If the Debye radius is larger than r_ζ, then the Debye spheres of the ions begin to overlap (see Fig. 3). From Fig. 3, one can see that this is the case for the studied concentration values. Moreover, the concentration at which the Debye spheres begin to overlap depends on the NaCl solution temperature and increases as this temperature grows. For instance, the Debye spheres begin to overlap at the inverse NaCl concentration of $\zeta = 300$ if $T = 25$ °C, and at $\zeta = 335$, if $T = 50$ °C. Therefore, within the analyzed interval of salt concentrations in the solution, the electrostatic field of charged ions affects the processes of establishing the equilibrium pH values in aqueous NaCl solutions.

So, we see that the electric field created by ions in the solutions can considerably change the value of $\tau_s(T,\zeta)$. The corresponding modification occurs, because this electric field substantially lowers the dissociation barrier for surrounding water molecules in the nearest environment of salt ions,

$$H_2O \rightleftharpoons H^+ + OH^-, \tag{5}$$

which leads to the concentration growth of hydrogen ions. As a result, the pH of the solutions increases with the increase of ζ (see Fig. 4), which agrees with the obtained experimental results.

Furthermore, the addition of NaCl leads to the appearance of a number of effects. The minimum of $\tau_s(T,\zeta)$, which is observed in water at $T = 42$ °C, shifts toward lower temperatures – namely, to 36.6 °C, which is a characteristic temperature of human life – if the salt is added. The relaxation time minimum $\tau_s(T,\zeta) \approx 25$ min corresponds to the maximum rate of physiological processes, which is characteristic of the optimal temperature for human vital functions [14]. By order of magnitude, this relaxation time value is close to the expected one [14].

Also important are the results presented in Fig. 5, where the dependences of $\tau_s(T,\zeta(\mathrm{pH}))$ on the pH are illustrated. If the salt concentration is maximum ($\zeta = 180$), the minimum of $\tau_s(T,\zeta(\mathrm{pH}))$ is observed at the temperature $T \approx 37$ °C and pH ≈ 6.3. This pH value is less by one than pH(opt) = 7.35, which is typical of a healthy person. But this value is expectable as well, because all macromolecules of the albumin type [21–24], as well as acid additives, also affect the equilibrium pH values.

6. Conclusions

Temporal changes in the temperature and concentration dependences of the acid-base balance in aqueous sodium chloride solutions contacting with atmospheric carbon dioxide have been studied experimentally. The measurements are carried out in a temperature interval of 294–323 K for ion concentrations corresponding to 180, 215, 270, and 360 water molecules per sodium or chlorine ion.

The pH relaxation time in aqueous NaCl solutions with dissolved atmospheric CO_2 and its dependence on the temperature and the ion concentration are calculated. The analysis of the obtained temperature dependences of the pH relaxation time testifies that they are similar for various concentrations of NaCl ions in water.

The principle of natural selection of the optimal states of aqueous salt solutions characterized by the temperature and the irreducible component of pH has been formulated for the first time. Optimal are those states in which the pH relaxation time in aqueous salt solutions is minimum. On the basis of this principle, the interval of human vital activity is determined to extend from a temperature of (30 ± 2) °C to a temperature of 42 °C.

It is shown that the optimal temperature for the vital activity agrees with its actual value for humans and most mammals. In the absence of contributions from albumin and other proteins, the optimal pH value agrees with its actual value only qualitatively, with their difference being equal to one.

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ТЕМПЕРАТУРНІ ТА КОНЦЕНТРАЦІЙНІ ЗАЛЕЖНОСТІ ПОКАЗНИКА КИСЛОТНО-ЛУЖНОГО БАЛАНСУ ВОДНИХ РОЗЧИНІВ ХЛОРИДУ НАТРІЮ ПРИ РОЗЧИНЕННІ У НИХ АТМОСФЕРНОГО ВУГЛЕКИСЛОГО ГАЗУ

У роботі експериментально досліджено зміни в часі температурних і концентраційних залежностей показника кислотно-лужного балансу розбавлених водних розчинів хлориду натрію, які перебувають у контакті з атмосферним вуглекислим газом. Вимірювання проведено в інтервалі температур 294–323 K для обернених концентрацій йонів, які відповідають 180, 215, 270 і 360 молекулам води на один йон натрію або хлору. Знайдено часи релаксації показника кислотно-лужного балансу водних розчинів хлориду натрію при розчиненні у них атмосферного вуглекислого газу в залежності від концентрації солі та від температури. Сформульовано принцип відбору оптимальних станів водно-сольових розчинів, які характеризуються температурою і незвідною складовою показника кислотно-лужного балансу: оптимальними вважаються саме ті значення, які забезпечують мінімальне значення часу релаксації показника кислотно-лужного балансу. На основі цього встановлено температурний інтервал життєдіяльності людини, який простягається від (30 ± 2) °C до 42 °C.

Ключові слова: водний розчин, хлорид натрію, показник кислотно-лужного балансу, вуглекислий газ, час релаксації.